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Property enhancement of kenaf fiber reinforced composites by in situ aluminum hydroxide impregnation

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ABSTRACT

This work was to apply the vacuum-assisted resin transfer molding (VARTM) process and use aluminum hydroxide inorganic nanoparticle impregnation (INI) to improve the mechanical properties and water resistance of the kenaf fiber reinforced composites. As a novel technique, a two-step aluminum hydroxide impregnation was used in the fabrication of composites, including the alkali retting and aluminum hydroxide loading. After the alkali retting, the cellulose content of the kenaf fibers went up to 94.2%, the ash content was 2.7%. The tensile strength and modulus of individual fiber were 810 MPa and 13.5 GPa, respectively. The results showed that, with a loading amount of 2.2% aluminum hydroxide, the modulus of elasticity, modulus of rupture, tensile modulus and tensile strength of the composites made with INI-treated fibers were increased by 67.2%, 55.7%, 32.5% and 36.3%, respectively, compared with those of composites made from un-treated fibers. When the INI-treated fibers were employed, the thickness swelling of 24-h water submersion was reduced from 19.7 to 6.3%, and the moisture contents of the composites after the conditioning and water submersion were reduced from 5.8% to 1.9%, and 18.3% to 6.6%, respectively. The improvement makes the kenaf fiber reinforced composites possible to replace the glass fiber composites for the automotive application.

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1. Introduction

Large quantities of potentially useful but largely underutilized kenaf (*Hibiscus cannabinus*, L. Malvaceae) fibers presently exist and are available for converting into value-added products (Kalaycıoglu and Nemli, 2006; Liu et al., 2007; Okuda and Sato, 2007). Kenaf is under the category of natural biopolymers due to its friendly properties of environmental aspects. Because of the excellent characteristics of low density, low cost, and availability, interest has recently arisen in the manufacturing of kenaf composites (Akil et al., 2011; Feng et al., 2002; Liang et al., 2013; Tsakonas et al., 2005; Zampaloni et al., 2007). Deka et al. (2013) developed "all green composites" from kenaf bio-fiber and poly bio-resin based on renewable resources. Tawakkal et al. (2014) improved the mechanical and thermal properties of PLA/kenaf/thymol composites by adding kenaf fibers. Bledzki et al. (2015) created polypropylene bio-composites reinforced with kenaf fibers. Papadopoulou et al. (2015) used bast fiber crops to produced value-added industrial products, such as, ideal feedstocks for the production of high quality yarns and paper, for rendering flame retardant properties to composites reinforced with them, for the production of energy via a sustainable way without environmental pollution and for the use as alternative materials to wood-based panels such as particleboards and fiber boards. Ali et al. (2015) used the response surface methodology to improve the dimensional stability of kenaf panels by post-manufacturing hydrothermal treatments.

The vacuum-assisted resin transfer molding (VARTM) technology is one of the well-established processes used to fabricate high-performance composites using synthetic fibers, such as glass or carbon fibers (Agirregomezkorta et al., 2012; Brouwer et al., 2003; Cecen and Sarikanat, 2008; Park et al., 2012). VARTM is also employed to manufacture natural fiber reinforced composites to increase the mechanical properties (Xia et al., 2015a). The mechanical properties of composites was significantly increased by the VARTM technology compared to the traditional hot-pressing

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process, owing to the porosity reducing in the composites (Xia et al., 2015a).

There have been some concerns regarding natural fiber reinforced polymers, e.g. matrix-fiber interfacial adhesion, fiber dispersion, and porosity of the fibers (Bledzki et al., 2005). The efficiency of the load transfer from matrix to fiber was reduced due to the bad bonding at the interface of the composites, resulting in poor mechanical properties of the composites and weak to environmental attacks (Wambua et al., 2003). Some studies were conducted for the improvement in mechanical properties of kenaf composites (Anuar and Zuraida, 2011; Yang et al., 2014). To enhance the interaction between natural fibers and matrix, various fiber treatment methods were explored. Among these treatment methods, the inorganic nanoparticle impregnation (INI) was an effective method to improve the compatibility between natural fibers and polymer matrices (Shi et al., 2011). As a promising surface treatment method, calcium carbonate nanoparticles have been used to address these concerns (Cheng et al., 2014a,b; Shi et al., 2011). In the previous studies, the mechanical properties of both kenaf fibers and kenaf fiber/polymer composites were increased by impregnating calcium carbonate nanoparticles into kenaf fibers (Liang et al., 2014).

With advantages of non-toxic, odorless and good dispersion, aluminum hydroxide ($Al(OH)_3$) is widely used as fillers in the papermaking industry (Take et al., 1988; Yan and Deng, 2002). As a hydrophilic inorganic compound (Vroman, 1962), $Al(OH)_3$ is expected to be hydrogen bonded with cellulosic fibers. In our previous research about calcium carbonate impregnated fiber composites (Xia et al., 2015b,c), a three-step process was carried out, including alkali retting, calcium chloride impregnation, and calcium carbonate forming by adding the sodium carbonate. To simplify the fabrication process and reduce the consumption of chemicals, a more efficient and economic two-step $Al(OH)_3$ impregnation was proposed in this study including the alkali retting and $Al(OH)_3$ formation.

