Influence of Hwangto on the Mechanical Properties of Wood Flour Reinforced High Density Polyethylene (HDPE) Composites[*1]

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ABSTRACT

The mechanical properties of wood flour, Hwangto (325 and 1,400 mesh per 25.4 mm) and coupling agent-reinforced HDPE composites were investigated in this study. Hwangto and maleated polyethylene (MAPE) were used as an inorganic filler and a coupling agent, respectively. The addition of Hwangto and MAPE to virgin HDPE also increased the Young's modulus in the smaller degree. The addition of wood flour and Hwangto to virgin HDPE increased the tensile strength, due to the high uniform dispersion of HDPE by high surface area of Hwangto in HDPE and wood flour. MAPE also significantly increased the tensile strength. When wood flour was added, there was no notable difference on the tensile properties, in terms of Hwangto particle size. Hwangto also improved the flexural modulus and strength of reinforced HDPE composites. With different particle sizes of Hwangto, there was no considerable difference in flexural modulus and strength of reinforced HDPE composites. The addition of Hwangto showed slightly lower impact strength than that of wood flour. However, the particle size of Hwangto showed no significant effect on the impact strength of reinforced composites. In conclusion, reinforced HDPE composites with organic and inorganic fillers provide highly improved mechanical properties over virgin HDPE.

Keywords : Hwangto, Young's modulus, tensile strength, elongation, flexural modulus, flexural strength, Notched impact strength, Maleated polyethylene

1. INTRODUCTION

Traditionally, inorganic fillers such as fiberglass, calcium carbonate and talc have been combined with plastics to make inexpensive composites for a wide range of applications. Recently, many researchers have developed wood-plastic composites (WPCs) using lignocellulosic fibers, on the basis of the advantageous properties of plastics, such as polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC) (Jana and Prieto, 2002; Keller, 2003; Mishira *et al.*, 2003; Nair *et al.*, 2001; López-Manchado *et al.*, 2000). Lignocellulosic fibers used in WPCs

are most often incorporated in particular form up to the loading level of 50% by weight (Clemons, 2002).

There has been an elevating utilization of composites in various fields ranging from a residential building to automobile industries. Although the Wood Polymer Composites (WPCs) industry is only a section of the total wood products industry, building applications are the largest and fastest growing market for WPCs (Leaversuch, 2000; Mapleston, 2001). Besides decking, products such as siding, fencing, floor materials, moldings, louver and wooden housing are manufactured. Moreover, considerable market growth is expected in the near future (Clemons, 2002).

On the other hand, the addition of wood flour to virgin polymer stiffens the polymer greatly, representing the increase of tensile modulus, but at the same time providing the decrease of tensile strength (Stark and Rowlands, 2003). The loss of tensile strength may be a handicap for using as materials requiring a high mechanical strength. It was also reported that coupling agent plays an important role in improving the compatibility and bonding strength between hydrophobic polymer and hydrophilic wood flour (Lu et al., 2000; Lee et al., 2004), thereby improving MOR. Moreover, the addition of other additives may be necessary to improve the mechanical strength of composites.

Hwangto as an inorganic material is extensively distributed in North America, Europe, North China (The Yellow River Valley) and Asian countries. Generally, Hwangto is a yellow or reddish brown soil which loam was weathered by water and wind for a long time (Bates, 1987; Cho et al., 2003). The Hwangto has been widely applied with some advantageous properties such as the radiation of far-infrared and adsorption-condensation-precipitation of a toxic substance (Chorover and Sposito, 1995). It was

also reported that Hwangto showed high sound and impact absorption, thereby showing proper flexibility and releasing the burden of human body and activating the metabolism. In order to use Hwangto effectively as reinforcement materials, information such as the particle size, distribution of particle, morphology, surface area, density and plasticity will be necessary.

Many researchers have worked for the development of new material for better indoor environment and for the partial substitution of cement and concrete, using Hwangto. As a matter of interest in green cement, the concrete mixed with Hwangto and cellulose fiber was studied (Yang et al., 2006). There are also attempts to develop the cement and concrete composites with Hwangto and to wide applications to construction materials (Choi et al., 2000; Ryu and Seo, 2000).

