Diamond formation by acidity changes during fluid-rock interactions in the upper mantle

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The traditional picture of diamond formation from CHON fluids in the upper mantle is inadequate. CHON fluids are modelled as molecular fluids representing a mixture of simple gas molecules. The model fluids contain only a small number of species (e.g. CO_2, CH_4, H_2, H_2O and N_2), there no ions, and fO_2 is the only link to the silicate rocks in which the fluids occur [1]. As a consequence, the only way diamond can precipitate from such a fluid is by redox changes. However, real fluids in the upper mantle exist in a silicate environment. Aqueous fluids at elevated pressures contain large amounts of dissolved silica, neutral species, ions, and metal complexes in various degrees of polymerization [2]. Furthermore, recent fluid inclusion studies, experiments and theoretical calculations have indicated substantial carbonate mineral solubilities at high pressures and that aqueous carbonate ions may play a role in the transport of carbon in upper mantle aqueous fluids [3]. A more comprehensive approach to fluid speciation in the upper mantle is needed.

By including HCl as a component, reactions involving scores of aqueous ions and metal complexes in upper mantle fluids can be considered and the model fluid chemistry is coupled to its silicate rock environment. Models of diamond formation can then include changes in acidity as well as redox. Here the new Deep Earth Water (DEW) model [4] was used to calculate equilibrium constants for incorporation in data files for aqueous speciation and chemical mass transfer calculations modelling diamond formation in cold subduction zones for the first time.

A fluid equilibrated with a model mineral assemblage representing the peridotitic mantle part of a subducting slab was reacted with the magnesite-bearing metabasaltic upper part of the slab at 600 °C and 5.0 GPa. Silicate-fluid reactions such as pyrope altering to chlorite consumed Mg^{2+} from the aqueous phase causing a drop in pH that favoured alteration of magnesite to diamond. In this way, acidity changes driven by water-silicate rock reactions can be a way to form diamond.

[1] Zhang & Duan (2009), *GCA* **73**, 2089-2102. [2] Manning *et al* (2010), *EPSL* **292**, 325-336. [3] Pan *et al* (2013), *PNAS*, **110**, 6646-6650; Frezzotti *et al* (2011), *Nature Geo.*, **4**, 703-706; Facq *et al* (2014) *GCA* (doi 10.1016/j.gca.2014.01.030). [4] Sverjensky *et al* (2014) *GCA*, **129**, 125-145.