Role of Paper Sludge Particle Size and Extrusion Temperature on Performance of Paper Sludge–Thermoplastic Polymer Composites

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ABSTRACT: The effect of paper sludge's particle size and extrusion temperature on the physical and mechanical properties of paper sludge–thermoplastic polymer composites was investigated. In the experiment three levels of particle sizes for the paper sludge and four extrusion temperatures were designed to examine the physical and mechanical properties of these composites. The ash contents of the paper sludge were about 73.7, 46.2, and 38.1% with particle sizes of below 0.15, 0.18–0.25, and 0.42–0.84 mm, respectively, which meant lower ash content and higher cellulose fiber content, in the larger particle size of paper sludge. As the particle size of the paper sludge decreased, the swelling thickness, water absorption, and tensile and flexural strengths of the composite improved; but the particle size of the paper sludge had no effect on its unnotched impact strength. With the increase of the extrusion temperature the thickness swelling and water absorption of the composites were slightly improved but not statistically different. A rise of the extrusion temperature generally had a positive effect on the tensile and flexural properties of the composite. The notched and unnotched impact strengths of the composite increased with the increase of the extrusion temperature from 190 to 230°C, but they decreased slightly at an extrusion temperature of 250°C. This low impact energy at an extrusion temperature of 250°C may be attributed to the excessively brittle fibers from thermal decomposition. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 82: 2709–2718, 2001

Key words: paper sludge; particle size; extrusion temperature; thickness swelling; water absorption; tensile properties; flexural properties; notched and unnotched impact strengths

INTRODUCTION

The U.S. pulp and paper industry generates approximately 45 kg of sludge per ton of pulp. An annual pulp production of 80 million tons produces 4 million dry tons of sludge per year.[1] The disposal of sludges from the production of pulp and paper is a difficult environmental problem because most sludge is landfilled. Although some sludge is mixed with hog fuel and incinerated, some is composted and sold, and some is applied to forest lands, these are not common practices.[2]

On the other hand, the waste wood fibers separated from paper sludge, besides the conventional inorganic reinforcing fillers such as clay, talc, calcium carbonate, and so forth, also have the feasibility of being effective fillers in the manufacture of thermoplastic polymer composites.

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The use of reinforcing fillers for the reduction of material cost and improvement of composite performance is constantly increasing in the area of thermoplastic polymer composites.

In the manufacture of composites cellulose fibers have recently attracted considerable interest as reinforcing fillers for thermoplastic polymers with relatively lower melting points like polypropylene (PP) and high- (HDPE) and low-density polyethylene (LDPE). Sludge from paper mills consists of mainly two components, fine cellulose fiber and inorganic materials; it can offer a number of benefits as a substitute for the typical inorganic reinforcing fillers in the manufacturing of thermoplastic polymer composites.

These benefits, in comparison with typical inorganic fillers, include a high aspect ratio for the cellulose fiber and the resulting minimal abrasion of processing equipment, relatively low composite density, and low production cost on a unit volume basis.

This article reports the results of an investigation to determine the effects of the paper sludge particle size and extrusion temperature on the dimensional stability and tensile, flexural, and impact properties of paper sludge–thermoplastic polymer composites. In addition, thermal analysis and microstructural investigations of the composite were conducted to provide information about the particle size of paper sludges.

EXPERIMENTAL

Materials

The PP was in the form of homopolymer spheres with a density of 0.91 g/cm^3 and a melt flow index of 12 g/10 min (at 230°C with 2160 g). The high impact PP (HIPP) comprised ethylene-propylene random copolymer spheres with high impact strength and a low melt flow index of 0.5 g/10 min. The HDPE consisted of homopolymer spheres with a density of 0.957 g/cm^3 and a melt flow index of 15 g/10 min, and the LDPE was homopolymer spheres with a density of 0.918 g/cm^3 and a melt flow index of 20 g/10 min. The reinforcing filler in the composites was paper sludge from a newspaper-making mill; the particle sizes were below 0.15, 0.18–0.25, and 0.42–0.84 mm.

Processing

The paper sludge was dried at 80–100°C for 24 h to adjust it to a moisture content of 1–2% and then stored over desiccant in sealed containers. Four types of thermoplastic polymers were blended with paper sludge or coupling agents in a two-roll rheomixer. Mixing was continued at 200°C for 15 min with a rotor speed of 20 rpm. A laboratory-size single-screw extruder was employed to compound the paper sludge with the PP matrix at 190, 210, 230, and 250 °C and with the PE matrix at 150, 170, 190, and 210°C. The extruded strand was pelletized and stored in sealed cans containing desiccant. Test specimens were prepared using an injection molding machine at 200°C, an injection pressure of 1200 psi, and a device pressure of 1500 psi. After molding, the specimens were stored over desiccant at room temperature for at least 7 days before testing.

Testing

The dimensional stability of the composites was according to ASTM D1037-95. The tensile, flexural, and Izod impact strength tests were conducted according to ASTM D638-91, D790-91, and D256-90, respectively. The crosshead speeds during the flexural and tension testing were 2 and 5 mm/min, respectively. An elemental analyzer was used to analyze the components by the particle size of the paper sludge. A thermogravimetric analyzer was used to determine the ash content and thermal decomposition temperature of the paper sludge. The heating rate was 20°C/min. A scanning electron microscope (Jeol JSM-5410LV) and an energy dispersive X-ray spectrometer were used to observe the distribution of cellulose fiber and inorganic materials by the particle size of the paper sludge and to analyze the inorganic elements of the fracture surfaces of the tensile samples.

