Preface

POLYMER MATRIX COMPOSITE MATERIALS are being selected for an increasing number of applications in the aerospace, automobile, and construction industries. On the other hand, thermal deformations that occur during the manufacturing processes of long fiber-reinforced composites, including thermoplastic composites, have been a continuing challenge for manufacturers. These deformations can be difficult to predict due to the complex thermal and mechanical behavior of composite laminates. Currently there are significant research efforts to understand and create specialized computer codes for performing process optimization and predicting dimensional distortions in composites. To date, however, commercially available codes have shown limited capability to model thermoplastic matrix materials, and the simulations are vastly dedicated to thermoset composite materials and their manufacturing processes.

In thermoplastic composite industries, trial-and-error methods are commonly used to optimize processing variables and to apply corrections to the original mould or tool shapes and ensure that the final part meets the desired design specifications. These corrections, however, must be made for each new part and often require several iterations of high-cost tooling, increasing the time and cost of process development.

We designed this book as a guide toward developing accurate process models for thermoplastic composites. We first discuss the most known fundamental mechanisms that lead to dimensional distortions during the manufacturing process of thermoplastic matrix composite parts. Subsequently, the book provides a step-by-step approach for making models to predict the distortion magnitude of such composites

via commercially available finite element (FE) codes. For the latter, we selected a case study on a multistage roll forming a glass/polypropylene-woven composite within the commercial FE code ABAQUS. It is also shown how such process models can be used for sensitivity and statistical analysis and eventually optimizing process variables. By reducing the need for trial-and-error processes,such models have the potential to reduce significantly the upfront time and costs associated with a new part and increase the competitiveness of thermoplastic matrix composite manufactures.

Finally, we would like to acknowledge that some sections of this book were published as a dissertation by Corey Lynam at the University of British Columbia (UBC). Financial and administrative support from Natural Sciences and Engineering Research Council of Canada, Industrial Materials Institute of the National Research Council of Canada, Mathematics of Information Technology and Complex Systems Research Network,all our colleagues at UBC Composites Group and the pan western Composites Research Network, AS Composites Inc., and DEStech Publications Inc.are greatly acknowledged.

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CHAPTER 1

Introduction

THERMOPLASTIC MATRIX COMPOSITES have attracted an increasing market share in a number of applications in leading industries such as aerospace, automotive, and construction. Developments in fabricating new composite architectures as well as new, rapid forming processes have allowed thermoplastic matrix composites to be competitive alternatives to thermoset composites. Commercial materials such as Twintex® are designed to overcome the relatively high viscosity of thermoplastic polymers at processing temperatures. In its initial fabric form, continuous glass fibers are commingled with continuous polypropylene (PP) fibers in bundles that are then woven into a fabric. Commingling the polypropylene and glass fibers in each yarn reduces diffusion distances of the matrix and, therefore, lower cycle times and pressure can be used. New composite manufacturing processes have also been developed that take advantage of these types of materials. Among others, roll forming is a continuous manufacturing process that has been implemented recently in industrial settings. In roll forming, unconsolidated laminates of a thermoplastic composite (e.g., Twintex®) are passed continuously through an oven and a series of compaction rollers. The oven heats the material above the melting temperature of polypropylene, then several pairs of rollers compact the laminate and distribute the melted polypropylene throughout the fabric. As the material cools below the crystallization temperature of the matrix, it consolidates into its final hard form. This thermoplastic matrix composite material and roll forming manufacturing process combination can demonstrate several advantages over traditional thermoset matrix composites. These include higher production rates, process automation, lower

costs, higher impact toughness, and simpler manufacturing processes. Because there is no release of volatile organic compounds when forming thermoplastic composite materials, there are also fewer health and safety concerns.

Thermal deformations can occur during the manufacturing process of thermoplastic matrix composites where the final part shape is different from the original tool shape (in the case of roll forming, different from the shape of the final set of rollers). Typically, these deformations are in the form of "spring-in," where the included angle between two surfaces on the part is smaller than the angle of the tool. These deformations can be significant for large components, in assemblies, or when strict dimensional tolerances are required. Spring-in is primarily due to the anisotropic thermal properties of the composite and the thermal cycle during the manufacturing process. The current practice in industry is to use "rule-of-thumb" compensations on the mould (tool) shapes based on previous experience. Typical compensations range from 1° to 2.5°. As a result, several iterations of tooling geometries are needed before a final part can be produced within acceptable tolerances. In turn, these tooling iterations can add a significant cost to the process-development phase of a new product and prevent it from being financially feasible. The ultimate goal of the present case study is to create a predictive model of the spring-in for a roll-forming process so that few, if any, trial-and-error iterations of tooling are needed.

