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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 (FeSi2, Fe3Si).

Secondary ion mass spectrometric (SIMS) analysis show that the ophiolite-hosted moissanite has a distinctive 13C-depleted isotopic composition (δ13C from -18 to -35 ‰, n= 36), much lighter than the main carbon reservoir in the upper mantle (δ13C 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).

**Keywords:** moissanite, SIMS, C-isotopes, ophiolites, Ray-Iz, Luobusa, Dongqiao, Semail
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 (Moore and Gurney, 1989; Otter and Gurney, 1989; Leung, 1990) from kimberlites, 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 it is not clear if this has 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 ultrahigh 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 ($\delta^{13}$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 ($\delta^{13}$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, 1-2 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; 2005; Malpas et al., 2003; 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 volume % chromite with Cr# (100*Cr/Cr+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 40Ar/39Ar 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 (Savel’ev and Savel’yeva, 1977; Garuti et al., 1999; Yang et al., 2007b). A summary of age data in the ophiolite belt (Savel’yeva 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# = 0.74-0.86) 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 dunites 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 grains hosting such high-pressure minerals are xenocrysts themselves and were transported by upwelling mantle and/or partial melts from much greater depths. Evidence for xenocrystic chromites was suggested based on observed Re-Os ages of chromitites that predate the host ophiolite at Dongqiao (Shi et al., 2007), and by the finding of coesite and clinopyroxene lamellae in chromite grains from Luobusa (Yamamoto et al., 2008), which requires crystallization of the host at high pressure. Clearly, this new evidence makes it easier to understand the finding of diamond and other high-pressure or chemically incompatible phases in mineral separates from these chromitites, and there may be implications for the formation of chromite ores more generally, but these are still isolated observations and there is no information yet available on how widespread xenocrystic chromite in ophiolites may be.

3 Sample descriptions
Moissanite is a rare mineral and our procedures for sampling and sample treatment were designed accordingly. Very large samples of chromite ore, 100 to 500 kg each, were collected from outcrops at mine sites. In the processing facility the samples were first cleaned, then ground to pass a 1 mm mesh sieve using machinery free of SiC components (steel jaw-crushers, steel ball or roller mills). Heavy minerals were separated using gravimetric and magnetic techniques, and moissanite was hand-picked from these separates under a binocular microscope. Mineral identification was based on optical properties and confirmed by Raman spectroscopy, SEM-EDS or X-ray diffraction. Although the studied grains were recovered from mineral separates, we are confident that the material reported is natural moissanite for the following reasons: (1) samples were collected directly from outcrop; (2) sample preparation (crushing and mineral separation) was done at different occasions in different laboratories: Freiberg, Saxony, the GFZ Potsdam and St. Petersburg for the Oman samples, and Zhengzhou, China, for Luobusa and Donqiao. Care was taken to avoid contamination during all stages of sampling and processing. The machinery was disassembled and carefully cleaned before use and none of the components used in processing contained SiC. In the case of Luobusa chromitite, a 100 kg sample of granite was processed before the chromitite in exactly the same way and went through the same mineral separation procedure but not one grain of SiC was found; (3) moissanite from Luobusa is intergrown with and/or contains inclusions of the Ca-Al phase gehlenite (Hu, 1999), which was also found along with anorthite, in moissanite from Turkey by Di Pierro et al., (2003). To the authors' best knowledge, Ca-Al silicates have never been reported from synthetic SiC.

