

# Effects of Processing Parameters on Axial Stiffness of Self-Reinforced Polyethylene Composites

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**ABSTRACT:** Self-reinforced polyethylene composites have proven to be promising candidate materials for a number of wear-resistance and bioimplant applications. In this study, we investigated the effects of processing parameters on the elastic modulus of self-reinforced high-density polyethylene (HDPE) composites. The processing parameters investigated were the cooling rate, processing pressure, temperature, and duration. Our results showed an optimum processing temperature, pressure, and duration that were matrix-dependent. In addition, for an HDPE matrix, the slower the composite cooling rate was, the higher the composite modulus was. © 2001 John Wiley & Sons, Inc. *J Appl Polym Sci* 81: 1136–1141, 2001

**Key Words:** Polyethylene, self-reinforcing composites

## INTRODUCTION

The growing demand for lightweight, high-performance materials coupled with escalating energy costs in the manufacture of other high-performance materials has been a key factor in the increasing interest in polymeric matrix composites (PMCs).<sup>1</sup> PMCs are one of the most developed classes of composites used widely in aerospace and commercial applications. A special class of PMC is the self-reinforced polymeric composite. Self-reinforced polymeric composites are those composites where both the matrix and reinforcement are made of the same polymer but with different mechanical properties. The difference in the mechanical properties between the reinforcement and the matrix is a direct result of the difference in polymer chain orientation. In the case of polyethylene (PE), for example, an increase of about 3 orders of magnitude in the elastic modulus can be achieved because of chain orientation. The concept of self-reinforced PE com-

posites was first studied by Capiati and Porter.<sup>2</sup> In this case, the difference in the chain orientation in the fibers and the matrix resulted in a difference in their melting temperatures. Highly oriented high-density polyethylene (HDPE) fibers were found to exhibit an increase in the melting point of about 5–9°C above that of the regular HDPE matrix.<sup>3</sup> The complete melting of the matrix accompanied by partial melting of the fibers at the surface aids in the formation of an interfacial bond between the matrix and fibers and helps the mechanical performance of the composite. A number of researchers have investigated the properties of self-reinforced PE.<sup>4,5</sup> A systematic study to investigate the effects of processing parameters on the resulting composites has not been done. In this study, we systematically investigated the effects of processing parameters on the composite stiffness. A modified rule-of-mixture formula was used to quantify the interfacial adhesion between the matrix and fiber.

## EXPERIMENTAL

The matrix used was Exxon's (Exxon, USA) commercially available HDPE with a melt index of

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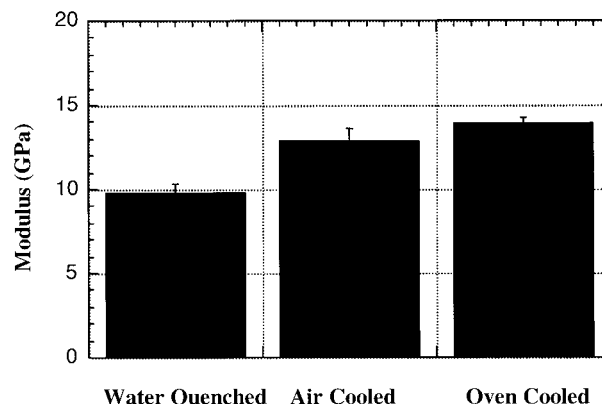
0.3, a density of 0.97, and a melting point of 130°C. The fiber used was Spectra® 1000 of Allied Signal Corp. USA, which is made of ultra-high-modulus PE with a diameter of 23  $\mu\text{m}$ . The fiber ultimate tensile strength and elastic modulus were 4.5 and 177 GPa, respectively. These experimentally measured values were in agreement with values reported by other authors.<sup>6</sup> Composite sheets were prepared in a Carver hot press (model 2915). Fibers were sandwiched between two matrix sheets and then processed in the hot press. Care was taken to spread the fibers uniformly over the matrix to get a uniform weight fraction. In all the specimens, the fiber weight fraction was 20–25%. Specimens for the tensile tests were cut with a steel mill die from 15  $\times$  15 cm composite sheets. The gauge length of all the samples was 50 mm, the width was 5 mm, and the thickness was an average of five values obtained from five different locations within a 50-mm gauge length. Tensile properties of the composite samples were determined with an Instron tensile testing machine (model 4500). Tensile testing was done according to ASTM D 1708-84 with a strain rate of 2.5%/min. While one of the aforementioned processing parameters was changed, the rest of the parameters were kept constant. The constant values for the processing time, load, and temperature were 2 h, 1 ton, and 140°C, respectively.

