Mechanics of post-fry structural, thermal and physicochemical changes in batter-coated food

by

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DEDICATION

This thesis is dedicated to these my loved ones: Shahana Akter, my mother, foremost teacher. Iqra Yeasmen, my child ‘little-mike’. Nushrat Yeasmen, my best friend and sweet wife. Liya Akter, my cute sister. And the memories of Md. Liaquat Ali Bhuiyan, my father, foremost idol.

ABSTRACT

Batter-coating is an effective strategy to reduce fat in fried products. To achieve consumer satisfaction, maintenance of batter-coated fried foods quality at post-frying stage is of crucial importance. Understanding of the mechanisms relating to batter-coated fried foods quality, is prerequisite to customize its quality attributes. Getting mechanistic insights on post-fry structural, thermal and physiochemical evolution of batter-coated food, is the focus of this study.

Wheat and rice flour-based batter systems of different solid-to-water ratio (1:1.1, 1:1.3, 1:1.5) was used to coat a meat analog model food, and were deep-fried at 180°C for 2, 4 and 6 minutes in canola oil. Evolution of thermo-mechanical properties of fried foods were assessed by differential scanning calorimetry, mechanical texture analyzer, scanning electron microscopy and colorimetric analysis. Solid-to-water ratio (SWR) of batter coatings negatively correlated with batter pickup (%), water retention ability (%), cooking yield (CY, %), moisture and structural properties (thickness, densities) of fried foods crust. SWR positively correlated with frying loss (FL,%), fat content, textural attributes (hardness, brittleness, crispiness), and glass-transition-temperature (Tg, °C) of the crust. The FL, crust thickness, fat content, Tg, and textural attributes positively correlated with frying time (FT); whereas CY, moisture content and crust densities are negatively correlated with FT. Higher SWR and FT diminishes post-fry mass redistribution, textural evolution and color changes; whereas lower SWR and FT retains juiciness of food core substrate. Both SWR and FT have impacts on surface microstructure (roughness, micro-opening) of batter-coated fried foods.

Hot air blow (HAB), pressure assisted absorbent paper (PAP) and centrifuge assisted absorbent paper (CAP) treatments were employed to reduce fat from fried foods. CAP treatment reduced crust total fat (CrF) to the highest extent, followed by PAP and HAB. The PAP, CAP and HAB treatments reduced oil from fried products, without distorting textural and color traits. Surface fat (SF) content of coated product is dependent to both batter formulation and frying time. Surface fat of batter-coated fried foods are comparatively static and removable during post-fry holding. Post-fry de-oiling treatment reduces SF, matrix fat (CrMF) and crust total fat (CrF). Effectiveness of the de-oiling treatments are dependent to surface microstructure of fried foods.

Internal microstructure of batter-coated fried foods were characterized by X-ray µCT based Multifractal analysis. Textural attributes of fried foods were assessed by mechanical texture analyzer. Batter formulations and frying time impacts the evolution of micropores. Internal microporosity of fried coatings is positively correlated with fat content. Fried batters having higher internal micropores provided hard, brittle and crispy texture. Lower extent of post-fry moisture redistribution and higher textural stability were observed in porous, low moisture and high fat batter coated product. Singularity and Rényi spectra depicts that the internal structure (pore distribution) of coated fried food is heterogenous and possesses multifractal scaling behavior. Higher heterogeneity of pore distribution were observed in outer crust region of coated foods. Heterogeneity of pore distribution slightly diminishes with the increase of frying time.

Glass-transition behavior of fried batter coatings were assessed by differential scanning calorimetry (DSC). Batter-formulation greatly impacts the glass transition temperature (Tg) of fried batters. Tg of rice-flour based fried batters were higher than wheat-flour based batters. Tg of fried batters are positively correlated with frying time. Tg are negatively correlated with moisture. Both moisture and fat acts as plasticizer in fried-batters. Tg are positively correlated with microporosity (%) of fried batters. Post-fry textural stability was directly impacted by Tg of fried batters. Tg value of fried batters were ranged between -20°C to -24°C. The lower Tg value of fried batter explains their post-fry textural changes (hard to soft, brittle to ductile, crispy to soggy) at room temperature (~25°C) and under IR heating (~65°C).

Microwave heating (MH), infrared heating (IH) and air frying (AF) were evaluated as cooking methods of parfried-frozen foods and compared with deep fat frying (DFF). MH, IH, and AF reduced fat content compared to DFF. Cooking loss (%) and moisture profile of finish-cooked foods are impacted by both batter-formulations and finish-cooking methods. Evolution of key quality traits (texture, color) of parfried coated products are associated with batter-formulations and finish-cooking methods, where AF has been found as a suitable substitute of DFF. Tg of finish-cooked products were found in sub-zero temperature zone (between -20°C to -23°C). Post-finish-cooking stability of quality traits are linked with batter-formulations and finish-cooking method.

Mass-transfer, texture and color development during air frying (AF) of parfried frozen coated foods, are dependent on AF parameters (time, temperature) and batter formulation. Moisture of parfried-frozen product declines with AF time. Newton, Page, modified Page, Henderson & Pabis model could satisfactorily predict moisture-loss. Zero-order kinetic models satisfactorily characterized changes in textural and color traits. Rate constant of moisture, texture and color changes are positively correlated with AF temperature. Arrhenius model sufficiently characterized the temperature dependency of rate constants (k) and effective moisture diffusivity (D_{eff}). The k, D_{eff} and activation energy (E_a) are correlated with AF temperature and batter formulation. Surface microstructure (surface opening, fractal dimension) of air-fried products are impacted by batter formulation and AF parameter.

RÉSUMÉ

L’enrobage de pâte est une approche efficace qui permet de réduire les matières grasses dans les aliments frits. Pour obtenir la satisfaction des consommateurs, le maintien de la qualité des aliments frits enrobés de pâte est d'une importance capitale. L’obtention d’informations mécanistes sur les caractéristiques structurelles, thermiques et physicochimiques des aliments enrobés de pâte après la friture est au centre de cette étude.

Les résultats de cette étude ont révélé que le rapport solide-eau (SWR) des pâtes d’enrobage est négativement corrélé avec la quantité de pâte qui adhère à l’aliment (%), la capacité de rétention d'eau (%), le rendement de cuisson (CY, %), l'humidité et les propriétés structurelles (épaisseur, densités) de la croûte de pâte frite. Le SWR est positivement corrélé à la perte de friture (FL, %), à la teneur en matières grasses, à la texture (dureté, fragilité, croustillant) et à la température de transition vitreuse (Tg, °C) de la croûte. Le temps de friture (FT) a montré un impact positif significatif sur la FL, l'épaisseur de la croûte, la teneur en graisse, la Tg et les attributs texturaux ; tandis que l'impact était négatif sur CY, la teneur en humidité et la densité de la croûte. Un SWR et un FT plus élevés diminuent la redistribution de la masse après la friture, l'évolution de la texture et les changements de couleur ; tandis que SWR et FT réduits conservent la jutosité du substrat de base alimentaire. Le SWR et le FT ont eu des impacts sur la microstructure de surface (rugosité, micro-ouverture) des aliments frits enrobés de pâte.

Dans cette étude, des traitements par soufflage d'air chaud (HAB), papier absorbant assisté par pression (PAP) et papier absorbant assisté par centrifugation (CAP) ont été utilisés pour réduire la teneur en matières grasses des aliments frits. Les résultats ont montré que la graisse de surface (SF) du produit enrobé est corrélée à la fois à la formulation de la pâte et au temps de friture. La graisse de surface des aliments frits enrobés de pâte est relativement statique et amovible pendant la conservation après la friture. Les traitements de déshuilage post-friture ont réduit le SF, la graisse de la matrice (CrMF) et la graisse totale de la croûte (CrF). Le traitement CAP a le plus réduit le CrF, suivi du PAP et du HAB. Les traitements PAP, CAP et HAB ont réduit l'huile des produits frits, sans déformer les traits de texture et de couleur. L'efficacité des traitements de déshuilage est corrélée à la microstructure de surface des produits frits.

Cependant, la microstructure interne des aliments frits enrobés de pâte a été caractérisée par une analyse multifractale basée sur les rayons X µCT. Les formulations de pâte et le temps de friture ont un impact sur l'évolution des micropores. La microporosité interne est positivement corrélée avec la teneur en graisse. Les pâtes frites ayant des micropores plus élevés ont fourni une texture dure, cassante et croustillante. Une plus faible étendue de la redistribution de l'humidité après la friture et une plus grande stabilité de texture ont été observées dans les produits enrobés de pâte à frire poreux, à faible teneur en humidité et à haute teneur en matières grasses. Les spectres Singularity et Rényi représentent la structure interne (distribution des pores) du produit enrobé non homogène et possédant un comportement de mise à l'échelle multifractale. Une plus grande hétérogénéité de la distribution des pores a été observée dans la région de la croûte des produits enrobés. L'hétérogénéité de la distribution des pores diminue légèrement avec l'augmentation du temps de friture.

Le comportement de transition vitreuse des revêtements de pâte frite a été évalué par calorimétrie différentielle à balayage (DSC). La formulation de la pâte a un impact important sur la température de transition vitreuse (Tg) des pâtes frites. La Tg des pâtes frites à base de farine de riz est comparativement plus élevée que la Tg des pâtes à base de farine de blé. La Tg des pâtes frites est positivement corrélée avec le temps de friture. Les Tg sont corrélées négativement avec l'humidité. L'humidité et la graisse agissent comme plastifiant dans les pâtes frites. Les Tg sont positivement corrélées avec la microporosité (%) des pâtes frites. La stabilité texturale post-friture est directement impactée par la Tg des pâtes frites. Le valeurs de Tg des pâtes frites qui sont inférieures à zéro expliquent les changements de texture post-friture induits par la redistribution de l'humidité (durs à mous, cassants à ductiles, croustillants à détrempés) à la température de la pièce ou sous chauffage IR.

Le chauffage par micro-ondes (MH), le chauffage par infrarouge (IH) et la friture à l'air (AF) ont été utilisés comme méthode alternative de cuisson de finition des produits surgelés et semi-frits, et comparés à la grande friture (DFF). MH, IH et AF ont tous réduit la teneur en matières grasses par rapport au DFF. La perte de cuisson de finition (%) et le profil d'humidité des produits cuits de finition sont influencés à la fois par les formulations de pâte à frire et les méthodes de cuisson de finition. L'évolution des caractéristiques de qualité clés (texture, couleur) du produit enrobé semi-frit est associée aux formulations de pâte à frire et aux méthodes de cuisson de finition, où l'AF s'est avéré un substitut approprié pour la DFF. La Tg des produits finis cuits a été dans la zone de température inférieure à zéro. La stabilité des caractéristiques de qualité après la cuisson finale est liée aux formulations de pâte et à la méthode de cuisson.

Le transfert de masse, le développement de la texture et de la couleur pendant la friture à l'air (AF) d'aliments enrobés surgelés semi-frits dépendent des paramètres AF (durée, température) et de la formulation de la pâte. L'humidité du produit surgelé semi-frit diminue avec le temps de AF. Le modèle de Newton, Page, Page modifié, Henderson & Pabis pourrait prédire de manière satisfaisante la perte d'humidité. Les modèles cinétiques d'ordre zéro ont caractérisé de manière satisfaisante les changements de texture et de couleur. La constante de taux d'humidité, la texture et les changements de couleur sont positivement corrélés avec la température de AF. Le modèle d'Arrhenius a suffisamment caractérisé la dépendance à la température des constantes de vitesse (k) et de la diffusivité effective de l'humidité (Deff). Le k, Deff et l'énergie d'activation (Ea) sont corrélés avec la température AF et la formulation de la pâte. La microstructure de surface (ouverture de surface, dimension fractale) des produits frits à l'air est affectée par la formulation de la pâte et le paramètre AF.

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‘Read in the name of your lord who created’

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This thesis is manuscript-based following the thesis preparation guidelines of the Graduate and Postdoctoral Studies of McGill University and consists of six research manuscripts (Chapter 3, 4, 5, 6, 7, and 8). Md. Hafizur Rahman Bhuiyan is the principal author of the study. He is the Ph.D. candidate who executed experimental setup, design of experiment, analytical work in the laboratory, data analysis, preparing the manuscripts and thesis. Professor Michael Ngadi is the research supervisor, who provided supervisory guidance, resource and constructive comments in relation to the laboratory experiments and reviewing of the thesis.

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NOMENCLATURE

Abbreviation/ Acronym	**Meaning**
a*	Redness
AF	Air frying
ANOVA	Analysis of variance
AOAC	Association of official agricultural chemists
au	Absorbance unit
b*	Yellowness
BP	Batter pickup
CAP	Centrifuge assistant absorbent paper
CLSM	Confocal laser scanning microscopy
CO	Canola oil
CoF	Core fat
CoM	Core moisture
CoP	Core microporosity
CrF	Crust total fat
CrM	Crust moisture
CrMF	Crust matrix fat
CrP	Crust microporosity
CY	Cooking yield
D_0	Capacity dimension
D_1	Entropy dimension
D_1/D_0	Entropy dimension to capacity dimension
D_2	Correlation dimension
db	Dry basis
D_{eff}	Effective moisture diffusivity
DFF	Deep fat frying
Dq	Generalized fractal dimensions
DSC	Differential scanning calorimetry
DW	Distilled water
Ea	Activation energy

$f(\alpha_q)$	Hausdorff dimension
FD	Fractal dimension
F_{FL}	Finish frying loss
FL	Frying loss
FT	Frying time
FTIR	Fourier transform infrared
GLM	Generalized linear model
HAB	Hot air blow
HD	Holding time
HD	Holding time
IA	Index of asymmetry
IH	Infrared heating
K	Rate constant
L	Lightness
MC	Methylcellulose
MD	Distance at maximum force
MD*	Normalized value of force at maximum distance
MF	Maximum force to break
MF*	Normalized maximum force
MFA	Multifractal analysis
MH	Microwave heating
MR	Moisture ratio
NC	Non-coated
PAP	Pressure assistant absorbent paper
PCA	Principal component analysis
PL	Parfrying loss
q	Moment order
R^2	Coefficient of determination
RB	Rice flour-based batter
ROI	Region of interest
RWB	Rice & wheat flour-based batter

S	Slope value at maximum force
S*	Normalized value of slope at maximum force
SD	Standard deviation
SEM	Scanning electron microscopy
SF	Surface fat
SO	Surface opening
SPI	Soy protein isolate
SSE	Sum square error
SWR	Solid-to-water ratio
TF	Total fat
T_g	Thermal glass transition temperature
TM	Total moisture
TP	Total microporosity
WB	Wheat flour-based batter
WG	Wheat gluten
WHC	Water holding capacity
WRA	Water retention ability
X-ray µCT	X-ray micro computed tomography
α_q	Hölder exponent
$\Delta D= [D_{-8}-D_{+8}]$	Difference between $D_{-8}-D_{+8}$
ΔE	Total color difference
$\Delta f\alpha= [f\alpha_{min}–f\alpha_{max}]$	Difference of Hausdorff dimension
$\Delta\alpha=[\alpha_{max}-\alpha_{min}]$	Difference of Hölder exponent

CHAPTER 1

GENERAL INTRODUCTION

1.1 Background

Fried foods are very popular around the globe. However, presence of higher amount of fat (up to 50% of total weight) in fried food is a major health concern, due to its association with obesity, coronary heart diseases, diabetics and so on (Zhang *et al*., 2020; Devi *et al*., 2020; Liberty *et al*., 2019). Under this circumstance the need for fat-reduction strategies is inevitable, to lower health risks. The strategy of food surface modification by batter coating could play effective role, as frying is majorly a surface phenomenon (Liberty *et al*., 2019; Rahimi & Ngadi, 2014). Tempura type batter coatings create a uniform outer-layer over food surface that influence heat and mass transfer process during frying (Devi *et al*., 2020; Zhang *et al*., 2020; Liberty *et al*., 2019; Rahimi & Ngadi, 2014; Mellema, 2003). Batter coating is a liquid matrix containing essentially flour base and water. Wheat flour is mostly used in batter preparation. However, its associations with celiac disease, gluten sensitivity, gluten ataxia and wheat allergy act as thriving force for the effort to find a replacement. In this context, rice flour would be a potential alternative. The formulation of batter systems could include other ingredients such as hydrocolloids, gums, seasoning, egg, spices, herbs and so on. In addition to water and flour, tempura type batter coatings contain chemical leavening agents ($NaHCO_3$, $Na_4P_2O_7$) that create a puffed texture during frying (Voong *et al*., 2018; Nasiri *et al*., 2012; Adedeji & Ngadi, 2011; Xue & Ngadi, 2007).

Frying is a simultaneous heat and mass transfer process. During frying, as food losses moisture, it takes up oil. Hence, initial moisture content plays crucial role in fat content of fried products (Dehghannya & Abedpour, 2018; Rahimi & Ngadi, 2015; Adedeji *et al*., 2009; Mellema, 2003). Thus, modification of solid-to-water ratio (SWR) of batter systems might be a pragmatic way of modifying initial moisture content of coated food. SWR might influence not only oil reduction functionality of batter coatings but also the textural attributes, as both are associated with loss of moisture. In addition, SWR could impact post-fry evolution of physicochemical properties of batter coated fried foods, by impacting the probable post-fry mass redistribution between core and crust region. However, detailed information on this aspect is still lacking.

Recent studies have shown that bulk of oil in fried foods are absorbed during post-fry stage, than during frying. Post-fry stage starts as soon as fried food is removed from the fryer. At post-fry stage, adhered surface oil penetrates (due to suction action) the porous structure left by water molecule and enters into food matrix (Devi *et al*., 2020; Liberty *et al*., 2019; Patsioura *et al*., 2016; Cortés *et al*., 2015; Mellema, 2003; Bouchon *et al*., 2003). Hence, post-fry de-oiling (removal of surface oil immediately after frying) could be an interesting way of removing fat from fried product. Post-fry oil recovery will offer economic advantages as well as positive health impact.

The elements that determine fried foods qualities, exist at microlevel (Adedeji & Ngadi, 2009; Ross et al., 2002). Understanding of physicochemical properties at microscopic scale would set the ground for development of new and high quality fried products, as well as assist in maintaining quality at post-fry stage. Internal pore structure could influence physical properties as well as impact transport and storage of fluids within fried foods structure, as mechanical properties of foods are influenced by porosity, moisture content, density (Ross et al., 2002). To get insight on textural attribute (properties which is associated with mass transfer event), characterization of internal pore structure is of crucial importance for understanding their post-fry behavior. Spatial distribution, local densities, lack of homogeneity in pore structures could contribute differently, which needs to be considered in obtaining detailed and realistic information on internal pore characteristics of coated product in relation to any specific aspects of quality attributes. In this context, X-ray microtomography (µCT) could be a rational choice to obtained three dimensional details of internal structure of coated products, as it has the capability of components differentiation based on density difference without any physical slicing, staining procedure, destruction of test specimen etc. (Wang et al., 2018; Nicoläi et al., 2014). However, study on statistical characterization of internal micropores (obtained through X-ray µCT images) of batter-coated fried products in relation to mass and textural evolution, is still lacking. Multifractal approach could be used in analyzing and classifying of digital images having complex structure (Ampilova et al., 2019; Jung & Yoon, 2017) as multifractal analysis (MFA) could be used in studying spatial distribution of pores in a solid matrix which is linked to diffusion of fluids (Han et al., 2020; Srocke, 2019; Lafond et al., 2012; Mendoza et al., 2010). However, use of MFA in studying internal pore microstructure of tempura-batter coated fried product is a novel scope of research, and to the best of my knowledge, is scarce in scientific literature.

It is crucial to mention, apart from impacting fat, tempura-batter coatings develop a peculiar organoleptic characteristic in fried product i.e., juicy inside and crispy outside (termed as crust). Loss of textural quality of the crust of coated fried products, is the prime cause of its consumer rejection (Jothi *et al*., 2020; Carvalho & Ruiz-Carrascal, 2018; Pariya *et al*., 2007). In this aspect, post-fry changes in textural attributes could be likened to glass-transition phenomena, as the state of the product might became glassy due to decrease in water content during frying, and both water and oil could influences textural attribute of fried products (Jothi *et al*., 2018; Homer *et al*., 2014; Pariya, 2007; Kayacier & Singh, 2002). Thus, to control and predict textural changes of batter-coated fried products, research is required to understand the nexus between glass-transition, textural change, and moisture-fat distribution, under different circumstances.

Par-frying is a unit operation where foods are partially cooked by frying in oil at high temperature (to inactivate enzymes & microbes) and for shelf stability, Par-fried products are mostly stored as frozen, until finish cooking (Rady *et al.*, 2019; Adedeji & Ngadi, 2018; Raj *et al*., 2016). Parfried-Frozen products are generally finish-cooked by deep-fat-frying and the two stage frying (i.e., par-fry, finish-fry) favors the presence of high amount of fat in finish-cooked foods which is a serious health concern. Under this circumstance, use of alternate finish-cooking process would be a pragmatic way of enhancing oil reduction functionality of batter coatings. Emerging heat processing techniques such as Infrared heating (IH), Microwave heating (MH), Air frying (AF) could be considered as finish-cooking methods to produce fried products with less amount of oil, as a substitute for atmospheric deep-fat-frying of frozen-parfried food (Zaghi *et al*., 2019; Rahimi *et al*., 2018; Ghaitaranpour *et al*., 2018; Albert *et al*., 2014; Shaker 2014; Andrés *et al*., 2013; Chen *et al*., 2008). However, there is paucity of knowledge on applying these techniques as finish-cooking method and their impact on quality attributes of batter coated food, in comparison to deep-fat-frying.

Coated products are composed of a food core-substrate and a batter system. The presence of a food core substrate is essential, where crust formation in coated products would be realistic (Shokrollahi *et al*., 2019). However, different characteristics such as moisture content, porosity, surface roughness etc. of food core substrate decrease the possibility of reproducible production of batter-coated fried products (Shokrollahi *et al*., 2019, Rahimi *et al*., 2019; Ansarifa *et al*., 2012; Visser *et al*., 2008). In these aspects, use of plant-protein based a restructured meat-analog model food system, as core substrate would be a pragmatic choice. In batter coated product, unexpected change in structural, thermal, and physiochemical attributes might happen during post-fry stages, although their mentioned attributes were quite good immediately after frying. Therefore, maintaining the physicochemical and thermal properties (that developed during frying) of batter coated products at post-fry stage, is crucial for achieving its consumer satisfaction. In this aspect, detail study on understanding the post-fry behavior of batter coated food is the demand of time.

1.2 Hypothesis

Batter formulation (flour, solid to water ratio), frying time, post-fry holding environment, post-fry holding duration, post-fry surface de-oiling technique and finish-cooking method influence the evolution of structural, thermal and physicochemical properties of batter-coated food.

1.3 Overall objective

The overall objective of this study is to understand post frying structural, thermal and physicochemical behavior of batter-coated food; by using a meat-analog model (as core substrate) and different techniques such as scanning electron microscopy (SEM), X-ray microtomography (X-ray µCT), ATR-FTIR spectroscopy, differential scanning calorimetry (DSC), mechanical texture analysis, rheometry, colorimetry, multifractal analysis, etc. The outcomes of this study would be useful in maintaining the quality of batter coated foods, during post frying handling.

1.4 Specific objectives

This research is focused on understanding and characterization of post-frying behavior of batter coated food product. The specific objectives of this study are as follows:

1. To study the influence of batter solid-to-water ratio and frying time on structural, thermal and physicochemical evolution of coated fried food.

2. To investigate the impact of post-fry de-oiling treatments on oil-redistribution in batter-coated fried food.

3. to characterize the relationship between internal microstructure, mass redistribution and textural evolution of batter-coated fried food.

4. To study the glass-transition behavior of batter coated fried foods in relation to textural evolution.

5. To study the impact of finish-cooking methods on structural, thermal and physicochemical evolution of parfried frozen batter-coated food.

6. To study the kinetics of mass, texture and color evolution during the air-frying of parfried frozen batter-coated food.

CHAPTER 2

LITERATURE REVIEW

2.1 Batter

Batter is a liquid matrix containing essentially flour base and water, into which a piece of food is dipped before frying (Martínez-Pineda et al., 2020; Zhang *et al*., 2020; Rahimi *et al*., 2019; Román *et al*., 2018). In addition to oil reduction by forming a continuous, uniform layer over the food substrate; batter may contain chemical leavening agents to create a puffed texture during frying (Mesias et al., 2020; Martínez-Pineda et al., 2020; Voong *et al*., 2018; Adedeji & Ngadi, 2011). Hence, batter coating is a method for preparing crispy fried product with less oil content, and value addition in vegetable, chicken, and fish based fried foods by modifying physicochemical attributes as well as improving cooking yield (Rahimi *et al*., 2019; Rahimi & Ngadi, 2016; Adedeji & Ngadi, 2011). Batter coating acts as a barrier to mass-transfer during frying (prevents moisture loss and reduces fat uptake), ensuring a final product that is tender and juicy on the inside and at the same time crispy on the outside (Mesias et al., 2020; Román *et al*., 2018; Carvalho & Ruiz-Carrascal, 2018). Batter coating also act on modifying product color, due to changes associated with non-enzymatic reactions involving sugars and proteins in coating materials (Ngadi *et al*., 2006).

2.2 Batters ingredients

Flour and water are the two main ingredients of any batter system and for tempura-type batter, presence of leavening agent (to create puffed texture) is the only required additional item (Zhang *et al*., 2020; Rahimi et al., 2019; Román et al., 2018; Carvalho & Ruiz-Carrascal, 2018; Yılmaz *et al*., 2017; Rahimi & Ngadi, 2014a; Adedeji & Ngadi, 2011). Mostly used leavening agents is the Na-compound such as Sodium bicarbonate ($NaHCO_3$) known as baking soda, and another is Sodium pyrophosphate ($Na_4P_2O_7$). It is noteworthy, while these two compounds have been used in batter preparation simultaneously, they were seemingly in a constant ratio i.e., $Na_4P_2O_7$:$NaHCO_3$ =1.78%: 1.32% of dry batter ingredient. In addition, use of NH_4HCO_3 and other salts in batter preparation is not unusual (Aykn et al., 2016). In practice, batter formulation is extremely flexible allowing for maximum adaptation to food product development, and list of batter ingredients has expanded with the addition of hydrocolloids, gum, seasoning, egg, starch, table salt, colorants, and other minor ingredients, which makes batter coating as a technological art (Carvalho & Ruiz-Carrascal, 2018; Voong *et al*., 2018; Adedeji & Ngadi, 2011). The fact that needs to be considered in formulating and applying batter coating is that; batter is a complex system in which their composition, concentration, and ingredients characteristic determines the final performance of coating on mass and heat transfer during frying (Nasiri *et al*., 2012; Sahin & Sumnu, 2009). It is important to mention, Mesias et al., (2020) studied the process contaminants in battered foods prepared at public food service establishments and reported that increase in complexity of batter formulation through the introduction of ingredients (aimed to improve palatability) leads to the development of heat induced process contaminants i.e., Acrylamide, hydroxymethylfurfural (HMF), and furfural. Hence, increase of harmful compounds during thermal treatment of coated product should be carefully considered during the formulation of batter coatings.

2.2.1 Water

Water is the core component of any batter system and its proportional contribution in preparation of batter coating is > 50% of its final total weight (Rahimi *et al*., 2019; Rahimi & Ngadi, 2016; Adedeji & Ngadi, 2011; Xue & Ngadi, 2009). Preferably, chilled water is used in preparing batter systems to avoid any unexpected changes (due to presence of leavening agents) in batter properties until frying and to achieve optimum batter functionality (Rahimi et al., 2017a; Xue & Ngadi, 2009, 2007). Water in batter preparation plays crucial role by impacting batter rheology and modifying the initial moisture content of coated food, consequently, impacts the quality attributes of fried product. Initial moisture content of batter coated food could influence the final fat content of fried products, as frying is a simultaneous heat and mass transfer process where the food losses moisture and uptakes frying oil. In this context, initial moisture reduction by (pre-)treatment such as drying/microwave heating etc. could be used to decrease fat content in fried products. Rahimi & Ngadi (2014) used pre-drying as a method for reducing initial moisture content of batter systems and reported that pre-drying decreased the amount of fat in fried batter coatings, compared to non-pre-dried batters. Longer pre-drying times showed greater fraction of spaces occupied by air, and smaller fraction of spaces occupied by moisture and fat, in fried batter systems (Rahimi & Ngadi, 2014). Ngadi et al., (2009) used Microwave (MW) heating as a method of initial moisture reduction from batter coated chicken nugget, and reported that, MW pre-treatment has significant effect on moisture loss and oil uptake of chicken nuggets during frying, where longer MW treatment reduced higher amount of moisture and consequentially uptakes less oil during frying.

2.2.2 Flour

The use of wheat flour in batter preparation is mostly reported (Pinkaew & Naivikul, 2019). The prominent use of wheat flour is due to its high protein (gluten) content, which helps to prepare a batter with unique viscoelastic properties, which favor the desired functionality of batter coating i.e., lower moisture loss, lesser fat uptake, and smoother crust surface (Oke *et al*., 2018). However, use of wheat flour is under question, as it is associated with health issues (due to gluten) such as celiac disease, allergy, etc. (Jackson et al., 2006). In this context, incorporation of gluten-free flour is suggested (Davarcioglu & Kolsarici, 2019; Pinkaew & Naivikul, 2019; Sansano *et al*., 2018; Jackson *et al*., 2006) and the use of rice, corn, rye, and soy flour as well as blend of flours in batter preparation has been practiced, in recent time. However, the type of flour used in batter preparation plays very crucial role in multiple aspects of batter coated fried food production. Adedeji & Ngadi (2009) reported, the proportions of rice or wheat flour in the batter preparation influences batter viscosity, batter pickup, as well as porosity, color, texture, moisture, and fat content of the fried batter system. In a comparative study, it is observed that fried batters with higher wheat flour showed higher moisture content, lower fat content, and lower porosity than fried batters with higher rice flour (Rahimi & Ngadi, 2016). However, Rahimi & Ngadi (2015) observed that total number of generated ruptures on the surface of fried batters was significantly influenced by the type of flours used in batter preparation. Their study reported that, only wheat-flour based fried batter showed highest number, whereas only rice-flour based fried batter showed least number of holes & cracks. Higher area of generated holes and cracks on fried surface was observed for the batter formulation that contains higher amount of rice flour. Rahimi & Ngadi (2014) reported, using higher amount of wheat flour in batter significantly decreased the fat uptake during frying, whereas higher rice flour content in batter significantly increased the fraction of inter-particle spaces occupied by air (SOA), and decreased fraction of spaces occupied by moisture (SOM) in fried batter.

2.2.3 Hydrocolloids/gel forming agents

Now a days, use of hydrocolloids in batter preparation is very common. Hydrocolloids are used in batter formulation to control viscosity, batter pickup, and water holding capacity (Liberty *et al*., 2019; Oke *et al*., 2018; Adedeji & Ngadi, 2011). This helps to create a network of structures that minimizes mass-transfer during frying, resulting in reduced oil uptake and the formation of a desirable texture (Liberty *et al*., 2019; Oke *et al*., 2018; Xue & Ngadi, 2009, 2007). Hydrocolloids such as MC (Methylcellulose), CMC (Carboxymethyl Cellulose), HPMC (Hydroxypropyl methylcellulose) is mostly used in batter preparation, as these cellulose derivatives are water soluble with good film-forming properties and forms gels when heated but return to their original viscosity when cooled. This gelation is what promotes the barrier-resistant effect to oil uptake and moisture loss during frying (Zhang et al., 2020; Liberty *et al*., 2019; Oke *et al.*, 2018; Xue & Ngadi, 2009 & 2007). A number of studies have reported that addition of MC and HPMC in batter has successfully reduced oil uptake in coated fried products such as chicken pieces, fish, vegetables, cheese, and cereal products (Xue & Ngadi, 2007). In literature, use of these hydrocolloids are reported by several authors, where their functionalities and performances varied based on other factors**.** It is evident, use of hydrocolloids in batter is not limited to MC, CMC or HPMC, as the use of pectin, starch and dextrin are also well mentioned. The list of hydrocolloids includes gums such as Xanthan gum, Gum Arabic and Guar gum, as well as protein isolates such as soy protein isolate and whey protein isolate. The list of ingredients, which play role in mass transfer phenomena during frying of batter coated foods are expanding day by day and recently the use of sodium alginate, carrageenan and chitosan have also been reported (Liberty *et al*., 2019; Oke *et al*., 2018; Naghavi *et al*., 2018; Martin Xavier *et al*., 2017). Selection of ingredients for preparing batter is becoming more and more complex day by day, as different hydrocolloids/gel forming components behave differently while they are used in combination with other batter ingredients (Liberty *et al*., 2019). However, low amounts of hydrocolloids (MC, CMC, HPMC, Gums etc.) were used in batters i.e., 0.2-1.5% of dry batter ingredients, and was sufficient to endow desired batter functionality. The performance of batter might be affected by its wetting properties associated with interfacial tension and become a mechanical barrier to lipids during frying (Liberty *et al*., 2019). The flour types have ability to influence performance of hydrocolloids and performance of hydrocolloids is product-dependent; for example, in comparison to CMC, HPMC was more effective in fat reduction of fried Green gram splits (*Vigna radiata*), whereas CMC outperformed HPMC when chickpea flour was used (Liberty *et al*., 2019; Xue & Ngadi, 2009).

2.2.4 Salt

Salt is an ingredient that is frequently used in preparation of batter coating as it plays dual role i.e., flavor enhancer and batter stabilizer (Xue & Ngadi, 2007). In addition to sensorial aspects, salt affects the rheological properties by modifying the water holding capacity of the batter systems. Xue & Ngadi (2006) reported that addition of salt (2.5%) to the flour (wheat, rice, corn) based batter formulations lowered their viscosity, and decreased storage moduli (G'_{max}) and loss moduli (G''_{max}) of the formulated batter systems. In addition to rheological property, salt greatly influence the thermal properties of batter systems. Xue & Ngadi, (2007) reported that salt in batter systems increased their gelatinization temperature (T_G) and depressed melting temperature (T_m). Batter system formulated with salt have shown a lower glass transition temperature (Tg) compared to the batter without salt (Xue & Ngadi, 2007). NaCl have been frequently used in batter preparation and the concentration of salt in batters are reported as within the range of 1-5.5 % of total dry weight of batter ingredient, however, most preferable concentration was $\leq$ 3% of total dry batter ingredient. In selecting the salt and its concentration for batter formulation should take into consideration the taste preference of consumer group as well as its impact on health. It is notable, salt not only have impact on sensory property but also affects mass-transfer process via osmosis principle as well as by boiling-point elevation, which might impact physical characteristics of batter coatings during frying. Salt concentration might play important role in dielectric e.g., microwave (re-) heating of batter coated product.

2.2.5 Miscellaneous

The use of sweetening ingredients like sugar and sugar-alcohol in batter coating is appearing as an effective approach to modify batter properties, as these ingredients have shown impact on leavening activity and porosity of batter coating upon frying, where CO_2 plays vital role (Carvalho & Ruiz-Carrascal, 2018; Rossi Marquez et al., 2014; Shih *et al*., 2010; Mukprasirt *et al*., 2000). The use of yeast in batter to achieve the functionality of puffing and use of egg or egg white powder in batter systems are also reported. In recent time, use of herb, spices or their extract in batter preparation is becoming popular form health point of view. It is notable, incorporation of ingredients is intertwined with achieving any specific functionality of used batter coatings.

2.3 Batters characterization

Rheological attribute is crucial to characterize the raw batter systems, as rheological properties affect the amount of batter to be picked up by food core substrate. And in case of insufficient batter pickup (BP) or weak adherence of batter coating to food substrate, the applied batter coating might not be able to properly serve the desired purpose of its use i.e., reduction of fat content in fried product. The BP of batter systems largely depends on their flow behavior (viscosity) and could be impacted by temperature, composition, amount of ingredients in recipe, and solid-to-water ratio; for example, wheat-flour based batter had shown higher wet pick-up than rice-flour based batter (Pinkaew & Naivikul, 2019; Yılmaz *et al*., 2017)**.**

2.3.1 Batter pick-up: Batter pick-up (BP) is used to denote the amount of batter that adhered to the piece of food, where the yield and quality of final fried product depend upon it (Mahdavian et al., 2016; Labropoulos et al., 2013; Yılmaz *et al*., 2017; Pinkaew & Naivikul, 2019) and calculated as follows:

$$\text{Batter pick-up, BP (\%)} = \left(\frac{\mathrm{CWT-UWT}}{\mathrm{UWT}}\right) * 100 \qquad 2.1$$

Where, CWT and UWT respectively represent weight after and before batter coating.

2.3.2 Water holding capacity: The water holding capacity (WHC) of a batter system depict its ability to retain its water content, while the batter passes through a vigorous process. The WHC of batter systems is generally evaluated via centrifugation process as such: specific amount of raw batter is put into a centrifuge tube, and the batter-containing tube undergo vigorous centrifugation, after centrifugation process, the supernatant is being separated and measured (Saleh, 2018; Yılmaz *et al*., 2017). WHC of batter is calculated as the percentage (%) of water released from batters, based on weight difference [i.e. (before centrifuge weight-after centrifuge weight) / before centrifuge weight]. Batter WHC affect moisture retention ability and consequent resistivity to fat uptake during frying. Chayawat & Rumpagaporn (2020) reported that addition of fortified defatted rice bran in batter formulation increased batters' water retention ability and consequently reduced oil content in fried chicken nugget. Yılmaz *et al*., (2017) reported that when hydrocolloid concentration in batter formulation increases, the water holding power of hydrocolloid chains rises, that consequentially results into higher WHC of batter systems. Water holding capacities of methylcellulose (MC) containing batter is found higher than hydroxy propyl methyl cellulose (HPMC) containing batter (Yılmaz *et al*., 2017).

2.3.3 Flow behavior: Viscosity plays critical role in characterizing flow behavior of batter coating. Apparent viscosity of batter could be measured by viscometer (Pinkaew & Naivikul, 2019). The flow behavior index (n) and consistency index (k) values of the batter coating is computable by applying power law model i.e., Ostwald de Waele Model (Yılmaz *et al*., 2017; Mahdavian *et al*., 2016; Soorgi *et al*., 2012) as follows:

$$\tau = K\gamma^{n} \qquad 2.2$$

where, τ, γ, K and n respectively represents shear stress (Pa), shear rate (s^{-1}), consistency coefficients (Pa s^n), and dimensionless flow behavior index; for example, "n" value below 1 indicates batter as "pseudo-plastic" substance (Yılmaz *et al*., 2017) and "n" value less than 1 represents "shear-thinning" behavior of batter system (Xue & Ngadi, 2007a).

It is notable, batters' flow behavior could be characterized by other models than the frequently used "power law". For example: Xue & Ngadi (2007a) stated that Herschel-Buckley model (Eq 2.3) adequately (R^2>0.87) describes the experimental data for flow curves of batter systems containing different combinations of flours and hydrocolloids. Their study showed that, the types of flour and hydrocolloid used in formulating batter significantly influences the flow behavior index (n) of batter system, where an increasing concentration of hydrocolloid lowered 'n' values and increased shear-thinning behavior of the batters. Addition of hydrocolloid (methylcellulose, xanthan gum) increases the consistency index (K) value in rice flour-based batters much more than in wheat-or corn flour-based batters. Methylcellulose (MC) and xanthan gum (XG) develops viscosity in batter systems and reduces the amount of available free water, that subsequently increases the yield stress required to initiate flow in the flour (wheat, rice, corn) based batter systems. The XG showed a higher effect on batter yield stress than the MC; and addition of XG lowered the n values, imparting a higher degree of pseudoplasticity to the batters compared to MC. Sansano *et al*., (2018) used Herschel-Buckley (HB) model to characterize the properties of rice and wheat flours-based batters and reported that, increasing replacement of wheat-flour by rice-flour decreases the consistency index and yield stress, while increasing flow behavior index; and the addition of chitosan to rice-flour based batter enhanced viscosity and consistency. Martínez-Pineda et al., (2020) used HB model to examine the effect partial ingredient substitution in tempura batter with maltodextrin and ethanol on the rheological characteristics; and reported that ethanol containing batted had a significantly higher consistency index, lower yield stress and flow index, than standard batter and maltodextrin containing batter. Cengiz & Dogan (2021) reported that HB model provides a good fit for the flow behavior of starch-hydrocolloids (CMC, xanthan gum, guar gum) based batters, where the hydrocolloid addition changes in apparent viscosity of batters at 25°C, and CMC added batter had the highest viscosity, followed by xanthan and guar.

$$\sigma=\sigma_0 + K\gamma^n \qquad 2.3$$

here σ is shear stress (Pa), σ_o is yield stress (Pa), K is the consistency coefficient (Pa s^n), γ is shear rate (1/s), and n is the flow behavior index (dimensionless).

Temperature dependency of consistency coefficient (i.e., indicator of viscous nature) is well assessable by Arrhenius model (Yılmaz *et al*., 2017; Mahdavian *et al*., 2016) as follows:

$$K=K_0 e^{-Ea/RT} \qquad 2.4$$

Where, k_0 represent proportionality constant (or consistency coefficient at a reference temperature, Pa.s^n), E_a is activation energy (J/mol), R is universal gas constant (J/mol K), T represent absolute temperature (K).

Linearization of equation (3) is proposed by Yılmaz *et al*., (2017) as equation (2.5); where, K_0 and Ea could be calculated from ln K versus $(\frac{1}{To} - \frac{1}{T})$ plot.

$$\ln K = \ln K_0 - \frac{Ea}{R}.\left(\frac{1}{To} - \frac{1}{T}\right) \qquad 2.5$$

2.3.4 Viscoelastic properties

Just like most foods, batter system might possess both liquid and solid properties and could be termed as viscoelastic materials (Okonkwo et al., 2021; Xue & Ngadi, 2007a). Determination of viscoelastic behavior often requires oscillatory rheological measurements to be conducted within a boundary (known as linear viscoelastic region, LVR) where the properties of viscoelasticity [i.e., storage modulus (G′), loss modulus (G″)] are unaffected by stress/strain (Okonkwo et al., 2021). Xue & Ngadi (2007a) studied the effects of hydrocolloids (xanthan gum, methylcellulose) on dynamic viscoelastic parameters of formulated batters, as function of temperature of the batter systems. Their study reported that xanthan gum and methylcellulose changed the onset temperature (T_{on}) of structure development, storage moduli (G'_{max}), and loss moduli (G''_{max}) of the batter systems. Xanthan gum increased both G'_{max} and G''_{max}, whereas, at higher concentrations methylcellulose increased G'_{max} but lowered G''_{max}. Xue & Ngadi (2007a) also reported that, a higher temperature and shorter time were required to gelatinize starch when the hydrocolloids were added to batter systems.

Jeong & Lee (2021) studied the viscoelastic properties of batters made from wheat and brown rice flour blends. Their study reported that a higher proportion of brown rice flour in the blends caused their viscoelastic parameters to increase, contributing to a more elastic nature. Cengiz & Dogan (2021) used dynamic rheological tests to assess the effect of corn starch-hydrocolloids (CMC, xanthan gum, guar gum) interactions on rheological properties of batter coatings. They observed a clear difference between storage moduli (G′) and loss moduli (G″) values of studied batter samples. They also reported that addition of hydrocolloids in batter systems makes them more gel like structure as the increment of G′ was always greater than G″. It was also reported, after 55°C, all batters showed thermo-gelling ability and at temperature range of 70–80°C all the studied batter showed highest complex viscosity values.

2.4 Batters mechanism of action

Frying is a simultaneous process of heat and mass transfer, that results in changes of food structure (Adedeji *et al*., 2011; Mellema, 2003). To the context of oil absorption during frying, several studies mentioned three major mechanisms i.e., water replacement, cooling phase effect, and surface-active agent theory (Ching et al., 2021; Jeong et al., 2021; Liberty et al., 2019; Ziaiifar et al., 2008). Water replacement mechanism demonstrates the oil uptake in food during frying as the outcomes of its moisture loss and pore formation. In surface-active theory, it is pointed that during frying in oil, different chemical reactions (hydrolysis, polymerization, oxidation) occurs that results in the formation of volatile and non-volatile compounds. These compounds cause reduction in surface tension between food and frying oil, which results in higher fat uptake during frying. Cooling-phase effect mechanism demonstrates the oil absorption in fried foods as the event of oil-migration from surface to inside. It is notable, microstructure (especially surface topography) of food influences oil uptake during frying, as both the heat and mass exchange between frying-oil and food occurs through the surface (Jeong et al., 2021; Ching et al., 2021; Liberty et al., 2019). Hence, the batter coating over a food substrate modifies the foods surface properties, which consequently impacts the mentioned mechanisms of oil absorption in fried food. In general, batter coating contributes in modifying the overall heat and mass transfer characteristics of the coated food (Zhang *et al*., 2020; Zaghi *et al.,* 2019; Xue & Ngadi, 2007; Ngadi *et al.*, 2006). Application of heat on batter coating during frying (which is the process of drying and cooking foods through contact with hot oil at a temperature above the boiling point of water) creates 3D-network structure resulting as crust of the fried product (Rahimi & Ngadi, 2016; Adedeji *et al*., 2011; Nasiri *et al*., 2012; Akdeniz *et al*., 2005). The crust that is developed as a result of moisture loss, are relatively impervious to the movement of moisture and oil; and provides a brittle texture with less oil content (Rahimi *et al*., 2019; Román *et al*., 2018; Carvalho & Ruiz-Carrascal, 2018; Nasiri *et al.*, 2012; Akdeniz et al., 2005). The ability of batters to form a crust is enhanced by the high initial amount of coating that adheres to food core, and the nature of the ingredients used as well as their interaction determines the final performance (Akdeniz *et al*., 2005). In addition to crust formation, protein denaturation and starch gelatinization in batter coating during frying are the other causes of oil transfer prevention; as changes in the structure of protein and starch develop a film that blocks oil transfer (Rahimi *et al*., 2019). However, due to versatility in batter ingredients functionality as well as batter coatings formulation, their overall mechanism of action might differ; nonetheless the core concept of impacting heat and mass transfer process during frying would remain unchanged.

2.5 Modeling of batter coated food production

Frying of batter coated food is a dehydration process that requires simultaneous heat and mass transfer; where mass transfer is characterized by dynamics moisture loss from food and fat uptake into food (Dehghan Nasiri *et al*., 2011). Moisture loss is considered as a diffusion-controlled process, and diffusion (Fick's law) equation is generally used to model mass-transfer phenomena during deep-fat-frying which is pertinent to batter coated food (Rahimi & Ngadi, 2014; Dehghan

Nasiri *et al*., 2011; Adedeji *et al*., 2009). Fick's law detailing diffusion-controlled process is expressed as follows:

$$\frac{\partial}{\partial \mathrm{L}}[\mathrm{D_{eff}}\frac{\partial \mathrm{M}}{\partial \mathrm{L}}]= \frac{\partial(\mathrm{M})}{\partial \mathrm{t}} \qquad 2.6$$

Where Deff. is effective diffusivity (m^2/s), M is instantaneous, initial and equilibrium moisture content (db), L is thickness of sample (m), t is time (s). Assuming (i) an infinite slab model because of smaller thickness of samples compared to the other dimensions (ii) an initial uniform distribution of moisture and temperature in the samples (iii) minimal shrinkage and gradients in temperature and moisture (iv) negligible external resistance to mass transfer and (v) mass transfer from both side of the samples, solution of the above partial differential equation was presented by Crank (1975) and reported in several relevant studies (Rahimi & Ngadi, 2014; Soorgi *et al*., 2012; Dehghan Nasiri *et al*., 2011; Adedeji *et al*., 2009) as following:

$$\mathrm{M_r}= \frac{(M-Me)}{(Mo-Me)} = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}\exp\,[(-2n+1)^2\frac{\pi^2}{4L^2}\mathrm{Deff.}{*}\mathrm{t}] \qquad 2.7$$

Where, M_r is moisture ratio (dimensionless), Me is equilibrium moisture content (db), Mo is initial moisture content (db), $D_{eff.}$ is effective diffusivity (m^2/s), t is time (sec), and L is thickness of sample (m), π (Greek symbol) is 3.1416, exp is exponential sign.

It is reasonable to assume that moisture content is negligible when equilibrium is reached in frying process, so Me=0 (Soorgi *et al*., 2012; Adedeji *et al*., 2009) and when *Me* is very small, only the first term in equation (n=1) is considered to calculate the values of the diffusion coefficients (Rahimi & Ngadi, 2014; Soorgi *et al*., 2012; Dehghan Nasiri *et al*., 2011). So, the equation (2.7) comes as follows:

$$\mathrm{M_r}= \frac{M}{M0} = \frac{8}{\pi^2}\exp\,(-\frac{\pi^2}{4L^2}\mathrm{Deff}{*}\mathrm{t}) = \frac{8}{\pi^2}\exp\,(-\mathrm{K}{*}\mathrm{t}) \qquad 2.8$$

Where, K is rate constant (per second) and the following equation determines the correlation between rate constant and effective moisture diffusivity as follows:

$$\mathrm{D_{eff}} = \frac{4KL^2}{\pi^2} \qquad 2.9$$

For modeling the fat uptake, a first (1^{st}) order kinetic model is a niche approach as reported (by Dehghan Nasiri *et al*., 2011; Adedeji *et al*., 2009) as follows:

$$\mathrm{F_C}= \mathrm{O_{eq}}\,[1-\exp(-\mathrm{K}*\mathrm{t})] \qquad 2.10$$

Where, Fc is the fat content (db) and C_0 is equilibrium fat content (db). And variation of apparent diffusion coefficient (D_{eff}) and equilibrium oil content (O_{eq}) with temperature can be determined by using Arrhenius type equation (Soorgi *et al.*, 2012; Dehghan Nasiri *et al.*, 2011; Adedeji *et al.*, 2009):

$$D_{eff} = D_0 \exp [- \frac{Ea}{RT}] \quad 2.11$$

$$O_{eq} = K. \exp [- \frac{Ea}{RT}] \quad 2.12$$

Where, D_0 is effective diffusivity (m^2/s) at high liquid concentration, Ea is activation energy (kJ/mol), T is absolute temperature (^{0}K), R is universal gas constant (0.0083143 kJ/mol ^{0}K)

Rahimi & Ngadi (2014) also mentioned about other kinetic models (Eq 2.13 - Eq 2.16) to describe fat uptake rate (penetrated fat uptake) of batter-coating during frying as follows:

Newton model: $FR=e^{-kt}$ 2.13

Page model, $FR=e^{-kt^n}$ 2.14

Modified Page model, $FR= e^{(-kt)^n}$ 2.15

Henderson & Pabis model, $FR= a* e^{-kt}$ 2.16

For the equations (13 -16), FR is fat content ratio, k is kinetic of oil penetration, n and a are constant numbers. Fat content ratio was calculated as in the similar manner of moisture ratio, as follows:

$$FR=\frac{Fe-F}{Fe-Fo} \quad 2.17$$

where F_0, F, Fe respectively represents initial-fat content (db), penetrated-fat content at time t (db), and equilibrium penetrated fat content (db).

As in overall, frying yield of batter coated product could be computed according to Labropoulos *et al*. (2013) and Mahdavian *et al.* (2016) as follows:

$$F_Y = \frac{\mathrm{Fwt}}{\mathrm{Iwt}} \quad 2.18$$

Where, F_Y represent frying yield, F_{WT} and I_{WT} respectively represent weight after entire processing (coating & frying) and weight before coating.

2.6 Frying oil

During frying, water escapes from food while oil migrates into food, which consequence into the addition of nutrients and flavors in fried product. Therefore, frying oil has the contribution as of being a heat transfer medium and an ingredient of fried batter coated products. In deep-fat-frying, heat and mass transfers occurs through oil, and there is a new trend of Air frying where air or inert fluid is used as frying medium (Zaghi *et al.*, 2019; Oke *et al.*, 2018; Santos *et al.*, 2017; Sansano *et al*., 2015). Undoubtedly, food frying industries solely rely on deep-fat-frying (DFF), due to its technological advantages over others. The choice of frying oil is based not only on its technological characteristics, but also other factors such as nutritional features, stability, resistivity to oxidation, cost of oil etc., as example, oils rich in saturated fatty acids and partially hydrogenated oils have improved stability profiles for prolonged frying (Oke *et al*., 2018; Gadiraju et al., 2015; Mellema, 2003). Choice of frying oil might also vary with the location, wherein batter coated fried food would be consumed and oil is produced; as an oil that is produced in any specific location will get priority to be used within and around that territory, as consumer are adopted and might have preference with that specific oil. Soybean, palm, canola, and sunflower oils represent 80% of world oil production (Ziaiifar *et al*., 2008) and it is apparent that most of research on batter coated fried food uses these plant-based edibles due to their better health effect over animal fats. Highly polyunsaturated fatty acids (PUFA) containing oils are nutritionally interesting but are very sensitive to oxidation, in contrary, saturated oil is better in terms of stability but that is highly causative to cardiovascular disease (Ziaiifar *et al*., 2008). Similarly, highly viscous oil has more adhesiveness, hence, oil content would be higher in finished product, than frying in low viscous oil. Saturated oil as frying medium increases final oil content of product, while enhance their physical attributes i.e., crispiness. Therefore, ideal frying oils could be a mix of different oils presenting complementary chemical properties as well as technological functionality (Ziaiifar *et al.,* 2008). In recent days, use of vegetable oil, corn oil, peanut oil, and the blends of oil (soybean, sunflower & cotton seed) is not surprising (Abtahi *et al*., 2016). It is notable, frying oil characteristics could affect the frying-yield (%) of batter coated food, which needs to be properly considered specially in large scale production. In concise, considerable facts of selecting oil for frying of batter coated foods would be the fatty acid profile, degree of hydrogenation, degree of saturation, melting point, viscosity, smoke point, stability, prone to the formation of carcinogenic compounds, etc.

2.7 Time-temperature combination of frying

Batter coated foods are generally processed through deep-fat-frying, to develop uniform product quality and to make the frying process faster. In deep-fat frying, batter coated foods are kept completely immersed in hot oil for specific period of time (Oke *et al*., 2018). The reported range of frying-oil temperature is 150°C -190°C (Zhang *et al*., 2020), whereas 180°C temperature has been used in most of frying related studies. Generally, time-temperature combination for frying of batter coated food is found as inversely correlated i.e., higher the oil temperature lowers the frying time, and vice versa. The considering facts for selecting the frying time-temperature combination are follows: (i) longer frying times favors higher oil content in fried product, due to longer exposure to frying oil (ii) frying at higher temperature favor lower oil content in finished fried product, due to rapid crust formation which act against oil uptake during frying (iii) duration of frying at certain oil temperature should achieve minimum desired temperature at the coldest point of batter coated food, which is generally linked with food safety issue. It is notable, immediately after dipping of batter coated food in oil, a sudden drop in frying-oil temperature could occur due to rapid heat-transfer from frying-oil to immersed product; wherein maintaining a good solid-to-oil ratio might minimize the temperature drop event. It is also recommended to pre-heat the frying oil and hold at the desire frying temperature for a considerable period, before putting batter coated food into oil. Several researchers pre-heated and maintained the oil temperature at 180±2°C for 1/2 h, before frying operation (Rahimi et al., 2017a; Román *et al*., 2018).

2.8 Pre/post-fry treatment of batter coated food

Pre-frying is a process, where batter coated food is partially processed for only a short period of time to (i) reduce finish-cooking time (ii) enhance storage life (iii) enhance stability by inactivation of enzymes and microbes (Ziaiifar *et al*., 2008). Generally, the temperature that would be used for finish-cooking is used for pre-frying operation, and duration of pre-frying operation is less than finish-cooking, as example: conventional par-frying time is ≤ 1 minute where finish-frying operation is mostly >1 minute. Pre-frying treatment mostly involves par-frying as well as other treatments are getting involved such as pre-drying, microwave assisted heating/frying, oven baking, IR heating etc., and these treatments could also be used as post-frying treatment of batter coated product. Rahimi & Ngadi (2014) studied the effect of batter formulation and pre-drying time on inter-particle space fractions in fried batter coatings and reported that longer pre-drying time significantly decreases fat uptake during frying. Rahimi & Ngadi (2016) studied the effects of pre-heating temperature on porosity, moisture content, and fat content of fried batters, and reported that batter pre-heated at 60°C showed higher moisture content, lower fat content and lower porosity than non-pre-heated batters. Ngadi et al., (2009) studied the effect of microwave

(MW) pretreatment on mass transfer during deep-fat-frying of batter coated chicken nuggets and reported that, MW pretreatment had significant effect on moisture loss and oil uptake of chicken nuggets during deep-fat-frying. Liberty et al., (2019) discussed that, bulk of oil in fried food is absorbed during their removal from fryer and good quantities of oil adhered to the surface of the fried product. Therefore, post-fry de-oiling from coated fried products would enhance the oil reduction functionality of batter coating. Hence, methods of removing adhered surface oil (while the food is still in the fryer and outside of fryer) would be an aspect of great interest in food industry, and this could be a novel area for further studies. However, there is lack of study detailing the effect of pre- and post- frying treatment on finished products, hence more investigation is needed, as theses could directly affects quality characteristics (structural, thermal, chemical, mechanical) and storage stability of batter coated products (Bechtel *et al*., 2018; Carvalho & Ruiz-Carrascal, 2018; Martin Xavier *et al*., 2017; Lalam *et al.*, 2013; Albert *et al.*, 2009).

2.9 Finish cooking methods

Finish-cooking is a unit operation by which pre/partially processed foods are prepared for consumption. For preparing the partially fried and frozen stored products to be consumed, finish-cooking is an inevitable process (Rady *et al.*, 2019; Raj *et al*., 2016).

2.9.1 Infrared heating: Infrared is part of electromagnetic spectrum in the range of 0.5-100 μm, and infrared (IR) could be absorbed by food compounds (mostly water molecules, ions) which result in temperature rising and IR-heating have been introduced in food industry (Rahimi *et al*., 2018). IR heating possesses few special attributes such as high heat flux, direct heat penetration, faster and uniform heating, lower degradation of nutritional components (Rahimi *et al*., 2018). IR heating is mostly used as various unit operation such as pasteurization, drying, thawing, and baking. However, IR treatment have been applied by Lloyd *et al*., (2004) to produce French fries and remarked that, IR heating could produce food having quality attributes like immersion frying, with lower fat content. Melito & Farkas (2013) used IR heating for donuts preparation, and concluded that, as finish-frying step IR heating had no considerable effects on sensorial aspects of gluten free donuts; and IR cooked sample had lower fat content in comparison to the fully fried donuts. Recently, Rahimi *et al*., (2018) mentioned that IR cooking can produce food having similar acceptance to the deep fat fried sample in addition to having significantly lower fat content..

2.9.2 Microwave heating: Electromagnetic spectrum between frequencies of 300 MHz and 300 GHz is represented by microwaves (Singh & Heldman, 2009). In MW treatment, heat is generated from inside of the food materials and Federal Communications Commission (FCC) of USA, has set two frequencies i.e., 915±13 MHz, and 2450±50 MHz for industrial and home use, respectively (Kirmaci & Singh, 2012; Singh & Heldman, 2009). Absorption of microwaves by a dielectric material, results in the microwaves giving up their energy to the material, with a consequential rise in temperature. Singh & Heldman (2009) describes the principle of microwave heating very nicely as follows: MW increase food temperature by two distinct mechanism (i) ionic polarization and (ii) dipole rotation. When MW is applied to food containing ions, the ions move at an accelerated pace due to their inherent charge, and the resulting collisions between ions cause the conversion of kinetic energy into thermal energy. Food materials contain polar molecules such as water, when an electrical field is applied, the molecules orient themselves according to the polarity of the field. In MW heating, polarity alternates rapidly (2450/ 915 MHz), and polar molecules rotate to maintain alignment with the rapidly changing polarity. Therefore, rotation of molecules leads to friction with the surrounding medium, and heat is generated. Several factors might influence the microwave heating of a food materials such as size, shape, state (water/ice), temperature, composition etc. (Kirmaci & Singh, 2012; Singh & Heldman, 2009; Barutcu *et al*., 2009). Microwave heating has many advantages such as shorter treatment time, saving of energy, improving product uniformity, and uniform moisture loss (Soorgi *et al*., 2012; Ngadi *et al*. 2009). MW benefit from the possibility of having pre-fried food in a freezer and preparing the final cooking/heating as soonest possible time (Albert *et al*., 2014). The microwaves offer tremendous advantages in certain food processing operations primarily short processing time.

Microwave (MW) is a method of heating, that can be used as pre-cooking, cooking as well as post-cooking method (Albert *et al*., 2014; Chen *et al*., 2008; Llorca *et al*., 2005) and as post-frying treatment it serves as rapid food preparation method. Microwave heating is preferred due to its rapid temperature generation ability and no need of additional preparation to perform it. Ngadi *et al*. (2009) used microwave as pre-cooking method and mentioned that the increase of pre-cooking time causes decrease of free moisture content and consequent less oil uptake in chicken nuggets. Soorgi *et al*., (2012) studied the effect of MW pre-treatment on mass transfer during deep fat frying of chicken nuggets. Demirok & Kolsarici

(2014) studied the effect of green tea extract and microwave pre-cooking on the formation of acrylamide in fried chicken drumsticks and chicken wings. Llorca *et al.*, (2005) used microwave treatment as an innovative process (replacing conventional pre-frying operation) to develop frozen batter coated squid rings and studied microstructural property upon final frying. Use of MW as core cooking method is seldom in literature, however, Barutcu *et al.*, (2009,a,b) studied the effects of microwave frying on acrylamide (human carcinogen) formation in the coating part of chicken and reported that microwave frying provides lower acrylamide content compared to conventional frying. Albert *et al*., (2014) studied the influence of MW heating on breaded chicken nuggets and Chen *et al.*, (2008) studied the influence of hydrocolloid and reported the ability of HPMC (Hydroxy propyl methyl cellulose) to inhibit moisture diffusion from core to crust during microwave reheating. The most common use of MW in relation to current day's food sector is food reheating. It is noteworthy, re-heating could affect the quality characteristics of previously cooked food and how MW heating (as finish cooking/ core cooking method) impacts the quality of partially fried batter coated frozen food is not well reported.

2.9.3 Air frying: The replacement of traditional frying process with a system capable of providing similar properties of food fried in deep fat while providing higher nutritional quality, as well as ease of use is the idea behind the air frying technology (Zaghi *et al*., 2019). Air-frying has emerged as an alternative process to deep-frying, producing fried food with small amounts of fat (Zaghi *et al*., 2019; Santos *et al*., 2017 & 2015; Shaker, 2015 & 2014; Andr´s *et al*., 2013). Air frying acts by direct contact between a fine mist consisting of oil droplets in the hot air and the product inside a chamber, where hot air (containing fine oil droplets) is circulated at high speed and foods are heated from all sides at once (Zaghi *et al*., 2019). The product is constantly kept in motion to promote homogeneous contact between both phases (Sansano *et al*., 2015; Andrés *et al*., 2013). It is not unusual in air frying, that oil is sprayed on the surface of food, where hot air is used as the heat transfer medium (Ghaitaranpour *et al*., 2018; Teruel *et al*., 2015). Heat transfer in air frying is extremely high and evenly distributed in the product, hence, minimizes quality variation (Zaghi *et al*., 2019; Santos *et al*., 2017).

