

## Formation of metastable lingunite

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Lingunite (hollandite-type NaAlSi<sub>3</sub>O<sub>8</sub>) has been frequently found in shocked meteorites with other high-pressure minerals (Liu and El Goresy, 2007). According to the laser-heated diamond anvil cell (LHDAC) experiment by Liu (1978), following the decomposition of albite (NaAlSi<sub>3</sub>O<sub>8</sub>) into jadeite (NaAlSi<sub>2</sub>O<sub>6</sub>) plus silica (SiO<sub>2</sub>) at 2-3 GPa, these phases recombine to form lingunite in the range of pressure between 21 and 24 GPa, and then it decomposes again into calcium ferrite-type NaAlSiO<sub>4</sub> plus stishovite at pressures above 24 GPa. Similarly, Tutti (2007) observed lingunite as a minor phase at 21-23 GPa and 2273K using LHDAC. In contrast to these LHDAC studies, high-pressure experiments using multi-anvil type (MA) apparatus revealed that the maximum solubility of NaAlSi<sub>3</sub>O<sub>8</sub> component in hollandite structure is limited to ~50 mol% at 14-25 GPa and 1073-2673K (Yagi et al., 1994, Liu, 2006) and NaAlSi<sub>3</sub>O<sub>8</sub> lingunite is not stable at least up to 2273K (Akaogi et al., 2010). This contradiction has not been solved yet, which makes it difficult to understand the shock conditions for the presence of lingunite in shocked meteorites.

To investigate the lingunite puzzle, we focused on the formation process of lingunite by conducting time-series experiments. We performed high-pressure experiments at 18-27 GPa and 1073-2023K using both LHDAC and MA apparatus. Powders of natural albite, oligoclase and labradorite are used as starting materials. Existing phases were identified by X-ray diffraction method.

The quenching experiments using MA apparatus revealed that lingunite does not form in 5 min, but forms in 60 min as a single phase from oligoclase at 20 GPa and 1473K. In situ X-ray diffraction study indicated that oligoclase becomes amorphous with increasing pressure and temperature. At 22 GPa and 1473K, lingunite first crystallizes from the complete amorphous oligoclase in 100 sec, and it decomposes into stishovite and CAS phase in 60 min. These results suggest that lingunite forms as a metastable phase by solid-state reaction after the amorphization of oligoclase, which might have also occurred with maskelynite in shocked chondritic meteorites (Tomioka et al., 2000). In contrast, lingunite was not observed when albite and labradorite were used as starting materials. The amorphization pressure increases with increasing albite component. The pressure condition for complete amorphization of albite is higher than that for the lingunite formation. No lingunite observed from the albite sample in this study implies that the complete amorphization is required for the metastable formation of lingunite by solid-state reaction. In the case of labradorite, lingunite was not formed even after the complete amorphization. This is consistent with the observation that lingunite with labradorite composition in martian shocked meteorites crystallized not by solid-state reaction but from plagioclase melt (e.g., El Goresy et al., 2013).