## RESULTS AND DISCUSSION

The goal of this work was to investigate the effects of processing parameters on the stiffness of self-reinforced PE composites. Parameters considered in this study were the cooling rate (from processing temperature to room temperature), the load applied during composite processing (processing pressure), the processing time, and the processing temperature.

### Effect of the Cooling Rate

Three different cooling rates were considered: a very fast cooling rate in which the samples were quenched in water after processing, a medium cooling rate in which samples were taken out of the hot press and left to cool in air, and a slow cooling rate in which samples were left in the hot press after the press off was switched off. Figure 1 shows the composite tensile modulus at the three different cooling rates. It is well known that the cooling rate will affect the crystallization of

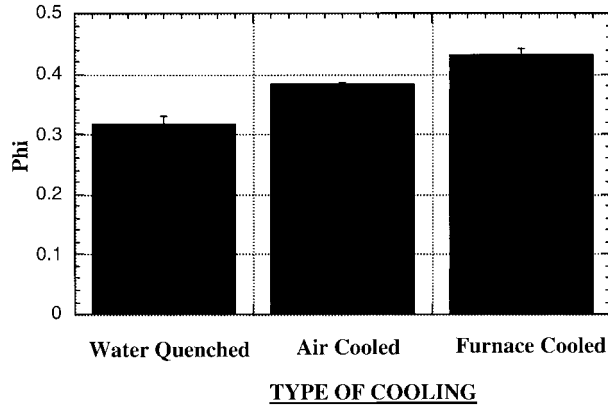


**Figure 1** Effect of the cooling rate on the tensile modulus of self-reinforced PE composites.

the matrix and the transcrystallization developed at the fiber/matrix interface.<sup>5,7–9</sup> The observed increase of the composite modulus as the cooling rate was reduced can result from the increase in the matrix modulus due to increased bulk crystallinity and/or the increase in interfacial interactions due to increased transcrystallinity. The contribution of the increase in matrix modulus, however, should be minimal compared to the contribution of the increase in interfacial interaction due to large differences between matrix and fiber moduli (1.15 and 177 GPa, respectively). In addition, the matrix modulus did not show much of an increase under the three cooling rates used in this study. The matrix modulus increased from  $1.15 \pm 0.2$  GPa for water-quenched samples to  $1.45 \pm 0.3$  GPa for oven-cooled samples. This should only increase the composite modulus by about 0.2 GPa under perfect adhesion conditions and was neglected in our calculations. A modified rule-of-mixture formula was defined to quantify the interfacial adhesion in the processed composite. The composite tensile modulus ( $E_c$ ) was calculated as

$$E_c = \Phi (E_f * V_f + E_m * V_m)$$

where,  $\Phi$  is the interfacial adhesion parameter;  $E$  and  $V$  are the elastic modulus and volume fraction, respectively; and the subscripts  $f$  and  $m$  refer to the fiber and matrix, respectively.  $\Phi$  can only assume values in the range  $1 \geq \Phi \geq 0$ . When the value of  $\Phi$  is 1, interfacial adhesion is perfect, and our modified equation would yield the same composite modulus as the linear rule-of-mixture equation that is already based on a perfect inter-