RESULTS AND DISCUSSION

Ash Content and Elemental Analysis by Particle Size of Paper Sludge

The ash contents of the paper sludge according to the particle size are shown in Table I. The table shows that the ash contents of the paper sludge were about 73.7, 46.2, and 38.1% in particle sizes below 0.15, 0.18–0.25, and 0.42–0.84 mm, respectively, which meant a lower ash content and thus a higher cellulose fiber content with the larger particle size of the paper sludge.

Table I Ash Content and Analysis of Paper Sludge Elements by Particle Size

Particle Size (mm)	Ash Content (%)	Carbon (%)	Hydrogen (%)	Oxygen (%)	Nitrogen (%)	Sulfur (%)
Below 0.15[a]	73.7	23.4	3.27	72.5	0.583	0.188
0.18–0.25[b]	46.2	25.4	3.45	70.5	0.564	0.155
0.42–0.84[c]	38.1	45.3	6.37	47.8	0.479	0.000

[a] Below 100 mesh.
[b] 60–80 mesh.
[c] 20–40 mesh.

Figure 1 shows the thermogravimetric curves of the paper sludge according to the particle size by thermal analysis. The figure also confirms that the ash content was increased with the decrease in particle size of the paper sludge.

Figure 2 shows a scanning electron micrograph and energy dispersive X-ray spectrometric curve of the paper sludge–PP composite. According to these results, we observed peaks for Al, Si, and Ca, which were derived from inorganic materials such as clay, talc, and calcium carbonate that were added during the paper-making process.

Effect of Paper Sludge Particle Size on Dimensional Stability

The dimensional stability of paper sludge–thermoplastic polymer composites as a function of the particle size of the paper sludge is shown in Figures 3 and 4. Generally, the thickness swelling and water absorption of the composites were increased with a decrease in the paper sludge particle size. The values of thickness swelling and water absorption of the paper sludge–thermoplastic polymer were very low because of the hydrophobic thermoplastic polymer matrix. However, there were some differences in the dimensional stability of the composites according to the paper

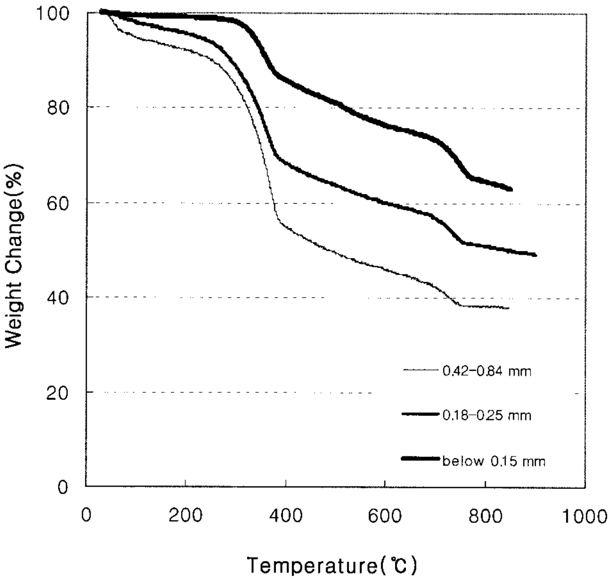

Figure 1 The thermogravimetric curve of paper sludge by its particle size.

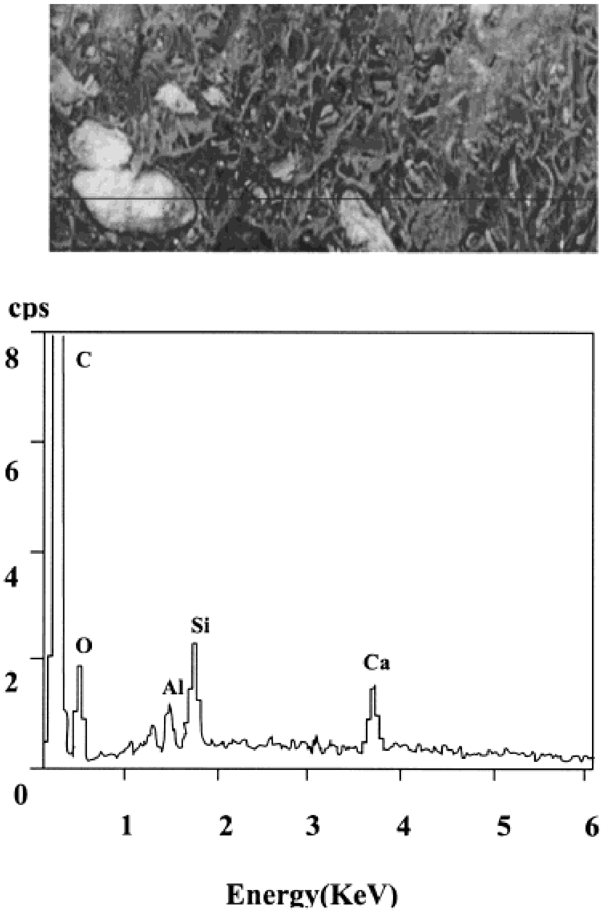

Figure 2 A scanning electron micrograph and energy dispersive X-ray spectrometric curve.