As an emerging technique, literature regarding the application of air frying and its impact on food quality is limited. Shaker (2015) compared traditional deep-fat frying and air-frying in the production of healthy fried potato strips and reported that the oil uptake was lower under air-frying. Ghaitaranpour *et al*., (2018) studied the effect of air frying on doughnuts physical properties and reported that crust properties like smoothness and irregularities were influenced by air frying. Tian *et al*., (2017) performed comparative study on microstructure and digestibility of potato strips produced by conventional frying and air-frying and reported that air fried potato showed less starch gelatinization, and the microstructure of the air-fried potato showed a compact and well- organized structure, compared to conventional frying. Abd Rahman *et al*., (2017) studied effects of air-frying on quality (composition, optical, textural, and structural properties) of sweet potato snack, and reported that, air fried sweet potato snack had very low-fat content i.e. decreased by 90.1%, lighter color and harder texture than deep fat fried snack. Ferreira *et al*. (2017) studied the impact of air frying on cholesterol and fatty acids oxidation in sardines and mentioned that, air frying significantly decreases the content of essential PUFAs, and increased the levels of cholesterol oxidation products (COPs). Santos *et al*., (2017) performed a comparative study of deep-frying and air-frying and mentioned that fried potatoes obtained by air-frying processes an average of 70% less fat. Similarly, Santos *et al*., (2015) studied the effect of air frying as a novel technology to control carcinogenic Acrylamide formation in fried potato; and mentioned that air-frying reduced acrylamide content by about 90% compared with conventional deep-oil-frying. Yu *et al*., (2020) studied the effect of air-frying time and temperature on quality of surimi and reported that, along with the increase of temperature and time during air-frying, the surimi became crispy out-side while the moisture was well preserved inside, and the lipids were easily oxidized and degraded to volatile compounds.

2.9.4 Deep fat frying: Generally, partially fried foods are finally cooked by deep fat frying at atmospheric pressure (1 atm). The oil type and temperature that are used in par-frying, are preferred to use that identical type of oil and oil temperature, in final deep fat frying, where only the duration of frying varies. Few examples are as follows: Hamza *et al*., (2016) use deep fat frying (180±1^{0}C, for 2 min) as the final cooking method, while investigated quality characteristics and storage stability of gluten-free coated chicken nuggets. Llorca *et al*., (2007) used deep fat frying (180±1^{0}C, for 3 min) for studying the structure of starch granules in fried battered products. Raj *et al*., (2016) used deep fat frying (180^{0}C, 1 min) as final cooking method, to study the evaluation of post-storage finish frying sensory quality of samosa.

2.10 Characterization of batter/coated fried product

To characterize the physical, chemical, thermal, and microstructural properties of batter and coated fried products several tools and techniques have been used as well as new techniques are getting involvement.

2.10.1 Chemical characterization

Frying is a process where product losses moisture and uptake oil. Hence, moisture and fat measurement are the very fundamental approach to understand the chemical nature of batter coated fried products. Moisture content of coated products have mostly determined through drying methods such as: convection oven drying, vacuum oven drying, freeze drying, etc. (Voong *et al.*, 2018; Rahimi *et al.*, 2017a). However, not only the moisture content but also its distribution has crucial impact on quality (e.g., textural) attributes of bater coated fried product. To evaluate spatial re-distribution of moisture in batter coated fried potato strips during post-fry holding at both room environment and IR-heating, Rahimi et al. (2017a) used microtome blade in separating different regions (crust, core) of fried product, afterward have followed freeze-drying approach to determine moisture.

Fat content (dry basis) has been mostly determined by extraction-based method, where petroleum ether, chloroform or hexane has been used as solvent. However, extraction-based method is not suitable to determine the distribution of fat (Mellema, 2003). Adedeji et al., (2011) successfully quantified fat distribution (%) in deep-fat-fried chicken nuggets batter coating by using confocal laser scanning microscopic image and reported a strong correlation between fat distribution (%) and conventionally measured fat content (g/g dry matter, db). Fat content of batter coated fried products could be regionally grouped as core-fat, crust-fat, matrix-fat, penetrated-fat, and surface-fat, where measurement of surface fat is tricky (Zhang *et al*., 2020; Rahimi et al. 2017a; Rahimi & Ngadi, 2014a). Complete immersion of the fried product in extraction-solvent (immediately after frying & for a very short time) and after that the vaporization of solvent is most pragmatic approach in determining the surface fat (Adedeji, & Ngadi, 2018).

Predicting oil uptake in coated food based on the raw batter property, is a very recent approach. Jeong et al., (2021) develops a successful machine learning workflow [based on amylose content, pasting parameters (peak time, peak temperature) and viscosity of batter] to predict oil uptake in rice-flour based batter system. They concluded that, the multilayer perceptron neural network is more efficient over multivariable linear regression, to predict oil uptake in rice flour-based batter. For evaluating the quality of batter coated foods' frying-oil, test of thiobarbituric acid reactive substances (TBARS) is suggested along with the peroxide value and *p*-Anisidine value determination; while the protein content of fried food is measured mostly by kjeldal / combustion method and ash content by Muffle furnace (Hauzoukim *et al.*, 2019; Davarcioglu & Kolsarici, 2019; Bechtel *et al.*, 2018; Martin Xavier *et al.,* 2017; Hamza Taşbaş *et al*., 2016; Cagdas & Kumcuoglu, 2015; Jackson *et al*., 2009). The gluten plays crucial role in batter rheology and gluten content of chicken nuggets were determined by using the ELISA method (Davarcioglu & Kolsarici, 2019). Mesias et al., (2020) reported that, Acrylamide and hydroxymethylfurfural (HMF) are the most common process contaminants in batter coated fried foods and could be determined by liquid chromatography-electrospray ionization-tandem mass spectrometry and HPLC-Diode array detector, respectively.

2.10.2 Physical characterization

To characterize the physical attributes of batter coated fried products, texture profile analysis is (TPA) the mostly used approach. For performing a texture test of coated fried product, few parameters need to be pre-set e.g., punching speed of the texture analyzer probe, travel distance of probe, minimum triggering force, etc. (Rahimi et al., 2017a). In a texture analysis experiment, the force versus displacement/time curves is generated, from where, maximum force to break (F), displacement at maximum force (MD), number of fracture peak (N), slope of curve (S), and so on is used to report textural properties of batter coated fried products. Along with conventional texture analysis, acoustic measurement (detailing number of sound peaks, sound peak height, sound peak width etc.) are appearing as a new trend of characterizing the physical attributes of batter coated fried products in relation to structure and quality at consumption (Voong *et al.*, 2019, 2018; Primo-Martín *et al.*, 2010; Albert *et al.,* 2009).Voong *et al.* (2018) had performed detailed study on physical characterization of deep-fried batter coatings, where simultaneous application of different instruments /methods was considered. However, as crispness is a fundamental sensorial property of deep-fried battered products, Voong *et al.,* (2018) evaluated crispness of deep-fried batter coatings of white prawn by the uniaxial compression, to acquire mechanical and acoustical measurements simultaneously.

Porosity is a crucial property for batter coated fried products, as pore influences textural, mechanical, and sensory properties as well as affects transport phenomena during frying and storage (Adedeji & Ngadi, 2011a; 2010). Pore or void structure in batter coated product develops during frying as a resultant effect of its moisture loss, which is attributed mainly to trapped vapor within coated food that gets extremely heated and forces it way out into frying oil; and pore formation is influenced by processing conditions (frying temperature & time), food composition, and pre- and post-frying treatments (Adedeji & Ngadi, 2011a). Physical pore structure is characterized mainly by porosity value as well as by pore size distribution, where porosity (%) is defined as a ratio of the void/space volume present in the sample to its overall volume (Rahimi & Ngadi, 2016, 2014; Adedeji & Ngadi, 2011a, 2010). Porosity not only affect physical properties of batter coated fried product but also the fat content, as porosity is positively correlated with the amount of fat absorbed during frying (Rahimi & Ngadi, 2016; Adedeji & Ngadi, 2011a, 2010). Adedeji & Ngadi (2010) used mercury intrusion porosimetry (MIP) technique to characterize pore properties of deep-fat-fried chicken nuggets. Adedeji & Ngadi (2011a) used helium pycnometer (HP) method to determine the porosity (%) of deep-fat-fried batter coatings and reported that porosity is significantly affected by frying temperature, frying time and batter formulation. Rahimi & Ngadi (2016) used helium pycnometer to characterize the effects of pre-heating temperature and batter formulation on porosity of fried batters. It is reported that, batter pre-heated at 60°C results lower porosity than non-pre-heated batter and batters pre-heated at 70 and 80°C; and batters with higher wheat flour content showed lower porosity than batters with higher rice flour (Rahimi & Ngadi, 2016). As emerging approach, Adedeji & Ngadi (2011) used X-ray micro-tomographic images to estimate the porosity of laboratory formulated fried batters and commercial batter coated fried chicken nuggets. Adedeji et al., (2011) used confocal laser scanning microscopic image to estimate the porosity of deep-fat-fried chicken nuggets batter coating, and reported that, porosity in coating layer increased with frying temperature and time. It is notable, shrinkage i.e., changes in size/volume (because of water loss) is a very common phenomenon during deep-fat frying of batter coated food (Parthasarathi & Anandharamakrishnan, 2014), and shrinkage can influence the porosity of fried product (Wang et al., 2010). Das et al. (2013) determined the area-shrinkage of battered coated fried chicken (prepared by pressure frying, conventional frying) by planimeter. Wang et al. (2010) determined the volume-shrinkage of deep-fat-fried chicken nuggets by volume displacement (Archimedes) pycnometer.

Color is one of the crucial surface properties of batter-coated fried products, as color is the quality parameter evaluated by consumer even before enters the mouth. The La*b* color system is device independent and color perception is uniform, hence, La*b* color measurement is mostly used in evaluating the color attributes of batter coated fried products. For example, La*b* color measurement was used to evaluate the surface color of fried chickens coating (Das et al., 2013), Infrared-heating based finish-cooked chicken nuggets (Rahimi et al., 2018).

2.10.3 Thermal characterization

Thermal property, most specifically the glass transition temperature (Tg) of batter is a highly influential factor that affects structural-chemical stability and chemical-mobility in batter coated fried foods, during frying as well as at post-frying handling. The Tg of batter systems is impacted by change in formulation (flour, hydrocolloid, moisture, etc.), frying condition (time, temperature), plasticizer, and so on. Thermal characterization of raw batter is crucial for process optimization of batter coated products, as batter coating changes the heat transfer characteristics of the composite products, while reduce fat uptake during frying (Xue & Ngadi, 2007). On the other hand, glass transition temperature of processed item is an important parameter that determines textural changes, quality, stability, product properties, and safety of food systems (Rahimi *et al.*, 2017a). Therefore, determination of thermal properties plays crucial role in batter coated fried food processing industries. In this context, Differential scanning calorimetry (DSC), Dynamic mechanical thermal analysis (DMTA), Thermal rheological analysis (TRA), Thermo mechanical compression test (TMCT), have been reported in literature (Jothi *et al.*, 2020, 2018; Sansano *et al.*, 2018; Rahimi *et al.*, 2017a; Xue & Ngadi 2009, 2007). However, the use of these methods was dependent on the objectives of research.

Xue & Ngadi (2007) used Differential scanning calorimetry (DSC) to determine some thermal properties of selected batters (based of wheat, rice, corn flour) that are commonly used in deep-fat-frying of chicken products. The DSC was used to measure the glass transition temperature (T_g), gelatinization temperature (T_G), ice-melting temperature (T_m), and enthalpy (ΔH_G) of the selected batter systems. Xue & Ngadi (2007) reported that, salt and hydrocolloid (methylcellulose, MC) greatly influenced the thermal properties of batter systems as by increasing Tg, while depressed

Tm. They also reported that, addition of rice and corn flours to wheat flour-based batter changed the thermal properties, while corn-flour based batters required considerably more energy for gelatinization during the cooking process. Similarly, Xue & Ngadi (2009) used DSC used to determine thermal parameters of batter systems i.e., gelatinization temperature (T_G), total enthalpies of gelatinization (ΔH_G), glass transition temperature (Tg), melting peak temperature (Tm), and total melting enthalpies (ΔH_m). Their study showed that, different thermal processes (CFT: cooking–freezing–thawing & FC: freezing–cooking) did not considerably affected either T_G or ΔH_G of batter systems but influenced the glass transition behavior and ΔH_m of the studied batter systems. The thermal processes (i.e., CFT, FC) showed distinctive effect on batter system containing different hydrocolloids such as methylcellulose (MC), carboxymethylcellulose (CMC), and xanthan gum (XG). The studied hydrocolloids shifted the T_G upwards, depressed the Tg, and increased Tm of the studied batter systems. The effect of MC, CMC, XG on Tg was more pronounced in raw samples (FC process) than in cooked samples, and effects of hydrocolloids increased with their increasing levels (Xue & Ngadi, 2009).

Sansano *et al.*, (2018) used DSC to study the influence of chitosan addition (up to 1g/100g), on thermal properties (glass transition temperature, temperature & enthalpy of gelatinization) of wheat and rice flour-based batters. Their study showed that, melting temperature (Tm) was not dependent on type of flours/chitosan percentage (%) in batter formulation. The influence of chitosan on melting enthalpy (ΔH_m), melting temperature (T_m), and non-freezable water content, was only noticeable in rice-flour batter. The incorporation of chitosan gradually decreases the ΔH_m & Tm, while increases the bound water content. Chitosan incorporation did not significantly modify either gelatinization peak temperature (Tp) and onset temperature (To) or gelatinization enthalpy (ΔH_G) of batter systems. Wheat-flour replacement of by rice-flour, increased the glass transition temperature (Tg) of the batters.

In understanding the relation between Tg and textural changes of batter coated product, Rahimi et al. (2017a) used DSC to study the change in glass transition temperature (Tg) of batter coated fried potato strips during post frying holding at room environment and under IR-heating. Rahimi et al. (2017a) reported that, Tg and textural attributes of the studied fried coated samples were affected by both the batter formulation and heat lamp duration.

Jothi *et al*., (2020) used Thermal rheological analysis (TRA) to study the effects of trehalose and corn starch on the mechanical glass transition temperature and texture properties of a deep-fried sample (particles of deep-fried batter) with varying water and oil contents. Their study reported that glass transition temperature decreased linearly with the increases in both water and oil contents, as attributable to their plasticizing effects. Whereas, Tg was elevated by the addition of trehalose, likely due to its anti-plasticizing effect.

2.10.4 Microstructural characterization

The elements that determine fried food qualities exist at microlevel (Adedeji & Ngadi, 2009). Hence, food researchers and manufacturers are focusing on understanding and establishing relationship between food microstructure and quality attributes. However, microstructural analysis of batter-coated product is relatively a new approach of understanding and characterization of structure, and several tools and techniques have been used to fulfill the mentioned purpose. Scanning electron microscopy (SEM), Confocal laser scanning microscopy (CLSM), and X-ray microcomputed tomography (X-ray µCT) are mostly used for microstructural characterization of batter coated products. However, purpose of the use of these techniques are found as versatile.

Rahimi *et al*., (2017b) used SEM to characterize oil spots and moisture pocket re-distributions between crust and core regions of potato strips during post-frying holding. Their study results indicated that the number of oil spots and moisture pockets were indirectly related to the area of the spots; and holding the samples for longer time under infrared (IR) heat lamp reduced the number of big oil spots, whereas increased their mean spot area.

Rahimi & Ngadi (2016) performed scanning electron microscopic image-based Fractal and Lacunarity analysis, to study the structure and irregularities of surface of fried batters of different formulations. Rahimi & Ngadi (2016) reported that, surface microstructure of fried batter was affected by flour type as such: higher amount of wheat flour in batter preparation resulted in higher fractal dimension (FD) values compared to batters with higher amount of rice flour. The FD of fried batter increased with duration of frying, showing a highly positive correlation between FD and fat uptake in fried batters. The SEM image based Lacunarity (i.e., a measure of degree of heterogeneity of batter ruptures) analysis showed that, size and shape of ruptures generated on the surface of batters during frying was approximately uniform (Rahimi & Ngadi, 2016). Jeong & Lee (2021) performed digital image-based surface roughness analysis of fried batters made from wheat and brown rice flour blends. They evaluated surface roughness by two surface roughness parameters i.e., the mean height of the roughness profile from its average line (R_a), and the root mean square roughness (R_q). Their study reported that, the degree of surface roughness distinctly became lower (R_a, R_q) with increasing levels of brown rice flour and the surface roughness parameters had a strong relationship with the reduced oil uptake.

Adedeji *et al.*, (2011) used Confocal laser scanning microscopy (CLSM) to characterize the pore properties and quantify fat distribution in fried chicken nuggets batter coating. Their study reported that, fat distribution obtained from CLSM image analysis was significantly affected by frying temperature and time, and fat distribution decreased within the depth of the sample thickness. It is also reported that, internal porosity was significantly influenced by frying temperature and frying process led to the formation of more micropores and bigger pores.

Adedeji & Ngadi (2011) used X-ray μCT to study the properties of deep-fat fried chicken nuggets, coated with different batter formulation. Their study reported that inclusion of hydrocolloid into batter system significantly influence the microstructural development as such: number of pores, fragmentation index, and structure model index (shape) of the fried coating increased with the addition of carboxymethyl cellulose (CMC), while the porosity decreased. Similarly, Adedeji & Ngadi (2009) used X-ray microcomputed tomography imaging technique to study the microstructural characteristics of deep-fat fried chicken nuggets and reported that, porosity of outer breading coating as well as number of pores increased with frying time, and the shape of pores were between rod-like and spherical structure. In addition, strong correlation is reported between porosity, frying time, fat uptake and moisture loss. However, structural analysis of batter coated products at microscopic level is apparently a recent approach in fried food research, which opens the scope for more future research.

CONNECTING TEXT TO CHAPTER 3

Literature review (Chapter 2) showed that frying is a simultaneous heat and mass transfer process, where "mass transfer" is characterized by moisture-loss from food and fat-uptake into food. Considering dynamic nature of mass-transfer, it could be hypothesized that modification of solid-to-water ratio (SWR) in batter preparation would subsequently affect the mass-transfer process of batter-coated food. It is notable, frying is a heat-induced mass transfer process. Considering this mutuality, it could also be hypothesized that duration of heating would affect mass-transfer associated physicochemical, thermal and structural properties of batter-coated food.

In Chapter 3, the impact of solid-to-water ratio of batter systems and frying time on physicochemical, thermal and structural properties of batter-coated fried foods are investigated. Parts of this chapter have been presented at scientific conference.

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CHAPTER 3

IMPACT OF BATTER SWR ON EVOLUTION OF THERMO-MECHANICAL PROPERTIES OF BATTERS COATED FOODS DURING FRYING AND POST-FRYING

3.1 Abstract

This study aimed to investigate the impact of batters solid-to-water ratio (SWR) and frying time (FT) on physicochemical, thermal and structural properties of meat-analog based batter coated fried foods. Wheat and rice flour-based batter systems of different SWR (1:1.1, 1:1.3, 1:1.5) was used to coat a meat analog model food, and were deep-fried at 180°C for 2, 4 and 6 minutes in canola oil. Results reveals, SWR of batter coatings are negatively correlated with batter-pickup (%), water-retention-ability (%), cooking yield (CY, %), moisture and structural properties (thickness, densities) of fried foods crust. SWR are positively correlated with frying loss (FL, %), fat, textural attributes (hardness, brittleness, crispiness) and glass-transition-temperature (Tg) of the crust. FT showed significant ($p<0.05$) positive impact on FL, crust thickness, fat content, Tg, and textural attributes, whereas negative impact on CY, moisture content, and crust densities. Higher SWR and FT diminishes post-fry mass redistribution, textural evolution, and color changes; whereas lower SWR and FT retains the juiciness of food-core substrate. Rice-flour based batter was favored by the changes in SWR and FT during frying, but was less responsive to post-fry changes. In contrast, wheat-flour based batter was comparatively responsive to post-fry physicochemical changes. Scanning electron microscopy (SEM) analysis reveals, both SWR and FT impacts surface microstructural properties (roughness, micro-opening) of batter coated fried products.

3.2 Introduction

Fried food contains high amount of fat, which is a major health concern. Food surface modification by batter-coating is an effective approach to reduce fat content in fried product, as batter-coating creates layer over food surface that influence mass-transfer process during frying (Devi *et al.*, 2020; Rahimi & Ngadi, 2014). In addition, batter-coating imparts unique organoleptic characteristic (juicy inside and dry outside) in fried food, and textural attributes of batter coated product is crucial in relation to its consumer satisfaction (Adedeji & Ngadi, 2018; Carvalho & Ruiz-Carrascal, 2018).

Frying is a simultaneous heat and mass transfer process, where mass transfer are characterized by moisture-loss from food and fat-uptake into food (Rahimi & Ngadi, 2015; Adedeji *et al*., 2009). Considering the dynamic nature of mass transfer, it could be hypothesized that reduction of water content before frying, will have favor lower fat uptake during frying. To this context, pre-drying, microwave pre-treatment, pre-cooking in oven, and ultrasound treatment have been suggested to reduce initial moisture content of food, before frying (Rahimi *et al.*, 2018; Rahimi & Ngadi, 2014; Soorgi *et al*., 2012; Adedeji et al., 2009; Moyano & Pedreschi, 2006). Suggested methods are energy and time consuming, in addition, these methods negatively impact the juiciness of food substrate. In this scenario, modifying the moisture content of batter-systems would be a pragmatic approach to produce coated-fried products of having crispy outside and juicy inside; as moisture is externally added to solid ingredients (during batter preparation) and the proposed approach will not impact the initial properties of food-core substrate until frying. Considering dynamic nature of mass-transfer during frying it could be hypothesized that, modification of solid-to-water ratio (SWR) in batter preparation would subsequently impact the oil-reduction functionality of batter coatings as well as textural evolution; as crust formation in fried foods is the results of its moisture loss (Rahimi *et al.,* 2017). However, to the best of our knowledge, there is no scientific study detailing the impact of batters SWR on mass distribution and textural evolution during frying and post-fry holding of batter-coated food products. It is notable, like oil uptake event, the development of textural attribute is associated with the loss of moisture during frying. Therefore, modification of frying time (FT) could be a way of achieving desired textural attributes in coated fried food as well as to modify their post-fry evolution (by impacting mass re-distribution). However, modification of FT might impact not only textural properties but also fat-reduction functionality of batter-coatings as both are linked with moisture-removal from food; hence it requires detailed investigation.

Flour is a core ingredient of batter systems, and at present, wheat-flour is most commonly used in batter preparation (Pinkaew & Naivikul, 2019). The relevancy of wheat-flour with some issues (celiac disease, non-celiac gluten sensitivity, wheat allergy, etc.) are acting as a driving force in search of its replacement in batter preparation, wherein use of rice-flour could be an option (Davarcioglu & Kolsarici, 2019; Pinkaew & Naivikul, 2019). However, the impact of SWR on functionalities of batters formulated with different flours has not yet been investigated.

Hence, the overall goal of this study was set to (i) evaluate the impact of solid-to-water ratio (SWR) of batter-coatings on process parameter, structural development, thermal properties and mass transfer process of coated food (ii) characterize the influence of wheat and rice flour-based batter systems on physiochemical, thermal, and structural properties of coated products (iii) assess the impact of batters' SWR and frying time (FT) on post-fry textural evolution and mass redistribution.

3.3 Materials and Methods

3.3.1 Materials

Soy protein isolate (SPI) was procured from MP Biomedicals (29525 Fountain Pkwy, OH, USA). Wheat gluten (WG) was supplied by Sigma-Aldrich Co. (Ontario, Canada). Sodium pyrophosphate ($Na_4P_2O_7$) crystals and Methylcellulose (MC) were received from Fisher Scientific (Fair Lawn, NJ, USA). Sodium bicarbonate ($NaHCO_3$) powder was supplied by Church & Dwight Canada Corp. (Ontario, Canada). Rice flour (Suraj®, composed of 83.32% carbohydrate, 6.67 % protein, 8.68% moisture and 1.33 % fat) was procured from a grocery store in Montreal, Canada. Wheat flour (Five Roses®, composed of 73.33% carbohydrate, 13.33 % protein, 1.33 % fat, 3.33% fiber and 8.68% moisture), NaCl (Sifto, Compass Minerals Canada Corp.) and Canola oil (Sans nom®, Loblaws Inc.**)** were purchased from a grocery store in Sainte Anne de Bellevue, Quebec, Canada.

3.3.2 Sample preparation

In this study, a meat-analog model was used as core substrate. Meat-analog model was formulated with wheat gluten (WG), soy protein isolate (SPI), distilled water (DW), and canola oil (CO).

Firstly, wheat gluten was mixed with fresh canola oil and a homogeneous slurry was prepared, to which soy protein isolate was added and mixed thoroughly. Then distilled water was added and mixed properly until a soft-textured dough was formed. In the prepared dough, respective proportion (g) of the ingredients WG: CO: SPI: DW were 12:15:22:73. The dough was filled in individual rectangular-shaped cavities of silicon mold (Freshware CB-115RD 24-Cavity Silicone Mini Mold, safe temperature range: -40 to +230°C). Filled and covered mold was held in complete horizontal position (surrounded by water, but not immersed) on top of rigid metal wire surface and heated at 70°C for 15 min in a water bath. At the end of heat treatment, silicon mold was allowed to reach equilibrium with room temperature. Then aluminum cover was detached from silicon mold, and gentle pressure (by finger) was applied at the back of each cavity, to bring out the formed substrates without any cracks. This formulated substrate was used as meat-analog model food, weighing an average weight of 16±0.2 g/substrate and having a uniform dimension of 5.6 cm x 2.5 cm x 1.3 cm. Prepared meat analog model was packed in zip-lock plastic bags and stored in refrigerator (4±2°C) for 24 hours, before its use.

Tempura type batters were used to coat the model food. To prepare batter coatings: crystals of Sodium pyrophosphate ($Na_4P_2O_7$) was gently crushed and grinded with mortar-pastel to obtain fine white powder. Sodium bicarbonate (1.4%), Methylcellulose (0.3%) and NaCl (1.5%) were added and mixed properly with Sodium pyrophosphate powder (1.8%), to form a homogeneous powder mix. Powder-mixture was further thoroughly mixed with dry-flour in a proportion of 5% (powder mix) to 95% (flour). Chilled distilled water was added to the solid ingredients with a total solid-to-water ratio (SWR) of 1:1.1 / 1:1.3 / 1:1:1.5 and mixed until a homogenous batter slurry was formed. Batter systems which were prepared with the SWR of 1:1.3, were used in studying the influence of frying time (FT). Wheat based batters (WB) were formulated using only wheat-flour whereas rice based batters (RB) were formulated with only rice-flour. Some samples were formulated using equal mixture of wheat and rice based batters (RWB). Prepared batter systems were kept for 5 min at room environment and then used to coat the model food. Prior to batter coating, model food was brought out of refrigerator and kept at room environment for 30 min. Model food was fully immersed in batter system for 1 minute and then the coated food was held on a kitchen-fork for about 30 sec, to drain the excess amount of batter, in order to obtain a uniform layer of coating around model food. Noncoated (NC) model food was used as control sample.

A programmable deep-fat-fryer (T-fal Compact, FF122851, China) was used to fry the samples at atmospheric condition. Fryer was filled with 1.5 L of fresh canola oil and temperature was set to 180°C. Canola oil was preheated for 1hr and stirred to minimize variation in oil temperature. Each time three (3) sample was transferred into the fryer and uninterruptedly fried for 2, 4, and 6 min, maintaining a sample to frying-oil ratio of 1:30. During frying, all surfaces of the samples were fully immersed in frying oil. At the end of frying, frying-basket was removed from oil and was shaken for 5 times to remove excess oil from the surface of fried products. To minimize variation in frying-oil properties (due to degradation during frying) each batch of oil was replaced with a new batch of fresh canola oil, after 30 min. of effective frying operation. For studying the post-fry quality evolution, the fried samples were individually placed on trays and kept at room environment (25°C) for a duration of 30 min.

3.3.3 Batter characteristics and process parameters

The flow behavior of batter systems were assessed by a strain/stress control rheometer (AR 2000, TA Instruments, Delaware, USA) equipped with a 40 mm diameter parallel plate. The procedure detailed by Adedeji & Ngadi (2011) was followed with slight modification. In brief: after calibrating rheometer, the gap between the plates (geometry & rheometer platform) was set to 1000 µm. Approximately 1.26 ml of instantly prepared batter was placed at the center of the rheometer platform. Excess amount of batter was discarded, and a steel cover was used to minimize water loss during the measurement. The flow behavior of batter systems were determined over a shear rate range of 2–150 s^{-1} at a constant experimental temperature of 25°C. Flow behavior was modeled by power law expression as follows:

$$\tau = K\gamma^n \qquad 3.1$$

where, τ is the shear stress (Pa); K is the consistency index ($Pa.s^n$); γ is the shear rate (1/s); and n is the flow behavior index.

Water retention ability (WRA) was defined as the capacity of batter systems, to retain its moisture within batter slurry under centrifugal force. With required modifications, the approach of Rahimi & Ngadi (2015) was used in determining the WRA of batter systems, as such: 30-33g freshly prepared batter was placed in falcon tube and was centrifuged at 3000 Xg (relative centrifugal force) for 1 min. The supernatant water was carefully removed and weighed. Water retention ability (WRA) was calculated as follows:

$$\text{WRA}= \left(\frac{Wi-Wc}{Wi}\right) * 100 \qquad 3.2$$

where, Wi, Wc, represent the initial weight of the water present in batter system, and the weight of supernatant water, respectively. Wi was calculated from the initial weight of batter before centrifugation.

Batter pick-up (BP) was calculated to denote the amount of raw batter that adhered to model food, which develop crust during frying. BP was calculated by dividing the weight difference (i.e., weight after coating–weight before coating) by the weight before coating, and the result was multiplied by 100 to express as percentage (%) of raw model food.

Cooking yield (CY) represent the total gain in weight after the complete processing i.e., batter coating + frying. Cooking yield is calculated by dividing the weight difference (i.e., weight of batter coated food after frying-weight of food before coating) by the weight of food before coating and express as % of noncoated model food.

Frying loss (FL) was calculated by dividing the weight difference (i.e., weight of coated food before frying-weight of coated food after frying) by the weight of coated food before frying and express as % of un-fried coated food.

3.3.4 Physicochemical properties

Textural attributes were evaluated by puncture/compression test, by using a texture analyzer (Stable Micro Systems Texture Analyzer, TA. HD PlusC, Surrey, UK). Fried samples were individually mounted on a flat rigid support. A puncture probe TA-52 (2mm dia.) and 500 N was used to punch test sample, at a constant test speed of 1mm/s and a travel distance of 5 mm. Puncture test were performed on three equidistance locations of each of the two longest-facets surface of the crust. Following the approach as adopted from Pedreschi *et al*., (2004) and detailed by Rahimi *et al*., (2017), Maximum force to break (N), displacement at maximum force (mm), and slope (N/mm) at maximum force of the puncture test data were chosen as parameters to evaluate crust textural properties i.e., hardness, brittleness, and crispness, respectively. Puncture test data were analyzed with TA. HD PlusC program (Exponent 6.1.14 v, Stable Micro Systems, UK).

Surface color of fried samples were assessed by a spectrophotometer (Minolta Spectrophotometer, CM-3500d, Japan). Surface color was determined in CIELab space (Illuminant D65, 10° viewing angle) from the reflection spectra between 400 nm and 700 nm. Three measurement was performed on each measurement point and three measurement point was chosen for each sample, and the mean value of each sample was recorded. Color parameters L (lightness-darkness), a* (redness-greenness), and b* (yellowness-blueness) was estimated at room environment (25°C) and color difference (ΔE) was calculated according to the following relation**:**

$$\text{Total color difference, } \Delta E = [\Delta L^2 + \Delta a^2 + \Delta b^2]^{1/2} \qquad 3.3$$

where, $\Delta L = L_0 - L$, $\Delta a = a_0 - a$, and $\Delta b = b_0 - b$. The L_0, a_0, b_0 and L, a, b, respectively represents their values immediately after frying and at any duration of post-fry holding.

Crust region of fried samples were carefully separated from core region, by using a microtome blade (Feather, C35 Type, Japan). A cork-borer (AP8326, Flinn Scientific, Canada) having a diameter of 1.15 cm was used to obtained specific portion (area, Ca=1.04 cm^2) of detached crust. The crust sample was collected from three equidistant location (left, middle & right position) of the fried samples. Thickness (Ct) of the procured crust portion was measured by a calibrated digital slide-caliper, and weight (wt.) was measured by an electronic analytical balance (Denver Instrument APX-100, Denver, USA) with the readability of 0.0001g. Crust area-density (ρ_{ca}, g/cm^2**)** was calculated by dividing the mean weight (wt.) of circular crust by it's area (Ca). Crust volumetric density (ρ_{cv}, g/cm^3**)** was calculated by dividing the weight (wt.) by geometric volume (Ca x Ct) of the crust.

Moisture content of fried samples were grouped into three categories namely crust moisture (CrM), core moisture (CoM) and total moisture (TM). Detached (by microtome blade) crust and core portion of fried samples were separately freeze dried in a freeze-dryer (Modulyod-115, Thermo Savant, NY, USA) at -50°C and 250 mbar for 48 hours and then transferred into a desiccator to equilibrate for 30 min. Weight of the sample before and after freeze-drying was measured and moisture content was calculated on dry basis (g/g dry matter). Separately, some fried samples were freeze dried without separating the crust from the core to measure the total moisture content of the samples.

Fat content was grouped into three categories, namely crust fat (CrF), core fat (CoF) and total fat (TF). Each freeze-dried sample was grounded to increase surface to volume ratio, and an amount of 3-5 g was placed in thimbles of a VELP SER 148 solvent extraction unit (Velp Scientifica, Usmate, Italy). Fat was extracted by following the Randall method, with petroleum ether at 130°C pre-set temperature. Complete fat extraction process was performed in three consequent steps: immersion (30 min), washing (30 min), and recovery (30 min), totaling of 90 min. Weight of the extracted fat was measured and fat content was computed on dry basis (g/g dry matter) i.e., by dividing the mass of extracted oil with the mass of the freeze-dried sample.

3.3.5 Thermal properties

Glass transition temperature (Tg) of the detached (by microtome blade) crust of fried samples were determined by using a differential scanning calorimeter (DSC Q250, TA Instrument, USA). Tg of samples were determined by following the method of Rahimi *et al*., (2017), in brief: around 10-15 mg sample was placed in sample pan, a lid was attached, and a mechanical device was used to make them as hermitically sealed. One sample holder with lid (but without crust sample) was used as reference pan. In DSC measurement chamber, sample was rapidly cooled to -40°C at a cooling rate of 20°C/min, held isothermally for 2 min, and then heated to +40°C at a rate of 2°C/min. DSC data were recorded and analyzed by DSC-Trios 5.1.1v software (TA Instruments, USA). From DSC thermogram, Tg were obtained as onset of the heat capacity change and were represented as Tg_{onset}. The Tg_{onset} was obtained as the change of the baseline slope, after a variation of heat flux occurred, and determined as the intersection of tangents to the transition curve

3.3.6 Scanning electron microscopy

Surface morphology of the fried samples were assessed by scanning electron microscopy (SEM). Surface washed (by three consecutive charges of petroleum ether, for a total immersion period of three min) fried samples were freeze dried and stored in desiccator until SEM imaging by a scanning electron microscope (Hitachi TM3000, Japan). SEM operational setting was adopted from Adedeji & Ngadi, (2018) and performed with required modification. In brief; 5 kV electron power (to prevent sample modification due to heat generated at higher voltage) was applied, auto contrast function was chosen, and composition mode of imaging was used. Small cut, rectangular shaped, defatted sample of about 10×10×5 mm in dimension was placed on the sample base with a sticky surface that comprises of carbon tab. After each sample was properly aligned by visual control using two sets of knobs and images shown in the control software on a computer screen, vacuum pressure was created in the sample chamber and images were quickly acquired to prevent charging and heating that could lead to artifacts in images. SEM images were acquired at a magnification of ×30. SEM micrograph was recorded as 8-bit image and surface microstructure was analyzed by Java-based public domain image processing software (ImageJ 1.52 v, National Institutes of Health, USA). Surface opening (SO) was estimated from SEM micrograph, as ratio of the area of total openings (holes, cracks, ruptures, crevices etc.) in surface to the total surface area. In estimating SO from SEM image, the image analysis approach of Rahimi & Ngadi (2015) was considered i.e., darker region represents holes, cracks, ruptures, crevices, while whiter region represents solid matrix. Following the method as detailed by Rahimi & Ngadi (2016b), fractal dimension (FD) of surfaces were estimated from 2D SEM images by box-counting method.

3.3.7 Statistical analysis

All experimental data obtained from triplicate of samples and mean ± standard deviation was reported. Tukey's honestly significant difference (HSD) was used in reporting significant ($p<0.05$) difference among mean values. Statistical analysis is performed by licensed statistical software (JMP 14.1v, SAS Institute Inc., USA).

3.4 Results and Discussion

Figure 3.1 depicts, rheological properties of batter systems and process parameters of batter-coated samples. Rheological properties of batter systems were greatly impacted by solid-to-water ratio (SWR). The apparent viscosity of batter systems decreased with the increase of SWR. Irrespective of SWR, wheat-flour based batter (WB) was more viscous in nature than rice-flour based batter (RB) and equal portion (50:50) of rice & wheat flour-based batter (RWB). Higher protein content of wheat-flour might have played key role behind the higher viscosity of WB batters, as gluten possess good film-forming ability in favor of the formation of strong elastoplastic dough (Rahimi & Ngadi, 2014). The studied batter systems were pseudoplastic fluids (Adedeji & Ngadi, 2011). The consistency index (K) of batter systems decreased with the increase of their SWR, and K values varied from 78.37 to 1.17 Pa.s^n. These K values are in close similarity with the values reported in relevant literature (Adedeji & Ngadi, 2011; Xue & Ngadi, 2007) for the batter-systems formulated with different hydrocolloids (CMC, HPMC, Xanthan gums). The flow behavior index (n) of batter systems were less than 1, confirming their shear thinning behavior, wherein n values (ranged between 0.56 to 0.25) have been found as inversely related with SWR. Water retention ability (WRA) and batter pickup (BP) were negatively correlated with the SWR of batter systems. Both WRA and BP of wheat-flour based batter were higher compared to rice-flour based batter. Higher WRA and BP has favored lower frying loss (FL) and higher cooking yield (CY) of WB coated samples. Frying loss increased with the increase of SWR, as batter systems having lower solid content (higher SWR) were less effective to retain their moisture under vigorous process. The frying loss (FL) of coated samples increased with frying time (FT), and higher frying loss resulted into lower cooking yield. Cooking yield of batter coated fried (for any duration) samples were significantly higher than noncoated (NC) sample. Overall, flour type, solid-to-water ratio, and frying time showed considerable impact on raw batters' properties and process parameters.

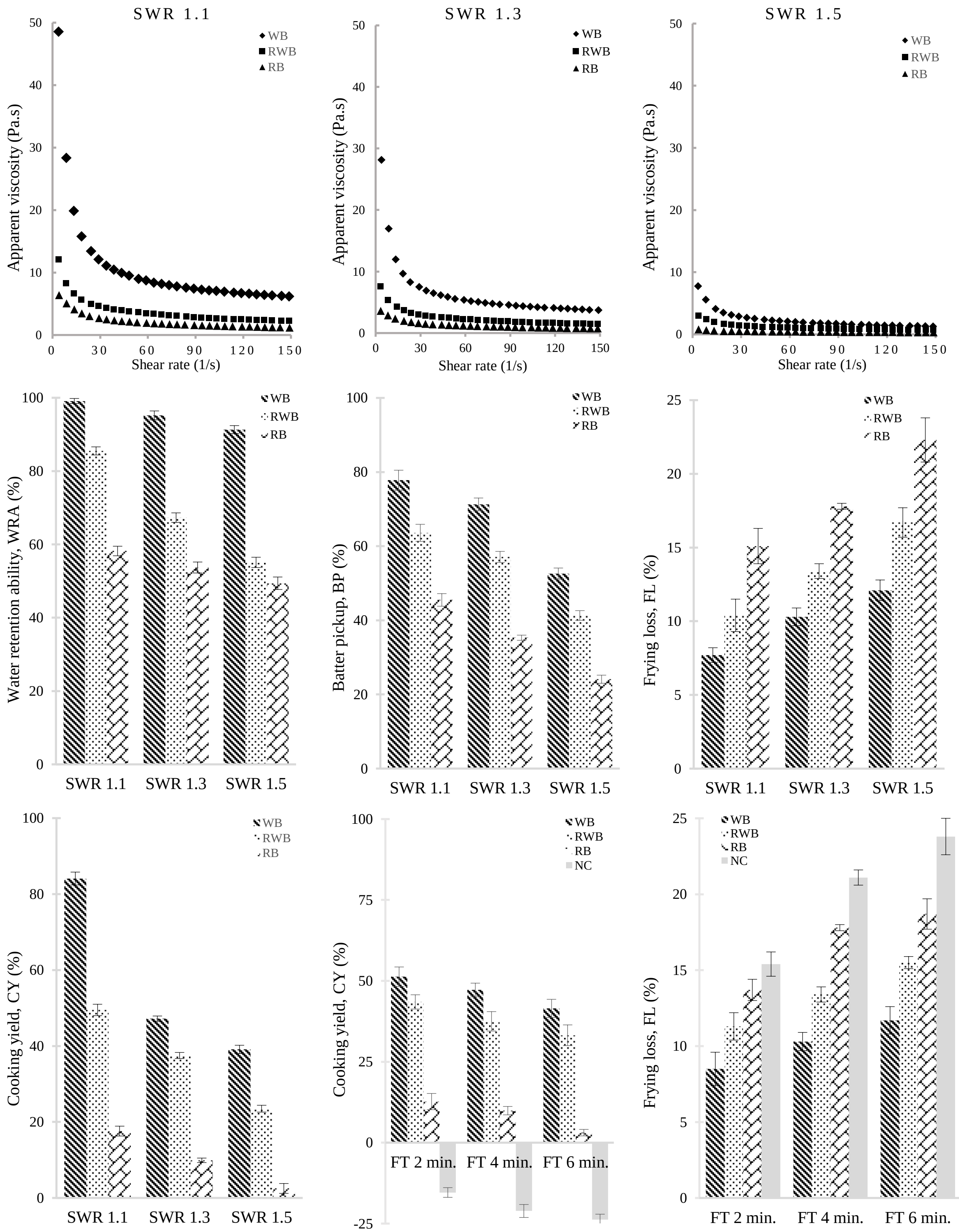


Figure 3.1: Rheological properties of batters and process parameters of batter-coated samples. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter/coated sample, respectively NC: noncoated model food. FT: frying time (minute). SWR: solid-to-water ratio.

Figure 3.2 depicts moisture-fat profile of fried samples. Moisture content of coated fried samples were substantially impacted by solid-to-water ratio (SWR) of batter systems. Crust moisture (CrM), core moisture (CoM), and total moisture (TM) of fried samples showed a decreasing trend with the increase of SWR. The SWR of batter systems has not only impacted crust moisture but also the juiciness of food-core substrate of fried samples, for example, higher CoM were observed for lower SWR of batter systems. The impact of SWR on moisture loss during frying could be understood as such: batter systems of lower SWR possessed not only higher adhesiveness (BP) but also higher water retention ability (WRA), and these attributes were in favor of their lower frying loss (FL). As net impact of these (BP, WRA) parameters , the presences of higher moisture (CrM, CoM) in coated fried sample was determined for the batter systems of lower SWR. However, moisture (CrM, CoM, TM) content of batter-coated fried samples showed a negative correlation with frying time (FT). Compared to noncoated sample (NC), batter coatings has shown impact in delaying the process of moisture-loss as a function of FT. This event could be attributed to lower moisture availability in batter-coated samples, as higher availability of moisture supports rapid loss of moisture during frying (Ngadi *et al.,* 2006).

Fat content of coated fried samples were greatly impacted by SWR. Crust fat (CrF), core fat (CoF), and total fat (TF) of fried coated samples showed an increasing trend with the increase of batters' SWR. For identical (flour based) batter system, modification of SWR has impacted the oil-reduction functionality of batter coatings, wherein lower SWR had favored the presence of low fat in fried products. This observation could be attributed to the initial moisture content of batter-coated samples; as due to dynamic nature of mass-transfer, initial moisture content of food could impact moisture-loss and oil-uptake (by forming pores) during frying (Dehghannya & Abedpour, 2018; Rahimi & Ngadi, 2014). The impact of SWR on fat uptake could be understood as such: during frying, cavities/pores were formed in samples due to moisture loss and frying-oil were substituted in the pore spaces. Hence, presence of high amount of water (in batters' of high SWR) might had resulted the formation of higher pores (due to lower water retention ability, WRA) that favored higher oil uptake. SWR of batter systems has not only impacted the crust fat but also the core fat of fried samples, for example, higher CoF and CrF were observed for higher SWR. Fat (CrF, CoF, TF) content of coated fried samples showed a positive correlation with frying time (FT), which could be linked to the formation of porous structure (Rahimi & Ngadi, 2016a; Adedeji

& Ngadi, 2011). Compared to noncoated (NC) sample, batter coatings has shown substantial impact in delaying the process of fat-uptake as a function of FT. Irrespective of SWR and FT, wheat-flour based batter (WB) had favored the presence of lower fat in fried coated products, which could be attributed to its higher water retention ability.

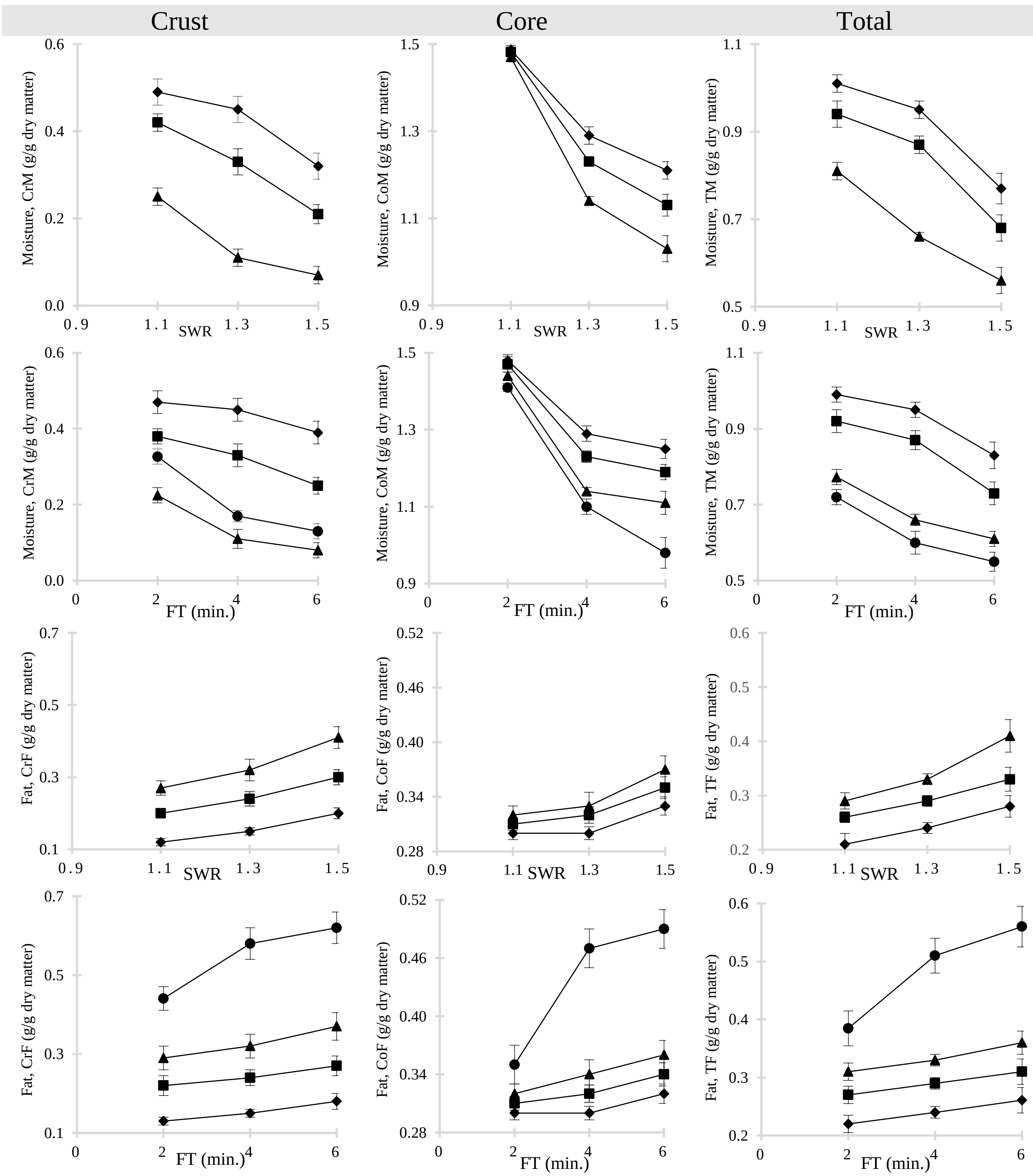
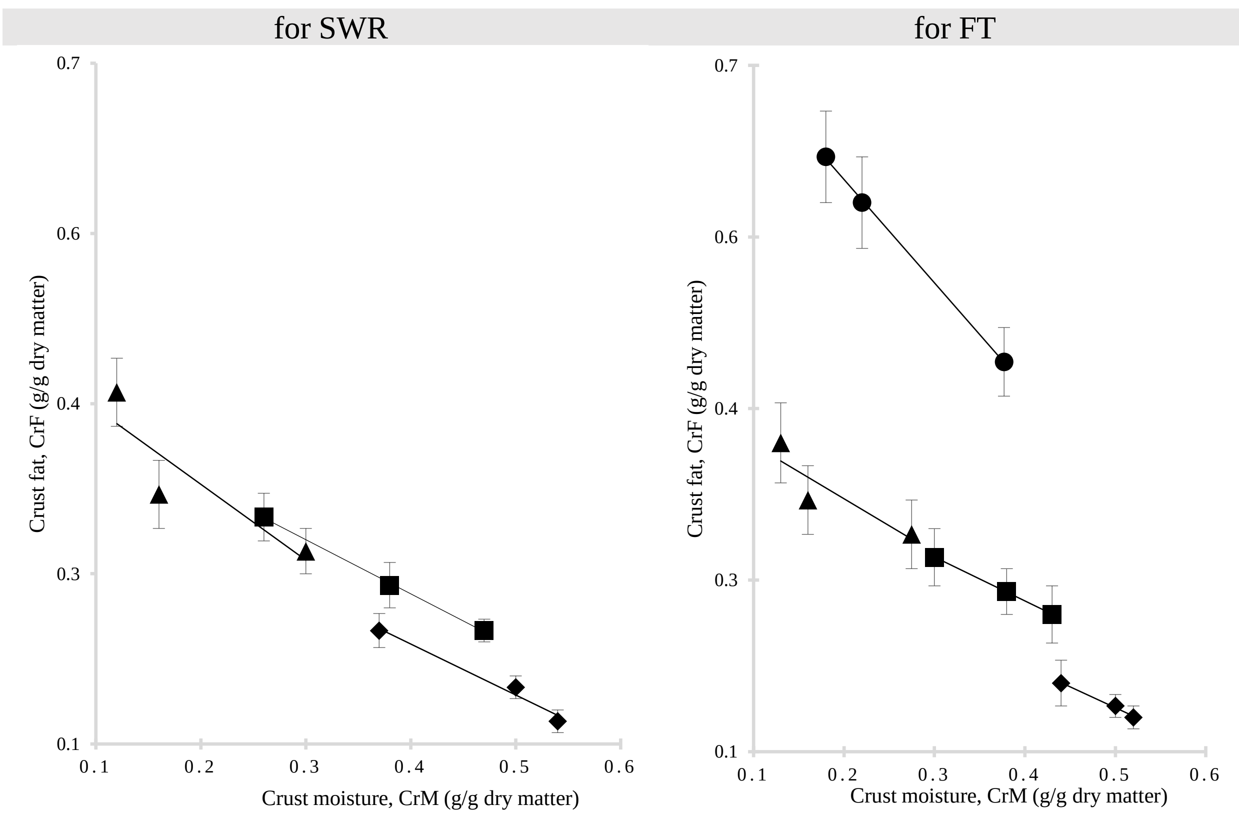


Figure 3.2: Moisture-fat profile of fried samples. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time (minute). SWR: solid-to-water ratio.

Figure 3.3 depicts, impact of batters SWR and FT on crust moisture-fat relation. Under all circumstances (SWR, FT) crust moisture and crust fat content have shown negative (-) correlation. For different SWR, fat absorption in coated samples showed great dependency on their moisture loss; where higher extent of dependency (slope value of the GLM) were observed for rice-flour based batter (RB) coated sample. Fat uptake in batter coated samples were not the exact equal of moisture loss, as indicated by a slope value less than 1. Overlapping (in left Figure 3.3) of the predicted GLMs indicates: modification in SWR of rice and wheat flour-based batters could act as substitutes to each other's in impacting moisture-loss and fat-uptake during frying, without modifying the FT. Comparative slope values of the GLMs indicate, to impact the mass-transfer process of batter-coated food, modification of batters SWR would be robust than the modification of FT.



	GLM	R^2
WB	CrF= -0.4494 CrM + 0.3454	0.98
RWB	CrF= -0.4775 CrM + 0.3995	0.99
RB	CrF= -0.6679 CrM + 0.4291	0.80

	GLM	R^2
WB	CrF= -0.3654 CrM + 0.3029	0.99
RWB	CrF= -0.3837 CrM + 0.3661	0.99
RB	CrF= -0.4718 CrM + 0.3919	0.80
NC	CrF= -0.9038 CrM + 0.7358	0.99

Figure 3.3: Moisture-fat interaction. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. SWR: solid-to-water ratio. FT: frying time (minute). GLM: generalized linear model.

Figure 3.4 depicts texture profile and structural features of fried samples, respectively. Textural attributes of coated samples were greatly impacted by solid-to-water ratio (SWR) of batter systems. Hardness (MF) and crispness (S) of coated fried samples have shown an increasing trend with the increase of batters' SWR, and their decrease in ductility (MD) indicates the increase of brittleness. Prominent change in textural attributes were noticed at SWR>1.3. Regardless of batter formulations (SWR, flour type), the hardness, brittleness, and crispiness of fried coated samples increased with the duration of frying (FT), and prominent changes were observed at FT > 4 min. Compared to wheat-flour based batter, considerably higher MF and S values with lower MD value indicates that rice-flour based batter coating develops hard, crispy, and brittle crust.

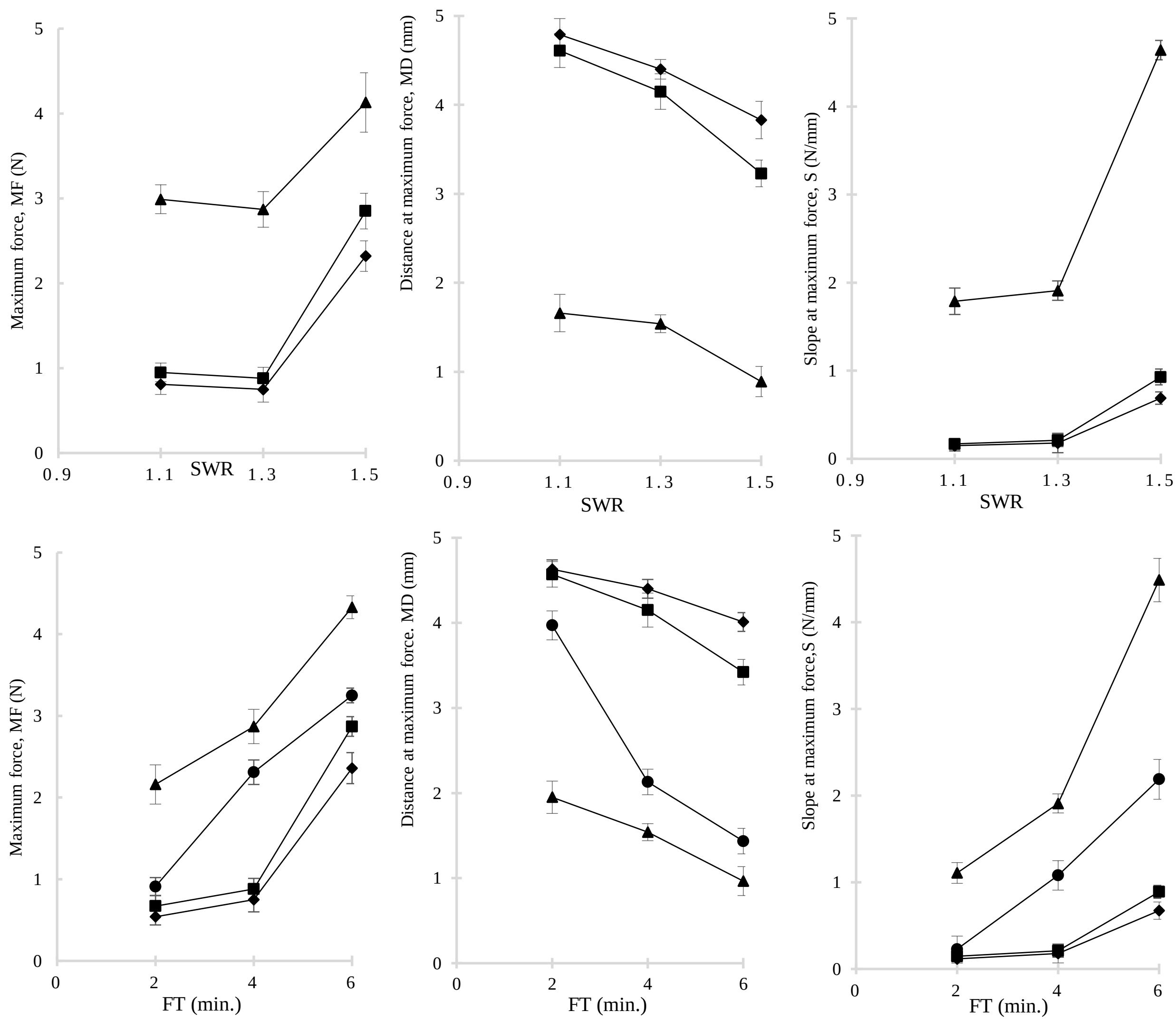


Figure 3.4: Textural attributes of fried samples. Textural attributes of variable SWR were measured after 4 minute of frying. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time (minute). SWR: solid-to-water ratio.

Table 3.1 depicts that crust's structural features such as thickness (Ct), area density (ρ_{ca}) and volumetric density (ρ_{cv}) decreased with the increase of solid-to-water ratio of batter systems. The area density and volumetric density of coated samples decreased with the increase of frying time (FT), whereas crust thickness increased with FT. Higher crust thickness and area density were found for wheat-flour based batter coated samples, compared to rice-flour based batter systems.

Table 3.1: Structural features of fried foods crust.

Frying time (FT)	Solid-to-water ratio (SWR)	Sample	Thickness, Ct (mm)	Area density, ρ_{ca} (g/cm^2)	Volumetric density, ρ_{cv} (g/cm^3)
4 min.	1.1	WB	3.04±0.05^{a}	0.45±0.02^{a}	1.87±0.07^{c}
		RWB	2.53±0.04^{b}	0.39±0.01^{b}	2.03±0.05^{b}
		RB	2.21±0.03^{c}	0.37±0.02^{b}	2.15±0.03^{a}
4 min.	1.3	WB	2.11±0.08^{a}	0.38±0.01^{a}	1.74±0.02^{c}
		RWB	1.83±0.05^{b}	0.35±0.01^{ab}	1.95±0.01^{b}
		RB	1.71±0.03^{c}	0.34±0.01^{b}	2.02±0.01^{a}
		NC	0.55±0.02^{d}	0.08±0.02^{c}	1.42±0.03^{d}
4 min.	1.5	WB	2.03±0.07^{a}	0.23±0.01^{a}	1.26±0.03^{c}
		RWB	1.37±0.05^{b}	0.19±0.02^{b}	1.43±0.02^{b}
		RB	1.13±0.02^{c}	0.17±0.01^{b}	1.57±0.05^{a}
2 min.	1.3	WB	2.05±0.09^{a}	0.44±0.02^{a}	1.82±0.07^{c}
		RWB	1.78±0.07^{b}	0.39±0.01^{b}	1.99±0.05^{b}
		RB	1.62±0.04^{c}	0.37±0.02^{b}	2.11±0.04^{a}
		NC	0.51±0.03^{d}	0.14±0.02^{c}	1.46±0.02^{d}
6 min.	1.3	WB	2.33±0.10^{a}	0.36±0.02^{a}	1.71±0.05^{c}
		RWB	1.95±0.07^{b}	0.33±0.03^{ab}	1.92±0.02^{ab}
		RB	1.79±0.05^{c}	0.30±0.01^{b}	1.97±0.03^{a}
		NC	0.94±0.03^{d}	0.07±0.02^{c}	1.34±0.02^{d}

WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coating, respectively, and NC represent non-coated model food. Lower case letters (a-d) rank significant difference ($p<0.05$) among samples of identical solid-to-water ratio and frying time.

Table 3.2 summarizes, post-fry evolution of moisture-fat profile and textural attributes. The normalized value (ratio of values at any duration of post-fry holding and their corresponding value immediately after frying) was used to characterize post-fry evolution of textural attributes. Hardness (MF*), brittleness (MD*), and crispiness (S*) of coated fried samples has changed considerably within the post-fry holding duration of 30 min. Extent and nature of post-fry textural evolution have varied with batter formulations (flour type, SWR). Hardness and brittleness of rice-flour based batter (RB) coated sample has increased during post-fry holding, while the change of crispiness were not significant. Post-fry changes in textural attributes of RB coated samples could be attributed to the starch retrogradation phenomena (Wani *et al*., 2017; Raj *et al*., 2016). Hardness, crispiness, and brittleness of wheat-flour based batter coated samples has reduced during post-fry holding. Extent of post-fry changes in textural properties of batter-coated fried samples were lower for higher SWR of batter-systems, and this could be understood as a fact of the presence of low moisture in their crust and core region. Coated samples that received longer frying treatment (FT) had developed stable crust, which were less sensitive to post-fry textural changes. In coated samples, significantly higher extent of moisture gain in crust were associated with the lower SWR of batter-coatings and shorter FT. There was no considerable changes in crust fat (CrF) content during post-fry holding, only a slight reduction in CrF were noticed for the samples of higher SWR. Hence, post-fry textural evolution of batter-coated food products could be considered as moisture-migration dependent event, wherein SWR and FT acts as influential factor.

Table 3.2: Normalized value of maximum force (MF*), normalized value of maximum distance (MD*), and normalized value of slope (S*) of the instrumental texture profile; and mean values (g/g dry matter) of crust moisture (CrM) and crust fat (CrF) content.