The moissanite grains recovered in this study are mostly conchoidal fragments (20 to 150 μm), probably produced during sample preparation. Some euhedral hexagonal grains were also recovered and many of the broken grains show one or more well-developed crystal faces. Some moissanite from the Luobusa ophiolite forms twinned crystals (Fig. 2). The moissanites are rarely colorless, more commonly light blue-gray to blue or green. Electron microprobe analyses for Al, Fe, Sc, Cr using enhanced counting times to improve sensitivity failed to find significant concentrations in most grains but in other cases, Al and Fe contents of up to 300 ppm have been reported (cf. Bauer et al., 1963; Marshintsev, 1990; Mathez et al., 1995; Shiryaev et al., 2008a). Some of the moissanite grains from the Semail, Luobusa and Ray-Iz
ophiolites contain rounded inclusions of native Si and, more rarely of Fe-Si alloys (Fig. 3b,c). In our example and in other published reports, the Fe-Si phase occurs at the boundary of native Si and the SiC host (Mathez et al., 1995; Di Pierro et al., 2003; Shiryaev et al., 2008a). The Fe silicides in our example are too small for electron microprobe analysis because of beam overlap with the SiC host, but analyses of individual Fe-Si grains from Luobusa revealed Fe$_3$Si and FeSi$_2$ (Bai et al. 2000).

The association of moissanite and native Si, with or without Fe-silicides, appears to be characteristic since it has been found in most studied moissanite localities (Marshintsev, 1990; Leung et al., 1990; Mathez et al., 1995; di Pierrro et al., 2003; Shiryaev et al., 2008; Xu et al., 2008). The Si inclusions are typically round (Fig. 2), which suggest a trapped melt. If so, this places limits on the trapping conditions. The melting point of Si (1410°C at surface) decreases with pressure by about -33°/GPa (Soma and Matsuo, 1982), so Si metal should be liquid at mantle depths below about 200 km (for the continental mantle geotherm).

4. Carbon isotope compositions

4.1 Methods

Carbon isotope analyses were performed on polished, gold-coated grain mounts of moissanite using a Cameca ims6f secondary ion mass spectrometer (SIMS). The analyses employed a $^{133}$Cs$^+$ primary beam operated at 400 pA and 10 kV, and a secondary extraction voltage of -7.5 kV with an energy window of 50 eV and no energy offset. A 20 µm contrast aperture was used in conjunction with a 750 µm field aperture (60 µm field of view). The mass resolving power of M/ΔM ≈ 3700 was adequate to resolve $^{13}$C from the isobaric interference by $^1$H$^{12}$C. The conductivity of SiC made electron flooding unnecessary. The beam diameter at the sample surface was ~5 µm. Each analysis consisted of a 10 minute preburn followed by 80 cycles of the peak stepping sequence - mass 11.9 (background, 0.1 s per cycle), $^{12}$C (2 s) and $^{13}$C (5 s). A deadtime correction of 16 ns was applied to all data. SIMS analyses were performed on 2-5 grains from each of the localities, giving a total of 36 analyses (Table 2). Carbon isotope compositions are reported as $\delta^{13}$C = [$^{13}$C/$^{12}$C$_{\text{sample}}$ / $^{13}$C/$^{12}$C$_{\text{RM}}$ - 1] x 1000) relative to the PDB reference ($^{13}$C/$^{12}$C =
Instrumental mass fractionation and analytical uncertainties were monitored using two synthetic SiC reference materials: ANMH 100578 (Mathez et al., 1995), and an in-house reference prepared from 80-mesh SiC abrasive (GFZ-S113). The C-isotope composition of ANMH 100578 is slightly uncertain because the procedure used in combusting the SiC resulted in yields of less than 50% (Mathez et al., 1995). The C-isotope composition of our in-house reference material GFZ-S113 was determined at the University of Lausanne using procedures described in detail in the appendix. Yields were between 90 and 100% for each of two duplicate analyses of the GFZ-S113 material, and the resulting δ\(^13\)C values are \(-28.71\)‰ and \(-28.79\)‰. Mathez et al. (1995) noted that their C-isotope determination for ANMH 100578 could be affected by the incomplete yield of sample combustion, but the agreement of both reference materials in our study show that this is not the case (Table 1). Based on repeated analyses of the reference materials (Table 1) the internal precision of our SIMS measurements is about 1 per mil and the repeatability is about 2 per mil (1 \(\sigma\) values).

