

Japan Academy Prize to:

Akira ISOGAI
Professor, Graduate School of Agricultural and Life
Sciences, The University of Tokyo

for “Studies on Completely Dispersed Cellulose Nanofibers
Originating from Plant”

***Outline of the work:***

Cellulose is the most abundant natural polymer, and it is one of the major plant components biosynthesized from atmospheric CO₂. Prof. Akira Isogai has succeeded in preparing new cellulose nanofibers (CNFs), from plant celluloses that are completely nanodispersible in water. The CNFs were prepared by catalytic reaction under aqueous conditions, which is similar to enzymatic reactions in living bodies. The dispersion mechanism and surface nanostructures of the CNFs were clarified from various analytical data. The CNFs can be converted into versatile functional bulk materials and composites owing to the unique nanostructures and morphologies of the CNFs. Prof. Isogai has, therefore, taken a leadership role in the scientific field of nanocelluloses based on his great achievements through domestic and international collaborations.

Crystalline cellulose microfibrils are the smallest elements of terrestrial plants and they have widths of ~3 nm and high aspect (i.e., length/width) ratios in plant cell walls. Each cellulose microfibril consists of 20–40 fully extended cellulose chains. Plant cellulose microfibrils coexist with non-crystalline hemicelluloses and lignin in cell walls at the molecular level. Such natural nanocomposite structures of plant cell walls inside plant living bodies allow plants to be resistant to weather, gravity, and biological attack. However, because cellulose microfibrils are tightly bound to each other through numerous hydrogen bonds in plant cell walls, individual cellulose microfibrils have not previously been separated and used as bio-nanofibers.

Prof. Isogai has investigated catalytic oxidation of cellulose using water-soluble and stable nitroxyl radicals under aqueous conditions (refs. 9, 30, and 31). When catalytic oxidation of plant cellulose fibers is performed, the oxidized celluloses maintain the original crystal structures, crystallinities, crystal sizes, and fibrous morphologies. Nevertheless, the sodium carboxylate content of the oxidized celluloses significantly increases by 170 times (refs. 4 and 9).

When the oxidized wood cellulose fibers are mechanically disintegrated in water, the fiber/water suspensions transform to transparent and highly viscous gels (refs. 4 and 9). Transmission electron and atomic force microscopy images show that the gels consist of nanofibers with homogeneous 3-nm widths and high aspect ratios, which originate from plant cellulose microfibrils. Therefore, Prof. Isogai was the first to prepare new CNFs from plant celluloses (refs. 4, 9, 30, and 31). Studies of the nanostructures and surface structures of the CNFs reveal that catalytic oxidation occurs position-selectively at the primary C6-hydroxy groups present on the crystalline cellulose microfibril surfaces, resulting in dense and regular formation of sodium C6-carboxylate groups on the cellulose microfibril surfaces (refs. 7 and 30).

All of the CNFs prepared from gymnosperm and angiosperm celluloses under optimum oxidation or nanofibrillation conditions have the same ~3-nm widths, irrespective of the plant species, which is scientifically interesting and also advantageous for use of CNFs as industrial nanomaterials (ref. 30). The

Young's moduli and tensile strengths of the single CNF elements are high and similar to those of multi-walled carbon nanotubes (ref. 14). Such high mechanical strengths of the CNFs originate from their crystalline nanostructures consisting of fully extended cellulose chains with high molar masses. Therefore, CNFs are new and unique bio-nanofibers with high mechanical strengths, large specific surface areas, and characteristic surface nanostructures and morphologies.

The CNF elements have high and stable nanodispersibility in water because electrostatic repulsion efficiently occurs between the CNFs owing to their high surface-anionic charges in water. CNF hydrogels prepared from aqueous CNF gels are self-standing and highly stiff owing to the high mechanical strengths of the CNFs and their high aspect ratios. CNF aerogels prepared from the CNF hydrogels by supercritical drying are transparent and stiff, and they can be used as new bio-based super insulators (refs. 19, 24, 26, and 27). Because abundant sodium carboxylate groups are densely present on the crystalline CNF surfaces, versatile ion-exchange of sodium to other metal and alkylammonium counterions can be achieved as for efficient modification of the CNF surfaces under aqueous conditions. This ion-exchange allows improvement of the mechanical strength and oxygen-barrier properties of the CNFs under high humidity conditions, efficient switching of the hydrophilic/hydrophobic properties, and stable/biodegradable characteristics (refs. 18 and 25). CNF-containing composites prepared using natural and synthetic polymers, carbon nanotubes, and metal nanoparticles have characteristic functionalities for practical applications (refs. 8, 12, 13, 15–17, 21, 22, and 28).

Large CNF production plants are already operating according to the procedure developed by Prof. Isogai. Various CNF-containing products have been commercialized, such as diapers with super-deodorant properties, dispersants, thickeners, functional packaging films, and electronic devices (ref. 30). Qualitative and quantitative expansion of CNFs produced from abundant wood resources in the world may contribute to creation of a sustainable and environmentally friendly society based on renewable biomass resources, hopefully leading to prevention of global warming (ref. 30).

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