FT	SWR	Sample	Textural properties						Crust moisture-fat profile			
			MF*		MD*		S*		CrM		CrF	
			0 min	30 min	0 min	30 min	0 min.	30 min	0 min	30 min	0 min	30 min
4 min	1.1	WB	1.0^{A}	0.85 ± 0.04^{bB}	1.0^{A}	0.87 ± 0.04^{bB}	1.0^{A}	0.79 ± 0.02^{bB}	0.49 ± 0.03^{aB}	0.61 ± 0.04^{aA}	0.12 ± 0.01^{cA}	0.12 ± 0.02^{cA}
		RWB	1.0^{A}	0.88 ± 0.03^{bB}	1.0^{A}	0.89 ± 0.03^{bB}	1.0^{A}	0.80 ± 0.02^{bB}	0.40 ± 0.02^{bB}	0.46 ± 0.03^{bA}	0.20 ± 0.02^{bA}	0.19 ± 0.03^{bA}
		RB	1.0^{B}	1.33 ± 0.05^{aA}	1.0^{B}	1.21 ± 0.01^{aA}	1.0^{A}	0.97 ± 0.03^{aA}	0.24 ± 0.02^{cA}	0.28 ± 0.013^{cA}	0.27 ± 0.03^{aA}	0.26 ± 0.02^{aA}
4 min	1.3	WB	1.0^{A}	0.83 ± 0.05^{cB}	1.0^{A}	0.89 ± 0.02^{cB}	1.0^{A}	0.81 ± 0.03^{cB}	0.45 ± 0.03^{aB}	0.59 ± 0.02^{aA}	0.15 ± 0.01^{dA}	0.15 ± 0.02^{dA}
		RWB	1.0^{A}	0.86 ± 0.04^{cB}	1.0^{A}	0.90 ± 0.02^{cB}	1.0^{A}	0.82 ± 0.02^{cB}	0.33 ± 0.02^{bB}	0.43 ± 0.01^{bA}	0.24 ± 0.02^{cA}	0.24 ± 0.01^{cA}
		RB	1.0^{B}	1.29 ± 0.04^{aA}	1.0^{B}	1.18 ± 0.01^{aA}	1.0^{A}	1.01 ± 0.01^{aA}	0.11 ± 0.02^{dA}	0.15 ± 0.01^{dA}	0.32 ± 0.03^{bA}	0.32 ± 0.03^{bA}
		NC	1.0^{A}	0.97 ± 0.02^{bA}	1.0^{A}	0.98 ± 0.02^{bA}	1.0^{A}	0.98 ± 0.02^{bA}	0.17 ± 0.02^{cA}	0.20 ± 0.02^{cA}	0.58 ± 0.04^{aA}	0.54 ± 0.02^{aA}
4 min	1.5	WB	1.0^{A}	0.90 ± 0.02^{bB}	1.0^{A}	0.93 ± 0.02^{bB}	1.0^{A}	0.92 ± 0.03^{bB}	0.32 ± 0.03^{aAB}	0.37 ± 0.02^{aA}	0.20 ± 0.01^{cA}	0.19 ± 0.02^{cA}
		RWB	1.0^{A}	0.94 ± 0.03^{bB}	1.0^{A}	0.95 ± 0.02^{bB}	1.0^{A}	0.94 ± 0.02^{bB}	0.21 ± 0.02^{bA}	0.23 ± 0.02^{bA}	0.30 ± 0.02^{bA}	0.29 ± 0.01^{bA}
		RB	1.0^{A}	1.04 ± 0.03^{aA}	1.0^{A}	1.03 ± 0.02^{aA}	1.0^{A}	1.02 ± 0.02^{aA}	0.07 ± 0.02^{cA}	0.07 ± 0.01^{cA}	0.41 ± 0.03^{aA}	0.38 ± 0.02^{aA}
2 min	1.3	WB	1.0^{A}	0.81 ± 0.04^{cB}	1.0^{A}	0.87 ± 0.04^{cB}	1.0^{A}	0.78 ± 0.05^{cB}	0.47 ± 0.03^{aB}	0.60 ± 0.05^{aA}	0.13 ± 0.01^{dA}	0.13 ± 0.02^{dA}
		RWB	1.0^{A}	0.82 ± 0.02^{cB}	1.0^{A}	0.89 ± 0.02^{cB}	1.0^{A}	0.80 ± 0.02^{cB}	0.37 ± 0.02^{bB}	0.45 ± 0.03^{bA}	0.22 ± 0.03^{cA}	0.22 ± 0.02^{cA}
		RB	1.0^{B}	1.23 ± 0.05^{aA}	1.0^{B}	1.12 ± 0.03^{aA}	1.0^{A}	1.01 ± 0.02^{aA}	0.22 ± 0.02^{dA}	0.26 ± 0.04^{dA}	0.29 ± 0.02^{bA}	0.28 ± 0.03^{bA}
		NC	1.0^{A}	0.94 ± 0.01^{bB}	1.0^{A}	0.96 ± 0.02^{bA}	1.0^{A}	0.95 ± 0.02^{bA}	0.33 ± 0.02^{cAB}	0.37 ± 0.02^{cA}	0.44 ± 0.03^{aA}	0.42 ± 0.04^{aA}
6 min	1.3	WB	1.0^{A}	0.88 ± 0.04^{cB}	1.0^{A}	0.91 ± 0.02^{bB}	1.0^{A}	0.89 ± 0.03^{bB}	0.39 ± 0.03^{aB}	0.47 ± 0.03^{aA}	0.18 ± 0.02^{dA}	0.18 ± 0.03^{dA}
		RWB	1.0^{A}	0.93 ± 0.02^{cB}	1.0^{A}	0.93 ± 0.02^{bB}	1.0^{A}	0.92 ± 0.02^{bB}	0.25 ± 0.02^{bAB}	0.30 ± 0.02^{bA}	0.27 ± 0.03^{cA}	0.26 ± 0.02^{cA}
		RB	1.0^{A}	1.03 ± 0.01^{aA}	1.0^{A}	1.03 ± 0.02^{aA}	1.0^{A}	1.01 ± 0.01^{aA}	0.08 ± 0.02^{dA}	0.09 ± 0.02^{dA}	0.37 ± 0.03^{bA}	0.35 ± 0.02^{bA}
		NC	1.0^{A}	0.98 ± 0.01^{bA}	1.0^{A}	0.99 ± 0.02^{aA}	1.0^{A}	0.98 ± 0.02^{aA}	0.13 ± 0.01^{cA}	0.14 ± 0.01^{cA}	0.62 ± 0.04^{aA}	0.55 ± 0.02^{aB}

Lower case letters (a-d) ranks significant ($p<0.05$) difference among samples for same attribute of the identical solid-to-water ratio (SWR) and frying time (FT). Upper-case letters (A-B) ranks significant ($p<0.05$) difference among samples for same attribute of identical sample at different post-fry holding time. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coating, respectively, and NC represent non-coated model food.

Figure 3.5 depicts, glass transition temperature (Tg_{onset}, °C) of the crust region of fried samples. Tg value of the studied samples were in negative (-) temperature zone ranged between -23.51°C to -20.16°C. In literature (Kerr, 2016), negative Tg has been reported for fried food matrix such as: donut (-18°C), carrot chips (-39°C) and french fries (-11.8°C). Tg values in the ranges between -15.1°C to -20.4°C has been reported for modified starch (potato, corn, tapioca) based fried batters (Rahimi et al., 2017). However, Tg of the studied crust samples were found far lower than the holding temperature (+25°C) at room environment. These could explain post-fry evolution in crust mass and texture profiles, as generally, a higher stability of qualities are anticipated at a temperature below the characteristic Tg of the samples. However, Tg of batter coated samples has shown a slight increasing trend with the increase of batters SWR and FT. These could be due to the presence of lower moisture in samples, as a negative correlation were observed between the Tg and crust moisture content. It has been reported, moisture in food matrix can acts as plasticizer and consequently lowers the Tg of samples (Rahimi et al., 2017).

for SWR	for FT

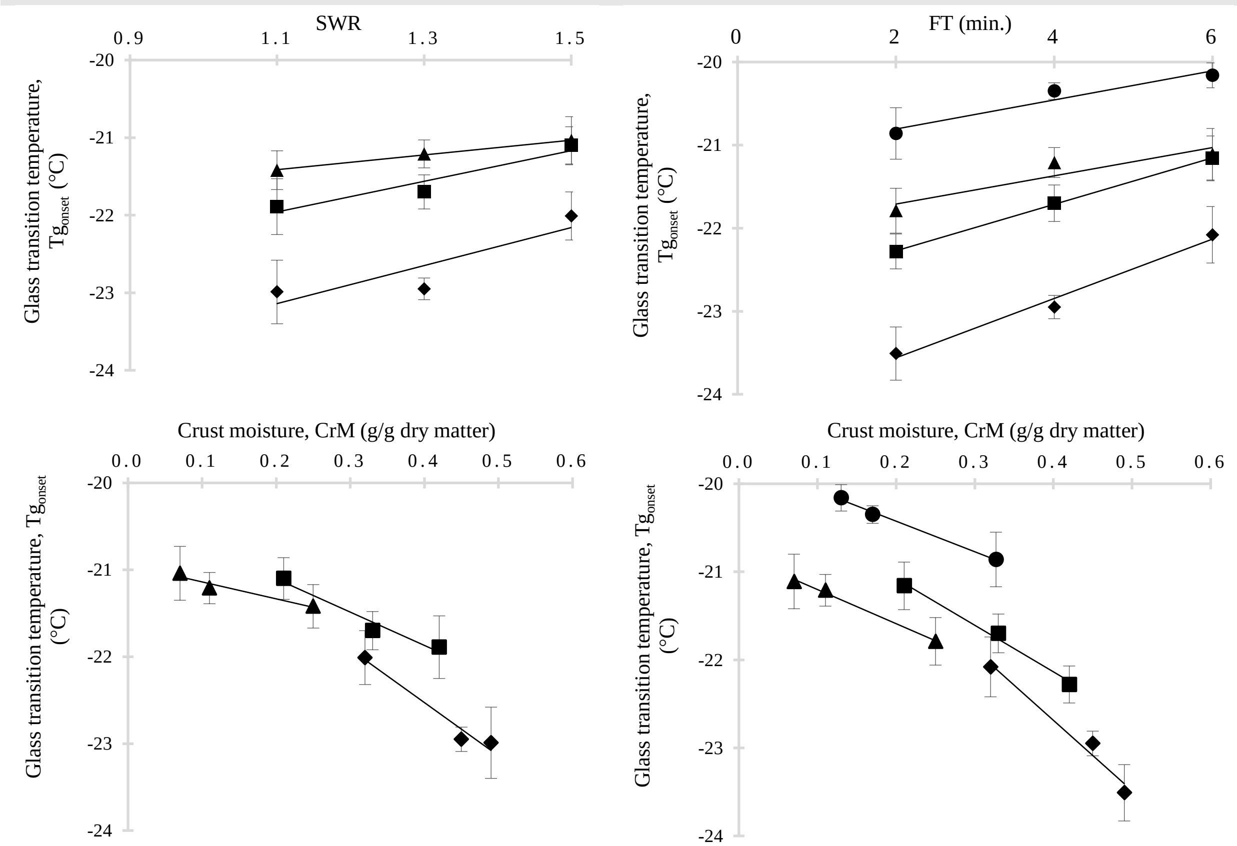


Figure 3.5: Glass transition temperature of the crust region of fried samples. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time (minute). SWR: solid-to-water ratio.

Figure 3.6 depicts, surface color features of fried samples. Lightness (L), redness (+a*), and yellowness (+b*) color tone of coated fried samples has showed an increasing trend with the increase of SWR. Lightness and yellowness showed a decreasing trend with frying time (FT), while redness increased with FT. Lightness and yellowness color tone of RB coated fried samples were considerably different than WB coated samples. Post-fry total color changes (ΔE value) were impacted by batters' SWR and frying time (FT), wherein change in color were not prominent for the samples of higher SWR and higher FT. However, changes in ΔE of fried foods mostly indicates the darkening of initial color.

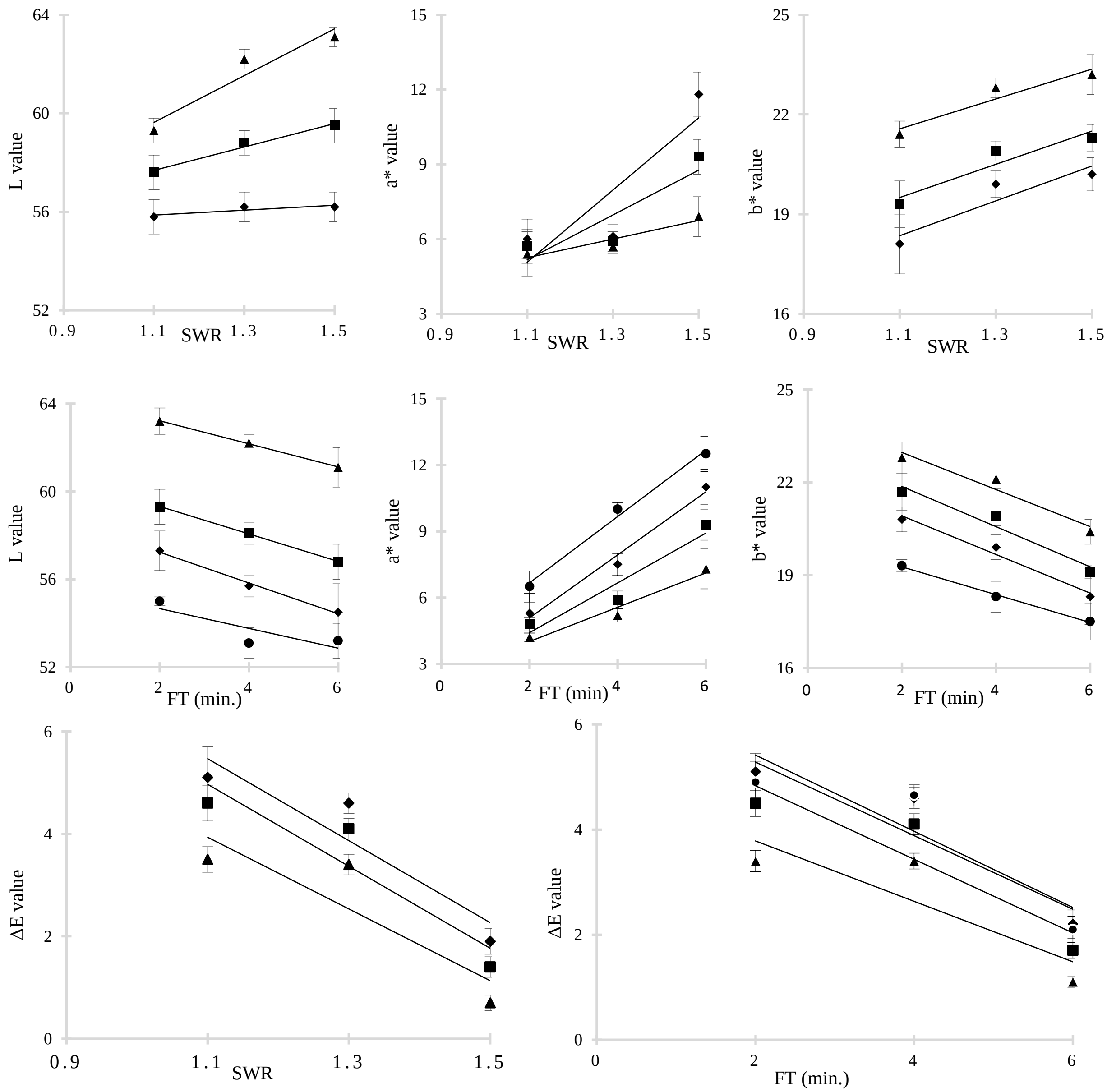


Figure 3.6: Color attributes of fried products. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. SWR: solid-to-water ratio. FT: frying time (minute).

Figure 3.7 depicts surface microstructure of de-oiled crust. Surface microstructure of batter coated fried samples were different than noncoated sample. There were presence of different irregularities (e.g., holes, cracks, ruptures, crevices) on surfaces of fried samples. Surface irregularities were prominent on the samples of higher SWR and FT, in compared to the samples of lower SWR and FT. Irregularities on surfaces were the resultant effect of samples moisture-loss during frying, and these irregularities has favored (acted as open access to) fat uptake in samples.

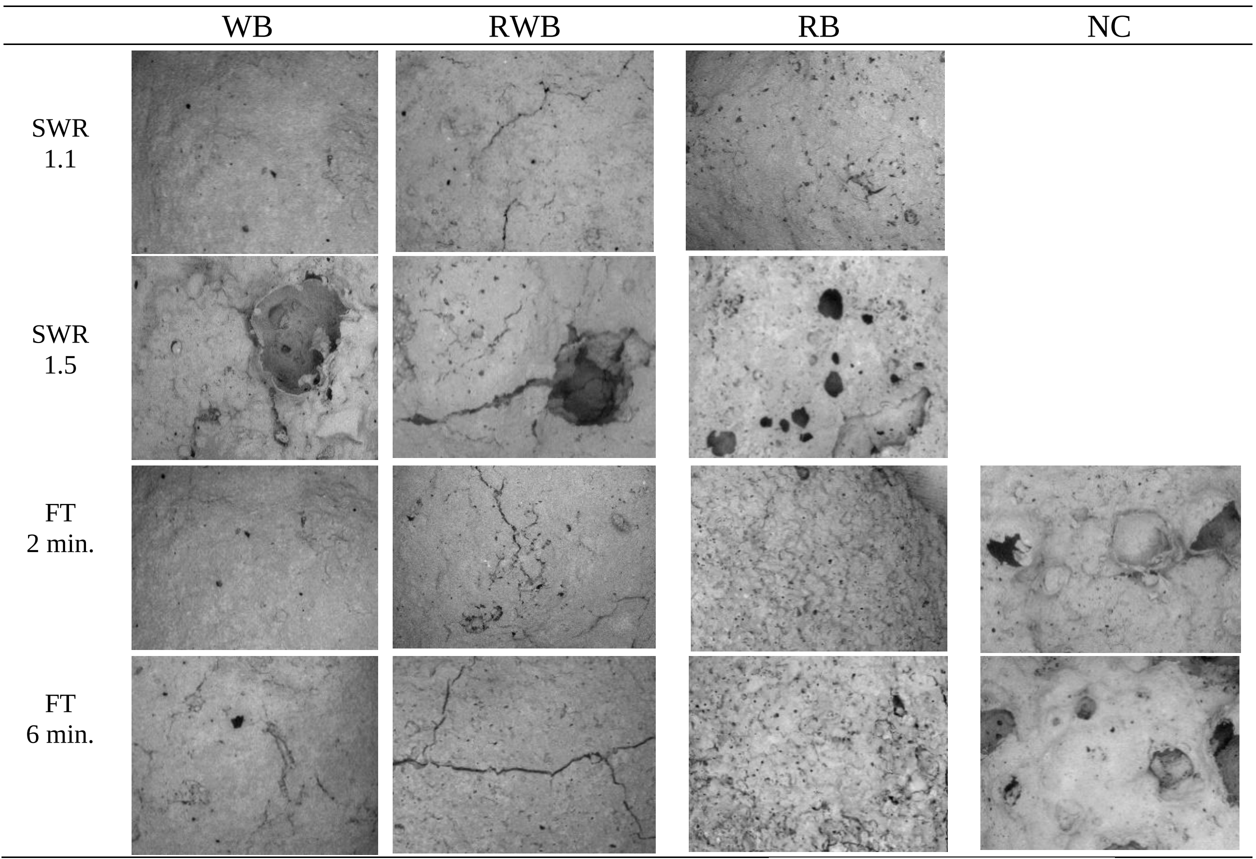


Figure 3.7: Scanning electron microscopy (SEM) image of fried de-oiled crust surface. NC, WB, RWB, RB represent non-coated model food, wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coated model food, respectively. FT: frying time (minute). SWR: solid-to-water ratio.

Batters' SWR and FT have shown influence on roughness of crust surface (Figure 3.8), which was reflected as change in fractal dimension (Figure 3.9) of SEM image. Fractal dimension (FD) is an index of surface roughness (Rahimi & Ngadi, 2016b) and surface roughness (represented by FD) of coated samples increased with frying. Comparatively smooth surface (lower FD) were observed for batter systems of higher SWR. For identical SWR and FT, wheat-flour based batter systems developed smoother surface (lower FD) compared to rice-flour based batters.

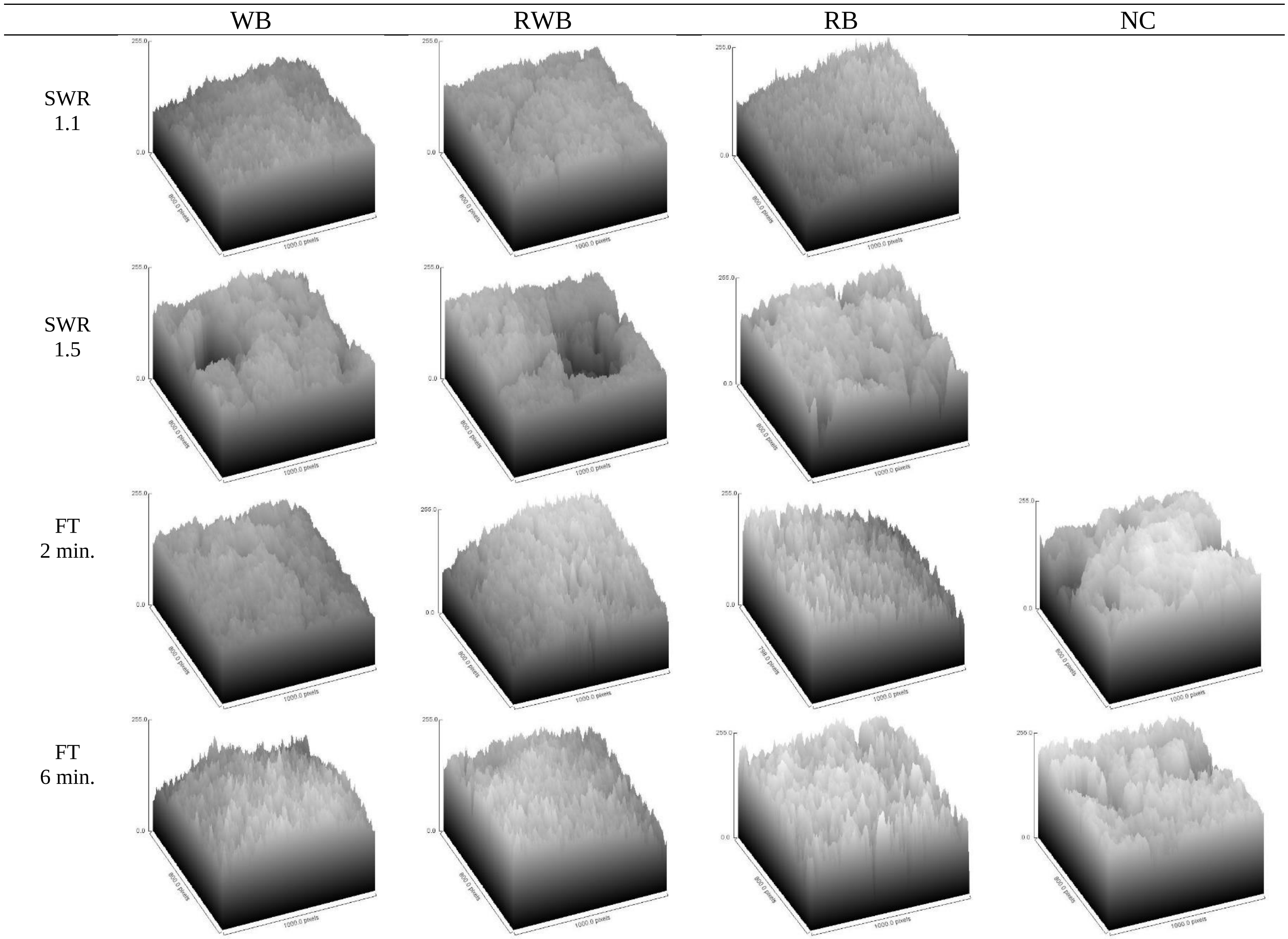


Figure 3.8: Surface-plot depicting roughness of fried de-oiled crust. NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. FT: frying time (minute). SWR: solid-to-water ratio.

Figure 3.9 depicts, correlation between solid-to-water ratio (SWR), frying time (FT), surface openings (SO), and fractal dimension (FD). Compared to noncoated sample, lower presence of SO in batter coated fried samples explains their lower moisture loss and consequent lower fat uptake. Presence of SO in coated samples have shown positive correlation with SWR of batter coatings, and SO of samples has shown an increasing trend with the increase of FT. Increasing trend of SO explains the presence of lower moisture and higher fat in fried samples of higher SWR and FT; as a positive correlation was observed between SO and CrF while the correlation between SO and CrM was negative (Figure 3.10). However, smooth surface (lower FD) having higher micro-opening (SO) gives the impression that loss of moisture from batter-systems of higher SWR were comparatively easy and smooth (due to lower water retention ability of the batter systems); and that had resulted in their higher fat content.

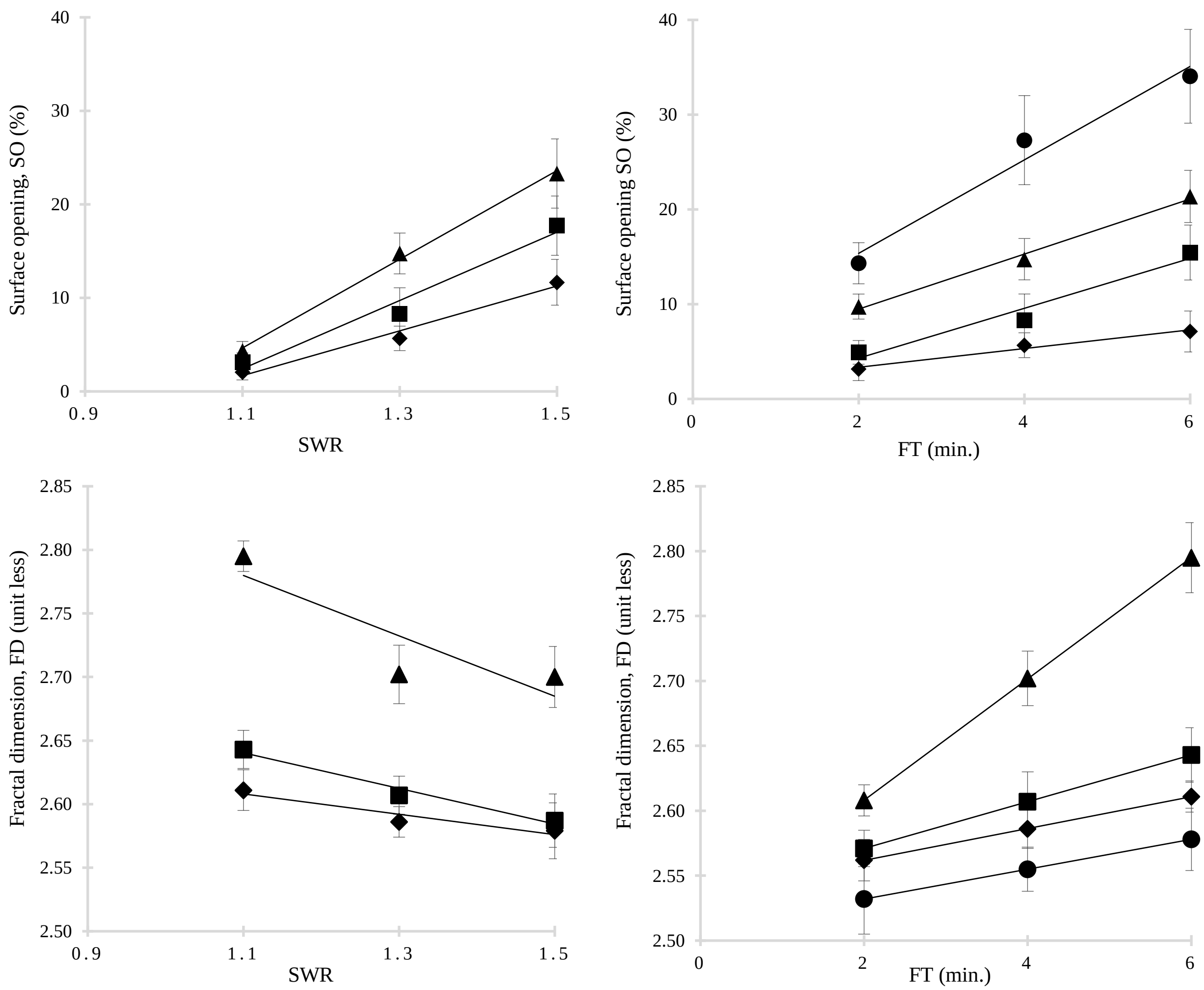


Figure 3.9: SEM image based estimated surface openings and fractal dimension. NC (●), RB (▲), RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time (minute). SWR: solid-to-water ratio.

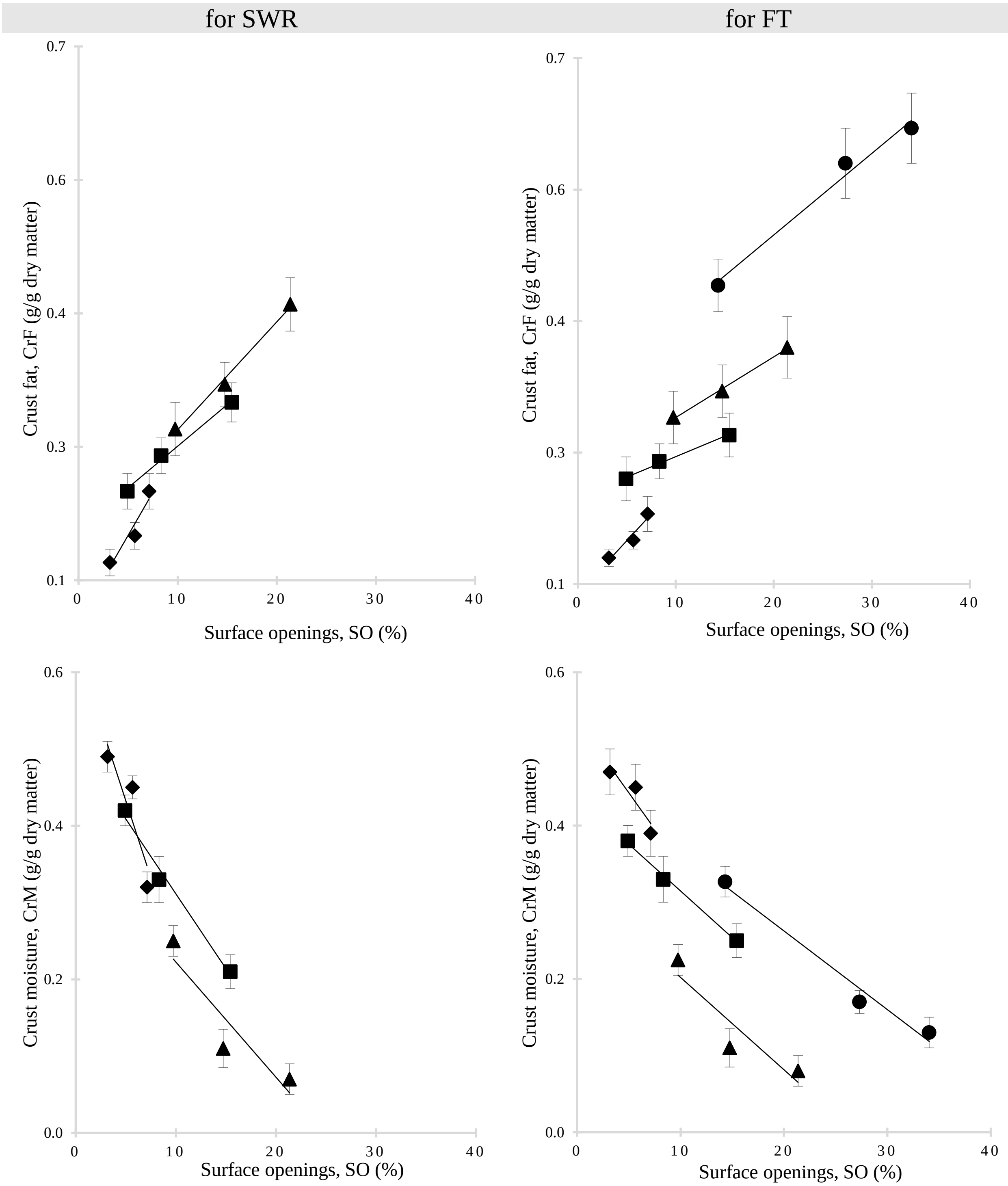


Figure 3.10: Plot of correlation between surface moisture-fat profile and surface openings. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time (minute). SWR: solid-to-water ratio.

3.5 Conclusion

Batter coatings of different solid-to-water ratio (SWR) have possessed distinctive rheological properties and water retention ability (%), that impacts batter pickup (%), frying loss (%), and cooking yield (%) of meat-analog based coated food. Batter coatings of higher SWR has developed crust of having higher textural attributes (hardness, brittleness, crispiness) and stable color features, whereas lower SWR had favored the retention of juiciness of food-core substrate. Higher textural and color stability of the crust were resulted by longer frying time (FT). The fat content of coated food significantly increased with the increase of FT. Both the SWR and FT have shown considerable influence on crust structure, thermal properties, and surface topography. Higher SWR and FT significantly reduced post-fry mass redistribution, textural evolution, and color changes. Lower SWR and FT favored retention of the juiciness of food core substrate, which results in undesirable post-fry changes in textural properties. Rice-flour based batter has shown higher responsiveness (moisture-fat interaction during frying) to the modification of SWR and FT, whereas wheat-flour based batter coated food has shown proneness to post-fry physicochemical changes. Modification of batters' SWR and FT could be used to produce fried products of having desirable and stable physicochemical, thermal and structural properties.

CONNECTING TEXT TO CHAPTER 4

Previous study (Chapter 3) showed that batter systems and process duration significantly affects the mass-transfer associated properties of coated fried foods, which include surface structure. Hence, it could be hypothesized that the surface characteristics of batter coated fried foods might affect post-frying oil redistribution as well as the effectivity of post-frying de-oiling techniques. In Chapter 4, interplay between surface microstructure of batter coated fried food and mass re-distribution under different external treatments are investigated.

Parts of this chapter have been presented at scientific conference.

Md. Hafizur Rahman Bhuiyan and Michael O. Ngadi. (2021). Post-fry surface de-oiling enhance oil-reduction functionality of tempura batter coating. Presented at American Society of Biological and Agricultural Engineering (ASABE) annual conference. Held virtually from July 11th –July 14th, 2021. No. 2100630. *Oral Presentation*.

CHAPTER 4

POST-FRY OIL DISTRIBUTION IN BATTER COATED FRIED FOODS

4.1 Abstract

Surface characteristics are important contributors to quality of fried foods. The goal of this study was to examine the functionality of batter coatings in terms of post-fry surface de-oiling. Wheat and rice flour-based batter systems were used to coat a meat analog model food. The product was deep fried at 180°C for 2, 4 and 6 mins in canola oil. Different post-fry surface de-oiling techniques namely hot air blow (HAB), pressure assisted absorbent paper (PAP), and centrifuge assisted absorbent paper (CAP) were investigated. Surface structure of the products were assessed by scanning electron microscopy. The results showed that batter coated samples had distinct spatial (surface, matrix and total) fat profiles compared to the non-coated sample. Surface fat (SF) of the coated samples were related to both batter formulations and frying time. Different post-fry de-oiling treatments reduced the SF, matrix fat (CrMF) and consequently crust total fat (CrF). The impact of post-fry surface de-oiling treatments were more obvious for samples with higher SF. Non-destructive, ATR-FTIR spectroscopic analysis revealed that surface fat of batter coated fried food was comparatively static and removable during post-fry holding. The CAP treatment reduced CrF to the highest extent, followed by PAP and HAB. Post-fry surface de-oiling treatments can reduce oil from fried batter coated food, without distorting textural and color attributes. Effectiveness of post-fry de-oiling treatments apparently were related to the surface microstructure of coated products.

4.2 Introduction

Deep-fat-fried products are very popular around the globe. However, presence of higher amount of fat in fried products is a major health concern due to its association with obesity, coronary heart diseases, diabetics, cancer causing compounds, and so on (Zhang *et al*., 2020; Dourado *et al*., 2019; Hauzoukim *et al*., 2019). Hence, in producing high quality fried product, the use of fat reduction technology is unquestioned. In this respect, food surface modification by batter coating is an effective approach for a number of reasons. Oil uptake in food occurs through their surfaces and tempura-batter creates uniform layer over food surface that influences heat and mass transfer process (Devi *et al*., 2020; Zhang *et al*., 2020; Liberty *et al*., 2019; Rahimi & Ngadi, 2014a; Ngadi *et al.,* 2006). In this context, surface topography is an essential attribute of solid food which could impact the behavior of coated food during processing and storage (Rahimi & Ngadi, 2016a; Quevedo *et al*. 2002). It is notable that fat on surface of fried foods may constitute up to 80% of its total fat content (Rahimi & Ngadi, 2016a). It has been reported that bulk of the oil in fried foods are absorbed during post-frying stage, and final oil content of fried products strongly depends on post-fry practices (Zhang *et al*., 2020; Devi *et al*., 2020; Liberty *et al*., 2019; Adedeji & Ngadi, 2018; Patsioura *et al*., 2016; Rahimi & Ngadi, 2014b). It is expected that removal of surface oil after frying would be an interesting way of reducing fat content of fried foods. Therefore, it could be hypothesized that post-fry surface de-oiling would enhance the effectiveness of batter-coating in reducing fat content of fried foods. However, there is paucity of scientific literature detailing this novel aspect in producing healthier fried foods. In addition, there is no information about the impact of post-fry de-oiling treatments on textural and color attributes of batter coated products, which are two crucial attributes regarding its consumer satisfaction.

Post-fry surface de-oiling is an emerging concept, hence, there is availability of only limited methods that could be used for the purpose. Zhang *et al*. (2020) and Liberty *et al*. (2019) mentioned the use of hot air as a potential de-fatting technique. Relevant studies projected that oil content of fried foods could be reduced by surface de-oiling with the aid of centrifugation (Zhang *et al*., 2020; Kim & Moreira, 2013; Sothornvit, 2011; Rosanag *et al*., 2009). It is conjectured, that holding the fried products in contact with high absorbent paper could remove surface oil and could reduce the final-fat content of fried foods (Zhang *et al*., 2020; Debnath *et al*. 2009). Further, literature verifies that de-oiling techniques have been applied mostly to thin-fried-food (e.g., potato chips) and preferably in association with vacuum-frying (Zhang *et al*., 2020; Liberty *et al*., 2019; Rosanag *et al*., 2009). Scientific studies on post-fry de-oiling of thick, crispy-moisty, atmospheric fried, multi-component batter-coated products (e.g., nuggets, fish strips, chicken strips, etc.) are not available; and most of the previous research on batter technology was focused on pre-frying and frying process. However, integrated contribution from food producer, service provider, and consumer are indubitable; to enjoy the taste of fried foods without the worries of its fat-related health consequences. In addition, post-fry oil recovery would be economically advantageous along with its positive health impact.

It is notable, conventional solvent-based fat determination methods are laborious, time consuming, requires sample preparation, and destructive in nature (Rohman & Che Man, 2013); which are major constrains in studying the post-fry oil re-distribution in fried samples. In addition, there is lack of method for assessing surface moisture of fried samples. Simultaneous assessment of fried foods surface-fat and surface-moisture based on non-destructive ATR-FTIR spectral analysis could be an effective means; as FTIR spectroscopy can produce high-quality spectra in a short time (<minute) without extensive sample preparation and also can provide information about the intensities & distribution of molecules within sample (Rahman & Yu, 2017; Liu & Yu, 2016).

Considering the paucity of scientific literature on the mentioned aspects, the overall goals of this study were set to (i) study the impact of batter coatings on post-fry surface de-oiling from fried food (ii) investigate the impact of post-fry surface de-oiling treatments on textural and color attributes of batter-coated fried product (iii) assess the effectivity of hot air blow, pressure-assisted absorbent paper, and centrifuge-assisted absorbent paper treatments as post-fry surface de-oiling method of batter-coated fried product (iv) study the impact of batter coating and frying-time on surface topography in relation to spatial fat profile and post-fry surface de-oiling.

4.3 Materials and Methods

4.3.1 Materials

Soy protein isolate was purchased from MP Biomedicals (29525 Fountain Pkwy, OH, USA). Wheat gluten was supplied by Sigma-Aldrich Co. (Ontario, Canada). Methylcellulose (MC) and Sodium pyrophosphate ($Na_4P_2O_7$) crystals were received from Fisher Scientific (Fair Lawn, New Jersey, USA). Sodium bicarbonate ($NaHCO_3$) was supplied by Church & Dwight Canada Corp. (Ontario, Canada). Rice flour (Suraj®, composed of 6.67 % protein, 83.32% carbohydrate, 1.33 % fat and 8.68% moisture) was procured from a grocery store in Montreal, Canada. Commercial wheat flour (Five Roses®, composed of 13.33 % protein, 73.33% carbohydrate, 3.33% fiber, 1.33 % fat and 8.68% moisture), NaCl (Sifto, Compass Minerals Canada Corp.) and Canola oil (Sans nom®, Loblaws Inc.) were purchased from a grocery store in Sainte-Anne-de-Bellevue, Canada.

4.3.2 Sample preparation

A meat analog model food was formulated with soy protein isolate (SPI), wheat gluten (WG), canola oil (CO) and distilled water (DW). Firstly, wheat gluten was mixed with canola oil, and as soon as homogeneous slurry was obtained, soya protein isolate was added and mixed thoroughly. Then distilled water was added and mixed properly until a very soft-textured dough was developed. The respective proportion (g) of the ingredients WG: CO: SPI: DW were 12:15:22:73. The prepared dough was used to fill individual rectangular-shaped cavities of silicon mold (Freshware CB-115RD 24-Cavity Silicone Mini Mold). Covered silicon mold was held in complete horizontal position (surrounded by water, but not immersed) on top of a flat metal wire surface and heated at 70°C for 15 min in a water bath to uniformly gelatinize the material inside each cavity of silicon mold. After heat treatment, mold was allowed to reach equilibrium with room temperature. Then aluminum cover was removed, and gentle pressure (by finger) was applied from the back of each cavity, to bring out the formed substrates without any fracture. This substrate was used as meat-analog model food, weighing an average weight of 16±0.2 g/substrate and having a uniform dimension of 5.6 cm x 2.5 cm x 1.3 cm. The prepared model-food was packed in zip-lock plastic bags and stored in refrigerator (4±2°C) for 24 hours, before its use.

Tempura-batter coatings were used to coat the model food. To prepare batter coatings: white crystal of Sodium pyrophosphate ($Na_4P_2O_7$) was gently crushed and grinded with mortar-pastel to obtain as fine white powder. Then Sodium bicarbonate (1.4%), Methylcellulose (0.3%) and NaCl (1.5%) were added and mixed properly with Sodium pyrophosphate powder (1.8%), to form a homogeneous powder mix. The powder-mixture was further thoroughly mixed with dry flour in a proportion of 5% (powder mix) to 95% (flour). Distilled water (at chilled condition) was added gently to the solid ingredients with a total solid to water ratio of 1:1.3 and mixed them properly until a homogenous tempura-batter slurry was obtained. Wheat based batters (WB) were formulated using only wheat flour whereas rice based batters (RB) were formulated with rice flour. Some samples were formulated using equal mixture of wheat and rice based batters (RWB). Batter systems were kept for 5 min at room environment and then used to coat the model food.

The model food was brought out of refrigerator and kept at room environment for 30 min, prior to batter coating. It was fully immersed in batter system for 1 minute and then removed from batter solution to make a complete coat. The coated product was held on a kitchen-fork for about 30 sec, to drain the excess amount, in order to obtain a uniform layer of coating around model food core. Non-coated (NC) model product was used as control sample.

A programmable deep fat fryer (T-fal Compact, FF122851, China) was used to fry the samples at atmospheric condition (at barometric pressure of 1 atm). Fryer was filled with 1.5 L of fresh canola oil and temperature was set to 180°C. Frying-oil was preheated for 1 hr and stirred to minimize variation in oil temperature. Each time three sample was transferred into the fryer and uninterruptedly fried for 2, 4 or 6 min maintaining a sample to frying-oil ratio of 1:30. It was cautiously monitored that all surface of the samples were fully immersed in frying oil. At the end of frying, the frying-basket was removed from the oil and was shaken 5 times, and immediately transferred to the de-oiling treatment facilities. To minimize variation in frying-oil properties (due to degradation during frying) each batch of oil was used for frying only seven batches of samples (three sample/batch) equivalent to around 30 min of frying operation; before the used frying-oil was replaced with a new batch of fresh oil.

4.3.3 De-oiling techniques

Hot air blow (HAB), pressure-assisted absorbent paper (PAP), and centrifuge-assisted absorbent paper (CAP) were used as post-fry surface de-oiling treatments and are detailed in the following section. For NSD (no surface-de-oiling), the fried samples were not treated by any of the mentioned de-oiling methods.

For HAB treatment, fried sample was placed on a horizontal wire mesh and an air blower system (Nalk & Rey pro compact, NRHD24AC-17, Massy Cedex, France) was used to blow hot-air over the surface of fried sample. Air-blower was installed on laboratory work bench and was arranged in such a way that blows hot-air to the surface of fried products at a contact angle of 45^0. Relative distance between nozzle of the air-blower and surface of the fried sample was fixed at 2.5 cm. To achieve constant air flow, the blower system was turned on 1 min before starting the post-fry surface de-oiling treatment. Hot air (65°C) was blown in “line pattern” on crust surface and during air blowing a relative movement (0.4 cm/s) of the sample against blower-nozzle was maintained. Hot air was blown for 15 sec on each of the two longest surfaces (by flipping over), totaling a surface de-oiling treatment of 30 sec.

To apply PAP treatment, 6 closely attached individual layers of high absorbent paper (Royale® facial tissue, Irving Consumer Products Ltd., E1A 6X4, Canada) were placed on the flat surface of a completely dry, stainless-steel tray. Fried sample was placed on center region of the absorbent paper arrangement (21.5 cm X 19.3 cm) as such, the longest surface of the fried sample was in direct contact with paper absorbent. Another six layers of absorbent paper was placed on top surface, making a sandwich like system. On top of that, a flat-solid-inert bar was carefully placed (mimicking human hand palm) to provide a constant-gentle pressure of 0.11 N/cm^2; to confirm and enhance the contact between absorbent paper and crust surface of fried food, in order to maximize surface de-oiling effect without impacting structure of the crust. After 15 sec of pressure application, which is equivalent to a total of 30 sec (as pressure acts on both two longest-facet surfaces) of pressure-assisted absorbent paper surface-de-oiling treatment, both the absorbent paper and applied pressure was carefully removed (confirming that no part of the paper absorbent was attached with crust surface), and treated sample was used for further studies.

CAP treatment was applied by placing fried samples in a centrifuge (Sorvall Legend XTR, 50119927-5, Thermo scientific, Germany) and centrifuging at the maximum setting of 9. Following the approach of Rosanag *et al*., (2009) and Khalilian et al., (2021) with required modification, the Xg (relative centrifugal force) at full speed of centrifugation was set to a value of 100. The centrifugal force treatment consisted of three sequential-uninterrupted steps i.e., speed acceleration (10 sec), full speed (10 sec) and decelerating speed (10 sec), totaling a duration of 30 secs. After centrifugation, samples were carefully brought out (without touching the oily surface) of centrifuge-cavity and the procedure detailed in PAP section were followed before further analysis.

4.3.4 Fat and moisture determination

Fat content of the fried foods crust was grouped into three categories, namely surface fat (SF), matrix fat (CrMF), and crust total fat (CrF). SF was defined as the fat that adhered on the outer surface of the fried product. CrMF was the fat that was present within the crust matrix whereas, CrF included both the surface fat and the matrix fat. These fractions of fat content were determined following the methodology detailed by Adedeji & Ngadi, (2018), Rahimi et al. (2017), Pedreschi *et al*. (2008), and Rahimi & Ngadi (2016a, 2014b), with required modifications. Firstly, to measure the SF, surface of the fried product was washed by immersing in 140 ml glass extraction vessel containing 100 ml petroleum ether (to fully cover the sample) for about 5 secs at 25°C. The mass of extracted (surface) oil were determined, by vaporizing (at 130°C) the solvent in a solvent extractor for 30 min and subsequently placing in desiccator for 30 min. The surface-washed (by petroleum ether) hot fried product was held at room environment to evaporate any residual petroleum ether from surface of the product. After that, outer crust portion of the surface-washed product was carefully detached by a microtome blade (Feather C35 type, Japan) from their core region. Detached crust was freeze dried in a freeze-dryer (Modulyod-115; Thermo Savant, NY, USA) at -50°C and 250 mbar for 48 hours and then transferred into a desiccator to equilibrate for 30 minute. Weight of the crust sample before and after freeze-drying was measured and crust moisture (CrM) content was calculated on dry weight basis (db, g/g dry matter). To determine crust matrix fat (CrMF), the detached freeze-dried crust sample was ground with a pestle and mortar, after which grounded sample was placed in a thimble for oil extraction in a VELP SER 148 (Velp Scientifica, Usmate, Italy) extraction unit using petroleum ether as solvent. Crust matrix-fat extraction process were performed in three consequent steps: immersion (30 min), washing (30 min), and recovery (30 min), totaling 90 minute.

The SF content was computed by dividing the mass of extracted-oil (from surface) by mass of the dry matter (including SF extracted) of detached freeze-dried crust. The CrMF was computed by dividing the mass of the extracted oil (from freeze-dried crust portion) by mass of the dry matter (including surface and matrix oil) of the crust. Total crust fat (CrF) was estimated as summation of SF and CrMF, divided by the total mass of the crust. All fraction of fat was computed on dry weight basis and reported as (g/g dry matter).

4.3.5 Texture and color assessment

Textural properties (hardness, brittleness, crispiness) were evaluated by puncture/compression test, using a texture analyzer (Stable Micro Systems Texture Analyzer, TA. HD PlusC, serial: 6025, Surrey, GU7 1YL, UK), by following the detailed approach of Rahimi *et al*. (2017). And the surface color (CIE Lab) was assessed by spectrophotometer (Minolta Spectrophotometer CM-3500d, Japan).

4.3.6 ATR-FTIR Spectroscopy

FTIR spectra of crust surfaces were recorded by an ATR-FTIR spectrometer (Thermo Scientific Nicolet iS5 FTIR, Thermo Fisher Scientific, USA). The operation of spectrophotometer, acquisition and manipulation of spectra were performed using the Omnic software (version 6.1, Nicolet, Madison, WI, USA). Crust surface of the fried product was in directed contact with ATR-crystal and spectra acquisition was at control ambient temperature of 25°C. ATR-crystal was cleaned with ethanol to remove any residual (especially fat) by the previous sample. FTIR spectra were collected over the wavenumber range of 4000–950 cm^{-1} by co-adding 32 scans and at resolution of 4 cm^{-1}. All spectra were normalized against background of air spectrum. After every 60 minute of operation, a new reference air background spectrum was taken. The spectra were recorded as absorbance values. According to Chen *et al*. (2015) and our preliminary study, absorption peaks at around 2922 cm^{-1} and 2852 cm^{-1} (asymmetrical and symmetrical stretching of –CH_2), 1743 cm^{-1} (C=O stretching), and 1157 cm^{-1} (-C-O stretch; -CH_2 bending) of the FTIR spectra were considered as surface fat response i.e., indication of the presence of frying canola oil on crust surface. In this aspect, considering relevant studies (Tarhan *et al*., 2017; Rahman & Yu, 2017;Rohman *et al*., 2016; Rohman & Che Man, 2013; Li-Chan *et al.*, 2006; Bouchon *et al.*, 2001), absorption area under the FTIR spectral ranges between 1130-1200 cm^{-1}, 1700-1775 cm^{-1}, and 2800-3000 cm^{-1} were calculated by Omnic software and their summed value was reported as surface fat response (SFR). According to Nicolaisen (2009) and Efimov *et al*. (2003), a broad peak around 3000–3700 cm^{-1} region was due to the components of asymmetric and symmetric stretching modes of the H_2O molecule. Absorption area in FTIR spectra within this (3000–3700 cm^{-1}) range as well as absorption between 1525-1725 cm^{-1} region were considered as surface moisture response (SMR).

4.3.7 Scanning electron microscopy

Surface morphology of fried product was assessed by scanning electron microscopy (SEM). Surface washed (by three consecutive charges of petroleum ether, for a total immersion period of three minute) fried samples were freeze dried and stored in desiccator until imaged by a scanning electron microscope (Hitachi TM3000, Japan). SEM operational setting was adopted from Adedeji & Ngadi, (2018) and performed with required modification. In brief; 5 kV electron power (to prevent sample modification due to heat generated at higher voltage) was applied, auto contrast function was chosen, and composition mode of imaging was used. Small cut, rectangular shaped, defatted sample of about 10×10×5 mm in dimension was placed on the sample base with a sticky surface that comprises of carbon tab. After each sample was properly aligned by visual control using two sets of knobs and images shown in the control software on a computer screen, vacuum pressure was created in the sample chamber and images were quickly acquired to prevent charging and heating that could lead to artifacts in images. SEM images were acquired at a magnification of ×30. SEM micrograph was recorded as 8-bit image and surface microstructure was analyzed by Java-based public domain image processing software (ImageJ 1.52 v, National Institutes of Health, USA). Surface opening (SO) was estimated from SEM micrograph, as ratio of the area of total openings (holes, cracks, ruptures, crevices etc.) in surface to the total surface area. In estimating SO from SEM image, the image analysis approach of Rahimi & Ngadi (2015) was followed i.e., darker region represents holes, cracks, ruptures, crevices, while whiter region represents solid matrix. Following the method as detailed by Rahimi & Ngadi (2016a), fractal dimension (FD) of surfaces were estimated from 2D SEM images by box-counting method.

4.3.8 Statistical analysis

All experimental data obtained from triplicate of samples, mean ± SD (standard deviation) was reported. Tukey's honestly significant difference (HSD) was used in reporting significant ($p<0.05$) difference among mean values. All statistical analysis is performed by licensed statistical software (JMP 14.1v, SAS Institute Inc., Cary, NC, USA).

4.4 Results and Discussion

Typical Attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectra of the fried samples are shown in Figure 4.1. ATR-FTIR spectral pattern for all the samples used in the study looked very similar, as expected since they were all fried in same oil. However, there were differences in the intensities of the peaks at the absorption bands of 2922, 2852, 1743, 1157, and 3000-3700 cm^{-1} for the different fried samples. Apparently the absorption peaks at 2922, 2852, 1743, and 1157 cm^{-1} indicate the presence of canola oil on the fried food samples (Chen *et al*. 2015). These bands are close to the absorption bands of reported for other lipids such as soybean, sunflower, avocado, corn, rice bran, palm oil (Tarhan *et al*., 2017; Rahman & Yu, 2017;

Rohman *et al.*, 2016; Rohman & Che Man, 2013; Bouchon *et al.,* 2001). Broad absorbance peak around 3000–3700 cm^{-1} region is attributed to the asymmetric and symmetric stretching modes of H_2O molecule (Nicolaisen 2009; Efimov et al., 2003). The broad absorbance peak (3000–3700 cm^{-1}) as well as peak around 1648 cm^{-1} indicate the presence of moisture on surface of fried samples. The differences in the absorbance peak characteristics (width, height) of different fried samples, represents the relative variation in concentrations.

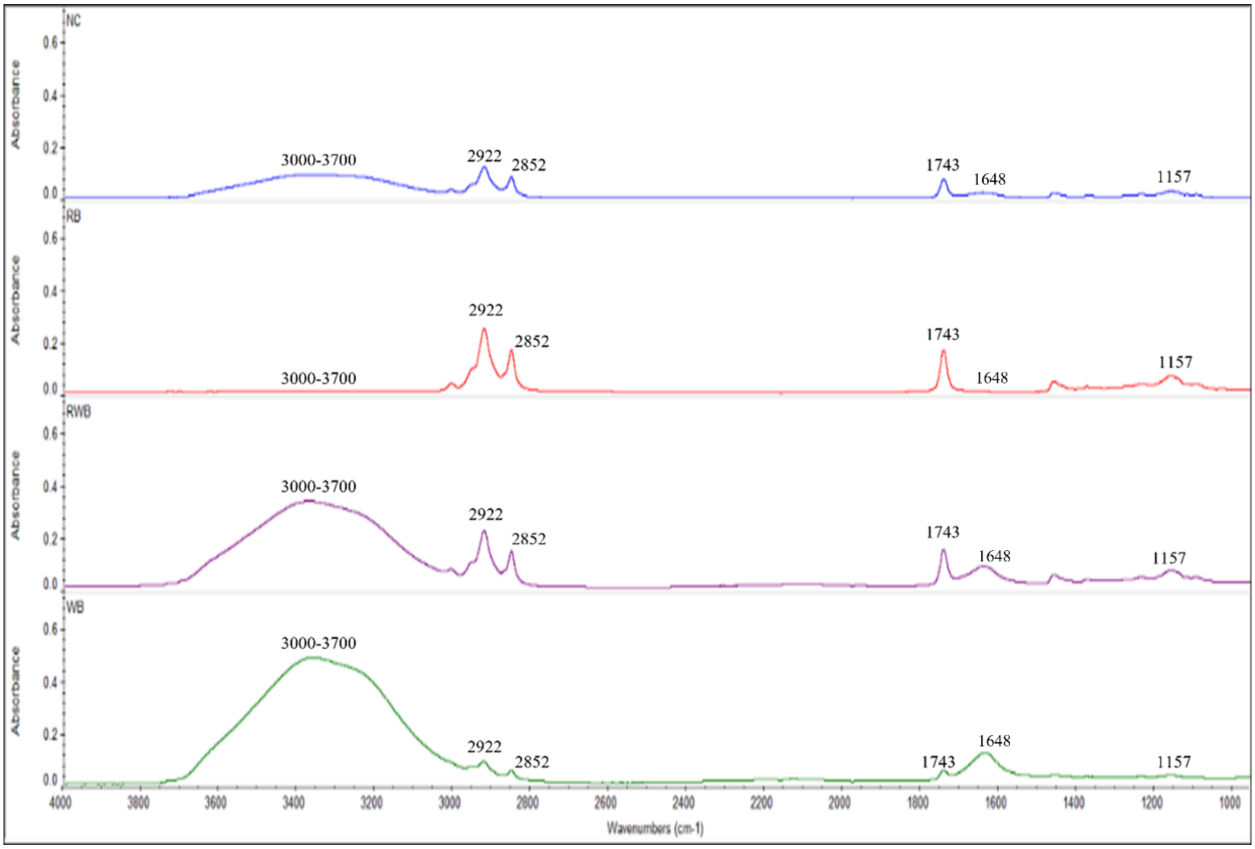


Figure 4.1**:** Typical ATR-FTIR spectra showing surface moisture-fat profile of fried samples. Downwards: NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively.

Figure 4.2 shows the correlation between surface fat (SF) content measured using conventional method and the surface fat response (SFR) obtained from the ATR-FTIR spectral signatures of the samples (fried for 2-6 min). The results showed that there was a strong positive relationship (R^2 >0.96) between the SF and SFR. Higher SFR (a.u.) were recorded for higher SF (g/g dry matter) content, irrespective of batter formulations. Generalized linear model fit value indicates, the surface fat assessment by non-destructive ATR-FTIR spectra collaborates well with the results obtained by the destructive assessment. This validates the use of ATR-FTIR spectral based method as an effective and robust means of studying post-fry oil-redistribution in batter coated fried foods.

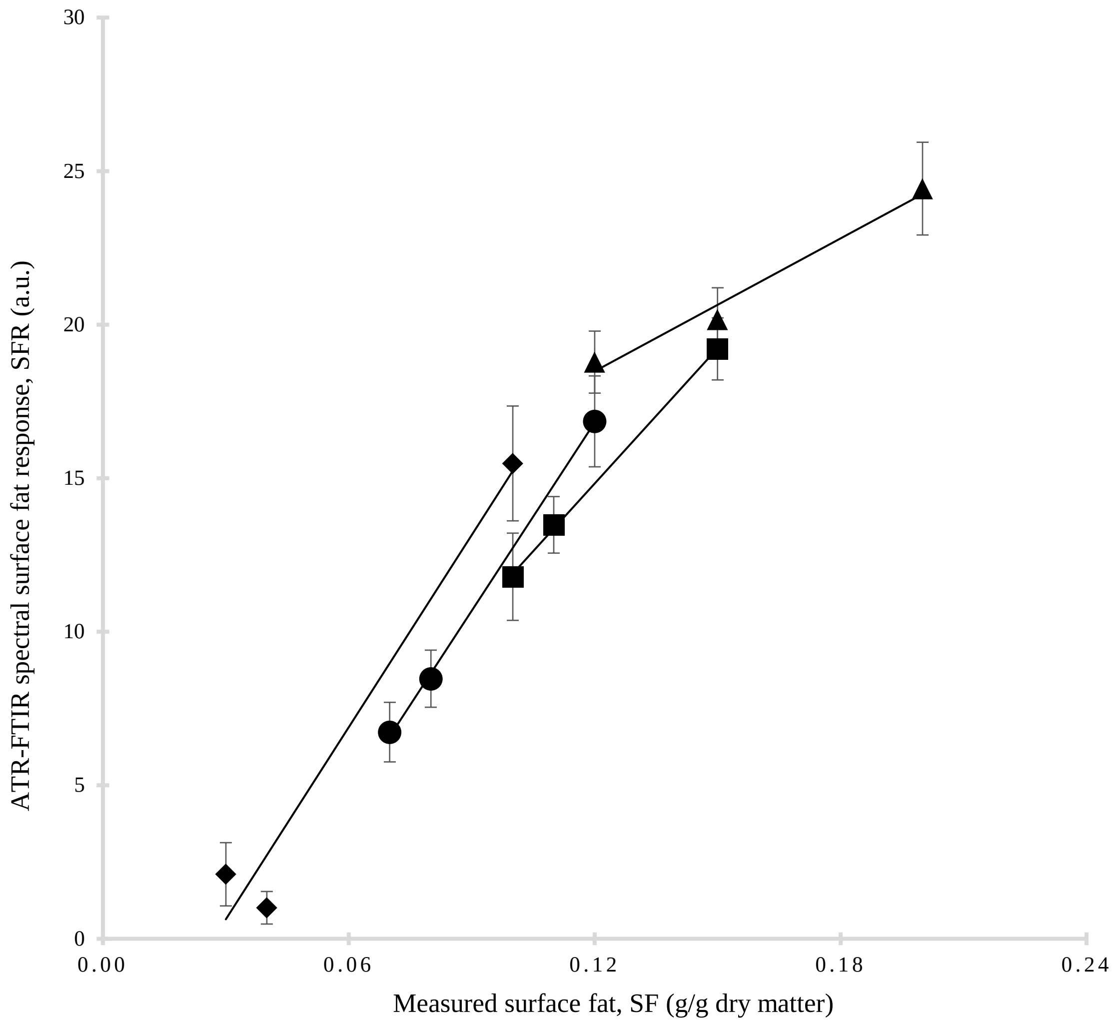


Figure 4.2: Plot of correlation between measured surface fat content and ATR-FTIR spectral surface fat response of fried samples. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively.

For each batter system, there was a high correlation (R^2>0.80) between ATR-FTIR spectral signals for surface fat response (SFR) and surface moisture response (SMR) as shown in Figure 4.3. A negative correlation between surface fat and surface moisture was observed for wheat-flour based batter (WB) and equal proportion of wheat & rice flour based batter (RWB) coated samples. The negative correlations between SFR and SMR are consistent with reported literature on relationship between moisture loss and fat uptake during frying of foods (Rahimi & Ngadi, 2014a & b; Ngadi *et al.*, 2009). However, rice-flour based batter (RB) coated sample showed a slightly different trend showing minimal change in moisture at various fat levels. This behavior of RB coated samples could be understood as the consequential impacts of heat-induced changes in surface structural-chemical properties or an artifact of ATR-FTIR based measurement technique.

