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Mechanical property and hydrothermal aging of injection molded jute/polypropylene composites

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Abstract This study discussed the effects of jute fiber content and hot water immersion on the tensile properties of jute fiber reinforced polypropylene (PP) composites. The jute/PP composite with different fiber contents was molded by injection molding by dry-blending of jute/PP and neat PP pellets in various mix ratios. Firstly, the quasi-static tensile test was performed. Then the specimens were aged in hot distilled water at 80 °C. After the fixed periods of aging, the changed weight and the tensile properties were investigated. It is found that with the increase of the jute fiber content, the tensile modulus is increased lineally. However, referring to the tensile strength, it is increased firstly followed by a decreased when the jute weight percent is over 30. Additionally, it is found that the weight gain by water absorption was significantly affected by the fiber content. The specimens with the jute fiber content of or over 30 wt% absorbed water easily and significant material loss by aging was also occurred. The tensile strength after aging decreased remarkably in these specimens with the jute fiber content of or over 30 wt%, and all the jute/PP composites showed the lower strength than neat PP after the aging of 1000 h. It is considered that the hydrophilic property of natural fiber decreases the resistance of the composite in humidity environment.

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Introduction

Until now fiber reinforced composites are commonly used in the aerospace, automotive, marine, and construction industries. The fibers are usually fiberglass, carbon, or aramid fiber. However, those petro-based fibers cost a lot of energy in manufacturing process and are difficult to be recycled. With the increasing high concerns on the sustainability issues represented by the environment and resource protection, therefore, recently, environmentally friendly material such as natural, green, recycle, or biodegradable material has been attractive. Representatively, lingo-cellulosic fibers possess low density and sometimes equivalent stiffness to glass fibers. Therefore, those natural fibers have been a growing interest as reinforcing fiber in polymeric composites [1–9].

According to the previous studies, it is found that although glass fibers do not absorb much water, the mechanical property of the glass fiber reinforced composite after hydrothermal aging still decreased because of the moisture absorbed by the polymer matrices or the glass/matrix interface [10–12]. That is to say, hydrothermal aging is an important evaluation method to investigate the long-term life of the composites. In particularly, unlike glass fiber, natural fibers are able to absorb water themselves. Therefore, because of the hygroscopic nature of natural fibers, it became primary importance to clarify the long-term properties including hot water aging of natural fiber reinforced composites.

Before now, Bledzki et al. [13] have studied the wood-filled thermoplastic composites. It is found that after storing the samples for 28 days in water, the moisture absorption of wood-filled plastics ranged from 3 to 9 wt% and the mechanical properties of the wood-filled thermoplastic samples stored in water decreased by 8 to 35%. The

environmental effect on bamboo-glass/polypropylene hybrid composites has been investigated by Thwe and Liao [14]. The specimens were prepared by compressing the mixture of bamboo fiber, glass fiber, and PP in a stainless steel mold. Similarly, the results show that the tensile and flexural strengths of the bamboo fiber reinforced composites reduce 7.5–32% with sorption time and temperature from 25, 50, and 75 °C, although the environmental degradation process could be delayed by adding a small amount of glass fiber. The water diffusion behavior of hybrid natural fiber including wood flour, rice hulls, newsprint fibers, and kenaf fibers reinforced PP were studied by Tajvidi et al. [15]. Results indicated a significant difference among different natural fibers, with kenaf fibers and new-print fibers exhibiting the highest and wood flour and rice hulls the lowest water absorption values, respectively. Additionally, water diffusion coefficients of the composites were found to be about three orders of magnitude higher than that of pure PP. And other researches were reported that water uptake and water diffusion coefficient also increase with fiber content [16–19].

Recently, injection molded composites are applied into the practice more and more widely because it is of high-quality products with low cost. Therefore, at this study, jute/polypropylene composites (Jute/PP) with different fiber contents were molded by injection moldings to investigate the effect of hot water immersion on the tensile property and acoustic emission behavior of the composites. The molded specimens were aged in hydrothermal environment. In particularly, after the aging, both the weight loss by water solution and the weight change by water absorption were under consideration. Based on the test results and observation on the fracture surface, the relationship between weight change and reduction of tensile properties were discussed.