We also conducted SIMS analyses on moissanite grains separated from the Turkish volcanic pebble previously studied by Di Pierro et al. (2003). The average of 9 analyses performed on the Turkish moissanite is \(-26.7\)‰, which agrees very well with the value of \(-28\)‰ reported in the earlier study. However, the range of δ\(^13\)C values in the SiC grains is from \(-21\) to \(-31\)‰ (Table 2), and even the within-grain variations are up to 9‰. This heterogeneity went unnoticed in the earlier study because C analysis was done on an aggregate sample by conventional mass spectrometry.

### 4.2 Results

The total range of δ\(^13\)C values from ophiolite-hosted moissanite is \(-18\)‰ to \(-35\)‰ (Table 2, Fig. 3). The mean value and standard deviation of 27 analyses comprising four localities are \(-28.2\ ±\ 3.9\)‰. A comparison of results for the four ophiolite localities Ray-Iz \((-32.7\ ±\ 2.1\)‰), Luobusa \((-28.4\ ±\ 3.3\)‰),
Dongqiao (-23.5 ± 3.4 ‰) and Semail (-28.1 ± 2.6 ‰) shows considerable overlap, with only two localities: Ray-Iz and Dongqiao being statistically distinguishable. The range of C-isotope compositions within each locality varies from 5 to 9 ‰, which is larger than analytical uncertainty even at the 2-sigma level (4 ‰). Typically, only a single spot on each moissanite grain was analysed in order to maximize the sample coverage and thus we have no information on within-grain variations for most samples. Exceptions are one grain from the Semail ophiolite (UAE1-2) and one from Luobusa, which were analyzed twice and showed less than 3 ‰ variation (Table 2). In their SIMS study of mosssanite from two kimberlites in Yakutia, Mathez et al. (1995) found a range of δ13C values of about 4 ‰ for the individual localities Aihkal and Mir. Those authors mentioned a weak correlation of the C-isotope composition with color of moissanite grains, but no such correlation was observed for the ophiolite moissanite in our study (Table 2). Shiryaev et al. (2008b) determined a range of about 3.5 ‰ among different moissanite grains from Yakutia kimberlites and inferred possible growth zoning but this is not demonstrable from their data coverage. Lacking more within-grain analyses and petrographic association we can offer no comment on the significance of C-isotope variations observed our study.

The most important result in our C-isotope study is that all moissanites from the studied ophiolites are strongly depleted in 13C relative to the mantle carbon reservoir represented by diamonds, carbonatites, mantle xenoliths and CO2 emissions from basaltic volcanoes with δ13C value around -5 ‰, as reviewed by Deines (2002). Furthermore, a data compilation of published C-isotope analyses of natural moissanite from the other settings (Fig. 3) shows that all terrestrial occurrences of moissanite yet described contain carbon that is strongly depleted in 13C. As pointed out earlier (Mathez et al., 1995; Di Pierro et al., 2003), and confirmed by our study (Table 1), synthetic SiC has low δ13C values in the same range as the natural mineral. Therefore, one cannot use carbon isotope compositions alone to distinguish natural from synthetic SiC. This does not refute our claim of a natural origin for the moissanites studied, for which other evidence was described above. Note also that Leung et al. (1990) found 13C-depleted carbon (δ13C -24 ‰) in moissanite inclusions in diamond from the Fuxian kimberlites, which are undeniably natural.
The host diamonds in that example yielded $\delta^{13}C$ values of -2.9 to -4.5‰, within the “normal” mantle range.

5 Discussion

With the new occurrences reported here, moissanite is now well documented from mantle-derived rocks of both the kimberlite-lamproite association (sub-cratonic lithosphere) and the ophiolite association (shallow upper mantle). In both cases, its occurrence is enigmatic because the low redox conditions required for SiC formation seem highly incompatible with the geologic settings in which it is found. The extremely reducing environment of formation is underscored by the common presence of native Si inclusions within moissanite grains. A second enigmatic feature of moissanite from the earth’s mantle is the rather narrow range of C-isotopic composition from all occurrences yet studied, and the fact that these compositions are strongly $^{13}C$-depleted compared with most diamonds and other carbon-bearing phases known from the mantle. It may be that the unusual redox conditions and isotopic composition of moissanite are both related to how the mineral forms in Earth, but we first consider the two characteristics separately.

