Mechanical performance of bacterial cellulose nanofibre-reinforced epoxy composites

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Abstract

This paper reports the preparation and characterization of a new type of natural fibre nanocomposite, which is composed of bacterial cellulose nanofibre and thermoset epoxy resin. Before combining with epoxy resin, the bacterial cellulose nanofibre was prepared by a freeze-drying process. The effect of nanofibre loading on the mechanical properties of the polymer composites has been investigated. The tensile strength of the composites increased with increasing fibre content, and reached the maximum tensile strength value of about 74 MPa, reaching almost twice the strength of the neat resin. This strength value was quite low as compared with a theoretical estimation. This discrepancy seemed to be derived from weak interfacial adhesion.
Keywords: bacterial cellulose fibre, composites, epoxy resin, strength, modulus, fracture, debonding.

1 Introduction

In recent years, much attention has been focused on plastic waste problems and resource depletion in a global scale [1]. To build a sustainable society, research and development of natural fibre reinforced polymer composites have been carried out over a worldwide scale [1]. Some kinds of natural fibre composites exhibit fully-biodegradable nature, and therefore they can be disposed off easily and at the same time, there is no additional green house gas emissions even after burning them (namely carbon neutrality) [2, 3].

Most of conventional studies on natural fibre composites have been made by using macroscopic natural fibre bundle or single natural fibre (e.g. pulp) as a reinforcement [4–10]. In the case of macroscopic natural fibres, they have often

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suffered from various kinds of damage introduced during manufacturing, resulting in lower mechanical performance with wide scattering. On the contrary, in the case of nanoscale fibre such as cellulose microfibrils, which are extracted from pulp by a high pressure homogenization, it is believed that they have relatively high performance. Therefore the cellulose nanofibres have attracted a great deal of researchers' attention [11–14].

Nakagaito and Yano fabricated high-strength micro-fibrillated cellulose (MFC) fibre-reinforced phenolic composites having tensile strength of 370 MPa and Young's modulus of 19 GPa [11]. Fully biodegradable cellulose nanofibre-reinforced starch-based composites were also developed, and reported that the mechanical properties of the composites were affected by the processing conditions [12]. Omrani *et al.* fabricated cellulose nanofibre/epoxy composites having a maximum fibre content of 5wt. %, however they did not carry out tensile tests, but only dynamic mechanical thermal analysis [13].

The purpose of this paper is to elucidate the applicability of the bacterial cellulose nano-fibre as a reinforcing phase in polymer composites. Bacterial cellulose nanofibre/epoxy composites were fabricated and their mechanical properties were evaluated by tensile tests. In conclusion, it can be seen that the bacterial cellulose nanofibre can be used as reinforcing phase in a natural fibre composite system.

2 Experimental methods

2.1 Materials

Bacterial cellulose pellicle was purchased from Fujicco Co. Ltd., Japan (Fig. 1). The pellicle in as-received condition was soaked in acetic acid solution. First the pellicle was washed with running water to remove acid (i.e. neutralize). The pellicle was cut into 100x45x15 mm, then frozen at -15°C in a home-use freezer.

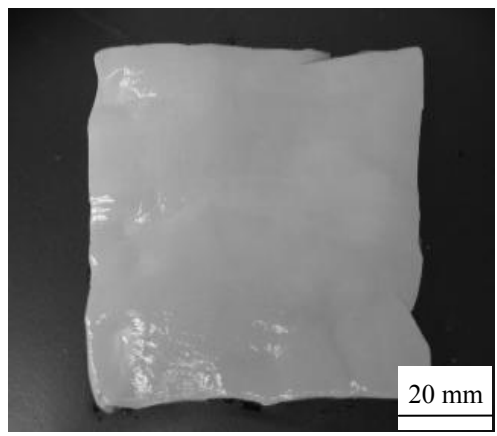

Figure 1: Photograph of bacterial cellulose pellicle.

This frozen bacterial cellulose pellicle was freeze-dried for four days by using a laboratory freeze-dryer (FDU-1200, Advantec Toyo Kaisha, Ltd., Japan).

Figure 2 shows a SEM photomicrograph of the freeze-dried bacterial cellulose nanofibre. It can be seen that the bacterial cellulose fibre has nano-scale diameter and web-like complicated microstructure as indicated elsewhere [11, 12]. Additionally we can see that each fibre is slightly curved (namely not straight) and branched at many points, and that its diameter varies from 50 nm to 200 nm. Low viscosity bisphenol-A epoxy resin (Low viscosity epoxy resin #7062, Refine-Tech. Co., Japan) was used as a matrix polymer.

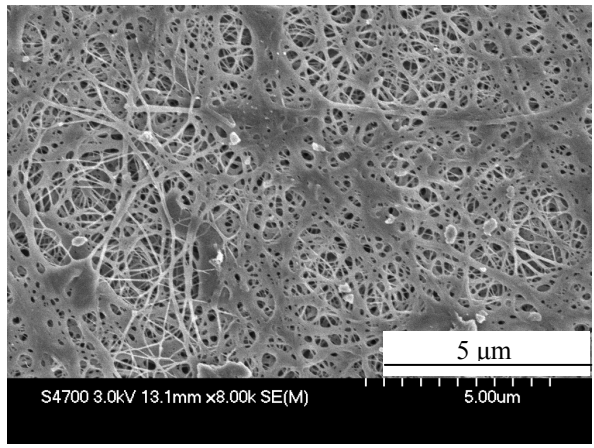

Figure 2: SEM photomicrograph of freeze-dried bacterial cellulose.