However, very little data were available on the relation between Hwangto as a inorganic filler and the mechanical characteristics of reinforced polymer composites. The objective of this study was to find the effect of Hwangto, wood flour and MAPE on the mechanical properties of the reinforced composites.

2. MATERIALS and METHODS

2.1. Sample Preparation

High density polyethylene (HDPE) was obtained from LG Petrochemical Co., Korea. HDPE has a melt flow index of 5.5 g/10 min and a density of 0.961 g/cm^3. Wood flour (WF) of 80~100 mesh per 25.4 mm was supplied from Il-Song Wood Flour, Korea. Wood flour was manufactured from mixed hardwood and softwood waste panels. Maleated polyethylene (MAPE) as a coupling agent was obtained from Honam Petrochemical Co, Korea. The melt flow index and density of MAPE are 1.0 g/10 min

Table 1. Chemical components of Hwangto (Unit: %)

SiO_2	Al_2O_3	TiO_2	Fe_2O_3	CaO	MgO	K_2O	Na_2O	Ig.loss
42.5	36.6	0.23	4.05	0.57	0.69	0.41	0.18	14.8

and 0.920 g/cm^3, respectively. Hwangto as a reddish-brown inorganic filler (325 and 1,400 mesh per 25.4 mm) was obtained from Hwangto Nara Co, Korea. Major chemical components of the Hwangto were shown in Table 1.

2.2. Manufacture of Pellets and Compounding

Wood flour was dried to the moisture content of 2~3% in a dry oven at a temperature of 80°C for 24 hours. Wood flour, Hwangto, and MAPE were blended with HDPE in a Hakke Rheomix (Germany) for 20 min (40 rpm and 170°C). HDPE (60, 70, 80, 90 and 100 weight % based on total weight), WF (0, 20, 30 and 40%), Hwangto (0, 10, and 20%) and MAPE (0 and 3%) were loaded at various combinations. The blended materials were cooled in air and then granulated in a cutting mill (Fritsch Co., Germany).

A single extruder (Brabender® stand-alone extruder, Germany) was used to extrude the granules. The extruder had a screw diameter of 19 mm and a L/D ratio of 25 and a circular nozzle of 6 mm in diameter. The extrusion temperature was 180°C. The extrudate in the form of strands was cooled in air and pelletized with Brabender® pelletizer (Germany). The resulting pellets for tensile, flexural and impact strength test specimens were dried at 80°C for 24 hr before using an injection-molding machine (Korea Manufacturing Technology Center, Korea). The molding temperature for all pellets was 195°C.

2.3. Flexural Testing

The flexural strength properties for WPC were measured at temperature of 23°C and humidity of 50% according to ASTM D-790. The dimensions of a specimen for flexural tests were 125.0 mm × 4.0 mm × 10 mm (length × thickness × width). The tests were conducted by a Zwick Universal Testing Machine at a crosshead speed of 200 mm/min on the samples with a span of 64 mm. Each test was repeated 5 times to obtain more representative results.

2.4. Tensile Testing

Static tensile strength tests were conducted according to ASTM D-638. The test specimens for the evaluation of tensile strength were molded in a mold whose cavity has the following dimensions; W = 3.23±0.06 mm, L = 9.62±0.08 mm, G = 7.52±0.07 mm and T = 3.35±0.05 mm, where W = width of narrow section, L = length of narrow section, G = gage length and T = thickness of narrow section. The tests were performed using a Zwick Universal Testing Machine (Germany) at a crosshead speed of 10 mm/min on the samples with an initial length of 40.0 mm. Each test was repeated 5 times.

2.5. Izod Impact Testing

Notched Izod impact tests were conducted aaccording to ASTM D-256. The dimensions of a specimen for impact strength measurement were 62.5 mm × 4.0 mm × 10 mm (length × thickness × width). The tests were performed with an XJ-40A pendulum-type apparatus using acutely notched specimens (notch depth: 2 mm). Each mean value represented an average of five samples.

