In situ XAS measurement of Zr, U and Th in silicate melts using the D-DIA facility at the Australian Synchrotron

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Geochemical variations in the Earth, such as the chemical differences between the core, mantle and crust, are fundamentally explained through an understanding of how elements are partitioned amongst metals, minerals and melts over a wide range of pressures, temperatures, and redox conditions. Abundant experimental evidence demonstrates that the coordination numbers of cations in silicate melts begin to increase at pressures as low as ~ 3 GPa, which implies that changes in partitioning behaviour may occur as a function of pressure. However, we routinely use ambient or low-pressure partitioning data to understand mantle processes. Changes in cation speciation are often cryptic, as they are not always faithfully preserved in glasses upon quenching. Without high quality in situ measurements, it is difficult to determine the utility of speciation data collected from quenched glasses, or glasses annealed at high pressure. To date, there are few extant in situ studies of element coordination as a function of pressure in liquids or glasses, and even fewer applying XAS. Here we present the results of experiments conducted using the D-DIA facility at the Australian Synchrotron, which has allowed us to collect XAS measurements of the speciation of key trace cations (U, Th and Zr) in geologically-relevant silicate melts in situ at high temperature and pressures up to 6 GPa. Specifically, XANES measurements on CMAS + Zr show a pressure-dependent shift in relative peak heights between 0-6 GPa. These data have been compared to glasses of the same composition annealed just above the glass transition by piston-cylinder experiments [1] and suggest that quenching glasses under these conditions can retain speciation information.

[1] Burnham A.D., Le Losq C, O'Neill H.S. & Wykes J.L. (2019) Goldschmidt Abstracts, 2019 436