목재공학 22(2) : 46~53(原著)
Mokchae Konghak, 22(2) : 46~53(Original), 1994

# 木纖維와 熱可塑性 플라스틱 複合材料의 機械的 性質*1

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# Mechanical Properties of Wood-Fiber Thermoplastic Composites*1

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## 要 約

본 硏究는 木纖維와 熱可塑性 플라스틱의 複合材料를 제조하고 그 機械的 性質을 평가하기 위하여 실행되었다. 강도가 높고 밀도가 낮아 플라스틱의 보강재료로써 잠재성을 갖는 목섬유를 2종의 열가소성 플라스틱(폴리프로필렌과 폴리에틸렌)과 混合하여 複合材料를 만들었다. 吸濕性인 목섬유와 非吸濕性인 플라스틱과의 친화성을 위해 界面活性劑를 사용하였다. 또한 낮은 밀도의 목섬유를 플라스틱내에서 혼합하기 위해 고속 플라스틱믹서를 사용하였다. 射出成形한 샘플을 사용하여 機械的 性質을 試驗한 결과 인장및 휨강도는 목섬유 혼합량에 따라 크게 향상되었다. 휨 強度는 引張強度보다 훨씬 크게 나타났으며 引張및 휨 彈性 係數는 플라스틱내 목섬유 혼합량과 비례적으로 증가하였다. 목섬유는 복합재료의 強度와 彈性係數를 향상시킴으로서 플라스틱을 보강할 수 있었다. 이와는 반대로 인장시험에서 시편 파괴점까지의 신장율과 파괴에너지는 목섬유 혼합량이 증가함에 따라 감소하였다. 衝擊強度 역시 유사한 경향을 보였다.

## ABSTRACT

This study was conducted to investigate a feasibility of manufacturing wood-fiber thermoplastic composites and to evaluate their mechanical properties. Wood fiber as a potential reinforcing filler was compounded with two thermoplastics (polypropylene and high density polyethylene) in high intensity thermokinetic plastic mixer aided with a wetting agent. It was found that wood-fiber thermoplastic composites could be manufactured by injection molding process. The tensile and flexural strength of injection molded specimens were improved greatly with increasing wood fiber concentration. Tensile and flexural modulus increased proportionately with wood fiber concentration. Wood fiber provided reinforcement with thermoplastics in terms of strength and modulus. However, the percent elongation at break and energy to break were reduced with increasing wood fiber loadings. Impact strength also showed similar trend.

*Keywords* : Wood fiber, PP, HDPE, composites, wetting agent, mechanical properties

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## 1. INTRODUCTION

Composite materials are made by combining two or more components to achieve desired properties that could be obtained with the separate components. As the use of thermoplastic polymer composites is tremendously increasing, considerable efforts have been made to find suitable reinforcing fillers. Using a reinforcing filler can reduce material cost but also improve certain properties. Among organic fillers, wood and cellulosic fibers were considered attractive alternates for reinforcing fillers in thermoplastics[1]. Thus interest has grown recently in thermoplastic composites reinforced with cellulosic fibers that have been used as extenders for thermosetting polymers such as phenolics. This development is being encouraged by the fact that cellulosic fillers had several advantages over inorganic fillers. In particular, wood-fiber thermoplastic composites are fascinating for low cost/high volume applications. Potential applications of the composites are furniture components, door molding, floor system for light-frame construction, and packaging pallets. A current major application is for the interior panels of automobiles[2].

Wood fibers give high aspect ratio and have high specific stiffness and strength[3]. In addition, wood and cellulosic fibers also offer number of benefits for wood fiber thermoplastic composites. These include: low hardness that minimizes abrasion of the equipment duirng processing, relatively low density, biodegradability, and low cost per unit volume basis[4]. Woodhams et al.[5] reported that in terms of stiffness, kraft pulp fiber polypropylene composite was a cost effective substitute for the same relative weight of glass fiber polypropylene composite or aluminum sheet.

