

H₂O-CO₂ solubility in mafic melts

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Water and carbon dioxide are the two most abundant volatile species in volcanic gases. Accurate laws describing their solubilities in silicate melts are therefore crucial to understand volcanic degassing and interpret melt inclusion entrapment depths. We present here new experimental data on H₂O-CO₂ solubility in mafic melts with variable chemical compositions (alkali basalt, lamproite and kamaufugite) that extend the existing database. We show that potassium and calcium-rich melts can dissolve ~ 1 wt% CO₂ at 3500 bar and 1200°C, whereas conventional models predicts solubilities of 0.2-0.5 wt%, under similar P-T conditions. These new data, together with those already existing in the literature, stress the fundamental control of melt chemical composition on CO₂ solubility. We present a H₂O-CO₂ solubility model for mafic melts, which employs simplified concepts of gas-melt thermodynamics and accounts for the combined effects of melt chemical composition and structure. The model is calibrated on a selected database consisting of 270 experiments with 43 different mafic compositions. The statistic analyses of the experimental data indicate that the structure and the chemical composition of the melt play a fundamental role in CO₂ solubility in mafic melts, whereas H₂O solubility is negligibly affected by melt composition and structure. CO₂ solubility mainly depends on the amount of non-bridging oxygen per oxygen (NBO/O) in the melt, but the nature of the cation bounded to NBO is also critical. Alkalis (Na+K) bounded to NBO result in a strong enhancement of CO₂ solubility, whereas Ca has a more moderate effect. Mg and Fe bounded to NBO have the weakest effect on CO₂ solubility. Other structural parameters, such as the aluminous index ($Al_2O_3 / [CaO + K_2O + Na_2O]$), are shown significant, though not as critical as NBO/O. Finally, we modelled the effect of water on CO₂ solubility and suggest that molecular H₂O enhances its solubility, whereas hydroxyls appear to have the opposite effect. In contrast with CO₂, H₂O solubility in mafic melts shows a weak correlation with NBO/O, and is statistically independent on melt composition.

Migration of europium and uranium in opalinus clay influenced by pH and temperature

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Introduction

The development of a disposal in deep geological formations for high-level radioactive waste is a very important task for the future. The migration of radionuclides through soil is one of the critical paths from leaked stored container to the environment [1]. As metals of interest, europium as homologue of americium and uranium as principal component of nuclear fuel elements were used.

Spotlight of our investigations is the sorption of europium and uranium onto Opalinus clay. To get a first understanding for the different sorption processes we used batch experiments. During this experiments we varied the conditions like pH and temperature.

Results

The sorption of the metal ions is strongly pH-dependent. At pH<7 the Eu sorption decreases strongly. The uranium sorption decreases strongly between pH 6 and 9. An explanation afford the formation of the neutral Ca₂UO₂(CO₃)₃ species, which doesn't sorbed onto the clay [2, 3]. The necessary carbonate and calcium was dissolved out of the Opalinus clay. For the investigations synthetic porewater was used. It has an ionic strength of 0.4 M and the concentration of cations is more than 7g·L⁻¹. These porewater cations influence the sorption of europium in a pH range <7 significantly. Due to the presence of competing cations the sorption of Eu is inhibited. The uranium sorption is nearly independent from the competing cations. The influence of pH and temperature is significant higher. With an increasing temperature from 298 to 333 K the sorption of the metals increases, too. Investigations at different temperatures allow it to calculate the entropies and enthalpies for the reactions with uranium and europium derived from Van't Hoff plots.

[1] Wang *et al.* (2011) *J. Radioanal. Nucl. Chem.* **287**, 231–237. [2] Liu *et al.* (2005) *Environ. Sci. Technol.* **39**, 4125–4133. [3] Meleshyn *et al.* (2009) *Environ. Sci. Technol.* **43**, 4896–4901.