EFFECT OF SURFACE MODIFICATION ON FLEXURAL PROPERTIES OF JUTE FIBER GREEN COMPOSITES

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1 Introduction

Green composites, consisting of an association of a biodegradable polymer matrix and a natural fiber as reinforcement have become popular due to both increasing social and economic pressure to conserve petroleum resource. Green composites offer environmental benefits such as biodegradability, less greenhouse gas emissions, and renewability of the base material.

Most of the natural fibers and reinforcement used for composites are hydrophilic nature, whereas synthetic polymers are hydrophobic. Poor adhesion between the natural fibers and polymer matrix often prevents the possibility of natural fibers to act as fillers, resulting in poor dispersion, inadequate reinforcement, and low mechanical properties [1-3]. Therefore, natural fibers require the addition of coupling agents or the chemical modification for industrial application in composites. Many research efforts have been made to green composites with high strength, modulus, durability estimated by tensile, flexural and fatigue test, with extensive attention being focus on the improvement of fiber-matrix interface compatibility, such as the use of coupling agent and surface modifications [4-6].

Creep is one of the principal properties which serve to be good reference when developing and using composite materials. It was well known that most materials respond differently depending on the time required to complete the mechanical test [7]. The creep behavior of composite materials is particularly important and a critical issue for many modern engineering application such as aerospace, biomedical and automotive industries

In spite of many reports on green composites, the analysis of creep behavior is still rare. Vazquez et al. [8] have performed flexural-creep test on bagasse fiber-polypropylene composites. They treated the fiber with an alkaline solution and the treated fiber improved the creep behavior of the composites due to the higher adhesion between the fiber and the matrix. B.A. Acha and coworkers [9] reported the effect of the interfacial adhesion on creep behavior of PP-jute composites. Maleic anhydride and a commercial lignin were added to the polypropylene in order to improve the compatibility between the jute fiber and the matrix.

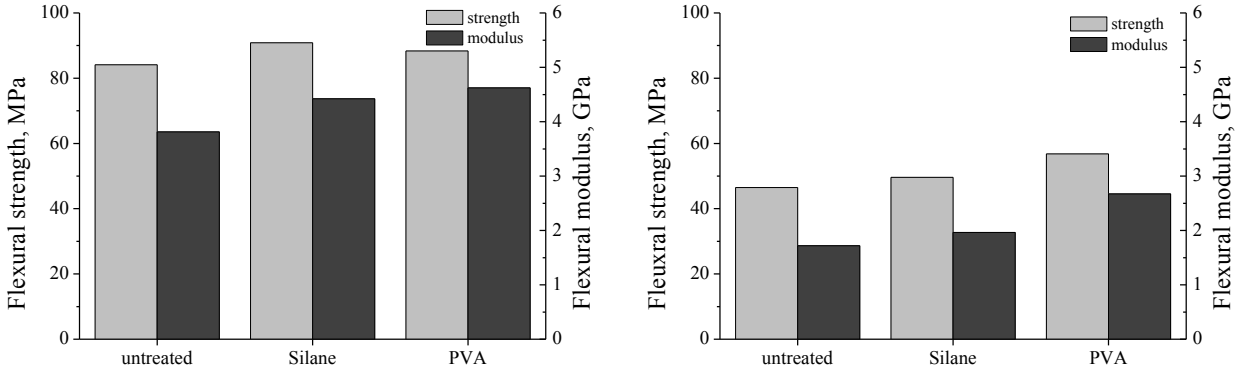
In this work, the interfacial adhesion between jute and poly(lactic acid) was modified in two ways: by the addition of silane coupling agent and by treating chemically the jute fibers with a commercial PVA (polyvinyl alcohol) prior to composite preparation. The effect of the surface modification on flexural behavior of jute fiber green composites was investigated.

2 Experimental details

2.1 Materials

The woven fabric jute fiber was supplied by Kawashima Selkon (Kyoto, Japan). According to supplier, Young's modulus and the density for jute fiber were 26 GPa and 1.44 (g/cm^3). Poly(lactic acid) sheet (Terramac SS300) was supplied by Unitika Ltd. (Tokyo, Japan). Polypropylene sheet (Kobepoly sheet PP-N-AN) was supplied by Shin-Kobe Electic Machinery Co. Ltd. (Kobe, Japan). The interfacial adhesion between fibers and matrix was modified using a silane coupling agent Z-6040 (Glycidoxypropyl-trimethoxysilane, $CH_2(O)CHC\text{-}H_2OC_3H_6Si(OCH_3)_3$) for PLA and Z-6300 (Vinyltrimethoxysilane, $C_2H_3Si(OCH_3)_3$) for PP supplied by Dow Corning Toray Co. Ltd. and poly(vinyl alcohol) (PVA), supplied by Wako pure chemical industries, Ltd.

