AN ENGINEERING ANALYSIS OF THE THERMOPLASTIC FILM EMBOSSING PROCESS

by

Andrew Haber

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Department of Chemical Engineering McGill University Montreal, Canada

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ABSTRACT

Film embossing is a mechanical process in which a flat film is transformed into an embossed product. During the process, thermal and stress fields are applied to the polymer, causing changes in the microstructure and physical dimensions of the material. The engineering analysis of the process requires the study of various aspects relating to the characterization of the microstructure before and after embossing. A variety of experimental techniques were employed to characterize the properties and microstructure of embossed films in relation to: crystallinity, orientation, mechanical properties and dimensions of the embossed films. Mathematical models were also developed to predict the temperature, stress and deformation in the plastic films during embossing.

The thermal treatment of the plastic film was shown to be the most significant factor in the process. The important aspects influencing the thermal treatment include the radiation heater temperature, preheat roll temperature, line velocity and film thickness. In general, the heat transfer mathematical model predictions were in agreement with the experimental data. The stress and deformation analysis, suggested that the embossing pressure and the rubber layer on the backup roll are significant factors in determining the dimensions and the stress distribution in the nip region.

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RESUME

Le gaufrage d'un film est un procédé mécanique dans lequel un film plat est transformé en un produit gaufré. Durant le procédé, des champs thermiques et de tension sont appliqués polymère, causant des changements dans sur le la microstructure et les dimensions physiques de la matière. L'analyse technogéniques du procédé requiert l'étude de différents aspects ayant rapport à la caractérisation de la microstructure avant et après le gaufrage. Une variété de techniques a été utilisée pour caractériser les propriétés et la microstructure de films gaufrés par rapport à: la cristallinité, l'orientation, les propriétés mécaniques et les dimensions des films qaufrés. Des modèles mathématiques ont aussi été développés afin de prédire la température, la tension et la déformation du film de plastique pendant le gaufrage.

Le traitement thermique du film de polymère s'est avéré le facteur le plus significatif du procédé. Les aspects importants influençant le traitement thermique comprennent en outre la température du chauffage par radiation, la température du rouleau pré-chauffé, la ligne de vélocité et l'épaisseur du film. En général, les prédictions du modèle mathématique du transfert de la chaleur concordèrent avec les données expérimentales. L'analyse de tension et de déformation ont montré que la couche de caoutchouc sur le rouleau de réserve et la pression du gaufrage sont des facteurs significatifs dans la détermination des dimensions et de la distribution de tension dans la région du resserrement.

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NOMENCLATURE

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<u>Symbol</u>	Definition	<u>Units</u>
A	Area	m ²
a_c	Punch Radius	m
2B	Nip Region Length	m
Bi	Biot Number	
BUR	Blowup Ratio	
C _⊽	Specific Heat	J/kg°C
d_c	Cylinder Diameter	m
$\mathbf{E}_{\mathbf{i}}$	Young's Modulus Material i	Ра
F	Shape Factor	
G'	Storage Modulus	Pa
G"	Loss Modulus	Ра
h	Heat Transfer Coefficient	W/m ²⁰ C
н	Film Thickness	m
k	Thermal Conductivity	W/m°C
k	Power Law Coefficient (Equation D-1)	Pa-s ⁿ
L	Length	m
MD	Machine Direction	
n	Power Law Index (Equation D-1)	
n	Sample Size	
N	Number of Increments	
N '	Applied Force	N/m
ND	Thickness Direction	

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<u>Symbol</u>	Definition	<u>Units</u>
Р	Pressure	Ра
đ	Heat Flux Vector	
R_i	Radius Roll i	m
t	Time	s
Т.	Temperature	°C
TD	Transverse Direction	
U	Displacement	m
v	Velocity Vector	
v	Velocity	m/s
W	Width	m
w	Weight	kg
x _c	Crystallinity	8
XYZ	Cartesian Co-ordinates	
rθz	Cylindrical Co-ordinates	
ijk	Finite Difference Co-ordinates	

Subscripts:

Α	Ambient	
am	Amorphous	
cr	Crystalline	
F	Film	
MIN	Minimum	
MAX	Maximum	
0	Initial	
Р	Preheat Roll	



<u>Symbol</u>	Definition	<u>Units</u>
R	Radiation Heater	
S	Surface	
XYZ	Cartesian Co-ordinates	
rθz	Cylindrical Co-ordinates	
ijk	Indices	

Greek Symbols

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ρ	Density	kg/m ³
ω	Frequency	s^{-1}
τ	Shear Stress	Ра
τ	Shear Stress Tensor	
γ	Shear Strain	8
σ	Normal Stress	Ра
ε	Emissivity	
α	Thermal Diffusivity	m²/s
υ	Poisson Ratio	
δ	Punch Penetration	m
λ,μ	Constants	Pa
$\dot{\gamma}$	Shear Rate	s ⁻¹
n	Shear Viscosity	Ра

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Chapter 1 INTRODUCTION

1.1. GENERAL INTRODUCTION

The dictionary definition of embossing is [1]:

"To cause to swell or protrude. To raise in relief from a surface either by carving, handiwork or more commonly by mechanical means by pressing against a steel die or engraved roller".

Embossed patterns may be two or three dimensional and can vary in size from several micrometers to centimeters. The raised or indented patterns used are often repeating geometrical shapes such as squares, rectangles, circles, lines or diamonds. The embossed pattern may appear on one or both sides of the final product. Furthermore, the surface being embossed can be of any thickness.

Steel is the material most frequently embossed. The common characteristic of all steel embossments is the well defined localized stretching of the material to create the desired shape. The most common way to identify steel embossing operations is by the function performed by the embossment [2]:

<u>Stiffening</u>: Beads and ribs are used to stiffen flat areas of sheet metal. These are long narrow depressions embossed in the sheet metal. <u>Functionality</u>: Often the product designer does not want the appearance of stiffening beads, instead a series of offsets are added for stiffness and other functional needs. The offset may be used to provide clearance for other components. <u>Decorative</u>: The embossments are intended for decorative purposes, such as for trade names, emblems or product specifications.

A plastic film can be defined as a planar sheet, thick enough to be self-supporting and thin enough to be flexed or creased without cracking. Depending on the characteristics of the polymer being used, plastic films generally have a thickness of 0.5 mm or less [3]. The embossing of plastic films can also be classified according to the function performed by the embossment:

Texturizing: The embossments are intended to change the texture of the flat plastic film. Plastic films are embossed to provide aesthetic softness. Often, properties associated with textile products, such as low gloss, smoothness, softness and limpness, can be imparted to the film by embossing. Property Modification: The embossing process can alter and possibly improve the optical, mechanical and thermal properties of the flat film. Surface Modification: The embossments can improve the surface characteristics of plastic films. Embossed films do tend not to stick together when in contact with other films or surfaces. The embossments also lower the tendency of a film to crease or wrinkle. The embossing process can be used to transform a film having surface defects into a film having an acceptable surface by concealing any processing imperfections.

The thermoplastic film embossing process offers an alternative method of producing plastic sheets and films with properties and surface patterns which can replace consumer products previously made of paper and foil. One particularly large scale use of embossed plastic films is for the fabrication of

disposable articles such as hospital pads, tablecloths, feminine pads, diapers and bags. Embossed films are also finding increased use in the packaging field, as protective coverings. The majority of embossed plastic films are made from polyethylene, polypropylene and polyvinyl chloride resins.

The production of an embossed film is usually only one of the many stages involved in the manufacture of the final product. For example, during the production of disposable diapers, the embossed film is only one of several components making up the final product. The embossed film serves as a waterproof backsheet. In this application, the film is combined with a nonwoven absorbent cellulose fiber layer to prevent any liquid from passing through the absorbing layer. During processing, the embossed film is glued to a nonwoven lining, cut to the desired shape, folded and packaged. The embossed film must not only withstand the stresses applied during the embossing process, but also those occurring during the diaper manufacturing stage. For this reason, the dimensions of the embossed film and physical properties must be carefully controlled during the embossing process.

1.2. THE FILM EMBOSSING PROCESS

The film embossing process can be classified as a secondary forming mechanical operation in which a flat plastic film, with a microstructure determined by the thermo-mechanical history experienced during the film forming operation, is transformed into an embossed product.

The simplest method of embossing a thermoplastic sheet continuously is by passing it between two rolls, one of which is hot and incorporates the design [4]. This process is known as hot embossing. The technique which is most widely used however, is that of feeding a hot sheet between two cooled rolls in contact [4]. This process is known as cold embossing. The temperature of the plastic film prior to passing between the two rolls must be high enough so that the film can be deformed easily before it freezes.

Embossing schemes can be classified as either continuous or batch. Continuous cold embossing systems can be of two types. In cast film embossing a plastic film is extruded directly into the nip region formed by the contact of the two embossing rolls. In the tubular film embossing process, a plastic film produced by the tubular blown film process is preheated prior to entering the nip region of the embossing system. Batch cold embossing systems consist of preheating rolls of plastic film that have been fabricated using either the tubular blown or cast film process prior to passing through the embossing zone. Continuous hot embossing systems apply only to tubular blown film processes. The most common hot embossing system, however, is the batch system, which can process either tubular blown or cast films. An advantage of the batch process over the continuous process is that the production rate using the batch process is not limited by the extruder output rate, as is the case with continuous processes.

In practice, the batch and continuous schemes have been modified to give a variety of processing configurations, which may be used to produce different types of embossed products. The remainder of this discussion will focus on cold embossing systems.

1.2.1. Cold Embossing Systems

A plastic film can be thermally treated using conduction, convection or radiation heaters. The most common types of heating systems used in embossing systems [5-7] are as follows: <u>Preheat Roll:</u> Oil or water is used to preheat a metal roll to 90-100°C. The film is heated by the conduction of heat from the preheat roll surface to the plastic film. Heat loss by convection occurs at the film free surface.

<u>Radiation Heater:</u> Radiation tube heaters are used to heat the film to 90-110°C. The film is heated by radiation absorbed at the surface, which is then conducted into the interior of the film. Heat transfer due to convection also occurs at the film free surface, due to the heating of the surrounding air by the radiation heaters.

<u>Convection Heater</u>: Forced air heaters are used to heat the film to 90-110°C. The film is heated by convection at the free surface. Subsequently, heat is conducted into the interior of the film. Air, heated by electrical or gas sources, is blown across the surface of the film.

The embossing unit can employ either contact or vacuum techniques, Table 1-1. Due to its simplicity, engraved roll embossing is the most commonly used configuration. When using such a system, the roll pattern can be transferred to the film in the following ways:

<u>Two engraved embossing rolls</u>: Two engraved embossing rolls are used to impress the desired pattern into the film. For thick sections, the patterns on the two rolls do not need to be compatible. However, for thin films, the embossing patterns must intermesh properly to avoid processing problems. The main limitation of this configuration is the difficulty in controlling the position of the two rolls to avoid misalignment of the pattern. In both cases, the two rolls do not come in contact and the gap between the rolls is kept constant.



Embossing Screen

Vacuum

Applied

Engraved Roll Embossing: A heat treated plastic film is passed between the nip of two embossing rolls in contact, of which at least one carries the embossing pattern [5-9].

<u>Vacuum Embossing</u>: A heat treated plastic film is passed over an embossing screen. A vacuum is applied to pull the film into contact with the screen to impress the embossing pattern into the film [8,10,11].

Polymer Film Support Roll Embossing Belt

Support Rolls

<u>Belt Embossing:</u> A continuous belt, containing the embossing pattern, presses the heat-treated polymer film against a rotating backup roll to impress the pattern into the film [12].

Table 1-1: Types of Film Embossing Systems.

Engraved metal roll with a metal backup roll: This configuration can only be used in cases where one surface is being embossed. Furthermore, a gap exists between the two rolls.

Engraved metal roll with a rubber covered backup roll: The metal roll deforms the rubber roll and a two-sided embossing pattern is achieved. The resulting embossed film has distinctive upper and bottom surfaces which depend on the applied pressure.

An engraved metal roll and rubber covered backup roll combination is the most commonly used system for embossing plastic films. Due to the deformation of the rubber covered backup roll, an embossed pattern appears on both sides.

The most common cold embossing schemes used in industry are based on continuously embossing either tubular blown or cast thermoplastic films using the engraved metal embossing roll and rubber covered backup roll design.

1.2.1.1. Tubular Film Embossing Systems

Early embossing operations were based on the tubular blown film process, due to the considerable expertise which existed with respect to the design, installation and operation of this process.

The tubular blown film process is widely used for the production of thin thermoplastic films [13]. The process consists of extruding a tubular parison from a circular die. Air is simultaneously injected within and around this tubular envelope to expand, orient and cool the extrudate into a blown film. The resultant film is collapsed into a flat sheet. Then it is edge-slit to provide two film sheets of equal width, as illustrated in Figure 1-1.



Air Supply

Figure 1-1: Tubular Blown Film Process Schematic.

Tubular blown film systems produce films with superior mechanical properties, as compared to films produced using the cast film process [8,13,14]. Consequently, tubular blown films exhibit a higher gloss and stiffness as compared to cast films. The width of tubular blown films can be easily controlled and larger layflats are possible in comparison to cast films. The installation and operating costs of tubular film systems are higher than those for a cast film system. The higher costs are due to the lower output rate and the additional hardware required to expand and thermally treat the film prior to embossing. Overall, the tubular blown film process is easier to operate than a cast film line.

1.2.1.2. Cast Film Embossing Systems

The cast film extrusion process utilizes a conventional slot extrusion die system to extrude the thermoplastic resin in the form of a thin sheet, which is then cooled either by passing over a cooling roll system or through a quench bath. The single web is subsequently edge-slit and wound.

Cast film embossing systems produce high quality embossed patterns at high output rates [8,13,14]. The mechanical properties of cast films are lower than the mechanical properties of tubular blown films, having the same thickness and using the same resin, due to the uniaxial deformation of the film. In addition, due to the higher cooling rates achieved using the cast film process, the optical quality of these films is better than that of tubular blown films. Film thickness is more uniform in cast films than in tubular blown films, since the die gap can be easily controlled in the cast film process. A problem in the cast film process is the possible adhesion of the film to the rubber backup roll at high film temperatures, thus limiting the output rate. This problem can be somewhat alleviated by using a water bath to cool the rubber backup roll. In addition, it is possible to add release agents to the water bath to further reduce film adhesion. Finally, cast processes offer new product possibilities in the area of co-extrusion, extrusion coating and thick films.

1.2.1.3. Batch Embossing Systems

Batch embossing systems are designed to process prefabricated rolls of plastic film. The flat film is thermally treated to raise the temperature close to the softening point of the polymer, which is then embossed to impress the pattern into the film. The analysis of a batch embossing system is similar to that of the tubular blown film embossing process.

1.3. PREVIOUS WORK

Apart from the extensive literature available directly related to blown film or cast film extrusion processes, no comprehensive study has been published to date which analyzes any type of plastic film embossing operation.

The majority of the published literature [5,6,9-12,15-20] is related to patent applications. These inventions relate to roll design, methods to thermally treat plastic films, methods to emboss plastic films and new applications for embossed plastic films. Only information relevant to the invention is presented in these patent applications.

The few published studies related to the embossing of plastics, focus only on very general aspects of the process. Hamel [21] discussed how to extrude embossed flexible profiles. An engraving wheel was located at the end of the extruder die. As the extrudate passed through the die it turned the engraving wheel, thus eliminating any mechanical drives. The embossed extrudate was cooled upon leaving the die. This process differs from the typical cast film embossing process in that the engraving wheel is part of the die, no backup roll was used and a gap existed between the extrudate and embossing wheel. The embossed pattern was also impressed only onto the top surface of the plastic extrudate. This process is applicable only to thick profiles and large patterns, up to 2.5 mm. Finally, no mention was made of how the embossing process variables were related to the final properties of the extrudate or how the pattern was transferred to the extrudate.

Process configurations of the type described by Hamel [21] are commonly used to emboss rigid polyvinyl chloride siding profiles. The extrudate leaving the die is embossed on one side to simulate a wood grain pattern on the part. The embossed sheet, whose temperature is still greater than the softening point of the material, is then shaped into the desired form using a mold and subsequently cooled to freeze it in that shape.

Akay [22] investigated the roll embossing of polypropylene films. Roll embossing was employed as an intermediate operation between melt casting and drawing a fiber from a film extrusion line. The embossing pattern consisted of concentric circles which transformed the flat cast film into a series of strands which were then drawn into fibers. The embossing roll was heated (i.e. hot embossing) to facilitate the forming of the fibers using the flat film. Material structure variations were identified using small angle x-ray scattering, small angle light scattering and wide angle x-ray diffraction techniques. Static and dynamic mechanical tests were performed using an Instron Universal Test Machine and a sonic apparatus. Roll embossing was shown to improve the tenacity of the final drawn tape by as much as 15%. The increase in mechanical strength was considered to be a result of the improved crystalline structure and longitudinal orientation in the plastic film. This process differs from the cast film

embossing process in that it is related to fiber production and not film texturing. Furthermore, the plastic film in most embossing applications is not drawn to alter the orientation or reduce the thickness, as was the case for this application.

Roberts, Lucchesi and Kurtz [23] discussed the problem of draw resonance in melt embossing, extrusion coating and extrusion processes. Draw resonance was defined by these authors as a sustained periodic nonuniformity of the plastic film gauge or weight in the machine direction. A draw resonance eliminator was developed by the authors to overcome draw resonance when processing linear low density polyethylene and high pressure low density polyethylene using a cast embossing system. Draw resonance was found to be a phenomenon that required two fixed points: the extrusion die and the embossing nip. It was found to occur at a critical draw ratio of 35:1, when producing a 25 μ m thick plastic film. Eliminating draw resonance resulted in higher output rates and higher draw ratios. The mechanical properties of the embossed films were compared keeping the line speed and film thickness constant for both resins. The low density polyethylene film exhibited superior puncture, impact and tear strength. No attempt was made to consider the effect of varying the embossing conditions.

Stein, Tse and Melngailis [24] investigated the Summa, embossing of square wave gratings into solid plastic films. The embossing pattern was transferred to the plastic film using a hydraulic press, i.e. stamping process. Summa et al presented an analytical model based on the closed die forging operation to describe the process, predict the approximate forming pressure and predict the temperature distribution. To preserve the piezoelectric properties of the material used, i.e. polyvinylidene fluoride, the maximum processing temperature possible was found to be 70°C. The analysis of this process was divided into two steps. The first step was considered as a backward extrusion step in which material was

pushed out from under the punch and into the channel. In the second phase, additional pressure was applied to fill the corners of the projection. The second phase was found to be the controlling step in determining the quality of the embossed grating. The additional pressure required during this phase determined the minimum forming load.

Kamal, Haber and Ryan [25] developed mathematical models of the dynamics and heat transfer of the tubular blown film embossing process. The thermal analysis around the preheat roll was determined using an unsteady state, two-dimensional heat conduction equation along with appropriate boundary conditions. The heat transfer occurring between the preheat roll and the embossing rolls was based on a one-dimensional analysis, including both convective and radiative effects. The theoretical treatment provided a reasonable description of the thermal history of the plastic film up to the nip region of the embossing roll. Chapter 4 describes in detail the mathematical models developed. The deformation in the nip region was analyzed for two different situations, where the the embossing roll and backup roll are separated by a fixed distance. For the case where the surface features, i.e. embossments, are small in comparison with the film thickness, a modified one-dimensional calendering analysis was given. For the case where the plastic film does not make complete contact with the surface of the embossing roll, the local deformation was determined by means of a simple one-dimensional cavity filling model. The analysis for the case of a Newtonian and power law model was considered.

The above studies illustrate the limited nature of the work done to date regarding thermoplastic embossing processes. The first three studies [21-23] focused on the cast film process and did not attempt to determine the interrelationships among the process variables or their effects on the embossed product. The study by Summa et al [24] considered compression molding embossing techniques, which are not used to produce commercial embossed films. Finally, the study by Kamal et al [25] studied the case where the two embossing rolls are separated by a fixed distance. This geometry is not the same as that used in the tubular blown film embossing process, where the embossing roll and backup roll are in contact. Ultimately, what is needed is a comprehensive analysis of the film embossing process.

1.4. ENGINEERING ANALYSIS

A polymer processing system is a combination of operations which affects the transformation of a resin, usually in the form of pellets or powder, into a commercial product. This transformation may be viewed as a multi-step process involving solids transport, heating, melting, mixing, melt conveying, shaping, cooling, solidification and finishing. The thermomechanical history experienced by the material during processing determines the microstructure developed in the finished part, which in turn, determines the ultimate properties of the final product. Figure 1-2 shows the various components of a plastics processing system. Generally, the engineering analysis of processes is carried out in order to:

(i) determine the significant process variables and factors which affect the final product properties,(ii) manipulate and control the final product properties,

(iii) optimize processing conditions to minimize material consumption and manufacturing costs,

(iv) design equipment,

(v) trouble shoot processing problems and

(vi) control the process.

An understanding of the various components comprising the film embossing process requires a combination of experimental and



Figure 1-2: <u>Components of a Polymer Processing System.</u>

mathematical modelling techniques. For example, the cold embossing engraved roll process can be subdivided into the following four parts:

(a) Flat Film Analysis: This is the starting point for either a batch or continuous embossing operation. At this point, the flat plastic film is characterized by a microstructure which is the result of the thermo-mechanical history experienced by the material in the extruder, die and film forming operation.

(b) Heat Transfer Analysis: The film is heated or cooled to a specified temperature which is close to the melt temperature of the polymer prior to embossing. The film is subsequently cooled on the embossing roll.

(c) Stress Analysis: The film is exposed to stress fields as it passes through the nip formed by the contact of the engraved embossing roll and the rubber covered backup roll.

(d) Embossed Film_Characterization: The end point of the process is the final embossed product with characteristic microstructure, geometry and properties.

In points (a) and (d) the solid state properties of the thermoplastic film must be determined experimentally and related to the processing conditions. In points (b) and (c) the relevant thermal and stress fields must be experimentally measured. In many cases, it is difficult to make direct measurements of certain quantities. For example, due to the physical dimensions of the embossed pattern, certain experimental techniques which applicable are to the characterization of flat films may not be applicable when characterizing embossed films. Moreover, the measurements of

the material temperature and the pressure distribution in the nip present some serious difficulties.

Mathematical analysis of the process involves the use of either statistical methods [26] or solving the conservation equations [27] to estimate the thermal and stress fields involved in the process. The key modelling constraints are related to the ability to measure the estimated variables and to the complexity of the models developed. It is necessary to measure the variables in order to test the accuracy of the model.

Analysis of the film embossing process, as illustrated above, requires the application of both experimental and mathematical modelling techniques. Due to the limited amount of work done in this area, the experimental aspects need to be emphasized initially in order to develop the required data for understanding the process.

1.5. THESIS CONTENT

The text of this thesis is divided into seven chapters. Chapter 1 deals with the general background and a review of the embossing of plastic films. Chapter 2 outlines the objectives of the research project. Chapter 3 describes the embossing systems used in this study. Descriptions of the embossing roll pattern, rubber backup roll properties, process variables and resin properties are included. Chapter 4 describes the experimental analyses of the process. This includes characterization of the flat film properties, embossed film properties, thermal fields and stress fields as functions of the processing conditions. Chapter 5 deals with the mathematical models derived to describe the heat transfer and material deformations during the thermal treatment and embossing stages. Chapter 6 includes comparisons between the experimental data and the predictions of the mathematical

models. Chapter 7 contains the general conclusions and suggestions for future work. Each section presents a review of literature relevant to that chapter.

Appendix A presents a description of the experimental methods used in this study. Appendix B presents tables containing all the experimental data and an analysis of the variability of the data. Appendix C presents a reprint of a paper presented at the Society of Plastics Engineers Technical Conference related to the morphology and orientation in polyethylene tubular blown films. Appendix D presents a description of the rheological properties of the low density polyethylene resin used in this study. Finally, copies of the computer programs developed for this study are available from Professor M.R. Kamal, Department of Chemical Engineering, McGill University, Montreal, Canada, upon request.

Chapter 2 SCOPE AND OBJECTIVES

2.1. PROJECT OBJECTIVES

In view of the above discussion, the present study was planned with the following objectives in mind:

1. To perform a preliminary engineering analysis of the film embossing process with special emphasis on the following aspects:

> (a) to determine, both experimentally and analytically, the relationships between the process variables and the material properties at various stages of the process.

> (b) to investigate the interrelationships between the process variables and the final properties of the embossed film.
> (c) to obtain mathematical models describing the heat transfer and mechanical stresses and strains involved in the embossing process.

2. To develop relevant techniques to characterize unembossed and embossed thermoplastic films.

Specifically, the purpose of this work has been to achieve an overall understanding of the embossing process. Since no systematic study of the embossing process has as yet been reported, it is hoped that the present study will represent a significant step towards a fundamental understanding of the process and serve as a framework for future work.

Chapter 3 PROCESS DESCRIPTION

This chapter describes the film embossing systems used in the present study. The discussion includes description of the continuous embossing system, batch embossing system, process variables, and experimental design.

Both a continuous tubular blown film embossing and a batch embossing process were used in this study. The analysis of the tubular blown film embossing process can be easily modified to include the analysis of the batch embossing process. Furthermore, the conclusions made from studying the tubular blown film embossing process can be extended to the cast film process, since the analysis of the phenomena in the nip region is identical for all the above three types of embossing configurations.

Appendix A presents a description of the experimental methods used in this study. Appendix B presents tables containing all the experimental data presented in this chapter and an analysis of the variability of the data.

3.1. CONTINUOUS TUBULAR FILM EMBOSSING SYSTEM

The continuous tubular blown film embossing system used in this study and illustrated in Figure 3-1, consisted of four parts:

- (a) Tubular blown film extrusion system
- (b) Thermal treatment zone
- (c) Embossing zone
- (d) Wind-up stage.

The following discussion describes the highlights of each of the above components of the embossing system.


Figure 3-1: Schematic Diagram of the Continuous <u>Tubular Blown Film Embossing Process.</u>

3.1.1. Tubular Blown Film Extrusion System

The molten plastic is first extruded through an annular die and then drawn upwards by the take-off device. Air is introduced at the bottom of the die, causing the parison to become inflated. Both the forward stretching and sideways blowup occur in the melt before the polymer starts to solidify at the freeze line. An air ring is used to cool and solidify the hot bubble. The inflated bubble is then flattened as it passes through the nip rollers, which also serve to provide axial tension to draw the film upwards and form an air-tight seal. Constant pressure is thus maintained in the bubble. A Black Clawson extruder with a 12.7 and 25.4 cm annular die was used to produce all the blown film samples used in this study.

The tubular blown film process is a complex system, in which several factors affect the final morphology of the film. Detailed description of the tubular blown film process is available elsewhere [13,28-30]. The key variables in the process are:

- (a) Resin rheological properties
- (b) Melt temperature
- (c) Draw ratio
- (d) Blowup ratio
- (e) Freeze line height
- (f) Cooling rate
- (g) Extruder screw speed
- (h) Die gap.

3.1.2. Thermal Treatment Zone

The heat treatment zone of the embossing system consisted of a preheat roll and a radiation heater system, as shown in Figure 3-2. A water heated steel roll, 1.07 m wide and 0.30



Figure 3-2: Schematic Diagram of the Continuous <u>Embossing Station.</u>

meters in diameter, was used to preheat the plastic film to a temperature ranging between 90 and 95°C.

The plastic film is heated by conduction of heat from the roll surface through the film. The heated plastic film then travels a distance of 0.19 m to the radiation heaters, where it loses heat by convection and radiation to the surroundings.

The final component of the heating system consists of two radiation units each containing four horizontal radiation tube heaters, spaced 5.08 cm from each other and 5.87 cm from the travelling film. Heat is radiated to the surface of the moving film and conducted from the surface towards the core of the film. Within the heat treatment zone, the plastic is not only heated, but is also stretched as a result of the tension applied to the travelling film between the various rolls. The combination of applied thermal and stress fields causes changes in the physical dimensions, microstructure and mechanical properties of the film.

3.1.3. Embossing Zone

After heating, the film passes into the nip between a water cooled engraved steel roll, 1.07 m in length and 0.15 m in diameter, and a rubber coated steel backup roll, 1.07 m in length and 0.20 m in diameter, to impress a pattern onto the surface. The rubber layer, 1.27 cm thick, is made of Neoprene rubber having 45 Shore A hardness. Force, which is controlled by a pneumatic mechanism, is applied to a ram 8.9 cm in diameter. The force controls the movement of the embossing roll with respect to the stationary backup roll. At the point of contact between the two rolls, the plastic film is compressed, sheared and cooled, thereby causing further changes in the material dimensions and microstructure (i.e. crystallinity, morphology, and orientation). These changes ultimately determine the final properties of the embossed film. Figure 3-3 presents a schematic describing the geometry of the plastic film.

The embossing pattern used in the continuous embossing system is presented in Figure 3-4. The diamond shaped pattern, as viewed from the top, is typically used for the production of embossed film for the backing of disposable diapers. The actual pattern shape is that of a truncated pyramid. The dimensions of the pattern face (Figure 3-4a) relate to the dimensions of the peaks of the pyramid. The distance between the diamonds is equal to the sum of the projection of the sides of the pyramid and the spacing between two consecutive embossing patterns.

3.1.4. Wind-Up Stage

Like an elastic band wrapped around a finger, polyethylene rolls build up large forces with each additional layer. In conventional winding operations, as long as the core support is not crushed and the roll remains flat, the product would be adequate. However, for embossed films the applied tension could cause the film near the core to be flattened, resulting in a low film gauge and loss of pattern definition. Thus, automated tension controls are used with embossing systems to obtain rolls of uniform quality.

3.2. BATCH EMBOSSING SYSTEM

The batch embossing system was based on the embossing station of a tubular blown film embossing system. It is illustrated in Figure 3-5. The radiation heating zone was enlarged to include two units of eight radiation tube heaters. The vertical spacing between the individual tubes was 5.08 cm. The travelling film was located 6.03 cm from the radiation tubes. Film feeding equipment was added to allow the processing of rolls of flat film. The configuration of the preheat roll,



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Figure 3-3: <u>Schematic Diagram of the Film Geometry.</u>



- a: 113.5 degrees
- b: 0.368 mm
- c: 0.356 mm
- d: 0.252 mm
- e: 0.242 mm
- f: 66.6 degrees



h: 49.5 degrees

i: 0.057 mm

g: 0.114 mm

Figure 3-4: Schematic Diagram of the Continuous System Embossing Pattern. ۰.

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Figure 3-5: Schematic Diagram of the Batch Embossing Station.

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rubber backup roll and embossing rolls remained the same. The embossing pattern used in the batch embossing system is presented in Figure 3-6. The diamond shaped pattern is similar to that used in the continuous embossing system.

3.3. PROCESS VARIABLES

The key process variables in both the continuous and batch embossing systems can be divided into three categories as follows:

Flat Film Extrusion Variables

Resin Type Film Thickness Film Blowup Ratio Film Draw Ratio

Pre-Embossing Variables Film Velocity Preheat Roll Temperature Radiation Heater Temperature Film Tension

Embossing Process Variables

Embossing Roll Temperature Embossing Pressure Backup Roll Temperature Rubber Hardness Embossing Pattern Roll Velocity.

The flat film properties are determined by the thermomechanical history experienced by the resin during processing. In most commercial applications, the resin grade, film thickness and film layflat width are specified. The film blowup and draw ratios are important in determining the ultimate properties and dimensions of the film, at a fixed thickness.

In the pre-embossing zone, the flat plastic film is treated thermally. The temperature distribution of the flat film prior to entering the embossing zone nip region is a function of



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i: 0.013 mm

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Figure 3-6: Schematic Diagram of the Batch System Embossing Pattern. the preheat roll geometry, preheat roll temperature setting, radiation heater geometry, radiation heater temperature setting, flat film velocity, line tension, film thickness, resin grade and flat film orientation.

Upon entering the nip of the embossing zone, the thermally treated flat film is sheared, compressed and cooled to impress the embossing roll pattern into the film. The temperature distribution of the backup roll, temperature distribution of the engraved embossing roll, film velocity, film thickness, resin grade and film orientation determine the rate at which the embossed film is cooled. The pressure distribution in the nip is determined by the backup roll rubber layer properties, engraved roll metal properties, pneumatic pressure, film properties, resin grade, embossing roll pattern, temperature distribution and film velocity. These variables also control the extent to which the embossing pattern is transferred to the flat film.

The process variables listed above can be further divided into two groups: fixed and independent variables. The fixed variables, which include design parameters and resin grade, cannot be altered for a given process operation. The heater temperature settings, film speed, pneumatic pressure, initial film orientation and initial film thickness can be considered as the independent process variables. The embossing process variables for the continuous and batch embossing systems are identical.

3.3.1. Extrusion Variables

The three most important extrusion process variables are: resin grade, film thickness and film blowup ratio.

3.3.1.1. Resin Properties

A low density polyethylene resin manufactured by Union Carbide, grade DFDY 4409, was used in this study. The ASTM classification [31] for the resin is LDPE Type 1, Class A and Category 3. It is commonly used to manufacture embossed films for disposable pad applications. This material will be referred to as LDPE-1 for the remainder of this thesis.

The resin characterization experiments were conducted using compression molded samples. Wide angle x-ray diffraction, birefringence, infrared spectroscopy and sonic velocity techniques were used to verify that the orientation of the crystalline and amorphous phases in these samples were random and isotropic.

The density of LDPE-1 at room temperature was found to be 0.917 g/cm³ using a density gradient column, corresponding to 43% crystallinity. The density values for the amorphous and crystalline phases, used to determine the crystallinity, were those quoted by Kavesh and Schultz [32].

The crystal structure of polyethylene is known to be based on an orthorhombic unit cell. Using Miller indices and based on the dimension of the unit cell and x-ray camera, the order of the x-ray diffraction rings appearing on the x-ray photographs are (2,1,0), (2,0,0) and (0,2,0) [33].

The infrared absorption spectrum for LDPE-1, measured at 0.05 mm, is presented in Figure 3-7. The corresponding infrared band assignments for polyethylene are described in Appendix A, Section A.4.2..

The total absorptance, reflectance and transmittance spectra of LDPE-1 [34], measured at a film thickness of 0.025 mm, are presented in Figure 3-8. The absorptance spectrum was





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Figure 3-8: Total Reflectance, Transmittance and Absorptance of LDPE-1, H=0.025 mm.

calculated based on the relation that the summation of the absorptance, reflectance and transmittance is equal to unity [35]. Average values were calculated by integrating the data as a function of wavelength. The average emissivity according to Kirchoff's Law [36], which states that the emissivity is equal to the absorptance, was found to equal 0.14 using the data presented in Figure 3-8. In addition, when the sample thickness was 0.05 mm, the average emissivity was found to values of the 0.23. Published emissivity for equal polyethylene [37-40] are reported to range between 0.15 and 0.60. The differences observed depend on the thickness of the sample used to determine the emissivity. The thicker the test sample, the larger the emissivity. The emissivity for films having a thickness of 0.025 mm was reported as ranging from 0.15 to 0.2. The values based on Figure 3-8 will be used in this thesis.