Experimental

Materials

Two kinds of pellets were used to molded dumbbell-shaped testing jute composites specimens by injection molding. They were jute/polypropylene (jute/PP) pellet (Calp Corp) and neat polypropylene (PP) pellet (Prime Polymer Co., Ltd) (here PP pellet is same type with the one used in the Jute/PP pellet). In this study, in particularly, the jute/PP pellet was fabricated by the long fiber-type pellet making machine with the use of the pultrusion technique developed by Kobelco Co. Ltd [20]. The pultrusion machine was able to twist the jute yarns and continuously fed through an extruder where the yarns were impregnated with a special high melt flow grade PP. The impregnated yarns were then cooled and cut to a desired length, in this case 12 mm, to produce long fiber pellets. To vary the jute content in the composite, high flow grade PP (J-3000GP) from Prime Polymer Co. Ltd. was dry-blended with the jute/PP pellets.

Table 1 Material composition

Specimen ID	Blend ratio (wt%)		Jute fiber (wt%)
	Jute/PP pellet	Neat PP pellet	
PP	0	100	0
J10	20	80	10
J20	40	60	20
J30	60	40	30
J40	80	20	40
J50	100	0	50

Those pellets were dried in an oven set at 80 °C for at least 3 h prior to being used for molding. The weight fraction of jute fiber in jute/PP pellet was 50. By dry-blending of jute/PP and neat PP pellet, five kinds of jute/PP dumbbell-shaped specimens with different fiber contents were prepared. Table 1 summarizes the details of the specimens, i.e., those with the jute fiber content of 10, 20, 30, 40, and 50 wt%. Referring to the injection molding process, dumbbell test pieces were fabricated by using a 30-ton TOYO PSS TI-30F6 injection-molding machine at a barrel temperature of between 190 and 200 °C.

Hydrothermal aging

In order to discuss the effect of water absorption on the weight changes and the mechanical properties, all the specimens were aged under hydrothermal environment. Before the aging, all the specimens were conditioned in an oven at 100 °C under vacuum condition for 24 h to remove the free water. (According the previous study on the water loss-dry time of jute/PP, it was found that about 0.56 and 0.85% water was evaporated from specimens J20 and J40, respectively, after 24 h in an oven at 100 °C under vacuum condition, and 0.57 and 0.88% after 48 h. Therefore, it is considered that it gives necessarily the true dry weight of the specimens.) After that the conditioned specimens were weighted at the first time and immersed into distilled water at 80 °C immediately for 3, 10, 30, 100, 300, 1000, and 3000 h. After the fixed periods of aging, the specimens were removed from the water, wiped off the surface water by a dry tissue paper and weighed at the second time. After that the specimens were dried at the oven again at 100 °C under the vacuum condition for 24 h and weighted at the third time for reference.

Weigh change measurement

In order to discuss the effect of hydrothermal aging, the weight change behaviors were evaluated. Before the aging, the weight of conditioned specimen, i.e., the dried weight of original specimens, W_o, was weighed. After the fixed periods of aging, the specimens were taken out of the water bath, wiped the surface water and weighted. Therefore, the weight of wetted specimen, W_w, could be obtained. Finally, the dried weight of wetted specimens after drying at the oven again, i.e., W_d, was measured. From W_o, W_w, and W_d the weight change parameters, M_g, and M_l, were evaluated by Eqs. 1 and 2

$$M_g = \frac{W_w - W_d}{W_o} \quad (1)$$

$$M_l = \frac{W_o - W_d}{W_o} \quad (2)$$

where M_g, i.e., moisture gain, is corresponded to the absorbed water and M_l corresponded to the weight loss by water-soluble matter.

Tensile test and acoustic emission measurement

In order to discuss the effect of hydrothermal aging on the mechanical properties, the tensile tests of un-aging and the wet specimens after aging and drying were performed. Three testing specimens were prepared for each type-aging condition. The tensile tests were carried out at a constant cross-head speed of 2 mm/min under room temperature on an Instron type universal testing machine (Autograph AG-50KNI, Shimadzu Corp., Japan). The gauge length is 115 mm. During the tensile tests acoustic emission (AE) was monitored simultaneously by 7600 series AE instrumentation (NF Corp., Japan). In AE measurement the transducer with the resonant frequency of 140 kHz was attached onto the central point of the specimen, and AE counts were monitored. The fixed gain was 40 dB and the threshold was set at 100 mV to avoid the inclusion of noise.

