ULTRASONIC CONSOLIDATION OF DRY CARBON FIBER AND POLYPHENYLENE SULFIDE FILM

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ABSTRACT

Ultrasonic welding of thermoplastics is a common practice in industry but its use for composite materials is not fully understood yet. Ultrasonic consolidation of carbon fiber/thermoplastic composites uses frictional and viscoelastic heating generated by ultrasonic vibrations to melt the thermoplastic matrix and infuse dry fibers. The advantage of this method is its high speed (<10 s) and low cost compared to traditional techniques such as compression molding. Ultrasonic consolidation was performed on polyphenylene sulfide (PPS) films and woven dry carbon fibers with a Rinco Dynamic 3000 welder. Consolidation pressures ranged from 0.48 MPa to 1.1 MPa. Weld duration was controlled with the welder's travel parameter, setting the distance of compression during the application of vibrations. Viable travel values were determined from fiber compressibility. Void content was obtained with optical microscopy and crystallinity was calculated using differential scanning calorimetry (DSC). Crystallinity was also verified with microhardness measurements. This study demonstrated that ultrasonic consolidation is a viable manufacturing method, producing CF/PPS parts with low void content (< 2 %) and crystallinity values that increased with consolidation pressure.

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1. INTRODUCTION

The need for lightweight structures in the aerospace and automotive industries creates a demand for cost effective manufacturing methods. Thermoplastic composites (TPCs) are well suited for this because of their short manufacturing time and their ability to be re-melted for assembly. Ultrasonic consolidation (USC) of thermoplastic composites is a promising manufacturing technique. This method has the advantages of rapidity, energy efficiency, and ease of automation. Ultrasonic consolidation uses frictional and viscoelastic heating generated by ultrasonic vibrations to melt the thermoplastic matrix and infuse the dry fibers. Ultrasonic welding of thermoplastic parts has been in use since the 1980's [1], but its application to joining thermoplastic composite parts has only been studied more extensively in the past 7 years [2]. There have been very few studies on the ultrasonic-assisted consolidation of composite materials, which is the focus of this study.

Gomer et al. [3] conducted a study on unidirectional dry carbon fibers (CF) and glass fibers (GF) with polyamide 6, polyethylene, and polypropylene films. They successfully demonstrated the feasibility of this method, requiring high tension in the unidirectional dry fibers during consolidation to prevent the fibers from spreading. They reported maximum tensile strengths of 1.2 GPa and a fiber volume fraction of 33 %, but did not investigate the microstructural outcomes. Another similar method was developed by Rizzolo et al. [4], who tested the use of ultrasonic heating for automated fiber placement of unidirectional GF/high-density polyethylene tape and unidirectional CF/polyethylene terephthalate tape. They accomplished this by placing the tapes onto a moving stage under a stationary sonotrode. They demonstrated that the stiffness of these layups decreased by approximately 50 % when the speed of the layup stage during consolidation was tripled.

Crystallinity plays an important role in the mechanical behavior and chemical resistance of a polymer. Higher crystallinity will lead to higher chemical resistance, stiffness, and strength, where an amorphous structure will be less brittle and more impact resistant [5]. Differential scanning calorimetry (DSC) is typically used to calculate the crystallinity of polymers. After ultrasonic heating, the polymer cools down from its meting temperature to room temperature in less than 10 seconds. Cooling rates this rapid are expected to result in a predominantly amorphous thermoplastic matrix [3]. Microhardness tests are a simple method for characterizing the microstructure of a polymer. The microhardness is directly related to the crystallinity of the sample [4] and may be used to compare the crystallinities of samples consolidated under different processing conditions.

During ultrasonic consolidation, pressure and vibrations are applied through a flat sonotrode. In theory, the optimum downward displacement of the sonotrode during consolidation (also called "travel") should be high enough to replace all of the air inside the dry CF plies with melted PPS. This idea is based on Helmus et al. who studied air evacuation from a partially impregnated microstructure [6]. The amount of air inside the CF plies is determined based on the thickness of the dry CF plies, the mass per area of the plies and the volume density of the carbon fibers. The volume fraction V_f of carbon fibers inside the plies is given by Eq. [1]:

$$V_f = \frac{\sigma}{th * \rho_f} \quad [1]$$

where σ is the surface area density of the plies, th is the thickness of the plies, and ρ_f is the volume density of the carbon fibers. The travel T required to impregnate the plies is given by Eq. [2]:

$$T = th * (1 - V_f) \quad [2]$$

where T is the travel required. Thickness of the plies refers to thickness just before the onset of consolidation once the consolidation pressure has been reached. Compression tests were conducted to determine the relationship between the consolidation pressure and the thickness of the dry CF plies.