The specific heat, thermal conductivity and diffusivity of LDPE-1, as functions of temperature, are presented in Figure 3-9. The thermal diffusivity can be considered constant when the reproducibility of the data is taken into account, even though it appears to be decreasing as a function of temperature. The main source of error in measuring the thermal diffusivity was due to the measurement of penetration time [41]. The variability in the measured penetration times was found to range from 20-30%. The peak observed in the specific heat data at 110°C corresponds to the melting point of the resin. Semicrystalline materials are known to have a melting temperature range instead of a single melting temperature, as in the case of perfectly crystalline materials. The melting characteristics of LDPE-1, determined by differential scanning calorimetry, were as follows:

Onset: 100.9°C Peak: 106.3°C Heat of Fusion: 104.6 J/gEnd: 112.3° C.



Figure 3-9: Thermal Properties of LDPE-1.

The melt index of LDPE-1 is 1.7, according to the manufacturer. The steady shear and oscillatory shear rheological properties of LDPE-1 are presented in Appendix D.

The tensile properties of LDPE-1, measured at room temperature, are listed below:

Tensile Modulus: 69.0 MPa Yield Stress: 8.9 MPa Elongation at Yield: 26.7%.

The compressive modulus of elasticity and yield stress, as functions of temperature, are presented in Figure 3-10. The elongation at yield may be considered to be independent of temperature when considering the variability in the data. The yield strength and elongation at yield were difficult to measure at temperatures greater than 100°C. The mean values of these properties over the temperature range were found to be as follows:

> Proportional Limit: 4.2 MPa Proportional Limit Compression: 11.4%.

Due to the small deformations encountered, the effect of sample barrelling (i.e. case where the shape of the sample is no longer cylindrical) was assumed not to be significant. The modulus of elasticity measured under tension and compression at room temperature were found to be unequal.

3.3.1.2. Flat Film Thickness

Embossed films are used mainly as an outer layer in the final product. In the case of disposable diapers, the embossed film serves as a waterproof back sheet. The thickness of films used in these applications should be as small as possible, while retaining the mechanical properties required for finishing



Figure 3-10: Mechanical Properties, Compression, of LDPE-1.

operations and end use. The flat film thickness for most personal products typically ranges between 0.019 and 0.032 mm.

In the tubular film process, the flat film thickness is controlled by the draw ratio, die gap, blowup ratio and cooling conditions. The thickness of the films evaluated in this study varied between 0.01 and 0.15 mm. Since it is also common practice to emboss the collapsed web (i.e. two layers) in a single embossing station, the thickness of the films ranged from a minimum 0.01 mm (single layer) to a maximum 0.3 mm (two layers).

3.3.1.3. Flat Film Blowup Ratio

The film blowup ratio, BUR, is defined as the bubble diameter divided by the die diameter. Blowup ratios used in the film industry commonly range from 2 to 4. Values greater than 4 and less than 2 are rarely used due to the difficulties encountered in maintaining a uniform bubble. The blowup ratio the film velocity, die diameter, cooling depends on conditions, draw ratio and internal bubble pressure, for a fixed film thickness. Due to the complex interactions between the various process variables, no unique relationship is known to exist between these parameters. In most applications, the blowup ratio is specified to optimize the film layflat width and film thickness. In the case of disposable embossed films, this may involve the use of a blowup ratio which results in a layflat width equivalent to the length of the embossing pattern on the embossing roll.

The blowup ratio is a parameter which relates only to the continuous embossing process. The flat films used for the batch embossing process can be produced either using a cast or tubular blown film process. All the flat films in this study were produced using the tubular blown film process and

the range of blowup ratios used varied from 1 to 5. The standard blowup ratio, using a 25.4 cm annular die, was 3.

3.3.2. Pre-Embossing Variables

The three most important pre-embossing process variables are: preheat roll temperature setting, radiation heater temperature setting and flat film velocity.

3.3.2.1. Preheat Roll Temperature

The diameter of an industrial scale preheat roll can range from 30 to 60 cm. Depending on the processing conditions, a single or double preheat roll system can be used. The temperature of the preheat roll is typically controlled by the circulation of hot water in the core of the roll. The preheat roll consists of a metallic outer cylinder and an inner core having a helical channel to allow the circulation of the heating fluid. The height of the flights on the inner core is such as to minimize leakage from the channels. The typical temperature drop across the length of a commercial preheat roll ranges between 1 and 3°C. A temperature controller is used to control the temperature of the heating fluid. The flow of heating fluid for a two preheat roll system, is in a counter current pattern so as to minimize the temperature variation across the plastic film width.

The preheat roll was used to raise the temperature of the flat film from approximately room temperature to 95°C. If the temperature of a polyethylene film is raised above 100°C, processing difficulties may occur because the flat film may stick to the surface of the preheat roll. In addition, when operating at temperatures above 100°C, steam or oil based heaters must be used. The factors which influence the final temperature of the flat film are the film velocity, film thickness, ambient air temperature and preheat roll surface temperature.

The temperature of the preheat roll could be controlled between 40 and 95°C. The experimentally measured surface temperature of the preheat roll, T_s, at various preheat roll controller settings, T_P, is presented in Figures 3-11 and 3variability of the temperature around 12. The the circumference of the preheat roll (Figure 3-11) was found to be negligible regardless of the axial position on the preheat roll. The dimensionless axial position, $Y/W_{P}/2$, is defined as the axial position relative to the center of the roll, Y, divided by half the roll width, W_P . A value of -1 refers to the water inlet side of the preheat roll, 0 to the roll center and 1 to the water outlet side. An angle of 0 degrees relates to the top of the preheat roll. The preheat roll surface temperature was found to vary by less than 1°C across the roll face. The temperature drop of the heating fluid across the preheat roll was found on average to vary between 1 and 2°C. The difference between the controller setting and the inlet water temperature was found on average to be 3°C. The variation of the ambient air temperature at various distances from the preheat roll is presented in Figure 3-13. At distances greater than 5 cm, the air temperature was found to equal the ambient air temperature.

The measured values of the preheat roll surface temperature in this study were used for all subsequent calculations and comparisons. The following linear equation was determined to express the relationship between the preheat roll surface temperature, T_s , and the preheat roll temperature setting, T_p , and position on the preheat roll, $Y/W_p/2$:

$$T_s = -5.74 + 1.04 T_p.$$
 (3-1)



Figure 3-11: Preheat Roll Surface Temperature <u>Distribution, $T_p=93.3^{\circ}C.$ </u>



Figure 3-12: Preheat Roll Surface Temperature Distribution.

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The value of the correlation coefficient was calculated to be 0.997. The axial position, $Y/W_P/2$, was found not to be a significant variable based on the use of a backward elimination technique to determine the best model. The partial F values were determined using a 95% confidence level.

3.3.2.2. Radiation Heater Temperature

The dimensions of an industrial scale radiation heater can range up to 2.5 m in length and 0.5 m in width. At such large lengths, it is not possible to orient the radiation heater tubes horizontally. In such cases, the radiation heater tubes are positioned between 80 and 90 degrees from the horizontal. The heater length is thus reduced substantially. However, the number of radiation tubes increases. As a result, the spacing between the radiation tubes must be adjusted to give a uniform temperature field.

The radiation heaters used in this study were used to raise the temperature of the plastic film from 90 to 105°C, just below the melting point. The film travels between two banks of heating elements to heat the film uniformly and quickly. Due to the high line speeds, the residence time of the plastic film in the heating zone is very small. For this reason, the temperature setting of the radiation heaters must be very high. If the temperature of the film is raised above 105°C, excessive film shrinkage can occur. This can result in necking and breaking of the film web. The factors which control the final temperature of the flat film are the film velocity, film thickness, ambient air temperature, radiation heater temperature setting and initial flat film temperature.

The radiation heater arrangements for the continuous and batch embossing systems were different. The batch embossing system radiation heater design incorporated four additional radiation tubes per side and an enclosure to insulate the system. The advantage of this arrangement was that the length of the heating zone was increased, which in turn allowed for an increase in line speed. The disadvantage of having an enlarged heating zone is that unless the line speed is properly controlled, film shrinkage can occur due to overheating of the plastic film.

An optical pyrometer was used to measure the surface temperature of the radiation heater tubes. Figure 3-14 presents the temperature distribution along a radiation heater tube. The dimensionless axial position variable, $Y/W_R/2$, is set to equal zero at the middle of the radiation heater tube. The width of the radiation heater element is W_R. The maximum temperature variation across any one of the radiation tubes was found to be 22°C, at a radiation tube setting of 815.6°C. As the radiation heater tube temperature was decreased, the temperature variation along the tube was also found to decrease. The continuous embossing system was used to take all the temperature measurements because it was not possible to take any measurements using the batch embossing system due to the configuration of the heaters.

The air temperature distribution around the radiation heaters was found to vary significantly. The measured air temperature distribution at a distance of 1 to 2 cm in front of the radiation heater tubes, as a function of distance from the preheat roll, Z, is presented in Figure 3-15. The Z coordinate was specified as being zero at the top of the preheat roll and 0.24 m at the separation point between the preheat roll and the plastic film. With respect to the radiation heaters, the top of the radiation heater tube bank facing the wind up rolls was specified as Z=0.43 m and the bottom of the radiation heater tube bank as Z=0.65 m. The air temperature was found to be independent of the film speed. The high surface temperature of the radiation heaters gave rise to complex air flow patterns around the heating tube bank. Since





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the transmittance of air is approximately equal to unity, the high air temperatures are due to natural convection effects. Due to the difficulty in making the air temperature measurements, the variability of the temperature data was quite extensive and was found to range from 10 to 15%. On average, the air temperature in front of the radiation heater unit was found to be 200° C.

The effect of the radiation heater temperature on the preheat roll air temperature is illustrated in Figure 3-16. The air temperature distribution at the separation point between the preheat roll and the plastic film on the preheat roll, Z=0.24m, was found to be greater than 100° C, especially in the center region. As the film travelled from this point to the top of the radiation heater unit, Z=0.43 m, it was heated by convection because the air temperature was greater than the preheat roll temperature.

The air temperature distribution, as measured from the top of the preheat roll to the embossing roll nip region, is presented in Figure 3-17. These data were used in all subsequent calculations and comparisons to represent the air temperature distribution.

3.3.2.3. Film Velocity

The film velocity for the tubular film embossing process is limited by the output rate of the extruder. On the other hand, the film velocity for the batch embossing process is limited by the film take off equipment and radiation heater geometry. As a result, higher line speeds are possible using the batch embossing process.

The typical operating line speed for a commercial film embossing line ranges between 60 and 70 m/min. The film velocity attainable using the continuous embossing system was







Figure 3-17: <u>Air Temperature Distribution.</u>

in the range of 10 to 30 m/min. The film velocity attainable using the batch embossing system was in the range of 10 to 50 m/min, with a standard processing velocity of 20.5 m/min.

Since it is not the objective of the film embossing process to reduce the film thickness, the linear speeds of the preheat, embossing and wind up rolls were set equal. Tension due to shrinkage as a result of the thermal treatment, helps to avoid any wrinkling in the film which can cause problems during the wind up stage. The film velocity was found to be identical to the various roll linear velocities. As a result, perfect contact was achieved between the process roll and film surface, i.e. no slip occurred at the interface.

3.3.3. Embossing Variables

The six most important embossing variables are the embossing roll pattern, backup roll rubber layer properties, embossing roll metal properties, pneumatic pressure, embossing roll temperature and embossing backup roll temperature distribution.

3.3.3.1. Embossing Roll Pattern

The dimensions of the embossing patterns presented in Figures 3-4 and 3-6 were those specified to the manufacturer of the embossing roll. The actual dimensions were measured by making a cast impression of the roll pattern and subsequently taking photographs of the casting using an optical microscope. The dimensions were not found to vary across the width or along the circumference of the embossing roll. In comparison to the specified pattern dimensions, the actual pattern did not have any sharp corners. The pattern edges were instead found to be rounded sections, resulting in a smooth contoured pattern. This was most noticeable for the space between the various embossments. The dimensions of the embossing pattern presented in Figures 3-4 and 3-6 will be used in all subsequent references to the embossing pattern.

3.3.3.2. Rubber Layer Properties

The rubber layer on the backup roll was manufactured of Neoprene rubber with a Shore A durometer hardness of 45. Some typical properties of Neoprene rubber are presented in Table 3-1. The modulus of elasticity was estimated using the probite curve in ASTM D 1415 [44]. According to this method, the durometer hardness is equivalent to the International Rubber Hardness Number.

The actual properties of the rubber layer on the backup roll can vary significantly from those presented in Table 3-1. Variations in the rubber layer properties will tend to occur due to the mechanical and thermal stresses imposed on the material which cause the rubber to toughen over time. Moreover, the data presented are based on calculations using the durometer hardness readings. The durometer hardness is measured by instruments which apply a force to a rigid indentor of known geometry. The amount of indentation has been shown to be related to the modulus of elasticity [45]. However, other factors such as the homogeneity of the rubber, sample thickness, nature of the sample surface, radius of the indentor and indentation force have been found to affect the durometer reading. The relation relating these factors is complex and only partially known. Since the experimental conditions used to measure the hardness of the rubber on the backup roll are not known, we can only assume that the durometer reading represents an approximation of the modulus of elasticity. The data presented in Table 3-1 will be used in all future references to the properties of the rubber on the backup roll.

Rubber Layer Properties^[42-44]

ASTM Name	Chloroprene
Commercial Name	Neoprene
Density	$1.23-1.25 \text{ g/cm}^3$
Thermal Conductivity	0.19 W/m°K
Tensile Yield Strength	24-28 MPa
Elongation at Yield	500-600%
Young Modulus	1.8 MPa
Poisson Ratio	0.49

Metal Roll Properties^[46]

Туре	SAE 4140
Hardness	Brinnell 190
Density	7.73 g/cm^{3}
Thermal Conductivity, 100°C	42.7 W/m°K
Thermal Conductivity, 200°C	42.3 W/m°K
Specific Heat, 150-200°C	473 J/kg°K
Tensile Yield Strength	417 MPa
Elongation at Yield	25.7%
Young Modulus	655 MPa
Poisson Ratio	0.29

Table 3-1: <u>Rubber Layer and Metal Roll Properties.</u>

3.3.3.3. Metal Roll Properties

The embossing roll and metal core of the backup roll were constructed of an annealed SAE 4140 carbon low alloy steel with a minimum Brinnell hardness of 190. Some of the important physical properties for this material are presented in Table 3-1. The data presented in Table 3-1 will be used in all future references to describe the properties of the metal rolls.

3.3.3.4. Embossing Pressure

The pressure distribution in the nip formed by the contact of the two embossing rolls is determined by the embossing pressure, metal roll mechanical properties, rubber layer mechanical properties and roll geometries.

The two embossing rolls are pressed together by using two pneumatic cylinders to control the position of the embossing roll. Air pressures commonly encountered in industry range up to 2.4 MPa, resulting in a force of 9.7 kN, assuming a cylinder head diameter of 15 cm. The air pressures used in this study ranged from 0.1 to 0.2 MPa. These pressures, based on the dimensions of the roll cylinder (cylinder head diameter of 88.2 mm) produce nip forces ranging from 0.6 to 1.2 kN on the embossing roll.

3.3.3.5. Embossing Roll Temperature

The embossing roll serves not only to impress a pattern into the plastic film but also to cool the film to a temperature at which the embossed pattern remains fixed, i.e. below the softening point of the resin. The design of the embossing roll is very similar to that of the preheat roll. The embossing roll is cooled by the circulation of water through the core. An industrial embossing roll can be as large as 2.5 m in width and 1 m in diameter. The circulating water is usually cooled by a cooling tower. As a result of seasonal variations, the temperature of the cooling water can vary by as much as 15° C.

In this study, municipal water was used to cool the roll. The temperature of municipal water supply varies seasonally and geographically. The seasonal variation in the temperature can range between 10 and 15°C, with winter being the coldest and summer the hottest. All the embossing experiments were conducted in the spring. The inlet water temperature on the embossing roll was on average 14°C, while the outlet temperature was 32°C.

The surface temperature of the embossing roll when the radiation heaters were turned off, was found to be uniform $(27^{\circ}C)$ along the length and circumference of the roll. During operation, the radiation heater had the effect of heating the surface of the embossing roll. Depending on the radiation heater temperature setting, the surface temperature of the embossing roll could be increased by as much as 5°C. The maximum variation in the temperature distribution along the embossing roll was found to be 4°C.

Due to the geometry of the embossing roll, the air temperature distribution at various circumferential positions could not be measured. On the basis of the limited number of data points available, it was decided to use air temperature values of 28°C when the radiation heaters were turned off and 100°C when they were set at 815.6°C.

3.3.3.6. Backup Roll Temperature

The design of a commercial backup roll is very similar to that of the embossing and preheat rolls. In this study, the backup roll was not cooled except by contact with the engraved roll. As a result, the temperature of the backup roll was found to
vary significantly. The backup roll surface temperature was found to be 27°C when the radiation heaters were turned off and 35°C when the radiation heater temperature setting was 815.6°C. This occurred because of the increase in the ambient air temperature and because a small portion of the backup roll was in direct view of the radiation heaters. The surface temperature of the backup roll was found to be uniform along its length. The air temperature distribution was found to be the same as that for the embossing roll.

Chapter 4 EXPERIMENTAL

This chapter describes the experimental work carried out to characterize the morphology, orientation, mechanical properties, optical properties and thermal properties of the flat and embossed films produced in this study.

Appendix A presents a description of the experimental methods used in this study, while Appendix B presents tables containing all the experimental data presented in this chapter.

4.1. FLAT FILM ANALYSIS

The objective in this section was to characterize tubular blown polyethylene films with regard to the effect of variations in the blowup ratio on the final flat film properties and to evaluate the effect of various factors on the morphology and orientation of tubular blown polyethylene films.

4.1.1. Previous Studies

4.1.1.1. Morphology

Over the past 30 years, several studies have been carried out to investigate and explain the orientation and morphology of polyethylene films. The type of orientation described in the early studies can be grouped into the following three categories:

<u>Type I:</u> *a*-axis orientation model proposed by Holmes, Miller, Palmer and Bunn [47]. <u>Type II:</u> Row model proposed by Keller [48-51]. Type III: Intermediate Type I-II model proposed by Holmes and Palmer [52].

Holmes et al [47] examined the structure of cast polyethylene films using x-ray, optical and polarized infrared techniques. It was proposed that the a-axis of the polyethylene unit cell was preferentially oriented towards the machine direction. The b and c axes of the polyethylene unit cell, in turn, were found to be randomly distributed in the plane perpendicular to the a-axis. Based on the x-ray, optical and polarized infrared absorption data, it was assumed that the orientation of the polymer chains in the amorphous region were perpendicular to the orientation of the chains in the crystalline region, i.e. parallel to the machine direction. This type of orientation was referred to as Type I or a-axis orientation. This model was identical to that proposed for relaxed drawn polyethylene samples and drawn fibers. The 729 cm⁻¹ infrared absorption band was ascribed to the crystalline regions of polyethylene, whereas the 721 cm⁻¹ was related to both the crystalline and amorphous regions. Information from both of these absorption bands was used to obtain a measure of the orientation in the films.

The findings by Holmes et al [47] were supported by Aggarwal, Tilley and Sweeting [53] who examined seven tubular and three cast polyethylene films using x-ray diffraction, infrared dichroism and birefringence techniques. They found that the a-axis was oriented parallel to the machine direction. The infrared dichroism data confirmed the a-axis orientation and indicated that the amorphous regions were unoriented. It was concluded that the orientation was of the Type I form even though the observed birefringence was found to be positive, which is contrary to the Type I model. The infrared absorption bands at 1300, 1340 and 1355 cm⁻¹ were used to measure the amorphous chain orientation. X-ray diffraction patterns were obtained for all three planes of the plastic film: machinetransverse (MD-TD), machine-normal (MD-ND) and transverse and normal (TD-ND) directions.

Keller [48-51] proposed an alternative model, the row orientation or Type II model. Most commonly, unoriented polymers consist of spherulites which themselves are constructed of closely coiled helical ribbons. During deformation, such as when the polymer is being drawn, the helical ribbons in the spherulites are pulled out. In processes such as film extrusion, spherulites are not always developed; instead, row-like aggregates form. Crystal growth starts from closely spaced nuclei which are situated at the surface of the melt. Each nucleus is the origin of radiating growth. However, due to the closely packed nature of the nuclei, growth can only take place perpendicular to the surface. The resulting texture is that of twisted ribbons, structure. this model. the b-axis is i.e. row In preferentially oriented perpendicular to the machine direction with the a and c axes rotated randomly about the b-axis. The direction of crystal growth was found to be in the b-axis direction. This type of orientation can also be referred to as the helix model. The orientation model proposed by Keller [48-51] was based on the examination of unoriented and drawn heat treated compression molded samples.

Both the Type I and Type II orientations result in similar xray diffraction patterns for the a and b axes. The c-axis orientation for the Type I model was proposed by Holmes et al [47] to be perpendicular to the machine direction, while the c-axis for the Type II model was proposed by Keller [48-51] to be parallel to the machine direction. Both models, however, are deficient in that the Type II model does not account for a negative birefringence and the Type I model does not account for a positive birefringence. In many cases, this discrepancy may be explained in terms of the orientations of the amorphous phase being parallel or perpendicular to the machine direction, thereby making a greater contribution to the birefringence than the crystalline phase.

The work by Stein and Sutherland [54,55] showed that the 721 cm⁻¹ infrared absorption band for polyethylene contains a strong crystalline component with its transition moment polarized parallel to the *b*-axis of the crystallites, in addition to the contribution from the amorphous regions. This work and that of Keller [48-51], as described previously, led Holmes and Palmer [52] to reconsider the orientation of the films analyzed in 1953. It was found that some of these films exhibited Type II orientation. Through examination of a large number of commercial polyethylene films with both positive and negative birefringences, a third orientation model, Type III, was proposed whereby the a-axis of the unit cell was tilted at a preferred angle to the machine direction. The value of this angle varied between 37 and 55 degrees, depending on the processing conditions. The b-axis remains oriented in the plane perpendicular to the machine direction. This type of orientation is intermediate to the Type I and Type II models. This model also tends to confirm the conclusions made by the authors in 1953 regarding the amorphous orientation which was postulated to be parallel to the machine direction even though the crystal chain axis was perpendicular to the machine direction. The value of the tilt angle of the a-axis was found to increase as the blowup ratio increased and as the haul-off ratio decreased, for а given polymer and extrusion temperature. Tubular blown films were found to exhibit varying orientations ranging from positive to negative birefringence.

Tobin and Carrano [56] used infrared dichroism to determine the orientation in polyethylene films. The dichroic ratios at the 719, 730 and 1300 cm⁻¹ bands were measured. The 1300 cm⁻¹ band was found to arise solely from the amorphous areas of the film. In extruded polyethylene films, the amorphous chain axes were found to have a slight preferential orientation in the machine direction. The *a*-axis of the unit cell was found to be parallel to the machine direction and the *b*-axis perpendicular to the machine direction. As the film samples were stretched, the *a*-axis was found to orient perpendicular to the machine direction.

Lindenmeyer and Lustig [57] were the first to study the orientation using the pole figure technique [58-60] instead of the flat plate x-ray technique. The pole figures showed that, in some cases, the orientation was biaxial rather than uniaxial. As a result, in these cases it was not possible to assume cylindrical symmetry about the machine direction as was done by previous authors. Furthermore, these authors demonstrated that the orientation of the *c*-axis cannot in general be deduced from measurements of the a and b axes distributions. In a single crystal, knowledge of any two axes immediately determines the orientation of all the other axes. However, with a distribution of crystal orientations, the knowledge of the orientation of the maxima in the a and b axes directions does not necessarily fix the location of the maximum in the *c*-axis distribution. Their results showed that, at low blowup ratios, the orientation of the a-axis was in the machine direction and was inclined approximately 45 degrees out of the plane of the film (MD-TD). As the blowup ratio increased, the a-axis orientation changed toward a uniaxial texture. At very high blowup ratios, the a-axis was oriented perpendicular to the plane of the film and the b-axis orientation changed from being perpendicular to the machine direction to being perpendicular to the plane of the film.

Pole figures graphically present the three dimensional orientation in a two dimensional framework. The characterization of polymer chain orientations can also be described in terms of mathematical quantities, i.e. orientation functions. Reviews of the methods used to

calculate uniaxial and biaxial orientation functions are available [61-73].

Desper [71-73] was the first to use orientation functions to study polyethylene films. He examined a series of low density polyethylene films using birefringence, infrared dichroism and x-ray techniques. The results indicated a helical morphology with a non-random distribution of the a and c axes about the b-axis. The orientation distribution of the a-axis was found to have a maximum in the machine direction, intermediate intensity in the transverse direction and a minimum orientation normal to the plane of the film. The b-axis orientation was found to show a maximum in the film normal direction and a minimum in the machine direction.

Judge and Stein [74] observed that as the elongation increased for recrystallized stretched crosslinked polyethylene, the crystallite orientation shifted from an *a*-axis type structure to a *c*-axis fiber orientation. This study led Keller and Machin [75] to re-examine the row orientation model and develop a more general hypothesis which took into account crystallization under stress. They postulated that if the stress during crystallization was small, then the orientation observed would be of the row model type. If the stress during crystallization was high, the c-axis of the crystals would be aligned in the draw direction, giving a *c*-axis type orientation similar to that found for drawn polyethylene fibers. Intermediate stresses would lead to an incomplete twisting of the ribbons. Crystal growth was said to occur along the b-axis and the orientation of the a and c axes about the b-axis was dependent on the stress during crystallization. Figure 4-1 is a representation of the orientations resulting from low and high stress conditions. It was also suggested by Keller and Machin that the amorphous chains were found on the surface of the crystals in the form of loose loops or tie molecules and that these amorphous chains become aligned along



Figure 4-1: Representation of the *a*-axis Orientation as <u>a Function of Stress [75].</u>

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the layer surface as a result of stretching, as the two ends are incorporated into different parts of the same crystal or into different crystals.

Using the same approach, Maddams and Preedy [76-78] have concluded that the observed morphology and orientation of polyethylene films represent the net effect from stresses developed at various stages of the film blowing process, as well as any reorientation from relaxation effects which might have occurred. When the molten plastic passes through the die, it is subject to shearing stresses. These shearing stresses will lead to partial orientation of the chain axes toward the machine direction. When the melt leaves the die, these shearing forces vanish and the melt experiences elongational stresses in the machine direction. As the melt is being simultaneously cooled and blown, its viscosity increases and the stress increases as a result of the reduction in film thickness which occurs. The material closest to the die experiences the highest strain rates. The effect of the elongational stresses is to cause the molecules to reorient they emerge from the die. Simultaneously, however, as reorientation may also occur due to relaxation which for polyethylene has been shown to shift the a-axis in the machine direction, i.e. low stress. The crystallization of the melt occurs between the die exit and the freeze line. The stresses developed during that phase of the operation will have the greatest influence on the crystallization mechanisms. Under very low stress conditions there is almost a pure a-axis orientation, and, with very high stresses, there is substantial *c*-axis orientation. Commercial blowing conditions give rather high stresses and the a-axis can be inclined up to 50-60 degrees from the machine direction in the plane normal to the machine direction. Since the cooling process for the tubular blown film process is gradual, it tends to favour relaxation and reorientation of the crystal axes, i.e. low stress. The conditions of lowest stress are predicted to occur

during the film blowing of low density polyethylene and will favour either pure a-axis orientation or one with the a-axis at an angle tilted away from the machine direction. Film blowing of high density polyethylene will lead to higher stress conditions with an increasing tendency for *c*-axis orientation. The occurrence of both the high and low stress orientations was established by analyzing a variety of tubular blown high density polyethylene films produced using a laboratory and commercial scale system. In general, both types of orientations were found to exist, often simultaneously. The proportions present were found to be a function of the type of resin used and the film blowing conditions. For the films where the blowup and draw ratios were kept constant, high stress crystallization orientation was favoured by a slower cooling rate. The data for the films examined were found to be insufficient to determine the relative importance of the machine parameters: melt temperature, blowup ratio, draw ratio and cooling rate.

Using infrared dichroic ratios, McRae and Maddams [79] investigated some of the films produced by Maddams and Preedy [76-78]. Dichroic ratios were determined for the 720, 730, 1080, 1303, 1352 and 1368 cm^{-1} bands. The 1080 cm^{-1} band is associated with the amorphous phase, i.e. extended tie chains. The 1368, 1352 and 1303 cm^{-1} bands are associated with the amorphous phase as well, i.e. gauche chain conformations. Their results demonstrated that the 1352 cm⁻¹ band could distinguish high stress and low stress crystallization orientation. Orientation of the loose chain folds was found to be greater for the high stress case, indicating that the crystal lamellae were aligned in a more regular fashion. The 1080 cm⁻¹ band was found to be greater than unity for the low stress crystallized samples. The 720 and 730 cm⁻¹ bands were found to be of limited use, except that a correlation to the tear strength was observed.

Using biaxial orientation functions, Choi, Spruiell and White [80] demonstrated that the resulting crystalline biaxial orientation functions for high density polyethylene films were unique functions of the stresses exerted on the bubble at the freeze-line. Equations for the kinematics and dynamics of the process between the die and freeze line were derived for cases of uniaxial, planar and biaxial extensions. These equations were used to estimate the stresses in the machine and circumferential directions. In addition, these equations were used to specify the process variables to yield either uniaxial, planar or biaxial orientation. The orientation was determined using wide angle x-ray scattering and birefringence. The orientation functions and birefringence were found to be correlated with the velocity ratio and stress difference. The data for the uniaxial samples indicated a row structure of the type proposed by Keller and Machin [75]. All the films produced had low to intermediate stress type orientations. The samples prepared using a constant blowup ratio exhibited a modified form of row structure. The orientation of the a-axis was found to concentrate in the machine and thickness directions. Finally, the orientation of biaxially oriented samples appeared to be equivalent to superimposed row structure.

Kwack, Han and Vickers [81] investigated the development of crystalline structure during the tubular film blowing of low density polyethylene using wide angle x-ray diffraction, small angle light scattering and scanning electron microscopy. The applied stress at the freeze line in the machine and transverse directions was determined using theoretical expressions derived in an earlier publication [82]. It was found that the magnitude of the stress in the machine direction was an important parameter influencing the crystalline orientation. In addition, the ratio of the machine direction stress to the transverse direction stress was much greater than unity. The effect of varying the blowup ratio had little influence on the x-ray diffraction patterns of the films studied. This was explained in terms of the machine direction stress, which was found to be independent of the blowup ratio. On the other hand, as the take-up ratio was varied, the stress in the machine direction increased and the level of crystalline orientation increased. The small angle light scattering patterns were indicative of the development of a rod-like structure oriented along the stretching directions. In some cases, the structure was found to consist of spherulites showing no preferential orientation. On the basis of these observations, it was concluded that at a low stress state (10^5 N/m^2) the crystalline structure of the low density polyethylene films was spherulitic. At a high stress state, a rod-like structure was achieved. It was also believed that the high stress state row structure model proposed by Keller and Machin [75] was valid when the applied stress exceeded 10⁷ N/m². None of the films evaluated exhibited this structure.

The x-ray patterns obtained by Kwack, Han and Vickers [81] are indicative of a low/intermediate stress crystallization orientation, following the model proposed by Keller and Machin [75].

4.1.1.2. Other Studies

Other studies [82-93] have attempted to evaluate the role of the rheological properties of film grade resins. In general, it was found that the rheological properties measured under steady shear conditions are less important than the elongational properties. For example, Winter [88] reported the results of a collaborative study on the relation between the film performance and rheological properties of two low and two high density polyethylene resins. The pairs of polyethylene studied were known to exhibit different processing characteristics but to have similar shear viscosities at high

shear rates. The uniaxial elongational viscosities of these resins were significantly different and seemed to explain the observed processing differences between the four materials.

Several attempts have also been made to study the kinematics, dynamics and heat transfer of the tubular blown film process in order to relate the bubble shape, blowup ratio and draw ratio to the processing conditions [38,94-97]. These studies used either experimental or theoretical techniques and yielded useful information regarding the thermo-mechanical history experienced by the melt during processing. For example, Farber [94] presented an experimental study of the film blowing of low density polyethylene and its relationship to the strain and thermal history experienced by the polymer melt between the die and the frost line. Results were obtained for the film machine direction strain rate, transverse temperature, direction strain rate and bubble shape between the die and the frost line. The melt emerged from the die at 185°C and cooled steadily until the film solidified at the frost line. The machine direction strain rate curve illustrated that the material near the die experienced the highest strain rates. As the melt was simultaneously blown and cooled as it emerged from the die, the machine direction stresses increased as a result of the reduction in the film thickness. The increase was accompanied by a steady decrease in the machine direction strain rate. The transverse direction strain rate implied that the stresses caused by the internal bubble pressure were sufficient to cause a significant strain in the transverse direction until the machine direction forces created a sufficient reduction in the film thickness.

4.1.2. Experimental

A total of 34 LDPE-1 tubular blown films were analyzed to evaluate the relationships between various film blowing process variables and flat film properties. Table 4-1 presents

Sample 	BUR	Screw Speed rpm	Melt Temperature	Film Speed m/min	Draw Ratio <u>%</u>	Freeze line Height Cm	Layflat Width	Average Thickness mm
UE6	2.5	74.0	200	17.7	20.0	36	99.0	0.027
7	2.5	74.0	200	17.7	20.0	36	98.5	0.031
8	1.4	74.0	200	29.7	34.0	91	65.0	0.025
9	1.5	54.0	200	21.7	24.0	38	60.0	0.027
10	1.4	54.0	200	21.2	24.0	38	56.0	0.026
11	1.0	54.0	200	21.2	46.0	38	39.5	0.025
12	1.6	56.0	200	22.9	25.0	38	56.0	0.026
13	1.3	56.0	200	22.9	25.0	38	52.0	0.025
14	1.5	72.0	185	17.4	20.0	51	58.0	0.035
20	0.9	80.0	186	-	55.0	-	36.5	0.027
21	1.3	80.0	186	22.0	-	-	53.0	0.028
22	1.6	80.0	186	23.0	35.0	51	65.0	0.024
23	2.1	80.0	186	23.8	29.0	64	82.0	0.024
24	2.0	80.0	186	21.0	25.0	64	81.0	0.027
25	1.5	80.0	186	22.0	26.0	-	61.5	0.026
26	1.9	80.0	186	22.0	26.0	-	77.0	0.026
27	2.0	80.0	186	22.0	26.0	-	79.0	0.025
28	2.0	80.0	186	22.0	26.0	-	78.0	0.026
29	2.0	80.0	186	22.0	26.0	-	78.5	0.027
30	1.9	80.0	186	22.0	26.0	-	77.0	0.022
31	1.9	80.0	186	22.0	26.0	-	76.0	0.021
32	1.9	80.0	186	22.0	26.0	-	76.0	0.028
33	2.1	66.5	186	12.8	15.0	58	85.0	0.036
34	2.1	66.5	186	12.8	15.0	58	82.0	0.033
35	2.1	66.5	186	12.8	15.0	58	83.5	0.034
36	2.1	66.5	186	12.8	15.0	58	83.5	0.036
37	2.1	66.5	186	12.8	15.0	58	82.0	0.032
48	1.1	-	186	-	-	-	21.5	0.030
49	2.2	-	186	-	-	-	44.0	0.030
50	5.1	-	186	-	-	-	102.0	0.021
60	4.1	•	200	24.4	30.0	61	81.3	0.028
61	4.1	-	200	24.4	30.0	61	81.3	0.029
62	2.0	-	200	-	50.0	-	40.6	0.028
63	2.0	-	200	-	50.0	-	40.6	0.022

· ·

Table 4-1: Flat Film Processing Conditions.

a summary of the processing conditions used to produce the LDPE-1 tubular blown films evaluated in this study. The film thickness, in most cases, was kept constant. The resulting screw rotational speed, frost line height, film speed and draw ratio were recorded. In some cases, the frost line height could not be seen and as a result was not reported. All the films were produced using processing conditions which resulted in a stable bubble. The cooling conditions, defined by the air flow rate and air ring geometry, were not measured. The die gap in all cases was set as 1 mm. Film samples UE6 to 37 were produced using a 25.4 cm annular die, whereas samples UE40 to UE63 were produced using a 12.7 cm die. Finally, samples UE7, UE10 and UE13 were produced using a rotating die (0.17 rpm). The LDPE-1 tubular blown films were evaluated to determine the following:

- (a) Crystalline Phase Orientation
- (b) Amorphous Phase Orientation
- (c) Thermal Properties
- (d) Mechanical Properties
- (e) Optical Properties
- (f) Physical Dimensions.

