

The carbon isotope composition of natural SiC (moissanite) from the Earth's mantle: New discoveries from ophiolites

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ABSTRACT

Moissanite (natural SiC) has been recovered from podiform chromitites of several ophiolite complexes, including the Luobusa and Donqiao ophiolites in Tibet, the Semail ophiolite in Oman and the United Arab Emirates, and the Ray-Iz ophiolite of the Polar Urals, Russia. Taking these new occurrences with the numerous earlier reports of moissanite in diamondiferous kimberlites leads to the conclusion that natural SiC is a widespread mineral in the Earth's mantle, which implies at least locally extremely low redox conditions. The ophiolite moissanite grains are mostly fragments (20 to 150 μm) with one or more crystal faces, but some euhedral hexagonal grains have also been recovered. Twinned crystals are common in chromitites from the Luobusa ophiolite. The moissanite is rarely colorless, more commonly light bluish-gray to blue or green. Many grains contain inclusions of native Si and Fe–Si alloys (FeSi_2 , Fe_3Si_7).

Secondary ion mass spectrometric (SIMS) analysis shows that the ophiolite-hosted moissanite has a distinctive ^{13}C -depleted isotopic composition ($\delta^{13}\text{C}$ from -18 to -35% , $n = 36$), much lighter than the main carbon reservoir in the upper mantle ($\delta^{13}\text{C}$ near -5%). The compiled data from moissanite from kimberlites and other mantle settings share the characteristic of strongly ^{13}C -depleted isotopic composition. This suggests that moissanite originates from a separate carbon reservoir in the mantle or that its formation involved strong isotopic fractionation. The degree of fractionation needed to produce the observed moissanite compositions from the main C-reservoir would be unrealistically large at the high temperatures required for moissanite formation. Subduction of biogenic carbonaceous material could potentially satisfy both the unusual isotopic and redox constraints on moissanite formation, but this material would need to stay chemically isolated from the upper mantle until it reached the high- T stability field of moissanite. The origin of moissanite in the mantle is still unsolved, but all evidence from the upper mantle indicates that it cannot have formed there, barring special and local redox conditions. We suggest, alternatively, that moissanite may have formed in the lower mantle, where the existence of ^{13}C -depleted carbon is strongly supported by studies of extraterrestrial carbon (Mars, Moon, meteorites).

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1. Introduction

Moissanite, the natural form of silicon carbide, is named after Nobel Laureate Henri Moissan (chemistry, 1906), who reported SiC grains in the Canyon Diablo meteorite (Moissan, 1904). Although listed as “doubtful or incompletely described” in an overview of meteorite mineralogy by Mason (1967), moissanite has since been confirmed to exist in many meteorites, some extrasolar (e.g., Alexander, 1993 and references therein). Natural SiC has been reported as a rare accessory phase in a wide variety of terrestrial rocks since the 1960s (reviewed in Lyakhovich, 1980). The geologic community was at first slow to accept

these findings because in most cases the moissanite was recovered from mineral separates and there was a possibility of contamination with synthetic SiC. Definitive proof of a natural origin came with the discovery of moissanite inclusions in diamond from kimberlites (Moore and Gurney, 1989; Otter and Gurney, 1989; Leung, 1990; Gorshkov et al., 1997) and such occurrences are now known from most cratons (South Africa, China, Siberia, North America, Brazil and Australia). In-situ occurrences of moissanite and/or intergrowths of moissanite with other natural minerals have also been documented in retrograde eclogites and serpentinites from the Dabie-Sulu belt in China (Qi et al., 2007; Xu et al., 2008) and in volcanic rocks (Bauer et al., 1963; Di Pierro et al., 2003; Zhang et al., 2006).

