ENHANCED PHYSICAL PROPERTIES OF NANOCELLULOSE FIBER-REINFORCED GREEN COMPOSITES

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ABSTRACT

As a potential nanoscale reinforcement, nanocellulose fibers (NCFs) have drawn much scholarly attention. The NCFs whose typical diameter is 5–100 nm exhibited potentially high mechanical properties, and its tensile strength and modulus were estimated to be 1.7 GPa and 140 GPa. These higher tensile strength of the NCF is reported to be equivalent to those of glass fiber. However, the resultant mechanical properties reported for the NCF reinforced composites are lower than estimated values. In this study, we tried to enhance their mechanical properties by applying the following two approaches. These approaches are optimization in the fibrillation condition of NCFs by using grinding treatment and ultrasonic treatment. The effectiveness of the proposed two approaches has been successfully demonstrated experimentally; tensile strength and Young's modulus of the NCF-reinforced composites ultra-sonicated for 60 min were improved by 70% and 55%, respectively, compared to that of the untreated composites.

Keywords: nanocellulose fiber, green composites, polyvinyl alcohol, dispersion.

1 INTRODUCTION

At present, many kinds of plastic products are used in our daily life. Among these plastic products, fiber-reinforced plastics (FRPs) are used as structural members in automobiles, aircraft, etc. However, the FRPs are generally non-biodegradable and thus difficult to dispose of. For this reason, FRPs have the disadvantage of having a significant impact on the environment when disposed of. In order to obtain alternative composite materials with low environmental impact, research and development of green composites using biodegradable resin as a base matrix material and natural fiber as a reinforcing phase are being conducted continuously [1]–[3].

In this study, we focused on green composites reinforced with nanocellulose fiber (NCF), which is abundant on the earth and has high strength. In general, it is known that the properties of nanocomposites vary not only with the individual properties of the matrix and the nanoscale reinforcement but also with various factors such as interfacial adhesion, uniform distribution, and orientation of the reinforcement. However, there are many unclear points about its concrete contribution to the mechanical performances of the nanocomposites.

In the past, there have been studies on the preparation and characterization of nanocomposite materials combining the biodegradable polylactic acid (PLA) resin and NCF [4]. This study reported that it is difficult to fabricate nanocomposite material at the nano-order level in the PLA resin because strong cohesive force acts among the NCF and the reinforcing effect of NCF was not sufficiently obtained. To overcome this difficulty in the dispersion of NCF, they are studying a new dispersing technology using melt kneading. They confirmed that composites with aggregates of NCF of several tens of μm had improved strength compared to those with aggregates of NCF of several hundred μm. They also reported that the uniform dispersion of finer reinforcement in the resin matrix greatly contributed to the improvement in the strength of NCF-reinforced composite materials.

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However, the details of the effect of the size and dispersion of NFC is still not elucidated. In this study, we used polyvinyl alcohol (PVA)/NCF composite as a model material and focused on uniform dispersion of NCF reinforcement in PVA matrix and aimed at the development of high-strength NCF-reinforced polymer composites.

In this study, we used PVA/NCF nanocomposite as a model material. We focused on the effect of uniform dispersion of the NCF reinforcement in the PVA matrix and orientation of the reinforcement and aimed at the development of high-strength nanocomposite.

2 EXPERIMENTAL METHOD

2.1 Materials

In this study, we used Celish (KY-100G, Daicel Co., Japan) as NCF. The Celish is commercially available NCF in which wood pulp is microfibrillated to nanoscale cellulose fibrils by high-pressure homogenization treatment. PVA, a water-soluble biodegradable polymer (162–16325, Wako Pure Chemical Industries, Ltd., Japan) was used as a matrix polymer. PVA has excellent water-solubility and has characteristics that hydrophilic reinforcing phase such as NCF can be easily dispersed in the resin.

