INVESTIGATING THE FORMATION CONDITIONS OF OLIVINES THROUGH LOW-TEMPERATURE HYDROTHERMAL PROCESSES USING MAGNESIUM-BEARING AMORPHOUS SILICATES. J. L. Maxwell1, E. Dobrica1, J. A. Nuth1, and A. J. Brearley1. 1Hawai‘i Institute of Geophysics and Planetology, School of Ocean, Earth Science, and Technology, University of Hawai‘i at Mānoa, Honolulu, HI (jmax@hawaii.edu). 2Solar System Exploration Division, Code 690, NASA Goddard Space Flight Center, Greenbelt MD 20771 USA. 3Department of Earth and Planetary Sciences, MSC03-2040, 1University of New Mexico, Albuquerque, NM.

Introduction: Iron-rich olivine is one of the major minerals in the matrices of unequilibrated ordinary (UOCs) and carbonaceous (CV, CK, CO) chondrites whose petrologic type is >3.1 [1-3]. Previous experiments provide clear evidence that hydrothermal growth of elongated, FeO-rich olivine can occur during thermal metamorphism in the presence of a fluid (220°C, 0.4 W/R mass ratio, 6 days, [4]), as described previously in meteorites and supported by thermodynamic calculations [5-7]. So far, hydrothermal alteration experiments have been successful at forming FeO-rich olivines with the compositions and textures observed in the matrices of chondrites only at 220°C [4]. Therefore, understanding the formation conditions of FeO-rich olivines at lower temperatures, durations, and water-to-rock (W/R) mass ratios, and using different reactants remains a key problem to understanding the effects of hydrothermal alteration on chondrite matrices.

We have investigated the formation conditions of olivines through hydrothermal processes using Mg-bearing amorphous silicates compared to previous studies that used Fe-bearing amorphous silicate [4].

Methods and results: We performed five hydrothermal alteration experiments in an attempt to synthesize FeO-rich olivines at low-temperature (200°C). We used four different reactants for these experiments: (1) amorphous silicates (~3.5 mg), (2) iron metal powder (~11 mg), (3) Fe-metal foil, and (4) different amounts of deionized water (18 megoHms) to simulate W/R mass ratios of 0.2, 0.5, and 1. We chose Mg-bearing amorphous silicates as the precursor material to understand the role of Mg in the formation of olivines. The reason is that most FeO-rich olivine in primitive chondrites contains magnesium (FeO-rich olivine: Fe50–100), and only rare, pure fayalite crystals were identified. The amorphous silicate grains were synthesized experimentally by disequilibrium condensation [8]. The composition of the silicate grains, which condense from the Mg-SiO-H: [9] vapor, is MgSiO (~10 at.% Si, and ~30 at.% Mg). Additionally, we added iron metal powder (spherical particles, <10 μm) to the experiments because previous studies [10] showed that fayalite forms from a FeO-rich fluid that is SiO2-poor and very reduced (log fO2) less than QFM conditions of ~35.7 at 300°C. According to thermodynamic calculations, fayalite could form at W/R ratios lower than 0.2 W/R [7]. However, the estimated W/R mass ratio for chondritic parent bodies range from ~0.1 to 0.6 (or even higher for CI chondrites), which is significantly lower than the solar value of 1.2 W/R mass ratio [11]. One experiment was performed using an iron foil (thickness 0.25 mm) as an iron source instead of the iron powder (Fig. 1). The advantage of the iron foil is that we know the exact amount of iron in contact with the other reactants, instead of a random distribution of iron powder, as for the other hydrothermal experiments.

We loaded the reactants in gold (Au) capsules, which were welded at room temperature and pressure in a glove box purged with nitrogen. We tested whether the experiments were successful or not (e.g., leaks) by weighing the capsules before and after the experiments. The capsules were placed in Parr bombs (reactor vessels) and heated in at 200°C for 14 days. The powder inside the gold tube was deposited onto carbon tape and was studied by scanning electron microscopy (SEM) on a FEI Quanta 3D Dualbeam® field emission gun (FEG) SEM/FIB operating at 30 kV. The powder inside the gold tube was deposited onto carbon tape. This powder and the surface of the iron foil were studied using the FEI Helios 660 dual-beam focused ion beam SEM (SEM/FIB instrument at the Advanced Electron Microscopy Center (AEMC) at the University of Hawai‘i at Mānoa. One transmission electron microscopy (TEM) section of a selected region containing the new phases in the experiment performed on the iron foil was prepared using the FIB technique.

New minerals were formed in all the experiments analyzed in this study, including those performed at low W/R mass ratio (0.2 W/R ratio). Magnetite (Fe³⁺Fe²⁺O₄), brucite [Mg(OH)₂], and greenalite [(Fe²⁺,Fe³⁺)₂Si₂O₅(OH)₄] were identified by quantitative EDS analysis. Figure 1 shows the distribution of reactants and products identified at the surface of the hydrothermal experiment performed on the iron foil. Brucite crystals have an acicular, platy habit (Fig. 1b-c). They vary in length from a few hundred nanometers up to 3 μm and in width from 80 nm to 300 nm. The iron foil was homogeneously transformed into iron oxide down to about 150 nm depth (Fig. 1d), which is expected since the foil was in contact with the aqueous fluid added on top of the Mg-bearing amorphous silicate. Additionally, euhedral magnetite grains were identified at the surface of the iron foil (Fig. 1a). All the new products formed during the experiment suggest that
the iron from the foil and magnesium from the amorphous silicate were mobile during the experiment. In the FIB section made in the experiment performed on the iron foil, we observed that areas that contain silicon are rare. Only one amorphous region (300 x 440 nm in size) with a stoichiometry similar to that of olivine (~2.0 Fe+Mg/Si ratio) was identified in the FIB section analyzed (Fig. 1d, am. Mg,Fe silicate fluffy region). However, no crystalline olivine was observed in the experiment performed on the iron foil. We suggest that the low abundance of silicon in the reactants (~0.3 Si/Mg) could be an important factor in the formation of olivine during the hydrothermal reactions in these experiments. Previous studies show that the chemical evolution of the hydrothermal fluid as alteration reactions proceed could trigger the formation of either fayalite or phyllosilicates depending on the Si/Fe ratios in the fluid [4]. Therefore, the Si/Fe,Mg ratio in the aqueous fluid will control the phase and composition of the reaction products, as indicated by [4]. Future studies will focus on detailed TEM characterization of the reactants and reaction products formed in the powder experiments that used the same Mg-bearing amorphous silicate.


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Figure 1. a-b) Backscattered (BSE) and secondary (SE) electron micrographs of the hydrothermal experiment products: amorphous (am.) silicate, brucite, and iron oxide formed at the surface of the iron foil. The black line shows the region where the FIB section was prepared for TEM studies. c-d) Bright-field TEM and dark-field STEM micrographs showing the texture, distribution, and mineralogy of the hydrothermal experimental products observed in the iron foil experiment. The amorphous Mg,Fe silicate material has a stoichiometry similar to that of olivine (~2.0 Mg+Fe/Si).