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Effect of chemical treatments on transverse thermal conductivity of unidirectional abaca fiber/epoxy composite

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ABSTRACT

Present paper investigated the effect of mercerization and silane treatments on the transverse thermal conduction properties of unidirectional abaca fiber–epoxy composite fabricated by resin transfer molding. As indicated by FTIR, XRD and SEM, the changes in chemical composition, crystalline and lumen structure of abaca fibers were introduced by chemical treatments. Transverse tensile test showed that the weakest linkage of unidirectional composite changed from interface between abaca fiber bundle and epoxy resin for untreated abaca fiber to interface between elementary fibers treated by mercerization and silanization. With the increasing of weakest linkages strength and the decreasing of void content, the transverse thermal conductivity (TCC) of the composite presents increasing trend. The changing of interfaces, cell wall and lumen derived from chemical treatments are the mainly factors affecting TTC. It was concluded that the abaca fiber composite with controllable transverse thermal conduction property can be designed by proper chemical treatment.

1. Introduction

In the last decades, a renewed interest toward the biomass materials has emerged due to the consumption of oil reserves and global environmental concern [1]. Plant fiber is one of the chief materials and has been often utilized in polymer-based composite because these fibers show the nature of biodegradability, renewability and low cost [2–4]. Specifically, bast and leaf fibers are considered as the high-performance fiber and show great potential as a reinforcement in green composite, such as bamboo, sisal and ramie . These cellulose fibers are porous and light weight, which makes plant fiber reinforced composite a good candidate as the thermal shield in building and vehicle [5,6]. However, few investigations have been carried out on the thermal insulation property of plant fiber reinforced composites compared to mechanical property.

For instance, fibers from hemp [7], flax [8], bamboo [9] and banana [10] have been introduced into polymers for studying the thermal conductivity of them. It has been reported that the composites show much lower transverse thermal conductivity (TTC) than longitudinal thermal conductivity, and that the addition of plant fiber promoted the thermal insulation property of composite in the direction across fiber. Further discussion in view of microstructure of plant fiber and composite has not been provided. Actually, plant fiber is essentially a multilayered composite in form of vascular bundle, of which rigid cellulose microfibrils are embedded in a soft lignin and hemicellulose matrix [11,12]. The chemical and structural status inside of plant fiber as well as the interfacial adhesion of hydrophilic plant fiber toward to polymers is both the factors influencing the properties of plant fiber and its composite [12]. Therefore, these factors without exception are necessary to be considered in studying the thermal conduction properties of plant fiber reinforced composite.

In our previous work, the TTC of plant fiber reinforced composite has been analyzed from the point of fiber cell wall and lumen structure with a conclusion that the TTC of unidirectional composites can be adjusted by selecting plant fiber with proper physical structure [13] (abaca fibers with high cellulose content and large lumen size tend to reduce the TTC of composite, while bamboo fibers with low cellulose content and tiny lumen size play a role of thermal conductor in the composite). Abaca fiber has been recognized with high quality such as: high aspect ratio, high cellulose content and high porosity, which offers a possibility of light composite in practical applications such as thermal insulation

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materials with excellent mechanical property [11,14,15], especially after fibers' pretreatment [2]. However, to our best knowledge, there is no paper reported on the microstructure change of abaca fiber through some treatments, not to mention the fiber composite. Therefore, the aim of this paper was to found an approach to change the surface and interior physicochemical structure of abaca fiber bundle chemically in order to regulate the TTC of unidirectional plant fiber–epoxy composite. In particular, it is expected that the mechanism of thermal conduction in unidirectional plant fiber composite can be known.

2. Experiments

2.1. Materials

Abaca fibers were supplied by Toho Tokushu Pulp Co. Ltd., Japan exported from Philippine with an average diameter of 186 µm. The fibers were extracted from the leaf sheathe of abaca plant by using decortication method [16] mechanically. Long abaca fibers were rubbed to remove the cuticle attached on the fiber surface and then twined around an aluminum thin sheet unidirectionally for straightening under running water. Finally, twined fibers were dried at 70 °C for 10 h and cut into 11 cm long.

The epoxy resin was a product of Mitsubishi Chemical Corporation (JER819, viscosity: 0.4 Pas at 25 °C, molecular weight: 180–200) with diethylenetriamine (DETA) as curing agent. Epoxy resin containing 7.0 wt% hardener was defoamed and mixed in vacuum mixer (Kurabo KK-250S) before used.

2.2. Fiber treatments

Abaca fiber was immersed in an aqueous solution of 1.0 wt% NaOH for 5 min (ATAF0) and 5.0 wt% for 30 min (ATAF), respectively, with fiber-to-solution weight ratio 1:30 at room temperature. The fiber was then washed thoroughly by water to remove the excess of NaOH, and dried at 70 °C.

