# PREDICTION OF SHRINKAGE AND THE EFFECT OF HEAT TREATMENT FOR INJECTION MOLDED PLATES

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A Thesis Submitted to the Faculty of Graduate Studies and Research in Partial Fulfilment of the Requirements of the Degree of Master of Engineering

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## ABSTRACT

Injection molded parts are susceptible to shrinkage effects which undermine the strict dimensional tolerances required when they are produced. Thus, it is important to predict the conditions under which shrinkage occurs. In this work, a 3-D thermoviscoelastic stress/strain model was used to analyze shrinkage for a flat rectangular injection molded plate under varying process conditions. Initially, the pressure, temperature, crystallinity and shear rate profiles were calculated, using an existing injection molding simulation program (McKam-IV). These data were then used to conduct the stress/strain analysis and to make shrinkage predictions for a rectangular plate represented by a 10 x 10 x 10 finite volume elemental mesh. The materials used were high density polyethylene (HDPE), a semi-crystalline material and polystyrene (PS), an amorphous material. The predictions were validated experimentally using a specially designed engraved mesh mold cavity. In-plane measurements showed good quantitative agreement for both materials. Thickness measurements for PS were limited by experimental factors. The molded plates were then subjected to a simple post-process heat treatment and the simulation program was modified to assess the effects of heating. Reasonable predictions were made for HDPE, because volumetric effects could be accounted for by the simulation program. Predictions were not as good for PS, possibly because the simulation code could not account fully for the effects of frozen-in orientation effects.

## RESUMÈ

Les pièces moulées par injection sont sensibles aux effets de rétrécissement qui minent les tolérances dimensionnelles strictes exigées quand elles sont produites. Ainsi, il est important de prévoir les conditions dans lesquelles le rétrécissement se produit. Dans ce travail, un modèle en trois dimensions thermo viscoélastique de stresse et contrainte a été employé pour analyser le rétrécissement d'un plat rectangulaire plat moulé par injection sous différentes conditions. Initialement, les profils de taux de pression, de température, de cristallinité et de cisaillement ont été calculés, en utilisant un programme existant de simulation de moulage par injection (McKam-IV). Ces données ont été alors employées pour effectuer l'analyse de stresse et contrainte ainsi que pour faire des prévisions de rétrécissement pour un plat rectangulaire représenté par la maille élémentaire d'un volume 10 x 10 x 10 fini. Les matières employées étaient du polyéthylène à haute densité (HDPE), un matériel semi cristallin et du polystyrène (PS), un matériel amorphe. Les prévisions ont été validées expérimentalement en utilisant une cavité gravée conçue spécialement de moule de maille. Les mesures ont montré un bon accord quantitatif pour les deux matériaux. Les mesures d'épaisseur pour le PS ont été limitées par les facteurs expérimentaux. Les plats moulés ont été alors soumis à un simple post-traitement thermique et le programme de simulation a été modifié pour évaluer les effets de la chaleur. Des prévisions raisonnables ont été faites pour le HDPE, étant donné que des effets volumétriques pouvaient être appliqués par le programme de simulation. Les prévisions n'étaient pas aussi bonnes pour le PS, probablement parce que le code de simulation ne pouvait pas expliquer entièrement les effets de relaxation de contrainte.

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# Acknowledgments

I would like to thank my supervisor, Dr Jeremy R. Cooperstock, for the invaluable research opportunities while working in the SRE Lab. I greatly appreciated his technical guidance, editorial advice, funding and patience as I completed this thesis. This research was supported by Fonds de recherche sur la nature et les technologies, Fondation J. Armand Bombardier and Valorisation-Recherche Québec; this support is gratefully acknowledged. I would like to thank Daniel Sud for implementing the dualprojector display system used for shadow removal and for the technical discussions during the integration of our work. Thanks also to Jianfeng Yin, Stephen Spackman and Wei Sun, whose technical insights pointed me in the right direction, as well as to François Rioux for helping with the French version of the abstract.

I would also like to express my appreciation to my friends for their encouragement. Especially to Amit Sihota, many thanks for being a great friend and study buddy, and to Patrick Di Nardo, I am truly grateful to have had his unconditional support.

Last but certainly not least- Very special thanks to Christopher Beltran, whose understanding, patience and support are a blessing. To my parents, Gisela Villamor and Virgilio Hilario, and to my sister, Cynthia "Ate" Hilario: this thesis is dedicated to my family, to whom I am grateful for their care, support and patience throughout the entire course of my studies.

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## **ABBREVIATIONS**

- 3-D 3-dimensional acrylonitrile-butadiene-styrene ABS high density polyethylene HDPE Ρ presssure PBT polybutylterathalate polycarbonate PC PS polystyrene PVT pressure-volume-temperature thermo-mechanical-history TMH Т temperature time t  $T_s$ solidification temperature Τg glass transition temperature Х X-direction or flow direction or in-plane
- Y Y-direction or cross-flow direction or in-plane.

## **1 INTRODUCTION**

## 1.1 Introduction to Injection Molding

Injection molding is a versatile process used to produce finished articles ranging from utensils to bathroom fixtures to automobile parts. A polymer resin is melted and injected into a mold cavity of a particular shape to obtain the desired product upon cooling. The high startup investment costs of the injection molding systems demand high production rates, while maintaining high product quality within strict tolerances of reproducibility and performance. A key issue in the quality of the molded parts is dimensional stability, that is, the extent to which a molded part retains its intended shape and dimensions after molding and under field use conditions. During the production process, the part may undergo shrinkage and warpage, as a result of the thermomechanical history experienced during the process. Furthermore, the development of orientation and residual stresses may lead to (further) deformation. The plastics engineer strives to eliminate these sources of dimensional instability, or alternatively, to predict and control the dimensional instability such that mold designs and molding conditions can account for the future deformation of the ejected part. Prediction of the ultimate properties requires an understanding of the complex interaction of numerous factors (Figure 1.1) that govern the thermo-mechanical history (TMH) experienced by the injection molded part during the process. The TMH strongly influences microstructure development and hence, the final properties. Additionally, the molded part may undergo a post-processing operation, such as a paint-bake cycle used to cure paint onto the surface. This post-process heat-treatment may further influence dimensions and properties of the product.



Figure 1.1. Some properties and parameters influencing the characteristics of an injection molded article.

### 1.1.1 Stages of the Injection Molding Process

Four stages are typically identified in injection molding:

- Plastication: a polymer resin is fed into a screw extruder, where it is melted as it is conveyed to the barrel exit (Figure 1.2).
- Filling: when sufficient melt has accumulated at the barrel exit, the screw moves forward and forces the polymer melt into the mold cavity through the nozzle and a system of sprues, runners and gate(s) (Figure 1.3).
- Packing/holding: once all the mold cavity surfaces have been contacted by the polymer melt, the packing/holding stage begins. Additional melt is forced into the mold cavity to compensate for shrinkage of the injected polymer melt as it cools, causing a rapid increase in the pressure in the cavity, until a plateau maximum is reached.

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• Cooling/ejection: When the polymer melt at the gate is completely solidified, the mold cavity becomes sealed and pressure in the cavity decreases as the polymer continues to cool and solidify. Upon solidification to a point, where sufficient mechanical strength has been attained, the part can be ejected. It continues to cool and equilibrate to ambient conditions outside the mold.



Figure 1.2. Cross-sectional schematic drawing of an injection molding machine (1).



Figure 1.3. Cross-sectional schematic drawing of the mold assembly and mold cavity (1).

## **1.2 Dimensional Stability**

The dimensional stability of a molded part is the extent to which it retains its dimensions and shape, from the instant of ejection out of the mold to the end of its lifespan. Three phenomena contribute to dimensional instability: (i) shrinkage is a reduction in the dimensions; (ii) warpage is a distortion of the shape; and (iii) residual stresses are frozen-in internal stresses that may lead to (further) deformation and other problems. Shrinkage, residual stresses, and warpage are interrelated in a complex manner. They are often studied together to gain an understanding of the dimensional stability of the molded article.

#### 1.2.1 Shrinkage

The uncontrolled shrinkage of plastic materials is the most significant challenge in designing and manufacturing quality injection molded parts. Shrinkage is defined as a change in the dimensions of the plastic product relative to some reference dimensions, usually the dimensions of the mold cavity (1). The typical shrinkage equation in one dimension is:

$$shrinkage = \frac{(d_m - d)}{d_m} \times 100\%$$

where  $d_m$  is the reference dimension, and d is the dimension of the molded sample. Shrinkage is the industry terminology for strain, except with the opposite sign. Thus, a positive shrinkage indicates that the dimension of the part has decreased. Shrinkage will be discussed in greater detail in the next chapter.

#### 1.2.2 Residual Stresses

Throughout the injection molding process, the injected polymer is subjected to various flow stresses, which are usually relaxed in the melt phase due to the high temperature and the corresponding short relaxation time. However, upon solidification, the increased relaxation time of the material causes the stresses to become "frozen" into the material. These residual stresses may have a variety of detrimental effects, such as distorting the product shape as it ages or causing environmental stress cracking (1). Residual stresses are intimately related to shrinkage. Appendix A provides a detailed introduction, with relevant references to the topic of residual stresses.

#### 1.2.3 Warpage

Warpage is the result of differential shrinkage in the molded article. Several causes of warpage can be identified, but it is primarily due to non-uniform mold temperatures or uneven flow paths and other factors that could lead to asymmetric thermo-mechanical history in the various parts of the molded article.

### **1.3** Post-processing

Post-processing operations involving heat treatment are sometimes employed. One such operation is annealing, where the product is heated for a specified time at a temperature below or above the glass transition temperature, but below the melting temperature, such that the mechanical integrity of the part is not compromised by deformation. The operation enhances the relaxation of molecular orientation and residual stresses, and promotes development of a uniform crystallinity distribution (2). The process of annealing can add considerable cost and time to the operation, and should be avoided if possible. However, in special circumstances, a combination of particular processing conditions and annealing becomes necessary.

Unlike annealing, paint-bake cycles are not used for the purpose of relaxing stresses and orientation in the article, but instead for the purpose of curing paints (which are polymers themselves) onto the surface of the plastic surface. Paint-bake cycles are optimized empirically to reduce negative effects of the heat treatment on the molded article. They involve different phases, where the article is heated and cooled at various temperatures for different times, such as the 3 coat process from GE Plastics shown in Figure 1.4 (3). Such heating cycles will affect the ultimate shrinkage and residual stresses of the article, and the paint itself may induce additional stresses at the surface of the part.





## **2 OBJECTIVES**

The objectives of this project are listed below.

- 1. To develop methodology for assessing the ability of existing injection molding software (McKam-IV) to predict shrinkage during molding and to validate the predictions for a flat rectangular plate (101.5 x 63.5 x 3 mm) (Figure 2.1).
- 2. To study the effect of different processing conditions on shrinkage.
- 3. To predict and validate the effects of a simple post-molding heat treatment.



Figure 2.1 Sketch of the flat rectangular injection molded plate used in the current work and directional terminology.

## **3 SHRINKAGE PREDICTION AND ANALYSIS**

## 3.1 Introduction to Shrinkage

Dimensional changes occur throughout the lifetime of an injection molded part. Shrinkage development normally takes place during the following three stages (4): (i) inmold shrinkage occurs during the packing and pre-ejection cooling stages, while the part is still in the mold; (ii) as-molded shrinkage occurs just after ejection as the part cools and equilibrates to ambient conditions; and (iii) post-molding shrinkage is the long-term shrinkage that occurs because of time effects during storage, such physical aging and recrystallization. Shrinkage is manifested by volumetric and linear dimensional changes, for both amorphous and semi-crystalline polymers. Volumetric shrinkage is determined by thermodynamic pressure-volume-temperature (PVT) relationships or the equation of state and is generally isotropic. Linear shrinkage is due to shear and normal stresses acting on the injected polymer, and it is generally anisotopic.

#### 3.1.1 Volumetric Shrinkage

Volumetric shrinkage is attributed to the changes in specific volume, as the resin is first melted and pressurized, then cooled and depressurized. During these processing steps, the specific volume may vary by up to 35%. The specific volume of the polymer at any point is governed by the PVT thermodynamic relationships (Figure 3.1 for amorphous and Figure 3.2 for semi-crystalline polymers).



Figure 3.1. Experimental and fitted PVT data for polystyrene (Styron 685D) (5).



Figure 3.2. Experimental and fitted PVT data for high-density polyethylene (Sclair 2807)(5).

One can approximate the volumetric shrinkage by tracing the thermodynamic pathway of the polymer on the PVT diagram, as shown in Figure 3.3. Between A and B, molten polycarbonate at 280 °C becomes pressurized as it flows into the mold cavity at constant temperature (filling stage). From B to C, the part is held at constant pressure during the holding stage. When sufficiently packed, the gate freezes off and cooling begins. The part cools at constant volume, the volume of the cavity. During cooling, from C to D, the pressure decreases slowly, while the temperature also decreases due to cooling. When the mold is opened, D to E, the part is allowed to shrink without constraint, as the part equilibrates to ambient conditions. The change in specific volume for this process from A to E was 10.7%, while the shrinkage was the change in specific volume from D to E, approximately 2.0%. Such an approximation is crude, because it assumes no in-mold shrinkage, neglects the complex pressure and temperature fields developed in the injected polymer, and ignores the deformation response of the material to the prevailing flow and thermal stresses. For example, the polymer in immediate contact with the mold wall solidifies rapidly at a relatively low pressure, while pressure in rest of the injected polymer increases throughout filling and packing. Subsequent layers freeze under increasing pressure, resulting in a differential volumetric shrinkage profile throughout the part. Coincidently, this differential shrinkage is the main cause of thermal residual stresses (See Appendix 1).



Figure 3.3. Trace of a thermodynamic pathway an injection molded polymer typically experiences during the process.

