Experimental tests on achieving equilibrium in synthetic fluid inclusions: Results for scheelite, molybdenite, and gold solubility at 800 °C and 200 MPa

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ABSTRACT

Synthetic fluid inclusions formed in high P-T experiments, which are subsequently analyzed with LA-ICP-MS, enable us to collect thermodynamic data to constrain metal transport in aqueous fluids as well as partitioning of metals between coexisting phases. The most essential prerequisite for such studies is to ensure that equilibrium conditions between liquid and solid phases are reached prior to the formation of synthetic fluid inclusions in the host mineral. Various methods have been proposed by different authors to achieve this goal, but to this point our knowledge on the best approach to synthesize equilibrated fluid inclusions under constrained pressure, temperature, and compositional (P, T, and X) conditions remains poor. In addition, information on the time needed to reach equilibrium metal concentrations in the fluid as well as on the timing of the onset of fluid inclusion formation in the host mineral are scarce.

The latter has been tested in a series of time-dependent experiments at 800 °C and 200 MPa using scheelite (CaWO₄), molybdenite (MoS₂) and metallic gold as dissolving phases and using different approaches to optimize the formation of equilibrated fluid inclusions. Both fO₂ and fS₂ were fixed during all experiments using the pyrite-pyrrhotite-magnetite buffer (PPM). As an intermediate in situ quenching of the sample charge plays an important role in the synthesis of fluid inclusions, we further tested the efficiency of such an intermediate quench for re-opening fluid inclusions formed at 600 °C and 200 MPa. Our results reveal that fluid inclusions start forming almost instantaneously and that equilibrium between fluid and solid phases occurs in the timescale of less than two hours for molybdenite and gold up to ca. 10 h for scheelite. The best approach to synthesize equilibrated fluid inclusions at 800 °C was obtained by using an intermediate quench on a previously unfractured quartz host. Experiments at 600 °C showed similar results and illustrate that this should be the method of choice down to this temperature. Below 600 °C pre-treatment of the quartz host (HF etching and/or thermal fracturing) becomes important to produce large enough fluid inclusions for the analyses via LA-ICP-MS and special care must be taken to prevent premature entrapment of the fluid.

Fluids with 8 wt% NaCl in equilibrium with scheelite, molybdenite and gold at 800 °C and 200 MPa have concentrations of ca. 7300 ppm W, 1300 ppm Mo, and 300 ppm Au, respectively, which is in good agreement with results from other studies or extrapolation from lower temperatures. It can be concluded that the formation of synthetic fluid inclusions from an equilibrated fluid is possible, but different experimental designs are required, depending on the investigated temperature. In general, dissolution of solid phases seems to be much faster than previously assumed, so that experimental run durations can be designed considerably shorter, which is of great advantage when using fast-consuming mineral buffers.

Keywords: Synthetic fluid inclusions, equilibrium, scheelite solubility in aqueous fluid, molybdenite solubility in aqueous fluid, gold solubility in aqueous fluid

INTRODUCTION

Magmatic and hydrothermal fluids play a crucial role in the formation of ore deposits, because they are the main transporting agents controlling mobilization and selective concentration of elements in the Earth’s crust and, among others, metals of economic interest. Fluids trapped as fluid inclusions in magmatic and hydrothermal minerals provide direct insight into the genesis and evolution of natural fluids at the conditions of mineral growth or fracture healing. The correct reconstruction of natural conditions, e.g., during ore formation, however, requires accurate and systematic quantification of the evolution of the fluid composition as a function of major parameters that control the properties of magmatic or hydrothermal systems. The main approach applied for such quantifications at high pressure (P) and temperature (T) is the experimental synthesis of fluid inclusions that were described in the pioneering studies of Roedder.