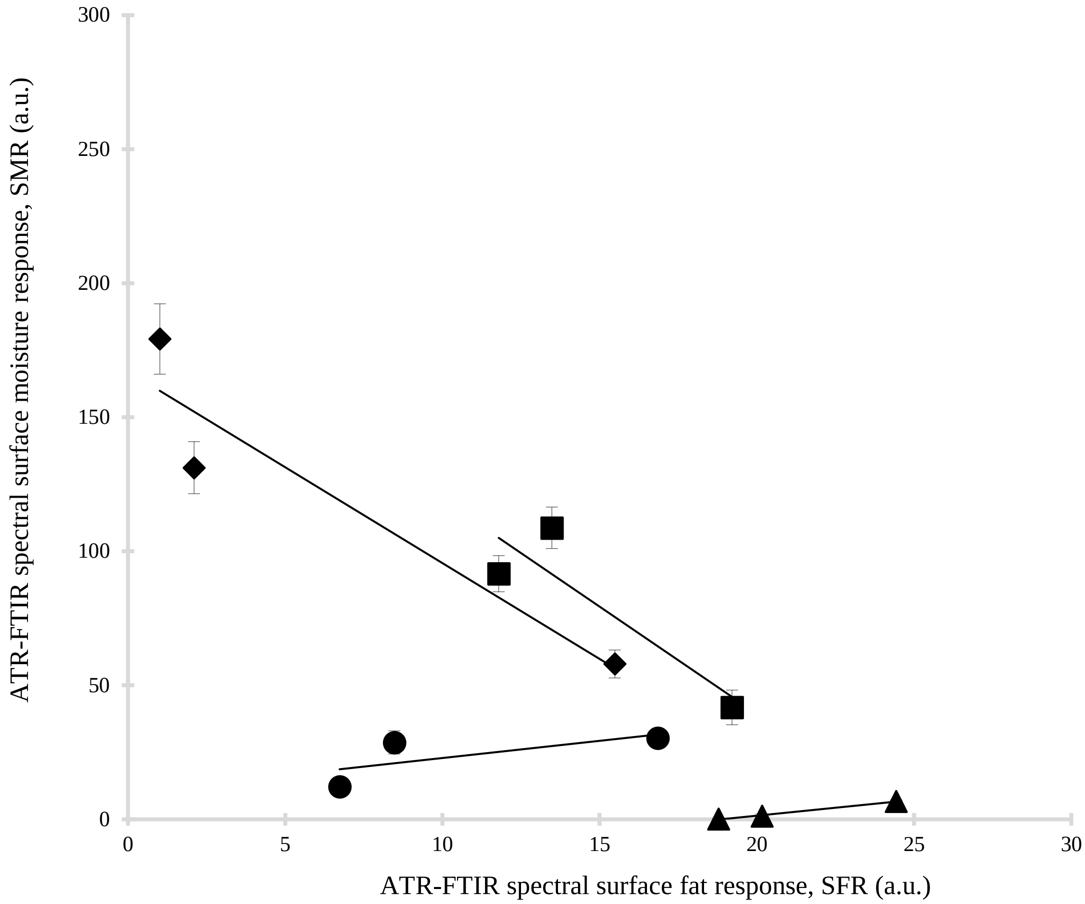


Figure 4.3: Plot of correlation between ATR-FTIR spectral surface fat response and surface moisture response. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated fried (2,4,6 min) samples, respectively.

Figure 4.4 represents simultaneous changes in surface fat and surface moisture of fried sample as function of frying time (FT). The measured surface fat response (SFR) of fried samples showed a decreasing trend with FT. However, surface moisture response (SMR) of NC and RB coated samples has shown a decreasing trend with frying time, whereas for WB and RWB coated samples an irregular overall pattern (e.g., first increase then decrease) was observed within the frying duration of 2 to 6 min. This irregular pattern could be understood as such: at early stage of frying, surface moisture rapidly evaporated (as soon as came in direct contact with hot frying oil) even before the start of any moisture migration from inside to outside surface region, therefore, lower SMR were observed at FT of 2 min. As frying progressed, moisture migration might have started from juicy inside (initial moisture content of 1.49 g/g dry mass) to outer (surface) region of the crust (initial moisture content of 1.3 g/g dry mass), therefore a higher value of SMR was observed at FT 4 min. After a long (6 min) frying treatment, there might be less amount of moisture on the surface which were reflected as lower SMR. Overall, the higher SMR of the WB and RWB coated fried samples could be linked with the higher water holding capacity of protein fractions of wheat flour (Adedeji & Ngadi, 2011; Moreno *et al*., 2010). And after long a longer (6 min) frying treatment, there might be less amount of available moisture on the surface and that was reflected as lower SMR. Overall, the higher SMR of WB and RWB coated fried samples could be associated with the water holding capacity of the formulated batters, as comparatively higher water holding capacity of wheat-flour based batter has been reported in literature (Adedeji & Ngadi, 2011). In contrary, the lower SMR of RB coated fried sample could be understood as such: frying creates void-spaces (pores), frying oil occupied the generated void-spaces and acted as a barrier against moisture-migration toward the outer surface and results in to lower SMR value. Positive correlation between porosity and fat content of fried samples has been reported (Rahimi & Ngadi, 2016b). However, from food processing point of view (i.e., to the aim of producing low surface-fat containing batter coated fried food products), wheat flour-based batter would be preferred over rice-flour based batter. Although longer frying treatment (FT), could be a way to produce batter-coated fried products of having lower fat on products surface, but proper attention should be on total fat content of the fried products as positive relation between FT and crust total fat (CrF) were observed (Figure 4.7). Positive correlation between frying time and fat content of deep-fat-fried samples has been reported in relevant literature (Rahimi & Ngadi, 2016b; Adedeji & Ngadi, 2011).

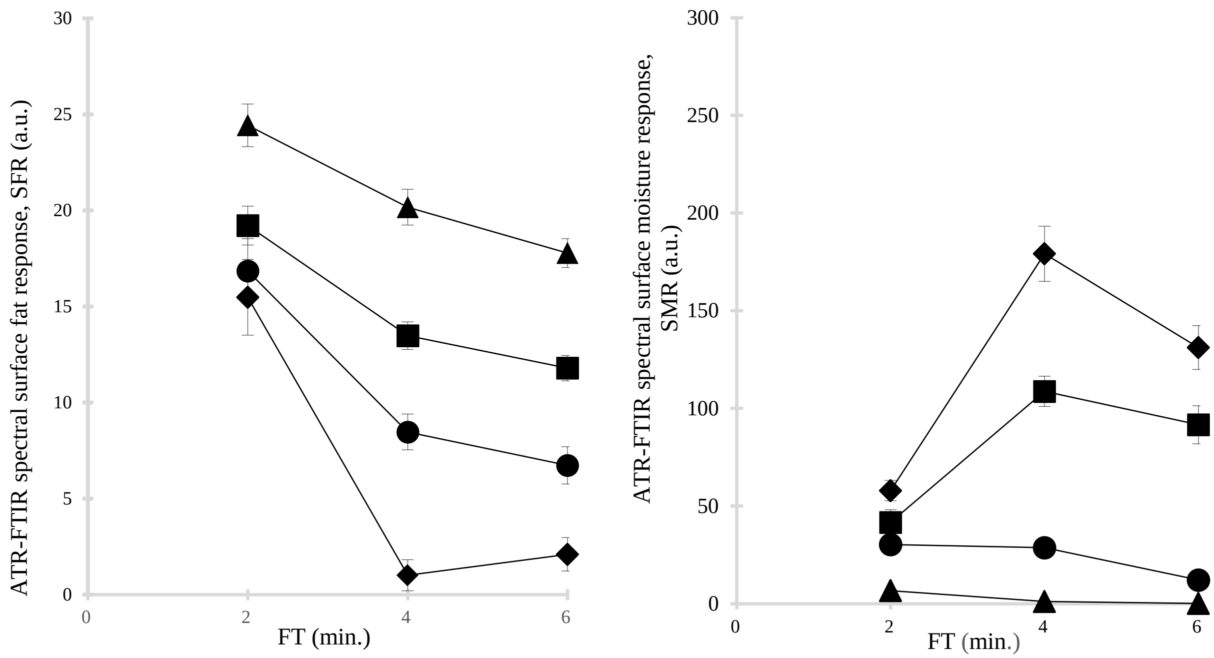


Figure 4.4**:** Surface fat and surface moisture of fried sample, as a function of frying time. NC (●), RB (▲), RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time (minute).

Figure 4.5 represent the evolution of crust (surface, matrix, total) fat and moisture profile of fried (4 min) samples under different post-fry de-oiling treatments. Post-fry de-oiling treatments has reduced the surface fat, matrix fat, and consequentially the total fat content of the crust of fried samples. The centrifuge-assisted absorbent paper (CAP) treatment was most effective in reducing surface, matrix and total fat of the crust, followed by the pressure-assisted absorbent paper (PAP) treatment and the hot air blow (HAB) treatment. The prominent effect of CAP treatment could be attributed to its two stages of oil reduction (i.e., by centrifugal force, and by absorbent paper). Under HAB treatment, the decrease in surface fat were interwoven with the slight increase of crust matrix fat. Though the applied post-fry de-oiling treatments has substantially reduced surface fat, matrix fat and total fat content of the crust, however, weren't able to completely erase the presence of fat on surface. Crust moisture (CrM) content of fried samples weren't considerably impacted by the HAB and PAP treatments, whereas CAP treatment caused slight increase of CrM. Under centrifugal force (in CAP treatment), moisture from inner region might migrated into the outer crust region, and even after the use of absorbent paper (reduces only surface moisture) moisture was present within crust matrix that reflected as increased CrM. However, surface moisture (SMR) of fried samples were considerably reduced by post-fry surface de-oiling treatments.

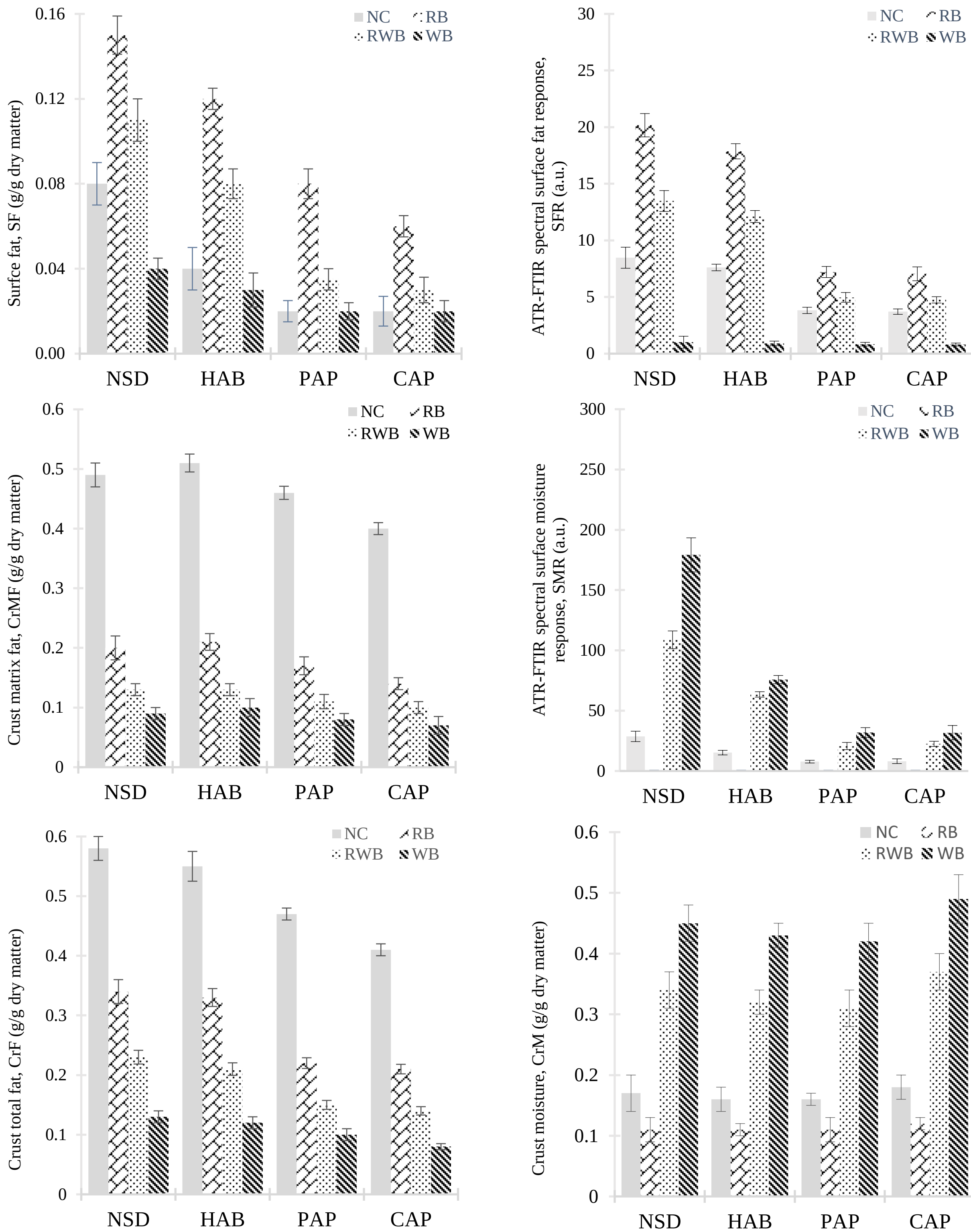


Figure 4.5: Evolution of crust spatial (surface, matrix, total) fat and moisture profile of fried (4 min) samples under different post-fry de-oiling treatments. NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food respectively. NSD, HAB, PAP and CAP represent, no surface-de-oiling, hot air blow, pressure assisted absorbent paper, and centrifuge assisted absorbent paper treatment, respectively.

Texture and color are two crucial attributes relating to the consumers satisfaction of batter coated fried food products. Mechanical texture analysis showed that, textural attributes (hardness, brittleness, crispiness) of batter-coated fried food samples weren't significantly ($p<0.05$) impacted by the applied post-fry de-oiling techniques. Similarly, color attributes (lightness, redness, yellowness) of fried samples weren't significantly impacted by post-fry de-oiling techniques. Non-significant changes in texture & color features are appreciable from consumers point of view, as this indicates the used post-fry de-oiling techniques were effective in reducing fat from fried products without compromising textural and color attributes.

Figure 4.6 depicts the changes in surface fat and surface moisture of fried (4 min) samples during post-fry holding. Surface moisture of wheat-flour based batter (WB) coated fried sample increased with the post-fry holding duration (HD), while the SMR of rice-flour based batter (RB) coated fried sample were relatively unchanged. This unchanged SMR of RB coated samples could be attributed to the presence of high oil content on the surface of rice-flour based batter, as fat (non-polar) and moisture (polar) are antagonistic in chemical nature. However, surface fat of WB and RWB batter coated fried samples were apparently unchanged during post-fry holding, whereas surface fat of non-coated (NC) and RB coated fried sample has shown a decreasing trend with the HD. Changes in surface fat during post-fry holding could be understood as the influence of moisture condensation-induced suction and/or the penetration of surface oil (via., surface openings, cracks, ruptures, interconnected microchannel) into the crust matrix. The changes in surface oil during cooling stage of fried samples such as potato strips (Adedeji & Ngadi, 2018), potato flakes and gluten based formulated products (Moreno *et al*., 2010) has been reported earlier in literature. Differences in the nature (i.e., unchanged in WB coated sample while changed in RB coated sample) of post-frying changes in surface fat could be understood as the resultant effect of the differences in protein content of batter systems, as such: wheat flour contains higher protein, and protein possess good film-forming capacity upon heating which develops less impermeable film i.e., film that blocks oil transfer (Rahimi et al., 2019). Hence, the surface fat of wheat-flour based batter (WB) coated fried samples were not able to move inwards and were comparatively unchanged during post-fry holding. However, compared to non-coated (NC) fried sample, the surface fat of batter-coated fried samples were comparatively stationary in nature, that is favorable to be removed by post-fry de-oiling treatment. However, due to higher availability of surface fat at early stage of post-fry holding, the application of surface de-oiling treatment at soonest possible (after frying) might be highly effective, to reduce fat from fried products.

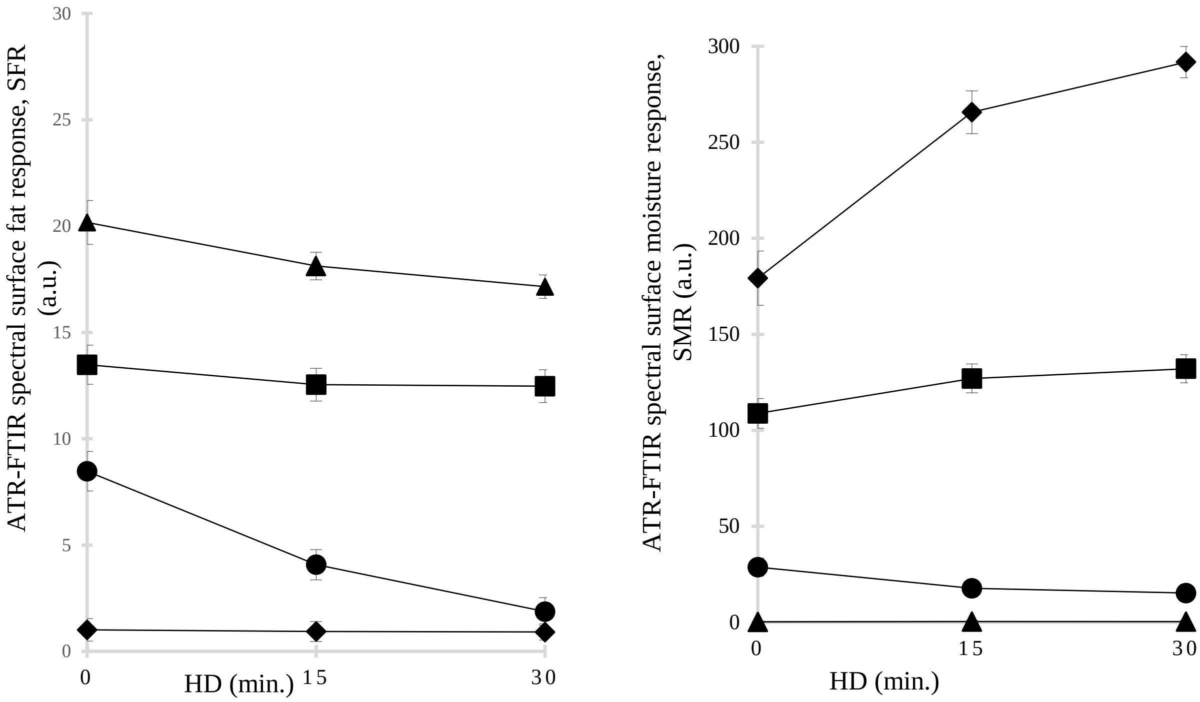


Figure 4.6: Evolution of surface fat-moisture of fried samples as a function of post-fry holding duration. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. HD: holding duration (minute).

For de-oiling of fried foods, the studied PAP treatment is convenient at consumer level. Figure 4.7 depicts the impact of frying time (FT) on effectiveness of post-fry PAP treatment as assessed by both the destructive and non-destructive means. PAP treatment showed prominent effect to reduce fat (SF, CrMF, CrF) from the samples that were fried for shorter time. Post-fry oil reduction effectivity of the PAP treatment were reduced with the increase of frying time i.e., the amount of residual fat (present in sample even after de-oiling treatment) increased with the FT. In a study on g-fried eggplants, Khalilian et al. (2021) observed seemingly high surface oil at the initial stages of frying and interpreted the observation as such: if the food sample is withdrawn from oil at initial stage of frying, the surface oil may easily be removed during the cooling step. However, for any frying duration and even after PAP treatment, all fraction of fat (surface , matrix, crust total) was higher in RB coated fried sample in comparison to WB coated sample. PAP treatment has effectively reduced fat content (g/g dry matter) of the crust without affecting crust moisture (CrM). ATR-FTIR spectral analysis revealed that irrespective of FT, PAP treatment has reduced surface moisture (SMR) of fried samples to a considerable level.

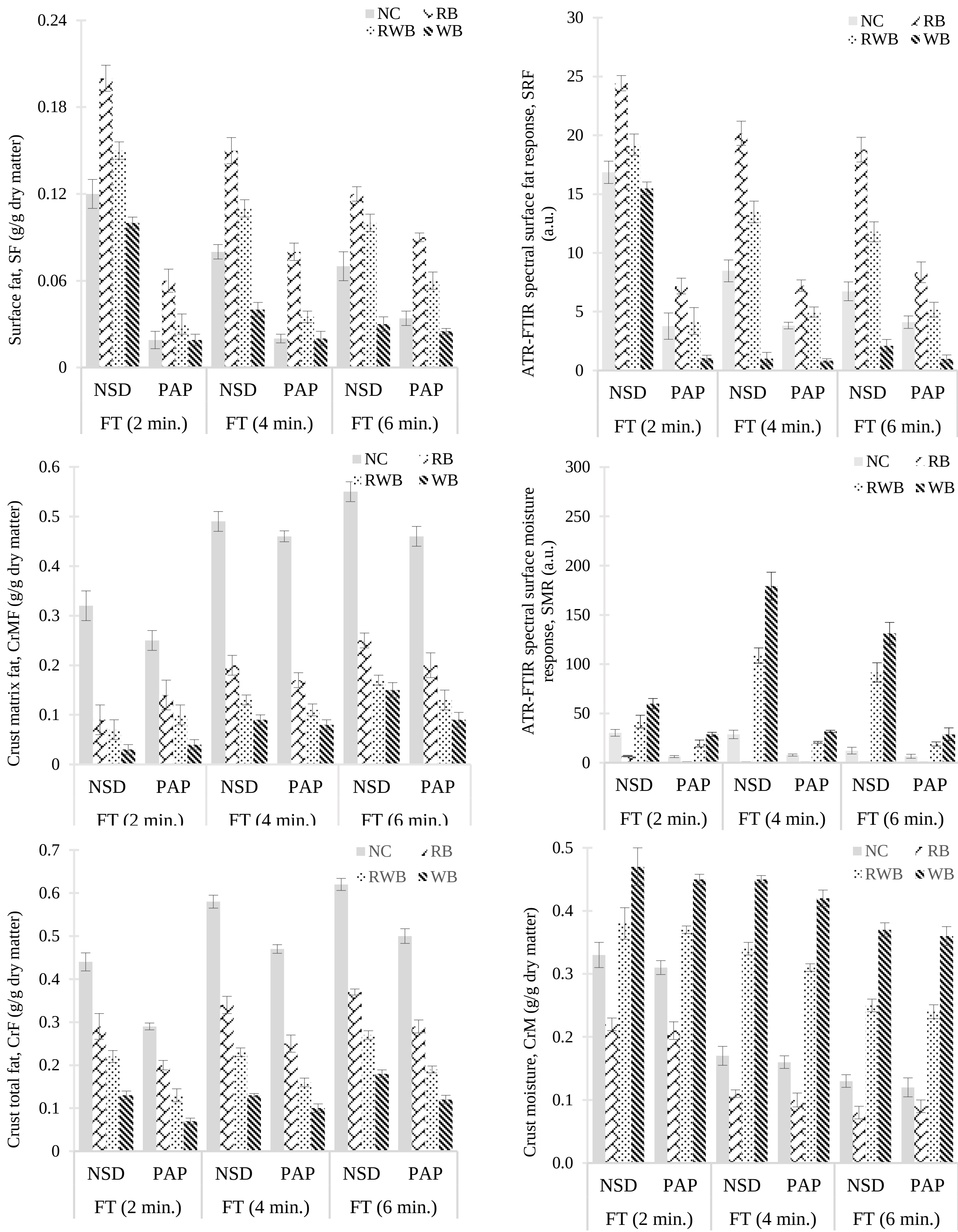
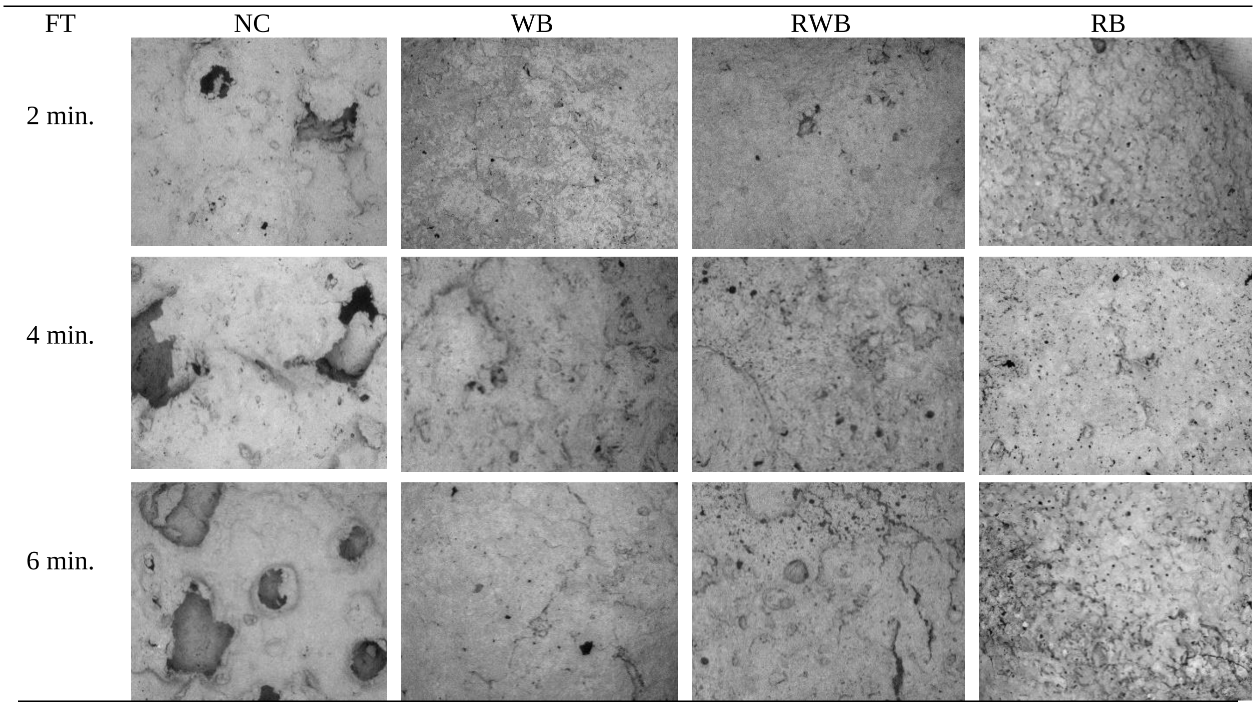


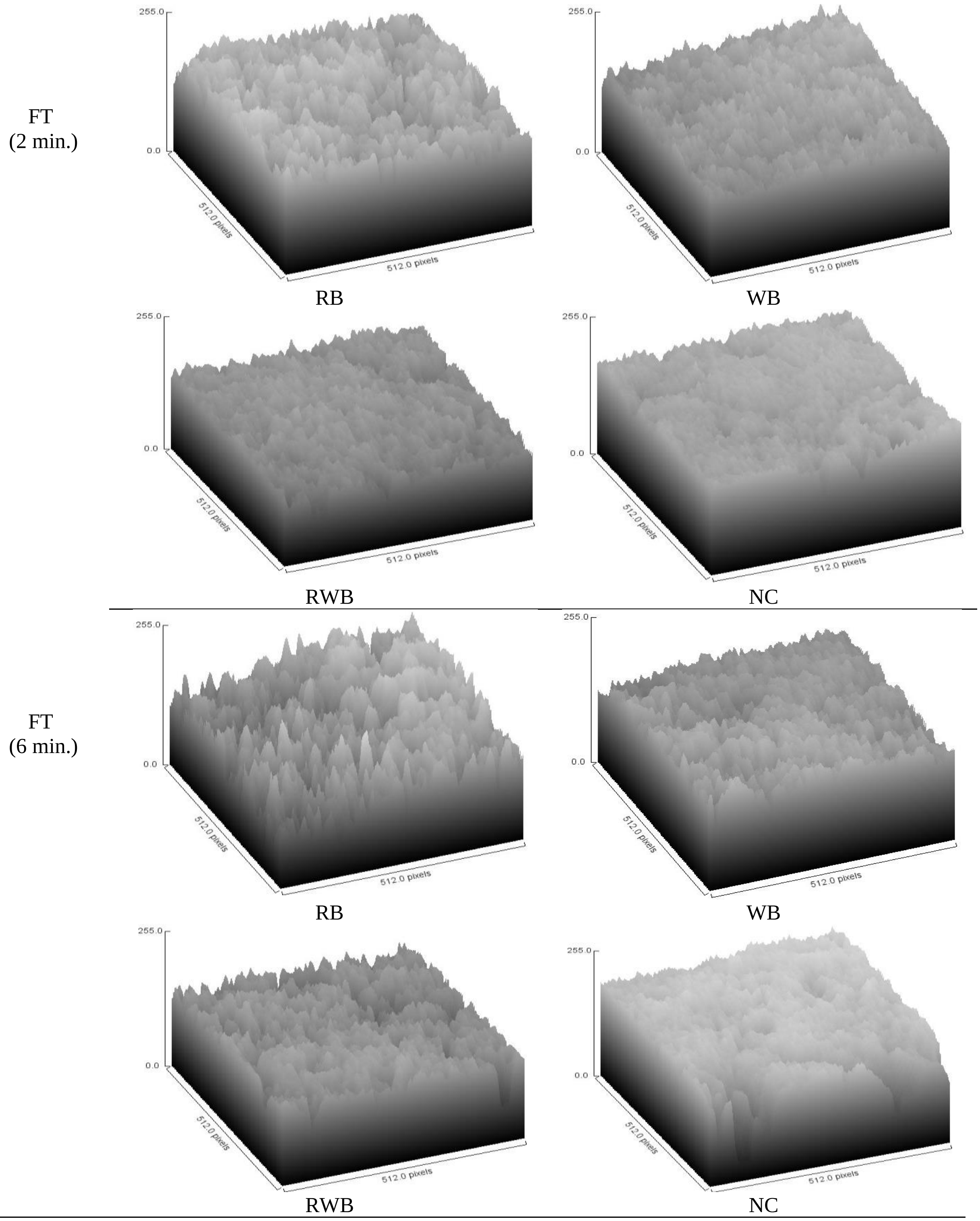
Figure 4.7: Moisture-fat profile as assessed after different frying time and post-fry de-oiling treatment. NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. NSD: no surface de-oiling, PAP: pressure assisted absorbent paper. FT: frying time (minute).

Figure 4.8 depicts, the surface structure of fried samples. Surface microstructure of batter-coated fried samples were different than non-coated sample, that explain their distinctive spatial fat profile. Surface opening (SO) of non-coated fried sample was higher than batter-coated fried samples. These surface openings (hole, crack, rupture, etc.) has favored the frying-oil to go inside the crust matrix and that resulted in higher fat (CrMF, CrF) content of NC samples. Among coated samples, higher SO were observed in RB coated fried sample and lower in WB coated sample. This confirms the influence of batter-formulations on surface microstructure to explain their distinctive crust fat profile (CrMF, CrF). Surface openings increased with the increase of frying (FT). Overall, increase of SO has resulted the decrease of surface fat (SF) and increase of crust matrix fat (CrMF) and crust total fat (CrF). It has been reported, an increase in porosity favors higher fat uptake (Adedeji & Ngadi, 2009).



Attribute	Sample	FT (2 min.)	FT (4 min.)	FT (6 min.)
Surface opening, SO (%)	NC	14.32±2.17^{a}	27.31±4.75^{a}	34.05±6.21^{a}
	RB	9.75±1.32^{b}	14.75±2.18^{b}	21.37±4.15^{b}
	RWB	4.92±1.25^{c}	9.33±2.75^{bc}	15.45±5.37^{bc}
	WB	3.17±1.23^{c}	5.67±1.31^{c}	7.12±2.16^{c}

Figure 4.8**:** SEM image and estimated surface opening of fried foods crust surface. Lower-case letters (a-c) ranks significant ($p<0.05$) difference among samples of identical frying time. NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. FT: frying time (minute).



		WB	RWB	RB	NC
Fractal dimension, FD (unit less)	FT (2 min.)	2.562	2.571	2.608	2.532
	FT (6 min.)	2.611	2.643	2.795	2.578

Figure 4.9: Surface plot depicting roughness of fried de-oiled crust. NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. FT: frying time (minute).

WB batter coating developed smoother crust surface in comparison to RB batter, and surface roughness (as represented by fractal dimension, FD) increased with frying time (Figure 4.9). Higher surface roughness of RB coated sample favored (by developing a strong bond between oil and surface matrix) the presence of higher fat on their surface. Rough-surface might favor higher grip-ability to retain its oil on samples surface, under post-fry surface de-oiling treatments. Smooth surface favored the slipping/removal of surface fat (due to week attachment between frying oil and solid matrix) during surface de-oiling treatments, and resulted in lower presence of fat on surface. Surface roughness, holes, crevices, and cracks in crust might have reduced the energy of applied hot air stream; that has resulted in lower effectivity of HAB treatment in removing fat. Longer frying (FT) favored the development of cracks/holes and these structures gave access the frying-oil, to go inside the crust, hence, increased in matrix and crust total fat were positively correlated with FT. In CAP treatment, fat from crust might came out through these openings (SO); that were in favor of its higher effectuality in fat reduction. With the increase of FT an increase in fractal dimension (FD) of surfaces were observed (Figure 4.9), that explains the lower-effectivity of post-fry surface de-oiling treatments for the sample that was fried for longer duration. Figure 4.10 depicts, correlation between FD and residual fat (i.e., fat which was present even after post-fry PAP treatment). Irrespective of batter formulations, residual fat has shown an increasing trend with the increase of FD.

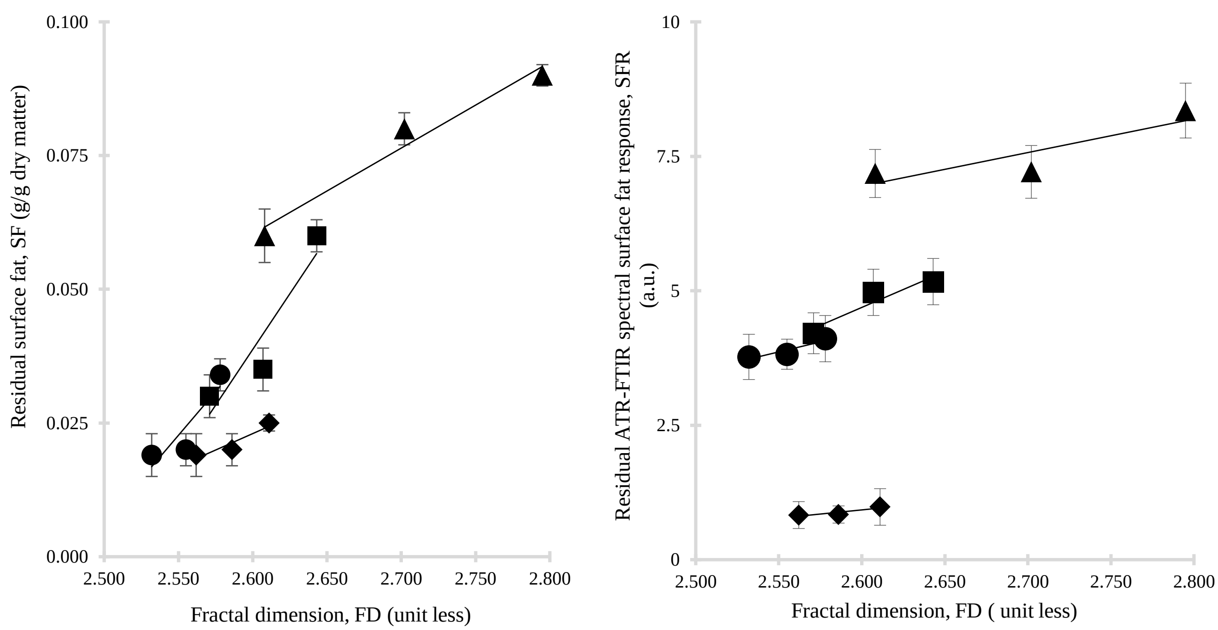


Figure 4.10: Plot of correlation between surface residual fat and surface fractal dimensions of post-fry PAP treated samples. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. PAP: pressure assisted absorbent paper.

4.5 Conclusion

A novel method (based on ATR-FTIR spectra) have been established to characterize the surface chemical nature of fried foods. The used ATR-FTIR spectra based method have shown consistency with the conventional method of assessing the surface attributes of batter coated fried foods. Surface structure-chemical characteristics, impacts the post-fry oil redistribution in batter coated fried foods. Batter-formulations and frying-time influences the surface microstructural-chemical nature and the effectivity of post-fry de-oiling treatments. Centrifuge-assisted paper absorbent (CAP) treatment have shown highest impact in reducing fat (surface, matrix, total) from batter coated fried foods, followed by pressure-assisted paper absorbent (PAP) and hot air blow (HAB) treatment. Findings of this study would provide scientific guidance to produce low-fat containing fried food product.

CONNECTING TEXT TO CHAPTER 5

Chapter 3 and Chapter 4 showed that microstructural traits of batter coated fried products are associated with mass-transfer process, and structural properties affect mass re-distribution. Chapter 5 established relationship between internal pore-structure and textural evolution of batter coated fried foods, based on X-ray µCT and Multifractal analysis of internal pore structures.

Parts of this chapter have been presented at scientific conference.

Md. Hafizur Rahman Bhuiyan and Michael O. Ngadi. (2022). X-ray Microtomography and Multifractal analysis of internal pore structures of fried coated food product. Presented at Canadian Society for Bioengineering (CSBE**)** Annual General Meeting. Held at Charlottetown, Prince Edward Island, Canada, from July 24-27, 2022. ID:1080**.** *Oral Presentation.*

CHAPTER 5

X-RAY µCT BASED MULTIFRACTAL ANALYSIS OF INTERNAL PORE STRUCTURE OF COATED FRIED FOODS

5.1 Abstract

Internal structure is crucial contributor to quality of fried food. This study aimed to characterize relationships between internal pore-structure and textural evolution in coated food products. Wheat and rice flour-based tempura-batters were used to coat a meat analog model food that was deep-fried at 180°C for 2, 4 and 6 minutes in canola oil. X-ray microtomography (µCT) and Multifractal analysis (MFA) were used to study internal pore-structure of fried products. Textural analysis was conducted by using a mechanical texture analyzer. Results of the study revealed that batter-coating influenced formation of internal microstructure during frying and also affected mass-redistribution leading to post-fry textural evolution. Batter-formulations and frying time (FT) impacted the evolution of microporosity (%) of coated food. Microporosity has positive correlation with fat content of coated fried product. Fried batter system having higher micropores provided hard, brittle, and crispy texture in coated food. Lower extent of post-fry moisture redistribution and higher textural stability are observed in porous, low moisture and high fat containing rice-flour based batter coated product. MFA outcomes i.e., Singularity spectra and Rényi spectra depicted that internal structure (pore distribution) of coated product is nonhomogeneous and possesses multifractal scaling behavior. Higher heterogeneity of pore distribution is observed in lower concentration of micropores at crust-region compared to core-region of coated product, and heterogeneity of pore distribution has shown slightly diminishing trend with increase of FT. Principal component analysis reveals varying extent of correlation between moisture-fat, textural attribute, microporosity and selected multifractal parameters ($\Delta\alpha$, $\Delta f\alpha$, R-L, D_1, D_2, ΔD).

5.2 Introduction

Fried food contains high amount of fat, which causes serious health issues. The use of batter-coating over food substrate, is an effective way of producing fried products having less oil. In addition to fat reduction, batter-coating develops a peculiar organoleptic characteristic (juicy inside & dry outside) in fried foods and textural attributes of coated-product is crucial for its consumer satisfaction (Hauzoukim et al., 2019; Rahimi *et al*., 2017; Adedeji, Liu, & Ngadi, 2011).

There is paucity of scientific information on post-fry mass-distribution and textural evolution in coated-product, which is very crucial to know for achieving consumer satisfaction by process-optimization and post-process quality maintenance. The element that determines fried food qualities exist at microlevel and microstructural properties are crucial in modeling mass-transfer process. Understanding of physicochemical properties at microscopic-scale would set foundation for the development of new and high-quality fried products as well as will assist in maintaining quality at post-frying stage. Microstructure that develops during frying defines the quality attributes of coated-products, and internal structure of foods are generally characterized by porosity (Adedeji et al., 2011; Adedeji & Ngadi, 2009). To get insight on physical properties (e.g., textural attributes) which is associated with mass-transfer event, the characterization of internal pore is of crucial importance for understanding the changes in textural attributes at post-frying stage. However, spatial distribution, local densities, lack of homogeneity in pore structure could contribute differently to their overall functionalities. These factors need to be considered in obtaining detailed and realistic information on internal pore characteristics of food, in relation to any specific aspects of quality attributes.

Advanced microscopic techniques (SEM, CLSM) have been used in obtaining microstructural information of fried-batters at surface/low depth. However, 2D microscopic-image has limitations in providing sufficient information to engineering aspects of food processing, as foods are three-dimensional (3D) object. In this context, X-ray microtomography (μCT) is a niche choice to obtained three-dimensional details of internal structure of batter-coated products. It is notable, X-ray μCT has the capability of components differentiation based on density difference without physical slicing, staining, or destruction of test specimen (Wang Z. et al., 2018; Adedeji & Ngadi, 2009). Adedeji & Ngadi (2009) investigated potential application of X-ray μCT for microstructural analysis of breaded-chicken nuggets, where centre of attention was the mean geometrical description of pores. However, a single average geometric measurement from digital images is not sufficient to characterize the heterogeneity of pore distribution in whole matrix (Zhang J. et al., 2020; Jiang et al., 2018; Mendoza et al., 2010). Study on statistical characterization of internal micropores (obtained through 3D X-ray μCT images) of batter-coated products in relation to post-fry mass and textural evolution, are still lacking.

Internal pore structure of porous media are generally very complex, and complex structures can't be effectively modeled by traditional Euclidean geometry (Zhang M. et al., 2021; Jiang et al., 2018). In these context, Multifractal approach could be used in analyzing and classifying of digital images having complex structure (Ampilova et al., 2019; Jung & Yoon, 2017). Multifractal analysis (MFA) have been used to study spatial distribution of pores in solid matrix which is linked to diffusion of water (Lafond et al., 2012; Mendoza et al., 2010). Recently, MFA have been used to characterize crystallographic image of milk (Ampilova et al., 2019) and rupture patterns of dried Laver (Jung & Yoon, 2017). Use of MFA in studying the complex internal pore microstructure of batter-coated fried product is a novel scope of research, as till now, there is no scientific literature in this aspect. It is hypothesized that 3D X-ray µCT image-based MFA of internal-pore structure of batter-coated foods would provide a better understanding on mass and textural evolution, at microscopic level. The overall objectives of this work were to (i) study the impact of batter formulation and frying time on the development and evolution of crust microstructure as described by microporosity obtained from high resolution X-ray µCT scanning images (ii) characterize spatial pore distribution in batter coated fried foods using multifractal analysis (iii) assess the relationship between multifractal parameters, moisture-fat distribution and textural quality changes in batter coated fried food.

5.3 Materials and Methods

5.3.1 Materials

Soy protein isolate and wheat-gluten was purchased from MP Biomedicals (29525 Fountain Pkwy, OH, USA) and Sigma-Aldrich Co. (Ontario, Canada), respectively. Sodium pyrophosphate ($Na_4P_2O_7$) crystals and Methylcellulose (MC) were received from Fisher Scientific (Fair Lawn, New Jersey, USA). Sodium bicarbonate ($NaHCO_3$) was received from Church & Dwight Canada Corp. (Ontario, Canada). Commercial rice flour (Suraj®, composed of 83.32% carbohydrate, 1.33% fat, 6.67% protein and 8.68% moisture) was purchased from a grocery store in Montreal, Canada. Wheat flour (Five Roses®, composed of 73.33% carbohydrate, 13.33% protein, 3.33% fiber, 1.33% fat and 8.68% moisture), NaCl (Sifto, Compass Minerals Canada Corp.) and Canola oil (Sans nom®, Loblaws Inc.) were procured from a grocery store in Sainte-Anne-de-Bellevue, Canada.

5.3.2 Sample preparation

Meat analog model food was formulated with the soy protein isolate (SPI), wheat gluten (WG), canola oil (CO) and distilled water (DW). Firstly, wheat-gluten was mixed with canola oil, and then soya protein isolate was added and mixed thoroughly. Then distilled water was added and mixed until a very soft-textured dough was developed. The respective proportion (g) of the ingredients WG: CO: SPI: DW were 12:15:22:73. Prepared dough was used to fill individual rectangular-shaped cavities of silicon mold (Freshware CB-115RD 24-Cavity Silicone Mini Mold). Covered silicon mold was held in complete horizontal position (surrounded by water, but not immersed) on top of a flat metal wire surface and heated at 70°C for 15 min in a water bath. After heat treatment, mold was allowed to reach in equilibrium with room temperature. After removing the top aluminum cover, gentle pressure (by finger) was applied from the back of each cavity to bring out the formed substrates. This substrate was used as model food, weighing an average weight of 16±0.2 g/substrate and having a uniform dimension of 5.6 cm x 2.5 cm x 1.3 cm. Prepared model-food was packed in zip-lock plastic bags and stored in refrigerator (4±2°C) for 24 hours, before its further use.

Tempura batter coatings were used to coat the formulated model food. To prepare batter coatings: white crystal of Sodium pyrophosphate was crushed and grinded, to obtain as fine white powder. Then sodium bicarbonate (1.4%), methylcellulose (0.3%) and NaCl (1.5%) were added and mixed properly with sodium pyrophosphate powder (1.8%), to form a homogeneous powder mix. The preform powder-mixture was further thoroughly mixed with dry flour in a proportion of 5% (powder mix) to 95% (flour). Distilled water was added to the solid ingredients with a total solid to water ratio of 1:1.3 and mixed properly until a homogenous batter slurry was obtained. Wheat based batters (WB) were formulated using only wheat flour whereas rice based batters (RB) were formulated with rice flour. Some samples were formulated using equal mixture of wheat and rice based batters (RWB). Formulated batter systems were kept for 5 min at room environment and then used to coat the model food. Prior to batter coating, model food was brought out of refrigerator and kept at room environment for 30 min. Food was fully immersed in batter system for 1 minute and then removed from batter solution to make a complete coat. Coated products were held on a kitchen-fork for 30sec to drain the excess amount, in order to obtain a uniform layer of coating around model food substrate. Non-coated (NC) model food was used as control.

A programmable deep fat fryer (T-fal Compact, FF122851, China) was used to fry the samples at atmospheric condition. Fryer was filled with 1.5 L of fresh canola oil and preheated for 1 hr at a set temperature of 180°C. Each time three sample was transferred into the fryer and uninterruptedly fried for 2, 4 or 6 mins maintaining a sample to frying-oil ratio of 1:30. At the end of frying, the frying-basket was removed from the oil and was shaken for 5 times. To minimize variation in frying-oil properties each batch of oil was used for frying only seven batches of samples (i.e., equivalent to around 30 min of frying) before the used frying-oil was replaced with a new batch of fresh oil.

5.3.3 X-ray μCT and image processing

A portion (20×20×5±1 mm) of the fried samples were procured by microtome blade (Feather, C35 type, Japan). The cut portions were lyophilized in a freeze-drier (Modulyod-115; Thermo Savant, Holbrook, USA) at -50°C and 250 mbar for 48 hours. After that, samples were scanned in a X-ray μCT scanner (SkyScan 1174, Bruker-Skyscan, Kontich, Belgium) with a preset voltage of 50 keV (no filter), current of 800 uA, exposure time of 2400 ms, and angular rotation step of 1.0^0 through 180^0. X-ray images were recorded as 16 bits Tagged Image File Format (TIFF files) and a cross-section pixel size of 8.9 μm was realized. X-ray images were reconstructed into a series of 2D images (1292*1292 pixels) by using a reconstruction software (NRecon, Version: 1.7.4.2, Skyscan, Belgium), that operates based on modified filtered back-projection algorithm. Reconstructed images were saved as 16 bits TIFF files.

The stack of 2D reconstructed images were loaded in CTAn software (Skyscan, Belgium) to visually identify a few top slices (of coating part) that need to be discarded in order to obtain an even crust region (Adedeji et al., 2011). Starting from preidentified image slice, a total of 128 consequential images were selected for further processing. A 128 image slices were chosen as it equals to the power of 2 (i.e., 2^7) which fulfills the fundamental requirement of fractal analysis; hence, no further pre-processing of data was required for subsequent multifractal analysis (Han et al., 2020; Rahimi & Ngadi, 2016). Selected images were imported as image sequences in Java-based public domain image processing software (Image J 1.52a, National Institutes of Health, USA). Imported image stack was converted to 8 bits grayscale images and the scale runs between 0 and 255. Smoothing (Gaussian filter) of grayscale images were performed to remove noise. From middle part of the images, a square shape region (512*512 pixels) was cropped and subsequently used for further analysis as region of interest (ROI).

In this study, the structure of interest was the internal void space (i.e., pore). To minimize the overestimation/underestimation of internal pores, Otsu algorithm was applied “Slice-by-Slice” in automatically finding a reasonable threshold value that delineates between the pores and the solid-matrix of each individual images of the image-stack (San et al., 2010). To remove any further noise from binarized images a morphological operation i.e., Despeckling (median filter) was applied cautiously; as too much treatment of image influences estimation of the structure of interest (Lafond et al., 2012). Microporosity (i.e., space occupied by void structures to the total space of interest) was estimated from the binarized images. Microporosity (fraction of void space) of each individual slices of the image-stack were estimated and a final 1D frequency curve (consists of 128 equally spaced data points) of estimated spatial microporosity were developed, that were used as input for subsequent multifractal analysis (Han et al., 2020; Lafond et al., 2012). To estimate mean volumetric microporosity (i.e., volume occupied by void structures to the volume of interest) the porosity information of each individual slice was used to calculate the total pore volume [{Σ (porosity of each slice*area of ROI) / Number of slice} * Length of image stack] relative to the total volume of interest (area of ROI* Length of image stack).

5.3.4 Multifractal analysis

A graphical-user-interface based multifractal analysis software namely MFA (developed by Liwen Han & Pierre Dutilleul, McGill University, Canada) is used in this study. MFA program was written and run in MATLAB platform (R2015a ver. 8.5, The Math-Works, USA). MFA software performed multifractal analysis on the basis of Singularity spectrum [estimated fractal dimensions f(α) for different values of estimated singularity exponents (α)] and Rényi spectrum [generalized dimensions (Dq) for different values of moment order (q)] from a curve with equally spaced data points (Han *et al*., 2020; Lafond et al., 2012). Taking the volumetric nature of data into account, 1D frequency curve of estimated microporosity (obtained from 3D X-ray µCT images) were used as input data to the MFA software (San et al., 2010; Lafond et al., 2012; Han *et al*., 2020). Multifractal parameters of singularity strength [α(q)] and the associated Hausdorff dimension [f(αq)] and of generalized dimensions (Dq) were estimated for moment orders (q) between -8 and +8 in increments of 1. Both figure and data table of Singularity spectra and Rényi spectra were used for subsequent data analysis and presentation of multifractal analysis results wherein only the multifractal parameter estimates for which the corresponding R^2 value was ≥ 0.9, were considered. In describing the information about the variability of the studied variable (internal pores) and the heterogeneity of distribution, different multifractal estimates such as α_{min}, α_{max}, $\Delta\alpha$, $f\alpha_{min}$, $f\alpha_{max}$, $\Delta f\alpha$, D_1/D_2, ΔD etc. were considered.

5.3.5 Mass and texture profile

Textural attributes of fried samples were evaluated by puncture/compression test, using a mechanical texture analyzer (Stable Micro Systems Texture Analyzer, TA. HD PlusC, UK). Fried samples were individually mounted on a flat rigid support. A puncture probe TA-52 (2mm dia.) and 500 N was used to punch the test specimen, at a constant test speed of 1mm/s and a travel distance of 5 mm. Puncture test were performed on three equidistance locations of each of the two longest-facets surface (5.6 cm x 2.5 cm) of the samples. Maximum force to break (N), displacement at maximum force (mm), and slope (N/mm) at maximum force of the puncture test were used to evaluate crust textural properties namely hardness, brittleness, and crispness, respectively (Rahimi *et al*., 2017). Puncture test data were analyzed with the texture analyzer Exponent ver. 6.1.14 software.

Moisture content were grouped into three categories, namely crust moisture (CrM), core moisture (CoM), and total moisture (TM). CrM was defined as the moisture present in crust-region, CoM represents the moisture present in core-region, and TM includes both crust and core moisture of fried samples. A microtome blade (Feather, C35 type, Japan) was used to detach crust-region from the core-region of the fried samples (i.e., remaining portion after the procuration for X-ray CT experiment). And the sample portion (used for nondestructive X-ray CT experiment) which was freeze-dried without separating the core from the crust were used to measure total moisture (TM). All samples were freeze dried in a freeze-dryer (Modulyod-115; Thermo Savant, Holbrook, NY, USA) at -50°C and 250 mbar for 48 hr. and then transferred into a desiccator to equilibrate for 30 minute. Weight of the sample before and after freeze-drying was measured and moisture content was calculated on dry weight basis (g/g dry matter).

Fat content was grouped in three categories, namely crust fat (CrF), core fat (CoF), and total fat (TF). CrF was defined as the fat present in crust, CoF represents the fat present in core, and TF includes both crust and core fat content. To determine fat content, freeze-dried samples were grounded to increase surface-to-volume ratio and grounded sample (3-5g) was placed in thimbles of a VELP SER 148 solvent extraction unit (Velp Scientifica, Usmate, Italy). Fat was extracted by following the Randall method, with petroleum ether at 130°C pre-set temperature. Complete fat extraction process was performed in three consequent steps: immersion (30 min), washing (30 min), and recovery (30 min), totaling of 90 minutes. Weight of extracted fat was measured and fat content was computed on dry weight basis (g/g dry matter), by dividing the mass of extracted oil with the mass of freeze-dried sample.

5.3.6 Experimental design and statistical analysis

A full factorial design was used where the factors were batter formulations and frying time. All experimental data obtained from triplicate of samples, mean ± standard deviation was reported. Tukey's honestly significant difference (HSD) is used in reporting significant ($p<0.05$) difference among mean values of estimates of destructive attributes. Principal component analysis (PCA) and all statistical analysis was performed by statistical software (JMP 14.1v, SAS Institute Inc., Cary, NC, USA).

5.4 Results and Discussion

Figure 5.1 depicts, X-ray µCT grayscale images and their corresponding binary images. Bright and dark spots in greyscale image, respectively represent denser components (e.g., carbohydrate, protein) and pores in fried samples (Adedeji & Ngadi, 2009). Processed binary image depicts the efficiency of Otsu segmentation algorithm in distinguishing internal micropores (dark) and solid matrix (white). Otsu based segmentation had reported higher threshold values (in scale of 0-255) for the images of crust-region, compared to the images of core-region. The differences in threshold values signifies, the density of inner and outer region of studied fried samples were different (Adedeji & Ngadi, 2009). It is visually noticeable, crust microstructure of fried samples (of different batter) possesses distinctive characteristics, and there is microstructural difference between crust and core region of the same sample. Difference between inner and outer microstructure of same sample (apple fruit) has been reported in literature (Mendoza et al., 2010).

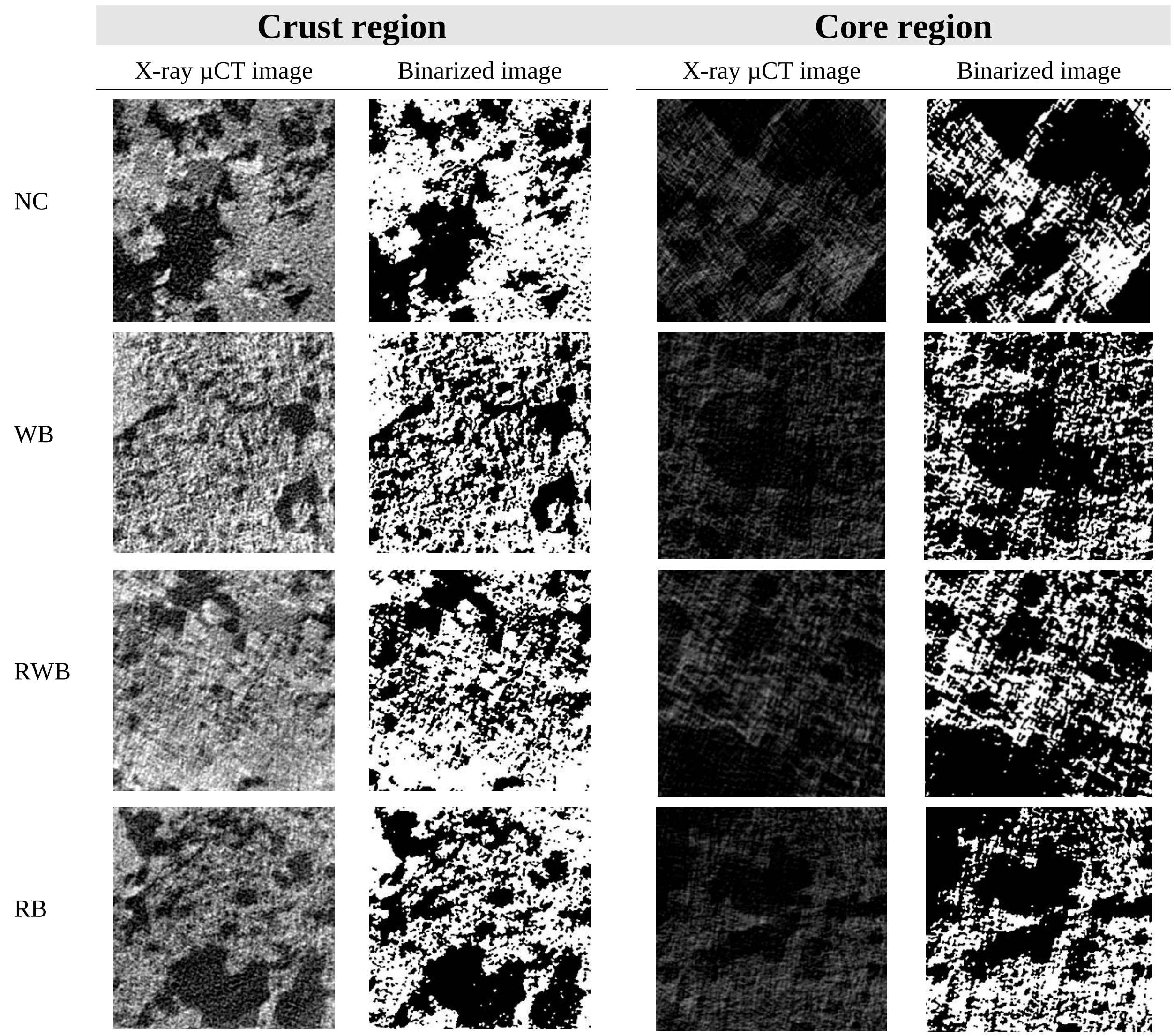


Figure 5.1: Representative X-ray µCT greyscale image and corresponding binary image of crust (left) and core (right) region of fried samples. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter, respectively. NC represents noncoated model food.

Figure 5.2 depicts, frequency curve (consist of 128 equally spaced data points) of estimated spatial microporosity of fried samples. Porosity has increased with increment of the order of slice position. Changes in microporosity were apparently higher at lower order of the slice position, while a relatively constant nature were observed at higher order. Irrespective of batter-formulations and frying time, all samples have shown this mentioned pattern. These indicates, development of internal pores in fried product is a spatially subjugated event and higher variability in pore structure is most probable in outer crust-region. Depth dependency of porosity within sample (apple, soil column) has been reported in literature (Mendoza et al., 2010; San et al., 2010). However, first thirty two slices (within the region of lower order of slice position) of X-ray µCT image stack were chosen to represent crust-region of fried samples and remaining were representing the core-region of the same sample. Under this consideration, higher porosity were observed for core-region of the samples, in comparison to their crust region. Higher porosity of core region could be attributed to higher (1.49 > 1.3 g/g dry matter) initial moisture content of the model-food substrate, in comparison to batter-coatings; as formation of pores are the resultant effects of the loss of moisture (as vapor) under heat treatment (Rahimi & Ngadi, 2015; Adedeji & Ngadi, 2011).

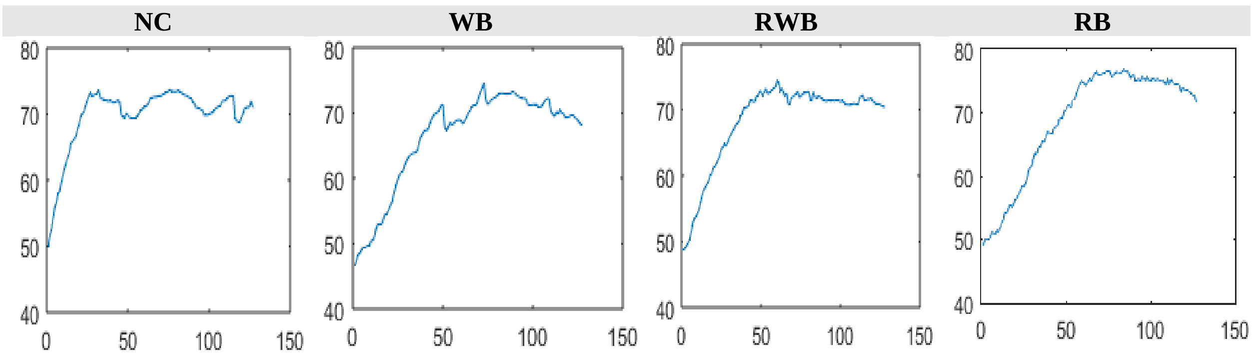


Figure 5.2: Representative frequency curve of spatial microporosity, obtained from stack of binarized X-ray µCT images. Vertical and horizontal axis represents microporosity (%) and position of image slice, respectively. Ordering of slice position starts (0) from outer crust-region and ends up (128) in inner core-region. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter system, respectively. NC represents noncoated model food.

Figure 5.3 depicts, evolution of microporosity as function of frying time (FT). X-ray µCT image based crust microporosity (CrP) of the batter-systems were ranged between 42–61% for the frying duration of 2 to 6 minutes. Non-destructive X-ray µCT image based porosity results of this study are in close similarity with the relevant literature (Voong et al., 2018; Adedeji & Ngadi, 2011; Adedeji & Ngadi, 2010), where the pore characterization techniques were mercury intrusion porosimeter (MIP) and helium pycnometer (HP). This assures, X-ray µCT image-based statistical characterization of internal pore-structure would provide realistic and acceptable information in understanding the quality aspects of batter-coated fried products, at micron level. Crust microporosity increased with the increase of FT. However, a faster increase in microporosity were observed at early stage of frying and after a certain period (4 min), the increase of microporosity had been slow down. The reduced rate of pore formation in batter-coated food samples might be due to lack of sufficient water in sample, as moisture leaves food during frying in the form of vapor and results the formation of internal pores. However, for an identical duration of frying, rice-flour based batter (RB) has developed more porous crust (higher CrP) than wheat-flour based batter (WB). Average core microporosity (CoP) values ranged between 51–67% for the frying duration of 2 to 6 minute. For identical duration of frying, CoP of non-coated sample was considerably higher than batter-coated samples; this gives an impression of lower extent of moisture loss from model food-core substrate while it was coated with batter-systems. In comparison to WB batter, RB batter had favored higher pore formation in core substrate, that display the lower capabilities of preventing moisture-loss from core-region of rice-flour based batter coated food samples. For identical duration of frying, core microporosity of studied samples were comparatively higher than their corresponding CrP. Like CrP, the CoP increased with frying time. As a resultant effect of the increase of crust and core microporosity, total porosity (TP) of coated samples increased with frying time. This indicates positive correlation between spatial microporosity in batter-coated fried samples. Higher TP of the RB batter coated samples could be attributed to the fact that the initial total moisture content (before frying) of rice-flour based batter coated model food sample was higher compared to other batter formulations and could be explained as such: for identical solid-to-water ratio of prepared batter systems, batter-pickup (%) of rice-flour based batter is generally lower (due to lower adhesiveness) than wheat-flour based batter (Adedeji & Ngadi, 2011). And initial moisture content of model food core substrate was (1.49 g/g dry matter) higher than the moisture content (1.3 g/g dry matter) of the formulated batter systems. Therefore, lower batter-pickup (BP) of rice-flour based batter was in favor of higher initial moisture content of RB coated model food sample, and higher availability of moisture had favored higher extent of moisture loss during frying which results in the formation of pores in fried sample.