This paper is focused on investigating the feasibility of manufacturing CF/PPS composites from woven dry fibers and PPS films using ultrasonic consolidation. This study determined the parameters under which ultrasonic consolidation was successful, and characterized the resulting

products. Void content and crystallinity were compared for samples manufactured under different consolidation pressures and travel values. This was done to understand the effects of the consolidation parameters on product characteristics, and to optimize product quality.

2. EXPERIMENTATION

2.1 Materials

PPS films, supplied by Professional Plastics, were 0.08 mm-thick and cut into 50 mm by 50 mm squares. The dry carbon fiber plies, supplied by Composite Envisions, were a 2x2 twill fabric with a surface density of 238 gsm, woven from high density Toray T-300 with a density of 1.76 g/cm^3. The dry fiber fabric was cut into 80 mm by 80 mm squares which were then taped around the edges to minimize fraying. The tape was applied to the CF fabric before the squares were cut out (Figure 1).

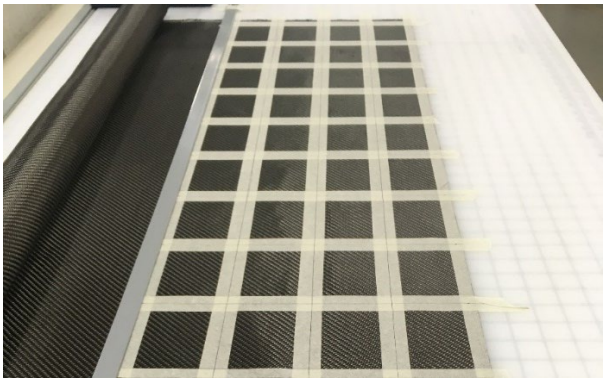

Figure 1. Tape was applied to 2x2 twill CF fabric (Toray T-300) before squares were cut out.

2.2 Ultrasonic consolidation

A Rinco Dynamic 3000 ultrasonic welder performed the consolidation. The welder operates at 20 kHz and has a maximum power output of 3000 W. A circular sonotrode with a diameter of 40 mm was used, all tests were conducted with a vibrational amplitude of 38.1 μm. The layup consisted of three plies of dry carbon fiber surrounded by four PPS films, as shown in Figure 2. All CF plies were taped to the jig to prevent motion during consolidation.

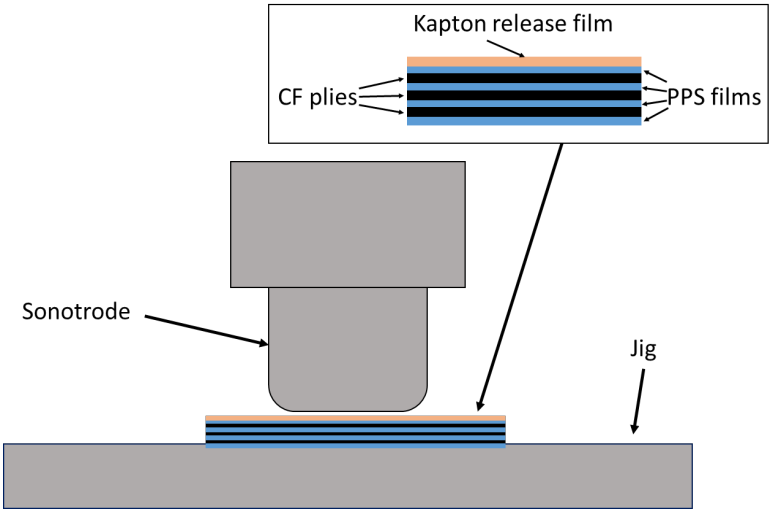

Figure 2. Diagram of ultrasonic consolidation layup.

