Interfacial Evaluation of Modified Jute and Hemp Fibers/Polypropylene (PP)-Maleic Anhydride Polypropylene Copolymers (PP-MAPP) Composites Using Micromechanical Test and Nondestructive Acoustic Emission

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Micromechanical 시험법과 음향방출을 이용한 Flax 와 Hemp 섬유 강화 에폭시 복합재료의 계면 물성 평가

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Keywords: Natural fiber, interfacial evaluation, dynamic contact angle, Interfacial shears strength (IFSS), acoustic emission (AE).

ABSTRACT

The surface energies and acid–base interaction between the untreated and treated Jute or Hemp fibers and different matrix compositions of polypropylene-maleic anhydride polypropylene copolymers (PP-MAPP) were investigated using dynamic contact angle measurement. The contribution of the acid-base property into the interfacial adhesion of the natural fibers/matrix systems were characterized by calculating the work adhesion coming from the acid-base interaction. On the other hand, microfailure mechanism of both single Jute and Hemp fiber bundles were investigated using the combination of single fiber tensile test and acoustic emission. Distinctly different microfailure modes of the different natural fiber/polypropylene systems were observed using optical microscope and determined indirectly by AE and their FFT analysis

Nomenclature

W_a	: Work of adhesion
γ^-	: Donor component
γ^+	: Acceptor component
γ^p	: Polar component
γ^d	: Dispersive component
γ^{LW}	: Lifshitz-van der Waals interactions
M	: A measured force
D	: Fiber diameter

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1. INTRODUCTION

In natural the plant fibers exhibit a high hydrophilic property as they are composed of lingo-cellulose, which contains strongly polarized hydroxyl groups [1,2]. Therefore, these fibers are inherently incompatible with hydrophobic polymer matrix materials and especially for poor interfacial adhesion between the hydrophilic natural fibers and resin matrices. The matrix materials are popularly used in biodegradable composites including thermoplastics such as a polypropylene and a polyethylene as well as even thermosets such as an epoxy, unsaturated polyester and cardanol based matrix [3,4]. Since the final mechanical behavior of a composite material depends to a great extent on the adhesion between the reinforcing fiber and the surrounding matrix, it is necessary to research the interfacial adhesion extensively [5].

Wetting is a perquisite to get a good interfacial adhesion.

However, interfacial adhesion also depends on the number of functional groups which contribute to acid-base interaction in interfacial area between the fiber and the matrix material. The acid-base interaction of the fiber surface is a significant factor in controlling the degree of adhesion. It is possible to find out the optimal combination of the fiber/matrix resin if the acid-base interaction of matrix resin is also determined. It is possible to evaluate the acid-base components using dynamic contact angle measurement; it follows the concept proposed by Fowkes which considers the short range hydrogen bonding is very important in adhesion [6]. In this work interfacial properties of natural fiber composites were evaluated micromechanical technique, wettability and AE.

2. EXPERIMENTAL

2.1. Materials

Hemp and Jute fibers (Carol Leigh's Hillcreek Fiber Studio, U.S.A.) were used as reinforcing fibers for green composites. Polypropylene (PP, Polymirae Inc., Korea) was used as thermoplastic matrix material and 1%, 3%, 5% modified maleic anhydride-polypropylene (MAPP-PP, Eastman Chemical Ltd.) were also used for comparison. Sodium hydroxide (NaOH) and Silane coupling agent, vinylbenzyl(trimethoxysilyl)propylethane-diaminehydrochliride (Z-6032, Dow Corning Corp.) were used. Four probe liquids used for contact angle measurement are double purified water, formamide (Dae Jung Chemicals & Kita-Ku., Ltd.), diodomethane (Tokyo Kasei Kogyo Co., Ltd.), ethylene glycol (Oriental Chemical Industries, Korea).

2.2. Methodologies

2.2.1. Contact angle measurement

Dynamic contact angles of Jute and Hemp fibers were measured using the Wilhelmy plate technique (Sigma 70, KSV Co., Finland). The basic equation for the Wilhelmy plate method is

$$F = mg + P\gamma_{LV}\cos\theta - F_b \qquad (1)$$

Where F is total force, m is mass of plate, g is a acceleration of gravity, F_b is buoyancy force, P is perimeter of fiber, γ_{LV} is surface tension of liquid, and the subtraction of $F-mg$ is equal to the measured force. Because the buoyancy force value is zero at the immersing interface. So the equation (2) can be arranged as follow.

$$Cos\theta = \frac{Mg}{\pi D\gamma_{LV}} \qquad (2)$$

In here M is a measured force. The acid-base component (or hydrogen bonding) includes electron acceptor γ^+ and electron donor γ^- components, which are defined as follow,

$$\gamma_S^{AB} = 2\left(\gamma_S^+\gamma_S^-\right)^{\frac{1}{2}} \qquad (3)$$

The calculation of these above components follows the modified young-Dupre equation of the work of adhesion which is given as.