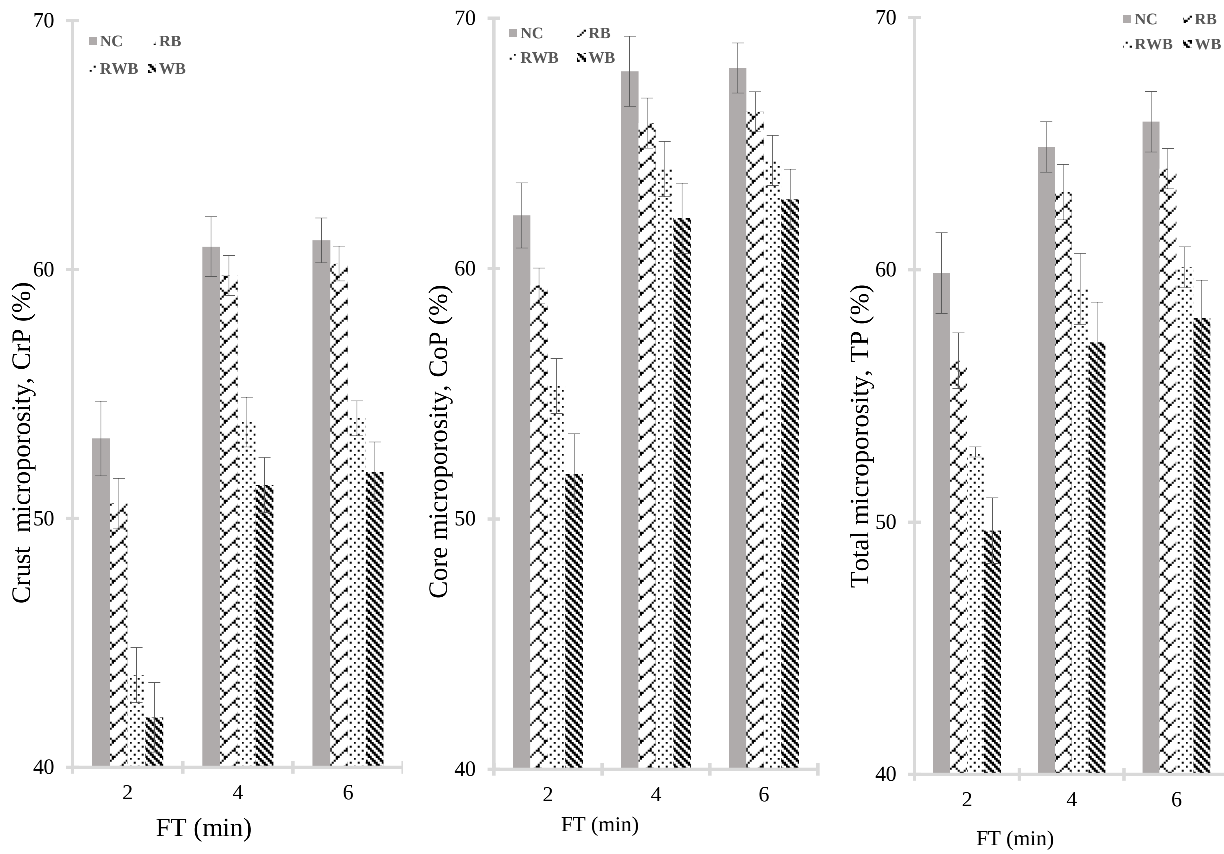


Figure 5.3: Microporosity as function of frying time. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter system, respectively. NC represents noncoated model food. FT: frying time.

The discussed microporosity (CrP, CoP, TP) information are the average value of the estimations. However, a single mean value is not sufficient to describe the heterogeneity of pore distribution in whole matrix of the studied samples (Zhang J. et al., 2020; Jiang et al., 2018; Mendoza et al., 2010). Hence, application of Multifractal analysis (MFA) were attempted in subsequent section. Motivation behind the use of MFA stands on the origin of its mathematical formulas, as this approach can characterize hidden order in apparently random signals of distributions coming from chaotic deterministic systems (Mendoza et al., 2010).

In performing the multifractal analysis (MFA), pore distribution was considered for inward direction as fat uptake during frying occurs through this direction i.e., from surface to core. Figure 5.4 depicts, Singularity spectra [$f\alpha$ against α] of the multifractal analysis. Shape of Singularity spectra depicts, internal pore structure of fried products is multifractal (i.e., multiscale structure); because a wide range of estimated fractal dimensions with a parabolic shaped concave graph have been observed instead of a one-point singularity spectrum concentrated at α_0, $f\alpha_0$ (San et al., 2010;

Mendoza et al., 2010). Studied battered samples has shown typical humped shape Singularity spectra that reveals, pore distribution of coated-products has multifractal scaling behavior. In this scenario, the curvature and symmetry of Singularity spectra could provide information on several aspects such as description of heterogeneity in pore distribution, concentration of multifractal measures, etc.

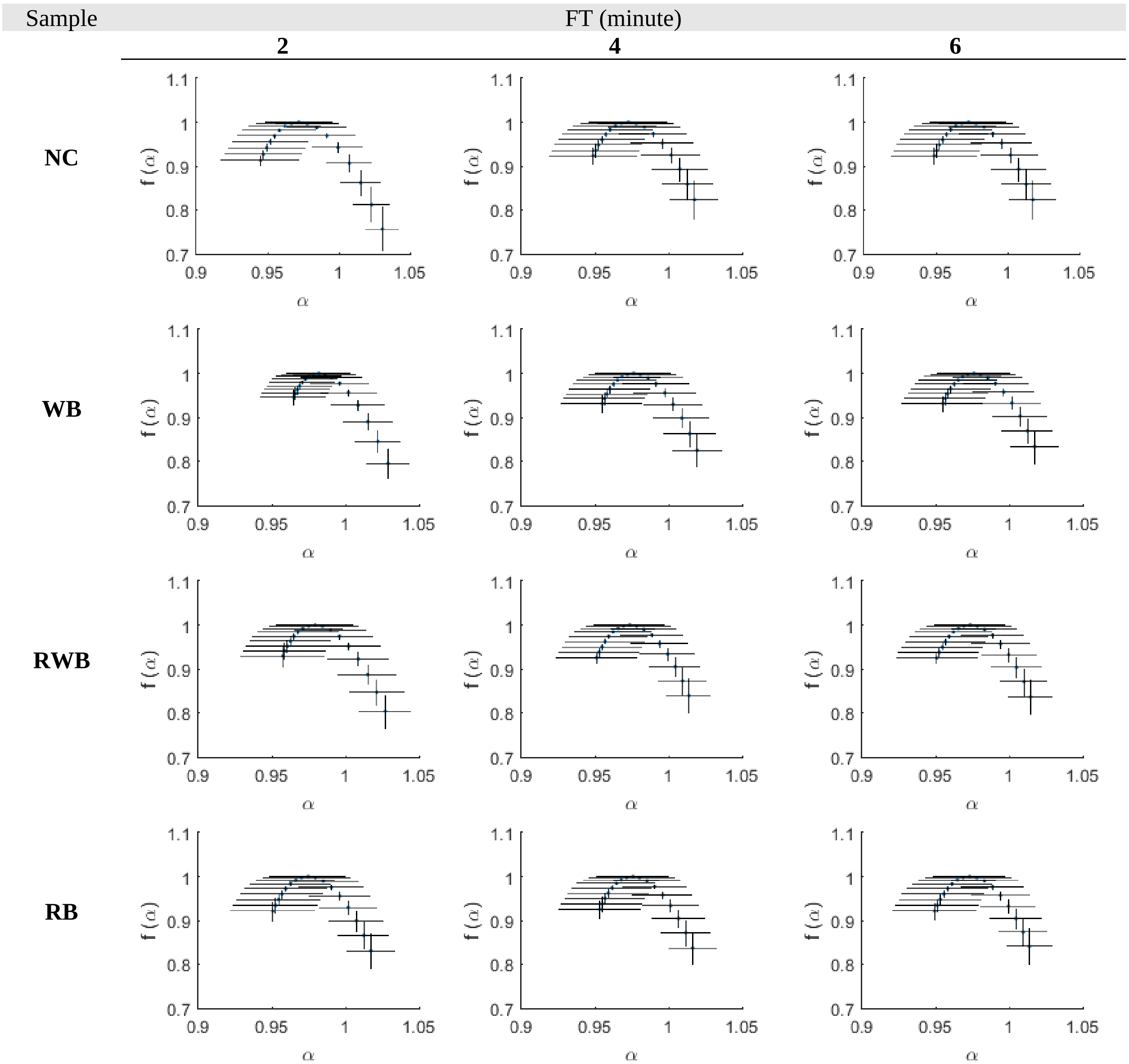


Figure 5.4: Singularity spectra in the view of common scale. Points and bars represent mean values and standard errors in the estimation of α (horizontal) and fα (vertical), respectively. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter, respectively. NC represents noncoated model food. FT: frying time.

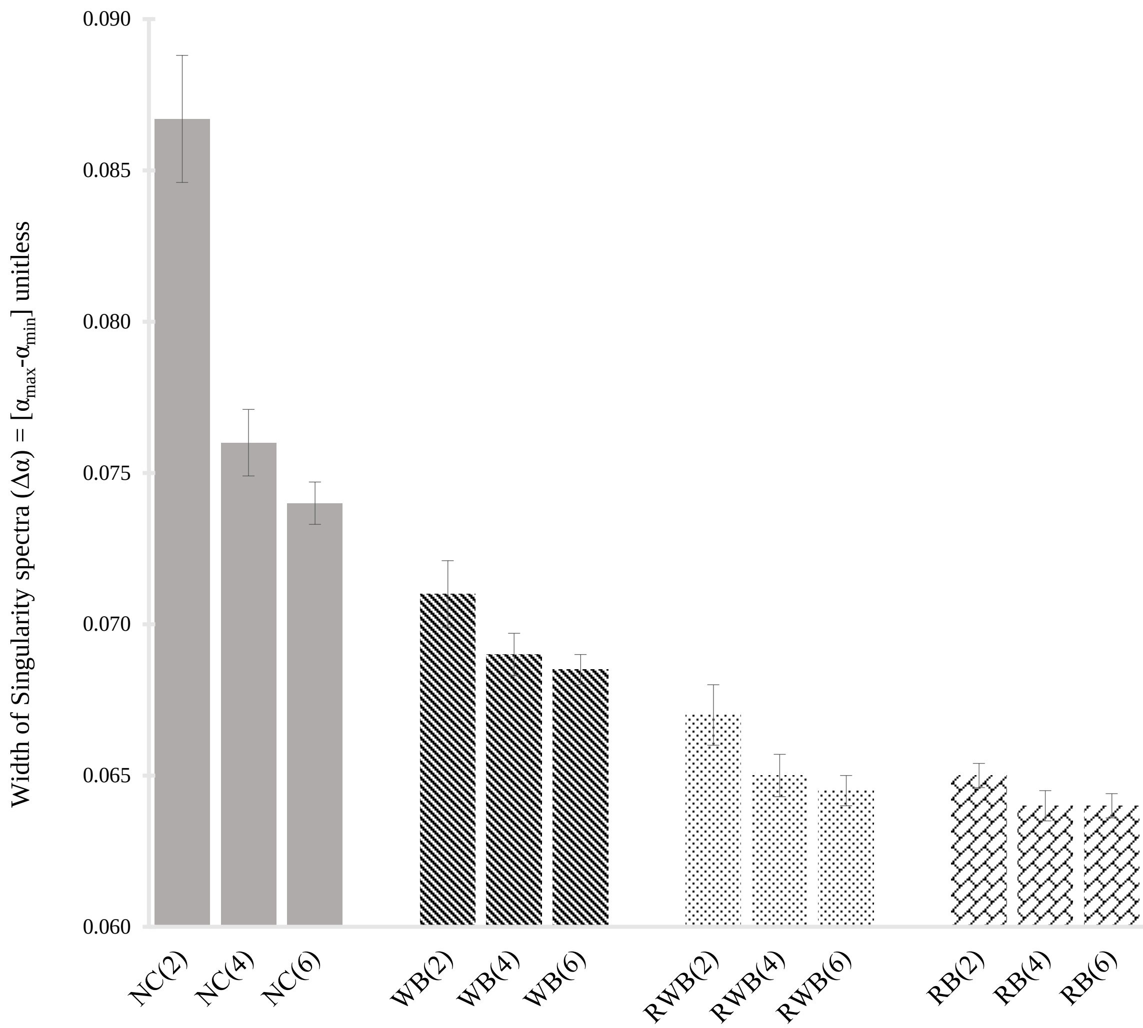
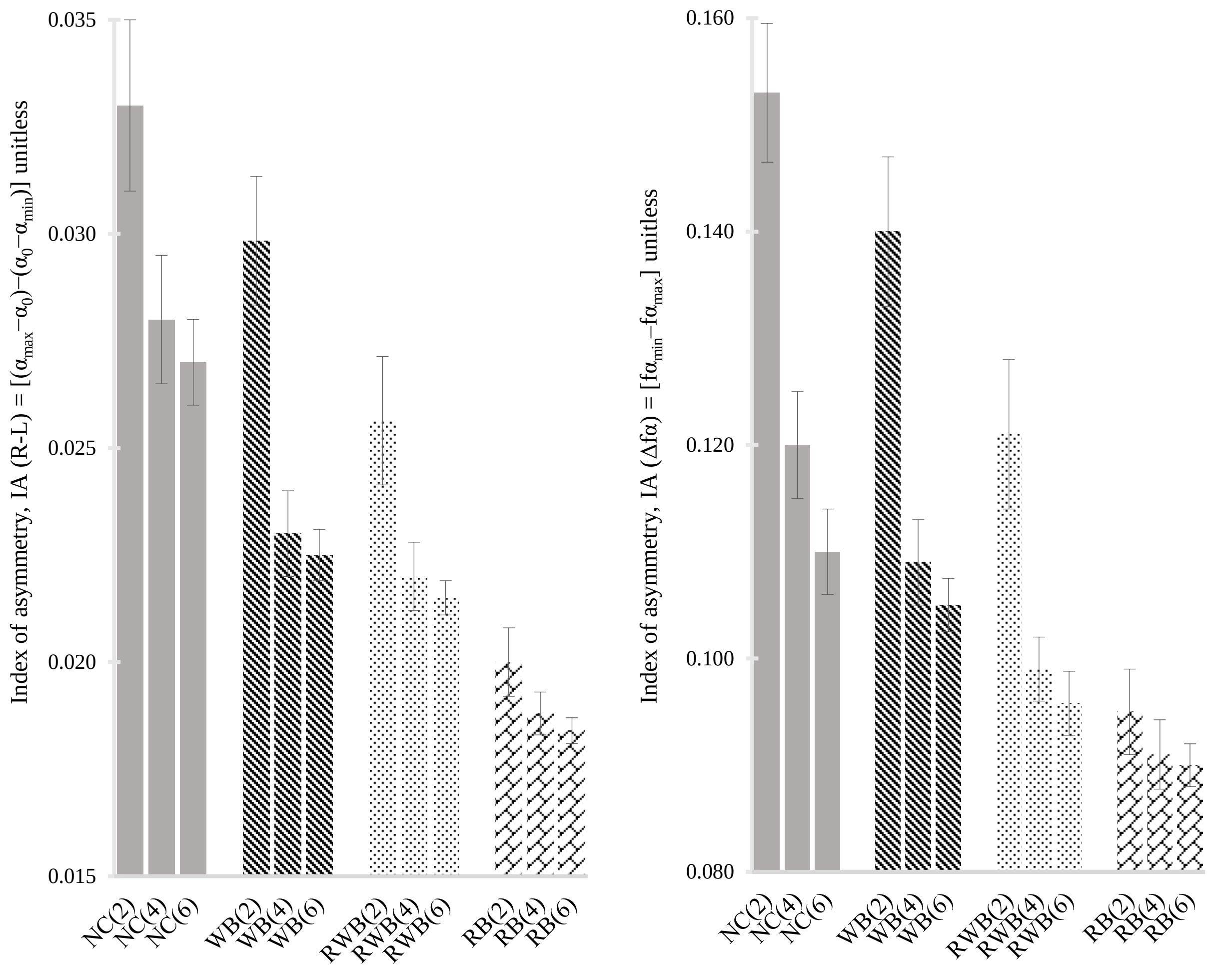


Figure 5.5: Width of Singularity spectra ($\Delta\alpha$) as measured by the difference between α_{max} and α_{min}. Number in parenthesis indicates FT (minute). WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter, respectively. NC represents noncoated model food. FT: frying time.

The fully developed Singularity spectra (having both left & right branch) indicates, degree of heterogeneity of pore distribution in fried coated samples can be reliably determined by width of Singularity spectrum. For estimated fractal dimensions $f(\alpha)$, width of singularity spectrum [α_{max}–α_{min}] provides information about scaling diversity of pore distribution (Wang Y. et al., 2022; Zhao et al., 2021; San et al. 2010). Figure 5.5 depicts, width of Singularity spectra ($\Delta\alpha$) as measured by the difference between α_{max} and $\alpha_{min.}$ The widest (highest value of $\Delta\alpha$) singularity spectra were observed for NC sample, indicating that the scaling diversity (number of fractal dimensions explaining the heterogeneity of micropores distribution) of non-coated fried sample was greater in comparison to batter-coated samples. The wider Singularity spectra represent higher heterogeneity of the pore distribution at all observation scales (Wang Y. et al., 2022; Zhao et al., 2021); and the heterogeneity of distribution implies that regions with high and low concentrations of pores scale differently (Mendoza et al., 2009). Heterogenicity of pore distribution in fried samples were impacted by batter-formulations, where a wider $\Delta\alpha$ were observed for wheat-flour based batter (WB) coated sample against rice-flour based batter (RB). The $\Delta\alpha$ have shown a slight decreasing trend with the increase of FT. This indicates, the scaling diversity of pore distribution decreased with the duration of frying. In physical sense, internal pore distribution of fried samples were impacted by the duration of frying and longer frying favors the formation of comparatively less-heterogenous pore structure; as homogeneous system exhibits a narrow singularity spectrum (Srocke, 2019; Lafond et al., 2012; Mendoza et al., 2009).

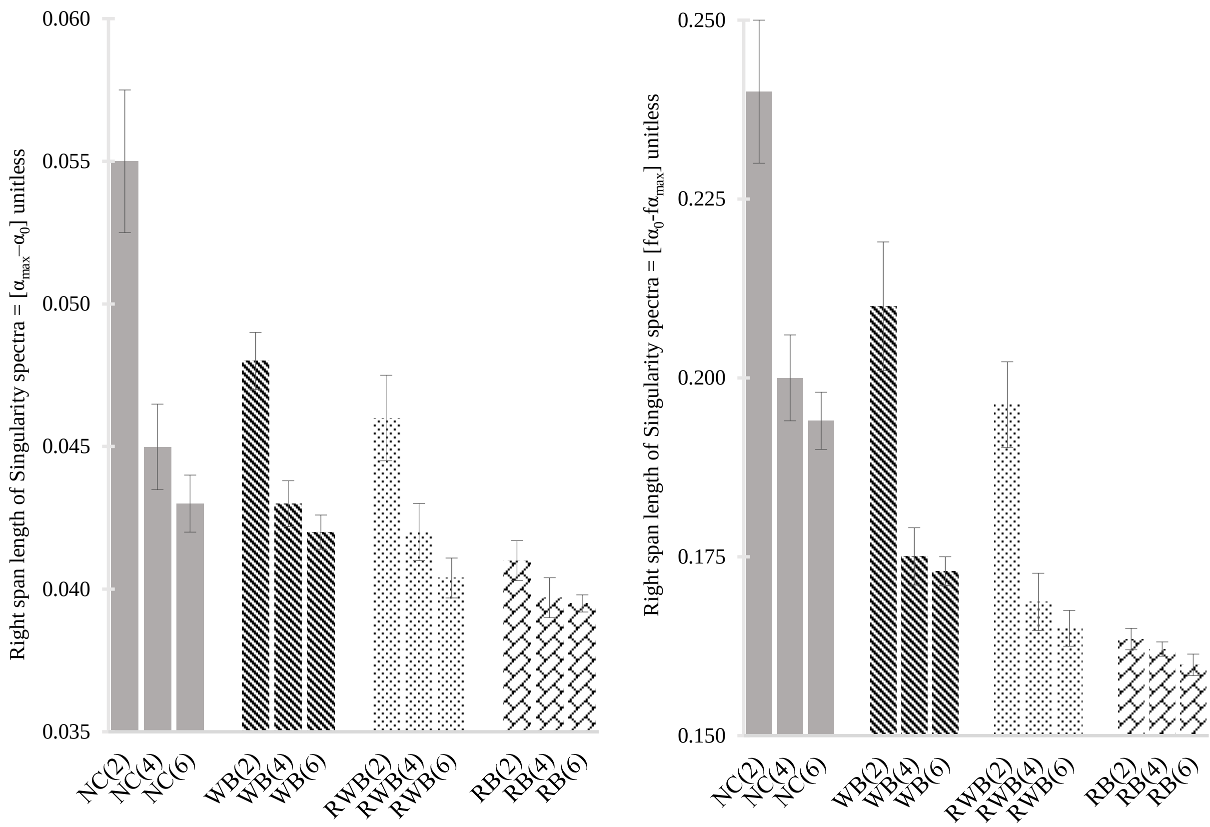
The symmetry of Singularity spectrum is useful in reviewing the concentrations/densities of the multifractal measure (Lafond et al., 2012; San et al., 2010). In this aspect, two indicators for assessing the asymmetry of Singularity spectra are proposed in relevant studies as such: (i) measures the symmetry in the horizontal axis of the range $[\alpha_{min}, \alpha_{max}]$ with respect to α_0 by the difference [R-L] between right ($R=\alpha_{max}-\alpha_0$) and left ($L=\alpha_0-\alpha_{min}$), following a vertical line crossing the Singularity spectrum at maximum spectrum value at α_0, $f\alpha_0$ (ii) measures vertical difference $[\Delta f\alpha=(f\alpha_{min}-f\alpha_{max})]$ between the two branches of the spectrum, where a negative or positive values mean a longer left or right component of the Singularity spectrum, respectively. Estimates of the mentioned two indicators (R-L and Δf) are represented in Figure 5.6. The non-zero and positive values of the Index of asymmetry (IA) confirms, all the samples had shown asymmetric Singularity spectrum with a longer right branch. Right branch of the Singularity spectra was wider ($R-L>0$) and longer ($f\alpha_{min}-f\alpha_{max}>0$) which manifests, the larger concentrations (smaller α-estimates) are less diverse and more common than smaller concentrations (larger α-estimates) (Wang Y. et al., 2022; Lafond et al., 2012; San et al., 2010). It was observed (Figure 5.3) that pores concentration in outer-crust region was comparatively less than the inner core-region of the coated fried samples. Therefore, outcomes of the index of asymmetry (IA) analysis could be realized as such: in batter-coated fried samples, heterogeneity of internal pore distribution at crust region were higher than that of core region. It has been reported in literature, fat distribution in fried chicken nuggets had varied with depth (Adedeji et al., 2011); and heterogeneity of pore size distribution of apple tissue near skin were higher than that of core region (Mendoza et al., 2010). These are in similarity with the observation of X-ray μCT images-based MFA of internal pore structures of coated fried samples of this study. The lower IA value represents, pore distribution of RB batter coated samples were less heterogenous (i.e., smaller densities of multifractal measures are diverse as larger densities) compared to WB batter coated samples. Frying time dependent evolution of IA was also lower for RB coated samples against WB. This alludes to the phenomenon that rice-flour based batter coated food develops an stable internal pore structure at early stage of frying which can withstand the impact of FT, in maintaining their initial distribution profile. This behavior of RB batters could be attributed to lower water retention ability of rice-flour based batter system, which favors easy moisture removal during frying. However, IA value of studied samples has shown a decreasing trend with FT, indicating that heterogeneity in pore distribution between crust and core region reduces to a more homogenous distribution while the samples were fried for a longer duration.



A B

Figure 5.6: Index of asymmetry (IA) of Singularity spectra as measured by A: [{R=(α_{max}-α_0)}-{L=(α_0-α_{min})}] and B: [$f\alpha_{min}$– $f\alpha_{max}$]. Number in parenthesis indicates FT (minute). WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter, respectively. NC represents noncoated model food. FT: frying time.

Multifractal parameters (α_{min} & $f\alpha_{min}$) and (α_{max} & $f\alpha_{max}$) respectively reflects the greatest and lowest density of pore distribution in solid matrix (Zhao et al., 2021; Lafond et al., 2012; San et al., 2010). Considering these as the basis and knowing (Figure 5.2 & 5.3) that pore concentration (%) in crust region were comparatively lower than core region of coated fried samples, the impact of frying duration on pore distribution in batter-systems were evaluated by determining length of the right span of Singularity spectra as such: (A) measures the difference between α_{max} and α_0 value of singularity spectra (B) measure the difference between $f\alpha_0$-$f\alpha_{max}$ value of the singularity spectra. The resulting outcomes are shown in Figure 5.7. Longer right span (higher value) indicates higher heterogeneity and shorter span length (lower value) represents less heterogenous pore distribution in crust region. With the increase of FT, crust of identical batter-coated samples became more homogenous. However, the decrease of right span length with the increase of FT favors the resultant decrease in IA value of Singularity spectra; while the length of left span was not so intensely affected by frying time.



A **B**

Figure 5.7: Right span length of Singularity spectra as measured by A: [α_{max}–α_0] and B: [$f\alpha_0$-$f\alpha_{max}$]. Number in parenthesis indicates FT (minute). WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter, respectively. NC represents non-coated model food. FT: frying time.

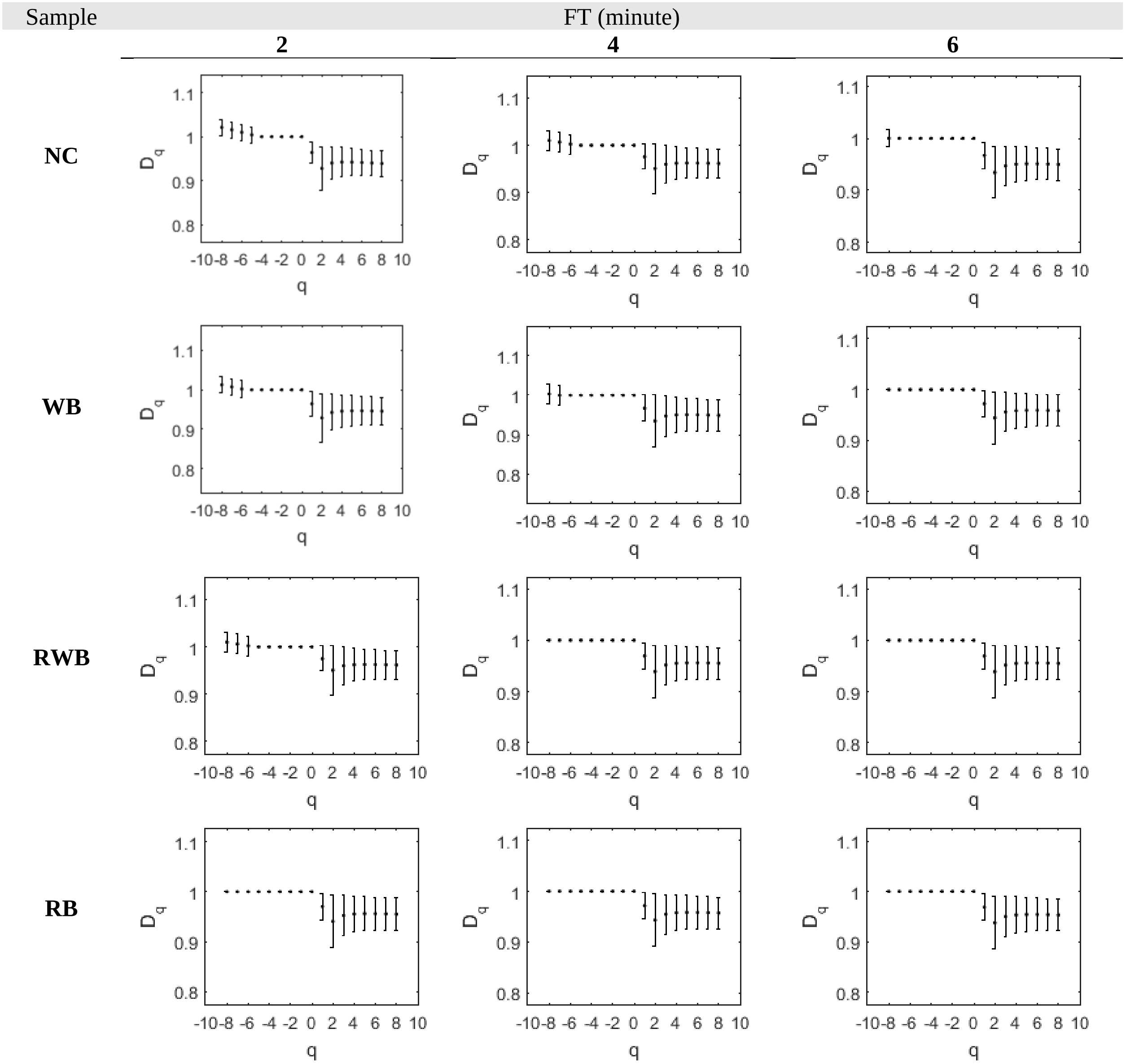


Figure 5.8: Rényi spectra in the view of common scale. Points and bars represent mean values and standard errors of Dq estimates. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter, respectively. NC represents noncoated model food. FT: frying time.

Figure 5.8 depicts, Rényi spectra [Dq against q] of multifractal analysis. Shape of Rényi spectra were non-horizontal with a clear asymmetry with respect to the vertical axis at D_0 (for q=0). Rényi spectra revealed the multifractality of pore distribution in coated fried samples as for a multifractal system the Dq changes with q and the values follow a non-increasing pattern (Wang Y. et al., 2022; Mendoza et al., 2010). Shape of Rényi spectra as well as the relationship [$D_{q=2} < D_{q=1} < D_{q=0}$]

between capacity dimension (D_0), entropy dimension (D_1), and correlation dimension (D_2) confirmed that internal pore structure of coated-fried samples has properties close to multifractal self-similarity measures; as the equality $D_2=D_1=D_0$ occurs only if the studied fractal is statistically/exactly self-similar and homogeneous (Zhang M. et al., 2021; Han et al., 2020; Mendoza et al., 2009).

Amplitude of Rényi spectra ($\Delta D=D_{-8}-D_{+8}$) with respect to frying time (Figure 5.9A) suggests that heterogeneity of pore distribution reduced with the increase of frying time, as lower ΔD indicates more homogenous structure (Mendoza et al., 2010 & 2009). Decreasing trend of ΔD with FT represents, pore structure of identical batter-coated food moved towards a less heterogenous structure while fried for longer duration. This event could be understood as such: longer frying favored higher (%) pore formation and increases in percentage of pores leads to better pore connectivity and more homogeneous pore structures (Zhang M. et al., 2021). The increase in porosity of fried batter-coated samples with the increase of FT, indicates the possibility of fusing of micropores to form a homogenous structure having lower numbered but bigger sized pores (Adedeji et al., 2011). Fusing of pores (consequent into the formation of crust structure having lower interconnectivity between pores) might act against mass redistribution during post-fry holding. Figure 5.9B depicts, evolution of the ratio of entropy dimension to capacity dimension (D_1/D_0) of Rényi spectra. D_1/D_0 ratio provides information about proportional variation instead of absolute variation (which is consider as similar to the coefficient of variation in statistics), and the values closer to 1 this ratio is, the more evenly distributed is pores (Mendoza et al., 2010). Comparatively higher value of D_1/D_0 ratio for RB coated samples indicate that rice-flour based batter coating was in favor of the development of evenly distributed pore structure. Irrespective of batter formulations, the increasing trend of this ratio with FT suggests that longer frying lead to the formation of evenly distributed internal pores.

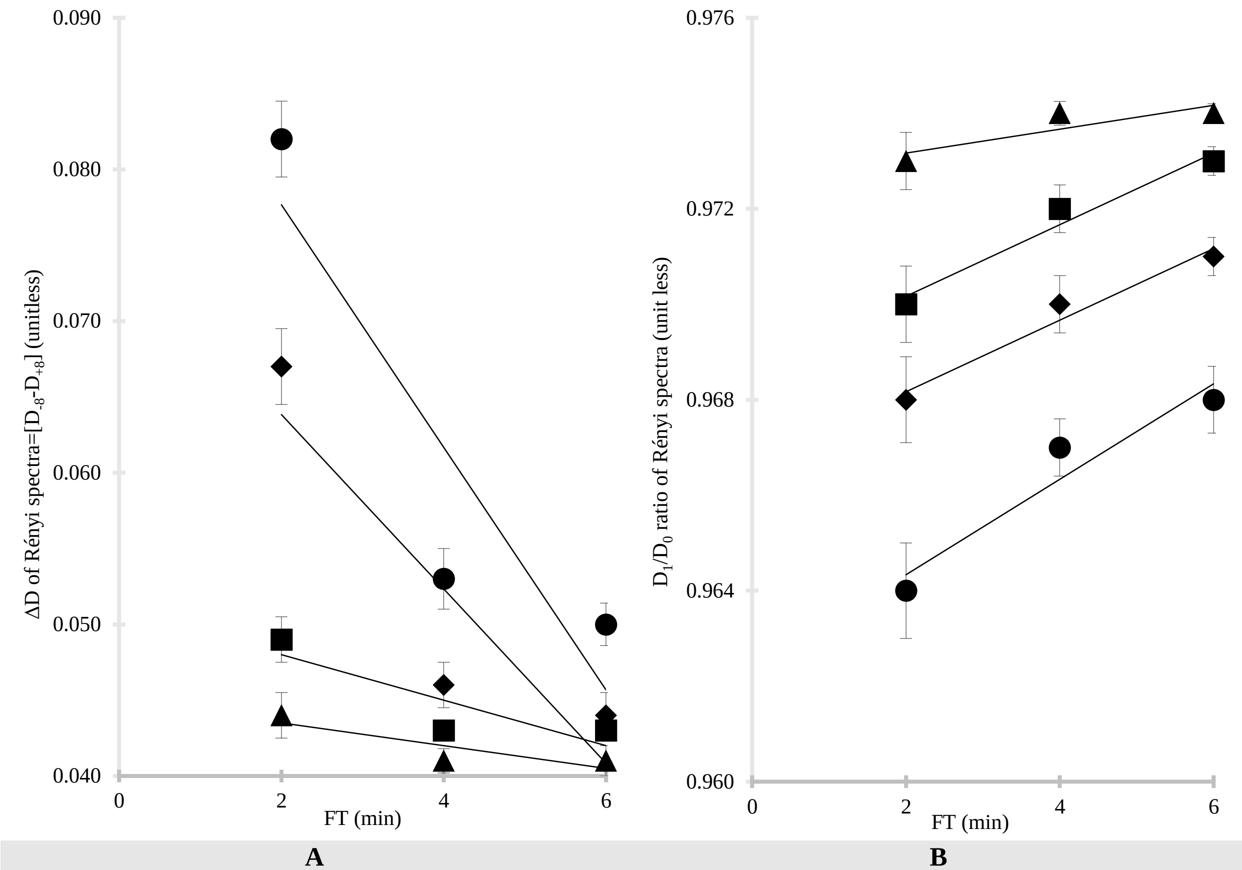


Figure 5.9: Selected Dq estimates of Rényi spectra as function of FT. **A:** ΔD= (D_{-8}-D_{+8}] and B: D_1/D_0= ratio of entropy dimension to capacity dimension of Rényi spectra. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time (minute).

Figure 5.10 depicts correlation between total porosity (TP) and selected multifractal parameters (obtained from both Singularity spectra & Rényi spectra). All the correlation suggest that increase of microporosity had favored the formation of homogeneous, less diverse, and evenly distributed pores in fried samples. Overall, the multifractal analysis have been found effective in reflecting major aspects of variability in internal pore structures of batter-coated fried products and provides unique quantitative characterization of the data spatial distribution.

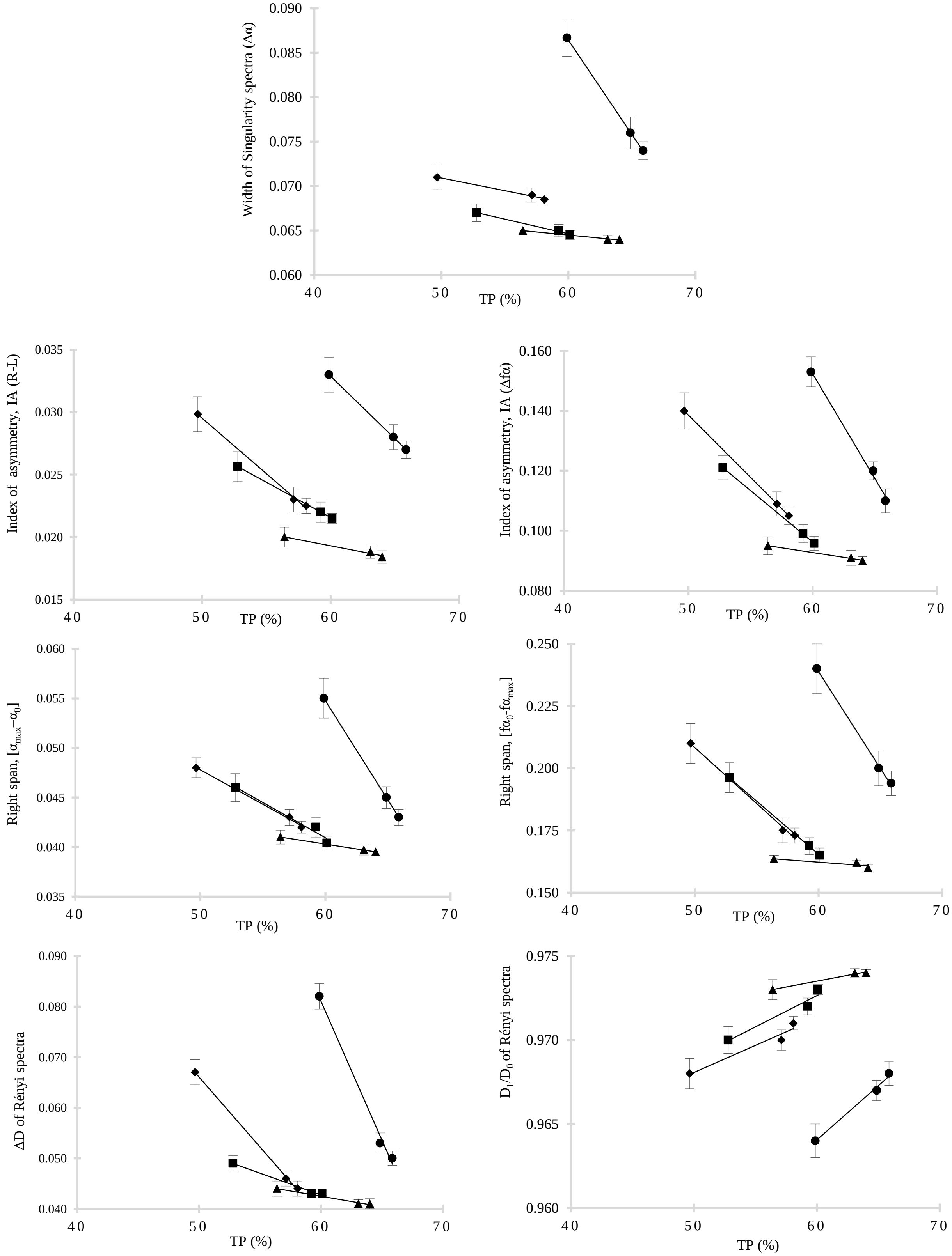


Figure 5.10: Plot of correlation between Singularity-Reiny spectral parameters and total porosity (TP). NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively.

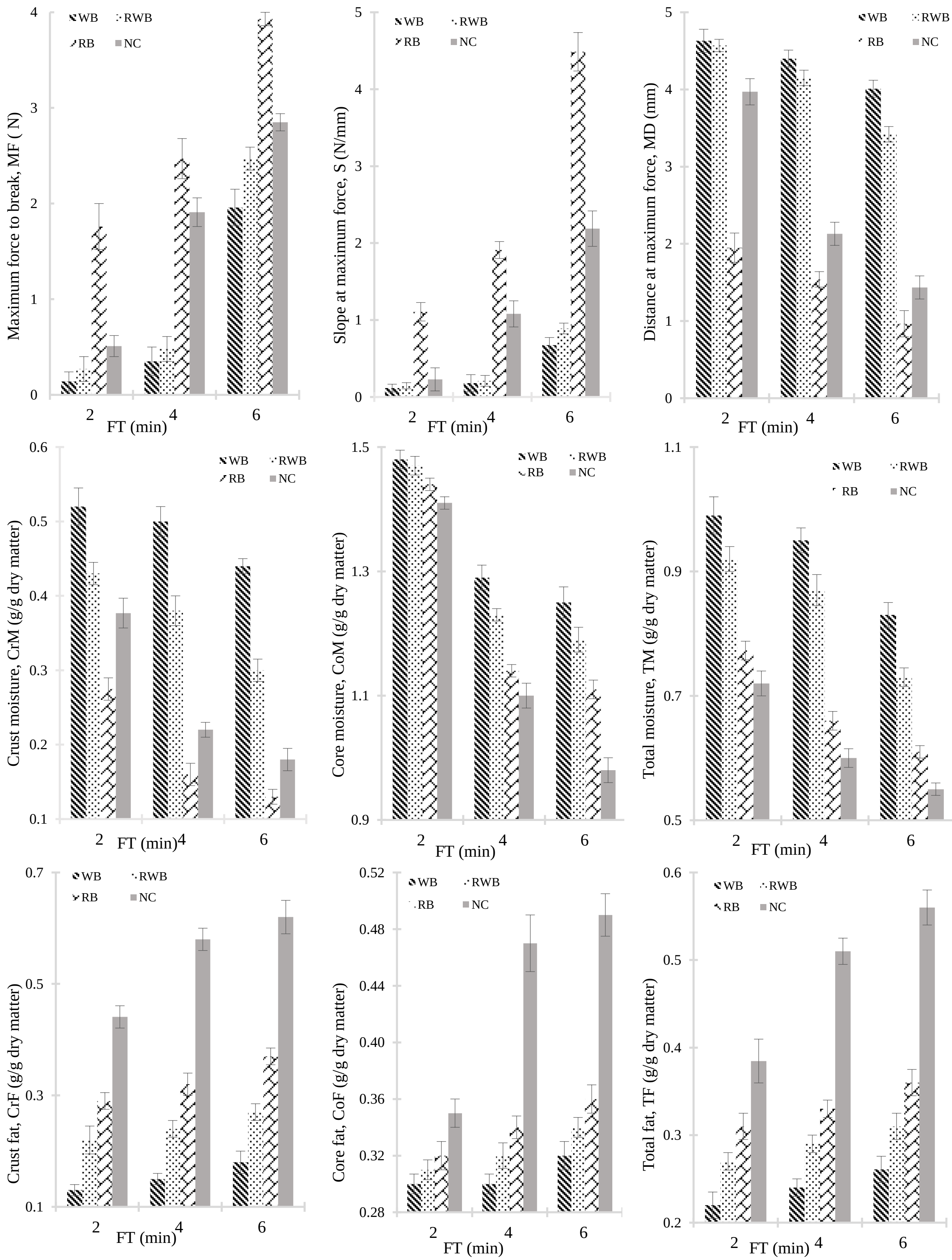


Figure 5.11: Moisture-fat profile and textural attributes of fried samples. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coating, respectively. NC represents noncoated model food. FT: frying time.

Figure 5.11 depicts, mass and textural evolution as function of frying time (FT). Crust moisture (CrM), core moisture (CoM), and total moisture (TM) of fried samples decreased with the increase of FT. Moisture profile of coated samples were different than non-coated control, that represents the impact of batter coatings on mass transfer (prevents loss of moisture) during frying. Moisture (CrM, CoM, TM) content of WB coated samples were higher than RB coated sample, this reflects higher moisture retention ability of wheat-flour based batter in comparison to rice-flour based batter systems. However, spatial moisture (CrM, CoM, TM) have shown positive correlation among themselves. CrM, CoM, and TM have shown decreasing trend with the increase of FT, while microporosity (CrP, CoP, TP) increased with the increase of FT. Thus, moisture and microporosities of coated fried samples are found as inversely correlated. Crust fat (CrF), core fat (CoF) and total fat (TF) content has shown positive relation with frying time. Fat content of coated fried samples were lower than non-coated sample, which assures the oil-reduction functionality (by impacting moisture loss) of batter-coatings. CrF, CoF, and TF have shown increasing trend with FT, while microporosity (CrP, CoP, TP) increased with the increase of FT. Thus, fat and microporosities of fried samples were found as positively correlated. Higher fat was observed for RB coated samples compared to WB batter. This could be considered from microstructural point of view as such: the micropores of RB coated samples might be highly interconnected to each other's, and strong interconnectivity of pores as well as their higher presence (% microporosity) had favored higher oil uptake during frying. It has been reported that addition of rice-flour into batter formulation decreases the fragmentation index (FI); and decrease in FI indicates increase in the degree of pores connectivity (Adedeji & Ngadi, 2010).

Hardness (MF) and crispiness (S) of fried batter coating (crust) increased with the increase of FT. Lowering of ductileness (MD) with the increase of FT, indicates the increase in brittleness of the crust (Rahimi et al., 2017). Increase in hardness, crispiness and brittleness are attributed to mass-transfer process, as crust formation in fried food is the result of changes in its original state. Development of textural attributes in batter-coatings were found as positively linked with moisture-loss and increase in fat-content. It was previously observed (Figure 5.3), crust microporosity increased with frying time. Lower crust moisture associated with higher crust fat and crust microporosity of rice-flour based batter coated fried samples have shown higher hardness, crispiness, and brittleness. The relation between micropores and textural attributes of fried coated samples could be understood as such: higher loss of moisture led to the formation of higher pores during frying and consequent higher fat uptake; and the porous, dried structure are generally crispy and brittle in nature (Voong *et al*., 2018). It has been proposed, mechanical properties could be related to the width (α_{max}–α_{min}) of the multifractal Singularity spectra (García-Armenta et al., 2016). In this study, higher hardness, crispiness, and brittleness is observed for RB coated fried samples, for whom lower width ($\Delta\alpha=\alpha_{max}-\alpha_{min}$) of Singularity spectra was observed. This gives the impression, comparatively homogenous internal pore structure favors the development of desired textural attributes (crispness, brittleness) in batter-coated food products.

Table 5.1 summarizes the post-fry textural evolution as represented by their normalized (ratio of values at any duration of post-fry holding and the initial value) values (Rahimi et al., 2017). The hardness, brittleness, and crispness of crust has changed within post-fry holding duration (HD) of 30 min, where extent and nature of evolution were intertwined with batter-formulations. Hardness (MF*) and brittleness (MD*) of RB crust increased considerably while change of crispiness (S*) was not significant. Increase of hardness and associated brittleness of RB crust might be the consequent result of starch-retrogradation (disaggregated amylose and amylopectin chains in gelatinized starch realign themselves to form more ordered structures), as starch content of rice-flour was higher. WB coated samples losses its crispiness and brittleness. This could be due to moisture migration from core to crust region of coated fried samples, as moisture reduces the crunchiness of food by weakening solid matrix (Voong et al., 2018). Changes in textural attributes of fried samples during post-fry holding have shown relation with their frying time. Coated samples that received longer frying treatment develops a crust which were less prone to post-fry textural changes. This could be understood as such: longer frying treatment reduces moisture content of coated samples to a greater extent, than lower FT. Hence, longer FT develops crust having less moisture and presence of lower moisture had favored their textural stability during post-fry holding.

Table 5.1: Normalized value of maximum force (MF*), normalized value of maximum distance (MD*), normalized value of slope (S*) as index of textural evolution of fried samples during post-fry holding.

Frying time, (FT)	Batter coating	Textural attributes at post-fry holding					
		MF*		MD*		S*	
		0 min	30 min	0 min	30 min	0 min	30 min
2 min	WB	1.0^{A}	0.81 ± 0.04^{B}	1.0^{A}	0.87 ± 0.04^{B}	1.0^{A}	0.78 ± 0.05^{B}
	RWB	1.0^{A}	0.82 ± 0.02^{B}	1.0^{A}	0.89 ± 0.02^{B}	1.0^{A}	0.80 ± 0.02^{B}
	RB	1.0^{B}	1.20 ± 0.05^{A}	1.0^{B}	1.12 ± 0.03^{A}	1.0^{A}	1.01 ± 0.02^{A}
	NC	1.0^{A}	0.94 ± 0.01^{B}	1.0^{A}	0.96 ± 0.02^{A}	1.0^{A}	0.95 ± 0.02^{A}
4 min	WB	1.0^{A}	0.83 ± 0.05^{B}	1.0^{A}	0.89 ± 0.02^{B}	1.0^{A}	0.81 ± 0.03^{B}
	RWB	1.0^{A}	0.86 ± 0.04^{B}	1.0^{A}	0.90 ± 0.02^{B}	1.0^{A}	0.82 ± 0.02^{B}
	RB	1.0^{B}	1.29 ± 0.04^{A}	1.0^{B}	1.18 ± 0.01^{A}	1.0^{A}	1.01 ± 0.01^{A}
	NC	1.0^{A}	0.97 ± 0.02^{A}	1.0^{A}	0.98 ± 0.02^{A}	1.0^{A}	0.98 ± 0.02^{A}
6 min	WB	1.0^{A}	0.88 ± 0.04^{B}	1.0^{A}	0.91 ± 0.02^{B}	1.0^{A}	0.89 ± 0.03^{B}
	RWB	1.0^{A}	0.93 ± 0.03^{B}	1.0^{A}	0.93 ± 0.02^{B}	1.0^{A}	0.92 ± 0.02^{B}
	RB	1.0^{A}	1.03 ± 0.02^{A}	1.0^{A}	1.03 ± 0.02^{A}	1.0^{A}	1.01 ± 0.01^{A}
	NC	1.0^{A}	0.98 ± 0.02^{A}	1.0^{A}	0.99 ± 0.02^{A}	1.0^{A}	0.98 ± 0.02^{A}

Upper-case letters (A-B) ranks significant ($p<0.05$) difference for the same attribute of identical samples at different post-fry holding time. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coating, respectively, and NC represent noncoated model food.

Figure 5.12 depicts, post-fry mass redistribution in crust region. As no post-fry de-oiling treatments were applied, hence, there was no considerable change in crust fat content. Considerable changes in crust moisture were observed. In coated samples, higher extent of moisture gain in crust was associated with shorter frying time, and this was also true for post-fry textural quality changes. Increase in crust moisture might be associated with moisture migration from juicy core to dry crust region, as this could be perceived by comparing their initial moisture content i.e., CrM<CoM. As post-fry changes in CrF was not notable while the change in CrM was so prominent, hence, post-fry textural evolution of batter-coated food products could be considered as moisture migration dependent event.

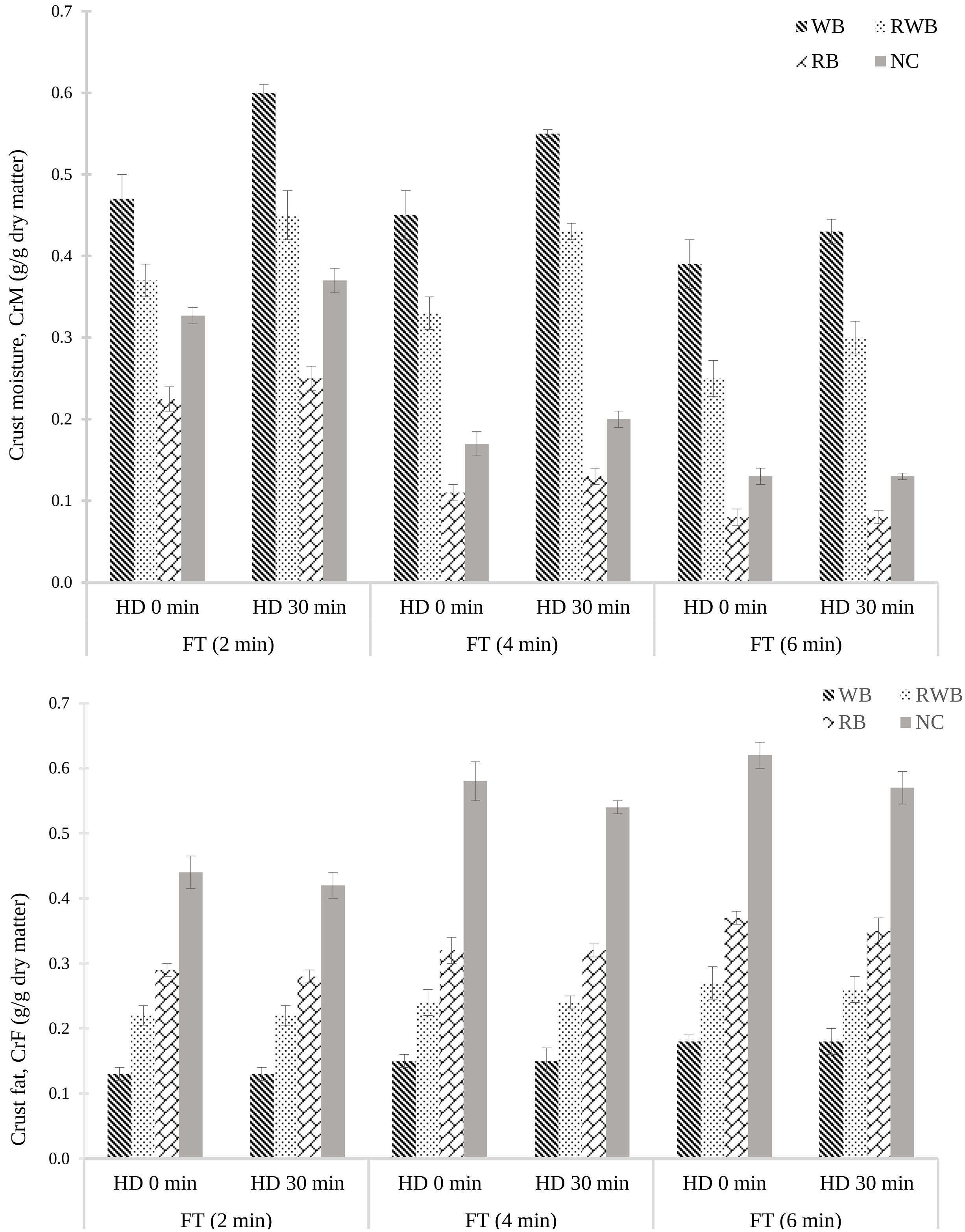


Figure 5.12: Post-fry evolution of crust moisture-fat profile. WB, RWB, RB represent wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coating, respectively, and NC represent noncoated model food. FT: frying time. HD: holding duration (minute).

From microstructural point of view, post-fry textural quality changes could be understood as such: during frying food losses its moisture and form pores, which subsequently fills up with frying oil. Longer frying treatment results low-moisture containing porous crust, which was mostly filled with frying oil. Therefore, lesser extent of moisture migration occurs between core and crust region, which favored higher post-fry textural stability of the crust. In contrary, at lower FT, pore formation in outer coating region might not impacted the core-moisture to a greater extent and the formed pores in crust might be slightly filed with oil, which offered available space to moisture in favor of their re-distribution. As consequence, considerable amount of moisture had migrated from juicy core to crust region via micropores, that consequently impacts textural stability of the crust during post-fry holding. In comparison to WB, the RB batter coatings develop crust having higher micropores & fat along with lower moisture. Hence, post-fry textural stability of RB batter coated samples was rationale according to the difference in mass-distribution and microstructural properties of WB coated samples. It is notable, internal pore structure of RB coated samples were comparatively less heterogenous than WB coated samples, and short FT had developed coated products having heterogenous pore structure, while longer FT was in favor of less heterogenous pore distribution. This indicates, moisture distribution in coated product (at immediately after frying) was also heterogeneous while they are fried for a shorter period, as pore formation in fried product is associated with the loss of moisture in the form of vapor (Rahimi & Ngadi, 2015; Adedeji & Ngadi, 2011). Therefore, higher extents of moisture re-distribution towards a homogenous distribution are most probable in coated-products which has received comparatively shorter frying treatment, and consequent higher extent of post-fry textural quality changes is anticipated for them.

Principal component analysis (PCA) are applied to depict the correlation among multi-dimensional data that obtained from destructive analyses and nondestructive X-ray μCT image based multifractal analysis. Correlation between moisture content (TM), fat content (TF), crispiness (S), porosity (TP), and selected multifractal parameters ($\Delta\alpha$, $\Delta f\alpha$, R-L, ΔD, D_1, D_2) were studied. PCA biplot (Figure 5.13) showing the relationship between samples, between attributes, and between attributes to samples. First two principal components (PC1 and PC2) accounts for 84.1% of the data variance. PCA bi-plot reveals clear discrimination between non-coated and batter-coated fried samples, as well as discrimination among batter-formulations. Negative correlation between TP and TM as well as positive correlation between TP and TF were expected, as because of the positive relation between moisture loss and fat update. Strong positive correlation between S and entropy dimension (D1) indicates, crispiness of coated samples were influenced by the degree of spatial heterogeneity of pore distribution (Lafond et al., 2012). Positive relations between (D1 & FT) and (D1 & TP) manifest, frying time have considerable impacts on the spatial heterogeneity of pore distribution as well as total porosity (%). Negative correlations between FT and multifractal parameters (Δα, Δfα, R-L, ΔD) and a very strong positive correlation between FT and S indicates, longer duration of frying favors the formation of less heterogenous pore structure which provides crispy texture in coated samples. Strong correlation of D2 to S and FT, suggest that a 2nd order power law might describe the scaling relationship (San et al., 2010; Lafond et al., 2012) between internal pore distribution and crispiness of coated fried products, where frying duration could play considerable role.

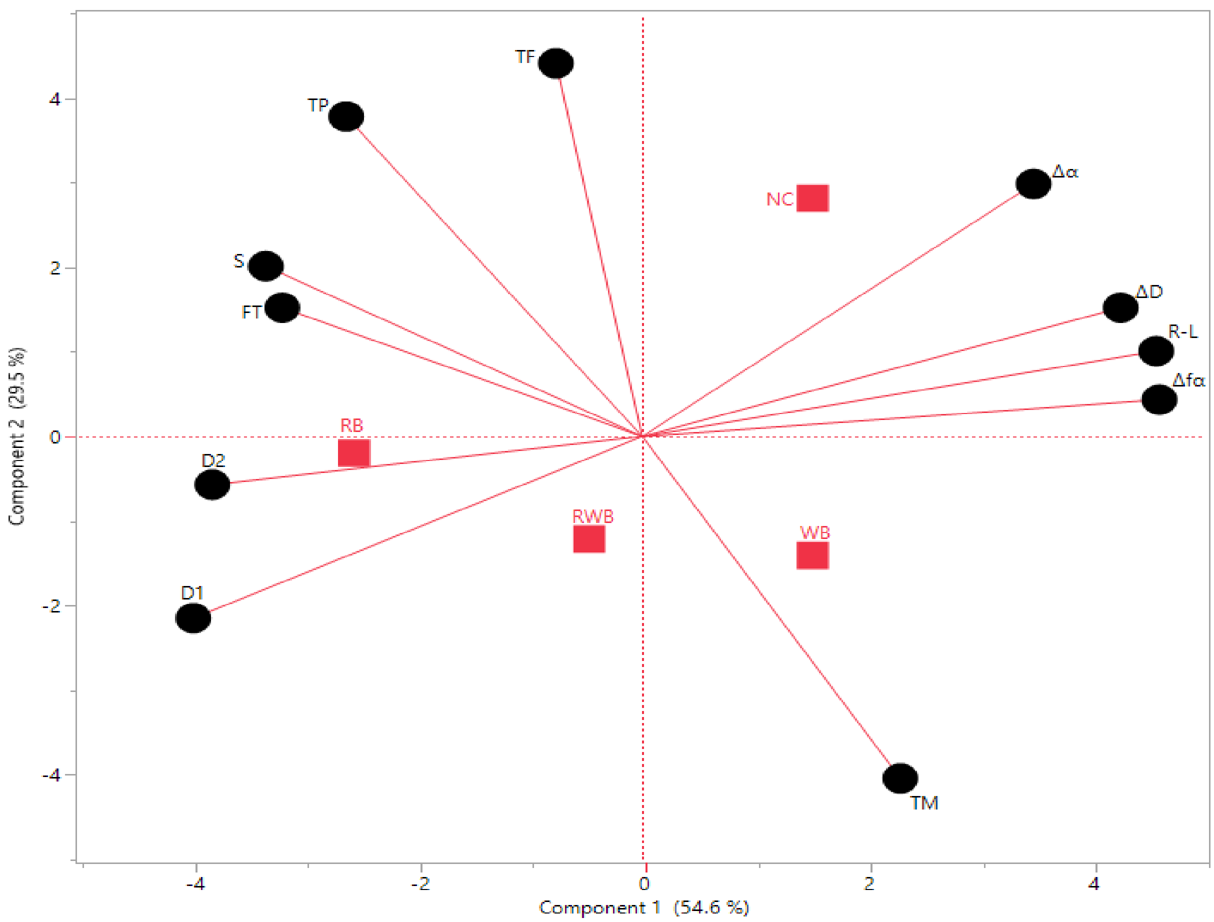


Figure 5.13: Bi-plot of PCA (principal component analysis). Black circles and red squares represent studied attributes and fried samples, respectively. NC, WB, RWB, RB represent noncoated, wheat-flour based batter, wheat & rice flour-based batter, rice-flour based batter coated model food, respectively.

5.5 Conclusion

Batter-formulations and frying time significantly impacted pore formation (% porosity) in coated food product, and consequently affects post-fry mass distribution and textural evolution. Irrespective of spatial location, pore formation in coated products were positively correlated with frying time. Pore distribution have shown considerable influences on mass (moisture) re-distribution between crust and core region of coated products during post-fry holding, that consequentially affects textural attributes. Compared to wheat-flour, lower extent of post-fry moisture redistribution and higher textural stability were observed in rice-flour based batter coated samples. Presence of higher pore space, lower moisture, and higher fat is in favour of stable textural attributes of batter-coated product, as these could limit moisture re-distribution between core and crust region. In this study, high resolution X-ray microtomographic images were used to investigate the internal structure of fried coated products and Multifractal analysis (MFA) has been used to characterize the distribution of micropores. 1D frequency curve of microporosity revealed that pore structure of coated products are highly impacted by spatial location. MFA outcomes i.e., Singularity spectra and Rényi spectra revealed that pore structure of batter-coated fried products have multifractal scaling behaviour and pore distribution within the analysed representative volume were inhomogeneous. Higher heterogeneity in pore distribution were observed in lower concentration of pores at crust region, compared to core region of batter-coated fried product. Increase of frying time have shown impact in favour of homogeneous pore distribution in crust of coated food product, and crust having homogenous pore structure were comparatively stable against post-fry mass re-distribution and textural evolution. Multifractal parameters of pore structure (at micron level) were estimated and have shown correlation with physicochemical properties (at macroscale) of batter-coated fried food products. This study demonstrated, X-ray μCT image-based MFA is an effective tool for characterizing internal pore distribution of fried foods, in understanding of how internal microstructure affect mass and textural evolution of coated products. This study improves the understanding on microstructural characteristic of coated products, to customize post-fry mass and textural evolution.

CONNECTING TEXT TO CHAPTER 6

Post-fry textural quality of batter-coated fried food is of crucial importance. Loss of textural quality of batter-coated fried food is the prime cause of its consumer rejection. Previous study (Chapter 5) has established relation between textural quality and internal microstructures of batter-coated fried foods. The quality and stability of food materials are greatly impacted by glass transition phenomenon. Chapter 6 investigated the glass-transition behavior of batter-coated fried foods, in order to better understand their post-frying textural evolution.

Parts of this chapter have been presented at scientific conference.

Md. Hafizur Rahman Bhuiyan and Michael O. Ngadi. (2023). Thermomechanical transitions of fried food batter coatings. Presented at American Society of Biological and Agricultural Engineering (ASABE) annual conference. Held at Omaha, Nebraska, USA, from July 9–12, 2023. No. 2300870. *Oral Presentation*.