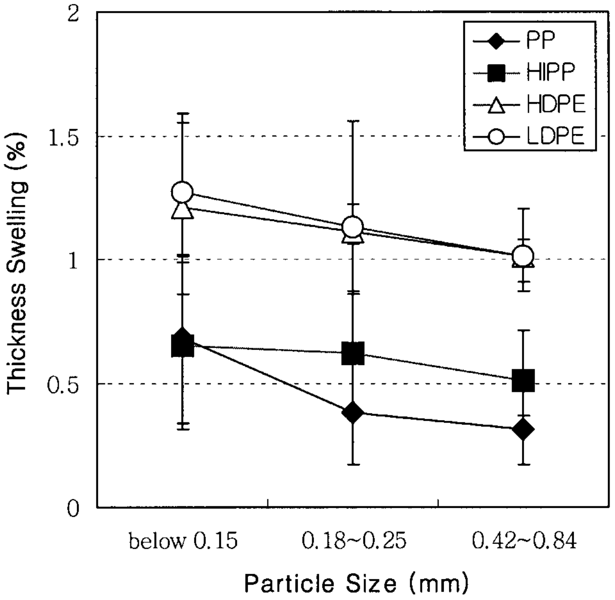

Figure 3 The thickness swelling of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the particle size of the paper sludge.

sludge particle size because the smaller particle sizes of the paper sludge, which consisted of mostly inorganic materials, had higher porosity than the larger particle sizes, which consisted of mostly fine cellulose fibers. Also, the thickness swelling of the paper sludge–PE composites was higher than that of paper sludge–PP composites. This was attributed to the weak interfacial adhesion between the PE chain and the paper sludge fiber, judging from the tensile and flexural property data of the paper sludge–thermoplastic polymer composites.

The relationship among the water absorption, thickness swelling, and particle size of the paper sludge is shown in Figure 4. In general, the thickness swelling of the paper sludge–thermoplastic polymer composite increased as the water absorption increased. Also, the coefficient of regression between the water absorption and thickness swelling of the composites filled with the largest particle size (0.42–0.84 mm) of paper sludge had the highest value. Because there might have been still higher cellulose fibers in the larger particle size of the paper sludge, the thickness swelling of the paper sludge–thermoplastic polymer composite increased as the water absorption increased according to general principles. However, the thickness swelling of the thermoplastic polymer composite filled with the smallest particle size of paper sludge showed little increase as the water absorption increased, which was due to a higher level of inorganic materials in the smaller particle size of the paper sludge, as previously mentioned in Table I.

Effect of Paper Sludge Particle Size on Mechanical Properties

The inset in Figure 5 shows the tensile strength of PP filled with paper sludge by its mixing ratio. As shown in the figure, the tensile strength decreased slightly with a rising mixing ratio of paper sludge.[3] In line with this tendency, Figures 5–8 show the tensile and flexural properties of thermoplastic polymer filled with 20 wt % paper sludge, according to the particle size of the paper sludge. Even though there were some outlying data, generally the tensile and flexural strengths and modulus of the composite increased as the particle size of the paper sludge increased. The higher observed values for the tensile and flexural properties in the larger particle size of the paper sludge may be attributable to the presence of many fine cellulosic fibers in the larger particle sizes. Another explanation might be a reduction of inorganic materials in the paper sludge that had a larger particle size. In other words, as shown schematically in Figure 9, there might be a good probability of interfacial adhesion linkages between thermoplastic polymer chains and cellulose fiber chains in paper sludge because of less

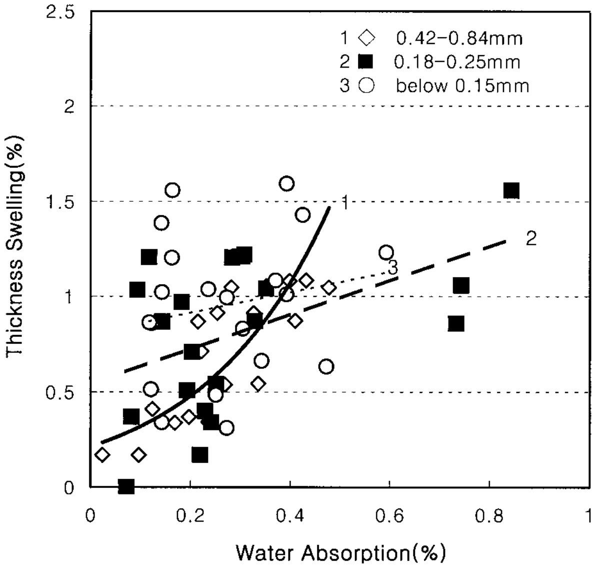

Figure 4 The relationship between the water absorption and swelling thickness according to the particle size of the paper sludge.

