

A Comprehensive Examination of Radiogenic Pb Distribution and Oxidation State in Natural Minerals and Ore by μ -XANES Spectroscopy

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The elemental and isotopic system of Pb is an important tool utilized in geochemistry, especially for use in geochronology and to quantify mass transfer processes during metamorphism and fluid-rock interaction. However, as of now, little is known about the controls that affect the distribution and coordination state of radiogenic Pb in important U-Th-Pb-bearing minerals^{1,2,3}, e.g. zircon, monazite, uraninite, and coffinite. Radiogenic Pb exists in a range of bonding environments amongst different minerals due to 1) metamictization and 2) the large difference in ionic size between oxidation states (Pb²⁺ vs. Pb⁴⁺), thus, controlling the compatibility in associated crystal structures. By understanding the distribution and speciation of radiogenic Pb, important constraints can be made on the individual reactivity of U- and Pb-bearing minerals and the chemical retentiveness of mineral phases upon metamorphism and fluid-rock interaction.

Thus, our experimental approach specifically examines a range of minerals that 1) either have U or Pb as a major element constituent and 2) have a large range in age, i.e. different concentrations of radiogenic Pb. μ -XANES spectra were collected at the Pb L_{III} edge on well-characterized grains of zircon, titanite, monazite, xenotime, uraninite, coffinite, and brannerite and also on natural ore from the Olympic Dam site in South Australia. Experiments took place at the GSE-CARS beam line at the Advanced Photon Source in Chicago.

Briefly, XANES and EXAFS spectra from high concentration Pb-bearing minerals indicate that Pb is concentrated along grain boundaries, where the speciation is dependent on mineral specificity and total radiogenic Pb concentration. With these data we can better predict the systematics of Pb distribution amongst minerals and mobility upon metamorphism and fluid-rock interaction.

¹Kramers et al., 2009, *Chemical Geology*, **261**, 4-11;

²Tanaka et al., 2010, *Physics and Chemistry of Materials*, **37**, 249-254 ³Watson et al., 1997, *Chemical Geology*, **141**, 19-31.