Table 2. Tensile, flexural and impact strength of wood flour, Hwangto (1,400 mesh), and MAPE reinforced HDPE composites

HDPE (%)	WF (%)	HT[1] (%)	MAPE (%)	Tensile properties			Flexural properties		Izod impact strength (kgcm/cm^2)
				Young's modulus (kgf/cm^2)	Tensile strength (kgf/cm^2)	Elong. (%)	Flexural modulus (kgf/cm^2)	Flexural strength (kgf/cm^2)	
100	-	-	-	28,857 (1,735)	281.1 (18.1)	6.9 (2.0)	7,504 (257)	243.3 (7.6)	8.3 (0.7)
90	10	-	-	33,029 (1,251)	266.7 (7.0)	5.2 (1.8)	10,040 (2,025)	266.1 (7.6)	5.0 (0.5)
90	-	10	-	29,432 (1,141)	278.1 (8.9)	5.1 (1.0)	10,050 (2,664)	283.8 (35.9)	3.3 (0.1)
80	20	-	-	41,593 (1,097)	264.3 (14.3)	3.1 (0.6)	13,640 (1,344)	294.0 (11.6)	4.2 (0.3)
80	-	20	-	33,991 (652)	268.4 (7.3)	4.2 (1.0)	10,600 (1,294)	250.9 (5.3)	3.3 (0.1)
80	10	10	-	38,941 (2,702)	282.6 (12.5)	2.2 (0.3)	17,840 (2,842)	456.2 (27.4)	2.6 (0.5)
80	10	10	3	32,641 (1,255)	285.2 (5.6)	4.1 (0.9)	10,900 (1,517)	390.2 (4.4)	4.7 (0.3)
70	30	-	-	51,261 (1,711)	248.3 (16.5)	1.6 (0.7)	15,800 (1,455)	324.4 (8.8)	3.2 (0.2)
70	20	10	-	43,895 (861)	300.9 (5.3)	1.8 (0.2)	13,030 (992)	354.8 (21.9)	3.6 (0.3)
70	20	10	3	42,872 (976)	308.0 (4.3)	1.9 (0.2)	14,140 (2,518)	365.0 (22.8)	3.9 (0.2)
60	40	-	-	59,726 (1,929)	232.7 (6.7)	0.9 (0.2)	20,840 (1,941)	357.4 (7.6)	3.1 (0.1)
60	30	10	-	59,709 (1,665)	308.7 (12.8)	0.9 (0.2)	19,930 (2,478)	339.6 (4.4)	2.9 (0.1)
60	30	10	3	45,805 (6,786)	319.8 (19.2)	1.7 (0.3)	14,370 (1,623)	370.0 (8.8)	3.8 (0.2)
60	20	20	-	50,643 (2,219)	240.8 (13.0)	1.4 (0.6)	18,690 (1,292)	357.4 (15.2)	3.4 (0.2)
60	20	20	3	43,921 (1,975)	303.0 (5.7)	1.9 (0.2)	15,240 (1,046)	397.9 (8.8)	3.6 (0.2)

