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FURAN-DERIVED NANOCRYSTALLINE CELLULOSE AS BIO-FILLER FOR SUSTAINABLE RUBBER COMPOSITES

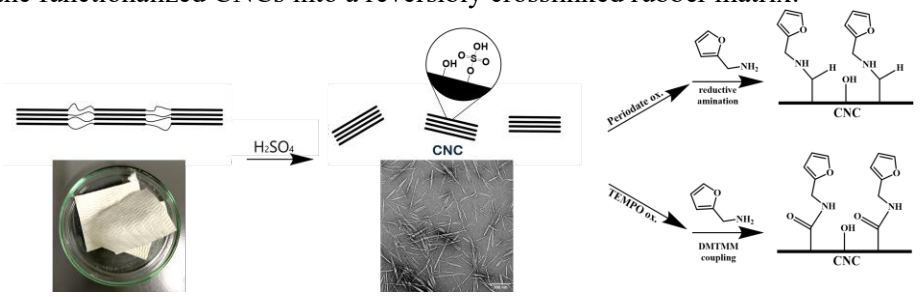
ANDREA RICCIONI[1], LUCREZIA CRISCUOLO[2], VINCENZINA BARBERA[2], ROBERTA BONGIOVANNI[1], ALESSANDRA VITALE[1].

[1] *Department of Applied Science and Technology, Politecnico di Torino, Torino, Italy*
[2] *Department of Chemistry, Materials and Chemical Engineering, Politecnico di Milano, Milano, Italy*

Email: andrea.riccioni@polito.it
PhD student 2nd year

Abstract

In recent years, nanocellulose (NC) has gained growing attention as a biobased material thanks to its high tensile modulus, large surface area, and complete renewability.[1] Moreover, the NC surface can be chemically modified to tailor its properties, making it a promising filler in the development of sustainable composites and elastomers.[2] Besides the replacement of traditional fillers, recycling of the polymeric matrix is key to composite/elastomers sustainability: dynamic covalent polymer networks (DCPNs) offer a solution by combining thermoset durability with thermoplastic reprocessability.[3] One widely used thermoreversible crosslinking strategy exploits the Diels–Alder (DA) reactions.
The present work describes the modification of cellulose nanocrystals (CNCs), in order to assure covalent and reversible interactions with a polymeric matrix through a DA reaction between furan and maleimide groups. In this perspective, different cellulosic materials (i.e., hemp pulp, micronized powder cellulose and microfibrillated cellulose) were treated with sulfuric acid to obtain CNCs. The extracted materials were then characterized via FT-IR spectroscopy, XRD analysis and TEM. Following this preliminary screening, hemp pulp was selected as the material of choice for further investigation. Two different strategies were then designed to graft furan moieties on CNC surface: one is based on reductive amination and the other on a coupling reaction (Figure). These synthetic approaches required pre-treatment of nanocellulose with sodium periodate or TEMPO, to introduce aldehydes or carboxylic groups, respectively. The presence of furane in the final products was confirmed by both FT-IR and Solid-state NMR, while the oxidation degree of the intermediate products was estimated using standard titration procedures. Preliminary tests were conducted to incorporate the functionalized CNCs into a reversibly crosslinked rubber matrix.



Extraction and functionalization strategies of hemp-derived cellulose nanocrystals (CNC)

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