2.2 Sample preparation method

The freeze-dried pellicle was cut into a rectangle bar of 100x15x15 mm by using a wheel-saw cutter. The bar was pressed in a metallic mould at various pressure and room temperature. This pressing process is needed to increase the fibre content of the resultant composite materials. The thickness of the samples ranged from 0.1 to 15 mm. The pressed sheet sample was immersed in epoxy resin in vacuum (about 0.01 MPa). After complete soaking of epoxy resin, the resin-impregnated sheet was took out, then placed in another metallic mould, and kept at 80°C for 12 min. After complete hardening of epoxy resin, the sample was post-cured at 80°C for 3 hours. Figure 3 indicates the macroscopic photos of the pressed freeze-dried bacterial cellulose sheets with different thicknesses and their composites with epoxy resin. The shape of the composite samples changes after resin-impregnation treatment. The changes in shape are small in the case of thinner sheet sample (i.e. 0.1 mm thickness). Finally the six surfaces of the sheet sample were ground with various sandpapers and then polished with fine alumina powders.

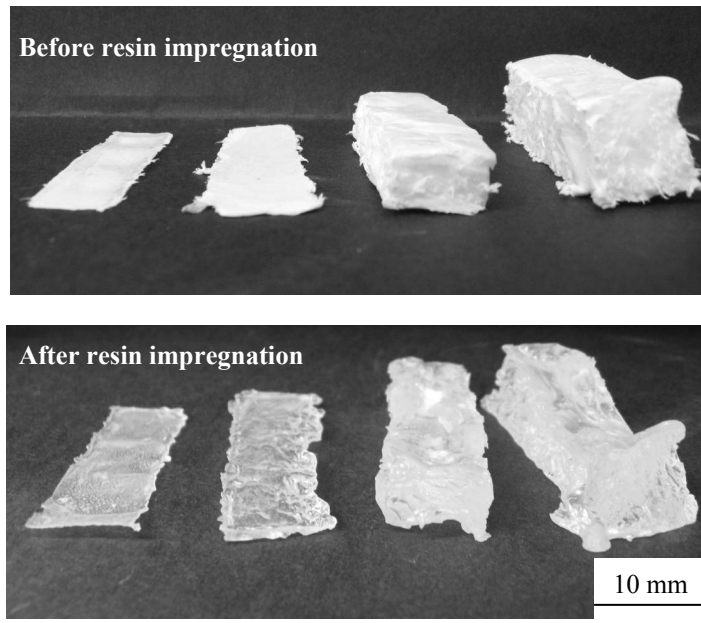

Figure 3: SEM photomicrograph of pressed freeze-dried bacterial cellulose sheets and composites; thickness =0.1, 1.0, 7.5, and 15 mm from the left).

2.3 Tensile tests

We carried out quasi-static static tensile tests in order to evaluate the mechanical properties and tensile fracture behaviour of the bacterial cellulose composites. The tensile tests were conducted using a universal testing machine (5567, Instron Co., U.S.A) at room temperature. The cross-head speed and gauge length were 1.0 mm/min and 30.0 mm, respectively.

2.4 Surface characterization

The fracture morphology of the samples after tensile was examined by a field emission-type scanning electron microscope (FE-SEM: S-4700, Hitachi Ltd., Japan). All samples were sputter-coated with platinum-palladium alloy using a sputter coater (E-1020, Hitachi Ltd., Japan) prior to SEM observation.

3 Experimental results and discussion

Figure 4 shows typical stress–strain curves for the epoxy composites reinforced with different bacterial cellulose fibre content. The stress–strain curve of neat resin is also shown in the same figure as a reference. It can be seen that the mechanical properties such as tensile strength and Young's modulus increase with increasing fibre content. The fracture strain of the composites ranges from

0.025 to 0.028. This strain value is almost similar to the fracture strain of macro-scale natural fibres such as hemp fibre and bamboo fibre. Therefore the fracture of the composites might be governed by the fracture of the bacterial cellulose fibres.

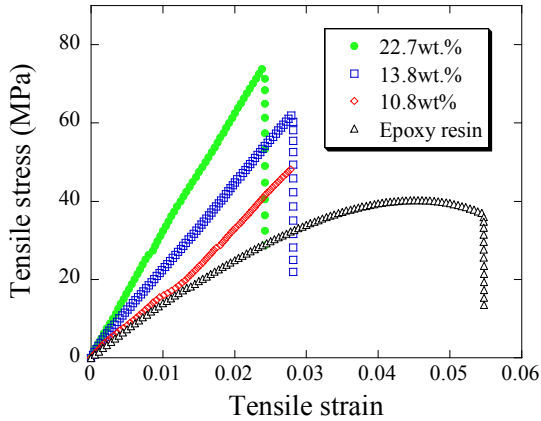

Figure 4: Typical stress-strain curves of the composites.