HT[1]: Hwangto, WF: wood flour, (1,400 mesh), MAPE: maleated polyethylene

3. RESULTS and DISCUSSION

3.1. Tensile Properties

3.1.1. Influence of Hwangto (1,400 mesh)

The tensile properties of wood flour, Hwangto (1,400 mesh), HDPE and MAPE reinforced composites by means of a Universal Testing Machine were summarized in Table 2. The Young's modulus and tensile strength of virgin HDPE were 28,857 and 281.1 kgf/cm^2, respectively. The maximum Young's modulus and tensile strength values were obtained from HDPE with wood flour of 40% or wood flour of 30% and Hwangto of 10%, respectively. The Young's modulus increased as the loading of wood flour increased up to 40%. As shown in Fig. 1, the Young's modulus of HDPE with wood flour of 40% showed 107% higher than virgin HDPE. It indicates that wood flour is much stiffer, more
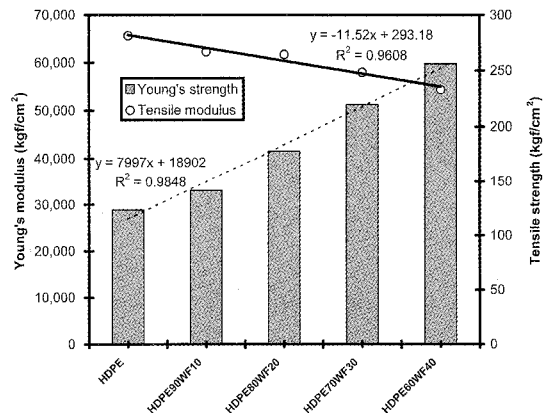

Fig. 1. Tensile properties of wood flour reinforced HDPE composites.

brittle, and stronger than virgin HDPE. The increase of the Young's modulus was well fitted by linear regression as a function of wood flour loading (R^2=0.9848).

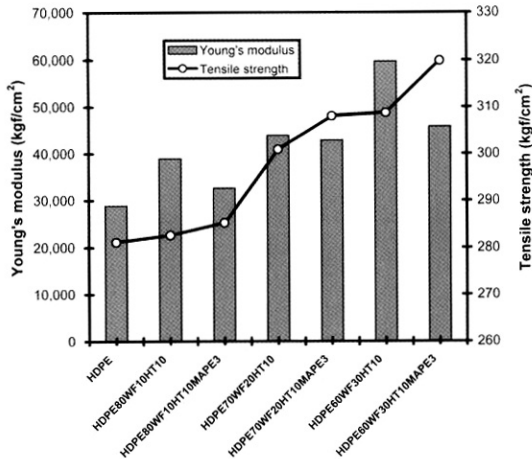The addition of Hwangto to virgin HDPE also increased Young's modulus, but in the small-

Fig. 2. Tensile properties of wood flour, Hwangto (1,400 mesh), and MAPE reinforced HDPE composites.

er degree, compared with the addition of wood flour (Fig. 2). The addition of Hwangto (10 and 20%) increased the Young's modulus in the range of 2.0 and 17.8%, respectively. The Young's modulus of HDPE/wood flour/MAPE/ Hwangto reinforced composites with MAPE was 2.4 to 30.0% lower than that of composites without MAPE.

On the other hand, the tensile strength de-

creased with increasing the loading of wood flour, due to the weak bonding strength between HDPE and wood flour. As shown in Fig. 1, the decrease of tensile strength was linearly fitted by regression as a function of filler loading (R^2 =0.9608). When Hwangto of 10 and 20% to virgin HDPE, the MOR reduced about $3 \sim 13$ kgf/cm^2, indicating that the higher Hwangto level showed the higher reduction of tensile strength (Fig. 2). However, the addition of wood flour (10, 20% and 30%) and Hwangto (10%) to virgin HDPE increased up to 30 kgf/cm^2, indicating that the Hwangto with high surface area improved the dispersion of HDPE to wood flour (Fig. 3(a) and (b)) and provided the higher stress distribution under the tensile load. As shown in Fig. 3(a) and (b), Hwangto of 1,400 and 325 mesh was widely distributed on the space between HDPE and wood flour.