CHAPTER 6

THERMOMECHANICAL TRANSITIONS OF FRIED FOOD BATTER COATINGS

6.1 Abstract

Glass-transition is a crucial phenomenon to control the quality and stability of food materials. This study was aimed to characterize thermomechanical transitions of fried food batter coatings. Wheat and rice flour-based batter systems were used to coat a meat-analog model food. Coated products were fried at 180°C in canola oil for 2, 4 and 6 min. Glass-transition behavior of the coatings were assessed by differential scanning calorimetry (DSC) directly after frying or after post-fry holding. Mechanical texture analyzer and X-ray µCT techniques were used to assess textural-attributes and internal-microstructure, respectively. This study reveals, batter-formulation greatly impacts the glass-transition-temperature (Tg) of fried batter coatings. Compared to wheat-flour, higher Tg values have been found for rice-flour based fried batter coatings. Tg of fried batter coatings were positively correlated with frying-time, in contrast, Tg values are negatively correlated with moisture content of fried batter coatings. Both moisture and fat acts as plasticizer in fried-batter coatings, and results into their negative values (<0°C) of the Tg. Internal microstructure greatly influences textural attributes of fried batter coatings, wherein Tg are positively correlated with microporosity (%). Post-fry textural stability of coated products, are directly impacted by Tg of fried batters. Subzero Tg value of fried batters explains their moisture-redistribution induced post-fry textural changes (hard-to-soft, brittle-to-ductile, crispy-to-soggy) at room-environment (25°C) and under IR-heating (65°C).

6.2 Introduction

The presence of high amount of fat in fried food products is a serious health concern. To produce fried products of having low-fat, foods surface modification by batter-coating play effective role (Liberty et al., 2019). In addition to fat reduction, batter-coatings imparts unique textural attributes (juicy inside and dry outside) in fried products. Textural attribute (specifically crispiness) of coated products is considered as the index of its freshness and loss of textural attribute is the prime cause of its consumer rejection (Carvalho & Ruiz-Carrascal, 2018; Kerr, 2016). Despite this fact, most of the past research on batter-coating technology have focused on reducing fat content in fried products, wherein their textural attribute hasn't got required attention. Hence, scientific insights and relevant information on batter-coated fried foods textural evolution, are still very rare.

Frying is a dynamic mass transfer process, where food loses its moisture and uptake oil. Hence, both water and oil could play crucial role on the textural traits of batter-coated fried foods. In these scenario, changes in textural qualities of batter-coated fried products could be linked up with the glass-transition phenomenon (glass to rubber transition and vice-versa) hypothesizing this as such: the state of coated fried product becomes glassy (due to substantial decrease of their initial moisture content during frying) and post-fry changes in textural property of coated products is the outcomes of mass-redistribution induced glass-to-rubber transitions. Pertinent literature pointed that several factors could influence the glass-transition of biomaterials, that includes temperature, time, moisture, matrix composition, structure, surrounding environment and so on (Jothi et al., 2020; Sogabe et al., 2018; Rahimi et al., 2017; Roos, 2010; Pereira & Oliveira, 2000). Therefore, to predict and control textural evolution of batter-coated fried foods, detailed investigation is required in understanding the nexus between glass-transition-temperature, textural attributes, internal microstructure, and moisture-fat distribution under different circumstances.

In plain words, the transformation of amorphous solid to supercooled liquid or vice-versa is defined as glass-transition and associated temperature is termed as glass-transition-temperature (Maidannyk et al., 2019; Roos, 2010). Glass-transition-temperature (Tg) of food materials could be determined based on the observation of changes in different properties including heat capacity (Jothi et al., 2020; Maidannyk et al., 2019; Sogabe et al., 2018; Roos, 2010). It is notable, glass-transition-temperature of food products and storage-temperature are closely related for determining the quality characteristics during their storage (Perdomo et al., 2009; Kayacier & Singh, 2002). It could be expected that study on batter-coated fried products' glass-transition behavior would provide detailed insights on their textural evolution at frying and post-frying holding. However, detailed information on Tg of batter-coated fried product is very rare.

Considering mentioned facts, the overall goal of this work was set to (i) investigate the effects of batter-formulations and frying time on glass-transition behavior of coated fried foods (ii) understand the nexus between glass-transition-temperature, internal microstructure, moisture-fat profile and textural evolution of batter-coated fried foods (iii) study the impacts of post-fry holding environment on Tg and texture of fried batter coatings.

6.3 Materials and Methods

6.3.1 Materials

Soy protein isolate was purchased from MP Biomedicals (29525 Fountain Pkwy, OH, USA). Wheat gluten was supplied by Sigma-Aldrich Co. (Ontario, Canada). Methylcellulose (MC) and Sodium pyrophosphate ($Na_4P_2O_7$) crystals were obtained from Fisher Scientific (Fair Lawn, NJ, USA). Sodium bicarbonate ($NaHCO_3$) was supplied by Church & Dwight Canada Corp. (Ontario, Canada). Rice flour (Suraj®, composed of 83.32% carbohydrate, 6.67 % protein, 8.68% moisture and 1.33 % fat) was procured from a grocery store in Montreal, Canada. Commercial wheat flour (Five Roses®, composed of 73.33% carbohydrate, 13.33 % protein, 8.68% moisture, 3.33% fiber and 1.33 % fat), NaCl (Sifto®) and Canola oil (Sans nom®, Loblaws Inc.**)** were acquired from a grocery store in Sainte-Anne-de-Bellevue, Canada.

6.3.2 Sample preparation

Soy protein isolate (SPI), wheat gluten (WG), canola oil (CO) and distilled water (DW) were used to prepare a restructured vegetable protein based meat-analog model food. Firstly, wheat gluten was mixed with canola oil, then soya protein isolate was added and mixed thoroughly. After that, distilled water was added and mixed properly until a very soft-textured dough was developed. In the prepared dough, the respective proportion (g) of ingredients i.e., WG: CO: SPI: DW were 12:15:22:73. Prepared dough was used to fill individual rectangular-shaped cavities of silicon mold (Freshware CB-115RD 24-Cavity silicone mini mold). The covered silicon mold was held in complete horizontal position (surrounded by water, but not immersed) on top of a flat metal wire surface and heated at 70°C for 15 min in a water bath. After heat treatment, the mold was allowed to reach equilibrium with room temperature. Then aluminum cover was removed, and gentle pressure (by finger) was applied from the back of each cavity, to bring out the formed substrates without any fracture. This substrate was used as meat-analog model food, weighing an average weight of 16±0.2 g/substrate and having a uniform dimension of 5.6 cm x 2.5 cm x 1.3 cm. Prepared model-food was packed in zip-lock plastic bags and stored in refrigerator (4±2°C) for 24 hour, before its use.

Tempura type batter coatings were used to coat the meat-analog model food. To prepare batter coatings: Sodium bicarbonate (1.4%), Methylcellulose (0.3%) and NaCl (1.5%) were added and mixed properly with fine white powder of Sodium pyrophosphate (1.8%), to form a homogeneous powder-mix. The powder-mix was further thoroughly mixed with dry flour in a proportion of 5% (powder-mix) to 95% (flour). Distilled water was added to solid ingredients with a total solid to water ratio of 1:1.3 and mixed them properly until a homogenous batter slurry was obtained. Wheat based batters (WB) were formulated using only wheat flour whereas rice based batters (RB) were formulated with rice flour. The RWB batter was formulated using equal mixture of wheat and rice flour. The model food was brought out of refrigerator and kept at room environment for 30 min, prior to batter coating. Model food was fully immersed in batter system for 1 min and then removed from batter solution to make a complete coat. The coated sample was held on a kitchen-fork for about 30 sec, to drain the excess amount, in order to obtain a uniform layer of coating around model food core. Noncoated (NC) meat-analog model food was used as control sample.

A programmable deep-fat-fryer (T-fal Compact, FF122851, China) was used to fry the samples at atmospheric condition. Prior to frying, fryer was filled with 1.5 L of fresh canola oil and temperature was set to 180°C. Frying-oil was preheated for 1 hr and stirred to minimize variation in oil temperature. Three samples were transferred into the fryer and uninterruptedly fried for 2, 4 or 6 min maintaining a sample to frying-oil ratio of 1:30. At the end of frying, the frying-basket was removed from oil and was shaken 5 times to remove excess oil from the surface of fried products. To minimize variation in frying-oil properties each batch of oil was used for frying only seven batches of samples (three sample/batch) equivalent to around 30 min of frying operation; before the frying-oil was replaced with a new batch of fresh canola oil.

Immediately after frying, fried samples were individually placed on trays and separately kept in two distinct environments: (i) room environment, RE (25±2°C) and (ii) IR-heating, IRH (Vollrath, model OHC-500, made in China). The IR-heating system was installed on laboratory workbench, that creates a hot environment with a consistent temperature of 65±2°C. Using an adjustable system, a fixed distance of 0.28 m between sample and IR-lamps were maintained. Fried samples were exposed to a steady heat flux of 4.17 KW/m^2.

6.3.3 Evaluation of moisture, fat, texture, and microstructure

Textural properties of fried samples were evaluated by puncture/compression test, using a mechanical texture analyzer (Stable Micro Systems Texture Analyzer, TA. HD PlusC, UK). Fried samples were individually mounted on a flat rigid support. A puncture probe TA-52 (2mm dia.) and 500 N was used to punch the test specimen, at a constant test speed of 1mm/s and a travel distance of 5 mm. Puncture test were performed on three equidistance locations of each sample. Following the method of Rahimi et al. (2017), maximum force to break (N), displacement at maximum force (mm), and slope at maximum force ($N.mm^{-1}$) of the puncture test were used to evaluate crust textural properties namely hardness, brittleness, and crispness, respectively. Puncture test data were analyzed with the texture analyzers' software i.e., Exponent (ver. 6.1.14).

Internal structure of fried samples were assessed non-destructively by X-ray microtomography (µCT). Immediately after frying, a portion (20×20×5 mm) of fried sample were procured by using a microtome blade. The cut portion of fried samples were dehydrated in a freeze-drier (Modulyod-115; Thermo Savant, USA) at -50°C and 250 mbar for 48 hours and then transferred into a desiccator. Dried samples were scanned in an X-ray µCT scanner (SkyScan 1174, Bruker-Skyscan, Kontich, Belgium) with a preset voltage of 50 keV, current of 800 uA, exposure time of 2400 ms, and angular rotation step of 1.0^0 through 180^0. X-ray images were recorded as 16 bits Tagged Image File Format (TIFF files) and a cross-section pixel size of 8.9 µm was realized. Obtained images were reconstructed into a series of 2D grayscale images using a reconstruction software (NRecon, Version: 1.7.4.2, Skyscan, Belgium), and reconstructed images were saved as 16 bits TIFF files. The stack of 2D images were imported as image sequences in Java-based public domain image processing software (ImageJ 1.52 v, National Institutes of Health, USA), and images were converted to 8 bits grayscale images. From the middle part of 2D images, a square shape region (512*512 pixels) was cropped and subsequently used for further analysis as region of interest (ROI). After discarding the first few image slices, a total of 32 images were chosen to estimate the microporosity. To minimize the overestimation/underestimation of porosity (based on binarized image), the Otsu algorithm was applied 'slice-by-slice' to delineate pores and non-pore portion within the image stack. To remove any further noise from binarized images a morphological operation i.e., despeckling (median filter) was applied. Crust microporosity (CrP) was calculated as follows: [{Σ (porosity of each slice*area of ROI) / Number of slice} * Length of image stack] ÷ total volume of interest (area of ROI* Length of image stack). The calculated value was multiplied by 100 to express the porosity in percentage (%).

Crust region of fried products were carefully separated from core region, by a microtome blade (Feather, C35 type, Japan). The detached crust were freeze dried in a freeze-dryer (Modulyod-115;

Thermo Savant, USA) at -50°C and 250 mbar for 48 hours and then transferred into a desiccator to equilibrate for 30 minute. Weight of the sample before and after freeze-drying was measured and moisture content was calculated on dry basis (g/g dry matter), by dividing the mass of weight loss with the mass of freeze-dried sample.

Freeze-dried sample was grounded to increase surface to volume ratio, and an amount of 3-5 g was placed in thimbles of a VELP SER 148 solvent extraction unit (Velp Scientifica, Usmate, Italy). Fat was extracted by following the Randall method, with petroleum ether at 130°C pre-set temperature. Complete fat extraction process was performed in three consequent steps: immersion (30 min), washing (30 min), and recovery (30 min), a totaling of 90 min. Weight of extracted fat was measured, and fat content was computed on dry basis (g/g dry matter) by dividing mass of extracted oil with the mass of dried sample.

6.3.4 Differential Scanning Calorimetry (DSC)

Glass-transition-temperature (Tg) of detached crust samples were determined by using differential scanning calorimeter (DSC Q250, TA Instrument, DE, USA). Prior to Tg determination, DSC instrument was calibrated using indium (melting point 156.6 ◦C, H_f = 28.45 J/g). Nitrogen (99.99% purity) was used as purge gas with a flow rate of 50 mL/min and "T Zero" holder was used. Tg of the crust was determined by following the method of Rahimi et al. (2017), with slight modification. Around 10-15mg crust portion was placed in sample pan, a lid was attached, and a mechanical device was used to make them as hermitically sealed. One sample holder with lid (but without crust sample) was used as reference pan. Both sample containing pan and reference pan was transferred into DSC measurement chamber. In DSC measurement chamber, sealed pan was rapidly cooled to -40°C at a cooling rate of 20°C/min, held isothermally for 2 min, and then heated to +40°C at a rate of 2°C/min. At the end of cycle, data was automatically saved and analyzed by DSC-Trios 5.1.1v software (TA Instruments, USA). From DSC thermogram, glass-transition-temperatures were obtained as onset, midpoint and endset of the heat capacity change; and were represented as $Tg_{onset,}$ $Tg_{midpoint}$ and Tg_{endset}, respectively. Tg_{onset} was obtained as the change of the baseline slope, after a variation of heat flux occurred, and determined as the intersection of tangents to the curve. $Tg_{midpoint}$ was obtained from the half height of the curve, and $Tg_{endsent}$ was the end point of transition curve. Temperature encompassed by Tg_{onset} and Tg_{endset} were presented (ΔTg) as thermal glass-transition temperature range.

6.3.5 Statistical analysis

All measurements were replicated at least twice and mean ± standard deviation was reported. Tukey's HSD (honestly significant difference) were used in reporting significant ($p<0.05$) difference among means. Statistical analysis was performed by statistical software JMP 14.1v (SAS Institute Inc., Cary, NC, USA).

6.4. Results and Discussion

Figure 6.1 depicts differential scanning calorimetry (DSC) thermogram of fried (4 min) samples. Endothermic peak in DSC thermogram represents glass–rubber transition (Kawai et al., 2014). Glass-transition-temperature (Tg) of flour-based batter coated fried foods' crust have found in subzero (far below 0°C) temperature zone. In literature (Kerr, 2016), negative Tg has been reported for fried food matrix such as: donut (-18°C), carrot chips (-39°C) and french fries (-11.8°C). Tg values in the ranges between -15.1°C to -20.4°C has been reported for modified starch (potato, corn, tapioca) based fried batters (Rahimi et al., 2017). The obtained Tg values of flour based fried-batter coatings, are considerably lower than the reported Tg values of wheat (-10.31°C) and rice (-7.29°C) flour based non-fried batters (Xue & Ngadi, 2007). Interestingly, the obtained Tg values of the studied fried-batters are very close to the reported Tg values (-15°C to -25°C) of wheat and rice flour based simulated (CFT: cooking-freezing-thawing) batters (Xue & Ngadi, 2009). These info gives the impression that, probable increase of Tg (due to moisture-loss during frying) of batter-systems were negatively impacted by absorbed frying-oil. Overall Tg depression in fried-batters could be understood as the resultant outcomes of the "oil plasticizing" and "starch gelatinization" effect. It has been reported that oil can acts as "plasticizer" in fried sample (Jothi et al., 2020; Jothi et al., 2018) and Tg of gelatinized-starch are lower than the native-starch (Pariya Thanatuksorn, Kazuhito Kajiwara, 2007). It is notable, the studied batter systems were prepared with NaCl, and presence of NaCl could also be a factor behind negative Tg of fried batters. Xue & Ngadi (2007) obtained lower Tg values for salt-containing fresh batters, compared to the batter without salt.

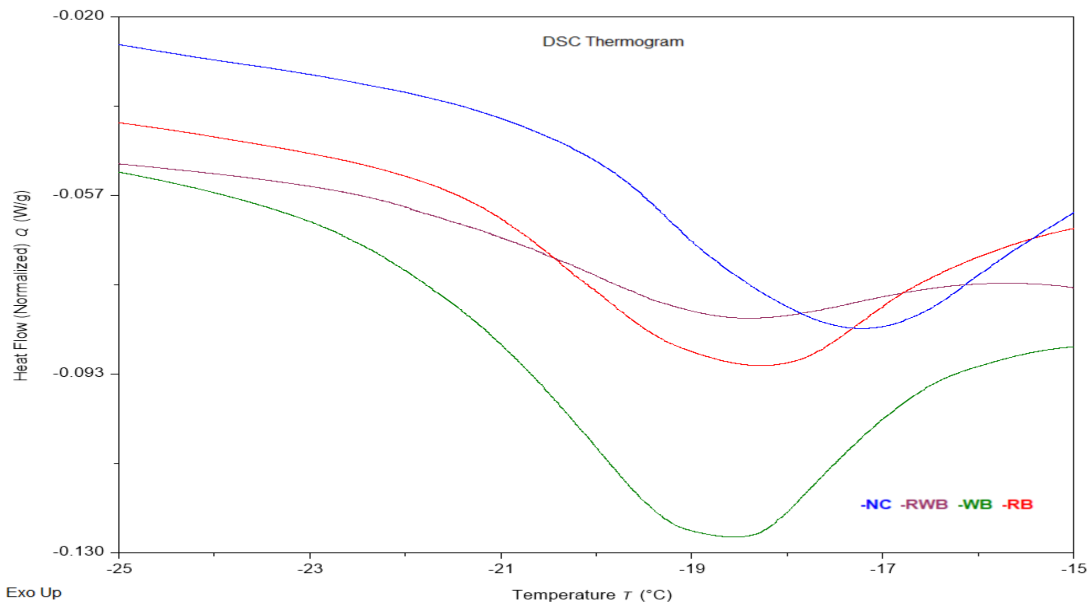


Figure 6.1: DSC thermogram of fried samples. NC, WB, RWB, RB represent non-coated model food, wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coated model food, respectively.

Tg of batter-coated fried samples were lower than the Tg of non-coated (NC) sample. This indicates, use of batter coating in producing low-fat containing fried product negatively affects the Tg of fried foods crust. The lowering of Tg is a great concern regarding the stability of fried products quality at post-fry holding; as a product is stable at glassy-state (below Tg) and its quality are prone to change at a temperature above their intrinsic Tg (Pereira & Oliveira, 2000). The Tg has a clear physical meaning and that could be understood as such: if the batter-coated fried foods are not being stored at a temperature lower than their intrinsic Tg value, viscosity of the fried foods crust matrix will be decreased drastically (due to increase in molecular mobility) that would influence the crust-structure to be collapsed and would consequently make changes in physical properties (Jothi et al., 2020; Sogabe et al., 2018). Batter-formulation showed considerable impact on Tg of the fried coatings. Compared to rice-flour based (RB) batter, lower Tg value was obtained for fried wheat-flour based (WB) batter. Observed discrepancy in Tg of fried WB and RB batters could be the effect of varying ratio of protein entity (gluten) in batter-system as well as the quantity and unique-size of their starch granules (Kerr, 2016; Xue & Ngadi, 2007). It is notable that water molecules can easily diffuse within starch–water mixture than gluten–water mixture; and water molecules is comparatively more protected against freezing within the gluten-water matrix (Xue & Ngadi, 2007). Hence, a lower Tg value could be anticipated for a gluten-dominant matrix compared to starch-based matrix.

Figure 6.2 depicts, glass-transition-temperatures (Tg) of fried batters were positively correlated with frying time (FT). Impact of FT on glass-transition-temperatures were comparatively obvious for WB batter. This could be attributed to higher water holding capacity (WHC) of wheat-flour based batter and could be understood as such: due to having higher WHC, the WB batter was able to hold much of its initial-moisture while were fried for a short time. In contrast, longer frying treatment resulted in a considerable loss of its moisture, consequently higher Tg were determined. It has been reported, glass-transition occurs over a temperature range (Roos, 2010; Kasapis et al., 2007). However, the range of glass-transition-temperature (ΔTg) weren't notably impacted by FT, while marginally impacted by batter-formulations.

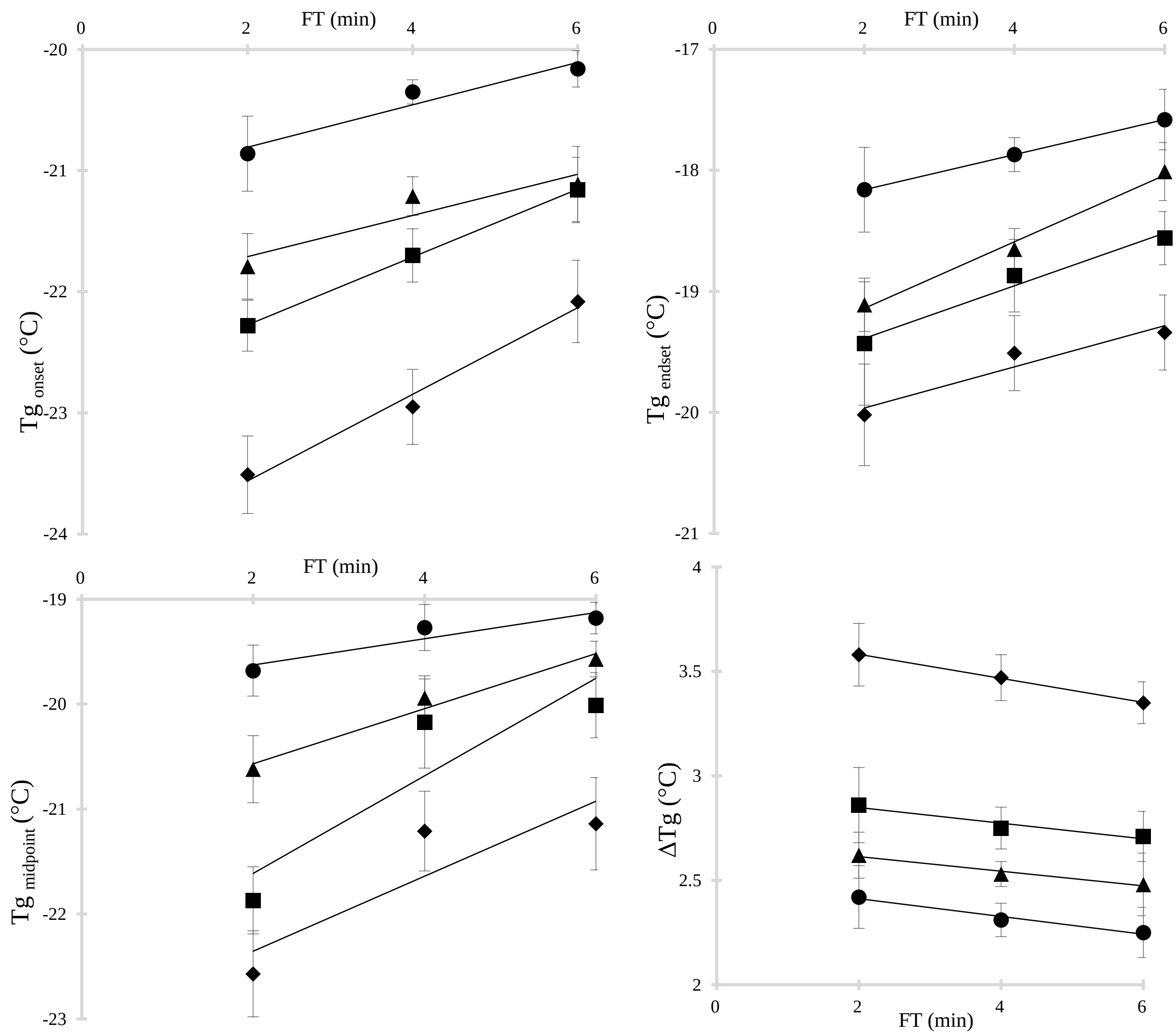
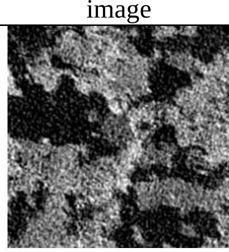
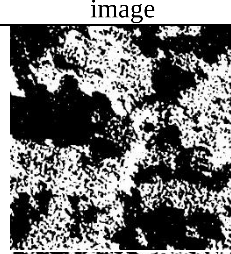
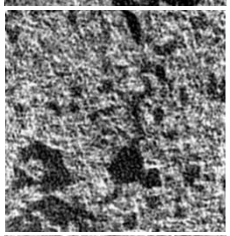
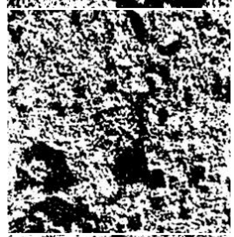
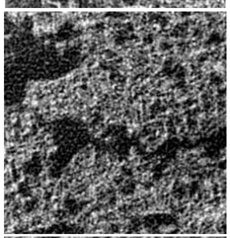
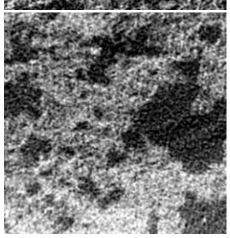


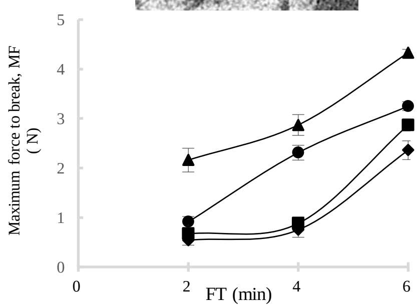
Figure 6.2: Plot of correlation between frying time and glass-transition-temperatures. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time

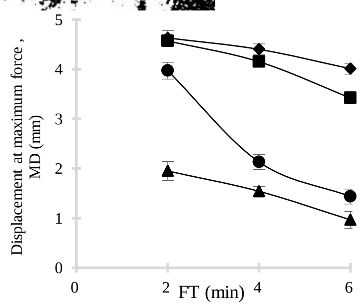
Table 6.1 depicts the microstructural, moisture-fat profile, and textural characteristics of fried foods crust. Dark color in X-ray μCT micrograph and binarized image represents internal micropores. X-ray image shows distinct difference in internal microstructure of the crust region of fried samples, where most dissimilarities were in porosity (%) and pore size. Crust microstructure of non-coated sample was characterized by higher porosity (%) while lower porosities were estimated for batter-coated samples. Compared to WB, higher microporosity was estimated for RB batter coated samples. Combine result (Table 6.1 & Figure 6.2) shows, higher Tg were found for high-porous RB coated samples, compared to low-porous WB coated samples. Compared to pellet sample (0% porosity), higher glass-transition-temperature (evaluated by thermal rheological analysis) were reported for porous (80% porosity) trehalose sample (Sogabe et al., 2018). Glass-transition-temperature (evaluated by dynamic mechanical analyzer) of starch-based extrudate porous (86.2%) samples were higher than their low-porous (35.5%) counterpart; where the decrease in pore size had resulted into lower glass-transition-temperature (Ross et al., 2002). Islam et al. (2015) reported larger pores size in dehydrated pear fruits had been in favor of a higher glass-transition-temperature. Compared to WB batter, fried RB batter was identified of having higher porosity (%) and bigger-size pores; along with these, higher Tg were determined for fried RB batters. Therefore, it could be remarked that modification of batter-formulation/frying-process to modify internal pore characteristics, could be a potential strategies to control the Tg of coated fried foods.

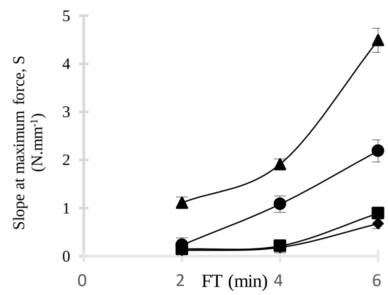
Crust microporosity (CrP) and fat (CrF) content were positively correlated with frying time (FT) of batter-coatings, while moisture (CrM) content was negatively correlated with FT. Positive relations were observed between FT and mechanical texture profile parameters [maximum force to break (MF, N) and slope at maximum force (S, $N.mm^{-1}$)], whereas a negative correlation was observed between FT and displacement at maximum force (MD, mm). These correlations suggests, while frying process continues, batter coatings continuously losses its initial moisture which results in the formation of internal-pores and uptakes more frying oil. This structural-chemical characteristics had favored the mechanical texture perception of higher hardness, brittleness, and crispiness of fried batters. Overall, internal microporosity (%) of fried batter coating has shown linear relation with textural attributes (hardness, brittleness, crispiness).

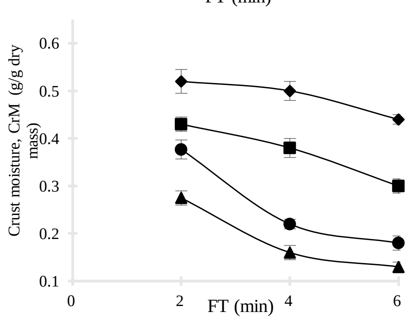
Table 6.1: Microstructural, moisture-fat profile, and textural characteristics of the crust of fried samples.

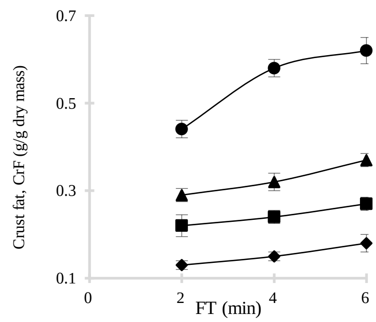
Sample	Representative X-ray µCT image	Binarized processed image	Crust microporosity, CrP (%)		
			FT 2 min	FT 4 min	FT 6 min
NC			53.21±1.98^{B}	60.92±2.06^{A}	61.17±2.84^{A}
WB			42.01±2.12^{B}	51.34±1.98^{A}	51.87±2.03^{A}
RWB			43.71±1.86^{B}	53.87±1.45^{A}	54.02±2.12^{A}
RB			50.61±1.28^{B}	59.76±1.13^{A}	60.24±1.87^{A}











NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. FT: frying time (minute). Upper-case letters represent significant ($p<0.05$) difference among means within row. FT: frying time.

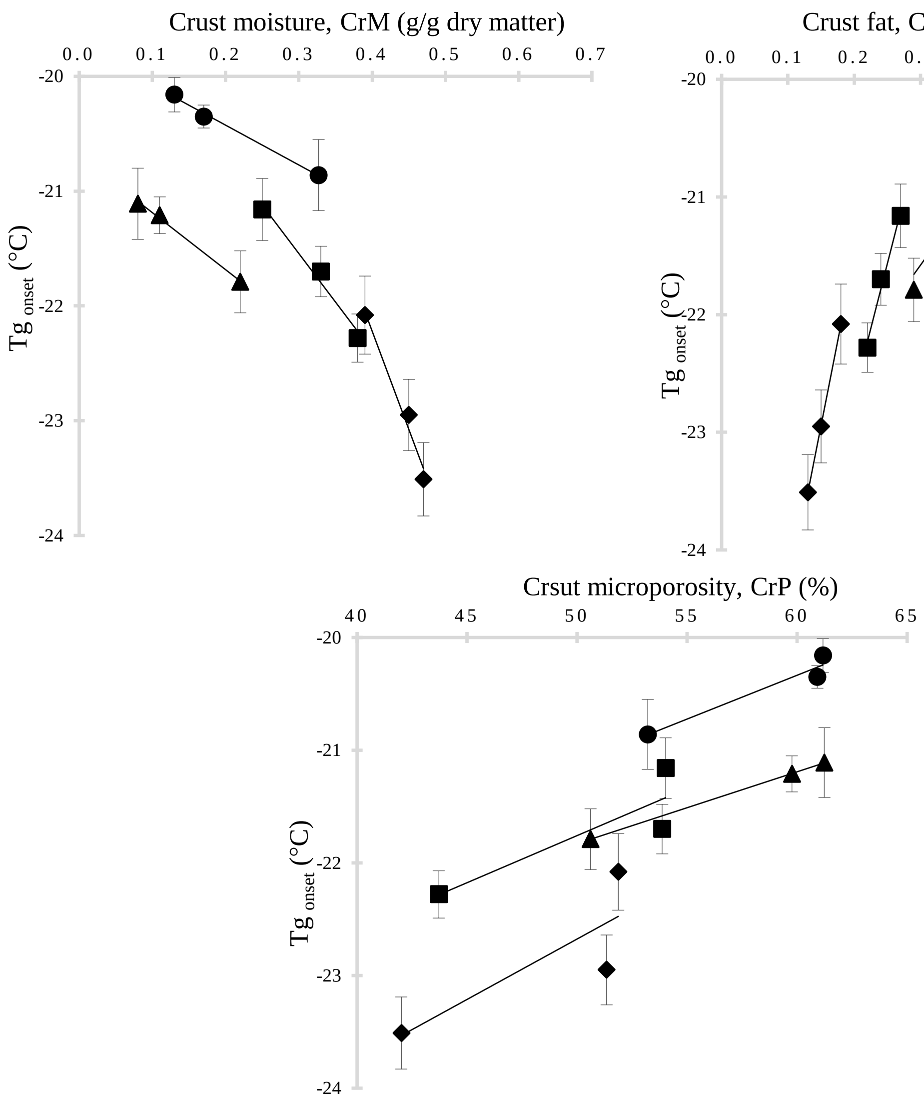


Figure 6.3: Plot of correlation between glass-transition-temperature with moisture, fat and microporosity. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively.

Figure 6.3 depicts the correlation between glass-transition-temperatures with moisture, fat, and microporosity. For all the samples, Tg were negatively correlated with moisture content (CrM). This corroborate to the "plasticizing effect" of water molecule (Jothi et al., 2020; Sogabe et al., 2018). It is notable, depending on moisture content of food matrix, water can show both "plasticizing" and "anti-plasticizing" effect. For example: in casava starch, water had acted as anti-plasticizing agent for the moisture content of 1–11%, in contrary, acted as plasticizing agent for the moisture content of higher than 23% (Kerr, 2016; Perdomo et al., 2009). However, negative correlation between moisture content and glass-transition-temperature of "dehydrated food matrix" such as bean powder (Kyomugasho et al., 2021) and pear (Islam et al., 2015) has been reported in literature. In this study, the degree of inter-dependency between moisture content and Tg were apparently higher (higher slope) for batter-coated samples, compared to non-coated sample. Within the studied moisture range, higher inter-dependency was noticed for high-moisture containing WB batter. Kerr (2016) pointed out that wheat-flour containing product might be rubbery at 20% moisture, but glassy at 5% moisture. In this study, WB batter was formulated with wheat-flour whose final moisture content was 0.35 (g/g dry matter) even after 6 min of frying. This explains their rubbery state and related textural attributes (lower hardness, crispiness, and brittleness) at the end of frying (Table 6.1). However, it was noticed that the formulations of batter-coating is a crucial regulating factor of the Tg and moisture content of fried foods. Hence, addition of selective ingredient in batter-formulation at preparation stage, could be a pragmatic option to obtain desired final properties in fried products through regulation of their Tg. For the water content of up to 8 % (db), Jothi et al. (2018) noticed higher Tg value for glucose-added samples than non-glucose added counterpart.

A linear-positive correlation was observed between fat (CrF) and Tg of fried foods. Higher fat was present in the crust of non-coated fried sample, whose Tg was also higher. However, for all the studied (coated & non-coated) samples the Tg were found in subzero temperature zone. This indicates both moisture and fat could play role as “plasticizer" on glass-transition of fried products, depending on the presence of one another’s. Madrigal et al. (2011) reported that water had showed better “plasticizing” effect (on cassava starch-corn oil blend) in the presence of lower corn oil, and apart from moisture, corn oil also has showed plasticizing effect via hydrophobic–hydrophilic interactions with the starch content of casava. In biopolymer, a plasticizer increases the free-volume between polymer chains (by reducing number of active centers that can support rigid polymer–polymer contacts, or by reducing macromolecular mobility of the polymers) and reduces the glass-transition-temperature (Jothi et al., 2020; Tavera-Quiroz et al., 2012; Li et al., 2010). Earlier, Perdomo et al (2009) reported that the plasticizing impact of water on food-biopolymers is low at low water activity (synonymous to lower moisture content). In this study, moisture content of batter-coated fried products crust were high whereas their fat content was lower. Hence, plasticizing effect of moisture might have not faced great hindrance, that consequentially resulted in their lower Tg value. In contrast, higher oil and lower moisture was present in the crust of NC fried sample. Hence, it could be presumed that oil has acted as key plasticizer for non-coated sample where the plasticizing effect of water might have faced some extent of hinderance. As consequence, a slightly higher Tg value was obtained for NC sample. It has been highlighted, not only the amount but also the type of fat could be a considerable factor that could have impact on glass-transition-temperature of food matrix (Jothi et al., 2020; Kawai et al., 2014). Similarly, Ngadi et al. (2007) reported that oil content of fried chicken nuggets had differed based on the degree of saturation of frying-oil. Therefore, the use of suitable frying oil/oil blends to control glass-transition temperature of batter-coated fried foods could be an interesting strategy and that requires more detailed investigation.

Positive correlations were observed between crust microporosity (CrP, %) and glass-transition-temperature (Tg). This observation is analogous to the study of Kasapis et al. (2007) where they established the relationship between porosity and glass-transition-temperatures of dehydrated apples. However, observed correlation between Tg and CrP affirms, both moisture and fat are prominent influential factors behind the glass-transition of batter-coated fried foods; as the pore-formation in fried foods are associated with the loss of moisture (as vapor) and uptake of frying oil (Liberty et al., 2019; Dehghannya & Abedpour, 2018).

Figure 6.4 depicts correlation between glass-transition-temperature and textural parameters. Overall, glass-transition-temperature, maximum force to break (MF), and slope at maximum force (S) has shown positive correlation. In contrast, a negative correlation was observed between Tg and displacement at maximum force (MD) of mechanical texture profile. These correlation indicates, hardness, crispiness and brittleness of fried samples increased with the increase of their intrinsic Tg. It is noticeable, slight change in Tg are associated with drastic changes in studied textural attributes (hardness, brittleness, crispiness). This drastic change in textural properties might be interlaced to the change in viscosity profile. It could be mentioned that a small difference in Tg could cause a large difference in viscosity (Sogabe et al., 2018). However, interdependency between Tg and textural properties of batter-coated foods might also be associated with the changes in moisture content and internal microstructure (porosity). It has been mentioned that microstructure (presence of pores, % porosity) could play vital role in physical/mechanical properties of dehydrated food materials (Sogabe et al., 2018; Rahimi et al., 2017; Islam et al., 2015). In this study, higher textural attributes as well as higher glass-transition-temperature were obtained for low-moisture-containing and porous fried RB batter coatings, compared to high-moisture-containing and low-porous WB batter. However, comparative higher Tg of fried RB batter explains their higher textural. A food with a high Tg is generally hard and brittle in nature, in contrast, a low glass-transition-temperature is synonymous to soft and rubbery texture (Kasapis et al., 2007; Kerr, 2016).

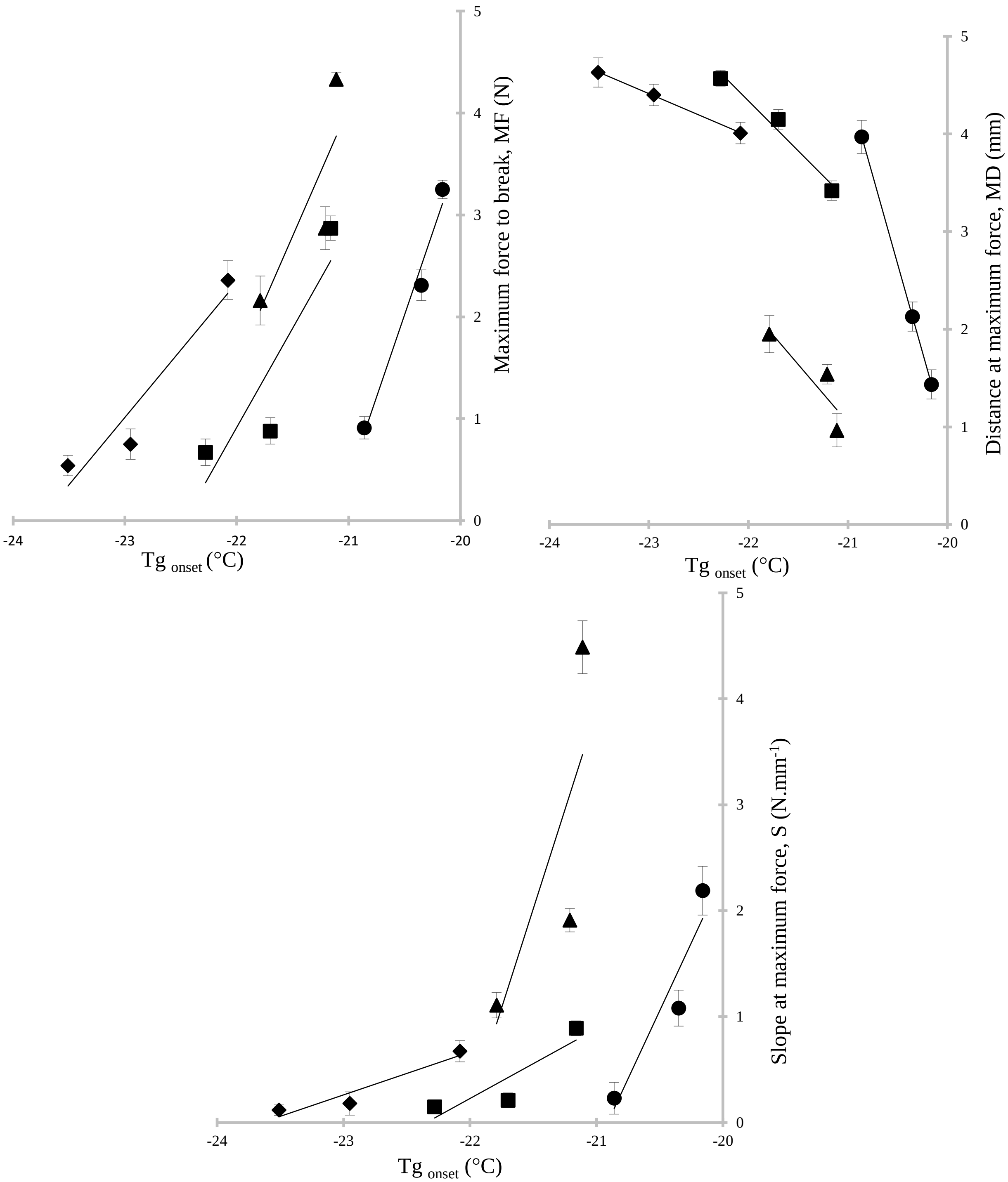


Figure 6.4: Plot of correlation between the glass-transition-temperature and mechanical textural attributes (hardness, ductileness, crispiness). NC (●) , RB (▲), RWB (■), and WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively.

Table 6.2 summarizes textural evolution of fried batter foods crust. From previously observed subzero Tg values, it is predictable that properties of batter-coated fried samples will change during post-fry holding at room environment (25°C) and under IR-heating (65°C). Hardness, brittleness, and crispness of fried coatings has changed within the post-fry holding duration of 30 minutes; where the nature of textural-changes were intertwined with batter-formulations. Hardness (MF*) and brittleness (MD*) of RB coated samples considerably increased while the change of its crispiness (S*) was not substantial. This could be the outcomes of "starch-retrogradation" event i.e., disaggregated amylose and amylopectin chains in gelatinized starch realign themselves to form more ordered structures, which is generally hard and brittle in nature (Wani et al., 2017). Hardness, brittleness and crispiness of WB batter coated fried samples changed to lower values. These changes might have been interconnected to the presence of high moisture in fried WB batter coated samples; as the loss of crispiness generally starts with the onset of mobility in water and small molecules (Kerr, 2016). To understand comparative textural differences between RB and WB batter coated sample; it could be mentioned that in food-biopolymers, glassy state forms earlier in starch-region compared to gluten-network (Kerr, 2016). And a food with high glass-transition-temperature is generally hard and brittle at ambient-temperature, in contrary, a low Tg means that at room-temperature the food will be soft and relatively elastic (Kasapis et al., 2007). Hardness, brittleness, and crispiness of batter-coated fried samples under IR-heating were seemingly slightly higher than their counterparts at room environment. These observation is analogous to the study of Rahimi et al. (2017).

Table 6.3 summarizes evolution of Tg and moisture content of fried foods crust. During post-fry holding, Tg values slightly changed but that weren't significant ($p<0.05$) and impact of IR-heating wasn't obvious. These observation are in close similarity with the study of (Rahimi et al., 2017). Earlier (Figure 6.4) it was observed that slight change in Tg is interwoven with drastic change in textural attributes. However, post-fry changes in Tg of fried batter coatings could be attributed to the changes in crust moisture (CrM). Presence of additional moisture (as consequence of mass-redistribution) in crust region might had enhanced the "plasticizing effect" of moisture and consequentially had resulted in slightly additional lower Tg value. It is notable, water-sorption can cause glass-to-rubber transition even at a constant temperature (Jothi et al., 2018; Kawai et al., 2014); and Tg of dehydrated cookies were influenced by water sorption (Sogabe et al., 2018).

Table 6.2: Normalized value of maximum force (MF*), normalized value of displacement at maximum force (MD*), and normalized value of slope (S*).

FT	Sample	MF*			MD*			S*		
		HD 0	HD 30		HD 0	HD 30		HD 0	HD 30	
			RE	IR-heating		RE	IR-heating		RE	IR-heating
2	NC	1.0	0.94±0.02	0.96±0.02	1.0	0.95±0.03	0.95±0.04	1.0	0.95±0.03	0.96±0.04
	RB	1.0	1.20±0.03	1.23±0.04	1.0	1.11±0.02	1.14±0.01	1.0	1.01±0.02	0.99±0.03
	RWB	1.0	0.81±0.02	0.85±0.03	1.0	0.88±0.03	0.90±0.03	1.0	0.81± 0.03	0.82± 0.03
	WB	1.0	0.80±0.04	0.83±0.05	1.0	0.86±0.04	0.89±0.05	1.0	0.79±0.04	0.81±0.05
4	NC	1.0	0.97±0.02	0.99±0.02	1.0	0.98±0.02	0.99±0.03	1.0	0.99±0.02	1.01±0.03
	RB	1.0	1.28±0.04	1.31±0.04	1.0	1.18±0.01	1.21±0.04	1.0	1.01±0.01	1.00±0.02
	RWB	1.0	0.86±0.03	0.88±0.02	1.0	0.91±0.02	0.93±0.02	1.0	0.82± 0.02	0.84± 0.03
	WB	1.0	0.83±0.05	0.87±0.05	1.0	0.88±0.03	0.89±0.04	1.0	0.80±0.03	0.82±0.04
6	NC	1.0	0.98±0.02	1.02±0.01	1.0	0.99±0.01	1.01±0.03	1.0	0.97±0.04	0.99±0.03
	RB	1.0	1.03±0.02	1.04±0.03	1.0	1.02±0.02	0.99±0.04	1.0	1.01±0.02	1.03±0.04
	RWB	1.0	0.93±0.03	0.95±0.02	1.0	0.93±0.04	0.92±0.02	1.0	0.91± 0.03	0.92± 0.01
	WB	1.0	0.88±0.05	0.90±0.04	1.0	0.90±0.03	0.92±0.03	1.0	0.88±0.04	0.88±0.04

WB, RWB, RB represents wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coating, respectively, and NC represent noncoated model food. HD: holding time (minutes), FT: frying time (minute).

Table 6.3: Glass-transition-temperatures (Tg) and crust moisture (CrM) content.

FT	Sample	Tg_{onset} (°C)			CrM (g/g dry matter)		
		HD 0	HD 30		HD 0	HD 30	
			RE	IR-heating		RE	IR-heating
2	WB	-23.51±0.32	-23.64±0.48	-23.58±0.53	0.47±0.03	0.61±0.04	0.58±0.03
	RWB	-22.28±0.21	-22.46±0.33	-22.33±0.26	0.38±0.02	0.47±0.02	0.44±0.02
	RB	-21.79±0.27	-21.83±0.27	-21.75±0.34	0.22±0.02	0.27±0.02	0.25±0.03
	NC	-20.86±0.31	-20.88±0.41	-20.95±0.51	0.33±0.03	0.37±0.03	0.38±0.04
4	WB	-22.95±0.31	-22.99±0.48	-22.93±0.54	0.45±0.03	0.56±0.03	0.53±0.04
	RWB	-21.70±0.22	-21.87±0.51	-21.76±0.42	0.33±0.02	0.43±0.02	0.41±0.02
	RB	-21.21±0.16	-21.25±0.20	-21.20±0.46	0.11±0.01	0.13±0.02	0.13±0.01
	NC	-20.35±0.11	-20.34±0.38	-20.28±0.51	0.17±0.02	0.20±0.02	0.19±0.03
6	WB	-22.08±0.34	-22.17±0.30	-22.14±0.44	0.39±0.03	0.43±0.03	0.41±0.04
	RWB	-21.16±0.27	-21.28±0.31	-21.20±0.25	0.25±0.03	0.34±0.04	0.30±0.03
	RB	-21.11±0.31	-21.20±0.16	-21.18±0.20	0.08±0.01	0.08±0.01	0.07±0.02
	NC	-20.16±0.15	-20.22±0.29	-20.19±0.28	0.13±0.02	0.13±0.02	0.12±0.03

WB, RWB, RB represents wheat-flour based batter, wheat & rice flour-based batter, and rice-flour based batter coating, respectively, and NC represent noncoated model food. HD: holding time (minutes), FT: frying time (minute)

6.5 Conclusion

Batter-formulations and frying time are the crucial parameters to control the glass-transition-temperature (Tg) of fried batter coatings. Tg of fried batter coatings are linearly correlated with the duration of frying. Batter-formulations and frying time influence the formation of internal microstructure, moisture-fat profile, and textural attributes (hardness, brittleness, crispness) of fried batters. Both moisture and fat acts as plasticizer in fried batter-coated foods, and results in negative Tg value. Positive correlations were observed between frying time, fat content, porosity and Tg, in contrast, moisture content are negatively correlated to these attributes. Crust moisture, texture and Tg of fried batter coated foods has changed during post frying holding, where external IR-heating has shown some extent of influence. Subzero Tg (°C) explains the moisture-redistribution induced textural changes in fried batter coatings during their post fry holding at room environment (25°C) and under external IR-heating (65°C). Based on observed results, it could be suggested that modification of moisture-fat profile and internal-structure through modification of batter-formulations and frying process would be an effective approach to obtain desired and stable textural qualities in batter-coated fried food foods. It is fundamentally important to understand the anti-/plasticizing effect of the ingredients that would be used in batter formulation, and this could be an aspect of future research.

CONNECTING TEXT TO CHAPTER 7

Chapter 3, 4, 5 and 6 successfully characterized the structural, thermal and physicochemical changes in deep-fat-fried batter coated foods. The evolution of aforementioned traits of parfried-frozen batter coated foods under different emerging finish-cooking process is crucial for achieving consumers satisfaction. In Chapter 7, comparative impacts of electromagnetic, air and fat frying on physicochemical, thermal and structural properties of parfried-frozen batter coated foods are investigated.

CHAPTER 7

COMPARATIVE ASSESSMENT OF ELECTROMAGNETIC, AIR AND FAT FRYING OF PARFRIED FROZEN BATTER COATED FOODS

7.1 Abstract

This study aimed to investigate the impact of batter-formulations and finish-cooking methods on physicochemical, thermal and structural evolution of parfried-frozen batter coated foods. Wheat and rice flour-based batters were used to coat a meat analog model food and were partially fried (at 180°C, 1 min) in canola oil, subsequently frozen (at -18°C) and stored for 1 week. Microwave heating (MH), infrared heating (IH) and air frying (AF) were used as alternate finish-cooking method of parfried-frozen products and were compared to deep-fat-frying (DFF). MH, IH, and AF reduced fat contents of parfried-frozen products compared to the DFF. Batter-coating considerably reduced fat uptake during DFF of parfried-frozen products, where the extent of fat-reduction was intertwined with batter-formulations. Finish-cooking loss (%) of parfried-frozen foods and moisture profile of finish-cooked products are impacted by both the batter-formulations and finish-cooking methods. Evolution of key quality attributes (texture, color) of parfried coated-product was allied with batter-formulations and finish-cooking methods, wherein AF is found as a suitable substitute of DFF. Low-moisture and high-fat containing parfried-frozen (rice-flour based) batter has develop light-yellowish color and crispy crust during finish-cooking. Post finish-cooking stability of the quality attributes are linked with batter-formulations and finish-cooking method. Glass-transition-temperature (Tg) of finish-cooked products were found in sub-zero temperature zone, where the Tg values has impacted by batter formulations and finish-cooking methods. ATR-FTIR spectroscopy and scanning electron microscopy (SEM) analysis revealed, surface' structural-chemical evolution of parfried-frozen batter coated product were impacted by both batter-formulation and finish-cooking method.

7.2 Introduction

Though fried foods are popular worldwide, the presence of high amount of fat in fried product is a major health concern. The use of fat-reduction technology is unquestioned, to produce less-fat containing fried product. In this circumstance, food surface modification by batter-coatings is an effective strategy, as the frying is majorly a surface phenomenon and batter-coating creates uniform layer over food surface that influences heat and mass transfer process during frying (Devi et al., 2020; Rahimi & Ngadi, 2014). Batter coating not only impacts fat-content of fried foods but also develops a peculiar texture (juicy inside, dry outside); and textural characteristic of coated-product is crucial for achieving its consumer satisfaction (Rahimi et al., 2017). It is notable, surface color is very crucial for any food items as this is the quality parameter that is evaluated by consumers even before the food enters the mouth.

Par-frying is a unit operation where foods are partially cooked by heating in edible oil at high temperature to inactivate enzymes and microbes (Raj *et al*., 2016). Parfried foods are generally stored as frozen and require finish-cooking for consumption (Rady *et al.,* 2019; Adedeji & Ngadi, 2018; Raj *et al*., 2016). Parfried frozen-foods are finish-cooked (heate up) mainly by deep fat frying (DFF), wherein two successive frying (par-frying & finish-frying) arises serious health concern; as the DFF results in the presence of excessively high amount of fat in finish-cooked product (Adedeji & Ngadi, 2018). In these scenarios, use of alternate (other than DFF) heat treatment as finish-cooking method could be a pragmatic approach in reducing total fat content of finish-cooked product. To this purpose, emerging heat processing techniques such as microwave (MW) heating, infrared (IR) heating, and convective air frying could be considered as alternate finish-cooking method.

Electromagnetic spectrum between the frequencies of 300MHz and 300GHz is represented as microwave, MW(Singh & Heldman, 2009). MW heating possess many advantages such as rapid temperature generation, shorter treatment time, improving product uniformity (Soorgi *et al*., 2012; Ngadi *et al*. 2009). Infrared (IR) is part of electromagnetic spectrum in the range of 0.5-100 μm, and IR is absorbed by food compounds which results in temperature rise (Rahimi *et al*., 2018). Air frying (AF) is an emerging cooking technique where hot air is used as heat transfer medium instead of edible oil (Cao *et al*., 2020). It has been reported in several studies (Cao *et al*., 2020; Sansano et al., 2018) that microwave and air frying results in less oil-deterioration and reduce acrylamide (human carcinogen) formation in products. However, the effects (as finish-cooking method) of electromagnetic (MW, IR) heating and air frying on parfried frozen batter coated food processing, is not yet investigated. In addition, scientific literature about post-finish-cooking stability of the quality attributes of coated products, are still rare.

The overall goal of this study was to (i) assess the effect of electromagnetic (MW, IR) heating and air frying as alternate finish-cooking methods of parfried frozen-batter coated product (ii) investigate the impact of batter-formulation on quality evolution during finish-cooking of parfried frozen-batter coated product (iii) study the effect of batter-formulations and finish-cooking methods on post-finish-cooking stability of physicochemical attributes.

7.3 Materials and Methods

7.3.1 Materials

Soy protein isolate was purchased from MP Biomedicals (29525 Fountain Pkwy, OH, USA). Wheat gluten was supplied by Sigma-Aldrich Co. (Ontario, Canada). Methylcellulose (MC) and Sodium pyrophosphate ($Na_4P_2O_7$) crystals were received from Fisher Scientific (Fair Lawn, New Jersey, USA). Sodium bicarbonate ($NaHCO_3$) was supplied by Church & Dwight Canada Corp. (Ontario, Canada). Rice flour (Suraj®, composed of 6.67 % protein, 83.32% carbohydrate, 1.33 % fat and 8.68% moisture) was procured from a grocery store in Montreal, Canada. Commercial wheat flour (Five Roses®, composed of 13.33 % protein, 73.33% carbohydrate, 3.33% fiber, 1.33 % fat and 8.68% moisture), NaCl (Sifto, Compass Minerals Canada Corp.) and Canola oil (Sans nom®, Loblaws Inc.**)** were purchased from a store in Sainte Anne de Bellevue, Canada.

7.3.2 Sample preparation

Meat analog model food was formulated with soy protein isolate (SPI), wheat gluten (WG), distilled water (DW) and canola oil (CO). At first, wheat gluten was mixed with fresh canola oil and as soon as homogeneous slurry was obtained, soya protein isolate was added and mixed thoroughly. Then water was added and mixed properly until a very soft-textured dough was developed. Respective proportion (g) of the ingredients WG: CO: SPI: DW were 12:15:22:73. Prepared dough was used to fill individual rectangular-shaped cavities of silicon mold (Freshware CB-115RD 24-Cavity Silicone Mini Mold). Covered silicon mold was held in complete horizontal position on top of a flat metal wire surface and heated for 15 min at 70°C in a water bath. After heat treatment, mold was allowed to reach equilibrium with room temperature. After that, gentle pressure (by finger) was applied from the back of each cavity to bring out the formed substrates without any fracture. This substrate was used as model food, weighing an average weight of 16±0.2

g/substrate and having a uniform dimension of 5.6 cm x 2.5 cm x 1.3 cm. The prepared model-food was packed in zip lock bags and stored in refrigerator (4±2°C) for 24 hours, before its use.

To prepare batter coatings: crystals of Sodium pyrophosphate was grinded to obtain as fine powder. Then Sodium bicarbonate (1.4%), NaCl (1.5%), and Methylcellulose (0.3%) were added and mixed properly with Sodium pyrophosphate powder (1.8%), to form a homogeneous powder mix. The powder-mixture was further thoroughly mixed with dry flour in a proportion of 95% (flour) to 5% (powder mix). Distilled water (at chilled condition) was added gently to the solid ingredients with a total solid to water ratio of 1:1.3 and mixed them properly until a homogenous tempura-batter slurry was obtained. Rice based batters (RB) were formulated using only rice flour whereas wheat four based batters (WB) were formulated with wheat flour. Equal proportion of flours were used to formulate rice and wheat flour based batters (RWB). The prepared batter system were kept at room environment for 5 min before its use to coat the model food. The model food was brought out of refrigerator and kept at room environment for 30 min, prior to batter coating. It was fully immersed in batter system for 1 minute and then removed from batter solution to make a complete coat. The coated product was held on a kitchen-fork for about 30 sec, to drain the excess amount, in order to obtain a uniform layer of coating around model food core. Non-coated (NC) model product was used as control sample.

A programmable deep fat fryer (T-fal Compact, FF122851, made in China) filled with 1.5 L of fresh canola oil was used to partially fry the sample. The oil was preheated for 1hr and were stirred to minimize any variation in oil temperature. Samples were uninterruptedly fried for 1 min at 180°C, maintaining a sample to frying oil ratio of 1:30. At the end of par-frying, the frying basket was removed from oil and shaken 5 times; and the samples were allowed to cool at room environment. Par-fried samples were packed in Ziplock bag and kept at -18°C for a week and frozen samples were used for finish-cooking without any prior treatment.

7.3.3 Finish-cooking

Microwave heating (MH), infrared heating (IH) and air frying (AF) were used as alternate finish-cooking methods, while deep-fat-frying (DFF) as reference finish-cooking method. The internal core-temperature of samples were considered as the critical parameter for configuring operational settings of the finish-cooking methods. Finish-cooking methods were aimed to achieve a temperature ≥75°C at the coldest point (at geometric center) within the samples. After each finish-cooking treatment, samples were in rest at room environment for 1 min, before further studies.

Microwave heating (MH): par-fried frozen samples were heated by a microwave facility (Hamilton Beach, EM720CPN, China) at 2450 MHz/700W. Actual microwave output power was 640W, as determined by IMPI 2-L test (Kang & Chen, 2015). Sample was placed on the center region of high absorbent paper based (Royale®, Canada) sandwich like system, where six individual layers of absorbent paper were closely attached with each side of the samples. The whole system was placed on a dry and horizontally rotating (360°) glass structure in microwave treatment chamber. MH system was programed to provide uninterrupted heating for a duration of 1.5 min, where sample was exposed to the microwave power density (PD) of ~25W/g. Microwave PD was computed based on the measured initial weight of sample and the actual power output of microwave heating (Kang & Chen, 2015; Adedeji *et al.*, 2009; Ngadi *et al.*, 2009).

Infrared heating (IH): a programmable infrared facility (Toast-R-Oven, TO1380SKT, China) was used in performing the infrared heating (IH) of parfried frozen samples. The frozen parfried sample were placed on a stainless-steel sample holder (39×15.5 cm) and the holder was placed in between of two cylindrical IR-lamps (1150W). The distance between sample and IR-lamps were adjusted to 8.9 cm and the sample were exposed to a heat flux of ~1.9 w/cm^2. To achieve and maintain constant heat flux, IR-heating unit was turned on 5 min before starting the finish-cooking treatment (Rahimi *et al.*, 2018). The IH system was programmed to maintain a constant temperature of 180°C for a total duration of 15 min, where the sample was flipped over at halfway.

Air frying (AF): a programable air fryer unit (Philips HD9240/90, China) were used for air frying (AF) of parfried-frozen samples. To reach a thermal equilibrium, AF unit were turned on for 5 min before its use to fry the parfried-frozen sample. Frozen-parfried sample was placed within the bucket (on top of were mesh) of AF unit. The AF unit was programmed to fry the sample at a constant temperature of 180°C for an uninterrupted total duration of 15 min. There was no need to rotate/flip over the sample during AF, since hot air covered all parts of the sample's surface. At the end of AF, samples were removed from frying basket and used for further studies.

Deep-fat-frying (DFF): deep fat frying (DFF) of parfried frozen samples were performed at atmospheric pressure, in a programmable fryer (T-fal Compact, SERIE F53-S3, China) filled with 1.5 L fresh canola oil. To maintain the set temperature of frying oil the DFF unit was turned on 1 hour before frying experiment. Parfried frozen samples were fried at 180°C for 3 min, maintaining a sample to frying-oil ratio of around 1:30. It was carefully monitored that the samples were fully immersed in oil, during the full length of DFF. At the end of DFF, frying basket were removed from oil and shaken for 5 times, and fried samples were used for further studies without any post-fry treatment. Each batch of used frying oil was replaced with a new batch of fresh oil, after thirty minutes of frying.

7.3.4 Process parameters

Batter pick-up (BP) was calculated to denote the amount of raw batter that adhered to model food. BP was calculated by dividing the weight difference (weight after model food coating–weight before model food coating) divided by the weight before model food coating. The result was multiplied by 100 and express as percentage (%) of raw-model food.

Par-frying loss (PL) was calculated by dividing the weight difference (weight of coated food before par frying- weight of coated food after par frying) divided by the weight of coated food before par-frying; and express as % of coated non-parfried food.

In order to determine finish-cooking loss (F_{CL}), parfried sample was weighed before finish-cooking (Wi) and after finish-cooking (Wf). F_{CL} (%) was calculated ($F_{CL}= \frac{Wi-Wf}{Wi}$X100) according to the method of Rahimi *et al*., (2018).

7.3.5 Physicochemical and thermal properties

Textural attributes were evaluated by puncture/compression test, using a mechanical texture analyzer (Stable Micro Systems Texture Analyzer, TA. HD PlusC, UK). Test samples were individually mounted on a flat rigid support. A puncture probe TA-52 (2 mm dia.) and 500 N was used to punch test sample, at a constant test speed of 1mm/s and a travel distance of 5 mm.

