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Thermal Conductivity of Thermoplastics Reinforced with Natural Fibers[1]

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With restrictions for environmental protection being strengthened, thermoplastics reinforced with natural fibers such as jute, kenaf, flax, etc., have replaced automotive interior materials such as chemical plastics. In this study, the thermal conductivity of several kinds of thermoplastic composites in the form of board composed of 48.5 mass% polypropylene (PP) and 48.5 mass% natural fiber (NF), and reinforced with 3.0 mass% maleated polypropylene (MAPP) and 0.3 mass% silane as the coupling agents, were measured at temperatures of -10, 10, and 30°C, using a heat flow meter apparatus. The results show that the thermal conductivity is in the range of 0.05–0.07 $W \cdot m^{-1} \cdot K^{-1}$, and the thermal conductivity increased about 10–15% by adding MAPP and about 10–25% by soaking in a silane aqueous solution. The tensile strength was also measured, and the result shows similar trends as the thermal conductivity.

KEY WORDS: coupling agent; natural fiber; tensile strength; thermal conductivity; thermoplastic composite.

1. INTRODUCTION

Natural fibers as a substitute for glass fibers in composite components, have gained renewed interest in the automotive industry. Carmakers are

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looking increasingly at thermoplastics reinforced with natural fibers to reduce weight and cost in interior and engine components [1]. To reduce vehicle weight, a shift away from steel alloys towards aluminum, plastics, and composites has led to predictions that in the near future, polymer and polymer composites would comprise about 15% of a car's weight. Of all the thermoplastic matrices available, polypropylene (PP) shows the most potential benefits when combined with natural fibers in making composites for industrial applications [2].

A notable shortcoming in the natural fiber–thermoplastic system is the poor bonding between the natural fiber and the plastic. This is due to the dissimilar chemical nature, i.e., the surface of the natural fiber is hydrophilic while that of the plastics is generally hydrophobic. In order to develop composites with better mechanical properties, it is necessary to impart hydrophobicity to natural fibers by suitable chemical treatments [3]. The selection of proper coupling agents is also important to improve fiber–matrix adhesion so as to produce composite materials with superior strength. Also, the energy saving in connection with car air-conditioning becomes very important; thus, the study about the effect of coupling agents to the thermal properties become important.

In this study, the thermal conductivity and tensile strength of thermoplastic polypropylene (PP) boards reinforced with natural fibers (NF) and coupling agents such as maleated polypropylene (MAPP) and silane are investigated.

2. MATERIAL PREPARATION

2.1. Coupling Agents

Epolene MAPP (maleated polypropylene) acts as a compatibilizing agent in polymer blends and is particularly effective when one polymer is hydrophilic and the other polymer is hydrophobic [4]. The compatiblization of the dissimilar polymers dramatically improves the properties of the polymer alloy or blend. Individual silane (aminoethylaminopropyltrimethoxy silane or AEAPTMS) coupling agent molecules which are supposed to attach to natural fibers form a continuous link. The long hydrophobic polymer chain of polymerized silane can adhere to polypropylene due to a van der Waals type adhesive force. As a result, silane coupling agents form cross links in the interface [5].

Both aqueous silane solution and MAPP were mixed to produce the required sizing for natural fiber treatments. In the case of hybrid sizing, the maleic anhydride functionality of MAPP reacts with the amino group

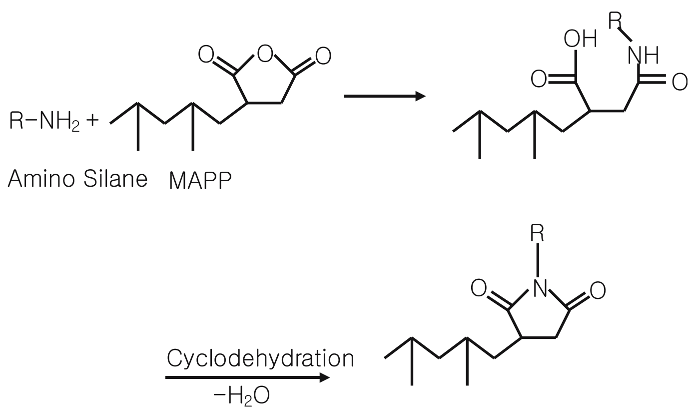

Fig. 1. Mechanism showing coupling reaction between maleic anhydride functionality of maleated PP with amino functionality of silane.

of silane as shown in Fig. 1 and thus creates a strong interface, which is expected to improve the fiber–matrix adhesion significantly [3,6].