On the other hand, the MAPE significantly increased the tensile strength, compared with the virgin HDPE. The addition of the MAPE to wood flour (30%) and Hwangto (10%) reinforced HDPE composites increased the tensile strength of 13.8%, because of the increased intra-molecular bonding strength between wood
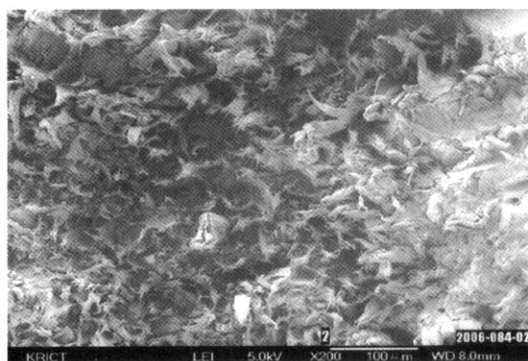
(a)
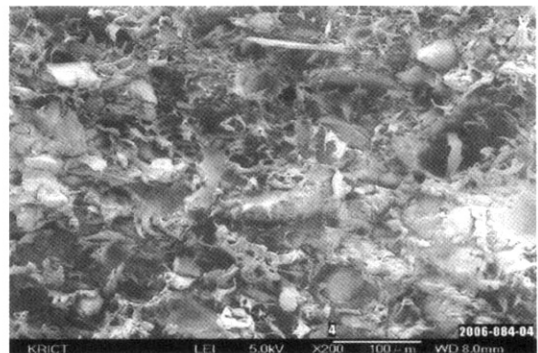
(b)

Fig. 3. SEM micrographs of the tensile fractured surface of (a) HDPE60-wood flour20-Hwangto20 (1,400 mesh) reinforced HDPE composites and (b) HDPE60-wood flour20-Hwangto20 (325 mesh) reinforced HDPE composites.

Table 3. Tensile, flexural and impact strength of wood flour, Hwangto (325 mesh), and MAPE reinforced HDPE composites

HDPE (%)	WF (%)	HT[2] (%)	MAPE (%)	Tensile strength			Flexural strength		Izod impact strength (kgcm/cm)
				Young's modulus (kgf/cm²)	Tensile strength (kgf/cm²)	Elong. (%)	Flexural modulus (kgf/cm²)	Flexural strength (kgf/cm²)	
90	-	10	-	31,667 (982)	275.0 (12.7)	5.9 (1.6)	9,903 (658)	296.5 (13.1)	4.6 (0.3)
80	-	20	-	39,931 (1,898)	316.6 (6.0)	3.4 (1.1)	13,400 (2,012)	339.1 (27.7)	4.3 (0.2)
80	10	10	-	37,941 (979)	256.8 (6.1)	2.5 (0.8)	11,840 (1,660)	313.2 (6.4)	4.2 (0.1)
80	10	10	3	32,252 (1,255)	284.6 (2.1)	3.4 (1.1)	11,320 (1,050)	325.4 (3.4)	4.5 (0.2)
70	20	10	-	44,324 (1,849)	266.1 (11.9)	1.7 (0.3)	14,060 (899)	336.1 (6.4)	3.7 (0.1)
70	20	10	3	39,333 (2,413)	308.4 (10.2)	2.3 (0.4)	13,640 (976)	371.0 (9.9)	4.0 (0.2)
60	30	10	-	54,935 (3,984)	307.7 (11.4)	0.9 (0.1)	21,790 (1,220)	343.7 (8.3)	2.9 (0.1)
60	30	10	3	48,023 (1,015)	320.8 (6.7)	1.7 (0.1)	16,000 (967)	408.1 (21.9)	3.9 (0.2)
60	20	20	-	51,946 (2,505)	223.0 (5.8)	1.1 (0.2)	16,910 (1,139)	329.5 (4.4)	3.2 (0.1)
60	20	20	3	47,765 (2,138)	301.7 (15.1)	1.9 (0.7)	15,020 (1,567)	372.6 (7.6)	3.8 (0.1)

HT[2]: Hwangto (325 mesh), WF: wood flour, MAPE: maleated polyethylene

flour and virgin HDPE, compared with the inter-molecular bonding of HDPE itself (Pritchard, 1998; Lu et al., 2000; Lu et al., 2002; Lee et al., 2004; 21). From these results, the interaction effect between wood flour and MAPE at Hwangto level of 10% was highly significant on the tensile strength.

The addition of wood flour significantly decreased the elongation, whereas the elongation of virgin HDPE was 6.9%. When the wood flour of 40% to virgin HDPE, the elongation was 0.9%. The addition of Hwangto (10 and 20%) to virgin HDPE also decreased the elongation. It represents the addition of wood four and Hwangto increased the stiffness of virgin HDPE, but the addition of MAPE increased slightly the elongation (Table 2).