Chapter 2 continues with a detailed description of thermoplastic composite materials and their manufacturing methods, and a comprehensive overview of the mechanisms that lead to spring-in. Chapter 3 provides a synopsis of currently published work in the field of thermoplastic spring-in modeling. Chapter 4 presents the results of material property testing conducted within an illustrative case study. Chapter 5 describes the example case for a multistage roll forming process and presents results of the spring-in angle measurements. Chapter 6 describes the finite-element modeling technique applied to this process and discusses the significance of the prediction results for manufacturers. This chapter summarizes earlier discussions of the book and outlines a few recommendations for future work in this magnificent field.

CHAPTER 2

Background

2.1. THERMOPLASTIC MATRIX COMPOSITE MATERIALS

CONTINUOUS FIBER-REINFORCED POLYMER (CFRP) composite materials can be categorized in two groups based on the matrix material. The first group is thermoset matrix composites, which is comprised of thermosetting polymers as the matrix material. The matrix is formed during the manufacturing process of the end-use component by an irreversible chemical curing reaction of two parts: a resin and a hardener. The advantage of this group is that the resin and hardener have a relatively low viscosity in their unreacted liquid state; it is relatively easy for the resin and hardener mixture to diffuse into a fabric of reinforcement material. In addition, since the cure reaction takes an appreciable amount of time after mixing and is typically thermally activated, there is sufficient time to assemble the part before cure. During cure, the glass transition temperature of the matrix material evolves from a very low value to a high value so that the final material is in a solid glassy state. One advantage of a high glass-transition temperature after cure is that thermoset matrix composite materials can often be used at temperatures above their cure temperature in service.

The second group of CFRP is the thermoplastic matrix composites, comprising of thermoplastic polymers as the matrix material. The thermoplastic polymer material is formed in a step prior to the end-use component-manufacturing step and can typically take the form of a solid sheet, tape, or fibers. A thermoplastic polymer will soften and melt with the application of heat, then crystallize and harden when cooled. When

processing a thermoplastic composite material, the matrix is heated above its melting temperature, allowed to diffuse into the reinforcement and then cooled below its crystallization temperature. The challenge in processing thermoplastic composite parts is that the matrix material has a much higher viscosity in its melted state than a precure thermoset matrix. It typically requires higher pressure and higher temperature to process a thermoplastic matrix composite material. In service, the composite cannot be used at temperatures that approach the processing temperature since the matrix will soften and remelt. However, there are reduced health and safety concerns when forming thermoplastic composite materials, since there is no chemical cure and therefore no release of volatile organic compounds.

The most common reinforcement materials for both thermoplastic matrix and thermo set matrix materials are glass fibers, carbon fibers, and aramid fibers. Given any of these reinforcement materials, a thermoplastic matrix composite will typically have a higher toughness and impact resistance, while a thermoset matrix composite will have a higher maximum operating temperature and a small strength and stiffness benefit. Traditionally, thermoset matrix materials have dominated CFRP composite industries due to their relative ease in manufacturing. However, with improvements in the manufacturing techniques of thermoplastic matrix composites, an increasing number of industries have been adopting thermoplastic matrix composites as an alternative.

2.1.1. Twintex®

The sample material used throughout the case study in this book is a thermoplastic matrix composite with the trade name Twintex®, commercialized by Saint-Gobain Vetrotex. It is a continuous fiber composite with constituents of a polypropylene matrix (40%) and E-glass fiber reinforcements (60%). It is produced in a woven fabric form with a balanced 2 × 2 twill weave pattern, has a specific weight of 1492 g/m^2 (44 oz/yd^2), and a nominal thickness of 1 mm (0.04 in) per lamina after consolidation. To overcome the manufacturing challenge described earlier—the high viscosity of the melted matrix during processing—the polypropylene is initially in fiber form and is commingled with the glass fibers in every fiber bundle (yarn). Figure 2.1 shows how this precommingling of the two composite constituents reduces the diffusion distances for the melted matrix and helps the matrix penetrate each bundle

and surround the glass fiber strands. With reduced diffusion distances, lower pressure is required to produce a composite part with a low void content (Borazghi, Boucher, Denault, & Fisa, 2008).

