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Biphasic lignocellulose fractionation for staged production of cellulose nanofibers and reactive lignin nanospheres: A comparative study on their microstructures and effects as chitosan film reinforcing

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ABSTRACT

The effective fractionation of lignocellulose and the enhancement of its components require formidable efforts. This study explored synergistic biorefinery routes for the holistic conversion of lignocellulose biomass to green nanomaterials through one-pot fractionation using recyclable *p*-toluenesulfonic acid (*p*-TsOH)/pentanol reagents. The potential of *p*-TsOH/pentanol pretreatment of aspen biomass under various temperatures (80–180 °C) for co-production of cellulose nanofibers (CNFs) and lignin nanospheres (LNSs) was investigated. The optimal pretreatment temperature, which minimizes cellulosic degradation and promotes the holistic conversion of biomass, was 120 °C. In addition to its highly digestible nature, the cellulose-rich residue produced high-quality CNFs with a diameter <100 nm, good crystallization, and thermostability, making it suitable for various applications. Lignin characterization, molecular dynamics simulation, and density functional theory analyses of the recovered lignin revealed well-protected β-O-4 bonds (47.3/100 Ar), uniform molecular weights (M*w*, 2815 g/mol; M*n*, 1704 g/mol; PDI, 1.65), < 1% sugar content, fewer condensed structures, and nanosize shapes (100–300 nm) due to the molecular interactions of *p*-TsOH and pentanol with lignin units. Moreover, 0.4% LNSs additions reduced the chitosan film thickness by 53% and enhanced its tensile strain and strength by 33 and 59%, respectively, as compared to the pure chitosan film. This study provides a sustainable platform for the co-production of biobased CNFs and LNSs using cutting-edge fractionation of LCB, which is a breakthrough toward industrial biorefineries.

1. Introduction

Achieving the United Nations Sustainable Development Goals, including energy, environmental, and climate security targets, requires new circular production systems. Accomplishing this goal requires cutting-edge research and the use of different waste/bio-resources [1]. Lignocellulose biomass (LCB), with an estimated annual global production rate of 200 billion metric tons, is the most biorenewable resource on the planet [2]. Converting LCB to biofuels is one of the most effective ways to address the imminent scarcity of fossil fuels and the environmental concerns caused by their combustion [3]. Biobased chemicals, biomaterials, biopolymers, cellulose fibers, and other high-value products can be obtained from LCB using biorefinery pathways [4]. In the integrated biorefinery pathway, pretreatment plays a crucial role in overcoming LCB recalcitrance for efficient fractionation and conversion of its main components, namely, cellulose, hemicellulose, and lignin [5,6].

Recently, lignin-first biorefinery approaches through biphasic

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