For the purposes of modeling and prediction, various equations of state have been employed. For example, in the current work, the Tait equation is used for representing the PVT data of polystyrene (6):

$$V(p,T) = (A_0 + A_1T) \left( 1 - 0.0894 \ln \left( 1 + \frac{p}{B(T)} \right) \right)$$
(3.1)  
$$T_g(p) = T_g(0) + s_1p + s_2p^2$$

where  $V(0,T) = (A_0 + A_1T)$  is the specific volume at zero pressure in cm<sup>3</sup>/g, T<sub>g</sub>(p) is the pressure dependence of the glass transition temperature, and A<sub>0</sub>, A<sub>1</sub>, B<sub>0</sub>, B<sub>1</sub>, s<sub>1</sub> and s<sub>2</sub> are constants for the material. For high-density polyethylene, since crystallization effects are associated with the transition from melt to solid, the PVT data are best represented by (7):

$$V(p,T) = \frac{a_1}{a_4 + p} + \frac{a_2}{a_3 + p} + a_5 e^{(a_6 - a_7 P)}$$
(3.2)

where a<sub>1</sub>, a<sub>2</sub>, a<sub>3</sub>, a<sub>4</sub>, a<sub>5</sub>, a<sub>6</sub>, a<sub>7</sub> are material constants.

#### 3.1.2 Linear Shrinkage

Linear shrinkage is determined primarily by the shear and extensional stresses acting on the polymer during the molding cycle. During filling, flow stresses cause the development of orientation in the melt by stretching and aligning the coiled, entangled structures of the polymer molecules (Figure 3.4).



Figure 3.4. Randomly oriented, partially oriented, and oriented polymeric structures.

The aligned structure is frozen into surface layers, upon rapid cooling of the melt along the mold wall. The low thermal conductivity of the frozen polymer skin insulates the core. Thus, the temperature remains high in this region, allowing enough time for stretched molecules return to their entropically favoured coiled, compact state. The relaxation of orientation is associated with a reduction in dimensions, referred to as linear shrinkage. Upon solidification of the melt, any remaining orientation will continue to relax, but generally, as room temperature is approached, the relaxation time will increase, leading to a slowdown of linear shrinkage effects. Orientation and relaxation phenomena are reflected in the visco-elastic properties of the polymer. Visco-elastic behaviour is intermediate between that of a purely viscous (recovers no strain upon release of the deforming stress) and a purely elastic material (recovers all strain upon release of deforming stress). Constitutive rheological equations have been developed to describe the behaviour of the polymer melts and solids under the influence of stress fields (and changes in temperature). For example, the simplest of these equations, the 1-D Maxwell model, describes the behaviour of the polymer as a spring and dashpot (a container with viscous liquid) in series. When a force is applied to the system, irrecoverable strain is represented by the dashpot, while the recoverable strain is represented by the spring (Figure 3.5).



Figure 3.5 Simple Spring and dashpot model of the 1-D Maxwell model used to demonstrate viscoelastic behaviour.

One characteristic of visco-elastic materials is that the stress required to maintain a fixed strain decreases with time. The constitutive equation for the Maxwell model has the form:

$$\sigma(t) = \sigma_0 \exp\left(-\frac{t}{\tau}\right)$$
$$\tau = \frac{E}{\eta}$$

where  $\sigma(t)$  is stress at time t,  $\sigma_0$  is the initial stress, E is Hooke's spring constant,  $\eta$  is the viscosity of the dashpot liquid, and  $\tau$  is the relaxation time. The Maxwell model is limited, though it illustrates the significance of a key visco-elastic parameter: relaxation time. This temperature dependent parameter determines the response of the material to stresses. Many different rheological equations have been developed for polymers, each with its advantages and disadvantages (8). In this work, the Leonov model is employed, and will be further elaborated in section 3.4.4.

While the primary forces acting on the injected molten polymer are flow stresses, other forces are present, such as pressure from the mold wall and mold/wall polymer friction. These external forces arise due to the packing pressure, the physical restraint of the part by the geometry of the mold, and the friction between the mold wall and the melt. Another source of orientation is associated with thermal changes in the material. Due to the visco-elastic nature of the polymer, some orientation development may occur as it thermally expands and contracts. However, it is believed that thermally induced orientation is an order of magnitude smaller than flow-induced orientation. Thus, it is generally ignored (9).

## **3.2** Development of Orientation and Crystallinity

#### 3.2.1 Crystallinity

In certain polymers, such as polyethylene, some of the molecules can form ordered, folded structures (Figure 3.6). These regions of high order are said to be crystalline, and exhibit properties distinctive from the non-ordered or amorphous regions of the polymer.



Figure 3.6. Depiction of amorphous and crystalline regions in polymers.

In particular, the crystalline regions have a smaller specific volume than the amorphous regions. For example, note the abrupt change that occurs in specific volume, during cooling between 120°C and 150 °C, for the semi-crystalline polymer shown in Figure 3.2,. compared with the behaviour of the amorphous polymer shown in Figure 3.1. The distribution, rate, and extent of crystallization are dependent on temperature, cooling rate, pressure and shear rate. The dependence of crystallization rate, during non-isothermal crystallization, on temperature and cooling rate can be described by Nakamura's equation:

$$\chi(t) = \chi(\infty) \left( 1 - \exp\left[ -\left( \int_{0}^{t} K[T(\tau)] d\tau \right)^{m} \right] \right) (3.3)$$

where  $\chi(t)$  is the extent of crystallinity at time t,  $\chi(\infty)$  is the ultimate crystallinity, K[T] is the temperature dependent rate constant, and m is the material dependent Avrami exponent. A temperature exists at which a maximum crystallization rate occurs (Figure 3.7). At low temperature slow molecular motions inhibit formation of crystal structures, while at high temperatures too much molecular motion prevents the crystals from forming. Thus, a slow cooling rate from melt to solid enhances crystallite formation.



Figure 3.7. Typical dependence of crystallization rate on temperature.

Due to rapid cooling and solidification at the mold wall, the surface of the molded product tends to have lower crystallinity. Figure 3.8 shows the typical evolution of crystallinity in an injection molded HDPE rectangular plate. Halfway through the packing stage, crystallinity at the surface has reached its peak value of ~25%. Meanwhile, near the core, cooling is much slower and the crystallinity continues to increase until ejection to a value of ~55%.

Shutlz (10) proposed a general model of morphology in molded parts. Crystallization of fibrillar cores occurs in regions of high melt strain and rapid cooling, which is near the mold walls. Subsequently, there is lateral overgrowth on the fibrillar structures, resulting in row structures, followed by development of a transcyrstalline layer in regions adjacent to the strain induced structures. The trans-crystalline layer continues to propagate, until the undercooling of the adjacent layer becomes large enough for the nucleation and growth of spherulites, in the core region of relatively low melt strain and slow cooling. Crystallization is influenced by the shear conditions in the melt. High shear rates tend to orient and align the polymer molecules, and in this state, formation of larger spherulitic crystalline structures is favourable (11).



**Figure 3.8.** The evolution of crystallinity near the gate of an injection molded HDPE plate, as predicted by the McKAM simulation program.

#### 3.2.2 Orientation

Considering flow into a simple rectangular geometry, the molecules will tend to be oriented in the flow direction. Orientation in the molded part will vary through the thickness. For example, Mendoza, Regnier, *et al* (12) analyzed the orientation developed in a 3mm thick injection molded semi-crystalline sample. They demonstrated that four distinctive layers are observed: the skin, the shearing layer, the post-filling layer, and the core. Orientation is low in the skin layer ( $30 - 50 \mu$ m, Figure 3.9), a result of fast cooling (nearly instantaneous) of the molecules that were just submitted to an extensional deformation in the flow front due to the fountain flow. The orientation rapidly increases in the shearing layer ( $200 \mu$ m), where shearing stresses orient the molecular chains in the flow direction, and cooling is fast enough to freeze this orientation into the solid. Once filling stops, shear stresses drop rapidly, however, a second deformation field is established due to slow polymer flow from packing. Thus, in the next layer, called the post-filling layer, a second peak is sometimes observed in the orientation function (Figure

3.9). Meanwhile, crystallinity increases as small spherulites are observed in this layer. Finally, once the gate solidifies and the flow stops completely, the core layer solidifies in the center, exhibiting a low degree of orientation, but high crystallinity with large spherulitic structures.



Figure 3.9. Sketch depicting orientation through the thickness of a 3mm semi-crystalline injection molded plate (adapted from Mendoza *et. al.* (12))

## 3.3 Post-Processing

After molding, the product may undergo a post-processing operation, such as a paint-bake heat treatment to cure paint on the molded surface. There are two ways in which post-process heating may affect the sample:

 frozen-in orientation is allowed to relax, as the temperature increases (a linear effect);
recrystallization effects may change the specific volume for semi-crystalline materials (a volumetric effect) Thus, a study of post-processing heat treatment effects on a molded article is partly an investigation of the influence of orientation and crystallinity on the dimensional stability of the molded article.

Linear effects will occur if the temperature increases sufficiently to permit relaxation of residual stresses and frozen orientation. Changes in orientation of polymer samples due to heating are manifested through dimensional changes. For example, a study of chain relaxation, using small angle neutron scattering while heating semicrystalline polymers showed that the overall chain extension decreases in a manner quantitatively proportional to macroscopic shrinkage (13). For semi-crystalline polymers, it is generally believed that macroscopic length reduction is driven by relaxation of non-crystalline segments of the polymeric chains (13,14). Conversely, the heat shrinkage method could be used to estimate the degree of molecular orientation (15). In this test, sections are micro-tomed from various locations of the polymer sample. The micro-tomed sections are then heated and cooled. Biaxial dimensional changes are observed and can be related to orientation in the flow and cross-flow directions.

Studies of post-process heating of injection molded samples have shown that post-molding shrinkage is much higher in the highly orientated surface layers than in the interior. Harrell and Elleithy (16) conducted annealing experiments on PVC resins to examine the effects of injection molding conditions, and the time and temperature of exposure to heating after molding. The outer layers of the test plates were cold milled and then subjected to the same heating as the unmilled plates. The milled plates exhibited 15 - 30 % less shrinkage than the unmilled plates, allowing for the conclusion that shrinkage after heating was due primarily to the outer layers. Accordingly, the frozen-in surface orientation should be of interest in post-processing paint-bake cycles, especially for amorphous polymers.

Volumetric changes are dominant for semi-crystalline polymers, where recrystallization effects could occur upon heating. At any particular point, crystallinity changes occur according to the prevailing crystallization kinetics. Since during post-process heating, the temperature is kept below the melting temperature  $(T_m)$ , both crystalline and amorphous phases will persist. The crystallinity will increase through

lamellar thickening. Furthermore, the crystallization kinetics will vary throughout the plate, due to the already existing different crystallinity distributions. The highest crystallnity is usually obtained in the center of the plate (core). Since the outer layers of the part tend to have lower crystallinity upon leaving the mold, they will be more affected by heating than the inner layers. Consequently, post-process heat treatment will tend to enhance the development of a more uniform crystallinity in the part (Figure 3.10). Drozdov *et. al.*(17) discuss in detail the morphological changes associated with heating an injection molded sample. The specific details are beyond the scope of the current work, but their observations are consistent with the notion of a more uniform crystallinity distribution developing across the thickness.



Figure 3.10. Changes in crystallinity after heating of an HDPE injection molded semicrystalline sample, measured using DSC analysis. Adapted from White *et. al.* (18).

### 3.4 Shrinkage Prediction

The focus of the current study is on shrinkage analysis. However, shrinkage is intimately related to residual stresses, and predictive models usually deal with both The prediction of shrinkage (synonymous with strain) is two-fold: phenomena. knowledge of the material response to pressure and temperature must be combined with the material response to deformational stresses. The approach to solving this problem requires selecting a constitutive model relating the stress and strain fields. Initially, models employed a simple elastic stress/strain analysis. However, as the understanding of polymers and computational methods improved, the more sophisticated thermo-viscoelastic stress/strain models were found to be more accurate. Furthermore, analyses were extended from simple one-dimensional models to complex shapes using computer-aided design. The fundamental difference between the two approaches is that an elastic stress model is independent of the strain history, while a visco-elastic model requires knowledge of the previous deformation state to make calculations for the new strain state. In the following discussion, a 3-D thermo-elastic model is first described because: (i) of its historical importance and (ii) it will provide clarity and help to demonstrate the significance and advantages of the 3-D thermo-visco-elastic model actually used in the current work.

#### 3.4.1 Elastic Models

Theories for predicting shrinkage have progressively evolved in complexity. Isayev (19) showed that following the P-V-T diagram from the glass transition temperature to ambient conditions, an average value could be obtained for the final product volume at a certain location. However, his model neglected the thermomechanical history, and his results were limited to predictions of isotropic shrinkage values. Greener (20) predicted, with some success, density distributions using P-V-T data, by considering the continuously changing solidification conditions, due to changing pressures. Titomanlio *et.al.* (21) recognized the importance of the melt pressure and incorporated a pressure term into their elastic equations predicting residual stresses in

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injection molded parts. Significant insight and progress was made by Jansen *et. al.*(22) who derived a comprehensive elastic model using several assumptions, including: (i) continuity of stress and strain at the solid-melt interface; (ii) shear stress components were neglected in the solid phase; (iii) uniform deformation of the solidified layer; (iv) normal stress is independent of z; (v) no out of plane deformation during solidification; (vi) the solid polymer is elastic, while the melt is unable to withstand tensile stresses and flow induced stresses can be neglected; (vii) T, P, position of the solid/melt interface are known; and (viii) crystallization shrinkage and reaction shrinkage are known. Analyses performed for several polymers provided satisfactory results in predicting shrinkage and residual stresses in the molded article (23). Furthermore, Jansen made the distinction between cooling in the mold and cooling outside the mold. During the former, the part is physically restricted by the mold cavity, possibly inducing internal stresses. Outside the mold, the part is free to shrink and warp.

Jansen and co-workers (24) found that a simple thermo-elastic model could describe all experimental results for amorphous polymers, but for semi-crystalline polymers, their model over-predicted the shrinkage. The reason was attributed to inmould shrinkage for the semi-crystalline polymers, violating the basic assumption of their model of no in-mold shrinkage. If in-mold shrinkage effects do exist for a particular set of processing conditions, they must be accounted for in the model to accurately predict the shrinkage. Typically, small in-mold shrinkage may occur in product parts that solidify under low holding pressure and are not restricted by rings or flanges or a rough mold surface. This influences both the final product dimensions and the residual stress distribution.