2.2. Material Mixing

Most work on natural fiber-PP composites is based on melt mixing of short length natural fibers and polypropylene matrix granules with subsequent injection molding. Such two-stage processing techniques expose natural fibers to high shear and thus damage the natural fibers. The best use of natural fibers will occur when processing methods reduce or eliminate fiber damage [7].

A twin screw co-rotating extruder (SCE) is developed for the direct incorporation of 48.5 mass% PP, 48.5 mass% natural fibers, and 3.0 mass% MAPP. The purpose of this equipment is to provide impregnation and homogenization between the polymer matrix and the natural fibers. The natural fibers are fed simultaneously with the melt of PP and MAPP. Inside of this equipment the natural fibers are impregnated, cut, and homogenized. Figure 2a shows a photograph of mat formed by mixing the PP, NF, and MAPP.

The chopped (50–80 mm) length natural fibers, e.g., kenaf/hemp/flax/sisal and micron size PP powder were used for composite fabrications. The main motivations for using natural fibers to replace glass fibers include the low cost (~1/3 of glass fibers), low density (~1/2 of glass), acceptable specific strength properties and enhanced energy recovery, CO_2 sequesterization, and bio-degradability [8].

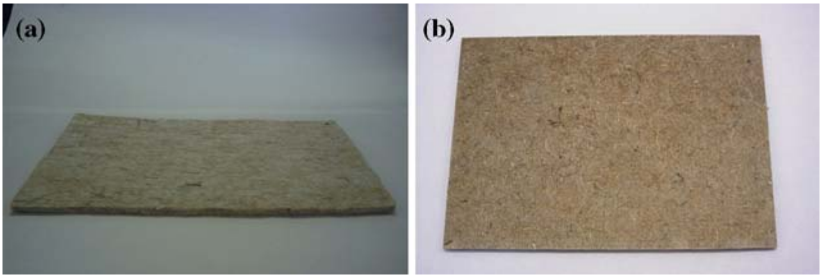

Fig. 2. (a) Photograph of a felt formed by mixing PP, NF, and MAPP and (b) board formed by compression molding.

2.3. Sample Boards

The chopped natural fibers and powder PP, after being mechanically mixed in a kitchen mixture, were subjected to compression molding processing. Then the materials were kept under contact temperature at 200°C for about 15 min under mild pressure followed by pressing at 350 to 360 $N \cdot m^{-2}$ pressure for about 2 min and cooling under the pressure to obtain the final composite plaques for testing. For natural fiber-PP composites, MAPP has been effectively used as an adhesion promoter to improve the physico-mechanical properties of the composites. Figure 2b is a photograph of the sample board produced by compression molding.

Figure 3 shows a micro-photograph of the three kind of samples (Sample A: PP(50 mass%) + NF(50 mass%), Sample B: PP(48.5 mass%) + NF(48.5 mass%) + MAPP(3 mass%), Sample C: same as Sample B, but soaked in 0.3% silane aqueous solution) prepared by compression molding. We can see that the micro-structures of each samples are different. The measured densities of each sample are 114.19, 125.09, and 136.28 $kg \cdot m^{-3}$, respectively. Therefore, we can assume that Sample A has a large porosity; however, Sample B has a slightly reduced porosity. Sample C has a much reduced porosity, and the fiber–matrix adhesion is significantly improved.