Cellulosic fibers can be combined with plastics as reinforcing filler. For example, wood fibers are mixed with thermoplastics in molten state. This melt-blending technology is being used for the manufacture of fiber-reinforced thermoplastic composites. In this process, cellulosic or wood fibers are blended with the melted thermoplastic matrix by shearing or kneading. For the application of melt-blending process, it is generally accepted that cellulosic fillers are suitable for four of the major classes of thermoplastics, i. g. polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC) which were melt at relatively low temperatures[6]. The melting temperatures of these thermoplastics are below 200℃ at which cellulose in wood fiber can be degraded thermally. Combined wood fibers provide reinforcement with thermoplastics. When wood fiber is incorporated into a polymer matrix, certain mechanical properties of the composite are improved with increasing the fiber concentration. In general, the tensile strength and modulus of the composites increased significantly compared to the unfilled polymer, while the elongation at rupture and impact strength decreased[7]. The mechanical properties are also dependent on the processing conditions at which the composites are being processed.[8]

However, there are some difficulties in reinforcing thermoplastics with wood fiber. One of them is to obtain satisfactory adhesion between the hydrophilic fibers and hydrophobic polymers[1]. The interfacial adhesion can be improved by using a wetting agent, especially maleic anhydride polypropylene (MA-PP) for PP-based composites[2, 4, 9]. The MA-PP is a polypropylne that has been reacted with maleic anhydride by free radical grafting reactions. It is assumed that the anhydride groups are attracted by the wood fiber whereas the PP segment provide compatability with the matrix PP. Myers et al[2]. investigated the influence of Epolene E-43 (a commercially available MAPP) on the mechanical properties of wood flour/PP composites. They observed a large increase in strength with addition of Epolene E-43. With an aid of various surface analysis techniques,

Felix and Gatenholm[10] confirmed that MAPP was bonded to the hydroxyl group of cellulosic fiber through esterification. Other special treatments such as grafting monomer or coating of polymer onto wood fiber also showed the increased strength of composites[11, 12].

Another difficulty is that wood fibers have low bulk density. Thus it is relatively difficult to compound fibers with polymer and to get a good dispersion of wood fibers in polymer resin when compared with mineral fillers (e. g. glass fiber, mica, talc, etc.). This might be overcome to some extent by using high intensity thermokinetic mixer (K-mixer)[13]. The other undesirable outcome of using wood fiber is demensional instability due to the absorption of moisture in humid environment[11].

This study was carried out to examine a feasibility of manufacturing wood-fiber thermoplastic composites and to evaluate their mechanical properties. The tensile and flexural properties (strength and modulus), and impact strength were measured for injection molded composites containing various wood fiber concentrations.

## 2.MATERIALS AND METHODS

### 2. 1 Materials and compounding

In the present paper, waste newspapars were used at the source of wood fiber. Newspaper fibers were prepared by the use of a standard hammer mill (American Pulverizer Type M), and used as reinforcement for two polymer resins. Approximate 100 newsprint fibers were used to measure the fiber length and diameter. The average fiber length and diameter were 1.64 mm and 0.034 mm respectively, giving an aspect ratio of 48 although they had wide variations in dimensions. PP (Profax, 6301, Himont Canada Inc.) and HDPE (Sclair, 2909, Du Pont Canada Inc.) were selected as homo polymers. The wetting agents employed were Epolene E-43 and C-18 (Eastman Chemical Products Inc.) which can aid the fiber dispersion in matrix resin, and facilitate adhesion between fibers and polymers. These wetting agents were low molecular weight polyolefins with terminal anhydride end groups. Epolene E-43 (density 0.934 g/cm$^3$, approximate mol. wt. 4500, and acid no. 47) and C-18 (density 0.905 g/cm$^3$, approximate mol. wt. 4000, and acid no. 5) were added to PP- and HDPE-based composites during compounding process, respectively.