Experimental results and discussion

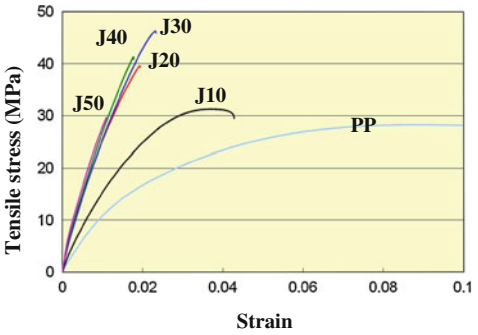
Tensile properties of virgin specimens

Figure 1a shows the tensile stress–strain curves of virgin specimens. The stress–strain relationship changed from ductile to brittle behavior with the increasing of the jute fiber content. However, stress–strain relationships were almost the same in the specimens with the jute fiber content of more than 20 wt%. Figure 1b shows the tensile strengths and moduli of each specimen. The tensile modulus increased with the increasing of the jute fiber content. Jute fibers show the reinforced effect to the PP resin in the composite structure. On the other hand, the tensile strength increased firstly with the increase of the jute fiber content. However, the strength of the specimens with jute fiber content over 30 wt% decreased with the increase of the fiber content. The strength of J50 even show similar to that of the neat PP. The observation results on the fracture surface of J30 and J50 after tensile test are shown in Fig. 1c, d. It is found that the jute fiber became inhomogeneous in J50. It is considered that jute has a microfiloplume and rough surface led to the collected situation at some degree in high fiber content composite. Many jute fibers are found revolved by the resin flowing and oriented to transversal direction (Fig. 1d). The highly anisotropic fiber orientation is considered main reason which led to the decrease of strength in high fiber content composites.

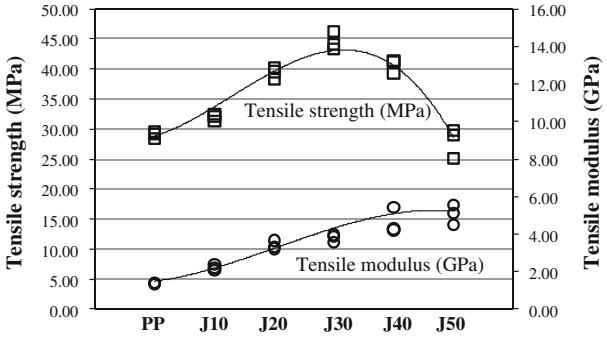
Figure 1e shows the tensile stress and cumulative AE count–time curves of J10, J30, and J50 specimens as samples. In those specimens, the stress–time relation was linear before the AE initiated. Afterward, it showed nonlinearity because of the generation of AE. After the AE initiation, the increasing slopes of AE counts of these three kinds of specimens were almost constant until final rupture. As summarized in Fig. 1f, cumulative AE count of J10 reached about 12,000. It is considered that J10 has a relative high resistant to the micro-fractures. However, cumulative AE count decreased remarkably with the increasing of the jute fiber content. In J40 and J50 specimens, the cumulative AE counts were very few. The composite became brittle with the increase of jute content. Brittle—there is few fractures before terminal broken would bring unexpected dangerous during the practical application. Regarding to the AE yield stress, i.e., the stress when the cumulative AE count reached 10, it increased with the increasing of the jute fiber content up to 40 wt%. However, it decreased in J50. In J50 the AE yield stress became lower and the cumulative AE count was a few. This might be derived from the complication fiber orientation.

Weight changes

In order to investigate the water diffusion in injection molded jute composites, the changes of the weight gain, M_g, and weight loss, M_l, against the square root of immersion time were investigated and illustrated in Figs. 2 and 3. In neat PP specimen the increase of M_g was a little. On the contrary, M_g of jute/PP specimens showed significant increase with the increasing immersion time. In the specimens with the fiber content below 20 wt% M_g increased gradually. On the other hand, for the specimens



(a)