The orientation of the crystalline phase was measured using wide angle x-ray diffraction and infrared dichroism. A conventional flat plate Laue x-ray camera was used with specially designed sample holders to position the plastic film normal, parallel and perpendicular to the x-ray beam. The xray photographs provided information regarding the a and b crystal axis orientations. The angle of the crystal axis was determined by direct inspection of the x-ray photographs. The limitation of this method is that it provides only qualitative identification of the orientation. The use of the flat plate technique to prepare pole figures and calculate orientation functions was attempted, but, due to the lack of reproducibility in the data, it was not used. The amorphous

phase orientation was measured using birefringence, infrared dichroism and sonic velocity techniques. The sonic modulus technique [63] has been used to characterize the orientation in fibers and injection molded samples. The sonic modulus technique measures the average orientation, just as the birefringence does, and has been shown to correlate with the orientation functions and the birefringence. The fractions of crystalline to amorphous material were estimated by measuring the sample density using a density gradient column. The thermal properties, specific heat and melting characteristics were measured using a Perkin Elmer differential scanning calorimeter. The optical properties were determined by measuring the 20 degree gloss, 60 degree gloss and contrast ratio of the flat films. The mechanical properties were determined by measuring the tensile modulus, tensile yield strength and impact strength. Finally, the physical dimensions of the flat films were obtained by measuring the film thickness and film layflat width.

To test the applicability of the stress crystallization model developed by Keller and Machin [75], a series of films were prepared using low, linear low and high density polyethylene resins. These materials will be referred to as LDPE-2, LLDPE and HDPE. The rheological properties of these resins were previously determined by Utracki et al [92,93,98]. Details of the experimental tests performed and the results are presented in Appendix C and References 99 and 100.

4.1.2.1. Sampling Procedure

The sampling procedure used to prepare the film samples for the various characterization tests was determined based on evaluating the variability of the orientation (crystalline and amorphous) and physical dimensions in the machine and transverse directions in the tubular blown films. The effect of rotating the extruder die was also considered. Flat film samples UE8 and UE10 were selected for this assessment.

Typically, commercial tubular blown films exhibit thickness variations ranging from ±5 to ±8% [14]. The film thickness was found to vary by only ± 2.8 and ± 3.8 % for samples UE8 and UE10, respectively, when evaluated over a 5 m section. The film width, measured as the layflat length, was found to vary by less than ±0.5% when measured over a 5 m interval for these films. Statistical tests were performed at a 95% confidence interval to see if the film thickness varied across the film width and along the film length. In all cases, the null hypothesis was found to be true, i.e. the means were found to be equal. The density of these film samples was found to vary by less than ± 1.5 % over a 5 m interval. In addition, the variation of the density across the film length and width was found to be negligible. The crystalline phase orientation, when assessed using wide angle x-ray diffraction and the 730 cm⁻¹ band infrared dichroic ratio, was found to be uniform regardless of where the test samples were taken from. The variability of the data was on average less than ± 10 %. Finally, the average orientation was also found to be uniform throughout the film, when assessed using the birefringence and sonic velocity.

The effect of rotating the blown film die was found to be negligible for the films evaluated. Rotation of the die is commonly used to minimize thickness and other variations which may exist across the film width. The variability in the test data for the films produced using a rotating die was found to be equal to the variability when the non-rotating die was used. The only difference noted when using the rotating die was that the x-ray diffraction patterns of the plastic films were tilted, up to a maximum 5 degrees, as compared to the xray diffraction patterns of the plastic films produced using the non-rotating die. This difference however, was not found to affect the results when the variability in the data was included in the analysis.

The above results suggest that it is fair to conclude that the properties of the flat films produced using the film blowing system in this study, were constant regardless of the location of the test sample. The effect of die rotation for the system used, was found not to be significant. In addition, the sampling procedure used involved the use of at least three samples, in most cases, evenly spaced across the collapsed film web. In cases where more than a single layer was evaluated, sections from both sides of the layflat were taken.

The effect of aging, as defined by the time taken to test the flat films after being produced, was evaluated by measuring the film density, sonic velocity, birefringence, infrared dichroic ratio and x-ray pattern after one day, one week, one month and six months. In all cases, no differences were noted between the films tested.

4.1.3. Results

Figure 4-2a illustrates the relationship between the blowup ratio and the flat film thickness, freeze line height, draw ratio and film speed used to prepare the LDPE-1 tubular blown films evaluated in this study. The degree of correlation between the blowup ratio and the freeze line height, draw ratio and film speed was found to be very small. On the other hand, the relationship between the draw ratio and film speed as shown in Figure 4-2b, was found to be linear, as expected.

Figure 4-3 presents the wide angle x-ray diffraction data obtained using the Laue flat plate camera technique. The xray patterns obtained for the thickness and transverse plane (ND-TD) showed random a and b-axis orientations. The angle of inclination of the a-axis from the plane of the film (MD-TD)



Figure 4-2a: LDPE-1 Film Blowing Conditions.



Figure 4-2b: LDPE-1 Film Blowing Conditions.



Figure 4-3a: X-Ray Diffraction Data, a-axis.



Figure 4-3b: X-Ray Diffraction Data, b-axis.

for the films evaluated, was found to vary from 35 to 85 degrees depending on the blowup ratio. The orientation of the a-axis in the plane of the film was found to be predominantly in the machine direction. The orientation of the b-axis was found to be in the transverse direction when measured in the MD-TD and ND-MD planes, regardless of the blowup ratio. This orientation is characteristic of the row structure model. For both the a and b axes, the spread of the diffraction pattern was found to increase as the blowup ratio increased.

No correlation was found between the blowup ratio and the dichroic ratios measured at 730, 1080 and 1352 cm⁻¹ (Figure 4-Dichroic ratios equal to unity indicate that the 4). orientation of the chains is equally distributed in the machine and transverse directions. Values greater than or less than unity, indicate that the orientation is more pronounced direction. The infrared dichroic ratios in the machine measured at the 730 cm^{-1} band, representing the *a*-axis orientation, were found to be greater than unity, thus indicating a-axis orientation. The correlation between the dichroic ratios measured at the 730 cm^{-1} band and the x-ray diffraction data for the a-axis, measured in the plane of the film (MD-TD), was low, i.e. R=0.28. This difference may be due to the low degree of orientation observed, i.e. large spread of the diffraction pattern. The dichroic ratios measured at 1080 and 1352 cm⁻¹ yield information with respect to the alignment of the tie chains and loose chain folds in the amorphous region. The orientation of the tie chains at 1080 cm^{-1} was found to be in the machine direction, similar to the a-axis orientation. The variability of the dichroic ratio data measured at the 1080 cm⁻¹ band was found, however, to be great because of the weak signal measured at this wavenumber. The orientation of the loose chains at the 1352 cm⁻¹ band indicated random orientation, i.e. dichroic ratios equal unity.



Figure 4-4: Infrared Dichroism Data.

The film density and crystallinity were found to be independent of the blowup ratio.

The sonic velocity data, represented as the ratio of the sonic velocity measured in the machine direction to the sonic velocity measured in the transverse direction (Figure 4-5) was found to be independent of the blowup ratio. The correlation between the infrared dichroic ratios and the sonic velocity ratio was also found to be less than 0.1 (i.e. poor correlation). The sonic velocity ratio in all cases was found to be less than unity, indicating that the average orientation of the amorphous and crystalline phases was slightly stronger in the transverse direction.

The birefringence was found to be related to the blowup ratio in a linear manner for blowup ratios less than 3, as illustrated in Figure 4-5. The birefringence of the films measured was found to include both positive and negative values. However, the majority of films tested had a negative birefringence. The correlation between the infrared dichroic ratios and the birefringence was found to be poor regardless of the wavenumber used to calculate the dichroic ratio. Since the birefringence and sonic velocity are measures of the average orientation, the degree of correlation between the two variables was found to be high as shown in Figure 4-6. As with the sonic velocity ratio, the birefringence data indicate that the average orientation is in the transverse direction. A strong correlation was also found to exist between the angle of inclination of the a-axis in the MD-ND plane and the birefringence (Figure 4-6).

Examination of the plastic films using a polarizing microscope did not show the presence of spherulites (i.e. characteristic Maltese cross pattern).



Figure 4-5: Sonic Velocity and Birefringence Data.





The tensile properties of the flat films as a function of the blowup ratio are represented in Figure 4-7. As the blowup ratio increased, the tensile modulus was seen to shift from a maximum in the machine direction towards a maximum in the transverse direction. The ultimate tensile strength and elongation were found to shift from a maximum in the transverse direction to a maximum in the machine direction. The tensile modulus when plotted against the birefringence (Figure 4-6), yielded a linear relationship. The same trend was observed when the tensile properties were plotted as a function of the angle of inclination of the a-axis from the MD-TD plane. No correlation was found between the impact properties of the flat films and the blowup ratio.

The surface gloss, measured at 20 and 60 degrees, was found to increase as the blowup ratio increased, as illustrated in Figure 4-8. No correlation, however, was observed when the contrast ratio was plotted against the blowup ratio.

The specific heat and melting characteristics of the flat films were found to be independent of the blowup ratio.

4.1.4. Discussion

The orientation of the crystalline phase of the films evaluated was found to be characteristic of a row type structure. The *a*-axis of the unit cell was found to be oriented in the machine direction at an angle of inclination ranging from 30-80 degrees from the plane of the film. The *b*axis of the unit cell was found to be oriented perpendicular to the machine direction. Cylindrical symmetry about the ND-TD plane was found to exist for all the films evaluated. Furthermore, according to the stress crystallization model of Keller and Machin [75], these films could be categorized as having an intermediate stress crystallization orientation (Figure 4-1). As the blowup ratio increased, the angle of

15-0 0 Ultimate Elongation 10-0 5 0 Ο 0 3 **Ultimate Strength** ο 0 2. 0 1 0 0 0 ο 1.2 Modulus 1.0-°0 0.8 0 Ο 0 0.6+-0 3 Blowup Ratio 2 5 4 1



6



Figure 4-8: <u>Surface Gloss Data.</u>

inclination of the *a*-axis of the polyethylene unit cell was found to increase when measured in the MD-ND plane. According to the stress crystallization model [75], this is indicative of an increase in the stress level at the freeze line as the blowup ratio is increased. Since the stresses developed in the film were not measured, it was not possible to verify this conclusion.

The high degree of correlation between the birefringence and the angle of inclination of the a-axis and the MD-TD plane is in agreement with the observations made by Holmes and Palmer [52]. The birefringence is related to the stress through the rheo-optical law, which has been shown to be valid for both melt systems and amorphous materials [80]. Although the rheooptical law is known not to be appropriate for semicrystalline qualitative means materials. it still provides a of illustrating the correlation between the a-axis orientation and the birefringence. The higher the stress level, the greater the angle of inclination of the *a*-axis from the plane of the plastic film (MD-TD) and the greater the birefringence.

The orientation observed in polyethylene films, as noted by Maddams and Preedy [76-78] can consist of different stress types superimposed on each other. This phenomenon was confirmed when it was observed that the x-ray diffraction patterns did not consist of a single point, but rather of an arc of varying intensities. The spread of the diffraction arc for the films evaluated, was at times found to be as large as 90 degrees, indicating that the chain orientation distribution included components which were perpendicular to each other. Figure 4-9 presents the intensity distribution for the a and b axes diffraction rings for sample UE47, measured in the MD-TD plane. The intensity distribution of the a-axis is seen to exhibit a maximum at 90 and 270 degrees. However, significant orientation is also observed at other angles. To explain this phenomenon, Maddams and Preedy [76-78] postulated that the



Figure 4-9: X-Ray Diffraction Pattern Intensity Distribution, UE47.

crystallization process begins with the very high molecular weight material and proceeds over a temperature interval until the lowest molecular weight material has solidified. Under these circumstances, reorientation can easily occur and some of the crystal axes will shift toward a low stress type orientation. The processing conditions play an important role in determining whether reorientation of the crystal axes occurs.

The orientation of the amorphous phase was found to be in the in the film plane (MD-TD), transverse direction using birefringence and sonic velocity techniques. The negative birefringence was thought to result mainly from the amorphous chains being oriented in the transverse direction, even though the c-axis of the crystallites is assumed to be oriented in the machine direction according to the row structure model. This orientation is consistent with the observations made by [47], i.e. the amorphous orientation is Holmes et al perpendicular to the crystallite chain orientation. The amorphous orientation based on the analysis of the tensile data confirms the conclusion made above. The orientation of the tie chains, measured using the dichroic ratios at 1080 cm⁻¹, was found to be slightly in the machine direction. The variability of the dichroic ratio data was found to be high for the 1080 and 1352 cm^{-1} bands and accounts for the different orientation suggested for the amorphous chains. The orientation of the loose chain folds, measured using the dichroic ratios at 1352 cm^{-1} , were found to be very low. These results are similar to those obtained by McRae and Maddams [79] and are consistent with the claim that the films produced have a low to intermediate stress orientation. McRae and Maddams [79] also concluded that at low stress levels, the dichroic ratios measured at 1080 cm⁻¹ are significant, whereas, when the stress during crystallization is high, it is the dichroic ratios measured at the 1352 cm⁻¹ band which are significant. The randomness of the loose chain folds

indicates that the lamellae are not aligned in a regular fashion, i.e. low degree of orientation. This conclusion is consistent with the x-ray data, where the spread of the intensity patterns was found to be high.

Gloss is a parameter directly related to the physical state of a surface and is the capacity of a surface to reflect light [101]. A high specular gloss can be associated to a smooth surface, while a low specular gloss to a rougher surface. The degree of surface roughness of the plastic films evaluated, based on the data in Figure 4-8, decreased as the blowup ratio increased. This trend can be attributed to two different mechanisms: crystallization induced surface roughness or die flow induced surface roughness [102]. When the gloss data was plotted against the birefringence and the angle of inclination of the a-axis from the plane of the film (MD-TD), Figure 4-10, a linear relationship was observed. This supports the first mechanism. When the gloss data was plotted against the draw ratio, Figure 4-10, a linear trend was also observed, supporting the second mechanism. Huck and Clegg [103] suggest that extrusion defects are smoothed out by drawing of the film, i.e. increasing the draw and blowup ratios. Clearly, the variations in the gloss data result from a combination of the crystallization induced surface roughness and die/film flow induced surface roughness.

The morphology and orientation in polyethylene films can be explained by comparing the elongational viscosity of the resins. For example, as described in Appendix C, the rheological properties of the linear low density polyethylene sample were very similar to those of the high density polyethylene sample. The elongational viscosity of these two resins was found to be lower than that of the low density polyethylene resin. Assuming that the elongational viscosity gives an estimate of the stresses at the freeze line, we would expect the linear low density polyethylene and the high



Figure 4-10: Correlation Between the X-Ray, Birefringence, Draw Ratio and Gloss Data.

density polyethylene samples to exhibit a low stress type of orientation and the low density polyethylene an intermediate type of stress orientation. These predictions were confirmed using x-ray, birefringence and infrared dichroism techniques. The orientation in the linear low density films, by virtue of the smooth nature of the stress strain curve, was predicted remain unchanged over a wide range of processing to conditions. This feature is one of the reasons why linear low density polyethylene resins can be down gauged significantly without breakage of the film bubble. Alternatively, the stress strain curve for low density polyethylene only allows for down gauging over a short range before high stresses lead to failure or yield. These observations, coupled with the fact that the cooling conditions used to produce the linear low density polyethylene films had to be adjusted, were verified by noting that the overall degree of orientation in the linear low density polyethylene films was lower than that found in high density films. In this study, it was found that bubble stability was achieved for low air cooling rates. This would lead to a longer relaxation time thereby allowing for reorientation. The exact angle of inclination of the a-axis from the MD-ND plane could not be estimated. Nonetheless, this approach was useful in confirming the results obtained.

The pole figures of Lindenmeyer and Lustig [57] and Desper [71], Kwack, Han and Vickers [81] showed a tendency of low density polyethylene toward a higher stress condition, as compared to high density and linear low density polyethylene, which tended toward a lower stress case. They support the postulation that based on the elongational stress behaviour and chain mobility of the resin, it is possible to predict the type of crystallization process, which in turn will lead to a specific orientation of the *a*-axis orientation in a film. The processing conditions also have an effect on the stresses exerted on the film.
If the rheological properties of a resin are known, it would be possible to obtain predictions of the stress distribution in the film as a function of distance from the die by using mathematical models. This information could then be used to see the effect of varying processing conditions on the film orientation.

4.2. EMBOSSED FILM ANALYSIS

The results presented in the previous section illustrated the relationship between several tubular blown film process variables and the optical, physical, morphological and mechanical properties of the flat films produced. This section will discuss how the techniques employed in the previous section were used to characterize embossed polyethylene films and to examine the effect of varying the embossing processing conditions on the final properties of the embossed films. In addition, the key process variables determining the temperature distribution, stress distribution, morphology and orientation of embossed films were identified.

4.2.1. Previous Studies

As discussed earlier, there is very little published literature available regarding the analysis of the film embossing process or the properties of embossed films. One possible reason is that the majority of technical work done in this area is proprietary in nature. Some general observations have been made, however [7]:

 As the preheat roll temperature is increased, the bulk thickness of an embossed film increases.
As the embossing pressure is increased, the bulk thickness of an embossed film increases.
As the hardness of the rubber on the backup roll is increased, the bulk thickness decreases. 4. As the temperature of the cooling water used to cool the engraved embossing roll is increased, the bulk thickness of an embossed film decreases.

An oriented semi-crystalline polymer will, upon heating, tend to retract toward its unoriented state. If the sample is constrained while the temperature is increased, a retractive force will develop [104,105] in a direction parallel to the fixed ends. The annealing of drawn polyethylene samples [104-107] has been shown to mobilize the extended amorphous tie molecules which try to assume a random configuration. The process observed is not random, but proceeds following a specific order, ultimately resulting in random orientation of the molecules. If a sample is annealed with free ends, no retractive forces are developed.

If the temperature of plastic film in the thermal treatment zone is too high, film shrinkage in the lateral direction may occur. If the degree of lateral shrinkage is high, then the film may tear due to high retractive stresses developed in the film. In addition, changes in the crystal orientation due to relaxation processes (i.e. annealing) can occur. The retractive stresses developed in the film are dependent on the properties of the material and the processing conditions. If the relaxation process is not controlled, not only can changes to the orientation of the crystal and amorphous chains occur, but the physical dimensions of the flat film prior to entering the embossing zone nip region may change. The degree of relaxation, of which the lateral shrinkage is a measure, is usually specified as zero by the processor to avoid problems.

The embossing stage of the embossing process can be viewed as a rolling operation. Previous authors who have analyzed the rolling process [106-116] have shown that as the rolling force is increased, which is defined by the degree of thickness reduction, the orientation of the crystalline phase in

polyethylene changes toward the *c*-axis being oriented in the rolling direction and the *a*-axis in the thickness direction. The yield and tensile strength values in the rolling direction usually increase as a result. The observed phenomena are not expected to apply to the present situation because the forces applied while embossing a plastic film are not intended to alter the dimensions of the plastic film. Therefore, we would not expect changes to occur in the orientation of the crystalline phase, amorphous phase and film thickness during the rolling stage of the process.

4.2.2. Experimental

A total of 106 embossed films were analyzed to evaluate the relationships between the process variables and the embossed film properties. Table 4-2 presents a summary of the processing conditions used to produce the embossed films evaluated in this study. All the flat films used to prepare the embossed films were produced using the tubular film process and LDPE-1. Flat films coded UE60 and 61 were used to produce samples E14-E98. Embossed films coded E1-13 were produced using the continuous embossing station, while samples E14-106 were produced using the batch embossing system. The temperature distribution of the plastic films was measured as a function of the processing conditions using the continuous embossing system and film samples UE14 to 37 and E1 to 13. The process variables analyzed were:

- (a) Film Thickness
- (b) Flat Film Blowup Ratio
- (c) Film Velocity
- (d) Embossing Pressure
- (e) Preheat Roll Temperature
- (f) Radiation Heater Temperature.

Embossed	Flat Film	Tp	$\mathbf{T}_{\mathbf{R}}$	Pressure	Velocity
<u>Sample #</u>	<u> </u>	<u>°C</u>	<u>°C</u>	setting	<u>_m/min_</u>
EI	-	49.7	830.0	max	-
2	-	79.0	832.2	max	-
3	-	81.7	832.2	max	-
4	-	84.9	829.4	max	-
5	-	86.7	821.7	max	-
6	-	90.0	822.2	max	-
7	30	37.8	676.7	max	-
8	31	37.8	815.6	max	-
9	32	37.8	537.8	max	-
10	34	65.6	off	max	-
11	35	65.6	815.6	max	-
12	36	93.3	815.6	max	
13	37	93.3	off	max	-
14	60	93.3	off	0.5	18.3
15	60	93.3	off	max	18.3
16	60	93.3	787.8	max	18.3
17	60	93.3	787.8	0.5	18.3
18	60	93.3	537.8	0.5	18.3
19	60	93.3	537.8	max	18.3
20	60	93.3	848.9	max	18.3
21	60	93.3	848.9	0.5	18.3
22	60	93.3	787.8	max	16.8
23	60	93.3	787.8	max	13.7
24	60	93.3	787.8	max	19.8
25	60	93.3	787.8	0.5	19.8
260	60	21.1	off	0.5	19.8
270	60	21.1	off	max	19.8
280	60	21.1	787.8	max	19.8
290	60	21.1	787.8	0.5	19.8
300	60	21.1	537.8	0.5	19.8
310	60	21.1	537.8	max	19.8
320	60	21.1	843.3	max	19.8
330	60	21.1	843.3	0.5	19.8
26	61	21.1	787.8	max	19.8
27	61	21.1	787.8	0.5	19.8
28	61	21.1	537.8	max	19.8
29	61	21.1	537.8	0.5	19.8
30	61	21.1	871.1	max	19.8
31	61	21.1	871.1	0.5	19.8
32	61	93.3	off	max	19.8
33	61	93.3	off	0.5	19.8
34	61	93.3	787.8	0.5	19.8
35	61	93.3	787.8	max	19.8

Table 4-2: <u>Embossed Film Processing Conditions.</u>

Embossed	Flat Film	$\mathbf{T}_{\mathbf{P}}$	$\mathbf{T}_{\mathbf{R}}$	Pressure	Velocity
<u>Sample #</u>	<u>UE#</u>	°Č	°C	setting	m/min
E36	61	93.3	537.8	0.5	19.8
37	61	93.3	537.8	max	19.8
38	61	93.3	843.3	0.5	19.8
39	61	93.3	843.3	max	19.8
40	61	65.6	787.8	max	19.8
41	61	65.6	787.8	max	16.8
42	61	65.6	787.8	max	13.7
43	61	65.6	815.6	max	19.8
44	61	65.6	815.6	max	16.8
45	61	65.6	815.6	max	13.7
46	61	65.6	843.3	max	19.8
47	61	65.6	843.3	max	16.8
48	61	21.1	off	max	19.8
49	61	21.1	off	0.5	19.8
50	61	21.1	787.8	max	13.7
51	61	21.1	787.8	max	16.8
52	61	21.1	815.6	max	19.8
53	61	21.1	815.6	max	16.8
54	61	21.1	815.6	max	13.7
55	61	21.1	843.3	max	19.8
56	61	21.1	871.1	max	19.8
57	61	21.1	882.2	max	19.8
58	61	21.1	882.2	max	18.3
59	61	21.1	882.2	max	16.8
60	61	21.1	882.2	max	13.7
61	62	21.1	off	max	19.8
62	62	21.1	off	0.5	19.8
63	62	21.1	787.8	0.5	19.8
64	62	21.1	787.8	max	19.8
65	62	21.1	537.8	max	19.8
66	62	21.1	537.8	0.5	19.8
67	62	21.1	879.4	max	19.8
68	62	21.1	879.4	0.5	19.8
69	62	93.3	787.8	max	19.8
70	62	93.3	787.8	0.5	19.8
71	62	93.3	537.8	0.5	19.8
72	62	93.3	537.8	max	19.8
73	62	93.3	871.1	max	19.8
74	62	93.3	871.1	0.5	19.8
75	63	93.3	off	max	19.8
76	63	93.3	off	0.5	19.8
77	63	93.3	787.8	max	19.8
78	63	93.3	732.2	may	16.8

Table 4-2: Embossed Film Processing Conditions.

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Embossed Sample_#	Flat Film <u>UE#</u>	T _P °C	T _R °C
E79	63	93.3	732.2
80	63	93.3	732.2
81	63	21.1	off
82	63	21.1	off
83	63	21.1	787.8
84	63	21.1	787.8

E/9	63	93.3	132.2	max	13./
80	63	93.3	732.2	max	19.8
81	63	21.1	off	max	19.8
82	63	21.1	off	0.5	19.8
83	63	21.1	787.8	max	19.8
84	63	21.1	787.8	0.5	19.8
85	63	21.1	537.8	0.5	19.8
86	63	21.1	537.8	max	19.8
87	63	21.1	787.8	max	19.8
88	63	21.1	815.6	max	13.7
89	63	21.1	815.6	max	19.8
90	63	21.1	843.3	max	19.8
91	63	21.1	871.1	max	19.8
92	63	21.1	787.8	max	16.8
93	63	93.3	787.8	0.5	19.8
94	63	93.3	537.8	0.5	19.8
95	63	93.3	537.8	max	19.8
96	63	93.3	835.0	max	19.8
97	63	93.3	835.0	0.5	19.8
98	63	93.3	871.1	max	19.8

note: A pressure setting of 0.5 corresponds to an air pressure of 0.1 MPa and max to an air pressure of 0.2 MPa.

Table 4-2: <u>Embossed Film Processing Conditions.</u>

Pressure Velocity <u>setting m/min</u>

The preheat roll temperature was varied between room temperature and 93.3°C, the radiation heater temperature between room temperature and 815.5°C, the embossing pressure between 0.1 and 0.2 MPa, the flat film blowup ratio between 2:1 and 4:1, the film velocity between 13 and 20 m/min and the film thickness between 0.025 and 0.055 mm. The film thickness was varied by embossing single and twin film webs. The standard operating conditions for the batch embossing process [111] were:

> Line velocity, V=20.5 m/min Embossing pressure, P=0.2 MPa Preheat roll temperature, $T_P=93.3^{\circ}C$ Radiation heater temperature, $T_R=787.8^{\circ}C$

The embossed films were evaluated using the techniques described in the previous section, to determine the crystalline phase orientation, amorphous phase orientation, mechanical properties, optical properties and physical dimensions. In addition to these tests, the film temperature distribution and shrinkage during processing were measured.

Embossed films, unlike flat films, have a three dimensional geometry. Therefore, certain standard characterization techniques cannot be used. The birefringence was difficult to measure because the variation in pattern height made it difficult to measure the extinction angle. Furthermore, due to the small pattern size, it was difficult to focus the optical microscope on a specific area of the film at high magnifications. When using x-ray or infrared dichroism to evaluate the morphology, the images and spectra measured represent only average values, i.e. encompassing several embossments. It was not possible using these techniques, to focus the x-ray or infrared source beam on a specific area of the film in order to measure the orientation distribution in the film. For example, the diameter of the sample area focused

upon using the wide angle x-ray diffraction camera was 0.75 mm. The dimensions of an embossment are smaller than the diameter of the source beam.

Table 4-3 presents data comparing the true length of an embossed film, L_0 , against the layflat length, L. The embossed films were analyzed using an optical microscope. Five embossed film cross sections were analyzed for each sample. Further, the film section being analyzed was divided into 19 segments and the length of each segment was added together to yield the true film length. The ratio of the true film length to the layflat length, L_0/L , for the films evaluated was found to range from 1.01 to 1.04. Due to the small differences observed, the layflat length was used in all calculations where the width and length of the test samples was required (e.g. the mechanical and sonic velocity calculations).

The thickness of embossed films can be specified in terms of a film and bulk thickness, as illustrated in Table 4-3. The film thickness refers to the actual thickness of the plastic film, whereas the bulk thickness refers to the thickness of the pattern as measured from the underside of the film to the top of embossed pattern. The bulk thickness is always greater than or equal to the film thickness. The bulk thickness is equal to the film thickness for flat samples. A microscope was used to measure the bulk thickness and the density gradient technique to measure the film thickness. A micrometer could not be used to measure the thickness because the embossed pattern interferes with the micrometer head and crushes the embossment. No problems due to knife effects were encountered using the microscope technique when cutting the embossed film. Although the accuracy of both techniques was approximately ± 0.005 mm, no other method could be found to determine the thicknesses more accurately.



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E810.4310.641.0E910.2210.291.0E1011.2011.511.0E1110.7110.881.0E1211.4311.851.0E1310.5910.741.0	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	1.02 1.01 1.03 3 1.02 5 1.04 4 1.01

Table 4-3: Embossed Film Length Determination.

The optical properties of the embossed films were measured using a HunterLab Gloss Meter at 20° and 60° . The gloss measurements were all made with the embossed pattern facing upwards and with the gloss meter source beam parallel to the machine direction. The direction of the source beam relative to the embossed film was found not to affect the gloss readings. The contrast ratio or opacity of the films [118] was measured using a Hunterlab Colorimeter. The contrast ratio is defined as the reflectance of the film sample over a black standard, CIE Y=0, divided by the reflectance of the same sample over a white standard, CIE Y=84.0.

Products made using embossed films should not only satisfy functional specifications, but should also satisfy some subjective requirements, particularly appearance and feel or touch, since embossed products usually replace products previously made of cloth. The measurement of the touch or feel of plastic materials is mainly qualitative and the results depend on the person taking the measurement. The texture and quality of the embossed films were evaluated using a qualitative rating system. The texture of the embossed films was ranked using a scale which rated a flat film as one and a film with the most cloth-like texture as five. Embossed film E98 was found to have the most cloth-like texture as compared to all the other samples produced. Pattern quality refers to how well the embossed pattern duplicates the metal roll pattern. Ideally the embossed pattern should be an exact duplicate of the engraved pattern. The pattern quality was rated upon visual analysis of photographs of the embossed films using a polarizing microscope. Photograph A-1 illustrates the scale used to make these measurements.

The shrinkage of the embossed films was measured using the following two techniques:

<u>On-line measurements</u>: The lateral film shrinkage was measured in two ways. The first compared the initial film width to the film width after processing. The second method was to mark two lines separated by a known amount on the flat film and to measure this separation after processing. The shrinkage of the film in the machine direction could only be measured using the second method.

<u>Static experiments:</u> Experiments were conducted using precut pieces of film which were heat treated in an oven under controlled conditions to determine the shrinkage as a function of temperature and time.

The surface temperature distribution of the plastic films as a function of the processing conditions was measured using the same procedures described in the previous chapter to measure the temperature of the preheat roll. These data will also be used in the next chapter to evaluate the mathematical models.

4.2.2.1. Sampling Procedure

The variability of the properties of the embossed films evaluated as a function of position, i.e. along the machine and transverse directions, was analyzed. The dimensions, properties and morphology of the films were found to be uniform regardless of the location of the samples taken. This is not surprising since the flat film properties and the air distribution in the nip region (Z=0.65 m) were also found to be uniform. Table 4-4 summarizes the variability of the density, shrinkage, thickness and mechanical property data measured at selected positions. Statistical tests were performed at a 95% confidence interval to see if the properties varied across the film width or along its length. In all cases, the null hypothesis was found to be true, i.e. the means were equal.

<u>Property</u>	<u>n</u>	Confidence <u>Interval (%)</u>
Tensile		
Modulus	20	4.6
Ultimate	20	8.4
Density	6	3.2
Shrinkage		
Lateral	4	1.2
Marker Method	12	0.6
Static	10	6.3
Thickness		
Microsope	15	3.0
Density Column	25	5.7

Table 4-4: Embossed Film Property Variability.

The same sampling procedure that was described in the previous section was used to characterize the embossed films.

4.2.3. Results

4.2.3.1. Temperature Distribution

The temperature of the film web before passing over the preheat roll was found to vary between 27.5°C and 29°C, depending on the day the films were produced. The initial temperature profile in the plastic films was found to be uniform across the film width, when the confidence intervals were calculated.

Figures 4-11a to 4-11c illustrate the flat film surface temperature distribution between the preheat and embossing rolls at various preheat roll and radiation heater temperature settings. Position Z=0 refers to the top of the preheat roll, Z=0.24 m at the separation point on the preheat roll, Z=0.43 m before the radiation heaters and Z=0.65 m after the radiation heater, based on the dimensions of the continuous embossing system.

As the preheat roll and/or radiation heater temperature setting was raised, the surface temperature of the plastic film increased. The temperature of the plastic film on the preheat roll when the radiation heaters were turned off, was found to be uniform and equal to the surface temperature of the preheat roll regardless of the film thickness, film velocity or position on the preheat roll. As the radiation heater temperature increased, the temperature of the film at the separation point on the preheat roll and beyond, was found to increase. The maximum film temperature measured was 105°C (Figure 4-11c) when the preheat roll temperature setting was 93.3°C and the radiation heater temperature setting was 815.6°C. The temperature distribution in the plastic film,



Figure 4-11a: Film Surface Temperature, T_p=37.8°C, <u>BUR=2:1, V=21.9 m/min and H=0.021 mm.</u>



Figure 4-11b: Film Surface Temperature, T_p=65.6°C, BUR=2:1, V=21.9 m/min and H=0.021 mm.



Figure 4-11c: Film Surface Temperature, T_P=93.3°C, BUR=2:1, V=21.9 m/min and H=0.021 mm.

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when the radiation heaters were turned on, was found to be uniform along the film width when the variation in the data was considered. Figure 4-12 plots data from Figure 4-11 against the machine direction position variable, i.e. Z, for a preheat roll temperature setting equal to 93.3°C. The film temperature was seen to increase in a linear manner, with the slope increasing as the radiation heater temperature setting was increased.

$\underline{\mathbf{T}}_{\mathbf{R}}$	Intercept	<u>Slope</u>	\underline{R}^2
off	91.15	0.15	0.08
537.8°C	91.06	11.34	0.98
815.6°C	91.28	21.86	1.00

No significant changes were observed in the film temperature as the film thickness and embossing pressure were increased. As the film velocity was increased, only a slight decrease was observed in the film temperature, i.e. less than 10%. The film velocity was maintained at a level where the film shrinkage was kept below 5% and, as a result, varied by less than $\pm 20\%$ from the standard conditions. The narrow velocity range accounts for the small differences observed.

The temperature of the plastic film entering the nip region was found to be below the melting temperature of LDPE-1. This was confirmed visually and by the fact that when embossing two layers simultaneously the two webs could easily be separated after processing.