Puncture test were performed on three equidistance locations of the crust. Following the methodology of Rahimi *et al*., (2017), the maximum force to break (N), displacement at maximum force (mm), and slope (N/mm) at maximum force of the puncture test data were chosen as parameters to evaluate textural properties namely hardness, brittleness, and crispness, respectively. Puncture test data were analyzed by TA. HD PlusC program (Exponent 6.1.14 v, Stable Micro Systems, UK).

Moisture content was grouped into two categories namely, crust moisture (CrM) and total moisture (TM). CrM was defined as the moisture that was present in outer crust region, and TM includes both the core and crust moisture. Crust region of finished-cooked sample were carefully separated from core region, by a microtome blade (Feather C35, Japan). Detached portion was freeze dried in a freeze-dryer (Modulyod-115; Thermo Savant, USA.) at -50°C and 250 mbar for 48 hours. Weight of the sample before and after freeze-drying was measured and moisture content was calculated on dry basis (g/g dry matter), by dividing the mass of moisture with the mass of the freeze-dried sample. Separately, some samples were freeze dried without separating the crust from core, to measure the total moisture.

Freeze-dried samples were grounded to increase surface to volume ratio, and an amount of 3-5 g was placed in thimbles of a VELP SER 148 solvent extraction unit (Velp Scientifica, Usmate, Italy). Fat was extracted by following the Randall method, with petroleum ether at 130°C pre-set temperature. Complete fat extraction process was performed in three consequent steps: immersion (30 min), washing (30 min), and recovery (30 min), totaling of 90 min. Weight of extracted fat was measured, and fat content was computed on dry basis (g/g dry matter) by dividing the mass of extracted oil with the mass of the freeze-dried sample.

To measure crust thickness (Ct), a cork-borer was used to obtain flat portions from the detached crust, and Ct was measured by a calibrated digital slide-caliper.

Color properties were assessed by using a spectrophotometer (Minolta Spectrophotometer CM-3500d, Japan). Color was determined in CIELab space (Illuminant D65, 10° viewing angle) from the reflection spectra between 400-700 nm. Color parameters L (lightness-darkness), a* (redness-greenness), and b* (yellowness-blueness) was estimated at room environment (25°C) and total color difference (ΔE) was calculated as follows:

$$\text{Total color difference, } \Delta E = [\Delta L^2 + \Delta a^2 + \Delta b^2]^{1/2} \qquad 7.1$$

where, $\Delta L = L_{pf} - L_{fc}$, $\Delta a = a_{pf} - a_{fc}$, $\Delta b = b_{pf} - b_{fc}$, and $_{pf}$ is parfried, and $_{fc}$ is finish cooked.

Glass transition temperature (Tg) of the detached (by microtome blade) crust of finish-cooked samples were determined by using a differential scanning calorimeter (DSC Q250, TA Instrument, USA). Tg of samples were determined by following the method of Rahimi *et al*., (2017) and in brief: around 10-15 mg sample was placed in sample pan, a lid was attached, and a mechanical device was used to make them as hermetically sealed. One sample holder with lid (but without crust sample) was used as reference pan. In DSC measurement chamber, sample was rapidly cooled to -40°C at a cooling rate of 20°C/min, held isothermally for 2 min, and then heated to +40°C at a rate of 2°C/min. DSC data were recorded and analyzed by DSC-Trios 5.1.1v software (TA Instruments, USA). From DSC thermogram, Tg were obtained as onset of the heat capacity change and were represented as Tg_{onset}. The Tg_{onset} was obtained as the change of the baseline slope, after a variation of heat flux occurred, and determined as intersection of tangents to the transition curve.

7.3.6 ATR-FTIR Spectroscopy

Attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectrometer (Nicolet iS5 FTIR, Thermo Fisher Scientific, USA) were used to record the FTIR-spectra of the surfaces of par-fried sample. The operation of spectrophotometer, acquisition, and manipulation of spectra were performed by Omnic software (Nicolet 6.1v., USA). Outer surface of the samples were in direct contact with ATR-crystal and spectra acquisition was at control ambient temperature at 25°C. ATR-crystal was cleaned with ethanol to remove any residual (especially fat) by the previous sample. FTIR spectra were collected over the wavenumber range of 4000–950 cm^{-1} by co-adding 32 scans and at resolution of 4 cm^{-1}. All spectra were normalized against background of air spectrum. After every 1 hr of operation, a new reference air background spectrum was taken. The spectra were recorded as absorbance values. According to Chen *et al*. (2015) and our preliminary study, absorption peaks at around 2922 cm^{-1} and 2852 cm^{-1} (asymmetrical and symmetrical stretching of $–CH_2$), 1743 cm^{-1} (C=O stretching), 1157 cm^{-1} (-C-O stretch; -CH_2 bending) of the FTIR spectra were considered as surface fat response i.e., presence of frying canola oil on crust surface. In this view, considering pertinent studies (Khudzaifi, et al., 2020;

Rohman *et al*., 2016; Tarhan *et al*., 2017; Li-Chan *et al.*, 2006), absorption area under the spectral ranges between 1130-1200cm^{-1}, 1425-1475cm^{-1}, 1700-1775cm^{-1}, and 2800-3000 cm^{-1} were calculated by Omnic software and their summed value was reported. According to Nicolaisen (2009) and Efimov *et al*. (2003), a broad peak around 3000–3700 cm^{-1} region was due to the components of asymmetric and symmetric stretching modes of the H_2O molecule, and the absorption area of the FTIR spectra within this range as well as absorption between 1525-1725 cm^{-1} region were considered as surface moisture response.

7.3.7 Scanning electron microscopy

Scanning electron microscopy (SEM) were used to assess surface morphology. Surface washed (by three consecutive charges of petroleum ether, for a total immersion period of three minute) par-fried and finish-fried samples were freeze dried and used for SEM imaging by a scanning electron microscope (Hitachi TM3000, Japan). SEM operational procedure of Adedeji & Ngadi (2018) were followed with required modifications, as such: 5 kV electron power, auto contrast function, and composition mode of imaging were used. Small cut, rectangular shaped, defatted sample of about 10×10×5 mm in dimension was placed on sample base with a sticky surface that comprises of carbon tab. After each sample was properly aligned by visual control using two sets of knobs and images shown in the control software on a computer screen, vacuum pressure was created in the sample chamber and images were quickly acquired to prevent charging and heating that could lead to artifacts in images. The SEM images were acquired at a magnification of ×30 and recorded as 8-bit joint photographic experts group (JPEG) file. Fractal dimension (FD) of surfaces were estimated from 2D SEM images by box-counting method, as detailed by Rahimi & Ngadi (2016).

7.3.8 Statistical analysis

All experimental data obtained from triplicate of samples, mean ± standard deviation was reported. Tukey's HSD (honestly significant difference) was used in reporting significant ($p<0.05$) difference among means. Statistical analyses were performed by a licensed statistical software (JMP 14.1v, SAS Institute Inc., NC, USA).

7.4 Results and Discussion

Figure 7.1 depicts, process parameters and attributes of par-fried samples. Average batter pickup (BP) of model food substrate were 71.3 % for wheat-flour based batter (WB) and 35.3% for rice-flour based batter (RB). Lower (6.51<11.37%) par-frying loss (PL) and concomitant higher (0.53>0.27 g/g dry mass) crust moisture (CrM) and higher (1.23>1.03 g/g dry mass) total moisture (TM) was in WB coated sample; whereas higher (0.24>0.09 g/g dry mass) crust fat (CrF) and higher (0.29 >0.19 g/g dry mass) total fat (TF) were found in RB coated sample. These differences could be explained as such: wheat-flour contains higher amount of protein (e.g., gluten) that can form 3D network during frying and prevents moisture loss and fat uptake; and methylcellulose was in favor of higher viscosity and moisture retention ability of wheat flour based batter (Adedeji & Ngadi, 2011; Rahimi & Ngadi, 2014). These properties had resulted in lower moisture loss from WB coated sample and consequent lower fat absorption during par-frying. PL, CrF, and TF of batter-coated parfried samples were significantly ($p<0.05$) lower than non-coated (NC) sample.

Finish-cooking loss (F_{CL}) of parfried-frozen samples were greatly dependent on their cooking methods as such: for identical sample, lowest F_{CL} were observed for conventional deep-fat-frying (DFF). Among alternate finish-cooking methods, highest F_{CL} was observed for microwave heating (MH) followed by air frying (AF), and lowest F_{CL} were found for infrared heating (IH). These disparities could be explained as such: DFF is a simultaneous heat and mass transfer process where samples have lost their moisture and uptakes frying-oil, which minimized the overall weight differences and were reflected as lower F_{CL}. Under IH, MH and AF treatments, parfried-frozen samples had only lost their moisture without up-taking any oil, as consequence higher F_{CL} were observed. Contribution of oil leaching out (as drip-loss) in favor of the weight loss of parfried-frozen samples, could also be a probable cause of higher F_{CL}. At frozen temperature (-18°C) the frying-oil (absorbed during par-frying) was in solid state and were strongly attached to solid-matrix of parfried samples. Under heat treatments, with the raise in frozen-samples' temperature the adhered oil melts and could start to flow; as viscosity of oil reduces with the increase of temperature (Fasina & Colley, 2008). Highest F_{CL} under MH might be due to its rapid heat generation, where the generated heat had moved out inner moisture & fat components, which was subsequently removed by absorbent-paper arrangements. F_{CL} under microwave-heating could be controlled by modifying power density (PD) i.e., sample volume to actual microwave power. Compared to IH, higher finish-cooking loss under AF treatment might be due to the intense removal of moisture & oil by high velocity convective hot air, along with the drip-loss phenomenon. Higher F_{CL} were observed for WB coated samples, compared to RB coated samples. This could be linked with the presence of higher moisture in WB coated parfried sample; as during finish-cooking of parfried-frozen samples the probable source of weight reduction was the loss of flowable components (e.g., moisture, fat). For the studied samples and methods of cooking, observed finish-cooking loss of batter-coated parfried frozen samples were < 20%. Under IR-treatments a weight loss of up to 27.3% has been reported (Rahimi *et al.*, 2018).

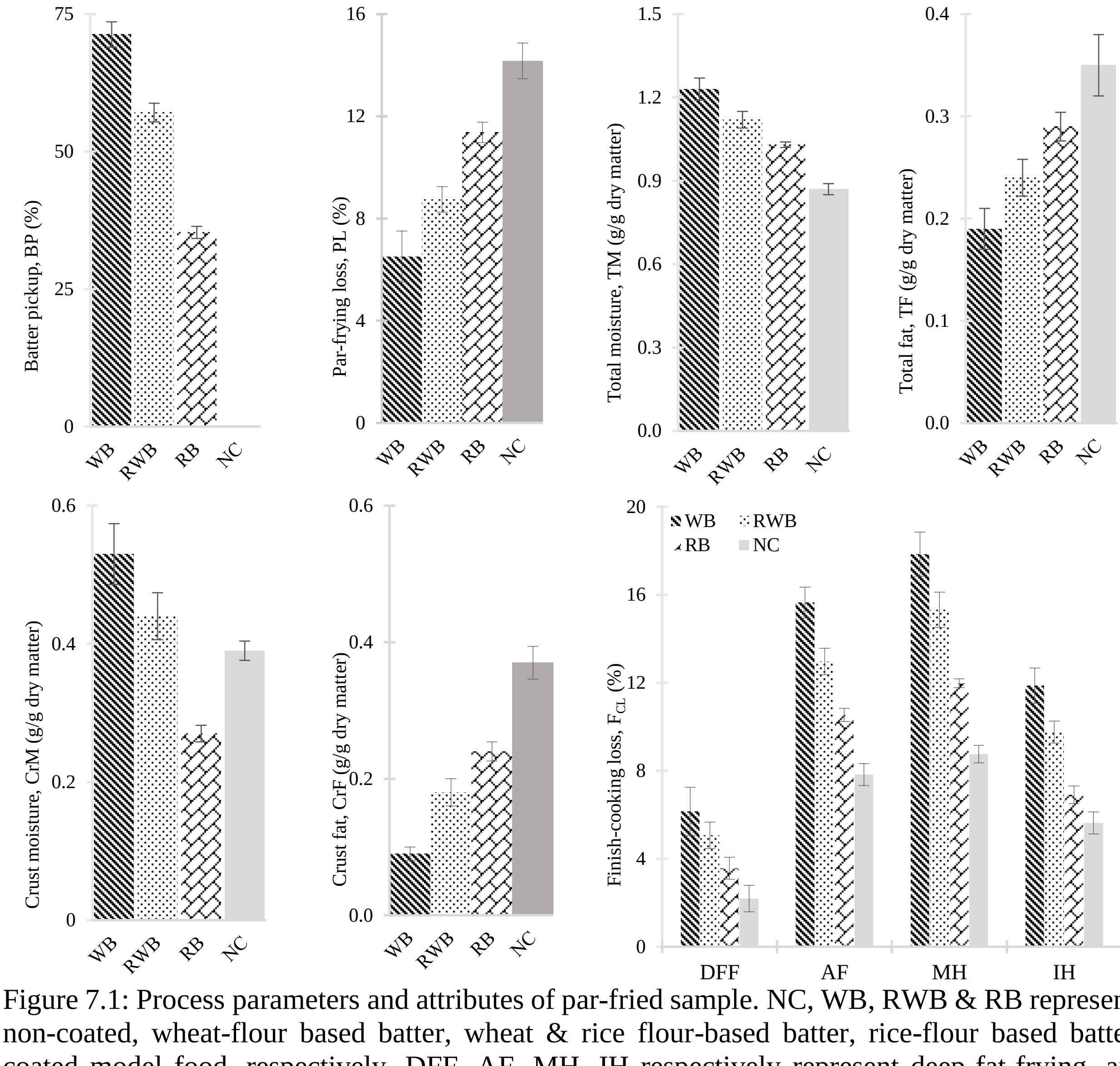


Figure 7.1: Process parameters and attributes of par-fried sample. NC, WB, RWB & RB represent non-coated, wheat-flour based batter, wheat & rice flour-based batter, rice-flour based batter coated model food, respectively. DFF, AF, MH, IH respectively represent deep-fat-frying, air frying, microwave heating, and infrared heating.

Figure 7.2 depicts moisture-fat profile of finish-cooked products. Compared to DFF, both the crust fat (CrF) and total fat (TF) content of alternate finish-cooked (by IH, MH, AF) samples were significantly ($p<0.05$) lower. MW, IH and AF could be considered as a healthier alternative of deep-fat-frying (DFF), to cook parfried-frozen products. IH, MH, and AF treatments weren't involved with any external oil addition, hence, the fat profile of alternate finish-cooked samples were either similar or lower than the fat content of their respective parfrying stage. Presence of lower fat (compared to parfried samples) in finish-cooked product indicates oil leaching out as drip loss (Cao *et al*., 2020; Rahimi et al., 2018). Higher fat profile for DFF was associated with the loss of moisture from parfried-frozen samples and consequent oil uptake during finish-cooking in oil. In DFF use of a suitable oil/oil blends might be a way to reduce fat content in finish-cooked product; as the degree of saturation of frying oil could affect the fat content of fried products (Ngadi et al., 2007). In DFF of parfried-frozen samples, significantly lower fat were observed in batter-coated samples in comparison to noncoated model food. This indicates, even during finish-cooking (by DFF) of parfried-frozen products, batter-coating successfully impacts the mass-transfer process. Among batter systems, fat content was lowest in WB coated sample which is attributed to higher water retention and fat-update prevention ability of wheat-flour based batter.

Moisture profile of parfried-frozen samples were impacted by finish-cooking methods, and extent of moisture loss from parfried-samples were interlaced with both the finish-cooking methods and batter-formulations. Lower crust moisture (CrM) were observed for DFF of parfried-frozen samples. Higher moisture loss at DFF is attributed to its higher rate of heat transfer; as frying is a simultaneous heat and mass transfer process where mass-profile is changed by the application of heat (Cao et al., 2020). Among alternate methods, higher extent of total moisture (TM) loss were found for MH, followed by AF and lower extent of moisture loss was for IH. Higher extent of moisture loss at MH was not surprising. The use of microwave power as a pre-treatment method to reduce initial moisture of samples has been reported in literature (Ngadi *et al*., 2009; Adedeji *et al*., 2009). For identical sample, moisture in crust region of finished-cooked food of DFF and AF were quite similar. IH was in favor of the presence of high amount of moisture in crust region.

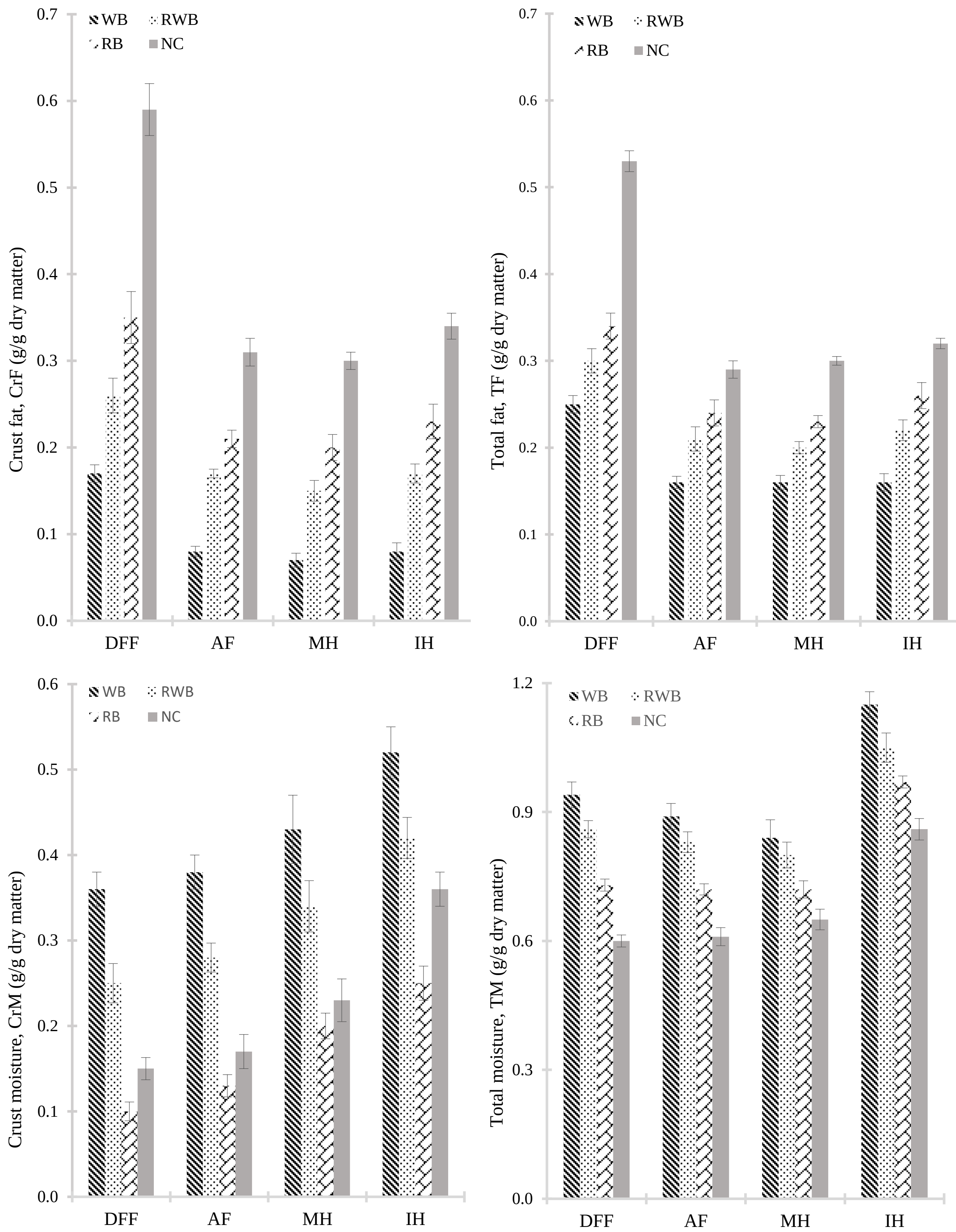


Figure 7.2**:** Moisture-fat profile of finish-cooked products. NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. DFF, AF, MH, IH respectively represent deep-fat-frying, air frying, microwave heating, and infrared heating.

Figure 7.3 depicts, texture profile and crust thickness of finish-cooked products. Hardness (MF), ductileness (MD) and crispiness (S) of finish-cooked samples greatly impacted by finish-cooking methods. Higher values of MF and S with lower values of MD of the samples for DFF and AF represents, both the deep-fat-frying and air-frying has produced crust having hard, brittle and crispy texture. These suggests, to develop desired textural attributes with lower fat in finish-cooked products, AF could be used as a suitable alternate finish-cooking method against conventional DFF. It is notable, applied cooking time of AF were (15 min) considerably higher than (3 min) the duration of DFF treatment. As AF treated samples had contained considerably lower fat, thus air-frying could be used as a healthier alternate of deep-fat-frying in preparing parfried-frozen batter coated product. In the same context, both MH and IH could also be considered as a healthier alternative of deep-fat-frying. In case of IH and MH, the modification of process parameters such as: longer processing time (for IH), use of susceptor materials (for MH), etc. could be considered to achieve desired textural attributes in finish-cooked products. Textural changes that occurs in foods during cooking are mainly due to the evaporation of water, protein denaturation, and starch gelatinization (Cao *et al*., 2020). In harmony with this phenomenon, a very similar moisture profile was observed while parfried-frozen samples were finish-cooked by DFF and AF (Figure 7.2). Presence of higher moisture in crust region of IH and MH treated samples might be the prime cause behind their lower crispy texture; as moisture and crunchiness of foods are generally negative correlated (Rahimi et al., 2017). Compared to IH, the MH produced hard crust, but that was not brittle and crispy. As moisture removal from parfried-frozen samples were higher in MH, therefore a hard crust was developed as the consequential result of the presence of lower moisture. However, crispiness development in battered product might not solely dependent on their moisture content, wherein their spatial distribution and overall microstructure might be influential contributors. Textural attributes of finish-cooked coated products has shown strong relation with batter-formulations as such: for identical finish-cooking method, hardness, brittleness, and crispiness of RB coated samples were considerably higher than WB coated samples. This disparity are associated with flour-composition of batter-systems and final-moisture in crust region. This could be understood as such: starch content of parfried-frozen batters gelatinizes under heat treatments (during finish-cooking), while the samples were removed from finish-cooking units, with the lowering of samples temperature it starts becoming hard, might be due to the occurrence of starch retrogradation, and starch (carbohydrates) content was higher in rice-flour.

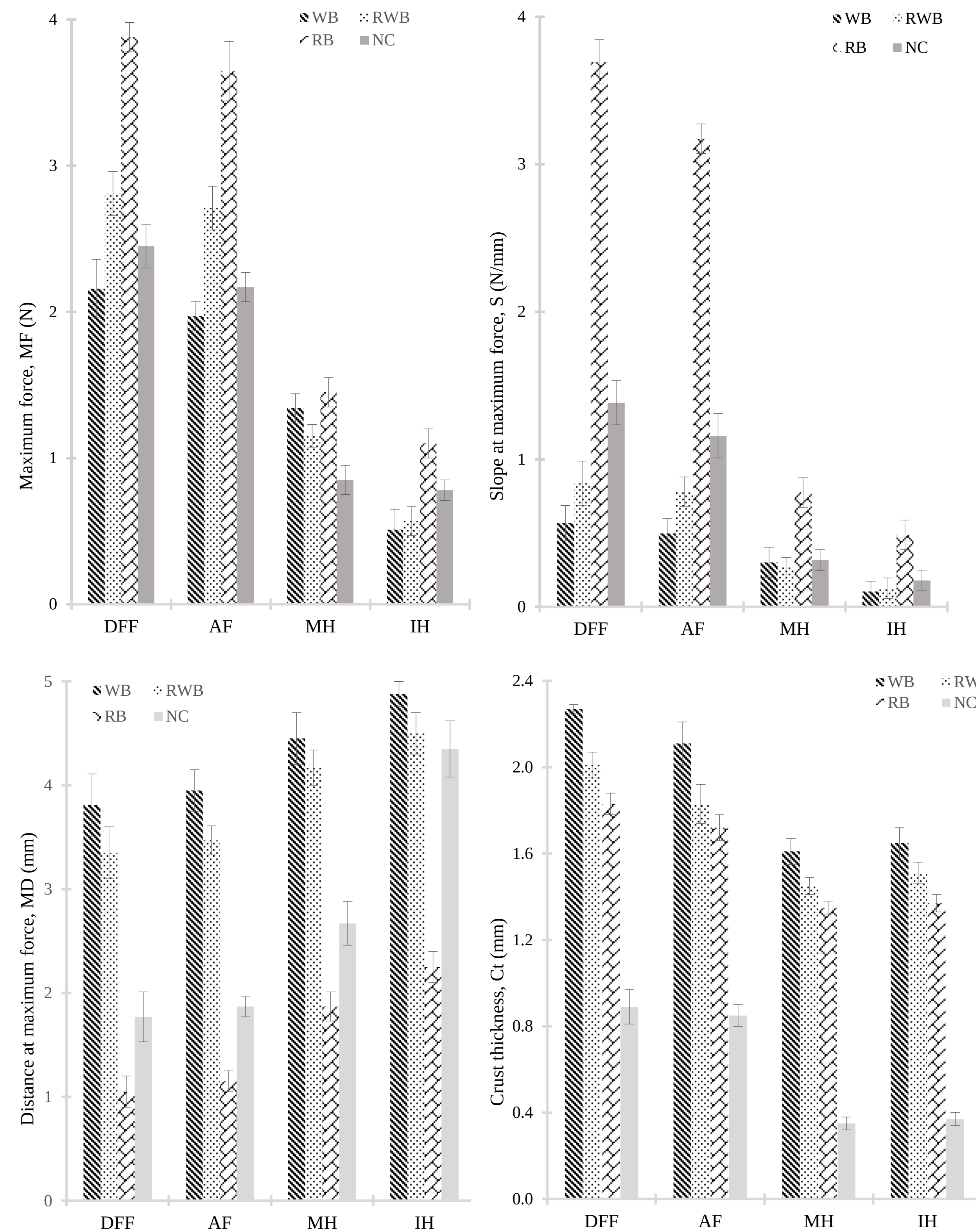


Figure7.3: Textural and structural profile of finish-cooked samples. NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. DFF, AF, MH, IH respectively represent deep-fat-frying, air frying, microwave heating, and infrared heating.

Crust thickness (Ct) of DFF and AF treated samples were like each other, while IH and MH treated samples were in close similarity. This indicates, during finish-cooking, effective outer boundary layer (which produces crust) changed with cooking methods. This changes were batter-formulation dependent, where wheat-flour based batter has produced a thicker crust and rice-flour based batter produced a thinner crust. This property can explain their others physiochemical properties as such: thicker WB crust was in favor of higher moisture retention and prevented fat uptake, while due to presence of lower moisture the RB-crust were crispier. Therefore, modification of the formation of crust structure via modification of batter formulation could be a way to develop desired textural properties in finish-cooked batter coated products.

Table 7.1 summarizes post-finish-cooking moisture and texture attributes. At post-finish-cooking stage, an increasing trend in crust-moisture were observed although total-moisture was unchanged. The extent of change in CrM was intertwined with batter-formulations and finish-cooking methods. Evolution in CrM indicates moisture re-distribution between spatial region (from high moisture containing zone, to low moisture region), and increase in CrM is attributable to the moisture migration from juicy-core region of finish-cooked samples. Post-finish-cooking moisture redistribution might be the continuous effect of finish-cooking, as moisture migration in product can continues (due to vapor pressure difference) even after removal from cooking unit (Shokrollahi Yancheshmeh *et al.*, 2019). Higher affinity of increase in CrM were observed for IH treated samples. This could be linked with their higher initial total moisture (TM) content, as moisture re-distribution in food is generally a diffusion-controlled phenomenon (Singh & Heldman, 2009). Batter-formulation has shown influence on post-finish-cooking moisture redistribution behavior as such: higher extent of moisture redistribution were observed in WB coated samples, compared to RB coated samples. This could be attributed with moisture absorption affinity of fibers, as good amount of fiber was present in wheat-flour in compared to rice-flour.

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Table 7.1:Post finish cooking evolution of moisture and texture profile.

Finish-cooking	Sample	Moisture content (g/g dry matter)				Textural attribute (normalized)					
		CrM		TM		MF*		MD*		S*	
		0 min	30 min	0 min	30 min	0 min	30 min	0 min	30 min	0 min	30 min
DFF	WB	0.36 ± 0.03^{a}	0.40 ± 0.02^{a}	0.94 ± 0.03^{a}	0.92 ± 0.02^{a}	1.0^{A}	0.87 ± 0.03^{B}	1.0^{A}	0.90 ± 0.02^{B}	1.0^{A}	0.92 ± 0.03^{B}
	RWB	0.25 ± 0.02^{b}	0.28 ± 0.03^{b}	0.86 ± 0.02^{b}	0.85 ± 0.03^{b}	1.0^{A}	0.90 ± 0.02^{B}	1.0^{A}	0.92 ± 0.03^{B}	1.0^{A}	0.94 ± 0.01^{B}
	RB	0.10 ± 0.01^{d}	0.11 ± 0.02^{d}	0.73 ± 0.01^{c}	0.73 ± 0.02^{c}	1.0^{B}	1.07 ± 0.02^{A}	1.0^{A}	1.02 ± 0.02^{A}	1.0^{A}	1.01 ± 0.02^{A}
	NC	0.15 ± 0.02^{c}	0.17 ± 0.02^{c}	0.61 ± 0.01^{d}	0.61 ± 0.02^{d}	1.0^{A}	0.98 ± 0.02^{A}	1.0^{A}	0.98 ± 0.02^{A}	1.0^{A}	0.97 ± 0.02^{A}
AF	WB	0.38 ± 0.03^{a}	0.43 ± 0.03^{a}	0.89 ± 0.02^{a}	0.88 ± 0.04^{a}	1.0^{A}	0.85 ± 0.03^{B}	1.0^{A}	0.89 ± 0.02^{B}	1.0^{A}	0.91 ± 0.03^{B}
	RWB	0.28 ± 0.02^{b}	0.32 ± 0.02^{b}	0.83 ± 0.02^{b}	0.81 ± 0.02^{b}	1.0^{A}	0.89 ± 0.02^{B}	1.0^{A}	0.91 ± 0.03^{B}	1.0^{A}	0.93 ± 0.02^{B}
	RB	0.13 ± 0.01^{d}	0.14 ± 0.03^{d}	0.72 ± 0.01^{c}	0.70 ± 0.03^{c}	1.0^{B}	1.09 ± 0.03^{A}	1.0^{A}	0.98 ± 0.02^{A}	1.0^{A}	0.98 ± 0.02^{A}
	NC	0.17 ± 0.03^{c}	0.29 ± 0.02^{c}	0.61 ± 0.02^{d}	0.60 ± 0.02^{d}	1.0^{A}	0.96 ± 0.03^{A}	1.0^{A}	0.97 ± 0.03^{A}	1.0^{A}	0.96 ± 0.03^{A}
MH	WB	0.43 ± 0.05^{a}	0.46 ± 0.04^{a}	0.84 ± 0.03^{a}	0.82 ± 0.04^{a}	1.0^{A}	0.89 ± 0.03^{B}	1.0^{A}	0.93 ± 0.03^{B}	1.0^{A}	0.89 ± 0.04^{B}
	RWB	0.34 ± 0.04^{a}	0.37 ± 0.03^{a}	0.80 ± 0.02^{ab}	0.79 ± 0.03^{ab}	1.0^{A}	0.93 ± 0.02^{B}	1.0^{A}	0.96 ± 0.04^{B}	1.0^{A}	0.91 ± 0.02^{B}
	RB	0.20 ± 0.02^{b}	0.21 ± 0.02^{b}	0.72 ± 0.03^{b}	0.71 ± 0.03^{b}	1.0^{B}	1.21 ± 0.05^{A}	1.0^{B}	1.05 ± 0.01^{A}	1.0^{A}	0.96 ± 0.04^{A}
	NC	0.23 ± 0.04^{b}	0.27 ± 0.03^{b}	0.65 ± 0.02^{c}	0.62 ± 0.03^{c}	1.0^{A}	0.95 ± 0.02^{B}	1.0^{A}	0.91 ± 0.06^{B}	1.0^{A}	0.92 ± 0.02^{B}
IH	WB	0.52 ± 0.03^{a}	0.59 ± 0.02^{a}	1.15 ± 0.02^{a}	1.15 ± 0.04^{a}	1.0^{A}	0.80 ± 0.02^{B}	1.0^{A}	0.85 ± 0.04^{B}	1.0^{A}	0.85 ± 0.03^{B}
	RWB	0.42 ± 0.02^{b}	0.48 ± 0.03^{b}	1.05 ± 0.02^{b}	1.04 ± 0.03^{b}	1.0^{A}	0.84 ± 0.03^{B}	1.0^{A}	0.89 ± 0.03^{B}	1.0^{A}	0.89 ± 0.02^{B}
	RB	0.25 ± 0.01^{d}	0.29 ± 0.02^{d}	0.97 ± 0.01^{c}	0.95 ± 0.03^{c}	1.0^{B}	1.13 ± 0.03^{A}	1.0^{A}	0.97 ± 0.03^{A}	1.0^{A}	0.96 ± 0.04^{A}
	NC	0.36 ± 0.02^{c}	0.38 ± 0.03^{c}	0.86 ± 0.03^{d}	0.84 ± 0.04^{d}	1.0^{A}	0.90 ± 0.02^{B}	1.0^{A}	0.93 ± 0.02^{B}	1.0^{A}	0.91 ± 0.03^{B}

Lower case letters (a-d) rank significant difference among moisture content of samples under same finish-cooking method and time. Upper case letters (A-B) rank significant difference for same textural attribute of the identical sample under same finish cooking at different holding time. NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. DFF, AF, MH, IH respectively represent deep-fat-frying, air frying, microwave heating, and infrared heating.

Post-finish-cooking changes in textural attributes were represented as normalized value of maximum force (MF*), normalized value of maximum distance (MD*), and normalized value of slope (S*). Textural attributes has changed during post-finish-cooking stage, where the extent and nature of textural evolution varied with batter-formulations and finish-cooking methods. DFF processed samples were less prone to textural changes. This could be understood as such: major physicochemical transformations of parfried-frozen batter ingredients occurred under intense heat treatment (during finish-cooking by hot oil), consequently, their post-finish-cooking changes were lesser. AF processed samples have shown very similar behavior as of DFF processed samples, as the processing time for AF was 15 min and its longer processing time might have mitigated the lackage of low heat-intensity of air. RB batter was less prone to post-finish-cooking textural (crispiness, brittleness) changes, as this sample was mostly impacted during finish-cooking (as shown by their higher initial values in Figure 7.3). Whereas WB coated product has shown proneness to post-finish-cooking textural changes, that could be due to their higher extent of moisture re-distribution between spatial region. Interestingly it was observed in RB coated samples that their hardness (MF) was prone to increase during post-finish-cooking stage, while they had been finish-cooked by MH & IH; whereas the extent of post-finish-cooking textural change was lower in DFF & AF treated samples. This could be understood as such: during IH and MH the starch of batter-coating was gelatinized to a lower extent due to shorter period (MH) and lower extent (IH) of heating; hence, remaining's undergo higher extent of starch-retrogradation during their post-finish-cooking stage and consequent changes in textural attributes were observed. Both the DFF and AF might had intensely impacted the starch content during finish-cooking stage; hence, post-finish-cooking textural changes were comparatively less for the DFF and AF cooked samples.

Table 7.2 summarizes, glass-transition-temperature (Tg_{onset}, °C) of the crust region of finish-cooked samples. Tg value of samples were in negative (-) temperature zone, and Tg of the finish-cooked samples were found far-lower than the holding temperature (25°C) at room environment. The lower Tg value explains post-finish-cooking evolution in crust moisture and texture profiles of different samples; as higher stability of qualities are anticipated at a temperature below the intrinsic Tg temperature of the studied samples. Comparatively higher Tg might has supported post-finish-cooking textural stability of DFF and AF processed samples.

Table 7.2 : Mean values with standard errors of glass transition temperature (Tg_{onset}, °C) of the crust of finish-cooked samples.

Finish-cooking method	WB	RWB	RB	NC
DFF	-22.04±0.29	-21.14±0.28	-21.01±0.24	-20.09±0.22
AF	-22.23±0.28	-21.46±0.29	-21.14±0.33	-20.18±0.25
MH	-22.86±0.31	-21.62±0.33	-21.18±0.28	-20.32±0.24
IH	-23.11±0.33	-21.81±0.32	-21.34±0.31	-20.51±0.33

NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. DFF, AF, MH, IH respectively represent deep-fat-frying, air frying, microwave heating, and infrared heating.

Figure 7.4 depicts color attributes of finish-cooked products. Color tones (lightness, redness, yellowness) of AF processed samples were quite similar to DFF processed samples. This similarity in color attributes might had been associated to their heat intensity and treatment duration. Rapid color evolution in DFF were due to its higher rate of temperature rise, and comparable color development in AF was supported by its longer treatment-time. The MH was in favor of retaining the initial color characteristics of par-fried samples (represented by lower ΔE value), which was characterize by higher lightness (L), lower redness (a*), and lower yellowness (b*) color tone. The color tone of MH treated finish-cooked samples indicates that, microwave heating caused lesser alteration in surface color of par-fried samples. To minimize the changes in color properties during finish-coking, MH is preferred over conventional deep-fat-frying. IH favored the formation of undesirable darker surface (lower L values). This could be due to longer processing time and direct exposure of IR-ray to food surface; as food component absorb electromagnetic energy and color of food became darker at higher intensity IR treatment (Rahimi *et al*., 2018). L and b* value of RB batter coated finish-cooked products were higher than WB batter. This indicates, rice-flour based parfried frozen batter has developed bright-yellow colored crust at finish-cooking. Higher yellowness (b* value) of RB crust might be associated with its higher CrF, as color of frying-oil (canola) were light-yellow. Higher redness (a* value) tone of color were noticed in wheat-flour based batter coated samples, while they were finish-cooked by the DFF, IH and AF.

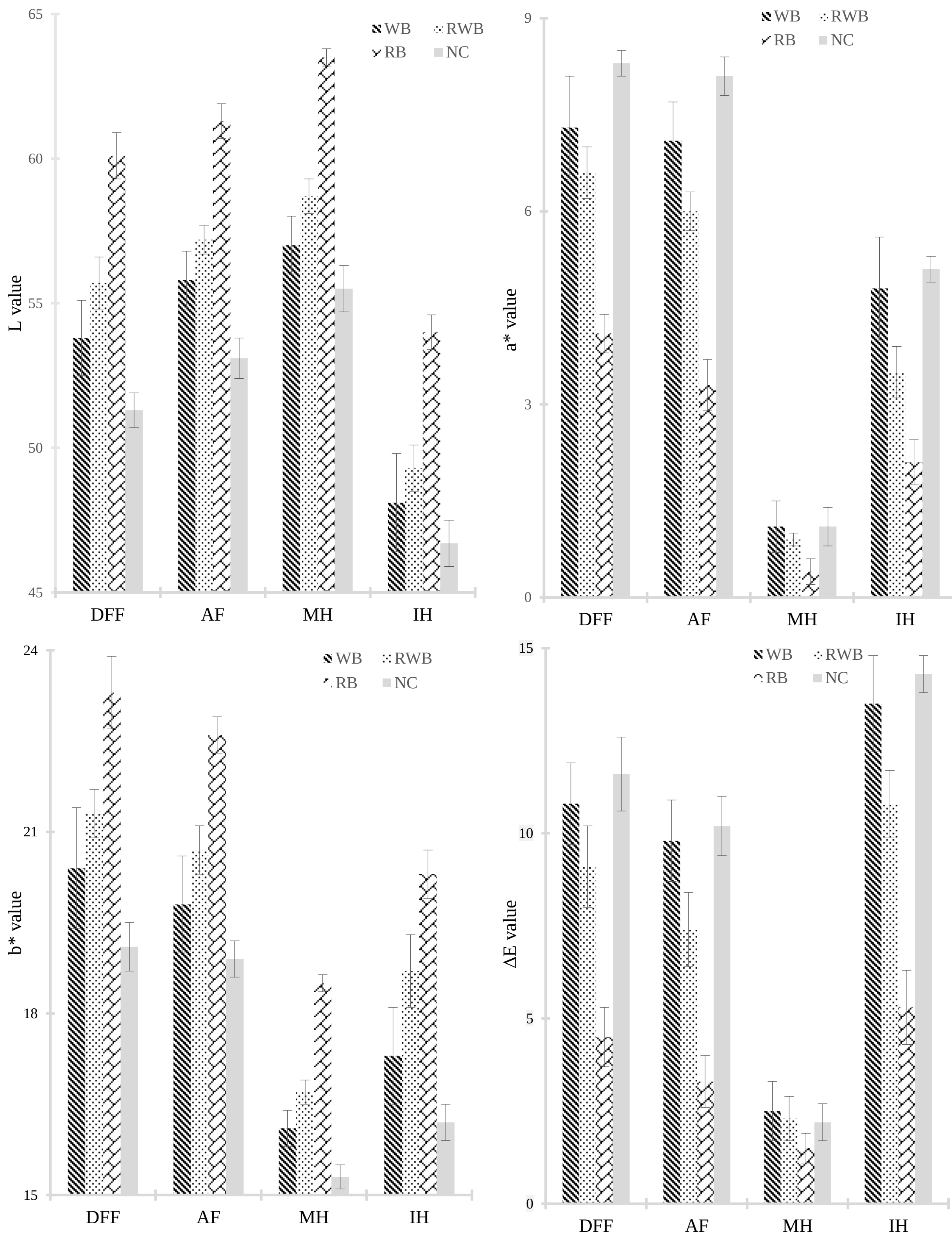
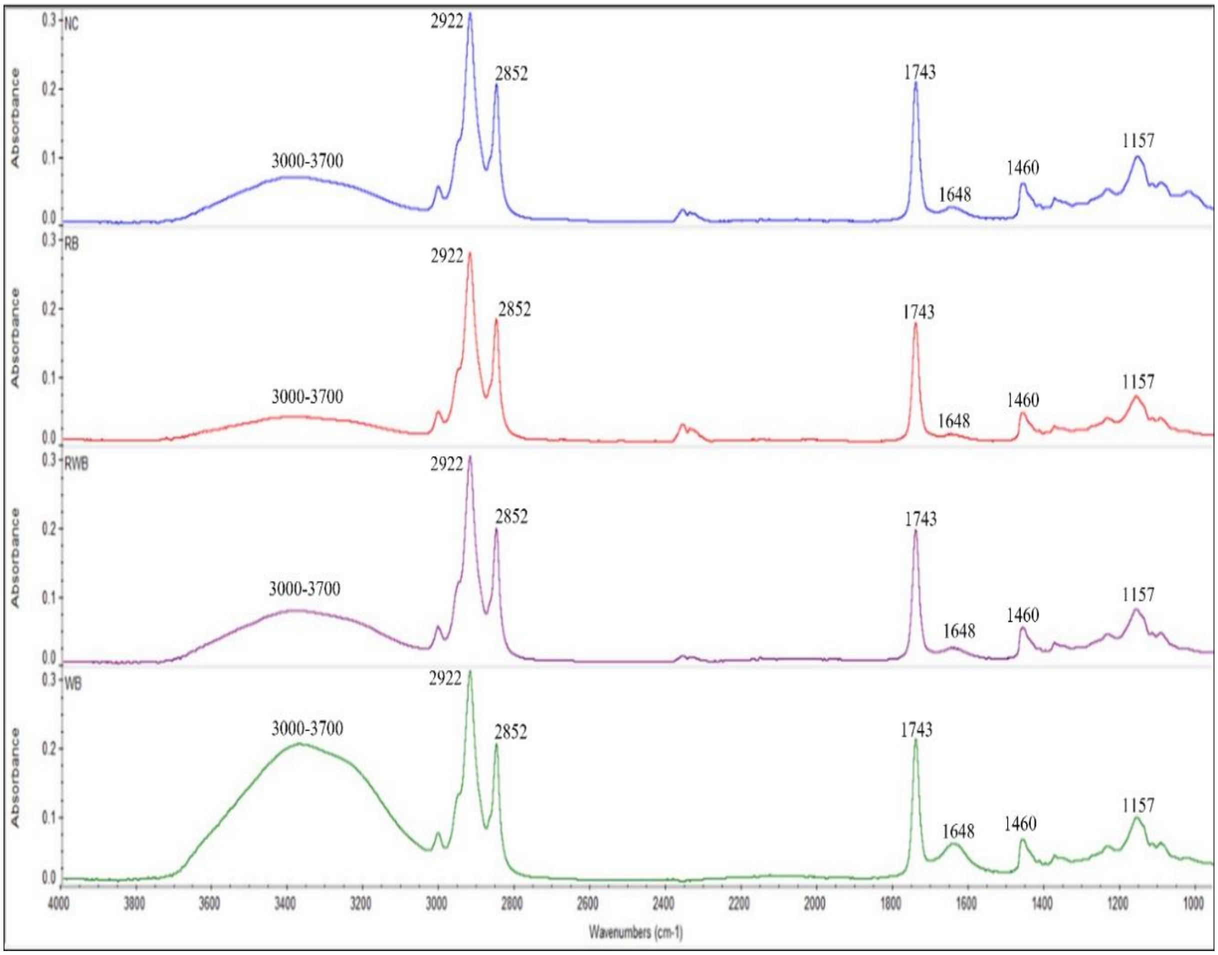


Figure 7.4**:** Color indices of finish-cooked samples. NC, RB, RWB & WB represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. DFF, AF, MH, IH respectively represent deep-fat-frying, air frying, microwave heating, and infrared heating.



Sample	Surface moisture response, SMR (a.u.)	Surface fat response, SFR (a.u.))
WB	88.35±5.68[a]	33.36±3.23[a]
RWB	34.08±3.89[b]	30.15±1.72[a]
RB	11.37±2.17[c]	24.65±1.03[b]
NC	29.24±3.20[b]	32.12±2.17[a]

Figure 7.5**:** ATR-FTIR spectra of parfried nonfrozen samples and estimated mean values of absorbance-area representing surface moisture and surface fat response. Downwards spectra: NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated sample, respectively. Lower case letters (a-c) represent significant ($p<0.05$) difference among means (within column).

Non-zero and higher ΔE value represents, initial color attributes of parfried-frozen samples had undergone prominent changes at IH, followed by DFF and AF. Impact of deep-fat-frying and air-frying on ΔE were quite similar, indicating AF as a suitable substitute to DFF in terms of color development in finish-cooked products. Compared to RB, higher extent of total color change was observed in WB coated samples. Analysis of parfried samples' surface chemistry by ATR-FTIR spectroscopy (Figure 7.5) provides more insight to understand the variation in total color change at finish-cooking. Absorption peaks around 2922, 2852, 1743, 1460, and 1157 cm^{-1} represents the canola oil on the surface of parfried samples (Chen *et al*., 2015). Presence of broad peak around 3000–3700 cm^{-1} and 1648 cm^{-1} region in ATR-FTIR spectra of WB indicates the presence of higher moisture. Presence of higher moisture might be a cause behind its lower L values, as color development during frying occurs, because of moisture loss. In addition to moisture, presence of higher protein in wheat-flour based batter (WB) might favors the formation of brown color via Maillard reaction (Rahimi et al., 2018), as this reaction requires amino acids which was abundant in wheat-flour.

Figure 7.6 depicts scanning electron microscopic images of raw, par-fried, and finish-cooked samples. Distinctly visible polygonal shapes structure were very prominent in SEM image of raw RB batter coated sample, these polygonal structures might be the starch granules (Adedeji & Ngadi, 2018). In SEM image of parfried RB sample, these polygonal shapes were not present and a flatter surface was observed. This indicate the occurrence of starch gelatinization of rice flour. At parfrying, surface of WB and RWB batter coatings have become smoother; that might be due to the denaturation of their protein entity under heat treatment. Parfrying process created surface-irregularities such as holes, rupture, and crevices. The presence of these structural irregularities were mostly noticed in parfried RB batter. These surface structures has acted as access way for moisture-loss and consequent fat-uptake during finish-cooking by DFF, whereas had favored loss of moisture as well as loss of oil under AF, MH, and IH treatments. Surface micrograph of finish-cooked noncoated (NC) sample showed higher extent of these structural irregularities, that explains their higher moisture loss and consequent higher fat uptake during deep fat frying.

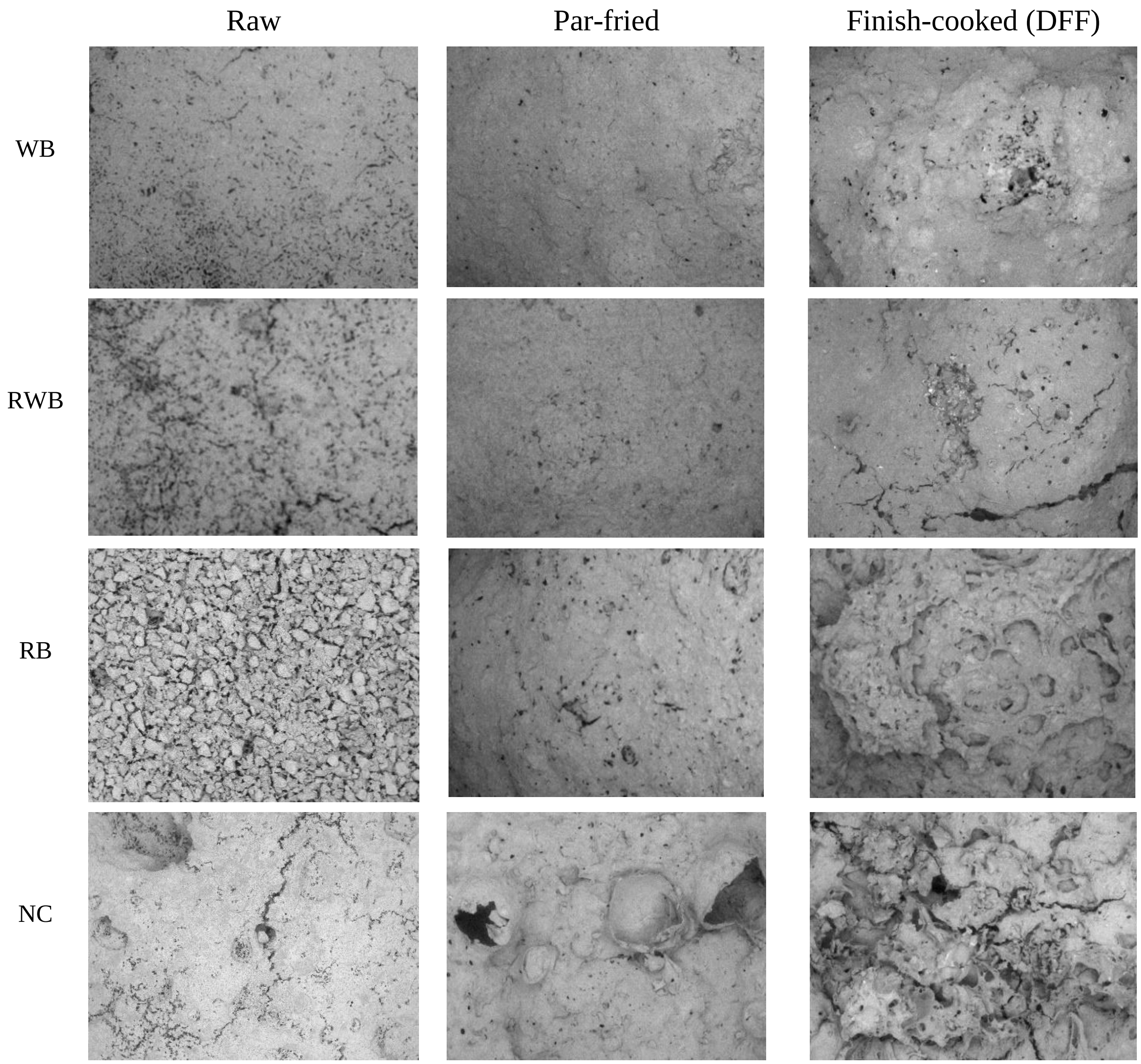


Figure 7.6: Micrographs of surface obtained by scanning electron microscopy. WB, RWB, RB & NC represent wheat-flour based batter, wheat & rice flour-based batter, rice-flour based batter, and noncoated model food, respectively. DFF: deep-fat-frying.

Figure 7.7 & Table 7.3 depicts the effect of batter-formulations and finish-cooking method on surface roughness. Higher surface roughness were observed for rice-flour based batter (RB) coated samples, compared to wheat-flour based batter (WB) coated samples; and these relatively higher roughness have been reflected as higher fractal dimension (FD) values of RB coated samples. Surface roughness of par-fried samples has increased (higher FD) due to finish-cooking, wherein deep-fat-frying (DFF) have caused in higher surface roughness in comparison to air-frying (AF).

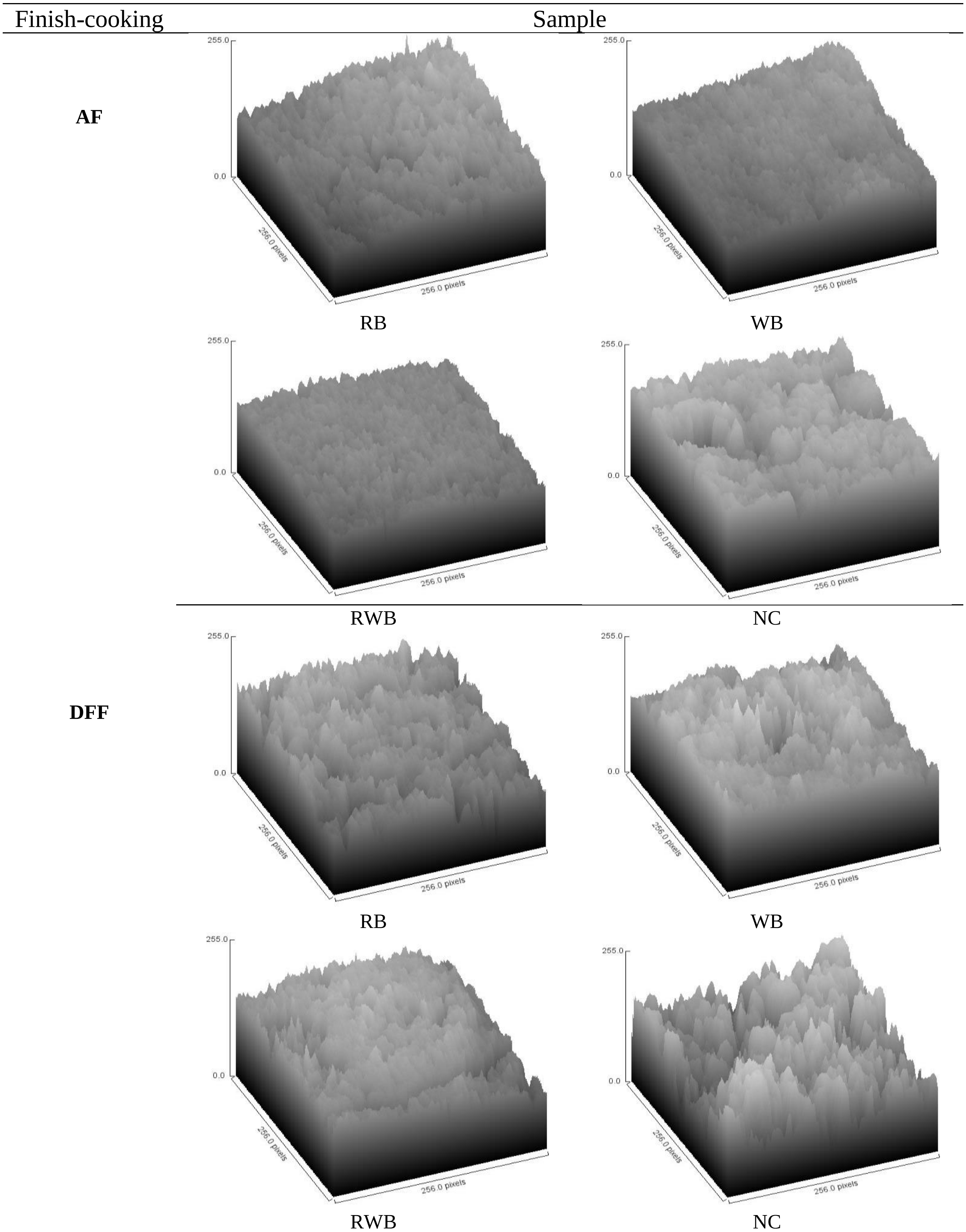


Figure 7.7: Surface plot depicting roughness of the samples. NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. AF: air-frying, DFF: deep-fat-frying.

Table 7.3: Estimated fractal dimension (FD) as an indicator of surface roughness.

Sample	WB	RWB	RB	NC
Par-fried	2.559±0.016	2.568±0.014	2.598±0.012	2.530±0.031
AF	2.588±0.012	2.597±0.015	2.613±0.011	2.644±0.024
MH	2.598±0.027	2.612±0.028	2.671±0.014	2.685±0.025
IH	2.613±0.032	2.658±0.037	2.704±0.022	2.738±0.036
DFF	2.827±0.018	2.854±0.021	2.921±0.020	2.948±0.024

WB, RWB, RB & NC represent wheat-flour based batter, wheat & rice flour-based batter, rice-flour based batter, and noncoated model food, respectively. DFF, AF, MH, IH respectively represent deep-fat-frying, air frying, microwave heating, and infrared heating.

7.5 Conclusion

Batter-formulation influences the process parameters and attributes of meat analog based coated food. Microwave heating (MH), infrared heating (IH), and air frying (AF) as alternative finish-cooking methods has caused higher finish-cooking loss (F_{CL}) of parfried-frozen batter coated product; and for deep-fat-frying (DFF) a lower F_{CL} were observed. Alternative finish-cooking methods (MH, IH, AF) significantly reduced spatial (crust, total) fat, compared to conventional DFF of parfried-frozen products. Parfried frozen-batter significantly reduced fat uptakes during finish-cooking by DFF. Spatial (crust, total) moisture profile of finish-cooked coated products are intertwined with batter-formulations and finish-cooking method. Evolution of textural and color attributes of parfried-frozen coated foods were associated with batter-formulations and finish-cooking methods. Rice-flour based parfried frozen batter coating have shown preference over wheat-flour based batter, to develop bright-yellowish colored and crispy crust during finish-cooking. AF have shown great potentiality to be used as a very suitable substitute of DFF, to finish-cook the parfried frozen-batter coated products. Electromagnetic heating (MH, IH) could be considered as healthier compared to DFF, while, in terms of textural attributes the later method has shown preference over the former two. Outcomes of this study would provide scientific guidance in serving fried products of having less oil.

CONNECTING TEXT TO CHAPTER 8

Chapter 7 reveals that air-frying (AF) could be used as a suitable alternate to deep-fat-frying of batter coated parfried frozen foods. However, mechanistic insights on quality evolution as well as prediction of change in major quality traits, is crucial to optimize the processing parameters. Chapter 8 investigated the effect of AF parameters on mass-transfer, texture, surface structure and color evolution of parfried-frozen batter-coated foods, and mathematically model their kinetics.

Parts of this chapter have been presented at scientific conference.

Md. Hafizur Rahman Bhuiyan and Michael O. Ngadi. (2023). Air-frying of meat-analog based parfried-frozen batter coated foods. Presented at Canadian Society for Bioengineering (CSBE**)** Annual General Meeting. Held at Lethbridge, Alberta, Canada, from July 23-26, 2023. ID:153. *Oral presentation*.

CHAPTER 8

AIR-FRYING OF PARFRIED FROZEN BATTER COATED FOODS

8.1 Abstract

Air-frying (AF) is a novel method of producing low-fat containing fried foods using hot-air. This study investigated the effects of AF parameters (time, temperature) on mass-transfer, texture, surface structure and color changes during air-frying of parfried-frozen batter coated product; and mathematically modeled their kinetics. Wheat and rice flour-based batters were used to coat a meat-analog model. Coated products were partially-fried at 180°C for 1 min in canola oil and subsequently frozen at -18°C before they were stored for 7 days. Frozen-products were fried for different time (0, 5, 10, 15, 20 min) by hot-air of different temperature (160, 170, 180, 190°C). This study reveals, mass-transfer, texture and color development during air-frying of parfried-frozen coated product, are dependent on both AF parameter and batter-formulation. At varied AF temperatures, moisture content of frozen-products declined with frying time. Non-linear regression analysis outcomes (R^2, sum square error) showed, Newton, Page, modified Page, and Henderson & Pabis model could satisfactorily predict moisture-loss of parfried-frozen products. Zero-order kinetic models satisfactorily characterized the changes in textural (hardness, brittleness, crispiness) and color (L, a*,b*, ΔE) traits. Rate-constant (min^{-1}) of moisture, texture and color changes were positively correlated with AF temperature. Arrhenius model sufficiently characterized the temperature dependency of rate-constants. Higher moisture-diffusivity (D_{eff}, m^2/min), higher rate constant (k, min^{-1}) of textural change, lower rate of total color change (ΔE) and lower activation energy (E_a, kJ/mol) values, were estimated for rice-flour based batter coated product. Microstructural analysis (surface opening, fractal dimension) demonstrated the impacts of frying-parameter and batter-formulation on mass-transfer during AF.

8.2 Introduction

Presence of high amount of fat in fried food product, is a serious health concern. To reduce fat content in fried product, different strategies like modification of product surface, modification of frying medium and modification of frying technique have been proposed in literature (Korkmaz et al., 2022; Devi et al., 2020; Liberty et al., 2019).

Batter coating is an effective strategy to modify the surface of food. And in frying process, both heat and mass-transfer occur through the surface of food. However, par-frying is a process where foods are partially cooked by heating in edible oil at high temperature. Par-fried (PF) foods are generally stored as frozen and that require finish-cooking before consumption. Par-fried-frozen (PFF) foods are finish-cooked mainly by deep-fat-frying (DFF). The DFF usually causes the presence of high amount of fat in fried product (Castro-López et al., 2023). However, health concern people are interested in low-fat or fat-free foods which could be prepared by novel air-frying technique (Fikry et al., 2021; Yu et al., 2020). It has been pointed, air-frying could reduce up to 90% the use of oil in compare to conventional deep-fat-frying (Castro-López et al., 2023; Fikry et al., 2021; Cao et al., 2020).

Frying could be considered as a dehydration process where heat and mass transfer occurs simultaneously in a counter-flux fashion. Air frying (AF) is an emerging cooking technique where hot-air is used as heat transfer medium instead of edible oil (Cao et al., 2020). In AF, the superheated air is circulated around the product and the overall process could be regarded as dehydration process, in which simultaneous heat and mass transfer happens that causes water-loss and chemical reactions in food (Fikry et al., 2022 & 2021; Shaker, 2014). It could be hypothesized that these changes are associated with the development of quality attributes in air-fried products. Therefore, study on the kinetics of mass-transfer and related quality attribute in air-frying process is of crucial importance, towards the production of low-fat-containing healthier fried food product.

Air-frying has been studied for several food products such as french fries (Gouyo et al., 2020), surimi (Yu et al., 2020), doughnuts (Ghaitaranpour et al., 2018), potato strips (Shaker, 2014), and falafel (Fikry et al., 2022 & 2021). The dynamics of mass-transfer during deep-fat-frying have been widely studied and reported in literature (Rahimi & Ngadi, 2014; Adedeji et al., 2009). However, to the best of our knowledge, study on the kinetics of moisture loss from parfried-frozen batter-coated products and their kinetics of quality (textural, color) development during air-frying, is not available in literature. To these context, this study evaluated the effect of parameters (temperature, time) of air-frying on kinetics of moisture-loss as well as on the development of textural and color attributes.

