Green Nanofibrillation of Hemp Cellulose via Deep Eutectic Solvent and Simple Shear Mixing: A Response Surface Approach to Process Refinement

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ABSTRACT: Sustainable production of cellulose nanofibrils (CNFs) from bast fibers offers a promising route to high-value biobased materials. Despite their high cellulose content (70–92%), hemp fibers remain underexplored for nanofibrillation. Here, hemp cellulose was nanofibrillated for the first time using a choline chloride–lactic acid deep eutectic solvent (DES) followed by shear mixing. Response surface methodology (RSM) was employed to model the effects of DES treatment and shear mixing time on the fibril diameter, yield, and aspect ratio. The quadratic models an exhibited excellent predictive capability ($R^2 > 0.99$), confirming their robustness. Fibril diameter decreased under harsher conditions, reaching 10–12 nm, while the aspect ratio increased up to ~145. Yield declined with increasing severity, highlighting

the trade-off between fibrillation efficiency and material recovery. SEM and AFM confirmed progressive fibrillation from coarse bundles under mild conditions to highly individualized nanofibrils with narrow distributions (<20 nm) under optimized conditions. Sedimentation and gel point analysis indicated enhanced suspension stability and network formation for the optimized sample, with gel points as low as 0.15 wt % and a zeta potential of −30.3 mV. Energy analysis revealed shear mixing as the dominant contributor to consumption, yet optimization achieved superior fibrillation at a reduced demand of ~1.5 kWh for the whole process. FTIR confirmed cellulose esterification by DES, while XRD verified retention of cellulose I structure with crystallinity indices of 71–79%. The solvent was successfully reused for three consecutive cycles without loss of the treatment performance. Overall, this study demonstrates an efficient, low-energy route for producing high-quality hemp nanofibrils by integrating DES pretreatment, shear processing, and statistical optimization.

KEYWORDS: bast fibers, choline chloride/lactic acid, DES-assisted fibrillation, response surface methodology, crystallinity
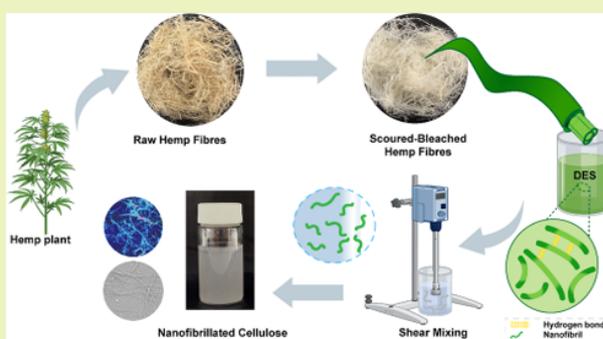
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