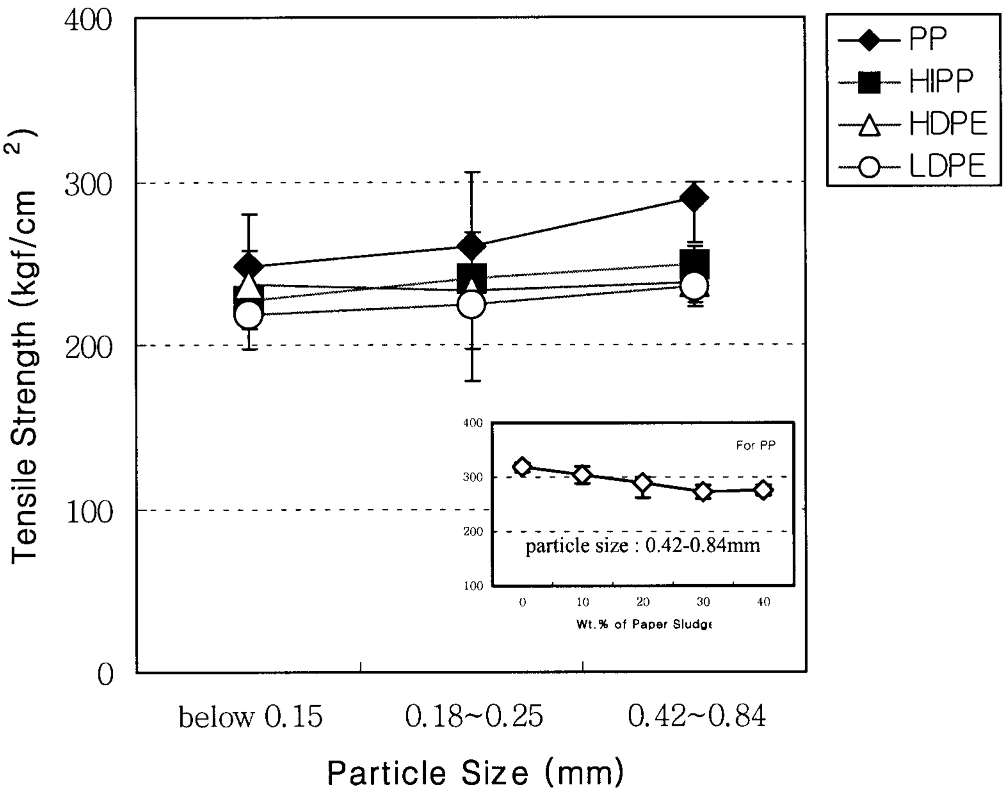

Figure 5 The tensile strength of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the particle size of the paper sludge.

dense inorganic materials in paper sludge, which has a larger particle size.

According to Myers et al.'s research,[4] on the other hand, the tensile strength and modulus were significantly increased by decreasing the particle size of wood flour for wood flour–PP (45/55 wt %) composites. Given good particle dispersion and some degree of particle–matrix bond-

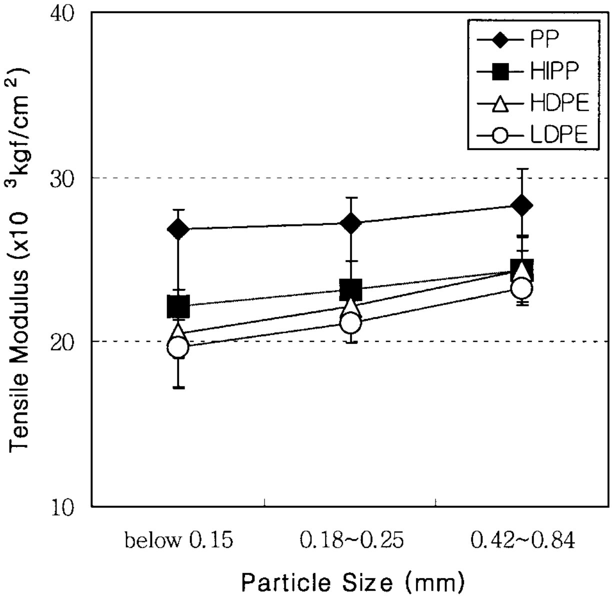

Figure 6 The tensile modulus of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the particle size of the paper sludge.

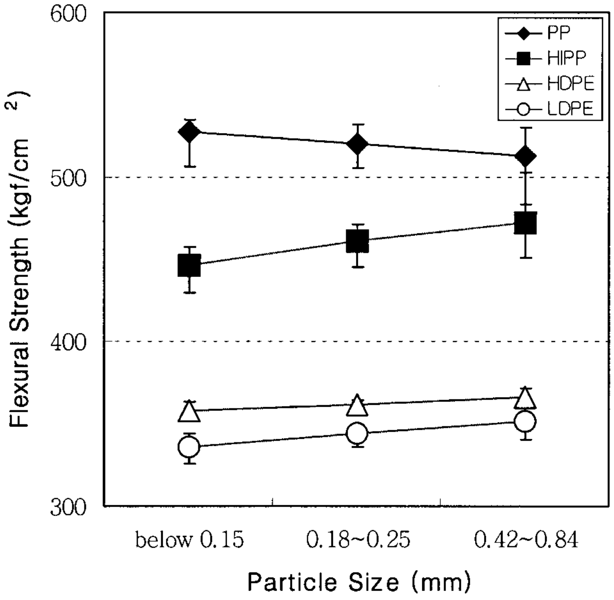

Figure 7 The flexural strength of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the particle size of the paper sludge.

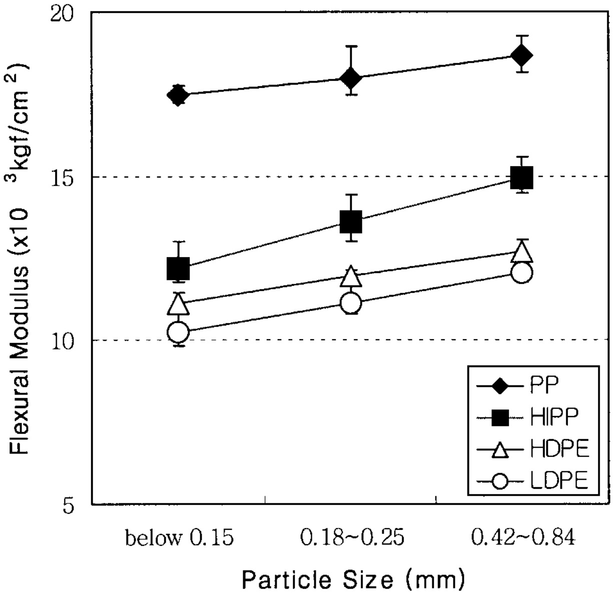

Figure 8 The flexural modulus of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the particle size of the paper sludge.

ing, they expected the larger particle system to possess greater stress concentrations and poorer particle–matrix stress transfer.[4] Their results were somewhat different than our results. This may be because of differences between the particle types in paper sludge and wood flour. The strength and modulus of a short fiber composite depends on several factors, including the fiber length, fiber orientation, fiber and matrix volume fractions, and the strength and modulus of both components. Because the matrix properties were assumed to remain the same regardless of the composite system, it was expected that all observed differences in the composite strength and modulus with different fiber types must be attributed to other factors such as the void content, void size, fiber length, and fiber orientation.

