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On the electronic structure of REE bearing crystalline compounds and silicate glasses determined by RIXS

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Core level spectroscopy has gained interest in earth science in the last decade because it provides deep insight into the electronic structure of elements in mineral phases and thus insights to their coordination environment. The electronic structure of rare earth elements (REE) is mainly determined by the interactions between electrons in the localised 4f and in the broad 5d bands. This is probed by L-edge XANES spectroscopy where the main absorption edge arises from 2p to 5d dipole transitions, whereas the fine structure at energies well above the edge is related to scattering of the created photoelectron by neighbouring atoms. In resonant inelastic x-ray scattering (RIXS) a core-electron is promoted to an excited state just as in XANES but also the energy dependence of the scattered or emitted photon is measured, which gives additional information about the intermediate and the final state. The final state, results in a M-core-hole whose lifetime broadening defines the energy resolution. Consequently, one may acquire the pre-edge fine structure, which corresponds to 2p to 4f quadrupolar transitions, with strongly enhanced resolution.

2p3d-RIXS spectra have been collected for the model compounds of La, Gd, Yb and for silicate glasses doped with REE (2wt% of La, Gd or Yb). Glass compositions are those of Prowatke and Klemme [1] as well as simplified natural compositions like haplogranite and haplobasalt. The RIXS spectra for both, model compounds and glasses show slightly different pre-edge features. For the model compounds the main spectral features are reproduced employing the FEFF 9 code in combination with the Missing Code [2]. For the glasses, the extracted RIXS spectra show an increase of the pre-edge intensity and a slight shift of the maximum to higher energies with increase of ASI (aluminium saturation index) or polymerisation. For Yb, these changes correlate well with changes in the coordination as observed by our EXAFS study [3]. This implies, that changes in 2p3d-RIXS may be used to trace changes in coordination of REE in silicate melts.

[1] Prowatke & Klemme (2005) Geochim. Cosmochim. Acta 69, 695 - 709.

[2] Kvashnina, Butorin & Glatzel (2011) J. Anal. At. Spectrom. 26, 1265 – 1272

[3] Simon, Wilke, Klemme & Chzernikov (2012) contribution to this conference