

## Diamond formation from magnesite in the presence of C-H-O fluid under high pressure and temperature

OHFUJI, Hiroaki<sup>1\*</sup>; IKAWA, Syunta<sup>1</sup>; KIMURA, Tomoaki<sup>1</sup>

<sup>1</sup>Geodynamics Research Center, Ehime University

Diamond formation in the Earth's mantle is believed to be a metasomatic process by fluid/melt with peridotitic and/or eclogitic rocks (e.g. Shirey et al., 2013). The ubiquitous occurrence of nano- to micro fluid inclusions in natural diamond and the result of recent experimental studies (e.g. Sokol et al., 2009) suggest that the presence of C-H-O fluid, which consist of a mixture of H<sub>2</sub>O, CO<sub>2</sub>, CH<sub>4</sub>, etc. plays an important role in diamond forming reaction. The composition of C-H-O fluid depends largely on the surrounding oxygen fugacity. Taking into an account the oxygen fugacity values estimated from diamond-bearing xenoliths, the most major components of such fluids at the depth of 150-200 km are likely H<sub>2</sub>O and CH<sub>4</sub> (Shirey et al., 2013). In the deeper mantle, the relative proportion of CH<sub>4</sub> is expected to increase with depth (Frost and McCammon, 2008). In the present work, we studied the diamond formation through the reaction between magnesite and reduced C-H-O fluid at high pressure and high temperature.

A series of high-pressure and high-temperature experiments (up to 30 GPa, 2000K) were performed by using laser-heated diamond anvil cell (LH-DAC). A small fragment of natural pure magnesite (either polycrystalline aggregate or single crystal) is loaded together with stearic acid (C<sub>18</sub>H<sub>36</sub>O<sub>2</sub>) or pure CH<sub>4</sub> in a sample chamber drilled in a pre-indented rhenium gasket. Upon laser-heating, the stearic acid is expected to produce CH<sub>4</sub>-rich fluid as a result of decomposition reaction (C<sub>18</sub>H<sub>36</sub>O<sub>2</sub> → 8CH<sub>4</sub> + 2H<sub>2</sub>O + 10C). A dual-beam CO<sub>2</sub> laser system (at Ehime Univ.) or a dual-beam fiber laser system (at SPring-8) was used for the heating. In-situ XRD observation at high pressure and high temperature was conducted in BL10XU of SPring-8. The samples after recovery were examined by XRD, Raman spectroscopy and scanning and transmission electron microscopies.

In-situ XRD observation showed that magnesite, MgCO<sub>3</sub> decomposes into periclase, MgO, brucite, Mg(OH)<sub>2</sub> and diamond in the presence of CH<sub>4</sub>-rich fluid at pressures above 18 GPa and at ~2000K. Brucite is likely to be a metastable product, because the formation is usually observed at the beginning of the reaction and its relative proportion decreases with time by being taken over by the formation of periclase. The formation of nanocrystalline diamond was also observed in the samples recovered from the experiments. Although diamond was produced by the decomposition reaction of stearic acid through the equation described above, the reduction of magnesite by CH<sub>4</sub>-rich fluid must also have resulted in diamond formation. Inclusion studies of super-deep (lower-mantle derived) diamonds reported that the most dominant mineral phase found in such diamonds is periclase (up to 60%). However, this is not consistent with the result of high-pressure experimental studies using pyrolytic composition, where Mg-perovskite (bridgmanite) is the most dominant. Our result implies that some of the periclase inclusions solely found in those diamonds might have formed as a result of the reaction of Mg-rich carbonate with reduced C-H-O fluid. This means that the information obtained from diamond inclusion does not necessarily reflect the bulk composition of the lower mantle. Furthermore, the formation of periclase at shallower condition (at 18 GPa) suggests that such formed periclase inclusions in diamond may not be used as indicator of lower-mantle origin.

Keywords: Diamond, Magnesite, C-H-O fluid, High pressure and high temperature