The process begins when the sonotrode descends onto the materials and presses down with increasing force until the pressure for consolidation is reached. Once that pressure has been reached ultrasonic vibrations are applied. The application of pressure and ultrasonic heating is continued until the sample has been compressed to a prescribed distance since the start of vibrations. This distance is revered to as travel. When the travel distance is met the sonotrode stops transmitting ultrasound and maintains the consolidation pressure for a holding time of 2 seconds. The ultrasonic welder is controlled by a microprocessor referred to as the automated control unit (ACU) which allows the welding parameters such as force, travel, and weld time to be monitored and controlled [7].

Temperature measurements were taken using type K thermocouples from Omega Engineering to record the cooling curves for some of the samples. This was done by placing the thermocouples underneath the layup and in the center of the sonotrode's contact area.

2.3 Dry fiber bed compression

The thickness of the dry CF plies determines the theoretical travel required to fully impregnate the CF plies with PPS. The relationship between the ply thickness and consolidation pressure was determined by placing 10 CF plies on the welding jig and increasing the pressure from 0 MPa to 1.6 MPa. The thickness and pressure were obtained from the sonotrode's displacement and force data generated by the ACU.

2.4 Void content

Cross sectional images of the samples were taken to measure void content. The samples were cut and mounted vertically in epoxy pucks which were then polished using 360, 600, 800, and 1200 grit sand paper, followed by pads sprayed with diamond particle (6 µm, followed by 1 µm). Images of the samples were then taken through a Meiji MT8100F optical microscope at 20x magnification.

The void content was found using ImageJ by selectively picking out voids using brightness thresholds for macro- and micro-voids, as illustrated in Figure 3.

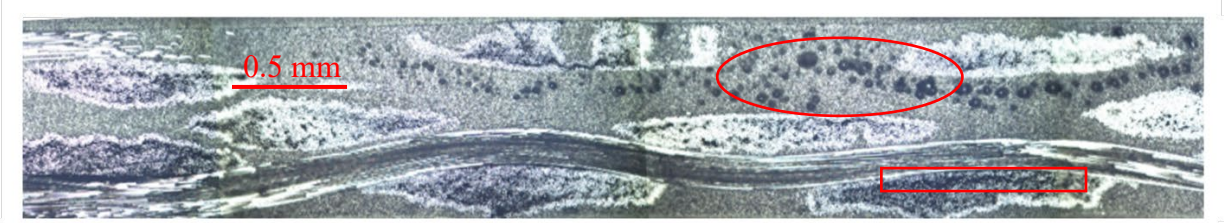

Figure 3. The macro voids are seen as spherical bubbles in the composite's matrix, examples are circled with an oval. Micro-voids were located inside the fiber tows, such as the section contained by the rectangle.

2.5 Crystallinity

A PerkinElmer DSC 4000 was used to determine the crystallinities of the different samples. The samples were cut into pieces between 5 mg and 10 mg, and sealed in a 5 mm diameter aluminum pan. The pan was placed in the DSC with a second empty aluminum pan for reference. The samples were heatup up to 300 °C at 10 °C/min, then remained at 300 °C for 1 minute. It was cooled down to room temperature at 10 °C/min. After 1 minute at room temperature, the cycle was repeated a second time.

Cold crystallization occurs at temperatures above the glass transition temperature (T_g), which is 90 °C for PPS [5]. Cold crystallization is an exothermic process where the polymer chains have gained enough mobility to cross link into semi-crystalline structures [8]. The melting temperature (T_m) of PPS is 285 °C [5], the latent heat of melting is associated with the potential energy of semi-crystalline structures (Figure 4). The degree of crystallinity is calculated using Eq. [3]:

$$X_c = \frac{\Delta H_m - \Delta H_c}{\Delta H_f^0} * 100 \quad [3]$$

where X_c is the percent crystallinity, ΔH_m is the specific melting enthalpy, ΔH_c is the specific energy of cold crystallization, and ΔH_f^0 is the specific melting enthalpy of an ideal crystal. The values for ΔH_f^0 vary greatly in literature, ranging from 50 J/g to 150.4 J/g [9]. The ΔH_f^0 value used to calculate these crystallinities (Eq. 3) was 100 J/g since that is the median of the range of possible values.

