

# Nanocellulose Fibril-Templated Preparation of $\text{Na}_{1+x}\text{Zr}_2\text{SixP}_3\text{-xO}_{12}$ (NZSP) NASICON Electrolytes

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Solid-state electrolytes (SSEs) can address the issues of safety, longevity, and energy density limitations in batteries, but an up-scaling challenge remains from high-risk processing parring with good ionic conductivity and interfacial contact.

Meanwhile, NASICON-type SSEs for sodium-ion batteries have been investigated widely for achieving a decent balance of stability for performance. Their progress nonetheless is still hampered by non-uniform structural design and impurity apparitions which reduces the  $\text{Na}^+$  ion transport efficiency.

Our group introduces cellulose nanofibrils (CNFs) as a biotemplate [3] for the arrangement of  $\text{Na}_{1+x}\text{Zr}_2\text{SixP}_3\text{-xO}_{12}$  ( $0 \leq x \leq 3$ , NZSP) electrolytes. The CNF's 3D fibre network permits the passage of NZSP sol, creating a systematic alignment of the NASICON grains. Material characterisation reveals a solid solution ( $x \sim 2.4$ ) consisting of monoclinic and rhombohedral NZSP crystals, with the CNF-templated NZSP favouring the latter. X-ray diffraction (XRD) and scanning electron microscopy (SEM) results reveal a reduction of impurities and more uniform NZSP grains from the CNF-NZSP.

Sintered CNF-NZSP pellets show higher ionic conductivities ( $\sim 3 \times 10^{-4} \text{ S cm}^{-1}$ ,  $25^\circ\text{C}$ ) compared to regular NZSP pellets, rendering them better candidates for NASICON-type SSEs. This work aims to determine the magnitude of ionic conductivity improvement and reduction of interfacial resistance with biotemplated NASICON SSEs. Alongside, this work has the potential to increase the valorisation of biomass components in energy storage application, speeding up the realisation of solid-state, sodium-ion batteries.