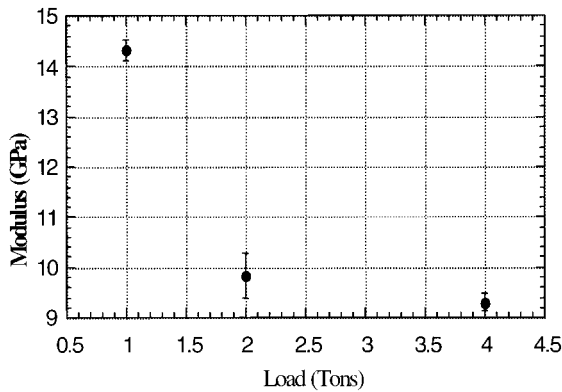


**Figure 2** Calculated interfacial adhesion parameter ( $\Phi$ ) at different cooling rates.

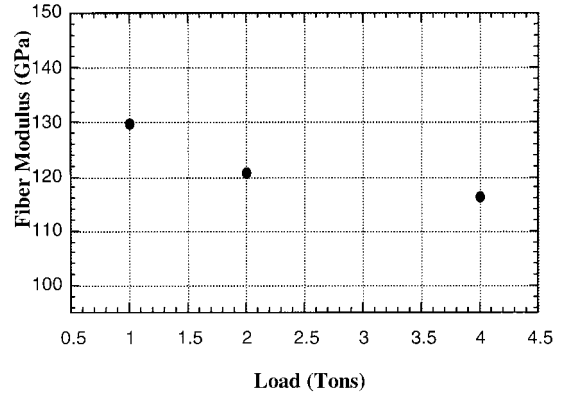
facial adhesion assumption.<sup>10</sup> Figure 2 shows the calculated interfacial adhesion parameter ( $\Phi$ ) at the three cooling rates considered in this study. It is clear that increased interfacial adhesion occurred as the cooling rate was reduced.

**Effect of the Processing Pressure**

Self-reinforced composites were processed under three different processing loads: 1, 2, and 4 tons. These loads correspond to processing pressures of 0.6, 1.2, and 2.4 MPa, respectively. Figure 3 shows the effect of the processing load on the tensile modulus of the self-reinforced PE composite. It is clear that an almost exponential drop in the composite modulus resulted as the processing pressure was increased. On the exposure of the reinforcing fibers to the same processing conditions and the change of the processing pressure, a similar reduction in the reinforcing fiber modulus



**Figure 3** Effect of the processing load on the tensile modulus of self-reinforced PE composites.

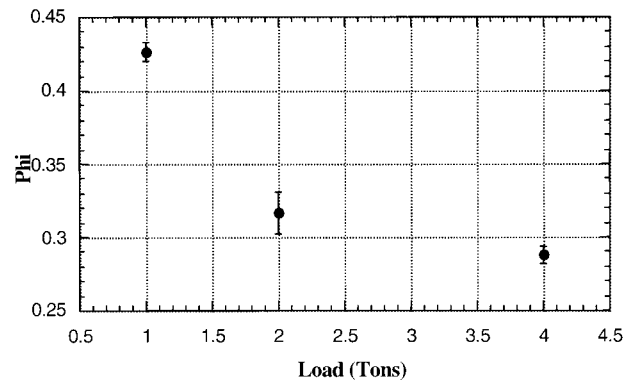


**Figure 4** Effect of the processing pressure on the fiber modulus.

was observed with an increase in the processing load (Fig. 4). For the first instance, this drop in the composite modulus can be understood as a direct result of the observed drop in the fiber modulus. The calculated interfacial adhesion parameter for composites processed under different processing loads (Fig. 5) showed that  $\Phi$  decreased as the processing load increased. This indicates that the observed drop in the composite modulus was a combined effect of a drop in the fiber modulus and a drop in interfacial adhesion. The observed drop in the fiber modulus may be because of a loss of molecular orientation in the fiber. Moreover, at applied loads less than 1 ton, not enough matrix penetration took place, and a composite could not be fabricated.