The glass transition temperature of the polypropylene in Twintex® is around –15°C and the melting temperature is around 163°C (see DSC results in Section 4.3). Since the glass transition temperature is below room temperature, the material will show viscoelastic behavior even at room temperature. With a melting temperature of 163°C, the applications of Twintex® are limited to lower temperatures. However, being based on E-glass and the commodity polymer polypropylene, Twintex® has a very low material cost and is generally very competitive for low, room, and modest temperature applications. Some of these applications include vehicle door panels, helmets and protective armor, small wind turbine blades, and residential building materials. For higher temperatures and more demanding applications there are other thermoplastic polymers that can be substituted for polypropylene, usually at an increased material cost.

2.1.2. Crystallization in Polypropylene

An understanding of the crystallization process of the polypropylene matrix material is fundamental to any manufacturing process model of Twintex®. The isotactic polypropylene in Twintex® is a linear polymer, such that propylene monomers (C_3H_6) link in long chains through covalent bonds. At temperatures above the melting temperature, these

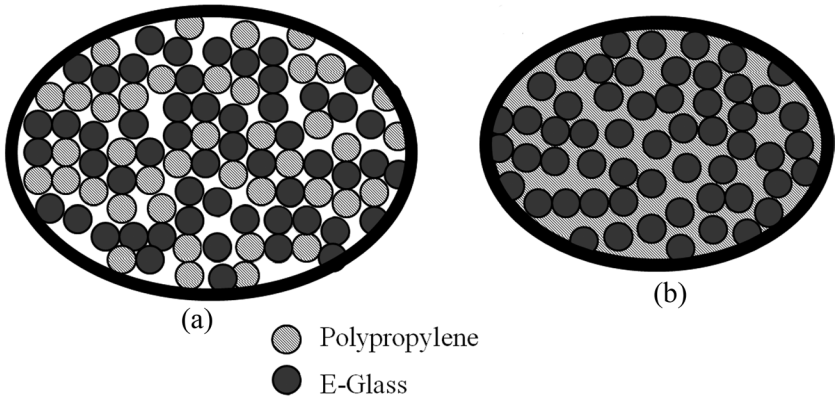
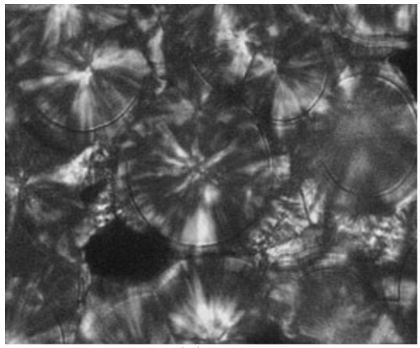
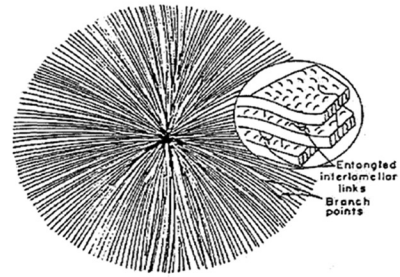


FIGURE 2.1. *Schematic of a cross section of a pre-consolidated (a) and consolidated (b) Twintex® bundle.*



(a)



(b)

FIGURE 2.2. *(a) Micrograph of polypropylene (Tian, Yu, & Zhou, 2007) and (b) Scematic of a spherulite (Advani & Sozer, 2003).*

chains are arranged in an irregular amorphous structure. If the polymer cools quickly, the amorphous arrangement of molecular chains can freeze in place due to van der Waals' forces. However, at slower cooling rates, the molecules will align themselves into their lowest energy state, which is a regular crystal formation. This crystallization process in thermoplastics occurs by repeated stacking of individual molecular chains. A single molecular chain can fold back onto itself many times within a crystal or extend between two adjacent crystals. Crystallization can occur to various degrees such that the microstructure is a certain combination of amorphous and crystalline zones. The polypropylene in Twintex® is typically described as a semi-crystalline material.