3. EXPERIMENTAL RESULTS

3.1. Thermal Conductivity

The thermal conductivity of three samples given above were measured by a heat flow meter apparatus (RK-30) following ASTM-C 518; Standard Test Method for Steady-State Thermal Transmission Properties by Means of the Heat Flow Meter Apparatus and ISO 8301; Thermal Insulation,

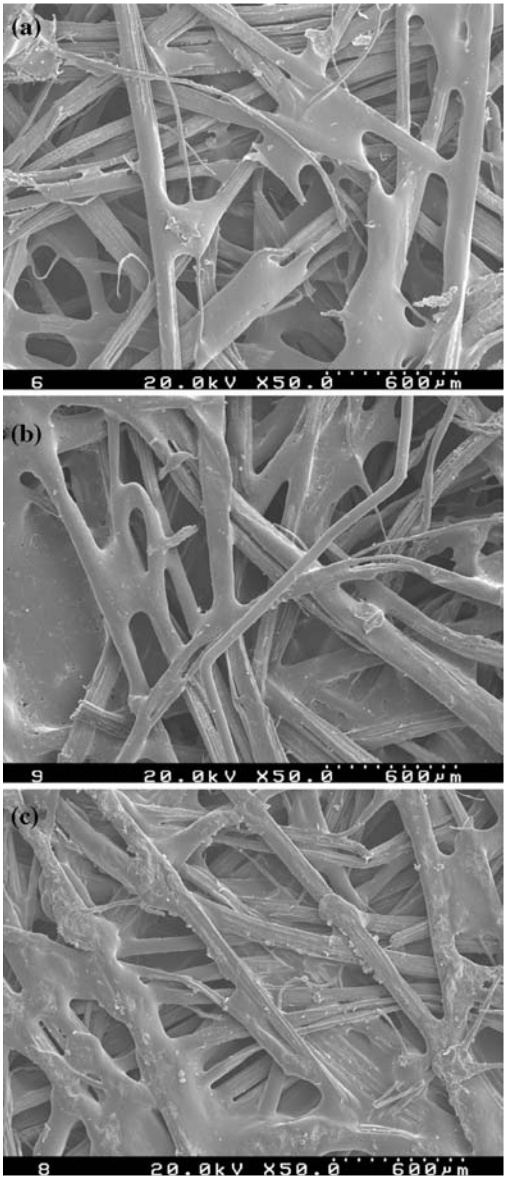

Fig. 3. Micro-photograph of the three kinds of samples (a) sample A: PP(50 mass%) + NF(50 mass%), (b) sample B: (48.5 mass%) + NF(48.5 mass%) + MAPP(3 mass%), (c) sample C: Sample B, also soaked in 0.3 mass% silane aqueous solution.

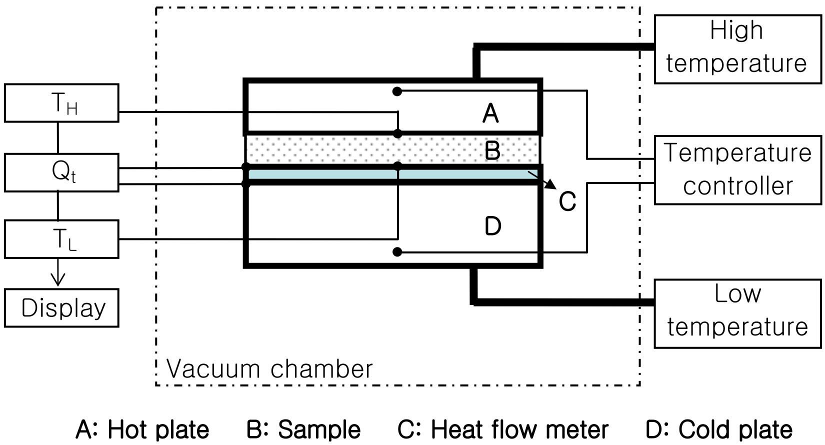

Fig. 4. Schematic of heat flow apparatus for measurement of thermal conductivity.

Determination of Steady-State Thermal Resistance and Related Properties. A schematic of the apparatus is shown in Fig. 4. The thermal conductivity can be calculated from the temperatures T_H and T_L, the heat flow Q_t, and the thickness of the sample [9]. The thermal conductivity was measured at temperatures of −10, 0, and 10°C, The measurements were made five times at each temperature. The sample size is 300 mm × 300 mm, and the thickness is 18–25 mm.