Prior to the addition of the matrix resin, air-dried wood fiber was first precoated with a wetting agent, Epolene E-43 or Epolene C-18 (3% relative weight of wood-fiber) in K-mixer (Wener and Pfleiderer Gelimat G-1) for 1.5 min. The matrix resin was compounded in the turbine mixer (running at the tip speed of 3, 300 rpm), and automatically discharged at the pre-set temperature of 190℃. The hot melt mass was cooled and subsequently granulated.

### 2. 2 Injection molding and testing methods

The granulated compounds containing various proportions of wood fiber and matrix resin were injection-molded using an Engel ES-28 machine equipped with a standard ASTM test specimen. The injection-molding conditions are summarized in Table 1. Injection molded specimens were normally black and had smooth surface. The molded test specimens were conditioned at the room temperature for two days prior to testing. The mechanical properties were measured according to ASTM standard procedures: tensile properties (D 638); flexural properties (D 790); Izod unnot-

Table 1. The Injection Molding Condition of Specimen Preparation

| Parameters | | Value |
|---|---|---|
| Injection pressure | (MPa) | 4.84 |
| Clamp pressure | (MPa) | 14.5 |
| Temperaure | (℃) | 200 |
| Nozzle | (%) | 60 |
| Injection times | (s) | 15 |
| Cooling times | (s) | 25 |
| Molding opening times | (s) | 2 |

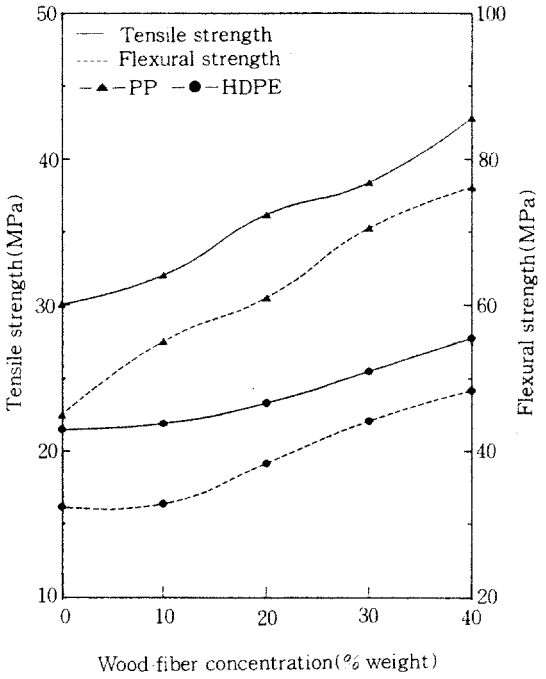


Fig. 1. Tensile and flexural strength of the composites containing various wood-fiber concentrations.

ched impact strength (D 256). The tensile and flexural properties were measured using computerized testing machine. The impact strength was measured by Izod impact test machine with unnotched specimen. All properties were measured at room temperature with six replications and the mean values were presented.

## 3. RESULTS AND DISCUSSION

### 3. 1 Tensile and flexural strength

Figure 1 shows the influence of wood fiber concentration on the tensile and flexural strength of the PP- and HDPE-based composites. The PP-based com- posites generally showed higher tensile strength than that of HDPE-based ones by a factor of 10. For PP-based composites, tensile strength increased to about 43 MPa with addition of 40% fiber concentration. HDPE-based composites at the same fiber concentration showed tensile strength of 28 MPa. This might be due to the inherent diffrence between two matrix polymers. The PP matrix resin used is isotactic, one with a high crystallinity when compared to HDPE [14].

