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Understanding microstructure-induced limitations of hydrogen transport in high temperature proton conductors: can nuclear microanalysis give an answer?¹

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High temperature protonic conductors (HTPC) are envisioned as electrolytes for fuel cells working at intermediate temperature (400 °C – 600 °C) to complement Y₂ZrO₇ electrolytes operating at 800 °C – 1000 °C. The most mature HTPC are doped perovskites (ABO₃) where tetravalent cation B is partially substituted by a trivalent one. Protons can be introduced in the lattice as point defects corresponding to hydroxyl groups on oxygen ion sites. In the temperature region of interest for technological applications, lattice vibrations allow the diffusion of protons by jumping and reorientation of O-H bonds (hoping mechanism). BaCeO₃ or SrCeO₃-based perovskites doped with a rare earth are the most widely studied compounds. However the proton conductance of these ceramics and their chemical stability are lower than the calculated values on single crystals and not sufficient to fulfill technological requirements. In most cases, the reasons for these discrepancies lie in uncontrolled microstructures with inter- and intra-granular defects that act as barriers for hydrogen diffusion but are preferential paths for chemical degradation by hydrolysis or carbonatation. Despite this crucial point, very few efforts are devoted to the optimization of microstructure of HTPC. Microstructure induced limitations are usually evidenced via impedance measurements which enable determination of respective contributions of bulk and grain boundaries to overall conductivity. Further information on hydrogen transport relevant for improvement of microstructure design requires local methods for hydrogen concentration measurement. Nuclear microanalysis, based on the use of MeV light ions microbeam, meets this demand. According to the chosen technique, nuclear reaction, elastic recoil or forward coincident scattering, the nuclear microprobe gives 2D-3D quantitative information on hydrogen distribution and diffusion within microstructure and enables to identify barriers and short-circuits.

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