

# Flexure and Water Sorption Properties of Wood Thermoplastic Composites Made with Polymer Blends

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**ABSTRACT:** Most WPC extrusion manufacturers utilize single polymer systems, where only one class of polymers is utilized. The concept of utilizing blended polymer systems can potentially provide lowered material costs, while maintaining or improving the composite properties. This cost savings can potentially be obtained in the ability to utilize mixed recycled plastics without the added costs of separation. Composites made of wood flour and binary blends of polypropylene (PP), high density polyethylene (HDPE), and polyvinyl-chloride (PVC) were extruded and evaluated for their mechanical and physical performance. The WPCs were also exposed to repeated extrusion runs to determine the influence of enhanced melt-blending of

the composite properties. Torque rheometry, flexural, and water sorption tests revealed subtle differences between the blends and single polymer composites. With increasing extrusion runs, the WPCs water diffusion was decreased, whereas the strength and stiffness of the blends and single polymer systems increased or showed no change. These results indicate that polymer blends can be successfully utilized for commercial WPCs. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 120: 1034–1039, 2011

**Key words:** blends; miscibility; wood plastic composites; torque rheometry; extrusion

## INTRODUCTION

The robust processing window associated with wood thermoplastic composite (WPC) extrusion has afforded many commercial manufacturers the ability to use a wide spectrum of resins. “Wide spec” and “off-spec” resins which fail quality control evaluations for narrow processing window methods, i.e., blow molding, film stock, etc., can be easily incorporated within a WPC extrusion process with minimal impact on production. This lack of process sensitivity, albeit in a relative sense, allows for the implementation of these wide-spectrum polymer systems to be utilized. Another attractive attribute of WPCs is their high level of recycled materials, with some manufacturers utilizing up to 90%. These composites primarily use recycled wood as well as some or all recycled thermoplastics. Currently, HDPE, PP, and PVC are the most commonly used plastics throughout the WPC industry.<sup>1</sup> The relatively low processing temperature of these commodity resins allows for the use of thermally sensitive lignocellulosic materials.

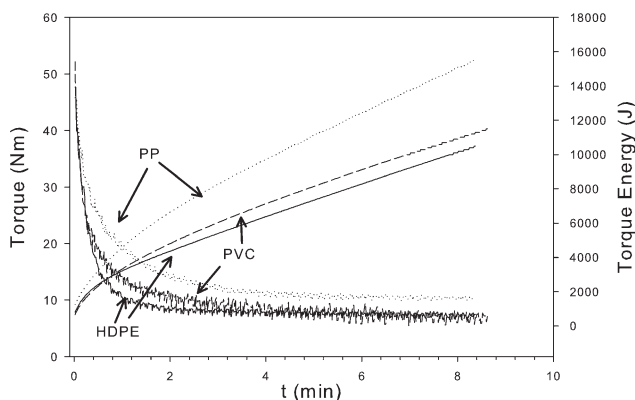
A majority of WPC and other polymer product manufacturers utilize a single polymer species. How-

ever, due to inherent processing flexibility, WPC's are likely candidates for the use of polymer blends. Polymers are generally immiscible due to the low combine entropy of the system preventing dispersion at a molecular level.<sup>2</sup> However, the separate phases within immiscible blends can impart a composite action that is comprised of the individual polymer attributes. The concept of designed polymer blends and the ability to reduce costs by accepting a range of polymer types into a manufacturing environment, provides incentives for using these alloys.

Blends of PP and HDPE have been studied extensively and have displayed good mechanical and physical properties.<sup>3–7</sup> The use of coupling agents and compatibilizers can aid in the miscibility of the blended polymers, which improves the final properties. The miscibility and morphology of a blend directly influences its mechanical properties.<sup>2,4,6</sup> Another option for improving the dispersion of polymers is through intensive mixing. With the high viscosities found in WPC melt blends, higher torque energy is required to mix the wood flour and the polymers, putting more shear energy into the mixture. This high shear energy could potentially aid in the improved dispersion of the polymers blends throughout the composite.