Figure 10 shows data on the composite notched impact strengths. The larger particle size of the paper sludge, which meant higher cellulose fiber contents in the paper sludge, increased the notched impact strength when the thermoplastic polymer was HIPP and HDPE. Meanwhile, an analysis of the unnotched impact strength data indicated that there was no trend with the paper sludge particle size according to thermoplastic polymer types. The existence of different trends for the two measurements of the impact strength was not surprising. The unnotched impact behavior is controlled to a considerable extent by fracture initiation processes that, in turn, are controlled by stress concentrations at defects in the system. By contrast, notched impact behavior is controlled to a greater extent by factors affecting the propagation of fracture initiated at the predominating stress concentration at the notch tip. For calcium carbonate filled PP composites the unnotched impact strength was shown to fall with increasing fractions of large particles or agglomerates, presumably as a consequence of the attendant stress concentration regions.[5] Thus, our observed decrease in the unnotched impact strength with paper sludge filled PP and HDPE may be attributable to a greater concentration of large particles (fiber type) or even agglomerates.

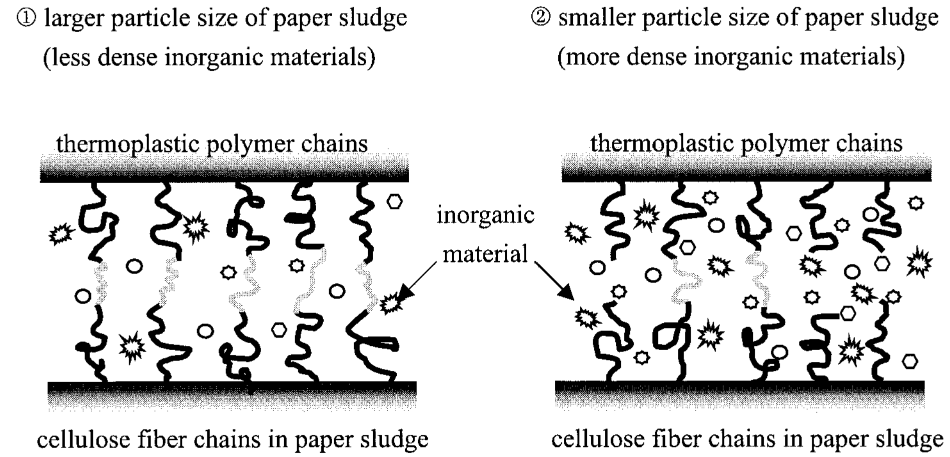

Figure 9 A schematic diagram showing the interfacial adhesion regions between the thermoplastic polymer chains and cellulose fiber chains in paper sludge according to the particle size of the paper sludge.

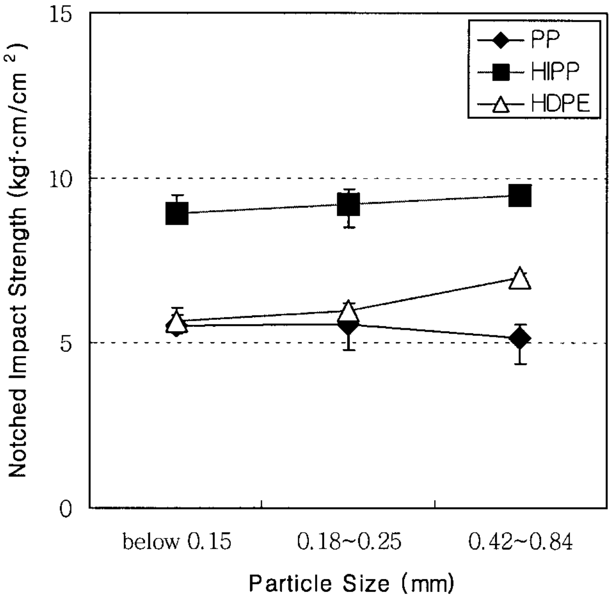

Figure 10 The notched impact strength of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the particle size of the paper sludge.

Effect of Extrusion Temperature on Dimensional Stability

Figures 11 and 12 show the dimensional stability of paper sludge–thermoplastic polymer composites as a function of the extrusion temperature.

