

Modelling phase behaviour in the geological storage of carbon dioxide

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Global warming is currently accepted as a key issue facing man. One potentially viable solution is carbon dioxide sequestration in shallow aquifers [1] where carbon dioxide is expected to exist as a supercritical phase above an underlying water phase [2]. It is essential therefore that this binary phase system is fully understood. One technique is to use trace inert proxies (i.e. noble gases) which can coexist as solute particles within both phases, the partitioning of which is affected by the extent of phase interactions (groundwater contact) and the magnitude/rate of carbon dioxide dissolution. These proxies can therefore be used to yield essential information on these processes [3].

It is however imperative that noble gas partitioning between the two phases is well constrained. We are constructing a Gibbs-Ensemble Monte Carlo simulation to model this binary phase system which will include noble gas tracers to generate the partitioning coefficients for all noble gases for the wide range of conditions expected in storage sites. This simulation simultaneously samples the molecular configurations in both water-rich and carbon dioxide-rich phases. Particles are exchanged between the two systems. At steady state the two phases are at thermodynamic equilibrium and the corresponding compositions and overall densities can be calculated. Models selected for water [4] and carbon dioxide [5] have already been extensively tested across the temperatures and pressure ranges of interest (40-150 Bar, 320-360 K) with average simulated densities being within 1.1% and 5% respectively of experimental values and thus are considered sufficiently accurate thermodynamically for incorporation into the model. Further ground-truthing for the model is being provided by lab experiments under simulated shallow aquifer conditions. We present the current progress in creating this model and discussion of the challenges which have arisen as a result of modelling such a complex system.

[1] IPCC (2005) [2] Holloway & Savage (1993) *Energy Convers. Mgmt* **34**, 925-932. [3] Ballentine & Burnard (2002) *RiMG* **47** 481-538. [4] Berendsen *et al.* (1987) *J. Phys. Chem* **91**, 6269-6271. [5] Zhu *et al.* (2009) *Chin. J. Chem. Eng.* **17**, 268-272.

Mantle heterogeneity constraints from abyssal peridotite sulfide Pb and Os isotopic compositions

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Convection over the history of the Earth has led to multiple cycles of mantle depletion and enrichment, which are preserved as ancient chemical compositional anomalies in the oceanic mantle. Studies of long-lived radiogenic isotopes in basalts, such as Pb, have indicated significant mantle heterogeneity in both the depleted upper mantle and the mantle source of ocean island basalts. The basalt data array defines a ~2 Ga age that has been interpreted as recording mixing of ancient subducted slabs with depleted mantle.

In contrast to basalts, abyssal peridotites from oceanic ridges have the potential to constrain the sub-kilometer lengthscale of mantle variability. Analysis of Pb isotopes in peridotites has been limited by the very low concentration of Pb in these residues of mantle melting. We have employed a technique for the determination of Pb and Re-Os isotopes and concentrations in peridotite sulfides using a modified version of the Re-Os technique for sulfide inclusions in diamonds.

The Pb and Re-Os isotopic composition of 21 sulfide grains were determined for abyssal peridotites from the Gakkel and Southwest Indian Ridges. Concentrations of Pb and Os in sulfides are correlated, with a correlation coefficient of ~0.8. The relatively lower mantle normalized concentrations of Pb (27, for an average of 4 ppm) with respect to Os (470, for an average of 1.6 ppm) indicates that Pb is much less compatible in sulfide during mantle melting than previously thought. Sulfide Pb concentrations are so low that sulfides cannot be the main reservoir of mantle Pb, storing only ~2%, with the remainder hosted in silicates.

Sulfide Pb isotopic compositions extend to unradiogenic values that plot to the left of the geochron and cover a wider range than associated basalts. Pb and Os isotopic compositions are correlated, also with a coefficient of ~0.8. The Re-Os data in sulfides fall along a 2 Ga model age, similar to the age given by the Pb-Pb system for the same sulfides. Multiple grains from the same peridotite have different isotopic compositions and elemental concentrations. Taken together, these results indicate that the mantle is heterogeneous down to the sub-sample lengthscale, with the Pb-Os correlation requiring that these heterogeneities were created by prior ridge melting events at ~2 Ga.