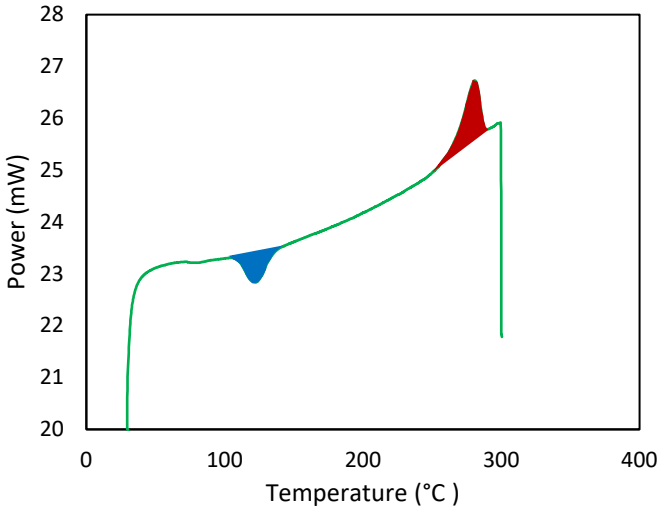

Figure 4. Power transmitted during DSC indicates cold crystallization at 125°C highlighted in blue and melting at 280°C highlighted in red.

ΔH_m and ΔH_c were found by plotting the power with respect to time and integrating the area between the peaks and the power curves that would have existed without the peaks (Figure 5).

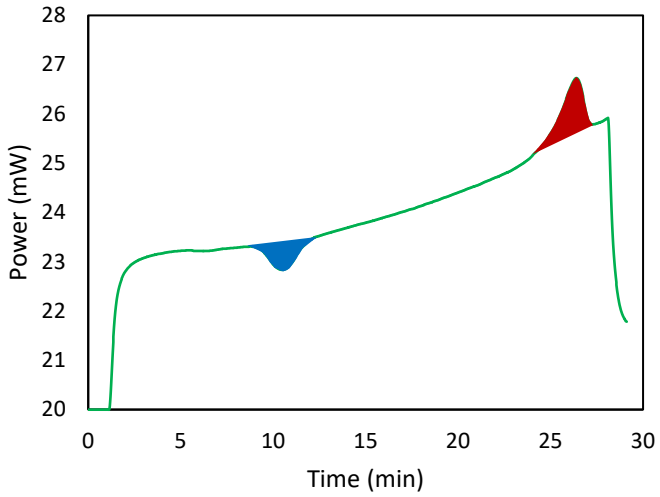

Figure 5. Power peaks are integrated over time for cold crystallization and melting. Highlighted areas are the areas integrated to obtain ΔH_m and ΔH_c.

2.6 Microhardness

A Clark CM-802AT microhardness tester was used to test the microhardness of the samples. Samples were mounted on epoxy casts and fastened onto the stage of the microhardness tester. An area on the sample with all PPS and no CF was found with a 10X objective lens. This area was then compressed for 15 seconds with a square pyramid shaped diamond tip under a load of 500 g. At the end of these 15 seconds the indent left by the tip is square shaped. The diagonals of the

square indents were measured through the microscope's 10X objective lens. The resulting microhardness was represented in the following equation:

$$HV = \frac{F}{d1 * d2} \quad [4]$$

HV is the Vicker's microhardness value, F is the force applied to the sample through the diamond tipped piece, and d1 and d2 are the lengths of the diagonals of the square indentation. Notice the units of HV are in N/mm^2, but it does not represent a pressure.

3. RESULTS

3.1 Compression tests of dry CF plies

The thickness of the dry fibers, the surface area density of the dry fibers (238 gsm), and the volume density of the dry fibers (1.76 g/cm^3) all determine the amount of air remaining in the dry fiber plies. The travel required to remove this air was defined by combining Eq. [1] and Eq. [2]:

$$T = th - \frac{\sigma}{\rho_f} \quad [5]$$

Compression tests on the dry fibers were carried out with the sonotrode to obtain the thickness of the plies as a function of consolidation pressure, which was then used to generate a relationship between consolidation pressure and optimum travel (Figure 6). The travel represents the downward vertical displacement of the sonotrode during the ultrasonic consolidation process.

