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Research Paper

Impacts of hemp fiber diameter on mechanical and water uptake properties of polybenzoxazine composites

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ABSTRACT

The waste hemp fibers (HF) were alkali treated; then shredded, ball milled, and sieved. The 20, 40, and 80 mesh collected HF was reinforced in polybenzoxazine. The effects of fiber diameter and vol.% loading on tensile, impact, flexural and water uptake properties of composites have been studied. On 30 vol.% loading of 20 mesh HF, an increase of 91%, 43%, 168%, 137%, and 73% in tensile strength, flexural strength, impact strength, tensile modulus, and flexural modulus, respectively. The additional enhancement was observed in all mechanical properties as fiber diameter was reduced from 311.5 ± 42.9 to 198.3 ± 40.7 µm (20–80 mesh). The similar behavior was confirmed by Halpin-Tsai model. The water uptake was also increased as vol.% loading increased and HF diameter decreased, 28.2% higher water uptake was observed on 20 vol.% loading as the diameter was reduced from 20 to 80 mesh.

1. Introduction

The rapid rise has been observed in the production of natural fiber (NF) reinforced composites mainly for the automotive and construction fields, due to the human desire to consume environment sustainable and friendly products. The reinforcement of NF such as hemp, jute, sisal, coir, etc; instead of glass fiber and carbon fiber brings many advantages. NF are biodegradable and renewable in nature, have lower density, produce lower quantity of harmful gases and solid residues on the combustion, and lower wear to molds and machinery was observed during processing. Therefore, research interests have been transferred towards the development of novel environment friendly materials with improved properties (Jabbar et al., 2016; Bismarck et al., 2002; Fortea-Verdejo et al., 2017; Vanleeuw et al., 2015; de Vega and Ligerode Vega, 2017; Hamid et al., 2012; Sam-Brew and Smith, 2015).

China leads the worlds in the production of hemp fiber (HF), during 2000–2015 the average annual production of 93,000 tons was recorded, as reported by the Food and Agriculture Organization of United Nations (FAO). Considerable quantities of solid residues are generated during the separation process of HF from plant, mainly regarded as a waste or low-value byproducts. The smaller length and lower diameter HF are discarded as waste (noil) HF, while hemp hurds and shives are low-value byproducts. These low-value byproducts are successfully utilized in several fields as reinforcement materials (de Vega and Ligerode Vega, 2017; Balčiūnas et al., 2016; Li et al., 2017). However, the noil HF are discarded as waste, the reinforcement of noil HF in polymeric composites can be a sustainable process for the utilization of waste HF.

Polybenzoxazine resins are well known for their advantages over the traditional epoxy and phenolic resins. These advantages include their excellent thermal and mechanical properties, low water absorption, high resistance to heat and chemicals, lower flammability, and near-zero volumetric change on curing (Brunovska et al., 2000; Dong et al., 2006; Jin Kim et al., 1999; Espinosa et al., 2004; Ning and Ishida, 1994; Yagci et al., 2009). Different kind of natural fillers are successfully reinforced in the polybenzoxazine resins, such as hemp (Dayo et al., 2017a; Dayo et al., 2017b), kenaf (Dansiri et al., 2002), sisal (Tragoonwichian et al., 2008), basalt fiber (Kim et al., 2012), ramie fabric (Li et al., 2015), chitosan (Ramdani et al., 2015), and crab shell particles (Ramdani et al., 2014).

In this study, the effects of HF diameter on the composite properties have been investigated. The fibers were distributed on the basis of their diameter and composites with varying vol.% were examined for their

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tensile, impact, flexural, and water uptake properties; and results were compared with pristine resin having no fiber. Furthermore, morphological study was conducted to find out the composites fracture behavior.

2. Material and methodology

2.1. Materials

The noil HF of the separation process from the hemp bark and bisphenol-A aniline based benzoxazine monomer (BA-a) were kindly received from Daqing Branch of Heilongjiang Academy of Science, Daqing and Jiangxi Huacui Advanced Materials Co., Ltd. (China), respectively. Ethanol, cyclohexane, and sodium hydroxide (NaOH) were procured from Shanghai Jingchun Reagent Co., Ltd. (China) and used as-received, deionized water was used throughout the experiments.

2.2. Treatment of noil hemp fiber

The NF are hydrophilic in nature, while the benzoxazine resin is hydrophobic, resulting poor fiber-matrix interfacial adhesion. It has been observed that the surface modification of NF can quietly overcome this problem. HF were subjected to the alkaline treatment with NaOH solution (5 wt.%). As-received HF were first subjected to washing cycle (i.e. with water, ethanol/cyclohexane (1:1, V/V), and water) to remove the attached physical as well as chemical impurities. Physical impurities include mud and silt, while chemical impurities means fats and waxes. Then the washed fibers were rinsed in the NaOH solution (5 wt.%) for 5 h. Afterwards, the alkali treated fibers were neutralized by washing several times with acetic acid solution (1 wt.%) and water, and overnight vacuum dried at 60 °C. Resultant dried fibers were passed from the shredder having 1 ± 0.20 mm opening. The obtained fibers were grinded in a planetary ball-mill (pulverisette 7, FRITSCH) in a ZrO_2 vessel at a speed of 400 rpm for 12 cycles; each cycle composed of 30 min in operation and 15 min in rest, while the ball-to-material weight ratio was kept as 15:1. Grinded HF were separated on the basis of their diameter by using vibrating sieve shaker, HF collected on 20, 40, and 80 mesh Tyler sieve standard are used in the study.