2.2 Surface modification and composites fabrication



(a) PLA (b) PP

Fig. 1 Flexural properties of surface modified jute fiber composites

To obtain an improved fiber matrix adhesion, the fibers were modified silane coupling agent and PVA treatment. Silane coupling agent treatment was carried out in distilled water with 5 % silane content for 1hr at room temperature, and dried in the oven for 24hrs. PVA treatment was performed same process with silane coupling agent.

Prior to composite fabrication, the woven jute fiber was completely dried at 50℃ in the oven. The fiber content of jute fiber green composites was 35 wt% and all green composites were made by a compression molding technique with vacuum. The woven jute fibers and PLA sheets was placed in an aluminum matched-die mold. The molding temperature, pressure and holding time were 190℃, 10 MPa and 10 min, respectively. And then the mold was cooled down to room temperature by water.

2.3 Flexural properties

The flexural tests were performed according to JIS K7017 standard using Autograph (AG-IS, Shimadzu corp.), and the crosshead speed was 2 mm/min.

The flexural creep tests were performed with handmade equipment using dry oven according to JIS K 7017. Creep tests were carried out with a constant load level of 40 N, temperatures were 25, 40, 50 and 60 ℃.

2.4 SEM study

The surface modified jute fiber and fracture surface of flexural tested samples were examined by scanning electron microscopy (S-4000 FE-SEM, Hitachi). Prior to SEM observation, all samples were sputter coated with a thin layer of gold to avoid electrical charging.

3 Results and discussion

3.1 Flexural properties

Figure 1 shows the relationship between the flexural properties with PVA and silane treated jute/PLA and jute/PP composites. The flexural properties, especially in modulus, of the surface modified jute fiber reinforced PLA and PP composites were compared with these of the untreated jute fiber. The flexural modulus of unmodified jute/PLA and jute/PP composites were increased about 20.7% from 3.81 GPa to 4.6 GPa and 56.1% from 1.7 GPa to 2.7 GPa by PVA treatment. The flexural modulus

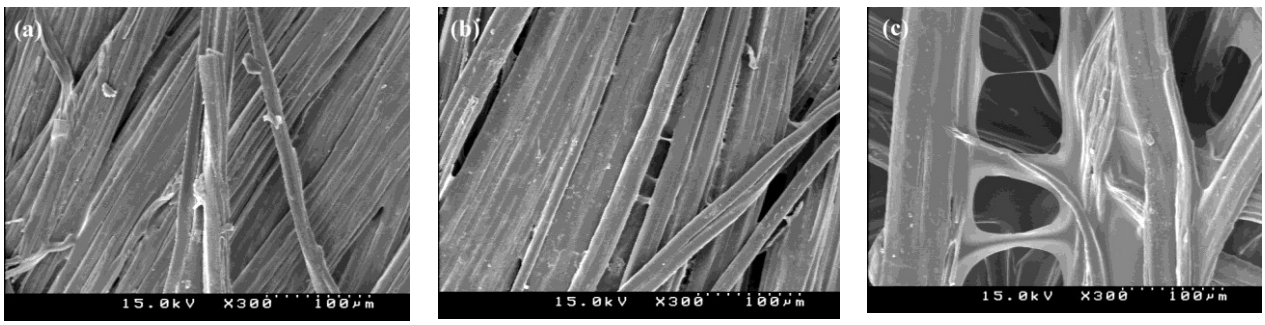


Fig. 2 SEM image of surface modified fiber: (a) untreated, (b) silane, (c) PVA

of untreated PLA and PP composites was improved after silane treatment, but the improvement rate of modulus was lower than PVA treatment.

PVA is suited for blending with natural polymers and is considered to be biodegradable. Because of this benefit, PVA has been extensively applied to improve mechanical properties in natural fiber composites [10-13]. Q. Liu et al. [13] reported the effect of PEG and PVA treatment as compatibilising agents to increase interfacial bonding between the flax fibers and the epoxy resin, PVA treatment lead to the increase of tensile modulus by the reduction in the cross-sectional area of fiber and the increase in the extent of fiber separation.

These results can be found in this study as shown in fig. 2. The diameter of surface treated fiber by silane and PVA was reduced compared with untreated fiber. In addition, the coated surface of jute fiber and the connection between fiber and fiber by PLA treatment were observed in this study. It is can be explain that the coated surfaces prevent the fiber damage from the fabrication processing and the connection of fibers lead to enhanced interfacial adhesion between the fibers and the matrix.