Generally, the strength of the composites increased with increasing wood fiber concentration. As expected, this result is similar to those of other investigators [2, 4, 5, 9, 15, 16]. The tensile strength of short fiber composites (e. g. wood-fiber reinforced composites) may be described by slip theory [17] in which the strength of short fiber reinforced composites tends to increase with fiber concentration, stiffness, aspect ratio, and fiber-matrix adhesion. Thus it is reasonable to expect that wood fiber concentration would improve the composite strength because a sinlge wood fiber has larger strength than those of PP and HDPE. Sanschargrin et al. [18] showed that the tensile strength of reinforced polystyrene with aspen fibers significantly increased as the aspect ratio increased from 5 to 25, and then reached a plateau. Furthermore, the tensile strength of wood-fiber filled thermoplastic composites depends on number of other factors such as processing method and condition, fiber types and polymer resin [8, 13, 19], and the interfacial adhesion between the fiber and matrix [8].

The flexural strength also increased with the incorporation of wood fiber into thermoplastic as in tensile strength. However, the flexural strength values are larger than the tensile strength. This might be attributed to the tendency of wood fibers to orient during injection molding. In other words, wood-fibers are becoming more parallel near the surface of injection-molded bar, and creating the effect of a sandwich laminate with an outer skin of more highly oriented fibers. Therefore, the apparent flexural strength of these molded test bars will be greater than in tension. Similar result was observed by Woodhams et al. [5]. Furthermore,the alignment of wood fibers in

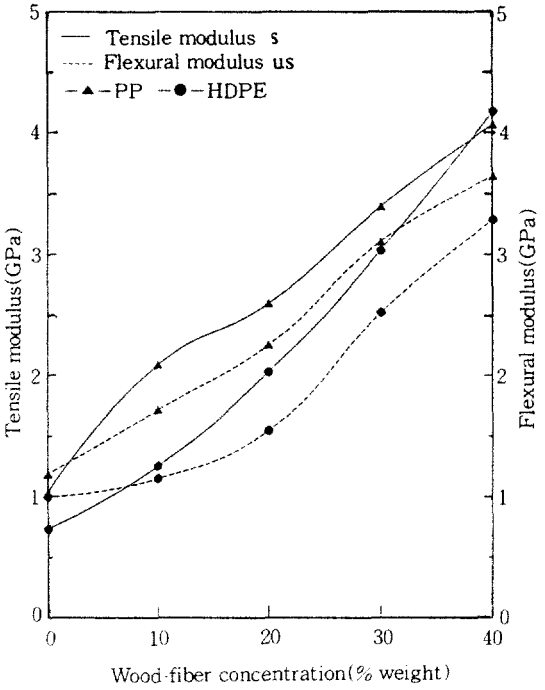


Fig. 2. The modulus of composites containing various wood-fiber concentrations.

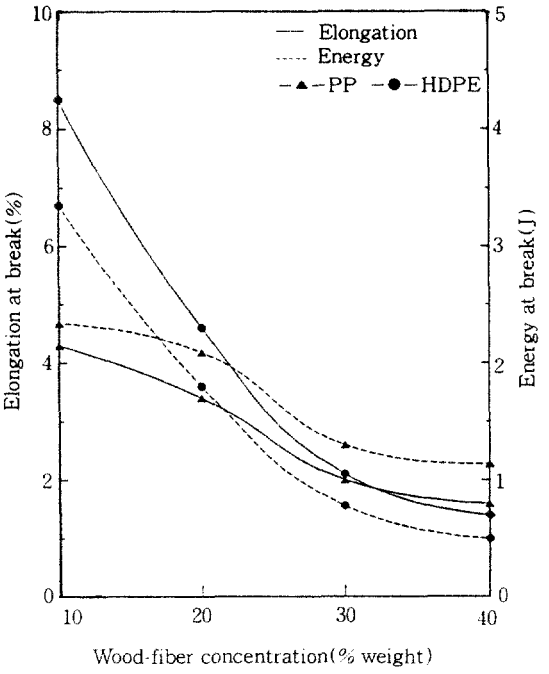


Fig. 3. The percent elongation and energy to break the composites containing various wood-fiber concentrations.

the skin of injection-molded specimen was confirmed by the investigation of microstructure of the specimen [6]. The available results of the flexural strength of wood fiber thermoplastic composites were limited compared to those of the tensile properties. The result of this study is, however, compatible with other studies [13, 16].