$$\gamma_L(1+\cos\theta) = 2\left(\gamma_S^{LW}\gamma_S^{LW}\right)^{\frac{1}{2}} + 2\left[\left(\gamma_S^-\gamma_L^+\right)^{\frac{1}{2}} + \left(\gamma_S^+\gamma_L^-\right)^{\frac{1}{2}}\right] \qquad (4)$$

2.2.2. Specimen Preparation and IFSS Measurement

The natural fibers were fixed with regularly separated distance in a steel frame. Microdroplets of polypropylene matrix were formed on each the natural fiber using a tip pin. A microdroplet specimen was fixed by the microvice using a specially designed micrometer. The IFSS was calculated from the measured pullout force, F using the following equation,

$$\tau = \frac{F}{\pi D_f L} \qquad (5)$$

Where D_f and L are fiber diameter and fiber embedded length in the matrix, respectively.

3. RESULTS AND DISCUSSION

3.1. Morphology and structure of natural fibers

Figure 1 shows morphology in diameter direction and crossed section of Jute and Hemp fibers, for the natural fibers its diameter shape is generally not circular and the diameter is not uniformed along their length with large scattering level

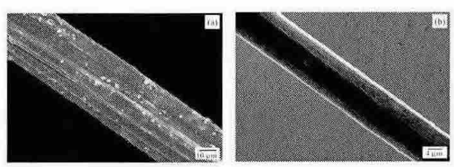

Figure 1. (a) Jute fiber, (b) Hemp fiber.

3.2. Analysis of strength distribution

Figure 2 shows the cumulative strength distribution of the untreated and treated (a) Jute and (b) Hemp fiber at the gauge length of 20 mm, respectively. The experimental strength data was estimated from $F(N) = i/(N+1)$, in here N is the total number of samples tested and i is the ith number in ascendingly ordered strength data. It was easily noted that for both Jute and Hemp fibers bimodal distribution curves were more fitted with measured data than unimodal distribution curves

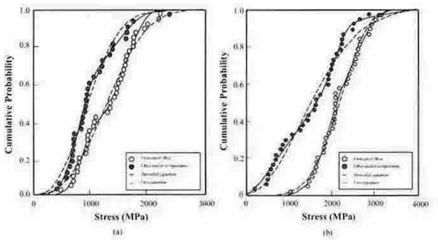

Figure 2. Uni- and bimodal Weibull distribution of the natural fibers :(a) Jute fiber; (b) Hemp fiber.

3.3. Microdroplet test

Figure 3 shows the plots of debonding force versus the embedded area for various natural fibers/polypropylene composites in microdroplet test.

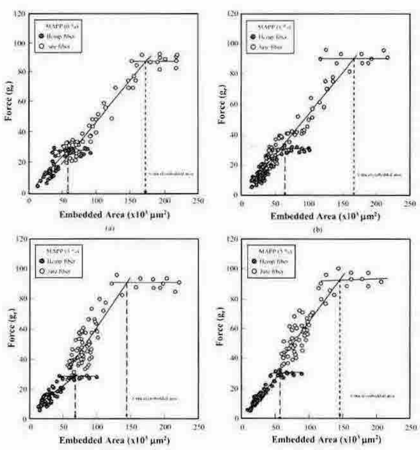

Figure 3. Plots of force vs embedded area for (a) MAPP (0%); (b) MAPP (1%); (c) MAPP (3%); (d) MAPP (5%).

It is noticed that the IFSS of given system can increase when the critical embedded area decreases. From the experimental plots the critical embedded area slightly shifted to the left hand side with an increase in MAPP content in PP-MAPP matrix material or after treating the natural fibers with alkaline solution and silane coupling agent. Figure 4 shows the IFSS of both Jute and Hemp fibers/PP-MAPP matrix composites with different treated conditions being calculated for each sample by the equation (3) and then determined by simply averaging.

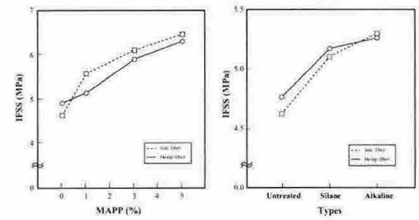

Figure 4. IFSS of Jute and Hemp fibers/PP with various treated conditions and contents of MAPP

From the experimental results the IFSS of the natural fibers/MAPP-PP significantly increases with increase in the content of MAPP introduced into the mixture of the materials as well as after treating with alkaline solution and silane coupling agent

3.4. Surface energy analysis and their wettability

Figure 5 shows the relationships between interfacial shear strength (IFSS) and the work of adhesion as a function of the content of MAPP. The IFSS increases with an increase in the work of adhesion for both Jute and Hemp fibers.

