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Hydrothermal and heating experiments: Implications for formation process of nepheline in carbonaceous chondrite

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Nepheline ($NaAlSiO_4$) and sodalite ($Na_4Al_3Si_3O_{12}Cl$) are widely distributed in chondrules and matrices of CO/CV chondrites (Kimura et al. 2014; Matsumoto et al. 2014). Because of the low condensation temperatures of such Na-rich minerals, these are likely not primary phases condensed from the solar nebula, but are secondary phases altered from melilite or plagioclase. According to previous petrographic studies (e.g. Russell et al., 1998; Tomeoka and Itoh, 2004), nepheline and sodalite are considered to form by hydrothermal processes on their parent body. Because of the high volatility and high mobility, Na element is potentially a powerful indicator reflecting aqueous environments of the chondrite parent bodies. In this study, we performed hydrothermal alteration experiments to understand the formation process of Na-rich minerals in chondrite parent bodies.

We used i) synthetic melilite (gehlenite, $Ca_2Al_2SiO_7$) + SiO_2, and ii) natural plagioclase (An_{48}) as starting materials. Four reaction solutions of 1N (mol/l)-NaOH (pH 0), H_2O (pH 7), 0.1N-NaOH (pH 13) and 1N-NaOH (pH 14) were reacted with three different water/rock ratios (10, 100 and 1000). Na^+ concentration in all solutions was maintained at 1 mol/l by addition of NaCl. Hydrothermal alteration experiments were performed with a PTFE reaction vessel loaded into steel autoclave and were carried out at temperature at 200 ℃ and pressure of 1.5 MPa for run duration time of 168 hours. The recovered samples were analyzed by XRD, SEM, TEM, and TG-DTA.

In the experiments using melilite+SiO_2 with W/R = 100 and pH 7-14 solutions, hydrogrossulor ($Ca_3Al_2[SiO_4]_{1.6}[O_4H_4]_{1.3}$) formed as a secondary phase with low SiO_2 contents, while analcime ($NaAlSi_2O_6H_2O$), nepheline-hydrate ($NaAlSiO_4H_2O$), basic-cancrinite ($Na_8Al_6Si_6O_{24}[OH]_2\bullet 2H_2O$) and tobermorite ($Ca_5Si_6O_{16}[OH]_2\bullet 4H_2O$) formed with high SiO_2 contents. Although no secondary phase was observed at high W/R (=1000), analcime and basic-cancrinite occurred at low W/R (=10). Under the pH 0 solution, melilite was dissolved and no secondary phases formed. In the experiments using plagioclase with W/R = 100 and pH 0-7 solutions, no secondary phases formed. On the other hand, under pH 13-14 solutions, analcime and nepheline-hydrate formed with W/R = 10 and 100.

A TG-DTA of nepheline-hydrate at heating rate 5 ℃/min in air showed that the dehydration started at ~100 ℃ and finished at ~800 ℃. From the DTA and XRD data of this condition, nepheline-hydrate is considered to be transformed into nepheline at ~800 ℃. From the TG-DTAs of nepheline-hydrate with different heating rates, we estimated that nepheline-hydrate could alter to nepheline for at least ~10^3 years at 400 ℃. A TG-DTA of analcime at heating rate 2 ℃/min in air showed that dehydration started at ~120 ℃ and finished at ~500 ℃. The XRD data showed analcime is transformed into nepheline and amorphous material below 1000 ℃, although we failed to estimate the required time of the transformation because of the DTA curves of analcime were too complicated.

The present results show melilite and plagioclase are easily altered into hydrous minerals like nepheline-hydrate and analcime in Na^+-rich high alkali solutions. Because the experimental condition demonstrated in the present study seems to be comparable to actual carbonaceous chondrite parent body, we infer that nepheline in carbonaceous chondrite formed initially as intermediate products like nepheline-hydrate and analcime during aqueous alteration, and these hydrous minerals was subsequently dehydrated to nepheline.

Keywords: nepheline, melilite, aqueous alteration, hydrothermal experiments, dehydration heating experiments, carbonaceous chondrite