### 3. 2. Tensile and flexural modulus

Figure 2 shows the tensile and flexural modulus of both PP and HDPE-based composites. The moduli were increased with an increase in the fiber concentration. Thus, the incorporation of wood fiber improved the stiffness of composite. This is attributed to the greater stiffness of wood fibers. PP-based composites were stiffer than HDPE-based ones reflecting the differences in the matrix polymer themselves as mentioned previously. Unlike the strength, the moduli showed almost similar magnitude in both tensile and flexural modulus scales. In addition, the moduli are strongly dependent on wood fiber concentration. Klason et al. [6] also found the modulus increased with increasing fiber concentration.

In contrast to tensile strength, the tensile modulus of aligned short-fiber composites is usually estimated by shear lag theory (also called elasticity theory) [17]. The plastic has a low elastic modulus while wood fibers are stiff. This difference in modulus between plastic and fiber gives rise to shear at the fiber-matirx interface when the composite is stressed. This shear transfers stress to the fibers, and result in reinforcement in the composites. The modulus of short-fiber composites depends on the volume fraction and modulus of fiber.

The dependence of flexural modulus of the composites on wood fiber concentration is also shown in Figure 2. In general, flexural modu-

lus increases proportionately as the fiber concentration increases. This result is compatible with other findings [2, 4, 5]. Our result suggest that the addition of wood fibers significantly enhance the stiffness of thermoplastic composites.

### 3. 3. The percent elongation and energy at break

The percent elongation and energy at break are shown in Figure 3. In general, the percent elongation was reduced with an rise in the fiber concentration, suggesting that the fiber concentration has large influence on the reduction of percent elongation of the composites. For HDPE-based composites, the elongation rapidly decreased from 8.3 % to 1.7 % as the wood fiber concentration increased from 10% to 40 %. Nevertheless, the elongation showed relatively small change at 40 % wood fiber concentration. In contrast, the percent elongation of PP-based composites were relatively small below 30 % fiber concentration when compared with HDPE-based composites. The decreasing rate of percent elongation is relatively moderate at more than 30 % fiber addition by weight. Other studies also reported the reduction of elongation at break by the addition of wood fiber into thermoplastics [11, 13, 16]. It is reasonable to assume that high strength and modulus composite has low elongation to its failure. In other words, PP-based composite show small elongation because of the steep slope and high peak stress of the stress-strain curve. However, HDPE-based composites have low strength and modulus, leading to large elongation at break.

The energy at break was measured in the tensile test as the area under the stress-strain curve. Along with the percent elongation, the fracture energy to rupture the specimen also decreased as the fiber concentration increased and showed similar pattern in changes to the percent elongation. At 10 % wood-fiber concentration, HDPE-based composites showed higher energy value than PP-based composites. However, PP-based composites have larger fracture energy than HDPE-based ones containing more than 20 % wood-fiber concentration. For all composites, the differences in energy changes were less than a factor of 1 J beyond the addition of 20 % wood-fiber. This result indicates that the area under the stress-strain curve is large in PP-based composites even though it has low percent elongation at break. On the contrary, HDPE-based composite showed small fracture energy despite its large elongation. This results might be due to small differences in the area under the stress-strain curve of the composites containing more than 20 % fiber concentration.

### 3. 4. Impact strength

Impact fracture most often takes place as a result of propagation of cracks originating at existing defects such as voids, foreign particles, and machining marks. The impact strength was measured by Izod impact test machine with unnotched specimens in the present study. For unnotched specimens, the impact energy includes the energy needed to initiate and to propagate a crack in the composites.