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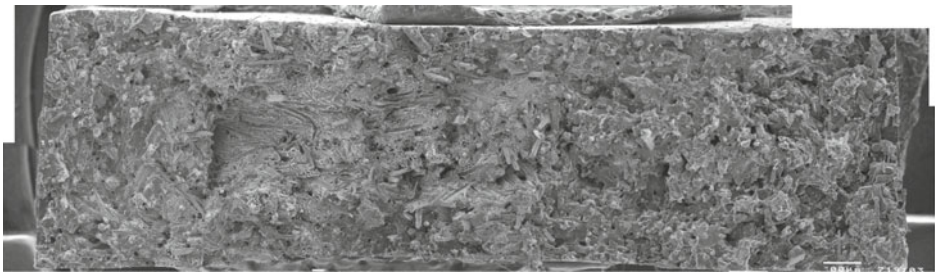

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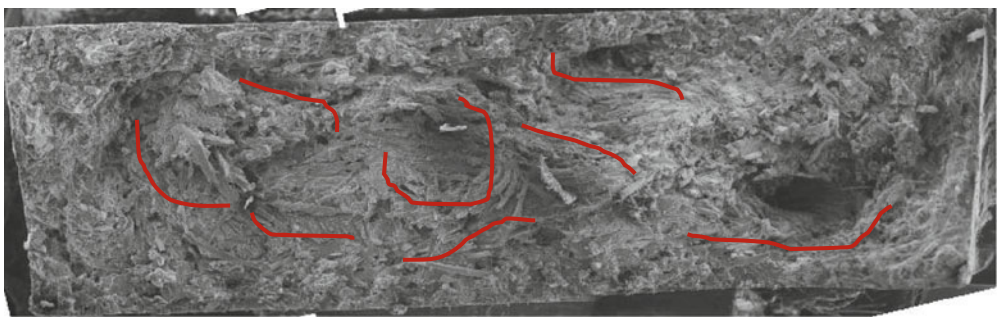

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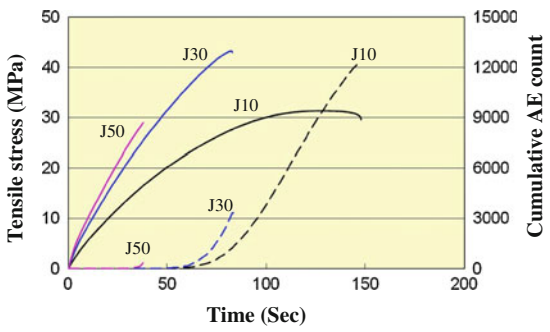
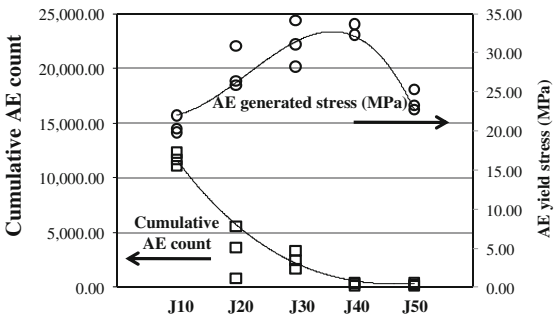


(e)



(f)

Fig. 1 Mechanical properties and fracture behavior of virgin injection molded jute composites. **a** Tensile stress–strain curves, **b** tensile strength and modulus, **c** SEM photograph on the fracture surface of J30 after tensile test, **d** SEM photograph on the fracture surface of J50 after tensile test illustrated the flowing directions of jute fibers which are collected at some degree, **e** tensile stress and AE count–time curves, and **f** cumulative AE count and AE yield stress

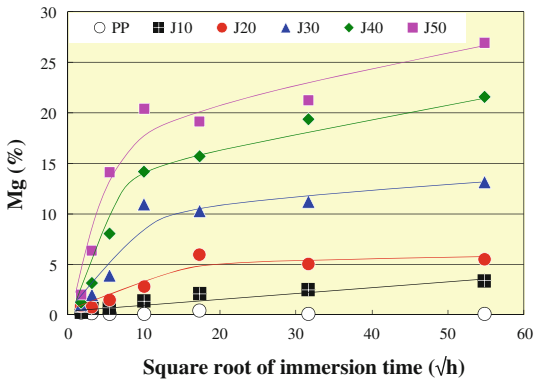


Fig. 2 Changes of the weight gain, M_g of immerged injection molded jute composites