3.2 Creep behavior

The creep behavior of the composites are a combined effect of the morphological change, such as orientation and crystallization, molecular weight and transition phenomena, such as glass transition and secondary transition [14].

Figure 3 shows the short-term creep behavior of jute fiber composites with PVA and silane treated jute/PLA and jute/PP composites at room temperature. With surface modification, the creep deflection were approximately 40-52% lower than that without surface modification, the coupling agent (Z-6040) was more effective than PVA treatment in jute/PLA composites. The creep deflection of jute/PP composites were approximately 33-60% lower than that without surface modification, the PVA treatment was more effective than coupling agent (Z-6300). These could be utilized in chemical interaction between jute fiber and PLA at the interface leading to better bonding. This behavior can be directly related with the interfacial adhesion. As compatibility of hydrophobic polymer and hydrophilic cellulose fiber by surface modification is improved, the fiber-matrix adhesion is increased. It leads to the enhancement of creep behavior.

Figure 4 shows the creep behavior of unmodified and surface modified jute fiber reinforced

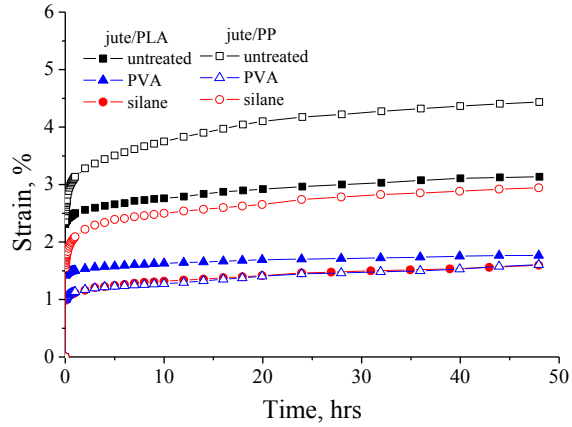
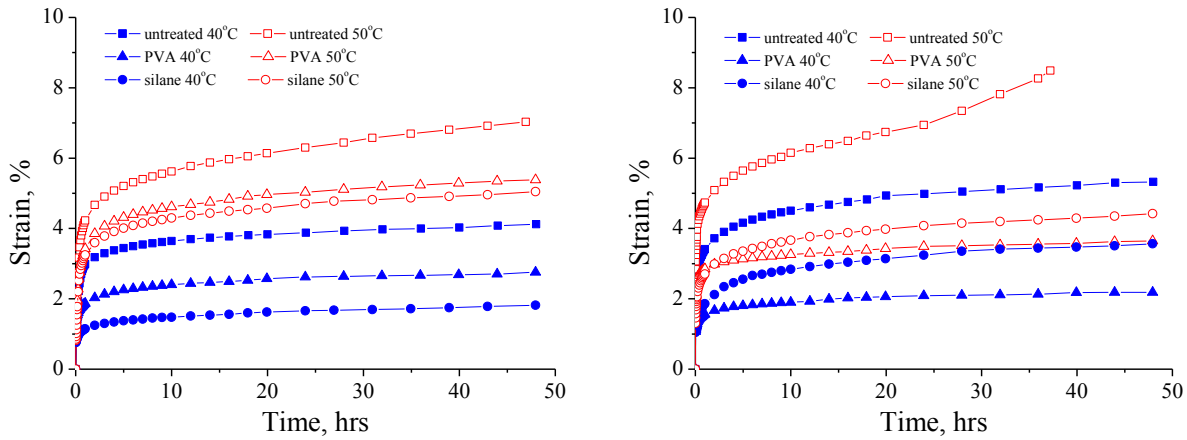


Fig. 3 Creep behavior of surface modified jute fiber composites



(a) PLA (b) PP

Fig. 4 Effect of temperature on creep behavior of surface modified jute fiber composites

composites at 40 and 50℃. As expected, as the temperature increases, the deformation of composites increases. Nevertheless, the use of the silane coupling agent and PVA modification were noticeably reduced the creep strain at the same temperature. The deformation of surface modified jute fiber/PLA composite at 50℃ was higher than that of unmodified fiber composites at 40℃. In spite of higher deformation at 50℃, the jute/PLA composites was not failed.