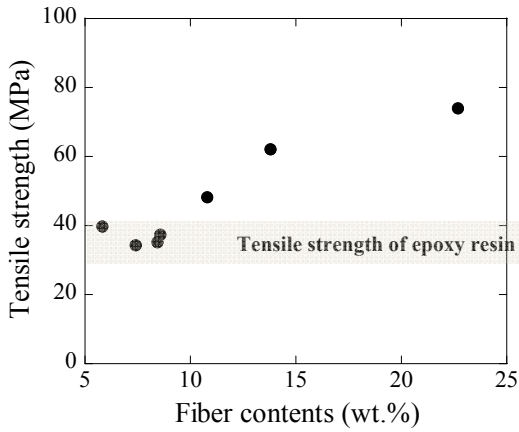

Figure 5: Relationship between tensile strength and fibre content.

The variation of tensile strength of the composites is indicated in Fig. 5. The tensile strength increases roughly linearly with fibre content, showing the maximum peak value of 74 MPa at 23 wt.%. This value is almost twice the strength of neat resin. A similar dependence was also reported in cellulose nanofibre/PLA composites, showing the maximum strength about 50wt. % [14]. It seems therefore that higher tensile strength might be obtained at higher fibre loading. The tensile strength of MFC was reported as 1700 MPa [15], therefore our result is quite low as compared with the theoretical estimation, which should be higher than 147 MPa.

The tensile fracture behaviour of neat resin and composites are depicted in Fig. 6. The fracture surface of epoxy resin is relatively flat and smooth (Fig. 6(a)), however that of composites with 11wt.% fibre is rather complicated and indicated that microscopic fracture occurs in the composites (Fig. 6(b)). Figure 6(c) shows enlarged image of fracture surface of the same composites,

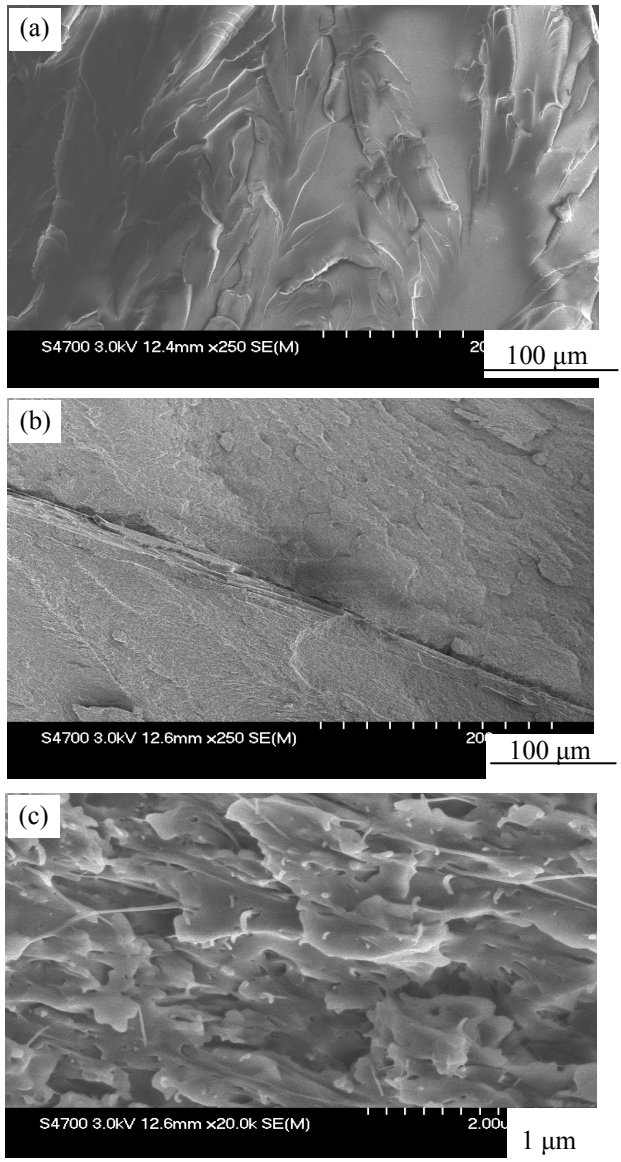

Figure 6: SEM photomicrographs of (a) neat epoxy resin, (b) composites with 11wt. % bacterial cellulose fibre, and (c) enlarged image of (b).

indicating the extensive pull-out of fibres as well as the corresponding holes. This suggests that the interfacial adhesion between cellulose nanofibre and epoxy resin is low in this system, resulting in the lower strength as compared with the theoretical estimation. Therefore additional surface treatment of the nanofibre is needed to get further strengthening.

4 Conclusions

In summary, the bacterial cellulose nanofibre reinforced epoxy composites were successfully fabricated by using freeze-dry method. Their tensile strength was almost comparable to that of glass-fibre reinforced plastics. Additional surface treatment of the nanofibre is needed to get further strengthening.

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