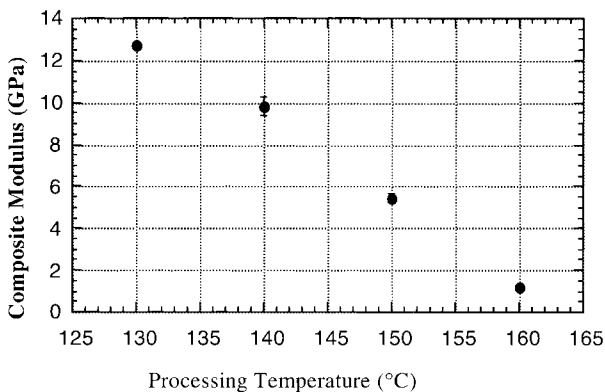
**Effect of the Processing Temperature**

For HDPE matrix self-reinforced composites, the possible processing temperature range was found

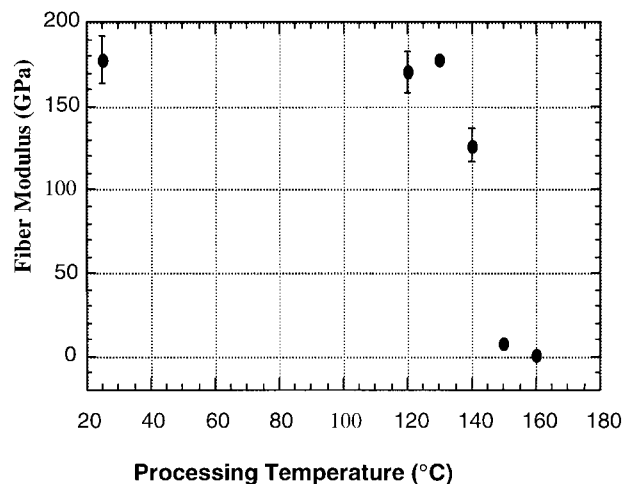


**Figure 5** Calculated interfacial adhesion parameter at different processing loads.

to be between 130 and 160°C. Below 130°C, the composite could not be manufactured because of incomplete melting of the matrix materials. However, at temperatures above 160°C, the reinforcing fiber completely melted, and the processed sheet did not contain any oriented molecules for reinforcement. The composite modulus showed a linear drop with an increased processing temperature in the range 130–160°C, as shown in Figure 6. The observed drop in the composite modulus was a direct result of the drop in the fiber modulus, which showed a linear drop with temperature increases above 130°C, as shown in Figure 7. The observed dependence of the fiber modulus on the processing temperature is reported here for the first time. The observed reduction in the fiber modulus can be understood on the basis of the surface cracks that were a direct result of fibrils shrinking away from one another because of molecular orientation disorder (Fig. 8). At 160°C (the melting point of fibers), the molecular orientation was mainly lost, and the fiber modulus was down to a matrix modulus of 1.15 GPa. The interfacial adhesion parameter,  $\Phi$ , shows that at temperatures up to 140°C, the same level of adhesion was maintained at 0.33. As the processing temperature increased, however, the partial melting of the fiber allowed better adhesion with the matrix. At 160°C, both the fiber and matrix became one phase;  $\Phi$  went to 1 (Fig. 9). Furthermore, these results on the linear low-density polyethylene matrix show that high-quality composites can be processed at lower temperatures (120°C). Such results indicate that the lower the processing temperature is, the better the composite modulus will be, as long as the melt viscosity of the matrix allows processing at such temperatures.



