

Reducing End Modification of Nanocellulose as a Novel Approach for High-performance Sustainable Composites

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The development of nanocellulose sustainable materials is considered as one of the most promising alternatives to address plastic pollution issues, as global plastic wastes may increase to 11 billion tonnes by 2025. However, how to achieve the homogeneous dispersion of nanocelluloses (CNCs) and strong interfacial interactions with matrix materials, while well maintaining its percolation networks, is a challenge in this field. As opposed to the conventional surface chemical modification strategy, the reducing end modification of CNCs as a novel approach provides an opportunity to achieve this objective, which also opens a new door for the design of stimuli-responsive CNC sustainable composites, such as vitrimer materials and stimuli-responsive Pickering emulsions.

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What is the Difference between Reducing End and Surface Modification of CNCs?

The reducing end modification of CNCs is different from its surface modification strategy in terms of the quantity and reactivity of reducing end aldehyde and surface hydroxyl groups. The surface modification primarily works on the three hydroxyl groups on the CNC backbone, resulting in the varied reactivity at different positions, e.g., C6-OH > C2-OH or C3-OH, while aldehyde groups at the reducing end modification have high reactivity, but the influence of steric hindrance is an important factor to be considered (Chemin *et al.* 2022). For the quantity of available groups in CNCs for chemical modification, the content of surface hydroxyl groups is much higher than that of aldehyde groups at the reducing end, but the reactive groups at the reducing end makes it possible to introduce some unique features by tailoring CNCs directly on one end (Imlimthan *et al.* 2019). The present work in term of reducing end modification primarily focuses on the group derivatization with introduction of new functional groups (Imlimthan *et al.* 2019) and polymer graftings with functional species on the reducing end of CNCs (Chemin *et al.* 2022).

How Does Reducing End Modified CNC Influence its Composite Performance?

Research in terms of CNCs-based sustainable composites has attracted researchers' great interest for the last few years. However, the mechanical performance of composites by adding CNCs is still far from expectations due to nonuniform dispersion and inadequate

interfacial interaction issues. Although the surface modification can make it possible to tailor its dispersion and interfacial adhesion with matrix materials, the consumption of surface hydroxyl groups of CNCs in conventional surface modification results in disrupting its percolation networks constructed by hydrogen bonding interactions in hydroxyl groups on the surface of CNCs (Habibi *et al.* 2010). Thus, researchers are faced with the question of how to achieve its homogeneous dispersion and strong interfacial interactions with a nonpolar matrix while maintaining CNC percolating networks. In terms of reducing end modification, the functional groups are only reacted with aldehyde groups at the end of CNCs, while its surface functional groups are well preserved, so its stable percolation networks are able to be established, thereby improving the composite performance. For instance, Lin's group achieved the well dispersion of CNCs and its excellent interfacial adhesion with the styrene-butadiene-styrene copolymer elastomer matrix by CNC reducing end modification, which was done in combination with a UV-induced thiol-ene click reaction strategy (Tao *et al.* 2019). This novel approach also preserved the surface property and ensured the formation of stable CNC percolation networks, thereby significantly enhancing composite mechanical performance.

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