## 3.4.2 3-D Thermo-Elastic Stress/Strain Analysis

The following thermo-elastic equations are well established and a comprehensive review can be found in numerous texts (25). Firstly, a body in equilibrium must satisfy the 3-dimensional force balance equation:
$$\frac{\partial \sigma_{ij}}{\partial x_i} + F_{bi} = 0$$
 (3.4)

where  $F_b$  is an external body force,  $\sigma$  are normal stresses for i = j and shear stresses for  $i \neq j$ , where i and j are x, y, and z directions as indicated in Figure 3.11.



Figure 3.11. Components of the stress tenor in Cartesian coordinates.

Secondly, the infinitesimal strain components for a volume element are given by:

$$\varepsilon_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$$
 (3.5)

or equivalently, in matrix form for a 3-D Cartesian coordinate system :

$$\begin{bmatrix} \boldsymbol{\varepsilon}_{xx} \\ \boldsymbol{\varepsilon}_{xx} \\ \boldsymbol{\varepsilon}_{xx} \\ \boldsymbol{\gamma}_{xy} \\ \boldsymbol{\gamma}_{yz} \\ \boldsymbol{\gamma}_{yz} \\ \boldsymbol{\gamma}_{xz} \end{bmatrix} = \begin{bmatrix} \frac{\partial}{\partial x} & 0 & 0 \\ 0 & \frac{\partial}{\partial y} & 0 \\ 0 & 0 & \frac{\partial}{\partial z} \\ \frac{\partial}{\partial y} & \frac{\partial}{\partial x} & 0 \\ 0 & \frac{\partial}{\partial z} & \frac{\partial}{\partial y} \\ 0 & \frac{\partial}{\partial z} & \frac{\partial}{\partial y} \\ \frac{\partial}{\partial z} & 0 & \frac{\partial}{\partial x} \end{bmatrix}$$
(3.6)

where  $\varepsilon$  are normal strains,  $\gamma$  are shear strains, u, v and w are displacements in the x, y and z direction, respectively Without showing the complete derivation, using the above equations, along with compatibility relationships, the stress/strain equation in the x direction is derived as:

$$\sigma_{xx} = \frac{E}{(1+\nu)(1-2\nu)} \left[ (1-\nu)\varepsilon_{xx} + \nu(\varepsilon_{yy} + \varepsilon_{zz}) \right]$$
(3.7)

where E is the Young's Modulus and v is Poisson's ratio. Similar equations are derived in the y and z directions. The shear stresses are derived in terms of the shear strains in the xy plane as:

$$\tau_{xy} = \frac{E}{1+\upsilon} \gamma_{xy} \qquad (3.8)$$

and similarly in the yz and xz directions. For elastic constitutive equations, the thermal and pressure effects are additive (25). Thus, representing the stress-strain equations in matrix form:

$$\begin{pmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sigma_{zz} \\ \tau_{xy} \\ \tau_{zx} \\ \tau_{yz} \end{pmatrix} = \frac{E}{(1+\nu)(1-2\nu)} \begin{pmatrix} 1-\nu & \nu & \nu & 0 & 0 & 0 \\ \nu & 1-\nu & \nu & 0 & 0 & 0 \\ \nu & \nu & 1-\nu & 0 & 0 & 0 \\ 0 & 0 & 0 & 1-2\nu & 0 & 0 \\ 0 & 0 & 0 & 0 & 1-2\nu & 0 \\ 0 & 0 & 0 & 0 & 1-2\nu & 0 \\ 0 & 0 & 0 & 0 & 1-2\nu \end{pmatrix} \begin{pmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \varepsilon_{zz} \\ \gamma_{xy} \\ \gamma_{zx} \\ \gamma_{yz} \end{pmatrix} - \frac{E}{(1-2\nu)} \begin{pmatrix} \varepsilon_{T} + \varepsilon_{P} \\ \varepsilon_{T} + \varepsilon_{P} \\ 0 \\ 0 \\ 0 \end{pmatrix}$$
(3.9)

where the pressure strain is defined by:

$$\Delta \varepsilon_P = \frac{(1-2\upsilon)}{E} \Delta P \qquad (3.10)$$

and the thermal strain is defined by:

$$\Delta \varepsilon_T = \alpha \Delta T \qquad (3.11)$$

The matrix equation can be summarized in the form:

$$\Delta \overline{\sigma} = \overline{\overline{D}} \cdot \Delta \overline{\varepsilon} + \Delta \overline{g} \qquad (3.12)$$

where  $\sigma$  is the stress tensor, D is the modulus matrix,  $\varepsilon$  is the displacement tensor, g is the load tensor containing the effects of temperature and pressure, and  $\Delta$  indicates the difference between two different times. Thus, a relationship between the strain (and hence shrinkage) and the forces, temperature change, and pressure change from one state to another is established.

#### 3.4.3 Visco-Elastic Models

An early theory visco-elasticy was first developed by Aggarwala and Saibel (26). Their four-parameter Maxwell model was used to calculate the residual stresses with temperature-dependent thermal expansion co-efficient. Modifications of this model by Struik (27) yielded reasonable agreement between theoretically predicted and experimentally measured surface stresses for quenched sheets of PMMA. Baaijens (28) and Douven (29) calculated the thermal stresses for PS and PC injection molded samples, using a linear thermo-viscoelastic model obtained from a linearization of the Leonov model. Zoetelife et.al. (30) used a similar formulation for calculation of thermal stresses with PS and ABS injection molded plaques. Bushko and Stokes (31) proposed a comprehensive formulation of a thermo-visco-elastic model for the solidification of a thermo-rheologically simple non-flowing visco-elastic melt, between two infinitely long parallel plates, and material properties were considered to be temperature and pressure dependent. Flow effects were neglected. A critical element of the model allowed material to be added to fill space created by the packing pressure applied during solidification. Thus, it could be used to evaluate packing pressure effects. While the results were not verified experimentally by the authors, the proposed model has served as a basis for subsequent work by others, who included flow effects. For example, Choi (32) used this 3-D thermo-visocoelastic approach to make predictions for shrinkage and residual stresses of amorphous materials. Rezayat and Stafford (33) developed a viscoelastic model, where only thickness shrinkage was considered. They suggested their model would be effective for shrinkage calculations of fibre reinforced injection molded plastics, because shrinkage is much greater perpendicular to the flow direction than in the parallel direction, where it is almost zero. A 3-D visco-elastic model for complex shapes has been proposed by Kabanemi *et. al.* (34) to predict, with some success, shrinkage for a polycarbonate box. However, in view of the complex flow analysis, simplifications in the calculation procedures were employed to save computational time, thus reducing the accuracy. Studies of visco-elastic stress/strain models for the purpose of predicting shrinkage have generally focused on amorphous materials, because the absence of crystallization phenomena simplifies the model problem. For semi-crystalline materials, few studies make direct comparisons between experimental data and 3-D thermoviscoelastic shrinkage predictions for HDPE, though some work has been done with polypropylene (35, 36, 37)

The 3-D thermo-visco-elastic model for a thermo-rheologically simple material for a flat rectangular plate used in this work is an extension of that proposed by Lai-Fook *et.al.* (38). A brief comparison is made to previous works in Table 3.1.

Author	year	Elastic (E) or Visco-Elastic (VE)	Dimension	Geometry
Titomanlio (21)	1987	Ε	1-D	Rectangular plate
Jansen (22)	1994	E	2-D	Rectangular plate
Bushko, Stokes (31)	1995	VE	3-D	Infinite parallel plates
Faroudi (39)	1997	E	2-D	Rectangular plate
Kambanemi (34)	1998	VE	3-D	Box
Choi (32)	2002	VE	3-D	Rectangular plate
Lai-Fook (38)	2002	VE	3-D	Rectangular plate

**Table 3.1**. Summary of some significant stress/strain analysis models, compared to that of the current work.

#### 3.4.4 3-D Thermo-Visco-Elastic Stress Analysis

The constitutive model used in the current work is the linearized compressible Leonov model. The thermo-visco-elastic equations used in the current analysis were first derived in 2-dimensional form by Douven (29) for the purpose of normal stress predictions across the thickness. The model was extended to 3-dimensions by Lai-Fook et. al.(38). In the derivation of the stress/strain equations, it is assumed the constitutive behaviour of the solidified polymer obeys the generalized Newtonian model:

$$\boldsymbol{\sigma} = P^h \mathbf{I} + \boldsymbol{\sigma}^d \qquad (3.13)$$

where  $\sigma$  is the Cauchy stress tensor, P<sup>h</sup> is the hydrostatic pressure, and I is the unit tensor. The final term,  $\sigma^d$ , is the dynamic or deviatoric part (flow induced stresses). It may be derived by using a multi-mode Maxwell model with a linearization of the compressible Leonov model:

$$\sigma^{d} = \sum_{i=1}^{m} 2 \int_{0}^{t} G_{i} e^{-(\xi(\tau) - \xi(\tau))/\theta_{i}} \varepsilon^{d} d\tau, \qquad \xi(\tau) = \int_{0}^{\tau} \frac{1}{a_{T}} ds \qquad (3.14)$$

where  $G_i$  is the shear modulus and  $\theta_i$  is the relaxation time of the i-th mode of the multimode Maxwell model, and  $a_T$  is the shift factor of the time-temperature superposition principle. For a thermo-rheological simple material, the shift factor is obtained from the WLF equation:

$$T \ge T_g :$$

$$\log a_T = \frac{C_1(T - T_0)}{C_2 + T + T_r}$$

$$T < T_g :$$

$$\log a_T = C_3(T - T_r)$$
(3.15)

where  $C_1$ ,  $C_2$ , and  $C_3$  are material constants,  $T_g$  is the glass transition temperature and  $T_r$  is the reference temperature from a set of master curves used to describe the modulus behaviour over a wide range of temperatures. From continuity and the effects of the rate of change of temperature and pressure on deformation, the hydrostatic pressure term may be obtained from equation (3.13):

$$P^{h} = -\frac{1}{3}tr(\sigma) = \int_{0}^{t} \frac{\alpha}{\kappa} \dot{T} - \frac{1}{\kappa}tr\left(\dot{\varepsilon}\right)d\tau \quad (3.16)$$

where  $tr(\sigma)$  is the trace of the cauchy stress tensor, T is the temperature,  $\alpha$  is the volumetric thermal expansion coefficient,  $\kappa$  is the isothermal compressibility coefficient, and  $\varepsilon$  is the linear strain tensor. For computational purposes, it is desired to express the above equations explicitly in terms of stress and strain tensors. Thus, after extensive rewriting of equation (3.13), the linear stress/strain equation is obtained in incremental form for the time interval  $\Delta t_{n+1} = t_{n+1} - t_n$ :

$$\sigma_{n+1} = \widetilde{\sigma} + \widetilde{K}tr(\Delta\varepsilon_{n+1})I + 2\widetilde{G}\Delta\varepsilon_{n+1}^{d} \qquad (3.17)$$

Where:

$$\widetilde{\sigma} = P_{n}^{h} I - \beta \Delta T_{n+1} + 2 \sum_{i=1}^{m} e^{-\Delta \xi_{n+1}/\theta_{i}} \int_{0}^{t} G_{i} e^{-(\xi(t) - \xi(\tau))/\theta_{i}} \dot{\varepsilon}^{d} d\tau \quad (3.18)$$

$$\widetilde{K} = \frac{1}{\Delta t_{n+1}} \int_{t_{n}}^{t_{n+1}} \frac{1}{\kappa} d\tau \quad (3.19)$$

$$\widetilde{G} = \frac{1}{\Delta t_{n+1}} \sum_{i=1}^{m} \int_{t_{n}}^{t_{n+1}} G_{i} e^{-(\xi(t) - \xi(\tau))/\theta_{i}} d\tau \quad (3.20)$$

$$\widetilde{\beta} = \frac{1}{\Delta t_{n+1}} \int_{t_{n}}^{t_{n+1}} \frac{\alpha}{\kappa} d\tau \quad (3.21)$$

Thus, the normal stress component in the X direction of the stress tensor may be written in incremental form as:

$$\sigma_{xx} = \sigma_{xx} + \widetilde{K} \left( \Delta \varepsilon_{xx} + \Delta \varepsilon_{yy} + \Delta \varepsilon_{zz} \right) + \widetilde{G} \left[ 2\Delta \varepsilon_{xx} - \frac{2}{3} \left( \Delta \varepsilon_{xx} + \Delta \varepsilon_{yy} + \Delta \varepsilon_{zz} \right) \right] \quad (3.22)$$

and similarly for Y and Z. The shear stress components in the xy direction are:

$$\tau_{xy} = \tau_{yx} = 2\widetilde{G}\Delta\gamma_{xy} \quad (3.23)$$

and similarly for the shear stresses in the zy and zx directions. Thus, the stress/strain equations become:

$$\begin{pmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sigma_{zz} \\ \tau_{xy} \\ \tau_{zx} \\ \tau_{yz} \end{pmatrix} = \begin{pmatrix} \tilde{a} & \tilde{b} & \tilde{b} & 0 & 0 & 0 \\ \tilde{b} & \tilde{a} & \tilde{b} & 0 & 0 & 0 \\ \tilde{b} & \tilde{b} & \tilde{a} & 0 & 0 & 0 \\ 0 & 0 & 0 & \tilde{G} & 0 & 0 \\ 0 & 0 & 0 & 0 & \tilde{G} & 0 \\ 0 & 0 & 0 & 0 & 0 & \tilde{G} \end{pmatrix} \begin{pmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \varepsilon_{zz} \\ \gamma_{xy} \\ \gamma_{xy} \\ \gamma_{zx} \\ \gamma_{yz} \end{pmatrix} - \begin{pmatrix} \tilde{\sigma}_{xx} \\ \tilde{\sigma}_{yy} \\ \tilde{\sigma}_{zz} \\ \tilde{\tau}_{xy} \\ \tilde{\tau}_{zx} \\ \tilde{\tau}_{yz} \end{pmatrix}$$
(3.24)

where:

$$\widetilde{a} = \widetilde{K} + \frac{4}{3}\widetilde{G}$$
(3.25)  
$$\widetilde{b} = \widetilde{K} - \frac{2}{3}\widetilde{G}$$
(3.26)

The stress/strain equation may be written in compact form as:

$$\sigma_{-n+1} = \mathbf{D}.\Delta\varepsilon + \mathbf{g} \qquad (3.27)$$

where  $\sigma_{n+1}$  is the stress tensor for any particular volume element at time step n+1, **D** is the modulus matrix,  $\varepsilon$  is the strain vector and **g** is the load tensor. The effects of temperature and pressure are taken into account by the thermal expansion coefficient,  $\alpha$ , and compressibility,  $\kappa$ , defined by:

$$\alpha(P,T) = \frac{1}{V} \left(\frac{\partial V}{\partial T}\right)_{P}$$
 (3.28)

$$\kappa(P,T) = \frac{1}{V} \left(\frac{\partial V}{\partial P}\right)_T$$
(3.29)

The effects of strain history are taken into account in the load vector  $\mathbf{g}$ . Thus, a relationship for stress and strain is obtained, where the components of the modulus matrix  $\mathbf{D}$  are time-, pressure-, and temperature-dependent. On the other hand, in the thermoelastic equation (3.9), the components of the modulus matrix are constant and strain history effects are not incorporated.

