RECENT RESEARCH ACTIVITIES

Novel process for fabricating high-modulus cellulose-based products

(Laboratory of Active Bio-based Materials, RISH, Kyoto University)

Kentaro Abe and Hiroyuki Yano

Cellulose is the most abundant natural polymer on earth. It is insoluble in water because of its inter- and intra-molecular hydrogen bonds and it occurs in plant cell walls in the form of crystalline nanofibers, the so-called cellulose microfibrils. Since this stable structure prevents it from dissolving even in common organic solvents, the fabrication of cellulose-based films and textiles often requires the use of special solvents. For example, toxic carbon disulfide has been used to prepare regenerated cellulose films, also known as Cellophane. However, the dissolution and regeneration processes deteriorate the Young's modulus of cellulosic products because of crystal conversion from Cellulose I to Cellulose II.

This study proposes a novel process for fabricating high-modulus films based on cellulose nanofibers. Instead of dissolution process, dried pulps were mechanically disintegrated into nanofibers in NaOH solutions. NaOH treatments loosened the hydrogen bonding between cellulose microfibrils in dried pulps. Furthermore, the preparation of a highly concentrated suspension (8%) of cellulose nanofibers using a ball-mill was attempted. In our previous study, a suspension containing a maximum of only 2% cellulose nanofibers was achieved by bead-milling because of high viscosity restrictions [1]. Therefore, in this study, a planetary ball-mill was used, which is a more powerful and effective piece of equipment for high viscous suspensions.

After ball-milling for 90 min a highly concentrated suspension (8%) of cellulose nanofibers with a uniform diameter of approximately 20–50 nm was prepared. The nanofiber suspensions prepared in the NaOH solution had both the crystal forms of Cellulose I and Cellulose II although Cellulose II gradually increased with increasing milling time. The suspensions were formed into hydrogels after neutralization and the formation of hydrogels is effective for the fabrication of cellulose nanofiber-based films. The hydrogel sheets were hot-pressed into thin films at 120 °C (Fig. 1). Young's modulus of the films was significantly higher compared to that of typical regenerated cellulose films due to some remaining Cellulose I (Table 1). Changes in the milling conditions and the apparatus used for nano-fibrillation need to be further optimized in order to maintain more cellulose I in the nanofibers leading to higher-modulus products. We believe that our method will facilitate the mass production of cellulose nanofibers as it also facilitates subsequent drying of hydrogels to fabricate films.

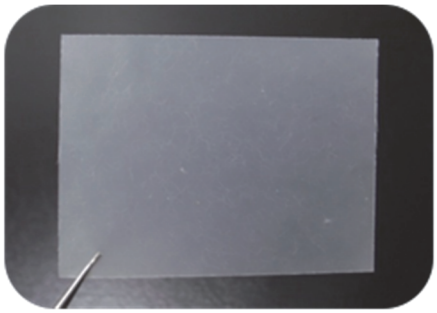

Figure 1. High-strength film based on cellulose nanofibers prepared by a ball-milling for 90 min in 8% NaOH solution.

Table 1. Average tensile property of cellulose nanofiber film prepared by a ball milling for 90 min in 8 % NaOH solution.

	Density (g/cm^3)	MOE (GPa)	MOR (MPa)	Break at strain (%)
8 %NaOH 90 min	1.46	9.3 (0.4)	100.2 (3.6)	2.4 (0.2)

Note: standard deviations are given in parentheses.

Reference

[1] Abe K, *Cellulose*, **23**, 1257-1261, 2016.