The temperature of the embossed film after processing was found to range from 28 to 34°C. In all cases the temperature distribution was found to be uniform across the embossing roll and film width, when measured at the separation point on the embossing roll.



Figure 4-12: Film Surface Temperature, $T_P=93.3^{\circ}C$, <u>Y/W_F/2=0, BUR=2:1, V=21.9 m/min and H=0.021 mm.</u>

Temperature, °C

4.2.3.2. Embossed Film Characterization

The embossed film characterization data are presented as a function of the following process parameters: thermal treatment, film velocity, embossing pressure, film thickness and flat film orientation. Figures 4-13 and B-1 to B-4 (Appendix B) summarize the results of these evaluations.

Effect of Thermal Treatment

The temperature of the plastic film prior to embossing should be as high as possible without changing the dimensions and structure of the flat film. If the temperature is too high, the plastic film will shrink thereby altering the structure, properties and dimensions of the initial flat film.

A detailed tabulation of the variation in the embossed film thickness, bulk thickness, shrinkage, *a*-axis orientation, crystallinity, infrared dichroic ratios, sonic modulus, tensile properties, pattern quality, pattern texture and surface gloss for different preheat roll and radiation heater temperature settings is presented in Figures 4-13a to 13f.

The lateral shrinkage, film thickness and bulk thickness as shown in Figure 4-13a, were found to increase as the temperature of the film increased. As the flat film temperature increases, the plastic film becomes softer and easier to emboss, resulting in an increase in bulk thickness. In addition, the higher the film temperature, the more the plastic film shrinks in the lateral direction, thus increasing the film thickness. Film shrinkage, as measured by the amount lateral shrinkage, was found to be significant for of conditions where the radiation heater temperature was greater than 500°C when embossing a single film web. Finally, the radiation heater temperature was found to have a greater









Static Shrinkage MD, mm

Static Shrinkage MD, mm







Figure 4-13d: Effect of Preheat Roll Temperature BUR=4:1, V=20.6 m/min, P=0.2 MPa and H=0.029 mm.



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Figure 4-13f: Effect of Preheat Roll Temperature BUR=4:1, V=20.6 m/min, P=0.2 MPa and H=0.029 mm.

effect on the amount of lateral shrinkage as compared to the preheat roll temperature.

The conditions in the thermal treatment zone are similar to those when annealing a plastic sheet with two ends fixed. In such a situation, as the temperature approaches the melting point of the polymer, the film will shrink more in the unconstrained direction. If the temperature is too high, the film sheet will tear. As the film is heated, the material tends to relax and attain a stress free random state. The temperature must be set so as to maximize the film temperature while at the same time minimizing film shrinkage. The film shrinkage, measured using the marker method, illustrated that no shrinkage occurred in the machine direction. Static shrinkage tests were also performed on the embossed films to obtain a measure of the time necessary to anneal the films. Figure 4-13b presents the shrinkage as a function of time at an annealing temperature of 100°C. The films processed at higher preheat roll temperatures exhibited a higher degree of shrinkage in the machine direction. No significant shrinkage was measured in the transverse direction. In all cases, no shrinkage occurred before 15 seconds. This period of time is still too long compared to what is required for heating and shrinkage effects to occur during processing, considering a minimum film velocity of 13 m/min.

X-ray patterns for the embossed films verified that, as the thermal treatment temperature increased, the crystal structure shifted from an *a*-axis type orientation to a completely random state (Figure 4-13c). Random orientation was found for cases where the shrinkage was greater than 10%, as illustrated in Figure 4-13d. In this figure, the x-ray data are plotted against the lateral shrinkage data. The 730 cm⁻¹ band dichroic ratio data support the results of the x-ray analysis by showing how the *a*-axis orientation changes toward the random state, i.e. dichroic ratio equal to unity. As indicated

previously, it was very difficult to measure the amorphous because weak band signals. The orientation of the crystallinity of the film samples was found to be constant up to a radiation heater temperature setting of 815.6°C (see Figure 4-13c). As the radiation heater temperature setting was increased, the crystallinity was found to decrease slightly. The sonic velocity, tensile modulus and ultimate tensile strength values were found to decrease as the thermal treatment temperature increased, until the machine and transverse direction values were equal, further indicating a random state condition at very high temperatures (see Figure 4-13e). The properties of the cold rolled films, $T_p=T_R=27^{\circ}C$, were found to be only slightly different from those of the flat film. No changes in the structure or physical dimensions were noted upon rolling. The thermal treatment of the plastic film determines the extent of change in the structure and properties of the flat film.

The pattern definition and texture were found to increase as the film temperature increased, as illustrated in Figure 4-13f. At the maximum temperature settings, the embossed films were found to have a coarse texture which is presumed to be due to the extensive amount of film shrinkage which occurred. At the minimum temperature setting, the film texture was plastic-like, which was due to the lack of pattern definition. The 60 degree surface gloss (Figure 4-13f) was found to decrease as the film temperature increased. Similar results were also obtained using a 20 degree measurement. As the film temperature increases, the pattern quality and bulk thickness increase. The surface of the film, as a result, becomes more contoured, i.e. rougher. The gloss, which is also a measure of the surface roughness, therefore decreases.

Effect of Film Velocity

The film velocity controls the residence time of the film in the thermal treatment zone. The lower the film velocity, the longer the residence time and the higher the temperature of the flat film. Ideally, it is desirable to operate at the maximum line velocity to ensure maximum production rates. The maximum line speed achievable using the batch embossing system was 20.6 m/min.

A detailed tabulation of the variation in the embossed film thickness, bulk thickness, crystallinity, sonic modulus, tensile properties, dichroic ratio, shrinkage and a-axis orientation for different film velocity settings is presented in Figures B-1a to B-1d (Appendix B). The results may be summarized as follows:

1. The lower the film velocity, slightly greater the bulk thickness and shrinkage values.

2. The orientation of the films analyzed in all cases was random. This is as expected, since the shrinkage value for all of these films was above 10%. The crystallinity and dichroic ratios were found to be constant.

3. The sonic modulus and tensile properties were found to remain fairly constant regardless of the velocity setting. This is not surprising since the x-ray results showed the orientation to be random in all cases.

4. The pattern definition and texture were found to remain constant over the entire velocity range tested.

5. The surface gloss was found to be sensitive (i.e. decrease) to changes in the velocity.

Effect of Embossing Pressure

After undergoing a specific thermal treatment, the plastic film is passed between the nip of an engraved steel roll and a rubber covered steel backup roll. A force is applied onto the embossing roll by the pneumatic system and the rubber roll. impressing a pattern into the plastic film. The embossing pressure will affect the bulk thickness of the embossed film. The lower the embossing pressure for constant thermal treatment conditions, the lower the bulk thickness should be, i.e. poorer pattern definition. The film properties are not affected by the embossing pressure, but as noted before, they are influenced by the thermal treatment. The embossing pressure is usually set at the maximum value during production in order to ensure the best possible pattern definition.

A detailed tabulation of the variation in the embossed film thickness, bulk thickness, crystallinity, sonic modulus, tensile properties, dichroic ratio, shrinkage and *a*-axis orientation for two different embossing pressure settings is presented in Figures B-2a to B-2d (Appendix B). The results may be summarized as follows:

 The bulk thickness was found to increase slightly as the embossing pressure increased, when the radiation temperature was greater than 600°C.
No differences were observed between the films prepared at a pressure setting of 0.1 MPa and those prepared at 0.2 MPa for all the measured film properties. This finding supports the conclusion that properties, or more specifically structural changes, are controlled by the thermal treatment.
The film properties for the twin web embossing system follow the same trends as those observed with the single web operation.

Effect of Film Thickness

The heat absorbed by the film is not only a function of the heater setting but also a function of the film thickness. The thicker the plastic film, the lower the overall film temperature will be for a given temperature setting and velocity.

A detailed tabulation of the variation in the embossed film thickness, bulk thickness, crystallinity, sonic modulus, tensile properties, dichroic ratio, shrinkage and *a*-axis orientation for two different film thickness values is presented in Figures B-3a to B-3d (Appendix B). The results may be summarized as follows:

 The lateral shrinkage was found to be greater when the film thickness was thinner.
Apart from the difference in the lateral shrinkage, no differences were observed between the films prepared using a single or double web configuration for all the measured film properties.
The film properties for the twin web embossing system followed the same trends as those observed with the single web operation.

Effect of Film Orientation

The film orientation determines the initial structure and properties of the flat film. However, for a specific resin and process geometry, it was very difficult to have films with substantially different structures. The differences in the two films analyzed were very small, as illustrated by the following:

		F1 <u>UE61</u>	lat Film	<u>UE63</u>	
a-axis orientatio	n spread maximum	120° 80°		45° 60°	
Tensile modulus	MD TD	157.2 M 236.7 M	IPa IPa	146.8 190.7	MPa MPa

A detailed tabulation of the variation in the embossed film thickness, bulk thickness, crystallinity, sonic modulus, tensile properties, dichroic ratio, shrinkage and *a*-axis orientation for two different blowup ratios is presented in Figures B-4a to B-4d (Appendix B).

The mechanical properties of the films produced at the two blowup ratios became equal when the orientation of the amorphous and crystalline phases changed to the random state. This was found to occur when the radiation heater temperature setting was greater than or equal to 787.8°C. Depending on the thermal conditions, the initial orientation may be unimportant.

4.2.4. Discussion

The primary objective of this part of the study was to evaluate and explain the relationships between the properties of embossed films and the processing conditions employed during the embossing process. The experimental results illustrated that the thermal treatment zone was the most significant factor. Moreover, by controlling the thermal treatment of the film, it was possible to manipulate the microstructure, properties and dimensions of the flat film. The thermal treatment was shown to be a function of the radiation heater temperature, preheat roll temperature, line velocity and film thickness. At radiation heater temperatures below 500°C, the preheat roll temperature was found to be a significant factor, while at temperatures greater than 500°C, the radiation heater temperature was the controlling factor. The film thickness, initial film orientation and line velocity were found to have a minor effect on the final properties of the embossed film. The main effect of the embossing pressure was on the bulk thickness of the embossed film.

The embossed film properties are determined, as illustrated in the previous chapter, by the transformations which have occurred during the thermal treatment phase. During the final phase of the process, the plastic film is embossed and simultaneously cooled as it passes through the nip of the engraved metal and backup roll system. The rolling operation was not found to affect the structure or orientation of the film and did not reduce the film thickness. There should be no change in orientation upon embossing because of the low degree of rolling and because there is no reduction in thickness.

The plastic film surface temperature was found to increase as the preheat roll and radiation heater temperature settings were increased. The data in Figures 4-11a to 11c were analyzed using statistical techniques and the following multiple regression model was found to yield the best fit to the data:

$$T_F = 0.82T_P + 0.03T_R + 19.74Z.$$
 (4-1)

The value of the correlation coefficient was calculated to be 0.992. The effect of including the axial position variable, i.e. $Y/W_F/2$, was found not to be significant. Equation 4-1 was determined using stepwise regression techniques. The critical F-value used to remove a coefficient, was specified as 4.0. Figure 4-14 presents graphically the relationship expressed in Equation 4-1 when the machine direction variable Z, is equal to zero, i.e. top of the preheat roll. As the preheat roll and radiation heater temperatures were increased, the plastic film surface temperature increased. Equation 4-1



Figure 4-14: Plastic Film Surface Temperature as Defined by Equation 4-1, Z=0.

is only valid for the continuous embossing system. However, the trends observed should be similar to those occurring when using the batch embossing system.

The relationship between the preheat roll surface temperature and plastic film surface temperature at the top of the preheat roll, i.e. Z=0, is illustrated in Figure 4-15. As the radiation heater temperature setting was increased, the air temperature distribution around the preheat roll increased and the film temperature became greater than the preheat roll temperature. This effect was most significant at low preheat roll temperature settings, i.e. less than 60°C. Above 60°C the film temperature was equal to the preheat roll temperature, i.e. $T_F/T_P=1$, if the variability in the data was included.

It was not possible to measure directly the film temperature at different positions along the process line using the batch embossing system, due to the design of the embossing system. For this reason, the film embossing data were presented as a function of the heater settings and not the actual film temperature. If the exact relationship between the processing conditions and the film surface temperature was known, then it would be possible to determine the processing conditions which would yield the desired embossed film properties. Chapter 5 will discuss theoretical methods to determine the film surface temperature distribution based on the equations of continuity, energy and momentum. The experimental data collected using the batch embossing system will be used to determine the accuracy of these models.

The data presented in Figures 4-13 and B-1 to B-4 were analyzed using statistical methods to determine the significant processing variables. The key process variables were determined using stepwise regression techniques. The following relationships were obtained:



Figure 4-15: Comparison of the Plastic Film Surface Temperature and Preheat Roll Surface Temperature, Z=0.

Lateral Shrinkage= $0.13T_{P}+0.02T_{R}-273.6H$ (4-2a)

 $Gloss=2.19V-0.14T_{P}-0.03T_{R}$ (4-2b)

$$a$$
-axis Orientation=2477.1H+0.30T_R (4-2c)

Bulk Thickness= $0.00043T_{p}+0.00007T_{R}+1.06H$ (4-2d)

Pattern Quality=
$$0.02T_p+0.003T_p$$
 (4-2e)

Film Thickness=1.44H (4-2f)

The value of the correlation coefficient was calculated to be 0.875, 0.962, 0.956, 0.996, 0.975 and 0.612, respectively for these equations. All the properties listed above are functions of the radiation heater temperature, except for the film thickness. In most cases, the preheat roll temperature was also a significant factor. The embossing pressure was found not to be a significant variable in all of the models evaluated.
CHAPTER 5 THEORETICAL ANALYSIS

This chapter describes the models used to predict the temperature and stress distributions in the plastic film during thermal treatment and embossing.

It was shown in the previous chapter that the thermal treatment was the most significant factor in determining the dimensions and orientation of the embossed film product. Due to design constraints in the equipment used, it was not possible to obtain detailed temperature measurements with the continuous embossing system or any temperature measurements with the batch embossing system. Mathematical models were developed to estimate the plastic film temperature as a function of the system geometry, preheat roll temperature, radiation heater temperature, air temperature, film thickness velocity order to explain and line in the final characteristics of the embossed film, in terms of the thermomechanical history, instead of the processing conditions and to estimate the effect of manipulating the processing variables on the film temperature. Again, due to equipment limitations, the deformations and pressure distribution in the nip region of the embossing roll system could not be measured and mathematical models were derived to estimate the deformations and stresses in the nip region of the embossing roll system.

5.1. HEAT TRANSFER ANALYSIS

Prior to embossing, the plastic film undergoes heat treatment by passing over a preheat roll and between two radiation heater tube banks. The purpose of the heat treatment step is to raise the temperature of the film close to the melting temperature of the material prior to passing through the nip region of the embossing and backup roll system. The heat transfer analysis was divided into four parts, i.e. modelling of the preheat roll system, radiation heater system, embossing roll system and the regions where the plastic film travels in the free space between the various process components. The preheat roll system involves mainly conduction and convection, the radiation heater system involves radiation and convection, the embossing system involves mainly conduction and, finally, the free space between components involves mainly convection.

The temperature distribution in the plastic film can be estimated by solving the equation of energy [27], which describes the transport of energy in a homogeneous fluid or solid, with appropriate initial and boundary conditions in conjunction with realistic material properties. The energy equation can be written in tensorial form as follows:

$$\rho \mathbf{C}_{\mathbf{v}} \mathrm{D} \mathbf{T} / \mathrm{D} \mathbf{t} = - (\nabla \cdot \mathbf{q}) - \mathbf{T} (\partial \mathbf{P} / \partial \mathbf{T})_{\mathbf{v}} (\nabla \cdot \mathbf{V}) - (\boldsymbol{\tau} : \nabla \mathbf{V}), \quad (5-1)$$

where ρ denotes the density, C_{v} the specific heat, T the temperature, t the time, **q** the heat flux vector, P the pressure, **v** the velocity vector and τ the viscous stress tensor.

Analytical solutions to the energy equation exist only for the simplest cases, e.g. one dimensional heat flow [28]. Kamal and Bata [119] describe in detail, current trends in the analysis of polymer shaping operations. Developments relating to the analysis and simulation of several important polymer processes are discussed, with due consideration to the elucidation of both thermo-mechanical history and microstructure development.

5.1.1. Preheat Roll System

The preheat roll is used to raise the temperature of the plastic film to the range of 90-93°C, when operating at the standard conditions. The plastic film is heated by conduction through contact with the roll surface and loses heat through convection and radiation at the free surfaces.

The following assumptions were made to simplify the energy equation, Equation 5-1, describing the preheat roll system:

1. The curvature of the cylindrical surface is negligible, since the film thickness was much smaller than the preheat roll diameter.

2. The thermal conductivity, specific heat and density of LDPE-1 were independent of temperature and pressure.

3. The film remained a solid, i.e. no phase change. As a result, the velocity components in the energy equation are zero.

4. The film velocity was constant and the time and film position components were interchangeable and related by the following equation:

where Z denotes the film position co-ordinate in the machine direction, V the film velocity and t the time.

5. Radiation heat losses are negligible, since the temperature difference between the preheat roll surface and ambient air was small, i.e. less than 80°C.

Based on the above assumptions and using the co-ordinate system defined in Figure 3-3, the two dimensional energy equation takes the following form:

$$\rho C_{v} \partial T / \partial t = k \left(\partial^{2} T / \partial X^{2} + \partial^{2} T / \partial Y^{2} \right), \qquad (5-3)$$

where k denotes the thermal conductivity. This equation was solved using the following initial and boundary conditions:

At
$$t=0$$

$$T(X, Y, 0) = f(X, Y)$$
 (5-4)

At Y = -W/2

$$-k\partial T(X, -W/2, t) / \partial Y = h(T(X, -W/2, t) - T_A(Y, t))$$
(5-5)
At Y=W/2

$$-k\partial T(X,W/2,t)/\partial Y=h(T(X,W/2,t)-T_A(Y,t))$$
(5-6)

At X=-H/2

 $T(-H/2, Y, t) = T_{P}(Y)$ (5-7)

At X=H/2

$$-k\partial T(H/2,Y,t)/\partial X=h(T(H/2,Y,t)-T_{A}(Y,t)), \qquad (5-8)$$

where W denotes the film width, $T_A(Y,t)$ the ambient air temperature, $T_{P}(Y)$ the preheat roll surface temperature, h the heat transfer coefficient, H the film thickness, X the coordinate position in the film thickness direction and Y the co-ordinate position in the film width direction. The initial condition, Equation 5-4, assumes that the temperature distribution in the plastic film at the initial point of contact with the preheat roll is known. The convective heat flux conditions at each edge of the film, Equations 5-5 and 5-6, are necessary since the temperature varies across the width of the film. Equation 5-7 indicates that the temperature of the plastic film at the point of contact with the preheat roll surface is known and is a function of the preheat roll temperature. Perfect contact is assumed between the plastic film and preheat roll surface. Equation 5-8 describes the convective heat flux at the top surface of the plastic film.

If the temperature of the preheat roll surface remains constant across the width of the preheat roll and edge effects are neglected, the heat transfer analysis reduces to the simpler one dimensional formulation:

$$\partial T/\partial t = \alpha \partial^2 T/\partial X^2$$
, (5-9)

where $\alpha = k/\rho C_v$, which denotes the thermal conductivity. The one dimensional mathematical description of the problem was solved using the following initial and boundary conditions:

At X=-H/2

$$T(-H/2,t)=T_{P}$$
 (5-11)

At X=H/2

$$-k\partial T(H/2,t) / \partial X = h(T(H/2,t) - T_A(t)). \qquad (5-12)$$

5.1.1.1. Numerical Solution

The two dimensional unsteady state heat conduction problem was solved using an implicit alternating direction numerical method [120-124]. The simplified form of the energy equation, Equation 5-3, written in a finite difference form and evaluated at the advanced point of time is as follows:

$$(T_{i,j,t+1} - T_{i,j,t}) / \Delta t = \alpha ((T_{i+1,j,t+1} - 2T_{i,j,t+1} + T_{i-1,j,t+1}) / \Delta X^{2} + (T_{i,j+1,t+1} - 2T_{i,j,t+1} + T_{i,j-1,t+1}) / \Delta Y^{2},$$
(5-13)

where $T_{i,j,t}$ denotes the film temperature at the grid positions i and j evaluated at time t. Figure 5-1 illustrates the grid system used to solve the two dimensional heat transfer problem. The subscript t+1 refers to the advanced time increment which is defined as t+ Δ t. To solve the finite difference equation having six unknowns, two finite difference equations were employed which were used over successive time



Preheat Roll Surface T=f(Y)

Figure 5-1: Finite Difference Grid System for the Preheat Roll System.

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steps, each of duration half the time increment, i.e. $\Delta t/2$. The first equation was implicit only in the X direction; the second was implicit only in the Y direction.

$$\frac{\text{Implicit X direction}}{(T_{i,j,t+1/2} - T_{i,j,t}) / \Delta t / 2 = \alpha ((T_{i+1,j,t+1/2} - 2T_{i,j,t+1/2} + T_{i-1,j,t+1/2}) / \Delta X^2 + (T_{i,j+1,t} - 2T_{i,j,t} + T_{i,j-1,t}) / \Delta Y^2)$$
(5-14a)

$$(T_{i,j,t+1} - T_{i,j,t+1/2}) / \Delta t / 2 = \alpha ((T_{i+1,j,t+1/2} - 2T_{i,j,t+1/2} + T_{i-1,j,t+1/2}) / \Delta X^{2} + (T_{i,j+1,t+1} - 2T_{i,j,t+1} + T_{i,j-1,t+1}) / \Delta Y^{2}).$$
 (5-14b)

These equations can be simplified as follows:

Implicit X direction

$$-T_{i-1,j,t+1/2} + (2\lambda_1+1)T_{i,j,t+1/2}/\lambda_1 - T_{i+1,j,t+1/2} = \lambda_2 T_{i,j-1,t}/\lambda_1 + (1-2\lambda_2)T_{i,j,t}/\lambda_1 + \lambda_2 T_{i,j+1,t}/\lambda_1$$
(5-15a)

Implicit Y direction

$$-\mathbf{T}_{i,j-1,t+1} + (2\lambda_{2}+1) \mathbf{T}_{i,j,t+1} / \lambda_{2} - \mathbf{T}_{i,j+1,t+1} = \lambda_{1} \mathbf{T}_{i-1,j,t+1/2} / \lambda_{2} + (1-2\lambda_{1}) \mathbf{T}_{i,j,t+1/2} / \lambda_{2} + \lambda_{1} \mathbf{T}_{i+1,j,t+1/2} / \lambda_{2},$$
(5-15b)

where $\lambda_1 = \alpha \Delta t / 2\Delta X^2$, $\lambda_2 = \alpha \Delta t / 2\Delta Y^2$, $\Delta X = H/N_X$, $\Delta Y = W/N_Y$, N_X denotes the number of increments in the thickness direction, X, and N_Y the number of increments in the width direction, Y. Equation 5-15a was solved at the intermediate time values and the resulting temperature distribution was then used in Equation 5-15b to evaluate the temperature at the end of the whole time increment. The initial and boundary conditions, Equations 5-4 to 5-8, written in finite difference form are as follows:

At t=0

$$T(i,j,0)=f(i,j)$$
 (5-16)
At Y=-W/2, j=1
 $T_{i,1,t+1}=(T_{i,2,t+1}+Bi_{Y}T_{A})/(1+Bi_{Y})$ (5-17)

At
$$Y=W/2$$
, $j=N_Y+1$
 $T_{i,NY+1,t+1}=(T_{i,NY,t+1}+Bi_YT_A)/(1+Bi_Y)$ (5-18)
At $X=-H/2$, $i=1$
 $T_{1,j,t}=T_P(j)$ (5-19)
At $X=H/2$, $i=N_X+1$

 $T_{NX+1,j,t+1} = (T_{NX,j,t+1} + Bi_X T_A) / (1 + Bi_X), \qquad (5-20)$

where Bi denotes the Biot number and is defined as: $Bi_x=h\Delta X/k$ and $Bi_y=h\Delta Y/k$. A series of linear equations were written for each half time step using Equations 5-15 to 5-20 and the resulting tridiagonal matrices for the ith row and jth column were solved using a Gaussian elimination method [120].

The implicit alternating direction method has been shown to be unconditionally stable and converge for any value of lambda, λ , when $\Delta X = \Delta Y$ [120,125]. The solution technique used in this study was one that was based on equal increment sizes, $\Delta X = \Delta Y$, and $\lambda_1 = \lambda_2 = 0.5$. This value of lambda represents the upper limit for which the explicit finite difference solution scheme converges [120]. The time increment ∆t was defined as $2\Delta X^2 \lambda / \alpha$ or $2\Delta Y^2 \lambda / \alpha$. This necessitated the number of increments in the width direction, N_v , to be greater than or equal to 50,000, depending on the film thickness. As a consequence, the solution matrix consisted of greater than 250,000 elements. The CPU time required, using a VAX 780 computer, was found on average to be 3.5 hours, with an execution time of 15 hours. A large amount of time was required because the CPU had to handle large amounts of memory in order to store, retrieve and update the solution matrix, not because of the number of computations required.

An implicit method was used to solve the one dimensional formulation of the problem. Equation 5-13 reduced to the following form:

$$T_{i,t+1} - T_{i,t} = \lambda \left(T_{i+1,t+1} - 2T_{i,t+1} + T_{i-1,t+1} \right), \qquad (5-21)$$

where $\lambda = \alpha \Delta t / \Delta X^2$. The initial and boundary conditions, written in finite difference form, are as follows:

At t=0

$$T(i,0)=f(i)$$
 (5-22)
At X=-H/2, i=1
 $T_{1,t}=T_{P}$ (5-23)
At X=H/2, i=N_X+1
 $T_{NX+1,t+1}=(T_{NX,t+1}+Bi_{X}T_{A})/(1+Bi_{X})$. (5-24)

5.1.1.2. Heat Transfer Coefficient

Equations 5-5, 5-6 and 5-8 define the convective heat transfer boundary condition between the plastic film and the ambient air environment in terms of an average heat transfer coefficient and the temperature difference between the film surface and ambient environment [36]. This form is misleading, because it is a definition of an average thermal conductance rather than a law of heat transfer convection. The convective heat transfer is actually a complicated function of the plastic film velocity, the properties of the polymer, properties of the preheat roll, geometry of the preheat roll, air velocity, air properties and temperature gradients. For example, the heat transfer coefficient for free convection from a horizontal cylinder to air at atmospheric pressure [36] can be expressed as follows:

$$h=1.32((T_s-T_A)/d_c)^{1/4},$$
 (5-25)

where h denotes the heat transfer coefficient, T_s the cylinder surface temperature, T_A the ambient air temperature and d_c the cylinder diameter. This equation is valid when the product of the Grashof and Prandtl numbers ranges between 10⁴ and 10⁹, i.e. laminar flow. The heat transfer coefficient between the plastic film and preheat roll was assumed equal to infinity, since the plastic film is moving at the same linear velocity as the preheat roll and is in perfect contact with the preheat roll surface, see Equation 5-7. In this study, the value of the heat transfer coefficient at the plastic film-air interface was specified as 20.3 W/m^{2o}C. This value is an average of several values quoted in the literature [36,38,126], ranging from 13 to 54 W/m^{2o}C.

5.1.1.3. Material Properties

Equation 5-3 was derived on the assumption that the thermal properties of the material were constant. The average thermal conductivity was estimated as $0.28 \text{ W/m}^{\circ}\text{C}$ and the average thermal diffusivity as $0.000848 \text{ cm}^{2}/\text{s}$, based on the data presented in Figure 3-9.

5.1.1.4. Model Accuracy

The implicit finite difference solution scheme used to solve the two dimensional unsteady state heat conduction problem was checked using the solution presented in Reference 120 for the unsteady state heat conduction in a long bar having a square cross section. The free surfaces in this example were maintained at a constant temperature.

No differences in the program estimates were observed when the value of lambda was varied from 0.25 to 1.0.

The effect of varying the heat transfer coefficient between the maximum and minimum values quoted was found not to be significant. Using these limits, the surface temperature of the plastic film was predicted and found to differ by less than 0.75°C. The higher the value of the heat transfer coefficient, the lower the film temperature at the plastic film-air interface.

No differences in the estimates were observed when the minimum and maximum values of the thermal conductivity and thermal diffusivity were used. In addition, simulation trials were conducted using linear relationships between the above material properties and temperature. At each increment, the thermal properties were updated based on the updated temperature profile prediction. Although this approach does not relax completely the assumption of constant thermal properties, it was felt that this would improve the accuracy of the model predictions. No differences were found between the predictions using the average values or the linear equations to estimate the thermal properties.

Figures 5-2 and 5-3 compare the estimated temperature distribution, using the two dimensional model, to the measured experimental values presented in Figure 4-11. The mathematical model predictions were found to equal the experimental values when the radiation heater was turned off, as shown in Figure 5-2. The average experimental error associated with the measured temperature values was found to be $\pm 1.5^{\circ}$ C, when calculated at a confidence level of 95%. When the radiation heaters were turned on, the predicted temperature distribution was slightly lower than the experimentally measured values. This was particularly evident when the radiation heater setting was 815.6°C, see Figure 5-3.

5.1.1.5. Discussion

The temperature gradient across the film thickness was found to vary by less than 0.25°C. If experimental error is included, the temperature of the plastic film can be assumed to be uniform along the thickness direction, i.e. thin film approximation.



Figure 5-2: Preheat Roll Mathematical Model Accuracy $\underline{Z=0.24 \text{ m and } T_R=\text{off.}}$



The temperature of the plastic film at the separation point on the preheat roll, Z=0.24 m, was found to equal the temperature of the preheat roll surface, when radiation effects were neglected. The time necessary to reach a steady state temperature in all cases was found to be very short. At conditions, the the standard operating steady state temperature was attained after only 10% of the total contact time had elapsed. Figure 5-4 presents a typical case, illustrating the temperature variation as a function of time on the preheat roll. The diameter of the preheat roll required to achieve the steady state value, as illustrated in Figure The diameter of the preheat roll when based 5-4, is 3 cm. only on thermal considerations, can be much smaller than that actually used. However, such factors as the mechanical strength of the preheat roll and the dimensions of the heat transfer fluid channel dictate a minimum limit of the preheat roll diameter.

The reason that the measured temperature of the plastic film was greater than the estimated temperature when the radiation heater temperature was turned on, was because the radiation heater was in direct view of the plastic film at the separation point on the preheat roll. The effect of including the radiation component in the boundary conditions will be considered in a subsequent section.

On the basis of the above findings, it seems that the value of the heat transfer coefficient used in the simulation trials was reasonable.

The most significant process variable affecting the temperature of the plastic film at the separation point on the preheat roll was the temperature of the preheat roll surface.

The significance of including the boundary condition describing the convective heat transfer at the film edges was



Figure 5-4: Time Required to Reach Steady State, $T_{P}=90.5^{\circ}C$, Y/W/2=0 and $T_{R}=off$.

determined by calculating the proportion of film which was affected by boundary conditions 5-5 and 5-6. In all cases, the penetration length was found to be less than 0.1% of the film width. In terms of the number of increments in the width direction, this represented less than 50 out of more than 50,000 increments. Consequently, edge effects were considered negligible. This point was not surprising, since the thickness of the plastic film and the temperature gradient across the film thickness were very small.

The temperature predictions made using the two dimensional model were found to be identical to those made using the one dimensional model, when the same preheat roll and ambient air temperature distributions were used. The time required to obtain the temperature distribution using the one dimensional model was less than 5 minutes using an IBM PC XT computer. The one dimensional model was used to model the temperature distribution across and along the preheat roll in all subsequent analysis presented in this study.

5.1.2. Free Moving Film

After leaving the preheat roll, the plastic film travels to the radiation heater system. During this phase, where the plastic film is not in view of the radiation heaters, it may gain or lose heat by convection. The case where the plastic film is in view of the radiation heaters is discussed in the next section. The simplified one dimensional form of the energy equation, i.e. Equation 5-9, was solved using the following initial and boundary conditions:

At t=0

$$T(X,0) = f(X)$$
 (5-26)

At X=-H/2

$$-k\partial T(-H/2,t) / \partial X = h(T(-H/2,t) - T_A(t))$$
(5-27)

At X=H/2

 $-k\partial T(H/2,t)/\partial X=h(T(H/2,t)-T_A(t)).$ (5-28)

The initial condition, Equation 5-26, assumes that the temperature distribution after leaving the preheat roll system is known. The boundary conditions, i.e. Equations 5-27 and 5-28, describe the convective heat fluxes at the two surfaces of the plastic film.

5.1.2.1. Numerical Solution

An analytical solution exists to solve the one dimensional energy equation, Equation 5-9, using the initial and boundary conditions represented in Equations 5-26 to 5-28 [28]:

At
$$X = \underline{+}H/2$$

$$\frac{T-T_A}{T_0-T_A} = \sum_{n=1}^{\infty} \frac{2 \operatorname{Bi} \cos(\beta_n)}{(\beta_n^2 + \operatorname{Bi} + \operatorname{Bi}^2) \cos(\beta_n)} e^{-\beta_n 2\alpha t/(H/2)2}$$
(5-29a)
 $\beta_n \tan(\beta_n) = \operatorname{Bi},$ (5-29b)

where T denotes the film surface temperature, Bi the Biot number (Bi=hH/2k), T_A the ambient air temperature, T_0 the initial film temperature, α the thermal diffusivity, t the time, h the heat transfer coefficient and H the film thickness.

5.1.2.2. Discussion

The surface temperature of the film at the separation point on the preheat roll, at a preheat roll temperature setting of 93.3°C and when the radiation heaters were turned off, was found to be 90°C. At these conditions, the temperature of the film was predicted to decrease by 19°C when the film velocity was 0.21 m/s and by 12°C when the film velocity was 0.37 m/s. Measurements of the temperature indicated that the film

possible temperature remained constant. One surface explanation for this finding could be that the value of the heat transfer coefficient used was too high. Trials were conducted using different values of the heat transfer coefficient. When the heat transfer coefficient was equal to 3 W/m^{2o}C, the estimated and measured temperature values were similar, taking into account a variability of $\pm 1.5^{\circ}$ C in the The fitted value of the heat transfer measured data. coefficient is smaller that the lowest value quoted in the literature [126]. The difference may be due to the polymer and system geometry used in this study.

The results presented in the previous section are unaffected by changing the value of the heat transfer coefficient. The temperature gradient in the plastic film using the new value would be only slightly smaller than that measured using the value stated in Section 5.1.1.2..

The implicit numerical method described in the previous section was also used to solve the one dimensional unsteady state convection problem. The initial and boundary conditions were rewritten following the form presented in Equations 5-22 and 5-24. The ambient air temperature distribution was considered as being variable and, therefore, was updated during each increment. The surface temperature estimated using the implicit numerical method was similar to the surface temperature calculated using the analytical solution.

5.1.3. Radiation Heater System

After leaving the preheat roll, the plastic film travels to the radiation heater system. The radiation heater system consists of two banks of horizontal tube radiation elements measuring 0.91 m in length. The arrangements of the batch and continuous radiation heaters were similar, except that the batch system incorporated four additional tube heaters per bank. Although the radiation heaters are located some distance from the preheat roll, it can be assumed that the plastic film immediately enters the radiation zone after separating from the preheat roll since the plastic film is always in view of at least one radiation tube in this region.