Crystals form in a radial direction from a central nucleus such that the microstructure contains an array of spherulites, as shown in Figure 2.2. Nucleation of stable spherulites is a diffusion-based process, as it requires the displacement of polymer chains. Thus, the nucleation process takes a certain amount of time before free growth of the crystals can begin. Free growth of the crystals will continue until the spherulites impinge on one another. Secondary crystallization process can then occur at sites other than those originating at the nuclei. However, secondary crystallization is slower and occurs at lower temperatures. Due to the nucleation and growth process, the crystallization kinetics and the degree of crystallinity will depend strongly on the rate of cooling. Even after secondary crystallization, the final microstructure will always contain some amorphous zones. It is, therefore, not possible to have a 100% crystalline microstructure.

2.2. THERMOPLASTIC MATRIX COMPOSITE MANUFACTURING METHODS

To transform a preconsolidated fabric of Twintex® into a rigid part, the laminate must be heated above the matrix material's melting temperature then cooled below its crystallization temperature. To reduce void content and to form the laminate to a desired shape, this thermal cycle occurs along with the application of pressure. Several manufacturing methods can be applied to Twintex®. The most common processes are compression moulding, vacuum bag and autoclave, roll forming, and stamp forming. Each of these methods is described in the following sections.

2.2.1. Compression Moulding

The most commonly used manufacturing process for thermoplastic matrix materials is compression moulding. In this process, the composite laminate is compressed between a male and female mould pair. The moulds are preheated to a temperature above the melting temperature of the matrix material by circulating oil or electrically, as shown in Figure 2.3. The laminate is loaded into the mould cavity and the mould pair is closed. Pressure is applied with a hydraulic or pneumatic piston and held for a specified amount of time. The entire assembly then cools to room temperature together. Compression moulding tooling can be expensive due to the required heating channels, but is generally more cost-effective than using an autoclave to reach high pressures. Process

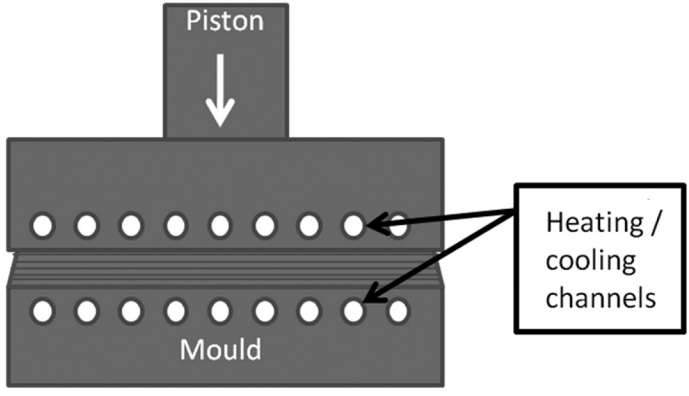


FIGURE 2.3. *A schematic of a compression moulding forming press.*

4.4. DYNAMIC MECHANICAL ANALYSIS

Dynamic mechanical analysis (DMA) can be used to test the viscoelastic behavior of the laminate. A wide range of tests can be conducted within the DMA family. For the purpose of this study, three-point bending tests on samples of roll-formed Twintex® are most appropriate. Three-point bending of a plate sample is the closest loading condition to the bending stresses that develop in the corner section of the V-shaped roll-formed sample. The fixture and sample are shown in Figure 4.11.

DMA testing involves imposing a sinusoidal strain on a sample and measuring the resulting stress. Similarly, a sinusoidal stress can be imposed and the resulting strain can be measured. For a perfectly elastic material and for small deformations, the sinusoidal stress and strain curves would be perfectly in phase so that there are no losses. Because there is a direct proportionality between stress and strain through Hooke's law, the ratio of the stress and strain amplitudes will be equal to the flexural modulus. For an ideal liquid with an applied sinusoidal shear strain, the sinusoidal stress response will be 90° out of phase because the stress is proportional to the strain rate. For viscoelastic materials, which exhibit intermediate behavior between that of a per-

