Functional Nanocellulose from Oil Palm Empty Fruit Bunches Pulp

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（オイルパーム空果房パルプ由来の機能性ナノセルロース）

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Thesis Summary

The use of agricultural residues as inexpensive raw materials to produce high-value natural nanomaterials such as nanocellulose is aligned with the concept of Sustainable Development Goals (SDGs) by pioneering its promising functions in several applications. Nanocellulose derived from oil palm empty fruit bunches (OPEFB) pulp has been successfully extracted by hydrochloric acid hydrolysis and TEMPO-mediated oxidation, respectively, affording cellulose nanocrystals (CNCs) and TEMPO-oxidized cellulose nanofibers (TOCNs). The CNC suspensions prepared from OPEFB by HCl hydrolysis remained stable without any sedimentation over 6 months. The OPEFB-derived CNCs exhibited high aspect ratios and superior thermal stability compared with those of woody CNCs. Very fine TOCNs in 3-nm thickness obtained from OPEFB pulps possessed high aspect ratios rather than 40 with various contents of carboxylates (1.0–1.5 mmol/g-TOCN), and showed good thermal stability.

Furthermore, highly-charged TOCNs from OPEFB were very effective in extractive fermentation to enhance butanol production. TOCNs were successfully applied to extractive fermentation using *Clostridium saccharoperbutylacetonicum* N1-4 as a butanol-producing strain. The butanol yield from free and immobilized cells remarkably increased in the presence of OPEFB-derived TOCNs. In the immobilized cell method, crosslinking of anionic carboxylate groups of TOCNs and alginate *via* Ca^{2+} ions to form alginate beads induced favorable 3-D networks for cell entrapment, and possibly provided better microenvironment for bacterial growth. The combination of OPEFB-derived TOCNs and alginate eventually yielded the highest total butanol production up to 37 g/L-broth.

From another viewpoint, the composite films of OPEFB-derived TOCNs and alginate performed as a pervaporation membrane for water/ethanol mixture, showing high flux as compared to PTFE commercial membrane. By applying pressure ranging from 30–40 hPa during the pervaporation, there was no micro-cracks observed on the membrane surface. Cross-linking between TOCNs and alginate presumably induced the change of pore sizes and surface areas, which are significant for adsorption capacity.

The use of CNCs and TOCNs from OPEFB as a reinforcing filler was tried to make polymer composites with poly(methylvinyl ether-*co*-maleic acid)–polyethylene glycol, and revealed different properties in surface roughness, optical transmittance, contact angles of water droplets, total color differences, thermal stability and mechanical properties. Fiber length of TOCNs affected the film properties, and induced higher mechanical performance as compared to other agricultural residues, confirming that the character of raw materials and the extraction process may play a critical role in

forming nanocellulose properties.

Overall, this work revealed the potential of low cost biomass, OPEFB pulp, as a source of functional nanocellulose in bioenergy applications and as additives in various polymer composites. The obtained findings could open up a wider utilization of agricultural residues and unused biomass into novel value-added products by advanced nanocellulose technology.