Probably the most frequently reported terrestrial host rock for moissanite is diamondiferous kimberlite, but is not clear if this has

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geological significance or if it simply reflects the fact that mineral concentrates from kimberlites are routinely scrutinized in detail. The compilation of moissanite occurrences by Lyakhovich (1980) shows an astonishing variety of host rocks including many of non-mantle origin like granites, salt deposits, limestones and bauxite. Accidental contamination by synthetic SiC may explain some of these occurrences, but it seems reckless to disclaim all of them, and in the case of moissanite from limestone (Gnoevaja and Grozdanov, 1965), a later study confirmed the occurrence using acid digestion where contamination can be ruled out (Shiryaev et al., 2008a). A surface origin of some moissanite is attested by its occurrence at meteorite impact craters (Ries: Hough et al., 1995; Berringer, Odessa: Miura et al., 1999) and in burn sites from forest fires (Sameshima and Rodgers (1990).

Here we report important new discoveries of moissanite from another geologic setting, namely, magmatic chromitite bodies in the upper mantle section of ophiolites. The first report of moissanite and diamond occurring in the ophiolite setting (Armenian Transcaucasus) is by Gevorkyan et al. (1976). Bai et al. (2000) found moissanite and microdiamond in podiform chromitites from the Luobusa ophiolite, Tibet, along with a number of other minerals which are interesting for their exotic stability conditions, requiring ultra-high pressure and/or very reducing environments (e.g., kyanite, coesite, metal alloys, native elements: Bai et al., 2000; Robinson et al., 2004; Yang et al., 2007a). Motivated by the Luobusa finds, we have now recovered moissanite from podiform chromitites and/or their peridotitic host, from the Dongqiao ophiolite in Tibet, the Semail ophiolite in Oman and the Ray-Iz ophiolite in the Polar Urals of Russia (Fig. 1). In an effort to understand the occurrence of SiC in ophiolites, and to compare these grains with those in kimberlites, we determined C-isotope compositions of the ophiolite samples by in-situ secondary ion mass spectrometry (SIMS). This investigation was motivated by the C-isotope studies of moissanite from diamondiferous kimberlites by Leung et al. (1990) and Mathez et al. (1995), which found ^{13}C -depleted compositions relative to the composition of most diamonds and other mantle-derived materials thought to represent the dominant mantle carbon reservoir ($\delta^{13}\text{C}$ near -5% ; see Deines, 2002; Cartigny, 2005). It turns out that the moissanite from all ophiolite localities sampled thus far have ^{13}C -depleted isotopic compositions; and indeed, this appears to be a characteristic feature of all terrestrial moissanite that has been studied to date ($\delta^{13}\text{C}$ from -18 to -35% , $n = 72$ from 12 occurrences).

2. Geologic background

The Luobusa ophiolite lies in the Yarlung–Zangbo suture in southern Tibet about 200 km ESE of Lhasa (Fig. 1). It is a fault-bounded slab, 1–

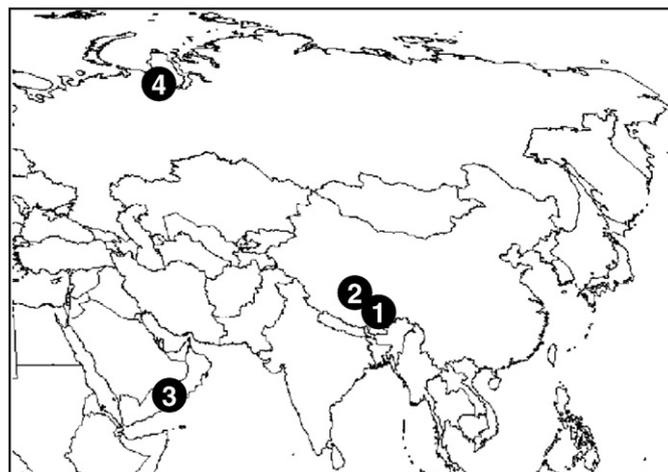


Fig. 1. Location map of the ophiolites sampled from this study; 1: Luobusa, 2: Dongqiao, 3: Semail, 4: Ray-Iz.