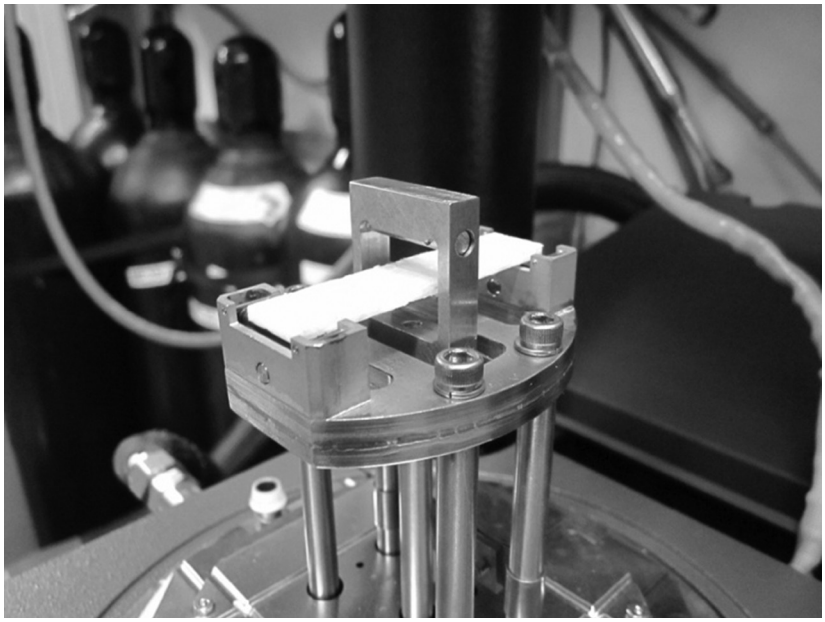


FIGURE 4.11. *Three-point bending fixture and Twintex® sample for DMA anlysis.*

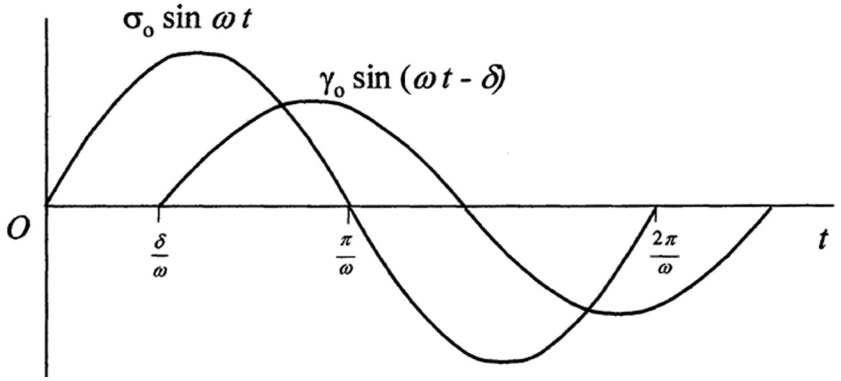


FIGURE 4.12. Sinusoidal stress and strain input and response curves for a viscoelastic material showing a phase lag (Mase & Mase, 1999).

fectly elastic and ideal liquid material, the sinusoidal stress (or strain) response will have some phase shift to the sinusoidal strain (or stress) input. Figure 4.12 shows the typical input and response. This phase shift, sometimes called a lag angle, is a measure of the viscous character of the viscoelastic material, where 0° represents an ideal elastic solid and 90° represents an ideal viscous liquid.

From the sinusoidal response we can define the storage modulus (E') and loss modulus (E''). The storage modulus represents the elastic character of the material where there are no losses and is given in Equation (4.2). The loss modulus represents the viscous character of the material and is given in Equation (4.3). Figure 4.13 shows, in vector form, how the storage modulus is based on the component of the stress response in phase with the strain input. Similarly, the loss modulus is based on the component of the stress response that is 90° out of phase with the strain

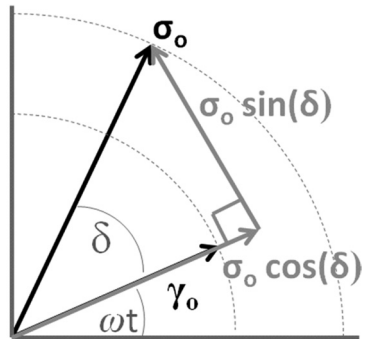


FIGURE 4.13. Sinusoidal stress and strain in vector form.