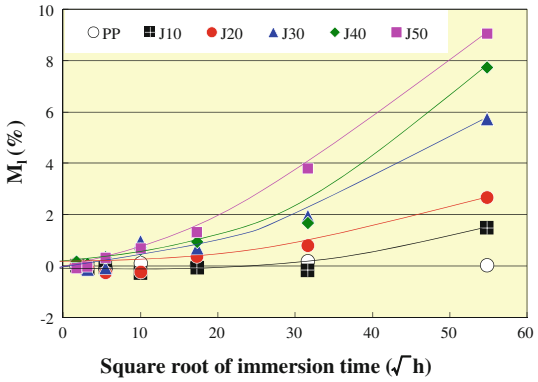


Fig. 3 Changes of the weight loss, M_l of immerged injection molded jute composites

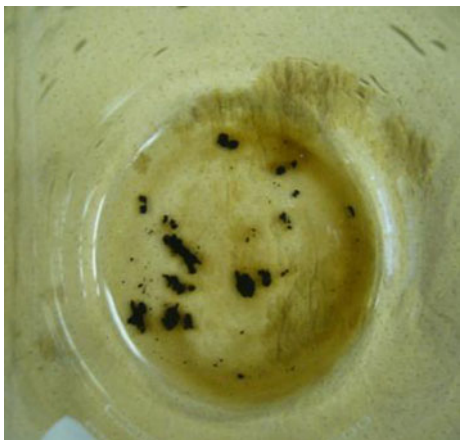


Fig. 4 Residue in the bottom of the container where jute composites have been immersed

with the fiber content above 30 wt%, M_g showed a rapid increase, and M_g at 1000 h reached more than 10%. Additionally, it has been observed that swelling was generated in those high jute content specimens, although the swelling could be recovered by keeping in air for a moment. According to the previous study, it is found that in case of the glass fiber/PP composite M_g reached less than 1%, and M_g reached saturation after 24 h immergence [21]. From these results it is easily concluded that high M_g was brought by the water absorption of jute fiber in jute/PP composites similar to the other researches [16–19].

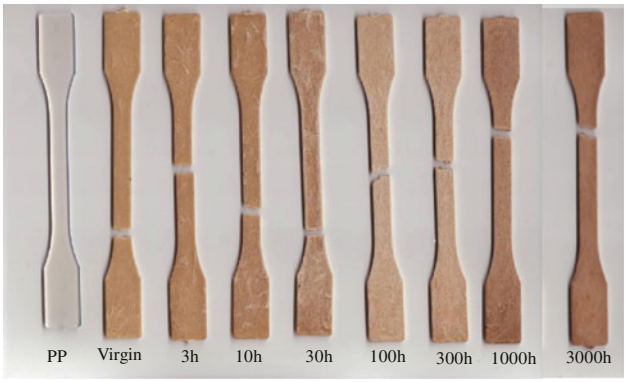


Fig. 5 The color change of the dried J30 as an example to illustrate the effect of hydrothermal aging

Referring to the weight loss, M_l, due to the water-soluble matter, unlike neat PP, those specimens with 20 wt% or more of jute fiber increased markedly. Neat PP did not show any material loss, and therefore the material loss of jute/PP composites was derived by the loss of jute fiber or semi-cellulose materials. The residue as shown in Fig. 4 was found in the bottom of the container where jute composites have been immerged.

Changes of tensile properties by aging

The pictures of the dried J30 specimens after tensile tests were compared in Fig. 5 as an example to illustrate the color change of injection molded jute/PP composite under the effects of hot water aging. Those hot water degraded specimens are all wet specimens, i.e., taken out of hot water and warped the surface water. It is clear that the color of J30 became white firstly before 300 h aging but return dark again after 1000 h aging. It is considered that during the first stage of aging, the delaminations between jute fiber and PP resin were generated and led to a light color. After that the further water absorption into the jute fiber recovered the original color then the specimens became dark again.