2.3. Poly(BA-a)/treated hemp fiber composites preparation

HF reinforced polybenzoxazine composites were produced by molding under isothermal heating and compression. HF composite with 20, 40, and 80 mesh size HF were developed, while the fraction of HF in the composite was kept as 10, 15, 20, and 30 vol.% for the composite. Eq. (1) was used to determine the fiber vol.% (V_f) in composite (Gibson, 2016).

$$V_f = (W_f \times \rho_r)/(W_r \times \rho_f + W_f \times \rho_r) \times 100\% \quad (1)$$

Where W and ρ represent the weight and density, respectively, while the subscripted f for fiber and r for resin.

Prior to the composite preparation the nonwoven mat of HF was prepared. The predetermined HF mass was vacuum dried in a vacuum oven at 65 °C for 2 h, hand layered on 150 × 150 mm^2 kraft paper and covered with kraft paper, and hydraulically compressed at 10 MPa, to form a nonwoven mat. The mold release agent was applied on steel mold and heated at 140 °C. The desired mass of BA-a monomer was degassed in a vacuum oven at 90 °C for 6 h and poured into the preheated mold in two steps, so that nonwoven fiber mat was sandwiched in between the BA-a resin. Then the mold was heated isothermally at 150 °C for 2 h, then, 10 MPa pressure was applied and heated at 165 and 180 °C for 2 h at each stage. The procedure followed for the HF treatment and composite making is represented in Fig. 1, while Bz/HFX (Y) coding standard was used for all samples, where Bz and HF are indicating the BA-a benzoxazine and hemp fiber, while X is vol.% of HF and Y is mesh size on which HF were collected.

2.4. Characterizations

The length and diameter of HF specimen were determined by an optical microscope (Nikon, ECLIPSE E600 POL). 150–160 readings for each sieved collected HF were noted and average values with standard deviation are reported. The tensile and flexural tests were recorded on Instron 5569 at 1 mm/min crosshead speed; the specimen size was about 50 × 10 × 2 mm^3. The Izod impact test was performed in accordance to ASTM D256-2010 on Tinius-Olsen impact-resistance device. The rectangular specimen of 63.5 × 12.7 × 3.2 mm^3 was notched at 45°, five samples were tested and mean value of absorbed energy was reported. Water absorption test was performed according to ASTM D570–98 standard and an average of five samples was reported as result. A rectangular specimen having 76.2 × 25.4 × 3 mm^3 dimensions were oven dried at 50 °C for 24 h, cooled in a desiccator, and weighed on 4 decimals balance. After that, the specimens were immersed in distilled water at 23 ± 1 °C and periodically removed, wiped dried, weighed, and immediately returned to the water bath. The amount of water absorbed was calculated from the following formula:

$$Wu = \frac{(M_t - M_0)}{M_0} \times 100\% \quad (2)$$

where Wu is water uptake, while M_t and M_o are the masses of the specimen at time t and initial condition when t was 0, respectively.

The morphology of tensile test fractures in corresponding samples was investigated by scanning electron microscope (SEM), CamScan MX 2600FE, Oxford Instruments, UK, at 20 kV accelerating voltage.

3. Results and discussion

3.1. Fibers aspect ratio

The fiber aspect ratio always remained a decisive factor for NF reinforced composites. The length distribution and fiber aspect ratio of as-received HF, alkali-treated HF, and collected HF on 20, 40, and 80 mesh sieves are presented in Fig. 2. Moreover, the summary of the average length, diameter, and aspect ratio of as-received HF, after alkali treatment, and after shredding and sieving are listed in Table 1.

A very minute difference was observed in the aspect ratio of as-received and alkali treated HF, due to the slight decrease in the average diameter. The microfibrils were also observed after the alkali treatment of HF, this suggests the removal of binder such as hemicelluloses and lignin from HF (Mohanty et al., 2004; Das and Chakraborty, 2008; Patel et al., 2010). A significant difference in the HF aspect ratio was observed due to the change in average fiber length. However, the average fiber diameter of the fiber was not much changed after shredding. The good difference was observed in the diameter of the fiber collected at different sieves; this induced the impact on aspect ratio, while a very minute change was observed in the average length of fiber collected on all sieves.

3.2. Tensile properties

The tensile properties of NF reinforced composites are mainly dependent on fiber-resin interfacial adhesion, fiber content, fiber length, and fiber properties. As we discussed earlier in fiber aspect ratio analysis that notable changes were observed in the HF diameter after alkalization, shredding, and ball milling process. This suggests a major role will be played by the HF diameter in the estimation of tensile properties of HF reinforced composites.

Tensile properties of HF composites with different diameter and composition are depicted in Fig. 3 and complete values are tabulated in Table 2. Aspect ratio and vol.% of HF loading in composites enhanced tensile strength and Young's modulus (E) of HF reinforced composites. The fiber fibrillation increases the effective fiber surface area available for fiber-resin adhesion, resulting in an enhanced mechanical