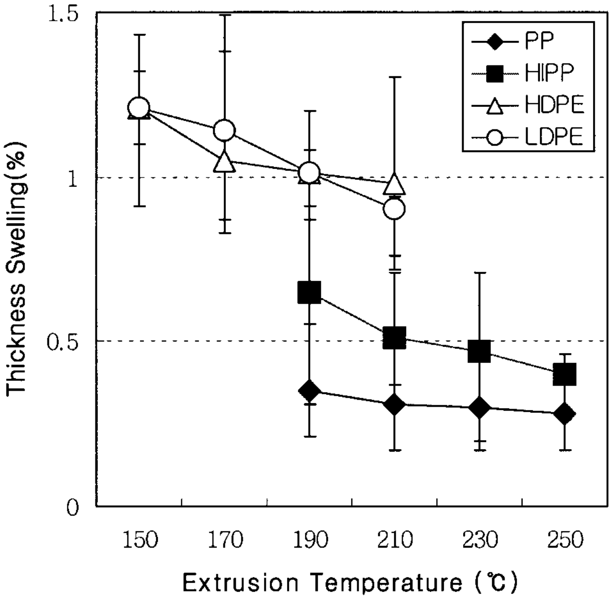

Figure 11 The thickness swelling of paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the extrusion temperature.

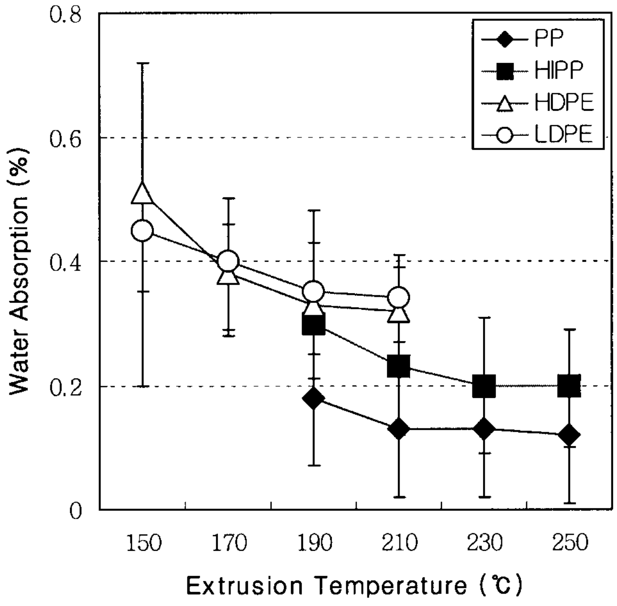

Figure 12 The water absorption of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the extrusion temperature.

For paper sludge–PP composites the values of the thickness swelling and water absorption were 0.28–0.35 and 0.12–0.18%, respectively, at extrusion temperatures from 190 to 250°C. For paper sludge–HIPP composites the value of the thickness swelling and water absorption were 0.40–0.65 and 0.20–0.30%, respectively, at extrusion temperatures from 190 to 250°C.

By contrast, the values of the thickness swelling and water absorption were 0.98–1.21 and 0.32–0.51%, respectively, with the increase of the extrusion temperatures from 150 to 210°C for paper sludge–HDPE. The values of the thickness swelling and water absorption were 0.90–1.21 and 0.34–0.45%, respectively, at extrusion temperatures ranging from 150 to 210°C for paper sludge–LDPE.

The dimensional stability of the paper sludge–thermoplastic polymer composites was somewhat improved with an increasing extrusion temperature, but they were not statistically different.

The thickness swelling of paper sludge–PE composites was also higher than that of paper sludge–PP composites. As shown schematically in Figure 13, this might be attributed to the weak interfacial adhesion between the PE chain and the paper sludge fiber.

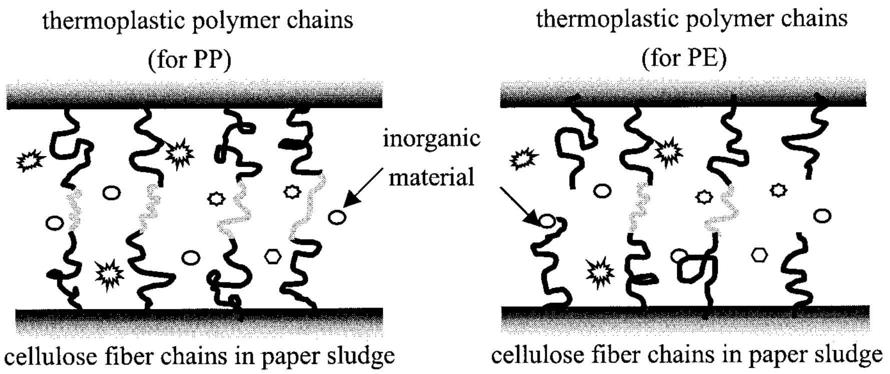

Figure 13 A schematic diagram showing the interfacial adhesion regions between cellulose fiber chains in paper sludge and polypropylene (or polyethylene).

Effect of Extrusion Temperature on Mechanical Properties

The tensile and flexural properties of the thermoplastic polymer filled with paper sludge by the extrusion temperature are shown in Figures 14–17. The tensile strength and modulus of the composite were significantly increased when increasing the extrusion temperature on four types of matrix composites. This was attributed to better wettability between the polymer chains and paper sludge fibers with increasing extrusion temperature. Also, increasing the extrusion temperature from 150 to 210°C had a slightly positive overall effect on the flexural strength and modulus properties of PE composites. As shown in Figures 14 and 15, there was some indication of an optimum in flexural properties in the 210–230°C extrusion range for paper sludge–PP and paper sludge–HIPP composites, and in the 170–210°C extrusion range for paper sludge–HDPE and paper sludge–LDPE. Generally, a rise of the extrusion temperature had a positive effect on the tensile and flexural properties of paper sludge–thermoplastic polymer composites.