In this region, energy from the radiation heaters is transmitted directly to the plastic film where it is absorbed, reflected and/or transmitted. Energy striking the film surface is absorbed and results in the generation of heat which is conducted towards the center of the film. Energy transmitted into the sheet can be absorbed as it passes through the sheet, reflected at the opposite interface where it is redirected into the film and/or transmitted through the film. A second possibility is that heat gains or losses due to convection at the film surface occur due to the air movement along the film surface. Since the transmissivity of air is approximately unity, the resulting air temperature is not greatly affected by the radiant heat flux. Instead, the air temperature distribution is affected by the high temperature of the radiation heaters, which heats the air by natural and forced convection.

Most radiation analyses assume relatively simple models for absorption of energy into the material. The most familiar approach is to assume that the material is a blackbody where all the energy is absorbed at the surface of the material. A second approach is to assume that the material is a grey body where only a fraction of the energy is absorbed by the surface of the material. The emissivity, described in Chapter 3, defines the fraction of energy absorbed relative to that of a blackbody. The latter approach was used in this study to model the radiation heater system. The one dimensional form of the simplified energy equation, Equation 5-9, was solved using the following initial and boundary conditions:

At t=0

$$T(X,0)=f(X)$$
 (5-30)
At X=-H/2
 $-k\partial T(-H/2,t)/\partial X=h(T(-H/2,t)-T_A(t)) + \epsilon F(t)_{-H/2}\sigma_{SB}(T_R^4-T^4(-H/2,t))$ (5-31)
At X=H/2
 $-k\partial T(H/2,t)/\partial X=h(T(H/2,t)-T_A(t)) + \epsilon F(t) + \epsilon F$

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 $\epsilon F(t)_{H/2} \sigma_{SB}(T_R^4 - T^4(H/2, t)), \qquad (5-32)$

where T_R denotes the radiation heater temperature setting, ϵ the emissivity, σ_{SB} the Stephan Boltzman constant and F(t) the shape factor. The initial condition, Equation 5-30, assumes that the temperature distribution after leaving the preheat roll system is known. The boundary conditions, Equations 5-31 and 5-32, describe the radiation and convective heat fluxes at the surface of the plastic film. The radiation heat flux term assumes that the emissivity is independent of wavelength and that all the radiation energy is absorbed at the film surface, i.e. -H/2 and H/2.

Alternative approaches [127-134] have been employed to solve radiation heat transfer problems. These approaches can be divided into cases where:

 The scattering and absorption characteristics of the material are taken into account. The common case is to use a two flux approach where one channel contains the diffuse flux in the forward direction and the second channel contains the diffuse flux in the reverse direction. Higher flux models can also be used which take into account collimated fluxes.
 The absorption and scattering characteristics as a function of the wavelength are taken into account.
 The radiation energy is assumed to have an appreciable volume absorptivity, resulting in a source term being included in the energy equation. An exponential form for the source is commonly used.

5.1.3.1. Numerical Solution

The boundary conditions, Equations 5-31 and 5-32, can be rewritten in a form similar to the convection boundary condition, Equations 5-27 and 5-28, using a modified heat transfer coefficient, h^* , as follows:

At X=-H/2 $-k\partial T(-H/2,t)/\partial X=h(T(-H/2,t)-T_A(t))+h_{-H/2}^{*}(T_R-T(-H/2,t))$ (5-33) At X=H/2 $-k\partial T(H/2,t)/\partial X=h(T(H/2,t)-T_A(t))+h_{H/2}^{*}(T_R-T(H/2,t)).$ (5-34)

The modified heat transfer, at each side of the film, is defined as follows:

$$h_{-H/2}^{*} = \epsilon F(t)_{-H/2} \sigma_{SB}(T_{R}^{2} + T^{2}(-H/2, t)) (T_{R} + T(-H/2, t))$$
(5-35)

$$h_{H/2}^{*} = \epsilon F(t)_{H/2} \sigma_{SB}(T_R^2 + T^2(H/2, t)) (T_R + T(H/2, t)). \qquad (5-36)$$

The analytical solution in Section 5.1.2. can be used to solve the radiation heat transfer problem. However, since the shape factor and air temperature are not constant, the travel length must be divided into increments where these variables are assumed to be constant. The film temperature is calculated at each increment and is then used as the initial temperature for the next increment.

The solution scheme used to solve the one dimensional unsteady state radiation problem (Equations 5-9, 5-30, 5-33 and 5-34) involved using the implicit numerical method described in the previous section. The radiation boundary conditions written in finite difference form are as follows:

At X=-H/2, i=1

$$T_{1,t+1}=(T_{2,t+1}+Bi^{1}T_{A}+Bi_{-H/2}^{2}T_{R})/(1+Bi)$$
 (5-37)
At X=H/2, i=N_x+1

 $T_{NX+1,t+1} = (T_{NX,t+1} + Bi^{1}T_{A} + Bi^{2}_{H/2}T_{R}) / (1+Bi), \qquad (5-38)$

where $Bi=Bi^2+Bi^1$, $Bi^1=h\Delta x/k$ and $Bi^2=h^*\Delta x/k$. Again, the same solution scheme described in the previous section was used to solve the set of equations describing the radiation heater system. The modified heat transfer coefficient, ambient air temperature and shape factor were updated following each increment.

5.1.3.2. Material Properties

The one dimensional form of the energy equation and boundary conditions were derived assuming constant material properties. The average values stated in the previous sections and chapters were used to predict the temperature distribution. The average emissivity of LDPE-1, as determined in Chapter 3 for a film thickness of 0.05 mm, was found to be 0.20. This thickness represents the case when two film sheets are embossed simultaneously, which is the configuration used in this study.

5.1.3.3. Shape Factor

The exposure regions for the continuous and batch radiation heater units were calculated based on the geometry of the radiation heater units. For both systems, the radiation exposure zone extended from the preheat roll to the embossing zone nip region. The amount of radiation incident on an element travelling from the preheat roll to the embossing zone was calculated using the following equation [135]:

$$F_{A1-A2} = (\tan^{-1}(Z/X) - \tan^{-1}(Y/X)) / (Z-Y), \qquad (5-39)$$

where X=c/d, Y=a/d and Z=b/d. This equation describes the shape factor from a sheet A_1 , having a length "b-a" and located "c" units from cylinder A_2 having a diameter "d", to a cylinder A_2 , Figure 5-5. The shape factor for a cylinder emitting to a sheet, i.e. A_2 to A_1 , was calculated using the following relation:

$$F_{A2-A1} = ((b-a)W_FF_{A1-A2})/\pi dW_R,$$
 (5-40)

where W_F denotes the film width and W_R the radiation tube length. For a rectangular film element, at a given machine direction position Z, a total shape factor was calculated based on the summation of the shape factors from each of the radiation heater tubes. Figure 5-6 illustrates the shape factor distribution obtained for the batch and continuous embossing systems.

5.1.3.4. Air Temperature Distribution

A good deal has been written about the heat transfer characteristics of cylinders in natural convection. The majority of these investigations, however, have been restricted to the study of single cylinder systems. When a horizontal cylinder is subjected to sudden constant heating [136], the heat transfer to the surrounding medium is initially by conduction. The hypothesis is that the conduction regime breaks down and global convective motion begins when the conduction field penetrates a certain critical depth. The fluid motion begins as the diffusion of heat from the heated cylinder establishes variations in the density field. Finally, the convection currents become established and dominate the heat transfer. The heat transfer characteristics exhibited by vertical arrays of heated horizontal cylinders cannot be predicted by simple superposition of single cylinder behaviour [137]. Instead, they are dependent upon the array spacing and Rayleigh number. Other factors which can affect the accuracy



Figure 5-5: Shape Factor Geometric Configuration, Equation 5-39.



Figure 5-6: <u>Shape Factor Distribution.</u>

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of the theoretical predictions are radiation heat transfer effects, draft or currents due to extraneous sources, a stratified environment, three dimensional effects and conductive heat transfer.

A rigorous analysis of the convective air motion between the radiation heater tube banks poses a formidable problem. A few, simple approaches were employed to predict the air temperature distribution in the vicinity of the radiation heaters. Thermal radiation effects were ignored, since, generally radiation passes through air without significant absorption taking place. The simplest approach used was to consider that the air surrounding the heaters was motionless and heat was conducted from the cylindrical heating surface to the air. Another approach considered natural convection on a vertical flat plate as the mode of heat transfer. In all cases, the temperature profiles obtained were significantly different from those measured experimentally.

For the purposes of this preliminary study, the ambient air temperature distribution used was based on the experimental data presented in Figures 3-15 and 3-16. The air temperature was updated following each time, as was done with the modified heat transfer coefficient and shape factor.

5.1.3.5. Model Accuracy

Figure 5-7 illustrates the accuracy of the one dimensional radiation heat transfer model for various initial film temperature values and radiation heater settings. The estimated temperature predictions were found to be slightly greater than the measured values. The maximum estimated film temperature was calculated to be 118°C, which is 15°C greater than the maximum temperature measured. The largest difference between the estimated and measured values was for the case



Figure 5-7: Radiation Model Accuracy, $\epsilon=0.2$ and Z=0.65 m.

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when the radiation heater temperature setting was 815.6°C and the initial film temperature was 91.4°C.

Trials were conducted in which the emissivity was varied from 0.0 to 0.2. The best fit, i.e. minimum least squares, between the estimated and measured values was obtained when the emissivity was equal to 0.1, as shown in Figure 5-8. The largest difference between the estimated and measured values, 8.1°C, was for the case when the radiation heater temperature setting was 537.8°C and the initial film temperature was 66°C.

The accuracy of the mathematical model in modelling the batch embossing system could not be determined since no experimental data were available.

5.1.3.6. Discussion

The temperature distribution across the plastic film thickness was found to be uniform after exiting the radiation heater zone regardless of the initial temperature profile. In addition, since the air temperature distribution was assumed to be uniform along the radiation heater length, the film temperature was found to be uniform across its width.

The differences observed between the measured and predicted temperature values may be due either to the use of an inaccurate air temperature distribution in the program or to the assumption of using an average emissivity. If the first explanation is correct, it could imply that the air temperature distribution assumed was too high when the radiation heater temperature setting was equal to 537.8°C.

Figure 5-9 shows the variation of the film surface temperature as a function of travel distance from the preheat roll. Separate effects due to convection and radiation are also shown in the figure. The radiation component is important in







Figure 5-9a: <u>Film Surface Temperature</u>, T_{R} =815.6°C.

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that it accounts for most of the temperature variation along the machine direction. The maximum variation due to convection i.e. no radiation, was 5.6°C for the case when the radiation heater temperature was 815.6°C and the initial film temperature was 91.4°C. As the radiation heater temperature decreases, the radiation effects become less significant.

Observations made during the experimental trials confirmed that the batch system temperatures were higher. The reason for this was that the batch radiation arrangement incorporated eight additional tube heaters. The temperature of the plastic film leaving the radiation unit, however, was still below the melt temperature of the polymer. The difference between the estimated maximum temperatures in the batch and continuous systems, when using the standard conditions, was 11°C. Since no experimental data were available, the emissivity and air temperature distribution could not be adjusted to provide accurate predictions.

5.1.4. Embossing Roll System

Following the thermal treatment, the plastic film travels to the embossing zone where it passes between the embossing and rubber covered backup roll system. The plastic film in this region, is first cooled by contact with the embossing and rubber backup rolls. Further, after passing through the nip region formed by the contact of these two rolls, the plastic film remains in contact with the embossing roll. The simplified energy equation was solved using the following initial and boundary conditions to describe the one dimensional representation of the first stage of the cooling problem:

At t=0 T(X,0)=f(X)

(5-41)

At $X=-H/2$	
$T(-H/2,t)=T_1$	(5-42)
At X=H/2	
$T(H/2,t) = T_2,$	(5-43)

where T_1 and T_2 denote the surface temperature of the embossing and backup rolls, respectively. The initial condition, Equation 5-41, assumes that the temperature distribution after leaving the radiation system is known. The boundary conditions, Equations 5-42 and 5-43, suggest that there is perfect contact between the surface of process rolls and the plastic film.

The solution to the second part of the cooling stage is identical to that used to solve the equations for the preheat roll system. The one dimensional unsteady state heat conduction problem was solved using the implicit numerical method described in the previous section.

The surface temperature of the plastic film at the separation point on the embossing roll was found to equal the temperature of the embossing roll. In fact, the temperature of the plastic film was found to equal the roll surface temperature, i.e. 27-32°C, after exiting the nip region formed by the contact of the embossing and backup rolls.

5.2. STRESS ANALYSIS

Unlike many other polymer processes, the film embossing process involves the deformation of a soft, solid plastic film and not that of a melt. As such, many of the techniques developed for the analysis of polymer processes (e.g. injection molding, film blowing, blow molding) do not apply.

The embossing of a soft sheet represents a special type of contact problem because it involves not only the contact

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between two rolls, but also the indentation of a sheet by a rigid body, i.e. embossment. Compared to the rolling process, the film embossing process does not result in a reduction of the plastic film thickness and the two rolls are not separated by a fixed distance.

The plastic film is compressed as it enters the nip region and is subject to plastic deformation resulting in the transfer of the pattern on the embossing roll to the plastic film. On leaving the nip region, some elastic recovery occurs. The dimensions of the nip region are determined by the forces applied to the engraved embossing roll in order to press it into the rubber covered backup roll. The extent of plastic deformation is determined by the geometry of the embossments and the forces applied to press the two rolls together.

The analysis of the embossing process was divided into two parts. The first section deals with the analysis of two cylindrical rolls in contact, with one roll having a rubber cover. Mathematical models were developed to estimate the pressure distribution in the nip region, dimensions of the nip region and the stress distribution in the backup roll. The second part deals with the problem of indentation, in order to determine the stress distribution in the plastic film and backup roll during embossment. Mathematical models were developed to estimate the pressure distribution under the punch and to determine the load required to achieve a specific penetration depth.

Existing models described in the literature [138-168] were used to solve the problems under consideration, as well as to gain a general understanding of the phenomena involved. Details regarding the derivation of the various equations can be found in the specific references cited. The following assumptions were made to simplify the system under analysis: 1. The plastic film is a solid as it enters the embossing system nip region.

2. The linear velocities of the plastic film, embossing roll and backup roll are equal, i.e. no tangential tractions.

3. The dimensions of the contact zone and the stress distribution in the nip region are unaffected by the presence of the plastic film. The stress distribution in the plastic film is assumed to equal the stress distribution at the embossing roll and rubber backup roll interface.

4. As demonstrated in the previous section, the plastic film is cooled in the nip region. The change in temperature as the plastic film passes through the nip region formed by the contact of the embossing and backup rolls is approximately 70°C. The dimensions of the nip region and pressure distribution in the nip, however, can be assumed to be independent of temperature since the rubber covered roll and embossing backup roll are maintained at constant temperature, а i.e. isothermal analysis. The temperature gradients in the plastic film influence the extent to which the pattern is transferred to the plastic film (will be discussed in the next chapter).

5. Constant and isotropic material properties.

6. The load applied to press the rolls together is uniform along the roll axis.

7. Static analysis can be used to analyze the rolling problem.

5.2.1. Contact Problem

The distribution of load over the contact area and the stresses produced when two bodies are pressed together is of special importance. In many practical applications, it is sometimes not possible to avoid highly concentrated loads, for example: knife edge contact (point force and a half plane), roller bearings in contact (spheres in contact), wheel contact (cylinder and a half plane) and cylinders in contact.

The analysis of the contact problem, as it relates to the film embossing process, was divided into two parts:

<u>Hertz Problem Analysis:</u> Contact between two homogeneous rolls, one of which is rigid. <u>Multi-Layer Roll Problem:</u> Contact between two rolls, one of which has an elastic cover.

The theory presented in this section assumes that the bodies in contact are smooth. The effect of the embossments is not considered to alter the predictions because the height of the embossments is orders of magnitude smaller than the roll radii.

5.2.1.1. Hertz Analysis

The Hertz theory [138-143] is concerned with the behaviour of two isotropic and linearly elastic bodies whose curvatures near the point of contact may be represented by parabolas when pressed together. The major assumptions used to derive the equations are as follows:

 The dimensions of the contact patch are small compared with the radii of curvature of the bodies, i.e. the contact zone is essentially plane.

2. Tangential tractions arising from differences in the elastic properties and system geometries are small.

3. The elastic limit is not exceeded in the deformed bodies.

4. The solids are isotropic.
Hertz completely solved the problem in terms of a Newtonian potential function. As a result, there is always an elliptical contact zone and the pressure distribution is given by the ordinates of a semi-ellipsoid centered over the contact region. A special case of the Hertz problem occurs when one of the contact dimensions is very much greater than the other, resulting in a two dimensional geometry, e.g. cylinders in contact. For parallel cylinders having radii, R_1 and R_2 , the ellipse of contact degenerates into parallel lines. The contact area becomes a rectangle having an area 2BW, where the nip dimensions are [139]:

$$2B = (16N'R_0/\pi E_0)^{0.5}, \qquad (5-44)$$

where 2B denotes the dimension of the contact zone, R_i the radius of cylinder i, N' the applied force, $1/R_0=1/R_1+1/R_2$, $1/E_0=(1-v_1^2)/E_1+(1-v_2^2)/E_2$, E_i the Young's modulus of body i and v_i the Poisson ratio of body i. The normal pressure distribution at the interface between the two rolls is:

$$\sigma_{\rm x} = \sigma_{\rm z} = P_0 \left(1 - {\rm Z}^2 / {\rm B}^2 \right)^{0.5}$$
(5-45)

$$\sigma_{\rm Y}=2\upsilon\sigma_{\rm X},\qquad(5-46)$$

where $\sigma_{\rm X}$ denotes the normal stress in the film thickness direction, $\sigma_{\rm Z}$ the normal stress in the machine direction, $\sigma_{\rm Y}$ the normal stress in the film transverse direction, Z the machine direction position co-ordinate and P₀=2N'/ π B. The shear stresses are equal to zero at the interface. Outside the contact zone, all the stresses vanish to zero. The stress distribution in the deformed solid can be calculated using the equations derived by Belayef [140], who derived equations relating the stresses on planes perpendicular to the coordinate axes in elliptical coordinates.

5.2.1.2. Multi-Layered Roll Analysis

The treatment of the contact problem, when the soft roll consists of a thin elastic cover on a hard supporting core, was first given by Hannah [144]. It was shown that both the dimensions of the nip and the form of the pressure distribution depended upon the cover thickness. Equations were derived assuming the condition of plane stress, with given displacement conditions on the inner boundary and subject to pressure by a body of given shape on its free face. The rubber layer was also assumed to be sufficiently thin so that the inner boundary would affect the stresses in the contact zone. The pressure distribution in the nip region was specified as follows:

$$P(Z) = w((1-Z^2/B^2)^{0.5} + \sum_{n=1}^{\infty} \cos((2N-1)Z\pi/2B)). \quad (5-47)$$

The values of w, B and a_N are determined using an iterative process.

Hannah's results illustrated that next to the diameter of the two rolls and their elastic modulus, the layer thickness is the most important factor determining the relation between loading and deformation. A greater load is required to produce the same contact length as that achieved when the two rolls are homogeneous. The pressure distribution over the contact zone is only slightly affected by the layer thickness.

Parish [145-147] extended the approach developed by Hannah [144] for the case of plane strain and made measurements of the pressure distribution in the nip region formed by the contact of a metal roll and a roll incorporating a thin rubber cover at various load conditions. Parish's results demonstrated that the pressure curves were very closely symmetrical and that their peaks laid along the center line of the rolls. A change in roller speed was found not to have a significant effect on the shape or magnitude of the pressure curves. Furthermore, the experimental results were in qualitative agreement with the trends predicted by Hannah [144]. Quantitatively however, the changes produced were significantly greater than those predicted. The author reasoned that nonlinearity in the elastic properties of the rubber was the most probable cause of the discrepancies observed. Nonlinearity in the elastic properties was said to be especially significant under dynamic conditions such as those in rolling contact systems.

Several other approaches have been developed to describe the deformation in multi-layered roll systems [148-151]. In general, these approaches involved specifying stress functions and using numerical techniques to determine the coefficients used to define the stress functions.

5.2.1.3. Material Properties

The average values presented in Table 3-1 were used to predict the stress distribution. The elastic modulus and poisson ratio of the rubber layer were estimated as 1.8 MPa and 0.49, respectively. The comparable values for the metal roll were 655 MPa and 0.29, respectively. These values were used, since it was not possible to measure the properties of the rubber on the backup roll without damaging the surface of the backup roll.

5.2.1.4. Discussion

The accuracy of the various model predictions could not be evaluated since it was not possible to obtain any experimental data relating the applied load to the nip dimensions and pressure distribution in the nip. The dimensions of the nip region, 2B, and the maximum pressure in the nip, P_{Max} were calculated using the Hertz theory (Equations 5-44 to 5-46) at the two embossing pressures used in this study:

<u>Pressure, MPa</u>	<u>2B, cm</u>	P _{Max} , MPa	<u>Load, kN</u>
0.1	0.72	0.10	0.61
0.2	1.02	0.14	1.22

The pressure distribution is parabolic according to Equation 5-45, as shown in Figure 5-10. The stress distribution in the deformed solid can be calculated using the equations derived by Belayef [140]. Figure 5-11 presents the stress distribution at a distance X=0.786B from the contact surface in the deformed rubber roll. The normal stresses in the thickness, σ_x , and roll axial, σ_y , directions exhibit maxima in the plane of symmetry along the cylinder center lines, Z/B=0. Both of these stresses decrease asymptotically to zero as one moves from the nip center to the edges, i.e toward $Z=+\infty$. The normal stress in the machine direction, σ_7 , has a saddle form, symmetrical about the center line of the two cylinders, when X>0.5B. In this case, during a loading cycle, the stress component in these layers passes twice through the maximum values. Finally, the shear stress distribution reaches a maximum, equalling 0.3P0 at a distance X=0.786B from the contact surface.

The dimensions of the contact zone, predicted using the equations developed by Hannah [140] when the rubber layer thickness was 0.0125 m, were found to approximately equal the predictions made using the Hertz theory for a metal roll in contact with a solid rubber roll. The pressure distribution in the nip region was the same as that predicted by the Hertz theory. The thickness of the rubber layer on the backup roll is so large that the Hertz theory, i.e. homogeneous roll analysis, can be used to approximate the dimensions of the contact zone and stress distribution in the nip region.



Figure 5-10: Stress Distribution, Nip Region.

 \Box



Figure 5-11: Stress Distribution, X=0.78B.

The effect of the plastic film in the nip region is considered to be negligible on the dimensions of the contact zone or pressure distribution in the nip, since the film thickness is several orders of magnitude smaller than the roll radii and the rubber layer thickness. Furthermore, the modulus of LDPE-1 and of the rubber used to cover the backup roll are both significantly smaller than the modulus of metal. As the plastic film enters the nip region, the temperature of the film is approximately 100°C and the elastic modulus of LDPE-1 at this temperature is similar to that of Neoprene rubber. As the film is cooled in the nip region, the modulus of LDPE-1 increases, until at a temperature of 25°C, it is greater than that of Neoprene rubber, but still much smaller than that of metal.

The Hertz theory of contact assumes that the surfaces are frictionless and do not transmit any tangential surface tractions across the interface. Contact stresses in tractive rolling have been considered by various authors [152-157]. When the elastic properties of the two bodies are the same, the shape of the contact zone remains symmetrical and unaffected by the tangential force. It has been recognized that a difference in the elastic constants of two contacting bodies leads to tangential tractions at the interface and the stress distribution may be asymmetrical. The magnitude of the shear tractions is generally an order of magnitude smaller than the normal pressure, so that, as long as the coefficient of friction at the interface is low, the assumption of no slip is justified. The tangential force is typically specified as a fraction of the normal force and this fraction is commonly defined as the coefficient of static friction. Parish [147] has shown that tangential forces for multi-layered roll systems cause the metal roller to have a higher apparent peripheral speed, which is attributed to extension of the rubber surface in the region of the nip. The extension of the rubber surface is due in part to the contact pressure as well as to the presence of shear strains that occur due to the transmission of torque through the nip. The effect of tangential tractions in the context of this study was assumed to be small, because the force applied to the rolls in order to press them together was small. The embossing process is only intended to impress the pattern into the soft film and not to decrease the film thickness as in rolling operations. The film velocities before and after the embossing rolls have been found to be equal and the linear speeds of the backup and embossing rolls have also been found to be equal, thus supporting the assumption of no tangential tractions.

The properties of the rubber layer can vary over time due to the rolling process and temperature variations in the radiation heaters. These effects can harden the rubber layer over time, resulting in an increase in the modulus and a decrease in the nip dimensions. According to Parish [147] these effects probably have the greatest impact on the accuracy of the estimates made. As mentioned above, in this study it was not possible to determine the true elastic modulus of the rubber layer; therefore literature values were used.

5.2.2. Indentation Problem

In 1885, Boussinesq [158] was the first to obtain an analysis of the state of stress in a homogeneous, isotropic elastic half space when its surface is deformed by a smooth rigid body, when he studied the problem of a pillar resting on a foundation. Using the method of potential theory, Boussinesq derived a solution for the axisymmetric problem in which the axis of the indenting solid of revolution was normal to the undeformed boundary of the elastic half space. When friction was ignored, the pressure distribution under the indentor, represented in cylindrical co-ordinates, was determined using the following equation:

$$P(r) = N' (a^2 - r^2)^{-0.5} / 2\pi a_c, \qquad (5-48)$$

where a_c denotes the cylinder radius, r the radius coordinate, P the pressure and N' the applied force. At the edges of the punch (r=a_c) the pressure is infinite, resulting in plastic deformation of the elastic half space.

In the present work, the analysis of the indentation problem as it relates to the film embossing process was divided into two parts: the analysis of single-layered systems and the analysis of multi-layered systems.

5.2.2.1. Single Layer Analysis

The simplest case is that where a flat cylindrical punch is pressed into a semi-infinite elastic half space. The following assumptions are usually made to reduce the complexity of the problem:

(a) linearly elastic material,

(b) material properties are isotropic,

(c) the elastic limit of the material is not exceeded,

(d) axial symmetry and

(e) no friction between the indentor and the elastic medium.

The resulting equations of elastic equilibrium [160], based on the above assumptions and expressed in cylindrical polar co-ordinates, are as follows:

$$\partial \sigma_r / \partial r + \partial \tau_{rz} / \partial z + (\sigma_r - \sigma_\theta) / r = 0$$
 (5-49a)

 $\partial \tau_{zr} / \partial r + \partial \sigma_z / \partial z + \tau_{zr} / r = 0,$ (5-49b)

where σ denotes the normal stress, τ the shear stress and r,z, θ the cylindrical co-ordinates. Equations 5-49a and 5-49b are solved using the following boundary conditions:

 $\sigma_{-}=0$ r>a_c

$$U_z = \delta \qquad 0 < r < a_c \qquad (5-50)$$

$$\tau_{zr}=0$$
 r>0, (5-52)

where U denotes the displacement, δ denotes the penetration depth and a_c the indentor radius. Boundary condition Equation 5-50 assumes that the surface displacement is specified over the contact region, Equation 5-51 assumes that the normal stress vanishes to zero outside the contact zone and Equation 5-52 that the tangential stresses are equal to zero on the boundary. The depth of penetration, according to Equation 5-50, is used to determine the resulting stress, strain and displacement fields in the elastic medium.

The above system of equations have been solved by Sneddon [159-162] in terms of Hankel transforms of an arbitrary function. He determined the arbitrary function by using a solution due to Titchmarsh [159,161] of the dual integral equations to which the mixed boundary value problem was reduced. An analytical solution was derived for this system. The equation estimating the total load required to produce a penetration [162] is as follows:

$$P=4\mu a_{c}\delta/(1-v), \qquad (5-53)$$

where δ denotes the penetration depth, P the load required to achieve this penetration, a_c the indentor radius, v the Poisson ratio of the elastic layer, E the Young's modulus of the elastic layer and $\mu = E/2(1+v)$. The penetration depth is a linear function of the applied load and decreases as the radius of the punch is increased.

(5-51)

5.2.2.2. Multi-Layered Analysis

Dhaliwal and Rau [163-165] obtained a solution to the axisymmetric Boussinesq problem for an elastic layer lying over an elastic foundation and under a rigid punch of an arbitrary profile. They employed a technique that reduced the mixed boundary value problem into a Fredholm integral equation of the second kind. The resulting stress and displacement relations for a homogeneous isotropic elastic medium, expressed in cylindrical co-ordinates, were given as:

$$\sigma_{zz}(\mathbf{r}, \mathbf{z}) = (\lambda + 2\mu) \partial \mathbf{U}_{z} / \partial \mathbf{z} + \lambda (\partial \mathbf{U}_{r} / \partial \mathbf{r} + \mathbf{U}_{r} / \mathbf{r})$$
(5-54)

$$\sigma_{rz}(r,z) = \mu \left(\frac{\partial U_r}{\partial z} + \frac{\partial U_z}{\partial r} \right)$$
 (5-55)

$$2\mu U_{r}(r,z) = \partial X/\partial r + z \partial \Omega/\partial r \qquad (5-56)$$

$$2\mu U_{z}(\mathbf{r}, z) = \partial X / \partial z + z \partial \Omega / \partial Z - (3 - 4v) \Omega, \qquad (5 - 57)$$

where E denotes the Young's modulus of the elastic medium, vthe Poisson ratio of the elastic medium, U the displacement, σ the normal stress, τ the shear stress, $\lambda = vE/(1+v)(1-2v)$, $\mu = E/2(1+v)$ and X(r,z) and $\Omega(r,z)$ harmonic functions represented as:

$$X(r,z) = H_{o}[A(\xi)e^{-\xi z} + B(\xi)e^{\xi z}; \xi - r]$$
(5-58)

$$\Omega(\mathbf{r},\mathbf{z}) = \mathrm{H}_{\mathrm{o}}[C(\xi) e^{-\xi \mathbf{z}} + D(\xi) e^{\xi \mathbf{z}}; \xi - \mathbf{z}]. \qquad (5-59)$$

A, B, C and D are arbitrary functions and H_o denotes the Hankel transform of order zero of the function $f(\xi,z)$ with respect to the variable ξ . The stress and displacement fields in the two layers were determined by solving the system of equations using the following boundary conditions:

$U_{Z}^{1}(r,-H) = \delta - f(r/a_{c})$	0 <r<a<sub>c</r<a<sub>	(5-60)
$\sigma_{zz}^{1}(r,-H)=0$	r>0	(5-61)
$\sigma_{\rm zr}^{1}(r,-H)=0$	r>0	(5-62)
$U_{z}^{i}(r,0) = U_{r}^{i}(r,0) = 0$	r>0 and i=1,2	(5-63)
$\sigma_{zz}^{i}(r,0) = \sigma_{zr}^{i}(r,0) = 0$	r>0 and i=1,2,	(5-64)

where δ denotes the penetration depth. The superscripts 1 and 2 refer to the top and foundation layers, respectively. Equation 5-60 represents the boundary condition relating the displacement of the elastic layer to the geometry of the punch. For the case of a cylindrical punch, the displacement of the elastic layer is equal to the penetration depth for the area under the punch. Equations 5-61 and 5-62 describe the condition where the stresses are equal to zero outside the contact zone. Equations 5-63 and 5-64 represent boundary conditions satisfying the continuity conditions between the elastic layer and the foundation, where perfect contact is assumed at the boundary. The values of the arbitrary constants were determined by Rau and Dhaliwal [163-165] using additional variables that satisfy the Fredholm integral equations.

The computing time needed to determine the load required to achieve a given penetration was between 1 and 2 CPU hours using a VAX 780 computer. The solution scheme involved numerical integration and series representations to solve the system of differential equations.

5.2.2.3. Discussion

Using Equation 5-53 derived by Sneddon [159-162], the calculated load required to achieve a penetration of 0.122 mm into the rubber layer is 0.06 N. This penetration level corresponds to complete penetration of the embossments described in this study. The diameter of the cylinder was specified to be equal the width of the top of the square embossment, i.e. 0.216 mm. The load required to achieve complete penetration is significantly less than the force used to press the two rolls together at the two pressure settings used in this study, even though the top of the embossments comprise 14.5% of the total available area in the nip region.

The load required to press a paraboloid of revolution into a semi-infinite elastic layer for complete penetration [162] was calculated as being 0.09 N. The diameter of the base of the paraboloid of revolution was specified to be equal to the width of the base of the embossment.

The load required to press a cylindrical punch into a semiinfinite solid having the same properties as LDPE-1, in order to achieve complete penetration as a function of the modulus, was as follows:

<u>Modulus, MPa</u>	<u>Temperature, °C</u>	<u>Load, N</u>
9	100	0.26
69	25	2.16

The load required to achieve complete penetration is again achieved at both pressure settings used in this study.

The normal stress distribution at the boundary between the elastic layer and punch for both punch geometries is presented in Figure 5-12. A high stress concentration occurs near the edges of the punch. Inside the deformed layer, at large distances from the punch, the isochromatic lines of stress approximate circles. Near the punch, these contour lines approximate ellipses touching the boundary of the deformed medium.

Using the methodology of Dhaliwal and Rau [163-165] the load required to achieve a complete penetration of an embossment, i.e. 0.122 mm, for the cylindrical punch into a rubber layer covering a metal foundation was found to be equal to that calculated when using the equations developed by Sneddon [159-162]. The effect of the metal foundation was found not to be significant, when the rubber layer thickness was equal to 1.27 cm.





The analysis of a case where two soft materials are in contact, with the top layer being very thin, was found to yield results similar to those predicted when only a single layer was considered. The magnitude of the load required to achieve complete penetration was of the same order of magnitude as that predicted using the equations derived by Sneddon [159-162] and was significantly smaller than the force used to press the two rolls together.

Unlike the solution scheme developed to analyze cases where the thickness of the elastic layer was significantly greater than that of the punch, the solution to the Fredholm equation was not straight forward when the elastic layer thickness was small. In this case, a series solution could not be obtained for the Fredholm equation. Due to the complexity of the solution technique required to solve the Fredholm equation [166,167], the values determined by Dhaliwal and Rau [164] were used to estimate the load required to achieve complete penetration.

No equations were found in the literature for the contact of a truncated pyramid and an elastic half space. However, based on the above simulation trials, it is felt that complete penetration occurs, i.e. full contact, at both pressure settings used in this study. The flat cylinder geometry represents the most severe case and requires the highest loads. This is due to the large flat surface of contact and sharp edges. The paraboloid of revolution requires a lower load due to the smooth contour. The embossments on the embossing roll have smoothed edges and only a small flat area of contact at the pyramid top and therefore should require a load between these two limits.

The results presented in this section illustrate that the rubber covered backup roll can be treated as a homogeneous

roll when determining the contact zone dimensions and pressure distribution. Furthermore, the presence of the plastic film in the nip region does not seem to have a significant affect on the dimensions of the contact zone or the pressure distribution, because of its small thickness and low modulus.

5.3. SUMMARY

Mathematical models were developed in this chapter to estimate the temperature and stress distributions in the plastic film as functions of the processing conditions.

The temperature of the plastic film was found to be a function of the preheat roll and radiation heater temperature setting. Due to the high film velocity, convection effects were found not to be significant.

The thickness of the rubber layer on the backup roll is so large as to render the effect of the metal core insignificant. The force required to impress an embossment in the plastic film and rubber backup layer is much smaller than the force necessary to press the two rolls together.

Finally, the following general comments can be made regarding the models presented in this chapter:

(a) The mathematical model predictions are strongly dependent on the material properties.