3.1.2. Influence of Hwangto (325 mesh)

Tensile properties of wood flour, Hwangto (325 mesh), HDPE, and MAPE reinforced composites were reported in Table 3. The addition of Hwangto (325 mesh) to virgin HDPE also increased the Young's modulus significantly. The influence of Hwangto (325 mesh) on the
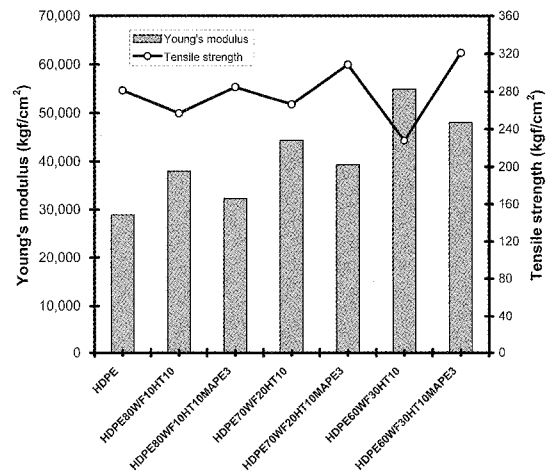

Fig. 4. Tensile properties of wood flour, Hwangto (325 mesh), and MAPE reinforced HDPE composites.

Young's modulus of reinforced HDPE was much bigger than that of 1,400 mesh Hwangto. When wood flour was added, however, there was no striking figure on the Young's modulus of wood flour reinforced HDPE in terms of Hwangto particle size. The addition of MAPE (3%) decreased the Young's modulus of reinforced HDPE (Fig 4).

The tensile strength of reinforced HDPE with Hwangto (325 mesh) at the loading level of 10% Hwangto was similar to tensile strength of reinforced HDPE with Hwangto of 1,400 mesh at the same loading level. However, the tensile strength of reinforced HDPE with Hwangto (325 mesh) at the loading level of 20% Hwangto was 18.0% higher than that of HDPE with Hwangto of 1,400 mesh at the same loading level (Table 3). The tensile strength at the loading level of 10% Hwangto (325 mesh) increased as the loading level of wood flour increased.

MAPE also increased the tensile strength of wood flour and Hwangto (325 mesh) reinforced HDPE (Table 3). At different Hwangto particle sizes of 325 and 1,400 mesh, there was no significant difference in the tensile strength of MAPE, wood flour and Hwangto reinforced HDPE composites.

3.2. Flexural Properties

3.2.1. Influence of Hwangto (1,400 mesh)

Flexural strength properties of wood flour, Hwangto (1,400 mesh), HDPE, and MAPE reinforced composites were reported in Table 2. The flexural modulus of elasticity (MOE) and flexural strength (MOR) of virgin HDPE were 7,504 and 243.3 kgf/cm^2, respectively. MOE of wood flour reinforced HDPE composites increased as the loading of wood flour increased up to 40%. As shown in Fig. 5, the flexural strength of reinforced HDPE with wood flour of 40% showed 177% higher than that of virgin HDPE, due to the increase of stiffness and brittleness. The increase of the flexural modulus is well fitted by a linear regression as a function of wood flour loading (R^2=0.9822).

The addition of Hwangto to virgin HDPE increased the flexural modulus, but the increase of the flexural modulus by Hwangto was rela-
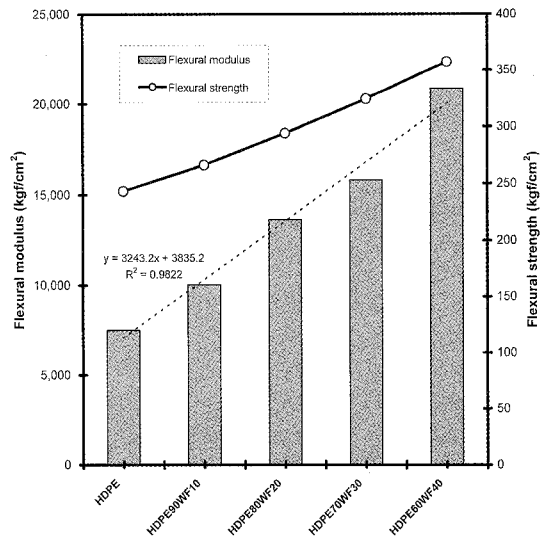
Fig. 5. Flexural properties of wood flour reinforced HDPE composites.