The notched and unnotched impact strengths of paper sludge–thermoplastic polymer composites as a function of the extrusion temperature

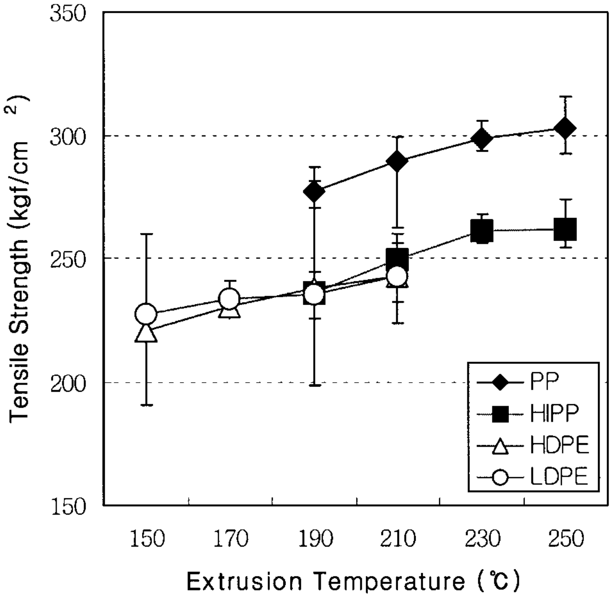

Figure 14 The tensile strength of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the extrusion temperature.

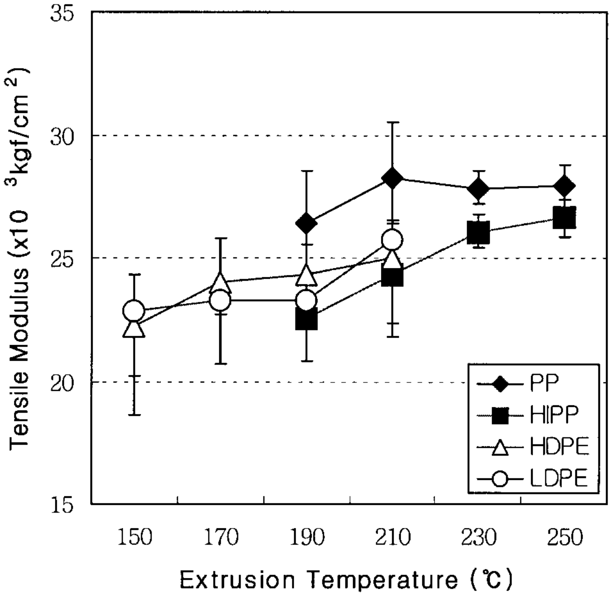

Figure 15 The tensile modulus of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the extrusion temperature.

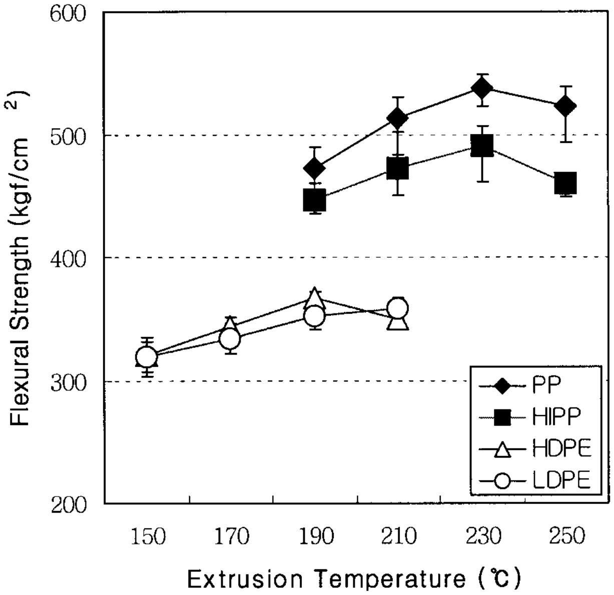

Figure 16 The flexural strength of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the extrusion temperature.

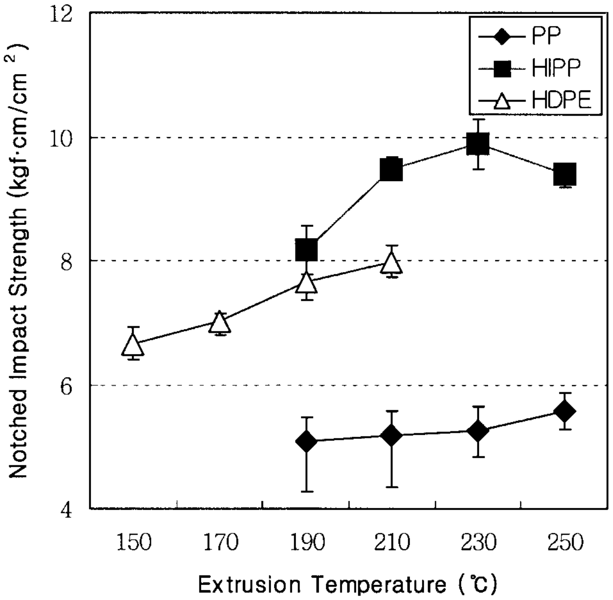

Figure 18 The notched impact strength of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the extrusion temperature.

are shown in Figures 18 and 19. For paper sludge–PP and paper sludge–HIPP composites the notched and unnotched impact strengths of the composites were increased as the extrusion temperature was increased from 190 to 230°C, but they decreased slightly in the range from 230 to 250°C. This lower impact energy at the extrusion temperature of 250°C might be attributed to excessively brittle fibers from thermal decomposition.[6] According to Kim et al.,[7] the interfacial adhesion between polymer and polymer was low because the chains of the polymer easily diffuse