(b) The models presented are based on static analysis.

(c) The equations are based on the theory of elasticity and plastic films are viscoelastic materials.

(d) No friction is assumed.

(e) The interaction between multiple punches is ignored.

CHAPTER 6 DISCUSSION

The primary purpose of this work was to present an engineering analysis of the film embossing process. In Chapter 4, experimental results illustrating the inter-relationships between the plastic film temperature, plastic film thickness, film velocity, film blowup ratio, preheat roll temperature setting, radiation heater temperature setting, embossed film physical dimensions, flat film physical dimensions, embossed film mechanical properties, flat film mechanical properties and embossed film morphology and flat film morphology were presented. In Chapter 5, mathematical models were developed to estimate the temperature and stress distributions as functions of the process variables.

The focus of this chapter will be to couple the information presented in Chapters 4 and 5 and to present a model which could account for the observed interactions when embossing a thin plastic film.

6.1. HEAT TRANSFER ANALYSIS

The experimental results presented in Chapter 4 suggested that the thermal treatment conditions were the controlling factor in determining the final properties and dimensions of the plastic film. Furthermore, the heat treatment conditions had a significant effect in determining the extent to which the pattern on the embossing roll was transferred to the plastic film.

The heat transfer models presented in Chapter 5 were used to estimate the film surface temperature as a function of the plastic film thickness, plastic film velocity, preheat roll temperature setting and radiation heater temperature setting. The calculated effects of the film velocity, film thickness, preheat roll temperature setting and radiation heater temperature setting on the plastic film surface temperature before entering the embossing zone nip region are illustrated in Figures 6-1 to 6-3. These measurements were estimated using the mathematical models developed for the batch embossing system. Although the temperature predictions are too high, they still illustrate the trends observed experimentally. The effects of manipulating these process variables can be summarized as follows:

 (a) At higher film velocities, the residence time of the plastic film in the heating zone is lower.
 This, in turn, results in a decrease in the film surface temperature. This effect is shown in Figure 6-1.

(b) When the process conditions remain the same, thicker plastic films require greater energy to reach a set temperature. As a consequence, the film temperature for the thicker film is lower than that of the thin film. This effect is shown in Figure 6-2.

(c) Higher preheat roll temperature settings increase the temperature of the plastic film entering the radiation heater system and embossing zone. The film temperature at the separation point on the preheat roll is equal to the preheat roll surface. These effects are shown in Figures 6-1 to 6-3.

(d) At higher radiation heater temperature settings,the film surface temperature is higher, see Figures6-1 to 6-3.

Heat transfer models were used to replot the embossed film characterization data presented in Figures 4-13a to 4-13f as a function of the estimated film surface temperature. Figures 6-4a to 6-4d illustrate the effects of the preheat roll



Figure 6-1: Effect of Film Velocity, H=0.050 mm.



Figure 6-2: Effect of Film Thickness, V=24.5 cm/s.



Figure 6-3: Effect of Preheat Roll Temperature, V=24.5 cm/s and H=0.050 mm.



Figure 6-4a: Effect of Preheat Roll Temperature, BUR=4:1, V=21.6 m/min, P=0.2 MPa and H=0.029 mm.













temperature and the estimated film surface temperature. The data collapse to a single curve for the two preheat roll temperatures. Furthermore, when all the lateral shrinkage, gloss and tensile modulus results were plotted as functions of the estimated film surface temperature, the data also fall on a single curve, as shown in Figure 6-5. As the film surface temperature increases, the lateral shrinkage increases, the gloss decreases and the ratio of the tensile modulus in the machine and transverse direction, approaches unity, indicating random orientation.

The thermal treatment conditions were found to be the most significant factor in determining the quality of the embossed film. Data for the two force levels applied to press the embossing rolls together, see Figure 6-6, illustrated that the pattern quality, as defined using the gloss and bulk thickness, was controlled by the thermal treatment conditions. The embossing force was not the controlling factor in determining the pattern quality. The mechanism involved in transferring the pattern from the embossing roll to the plastic film is discussed further in the next section.

6.2. STRESS ANALYSIS

The temperature of the plastic film as it enters the embossing zone nip region determines it's yield characteristics, while the force used to press the two rolls together determines the force applied to transfer the embossing pattern onto the plastic film. The extent to which the embossing pattern is transferred to the plastic film can be determined using a failure theory which relates the stress distribution in the nip region, as was estimated in Chapter 5, and the yield characteristics of the plastic, as measured experimentally in Chapter 4.



Figure 6-5: Relationship Between the Film Surface Temperature and Film Properties.



Figure 6-6: Effect of Embossing Pressure.

It is of practical importance in the embossing analysis to determine whether the rubber roll layer or plastic film yields under the loading. Various failure theories exist and are described by Urgural and Fenster [168]. In general, these theories focus primarily on comparing the principal stresses to a critical limit, in order to determine whether yielding has occurred.

The maximum distortion energy theory, proposed by M.T. Huber [168] and further developed by R. Von Mises and H. Hencky [168], states that failure by yielding occurs when, at any point in the body, the distortion energy per unit volume in a state of combined stress becomes equal to that associated with yielding in a simple tension test. The Mises-Hencky yield criterion is described by the following equation:

$$\tau_{\rm oct} = 1/3 \left(\left(\sigma_1 - \sigma_2 \right)^2 + \left(\sigma_2 - \sigma_3 \right)^2 + \left(\sigma_3 - \sigma_1 \right) \right)^{0.5} = 0.47 \sigma_{\rm yp}, \qquad (6-1)$$

where τ_{oct} denotes the octahedral stress, σ_1 , σ_2 and σ_3 the principal stresses and σ_{yp} the critical stress. The critical stress, which is equal to the yield stress for LDPE-1 when measured in tension at room temperature, was found to be 8.94 MPa. The critical stress measured in compression was found to range from 6.8 MPa, when measured at room temperature, to 3.1 MPa, when measured at 75°C.

The plastic film and rubber layer on the backup roll both undergo different loading cycles. The plastic film undergoes a single load cycle, while the rubber layer undergoes a cyclical loading each time it passes through the nip region. The distribution of the octahedral stress in the nip region, calculated using the Hertz theory, is presented in Figure 6-7. Regardless of the force applied to press the two rolls together, the maximum stress value obtained at the nip centerline is below the critical yield value for LDPE-1. The octahedral stress distribution in the contact area under a





cylindrical punch is below the critical yield value for LDPE-1, except at the edges of the punch. At the edge of the cylindrical punch, where the stresses approach infinity, the yield characteristics of LDPE-1 are exceeded and plastic deformation occurs. The deformation that occurs, is localized to the area near the edge of the punch, while the area directly under the punch is not deformed. This accounts for the reason that no changes in the film thickness are observed during embossing.

The extent to which the embossing pattern is transferred to the plastic film is a function of the film temperature, which in turn determines the yield limit, and is a function of the stress distribution in the nip region which is controlled by the force applied to press the two rolls together. Based on the characterization data presented in Chapter 4, the true critical yield value of LDPE-1 must be significantly less than the stress distribution at the embossing pressures and temperatures used in this study. It must be remembered that as the plastic film passes through the nip region it is cooled, and, as a consequence, the yield values of LDPE-1 increase. The stress distribution estimated in Chapter 5 is too low to correctly predict the observed trends.

6.3. FILM EMBOSSING PROCESS DESCRIPTION

The process of embossing a plastic film and the corresponding changes that occur during embossing can be divided into two steps. The first step involves the thermal treatment effects, while the second step involves the phenomena occurring in the embossing zone nip region.

The temperature of the plastic film increases as it passes over the preheat roll and between the radiation heaters. The physical dimensions, mechanical properties and morphology of the plastic film change in the same manner as if the film were allowed to anneal at a temperature close to the melt temperature. During such a process, the plastic film shrinks in the unrestrained directions and the orientation shifts toward the random state. Furthermore, as the temperature increases, the modulus and yield strength decrease, thus making it easier to transfer the pattern from the embossing roll onto the plastic film.

The force applied to press the embossing roll onto the backup roll determines the dimensions of the nip region and stress distribution in the nip region between the embossing and rubber covered backup rolls. As the plastic film passes through the nip region, it is simultaneously cooled and stressed. When the pressure at any point in the nip region exceeds the yield point of the polymer, plastic deformation of the material and permanent deformation of the film occur. If the stress is below the yield point of the plastic film, elastic recovery will occur and the film will maintain its original state. If the yield stress is not exceeded everywhere in the material, then the pattern will contain regions of permanently deformed and undeformed material. The pattern definition reflects the balance between the amount of material that is frozen (deformed) into the embossment shape and the amount of material that remains undeformed.

The observations made in this study relating the effect of the various process variables on the final properties of the embossed film are summarized below:

(a) Mechanical Properties: The thermal treatment conditions determine the temperature of the plastic film. If the plastic film temperature is high, the film undergoes annealing and the properties change toward the random state. As the temperature of the plastic film increases from room temperature toward the softening temperature of the resin, the properties shift away from the initial values determined during the film blowing process.

(b) Physical Dimensions: If the plastic film temperature is high, the degree of lateral shrinkage is also high. Thus, the film width decreases and the film thickness increases.

(c) Morphology: If the plastic film temperature is too high, the film undergoes annealing and the orientation shifts toward the random state.

(d) Pattern Quality: For a given nip stress profile, a high plastic film temperature causes a greater proportion of material to exceed the critical yield point of the film. This produces a higher pattern quality as compared to a film processed at a lower temperature. At low embossing pressures, the stress distribution in the nip region is small. In addition, the proportion of material exceeding the critical yield criterion is low, and, as a result, the pattern definition is poor. The softer is the rubber on the backup roll, the easier it will be for the engraved pattern to be impressed into the rubber, enlarging the contact area between the two rolls. This results in a film having both sides embossed with the pattern image. If the roll is too hard, only the top surface of the embossed film would contain the pattern and the bottom surface would be smooth. If the rubber layer on the backup roll is too soft, there is also a chance that permanent damage can be done to it if the yield properties of the rubber are surpassed. Finally, if the temperature of the engraved embossing roll increases, the temperature of the film leaving the roll is higher and the possibility that the film may relax or anneal is greater, once it is wound onto rolls for storage, thus resulting in a decrease in the bulk thickness.

CHAPTER 7

CONCLUSIONS, ORIGINAL CONTRIBUTIONS AND FUTURE WORK

7.1. GENERAL CONCLUSIONS

The analysis of the film blowing process requires characterization of the flat film structure and properties, in addition to the rheological properties of the resin and the thermo-mechanical history experienced by the melt during the process.

Tubular blown films produced using LDPE-1 were found to exhibit an intermediate stress type orientation, with the *a*axis of the polyethylene unit cell inclined at an angle ranging between 30 and 80 degrees from the plane of the film (MD-TD). The amorphous phase orientation was found to be in the transverse direction in the plane of the film.

As the blowup ratio increased, the angle of inclination of the a-axis of the polyethylene unit cell away from the plane of the film (MD-TD) was found to increase.

The birefringence, surface gloss, sonic velocity and tensile properties correlate with the angle of inclination of the *a*axis in relation to the plane of the film (MD-TD). The morphology and orientation in polyethylene films are closely related to the elongational viscosity of the resins via the stress crystallization model of Keller and Machin [75].

The thermal treatment phase of the embossing process was the most significant variable in determining the structure, dimensions and properties of the embossed film. The thermal treatment was shown to be a function of the preheat roll temperature, radiation heater temperature, film velocity and film thickness. At radiation heater temperatures below 500°C, the preheat roll temperature was the significant factor in determining the film temperature. At radiation temperatures greater than 500°C, the radiation heater temperature was the significant factor. The film thickness, initial film orientation and film velocity had a minor effect on the final properties of the embossed film. The force applied to press the embossing and backup rolls together had a small influence on the pattern quality. Finally, the rolling process did not affect the structure or orientation of the films, since it did not result in the reduction of the film thickness.

The temperature of the plastic film was found to increase as the preheat roll and radiation heater temperatures increased. The temperature of the plastic film leaving the preheat roll was equal to the surface temperature of the preheat roll. The diameter of the preheat roll required to heat the plastic film to the temperature of the preheat roll surface was found to be only 3 cm. The maximum air temperature measured in front of the radiation heaters was 207°C. The air temperature was found to vary in front of the radiation heaters, with the maximum temperature measured along the centerline of the heaters. The air temperature distribution had a significant effect at low preheat roll temperatures.

At higher plastic film temperatures, both the degree of lateral shrinkage and the tendency for the orientation of the crystalline and amorphous phases to become random are greater. Also at higher plastic film temperatures, it is easier to impress the pattern from the embossing roll onto the plastic film. Surface gloss of the embossed films correlates well with the pattern quality and texture.

Mathematical models were developed to predict the temperature, stress and deformation distributions in the plastic film during embossing. The thermal treatment zone was divided into the preheat roll system, the radiation heater system and the embossing system for a free moving film. The embossing phase
of the process was divided into two parts: the contact between two cylinders and the indentation of a punch into an elastic half space.

The analyses indicated that the accuracy of one dimensional heat transfer models was comparable to that obtained with two dimensional models. Further, the mathematical model predictions were in general agreement with the experimental data. The analysis suggested that temperature variations across the film width and thickness are very small. Also, natural convection effects has a negligible affect on the temperature of the plastic film. The temperature of the film as it exits the nip region, equals the temperature of the embossing roll.

The dimension of the nip region formed by the contact of the engraved embossing roll and rubber covered backup roll and the stress distribution in the nip region depend on the properties of the materials used to construct the two rolls, the thickness of the rubber layer on the backup roll and the applied force. The load required to impress an embossment into the rubber covered backup roll is significantly less than that applied to press the embossing roll and backup roll together. The shape of the embossment is the most significant factor in determining the pressure distribution under the embossment. Smoother contours produce more uniform stress distributions.

The maximum distortion energy failure theory [168] could be used to describe embossing. The force applied to press the embossing roll into the backup roll determines the dimensions of the nip region and stress distribution in the nip region between the embossing and rubber covered backup rolls. The thermal treatment determines the yield characteristics of the plastic film. As the film passes through the nip region, it is simultaneously cooled and stressed. When the pressure at any point in the nip region exceeds the yield point of the polymer, then plastic deformation of the material occurs and permanent deformation of the film occurs. If the stress is less than the yield point of the plastic film, elastic recovery will occur and the film will maintain its original state. If the yield pressure is exceeded only in parts of the material, then the pattern will contain regions of permanently deformed and undeformed material. The pattern definition is based on the amount of material that is frozen (deformed) into the embossment shape and the amount of material that remains undeformed.

7.2. ORIGINAL CONTRIBUTIONS

In general, the present work represents a significant contribution to the understanding of the film embossing process. It has employed a combination of detailed characterization of thermoplastic tubular blown films and mathematical models for the understanding and simulation of the temperature distributions and deformations applied to the plastic film during processing. The specific original contributions made in this work may be summarized as follows:

1. This study represents the first comprehensive analysis of the tubular blown film embossing process. The effect of varying the process variables was illustrated by measuring the structure, dimensions and properties of the embossed film product. Furthermore, measurements were taken of the plastic film temperature, air temperature, film velocity and roll pattern for a cold embossing system.

2. The stress crystallization model of Keller and Machin [75] was used to predict the orientation in tubular blown films. Differences in the orientation 202

of tubular blown LDPE-1, LDPE-2, LLDPE and HDPE films were correctly predicted.

3. Experimental techniques were employed and developed to characterize the microstructure, physical dimensions, optical properties and mechanical properties of embossed films. This included using surface gloss as a measure of pattern quality and texture.

4. Heat transfer models were developed to estimate the temperature distribution of the plastic film as it passes through a continuous and batch cold embossing system. Mathematical models were also developed to estimate the dimensions of the nip region and stress distribution in the nip region for an engraved roll embossing system.

7.3. RECOMMENDATIONS FOR FUTURE WORK

The following recommendations are proposed, as a result of this study:

1. Experimental studies should be carried out to measure the air temperature distribution in front of the radiation heaters.

2. Techniques should be developed to determine the microstructure and physical dimensions of embossed films for very small areas.

3. Mathematical models are needed to more accurately predict the stress distribution in the nip for an engraved metal roll embossing system. Instead of decoupling the analysis of the deformations, finite element techniques should be used to model the system.

4. A similar type of analysis may be performed to describe the cast embossing system.

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REFERENCES

- 1. B.S. Cayne, editor, The New Lexicon Webster's Dictionary of the English Language, Lexicon Publications, Inc., New York (1987).
- D.F. Eary and E.A. Reed, Techniques of Pressworking Sheet Metal, 2nd edition, Prentice-Hall, Inc., New Jersey (1974).
- 3. C.J. Benning, *Plastics Films for Packaging*, Technomic Publishing Co., Inc., Pennsylvania (1983).
- 4. S.H. Pinner and W.G. Simpson, *Plastics: Surface and Finish*, Chapter 2, Buttersworths, London (1971).
- 5. J.M. Adams, R.H. Hopper, R.R. Pease and G.E. Raley, Method for Embossing Plastic Material, U.S. Patent No. 3950480 (April 13, 1976).
- 6. R.H. Hugger, Embossing Method and Apparatus, U.S. Patent No. 2928124 (March 15, 1960).
- 7. B. Bowman and J. Cleminson, #1 Embossing Line Operator Training Manual, Internal Report, Technology Group, Union Carbide, Quebec (March 18, 1975).
- 8. R.A. Offermann, Embossed Polyethylene Film Process Selection and Description for Drydees Disposable Diapers, Internal Report, Research and Development Department, Union Carbide Corporation, Illinois.
- 9. E. Mittman, Embossed Material and Method and Apparatus of Producing Same, U.S. Patent No. 3176058 (March 30, 1965).
- 10. F.H. Bussey, Jr. and J.K. Rutherfoord, Apparatus for Embossing Film, U.S. Patent No. 3966383 (June 29, 1976).
- F.H. Bussey, Jr. and J.K. Rutherfoord, Apparatus for Embossed Film Manufacture, U.S. Patent No. 3957414 (May 18, 1976).
- R. Gersbeck and M. Krohn, Apparatus, Including an Endless Belt, for Continuously Hot Pressing a Web of Material, U.S. Patent No. 4269585 (May 26, 1981).
- 13. J.H. Briston, *Plastics Films*, 2nd edition, George Godwin, New York (1983).

- R. Keller, Sixth International Conference on Polyolefins Technical Papers, Texas, page 261 (February 1989).
- 15. S.C. Frowde, Method for Producing Embossed Thermoplastic Sheet, Canadian Patent No. 847768 (July 28, 1970).
- J.M. Adams, R.H. Hopper, J.K Rutherfoord and G.E. Raley, Apparatus for Embossing Plastic Material, U.S. Patent No. 3849050 (November 19, 1974).
- 17. T. Kloender, Apparatus for Hot Embossing Continuous Webs, U.S. Patent No. 3246365 (April 19, 1966).
- 18. F. Bustin, Method of Embossing Thin, Limp Plastic Film and Disposable and Embossed Plastic Bag Product, Canadian Patent No. 897921 (April 18, 1972).
- 19. F.R. Cramton, Crimped Binding Strap and Method of Making Same, U.S. Patent No. 3283378, November 8 (1966).
- 20. J.K. Rutherfoord, Embossed Thermoplastic Material, Canadian Patent No. 1052968.
- 21. P. Hamel, Plast. Eng., p. 34 (June 1980).
- 22. M. Akay, Polymer, 22:5 (January 1981).
- 23. E.H. Roberts, P.J. Lucchesi and S.J. Kurtz, Adv. Poly. Tech., 6(1):65 (1986).
- 24. D. Summa, J. Stein, M.K. Tse and J. Melngailis, S.P.E. Tech. Pap., 32:749 (1986).
- 25. M.R. Kamal, A. Haber and M.E. Ryan, Poly. Eng. Sci., 25(11):698 (1985).
- 26. G.E.P. Box, W.G. Hunter and J.S. Hunter, *Statistics* for Experimenters, John Wiley & Sons, New York (1978).
- 27. R.B. Bird, W.E. Stewart and E.N. Lightfoot, *Transport Phenomena*, John Wiley & Sons, Inc., New York (1976).
- Z. Tadmor and C.G. Gogos, Principles of Polymer Processing, John Wiley & Sons, New York (1979).
- 29. O.J. Sweetings, editor, The Science and Technology of Polymer Films, Vol. 1, Interscience Publishers, New York (1968).

- 30. W.R.R. Park, *Plastics Film Technology*, Robert E. Krieger Publishing Company, New York (1973).
- 31. ASTM D 1248-84, Annual Book of ASTM Standards, 8.01:584 (1986).
- 32. S. Kavesh and J.M. Schultz, J. Poly. Sci.: Pt A-2, 9:85 (1971).
- 33. S. Kavesh and J.M. Schultz, J. Poly. Sci.: Pt. A-2, 8:243 (1970).
- 34. R. Day, Internal Memo, Tioxide Group PLC, England, (April 19, 1989).
- 35. F. Grum and R.J. Becherer, Optical Radiation Measurements, Vol. 1, Academic Press, New York (1979).
- 36. J.P. Holman, *Heat Transfer*, 4th edition, McGraw-Hill Book Company, New York (1976).
- 37. W.O. Predöhl, Kunst., 61:489 (July 1971).
- 38. G. Menges and W.O. Predöhl, Poly. Eng. Sci., **15**(5):394 (1975).
- 39. C.J.S. Petrie, Plast. & Polym., p. 259 (December 1974).
- 40. Plastic Film Measurement, Technical Note: TN100, IRCON, Inc., Illinois (1979).
- 41. N. Toutoungi, A Study of Thermal Conductivity and Diffusivity of Polymers, M. Eng. Thesis, McGill University, Department of Chemical Engineering, (May 1983).
- 42. C.A. Harper, editor, Handbook of Plastics and Elastomers, McGraw-Hill Book Company, New York (1975).
- 43. G.S. Whitby, C.C. Davis and R.F. Dunbrook, editors, Synthetic Rubber, John Wiley & Sons, Inc., New York (1954).
- 44. ASTM D 1415-88, Annual Book of ASTM Standards, 9.01:223 (1990).
- 45. R.D. Steihler, G.E. Decker and G.W. Bullman, Rubb. Chem. Tech., 52:255 (1979).
- 46. H.E. Boyer and T.L. Gall, editors, *Metals Handbook*, American Society for Metals, Ohio (1985).

- 47. D.R. Holmes, R.G. Miller, R.P. Palmer and C.W. Bunn, *Nature*, **171**(4364):1104 (1953).
- 48. A. Keller, J. Poly. Sci., 11(6):567 (1953).
- 49. A. Keller, Nature, 174(4437):926 (1954).
- 50. A. Keller, J. Poly. Sci., 15:31 (1955).
- 51. A. Keller and I. Sandeman, J. Poly. Sci., 15:133 (1955).
- 52. D.R. Holmes and R.P. Palmer, J. Poly. Sci., 31:345 (1958).
- 53. S.L. Aggarwal, G.P. Tilley and O.J Sweeting, *J. Appl. Poly. Sci.*, **1**(1):91 (1959).
- 54. R.S. Stein and G.B.B.M. Sutherland, J. Chem. Phys., 21(2):370 (1953).
- 55. R.S. Stein and G.B.B.M. Sutherland, J. Chem. Phys., 22(12):1993 (1954).
- 56. M.C. Tobin and M.J. Carrano, J. Poly. Sci., 24:93 (1957).
- 57. P.H. Lindenmeyer and S. Lustig, J. Appl. Poly. Sci., 9:227 (1965).
- 58. ASTM E 81-89, Annual Book of ASTM Standards, 3.01:238 (1990).
- 59. M. Kakudo and N. Kasai, X-Ray Diffraction by Polymers, Elsevier Publishing Company, New York (1972).
- 60. L.E. Alexander, X-Ray Diffraction Methods in Polymer Science, Robert E. Krieger Publishing Company, New York (1979).
- 61. J.L. White and J.E. Spruiell, Poly. Eng. Sci., 21(13):859 (1981).
- 62. J.L. White and J.E. Spruiell, Poly. Eng. Sci., 23(5):247 (1983).
- 63. R.J. Samuels, Structured Polymer Properties, John Wiley & Sons, New York (1973).
- 64. J.V. Dawkins, editor, Developments in Polymer Characterization, Vol. 4, Ch. 6, Applied Science Publishers, New York (1988).

- 65. B. Ke, editor, Newer Methods of Polymer Characterization, Vol. 6, Ch. 4, Interscience Publishers, New York (1964).
- 66. R.S. Stein and F.H. Norris, J. Poly. Sci., 21:381 (1956).
- 67. R.S. Stein, J. Poly. Sci., 31:327 (1958).
- 68. R.S. Stein, J. Poly. Sci., 31:335 (1958).
- 69. R.S. Stein, J. Poly. Sci., 34:709 (1959).
- 70. R.S. Stein, J. Poly. Sci., 50:339 (1961).
- 71. C.R. Desper, A Study of Crystallization and Orientation Mechanisms in Polyethylene, Ph.d. Thesis, University of Massachusetts, Department of Chemistry, (August 1966).
- 72. C.R. Desper, J. Appl. Poly. Sci., 13:169 (1969).
- 73. C.R. Desper and R.S. Stein, J. Appl. Phys., **37**(11):3990 (1966).
- 74. J.T. Judge and R.S. Stein, J. Appl. Phys., **32**(11):2357 (1961).
- 75. A. Keller and M.J. Machin, J. Macromol. Sci.; Phys., B1(1):41 (1967).
- 76. W.F. Maddams and J.E. Preedy, J. Appl. Poly. Sci., 22:2721 (1978).
- 77. W.F. Maddams and J.E. Preedy, J. Appl. Poly. Sci., 22:2739 (1978).
- 78. W.F. Maddams and J.E. Preedy, J. Appl. Poly. Sci., 22:2751 (1978).
- 79. M.A. McRae and W.F. Maddams, J. Appl. Poly. Sci., 22:2761 (1978).
- 80. K.J. Choi, J.E. Spruiell and J.L. White, J. Poly. Sci.: Poly. Phys., 20:27 (1982).
- 81. T.H. Kwack and C.D. Han, J. Appl. Poly. Sci., 35:363 (1988).
- 82. C.D. Han, T.H. Kwack and M.E. Vickers, J. Appl. Poly. Sci., 28:3399 (1983).

- C.D. Han, Y.J. Kim, H.K. Chuang and T.H. Kwack, J. Appl. Poly. Sci., 28:3435 (1983).
- 84. T.H. Kwack and C.D. Han, J. Appl. Poly. Sci., 28:3419 (1983).
- 85. C.D. Han and J.Y. Park, J. Appl. Poly. Sci., 19:3257 (1975).
- 86. C.D. Han and J.Y. Park, J. Appl. Poly. Sci., 19:3277 (1975).
- 87. C.D. Han and J.Y. Park, J. Appl. Pol. Sci., 19:3291 (1975).
- 88. H.H. Winter, Pure & Appl. Chem., 55(6):943 (1983).
- 89. J. Meissner, Pure & Appl. Chem., 42:552 (1975).
- 90. W. Minoshima and J.L. White, J. Non-Newt. Fl. Mech., 19:251 (1986).
- 91. W. Minoshima and J.L. White, J. Non-Newt. Fl. Mech., 19:275 (1986).
- 92. L.A. Utracki and J. Lara, Melt Rheology of PE; Part I Correlation Between Extensional and Shear Flows, Paper Presented at the International Workshop on Extensional Flows, Mulhouse-La Bresse, France (January 1983).
- 93. L.A. Utracki, A. Catani, J. Lara and M.R. Kamal, *Processability and Flow of Polyethylenes*, Paper presented at the European meeting on Polymer Processing and Properties, Capri, Italy (June 1983).
- 94. R. Farber and J. Dealy, Poly. Eng. Sci., 14(6):435 (1974).
- 95. T. Kanai and J.L. White, Poly. Eng. Sci., 24(15):1185 (1984).
- 96. C.J. Petrie, Poly. Eng. Sci., 15(10):708 (1975).
- 97. R.K. Gupta, A.B. Metzner and K.F. Wissbrun, *Poly. Eng.* Sci., **22**(3):172 (1982).
- 98. L.A. Utracki and R. Gendron, Pressure Oscillation During Extrusion of Polyethylenes, Part II, Internal Report, Industrial Material Research Institute, National Research Council of Canada, Quebec (1984).

- 99. A. Haber and M.R. Kamal, S.P.E. Tech. Pap., 33:446 (1987).
- 100. A. Haber and M.R. Kamal, Plast. Eng., p. 43 (October 1987).
- 101. L.A. Simpson, *Measuring Gloss*, Ti-INFO System, Section 3.3.3., Tioxide Group PLC, England (1989).
- 102. H. Ashizawa, J.E. Spruiell and J.L. White, Poly. Eng. Sci., 24(13):1035 (1984).
- 103. N.C. Huck and P.L. Clegg, S.P.E. Trans., 1:121 (1961).
- 104. D. Hansen, W.F. Kracke and J.R. Falender, J. Macromol. Sci.; Phy., B4(3):583 (1970).
- 105. A. Peterlin, Poly. Eng. Sci., 18(6):488 (1978).
- 106. I.L. Hay and A. Keller, J. Matl. Sci., 1:41 (1966).
- 107. I.L. Hay and A. Keller, J. Matl. Sci., 2:538 (1967).
- 108. D.P. Pope and A. Keller, J. Poly. Sci.: Poly. Phys., 13:533 (1975).
- 109. D.P. Pope and A. Keller, J. Matl. Sci., 6:453 (1971).
- 110. Z.W. Wilchinsky, J. Appl. Poly. Sci., 7:923 (1963).
- 111. D.M. Brigg, E.G. Smith, M.M. Epstein and R.J. Fiorentino, S.P.E. Tech. Pap., 28:74 (1982).
- 112. S. Bahadur and A. Henkin, Poly. Eng. Sci., 13(6):422
 (1973).
- 113. A. Kaito, K. Nakayama and H. Kanetsuna, J. Appl. Poly. Sci., 28:1207 (1983).
- 114. D. Lewis, E.J. Wheeler, W.F. Maddams and J.E. Preedy, J. Poly. Sci., 10:369 (1972).
- 115. S.J. Krause and W.F. Hosford, J. Poly. Sci.: Pt. B Phys., 27:1867 (1989).
- 116. A. Kaito, K. Nakayama and H. Kanetsuna, J. Macromol. Sci.; Phys., **B26**(3):281 (1987).
- 117. A.D. Nercessian, Procedure for Operating 40" Embosser, Internal Report, Technology Group, Union Carbide, Quebec (August 24, 1983).

- 118. R.S. Hunter, The Measurement of Appearance, A Wiley-Interscience Publication, New York (1975).
- 119. M.R. Kamal and G.L. Bata, *Current Trends in the Analysis of Polymer Shaping Operations*, Paper Presented at the European Meeting on Polymer Processing and Properties, Capri, Italy (June 1983).
- 120. B. Carnahan, H.A. Luther and J.O. Wilkes, Applied Numerical Methods, John Wiley & Sons, New York (1969).
- 121. H.S. Carslaw and J.C. Jaeger, Conduction of Heat in Solids, 2nd edition, Oxford University Press, New York (1959).
- 122. G. Birkhoff and R.S. Varga, Amer. Math. Soc. Trans., 92:13 (1959).
- 123. J. Douglas, Jr., J. Soc. Indust. Appl. Math., 3(1):42 (1955).
- 124. D.W. Peaceman and H.H. Rachford, Jr., J. Soc. Indust. Appl. Math., 3(1):28 (1955).
- 125. J. Douglas, Jr., J. Soc. Indust. Appl. Math., 4(1):20
 (1956)
- 126. Transient Heating of a Film in a Combined Mode, Internal Report, Research and Development Department, Union Carbide Corporation, Illinois (1971).
- 127. H.A. Lunka, S.P.E. Journal, 26:48 (January 1970).
- 128. R.C. Progelhof, J. Franey and T.W. Haas, J. Appl. Poly. Sci., 15:1803 (1971).
- 129. R.C. Progelhof and J.L. Throne, Poly. Eng. Sci., 14(11):760 (1974).
- 130. R. Gardon, J. Amer. Cer. Soc., 41(6):200 (1958).
- 131. R.C. Progelhof, J. Quintiere and J.L. Throne, J. Appl. Poly. Sci.: Pt. 2, 17:1227 (1973).
- 132. M. Perez and A.F. Balso, J. Franklin Inst., 285(6):424 (1968).
- 133. R.C. Progelhof and J.L. Throne, Appl. Optics, 9(10):2359 (1970).

- 134. P.S. Mudgett and L.W. Richards, Appl. Optics, 10(7):1485 (1971).
- 135. E.M. Sparrow and R.D. Cess, *Radiation Heat Transfer*, Brooks/Cole Publishing Company, California (1966).
- 136. J.R. Parsons, Jr. and J.C. Mulligan, J. Heat Transfer, 102:636 (November 1980).
- 137. G.F. Marsters, Intl. J. Heat Mass Transfer, 15:921 (1972).
- 138. E.I. Radzimovsky, Univ. of Ill. Eng. Exptl. Station Bulletin, Series No. 408, Urbana (1953).
- 139. W. Flugge, editor, Handbook of Engineering Mechanics, Chapter 42, McGraw-Hill Book Company, Inc., New York (1962).
- 140. S. Way, J. Appl. Mech., 57:A-49 (1935).
- 141. H.R. Thomas and V.A. Hoersch, Univ. of Ill. Eng. Exptl. Station Bulletin, Series No. 212, Urbana (1930).
- 142. A.E.H. Love, A Treatise on the Mathematical Theory of Elasticity, 4th edition, Dover Publications, New York (1944).
- 143. A. Sackfield and D.A. Hills, J. Strain Anal., 18(2):101 (1983).
- 144. M. Hannah, Qtly. J. Mech. Appl. Math., 4:94 (1951).
- 145. G.J. Parish, Brit. J. Appl. Phy., 6:256 (July 1955).
- 146. G.J. Parish, Brit. J. Appl. Phy., 9:158 (April 1958).
- 147. G.J. Parish, Brit. J. Appl. Phy., 9:428 (November 1958).
- 148. T.C. Soong and C. Li, Intl. J. Mech. Sci., 23:263 (1981).
- 149. T.C. Soong and C. Li, J. Appl. Mech., 47:841 (December 1980).
- 150. T.C. Soong and C. Li, J. Appl. Mech., **48**:889 (December 1981).
- 151. H.T. Hahn and M. Levinson, Intl. J. Mech. Sci., 16:489 (1974).

