

## Measuring osmium isotopic composition of natural polycrystalline diamond (carbonado) and implications for its origin

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The origin of natural polycrystalline diamond, carbonado, has long been enigmatic. Carbonado is characterized as high porosity, no genetic relations to kimberlites, light carbon isotope ratio, and lack of mantle-derived mineral inclusions. Based on these observations, several hypotheses about the origin of carbonado have been proposed: transformation of subducted organic carbon into diamond in a cold slab (Robinson, 1978); shock metamorphism of organic carbon by meteorite impact (Smith and Dawson, 1985); radiation-induced diamond formation by spontaneous fission of uranium in crustal environment (Ozima *et al.*, 1991); formation in an interstellar environment (Garai *et al.*, 2006); crystallization from C-O-H fluid in cratonic upper mantle (Ishibashi *et al.*, 2012). However, no conclusive evidence has been provided to settle a controversy about the origin of carbonado. In this study, we first tried to measure Os isotopic composition of carbonados collected from placer deposits in the Central African Republic in order to identify its origin.

Natural samples have a wide variety of Os isotopic ratios,  $^{187}\text{Os}/^{188}\text{Os}$ , depending on their origin because  $^{187}\text{Re}$ , the parent nuclide of radiogenic  $^{187}\text{Os}$ , is a mildly incompatible element during mantle melting whereas Os is a strongly compatible element.  $^{187}\text{Os}/^{188}\text{Os}$  ratio of upper continental crust ranges from 1.0 to 1.4 (Peucker-Ehrenbrink and Jahn, 2001), whereas that of primitive upper mantle is about 0.13 (Meisel *et al.*, 1996). Os isotopic ratio of the micro diamond crystal itself can reflect the environment where diamond grains crystallized. Carbonado is a porous aggregate of micrometer-size diamond crystals and original chemical characteristics of the grain boundaries could be heavily altered after the diamond growth.

This study was designed to determine Os isotopic ratios within diamond crystals of carbonados and in the grain boundaries separately. Two-step sample chemical leaching was carried out by Carius tube method (Shirey and Walker, 1995). First, carbonado samples were crushed to submillimeter grains and were sealed in a Carius tube with spike solutions and inverse aqua regia ( $\text{HCl} + 3 \text{HNO}_3$ ). The solution was heated at 220 °C for 24 hours. This procedure was for extract Os in the grain boundaries. Second, the residue of solid samples was heated in a vacuum chamber to convert diamond to graphite. A graphitized sample was decomposed in acid solution in the same way as the first leaching process. The second process was to extract Os within diamond grains. Osmiums in the both solutions were purified with the solvent extraction (Cohen and Waters, 1996) and microdistillation (Roy-Barman, 1993). Osmium isotopic compositions of the samples were determined using thermal ionization mass spectrometry (TIMS). Blank levels of Carius tubes and inverse aqua regia solutions prepared from several chemical reagents were checked. As a result, a quartz glass tube was found to have the lowest blank level compared with other glass tubes made from borosilicate glass.

In the presentation, we will report preliminary results of Os isotopic ratios of the carbonado, which have the potential for a decisive evidence to close the debate on the origin of carbonado.

Keywords: carbonado, TIMS, Os isotope, diamond