input. The phase lag (*d*) can be calculated using Equation (4.4). In addition, the storage and loss moduluses can be expressed in complex form, where the storage modulus exists in real space and the loss modulus exists in imaginary space, as shown in Equation (4.5).

$$E' = \frac{\sigma_o \cos(\delta)}{\gamma_o} \tag{4.2}$$

$$E'' = \frac{\sigma_o \sin(\delta)}{\gamma_o} \tag{4.3}$$

$$\tan(\delta) = \frac{\sigma_o \sin(\delta)}{\sigma_o \cos(\delta)} = \frac{E'}{E''} \tag{4.4}$$

$$E^* = E' + iE'' = \frac{\sigma_o}{\lambda_o}[\cos(\delta) + i\sin(\delta)] = \frac{\sigma_o}{\lambda_o} e^{i\delta} \tag{4.5}$$

A Rheometric Scientific Model DMTA MKA machine was used to perform DMA tests of roll-formed Twintex® samples. Three-point bending tests were conducted on samples of two-layer, 44 oz/yd^2 Twintex® produced by roll forming. The tests were done at a constant frequency of 1 Hz while the temperature was swept from room temperature to the melting temperature of Twintex®. The results from two samples are shown in Figure 4.14. It can be observed that the storage modulus

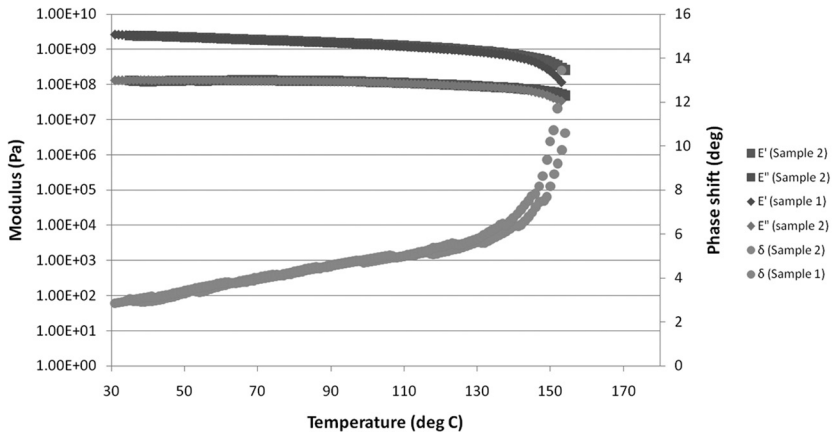


FIGURE 4.14. *DMA results; three-point bending, increasing temperature sweep at 1 Hz.*

and loss modulus are quite constant over the temperature range. Additionally, the phase shift is small, less than 6°, until the temperature approaches the melting point of the material and it begins to soften. A small phase shift suggests that within the range between crystallization and room temperature, the Twintex® laminate behaves much closer to an ideal elastic solid than an ideal viscous liquid.

4.5. STRESS RELAXATION

In the context of process modeling, we are interested to know the thermal mechanical behavior of the laminate during the cool-down cycle. It is very challenging to perform DMA testing with a cool-down thermal cycle due to the difficulty in holding the sample at temperatures above the melting temperature. Furthermore, above the melting temperature, the laminate tends to delaminate and deconsolidate so that if the cooling cycle is performed while the laminate is not under pressure, the sample may develop a higher void content and lower consolidation compared to the real process. To address these challenges, three-point bending stress relaxation tests were performed with a TA-Q800 DMA. During testing, the Twintex® samples were heated to 168°C and held for 5 minutes before cooling. Based on the DSC results, at this temperature and hold time, the samples should be uniformly above the melting temperature while the amount of deconsolidation or delamination of the sample is minimized. While performing these tests are challenging, they provide insight into the thermal mechanical behavior of Twintex® on a decreasing temperature cycle.