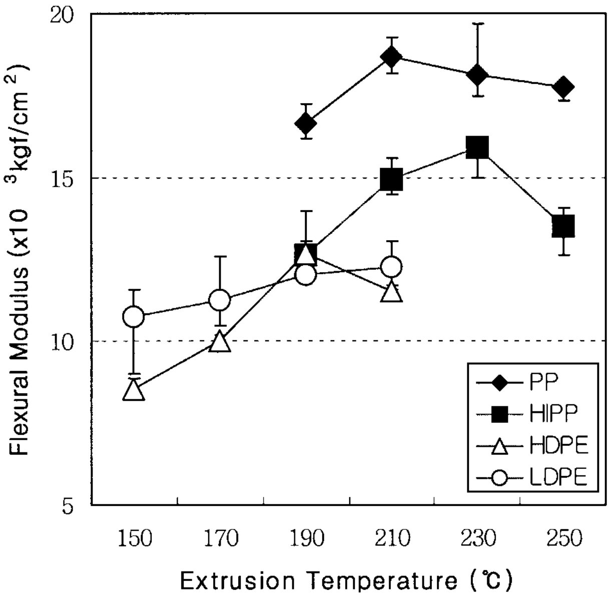

Figure 17 The flexural modulus of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the the extrusion temperature.

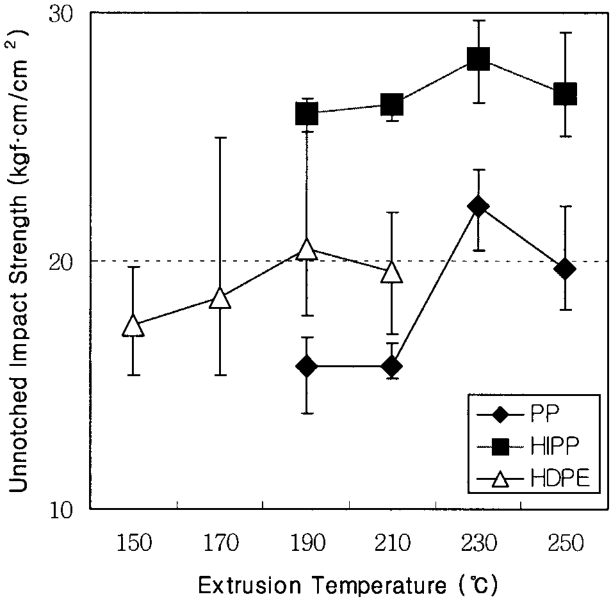

Figure 19 The unnotched impact strength of the paper sludge–thermoplastic polymer (20/80 wt %) composite as a function of the extrusion temperature.

into the bulk at high temperature. Judging from this phenomena, we supposed that the impact strengths of the composites were low at too high a temperature, which was due to weak interfacial adhesion because the chains of cellulose may diffuse into the thermoplastic polymer (matrix).

As shown in Figure 1, the residual weight of cellulose fibers in the paper sludge, which had particle sizes of 0.42–0.84 mm, started to decrease at 250°C and then sharply decreased from 290 to 350°C. From these results we inferred that thermal decomposition occurred in cellulose fibers of paper sludge at high extrusion temperatures. The composites also became darker in color with higher temperature.

On the other hand, for paper sludge in HDPE and LDPE, the notched and unnotched impact strengths of the composites increased with an increase of the extrusion temperature from 150 to 210°C.

CONCLUSIONS

A study was designed and performed to determine the influences of the particle size of paper sludge (below 0.15, 0.18–0.25, and 0.42–0.84 mm) and the extrusion temperature (190, 210, 230, and 250°C for PP and HIPP; 150, 170, 190, and 210°C for HDPE and LDPE) on the physical and mechanical properties of paper sludge–thermoplastic polymer composites.

In addition, thermal analysis and microstructural investigation of the composite were conducted to provide information about the particle sizes of paper sludges.

The results obtained are summarized as follows:

1. The ash contents of the paper sludge were about 73.7, 46.2, and 38.1% with particle sizes of below 0.15, 0.18–0.25, and 0.42–0.84 mm, respectively, which meant lower ash content and thus higher cellulose fiber content with a larger particle size of the paper sludge.
2. The thickness swelling and water absorption of the composites were increased with a decrease in the paper sludge particle size.
3. The tensile and flexural strengths and modulus of these composites were significantly increased as the particle size of the paper sludge was increased. The larger particle size of paper sludge had higher cellulose fiber contents and increased notched impact strength. In addition, an analysis of the unnotched impact strength data indicated that there was no trend with the paper sludge particle size according to thermoplastic polymer types.
4. The dimensional stabilities of paper sludge–thermoplastic polymer composites were somewhat improved with an increase in the extrusion temperature, but they were not statistically different.
5. The tensile strength and modulus of the composite was significantly increased as the extrusion temperature increased with the four types of matrix composites.
6. The notched and unnotched impact strengths of the composites increased as the extrusion temperature increased from 190 to 230°C, but they slightly decreased in the range from 230 to 250°C for PP-based composites. The notched and unnotched impact strengths of the composites increased with an increase of the extrusion temperature from 150 to 210°C for PE-based composites.
7. Paper sludge was very useful as a reinforcing filler in the thermoplastic polymer in terms of the use of cellulose-rich fibers from recyclable waste resources. Although we did not carry out biodegradability tests, we anticipate that biodegradability will be one of the significant parameters in favor of cellulose-based composites in the future.

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