# 3.5 Effect of Processing Conditions on the Development of Thermo-Mechanical History

Thermo-mechanical history (TMH) refers to the transient flow, pressure, and temperature fields that the polymer experiences from the beginning of the filling stage until it has cooled to ambient conditions outside the mold. It is important to obtain a good estimate of the distributions of stresses, pressure and temperature in the polymer, because the calculation of shrinkage (strain) depends on these variables. Volumetric shrinkage is primarily dependent on pressure and temperature, while linear shrinkage is primarily dependent, though not exclusively, on the stress field. The components of TMH are highly coupled and interrelated. However, analysis of each variable individually can provide insight into the development of part microstructure, which is ultimately manifested in the bulk properties.

Processing conditions are the particular machine settings that the operator can control and manipulate during the production process. The processing conditions of most significance include: injection pressure, holding pressure, holding time, injection velocity, melt temperature and mold temperature. Among other processing parameters (Figure 1.1), these processing conditions are usually manipulated to control the thermomechanical history (TMH). One aspect of the present work attempts to evaluate the influence of injection/holding pressure and injection melt temperature on shrinkage. Thus, these two processing conditions as well as the others are discussed in the context of their influence on the development of temperature, pressure, and flow/stress gradients in the polymer, and consequently, on volumetric and linear shrinkage.

### 3.5.1 Injection/Holding Pressure, Holding time and Pressure History

Injection pressure, the pressure applied by the reciprocating screw to force the polymer melt into the mold cavity, directly influences the flow rate of the molten polymer and, consequently, the pressure fields that develop in the melt during the filling stage. Due to the high viscosity of polymer melts, injection pressure can exceed 200 MPa under some molding conditions. The development of the pressure field in the melt is quite complex. It depends on the flow rate, rheological properties and the dimensions

and geometry of the flow channel, which are constantly changing during filling. Thicker sections of the cavity promote a higher volumetric flow, and conversely thinner sections restrict flow, creating a greater pressure drop. In a flat rectangular cavity, for example, the pressure will be a maximum in the runner, with a large drop at the gate and will be atmospheric at the advancing mold front throughout the filling stage (Figure 3.12).



Figure 3.12. Typical pressure distributions in a rectangular injection molded plate during various stages of filling, as predicted by the McKAM simulation program (5).

Upon completion of filling, flow is maintained by injecting (packing) additional material into the mold cavity, although at a much slower rate. The pressure will rise in the cavity to reach a plateau maximum, known as the holding pressure (Figure 3.13). The pressure is transmitted from the incoming melt at the gate through the core and to the mold walls. The time during which the holding pressure is maintained, known as the holding time, is important. Longer holding times lead to the injection of more material into the mold cavity and a reduction of shrinkage.

Once the polymer at the gate freezes, cavity pressure starts to decay as the article continues to cool down (Figure 3.13). Depending on the mold properties, the mold cavity may itself expand as pressure rises during packing and holding. As melt pressure decays, recovery of strain in the metal mold exerts additional forces onto the injected material. The packing pressure and packing time are very important. Underpacking results in sink marks and voids in thick regions of the mold cavity, as well as excessive shrinkage. Overpacking could lead to premature mold opening (flashing), difficulties in part removal (sticking), and excessive residual stresses resulting in warpage.

The effect of processing conditions has been extensively studied, with most studies concluding that the holding pressure is the most important parameter affecting shrinkage. Holding pressure and holding time are the only two conditions for which the results are completely predictable: increasing holding pressure always decreases shrinkage, and lengthening holding time always decreases shrinkage until it reaches a constant value at gate-freeze. For example, in a recent study, Chang (40) investigated experimentally shrinkages in the flow and cross flow directions for polystyrene (Figure 3.14) and HDPE (Figure 3.15). For the amorphous material, the shrinkage decreased in the flow direction from 0.8% to 0.7% as the holding pressure was increased. For the semi-crystallline material, shrinkage decreased in the flow direction from 2.9% to 2.6% with increasing holding pressure. Other processing conditions produce variable effects in the flow and cross-flow directions for different materials.



Figure 3.13. Typical gate pressure observed during the stages of injection molding, as predicted by the McKAM simulation program.



Figure 3.14. Effects of various processing conditions on the overall shrinkage of PS in the flow and cross-flow directions (40).



Figure 3.15. Effects of processing conditions on overall shrinkage of HDPE in the flow and cross-flow directions (40).

#### 3.5.2 Injection Melt Temperature, Mold temperature and Temperature History

The temperature of the polymer melt as it enters the mold is known as the injection melt temperature. It is controlled by the heaters and cooling system surrounding the barrel of the plastication unit (Figure 1.2). The combined effects of melt temperature, material thermal conductivity, mold wall temperature, and part thickness lead to the development of thermal gradients in the polymer during the injection molding process (Figure 3.16). Both the flow rate and the pressure fields developed during filling are strongly dependent on the injection melt temperature in addition to the injection pressure, since melt viscosity is a function of melt temperature (Figure 3.17).



Figure 3.16. Typical thermal gradients through the thickness of an injection molded plate, during filling, packing, cooling, and ejection, as predicted by the McKAM simulation program.



Figure 3.17. Dependence of melt viscosity on temperature and shear rate for PS (Dow Styron 58D). Experimental data fitted using the WLF cross model (5).

On the other hand, flow influences temperature gradients by convection and viscous dissipation. Wherever flow hesitates, cooling is enhanced, while with accelerated flow, the temperature of the core material remains high. Runner and cavity geometry and dimensions are significant factors. Thin sections will cool faster and solidify at high pressure. Thus, they exhibit lower shrinkage. Thick sections will exhibit more shrinkage, since they cool more slowly and solidify at lower pressures. Non-uniformities in the thermal gradients result, if there are non-uniformities in the part wall thickness or the mold wall temperature.

The knowledge of thermal gradients is important, because they are responsible for the development of thermal strain in a particular volume element, as governed by the PVT relationships for the particular polymer. At lower temperatures, the polymer will have lower specific volume. Consequently, the temperature gradients have direct influence on the pressure fields during the holding/packing stage. At higher temperature, thermal expansion of the polymer causes increased pressurization, due to the physical restrictions of the mold cavity. Typically, a large thermal gradient occurs between the core and the surface, because the polymer in immediate contact with the mold wall solidifies rapidly to form a low thermal conductivity skin layer, while the core of the plastic is at the melt temperature (Figure 3.16). The low thermal conductivity of the polymer melt maintains a relatively slow cooling rate in the core compared to the surface.

The effect of varying melt temperature depends on the material and experimental conditions. From inspection of the PVT diagram, it is expected that higher injection melt temperature should lead to greater thermal strain and higher shrinkage. However, at low temperatures, for some systems, high viscosity and early gate freeze hinder pressure transmission in the mold cavity and reduce holding time. For example, the results of Chang (38) showed that the shrinkage, for HDPE, was a minimum at the intermediate melt temperature of 216 °C (Figure 3.15), while, for PS, the shrinkage decreased as melt temperature increased (Figure 3.14).

#### 3.5.3 Flow history

Knowledge of the flow history is important, because it is directly related to the stresses applied to the polymer melt. To correctly analyze shrinkage, it is important

understand the visco-elastic response of the melt to variations in shear and elongational stresses along its flow path in the mold, because these stresses produce orientation in the molded product and influence crystallinity development for semi-crystalline materials.

The filling stage involves non-isothermal, non-Newtonian flow of the polymer melt into a mold cavity held at a temperature below the solidification temperature of the polymer. Typically, shear rates during filling will be very high near the wall and decrease towards the center of the plate (Figure 3.18).

The viscosity of the polymer melt, the key material property controlling flow, is dependent on shear rate, temperature, and to a lesser extent, pressure (Figure 3.17). Thus, injection pressure will be the processing condition with the most influence over shear rate distributions. The shear rate distributions are also dependent on melt temperature. but to a lesser extent. As discussed in sections 3.1.2 and 3.2, the forces acting on the polymer melt lead to the development of orientation, which is related to linear shrinkage. The effects of processing conditions on shear rate development, orientation and linear shrinkage are complex and vary for amorphous and semi-crystalline polymers. In the work of Chang (40) shrinkage increased with increasing injection pressure for both HDPE and PS in the flow direction, but not in the cross-flow direction, consistent with higher shear rates in the former.



Figure 3.18. Typical shear rate distribution through the thickness during the filling stage.

The effect of melt temperature on shear rates and linear shrinkage is difficult to quantify, because melt temperature has a greater effect on volumentric changes, especially for semi-crystalline materials. For example, Velarde and Yeagley (41) used a modular mold to evaluate orientation and linear shrinkage by comparing the effects of length to width ratios of mold cavities for 2 different thicknesses and 4 different polymers. The study considered the shrinkage differences between amorphous and semicrystalline polymers, as well the shrinkage effects of process variables and wall thickness. The 2 mm thick cavity resulted in ~15% less shrinkage, for semi-crystalline materials than for a 3 mm thick wall cavity, despite an increase in shear stresses. The thinner cavity resulted in faster cooling and less crystallization, and hence less volumetric The contribution of flow deformation to the shrinkage of injection molded shrinkage. PVC plates was analyzed by Harrell et. al. (16), using two types of resins: one low flow resin and another with lower viscosity. Mendoza, Regnier, et. al. (12) used mold cavities of varying thickness. While the thickness of the shear layers was almost the same, the chain orientation in the 1 mm molding was twice that in the 3 mm plate. There was a small influence of injection speed on the maximum of the chain orientation, when speed was tripled. This suggests that part thickness is a critical parameter that governs the global level of crystallization. In other words, cooling rate was the most influential parameter on the flow induced crystallization in the injection molding of semi-crystalline polymers. These results lead to the conclusion that volumetric effects will be dominant for semi-crystalline materials, and linear effects will be significant for amorphous materials.

Finally, a few additional notes on viscosity and flow fields. Polymer melts are usually shear-thinning, non-Newtonian fluids. Cooling rates are relatively slow in the core, due to the insulating frozen skin layer formed at the mold wall (Figure 3.19). As a consequence, during filling, melt viscosity remains rather low, maintaining a fast flow. Additionally, the frozen layer thickness contributes to a smaller cross-sectional area, increasing the velocity. High shear rates associated with increased speeds will cause frictional heating, further reducing the viscosity. These effects are illustrated by considering the pressure-fill time curve for a thermoplastic, in comparison with a common low viscosity Newtonian fluid, such as oil (Figure 3.20). For fast filling times, high pressure is required for both oil and the thermoplastic. As the filling time is decreased, the pressure also decreases for both materials. However, for the thermoplastic, the pressure will reach a minimum and subsequently increase, since the viscosity rises with slower flow.

A phenomenon of great importance during filling is fountain flow (42,43), a mechanism used to explain advancement of the polymer front. The polymer melt, in the front region, spreads toward the walls (Figure 3.19) as is it advances in the mold cavity during filling. Fountain flow is known to influence the part microstructure. Mavridis *et.al.*,(44), discussed effects of fountain flow on orientation in great detail, and demonstrated that orientation in the skin layer near the wall is produced during the moldfilling stage. Long, thin cavities will be less affected by fountain flow than thicker, shorter cavities.



Figure 3.19. Fountain flow and formation of a skin layer in the as the polymer melt advances in the runner system during the filling stage.



Figure 3.20. Typical filling pressure vs. filling time curves for a thermoplastic and a Newtonian fluid such as oil.

## **3.6 Computational Aspects**

### 3.6.1 Characterizing and Modeling the Injection Molding Process.

Prediction of shrinkage first requires a suitable simulation of the injection molding process that can predict the transient thermo-mechanical history experienced by the material under specified processing conditions. Developments in computer technology over the last 25 years have made it possible to obtain numerical solutions to many complex physical problems, such as the injection molding process. The details of the McKAM-4 injection molding simulation program used for the current work are discussed extensively in the literature (5,45). Briefly summarizing, the McKAM simulation program provides a complete thermo-mechanical analysis of a planar (in this case rectangular) injection molded plate. The modeling assumptions of the program describe the injection molding process in terms of the equations of conservation of mass, momentum, and energy, coupled with thermodynamic relationships, constitutive equations, crystallization models, and appropriate assumptions and boundary conditions. The flat shape is studied because it allows for simplifying assumptions when modelling the flow behaviour and hence, faster computational times. Furthermore, the absence of complex flow fields allows the researcher to focus on the effects of processing conditions.

Solutions for melt flow are calculated using a moving nodal finite volume mesh to represent the advancing melt front. The melt flow has two components. Firstly, there is flow through the melt delivery system, consisting of the runner and gate, and secondly, there is an abrupt change in flow path dimensions for flow into the mold cavity. The noslip condition is employed along the runner/mold wall boundary and fountain flow effects are incorporated, adding mathematical complexities to the flow equations. When the melt front reaches the end of the mold, the nodal mesh becomes stationary, signifying the start of the packing stage. The simulation assumes that the melt temperature and nozzle pressure are known at the start of filling, time =  $t_0$ . These are user specified parameters. The program outputs nodal values of temperature, pressure, crystallinity, and shear rate at time  $t_n$  for a nodal mesh covering the rectangular plate of 31 x 21 x 11 (55 x 21 x 11, including the runner) (Figure 3.21). The time step of the calculation,  $\Delta t = t_{n+1}-t_n$ , is user

specified and is typically 0.01 for a total time injection molding cycle of 60 s, to produce the best combination of solution accuracy with computational time. Thus, the transient thermo-mechanical profile at 7161 nodal points throughout the rectangular plate can be predicted. The predicted results for pressure (eg. Figure 3.13), temperature (eg. Figure 3.16), crystallinity (Figure 3.8), and shear rates (Figure 3.18) are then used by the simultaneously running stress/strain calculations to solve for stresses and strains in a particular volume element (Figure 3.22).