On the other hand, the deformation of the surface modified jute/PP composites was lower than that of jute/pp composite. Also, the deformation of the surface modified jute/PP composites at 50℃ was lower compared with unmodified fiber composites at 40℃. And the failure of unmodified jute/pp composites was observed at 50℃. In spite of temperature increasing, the reason that the deformation of PVA modified fiber composites is lower is due to enhanced compatibility and thermal stability between the surface modified fiber and the matrix. Q. Liu et al. [13] and L.E. Millon et al. [15] revealed that the PVA was increased the number of hydrogen bonding between fiber and matrix, it leads to high mechanical and thermal properties for composites. In this case, it can be explained that the coated fiber by PVA increase physical and chemical cross-linking between the jute fiber and polypropylene, it occurred the enhanced compatibility and thermal stability between the fiber matrix. It should lead to reduce the creep deformation in spite of temperature increasing.

Figure 5 shows the creep behavior of jute/PP composites at 60℃. The failure of unmodified and silane modified jute fiber/PP composites were observed, but the PVA modified jute fiber/PP composites was not failed. In case of jute/PLA composites, the specimens were failed in all conditions within 30 minutes.

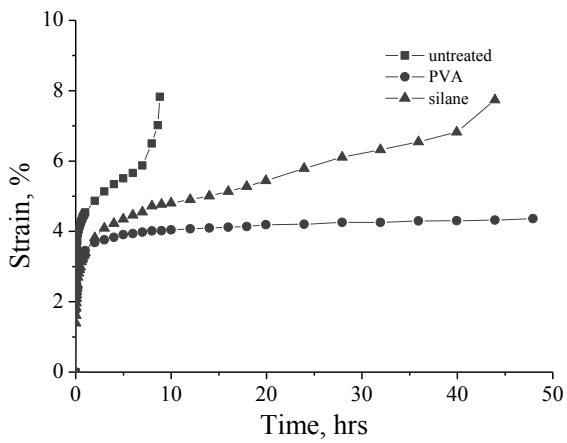


Fig. 5 Creep behavior of jute/PP composites at 60℃

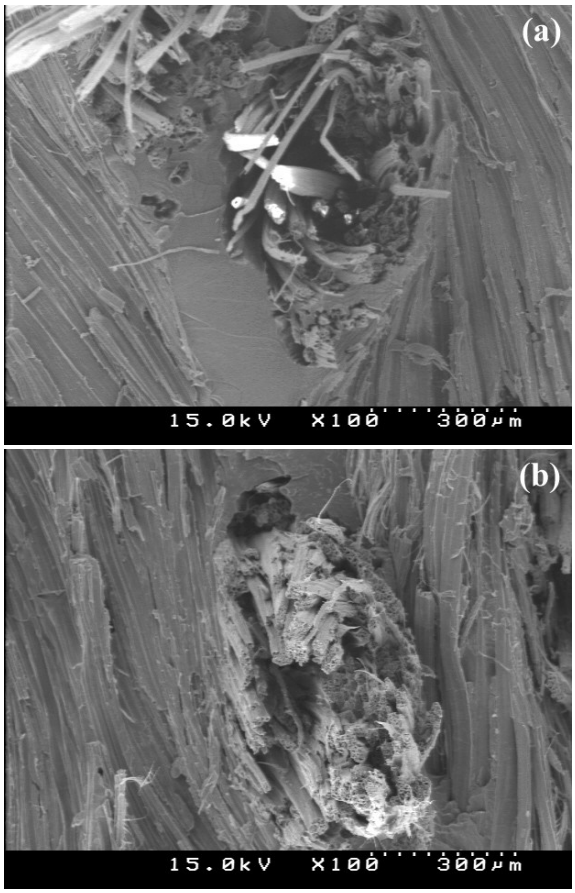


Fig. 6 Flexural fracture surface of PVA modified jute/PLA composites

3.3 SEM study

Flexural fracture surface of unmodified and surface modified jute/PLA composites were displayed in fig. 6. The modified jute/PLA composites shows the better retention of resin on failed fiber, while the unmodified fiber shows uncoated surface and was released the weaving. Thus, the indication of better fiber-matrix bonding in the case of PVA modified fiber is further supported by SEM study.

4 Conclusions

In this study, the effect of the surface modification on creep behavior of jute fiber green composites was investigated. The use of silane coupling agent and PVA were effective in improving the compatibility

of jute fiber with the matrix, and resulted in further enhancements of mechanical properties of jute fiber green composites. The enhanced interfacial adhesion between the fiber and the matrix by PVA treatment increased the flexural strength and modulus of the jute fiber composites, due to the coated surface and the reduction in cross-sectional area of the fiber. After surface modification by PLA and coupling agent, the improved compatibility between the fiber and the matrix was decreased with the creep deformation of jute fiber composites at room temperature. The surface modified jute fiber composites show the lower creep deformation than unmodified fiber composites due to the enhanced thermal stability by the increasing of physical and chemical cross-linking between the fiber and the matrix.

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