The objectives within this study were to evaluate the role of melt blending on the final properties of a wood filled binary blend of HDPE, PP, and PVC. Repeated extrusions were utilized to blend the

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**Figure 1** Torque rheometry data for the wood filled polymer melts.

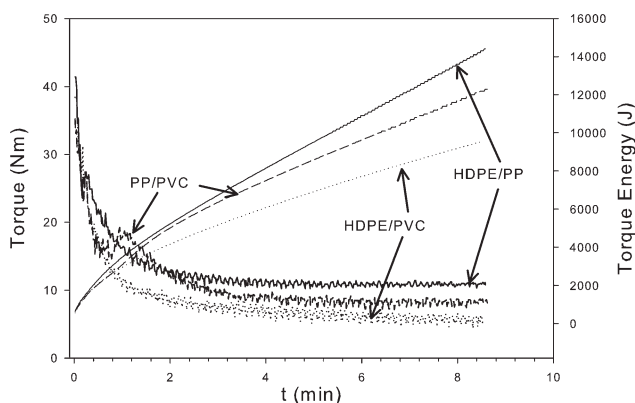
materials and static flexure, and water sorption tests were performed to evaluate the mechanical and physical performance of the composites, respectively. Torque rheometry was also performed on the polymer blends and wood flour to characterize the melt-blending behavior of the composites.

**MATERIALS AND METHODS**

Three thermoplastic resins; HDPE, PP, and PVC were used individually and in binary blends for wood plastic composites. All three polymers were in a granulated flake or powder form. The melt flow index for the HDPE and PP were 0.5 and 4.0 g/10 min, respectively. The wood flour utilized was a commercial 60-mesh eastern white pine (*Pinus strobus*), which was dried to a moisture content below 2%. Lubricants, ethylene bis stearamide (EBS) wax, and zinc stearate were added to the extrusion blends for improved processability of the composites.

**Torque rheometry**

A 120 cc torque rheometer chamber was used with roller rotor mixing elements. No lubricants were added



**Figure 2** Torque rheometry data for the wood filled polymer blends.

to the 60 : 40 wood polymer formulations. Two groups of specimens were run: binary polymer blends and wood-polymer composites. The binary polymer blends were used at a 50 : 50 ratio in both the wood-filled and non wood composites. The polymer and wood flour were dry blended by hand and fed into the torque rheometer and mixed for 10 min at screw speed of 20 rpm and a temperature of 180°C. For each formulation, three samples were run on the torque rheometer. A representative curve was taken from each sample set and used for graphical comparisons in Figures 1 and 2.

**Extrusion**

A 35-mm conical counter-rotating twin-screw extruder was used to process the blended composites. The individual components were weighed and dry-mixed in a drum blender according to the formulations outlined in Table I. The dry-blend was then fed into the extruder, profiled to a 38 × 9.5 mm x-section, and cooled via a water spray tank. The extruder, screw, and profile die temperatures were held constant at 180°C, while the screw rate was 20 rpm for the first dry-blended extrusion. The material had a dwell time of 3–5 min under an amperage load ranging from 6 to 12 (22 amps is the manufacturers suggested maximum). Flexure and water sorption test specimens were cut randomly throughout the run. The remainder of the material was then exposed to further melt-blending by granulating with a knife mill with a 4.7-mm screen and then fed through the extruder to make the same profiled composite. This process was repeated another two times for a maximum of four extrusions. To keep the volumetric output similar to the dry blend extrusion, the granulated material was processed at a lower screw rate of 10 rpm.