Sequently, 1.0 wt% acetic acid and 1.0 wt% γ-glycidoxypropyl-trimethoxy silane (GPS, KBM-403, Shin-Etsu Chemical Co., Ltd. Japan) was dropped slowly into 98.0 wt% mixture of water and alcohol with weight ratio of 1:1 in sequence, and stirred for 60 min in an sealed opaque PVDF container. Then untreated (UTAF) and mercerized abaca fibers were soaked in the solution. The reaction was carried out for 24 h at a PH = 5.3. After that, the fibers were washed with distilled water and kept in air for 30 min to hydrolyze the coupling agent. Eventually, the reaction between hydroxyl groups of abaca fiber and silanols of KBM-403 was induced in an oven at 100 °C for 2 h. By using similar method, fiber samples of STAF, ASAF0, ASAF were obtained as shown in Table 1.

Table 1
Physical definition of fiber abbreviation.

Fiber abbreviation	Physical definition
UTAF	Untreated abaca fiber
ATAF0	Abaca fiber treated by aqueous solution of 1 wt% NaOH for 5 min at RT
ASAF0	Abaca fiber treated by aqueous solution of 5 wt% NaOH for 30 min at RT
ATAF	Abaca fiber treated by aqueous solution of 1 wt% NaOH for 5 min and then by silane at RT
ASAF	Abaca fiber treated by aqueous solution of 5 wt% NaOH for 30 min and then by silane at RT
STAF	Abaca fiber treated by silane at RT

2.3. Preparation of unidirectional abaca fiber–epoxy composite

The unidirectional abaca fiber composites were fabricated by a method [13] based on resin transfer molding (RTM) as shown in Fig. 1. The resin was one-way injected through the soft silicon tube into the home-made mold filled with unidirectional arranged fibers under the action of roller pump. The whole process was performed within 30 min before resin curing. The raw composite samples were post-cured at 100 °C for 2 h, and then grided and polished to the final size: 10 × 10 × 1 mm for thermal conduction test with stripe shape and 100 × 10 × 2 mm for tensile test with square shape. Since higher fiber content leads to longer processing time and thus higher resin viscosity [17,18], the fiber content was designed less than 40.0 wt%.

2.4. Characterization

In order to identify the effect of treatment on the chemical components of abaca fiber, Fourier transform infrared (FTIR) spectra of fibers were measured by Bruker 270 infrared spectroscopy. Wide angle X-ray diffraction (XRD) spectra were collected by a Bruker D8 ADVANCE X-ray diffractometer with graphite monochromatized Cu $K\alpha$ radiation (λ = 1.5406 Å) to detect the crystalline structure of fibers and composites. The crystallinity of abaca fiber (exactly cellulose I) was calculated by Eq. (1) [19]:

$$\text{Crystallinity} = (I_{002} - I_{18.5°})/I_{002} \times 100\% \tag{1}$$

where I_{002} and $I_{18.5°}$ are the diffracted intensity by (002) plane and the diffracted intensity at 2θ = 18.5°, respectively.

The cross section of fibers and composite specimens were obtained by fixed-point tensile breaking and then observed in SEM (JSM-7000F). Prior to examination, the specimens were sputtering coated with gold. The cross-sectional area of abaca fibers bundles was estimated using a statistical method assisted by a laser microscopy [20], and then the density was calculated using mass divided by volume (cross-sectional area multiplied by length) [21].

The transverse tensile test was conducted to verify the effect of chemical treatments on the strength of weak linkages in transverse direction of composite. The tests were performed at a speed of 1.0 mm/min according to ISO 527-4 [22]. For each case, four specimens were tested at a gauge length of 40 mm.

The thermal conductivities of specimens were measured using a flash method. Firstly, a coating of graphite was then applied to the specimens. And then the test was performed at 25 °C using a xenon light flash diffusivity system (LFA 447 Nanoflash, Netzsch Instruments, Inc, Germany) according to the ASTM standard E-1461 [23,24]. The thermal conductivities (K) of specimen were calculated using Eq. (2):

$$K = \alpha \cdot \rho \cdot C_p \tag{2}$$

where α the measured diffusivity, ρ the density and C_p the specific heat capacity of composites. The density of the specimens was measured using an Archimedes method. The specific heat capacity was obtained using the rule of mixture in Eq. (3) [7]:

$$C_p = V_f C_{pf} + (1 - V_f)C_{pe} \tag{3}$$

where C_{pf} and C_{pe} are the specific heat capacities of fiber and epoxy, respectively, measured in a Netzsch Simultaneous Thermal Analyzer STA 449C Jupiter with sapphire as a heat capacity reference. V_f is the volume fraction of abaca fiber bundle calculated using Eq. (4) [21]:.

$$V_f = \frac{w_f \rho_m}{w_f \rho_m + w_m \rho_f} \tag{4}$$

where w_f and w_m are the weight percent of abaca fiber and epoxy resin, ρ_m and ρ_f are the densities of abaca fiber and epoxy resin.