Figure 6 shows the changes of the tensile moduli by aging. In neat PP modulus was almost constant even after the aging. In J10, J20, and J30, moduli decreased gradually with increasing the aging time. On the other hand, the moduli of J40 and J50 decreased remarkably before 30 h aging. The moduli of virgin specimens depended on the jute fiber content, however, the moduli after 1000 h aging became lower with increasing jute fiber content. The same

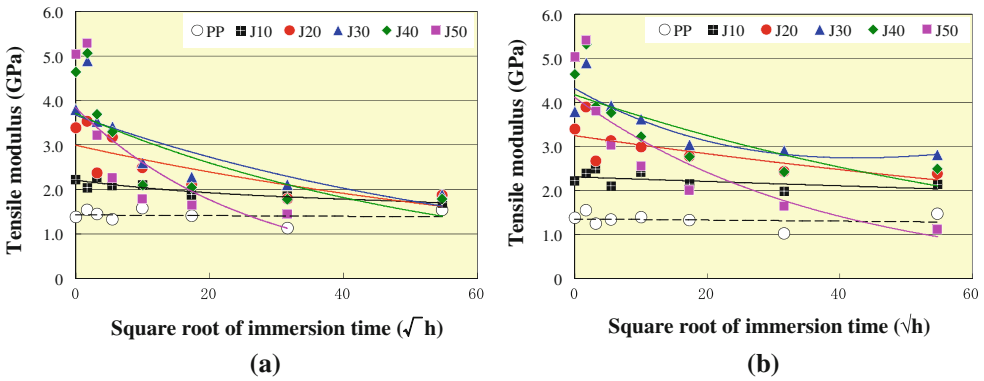


Fig. 6 Changes of the tensile moduli of the specimens after hydrothermal aging. **a** Wet specimens and **b** dried specimens

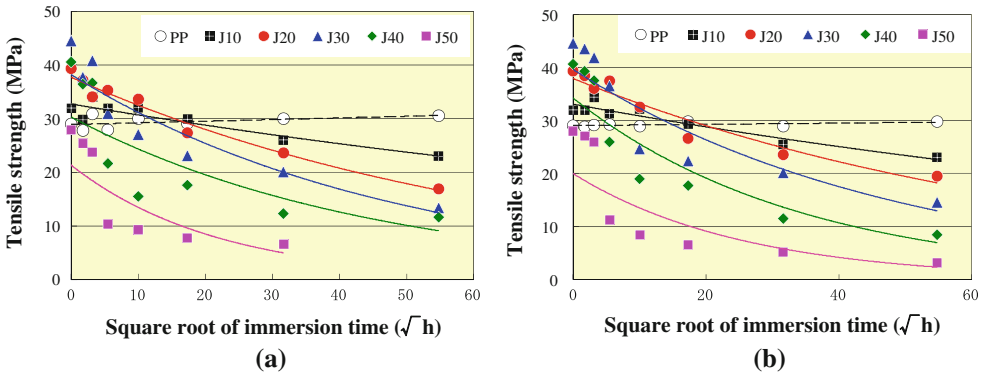


Fig. 7 Changes of the tensile strength of the specimens after hydrothermal aging. **a** Wet and **b** dried specimens

tendency could be found for the dried specimen after aging (Fig. 6b). On the other hand, although, the moduli after 1000 h again were still higher than neat PP, the wet specimens after 3000 h aging have the similar modulus to the neat PP, in particularly, the wet specimen J50 after 3000 h aging were broken during the installing in the test machine. (Therefore, the mechanical property of wetted J50 after 3000 h aging did not obtain.) The specimens after aging jute fiber absorbed a lot of water through the interface. It is though that before 300 h aging, the interface between jute and PP polymer is becoming larger and larger which lead to a white color change from the outside view on the specimens. And after that, the jute fibers absorb the water and then the color of the specimens changed to original dark.

Figure 7 shows the changes of the tensile strength by aging. Similar to the tensile modulus, the strength of neat PP did not change even after aging. However, in jute/PP specimens the strength showed lower value than the neat PP after long aging. For such jute/PP specimens with higher jute fiber weight content, after they are immerged in the water, their strength became lower than neat PP in a shorter immerge time. After 1000 h aging the strength became lower with increasing jute fiber content. Such tendency was the same in case of the dried specimen after aging. From these results, it is thought that the strength