**Figure 6** Effect of the processing temperature on the composite modulus.



**Figure 7** Effect of the processing temperature on the fiber modulus.

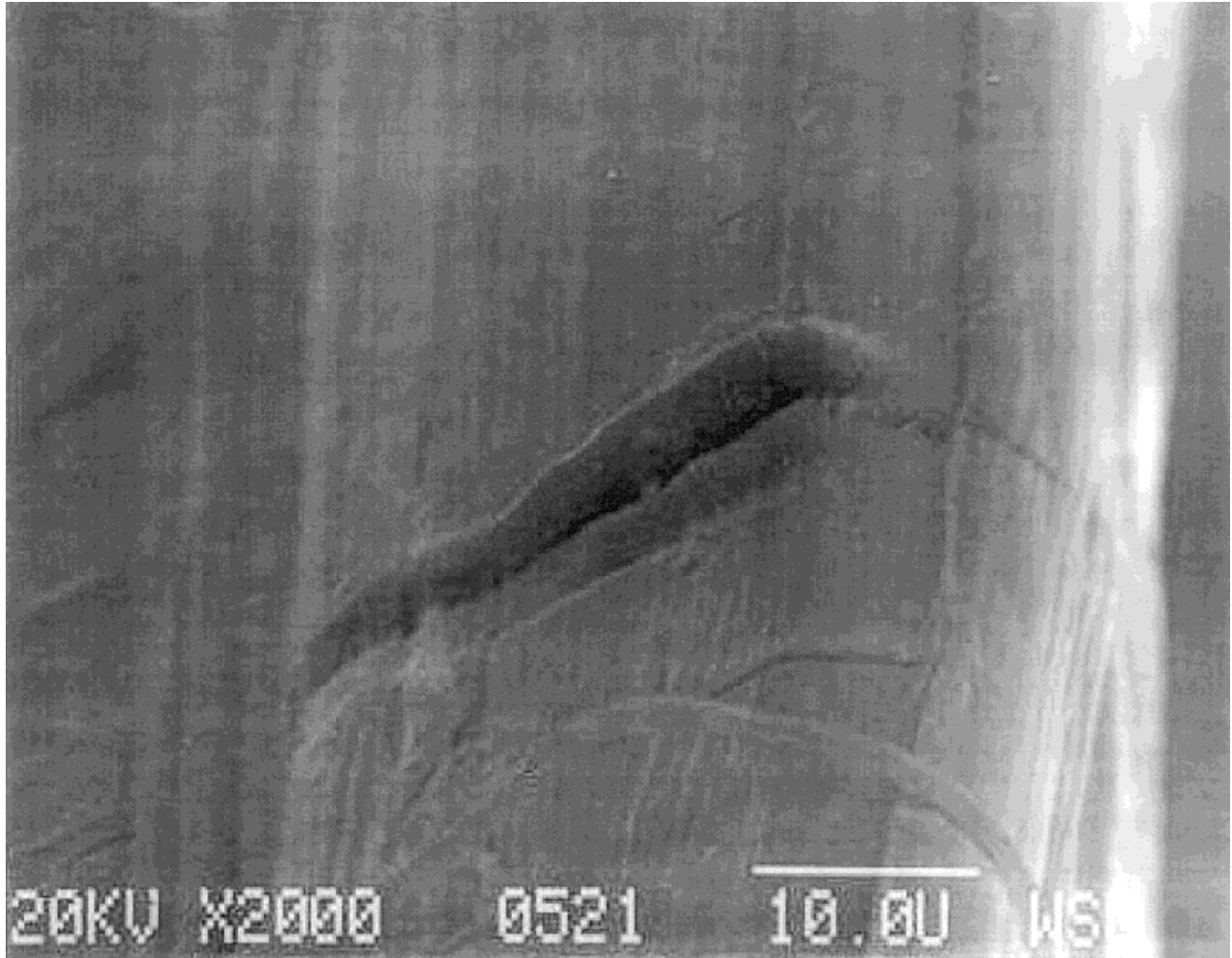
### Effect of the Processing Duration

The composite modulus increased as the processing duration increased up to 2 h. Further increases in processing duration resulted in a decrease in the composite modulus (Fig. 10). This may have resulted from either a similar dependence of the fiber modulus on processing duration and/or a similar dependence of interfacial adhesion. The fiber modulus showed almost no change (within the experimental error) on the processing duration within the duration limit investigated.  $\Phi$ , however, showed that interfacial adhesion in the composite increased up to 2 h and then decreased (Fig. 11). The increase in interfacial adhesion corresponding to processing duration can be understood from the viewpoint of an increase in the interdiffusion of fiber/matrix molecules as the processing duration increased. The subsequent reduction in the interfacial adhesion is not yet very well understood. PE oxidation and/or degradation at the processing temperature and pressure can be, however, thought of as a possible reason for the observed reduction.

### CONCLUSIONS

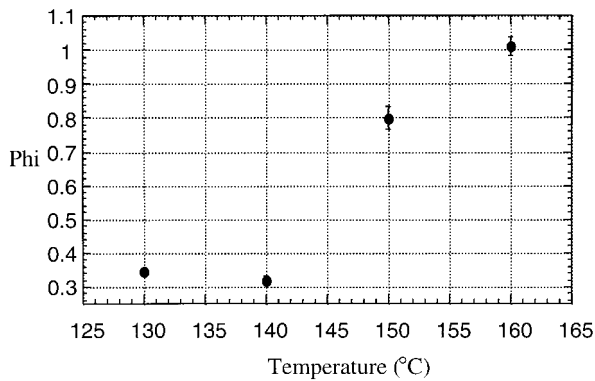