**Figure 3.21**. Top and side views of the finite elements nodal mesh generated by the McKAM simulation program. IX, IY, IZ are increasing nodal mesh values in the X, Y, and Z directions (Figure 2.1), respectively.



Figure 3.22. Schematic depicting inputs and outputs of the parallel running programs of the McKAM simulation program.

#### 3.6.2 Numerical Solution of Stress/Strain Equations.

The following numerical procedure was used to solve the 3-D thermo-viscoelastic stress/strain equations. Equation (3.6) is written in vector form as:

$$\Delta \boldsymbol{\varepsilon} = \mathbf{A} \cdot \Delta \mathbf{d} \qquad (3.30)$$

where d is the displacement vector, and A is the matrix of components of derivatives relating the strain and displacement. The force equilibrium equation (3.4) can be expressed as:

$$A^T \cdot \Delta \boldsymbol{\sigma} + F = 0 \qquad \textbf{(3.31)}$$

Substituting the stress equation (3.27):

$$A^{T} \cdot [D \cdot \Delta \varepsilon + g] + F = 0 \qquad (3.32)$$

Finally eliminating the strain by substituting equation (3.30) into equation (3.32), equation (3.33) is obtained in terms of displacements, u, v, w (see equation (3.6)):

$$(A^T \cdot D \cdot A) \cdot \Delta d = -F - A^T \cdot g$$
 (3.33)

Application of the Galerkin finite element method to solve the partial differential equations (3.33) numerically involves discretizing the equations over each element using shape functions [N], with iso-parametric normalized eight-noded brick-shaped elements (with local coordinates of each element,  $\xi$ ,  $\eta$ ,  $\zeta$ , varying form –1 on one face to + 1 on the opposite face), leading to the stiffness equations for a typical element as follow:

$$\left[K^{n}\right]\Delta d = -\int_{V} \left[B^{n}\right]^{T} g dV - \Delta F^{n} \qquad (3.34)$$

where subscript (n) denotes a time dependence, dV is the element volume,  $[B^n]$  is the matrix of components of the shape function derivatives [A.N.], and the initial stiffness matrix is:

$$\begin{bmatrix} K^n \end{bmatrix} = \int_V \begin{bmatrix} B^n \end{bmatrix}^T \begin{bmatrix} D^n \end{bmatrix} B^n dV \qquad (3.35)$$

Then, applying relevant boundary conditions, performing numerical integration using Gaussian quadrature, assembling the local stiffness matrices into a global stiffness matrix, and using the Gaussian elimination procedure on the global stiffness matrix and load vector, the displacements are calculated at time step,  $\Delta t$ . For each time interval, incremental displacements were determined, and the resulting stresses and strains were recovered via the matrix-vector equations at the centroid of each element.

#### 3.6.3 Boundary Conditions and User Specified Parameters.

The McKAM simulation program is highly complex document, containing over 300 pages of FORTRAN computer code and various boundary conditions to solve the coupled differential equations. While most of the code is fixed, there are specific boundary conditions and specified parameters the user can manipulate to simulate different experimental conditions. These user-specified processing conditions include mold wall temperature ( $T_{mold}$ ), melt temperature ( $T_{melt}$ ), nozzle pressure ( $P_{nozzle}$ ), and injection velocity ( $U_{ram}$ ). Details of these boundary conditions are summarized in Table 3.2. The program uses  $T_{mold}$  and  $T_{melt}$  as a basis to compute the temperature distributions,  $P_{nozzle}$  to compute pressure distributions during packing, and  $U_{ram}$  to compute pressure during filling. The program is designed such that values for these parameters directly correlate to experimental machine settings.

Secondly, the displacement boundary conditions must be defined for the three normal components of displacement, u, v, w (equation (3.6)). A constraining type of condition on the displacement is used, while the product is still in the mold, because it is

assumed that the product is physically restricted by the mold walls. Such an assumption is implemented by defining the displacement to be zero at the boundary nodes:

$$u = v = w = 0$$
 (3.36)

where u, v, and w are the displacements from equation (3.6). This assumption has practical value, since knowledge of the boundary nodes decreases the number of unknowns in the system, decreasing computational time. The solution procedure prevents these components from ever being assembled into the stiffness matrices of the final system, and only non-zero nodal values are calculated. The second possibility is a free displacement boundary condition:

 $\mathbf{u} \neq \mathbf{v} \neq \mathbf{w} \neq \mathbf{0}$  (3.37)

The free displacement boundary condition is required when the product is outside the mold, since it is no longer physically restricted to move. The time at which the free displacement boundary condition is applied is of critical importance prediction of stresses and strains in a plastic sample. For instance, consider the effect on residual stresses for free quenching conditions verses injection molding conditions (Appendix A). For free quenching, the surface of the material is allowed to move throughout cooling, resulting in a parabolic stress profile. The stresses are tensile towards the center and compressive towards the surface. For injection molding conditions, internal melt pressure and mechanical constraints of the mold result in a typical stress distribution with tensile stresses at the surface of the sample, then a region of compressive stresses, followed by tensile stresses in the core. The effect of the displacement boundary condition on shrinkage predictions will be further discussed in section 5.1.2.

Boundary	Location where	Time at which	Typical Values
Condition	it is defined	it is defined	
T <sub>mold</sub>	All boundary	All timesteps	298 °C
	nodes, eg. IX =		
	0, 31, IY = 0,		
	21, and $IZ = 11$		
	(Figure 3.21)		
T <sub>melt</sub>	Nozzle	$\mathbf{t} = 0 \mathbf{s}$	180 °C to 220
			°C, and 220 °C
			to 260 °C, for
			HDPE and PS,
			respectively
P <sub>nozzle</sub>	Nozzle	End of filling	20 MPa – 40
		-	MPa
U <sub>ram</sub>	Nozzle	T = 0 s	1.0e-3
			(dimensionless)

Table 3.2. User-specified processing conditions in McKAM.

# 3.7 Modification of the Simulation Model.

#### 3.7.1 Post-Process Heat Treatment

To simulate the heating process, temperature boundary conditions were manipulated. The parameter  $T_{mold}$  was changed from 298 °C to 363 °C at time  $t_1$  after the injection molding cycle was complete and the part had cooled to room temperature. At time  $t_2$ ,  $T_{mold}$  was changed back to 298 °C, analogous to instantly removing the part from the heating oven. During the heating phase,  $t_2 - t_1$ , the size of the timestep was increased to allow for larger heating times (up to 30 min) without needing unreasonably long computational times.

Secondly, analysis of the post-process heat treatment required a change in the displacement boundary conditions. Once removed from the mold, the part was no longer physically restricted to move by the mold walls. The post-process heat treatment analysis was therefore analogous to free quenching, and it was necessary before time  $t_1$  to change from the constrained boundary condition to the free displacement boundary condition (equation 3.37).

#### 3.7.2 Melt Strain

The simulation model presented was previously used strictly for the purpose of predicting residual stresses (38). A crucial assumption for calculating residual stresses is that the stationary polymer melt can not sustain any developed stresses (for example, see section 3.4.1, assumption 6 from Jansen (23)). Such an assumption is implemented numerically through the use of a solidification temperature, a no-flow temperature at which the polymer was deemed to have the mechanical strength to sustain stresses. For PS and HDPE,  $T_s = 90$  °C and 100 °C, respectively. Computationally,  $T_s$  is implemented by setting all modulus values of the symmetric matrix  $\mathbf{D}$  (equation (3.24)) to a value of zero until the particular element has cooled to the solidification temperature. Thus, both the stresses and strains are calculated with respect to this initial stress-free state of the solidified polymer and not the dimensions of the mold. This procedure is required for calculating stresses, since inclusion of stresses in the melt would erroneously increase the predicted values. However, the polymer melt is actually contracting volumetrically and linearly throughout the filling and packing stage. A more realistic assessment of strain in reference to the mold dimensions would consider melt strain effects which could be as great as 15-20% simply from looking at the PVT diagrams. The calculation procedure is modified to include melt strain effects by eliminating the usage of the solidification temperature, such that the stress/strain calculations start immediately upon the completion of filling for all volume elements. This also negates the arbitrary usage of solidification temperature, which is incorrectly assumed to be constant and independent of pressure. Such a procedure is acceptable provided that the time-temperature shift function appropriately accounted for the transition from liquid to solid state.

#### 3.7.3 Strain History Implementation

A key component of the visco-elastic simulation is the strain history. The original simulation model used the following to implement the strain rate at time step  $t_n$ :

$$\dot{\varepsilon}^{a} = shearrate_{n} - shearrate_{n-1}$$
 (3.38)

where  $\dot{\varepsilon}^{d}$  is the strain rate used for equation (3.18) and shearrate is the nodal shear rate value computed by the injection molding simulation. The consequence of this implementation of the strain history is that the shear rate decreased rapidly at the end of

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the filling stage once flow had stopped, and is zero for any solidified polymer. Such a formulation is essentially a pseudo-elastic calculation since the strain history is negated and the load vector  $\mathbf{g}$  (equation (3.24)) is nullified in the solid phase. The implementation of the term  $\dot{\varepsilon}^d$  is reformulated to:

$$\dot{\varepsilon}^{d} = \frac{\varepsilon_{n-1}}{t_n - t_{n-1}}$$
 (3.39)

where  $\varepsilon_{n-1}$  was the strain from the previous timestep as computed.

Model	Melt Strain	Strain History	
Original	No	Equation (3.38)	
Modified	Yes	Equation (3.39)	

 Table 3.3.
 Summary of modifications to simulation program.

# **4 EXPERIMENTAL**

## 4.1 **Processing and Production of Plates**

#### 4.1.1 Materials

The materials used in the current work were semi-crystalline high-density polyethylene (HDPE; Sclair 2907), and amorphous polystyrene (PS; Dow Styron 685D). All relevant material properties for experimental work and process modeling, including pressure-volume-temperature (PVT), viscosity, thermal conductivity and crystallization data are summarized in previous works (5,38).

#### 4.1.2 Injection Molding Equipment and Procedure

Rectangular plates were molded using a 60-ton Danson Metalmec reciprocating screw injection molding machine. The mold assembly employed a cold runner system. Mold cavity dimensions were 101.5 mm x 63.5 mm. x 3 mm. A 10 x 10 mesh was engraved into the mold cavity with line width of 0.025 mm and a depth of 0.025 mm (Figure 4.1), such that the mesh markings would protrude from the surface of the finished product. These markings allow the measurement of the incremental shrinkage profile across the X and Y directions.



Figure 4.1. Photo of the engraved mesh in the mold cavity.

At least 30 parts were produced for each set of processing conditions, of which 5 plates were randomly selected after the first 10 were produced to assure that steady state conditions were attained at the barrel exit. The only variable processing conditions were holding pressure and injection melt temperature. Sensors were installed at appropriate places around the nozzle and gate to monitor these variables. The choice of holding pressures was dictated by the physical limitations imposed by the equipment. Holding pressures below 20 MPa resulted in short shots, while pressures above 40 MPa resulted in excessive flashing. The mold temperature was constant at 28 °C. The parts were ejected and allowed to equilibrate at ambient conditions for 24 hours before shrinkage measurements.

### 4.1.3 Post-Process Heat Treatment

The molded samples were subjected to a simple post-molding heat treatment by placing them in a conventional oven at 90 °C for 30 min. The samples were removed and allowed to equilibrate to ambient conditions for 24 hrs before measurements were made again.

## 4.2 Shrinkage Characterization

The standard method for measuring shrinkage is ASTM D955-89. The measurements were made in the X and Y directions by using a Nikon traveling microscope to measure, element by element, dimensions of the surface mesh markings to establish a shrinkage profile in the X and Y directions.

Overall X and Y shrinkage measurements were made with standard measurement calipers and corroborated with the sum of the elemental measurements. The overall thickness measurements were made using a digital caliper at various locations on the molded plates, and the data were compiled using an Excel spreadsheet.



Figure 4.2. Schematic of molded plate indicting convention for directional terminology.

Experimentally, shrinkage was defined relative to the mold and engraved mesh dimensions according to the following equation:

$$shrinkage = \frac{(d_m - d)}{d_m} \times 100\%$$
 (4.1)

where  $d_m$  was the dimension of the mold or elemental mesh and d was the dimension of the part. For this study, shrinkage after post-process heating was still defined relative to the original dimensions of the mold, because simulation results were defined in this way as well.

# **5 RESULTS AND DISCUSSION**

## 5.1 Development of Simulation Program for Predicting Strains

The first objective was to assess the quality of the predictions for shrinkage and analyze reasons for discrepancies. The analysis involved the assessment of three issues: (i) modeling factors: how the model equations and assumptions affect the results; (ii) computational factors: how the computational method affected the results (eg. numerical implementation of boundary conditions); and (iii) experimental factors: consideration of errors in measurement of experimental variables (eg. temperature and pressure) and errors in dimensional measurements of the sample itself.

#### 5.1.1 In-Plane

The simulation program yielded average stress and strain values in the X, Y, and Z directions (Figure 4.2) for all volume elements for the 10 x 10 x 10 elemental mesh (Figure 3.21) representing the rectangular plate. Initially, the predictions of the original simulation program were compared against those of the modified program (Table 3.3) for the in-plane directions (X, Y). In some cases, the original simulation produced somewhat erroneous, and erratic curves in both the Y and X directions for PS (Figure 5.1). For example, at the surface (Z= 1, Figure 5.1), the shrinkage varied from -175% to 75%. This was attributed to computational irregularities resulting from the arbitrary usage of melt solidification temperature T<sub>s</sub>. The finite elements solution procedure involved a series of iteration loops. As a particular element temperature approached the specified T<sub>s</sub> (the temperature at which the stress/strain calculations were started) in a particular element, the elemental temperature was above T<sub>s</sub> in one iteration loop and below it in another loop, producing in some cases, an erroneous solution. For the case of the modified simulation model, such irregularities due to the discontinuity of arbitrary T<sub>s</sub> were

eliminated (Figure 5.2). The curves became much smoother and the results more reasonable, varying from 0.5% shrinkage in the middle of the plate to  $\sim 1\%$  shrinkage at the surface.