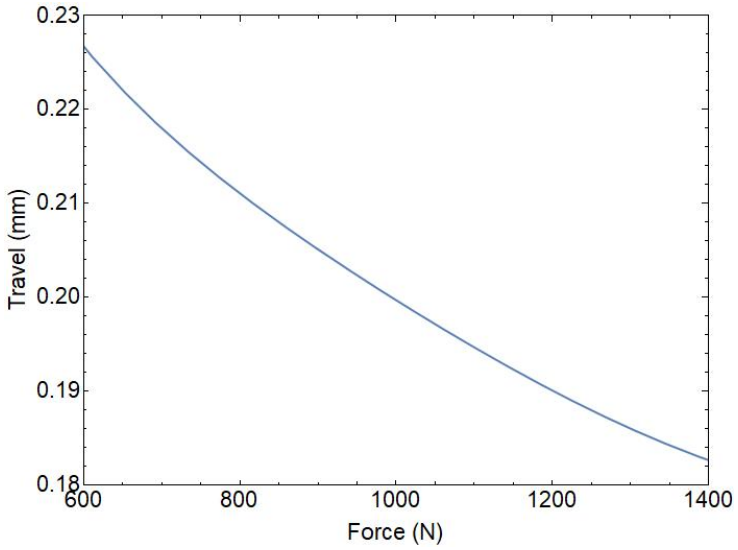

Figure 6. The travel required as a function of Force is found from the compressibility of the dry CF plies.

The resulting optimum travel values for a 3 ply layup at the tested consolidation pressures are shown in Table 1.

Table 1. The optimum travel value is listed for each consolidation pressure. These travel values vary slightly, ranging from 0.18 mm to 0.23 mm.

Consolidation pressure (MPa)	0.48	0.64	0.80	0.95	1.11
Sonotrode Force (N)	600	800	1000	1200	1400
Optimum Travel (mm)	0.23	0.21	0.2	0.19	0.18

3.2 Void content

All samples were consolidated under forces from 600 N to 1200 N in 200 N increments with a 40 mm diameter circular sonotrode, resulting in consolidation pressures of 0.48 MPa to 1.11 MPa. All these samples had travel values of 0.18 mm, 0.20 mm, 0.22mm, or 0.24mm in order to create consistency between different consolidation pressures and to cover the range of values shown in Table 1. Void contents for these samples are shown below (Figure 7).

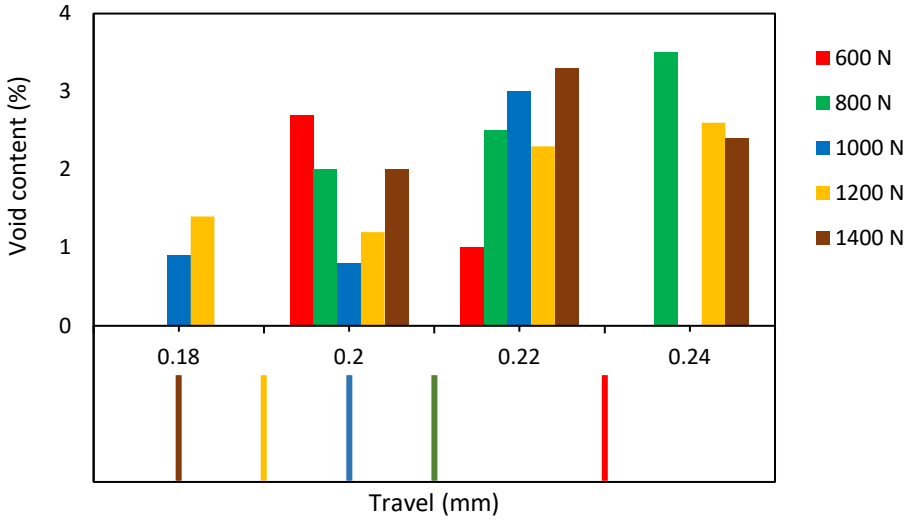

Figure 7. Resulting void contents for samples of varying force and travel values. Theoretical optimum travel values for each force are indicated with a vertical line under the bar graph.

The theoretical travel values for each force value is displayed under the bar graph. The travel values of lowest void content show a general agreement with the theory. The sample with the lowest void content for any given consolidation pressure also has a travel value that is closest to the optimal value. The void contents were averaged for the different forces and compared in Figure 8.

