## Aqueous interaction between olivine and organic compounds to imply spatial distribution of organics in meteorites

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## Introduction

Carbonaceous meteorite is relatively enriched in volatile materials including water and organic matter, recording the chemical evolution of organic compounds in the primordial solar system. The organic compounds could be absorbed on and/or intercalated in clay minerals formed from anhydrous silicates during aqueous alteration in a meteorite parent body. However, it is not well understood how organic compounds interacted with anhydrous silicates during the alteration. The spatial distribution of alkylimidazole ( $C_nH_{2n-2}N_2$ ) and alkylpyridine ( $C_nH_{2n-5}N$ ) was distinct in the Murray meteorite (CM2) depending on their species and homologues (Naraoka & Hashiguchi, 2018). The result may infer aqueous interaction between mineral and organic compounds in the meteorite parent body. In this study, we examined interactions between the CHN compounds and olivine, which is one of the major anhydrous silicates in chondritic meteorites.

## Materials and Methods

The powder of San Carlos olivine (particle size;  $~\mu$ m to  $~hundred ~\mu$ m) was packed in a stainless-steel tube (1.7 mm i.d. ×5.0 cm in length) to prepare a column for liquid chromatography. A mixed standard solution of four CHN compounds; i) imidazole, ii) 2-methylimidazole, iii) 2-methylpyridine, and iv) 3-methylpyridine was injected into the column connected to a mass spectrometer. The interaction between the organic compounds and olivine was evaluated by comparing their retention times. The same chromatography without olivine powder was also examined as a reference. The following three experiments (Exp. 1 to 3) were performed depending on pH and temperature. Exp.1: Neutral eluent conditions (H<sub>2</sub>O) at 40°C. Exp. 2: Effect of eluent pH using CH<sub>3</sub>COONH<sub>4</sub> buffer (pH 8.95), NH<sub>3</sub>aq (pH 10.57), and formic acid solution (pH 2.57) at 40°C. Exp. 3: Effect of temperatures; H<sub>2</sub>O eluent at 20°C and 30°C, CH<sub>3</sub>COONH<sub>4</sub> buffer (pH8.92) and NH<sub>3</sub>aq (pH10.35) at 20°C.

## **Results and Discussion**

Exp. 1: The four compounds had an identical retention time in the absence of olivine powder. In contrast, the retention times by the olivine column ranged in the order; iii) 2-methylpyridine  $\sim$  iv) 3-methylpyridine < i) and indicate < ii) 2-methylpinidazole. This order is correlated with the acid dissociation constant (pKa) of each compound, suggesting that the CHN compounds are present as cations (e.g.  $C_3H_5N_2^+$  for imidazole) interacting more strongly with the oxyanions of silicic acid in olivine.

Exp. 2: Under alkaline conditions, the retention times of all compounds decreased with the increase of pH. This is probably because the cation strength of the CHN compounds became weaker by decreasing the amount of protons in the eluent. Under acidic conditions, the retention times of imidazoles were shorter and those of methylpyridines were longer. The mechanism of this phenomenon is under consideration.

Exp. 3: Under neutral conditions, the retention times of all compounds increased with the decrease of the temperature (i.e.  $20^{\circ}C > 30^{\circ}C > 40^{\circ}C$ ). The compounds retained longer in the column at  $40^{\circ}C$  showed larger increase in retention times than those shorter retained at lower temperature. Under alkaline conditions, the retention times did not change significantly with the decrease of temperatures probably due to the weak strength of ionic bonds even at lower temperature. The results indicate that the CHN

compounds have a stronger ionic association with olivine under neutral conditions because it is difficult for the ions on the surface of olivine to transfer into the mobile phase at lower temperatures. These observations suggest that olivine of the carbonaceous meteorites could affect the interaction between organic matter and water and might have influenced the spatial distribution of organic compounds on the meteorite parent bodies.

Keywords: Carbonaceous meteorite, Olivine, Organic compounds, Aqueous alteration, Ionic interactions