ESRF	**Experiment title:** The local structure of silicate melts by in-situ x-ray Raman scattering	**Experiment number**: ES-55
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Names and affiliations of applicants (* indicates experimentalists):

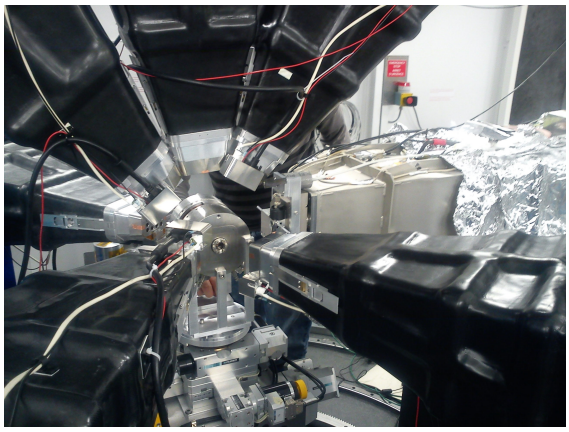
M. Wilke[*a)], K. Mende[*b)], Ch. Schmidt[*a)], A. Nyrow[*b)], J. Nyrow[*b)], Ch. Weis[*b)], C. Sternemann[*b)]

a) Helmholtz-Zentrum Potsdam, Deutsches GeoForschungsZentrum GFZ, D-14473 Potsdam, Germany.
b) Fakultät Physik/DELTA, Technische Universität Dortmund, D-44221 Dortmund, Germany.

Report: The aim of experiment ES-55 was to study the local coordination of sodium in dry and hydrous melts using in-situ X-ray Raman scattering at the sodium and oxygen K-edges together with resistive heated diamond anvil cell sample environment. This way we wanted to get insight into the role of H_2O in the structure formation in hydrous silicate melts at pressure and temperature. These melts are able to store high amounts of water, which is highly relevant for the understanding of e.g. ascent of magma.

We applied x-ray Raman scattering (XRS) to study the sodium and oxygen K-edges of glasses and melts at ex-situ and in-situ conditions. The focus was set on dry and hydrous albite ($NaAlSi_3O_7$) and NS3 ($Na_2Si_3O_7$) with up to 5 % and 10 % water content, respectively. The ex-situ measurements on quenched samples were performed for a natural sample of crystalline albite (Brazil), crystalline albite powder (SP1051), albite glass with 5 % water, NS3 glass, and NS3 glass with 10 % water content. Spectra of NS3 hydrous samples show small but distinct changes in overall shape compared to the dry glass indicative for an increased polymerization induced by the H_2O incorporation, a direct evidence of the effect of H_2O on the structure formation in sodium silicate glasses.

The in-situ measurements were carried out using a modified resistively heated diamond anvil cell using reducing atmosphere instead of vacuum to avoid aging and thus damage of heating wires and thermo couples. Hydrous glasses were loaded together with ultrapure water into the diamond anvil cell with a rhenium gasket. One of the diamonds had a recess hole to probe the melt with significantly reduced pathlength of the incident and scattered x-rays through the diamonds. Melts were produced at pressures around 250 MPa and temperatures in between 600 °C to 800 °C for NS3 and albite, respectively. We used the Si 311 analyzer reflection to measure both edges with a high energy resolution of about 0.7 eV. Although we were able to prepare the melts for in-situ measurements in the diamond anvil cell, the sample position of the NS3 melt within the gasket's hole and the recess was highly unstable showing dynamic fluctuation of the melt within the sample volume. Owing to these fluctuations it was not possible to acquire consistent spectra of the oxygen's and sodium's K-edges. Changing the sample from NS3 to albite hydrous melt did not improve the stability. Hence, we were not able to carry out the in-situ measurements as planned. The final alternative was to change the monochromator to Si 111 which significantly enhances the flux on the sample with the aim to achieve much shorter data acquisition times to yield consistent results for different measurements. However, the problem here was, that the x-ray intensity was too high, that strong bubble formation in the hydrous melt was induced. Consequently, the occurrence of radiation damage is likely and the corresponding data have to be critically evaluated. The analysis of in-situ data is still in progress, but to this point no final results can be presented. Possibly these measurements have to be repeated with a complete new design of the sample environment using e.g. pre-loaded gasket and diamonds without recess to avoid sample fluctuation and to guarantee proper data quality with adequate normalization.



Concerning the ex-situ measurements molecular dynamics simulations are performed and corresponding XRS spectra of the Na K-edge will be calculated to model the effect of H_2O incorporation on the degree of polymerization in the melts.