2.2 Preparation of preform sheets

First, the PVA powder of 25 g was poured into room temperature water of 475 g with stirring. The mixture (PVA content = 5 wt%) was then heated and dissolved using a mantle heater (HB-1000T, As One Corporation, Japan) with stirring. The Celish was in semi-solid form and the NCFs are highly agglomerated, so it was stirred with distilled water for 24 hours to prepare a 1 wt% NCF suspension. The aqueous PVA solution and the NCF suspension were mixed and stirred for another 24 hours. The resultant mixed suspension was cast into a polystyrene container and dried in a drying oven at 30°C for 24 hours to prepare PVA/NCF preform sheet, and then a further 100 g of the mixture solution was added on the PVA/NCF preform sheet which was not completely dried. This treatment was repeated six times to produce a thick preformed sheet. The final NCF content in the PVA/NCF composite sheet was 30 wt%.

2.3 Grinding treatment

The mixed suspension produced (NCF content = 1 wt%) was passed through a grinder (Supermasscolloider MKCA6-2, Masuko Sangyo Co., Ltd., Japan, Fig. 1) with a rotation speed of 1,500 rpm to refine NCF. This grinding process was performed up to twice.

2.4 Ultrasonication treatment

An ultrasonic oscillator (UH-150, SMT Co. Ltd., Japan, Fig. 2) was used to microfibrillate the NCF while stirring the PVA/NCF mixture (NCF content = 30 wt%) with a stirrer. A set of this microfibrillation treatment was ultrasonication treatment for 15 min. and cooling for 15 min. and this treatment was repeated up to four times (total ultrasonication time was 60 min.).

Figure 1: A grinder used in this study [5].

Figure 2: An ultrasonication machine used in this study.

3 RESULTS AND DISCUSSION

The results of the tensile test for the PVA/NCF nanocomposites are shown in Fig. 3. It can be seen that both the tensile strength and Young's modulus increase as the number of grinding treatment increases. Comparing the results of untreated specimens and specimens with twice grinding treatment, the tensile strength and Young's modulus of the treated specimen are improved by 43% and 28%, respectively.

Fig. 4 shows the results of SEM observation of the degree of fibrillation of NCF in order to investigate the effects of grinding treatment. We can see that some thick NCFs still remain in the untreated condition, on the other hand much finer NCFs are produced after grinding treatment. It should be noted that almost no thick NCF exists after twice grinding treatment. Similar experimental results regarding such refinement were also reported elsewhere [5].

Fig. 5 depicts the effect of ultrasonication treatment on the tensile properties of PVA/NCF nanocomposites. Both tensile strength and Young's modulus increase with increasing ultrasonication time. The highest tensile properties are obtained in the PVA/NCF nanocomposites ultrasonicated for 60 min. As compared with the untreated PVA/NCF

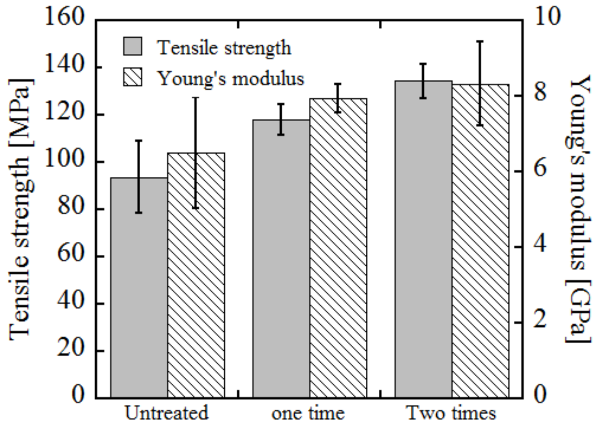

Figure 3: Tensile properties of the nanocomposites after grinding treatment.

