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Proton dynamics in high temperature polymer electrolyte fuel cell membranes

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Polybenzimidazole (PBI, poly(2,2'-(m-phenylene)-5,5'-bibenzimidazole) membranes with phosphoric acid impregnation are a new promising material for high-temperature polymer electrolyte membrane fuel cells (HT-PEFC). PBI as membranes for fuel cells was introduced in the middle of the nineties as replacement for the well-investigated Nafion membranes, whose use is limited by the boiling temperature of water and thus, water-thermal management. After being impregnated with phosphoric acid (PA) PBI membranes show very good proton conductivity. Besides, the operating temperature of about 160℃ of HT-PEFC results in a high CO tolerance of 1-2%. However, although PBI-based PEMFCs are well studied on macroscopic level (conductivity, rheology etc. [1, 2]), the microscopic dynamics associated with the conductivity has not been investigated in detail except from molecular dynamics simulations [3, 4]. A method predestined for this purpose is quasielastic neutron scattering because of the high incoherent scattering cross section of protons and the spatial resolution of the technique.

In order to investigate the microscopic proton dynamics in these PA impregnated membranes over a wide dynamic range, data from three neutron scattering instruments were combined in time range after Fourier transform: the time-of-flight spectrometer CNCS at SNS, Oak Ridge (0.2⋯30 ps), the TOF-backscattering spectrometer BASIS at SNS, Oak Ridge (20⋯800 ps), and the backscattering spectrometer SPHERES at MLZ, Garching (70 ps⋯5 ns). To suppress the scattering from the polymer matrix, the difference between samples with PA and deutero-PA was used as the basis of the evaluation.

A first description of the data using a model based on a distribution of relaxation times gave satisfactory results but left the microscopic picture open. Analysing the Q dependence of the characteristic times we could identify a region with an anomalous power law behaviour indicating diffusion on a fractal structure. A scattering function derived from a fractal-based model enabled us to describe the scattering on certain length scales.

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