The mobility of U and Pb in cooling aqueous fluid evolving from reduced to oxidized state

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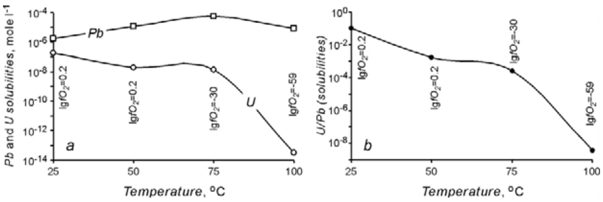
Solutions that have deposited low-temperature hydrothermal minerals within the vadose zone of Yucca Mountain, Nevada were likely evolving from low to high oxidation states as the temperature of fluids decreased with time (from about 85 to less than approximately 30-50ºC). Knowledge of solubilities of Pb and U in such solutions may prove important in interpreting the results of the U-Pb radiometric dating of the subject minerals. The "HCh" code (Shvarov, 1999) was used for thermodynamic modeling of the solution-mineral equilibria in the H-O-C-Ca-Si-U-Pb system. The roles of *T*, *Eh*, *pH*, and pCO_2 were examined.

The modeled evolution path was: from water equilibrated with silica, and dissolved CH_4 and CO_2 at equilibrium ($\lg fO_2$=-59) at *T* = 100 ºC, through methane-free water ($\lg CO_2$=-3.5 $\lg fO_2$=-30) equilibrated with calcium carbonate at *T* = 75ºC, to oxygenated water ($\lg CO_2$=-3.5 $\lg fO_2$=0.2) buffered by calcium carbonate at *T* = 50 and 25ºC. The path depicts evolution of a hypothetical mineral-forming fluid, early portions of which represent deep-seated fluid injected into the vadose zone, after which intermixing with more-oxygenated aquifer waters and cooling occurred.



The Figure *a* shows that the solubility of Pb after a one-order of magnitude increase related to the transition from silicate- to carbonate-equilibrated fluid, steadily decreases with cooling of the fluid. Solubility of U drastically increases as the fluid evolves from hot reduced to cool oxidized one. Respectively (Figure *b*), the U/Pb solubility ratio increases by some 8 orders of magnitude.

Solubility of Pb in modeled system varies between $1.8\cdot 10^{-6}$ and $5.5\cdot 10^{-5}$ mole$\cdot$l^{-1} (0.4 to 11 ppm). The early reducing fluids are thus expected to carry Pb derived from deep levels of the crust, whereas late oxidized fluids would carry Pb from shallower aquifer rocks. By contrast, solubility of U in early reduced fluids is so low ($3\cdot 10^{-14}$ mole$\cdot$l^{-1} or $7.2\cdot 10^{-6}$ ppb) that little U from the deep levels is expected to be brought. At U-solubility as high as $1.9\cdot 10^{-7}$ mole$\cdot$l^{-1} (45 ppb), late oxidizing fluids would be capable of mobilize U from shallow aquifer-hosting rocks.

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"Plum-cake" subcontinental mantle beneath SE Alps as resulting from the geochemistry of mantle xenoliths

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Preliminary geochemical and petrological data on the Tertiary ultramafic xenoliths from the Veneto Volcanic Province (VVP; SE Alps, Italy) suggest the presence of a "plum-cake" mantle region at P ≅ 1.5-2.5 Gpa and T ≅ 1250 °K (spl-lherzolite field). Major elements geochemistry of the spl-harzburgite and -lherzolite xenoliths reflects a depleted mantle source. Nevertheless, the VVP xenoliths are also characterized by "cryptic metasomatism", with varied enrichment in LREE, K, Rb, Sr, and P, indicating a re-fertilized mantle source after the depletion episode(s). Their PM-normalized spiderdiagrams are comparable to those observed for the VVP alkaline basalts having a plume-related origin (HIMU-DM; Macera et al., 2002). Disregarding their mineralogy, the VVP xenoliths do not show uniform Nb and Ta anomalies, suggesting variable interactions with crustal material at depth in the mantle. Finally, the VVP mafic xenoliths display a large spread in incompatible trace elements ratios, testifying to the heterogeneity of their mantle source. These features may be explained in terms of a plume-induced metasomatism (HIMU-type mantle upwelling) on a variably depleted subcontinental lithosphere (DM) beneath SE Alps, where a not perfectly homogenized crustal component was also present. We suggest that plume material has fed the VVP upper mantle starting at least 65-60 Ma ago. Different textures shown by the VVP xenoliths, ranging in general from protogranular in the Adige Valley outcrops to porphyroclastic in the Lessini area, may be a useful tool to estimate width and intensity variation of the shallow expression of mantle diapirism beneath SE Alps at Tertiary time.

References

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