

In-situ experiment of aqueous alteration for amorphous silicate in the MgO-SiO₂ system

*Yohei Igami¹, Tomoya Yamazaki², Yuki Kimura², Akira Tsuchiyama¹

1. Graduate School of Science, Kyoto University, 2. Institute of Low Temperature Science, Hokkaido University

Introduction

Based on infrared astronomical observations, >98% of interstellar silicate dusts are considered to be amorphous (Kemper, 2004), and these amorphous silicates are believed to be a main precursor material of the solar system. Moreover, the amorphous silicates are abundantly observed from cometary dust and primitive carbonaceous chondrite (e.g., Keller and Messenger, 2012; Leroux et al., 2015). On the other hand, silicates in most carbonaceous chondrite has more or less experienced the aqueous alteration, even if they are classified as the most primitive one (e.g., Abreu and Brearley, 2010). The aqueous alteration of amorphous silicate is considered to be a process which played an important role in the material evolution of primitive solar system.

For understanding the aqueous alteration of amorphous silicate, experimental studies have conducted in addition to detailed observation of carbonaceous chondrite. For example, Nakamura-Messenger et al. (2011) conducted hydrothermal experiments for GEMS (glass embedded with metal and sulfides) in cometary dust with aqueous solution of various pH, and observed phyllosilicate similar to those observed in carbonaceous chondrite. However, their samples should have experienced not only hydrothermal (aqueous alteration) process but also subsequent drying process. In a strict sense, they could not indicate whether their observed phyllosilicates had formed by the aqueous alteration process or the drying process. In order to obtain accurate information from carbonaceous chondrites, the two processes are required to be discussed separately. In this study, therefore, we conducted in-situ observation for aqueous alteration of amorphous silicate by establishing the method effective to analyze wet samples, and investigated time evolutions of aqueous alteration.

Methods

Samples were amorphous silicates in the MgO-SiO₂ binary system, with the composition of MgO/SiO₂ = 2.02 (sample name: Fo-am) and 1.15 (En-am). They were very fine spherical particles with ~70nm in diameter synthesized by thermal plasma torch (Nisshin Flour Milling Inc.) by Imai (2012, Doctoral thesis). These were mixed with distilled water, and analyzed at room temperature by transmission electron microscopy (TEM) and X-ray diffraction (XRD) experiment. For the TEM observation, the wet samples were sealed by using dedicated holder, Poseidon (Protochips, ILTS, Hokkaido Univ.), and time evolution of particles for 1-24 hours were observed. For the XRD experiments, the wet samples were covered by polyimide film, and the appearance and time evolution of XRD peaks for 0-48 hours were observed. After all the experiments, the samples were dried in vacuum, and then re-analyzed by TEM and XRD to investigate the change by the drying process.

Results and Discussion

As the results of TEM observations, particles gradually expand in water, although fine textures are unclear because of strong electron scattering by water. The observations after drying shows that most of Fo-am particles lose original shape, and surrounded by fibrous phyllosilicates. On the other hand, En-am particles relatively keep original spherical shape with rough surfaces, and have a few phyllosilicates with very bad crystallinity.

As the results of XRD experiments, amorphous peaks of all the samples show time evolution in shift toward the position of SiO₂ amorphous. This implies that Mg²⁺ is preferentially dissolved from the amorphous silicate. Appearance and time evolution of phyllosilicate peaks are also detected from both

samples, but their intensity-increasing speeds are different. The speed in Fo-am at 1-5 hours after mixing water are particularly large. This indicates that much ions are dissolved to make supersaturated solution temporarily, and then phyllosilicates rapidly precipitated. The samples after vacuum drying shows increasing the phyllosilicate peaks, in particular from Fo-am. This seems to be caused by precipitation from the aqueous solution by the drying process. The peak position of this phyllosilicate is slightly different from that of the phyllosilicate formed by aqueous alteration process.

From the above, it is revealed that phyllosilicates can be formed by both at the aqueous alteration and the drying process. Unless this is taken into account, one may lead to even inaccurate conclusions. However, our XRD results imply that these two types of phyllosilicates have slightly different features crystallographically. There is a possibility that careful analysis of phyllosilicate in carbonaceous chondrites can constrain their formation history.

Keywords: amorphous silicate, aqueous alteration, chondrite, in-situ experiment