

AQUEOUS ALTERATION EXPERIMENTS OF CHONDRULE ANALOGUE AND IRON SULFIDE MIXTURE WITH H₂O-CO₂ FLUID. K. Watanabe¹ and H. Isobe¹, ¹Dept. Earth Envi. Sci., Grad. Sch. Sci. Tech., Kumamoto Univ., Kurokami, Kumamoto, 860-8555, Japan. e-mail: isobe@sci.kumamoto-u.ac.jp

Introduction: CM carbonaceous chondrites contain various minerals produced by aqueous alteration processes. The aqueous alteration processes involved in the CM chondrite materials took place on precursor materials in parent bodies [1] or in the solar nebula before accretion to the parent bodies [2]. The precursor materials of aqueous alteration should be chondrite materials including chondrules. Chondrules consist of phenocrysts of silicate minerals, dominantly olivine, and glassy mesostasis phase. Mesostasis is the most sensitive material to aqueous alteration, and it is not observed in highly altered meteorites [1]. Serpentine and saponite have been reported as aqueous alteration products replacing for mesostasis [3-5]. The typical composition of precursory mesostasis is, however, different from that of phyllosilicate in respect of Mg/Fe ratio. It is suggested that aqueous alteration of mesostasis progresses with exchanging elements with constituents other than chondrules.

Opaque phases including Fe-Ni metal and sulfides are relatively sensitive to aqueous alteration and possible constituent involving Mg/Fe exchange during aqueous alteration. Alteration products from the opaque phases have been reported as magnetite, tochilinite and cronstedtite [4-6]. Sulfate phases also occur in carbonaceous chondrites. Fe sulfide may be a source of sulfur to various sulfur-bearing minerals by oxidative aqueous alteration processes.

Olivine, mesostasis glass and iron sulfide phases should be essential precursors of the aqueous alteration processes in the early solar system. Carbonate minerals with various chemical compositions also occur in carbonaceous chondrites [6-9]. Carbon dioxide may have important role in oxidative aqueous alteration processes. In this study, we carried out aqueous alteration experiments of mixed powder of olivine, artificial mesostasis glass and pyrrhotite with H₂O-CO₂ fluid.

Experimental: Mixed powder of the natural olivine, synthesized glass material and natural pyrrhotite was used as the starting material. We used olivine crystals of Fo₉₀ representing Mg-rich olivine phenocrysts in chondrules. For mesostasis glass synthesis, we adopted averaged composition of mesostasis glass of chondrules in the Allende meteorite (Taura, S., personal communication) as anhydrous precursor. For the iron sulfide phases, we used natural pyrrhotite (Fe_{1-x}S) as an alternative to troilite in chondritic meteorites.

The starting material was mixed so that the bulk ratio of Si-Mg-Fe, major component, to be the solar

abundance. About 5mg of the starting material was put into Au capsules with 5mg of distilled water. And Ag oxalate was sealed in the capsules so that H₂O/CO₂ molar ratio to be 9/1 in the fluid. Silver oxalate decomposes to Ag and CO₂ at about 250 °C. Sealed Au capsules were heated at 250 °C for one hour to decompose Ag oxalate.

Experimental temperatures are from 100 to 300 °C every 50 °C. Run durations are 4, 8, 16 or 32 weeks at 100 and 150 °C, and 2, 4, 8, 16 or 32 weeks at 200 and 250 °C, and 2, 8 or 32 weeks at 300 °C. Run products were observed with a scanning electron microscope (SEM) and analyzed with an energy dispersive X-ray spectroscopy (EDS).

Results and Discussion: Secondary sulfides and carbonate phases occur in the experiments at 100 °C for over 8 weeks. The secondary sulfide was observed as the rim of the pyrrhotite grains. EDS analysis revealed that the secondary sulfide is pyrite. Carbonates in the run products at 100 °C show layered structure (Fig. 1). The layered structure has sharp boundary. The core of the layered carbonates is siderite and the rim is magnesite (Fig. 2). Amorphous silica phases occur over 16 weeks run duration.

In the experiments of 150 °C, pyrite occurs in the run products of all run durations. Carbonates with three different compositions in Fe-Ca-Mg system occur in the run products over 8 weeks. Those are Fe-rich carbonate, Ca-rich dolomitic carbonate and Ca carbonate and show layered structure or are isolated grains. Phyllosilicate occur in the run products for over 16 weeks as a minor phase.

In the run products of 200 °C, pyrite, carbonates and phyllosilicates occur. Calcium carbonates can be found in the run products less than 4 weeks, and Ca carbonates, dolomites and magnesites show layered, zoned or isolated textures over 8 weeks. Phyllosilicate abundance is lower than carbonate in samples of 32 weeks. The composition of the phyllosilicate corresponds to Ca-bearing saponite, and it is getting more Mg-rich with longer run durations.