Secondly, it was important to determine how the simulation results may be related and compared to the experimental results, since in the X and Y directions strain profiles were available at 10 different depths from the surface to the middle of the plate. Physically, it was only possible to determine the strain profile across the surface of the plate. Upon inspection, it was elucidated that the average of the shrinkage predictions at the 10 different depths (ie. from the middle of the plate, Z = 1 to the surface of the plate Z = 10) from the modified simulation program produced the most accurate predictions compared with experimental results (Figure 5.3). Therefore, from here on, all simulated in-plane shrinkage predictions that were presented were thickness averaged X and Y shrinkages.

Similar results were obtained for HDPE (Figure 5.4 - Figure 5.6). The processing conditions for all figures in section 5.1 are summarized in Table 5.1.

Figure(s)	Holding	Injection Melt	Holding Time	Mold
	Pressure	Temperature	_	Temperature
5.1, 5.2, 5.3, 5.10, 5.14	30 MPa	200 °C	30 s	28 °C
5.4, 5.5, 5.6, 5.11, 5.15	30 MPa	240 °C	30 s	28 °C

 Table 5.1. Summary of processing conditions used for figures in section 4.1.



Figure 5.1. Simulated shrinkage profiles for PS at various depths from the middle of the plate (Z = 1) to the surface of the plate (Z = 10) (a) across the Y direction and (b) along the X direction, using the original simulation program.



Figure 5.2. Simulated shrinkage profiles for PS at various depths from the middle of the plate (Z = 1) to the surface of the plate (Z = 10) (a) across the Y direction and (b) along the X direction, using the modified simulation program (see Table 3.3).



Figure 5.3. Thickness averaged simulation profiles for PS in the (a) Y direction and (b) X direction.



Figure 5.4. Simulated shrinkage profiles for HDPE at various depths from the middle of the plate (Z = 1) to the surface of the plate (Z = 10) (a) across the Y direction and (b) along the X direction, using the original simulation program



Figure 5.5 Simulated shrinkage profiles for HDPE at various depths from the middle of the plate (Z = 1) to the surface of the plate (Z = 10) (a) across the Y direction and (b) along the X direction, using the modified simulation program



Figure 5.6. Thickness averaged simulation profiles for HDPE in the (a) Y direction and (b) X direction, using the original and modified simulations, compared with experimental shrinkage profile.
#### 5.1.2 Effect of Displacement Boundary Condition on In-Plane Shrinkage

For the most accurate results, one must assess the possibility of in-mold shrinkage for a particular system. Considering the simulated temperature and pressure history for one injection molding cycle (Figure 5.7), there were two possible times to implement the free displacement boundary condition (section 3.6.3). The first possibility was at the end of the molding cycle, when the product was ejected from the mold (time = 60 s, location B, Figure 5.7). This formulation assumed no in-mold shrinkage. That is, as long as the product was within the physical restrictions of the mold cavity it was not allowed to freely deform. The second possibility was to implement the free displacement boundary condition at 32 seconds (location A, Figure 5.7), when the pressure gradients throughout the molding had reached zero. Such a formulation assumed in-mold shrinkage was possible. Since the temperature at point B was still well above room temperature (~70 °C), there would be prediction of greater shrinkage due to the free movement of the boundary nodes resulting from thermal contraction of cooling. Inspection of the experimental shrinkage profile (Figure 5.8) showed that in-mold shrinkage effects were significant. In the vicinity of the edges, the local shrinkage was sometimes twice the shrinkage near the middle of the plate. Implementation of the free displacement boundary condition B produced a "better shaped" shrinkage profile and usually more accurate results for HDPE, particularly towards the edges, where results were largely underpredicted at the edges using boundary condition A. Considering the overall shrinkages for this case, condition B resulted in 3.40% and 3.04% and condition A resulted in 3.14% and 2.91%, compared with experimental values of 3.70% and 2.58%, in the Y and X directions, respectively (Table 5.2). The predicted in-mold shrinkage, the difference between overall shrinkage for conditions A and B, was 0.26% in the Y direction and 0.13% in the X direction (Table 5.2). For this case there was drastic improvement in the Y-Shrinkage prediction, but not in the X –direction. Inspection of all results found that the free displacement boundary condition B were generally more accurate for HDPE in both the flow and cross-flow directions. Consequently, free-displacement boundary condition B was used throughout the current work. The change in free displacement boundary condition did not have any significant effect for shrinkage predictions of PS.

Direction	Experimental Overall Shrinkage (%)	Simulated Shrinkage (%) (condition A)	Simulated Shrinkage (%) (condition B)	Predicted In- Mold Shrinkage (%) (B - A)
Y	3.6980	3.1374	3.4033	0.2609
Х	2.5830	2.9180	3.0480	0.1300

**Table 5.2.** Overall shrinkages for free-displacement boundary conditions A and B forHDPE molded plates with the processing conditions in Table 5.1.



**Figure 5.7**. Simulated evolution of holding pressure and temperature near the gate for HDPE injection molded plates molded under the conditions specified in Table 4.1.



Figure 5.8. Effect of displacement boundary condition on simulated shrinkage profile for HDPE in the (a) y-direction and (b) x direction.

#### 5.1.3 Thickness

The simulation program yielded stress and strain values in the Z direction (Figure 4.2) for all volume elements for the  $10 \times 10 \times 10$  elemental mesh (Figure 3.21). Thus, the Z shrinkage profile could be established for any location in the plate (constant X and Y). Generally, the predicted Z shrinkage profiles indicated that shrinkage was the greatest towards the center of the plate (core) and decreased towards the surface, sometimes even becoming negative. At a location near the gate, for example (Figure 5.9), the shrinkage for PS was predicted to be ~0.8% toward the center and decreased to ~0.3% towards the outer edge, while for HDPE, the predicted shrinkage was large at the center (~3.5%) and increased to negative values near the surface. High shrinkage in the core was consistent with high crystallinity development.



Figure 5.9. Simulated shrinkage profiles in the Z direction near the gate for PS and HDPE.

However, these thickness shrinkage profiles could not be validated, since it was not physically possible to measure the strain profiles for a plate thickness of 3 mm. Experimentally, only overall thickness values were attainable. Thus, only the integrated Z strain profile, representing the overall thickness shrinkage values, will be presented hereafter. These overall thickness predictions were mapped across the Y direction and along X direction for PS (Figure 5.10) and HDPE (Figure 5.11). The discussion of the two simulation models (Table 3.3) applies to the thickness shrinkage regarding the modified simulation model (section 5.1.1). The simulated results of the modified model did not seem to show improved agreement with the experimental results over those obtained with the original simulation program. This is probably due to experimental limitations, rather than to the relative accuracy of either of the simulation models. The experimental limitations for thickness shrinkages will be discussed in section 5.2.2.



**Figure 5.10**. Simulated overall thickness shrinkage profile for PS (a) across the Y direction and (b) along the X direction, using the original and modified simulation.



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Figure 5.11. Simulated overall thickness shrinkage profile for HDPE (a) across the Y direction and (b) along the X direction, using the original and modified simulation.

# 5.1.4 General Comments on The Experimental Results and Error

A significant challenge in this work was compiling the results and presenting them concisely because of the large amount of data available. For each plate, measurements could be made for every rectangle of the 10 x 10 rectangular engraved mesh surface, both in the X and Y direction, for a total of 200 measurements. Thus, shrinkage profiles over the entire plate were established for PS (Figure 5.12) and HDPE (Figure 5.13). After inspection, it was decided that such a complete analysis for every plate would be cumbersome and unnecessary. A good representation of shrinkages throughout the plates was obtained by using just the measurements in the X direction along the plane Y = 3 and, in the Y direction, the plane X = 7. Defects in the molded plates due to various markings (eg. temperature and pressure transducers, ejection pin marks, gate markings) were minimal at these locations, thus improving the reproducibility of the results. Thirdly, for the Y measurements, the results would relate directly to the simulated predictions, because the curvilinear finite-element mesh used in the simulation "became straight" at the plane X = 7 (Figure 3.21). Thus, from here on, all results presented in the X direction are along the plane Y = 3 and all results in the Y direction are across the plane X = 7.

Significant errors were present in the measurement of melt temperature. A temperature thermocouple was located at the exit of the injection unit, but it was estimated that the error in these readings was  $\pm 5$  °C. Pressure measurements were estimated to be  $\pm 3$  MPa. The experimental pressure profiles corresponded very well to the simulated pressure profiles (Figure 5.14), consistent with the work of Hernandez-Augilar (5) and Lai-Fook (38). Measurements of the shrinkage profiles resulted in errors not only from the limitations on the reproducibility of the plates, but also due to human error measurements of these properties within making measurements using the travelling base microscope. Measurements for overall shrinkages were more accurate because the use of callipers allowed for correlation with the integrated shrinkage profiles. Estimated relative measurement errors were summarized in Table 5.3.



Figure 5.12. Experimental shrinkages profiles measured across the surface of PS molded plates in the (a) Y direction and (b) X direction, for the processing conditions summarized in Table 5.1.



Figure 5.13. Experimental shrinkages profiles measured across the surface of HDPE molded plates in the (a) Y direction and (b) X direction, for the processing conditions summarized in Table 5.1



Figure 5.14. Simulated and experimental pressure history for (a) PS and (b) HDPE at the nozzle and gate, using the processing conditions summarized in Table 5.1.

Measurement	Estimated Error	
X – profile	5%	
Y – profile	7%	
X – overall	4%	
Y – overall	4%	
Thickness (Z overall)	6%	

Table 5.3. Summary of measurement errors in various shrinkage measurements.

## 5.1.5 Residual stresses

The effect of modifications to the simulation program (Table 3.3) on residual stress predictions was briefly investigated. Predictions of the modified simulation program were compared to experimental results and original simulation predictions from previous work (Lai-Fook et.al (38)), at a position near the gate for PS (Figure 5.15) and HDPE (Figure 5.16). Stress predictions from the modified program were over-predicted towards the core (z/b = 0) by ~3 MPa and ~7 MPa, for PS and HDPE, respectively. Towards the surface (z/b = 1), the stresses were predicted to become compressive, similar to free-quenching (Appendix A), in contrast with the original simulation which more correctly predicted surface stresses to become tensile. The discrepancy in stress prediction between the two programs was because of the addition of melt strain effects (section 3.7.2). During calculations in the melt phase, the modulus component  $\widetilde{G}$ (equation 3.20) was likely very small, however the pressure component  $\widetilde{K}$  (equation 3.21) was very large due to the high pressures, thus, when explicitly calculating stresses using equation 3.27, erroneous values were calculated in the melt. The basic assumption of residual stress analysis that the melt phase was unable to sustain any developed stresses was violated.

The work of Lai-Fook, however, considered only through thickness stresses and presented experimental results for one position near the gate. The current work was more comprehensive, as it considered in-plane and thickness shrinkages throughout the plate. Thus, the validation of the residual stress predictions, using the modified simulation, required the measurement of stress profiles throughout the plate in all directions. However, such a stress analysis would require extensive complicated measurements that were outside the scope of the present study. It appears, however, that the modified model would be better for predicting shrinkage, while the original simulation would give better predictions of residual stresses. Thus, it is suggested that accurate stress/strain analysis of the injection molded plates might involve two sets of calculations: one where melt strain is not calculated, providing more accurate stress predictions, while in the other, melt strain is calculated, providing more accurate results for the overall strain with respect to the dimensions of the mold cavity.



Figure 5.15. Residual stresses near the gate for the PS (processing conditions in Table 5.1.)



Figure 5.16. Residual stresses near the gate for HDPE.

## 5.2 Effect of Holding Pressure

## 5.2.1 In-Plane

#### 5.2.1.1 Experimental Results

Holding pressure was increased successively from 20 MPa to 40 MPa for PS and HDPE at injection melt temperatures of 240 °C and 200 °C, respectively. For the amorphous material, overall shrinkage decreased from 0.82% to 0.56% in the Y direction and 1.23% to 0.67% in the X direction as the holding pressure was increased from 20 MPa to 40 MPa (Table 5.4). For HDPE, overall shrinkage decreased from 4.15% to 2.65% in the Y direction and from 3.8% to 2.37% in the X direction, as the holding pressure was increased from 20 MPa to 40 MPa. The results were in agreement with literature values for moldings of similar geometry, molded under similar conditions. For example, typical shrinkages measured by Jansen *et.al* (24) were in the range 0.8% to 0.35% for PS and 3 to 1.8%, as the packing pressure was increased. Chang (40) reported similar findings (Figure 3.14 and Figure 3.15). It is important to note that the different injection molding systems and machine were not identical, making direct comparisons difficult.

Examination of the shrinkage profiles (Figure 5.18 for PS, Figure 5.19 for HDPE) revealed that shrinkage was greatest near the edges, sometimes more than double that in the vicinity of the middle of the plate. As pressure was increased, the discrepancy between the edges and the middle of the plate tended to decrease, consistent with the decrease of in-mold shrinkage at higher pressures. Also, for the Y direction, shrinkage was consistently greater on the left side than on the right side. Two possible factors may cause such differences. Firstly, the cooling system of the mold assembly was asymmetric; cold water inlets were on the left side of the mold assembly, and the outlets were on the right. Thus, the higher cooling rates on the left resulted in higher local shrinkage on that side. Temperature transducers where not installed to measure the mold wall temperature disparity. Thus, it is not possible to evaluate this effect. Additionally, it is possible that a small error in locating the center of the mold base during grid engraving (Figure 4.1) could cause the grid elements to be somewhat smaller on the left side of the cavity. Consequently, measured shrinkage on the left side would appear to be larger than

the true value. The opposite is true for the right side of the cavity. However, an offcenter engraved mesh should not affect the overall shrinkage measurements.