**Flexural testing**

Flexure tests were conducted in accordance with ASTM D 790<sup>8</sup> requirements. Six samples of a nominal 203 × 38 × 9.5 mm dimension were stored 24 h in a 20°C and 50% relative humidity conditioning room. The single-point test span was set at 152 mm based upon a span : depth ratio of 16. A universal testing

**TABLE I**  
Weight Percent Composition of the Extruded WPCs

	Blend 1 (%)	Blend 2 (%)	Blend 3 (%)	Blend 4 (%)	Blend 5 (%)	Blend 6 (%)
Wood	58	58	58	58	58	58
HDPE	39			19.5	19.5	
PP		39		19.5		19.5
PVC			39		19.5	19.5
Zinc stearate	2	2	2	2	2	2
EBS wax	1	1	1	1	1	1

apparatus was utilized for the test, while load and deflection were collected in real-time and used to calculate the stiffness (MOE), maximum bending stress (MOR), and the strain at break ( $\epsilon_{\text{break}}$ ).

### Water sorption

The composites obtained from the four extrusions were planned to a thickness of  $\sim 6$  mm to remove the polymer-rich surface of the boards and cut into specimens of  $127 \times 25$  mm. The samples were then immersed in distilled water at ambient temperature ( $20^\circ\text{C}$ ) for 16 weeks. The thickness and weight of the samples were measured throughout the 16 week duration to determine their thickness swell and water sorption, respectively. Before any measurement, the surface water was wiped away to avoid any additional mass. Diffusion coefficients were determined for the composites based upon a Fickian diffusion model.<sup>9</sup>

### Statistical analysis

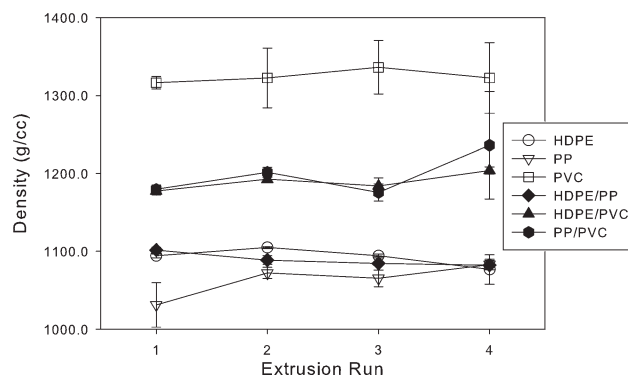
An analysis of covariance (ANCOVA) was run to analyze the flexural data. Density was used as a covariate in determining the significance of extrusion runs on the MOE, MOR, and  $\epsilon_{\text{break}}$ .

## RESULTS AND DISCUSSION

### Torque rheometry

To minimize the variables within the experimental design, a consistent temperature of  $180^\circ\text{C}$  was utilized for the extruder and torque rheometry profile. To assess the mixing performance of the wood filled polymer and polymer blends, torque rheometry was utilized. Although torque rheometry is not the most accurate measure of a polymers rheological properties, it does provide a realistic interpretation of the processing characteristics, especially for extrusion.<sup>10,11</sup> This procedure measures the torque or energy needed to melt-blend a polymer system or composite and reflects the resistance of the material to shear deformation during the mixing process. Torque-time curves provide an equilibrium torque ( $T_e$ ) and graphically represent the melt blending characteristics. The convergence of the torque curve to a consistent value represents the  $T_e$ , which indicates when the polymer has completely melted or softened and when the fillers have reached maximum dispersion.

With PP having the highest processing temperatures ( $180$ – $250^\circ\text{C}$ ) of the three polymers, an expected longer mixing time to reach  $T_e$  was anticipated. The results found in Figure 1 show the torque values for the single polymer wood filled composites. The HDPE composite showed the lowest energy



**Figure 3** Density values for the various WPCs exposed to repeated extrusions.

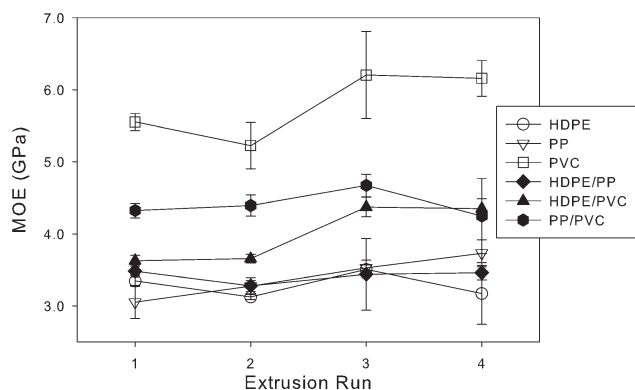
required for melting and dispersing the wood flour and also the quickest time to reach  $T_e$ . PP composites had a substantially higher energy requirement and also a longer dwell time prior to converging to  $T_e$ , however after 3 min the PP composites did reach  $T_e$ , which is similar to the estimated time within the extrusion barrel.