- 152. R.H. Bentall and K.L. Johnson, Intl. J. Mech. Sci., 9:389 (1967).
- 153. K.L. Johnson, Proc. Royal Soc. Lond., A230:531 (1955).
- 154. A. Sackfield and D.A. Hills, J. Strain Anal., 18(2):107 (1983).
- 155. E. M'Ewen, Phil. Mag., 40:454 (1949).
- 156. J.O. Smith and C.K. Liu, J. Appl. Mech., 75:157 (1953).
- 157. H. Poritsky, J. Appl. Mech., 72:191 (1950).
- 158. J. Boussinesq, Applications des Potentiels a L'Etude de L'Equilibre et du Mouvement des Soildes, Paris (1885).
- 159. I.N. Sneddon, Proc. Camb. Phil. Soc., 42:29 (1946).
- 160. J.W. Harding and I.N. Sneddon, Proc. Camb. Phil. Soc., 41:16 (1945).
- 161. I.N. Sneddon, Proc. Camb. Phil. Soc., 44:492 (1948).
- 162. I.N. Sneddon, Intl. J. Eng. Sci., 3:47 (1965).
- 163. R.S. Dhaliwal, Intl. J. Eng. Sci., 8:273 (1970).
- 164. R.S. Dhaliwal and I.S. Rau, Intl. J. Eng. Sci., 8:843 (1970).
- 165. I.S. Rau and R.S. Dhaliwal, Intl. J. Eng. Sci., 10:659 (1970).
- 166. C.T.H. Baker, L. Fox, D. Mayers and K. Wright, Comput. J., 7:141 (1964).
- 167. L. Fox and E.T. Goodwin, Phil. Trans. Roy. Soc., A245:501 (1953).
- 168. A.C. Ugural and S.K. Fenster, Advanced Strength and Applied Elasticity, Elsevier North Holland, New York (1981).
- 169. Temperature Measurement Handbook, Omega Engineering, Inc., Connecticut (1982).
- 170. Operating Instructions and Maintenance Manual for the Pyro Optical Pyrometer, The Pyrometer Instrument Co., Inc., New Jersey (1976).

- 171. ASTM D 1928-80, Annual Book of ASTM Standards, 8.02:169 (1990).
- 172. ASTM E 252-84, Annual Book of ASTM Standards, 8.03:763 (1990).
- 173. Large Polarization Microscope "Zetopan-Pol" Instruction Manual, Reichert, Austria (1972).
- 174. F.H. Moy, Microstructure and the Distribution of Tensile Properties in Injection Molded Polyethylene, Ph.d. Thesis, McGill University, Department of Chemical Engineering (February 1980).
- 175. R. Zbinden, Infrared Spectroscopy of High Polymers, Academic Press, New York (1964).
- 176. The IR8 Infrared Spectrophotometer, Beckman Instructions 1213-D, Beckman Instruments, Inc., California (1967).
- 177. ASTM E 168-88, Annual Book of ASTM Standards, 14.01:9 (1990).
- 178. W.W. Moseley, Jr., J. Appl. Poly. Sci., 3(9):266 (1960).
- 179. Rapid, Accurate Measurement of Elastic Modulus in Fibers and Filaments, Films and Papers, H.M. Morgan Co., Inc., Massachutetts (1977).
- 180. A. Ehringhaus, Rotary Compensator with Quartz Combination Plates Instructions.
- 181. ASTM D 1505-85, Annual Book of ASTM Standards, 8.01:458 (1990).
- 182. J.M. Dealy, Rheometers for Molten Plastics, Van Nostrand Reinhold Company, New York (1982).
- 183. T. Samurkas, A Rheological Study of Three Linear Low Density Polyethylene Film Resins, M. Eng. Thesis, McGill University, Department of Chemical Engineering, (September 1985).
- 184. V. Shah, Handbook of Plastics Testing Technology, A Wiley-Interscience Publication, New York (1984).
- 185. ASTM D 882-83, Annual Book of ASTM Standards, 8.01:317 (1990).
- 186. ASTM D 695-89, Annual Book of ASTM Standards, 8.01:197 (1990).

- 187. High Speed Impact Tester: A Preliminary Manual, Rheometrics, Inc., New Jersey (1978).
- 188. E.F.H. Chu, Crystallization Kinetics and Morphological Studies of High-Density Polyethylene, M. Eng. Thesis, McGill University, Department of Chemical Engineering, (March 1983).
- 189. Operating Manual: Differential Scanning Calorimeter, Perkin Elmer Corporation, Connecticut.
- 190. J.L. McNaughton and C.T. Mortimer, Differential Scanning Calorimetry, Perkin Elmer Corporation, Connecticut (1975).
- 191. J.P. Cali, Standard Reference Material 720: Synthetic Sapphire, National Bureau of Standards, Washington D.C. (1970).
- 192. ASTM D 3418-82, Annual Book of ASTM Standards, 8.03:85 (1990).
- 193. ASTM D 3417-83, Annual Book of ASTM Standards, 8.03:81 (1990).
- 194. Instruction Manual HunterLab Glossmeter Model D48D, Hunter Associates Labortory, Inc., Virginia (1989).
- 195. ASTM D 2805-88, Annual Book of ASTM Standards, 6.01:409 (1990).
- 196. Instruction Manual HunterLab Colorimeter Model D25A-9, Hunter Associates Labortory Inc., Virginia (1989).

Appendix A EXPERIMENTAL PROCEDURES

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A.1. PROCESS MEASUREMENTS

A.1.1. Temperature

The temperature distribution of the preheat roll, radiation heaters, embossing roll, tubular blown films, embossed films and ambient air were measured using contact and optical thermometry.

A.1.1.1. Preheat Roll

The surface temperature of the preheat roll was measured using an Omega Engineering Surface Temperature Probe, Model 68203, with a chromel-constantan thermocouple (Type E) and an Omega Engineering Model MCJ cold junction compensator. The thermocouple millivolt output was recorded using a Hewlett Packard two channel strip chart recorder, Model 17503A. The calibration data for the thermocouple are available elsewhere [169].

The following step by step procedure was used to measure the surface temperature of the preheat roll:

1. The temperature setting for the preheat roll was set to the desired set point temperature and the preheat roll was allowed to reach steady state, (minimum of one hour).

2. The preheat roll was rotated for at least five minutes prior to taking any measurements.

3. The thermocouple surface probe was pressed against the surface of the preheat roll at the desired position, and the millivolt output from the thermocouple recorded for a period of at least 30 seconds.

A.1.1.2. Radiation Heater

A Pyro Optical Pyrometer, Model 85-C, was used to measure the temperature of the radiation heater tubes. The calibration and operation of this instrument are described in detail elsewhere [170].

The following step by step procedure was used to measure the temperature of the radiation heater tubes:

1. The pyrometer was calibrated following the recommended procedures [170].

2. The pyrometer was focused on the area to be measured.

3. The small illuminated test spot in the field of vision was made to disappear by varying the brightness of the target and rotating the pyrometer's knurled wedge ring.

4. The temperature, as indicated on the direct reading scale on the pyrometer, was recorded.

A.1.1.3. Embossing Roll

An Omega Engineering Surface Temperature Probe, Model 68221, with а chromel-constantan thermocouple and Omega an Engineering Model MCJ cold junction compensator were used to measure the surface temperature distribution of the embossing roll. The thermocouple millivolt output was recorded using a Hewlett Packard two channel strip chart recorder, Model 17503A. The procedure used was the same as that used to measure the surface temperature of the preheat roll. The temperature setting for the embossing roll was controlled by the cooling water flow valve, which was set completely open for all the tests performed in this study.

A.1.1.4. Backup Roll

The procedure and equipment used to measure the surface temperature of the backup roll, was the same as that used to measure the surface temperature of the preheat roll.

A.1.1.5. Plastic Film Surface

The surface temperature of the tubular blown and embossed films, as they were being produced using the continuous embossing system, were measured using Omega Engineering Surface Temperature Probe, Model 68221, with a chromelconstantan thermocouple and an Omega Engineering Model MCJ cold junction compensator. The thermocouple millivolt output was recorded using a Hewlett Packard two channel strip chart recorder, Model 17503A.

The following step by step procedure was used to measure the surface temperature of the plastic films:

1. The tubular blown film process was allowed to stabilize for at least five minutes, for a given combination of processing conditions.

2. The thermocouple surface probe was pressed against the surface of the plastic film and the millivolt output from the thermocouple was recorded for a period of at least thirty seconds. Care was taken not to tear the film web, while at the same time ensuring that the temperature probe surface was flush against the surface of the moving film.

A.1.1.6. Air Temperature

The air temperature distribution, except in the vicinity of the radiation heaters, was measured using a mercury thermometer. The air temperature distribution in front of the radiation heaters was measured using a shielded thermocouple, chromel-constantan. A ceramic sleeve and a piece of aluminum foil were used to shield the thermocouple from the radiating heat source. The thermocouple probe was positioned in front of the radiation heaters and the millivolt output from the thermocouple was recorded for a period of at least thirty seconds.

A.1.2. Velocity

The linear velocity of the preheat, embossing and backup rolls was measured using a Power Instrument digital tachometer, Model TAK-ETTE. The accuracy of this instrument was ± 0.5 rpm. The tachometer head was pressed against the rotating roll, the start button pressed, and after five seconds the rotational speed of the roll was recorded. The linear velocity was calculated using the following equation:

$$V=R\pi d_{c}, \qquad (A-1)$$

where V denotes the velocity, R the number of rotations per minute and d_c the roll diameter.

A.1.3. Pattern Impression

The dimensions of the batch embossing roll pattern were determined using an alginate impression material, Jeltrate Chalk. The impression material was mixed with water and pressed against the embossing roll surface for 45 to 60 seconds, allowing the compound to set. The impression was then removed and a cast impression, using Polyfill Cellulose Filler, was made immediately. This was done to minimize the shrinkage that occurs as the impression material dries.

The dimensions of the embossing roll pattern were measured by taking photographs of the casting using a Leitz Wetzlar microscope, Model Dialex 20, equipped with a polaroid camera at low magnifications, i.e. 4-40x. A copper wire was placed next to the cast as a reference. The diameter of the copper wire was measured using a Mitutoyo Combinike micrometer, Model No. 159-211. the accuracy of this instrument was ± 0.0025 mm.

A.2. SAMPLE PREPARATION

A.2.1. Compression Molding

The test samples used to characterize the resin properties were prepared by compression molding the resin pellets. The general procedures outlined in ASTM D 1928-80 [171] were followed. A Dake Laboratory Molding Press, Model 44-325, was used to prepare the test samples, using the following step by step procedure:

1. The power set point for the upper and lower platen heaters on the compression molder were set at a temperature of 150°C. The temperature of the two platens on the compression molder was allowed to reach steady state (minimum 15 minutes).

2. The two aluminum back plates, 15 cm by 15 cm by 2 mm, were covered with aluminum foil, which served as a parting agent. Care was taken so as not to wrinkle the aluminum foil. The sample mold was placed on one of the plates and resin added to fill the mold. The second aluminum plate was then placed on top of the sample mold.

3. The sample mold with the two backing plates was placed in the compression molder and the two platens were brought together until the top platen was in contact with the top backing plate. A time of five

minutes was allowed for the samples to reach the set point temperature.

4. A pressure of 206.9 MPa was applied and maintained for three minutes.

5. The power to the temperature controller of the compression molder was turned off and cooling water passed through the platens to cool the test sample. 6. The plastic test sample was removed from the sample mold and visually analyzed to ensure that no air was entrapped and to ensure that the test sample dimensions were uniform.

7. The thickness of the test sample was measured using a Mitutoyo Combimike micrometer, Model No. 159-211.

A.3. PHYSICAL DIMENSIONS

A.3.1. Film Width

The width of the plastic films was measured using a measuring tape, having an accuracy of ± 0.5 mm. The plastic films were laid flat to avoid any wrinkling prior to measuring the film width. Measurements were taken at intervals of no less than one meter in the machine direction.

A micrometer, Mitutoyo Combimike Model No. 159-211, was used to measure the width of the plastic films when the width was less than 1 cm.

A.3.2. Film Thickness

The thickness of a plastic film, as illustrated in Table 4-3, can be defined in terms of a bulk and film thickness. The bulk thickness is the distance between the bottom and top surfaces of the film. The film thickness refers to the distance between the bottom and top sides of the film. In the case of a flat film, the bulk thickness is equal to the film thickness. In the case of an embossed film, the bulk thickness is always greater than, or equal to, the film thickness.

A.3.2.1. Film Thickness

The film thickness of the plastic films was measured using the density gradient technique. This approach is similar to that described in ASTM E 252-84 [172]. The density of the sample is determined and the film thickness calculated using the following equation:

$$H=w/A\rho, \qquad (A-2)$$

where H denotes the test sample thickness, w the test sample weight, A the area of the test sample and ρ the test sample density. The area of the test samples evaluated ranged between 1 and 3 cm². The dimensions of the test samples were measured using a Mitutoyo Combinike micrometer, Model No. 159-211. The sample weight was measured using a Mettler HIOT analytical scale, having an accuracy of ± 0.0001 g.

Section A.4.5. describes in detail the methodology used to prepare the density column and the procedure used to determine the density.

A.3.2.2. Bulk Thickness

The bulk thickness of the flat films was measured using a Reichert Zetopan-Pol microscope, at low magnifications (80-125x). The operation of the Reichert microscope is described in detail elsewhere [173]. The procedure used was to view a cross section of the film and then, using a calibration slide with an accuracy of ± 0.005 mm, determine the bulk thickness.

A.3.3. Film Length

The true length of the embossed films, was measured using a Reichert Zetopan-Pol microscope. The procedure used was to view a cross section of the film and then using a calibration slide with an accuracy of ± 0.005 mm, determine the length of the film. Since the embossed films were not flat, the length was divided into increments where the film was flat. The average number of increments used was 19.

The true length of the flat films was measured using a ruler.

A.3.4. Pattern Quality

The quality of the embossed film pattern was determined using photomicrography. A Reichert Zetopan-Pol microscope was used with a polarizing unit, polarizer and analyzer plates. The micrographs were visually analyzed and compared to a series of standards, Photograph A-1, to determine the pattern quality. A rating of 1 was indicative of a flat film, while a rating of 5 was indicative of a high quality embossed film. The standards used were chosen from the samples produced in this study.

The following step by step procedure was used to obtain the polarized light micrographs of the embossed films:

1. The magnification of the Reichert microscope was set at 100x.

2. The polarizer and analyzer were set at right angles to each other.

Rating















3. The embossed film sample was placed on the rotating stage of the microscope and viewed between the crossed polarizers.

4. The sample stage was rotated until extinction was achieved, i.e. maximum darkness.

5. The rotating stage was rotated 45 degrees to a position of maximum brightness.

6. The microscope image was then photographed using a polaroid camera.

A.4. MICROSTRUCTURE

A.4.1. Wide Angle X-Ray Diffraction

Wide angle x-ray diffraction was used to characterize the morphology and orientation of the a and b crystallographic axes of the polyethylene unit cell, with respect to a set of specified axes (MD-TD, MD-ND and TD-ND). A flat film photographic method (Laue method [174]) was used to obtain the average orientations of the a and b crystallographic axes. The Laue method employs monochromatic radiation which passes through the film sample to produce a photographic picture of the diffraction pattern.

The wide angle x-ray diffraction patterns were obtained using a Philips x-ray generator, Model PW 1120, supplied by Philips Electronics Industries Ltd., and a transmission pinhole camera manufactured in the Chemical Engineering machine shop at McGill University [174]. The Philips x-ray generator features 3kW maximum power, 4-80 mA continuously variable tube current and 10-60 kV continuously variable high voltage. CuK_{α} radiation with a nickel filter was used. Sample holders were available to obtain x-ray diffraction patterns related to the MD-TD, MD-ND and TD-ND planes of the plastic film. The following step by step procedure was used to obtain the x-ray diffraction pattern of a film sample:

1. The film sample was folded in the transverse direction to obtain pieces consisting either of 8 layers, or a maximum thickness of 0.2 mm.

2. Test pieces, approximately 2 cm by 3 cm, were cut from the folded film in the MD-TD plane, with the MD being the largest dimension. If the TD-ND or MD-ND planes were exposed (sample size 2 mm by 5 mm), a piece of scotch tape was used to keep the film layers together.

3. The test sample was mounted on the sample holder and subsequently mounted in the x-ray camera.

4. Under dark room conditions, the x-ray photographic film, Kodak Medical X-Ray Film NS-54T supplied by Eastman Kodak Co., was loaded into the x-ray camera and the camera sealed.

5. The x-ray camera was mounted on the x-ray unit and its alignment was checked to ensure that the xray beam was normal to the test sample. This was accomplished by turning on the x-ray unit to minimum power, and using the adjustment screws of the camera mount to align the x-ray beam; this was represented by a bright spot on the zinc sulphide based paper back stop.

6. The power to the x-ray unit was adjusted to have a current and voltage of 20 mA and 40 KV.

7. After the required exposure time, typically 8 hours, the x-ray unit was turned off and the x-ray camera removed.

8. Under dark room conditions, the film was removed from the x-ray camera.

9. The x-ray photographic film was developed by placing the x-ray photographic film in successive

batches containing Kodak D-19 developer for 4 minutes, Kodak stop bath (cat. 146 4593) for 1 minute and Kodak fixer (cat. 123 8146) for 20 minutes. The film was washed finally in cold running water for 4 hours, after which it was hung to dry overnight.

The x-ray photographs were examined to determine the position of maximum intensity on the diffraction ring and the broadness of the diffraction pattern of the (2,0,0) and (0,2,0) crystal planes. The x-ray photographs were placed on a light box to illuminate the x-ray photograph and the various angles measured using a compose, having an accuracy of ± 0.25 degrees.

A.4.2. Infrared Dichroism

Infrared absorption spectroscopy was used to determine the orientation of the a-axis of the polyethylene unit cell and the orientation of the amorphous phase in the plastic films. The data were represented in terms of dichroic ratios.

The dichroic ratio can be defined as the ratio of the absorption bands, at a specified wavelength and sample orientation to the infrared beam, in the longitudinal and latitudinal directions. Dichroic ratios equal to one indicate that the distribution of the orientation is equally dispersed in the plane of the film. Dichroic ratios greater than one indicate a preferential orientation in the machine direction. Dichroic ratios less than one indicate a preferential orientation in the transverse direction. As with wide angle x-ray diffraction, information from more than a single exposure is required to measure the biaxial orientation. However, due to the nature of the test method, it was not possible to obtain dichroic ratios for the ND-MD and TD-ND planes of the plastic film. The use of this technique to characterize the orientation of oriented plastic parts is

described elsewhere [63,174]. The following list summarizes the key infrared bands for polyethylene [175]:

Wavenumber cm ⁻¹	Туре	of Vibration	Contribution	Crystal Axis
2010	CH ₂	Extended Trans Sequences	Crystalline Amorphous	
1894	CH_2	Rocking	Amorphous	
1368	CH_2	Wagging	Amorphous	
1352	CH_2	Wagging	Amorphous	
1303	CH_2	Twisting	Amorphous	
1080	c-c	Stretching	Amorphous	
731	CH_2	Rocking	Crystalline	а
720	CH ₂	Rocking	Crystalline	Ъ

Dichroic ratios at 720, 730, 1080, 1303, 1352, 1368 and 1894 cm⁻¹ were measured using a Perkin Elmer Infrared Grating Spectrophotometer Model 467 with a Sample Space Wire Grid Polarizer. The spectrophotometer is capable of scanning from 250 cm⁻¹ to 4000 cm⁻¹. The wire grid polarizer contains a polarizing element, which is a gold wire grid vapour deposited on a silver bromide substrate, for use in the 2.5 to 35 μ m spectral range.

The following step by step procedure was used to measure the infrared absorption spectra in the machine and transverse directions of a plastic film:

1. A test sample, 2 cm by 5 cm, was cut from the film, with the larger dimension being in the machine direction.

2. The test piece was then mounted on the infrared spectrophotometer sample holder at a 45 degree angle from the vertical.

3. The infrared spectrophotometer was allowed to warm up for a period of at least 15 minutes, prior to making any measurements. 4. The infrared spectrophotometer was calibrated following the procedures outlined in the manufacturers operating manual [176].

5. The beam attenuator was set to 100% transmission at the 700 cm^{-1} band position, the scan mode set to "F" and the slit to "N".

6. The sample holder was placed into the spectrophotometer and the polarizer was set at the 45 degree position to measure the absorption in the longitudinal direction. The infrared spectra was then measured.

7. The polarizer was set to the 315 degree position to measure the absorption in the transverse position. The infrared spectra was then measured.

The infrared dichroic ratio, A_{MD}/A_{TD} , was calculated at selected wavelengths. The base line method was used to measure the absorption [177].

A.4.3. Sonic Velocity

The average orientation in the plastic films was determined using the sonic velocity, or pulse propagation technique [178]. This technique does not make use of electromagnetic radiation. When acoustic waves are sent across an array of parallel molecules, sonic energy is presumed to be transmitted from one molecule to the other by the stretching of intermolecular bonds. If sound is sent along the length of a bundle of parallel molecules, the sonic energy is presumed to be transmitted principally by stretching of the chemical bonds in the backbone of the polymer chain. In the case of partially oriented polymer molecules, the molecular motion due to sound transmission is presumed to have right angle components along and across the direction of the molecular axis. The magnitude of either of these two components is taken to be a function of the angle between the molecular axis and the direction of

sound propagation. Based on these mechanisms, the rate of sound propagation will be much greater when directed along the chain rather than when directed perpendicularly to the chain. The use of the sonic modulus technique to characterize the orientation in plastic parts has been described by various workers [63,174,178]

A convenient way to express the sonic velocity data is to compare the difference, or ratio, of the sonic velocity measured parallel and perpendicular to the machine direction, in the plane of the film (MD-TD plane).

A Dynamic Modulus Tester, Model PPM-5R, having an accuracy of ± 1.0 microsecond was used to determine the average orientation in the plastic films. The PPM-5R is a complete system comprising the tester, strip chart recorder, sample mount and transducers. The transducers contain a piezo-electric ceramic crystal with a natural frequency of 5 kHz. The operating temperature range of the transducers is from 0 to 45° C. The velocity of a longitudinal wave in the material is determined by measuring the transit time of a sonic pulse between the two transducers in contact with the sample. By varying the transducer separation distance and recording the response time, the sonic velocity can be calculated.

The following step by step procedure was used to measure the sonic velocity:

1. The power to the PPM-5R was switched on and the apparatus was allowed to warm up for at least 15 minutes.

2. Test samples, 1 cm by 12 cm, were cut from the film, parallel and perpendicular to the machine direction.

3. The range switch was set at 400 microseconds and the chart recorder speed at 1.91 cm/minute. The PPM-5R was calibrated following the procedures outlined in Reference 179.

4. The test sample was placed on the sample mount and the two ends of the sample were taped to the sample mount for support, using masking tape.
5. The distance between the two transducers was set at 10 cm and the lever on the sample mount was pressed until both transducers touched the surface of the test sample. The transit time, recorded on the chart recorder, was measured for 5 seconds.
6. The above procedure was repeated, for separation distances of 8, 6, 4 and 2 cm.

The sonic velocity was obtained by plotting the transit time as a function of the distance between the transducers. The relationship between the transit time and transducer separation distance is linear. The slope of this line is the reciprocal of the sonic velocity.

A.4.4. Birefringence

Birefringence, as does the sonic velocity technique, provides a measure of the total molecular orientation in a system. It is defined as the difference between the principle refractive indices, parallel and perpendicular to the plane of symmetry. The refractive index, in turn, is a measure of the velocity of light in the medium and is related to the polarizability chains. of the Thus, the final measurement of the birefringence is a function of contributions from the polarizabilities of all the molecular units in the sample. The use of this technique to characterize oriented plastic parts is described elsewhere [63,174].

A Reichert Zetopan-Pol microscope together with a Ehringhaus rotary compensator, having an accuracy of ± 0.05 degrees, was used to measure the phase difference of the light waves after passing through the test sample. The compensator is located after the test sample and between the polarizer and analyzer on the microscope.

The following step by step test procedure was used to measure the birefringence:

1. Test samples, 2 cm by 2 cm, were cut from the film and the orientation of the test sample relative to the film sheet was noted.

2. The thickness of the test samples was measured using a Mitutoyo Combimike Micrometer, Model No. 159-211.

3. The magnification on the microscope was set at 100x.

4. The polarizer and analyzer were set at right angles to each other.

5. The test sample was placed on the rotating sample mount of the microscope.

6. The sample mount was rotated until extinction was achieved.

7. The sample mount was rotated 45 degrees in the clockwise direction, to a position of maximum brightness.

8. The compensator was used to measure the retardation angle. Readings were taken in both the clockwise and counter clockwise rotation directions. The average retardation angle was calculated. If the retardation angle could not be seen, the compensator was reset to zero, the sample mount rotated counterclockwise to the position of extinction, the sample mount rotated 45 degrees counterclockwise and the retardation angles measured.

9. The orientation of the test sample relative to the axis of the compensator was recorded.
10. The measured retardation angle was converted to the phase difference using a table containing the phase differences for light of the wavelength of the e-line of the solar spectrum and for inclinations of the quartz plate from 0 to 70 degrees in intervals of tenths of a degree [180].

The birefringence was determined by dividing the phase difference by the sample thickness. If the machine direction of the film sample was parallel to the axis of the compensator, the birefringence was calculated as being positive. If the transverse direction of the film was parallel to the axis of the compensator, the birefringence was calculated as being negative.

A.4.5. Density

The density of the flat and embossed plastic films was determined using the density gradient technique, ASTM D 1505-85 [181]. Using this technique, the height of the test sample in the density column relative to two reference floats of known density, defines the sample density.

The density gradient technique employs a glass cylinder which is filled with a mixture of isopropanol and water. Filling of the column is carried out such that the liquid concentration varies continuously with the cylinder height. The concentration of water, and hence the density of the mixture, tends to be higher toward the bottom of the cylinder. In addition, a number of colored calibration floats of known density are placed such that the actual density variation with the column height can be established. The following calibration floats were used in this study:

		<u>Density q/cm³</u>
Float	1	.900
Float	2	.910
Float	3	.916
Float	4	.922
Float	5	.924

The following step by step procedure was used to determine the density of the flat and embossed plastic films:

1. The density column was prepared using standard procedures [174,181].

2. Film samples, approximately 1 cm by 0.5 cm, were cut from the flat or embossed films. Each of the samples was cut into a different shape for identification purposes.

3. Using a pair of tweezers, each sample was wetted in the light density mixture of isopropanol and water.

4. The sample was dropped into the column using tweezers. Twelve hours were allowed to pass, allowing the samples to attain an equilibrium position in the column.

4. The relative heights of each sample and of the calibration floats were measured using a cathetometer.

The density of the samples was calculated by linear interpolation, using the following equation:

$$\rho_{s} = \rho_{a} + (\rho_{b} - \rho_{a}) (h_{a} - h_{s}) / (h_{a} - h_{b})$$
(A-3)

where ρ denotes the density, h the height measured by the cathetometer, a the float above the sample and b the float immediately below the sample.
A.4.6. Crystallinity

Polyethylene is known to crystallize only partially and to consist of a crystalline portion described by the ideal crystal structure, and an amorphous portion described by the ideal melt. The volume fraction crystallinity is given by the following equation [63]:

$$X_{c} = (\rho_{s} - \rho_{am}) / (\rho_{cr} - \rho_{am})$$
(A-4)

where X_c denotes the crystallinity, ρ_s the sample density, ρ_{am} the density of a 100% amorphous sample and ρ_{cr} the density of a 100% crystalline sample. The values of ρ_{cr} and ρ_{am} were taken to be 1.001 and 0.856, respectively, in this study [32].

A.5. RHEOLOGICAL PROPERTIES

A.5.1. Shear Viscosity

Two techniques were used to determine the shear viscosity of the resins used in this study, as a function of shear rate and temperature. High shear rate viscosity measurements were made using an Instron Capillary Viscometer, while low shear rate viscosity measurements were made using a Rheometric Mechanical Spectrometer with a cone and plate attachment.

A.5.1.1. Cone and Plate Geometry

The cone and plate geometry is used extensively for the rheological characterization of viscoelastic materials. The sample is contained between a cone and a plate, both having the same radius and one of which rotates at a fixed angular velocity. Two measurements are usually made: the torque that is required to turn the rotating member, or to hold the stationary member in place, and the total force that is exerted by the fluid on the plate or cone. Cone and plate rheometers are commonly used to study materials limited to a maximum shear rate in the range of 0.1 to 10 s⁻¹. In order to derive a simple expression for the shear rate in cone and plate geometry, it is necessary to make a number of assumptions and approximations, which are only valid for certain combinations of cone angle, rotational speed and fluid properties. The assumptions and equations used to derive the flow pattern in this geometry are described in detail in standard references [182,183].

Viscosity measurements at low shear rates were made using a Rheometrics Mechanical Spectrometer, Model 605, with a cone and plate attachment. The cone angle was 0.04 radians and the cone and plate fixtures had a radius of 12.5 mm. As is typical of cone and plate units, the apex of the cone is truncated to prevent any damage to the fixtures when the cone is rotated. The temperature of the test sample is controlled by a forced convection oven.

The following step by step procedure was used to make all the measurements:

1. The forced convection chamber was preheated to the test temperature.

2. The micrometer was adjusted to read zero, when the cone tip and plate were in contact.

3. Once the gap was set, the convection oven gas supply was switched from air to nitrogen. Nitrogen was circulated for five minutes before the sample was inserted.

4. The sample was placed on the plate and was allowed to reach steady state.

5. Since the test sample is much thicker than the gap spacing, the cone tip was brought down to 100 μ m from the plate, and the excess material removed using a scraper. Subsequently, the gap was set at

50 μ m. Sufficient time was allowed until the temperature reached steady state. The time between the insertion of the sample and start of the test ranged between 10 and 20 minutes.

6. The mode of operation (rate sweep), shear rate and temperature were set, and the experiment was carried out. The first normal stress and viscosity are outputted directly to the system printer as functions of the shear rate and time.

A.5.1.2. Capillary Viscometer

The measurement of the flow rate in a tube, as a function of pressure drop, is the most popular technique to study the rheological properties of liquids because it is the viscometric flow most easily generated in the laboratory. Tube flow is commonly referred to as capillary flow. The assumptions and equations used to describe the flow pattern for this geometry can be found in standard references [182,183].

Viscosity measurements at high shear rates were made using an Instron Universal Testing Machine, Model TT-CV, equipped with a capillary viscometer [183]. The viscometer consisted of a hardened steel barrel with an accurately ground 0.953 cm inside diameter, mounted on a support assembly underneath the moving crosshead of the Instron tester. A plunger, which was made to fit inside the barrel, is driven by the moving crosshead of the machine. The force on the sample is detected by a strain gauge load cell mounted on the moving crosshead. The capillary is inserted in the bottom of the barrel and is held in place with a clamping nut. The barrel and capillary wall are maintained at a constant preset temperature. The capillaries used in this study had diameters of 0.762 mm and 1.321 mm, L/D ratios of 10, 20 and 40 and an entrance angle of 90 degrees. The following step by step procedure was used to make all the measurements:

1. The load cell, having an accuracy of ± 0.5 kg, was calibrated by hanging calibration weights on a fixture. The reproducibility of the measurements was checked by alternately adding or removing the weights.

2. The barrel temperature was set by adjusting the barrel temperature controller. One to two hours were allowed for the barrel temperature to reach steady state.

3. The barrel was filled with resin pellets, while occasionally pressing the material down to avoid air bubbles. Approximately 10 minutes were required to melt the resin and reach the test temperature.

4. The piston was inserted and lowered to compact and pressurize the material. To release the air bubbles, it was necessary to extrude some of the polymer and wait until the pressure had returned to approximately zero.

5. The experiment was started at the selected cross head speed, which ranged from .05 cm/s to 20 cm/s. The force as a function of time was recorded on the chart recorder.

6. When the force reading on the chart recorder became steady, the test was stopped and a new speed selected.

A computer program was developed to analyze the raw data and calculate the viscosity as a function of temperature and shear rate. The equilibrium force values were converted to pressure drop values using the barrel area. Bagley corrections were made at each shear rate using the pressure drops at the different L/D ratios. In cases where the Bagley corrections were smaller than the standard error, they were taken to be zero. Rabinowitsch correction was employed to convert the apparent shear rates into shear rates at the wall, from which the true viscosities were calculated.

A.5.2. Storage and Loss Moduli

The shear storage and loss moduli were determined using a Rheometric Mechanical Spectrometer with a cone and plate attachment. To operate the spectrometer in the oscillatory mode, the mode of operation was set to either time or temperature sweep and to the specified strain. The procedure outlined in section A.5.1.1. was followed.

A.6. MECHANICAL PROPERTIES

There are three basic mechanical properties which are of interest when working with plastics [184]: stiffness, strength and toughness. Stiffness represents resistance to deformation, while strength and toughness relate to failure of the material. Strength is the ultimate stress which a material can withstand before it fails. Toughness represents the work required to fracture a material.

A.6.1. Tensile Properties

In general, the tensile test evaluates the capacity of a material to withstand forces that tend to pull it apart and the extent to which the material stretches before breaking. Tensile stress-strain measurements are generally made by stretching the sample at a uniform rate, while recording the force required to stretch the sample. The Young's modulus, ultimate strength, yield strength and elongation at yield are among the most important indications of strength in a material and are the most widely specified properties [184]. The tensile tests performed in this study were carried out following the methodology outlined in ASTM D 882-83 [185]. This procedure allows for measuring the tensile properties of thin films.

An Instron Universal Testing Machine, Model 1123, was used to determine the tensile properties. A tensile load cell, type CM manufactured by Instron, and having full scale ranges of 1, 2, 5, 10, 20 and 50 Kg, was used in conjunction with the Instron Tester. The accuracy of the load system used was ± 0.5 % of the indicated load, or ± 0.25 % of the recorder scale in use, whichever was greater. The positional servo-drive provided a speed accuracy of ± 0.1 % of the set speed, at any load and speed within the overall range of the instrument. Two grips, manufactured by Instron, were used to hold the test samples. The grips had a load capacity of 100 Kg and the maximum sample thickness which could be used was 0.64 cm.

The following step by step procedure was used to measure the tensile properties of a plastic film:

1. The load weighing mechanism of the Instron tester was calibrated using a full scale load of 1 Kg. 2. A 1 cm by 14 cm test sample was cut from the plastic film. The thickness and width of the test samples was measured using a Mitutoyo Combinike Micrometer, Model No. 159-211. The length was measured using a ruler.

3. The distance between the two grips was adjusted to 10 cm.

4. The test sample was first mounted into the top grip and then carefully aligned in the second grip to ensure that the film remained flat and vertical.

A-27

5. The test specimen was extended at a strain rate of 5 mm/min. The chart recorder speed was set at 1 mm/min. The chart recording showed the load as the abscissa and the elongation as the ordinate.

Using the stress-strain curve and knowing the initial cross sectional area and the gauge length of the test sample, the ultimate tensile strength, elongation at the ultimate point and Young's modulus were determined. The strain was calculated as follows:

$$\epsilon = L - L_0 / L_0, \qquad (A-5)$$

where ϵ denotes the strain, L the length of the stretched sample and L₀ the initial length of the sample. Generally, the stress is calculated on the basis of the original cross sectional area of the sample. However, the cross sectional area of the test sample decreases as it is stretched. As a result, the sample area at any time, or strain, was expressed as:

$$A=A_0L_0/L, \qquad (A-6)$$

where A denotes the area at any time during elongation and A_0 the initial cross sectional area of the test sample.

A.6.2. Compression Properties

The compression tests were carried out following the methodology outlined in ASTM D 695-89 [186]. An Instron Universal Testing Machine, Model 1123, in conjunction with an Instron Environmental chamber, Model A74, was used to determine the compression properties. A load cell, type DRM manufactured by Instron, and having full scale ranges of 10, 20, 50, 100, 200 and 500 Kg was used in conjunction with the Instron tester. Two parallel plates, having a diameter of 15.24 cm and a thickness of 1.91 cm, supported by adapter rods were used instead of the tensile grips. The adapter rods were designed so that they could be attached in the same way as the tensile grips.