**Table 5.4.** Summary of simulated and experimental overall shrinkage in X and Y directions at variable holding pressures, for PS (injection temperature =  $240^{\circ}$ C) and HDPE (injection temperature =  $200^{\circ}$ C)

Material	Pressure (MPa)	Direction	Experimental Overall	Simulated Overall			
			Shrinkage (%)	Shrinkage (%)			
PS	20	Y	0.8710	0.7236			
PS	20	X	1.2286	0.6745			
PS	30	Y	0.8856	0.6428			
PS	30	X	0.8883	0.6158			
PS	40	Y	0.5468	0.5858			
PS	40	X	0.6798	0.5575			
HDPE	20	Y	4.1500	3.5391			
HDPE	20	X	3.8050	3.1914			
HDPE	30	Y	3.6980	3.4033			
HDPE	30	X	2.5863	3.0706			
HDPE	40	Y	2.9940	3.2689			
HDPE	40	Х	2.3788	2.9630			

#### 5.2.1.2 Simulated Results

The predicted overall shrinkages were in the range of 0.54% - 0.72% for PS and 2.3% to 4.1% for HDPE (Table 5.4), within acceptable values for shrinkage of these materials (46). The simulation results correctly predicted decreasing shrinkage with increasing holding pressure. For PS the predictions were in good agreement with simulated results of Jansen (24), a thermo-elastic analytis, and Choi (32), a thermo-visco elastic analysis (Figure 5.17). For the semi-crystalline material, predictions were better than those of Jansen, whose thermo-elastic model largely over-predicted shrinkage for HDPE. At lower pressure, 20 MPa, there was under-prediction of shrinkage (Table 5.4). From examination of the shrinkage profiles in the X and Y directions for both PS (Figure 5.18) and HDPE (Figure 5.19), it appeared the under-prediction was due to excessive inmold shrinkage. At the edges, even with the adjusted free displacement boundary condition (section 5.1.2), local shrinkage was experimentally 2-3% greater than the predicted values. Meanwhile, at a higher pressure of 40 MPa, there was an over-

prediction shrinkage by 0.56% in the Y-direction and 0.27% in the X-direction for HDPE. This was attributed to mold-wall/polymer friction effects: contact between the mold wall and polymer has been known to provide resistance to movement of the sample while in the mold. It is known that friction between the mold wall and polymer increases with increasing holding pressure (47). The engraved mesh contributed to the roughness of the surface of the mold cavity, further increasing the friction between the mold wall and polymer. Visual inspection of the moldings showed that plates molded at high pressure had markings strongly protruding from the surface. This could have caused a "lock in effect", which reduced the ability of the material to shrink between the engraved grid lines. Products molded at 20 MPa were smooth, and the interlocking effect was expected to be negligible. Simulated pressure, temperature and crystallinity profiles are provided in appendix B.



Figure 5.17. From Choi *et. al.* (32): predicted shrinkage predictions for PS injection molded plates (using a 3D thermo-viscoelastic stress/strain model), compared with experimental data.



Figure 5.18. Simulated shrinkage profiles for PS at different pressures (a) across the Y direction and (b) along the X direction.



Figure 5.19. Simulated shrinkage profiles for HDPE at different pressures (a) across the Y direction and (b) along the X direction.

# 5.2.2 Thickness

### 5.2.2.1 Experimental Results

Overall thickness shrinkages were mapped across the Y direction and along the X direction for PS (Figure 5.20) and HDPE (Figure 5.21). For PS, negative shrinkages were observed ranging from -1% at low pressure, up to -7% for high pressure. In other words, the plates expanded. For HDPE, shrinkages were in the range of 3% to -0.2% at high pressure. From the literature, there is a great discrepancy of experimental results. Some authors report thickness shrinkage of the same order of magnitude as in-plane shrinkage, while others mention thickness shrinkages many times larger (and in some cased of opposite sign) than length and width shrinkage. Leo et. al., for example, report thickness shrinkages on the order of -4% for injection pressures of up to 60 MPa (48). Jansen (24) reported values of up to -7% for holding pressures as high as 80 MPa. The negative shrinkages observed are primarily attributed to mold deformation and flashing effects. As pressure increased, these effects become more prominent, consistent with greater deviations in the experimental shrinkage values from the simulated results. Physical examination of the molded specimens confirmed evidence of flashing, indicating that the mold clamping force was not sufficient at high pressure. For some cases, shrinkage was always the lowest in the middle of the molded plate, consistent with the possibility of mold bending under high pressure inside the mold. It has been noted that mold elastic deformation can play a significant role in the cavity pressure-time history, even for a seemingly stiff mold construction (48).

Another contributing factor could be due to the observation that linear shrinkage was positive and largest in the flow direction and, to a lesser extent, in the Y direction. Thus, to conserve volume (or mass), expansion (or negative shrinkage) occurs in the Z direction. This effect would be important for polystyrene, compounding the effects of mold deformation and flashing. On the other hand, linear shrinkage effects should not be so important for HDPE, since, for semi-crystalline materials, linear shrinkage occurs mainly in the amorphous regions of the polymer. Furthermore, volumetric shrinkage should be dominant, due to the large volumetric shrinkage during crystallization. Thus, positive shrinkage is likely to be obtained. In our experiments, no large negative values of shrinkage were observed for HDPE, in accordance with the above arguments.

It should be pointed out that HDPE showed significant variations in shrinkage along the flow path and across the plate. This is attributed to variations in crystalline development (49). On the other hand, PS, which is an amorphous material, did not exhibit such large variations in shrinkage in the X- and Y- directions.

#### 5.2.2.2 Simulated Results

For PS, the predicted shrinkage values were approximately 0.6% with a change of 0.05% with increasing pressure (Figure 5.20). For HDPE, they were approximately 3% with a change of 0.2% with increasing pressure (Figure 5.21). In contrast with the experimental results, the simulated results showed little variation with changes in pressure. Simulated shrinkages in the thickness (Z) direction were over-predicted, for both PS and HDPE because of the experimental limitations discussed above. The accuracy of the predictions in the Z direction would be expected to improve, if mold deformation effects were incorporated into the simulation model. For example, in an analysis by Jansen (47), even a simple 2-D elastic model taking into account mold deformation and friction between the polymer and the mold wall, predicted expansion of the molded plate in the thickness direction.



Figure 5.20. Simulated overall thickness shrinkage for PS at different holding pressures (a) across Y direction and (b) along the X direction.



Figure 5.21. Simulated overall thickness shrinkage for HDPE at different holding pressures (a) across Y direction and (b) along the X direction.

# 5.3 Effect of Injection Melt Temperature

## 5.3.1 In-Plane

## 5.3.1.1 Experimental Results

The influence of injection melt temperature on shrinkage was investigated by molding at various temperatures and pressures. Consistent with comments from section 5.2.1, as pressure was increased at constant melt temperature, the overall in-plane shrinkage always decreased, for both PS (Figure 5.22) and HDPE (Figure 5.23). However, with increasing melt temperature at constant pressure, no consistent trend could be established. Intuitively, it was expected that with increasing melt temperature, the shrinkage would increase due to greater thermal strain. Such was the case, for example, for PS in the Y direction for a holding pressure of 20 MPa, where the shrinkage increased from 0.62% to 1.12% with increasing melt temperature (Figure 5.22). For other cases, different tendencies were observed and attributed to early gate freezing at the lower melt temperatures, reducing the holding time and transmission of packing pressure. For example, HDPE shrinkage in the X direction at 20 MPa was greatest at an injection melt temperature of 180 °C (Figure 5.23). At higher holding pressures, the early gate freeze effect was less evident.

There was a tendency at any particular temperature or pressure for shrinkage to be greater in the X direction, compared with that in the Y direction for PS. On the other hand, HDPE shrinkage in the X direction was lower than that in the Y direction. For HDPE, flow induced-orientation causes crystalline chains to align in the flow direction, leading to closer packing in the cross-flow direction. For PS, however, linear shrinkage effects due to relaxation of flow-induced orientation occur mostly in the X direction, causing flow shrinkage to be greater (40). The effect of crystallization on cross-flow shrinkage in HDPE was demonstrated by Velarde (41). Increased cooling rates by changing the mold cavity thickness from 3 mm to 2 mm caused a reduction in crystallinity. Shrinkage in the flow direction for HDPE was almost the same, but shrinkage in the cross-flow direction was lowered by 70%.



Figure 5.22. Overall shrinkage at various injection melt temperatures and holding pressures for PS in the (a) Y direction and in the (b) X direction.



Figure 5.23. Overall shrinkage at various injection melt temperatures and holding pressures for HDPE in the (a) Y direction and in the (b) X direction.

# 5.3.1.2 Simulated Results

For all temperatures, agreement between experimental results and model predictions of shrinkage tended to be better at lower holding pressures, for PS (Figure 5.22) and HDPE (Figure 5.23). This is consistent with the consideration that mold wall/polymer friction effects become more prominent with increasing pressure, as discussed in section 5.2.1. The simulation program did not correctly predict the early gate freezing effect, since the simulated packing pressure curves were relatively unaffected by changes in melt temperature. Consequently, greater shrinkage was predicted for increasing melt temperatures. For HDPE, shrinkage was correctly predicted to be less in the X direction than in the Y direction. The model predictions were the least accurate in the X direction for PS, indicating that some important contributors to linear shrinkage may not have been taken into consideration.

Another factor may have contributed to some discrepancies in the prediction of shrinkage for the case of HDPE. Very short cooling times are encountered in injection molding. Thus, the crystallinity tends to be lower than values obtainable under slower cooling conditions. Accordingly, the simulation, which employs data based on measurements of crystallization kinetics made at relatively low cooling rates, would be expected to yield relatively higher shrinkage predictions. Most of the current available PVT data were obtained using cooling rates of the order of 3 °C/min. Typical cooling rates during injection molding are much faster, up to 5000 °C/min for thin walled mold cavities. The effect of cooling rates is especially important for semi-crystalline polymers. The specific volume is also influenced by both the crystallinity and crystalline structure, which are influenced by the pressure, temperature, stress, and flow history. Studies by Zudiema et. al.(50) on the effects of cooling rate on the PVT data of polypropylene shows a shrinkage of 12.45% when cooling from 500 k (P = 80 MPa) to 300 k (P = 0.1 MPa) at a rate of 0.17 k/s. On the other hand, only 10.84% shrinkage is observed when cooling at a rate of 80.0 k/s. In general, faster cooling rates, produce less crystallinity, or a larger amorphous component and lower shrinkage. For improved numerical simulation of the injection molding process, it is important to incorporate the effect of cooling rates on crystallization and solidification behavior of the material.



Figure 5.24. Simulated shrinkage profiles for PS for different injection melt temperatures (a) across the Y direction and (b) along the X direction, at holding pressure = 20 MPa.



Figure 5.25. Simulated shrinkage profiles for PS for different injection melt temperatures (a) across the Y direction and (b) along the X direction, at holding pressure = 30 Mpa



Figure 5.26. Simulated shrinkage profiles for PS for different injection melt temperatures (a) across the Y direction and (b) along the X direction, at holding pressure = 40 Mpa.



Figure 5.27. Simulated shrinkage profiles for HDPE for different injection melt temperatures (a) across the Y direction and (b) along the X direction, at holding pressure = 20 Mpa



Figure 5.28. Simulated shrinkage profiles for HDPE for different injection melt temperatures (a) across the Y direction and (b) along the X direction, at holding pressure = 30 Mpa



Figure 5.29. Simulated shrinkage profiles for HDPE for different injection melt temperatures (a) across the Y direction and (b) along the X direction, at holding pressure = 40 MPa.

## 5.3.2 Thickness

#### 5.3.2.1 Experimental Results

Similar to the results in section 5.2.2, the experimental thickness shrinkages for PS were negative for all melt temperatures. There was no significant effect of the injection melt temperature, though sometimes the shrinkage was less negative at the highest melt temperature. For example, in the Y direction at P = 40 MPa (Figure 5.32), the shrinkage was approximately -7% for melt temperatures 220 and 240 °C, and -6% for a melt temperature of 260 °C, most likely due to increased shrinkage associated with increased thermal strain. Generally, there was no significant variation across the plate and along the flow path.

For HDPE, at lower pressure (Figure 5.33), the shrinkage was in the range of 2.5 to 4% and even as high as 8% near the edges in the Y direction. No clear trend could be observed regarding the effect of melt temperature. However, as the pressure was raised, the shrinkage was greater with increased melt temperature, At 30 MPa (Figure 5.34), the thickness shrinkage was ~1.5% at 180 °C and increased to ~3% at 220 °C. The differences in shrinkage obtained at different melt temperatures were greater at 40 MPa. For this pressure, a negative shrinkage was observed at the lowest melt temperature (-1%) and as the temperature increased, the shrinkage increased to 1 %. It was only at high pressure and low temperature that negative shrinkage was observed. This confirms that mold deformation effects did exist for HDPE and that volumetric shrinkage compensated for negative shrinkage. Thus, it is likely that shrinkage would be larger if mold deformation and flashing effects were not present, as reported by Bain et. al. (51). In contrast with PS, HDPE showed substantially more variation across the plate and along the flow path. The shrinkage was very high along the edges and showed variation along the flow path. Near the gate, it exhibited a minimum, and generally increased towards the middle and the far end, likely due to differences in the evolution of crystallinity in different regions of the plate.

#### 5.3.2.2 Simulated Results

Shrinkage predictions in the thickness direction were inherently limited by the effects of mold deformation and flashing, which were greater at higher pressures. Thus, model predictions were more dependable at lower pressures. Therefore, the following discussion will emphasize predictions for holding pressure 20 MPa. However, results for all pressures are presented. Model predictions showed much smaller changes in shrinkage, in response to changes in melt. For PS, all the predicted shrinkages were approximately 0.6% (Figure 5.30). The predicted shrinkage was constant along the flow path and across the plate. Predicted values for HDPE showed some variation with melt temperature, but still not to the same as for the experimental values (Figure 5.27).

