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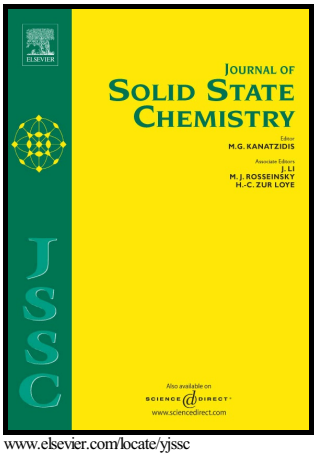

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Sodium superionic conduction in tetragonal Na_3PS_4

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Abstract

Sodium superionic conductors are highly promising as solid electrolytes for all-solid-state sodium batteries, with the capacity to improve energy and power density of liquid-electrolyte-based batteries and to address their safety issues. However, the ionic conductivity of sodium electrolytes remains roughly one order of magnitude lower than that of lithium systems. Herein, we report a synthesis method for a sodium superionic conductor, tetragonal Na_3PS_4, with an ionic conductivity of 3.39×10^{-3} Scm^{-1} at 25 °C—the highest value yet reported among sulfide-type sodium conductors. The high conductivity is achieved through quenching from 700 °C, which introduces sodium vacancies, and annealing at 450 °C, which increases vacancies and expands lattice volume. The phase is comprised of a framework of isolated PS_4 tetrahedra, with partial distribution of Na over 3D pathways. The high intrinsic conductivity together with low grain boundary contribution and soft mechanical properties make Na_3PS_4 a promising candidate for sodium solid electrolytes.