2 km thick, consisting largely of harzburgite that is believed to have formed at a mid-ocean ridge at ~ 177 Ma and then been modified by boninitic melts above an intraoceanic subduction zone at ~ 126 Ma (Zhou et al., 1996; Malpas et al., 2003; Zhou et al., 2005; Yamamoto et al., 2007). Luobusa hosts the largest active chromite mine in China. Numerous lenses and pods of chromitite occur in the harzburgite where they are surrounded by envelopes of dunite formed by melt–rock reaction in the upper mantle (Zhou et al., 1996, 2005). Our samples from orebodies 31 and 74 contain 80–90 vol.% chromite with Cr# ($100 \times \text{Cr}/(\text{Cr} + \text{Al})$) of 79–93 (Zhou et al., 1996). The Luobusa chromitites have become well-known for the occurrence of ultra-high pressure minerals such as diamond and coesite (IGCAGS, 1981; Robinson et al., 2004; Yang et al., 2007a).

The Dongqiao ophiolite is located in the Bangong Lake–Nujiang suture zone in central Tibet. It consists of an 18-km-long block of harzburgite hosting lenses and pods of chromitite with compositions similar to those of Luobusa (Cr# of 70–80: Shi et al., 2007). Amphibolites at the base of the ophiolite have $^{40}\text{Ar}/^{39}\text{Ar}$ ages of 175–180 Ma (Zhou et al., 1997), suggesting formation in the Middle Jurassic. The bulk sample from which SiC was separated originated from orebody Nr. 10.

The Semail ophiolite in Oman and the United Arab Emirates forms part of the Tethyan ophiolite belt that extends from Europe through the Middle East to Tibet (e.g. Searle and Cox, 1999). Podiform chromitites occur in both the mantle transition zone and deeper in the mantle section (e.g., Ahmed and Arai, 2002; Rollinson, 2008), where they form small pods and lenses. We collected samples from two localities in the Semail ophiolite. Sample OM-2 comes from the Shamis 2 orebody in Wadi Rajmi and consists of massive chromite (Cr# = 64, Rollinson, 2008) with interstitial olivine and orthopyroxene, minor clinopyroxene and amphibole. Sample UAE-1 is massive chromite ore with minor olivine taken from a 1- to 1.5-meter-thick chromitite lens in harzburgite from the defunct mine at Wadi Al-Hayl.

The Ray-Iz ophiolite occurs at the NE end of the Paleozoic Voikar-Syninsk ophiolite belt (Savelev and Savelyeva, 1977; Garuti et al., 1999; Yang et al., 2007b). A summary of age data in the ophiolite belt (Savelyeva et al., 2007) indicates early Ordovician to late Cambrian ages. The Ray-Iz ophiolite covers over 400 km² and consists chiefly of tectonized harzburgite and dunite in the upper mantle section, with minor amounts of gabbroic cumulates above the Moho. More than 200 podiform chromitite orebodies occur, hosted chiefly in the harzburgite–dunite complex and consisting of high-Cr (Cr# = 74–86) chromite (Pervozhikov et al., 1990).

2.1. Formation of podiform chromitites

Any attempt to explain the occurrence of moissanite in podiform chromitites must take into account the current models for the formation of these bodies. The mineralogical and geochemical–petrological features of podiform chromitites in the Luobusa, Dongqiao, Semail and Ray-Iz ophiolites are well established from previous work cited in the descriptions above. All are hosted by harzburgite in the upper mantle section and all are associated with dunite envelopes and veins. The favored model for massive chromite formation in this setting is a sudden chromite precipitation from basaltic (or boninitic) melts triggered by melt–rock interaction that produced the dunite veins and envelopes (Zhou et al., 1996). Many of the chromitites and associated dunitites show evidence of brittle fracture, supporting the litho-stratigraphic evidence for formation at shallow depths (Zhou et al., 1996; Robinson et al., 2004). There is no doubt that in these ophiolites, the chromitite bodies accumulated at shallow mantle depths, but the occurrence of high-pressure minerals within them is highly enigmatic. The high-pressure phases like diamond or coesite (Yang et al., 2007a) presumably occur as inclusions in the chromite, and it is impossible to conceive that they could survive in a low-pressure basaltic melt crystallizing chromite, without some kind of armoring. One possibility is that the chromite

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