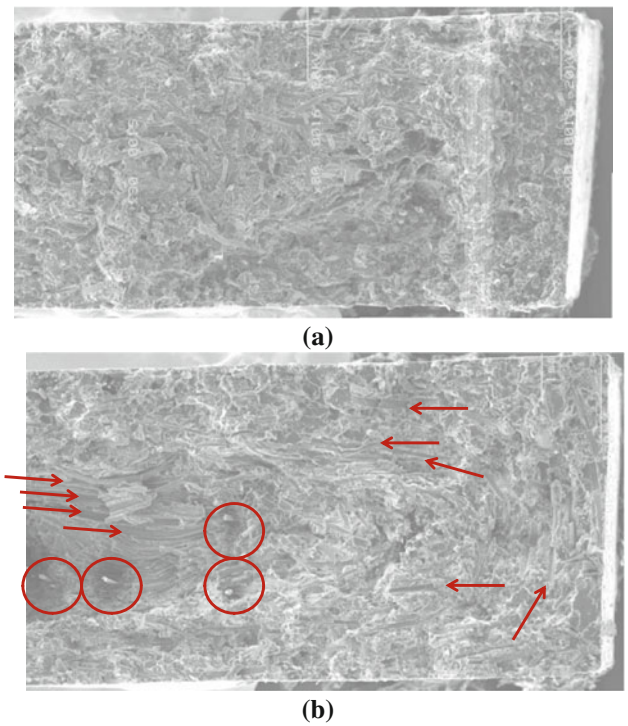


Fig. 8 SEM photographs of fracture surface of J30, illustrating many debondings between jute fiber and PP polymer were generated after the hydrothermal aging. Fracture surface of J30 **a** without hot water immersion and **b** after 3000 h hot water immersion

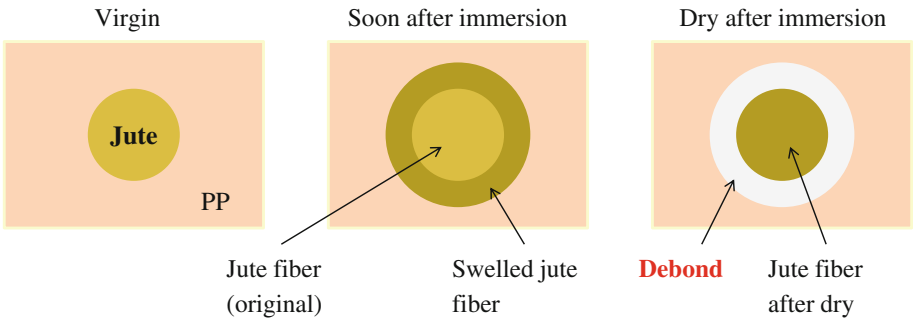


Fig. 9 Interfacial debonding mechanic of Jute/PP in hot water

reduction was not affected by the absorbed water itself. The strength of jute/PP after long aging reached much lower than the neat PP, and this result suggested jute fiber did not play as reinforcement of PP. Jute fiber did not carry the applied load, and it might exist as making defect in PP matrix.

The SEM observation results of J30 without immersion and after 3000 h aging are illustrated in Fig. 8 as an example to explain the effect of hot water. As compared to the original specimen (a), there are many tracks after fiber delaminated from the resin and big skip between the fiber and resin from the observation on the fracture surface of tested specimen after 3000 h immersion (b). The fracture initiated from the interface between jute fiber and PP, and it brought quite lower strength after aging. Similarly, the modulus reduction is also considered to be caused by the degradation of the interfacial adhesion between jute fiber and PP polymer. As illustrated in Fig. 9, it is considered that the jute fibers absorb the water and swelled which lead to a damage on the interface area. After drying, fiber diameter recovered. However, the debonding between jute fiber and PP remained and led to the modulus and strength reduction. In a word, it is considered that the hydrophilic property of natural fiber decreases the resistance of the composite in humidity environment. Suitable interface treatment method should be developed to increase the bonding between natural fiber and matrix under the considerations on temperature, water and so on.

Conclusion

This study dealt with the effect of fiber content and hot water immersion on weight changes and tensile properties of injection molded jute/PP. It is found that with the increase of the jute fiber content, the tensile modulus is increased lineally. However, referring to the tensile strength, it is increased firstly but decreased late after the jute is over 30 wt%. In jute/PP composites, significant increase of weight gain generated due to hydrophilic property of natural vegetable fiber. The tensile modulus and strength decreased significantly by aging in jute/PP with higher fiber content. Seriously, after 300 h hot water immersion, the strength even showed low value than that of neat PP. It is found that the main reason is the degradation of the interfacial adhesion between jute fiber and PP based on the SEM observation on the fracture surface of the tested specimen after hydrothermal aging. It is considered that the hydrophilic property of natural fiber decreases the resistance of the composite in humidity environment.

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