Interestingly, the PVC-wood filled composite did not exhibit a characteristic increase in torque due to fusion in PVC-wood or in the HDPE/PVC-wood material. However, the PP/PVC-wood blend did show a very distinct and quite repeatable fusion peak as seen in Figure 2. Matuana and Kim<sup>12</sup> found changes in the fusion characteristics when various wood loadings and particle size distributions were used in their torque rheometry study. PVC formulations can be significantly different, and often proprietary, therefore comparisons between different PVC resins is difficult. The PVC utilized in our study may have a more subtle fusion that cannot be detected with the torque rheometer until it was enhanced by the presence of PP. The torque load and energy was the highest for the PP based blends, with the HDPE/PVC having the lowest energy and quickest convergence to a  $T_e$ .

### Flexural performance

The flexural performance and density were measured using the extruded samples. The values for density, MOE,  $\epsilon_{\text{break}}$ , and MOR are represented graphically in Figures 3–6, respectively, based upon the enhanced mixing through increased extrusion runs. Overall, the PVC-based composite showed the highest MOE and MOR, while also having the lowest strain values.

Density values for WPCs often show limited variability when extruded or processed in a consistent manner, however wood composites in general can be dramatically altered with slight density fluctuations. The composites extruded within this work showed density had positive correlation with

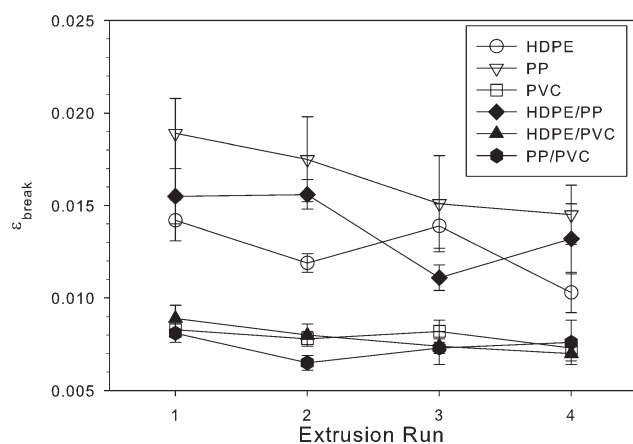


**Figure 4** MOE values for the various WPCs exposed to repeated extrusions.

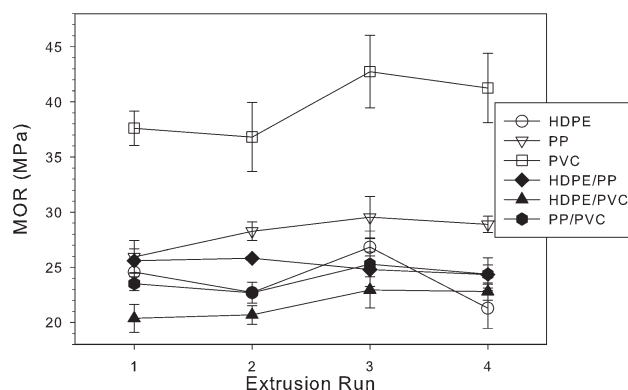
repeated extrusion runs for all of the PP-based composites. Only the single polymer systems of HDPE and PVC did not show a significant influence from density due to additional extrusions. The PVC showed a generally trend to a higher density, however the variability within the data provided no statistical significance.

