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Efficient lipase-catalyzed esterification of carbohydrate polyols in reactive natural deep eutectic solvents

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PURPOSE OF THE ABSTRACT
Biocatalytic pathways for esterification of carbohydrates and carbohydrate polyols show the significant advantage of a single step operation, but also face the challenge of finding the appropriate solvent to enhance simultaneously the solubilization of sugars and the stability and regioselectivity of the biocatalyst in the reaction medium, while minimizing the environmental burden. Natural deep eutectic solvents (NADES) emerged recently as a novel class of green solvents and a prospective solution for most of the problems related to the reaction media in biocatalytic transformations, being nontoxic, biodegradable, and biocompatible. In this study, we present a new route for the enzymatic esterification of carbohydrate polyols using a reactive NADES (R-NADES) as reaction medium, acting both as a solvent and as a reagent pool.
Binary hydrophilic R-NADES consisting of choline chloride (ChCl) as hydrogen bond acceptor (HBA) and sugar alcohols (i.e., D-sorbitol, xylitol, D-arabitol) as hydrogen bond donors (HBD) were prepared and characterized to determine the freezing temperature (Tf), the thermal stability and the viscosity-temperature profile. All polyol-based R-NADES were stable fluids at temperatures up to 300□C and showed low viscosities between 40-80□C, that is the optimal temperature range for lipase-catalyzed reactions. Polyol-based R-NADES were compatible reaction media for a number native and immobilized lipases. Lipase B from Candida antarctica immobilized on acrylic resins (LAR) showed significant esterification activity and high thermal stability in tested R-NADES, maintaining more than 90% of activity upon incubation at 70□C for 72 hours. Moreover, LAR effectively catalyzed the synthesis of lauryl esters of carbohydrate polyols in the binary ChCl/polyol R-NADES tested and consequently, the synthesis of lauryl esters of D-arabitol, xylitol and D-sorbitol was successfully achieved. The design of experiments for D-arabitol esterification with lauric acid (LA) catalyzed by LAR revealed that enzyme load and temperature are decisive reaction parameters for conversion increase. Preparative reaction at optimal conditions achieved 80% conversion of lauric acid after 24 h. The major product was identified as 1,5-dilauryl-D-arabitol based on ESI-MS, FTIR, 1D- and 2D-NMR. The 1,5-DLDA product was isolated in 95 % yield. Minor amounts of monolauryl-D-arabitol were detected by RP-HPLC, showing an almost quantitative conversion into the diester. At similar conditions, 1,5-dilauryl-xylitol (1,5-DLX) and 1,6-dilauryl-D-sorbitol (1,6-DLS) were obtained, at a LA conversion of 56 mol% and 62 mol%, respectively. The results are unprecedented in literature reports and open the way to further develop green processes for the enzymatic synthesis of glycolipids.
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FIGURES

FIGURE 1

FIGURE 2

KEYWORDS
reactive NADES | lipase | arabitol esters | green processes

BIBLIOGRAPHY