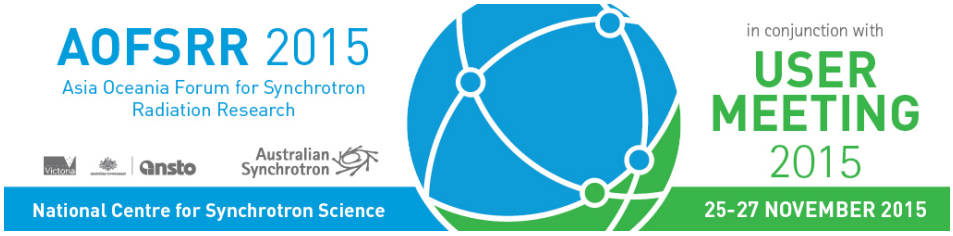

Contribution ID : **147** Type : **Oral**

Molecular-level understanding of metals in geo-fluids: combination of synchrotron-based XAS and ab initio molecular dynamics

Thursday, 26 November 2015 12:10 (20)

Aqueous fluids under wide range of T-P conditions are an essential medium for transporting metals in the Earth's crust. Metals are dissolved by forming complexes with ligands such as chloride and bisulfide, and understanding the nature and thermodynamic properties of these complexes is crucial for predicting solubilities of minerals and the mechanics of ore formation. Synchrotron based in-situ X-ray Absorption Spectroscopy (XAS) can provide insights into the molecular structures and thermodynamic properties of metal complexes in situ up to conditions beyond the critical point of water. With the advance of high-performance computing techniques, ab-initio Molecular Dynamics (MD) provides an independent means to determine the nature and stabilities of metal complexes, and particularly delivers independent crosschecks and reliable molecular models to help interpreting XAS data.

Here we demonstrate our recent studies of combining XAS and ab-initio MD in understanding the speciation, structural properties and thermodynamic stability of Zn(II)-Cl/HS, Pd(II)-Cl/HS, and Pb(II)-Cl complexes. The bond distances, coordination numbers and Debye-Waller factors for these metal complexes obtained from ab-initio MD are broadly consistent with the XAS results. The complex geometries and stoichiometries calculated from MD also agree with the XANES measurements. By combining the results from MD and XAS with existing solubility data, we recalculated the thermodynamic properties of Zn(II)-Cl/HS and Pd(II)-Cl/HS complexes, and predicted the solubilities of zinc and palladium in hydrothermal fluids with improved accuracy and reliability.

Keywords

XAS; ab initio MD; metal complexation; thermodynamics; ore deposit

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Session Classification : Energy Materials II

Track Classification : Energy Materials