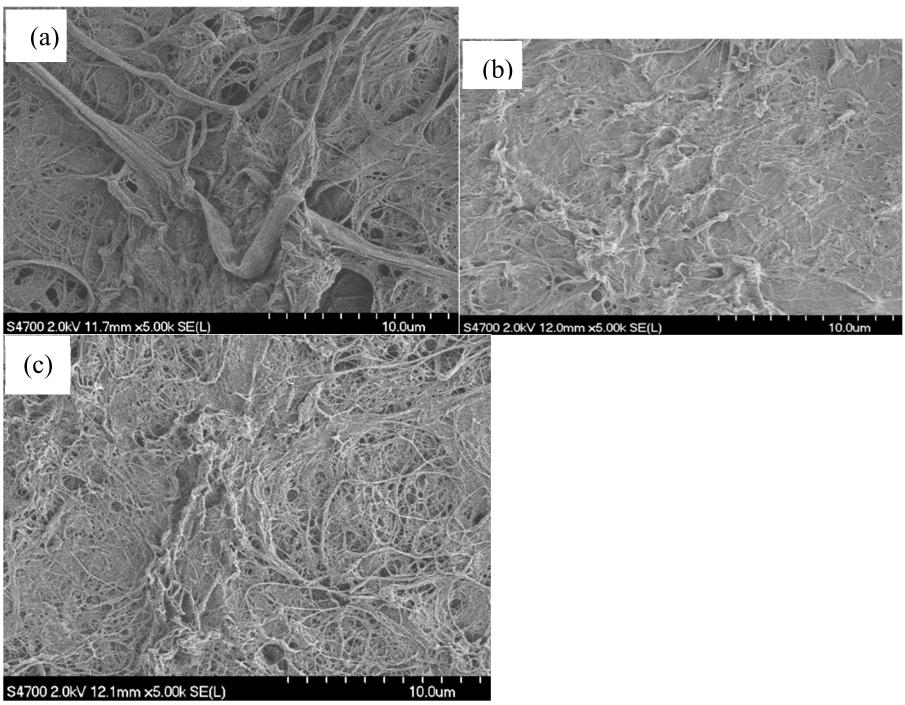

Figure 4: SEM photomicrographs of (a) Untreated NCF and grinding treatment of (b) One time and (c) Two times.

nanocomposites, the tensile strength and Young's modulus of the nanocomposites ultrasonicated for 60 min. are improved by 70% and 55%, respectively. The strength level of the nanocomposites ultrasonicated for 60 min was comparable to that of mechanical extension-treated nanocomposites with 30 wt% NCF [6].

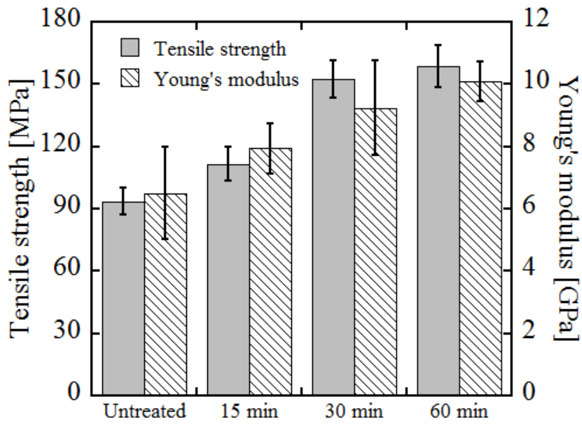

Figure 5: Tensile properties of the nanocomposites after ultrasonication treatment.

As in the case of the grinding treatment, the NCF subjected to the ultrasonic treatment has a smaller thick fiber instead. It can be seen that thick cellulose fibers are reduced in the NCF after the ultrasonic treatment as in the case of the grinding treatment. The following are conceivable causes of the improvement of the strength characteristics accompanying the finer fibers. As the fibers are finely fibrillated, the surface area of the fibers increases. Therefore, the bonding area at the interface between the resin matrix and NCF is increased, and the interfacial region where hydrogen bonding occurs is increased so that the strength is improved.

4 CONCLUSIONS

By performing the grinding treatment and the ultrasonic treatment, the tensile strength and Young's modulus of the PVA/NCF nanocomposite material are improved. It was suggested that the cause of the improvement in the mechanical properties of the PVA/NCF nanocomposites was related to the refinement of the NCF and the improvement of the uniform dispersion of the NCF in the PVA matrix.

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