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Intrinsic Proton Dynamics in Hydrus Silicate Melts as seen by QENS at elevated Temperature and Pressure

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We present quasielastic neutron scattering results on hydrous silica, sodium aluminosilicate, and sodium trisilicate melts with 10mol% total water content, studied at high temperature under high pressure. Combining neutron time-of-flight spectrometry with neutron backscattering, intrinsic, microscopic proton dynamics is investigated on a timescale from 0.2 ps up to 1 ns between 850K and 1250K. All three hydrous silicate melts exhibit a relatively slow proton dynamics, although the melt viscosity is drastically reduced upon water dissolution. The self-diffusion coefficient of proton in the hydrous sodium trisilicate melt is on the order of 10^{-11} m²s⁻¹, two orders of magnitude slower than the sodium dynamics in the corresponding dry melt. The proton dynamics in hydrous silica and albite is not faster than that time scale. We show that the transport mechanism involving not only -OH but also molecular water species. All protons are mobile during the transport of the water instead of diffusion of a specific water speciation. These characteristics of the proton structural relaxation in the melt can be attributed to a transport in a complex H-bonding environment involving water and the Si-O matrix.

Primary author: Dr YANG, Fan (Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt, Köln)

Co-authors: Prof. MEYER, Andreas (Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt, Köln); Prof. DINGWELL, Donald (Department für Geo- und Umweltwissenschaften, Ludwig Maximilians Universität München); Dr MAMONTOV, Eugene (Spallation Neutron Source, Oak Ridge National Laboratory, Oak Ridge, USA); Dr HESS, Kai-Uwe (Department für Geo- und Umweltwissenschaften, Ludwig Maximilians Universität München); Prof. UNRUH, Tobias (Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), Garching)

Presenter: Prof. MEYER, Andreas (Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt, Köln)

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