Doctoral Thesis

Utilization of Cellulosic Biomass towards sustainable Chemicals and Novel Biomaterials

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Abstract

It is predicted by the United Nations that by year 2030 the world will need at least 50 percent more food, 45 percent more energy and 30 percent more water. The emissions of carbon dioxide from combustion of fossil fuels and waste are also increasing. At the same time, the demand for natural resources has never been higher and the planet is under unprecedented stress. This increasing awareness and concerns also drive and accelerate the research to facilitate switching the fossildependent economy to biobased economy. In this premise, forest industry plays a significant role, from leading the sustainable development to providing more materials to meeting the expanding demand. Moreover, the forest industry is a crucial part of the solution to global warming. The utilization of forest product has a long history, and the efforts of converting the biomass into value-added products or innovative applications have never been more stimulated than now.

This thesis presents some examples of the exploration of lignocellulosic biomass based on the fractionation of lipophilic extractives and utilization of non-derivatized cellulose in novel materials. In the first part of thesis, the biorefinery of thermo-mechanical pulping (TMP) process water for lipophilic extractives was investigated as a way to extract the dissolved and colloidal substance (DCS). It was found that induced air flotation (IAF) combined with the foaming agent dodecyl trimethylammonium chloride (DoTAC) can efficiently remove the unwanted lipophilic extractives (Paper I) and retain valuable hemicelluloses (Paper II) in the TMP process water. By applying 80 ppm of DoTAC at a pH of 3.5 and 50 °C with induced air flotation, 94% of the lipophilic extractives were refined from the process water. The efficient biorefining of lipophilic extractives not only enabled the purification of TMP process water, but also facilitate the selective harvesting of hemicelluloses with very low impurities.

In the second part of the work, non-derivatized cellulose (sulfite pulp) dissolved in LiOH/urea was used as substrate to fabricate spherical nanocomposite particles (Paper III), pH-responsive nanocomposite films (Paper IV) and crosslinked cellulose hydrogel (Paper V), respectively. The cellulose-chitosan nanocomposite particles were prepared in three different ways: instantly by dripping alkaline cellulose solution into dissolved chitosan in diluted acetic acid, and by mixing and emulsifying the biopolymer solutions to a water-in-oil emulsion, with or without addition of a crosslinking agent. Spherical cellulosechitosan nanocomposite particles in the size from millimeter to micrometers were successfully prepared. It was demonstrated that some properties of the spherical particles, for example, morphology and size distribution, could be tuned by choosing between the different routes of preparation. In a different application of LiOH/urea dissolved cellulose, in the form of cellulose-chitosan nanocomposite films with pHresponsive swelling, were shown in the thesis. The nanocomposite film with 75% chitosan content exhibited maximum swelling ratio of 1500% and weight loss of chitosan of 55 wt% after 12 hours at pH 3. The utilization of the non-derivatized cellulose continued with cross-linking the macromolecules with methylenebisacrylamide (MBA) to form a robust hydrogel with superior water absorption properties. The cellulose hydrogel cured at 60 °C for 30 minutes, with a [MBA]/[glucose] molar ratio of 1.05, exhibited the highest water swelling capacity absorbing ca. 220 g H2O/g dry weight. This innovative procedure based on the direct dissolution of unmodified cellulose in LiOH/urea followed by MBA cross-linking provides a simple and fast approach to prepare chemically cross-linked cellulose hydrogels of high molecular weight with superior water uptake capacity.

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