The MOE of the WPCs was found to increase slightly with additional extrusion runs. The PVC based WPCs showed the most dramatic increase in stiffness, with the single polymer system improving the most. The PP and HDPE WPCs showed some limited improvement. Statistical analysis also showed a significant effect when density was run as a covariate. For the MOE results, density significantly influence all of the single polymer composites and the HDPE/PVC blended composite. The number of extrusion runs significantly influenced the PP, PVC, HDPE/PP, and HDPE/PVC composites with increasing values after repeated runs.

The  $\epsilon_{\text{break}}$  often indicates the toughness or brittle nature of a composite. With all of the WPCs, a decrease in  $\epsilon_{\text{break}}$  was seen with an increase in extrusion runs indicating an enhancement of a brittle



**Figure 5** The  $\epsilon_{\text{break}}$  values for the various WPCs exposed to repeated extrusions.



**Figure 6** MOR values for the various WPCs exposed to repeated extrusions.

behavior. These results are similar to Abad et al.,<sup>13</sup> in their work with multiple extruded PE's with stabilizers. The PP and HDPE homopolymer and blends showed the most dramatic decrease in strain, while the PVC composites remained lower but showed little decrease with increased extrusion runs.

The bending strength was also significantly altered with repeated extrusion runs (Fig. 6). A general trend of improved strength with increased extrusion runs was observed in all cases except the HDPE/PP and HDPE WPCs. The PVC-based WPCs exhibited the most change, however the variability was also quite substantial, causing the extrusion effect to be statistically non-significant.

The additionally extrusion runs will likely cause an increase in thermal, oxidative, and mechanical degradation that will cause chain scission, which results in a reduction in molecular weight (MW) or subsequent chain branching.<sup>14,15</sup> PP generally shows a reduction in MW during thermal processing, whereas HDPE can show an increase or decrease, depending upon the type and the amount of branching formed and the potential for crosslinking.<sup>14,16</sup> The degradation differences between processing PP and HDPE can also be observed in their final properties with PP showing significant reduction in properties with repeated extrusions and HDPE showing limited or no change in structural performance.<sup>13,17</sup>

### Moisture sorption

The repeated extrusions resulted in diffusion behavior changes for all of the WPCs, with the blended polymer systems having the greatest change as seen in Figure 7. The percent of sorbed water was based upon the initial weight of the composite. When looking at the influence of the number of extrusion runs on the final moisture content ( $M_m$ ) of the various WPCs, the HDPE/PP composites leveled off to a similar range of values. However, the PP/PVC and the HDPE/PVC composites all showed a decrease in