The average values of the thermal conductivity obtained in the present study are shown in Table I. As the temperature increases, the thermal conductivity of all the samples increases because in this case the vibration of the phonons is the thermal carrier and the moisture in the natural fiber begins to evaporate and escapes from the sample. The thermal conductivity of Sample B increased about 10–15% by adding 3 mass% of MAPP as a coupling agent and that of Sample C increased about 15–20%. These results can be understood by the change of density and micro-structure of the samples induced by the coupling agent effects as shown in Fig. 3. As expected in Fig. 1, the coupling agents of MAPP and silane form cross links in the interfaces of PP and NF and the porosities are reduced.

3.2. Tensile Strength

Tensile tests were performed using an Instron Model 4206 Universal Testing Instrument. A 150 kN load cell with a load weighing system accuracy of 1.0% of reading was used. Five samples were prepared for each case. The size of the sample is 120 mm × 25 mm, and the shape is shown in

Table I. Experimental Thermal Conductivities of the Thermoplastics Reinforced with Natural Fibers (compositions in mass%)

Temperature (°C)			Thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$)		
Average Temperature	Cold Plate	Hot plate	Sample A PP(50%) + NF(50%)	Sample B PP(47.5%) + NF(47.5%) + MAPP(3%)	Sample C PP(47.5%) + NF(47.5%) + MAPP(3%) + Soaked in silane solution
−10	−20	0	0.0500 ± 0.0027	0.0540 ± 0.0024	0.0597 ± 0.0036
10	0	20	0.0516 ± 0.0031	0.0583 ± 0.0030	0.0648 ± 0.0038
30	20	40	0.0550 ± 0.0022	0.0633 ± 0.0038	0.0692 ± 0.0037
Thickness (mm)			24.487	19.674	18.839

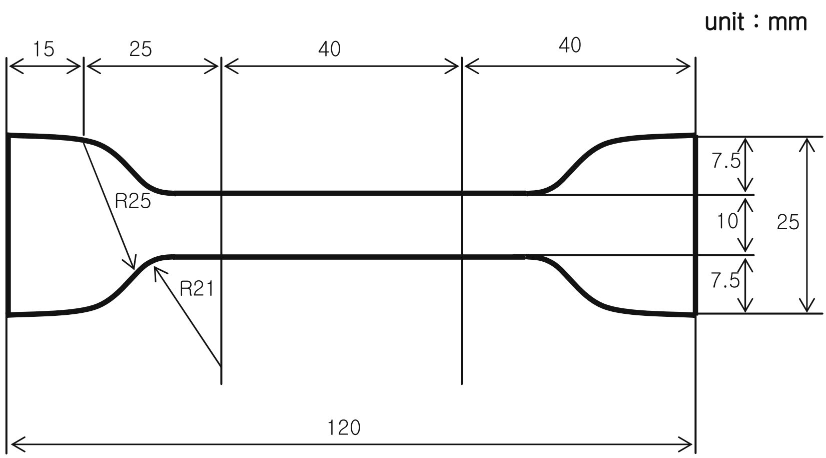

Fig. 5. Size and shape of the sample for measurement of tensile strength.

Fig. 5. Table II shows the measured tensile strengths of the samples [10]. The result shows that the tensile strength increases on adding the coupling agents, and this trend is quite similar to the case of thermal conductivity because the coupling agents formed cross links in the interfaces of PP and NF.

Table II. Experimental Tensile Strength of the Thermoplastics Reinforced with Natural Fibers

Samples		Tensile Strength ($N{\cdot}cm^{-2}$)	Thickness (mm)
Sample A	1	40.11	3.75
	2	45.43	3.78
	3	40.20	3.78
	4	42.74	3.68
	5	46.93	3.71
Sample B	1	48.83	4.68
	2	49.50	4.03
	3	47.01	4.15
	4	50.08	4.13
	5	47.24	4.40
Sample C	1	64.90	4.79
	2	64.80	4.79
	3	57.70	4.88
	4	55.90	4.86
	5	58.67	4.88

4. CONCLUSIONS

The thermal conductivity and tensile strength of several kinds of thermoplastic composite board composed of 48.5 mass% polypropylene (PP), 48.5 mass% natural fibre (NF), and two kinds of coupling agents were measured. The results show that both the thermal conductivity and tensile strength increase on adding coupling agents. These thermophysical data have significant potential benefit for thermal design of automobiles.

5. ACKNOWLEDGMENT

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