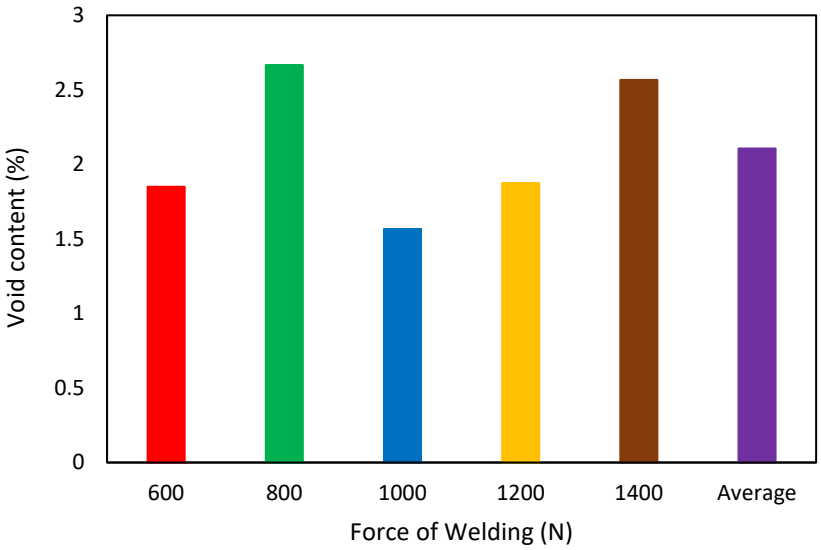

Figure 8. Void contents for each force are compared. The average void content for all samples shown on the right is 2.1 $\pm$ 0.9 %.

A consolidation pressure of 0.95 MPa resulting from a force of 1200 N generated samples with the lowest average void content. Repeated trials are underway to test for repeatability. It should be noted that a void content of 2 % is the quality threshold for the aerospace industry. Most of the samples fell within that threshold, and all samples were manufactured in a short period of time (4 to 8 seconds), making ultrasonic consolidation a high-speed process which does not sacrifice quality.

3.3 Temperature measurements

The temperature profiles for the cooling of the samples immediately after the application of vibrations showed an impressively rapid cooling rate of 125 °C/s between T_m and T_g. This was not surprising because the samples had thicknesses between 0.7 mm and 0.75 mm and remained under consolidation pressure for a holding time of two seconds after the vibrations had stopped. The samples submitted to temperature measurements all had travel values of 0.22 mm and all displayed similar cooling behaviors (Figure 9).

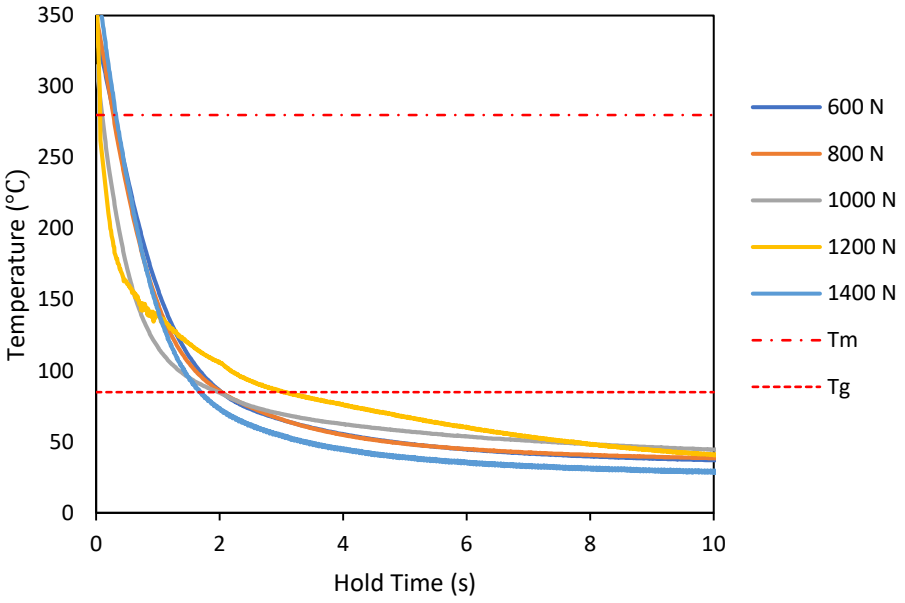

Figure 9. Cooling profiles for consolidated samples immediately after the application of ultrasonic vibrations.

