

Formation process of nepheline in carbonaceous chondrites: Reproductive experimental approach

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In CO/CV chondrites, nepheline is widely distributed in the Ca, Al-rich inclusions (CAIs), the chondrule mesostases and the matrices. Recent petrographic studies have shown abundant evidence suggesting that the nepheline is secondary phases altered from melilite and/or plagioclase by a chemical reaction with Na-rich fluids, and the reaction (Na-metasomatism) occurred in the chondrite parent bodies. Because of the high volatility and mobility, Na is potentially a powerful indicator reflecting aqueous environments of the parent bodies. While many petrographic studies of natural meteorites had made considerable progress on revealing behaviors of Na-metasomatism, quantitative uncertainty for the aqueous condition still remains. In the present study, we performed a number of hydrothermal experiments using melilite and plagioclase as starting materials, and made crystal identifications, micro-textural observations, chemical analyses and thermal analyses for run products. The purpose of this study is constraint of environment in carbonaceous chondrite parent bodies by revealing process of nephelinization.

We used i) synthetic melilite ($\text{Ca}_2\text{Al}_2\text{SiO}_7$) + SiO_2 , and ii) natural plagioclase ($\text{Na}_{0.5}\text{Ca}_{0.5}\text{Al}_{1.5}\text{Si}_{2.5}\text{O}_8$) as starting materials. The reaction solution of 1N (mol/l)-HCl (pH 0), H_2O (pH 7), 0.1N-NaOH (pH 13) and 1N-NaOH (pH 14) were prepared for the hydrothermal experiments with different water/rock ratios (4.67, 46.7 and 467 ml/g). Na^+ concentration in all solutions is maintained at 1 mol/l by addition of NaCl. Hydrothermal alteration experiments were performed with a PTFE reaction vessel loaded into steel autoclave at 200 °C and 1.5 MPa for run duration time of 168 hours. The recovered samples from the hydrothermal and thermal experiments were analyzed by XRD, SEM, TEM, and TG-DTA. The experiments using melilite without SiO_2 under the conditions of WR 46.7 and pH 7-14 showed that hydro-grossular ($\text{Ca}_3\text{Al}_2[\text{SiO}_4]_{1.5}[\text{OH}]_6$) forms as a secondary altered phase. Under the same WR and pH conditions, as the proportion of SiO_2 increases, various Na-rich zeolitic materials were observed in the run products including analcime (Anl, $\text{NaAlSi}_2\text{O}_6\text{H}_2\text{O}$), nepheline-hydrate (NephH, $\text{NaAlSi}_4[\text{H}_2\text{O}]$) and hydroxy-cancrinite (Canc, $\text{Na}_8\text{Al}_6\text{Si}_6\text{O}_{24}[\text{OH}]_2[\text{H}_2\text{O}]_2$). At the high W/R (=467) condition, no secondary phase was observed, while Anl and Canc occur at the low W/R (=4.65) under pH 14. In the experiments using plagioclase with high W/R ratios and pH 0-7 solutions, no secondary phases formed. Anl and NephH formed under pH 13-14 conditions. At low W/R ratios, secondary phases were identified as Anl, and NephH.

The TG-DTA and XRD analyses for nepheline hydrate showed that NephH is transform to nepheline at 796-841 °C at the heating rate ranging of 1-14 °C/min. From the analyses of the reaction kinetics, we estimated that NephH alter to nepheline for $\sim 10^5$ years at 515 °C. Although we failed to analyze the reaction kinetics for Canc and Anl because of their complicated DTA curves, thermostatic experiments suggested that both material also change to nepheline at 760 °C (Canc) and 800 °C (Anl).

The present results suggest that melilite and plagioclase are easily altered into zeolitic materials in Na^+ -rich high alkali solutions. The present experimental condition demonstrated in the present study seems to be comparable to actual carbonaceous chondrite parent body. Thus, we infer that nepheline in carbonaceous chondrite formed initially as zeolitic materials during aqueous alteration, and they were subsequently dehydrated to nepheline.

Keywords: nepheline, chondrite, aqueous alteration