8.3 Materials and Methods

8.3.1 Materials

Soy protein isolate was purchased from MP Biomedicals (29525 Fountain Pkwy, OH, USA). Wheat gluten was supplied by Sigma-Aldrich Co. (Ontario, Canada). Methylcellulose (MC) and Sodium pyrophosphate ($Na_4P_2O_7$) crystals were received from Fisher Scientific (Fair Lawn, New Jersey, USA). Sodium bicarbonate ($NaHCO_3$) was supplied by Church & Dwight Canada Corp. (Ontario, Canada). Rice flour (Suraj®, composed of 6.67 % protein, 83.32% carbohydrate, 1.33 % fat and 8.68% moisture) was procured from a grocery store in Montreal, Canada. Commercial wheat flour (Five Roses®, composed of 13.33 % protein, 73.33% carbohydrate, 3.33% fiber, 1.33 % fat and 8.68% moisture), NaCl (Sifto, Compass Minerals Canada Corp.) and Canola oil (Sans nom®, Loblaws Inc.**)** were purchased from a grocery store in Sainte Anne de Bellevue, Canada.

8.3.2 Sample preparation

Meat analog model food was formulated with soy protein isolate (SPI), wheat gluten (WG), distilled water (DW) and canola oil (CO). At first, wheat gluten was mixed with fresh canola oil and as soon as homogeneous slurry was obtained, soya protein isolate was added and mixed thoroughly. Then water was added and mixed properly until a very soft-textured dough was developed. Respective proportion (g) of the ingredients WG: CO: SPI: DW were 12:15:22:73. Prepared dough was used to fill individual rectangular-shaped cavities of silicon mold (Freshware CB-115RD 24-Cavity Silicone Mini Mold). Covered silicon mold was held in complete horizontal position (surrounded by water, but not immersed) on top of a flat metal wire surface and heated for 15 min at 70°C in a water bath. After heat treatment, mold was allowed to reach equilibrium with room temperature. After that, gentle pressure (by finger) was applied from the back of each cavity to bring out the formed substrates without any fracture. This substrate was used as model food, weighing an average weight of 16±0.2 g/substrate and having a uniform dimension of 5.6 cm x 2.5 cm x 1.3 cm. The prepared model-food was packed in zip lock bags and stored in refrigerator (4±2°C) for 24 hours, before its use.

To prepare batter coatings, crystals of Sodium pyrophosphate was grounded to obtain as fine powder. Then Sodium bicarbonate (1.4%), NaCl (1.5%), and Methylcellulose (0.3%) were added and mixed properly with Sodium pyrophosphate powder (1.8%), to form a homogeneous powder mix. The powder-mixture was further thoroughly mixed with dry flour in a proportion of 95% (flour) to 5% (powder mix). Distilled water was added gently to the solid ingredients with a total solid to water ratio of 1:1.3 and mixed them properly until a homogenous tempura-batter slurry was obtained. Rice based batters (RB) were formulated using only rice flour whereas wheat four based batters (WB) were formulated with wheat flour. Equal proportion of flours were used to formulate rice and wheat flour based batters (RWB). The prepared batter systems were kept at room environment for 5 min before its use to coat the model food. The model food was brought out of refrigerator and kept at room environment for 30 min, prior to batter coating. It was fully immersed in batter system for 1 minute and then removed from batter solution to make a complete coat. The coated product was held on a kitchen-fork for about 30 sec, to drain the excess amount, in order to obtain a uniform layer of coating around model food core. Non-coated (NC) meat analog model was used as control sample.

A programmable deep fat fryer (T-fal Compact, FF122851, made in China) filled with 1.5 L of fresh canola oil was used to par-fry (partially fry) the samples. The oil was preheated for 1hr and were stirred to minimize any variation in oil temperature. Samples were uninterruptedly fried for 1 mine at 180°C, maintaining a sample to frying oil ratio of 1:30. At the end of par-frying, the frying basket was removed from oil and shaken 5 times; and the samples were allowed to cool at room environment. Par-fried (PF) samples were packed in Ziplock bag and kept at frozen storage (-18°C) for a week. Par-fried-frozen (PFF) samples were used for subsequent air-frying (AF) experiment, without any prior treatment. Approximate mean thickness of the parfried-frozen WB, RWB, RB and NC samples were about 1.46, 1.42, 1.36 and 1.28 cm, respectively.

A programable air fryer unit (Philips HD9240/90, China) were used for air-frying (AF) of the par-fried-frozen (PFF) samples. To reach a thermal equilibrium, AF unit were turned on for 5 min before its use to fry the PFF samples. The PFF sample was placed within the bucket (on top of were mesh) of air-frying unit. The air-frying process was conducted at four different temperatures (160, 170, 180 & 190°C) for different times (0, 5, 10, 15 & 20 min). There was no need to rotate/flip over the sample during air-frying, since hot air covered all parts of the sample's surface. At the end of air-frying, fried samples were in rest at room environment for 1 min, before further analysis.

8.3.3 Assessment of moisture, texture, color and surface structure

To determine the moisture content, air-fried samples were freeze dried (Modulyod-115; Thermo Savant, USA.) at -50°C and 250 mbar for 48 hours. Weight of the sample before and after freeze drying was measured and moisture content was calculated on dry basis (g/g dry matter), by dividing the mass of moisture with the mass of the freeze-dried sample.

Color attributes of samples were assessed by using a spectrophotometer (Minolta Spectrophotometer CM-3500d, Japan). Color was determined in CIELab space (Illuminant D65, 10° viewing angle) from the reflection spectra between 400-700 nm. Color parameters L (lightness-darkness), a* (redness-greenness), and b* (yellowness-blueness) was estimated at room environment and total color difference (ΔE) was calculated as follows:

$$\text{Total color difference, } \Delta E = [\Delta L^2 + \Delta a^2 + \Delta b^2]^{1/2} \qquad 8.1$$

where, $\Delta L = L_i - L_t$, $\Delta a = a_i - a_t$, $\Delta b = b_i - b_t$, and subscript "i" and "t" denotes initial value at time zero and at any duration of air-frying, respectively.

Textural attributes of air-fried samples were evaluated by puncture/compression test, using a mechanical texture analyzer (Stable Micro Systems Texture Analyzer, TA. HD PlusC, UK). Test samples were individually mounted on a flat rigid support. A puncture probe TA-52 (2 mm dia.) and 500 N was used to punch test sample, at a constant test speed of 1mm/s and a travel distance of 5 mm. Puncture test were performed on three equidistance locations of the air-fried sample. Following the detailed methodology of (Rahimi et al., 2017), the maximum force to break (N), displacement at maximum force (mm), and slope (N/mm) at maximum force of the puncture test data were chosen as parameters to evaluate textural properties namely hardness, brittleness, and crispness, respectively. Puncture test data were analyzed by TA. HD PlusC program (Exponent 6.1.14 v, Stable Micro Systems, UK).

Scanning electron microscopy (SEM) were used to assess surface morphology. Surface washed (by three consecutive charges of petroleum ether, for a total immersion period of three minute) fried samples were freeze dried and used for SEM imaging by a scanning electron microscope (Hitachi TM3000, Japan). SEM operational procedure was as such, 5 kV electron power, auto contrast function, and composition mode of imaging were used. Small cut, rectangular shaped, defatted sample of about 10×10×5 mm in dimension was placed on sample base with a sticky surface that comprises of carbon tab. After each sample was properly aligned by visual control using two sets of knobs and images shown in the control software on a computer screen, vacuum pressure was created in the sample chamber and images were quickly acquired to prevent charging and heating that could lead to artifacts in images. The SEM images were acquired at a magnification of ×30 and recorded as 8-bit joint photographic experts group (JPEG) file. The surface microstructure was analyzed by Java-based public domain image processing software (ImageJ 1.52 v, National Institutes of Health, USA). Surface opening (SO) was estimated from SEM micrograph, as ratio of the area of total openings (holes, cracks, ruptures, crevices etc.) in surface to the total surface area. In estimating SO from SEM images, the image analysis approach of (Rahimi & Ngadi, 2015) was followed i.e., darker region represents holes, cracks, ruptures, crevices, while whiter region represents solid matrix. Fractal dimension (FD) of surfaces were estimated from the SEM images by box-counting method, as detailed by (Rahimi & Ngadi, 2016).

8.3.4 Modeling of mass transfer, texture and color change

Following relevant studies (Singh et al., 2013; Rahimi & Ngadi, 2014), the effects of air-frying (AF) parameters on moisture-loss kinetics was evaluated on the basis of mathematical models of thin layer drying, including Newton, Page, Modified page, Henderson and Pabis model as follows (Eq 8.2 to Eq 8.5):

Newton	$MR = e^{-kt}$	8.2
Page	$MR = e^{-kt^n}$	8.3
Henderson and Pabis	$MR = a\ e^{-kt}$	8.4
Modified Page	$MR = e^{(-kt)^n}$	8.5

Here, MR is moisture ratio ($\frac{M_t}{M_0}$). M_0 *and* M_t represents initial moisture content (dry basis), and moisture content (dry basis) at any time t (min). In equations, k represents the moisture-loss rate constant (min^{-1}). The n and a are coefficients (unitless) which have values depending on the curve and equation.

Moisture loss during AF could be considered as diffusion controlled process. To model the phenomenon of mass-transfer in fried foods, Fick's second law of diffusion (Eq 8.6), can portray a simple description of the moisture alteration throughout frying (Fikry et al., 2022; Adedeji et al., 2009; Rahimi & Ngadi, 2014).

$$\frac{\partial}{\partial L}[D_{eff}\ \frac{\partial M}{\partial L}] = \frac{\partial(M)}{\partial t} \qquad 8.6$$

Under several assumptions (i.e., initial uniform moisture content, negligible external resistance to mass transfer, minimal shrinkage, mass transfer took place from both sides of the sample) the above partial differential equation could be simplified (Eq 8.7) as per solution of Crank (1975) for infinite slap shape geometry, which has been used in numerous studies (Rahimi & Ngadi, 2014; Adedeji et al., 2009).

$$MR = \frac{M_t - M_e}{M_0 - M_e} = \frac{8}{\pi^2}\sum_{n=0}^{\alpha}\frac{1}{(2n+1)^2}exp\left[\frac{-(2n+1)^2\pi^2 D_{eff}t}{4L^2}\right] \qquad 8.7$$

Where MR is moisture ratio (dimensionless). M_0, M_e, *and* M_t represents initial moisture content (dry basis), equilibrium moisture content (dry basis) and moisture content (dry basis) at time t, respectively. The D_{eff} represent effective moisture diffusion coefficient (m^2/min), and L is the half-thickness (m) of parfried-frozen sample. Eq 8.7 can be simplified as Eq 8.8, considering the M_e value as very small (~0).

$$\text{MR}= \frac{M_t}{M_0} = \frac{8}{\pi^2}exp^{[-\pi^2 D_{eff}\ t/4L^2]} = \frac{8}{\pi^2}exp^{-kt} \qquad 8.8$$

where, k = $\frac{\pi^2 D_{eff}}{4L^2}$, and k represents the moisture-loss rate constant (min^{-1}) 8.9

To mold the changes in color traits (L, a*, b*, ΔE value) and textural traits (hardness, ductileness, crispiness) during air-frying, the Zero-order and First order kinetic models (Equation 8.10 and 8.11) were considered.

Zero-order model $P_t = P_0 \pm kt$ 8.10

First-order model $P_t = P_o\ exp^{(\pm kt)}$ 8.11

Where P_0 means the initial trait at zero-time, P_t is the trait at time t, k refers to rate constant (min^{-1}) and t represents frying time (min), + represents growth and – represents deterioration.

Temperature-dependency of the parameters (effective moisture diffusivity, rate constants) were modeled by using the Arrhenius-type expression as Eq 8.12 and Eq 8.13 (Castro-López et al., 2023; Fikry et al., 2022; Adedeji et al., 2009).

$$D_{eff} = D_0 \exp[-\frac{E_a}{RT}] \quad 8.12$$

$$k = k_0 \exp[-\frac{E_a}{RT}] \quad 8.13$$

Where, E_a refers to activation energy (kJ/mol), D_0 / k_0 refers to pre-exponential coefficient, T refers to the absolute frying temperature (K), and R signifies universal gas constant (8.314 kJ/mol. K).

To assess the suitability of mathematical models in fitting experimental data, statistical parameters i.e., reduced sum square error (SSE) and coefficient of determination (R^2) were estimated (Eq 8.14 and Eq 8.15) and considered as the criteria for goodness of fit (Fikry et al., 2022; Singh et al., 2013).

$$R^2 = 1 - \frac{\sum_{i=1}^{i=n} \left(X_i^{exp} - X_i^{pre}\right)^2}{\sum_{i=1}^{i=n} \left(X_i^{exp} - \bar{X}^{exp}\right)^2} \quad 8.14$$

$$SSE = \frac{1}{n} [\sum_{i=1}^{i=n} (X_i^{exp} - X_i^{pre})^2] \quad 8.15$$

Where, x^{exp}, x^{pre} and $\bar{x}^{exp}$ denotes experimental, predicted, average experimental value, respectively.

8.3.5 Data analysis

All experimental data obtained from triplicate of samples, mean ± standard deviation was reported. Fitting of model equations and estimation of the model parameters were performed in Microsoft Excel 16 (Microsoft Corporation, USA) environment using the Data “Solver Add-in” program. Statistical analysis were performed by a licensed statistical software (JMP 14.1v, SAS Institute Inc., Cary, NC, USA).

8.4 Results and Discussion

Figure 8.1 depict, changes in moisture of parfried-frozen samples during air-frying (AF). Analysis of variance (ANOVA) showed, both the batter-formulation and frying time significantly impacted the moisture content of air-fried coated food products. Initial moisture content of parfried-frozen products has reduced as AF processed. Parfried-frozen samples with higher initial moisture content before AF, generally had higher final moisture after identical duration of air-frying. This observation is similar to the deep-fat-frying of microwave pretreated chicken nuggets (Adedeji et al., 2009). The disparity in moisture content of air-fried batter-coated products (higher in WB and lower in RB coated samples) might be due to the presence of higher gluten in wheat-flour. It has been reported, gluten supports good film-forming and water-holding-capacity of wheat flour based batters to prevent moisture loss during deep-fat-frying (Rahimi & Ngadi, 2014). Overall, moisture loss profiles during the AF of parfried-frozen product has showed slight difference to the typical pattern of moisture loss (rapid loss at the beginning) in deep-fat-frying (DFF). Moisture loss from parfried-frozen samples were lower at early stage of AF, after that, substantial moisture loss were observed as frying progressed. This initial time could be understood as the required time to heat-up the frozen-samples (were -18°C) in favor of melting their ice-crystals and consequently evaporate them as moisture vapor. It could be mentioned, in coated food products, the moisture content of outer coating layer is generally lower than the inner core. It has been reported (Ngadi et al., 2006) that the moisture loss of oven-baked chicken nuggets mainly occurs in the coating region at the early stage of heating and occurred in the core region at the later stage of heating. In this study, the observed moisture-loss pattern (slow decrease at early stage and fast decrease at later stage) of parfried-frozen batter coated products during the AF, are analogous to relevant studies (Castro-López et al., 2023; Cao et al., 2020; Ghaitaranpour et al., 2018).

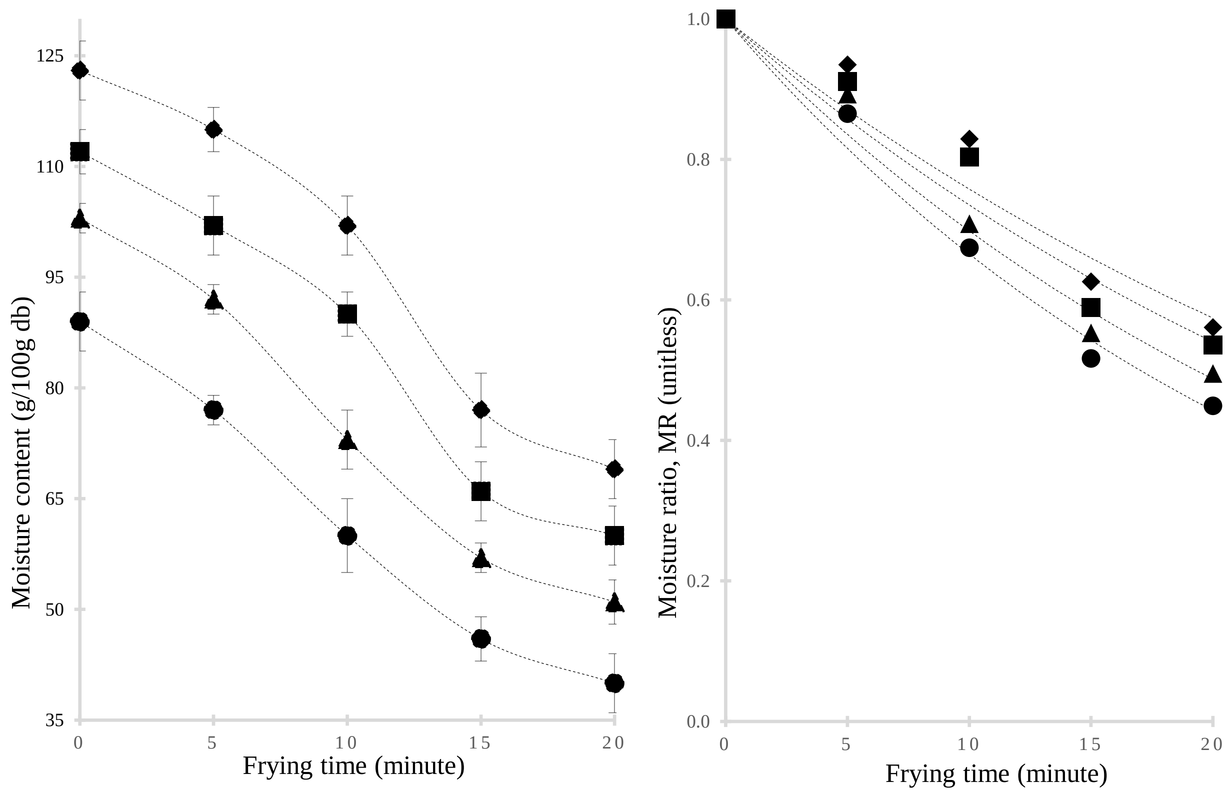


Figure 8.1: Evolution of moisture during air-frying at 190°C. NC (●), RB (▲), RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. Symbol and dashed-line represents experimental value and prediction line, respectively.

To predict moisture loss during AF of parfried-frozen products, experimental moisture ratio (MR) were represented by the proposed models (Eq 8.2 to Eq 8.5) by employing non-linear regression analysis (NLRA) technique. Table 8.1 summarizes coefficients and statistical parameters of the studied models for the samples fried at 190°C. The outputs of NLRA showed, proposed mathematical models exhibited goodness of fit, judging R^2 values higher than 0.945 and *SSE* values lower than 0.010. These indicates, AF behavior of meat-analog based parfried-frozen batter coated products could be satisfactorily predicted by the Newton, Page, Henderson & Pabis, and Modified Page model. This observation are similar to the study of Fikry et al. (2022), where they studied the moisture loss of non-parfried and non-frozen Falafel within the air-frying temperature range of 140-200°C. Irrespective of models, estimated value of the coefficient (k, min^{-1}) was higher for NC sample, followed by RB, RWB and WB coated sample. This could be due to variation in their formulation and microstructural properties. In deep-fat-frying process, Rahimi & Ngadi

(2014) observed that the moisture-transfer kinetic were significantly ($p<0.05$) affected by the changes in the amount of wheat and rice flour in the formulation of batter systems.

Table 8.1: Coefficients and statistical parameters obtained for different mathematical models.

Sample	Coefficient / Statistics	Model			
		Newton K (min^{-1})	Page K (min^{-1}) / n	Modified page k (min^{-1}) / n	Henderson & Pabis a / k (min^{-1})
WB	Coefficient	0.027	0.006/1.524	0.025/1.089	1.039/0.029
	R^2	0.948	0.979	0.948	0.945
	SSE	0.010	0.003	0.010	0.008
RWB	Coefficient	0.030	0.010/1.403	0.026/1.150	1.032/0.032
	R^2	0.956	0.976	0.956	0.954
	SSE	0.009	0.040	0.009	0.007
RB	Coefficient	0.035	0.021/1.200	0.028/1.255	1.023/0.037
	R^2	0.981	0.987	0.981	0.981
	SSE	0.004	0.003	0.004	0.004
NC	Coefficient	0.040	0.026/1.163	0.030/1.337	1.020/0.417
	R^2	0.988	0.992	0.988	0.988
	SSE	0.003	0.002	0.003	0.003

NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. *SSE*: Sum square error, R^2: coefficient of determination.

Relevant studies showed that during conventional frying process, the moisture ratio data of different food categories such as shrimp nugget (Dehghan Nasiri et al., 2011), chicken nugget (Castro-López et al., 2023; Adedeji et al., 2009), and batter-coating (Rahimi & Ngadi, 2014) well-fitted to the exponential model (as Eq 8.8) which was obtained based on the solution of Fick's 2nd law of diffusion. To estimate experimental effective moisture diffusivity (D_{eff}) of parfried-frozen products based on rate constant (k) of moisture loss, the exponential model (Eq 8.8) was considered. Moisture ratio (MR) versus air-frying time (t) were plotted in semi-logarithmic fashion (ln MR vs t). The slope of Eq 8.8 represents rate constant (K) of moisture loss (Rokib et al., 2021; Rahimi et al., 2018). In estimating K, value of the ($8/\pi^2$) term of Eq 8.8 was considered as unity, to obtain a MR value of 1 at time t=0 (Rahimi & Ngadi, 2014). Finally, the D_{eff} value was calculated from the rate constant according to the relationship as shown in Eq 8.9.

Table 8.2 summarizes, rate constant of moisture-loss and effective moisture diffusivity at varied AF temperature. Rate constant (k) of moisture loss of parfried-frozen products were impacted by AF temperature. The temperature of hot air and k-value showed positive correlation. Rate of moisture loss from batter-coated parfried-frozen products were interwind with the formulation of batter-coatings. Higher k values were estimated for RB batter coated product, in comparison to WB batter coated product. This observation are analogous to the study on deep fat frying of batter systems (without food core) of having different ratio of wheat and rice flour (Rahimi & Ngadi, 2014). Overall, higher K values were estimated for non-coated sample, in comparison to batter-coated samples. This signifies batter-coating impacted their rate of moisture-loss during the AF of parfried-frozen products.

Table 8.2: Kinetic model parameters of moisture-transfer.

Sample	Air-frying temperature, (°C)	Rate constant, K (min^{-1})	Effective moisture diffusivity, $D_{eff} \times 10^{-7}$ (m^2/min)	R^2
WB	160	0.005	1.07	0.92
	170	0.013	2.81	0.94
	180	0.025	5.39	0.93
	190	0.028	6.05	0.95
RWB	160	0.006	1.23	0.91
	170	0.016	3.27	0.94
	180	0.027	5.52	0.92
	190	0.031	6.33	0.96
RB	160	0.012	2.25	0.95
	170	0.018	3.38	0.93
	180	0.032	6.00	0.96
	190	0.036	6.75	0.98
NC	160	0.015	2.49	0.93
	170	0.023	3.82	0.95
	180	0.039	6.48	0.94
	190	0.041	6.81	0.99

NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. R^2: coefficient of determination.

Effective moisture diffusivity (D_{eff}) of foods characterizes its intrinsic moisture-transport characteristics that includes molecular, liquid and vapor diffusion as well as hydrodynamic-flow and any other probable mass transport phenomenon (Rahimi et al., 2018). D_{eff} values of noncoated

(NC) sample ranged between 2.49×10^{-7} m^2/min to 6.81×10^{-7} m^2/min for the frying temperature range of 160-190°C. D_{eff} values ranged between 1.07×10^{-7} m^2/min to 6.05×10^{-7} m^2/min for WB batter coated parfried-frozen samples and 2.25×10^{-7} m^2/min to 6.75×10^{-7} m^2/min for RB batter coated samples. For identical air-frying temperature, higher D_{eff} was estimated for RB coated samples in compared to WB coated samples. In a separate study on deep-fat-frying, compared to wheat-flour based batter system significantly ($p<0.05$) higher moisture diffusion coefficient (2.57×10^{-6} m^2/min < 2.85×10^{-6} m^2/min) were estimated for rice-flour based batter (Rahimi & Ngadi, 2014). However, comparatively lower D_{eff} values of the studied WB coated parfried-frozen products could be attributed to their higher water retention ability, which might be favored by the good film-forming ability of wheat-gluten. Overall, the D_{eff} of parfried-frozen products increased with the increase of AF temperature (°C). This could be associated with the increase of heating-rate which might had accelerated the water molecules to be diffused. This observation is analogous to the findings of the study on air-fried falafel (Fikry et al., 2022) and chicken nuggets (Castro-López et al., 2023). However, the estimated D_{eff} values of batter-coated parfried-frozen products of this study were comparatively lower than the recently reported average moisture diffusivity values for non-frozen commercially produced breaded chicken nuggets (Castro-López et al., 2023). The observed lower D_{eff} could be attributed to the differences in product-formulations and state/condition of the products; as initial-state of the samples of this study were at frozen (-18°C) condition and were batter-coated instead of bread-crump. In addition, initial moisture content (dry basis, db) of parfried-frozen samples of this study were lower than the samples used in the study of Castro-López et al. (2023). It could be mentioned, Rahimi & Ngadi (2014) observed significantly ($p<0.05$) lower D_{eff} value for identical batter system which was dried for 60 minutes prior to deep-fat-frying, in compared to non-dried counterpart.

Color is an important sensory attribute of fried foods that affects its consumer acceptance, even before enters into mouth. Figure 8.2 depicts, color traits of air-fried (at 190°C) products. L value refers to the degree of lightness, and this parameter is generally the first quality attribute of fried foods evaluated by its consumers (Fikry et al., 2022; Cao et al., 2020). L value of air-fried products has shown a decreasing trend with frying-time. This signifies, during AF, the initial lightness of parfried-frozen product changed towards a dark color tone. This observation is analogous to relevant studies, where alteration in lightness were explained as the the resultant effect of moisture-loss, protein-denaturation and browning reaction (Castro-López et al., 2023; Fikry et al., 2022; Cao et al., 2020). The a* value (redness color tone) of air-fried product increased with the duration of AF. Increase in a* value might be a considerable issue, from both the viewpoints of color-perception and health-consciousness. Generally, in fried food products, a* value have shown positive relation with the formation of carcinogenic (acrylamide) compounds (Cao et al., 2020). Within the air-frying temperature range of 140°C-200°C, the increase of redness color tone of chicken nuggets and falafel has been reported in literature (Castro-López et al., 2023; Fikry et al., 2022). During AF of parfried-frozen products, a slight increase in yellow color tone was noticed with the increase of frying time. Castro-López et al. (2023) reported similar observation for non-frozen chicken nuggets. Due to changes in L a* b* color tones, the changes in total color difference (ΔE) value increased with the duration of AF. Notably, color attributes of air-fried coated products were impacted by the formulation of batter coatings. In compared to RB batter coated sample, WB coated sample was more prone (represented by higher ΔE) to color changes during AF. This could be linked with the presence of higher protein content in formulated wheat-flour based batter; as the color of fried food is strongly correlated to nonenzymatic browning (Maillard) reaction, and that requires protein entity (Castro-López et al., 2023; Cao et al., 2020; Rahimi et al., 2018).

Texture is one of the critical quality indicators of fried products, and crispy fried foods is preferred by consumers (Cao et al., 2020; Fikry et al., 2022). Figure 8.3 depicts, experimental data of the texture traits (hardness, ductileness, crispiness) of air-fried (at 190°C) products. Hardness (MF), ductileness (MD) and crispiness (S) of air-fried products were impacted by both the duration of air-frying and batter-formulation. After an initial sharp-drop in hardness of parfried-frozen samples (at the beginning of AF), the hardness increased as AF progressed. Initial sharp-drop of hardness of parfried-frozen samples could be considered as the "thaw effect", while the subsequent increase of hardness could be considered as the “moisture-loss effect”. It has been reported, the hardness of chicken nuggets and vegetable-based falafel increased with the duration of air-frying (Castro-López et al., 2023; Fikry et al., 2022; Cao et al., 2020). Similarly, crispiness and brittleness (opposite of ductileness) of air-fried products showed a positive relation with frying time. The observed relationship between AF time and textural changes of parfried-frozen samples could be understood as the events of heating-induced moisture-loss, starch-gelatinization and protein-denaturation. Overall, textural traits (hardness, brittleness, crispiness) of air-fried coated products were impacted by the formulations of batter systems, wherein the aforementioned attributes were prominent for formulated rice-flour based batter coated sample in compared to wheat-flour based batter.

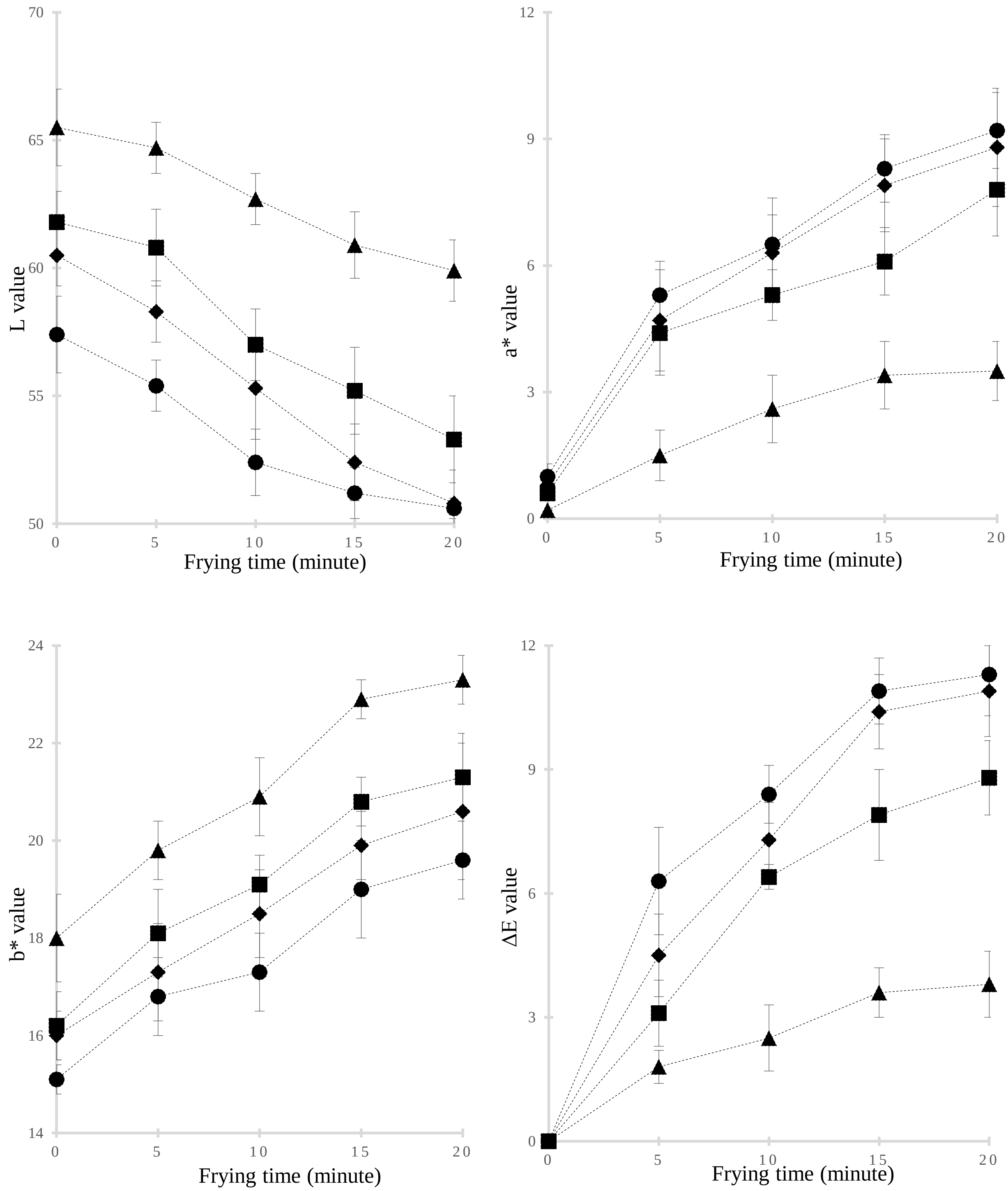


Figure 8.2**:** Changes in color attributes as function of frying-time at 190°C. NC (●), RB (▲), RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively.

Both the texture and color traits of fried products are known as prime quality indices in fried-food industry, as they play vital role in consumers acceptance and controlling the frying process (Castro-López et al., 2023; Fikry et al., 2022). Kinetic modelling of the changes in textural and color traits of parfried-frozen coated products could be useful, in predicting/optimizing quality characteristics of food prepared by the AF. To model color evolution as function of AF time, zero-order and first-order kinetic models (Eq 8.10 and Eq 8.11) were fitted to experimental data. Statistical parameter reveals (judging by R^2), the zero-order kinetic model fits well, compared to first-order kinetic model. Due to irregular pattern (i.e., first decrease, and then increase) in textural evolution, the zero and first order kinetic model was applied synchronously to characterize the evolution of textural traits of parfried-frozen samples. The initial sharp-decrease was satisfactorily modeled by first-order kinetic model, while the increasing region was modeled by zero-order kinetic model. It could be noted, first-order model was fitted to the data for 0 to 10 minutes of AF, while zero-order model was fitted to the data for the AF time of 5 to 20 minutes. Table 8.3 summarizes the rate constant (obtained from zero-order kinetic models) of color and textural changes at AF temperature of 190°C. Statistical parameter (R^2 >0.88) showed that, zero-order kinetic model can satisfactorily described the color (lightness, yellowness, redness, ΔE) and textural (hardness, crispiness, ductileness) changes of parfried-frozen batter coated products. Fikry et al. (2022) reported, the textural and color changes during air-frying of Falafel were better-fitted to zero-order kinetic model, compared to first-order kinetic model. For identical attribute, difference in rate constant (k, min^{-1}) values were noticed between WB and RB batter coated products. Higher rates of textural change (hardness, crispiness) were estimated for rice-flour based batter coated samples, while higher rates of color change (lightness, redness, total color) were estimated for wheat-flour based batter coated samples. The prominent textural changes of RB coated samples could be intertwined with the presence of higher starch in rice flour; while prominent changes in color attribute of WB coated samples could be associated with their higher protein (gluten) content.

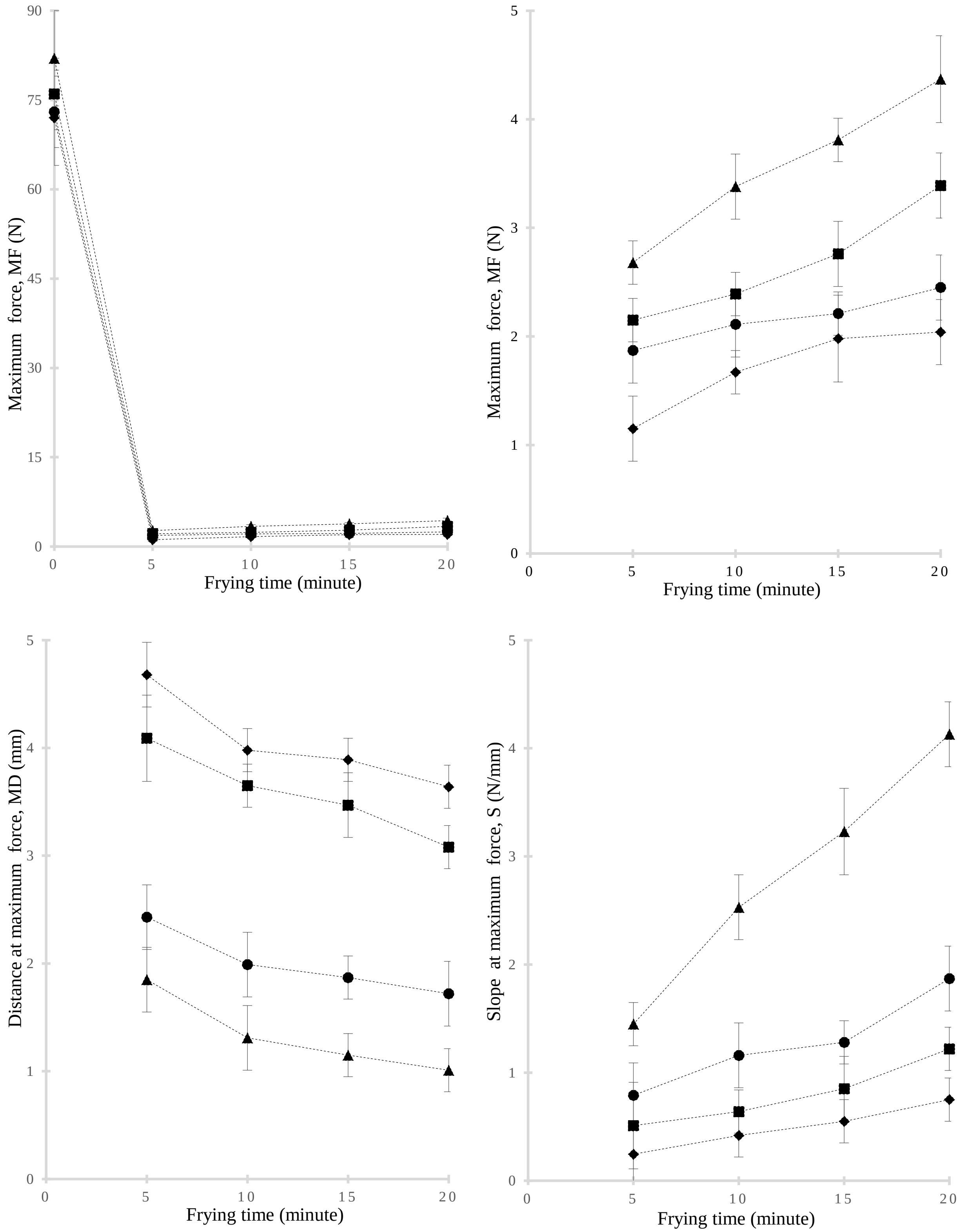


Figure 8.3: Changes in textural attributes as function of frying-time at 190°C. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively.

Table 8.3: Rate constant (k, min^{-1}) and R^2 value of the zero-order models for color and textural attributes.

Sample	Attribute						
	Hardness MF	Ductileness MD	Crispiness S	Lightness L	Redness a*	Yellowness b*	Total color ΔE
WB	+ 0.047 (0.893)	- 0.053 (0.865)	+ 0.022 (0.994)	- 0.505 (0.991)	+ 0.455 (0.926)	+ 0.243 (0.991)	+ 0.626 (0.978)
RWB	+ 0.049 (0.956)	- 0.045 (0.978)	+ 0.028 (0.949)	- 0.429 (0.973)	+ 0.390 (0.902)	+ 0.279 (0.971)	+ 0.499 (0.979)
RB	+ 0.077 (0.992)	- 0.044 (0.885)	+ 0.122 (0.994)	- 0.284 (0.982)	+ 0.193 (0.930)	+ 0.290 (0.970)	+ 0.219 (0.972)
NC	+ 0.026 (0.893)	- 0.036 (0.903)	+ 0.044 (0.937)	- 0.385 (0.941)	+ 0.467 (0.913)	+ 0.239 (0.970)	+ 0.673 (0.953)

NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. "+" and "− "signs denote increase and decrease, respectively. Numbers in parenthesis are coefficient of determination (R^2).

Temperature dependency of effective moisture-diffusivity and rate-constant of different physiochemical attributes were characterized by Arrhenius-expression (Eq 8.12 and Eq 8.13). Activation energy (E_a) is realized as the energy required for the starting of a process, and E_a value gives an indication about the temperature-dependency of the studied process (Rokib et al., 2021; Ghaitaranpour et al., 2018). In air-frying process, E_a could be understood as the energy needed for the beginning of moisture-diffusion as well as beginning of the changes in textural and color properties (Fikry et al., 2022). The natural logarithm (ln) of moisture-diffusivity (D_{eff}) / rate-constant (k) were plotted against the reciprocal of absolute temperature (K^{-1}); where the slope of straight line represents the ratio of activation energy (E_a) and universal gas constant (R) (Rokib et al., 2021; Adedeji et al., 2009). Negative slop of the generated-line indicates, rate constant (k) of color and textural changes of parfried-frozen products increased as AF temperature increased. Similarly, moisture diffusivity (D_{eff}) increased with the increase of AF temperature. These observation are analogous to relevant studies (Castro-López et al., 2023; Fikry et al., 2022).

Table 8.4, summarizes the estimated E_a values of different attributes. The coefficient of determination ($R^2 > 0.86$) value indicates, temperature-dependency of the studied attributes were well fitted to the Arrhenius-expression. Estimated higher E_a values, represents the relatively higher temperature dependency of moisture-diffusivity, compared to textual (hardness, crispiness, brittleness) and color (lightness, redness, yellowness, total color) changes. This signifies that in AF of parfried-frozen products, in compared to textural and color changes the moisture-loss event was much sensitive towards frying-temperature change. Estimated comparative ranks of temperature-dependency (i.e., moisture diffusivity> textural change> color change) during AF of parfried-frozen products are similar to the findings of relevant studies on other food categories, such as air-fried falafel (Fikry et al., 2022) and deep-fat-fried peas (Manjunatha et al., 2019). However, estimated E_a values (61.48-101.53 kJ/mol) for moisture-diffusivity of this study, are close to the reported E_a value (70.12 kJ/mol) for air-fried Falafel (Fikry et al., 2022). The observed higher E_a values could be understood as the effect of differences in composition and structure of studied samples as well as the difference in the initial state of the studied samples (frozen vs non-frozen). The substantial differences in E_a values (for different attributes) of RB and WB batter coated sample indicates, the formulations of batter-coating play crucial role during AF of parfried-frozen batter coated products. And their impact might be associated with the properties of formulated batter systems (i.e., water holing capacity, oil binding ability, protein content, starch content, etc.) as well as with the internal microstructure and surface properties of parfried-frozen samples.

Table 8.4**:** Activation energy (E_a, KJ/mol) of physicochemical attributes.

Sample	Attribute							
	Moisture diffusivity	Hardness	Ductileness	Crispiness	Lightness	Redness	Yellowness	Total color
WB	101.53 (0.91)	28.78 (0.95)	18.07 (0.98)	26.19 (0.96)	5.32 (0.94)	7.32 (0.98)	6.70 (0.95)	6.54 (0.97)
RWB	90.47 (0.92)	27.15 (0.97)	10.37 (0.97)	17.58 (0.97)	3.39 (0.94)	5.86 (0.95)	5.18 (0.96)	5.60 (0.97)
RB	66.76 (0.96)	18.26 (0.95)	10.21 (0.96)	16.37 (0.97)	6.32 (0.97)	3.83 (0.98)	3.51 (0.97)	2.74 (0.93)
NC	61.48 (0.93)	21.04 (0.94)	9.01 (0.97)	20.74 (0.96)	3.15 (0.86)	4.51 (0.95)	4.33 (0.93)	5.71 (0.97

NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively. Numbers in parenthesis are coefficient of determination (R^2).

Figure 8.4 depicts, surface microstructure of air-fried products. Surface structure of batter-coated samples were visually different than non-coated sample. Presence of surface openings, SO (hole, crack, rupture, etc.) in noncoated sample was higher than batter-coated samples; this explains higher moisture-loss of noncoated products during air-frying. Among coated products, higher surface openings were observed in RB coated sample compared to WB coated sample. This signifies the influence of batter-formulations on surface microstructure of air-fried products, and explains the distinctive rates (higher for RB and lower for WB) of moisture-loss during air-frying (Table 8.2).

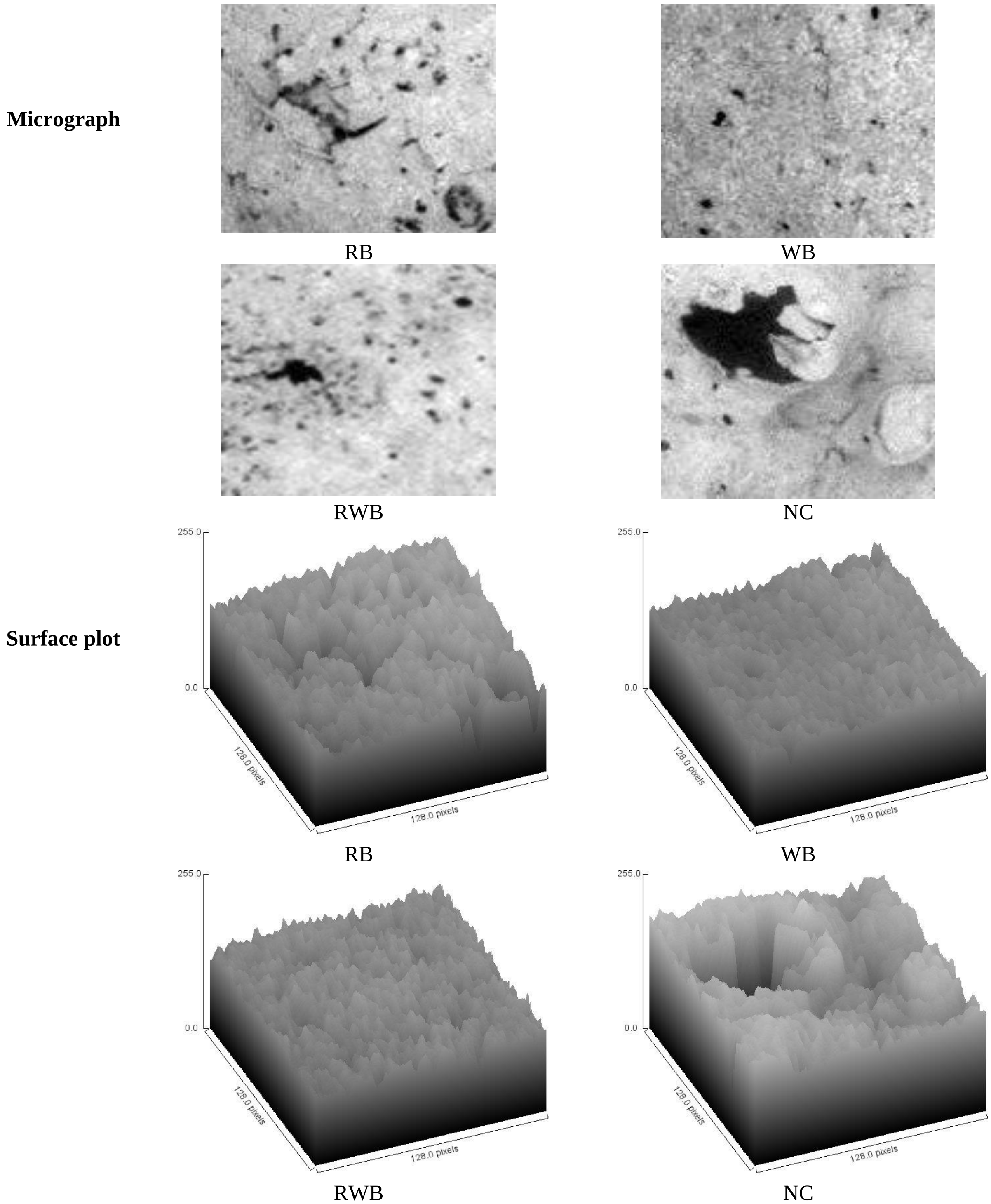


Figure 8.4: SEM micrographs and surface-plot depicting the surface openings and roughness of air-fried (190°C, 20 min) products. NC, RB, RWB & WB represent noncoated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food, respectively.

Surface roughness of fried food is one of the important textural features (Ghaitaranpour et al., 2018). To characterize surface-roughness of fried samples, fractal dimension (FD, unitless) has been used as an index of roughness (Rahimi & Ngadi, 2016; Ghaitaranpour et al., 2018). Figure 8.5 depicts the changes in surface microstructural properties (SO, FD) during AF at 190°C. Surface microstructure of parfried-samples changed during AF, and the changes were reflected as higher SO and FD values. Presence of higher SO in air-fried products indicates themself as the gateway of moisture removal (as vapor) from parfried samples under intense hot-air treatment. Overall, frying time were positively related to the SO and FD. Positive relation between frying time and FD of air-fried doughnuts crust has been reported; and observed surface structural changes were explained as the results of water-loss and creation of cracks during air-frying (Ghaitaranpour et al., 2018). It was observed, parfried-frozen WB batter developed smoother surface (lower FD) in compared to RB batter. This could be linked to their respective moisture-loss; a positive relation between moisture-loss and surface structural damages in deep-fat-fried batters has been reported in literature (Rahimi & Ngadi, 2016 & 2015). However, in this study, the estimated FD values of the air-fried products were found as comparably lower than the reported FD values of deep-fat-fried batter systems (Rahimi & Ngadi, 2016). This could be attributed to the differences in batter-formulations and the cooking methods (air-frying vs deep-fat-frying). Ghaitaranpour et al. (2018) observed that at identical frying temperature, air-frying process has produced smoother doughnuts crust surfaces in comparison to deep-fat-frying.

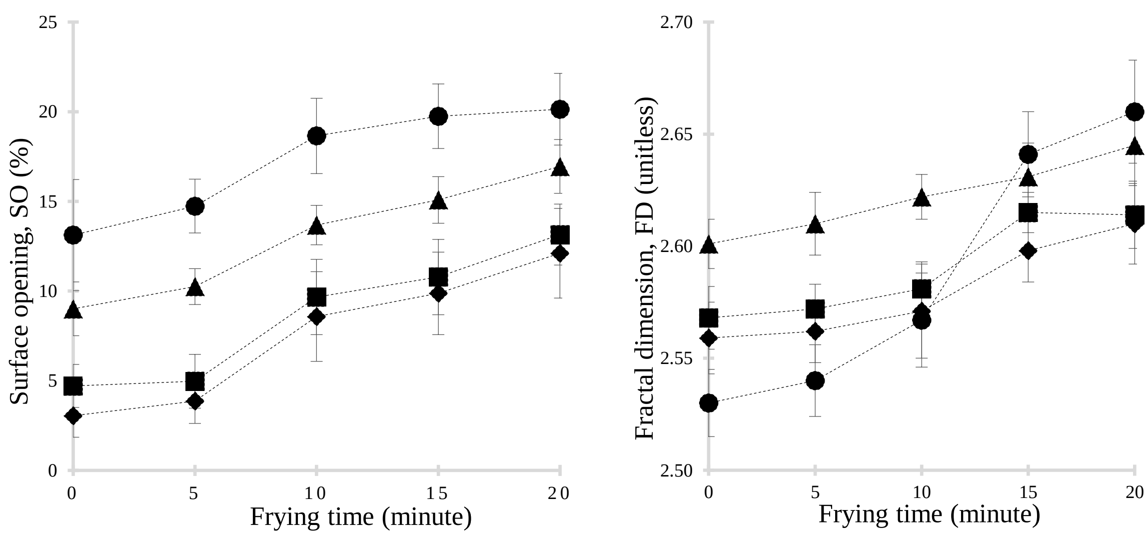


Figure 8.5: Changes in surface microstructural properties during air-frying at 190°C. NC (●) , RB (▲) , RWB (■), & WB (◆) represent non-coated, rice-flour based batter, wheat & rice flour-based batter, wheat-flour based batter coated model food sample, respectively. Symbol and dashed-line represents experimental values and prediction line, respectively.

8.5 Conclusion

Results showed that, both the batter-formulation and frying-parameter (time, temperature) has considerable influence on mass-transfer, surface-structure, texture and color development during the air-frying (AF) of parfried-frozen coated products. It is found, Newton, Page, modified Page, and Henderson & Pabis model can satisfactorily predict the moisture-loss of parfried-frozen products during the AF process. Zero-order kinetic models satisfactorily characterized the changes in textural (hardness, brittleness, crispiness) and color (L, a*,b*, ΔE) traits of parfried-frozen products during the AF process. In AF, temperature-dependency of both the moisture-diffusivity (D_{eff} , m^2/min) and rate constant (k, min^{-1}) were agreeably predicted by the Arrhenius-expression. Estimated activation energy (E_a, kJ/mol) values revealed that mass-transfer were comparatively more temperature-dependent, in comparison to textural (hardness, brittleness, crispiness) and color (lightness, redness, yellowness) changes. Studied quality attributes of air-fried coated products as well as parameters of the kinetic-models were substantially impacted by batter-formulations. Findings of this study could be useful in optimizing mass-transfer associated quality development during air-frying of batter coated parfried-frozen food product.

CHAPTER 9

GENERAL SUMMARY AND CONCLUSION

Presence of higher amount of fat in fried food is a major health concern. Food surface modification by batter-coating play effective role in reducing fat in fried products. In batter-coated product, changes in structural, thermal and physiochemical attributes may happen during post-fry stage. Therefore, maintaining the quality (that developed during frying) of batter coated products at post-fry stage, is crucial for achieving its consumer satisfaction. The main goal of this study was to get mechanistic insights on post-frying behavior of batter coated food products. In order to get deeper understanding of post-fry behavior, the evolution of structural, thermal, and physicochemical properties of batter coated fried food products were thoroughly studied. In this study, raw batter characteristics were assessed to know their subsequent influences on processing parameters. ATR-FTIR spectroscopy technique were used to study the post-fry surface chemical nature of fried products. Scanning electron microscopy (SEM) based image analysis was performed to characterize the surface microstructural properties of the fried products. Surface color were evaluated by CIELab calorimetry method. X-ray µCT technique was used in obtaining internal microstructural information of fried foods, and statistical approach i.e., Multifractal analysis (MFA) were performed to correlated internal pore-structure with textural properties. Mechanical texture analyzer was used in assessing textural (hardness, brittleness, crispiness) evolution of fried products. The glass-transition behavior of fried products were evaluated by differential scanning calorimetry (DSC). Deep fat frying, microwave heating, air frying, and infrared heating were employed as cooking methods for the preparation of batter coated products. The processed products quality evolution were thoroughly investigated and mathematically modeled. The quality evolution of products at room environment and under IR heating environment, were studied.

This research was focused on understanding and characterization of the post-frying behavior of batter coated food product. To maintain uniformity in the properties of core substrate, a meat-analog was used as model food. wheat and rice flour based tempura batter systems were used to coat the model food substrate. The specific objectives of the research described in this thesis are (i) study the influence of batter solid-to-water ratio and frying time on structural, thermal and physicochemical properties of coated fried food (ii) investigate the impact of post-fry de-oiling treatments on oil-redistribution in batter coated fried food (iii) characterize the relationship among internal microstructure, mass redistribution and textural evolution of batter coated fried food (iv) study the glass-transition behavior of batter coated fried foods in relation to post-frying textural evolution (v) study the impact of finish-cooking methods on structural, thermal and physicochemical evolution of parfried frozen batter coated food (vi) study the kinetics of mass, texture and color evolution of parfried frozen batter coated food.

The following conclusions were drawn from this research:

(i) Batter coatings of different solid-to-water ratio (SWR) possesses distinctive rheological properties and water retention ability; that influences batter pickup, frying loss, and cooking yield of coated food. Batter coatings of higher SWR developed crust of having higher textural attributes (hardness, brittleness, crispiness) and stable color features. Lower SWR was in favor of the retention of juiciness of food-core substrate, in coated fried foods. Higher textural and color stability of fried foods' crust were resulted by longer frying time (FT). Fat content of coated product increases with the increase of FT. Both SWR and FT considerably influenced fried foods' crust microstructure (surface opening, roughness) and glass-transition temperature (Tg). Higher SWR and FT substantially reduced post-fry mass redistribution and textural evolution. Lower SWR and FT favored retention of the juiciness of food core substrate, which caused to result in undesirable post-fry changes in textural properties. Rice-flour based batter has shown higher responsiveness (moisture-fat interaction during frying) to the modification of SWR and FT, whereas wheat-flour based batter coated food has shown proneness to post-fry physicochemical changes. Modification of batters' SWR and FT could be used to produce fried products of having desirable and stable quality traits.

(ii) A novel method (based on ATR-FTIR spectroscopy) have been established to characterize the surface chemical nature of batter-coated fried foods. ATR-FTIR spectra based method have shown consistency with the conventional method of assessing surface attributes of batter-coated fried foods. Surface fat (SF) of coated fried foods are related to both batter formulations and frying time. Post-fry de-oiling treatments reduced the SF, matrix fat (CrMF) and consequently crust total fat (CrF) content. Surface structure-chemical characteristics impacts the post-fry oil redistribution in batter coated fried food. Batter-formulations and frying-time influences the surface microstructural-chemical nature and the effectivity of post-fry de-oiling treatments. Post-fry surface de-oiling treatments can reduce oil from fried batter coated foods, without distorting textural and color attributes. Effectiveness of post-fry de-oiling treatments are related to surface microstructure of coated foods. The impact of post-fry surface de-oiling treatments are more obvious for samples with higher SF. Centrifuge-assisted paper absorbent (CAP) treatment have shown highest impact in reducing fat (surface, matrix, total) from batter coated fried foods, followed by pressure-assisted paper absorbent (PAP) and hot air blow (HAB) treatment.

(iii) Batter-formulations and frying time significantly impacts pore formation (% porosity) in coated fried food, and consequently affects post-fry mass distribution and textural evolution. Pore formation in batter-coated foods is positively correlated with frying time. Pore distribution have considerable influences on mass (moisture) re-distribution between crust and core region of batter-coated fried products during post-fry holding; that consequentially affects textural attributes. Compared to wheat-flour, lower extent of post-fry moisture re-distribution and higher textural stability were observed in rice-flour based batter coated samples. Presence of higher pore space, lower moisture, and higher fat was in favor of stable textural attributes of batter-coated products; as these could limit moisture redistribution between core and crust region. X-ray microtomographic images based Multifractal analysis revealed, pore structure in coated products are highly impacted by spatial location. Pore structure of batter-coated fried foods possesses multifractal scaling behavior and pore distribution are generally inhomogeneous. Higher heterogeneity in pore distribution were present in lower concentration of pores at crust region, compared to core region of batter-coated fried food. Increase of frying time have shown impact in favor of homogeneous pore distribution in crust of batter-coated fried foods, and crust having homogenous pore structure were comparatively stable against post-fry mass redistribution and textural evolution.

(iv) Batter-formulations and frying time are crucial parameters for glass-transition-temperature (Tg) of fried batters. Tg of fried batters are linearly correlated with the duration of frying. Both moisture and fat acts as plasticizer in batter coated fried foods, that results in subzero Tg of fried batters. Positive correlations were observed between frying time, fat content, porosity and Tg, in contrast, moisture content was negatively correlated to these attributes. Crust moisture, texture and Tg of fried-batter changes during post frying holding, where the external IR-heating has shown some extent of influence. Subzero Tg value explains the moisture-redistribution induced textural changes in fried foods batter coatings during their post-frying holding at room environment (25°C) and under external IR-heating (65°C). It could be suggested that modification of moisture-fat profile and internal-structure (through modification of batter-formulations and frying process) would be an effective approach to obtain (via modifying glass-transition-temperature) desired and stable textural qualities in batter-coated fried foods.

(v) Compared to conventional deep fat frying (DFF), microwave heating (MH), infrared heating (IH) and air frying (AF) as alternate finish-cooking method causes higher finish-cooking loss of parfried-frozen batter coated food. MH, IH, AF significantly reduces spatial (crust, total) fat, compared to conventional DFF of parfried-frozen products. Parfried frozen-batter is effective in reducing fat uptakes during finish-cooking by DFF. Spatial (crust, total) moisture profile of finish-cooked coated products are intertwined with batter-formulations and finish-cooking method. Evolution of textural and color attributes of parfried-frozen coated foods are associated with batter-formulations and finish-cooking methods. Rice-flour based parfried frozen batter coating develops bright-yellowish color and crispy crust during finish-cooking. AF is a suitable substitute of DFF, to finish-cook the parfried frozen-batter coated foods. MH and IH could be considered as healthier choice compared to DFF, while, in terms of textural attributes the later method has shown preference over the former two.

(vi) Both the batter-formulation and frying-parameter (time, temperature) has considerable influence on mass-transfer, surface-structure, texture and color development during air-frying (AF) of parfried-frozen coated foods. Newton, Page, modified Page, and Henderson & Pabis model can satisfactorily predict moisture-loss of parfried-frozen foods during AF. Zero-order kinetic models satisfactorily characterizes the changes in textural (hardness, brittleness, crispiness) and color (L, a*,b*, ΔE) traits of parfried-frozen products during AF. In AF, the temperature-dependency of moisture-diffusivity (D_{eff} , m^2/min) and rate constant (k, min^{-1}) could be agreeably predicted by the Arrhenius-expression. Estimated activation energy (E_a, kJ/mol) values revealed that mass-transfer are comparatively more temperature-dependent, in compared to textural (hardness, brittleness, crispiness) and color (lightness, redness, yellowness) changes. Quality attributes of air-fried coated products as well as parameters of the kinetic-models are substantially impacted by the batter-formulations. Surface microstructural properties of air fried products impacted by batter formulation and AF parameter.

CHAPTER 10

CONTRIBUTION TO KNOWLEDGE AND RECOMMENDATION FOR FUTURE RESEARCH

The presence of high amount of oil in fried foods, is a major health concern. “Batter-coating” is a technique that impacts heat and mass transfer process (during frying) to reduce fat in fried products and develops peculiar (juicy inside & crispy outside) textural attributes in fried foods. Post-frying quality of batter-coated foods are of crucial importance relating to its consumer satisfaction. Unexpected changes in structural, thermal and physicochemical properties in batter-coated fried foods negatively influences its consumers choice. These aspects need to be properly understood and investigated, in applying batter-coatings to produce low-fat containing fried products as well as to ensure their post-frying stability of quality attributes. This study provides mechanistic insight on structural, thermal and physicochemical properties of fried batter-coatings, in favor of the production of low-fat containing fried product. Knowledge obtained from this study will mostly be applicable in developing and maintaining the quality of low-fat containing batter-coated fried foods, and it is expected to contribute immensely on following aspects:

1. To the post-fry oil reduction from fried food products, without affecting their textural quality and color attributes.

2. To customize the production process (through batter formulation and/or process modification) of batter coated products, in achieving desired stability of quality attributes at post-fry stage.

3. To optimize the alternate cooking-methods (air frying, microwave heating, infrared heating) in final preparation of parfried-frozen batter coated food products, as effective oil reduction strategy.

4. To customize the foods surface characteristics by regulating batter-formulations and processing parameters in order to achieve desired oil-reduction functionality of the batter systems.
5. To improve/modify the textural quality of batter-coated fried foods, by regulating internal microstructure.

6. To customize the stability of batter-coated fried product quality traits (specifically texture) through controlling the glass-transition temperature of finish fried foods.

7. To utilize FTIR spectroscopy based techniques in simultaneous study the distributions of different components (specifically moisture-fat) in other fried food products.

From this work the following areas of further research have been identified:

1. In this study, laboratory formulated vegetable-protein based restructured meat-analog model was used as food-core substrate. Study on the interaction between real food (as food-core substrate) and batter-coatings could be a horizon of future research.

2. This study considered wheat and rice flour-based batter coatings. Post-frying behavior of other flour (corn, rye, maize, oats, barley) based batter coatings could be studied in future.

3. This study revealed internal microstructural information of fried foods in relation to their textural evolution. These information could be used in developing customized (e.g., 3D printed food) and low fat-containing fried foods, that would have desired textural properties.

4. This study gathered information about the glass-transition-temperature (Tg) of fried food batter coatings. Customizing the post-frying stability of the quality (textural) attribute of coated fried foods by modifying the Tg could be a topic for future research.

5. Frying-oil becomes an unavoidable constituents of batter-coated fried foods. Enrichment and/or fortification of frying-oil with fat soluble micronutrients could improve nutritional status of batter coated fried foods. However, their functionality on sensorial quality (e.g., texture, flavor, color, juiciness) needs detailed investigation and that could be a topic for future research.

6. This study investigated the comparative effects of electromagnetic, hot-air and deep-fat frying as “finish-cooking” method of parfried frozen foods. Use of other novel cooking techniques (g-frying, microgravity-frying, etc.) in producing batter-coated foods could be a topic of future research.

CHAPTER 11

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APENDIX

Model Food