tively lower than that by wood flour. The interaction effect of wood flour and Hwangto was significant on the increase of the flexural modulus. It was observed that the MAPE did not show the increase of the flexural modulus, indicating that the flexural modulus is not related to the increase of internal bonding strength between wood flour and HDPE, but to the increase of stiffness.

The addition of wood flour to virgin HDPE significantly increased the flexural strength as wood flour was incorporated up to the loading level of 40%. When wood flour of 20% and 40% was incorporated to virgin HDPE, the flexural strength increased 20.8% and 47.9%, respectively.

Hwangto also increased the flexural strength, but in the smaller range, compared with wood flour (Fig. 6). When Hwangto at the loading level of 10% and 20% was added, the flexural strength increased 16.6% and 3.1%, respectively. The flexural strength of wood flour/Hwangto reinforced HDPE increased considerably, representing that Hwangto was incorporated well
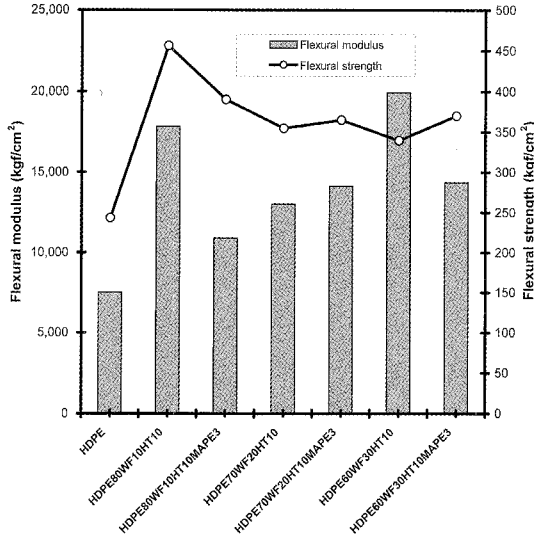
Fig. 6. Flexural properties of wood flour, Hwangto (1,400 mesh), and MAPE reinforced HDPE composites.
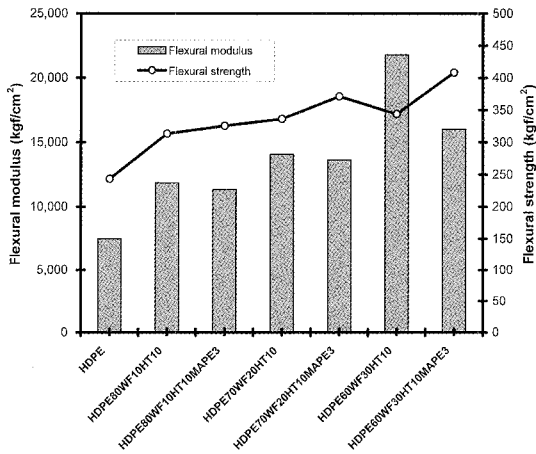

Fig. 7. Flexural properties of wood flour, Hwangto (325 mesh), and MAPE reinforced HDPE composites.

with wood flour and HDPE. The addition of 3% MAPE fairly increased the flexural strength of composites when wood flour of 20% and 30% was added.
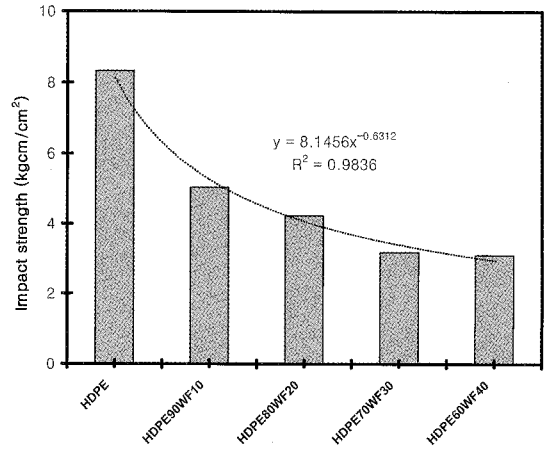

Fig. 8. Izod impact strength of wood flour reinforced HDPE composites.