On the basis of the aforementioned results and discussion, the following conclusions can be drawn:

1. The Spectra® fiber modulus is dependent on the processing temperature. Processing



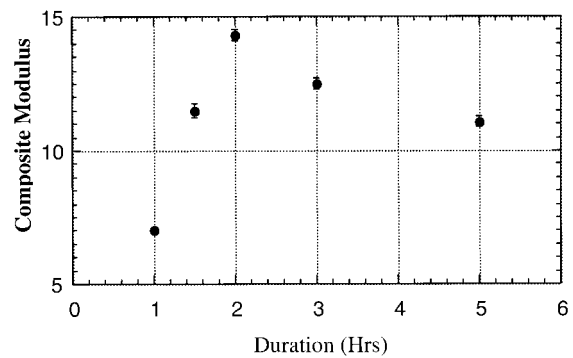
**Figure 8** Surface cracks observed in a Spectra fiber treated at 130°C.

above 130°C causes a large reduction in the fiber modulus.  
 2. The optimum processing temperature for a

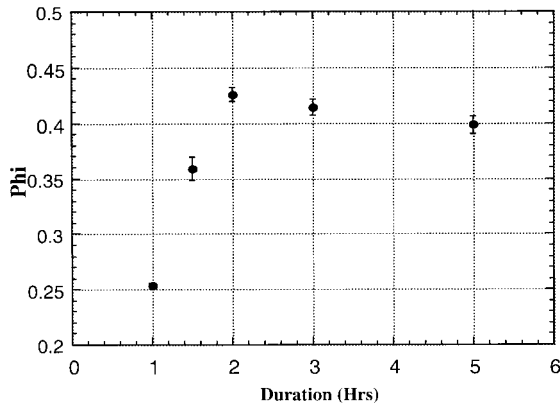


**Figure 9** Interfacial adhesion parameter as a function of the processing temperature.

self-reinforced PE depends on the type of PE matrix used and increases as the polymer melt viscosity increases.  
 3. The fiber modulus decreases as the pro-



**Figure 10** Composite modulus as a function of the processing duration.



**Figure 11**  $\Phi$  as a function of the processing duration.

cessing load increases. Hence, the optimum processing load is the minimum load sufficient for matrix penetration to the fibers.

4. There is an optimum processing duration that is matrix-dependent. The optimum duration is 2 h for HDPE.
5. The fact that an optimum duration was observed may draw attention to a possible

phase separation that takes place at the interface beyond the optimum processing duration.

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