Melt temperature has a significant effect on thickness shrinkage, due to its influence on the development of skin thickness. The skin layer is produced by fast cooling of polymer melt submitted to extensional deformation in the flow front due to the fountain flow. This layer, particularly important for semi-crystalline materials, is 30-50µm thick. It seems to have a very high nucleation density, because no spherulites are observed. It has been reported that, in injection molding of nylon-6, the thickness of the skin layer decreased from about 1.25 mm to 0.75 mm (39% to 23% of total thickness) as the melt temperature increased from 225°C to 310°C (52). The thickness of nonspherulitic skins of injection molded polybutylene terephthalate (PBT) were found to vary inversely with melt temperature (53). The skin layer was non-oriented and had very low crystallinity, due to rapid cooling. Thus, in semi-crystalline materials, underprediction of the thickness of the skin layer would result in under-prediction of thickness shrinkage. Such an effect may have contributed to under-prediction of shrinkage at the holding pressure of P = 20 MPa (Figure 5.33) (where mold deformation and flashing effects should be unimportant).



**Figure 5.30.** Effect of Melt Temperature on overall thickness shrinkage for PS in the (a) Y direction and (b) X direction at a holding pressure = 20 MPa.



**Figure 5.31.** Effect of Melt Temperature on overall thickness shrinkage for PS in the (a) Y direction and (b) X direction at a holding pressure = 30 MPa.


Figure 5.32. Effect of Melt Temperature on overall thickness shrinkage for PS in the (a) Y direction and (b) X direction at a holding pressure = 40 MPa.



**Figure 5.33**. Effect of Melt Temperature on overall thickness shrinkage for HDPE in the (a) Y direction and (b) X direction at a holding pressure = 20 MPa



**Figure 5.34.** Effect of Melt Temperature on overall thickness shrinkage for HDPE in the (a) Y direction and (b) X direction at a holding pressure = 30 MPa.



**Figure 5.35.** Effect of Melt Temperature on overall thickness shrinkage for HDPE in the (a) Y direction and (b) X direction at a holding pressure = 40 MPa.

## 5.4 **Post-Processing**

#### 5.4.1 In-Plane

#### 5.4.1.1 Experimental Results

The effects of post-process heating on overall shrinkage were examined at constant melt temperature. For PS, the experimental overall shrinkage values in the X and Y directions (Figure 5.36) tended to increase, in comparison with the pre-heat treatment overall shrinkage, consistent with linear shrinkage occurring due to relaxation of residual orientation. For instance, at 20 MPa in the Y direction, the shrinkage increased 0.82% to 0.96%. Although the development of molecular orientation is primarily attributed to injection pressure and injection speed, which were kept constant for this work, holding pressure is also known to affect the orientation of injection molded parts (54). Thus, it was expected that the shrinkage differences would increase as holding pressure was increased. This was not observed in the current results. Secondly, since orientation development was primarily along the X direction, the shrinkage increase was expected to be the greatest in this direction. The above trends were not observed in the experimental work, because there was not a large enough difference between the lowest and highest holding pressures, considering the data scatter due to experimental errors. In the work of Pontes et. al. (54), the range of holding pressures investigated was from 12 MPa to 110 MPa, a much greater range than the 20 MPa to 40 MPa used in the current work. Inspection of the shrinkage profiles showed that shrinkage tended to increase throughout the plates (eg. Figure 5.40).

For HDPE, experimentally, the overall shrinkage increased significantly for all holding pressures and injection melt temperatures (Figure 5.37). The shrinkage increase was in the range of 0.3% - 0.8%. No consistent relationship was observed between changes in shrinkage in the X and Y directions for the amorphous PS or for HDPE. The shrinkage profiles showed that shrinkage increased uniformly throughout the plate (Figure 5.41,Figure 5.42,Figure 5.43). Presumably, recrystallization effects, throughout the plate, contributed to increase in shrinkage by annealing crystallinity.

#### 5.4.1.2 Simulated Results

For PS, very little changes were predicted for overall shrinkage (Figure 5.36). It was apparent the simulation program could not respond properly to orientation relaxation effects. It was not clear whether the small predicted changes ( $\sim 0.03\%$ ) were due to predicted dimensional changes or computational differences.

Conversely, the predictions were much better for HDPE. For overall shrinkage was typically predicted to increase ~0.2% (Figure 5.41-Figure 5.43). Inspection of the shrinkage profiles showed shrinkage increased towards the edges but not towards the center. This was consistent with the predicted change in crystallinity by the simulation program. For instance, at a holding pressure of 30 MPa (Figure 5.44), the crystallinity in the core (z/a = 0) was predicted to increase from 55% to 62% towards the middle, but at the edges, it was predicted to increase from 30% to 62%.



**Figure 5.36**. Effect of heating on overall shrinkage of PS in the (a) Y direction and (b) X direction for at different holding pressures for an injection melt temperature of 240 °C.



**Figure 5.37**. Effect of heating on overall shrinkage of HDPE in the (a) Y direction and (b) X for at different holding pressures for an injection melt temperature of 200 °C.



**Figure 5.38.** Experimental and simulated shrinkage profiles across for PS at holding pressure = 20 MPa after post-process heating, along the (a) y-direction and (b) x-direction.

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**Figure 5.39.** Experimental and simulated shrinkage profiles across for PS at holding pressure = 30 MPa after post-process heating, along the (a) y-direction and (b) x-direction.



**Figure 5.40**. Experimental and simulated shrinkage profiles across for PS at holding pressure = 40 MPa after post-process heating after post-process heating, along the (a) y-direction and (b) x-direction.



Figure 5.41. Experimental and simulated shrinkage profiles across for HDPE at holding pressure = 20 MPa after post-process heating, along the (a) y-direction and (b) x-direction.



**Figure 5.42**. Experimental and simulated shrinkage profiles across for HDPE at holding pressure = 30 MPa after post-process heating, along the (a) y-direction and (b) x-direction.



**Figure 5.43**. Experimental and simulated shrinkage profiles across for HDPE at holding pressure = 40 MPa after post-process heating, along the (a) y-direction and (b) x-direction.



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Figure 5.44. Predicted crystallinity profiles for HDPE molded plates at different depths through the thickness (a) in the Y direction and (b) in the X direction, before and after heat treatment. Processing conditions are P (holding) = 30 MPa and T (melt) = 200 °C.

### 5.4.2 Thickness

#### 5.4.2.1 Experimental Results

Experimental results for PS revealed that, upon heating, shrinkage tended to decrease both across the plate and along the flow direction (Figure 5.45-Figure 5.47). Generally, uniform plate expansion was observed on the order of 0.1% to 1%, with no preference to location along the flow path. At higher holding pressure (Figure 5.47), there was greater expansion than at lower holding pressure (Figure 5.45).

In contrast to experimental results, shrinkage predictions for HDPE were inconclusive about the effects of heating on the plates (Figure 5.48-Figure 5.50). Sometimes the shrinkage increased, while at other locations the shrinkage decreased.

The fact that expansion in the thickness direction was greater for PS than for HDPE supports the previous conclusion (section 5.2.2.1) that linear shrinkage effects due to contraction in the X and Y direction cause expansion in the thickness direction. These effects are more significant for PS than for HDPE for two reasons: (i) linear shrinkage is not as important for semi-crystalline materials, because only the amorphous regions tend to be susceptible to linear effects; and (ii) volumetric effects in HDPE are dominant over the linear effects. Any positive shrinkage that may occur is counter-balanced by negative shrinkage due to recrystallization.

#### 5.4.2.2 Simulated Results

Simulation results for PS were similar to in-plane predictions, showing no change after heating. HDPE shrinkage profiles were also similar to in-plane results, showing the shrinkage to increase towards the edges because of increased crystallinity.



Figure 5.45. Effect of heating on thickness shrinkage of PS molded plates (a) in the Y direction and (b) in the X direction for processing conditions of P (holding) = 20 MPa, and T (melt) = 240 °C.



Figure 5.46. Effect of heat.ing on thickness shrinkage of PS molded plates (a) in the Y direction and (b) in the X direction for processing conditions of P (holding) = 30 MPa, and T (melt) = 240 °C.



Figure 5.47. Effect of heating on thickness shrinkage of PS molded plates (a) in the Y direction and (b) in the X direction for processing conditions of P (holding) = 40 MPa, and T (melt) = 240 °C.



Figure 5.48. Effect of heating on thickness shrinkage of HDPE molded plates (a) in the Y direction and (b) in the X direction for processing conditions of P (holding) = 20 MPa, and T (melt) = 200 °C.



Figure 5.49. Effect of heating on thickness shrinkage of HDPE molded plates (a) in the Y direction and (b) in the X direction for processing conditions of P (holding) = 30 MPa, and T (melt) = 200 °C.



Figure 5.50. Effect of heating on thickness shrinkage of HDPE molded plates (a) in the Y direction and (b) in the X direction for processing conditions of P (holding) = 40 MPa, and T (melt) = 200 °C.

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# **6** CONCLUSIONS AND RECOMMENDATIONS

### 6.1 Conclusions

The first step in the simulation is to obtain a mathematical model of the process, accounting for the major phenomena involved. Then, by employing appropriate boundary conditions and devising a computational scheme, validation of the simulation is sought by comparison of model predictions to experimental results. This has been accomplished in the current work. Reasonable, though not always similar, quantitative and qualitative predictions were obtained using a 3-D thermo-visco-elastic stress/strain simulation (McKAM-4) for shrinkage analysis of PS and HDPE injection molded rectangular plates. Subsequently, the research explored other factors to obtain more precise predictions that contain the subtle nuances of experimental trends. The current work explored the effect of various modelling, computational, and experimental variables that affected the experimental and simulated results and that might lead to discrepancies between the two. Some of the important factors evaluated are listed below.

- 1) inclusion of melt strain
- 2) modifying the strain history term used in the visco-elastic equations
- 3) mold deformation
- 4) mold wall/polymer friction
- 5) high cooling rates
- 6) development of the skin-layer
- 7) early gate freeze.

Among the processing conditions, holding pressure was the most important factor affecting the shrinkage. Consistent with the literature, both the experimental and predicted shrinkage decreased with increasing holding pressure. The agreement was best at lower pressures, since, at higher pressures, factors such as mold/polymer friction, mold deformation and flashing became more significant and prominent. Increasing melt temperature had variable effects, because of early gate freeze effects observed at lower melt temperatures.

Finally, the simulation proved to be effective in predicting shrinkage for a simple post-processing heat cycle for HDPE, because it could account for volumetric recrystallization effects, which dominated over linear effects. It was not sufficiently comprehensive to account for orientation in the molded plates. Thus, it was not as effective for predicting the effect of the heat cycle on shrinkage of the amorphous PS.

## 6.2 **Recommendations**

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It is recommended to carry out a more extensive experimental program. Some of the important aspects to be considered include:

(a) Varying injection speed and pressure (which was not possible in the present equipment) to investigate more thoroughly the effect of orientation development on linear shrinkage for the amorphous plates.

(b) Cold milling the PS molded plates to confirm that relaxation of outer orientation is the primary source of linear shrinkage due to heating, as done by Harrell and Elleithy (16).

(c) Making crystallinity measurements of the HDPE plates before and after heating to confirm that the change in shrinkage is attributable to recrystallization effects.

(d) Extending the post-process heat treatment to higher temperatures and longer times.

(e) Performing heat treatment experiments on plates molded at different injection melt temperatures.

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## APPENDICES

# A. Introduction to Residual stresses

During the injection molding process the material experiences a variety of stresses that relax and disappear quickly at temperatures around the melting or glass transition temperature. However, as the material cools and solidifies, the stress relaxation time becomes much longer, and once the material attains sufficient mechanical strength to resist deformations, the stresses become "frozen" into the material. A good illustration of stress development is provided by Douvan *et. al* (1).

Residual stresses may be classified according to their source: thermal and flow stresses. In general, the thermal and flow stresses are coupled (2), but for the purposes of simplifying the analysis, they are regarded as uncoupled; furthermore, the flow stresses are even ignored, since they have been found to be 2 orders of magnitude lower than the thermal stresses (3).

Thermal stresses arise in during the cooling of a molten polymer sample as a consequence of the thermal gradients present during solidification. Flow stresses are induced by the frozen molecular orientation as a result of the visco-elastic nature of the polymer material. Flow stresses are primarily present within the surface layers since a large part of the orientation has already relaxed in the core during the packing stage.

The stress profile observed for freely quenched parts is parabolic; rapid inhomogeneous cooling of the surfaces forms a rigid shell, which prevents free contraction of the core layers as they cool and solidify successively, resulting in compression at the surface, and tension in the middle (Figure 0.1) (4). In contrast with free quenching, the stress distribution for injection molded parts is significantly different from the ideal parabolic form (Figure A- 1) because the quenching conditions are complicated by the melt pressure history and mechanical constraints of the mold. A typical stress distribution will have tensile stresses at the surface of the sample (region I, Figure A- 2), then a region of compressive stresses (region II), followed by tensile stresses in the core (region III) (5).



Figure A-1. Typical residual stress distributions through a freely-quenched plastic sample.



Figure A-2. Typical residual stress distributions through an injection-molded plastic sample.

# **B.** Simulated Temperature, Pressure and Crystallinity Profiles



Figure B-1. Simulated and experimental gate pressure profiles for PS, at a holding pressures of (a) 20 MPa, (b) 30 MPa, and (c) 40 MPa. The simulated pressure curves are not significantly affected by a change in injection melt temperature.



**Figure B-2**. Simulated and experimental gate pressure profiles for HDPE, at a holding pressures of (a) 20 MPa, (b) 30 MPa, and (c) 40 MPa. The simulated pressure curves are not significantly affected by a change in melt temperature.



**Figure B- 3.** Temperature evolution through the thickness of a HPDE injection molded plate, for injection melt temperatures of (a) 180 °C, (b) 200 °C, and (c) 220 °C (holding pressure independent), as predicted by the McKam simulation program, for a molding cycle of 60 s.



**Figure B- 4.** Temperature evolution through the thickness of a HPDE injection molded plate, for injection melt temperatures of (a) 180 °C, (b) 200 °C, and (c) 220 °C (holding pressure independent), as predicted by the McKam simulation program, for a molding cycle of 60 s.



**Figure B- 5.** Crystallinity evolution through the thickness of a HPDE injection molded plate, for injection melt temperatures of (a) 180 °C, (b) 200 °C, and (c) 220 °C (holding pressure independent), as predicted by the McKam simulation program, for a molding cycle of 60 s.

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