RECENT RESEARCH ACTIVITIES

Cellulose nanofiber-based hydrogels with high mechanical strength

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Cellulose nanofibers, which can be isolated from any plant sources by mechanical and/or chemical fibrillation processes, have great potential for reinforcing polymer matrices because of excellent mechanical properties, low weight, large specific surface areas and high aspect ratios. Furthermore, the crystalline nanofibers behave in water like dissolved polymers and homogeneously disperse in water without aggregation and sedimentation although cellulose is insoluble in water.

By taking advantage of the unique nature of the nanofiber suspension, we recently found the easy preparation of hydrogels from cellulose nanofibers through alkaline treatment [1, 2]. Unlike traditional polymer hydrogels, the nanofiber hydrogels could be prepared without dissolving in specific solvents and consisted of a highly porous and crystalline nanonetwork capable of having two different kinds of crystal form of cellulose I and II in response to the increasing concentration of alkaline solutions.

The two types of hydrogels with different crystal forms (cellulose I and II) exhibited high Young's modulus and high tensile strength because of the crystalline network in the gels (Figs. 1 and 2). The nanofiber hydrogel with a cellulose II crystal structure had a continuous network formed by the interdigitation of the neighboring cellulose nanofibers and showed higher tensile properties than the hydrogel with a cellulose I crystal structure. Although recently there have been a few other reports on the preparation of stiff hydrogels from cellulose nanofibers, the gel formation by the interdigitated nanofibers is a unique and interesting finding in this study.

Hydrogen bonds or chemical linkages between cellulose nanofibers are probably not powerful enough to support a high tensile force acting on cellulose nanofibers; however the rigid interdigitated linkage can support the high tensile property inherent in cellulose nanofibers. Paradoxically, the high mechanical properties of the nanofiber hydrogel with a cellulose II crystal structure provides supportive evidence for the interdigitation between cellulose microfibrils with opposite polarities.

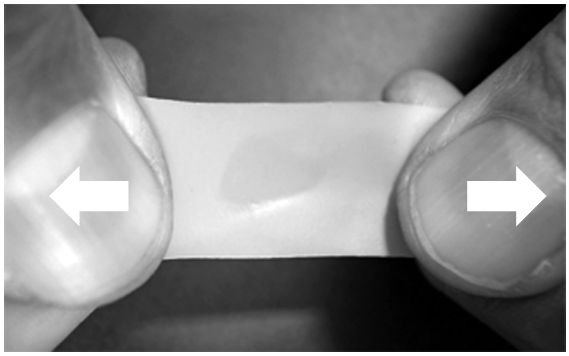

Figure 1. Appearance of the nanofiber hydrogels prepared in 15 wt% NaOH

Cellulose nanofibers are quite strong under tensile stress but rather weak when subjected to compression or bending, resulting in low compressive properties of the hydrogels prepared from cellulose nanofibers. The double-network method, which is a combination of the cellulose nanofiber gels with natural polymers and polysaccharides, is practical and useful for improving poor mechanical properties such as compressive modulus and strength.

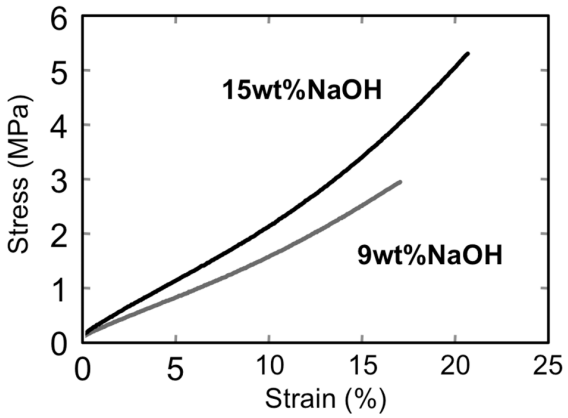

Figure 2. Tensile stress–strain curves of the nanofiber hydrogels prepared in 9 and 15 wt% NaOH.

References

[1] Abe, K., Yano, H. "Formation of hydrogels from cellulose nanofibers" *Carbohydrate Polymers* **85**, 733–737, 2011

[2] Abe, K., Yano, H. "Cellulose nanofiber-based hydrogels with high mechanical strength" *Cellulose*, DOI 10.1007/s10570-012-9784-3, 2012.