In the stress relaxation tests, a small, near instantaneous strain is applied to the sample and resulting force values are recorded so that the change in the modulus can be measured with time. The procedure for the experiment involved:

- Loading the sample into the fixture
- Bringing sample temperature to 30°C and holding for 5 minutes
- Applying a center piston displacement of 680 μm, holding for 30 minutes and recording load
- Raising the temperature to the following inclement and holding for 5 minutes to equalize
- Repeating the two steps above until the melting temperature of the matrix is reached

Stress relaxation tests were performed for both increasing and de-

creasing temperature cycles at 5°C increments from 30°C to 165°C. Figures 4.15 and 4.17 show the results for the increasing and decreasing temperature cycles, respectively. It can be observed that there is an initial modulus that relaxes over time. Figures 4.16 and 4.18 show the isochronal cross sections of the stress relaxation results. For the isochronals, the relaxation modulus is plotted as a function of temperature for a given value of time.

From the stress relaxation results it can be observed that the change in modulus due to a change in temperature is much greater than the change in modulus due to holding for an amount of time. This suggests that Twintex® behaves much more thermo-elastic than thermo-viscoelastic. In the short time scales of roll forming, these results suggest that any viscoelastic effect will have a small impact on the modulus development. This trend is consistent with the small phase lag angle as seen in the DMA testing in Section 4.4.

In addition to the viscous effect, Figure 4.18 shows the presence of a small but meaningful modulus at temperatures above the crystallization temperature range. For a decreasing thermal cycle, it was previously expected that the modulus was negligible before the crystallization process. However, these results suggest that the precrystallization, amorphous, supercooled matrix material begins to develop a significant modulus when cooled to temperatures below its equilibrium melting temperature. A precrystallization modulus development can have an effect on the spring-in angle predictions described in Section 2.3.1.4. Two

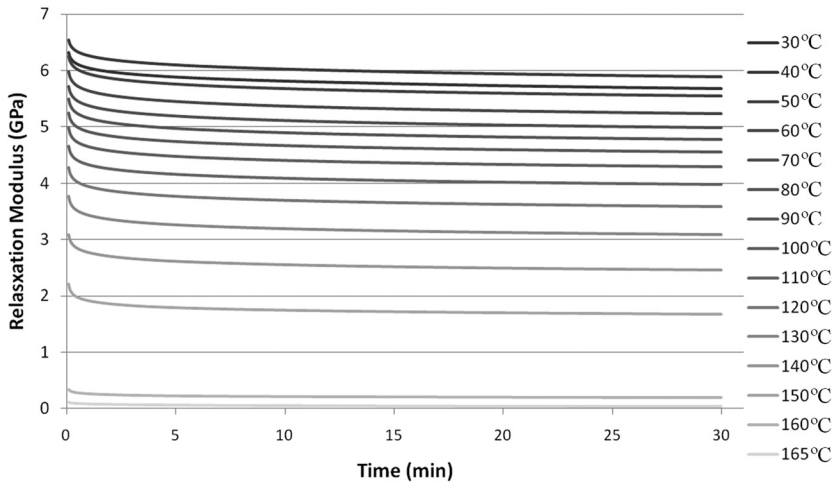


FIGURE 4.15. *Stress relaxation results: three-point bending, increasing temperature.*

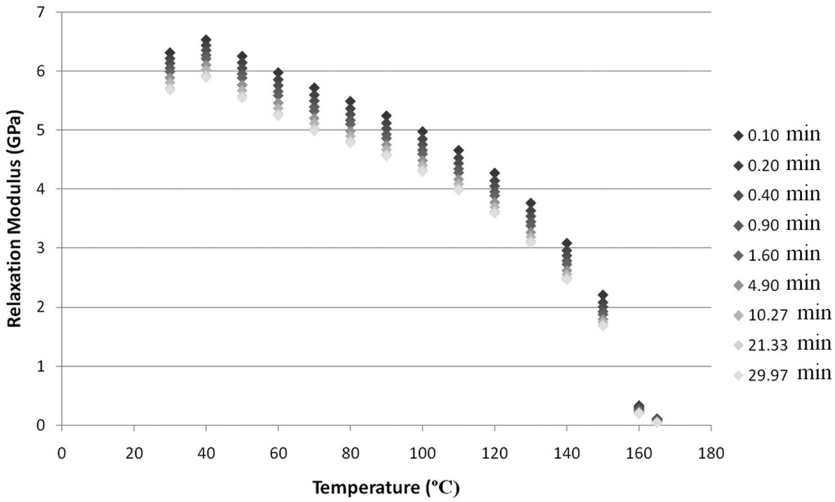


FIGURE 4.16. Stress relaxation isochronals: three-point bending, increasing temperature.