Most samples cooled from T_m to T_g in the first two seconds of hold time with an exception for the 1200 N samples, which took three seconds. They all displayed high cooling rates, but they had no consistent relationship with consolidation force.

3.4 Crystallinity and microhardness

DSC measurements were conducted on the same samples that were subjected to temperature measurements. Cooling rates appeared to be independent of the consolidation force (Figure 9), which suggests that the relationship between consolidation force and crystallinity must be caused by other factors.

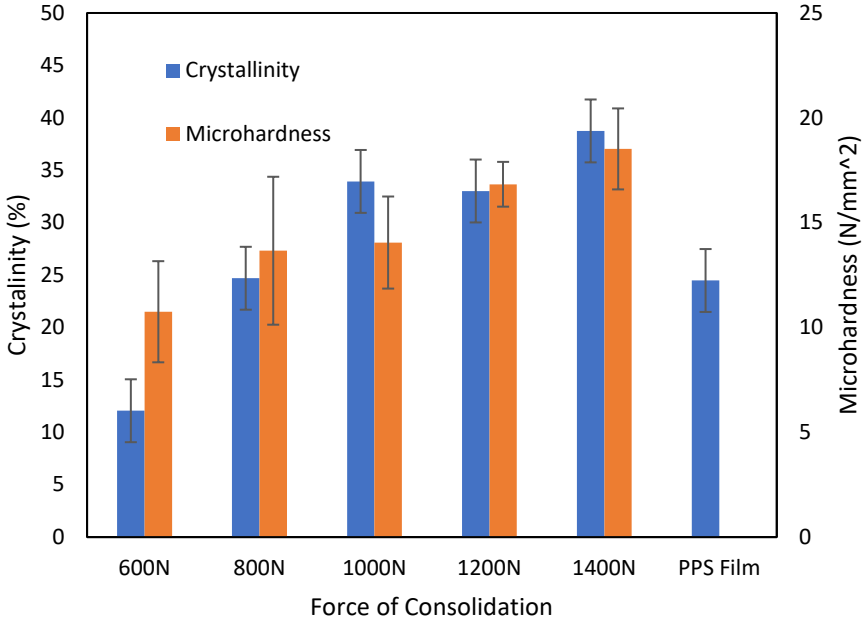

Figure 10. Both crystallinity and microhardness increased with consolidation force. The crystallinity of the unprocessed PPS film is displayed on the right.

The resulting degrees of crystallinity are unexpected considering the rapid cooling rate of these samples. The direct relationship between crystallinity and consolidation force is supported by a similar relationship between microhardness and consolidation force, this is because higher hardness is associated with higher crystallinity [4]. This is likely due to strain induced crystallization [10], which would be greater under higher consolidation forces. Since the ΔH_f^0 values used to calculate crystallinity (Eq. 3) vary greatly in literature [9], further work needs to be carried out to determine this value for the PPS used in these experiments. It should be noted that the trend of increasing crystallinity with increasing consolidation forces remains intact, regardless of the value for ΔH_f^0 used in Eq. 3.

4. CONCLUSIONS

Ultrasonic consolidation of CF/thermoplastic composites is a promising new manufacturing method which has the advantage of its speed and low cost. The samples fabricated for this paper showed low void volume fractions (< 2 %).Void content could be minimized in a predictable fashion that agreed with theory. However, future work is needed to test the repeatability of these void volume fractions.

Increasing consolidation force increased the crystallinity of the PPS matrix and microhardness of the matrix. This relationship between the consolidation force and the matrix crystallinity is a potentially useful tool for controlling the crystallinity of the manufactured product. An accurate value for the specific melting enthalpy of an ideal PPS crystal (ΔH_f^0) should be determined in future work.

Additional future works will include: 1) stiffness and strength tests, under tension and 3-point bending, of ultrasonically consolidated composites in comparison with their compression molded counterparts, 2) further investigation of temperature profiles and crystallinity with respect to ultrasonic consolidation parameters, and 3) real-time flow visualization through an optical welding frame.

5. REFERENCES

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