

Mobility of U and Th in subduction zone fluids – A synthetic fluid inclusion study

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The mobility of U in aqueous solutions at ambient conditions and in hydrothermal systems depends strongly on its oxidation state, the fluid pH, and fluid salinity, whereas the solubility of Th is less sensitive to these factors [1, 2]. Based on these observations it has been proposed that the enrichment of U over Th in arc lavas is due to the presence of oxidizing aqueous fluids in the source region. However, at present no experimental data exist at relevant pressures and temperatures. To test this hypothesis, we carried out U and Th solubility experiments at a pressure of 27.5 kbar and a temperature of 800°C. The aqueous fluid was trapped as synthetic fluid inclusions in quartz and analyzed by LA-ICPMS. The oxygen fugacity was controlled by iron-wustite, Co-CoO, Ni-NiO and Re-ReO₂ oxygen fugacity buffers, and the fluid salinity was varied between 0. and 13.7 wt% NaCl.

Our study suggests that the solubility of Th in aqueous fluids is low and independent of oxidation conditions, and increases only slightly with fluid salinity (from 4.5 to 8.9 ppm when fluid salinity changes from 0 to 13.7 wt%). In contrast, the solubility of U strongly depends on the oxidation conditions. At conditions buffered by iron-wustite U-solubility is below 2 ppm, while at conditions between Co-CoO and Re-ReO₂ buffers it increases linearly with logfO₂ from 7.0 to 20.4 ppm. Furthermore, U-solubility dramatically increases with increasing fluid salinity from 7.0 to 226.9 ppm and 20.4 to 968.0 ppm at conditions buffered by Co-CoO and Re-ReO₂, respectively.

In terms of U/Th decoupling, normalized to MORB concentrations, these results suggest that in pure aqueous fluid U can get enriched over Th by a factor of 5 to 10 if logfO₂(ΔFMQ) is between -0.5 and +3.5. For stronger U-enrichment more saline fluids might be responsible. On the other hand, Th enrichment over U in aqueous fluid can occur only in very reducing conditions where logfO₂(ΔFMQ) < -4.0, which is not representative for the source region of subduction zone magmas.

[1] Langmuir (1978) *GCA* **42**, 547-569. [2] Keppler & Wyllie (1990) *Nature* **348**, 531-533.

Lead in the core – The petrologic perspective

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Until recently, it was an accepted dogma that Pb is siderophile and that the Pb deficit in Earth's mantle has a complementary reservoir in the core. When it became clear that Pb is not siderophile and that the U-Pb model ages of the mantle significantly postdate the Hf-W ages, it was speculated that core formation involved a late sulfide melt segregation event, perhaps triggered by a giant impact, sequestering the missing Pb to the core [1].

From a petrologist's perspective, early segregation of a discrete sulfide melt is speculative at best. (1) Was there sufficient sulfide to segregate a sulfide melt? This is unlikely; the mantle after early core formation was virtually S-free (S being a siderophile element), and a Mars-sized impactor 50 Myr after solar system formation was almost certainly differentiated in an S-poor mantle and a metallic core, with S residing in its core. This is not a condition favorable to replenish proto-Earth's mantle with S. (2) Was a sulfide melt stable? Rohrbach *et al.* [2] showed that the present-day mantle below 250 km is Fe metal-saturated, hence too reduced to sustain a sulfide melt in the strict sense, i.e. with an atomic metal/S around unity. (3) Is Pb chalcophile enough such that the present-day ²³⁸U/²⁰⁴Pb (μ) of the mantle can be reproduced by sulfide segregation? Presumably not; Lagos *et al.* [3] derived D_{Pb} (sul/sil) of 2.5 ± 0.8 (IW-1), and the Ds published by Wood *et al.* [4] are not different when recast to a relative f_{O2} reasonable for a sulfide melt to be stable (> IW-2). Hence, the D values derived by experiment are two orders too low in order to reproduce the U/Pb ratio and ²⁰⁷Pb/²⁰⁶Pb isotope signature of Earth's mantle by sulfide segregation.

We cannot specify where the Pb deficit of the mantle is to be sought but it is unlikely to reside in the core. The broad coincidence of the U-Pb model ages of the mantle with the age of the Moon may suggest that Pb was lost from the mantle by high-temperature degassing from a magma ocean, and removed from Earth via impact erosion.

[1] Wood & Halliday (2005) *Nature* **437**, 1345-1348.

[2] Rohrbach *et al.* (2007) *Nature* **449**, 456-458. [3] Lagos *et al.* (2008) *Nature* **456**, 89-92. [4] Wood *et al.* (2008) *Earth Planet. Sci. Lett.* **269**, 326-336.