external sources could have caused this small modulus. First, it is possible that in the experiment, the sample did not completely melt before the cooling procedure started. This could leave some residual, pre-melt modulus. Second, due to the very slow cooling rate, crystallization in the sample would have started much earlier than for the fast cooling rate used in a real roll forming process. It may be that some crystallization has started leading to the apparent modulus. At this time, these results

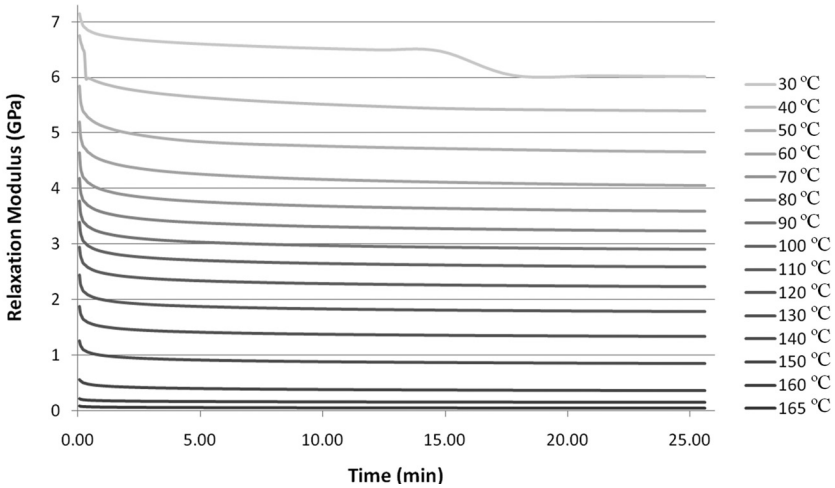


FIGURE 4.17. Stress relaxation results: three-point bending, decreasing temperature.

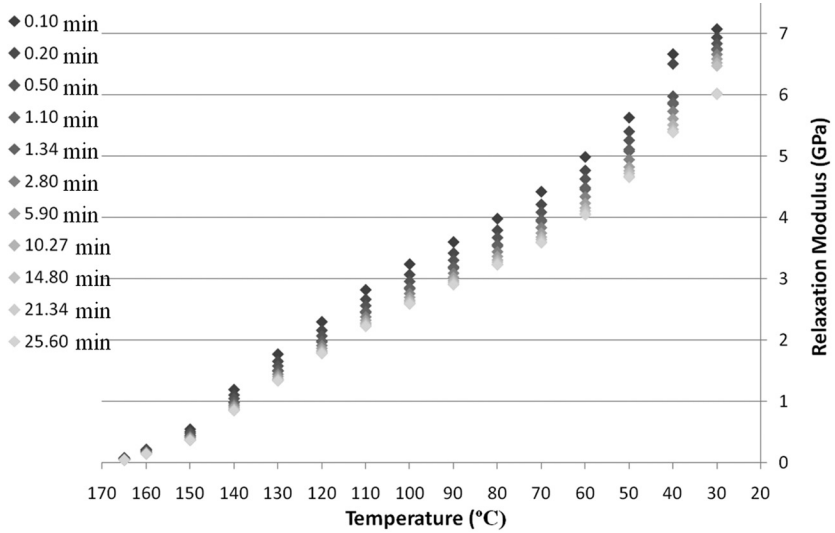


FIGURE 4.18. *Stress relaxation isochronals: three-point bending, decreasing temperature.*

are considered preliminary and it remains for future work to investigate the precrystallization modulus development of the matrix material more closely.

Overall, the results of DMA and stress-relaxation testing have revealed the complex thermal mechanical behavior of Twintex®. In general, both experiments have shown that Twintex® behaves much more thermoelastically than thermoviscoelastically and that, particularly in the time scales of roll forming, the viscous component can likely be neglected. A complete characterization of the thermoviscoelastic behavior of Twintex® is to be completed in the future, as it appears more applicable to manufacturing methods other than roll forming. Nonetheless, in Sections 6.3 and 6.4, a hypothetical viscoelastic model will be adopted with the purpose of demonstrating how its inclusion can affect the FE modeling results.

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