In the run products of 250 °C, a minor amount of secondary sulfides with identical composition to pyrrhotite occur. In addition, Fe oxides exist in run products of 32 weeks. These Fe-bearing phases indicate oxidative condition over 250 °C. Phyllosilicates and carbonates were abundantly observed. The composition of the phyllosilicates changes from saponite-like within 16 weeks to intermediate between saponite

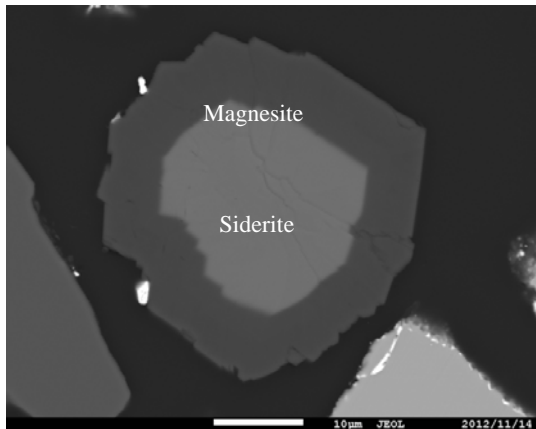


Fig. 1 Layered structure of carbonate grains in the run product of 100 °C, 32 weeks.

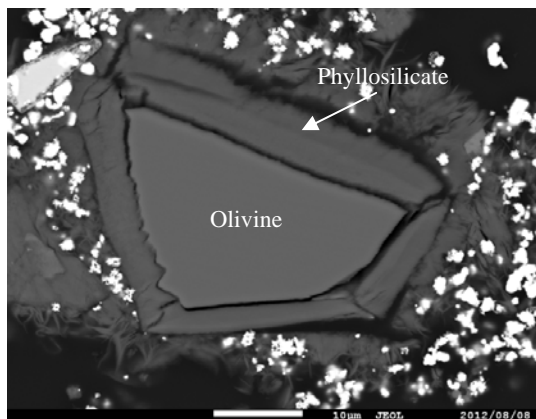


Fig. 3 Phyllosilicate formed around olivine grain in the run product of 300 °C, 32 weeks.

and serpentine at 32 weeks. The composition of the carbonates was Ca carbonate in the sample of 2 weeks and Ca carbonate, dolomite or magnesite in the samples over 8 weeks.

In the samples of 300 °C, abundant phyllosilicate and minor carbonate were found (Fig. 3). Phyllosilicate has saponite-like composition in the sample of 2 weeks and it is getting Si-Al-poor with longer run durations. Only Ca carbonates occur in the samples with all run durations.

The results of the experiments show that the composition of carbonates depends on temperatures and run durations (Fig. 2). Fe-rich carbonates occur at relatively low temperature from 100 °C to 150 °C. In the samples at 100 °C, no Ca carbonates observed. Ca carbonates occur over 150 °C. Mg-bearing carbonates occur under 250 °C. Experiments at 100 °C, evidence for dissolution of glass is not observed. Glass phase with mesostasis composition is an only source of Ca in the starting material. Occurrence of Ca carbonates formation may depend on dissolution of the glass phase depending on grain size distribution. Pyrrhotite

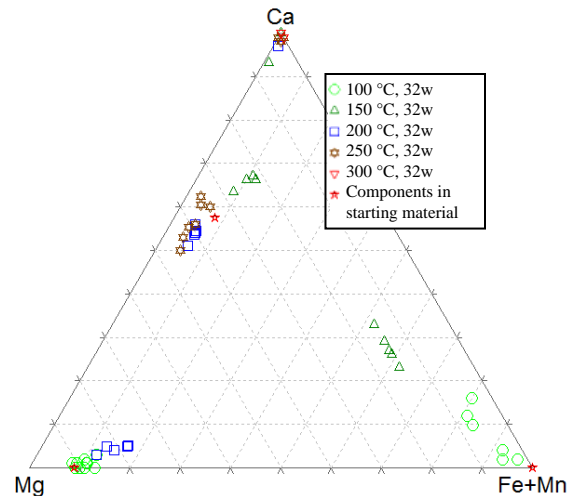


Fig. 2 Ca-Mg-(Fe+Mn) ternary diagram of carbonates in the run products.

w: weeks

and olivine dissolved gradually at all experimental temperatures and durations. In the run products, Fe is distributed to carbonate (100-150 °C), sulfide (all temperatures), phyllosilicate (over 150 °C) and oxide (over 250 °C). Magnesium is distributed to carbonate (100-250 °C) and phyllosilicate (over 150 °C). Distribution of Mg and Fe in aqueous alteration derived phyllosilicate and carbonate can be excellent indicator to constrain alteration conditions of carbonaceous chondrites.

Phyllosilicate occur in run products over 150 °C for long run durations and it tends to be Si-Al-poor with temperature increasing. At temperatures over 150 °C, glass phase with high Si-Al content is more vulnerable than olivine with Si-poor and Al free composition. Experiments over 200 °C for long run durations, glass phase dissolved completely. Dissolution kinetics depending on temperature conditions can control Si and Al concentrations in aqueous solution. Phyllosilicate mineral species may indicate aqueous alteration conditions on CM chondrite parent bodies.

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