3.2.2. Influence of Hwangto (325 mesh)

The addition of Hwangto (325 mesh) to virgin HDPE increased the flexural modulus of composites fairly. The addition of Hwangto (325 mesh) increased the flexural modulus of reinforced HDPE, compared with virgin HDPE. As a whole, there was no notable difference on the flexural modulus of wood flour reinforced HDPE in terms of Hwangto particle size. The addition of the MAPE (3%) provided somewhat better effect on the flexural modulus in the smaller particle size of Hwangto (Table 3 and Fig. 7).

The flexural strength of reinforced HDPE with Hwangto of 325 mesh at the loading level of 10% and 20% Hwangto was higher than that of HDPE with Hwangto of 1,400 mesh at the same loading level (Fig. 8). However, the smaller Hwangto size generally showed lower flexural strength of wood flour reinforced HDPE at the Hwangto levels of 10 and 20%. The MAPE also improved the flexural strength of wood flour and Hwangto (325 mesh) reinforced HDPE (Table 3). At different Hwangto particle sizes, there was no considerable difference of the flexural strength in wood flour,
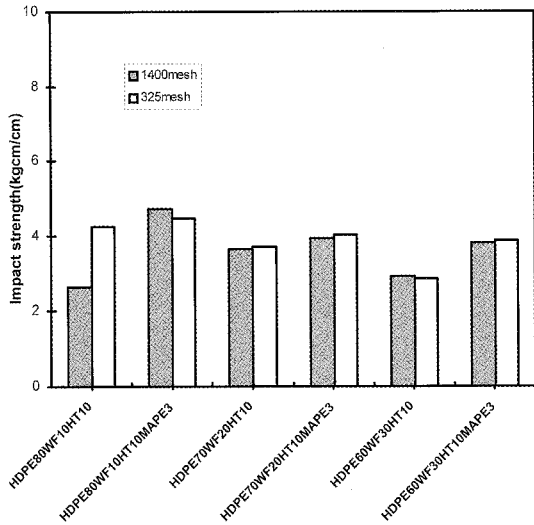
Fig. 9. Izod impact strength of wood flour, Hwangto (1,400 and 325 mesh), and MAPE reinforced HDPE composites.

Hwangto and MAPE reinforced HDPE composites.

3.3. Impact Strength

Notched impact strength properties of wood flour, Hwangto (325 and 1,400 mesh), HDPE and MAPE reinforced composites were reported in Tables 2 and 3. Impact strength of HDPE was 8.3 kgcm/cm^2. As the addition level of wood flour increased, impact strength decreased gradually. As shown in Fig. 9, the relationship between impact strength and addition level of wood flour followed a decaying exponential function (R^2=0.9836).

The addition of Hwangto showed slightly lower impact strength than that of wood flour. Impact strength of Hwangto of 10% and wood flour of 10~30% reinforced HDPE composites was lower than that of only wood flour reinforced HDPE composites. MAPE improved the impact strength of reinforced HDPE composites. However, the particle size of Hwangto showed no significant effect on the impact

strength of reinforced HDPE composites.

4. CONCLUSIONS

Mechanical strength properties are a requirement and a real solution in many application fields. The strength properties can be achieved in relatively simple and inexpensive ways in this study. The addition of Hwangto to virgin HDPE significantly increased tensile and flexural strength properties. However, there was no notable impact of Hwangto particle size on both properties. Conclusively, reinforced HDPE composites with organic and inorganic fillers provide highly improved mechanical properties over virgin HDPE.

ACKNOWLEDGEMENT

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