No study was reported on the manufacturing of kenaf composites by means of $Al(OH)_3$ impregnated natural fibers using the VARTM process. The objectives of this study were to fabricate composites using $Al(OH)_3$ impregnated kenaf fibers through VARTM process and explore the effect of $Al(OH)_3$ on the mechanical properties and water resistance behavior of the composites.

2. Materials and methods

Sourced from Kengro Corporation (MS, USA), the kenaf bast fibers were chopped into approximate 50.8 mm in length with a heavy duty paper trimmer. Sodium hydroxide (NaOH) solution (5%, w/v) was prepared using NaOH beads (≥97%, Acros Organics) and deionized water that was from the Millipore Milli-Q Integral Water Purification System. The 0.05 mol/L solution of aluminum chloride ($AlCl_3$) was prepared from $AlCl_3{\cdot}6H_2O$ (99%, Sigma–Aldrich). Unsaturated polyester AROPOL Q6585 (30% styrene, Ashland Chemicals) and *tert*-butyl peroxybenzoate (*t*-BP, 98%, Acros Organics) were used to fabricate the fiber reinforced composites.

2.1. $Al(OH)_3$ loading through INI processes

A mixture of 100 g kenaf bast fibers with a moisture content of 9.11% (measured by Mettler-Toledo HB43-S Moisture Analyzer) and 1.8 L NaOH solution (5%, w/v) was added into a hermetical reactor (251 M, Parr Instrument Co., USA). This alkali retting process was carried out at 160 °C for one hour with the mechanical stirring, with a saturated vapor pressure of 0.60 MPa. After cooling to room temperature (~4 h), the excessive ionic solution was removed from kenaf fibers firstly by gravity, and then by hand-squeezing. The retted fibers were measured as 36.6 ± 1.2% of the original ones, including 94.2% holocellulose, 92.3% α-cellulose, 0.9% hemicellulose, 0.24% lignin, and 2.7% ash (Shi et al., 2011). The mechanical properties of individual digested fiber were measured to be 287.91 MPa surface hardness and 5.24 GPa elastic modulus from nanoindentation tests, and 13.5 GPa modulus, 810 MPa strength and 5.6% elongation from tensile tests (Shi et al., 2011).

The in situ $Al(OH)_3$ loading process was carried out by mixing the fresh retted fibers which were hand-squeezed without washing, and 1.8 L $AlCl_3$ solution (0.05 mol/L) into the hermetical reactor. The reactor was then heated to 100 °C with a saturated vapor pressure of 0.10 MPa and maintained for 1 h. After cooling to room temperature, the mixture was filtered, washed with running water, and then dried at 105 °C for 24 h in a frame with a dimension of 100 × 165 mm (width and length) to form a mat preform with a thickness of approximate 10 mm. The $Al(OH)_3$ impregnated fiber (INI-treated) preform were prepared for the fabrication of composites.

2.2. $Al(OH)_3$ content determination

According to the difference in ash contents between the retted kenaf fibers and $Al(OH)_3$ impregnated kenaf fibers, the $Al(OH)_3$ contents in the composites were estimated. The ash contents were determined by burning the samples in a muffle furnace first at 400 °C for 30 min, and then at 850 °C for 45 min. The TGA data showed that $Al(OH)_3$ was fully decomposed into Al_2O_3 at 850 °C (Wang et al., 2013b). Based on the Al_2O_3 amount obtained by the ash difference, the $Al(OH)_3$ content was determined to be 2.2 ± 0.2% in the $Al(OH)_3$ impregnated kenaf fibers.

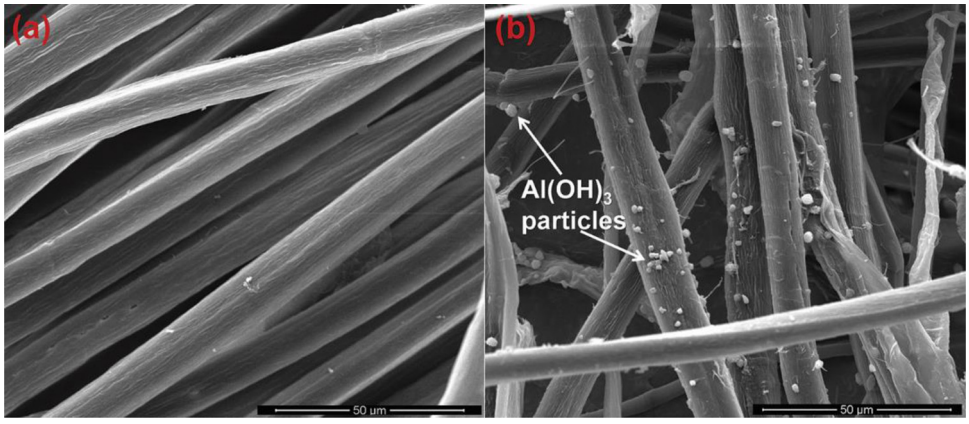

Fig. 1. SEM observations of the un-treated fiber (a) and INI-treated fiber (b).