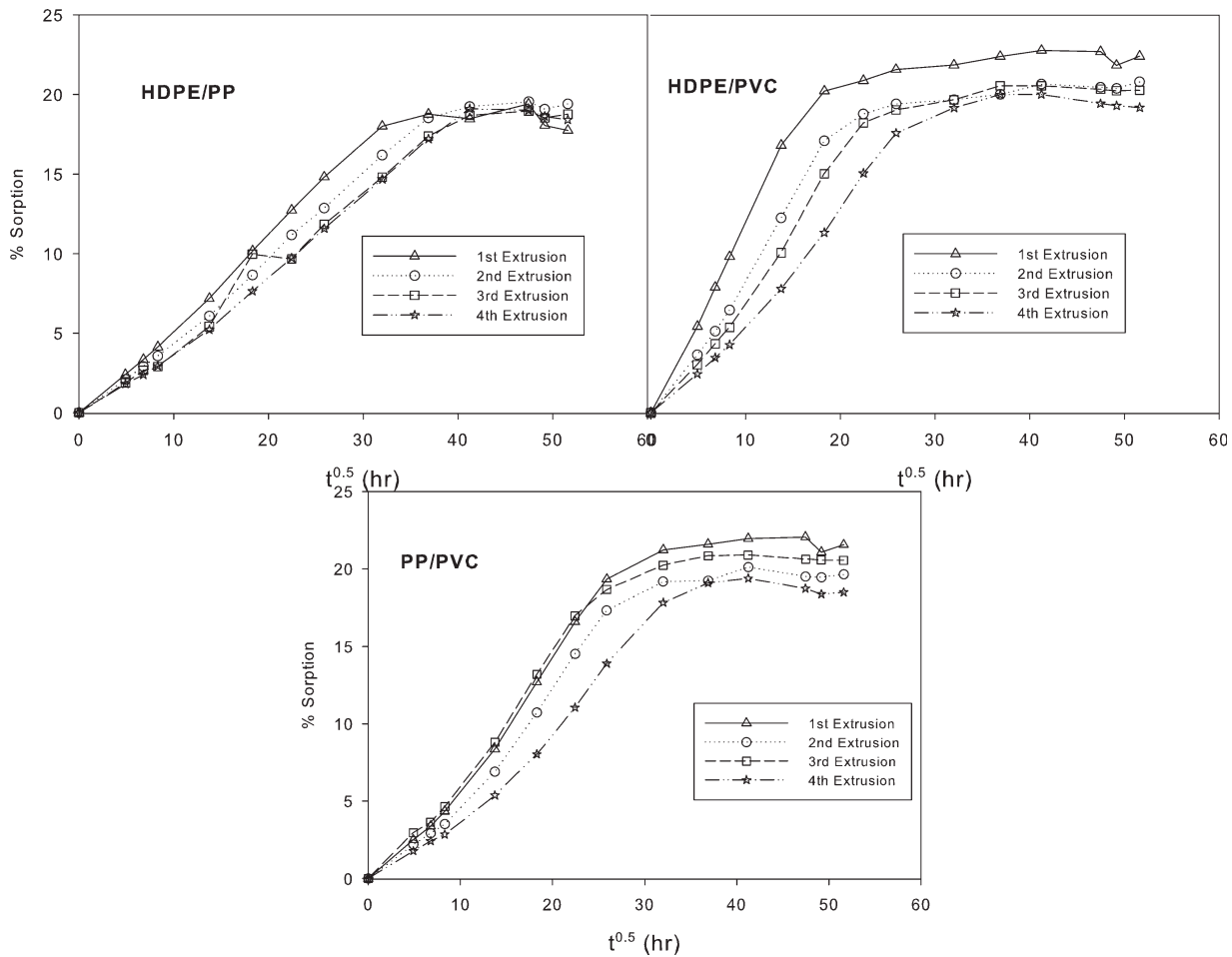


Figure 7 Percentage of water sorption with the blended polymer WPCs.

$M_m$  as they were exposed to more extrusions. This would likely indicate that these blends were not as sufficiently mixed, and that the repeated extrusions improved the dispersion and minimized the amount of total water uptake.

The sorption curves also showed a relatively linear behavior in the initial stages of the test when plotted with respect to the square root of time ( $t$ ) indicating that a Fickian diffusion model can be used to characterize the sorption behavior.<sup>18</sup> Fick's second law of diffusion can be briefly explained through eqs. (1)–(3). In a 1-dimensional solution for thin plates, the apparent diffusion ( $D_A$ ) can be described in eq. (1), where the thickness ( $h$ ) is substantially less than the length ( $l$ ) or width ( $w$ ) of the specimen.