The following step by step procedure was used to measure the compression properties of a plastic material:

1. Compression molded test samples were made using an eight cavity mold following the procedures outlined in Section A.2.1.. The molds used allowed for the preparation of cylindrical samples having diameters of 0.635, 1.27 and 2.54 cm and thicknesses of 2.54 and 5.08 cm.

2. The diameter and length of the cylindrical samples were measured using a Mitutoyo Combimike Micrometer, Model No. 159-211.

3. The load weighing mechanism of the Instron tester was calibrated using a full scale load of 10 Kg.

4. The environmental chamber was set to the desired set point temperature and allowed to reach steady state (minimum 30 minutes).

5. The test sample was placed on the bottom plate. Care was taken to ensure that the sample was positioned at the center of the plate.

 The top plate was lowered manually until it was in contact with the test sample. Care was taken to ensure that the test sample was not under a load. The cross head position indicator was reset to zero.
 The test sample was allowed to reach the steady state temperature (minimum 15 minutes). 8. The test specimen was compressed at a strain rate of 0.5 or 1.0 mm/min. The chart recorder speed was set at 1 mm/min. The chart recording showed the load as the abscissa and the compressive strain as the ordinate.

By knowing the initial cross sectional area and the gauge length of the test sample, the yield point, proportional limit and modulus were determined. The sample area was corrected using Equation A-6.

A.6.3. Impact Properties

An instrumented Rheometrics High Rate Impact Tester, Model RHIT 8000, was used to measure the impact properties. The impact tester is built around a linear displacement, velocity controlled, hydraulically driven mechanism. The instrument controls the rate at which a cylindrical probe with a spherical tip, punctures the test sample. The instrument is equipped with a data acquisition system, which presents the results in the form of a force-deflection curve. A detailed description of the impact tester is given elsewhere [187].

The following step by step procedure was used to measure the impact properties of a plastic film:

 The impact tester was turned on and allowed to warm up for at least 30 minutes.
 A 7.5 cm by 7.5 cm test sample was cut from the plastic film. The sample width and length were measured using a ruler, while the sample thickness was measured using a Mitutoyo Combinike Micrometer, Model No. 159-211.

3. The test sample was clamped in the sample holder.

A-30

4. The impact velocity was set at 0.212 m/s and the test conducted.

5. The generated impact data after each test were displayed on a CRT display. The points representing the yield and break points and the appropriate range for the modulus determination were individually selected. The modulus and ultimate stress were calculated using the instrument microprocessor and outputted on the instrument printer.

A.7. THERMAL PROPERTIES

The three principal thermal properties which are used to describe heat transfer through a material are [41]: thermal diffusivity, thermal conductivity and specific heat. The thermal conductivity is essential for the prediction of heat flow rates and temperature distributions for steady state conditions. The specific heat describes the amount of heat needed to raise the temperature of the material by one degree. The thermal diffusivity determines the time dependent conditions for the unsteady state.

A.7.1 Thermal Conductivity

Thermal conductivity is defined as the quantity of heat, at the steady state condition, passing through an area forming part of a slab of uniform material of infinite extent with flat and parallel faces [41]. For the heat flow in one direction, the heat flux is related to the temperature gradient by Fourier's law as follows [41]:

$$q=-k(\partial T/\partial X)$$
, (A-7)

where q denotes the heat flux, k the thermal conductivity and $\partial T/\partial X$ the temperature gradient in the thickness direction. The

negative sign indicates that the heat flow is in the direction opposite to the temperature gradient. The methods commonly used to measure the thermal conductivity of plastic materials are reported elsewhere [41].

The apparatus used to measure the thermal conductivity of LDPE-1 was developed in the Chemical Engineering Laboratories at McGill University [41]. This instrument is based on a steady state technique to measure thermal conductivity and permits measurement of the thermal conductivity and the thermal diffusivity of materials in the solid, as well as in the melt state. The apparatus consists of an insulated constant temperature chamber with two metal fixtures to keep the two stainless steel sample holder plates at the top and at the bottom of the conductivity cell, in position. The design allows for testing of two samples simultaneously. A nichrome wire heater is rolled over a mica sheet between the two stainless steel conductivity cells in order to ensure a symmetrical distribution of heat on both sides of the conductivity cells. coated copper-constantan Kapton thermocouples are positioned inside grooves provided on the inside part of the conductivity cell. The thermocouple tips are placed flush with the metal surface to record the sample surface temperature as a function of time during the experiments. A Dymec digital voltmeter and a Beckman digital ammeter were used to measure the voltage and the current required to heat the samples.

The following step by step procedure was used to measure the thermal conductivity of the resins evaluated in this study:

1. Compression molded test samples were made following the procedures outlined in section A.2.1., using a four cavity mold with a disc geometry (5 cm diameter and 1 mm thick). 2. The thickness of the compression molded samples was measured using a Mitutoyo Combinike Micrometer, Model No. 159-211.

3. The compression molded samples were placed in the conductivity cells. The bottom sample was placed on the male bottom plate and centered to ensure proper contact with the thermocouple. The female lower plate was then lowered onto the sample. Support screws were used to confine the sample and close the lower part of the conductivity cell. A similar procedure was used to secure the top sample. 4. The conductivity cells were secured in the constant temperature chamber.

5. The constant temperature convection chamber was turned on and the temperature set. After 10 to 15 minutes, the temperature of the chamber and sample to reached steady state.

6. The direct current power supply was turned on, and a high intensity current was applied to the nichrome wire. An immediate rise in temperature was detected in both thermocouples 2 and 3. The time response for the initial temperature rise and penetration time for thermocouples 1 and 4 were recorded. The penetration time was determined as the time when the temperature increase was found to be greater than or equal to 0.1 C.

7. After 15 minutes, the temperature gradient between thermocouples 1-2 and 3-4 was measured.
8. A new temperature setting was chosen and the procedure repeated.

The value of thermal conductivity at each temperature was computed using the following equation:

$$k = 0.005422 V I_H,$$
 (A-8)
 ΔT

where k denotes the thermal conductivity, V the voltage, I the amperage, H the sample thickness and ΔT the average temperature difference between thermocouples 1-2 and 3-4.

A.7.2. Thermal Diffusivity

The thermal diffusivity determines the time dependent temperature distribution in the material. The mathematical definition of the thermal diffusivity is [41]:

$$\alpha = k / \rho C_{\rm P}, \qquad (A-9)$$

where α denotes the thermal diffusivity, k the thermal conductivity, ρ the density and C_{v} the specific heat. The thermal diffusivity was measured using the penetration time data obtained while measuring the thermal conductivity. A computer program was used to analyze the data [41].

A.7.3. Specific Heat

A Perkin Elmer differential scanning calorimeter, Model DSC-1 and DSC-7, was used to measure the specific heat as a function of temperature for the resins evaluated in this study. Comprehensive description of the instrument and its operation is given elsewhere [188,189].

Differential scanning calorimetry is used to carry out direct calorimetric measurements of the changes in enthalpy of a polymer during heating and cooling cycles. The sample and reference compartments in the calorimeter are kept at identical temperatures by means of two separate heaters which are located under the sample and reference holders. Reference and sample temperatures are measured by platinum resistance thermometers and are fed into the differential amplifier whose output adjusts the necessary differential power increment to keep both sample and reference channels at the same temperature throughout the analysis. A signal proportional to the differential power output is traced out on a Hewlett Packard strip chart recorder (DSC-1) or plotter (DSC-7).

Aluminum pans, supplied by Perkin Elmer, were used to hold the plastic samples and indium crystals used to calibrate the system. A Cahn-RG electrobalance, with a capacity of 2.5 grams and a sensitivity of ± 0.1 micrograms, was used to weigh the samples. The calorimeters can be used to perform experiments under isothermal or non-isothermal conditions with constant rates of heating or cooling.

The following step by step procedure was used to measure the specific heat of LDPE-1:

1. Samples were cut from compression molded sheets using a punch with a diameter of 6 mm, approximately equal in size to that of a DSC sample pan, supplied by Perkin Elmer Co. The compression molded sheets were prepared following the procedure outlined in section A.2.1..

2. The test samples were weighed using the electrobalance. The test samples were then placed and sealed in the aluminum pans.

 The differential scanning calorimeter was turned on and allowed to warm-up for at least 30 minutes.
 The calorimeter was calibrated for both average and differential temperatures using the procedure outlined in Reference 188.

5. The two aluminum pans, one of which contains the sample and the other remaining empty, were placed in the assigned compartments and heated to 150°C. The samples were maintained at this temperature for 10 minutes to ensure total relaxation of the sample and eliminate traces of previous crystallinity. 6. The samples were cooled at 10°C/min to 30°C.7. The plastic sample was removed, and the scanning curve analyzed.

The specific heats for the resins were obtained using the ratio method [190]. The reference material used to calculate the specific heat was sapphire [191].

A.7.4. Melting Characteristics

It is possible to gain information regarding the melting behaviour of semi-crystalline materials by analyzing the nonisothermal differential scanning calorimetry data. The peak temperature is the temperature corresponding to the peak of the thermogram, while the onset temperature relates to the temperature at which melting starts. The onset temperature is determined by extending the slope of the thermogram near the peak, to intersect with the baseline [192]. The heat of fusion is determined by calculating the area under the thermogram [193].

The melting characteristics of LDPE-1 were measured using a Perkin Elmer DSC. The operating and calibration procedures for this instrument are outlined in section A.7.3..

A.8. OPTICAL PROPERTIES

When light falls onto a surface, four processes can occur: reflection, refraction, scattering and absorption. If the surface of the film is smooth, then the reflected light component, which occurs at an angle equal to the angle of incidence, is wholly responsible for the intensity of the reflected image, i.e. the glossiness of the surface. Light that penetrates the surface of the film is refracted, scattered or absorbed. Since these processes help to obscure the substrate, they play a major role in determining the opacity of a film or sheet.

The optical properties of the plastic films were assessed by measuring the surface gloss and opacity.

A.8.1. Gloss

Gloss is a property directly related to the physical state of the surface of a plastic film. Gloss can be regarded as the capacity of a surface to reflect light and has been defined by Hunter [118] as the degree to which a surface simulates the reflecting power of a perfect mirror. Hunter defines four types of gloss (specular, absence of bloom, distinctness of image and contrast gloss) depending on the intensity and shape of the reflected light profile, which itself is related to the physical nature of the surface.

A Hunterlab Model D48D Specular Glossmeter, having an accuracy of ± 0.05 , was used to measure the gloss of the plastic films. Specular gloss is a measure of the brightness of the reflected image. A parallel or slightly convergent beam of light is incident on the film under study at a fixed angle of incidence. A photodetector and collimating system are likewise fixed in a position corresponding to the mirror image angle.

The following step by step procedure was used to measure the gloss of a plastic film:

1. The glossmeter was turned on and allowed to warm up for at least 30 minutes,

2. The glossmeter was zeroed and calibrated using the procedures described in the operating manual [194] and a HunterLab gloss standard, D33G-5626. The specular gloss of the standard, measured at 20° and 60° , was 88.5 and 93.4 respectively. 3. The plastic film sample, 10 cm \times 10 cm, was placed under the glossmeter optical unit with the embossed pattern facing upwards and the machine direction parallel with the light source.

4. The digital readout on the glossmeter was recorded.

5. Measurements were taken using the 20 and 60 degree angle of incidence optical units.

A.8.2. Contrast Ratio

The opacity of a plastic film was determined in terms of the contrast ratio [195] in this study. The contrast ratio is defined as the ratio of the CIE Y value of the plastic film over a black substrate and the plastic film over a white substrate. A HunterLab Model D25A-9 colorimeter was used to measure the contrast ratio. The procedure used to zero, calibrate and operate the Hunterlab colorimeter is described elsewhere [196]. The CIE Y values of the white and black substrates used in this study were 84.0 and 0, respectively.

A.9. AESTHETIC PROPERTIES

A.9.1. Pattern Texture

The feel of a plastic film can be considered as the sensation experienced by the observer when touching the film. It therefore is a subjective measurement, and different observers may evaluate the texture of the film differently. The texture of the plastic films was determined by rubbing the film between one's fingers.

The following step by step procedure was used to determine the texture of a plastic film:

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1. A piece of plastic film, 10 cm x 10 cm, was placed between the thumb and fingers of the right hand, with the pattern facing upwards and with the machine direction facing toward the tips of the fingers.

2. The thumb was rotated in the clockwise direction for at least five seconds.

3. The feel, or texture, of the film was determined by comparing the sensation felt against a series of standards (Photograph A-1).

4. A rating from 1 to 5 was given based on the sensation felt. The same standards used to evaluate the pattern quality were used. The films were rated on a scale of 1 to 5, with 1 being indicative of a flat film and 5 indicative of the film believed to have the highest embossed film pattern quality.

No more than 10 films were evaluated at a time, in order to avoid overstimulating the nerve endings in the finger tips.

APPENDIX B EXPERIMENTAL DATA

This appendix presents a summary of the experimental data measured in this study. The data has been grouped into the following categories:

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The data tables can be found in the file named **APPENDIX.B** on the enclosed computer disk. The computer disk is formatted for use with an IBM PC XT computer or equivalent. The file **APPENDIX.B** is formatted using WORDPERFECT Version 5.0 for use with a HEWLETT PACKARD Laserjet Series II laser printer with a HEWLETT PACKARD 92286J MATH ELITE font cartridge. This file can be printed by selecting the print menu, **shift-F7**, and selecting the **Full** Document option.

The confidence intervals were calculated at a 95% significance level. Reference 26 describes in detail the equations used to calculate the mean, standard deviation and confidence interval based on n measurements.

B.5. FIGURES

B.5.1. Effect of Film Velocity





























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B.5.4. Effect of Initial Film Orientation





Radiation Heater Temperature Setting, °C

Figure B-4b: Effect of Initial Film Orientation $T_p=93.3^{\circ}C$, P=0.2 MPa, H=1 layer and V=20.6 m/min.



Figure B-4c: Effect of Initial Film Orientation $T_P=93.3^{\circ}C$, P=0.2 MPa, H=1 layer and V=20.6 m/min.



Figure B-4d: Effect of Initial Film Orientation $\underline{T}_P = 93.3^{\circ}C$, P=0.2 MPa, H=1 layer and V=20.6 m/min.
APPENDIX C

MORPHOLOGY AND ORIENTATION IN POLYETHYLENE TUBULAR BLOWN FILMS

Paper presented at the Society of Plastics Engineers Annual Technical Conference (ANTEC), Los-Angeles, May (1987), published in the SPE Technical Papers, Vol. 33, page 446 (1987) and published in Plastics Engineering, page 45, October (1988).

INTRODUCTION

The most common method for the production of plastic films is the tubular film blowing process, Figure C-1. Molten polymer is extruded through an annular die and the resulting parison is drawn upwards by the take-off rolls. Simultaneously, air is introduced at the bottom of the die, thereby inflating the parison. Both the forward stretching and sideways blowup occur in the melt before the polymer solidifies at the freeze line. An air ring is used to cool and solidify the hot bubble. The inflated bubble is flattened as it passes through the nip rollers, which also serves to provide the axial tension to draw the film upwards and form an air tight seal so that a constant pressure is maintained in the bubble. The key process variables are:

- 1. Melt temperature
- 2. Draw ratio
- 3. Blowup ratio
- 4. Freeze line height

The most common methods used for the characterization of orientation and morphology of polymer films are: birefringence, infrared dichroism and x-ray diffraction. The latter yielding the most useful information in the case of semi-crystalline materials, such as polyethylene. Maddams and Preedy [1,2,3], present possibly the most comprehensive experimental and x-ray analysis of the film blowing of polyethylene resins and describe in detail the previous work done in this area.

The stresses experienced by the polymer melt during the blowing process, are the combined result of several factors determined by the blowing conditions and the rheology of the resin. The thermo-mechanical history experienced by the melt is what determines the morphology and orientation of the crystalline and amorphous phases. When the polymer passes through the die, it is subject to shearing stresses, which result in partial orientation of the chain axis toward the machine direction, MD. When the melt leaves the die, these shearing forces vanish and the melt experiences biaxial stresses. As a result of the reduction in thickness and an increase in viscosity, due to cooling of the film bubble, these stresses increase. Moreover, the stresses favour *c*-axis orientation along the machine direction. Simultaneously however, reorientation may also occur due to relaxation, which for polyethylene has been shown to shift the c-axis away from the machine direction, resulting in a-axis orientation in the machine direction [4,5,6]. The overall effect, therefore, may be rather complex.

The objective of this study was to characterize the structure and properties of tubular blown polyethylene films processed at a variety of conditions. An attempt is made to explain the morphology based on the resin rheological properties and stresses during processing.

EXPERIMENTAL

Three commercial Union Carbide polyethylene resins were used in this study, 0774 Low Density Polyethylene (LDPE), 6039 High Density Polyethylene (HDPE) and 7047 Linear Low Density Polyethylene (LLDPE). The flat tubular films were blown using a Black Clawson extruder with a five inch diameter annular

die. Table C-1 outlines the processing conditions used in this study. The following methods were used to characterize the microstructure and properties of the flat films.

1. Wide angle x-ray diffraction (WAXD) to determine crystalline orientation and morphology.

2. Infrared spectroscopy (dichroic ratio) to determine *a*-axis orientation.

3. Sonic modulus and birefringence to determine average orientation.

4. Density column to determine crystallinity.

5. Tensile and impact tests to determine mechanical properties.

A conventional flat plate Laue camera was used with specially designed sample holders which positioned the film normal, parallel and perpendicular to the x-ray beam. The x-ray photographs provide information regarding the a and b axes orientation. The angle of the crystal axis can be determined from direct inspection of the x-ray photographs. The limitation of this method is that one obtains only qualitative identification of the orientation. The use of the flat plate technique to determine the pole figures for the various samples was found to be inaccurate due to the lack of reproducibility.

The rheological properties for the three resins studied are described in References 7 and 8.

BACKGROUND

The stress crystallization model of Keller [4,9] has been widely accepted to describe the crystallization and reorientation process for polyethylene. Polyethylene contains molecules covering a range of molecular weights. During processing, in the die, the polyethylene melt has an extended

chain form with the *c*-axis parallel to the machine direction. When the melt exits the die and cooling occurs, these extended chains act as nucleating points for bulk crystallization. As this takes place along the length of the extended chain, new crystallization can occur only in the radial direction. This results in the preferential orientation of the b-axis perpendicular to the machine direction, i.e. transverse This secondary crystallization occurs as the direction. conventional chain folded lamellae twist completely. When the stress is high, the molecules become aligned along the thickness direction and the c-axis aligned parallel to the machine direction. Under low stress conditions, the a-axis will have a maximum along the machine direction, but with a broad spread all round the ring. It is possible for the a-axis to be oriented at an angle to the machine direction for intermediate stress levels. Figure C-2 summarizes the stress crystallization model of Keller. The direction of the a-axis represents the mean orientation, which for a particular inclination from the machine-transverse direction plane lies in the thickness-machine direction plane and is symmetrical about the thickness-machine direction plane.

Conventional films polyethylene demonstrate low to intermediate stress crystallization orientation, with the angle of inclination varying from 20-50 degrees. It has been shown, by Maddams and Preedy [1,2,3] that both low and high stress crystallization orientation can occur in varying proportions in the film. This is determined by the particular polymer used. The lower the chain mobility, of which the elongational viscosity is a measure, the greater should be the stress during crystallization. As the temperature decreases or the degree of long chain branching increases, the polymer mobility decreases and the stress increases.

The crystallization process will begin with the very high molecular weight material and will proceed over a temperature

interval, until the lowest molecular weight material has solidified. Under these circumstances, reorientation can easily occur and the *c*-axis will shift further away from the machine direction.

This model can be used to explain the results of previous film studies using polyethylene, as well as clarify the controversy during the 1950's and 1960's where three models were postulated. The models, Type I, II and III [9,10,11] can be explained by considering different levels of stress and relaxation.

RESULTS AND DISCUSSION

Rheological Properties:

As the polymer melt approaches the freeze line, the changes in the bubble radius become smaller. As a result, near the freeze line, we would expect the extensional forces to dominate over the hoop and shear forces. Rheological data based on viscometric flow can be used to describe the flow of the melt through the die. Since the shearing history in the die is more or less erased by the time the melt reaches the line [12,13,14], viscometric based rheological freeze functions should provide little or no information as to the stress level at the freeze line region. Extensional flow rheological functions, on the other hand, should be more useful in yielding information as to the stress level at the freeze line. It is important to note that the majority of experimental techniques are limited to making measurements in the linear viscoelastic range, while during processing the deformation rates are large and nonlinear.

Figure C-3 presents extensional data for the three polyethylene film resins at 150°C, linear low density polyethylene exhibits strain softening behaviour and the

extensional viscosity is lower than that for high and low density polyethylene. Low density polyethylene exhibits strain hardening behaviour, while high density polyethylene is intermediate between the two. The chain mobilities of the resins, low density polyethylene exhibiting the lowest and linear low density polyethylene the highest, are consistent with the observed rheological behaviour.

Due to the lower extensional viscosity, linear low density polyethylene can easily be downgaged. On the other hand, linear low density polyethylene offers less resistance to the air cooling stream and thus, in many instances, bubble instability is encountered. Incorporating co-current air flow instead of impinging air flow in the air ring design or reducing the air velocity has helped in reducing these difficulties.

Based on these results, we would expect low density polyethylene to exhibit the highest stresses at the freeze line, followed by high density polyethylene, and linear low density polyethylene showing the lowest. This translates using the model of Keller, for linear low density polyethylene to exhibit the lowest stress related structure, followed by high density polyethylene and low density polyethylene with higher stress type structures. The reduced air velocity used in processing linear low density polyethylene results in longer relaxation times and leads to even a higher degree of *a*-axis orientation.

These conclusions are based on comparison of the rheological data at 150°C. The processing temperature for all three resins differ, with high density polyethylene processed at 40°C higher than either linear low and low density polyethylene. At the freeze line, the temperature of high density polyethylene will be higher than for both low density resins. As a result, the elongational viscosity of high density

polyethylene will be lower and exhibit a lower stress related structure as that predicted above. Figures C-4 and C-5 illustrate the effect of temperature on the elongational data. For all temperatures, low density polyethylene exhibits strain hardening behaviour, while high density polyethylene is less sensitive to temperature variations. The overall ranking with respect to the stress levels remains unchanged even after considering temperature effects.

An analysis incorporating the simulation of the dynamics, kinematics and heat transfer in the process, combined with experimental data, would allow for quantitative verification of the conclusion made above.

For comparative purposes, the viscous properties for the three resins were examined. The shear viscosity of the three resins were very similar, with high density polyethylene having the highest and linear low density polyethylene the lowest shear viscosity. Unlike the elongational data, the three resins all exhibit shear thinning behaviour and little information can be inferred form them.

Film Structure and Properties:

The film blowing process involves the complex interaction between several variables. By comparing the blowup ratio for example, the draw ratio, film velocity, freeze line height and cooling air velocity can vary.

Tables C-2 to C-4 summarize the measured properties of the various films tested. The variability of the properties of the flat film was analyzed for film samples at different locations. It was found that the dimensions, properties and structure were fairly uniform regardless of where the samples were taken.

The x-ray data support the prediction that linear low density polyethylene exhibits low stress orientation, a-axis parallel to the machine direction. Due to the low stress levels, the orientation was found to remain constant regardless of the blowup ratio. Although the a-axis has preferential orientation in the machine direction, there is still a large spread of aaxis orientation in all directions, i.e random. The overall level of orientation in these films is very low, indicating a very low degree of stress crystallization orientation. This can be attributed not only to the rheological properties of the resin, but also to the processing conditions which had to be adjusted to avoid bubble instability. The cooling air and film velocities settings were lower in comparison to those used for the low and high density polyethylene films produced. High density polyethylene also exhibited low stress level orientation, while low density polyethylene an intermediate value. The a-axis for all the low density polyethylene samples, were inclined at an angle from the TD-ND plane in the ND direction. For all the samples, symmetry was noted about the ND-MD plane in the TD-ND plane. Some degree of a-axis orientation was observed, for all the films. in all directions. No sample showed pure a-axis orientation. This is as expected due to the crystallization process occurring over a temperature range. For all the samples tested, the b-axis orientation was in the transverse direction. The degree of spread around the transverse direction was small. The observed b-axis orientation is consistent with the row structure model.

The infrared dichroism data are in agreement with the x-ray predictions for the *a*-axis orientation. Surprisingly, high density polyethylene seems to exhibit higher *a*-axis orientation than linear low density polyethylene, which should have the lowest stress related structure. This can be due to the increased crystallinity of the film which results in a higher overall orientation for high density polyethylene. The crystallinity of the film samples was found to remain constant regardless of the blowup ratio. All but one film exhibited a negative birefringence. For linear low and low density polyethylene, as the blowup ratio increased the birefringence increased. The birefringence data are supported by data regarding the ratio of the sonic velocity in the machine direction to the transverse direction. Birefringence and sonic velocity techniques measure the average orientation of the film. The negative birefringence results mainly from the amorphous chains orienting in the transverse direction. The crystalline contribution is low due to the random nature of the crystallites.

It was not possible to determine the orientation of the c-axis from the x-ray data. Although the a and b axes orientations are defined, one cannot deduce the c-axis orientation since we are dealing with orientation distributions. The orientation of the amorphous phase in the transverse direction at first might seem anomalous, however, if we associate the amorphous orientation with the crystallization process and consider the amorphous chains as tie molecules, it is possible for these chains to become oriented in the transverse direction [4].

The tensile data, in most cases, are consistent with the negative birefringence results, indicating transverse direction amorphous orientation. The transverse direction properties are slightly higher than the machine direction values. When the blowup ratio is less than unity, the draw ratio is high and the deformations are analogous to uniaxial drawing. This accounts for the high degree of orientation and tensile properties in the machine direction for low density polyethylene with a blowup ratio of 0.8:1. As the blowup ratio varies, however, no specific trend is observed. This is not surprising considering the complexity of the process. If the c-axis of the crystallite were oriented in the transverse direction, the differences in the machine and transverse

direction tensile results would be more significant.

The impact properties for all the resins are fairly constant. This can be misleading since the data do not take into account the thickness of the film. As the film thickness increases, the impact yield stress should become higher.

SUMMARY

The analysis of the film blowing process involves not only characterization of the flat film structure and properties, but also a knowledge of the rheological properties of the resin and thermo-mechanical history experienced by the melt.

Three sets of polyethylene were analyzed in this study. The crystallization model of Keller was used to explain the morphology of the blown films. Resin rheological properties and their dependence on temperature can be helpful in assessing the stress level during crystallization and in turn the orientation. Linear low density polyethylene, due to its strain softening behaviour exhibited the lowest stress orientation (*a*-axis parallel to the machine direction), while low density polyethylene, due to its strain hardening behaviour, exhibited an intermediate orientation (*a*-axis inclined from the plane of the film). The majority of the films exhibited a low degree of orientation.

The effect of blowup ratio on the mechanical properties of the films was evaluated. However, no simple relationship could be established between the blowup ratio and the mechanical properties.

REFERENCES

- W.F. Maddams and J.E. Preedy, J. Appl. Poly. Sci., Vol. 22, page 2721 (1978).
- W.F. Maddams and J.E. Preedy, J. Appl. Poly. Sci., Vol. 22, page 2739 (1978).
- W.F. Maddams and J.E. Preedy, J. Appl. Poly. Sci., Vol. 22, page 2751 (1978).
- 4. A. Keller, J. Poly. Sci., Vol. 25, page 31 (1955).
- 5. T.T. Li, R.J. Volungis and R.S. Stein, J. Poly. Sci., Vol. 20, page 199 (1956).
- J.T. Judge and R.S. Stein, J. Appl. Phys., Vol. 32, page 2357 (1961).
- L.A. Utracki and J. Lara, Paper presented at the International Workshop on Extensional Flow, Mulhouse-La Bresse France, Jan. 24 (1983).
- L.A. Utracki, A. Catani, J. Lara and M.R. Kamal, Paper presented at the European Meeting on Polymer Processing and Properties, Capri, June 13-16 (1983).
- 9. A. Keller and M.J. Machin, J. Macromol. Sci., Phys. Ed., Vol. 81, page 41 (1967).
- D.R. Holmes, R.G. Miller, R.P. Palmer and C.W. Bunn, Nature, Vol. 171, page 1104 (1953).
- 11. D.R. Holmes and R.P. Palmer, J. Poly. Sci., Vol. 31, page 345 (1958).
- 12. R. Farber and J. Dealy, Poly. Eng. Sci., Vol. 14, page 435 (1974).
- 13. K.J. Choi, J.E. Spruiell and J.K. White, J. Poly. Sci., Phys. Ed., Vol. 20, page 27 (1982).
- P.R. Soskey and H.H. Winter, SPE Technical Papers, Vol. 28, page 47 (1982).

<u>Resin</u>	Die Gap (mm)	Processing Temperature (°C)	Blowup Ratio	Thickness (mm)
LLDPE	2.54	190	1.3:1	0.027
			2.4:1	0.025
			2.8:1	0.017
LDPE	2.54	190	0.8:1	0.054
			1.9:1	0.026
			3.4:1	0.023
HDPE	2.54	230	2.0:1	0.023
			3.5:1	0.020
			4.6:1	0.021

TABLE C-1: Film Blowing Conditions.

		X_		Sonic Velocity (km/s)			
<u>Resin</u>	BUR	_(%)	<u>Birefringence</u>	MD	<u>TD</u>	MD/TD	
LLDPE	1.9:1	0.912	-0.75x10 ⁻³	0.83	1.01	0.82	
	2.4:1		1.99	0.82	0.95	0.86	
	2.8:1	0.917	0.00	0.84	0.72	1.20	
LDPE	0.8:1	0.918	-9.79	0.95	1.13	0.85	
	1.9:1		-8.23	0.87	1.00	0.87	
	3.4:1	0.918	-3.07	0.85	1.00	0.85	
HDPE	2.0:1	0.948	-1.61	1.49	1.67	0.89	
	3.5:1	0.947	-1.70	1.50	1.66	0.90	
	4.6:1		-2.74	1.49	1.48	1.04	

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TABLE C-2: Blown Film Properties.

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			Tensile Properties				
	BUR	Impact Yield <u>(kq)</u>	(MPa		Pa) Vie	i) Viold	
<u>Resin</u>			MD	TD	_TD_	<u>TD</u>	
LLDPE	1.3:1	2.6	123.0	143.3	10.6	10.9	
	2.4:1	2.1	113.5	137.3	9.8	10.6	
	2.8:1	3.0	63.4	86.5	5.2	7.2	
LDPE	0.8:1	4.4	187.7	164.5	14.8	9.3	
	1.9:1	3.3	100.7	126.1	13.5	8.3	
	3.4:1	3.2	130.1	148.9	9.2	8.5	
HDPE	2.0:1	3.3	388.8	422.6	18.2	21.0	
	3.5:1	1.8	499.9	456.9	22.2	27.5	
	4.6:1	2.9	277.6	396.3	18.9	26.3	

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TABLE C-3: Blown Film Mechanical Properties.

		Dichroic	X-Ray			
		Ratio	<u>a</u> -axis	•	<u>b</u> -ax	ris
<u>Resin</u>	BUR	730 cm^{-1}	MD	<u>TD</u>	TD	\underline{TD}
LLDPE	1.3:1	1.3	90	90	0	0
	2.4:1	1.4	90	90	0	0
	2.8:1	1.2	90	90	0	0
LDPE	0.8:1	1.2	45-50	45-60	0	0
	1.9:1	0.9	45	45-60	0	0
	3.4:1	1.1	70	60-70	0	0
HDPE	2.0:1	1.4	90	90	0	0
	3.5:1	1.4	90	90	0	0
	4.6:1	1.5	90	90	0	0

note: The x-ray angles are measured from the horizontal axis and is the point of maximum intensity along the diffraction ring.

TABLE C-4: Blown Film Crystal Orientation.

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Figure C-1: <u>Tubular Blown Film Process Schematic.</u>

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Figure C-2: Effect of Stress During Crystallization on Crystalline Orientation.

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Figure C-3: Elongational Viscosity Data of LDPE, HDPE and LLDPE at 150°C for Various Strain Rates.



Figure C-4: Elongational Viscosity of LDPE at Various Strain Rates and Temperatures.



Figure C-5: Elongational Viscosity of HDPE at Various Strain Rates and Temperatures.

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Appendix D RHEOLOGICAL PROPERTIES OF LDPE-1

This chapter presents a description of the linear viscoelastic rheological properties of LDPE-1. Appendix A presents a description of the experimental methods used and Appendix B presents tables containing all the experimental data.

The melt index of LDPE-1, as quoted by the manufacturer, is 1.7. The rheological properties of LDPE-1, evaluated at 150° C and 180° C, are presented in Figures D-1 to D-4.

The processing stability of LDPE-1 was assessed using a Rheometrics Mechanical Spectrometer in the time sweep mode. Thus configured, the linear viscoelastic properties of the sample in oscillatory shear were measured as a function of time, Figure D-1. While evaluating several linear low density polyethylene resins, Samurkas [183] found that the linear viscoelastic function which was most sensitive to thermal degradation was the storage modulus, G'. The criterion used to determine if the resin had started to degrade was the time at which the storage modulus was observed to vary by more than 5%. At 150°C and 180°C, the storage modulus was found to be stable up to a time of 60 minutes. The time required to conduct the experiments was always less than 60 minutes. The data presented in Figure D-1 was measured at a strain, γ , of 10% and at a frequency, ω , of 1 rad/s. The stability of the LDPE-1 at a strain of 15% was also found to be stable up to 60 minutes.

Prior to measuring the storage and loss moduli, strain sweep experiments were carried out to determine at which strain amplitudes the storage moduli of LDPE-1 began to show a dependence on the strain amplitude. The strain amplitude dependence of LDPE-1, at 150°C and 180°C, is presented in Figure D-2. The strain amplitude dependence of the storage modulus was first observed at a strain amplitude of 15%. Based on this result, in all the oscillatory shear experiments, a strain amplitude of 10% was used.

The storage and loss moduli, as a function of frequency, w, are presented in Figure D-3. In all cases, the magnitude of the storage modulus increases with increasing frequency, indicating that the elasticity of the material becomes more important. A similar trend was observed for the loss modulus data. At low frequencies, the loss modulus was found to be greater than the storage modulus. At higher frequencies however, the reverse was observed due to the predominance of elasticity. Furthermore, as the temperature increased, both moduli were found to decrease.

The steady shear viscosity of LDPE-1 is presented in Figure D-4. The steady shear viscosity was measured using a Rheometrics Mechanical Spectrometer at low shear rates, while at high shear rates the steady shear viscosity was measured using an Instron Capillary Rheometer. As observed in Figure D-4, the data measured using the two methods overlapped smoothly. LDPE-1 displayed the typical shear thinning behaviour commonly observed for polyethylene resins. The power law coefficients [182], using the Instron Capillary Rheometer data, were calculated using the following equation:

$$\eta = k \dot{\gamma}^{n-1}, \qquad (D-1)$$

where η denotes the viscosity, k a constant, $\dot{\gamma}$ the shear rate and n the power law index, and using linear regression techniques. The values of the power law index, constant and corresponding correlation coefficient, calculated at 150°C and 180°C, were as follows:

	<u> </u>	<u>k (Pa-sⁿ)</u>	R^2
150°C	0.396	13980.7	0.999
180°C	0.413	9403.7	0.999







Modulus, kPa





Figure D-3: Frequency Sweep Experiments, $\gamma = 10$ %.



Figure D-4: Shear Viscosity of LDPE-1.