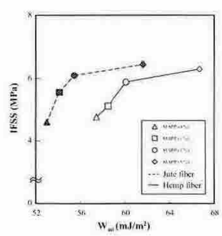

Figure 5. The relationship between the IFSS and work of adhesion of both Jute and Hemp fibers/MAPP-PP matrixs.

Figure 6 shows the relationships between the IFSS and work of adhesion as a function of the polar surface free energy and the acid-base interaction.

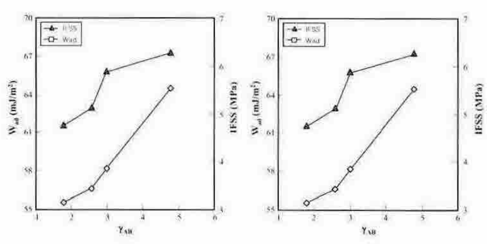

Figure 6. Plots of work of adhesion and the IFSS with acid – base interaction of the atrix materials: for (a) Jute fiber; (b) Hemp fiber.

The work of adhesion increases with an increase in the polar surface free energy and the acid-base interaction.

The increase in the work of adhesion and the IFSS may result from an increase in the acidic-basic character of the matrix materials in the order MAPP (5%)> MAPP (3%)>MAPP (1%) > MAPP (0%), respectively.

3.5. AE Outcomes correlating with their Microfailure Modes.

Figure 7 shows the different microfailure modes of single Jute and Hemp fibers after single fiber tensile testing for both low portion and high portion strength. From the photos can show that the microfailure mode of the high portion can be caused by slipping the elementary fibers inside of crystal material area where can respond to the high tensile strength before fracture occurs. In where the microfailure modes of the low portion can be caused by microfailure process inside of amorphous material area in which can responds to the low tensile strength.

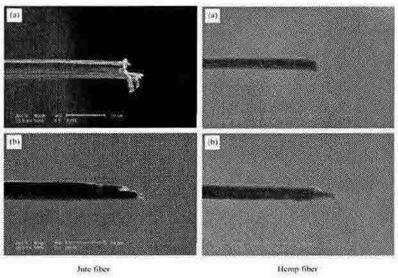

Figure 7. Microfailure modes of neat single Hemp fiber under tension: (a) high portion; (b) low portion

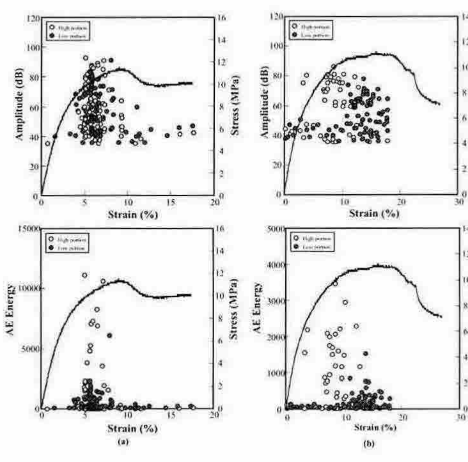

Figure 8. AE amplitude and AE energy of single Flax fiber bundle.

Figure 8 shows AE amplitude and AE energy of the two natural fibers/polypropylene (PP) composites for (a) Jute and (b) Hemp fibers, respectively. The experimental results showed that the amplitude and the energy of AE signals emitted by fractured process of the high strength fibers are significant higher than those of the low strength fibers. It may be because the microfailure process of the high portion happened through the crystal areas, whereas one of the low portions occurred through the amorphous areas.

4. CONCLUSION

Interfacial evaluation of Jute and Hemp fibers/various matrix material composites was performed using microdroplet test. With increasing the content of MAPP in the blend of PP made the IFSS increased. The IFSS also increased after treating the natural fibers with alkaline solution and silane coupling agent. The surface energy components of the fibers slightly decreased with silane coupling agent due to the high energy sites blocked, but increased after treating alkaline treatment. It may be because under alkaline solution the weaker boundary layers were removed and increased the surface area. On the other hands, the surface energy components increased with an increase in the MAPP content due to the number of the high energy sites introduced continuously when the content of MAPP increased. The experimental results of mechanical properties of the natural fiber show that bimodal Weibull distribution was fitting better with measured data than the unimodal distribution. The mechanical properties of the natural fiber significantly decreased under high temperature and slightly increased after treating with alkaline solution. Microfailure mechanisms of the single Jute and Hemp fibers with MAPP-PP systems were clarified consistently using nondestructive acoustic emission technique.

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