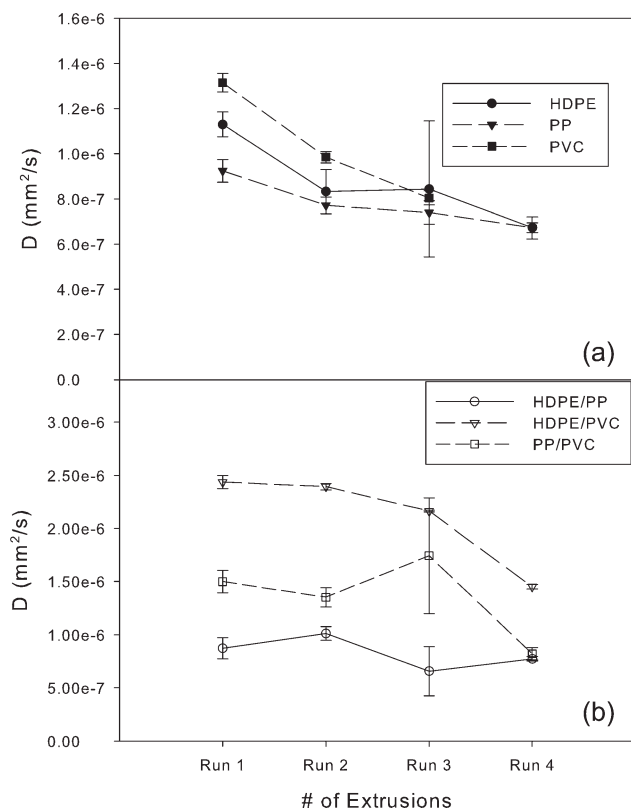
$$D_A = \pi \frac{h^2}{16M_m} \left[ \frac{dM_t}{d\sqrt{t}} \right]^2 \quad (1)$$

The  $D_A$  only looks at diffusion through the face of the plate, to compensate for sorption through the sides and ends an edge correction factor (ECF) is often applied [eq. (2)].<sup>9</sup> The resulting diffusion coefficient ( $D$ ) is then calculated through eq. (3).

$$ECF = \left[ 1 + \frac{h}{l} + \frac{h}{w} \right]^2 \quad (2)$$

$$D = \frac{D_A}{ECF} \quad (3)$$

The initial linear section of the sorption curves in Figure 7 can be attributed to the penetration of water into the hydrophilic wood flour and to the voids developed during the manufacture of the composites. In a sorption study by Chateauinois et al.<sup>18</sup> with glass fiber/epoxy composite they attributed the initial sorption to penetrate water into the epoxy resin and the voids between the fiber and matrix. Najafi et al.<sup>7</sup> attributed the water sorption mechanics of WPCs to sorption by capillarity because of gaps and flaws at the interface between wood and polymers, in addition to the natural sorption of the wood particles themselves. The latter nonlinear section of the curve the sorption mechanisms are likely associated to the water filling of the sorption-induced voids. Carfagna and Apicella found that with thin epoxy sheets, there were two distinct mechanisms of sorption where the initial



**Figure 8** Diffusion coefficients for the single (a) and blended (b) polymer WPCs with respect to extrusion runs.

stage results in no damage to the material and the second stage results as a consequence of damage to the material structure.<sup>19</sup> Chateauminois et al. concurred with this secondary damage in their work with composite structures.<sup>18</sup>

With the repeated extrusion runs, all single and blended polymer WPCs exhibited a decreasing trend in  $D$  as seen in Figure 8. The single polymer systems appeared to be influenced more dramatically than the blended materials. This decreasing trend is likely attributed to the improved dispersion of the wood flour within the polymer matrix and potentially the reduction of voids throughout the composite structure.

## CONCLUSIONS

The repeated extrusion runs were utilized to determine the influence of melt-blending on the flexural and water sorption properties of single and blended polymer systems. As the extrusion runs increased, flexural properties trended slightly higher or remained the same for the MOE and MOR. However, the  $\epsilon_{\text{break}}$  did show significant reductions indicating the development of a more brittle composite.

Although some improvement was seen in mechanical properties during the additional melt-blending, the polymer blends showed similar properties as the single polymer systems, indicating that the need for more intensive mixing may not be required for WPCs.

An improvement in water resistance was observed with increasing extrusions, which likely indicating enhanced wood flour dispersion and the minimization of voids and capillary pathways. The additional melt-blending showed lower  $D$ , especially in the single polymer systems, which could potentially assist in the protection of WPCs when exposed to moisture and subsequent bio-deterioration.

Using polymer blends to create WPCs did not show any particular disadvantage. The mechanical and physical properties observed depended on the components of the blends, not the fact that polymer blends were used. WPCs containing polymer blends often exhibited intermediate properties that depended on the blends they were made of, leaving no real objection to the use of several polymers to manufacture a WPC.

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