Figure 4 shows the unnotched impact strength of PP-based and HDPE-based composites as the function of wood fiber concentration. The impact strength of all composites decreased with the addition of wood fibers. PP-based composites showed higher impact energy than HDPE-based composites. This might be attributed to the inherent differences in crystallinity between two thermoplastics. The impact energy is more dependent on the fiber concentrations than types of plastic resins. In other words, the addition of wood fiber has larger influence compared with types of matrix resins. The unnotched impact specimen needed relaltively large amount of energy to initiate a crack in the skin. In this case, cracks nucleate at imperfections in the composites.

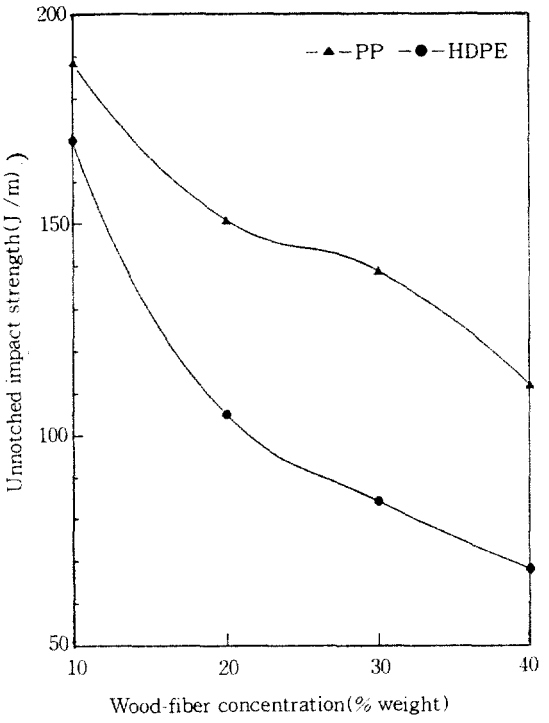


Fig. 4. Izod impact strength of the composites containing various wood-fiber concentrations.

Klason et al.[16] also found that unnotched Charpy impact strength fell sharply when wood flour concentration increased. Repeating the number of compounding cycles regardless of the compounding methods, Boldizar et al.[20] reported that the better dispersion gives rise to an increased fracture energy. However, no improvement was observed when the filler concentration was increased to 30 weight percent. Woodhams et al.[13] reported high Izod unnotched impact strength versus notched impact values, and attributed this to more highly oriented fiber in the outer skin caused during injection molding.

According to Nielsen[21], there are two mechanisms by which the fibers can reduce the impact strength of composites: (1) Fibers tend to reduce the elongation to break and may reduce the area under the stress-strain curve. (2) Stress concentrations may occur at regions around fiber ends, areas of poor adhesion, and regions where fibers contact one another. When a composite material is cracked by an impact, a crack exposes the fibers as the matrix separates. The crack then either continues through the fiber causing fiber breakage or travels along the fiber/matrix interface, resulting in fiber pull-out. Particularly in wood-fiber thermoplastic composites, however, the large aspect ratio of most wood fibers and their tortuous configuration (twisted ribbons) tend to inhibit fiber pullout. So a brittle fracture is normally observed even without coupling agents [13].

## 4. CONCLUSION

Wood fibers were successfully compounded with thermoplastsics by using a high intensity thermokinetic mixer (K-mixer). The incorporation of wood fiber improved the strength and modulus of PP- and HDPE-based composites. Normally the flexural strength has larger than tensile strength because of the fiber alignment in the skin of injection molded specimens. The tensile and flexural modulus proportionately increased with the fiber concentration. The percent elongation and energy at break was reduced by the addition of wood fibers. Izod unnotched impact strength also decreased with the added wood fibers. The potential of wood fibers as an reinforcement in thermoplastics was found in terms of mechanical properties of the composites, especially strength and modulus whereas the reinforcement increased the brittleness. Further study is needed first to investigate the influence of processing conditions on wood-fiber reinforcement in thermoplastics and secondly to evaluate the dimensional stability in high humid and variable temperature environments. Other studies may include the improvement of impact toughness and the understanding of the time-dependent deformation of the composites.

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