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Coupling between magnetite formation and H_2 production in aqueous metamorphic fluids: new experimental constraints

B. Malvoisin (1), V. Milesi (2), F. Brunet (1), J. Carlut (2), O. Vidal (1), and F. Guyot (2)

(1) ISTerre - CNRS, Université Grenoble 1, France (fabrice.brunet@ujf-grenoble.fr), (2) Université Paris Diderot, IPGP-CNRS, Paris, France

The reduction of natural hydrous fluids in metamorphic environments is often accounted for by the presence of carbonaceous matter which is considered as a fO_2 buffer. However, reducing conditions can also be generated by the oxidation of Fe^{2+}, a major element in rock systems. The oxidation of ferric iron in the course of mineralogical reactions is driven by the stability of magnetite, $Fe^{2+}Fe^{3+}_2O_4$, which can form in a wide range of P-T-fO_2 conditions. The hydrothermal decomposition of olivine into serpentine + magnetite + brucite is an example of H_2-producing reaction in relation to iron oxidation from ferric (olivine) to ferrous (magnetite). Such reaction is believed to account for the formation of dihydrogen and, indirectly, hydrocarbons in some hydrothermal fields of the Mid-Atlantic ridge[1]. The reducing conditions imposed by serpentinization reactions have also recently been invoked to account for the reduction of Ca-carbonate into carbonaceous matter[2] in oceanic metasediments. The hydrothermal decomposition of siderite, $FeCO_3$, is another example of H_2-producing reaction related to the formation of magnetite[3]. Note that $FeCO_3$ decomposition also produces CO_2 which, in turn, reacts with H_2 to produce hydrocarbons. In both cases of reaction, the aqueous fluid composition changes as the reaction proceeds (reaction path) and the fO_2 in the solution cannot be simply derived by considering oxygen buffering reaction(s) as it is the case when carbonaceous matter is present. It is therefore important to monitor these reactions on a time-resolved basis. San Carlos olivine of calibrated size was reacted with pure water in sealed gold capsules at 50 MPa in a range of temperatures. Reaction progress has been monitored directly on the encapsulated samples using the magnetic properties of magnetite, the time-resolved production of which was determined by measuring the saturation remanent magnetization (Jrs) and the saturation magnetization (Js, collected at the end of the run). Such magnetic monitoring is sensitive to variations in magnetite content at the hundred of ppm level. It can therefore allow a precise quantification of the reaction kinetics[4] as a function of grain size and temperature and hydrogen production can be estimated using additional thermochemical modeling. In parallel, the gas produced by the reaction and trapped within the capsule was analyzed using gas chromatography (GC) when the capsule was opened, i.e. at the end of the experiments. GC analysis of H_2-bearing gases produced at 50 MPa and 300˚C was also tested on siderite reacted under hydrothermal conditions in order to investigate the kinetics of $H_2 – CO_2$ chemical interactions under pressure and temperature.

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