5.1 Redox conditions and moissanite formation

The redox conditions in the upper mantle are well established by many lines of evidence, and lie within one or two log units of the forsterite-magnetite-quartz (FMQ) buffer (see review by Frost and McCammon, 2008). Certainly in the shallow upper mantle where ophiolites are formed, particularly in suprasubduction zone settings, one can expect conditions near the FMQ buffer. Studies of mineral inclusions in kimberlitic diamonds indicate that the great majority grew at the base of thick mantle lithosphere between about 140 and 200 km depth with exceptional cases suggesting formation in the mantle transition zone (410 to 660 km) or the shallow part of the lower mantle (Stachel et al., 2005). Oxygen fugacity estimates vary but many diamonds appear to have formed at $fO_2$ conditions around the iron-wüstite (IW) buffer (Jacob et al., 2004).
The redox stability of moissanite was estimated from thermodynamic analysis by Essene and Fisher (1986) and by Mathez et al. (1995), and was studied experimentally by Ulmer et al. (1998). All of these studies conclude that moissanite formation requires oxygen fugacities several orders of magnitude lower than that of the iron-wüstite buffer. Ulmer et al. (1998) found no important change in the relative positions of metal-oxide buffers and moissanite-forming reactions to pressures as high as 9 GPa. Dobrzhinetskaya and Green (2007) produced moissanite during diamond synthesis experiments in a graphite-silicate system with $f_{O_2}$ conditions nominally at the iron-wüstite buffer (1450-1500°C, 8.5 GPa). The authors took the formation of moissanite to mean that their oxygen fugacity was much lower than intended, but it is also possible that moissanite stability at high pressure needs further scrutiny. However, the common occurrence of native Si inclusions in natural moissanite supports the inference of redox conditions far below the iron-wüstite buffer. Experiments and calculations of Fe and Si metal-oxide equilibria by Malavergne et al. (2004) place the Si-SiO$_2$ buffer about 8 log units below the iron-wüstite buffer at 20 GPa and 2000°C, and the difference between the two buffers increases at lower temperature.

Stable growth of moissanite in the upper mantle can therefore be ruled out unless there are special redox conditions (see below). The bulk chemical composition and redox conditions of the lower mantle are not known from direct observation on xenoliths or igneous rocks, and it is not clear whether SiC and other reduced phases like metallic Si would be stable under “normal” lower mantle conditions, but prevailing thermodynamic and mineralogic models would suggest not. Frost and McCammon (2008) calculated the depth variations in oxygen fugacity for a 4-phase garnet peridotite along the cratonic geotherm. According to this model, there is a steady decrease of $f_{O_2}$ with depth until the IW buffer is intersected at about 8 GPa, but once Fe-Ni alloy forms, further decrease in $f_{O_2}$ with depth is small and conditions of 5 or more log units below IW are not achieved anywhere in the lower mantle. The metastable extension of Frost and McCammon’s $P-f_{O_2}$ relation without Fe-saturation reaches $f_{O_2}$ values 5 log units below IW at around 25 GPa. However, this seems an unrealistic proposition based on work by Rohrbach et al. (2007) and given that Fe-Ni alloys are relatively common in mineral separates from the ophiolite chromitites we have studied (e.g., Bai et al., 2000; Robinson et al., 2004; Shi et al., 2007).
In summary, the available evidence suggests that conditions are too oxidizing for moissanite (and native Si) formation in the upper mantle. Conditions in the lower mantle also seem hostile to moissanite stability from current understanding, but this is much less certain. The alternative is to propose local, highly reducing microenvironments in the mantle. One well-known example of this kind in the upper mantle is the local reduction of Fe-silicates to form Fe-Ni metal (awaruite) due to release of H₂ during serpentinization (e.g., Dick, 1974; Sleep et al., 2004). The high temperatures required for moissanite to form rule out this specific explanation for SiC, but there can be analogous hydration reactions at higher P and T involving DHMS phases (dense hydrous magnesian silicates), which are known to be stable above 22 GPa and 1200°C (Ohtani et al., 2001). Another example of a reducing microenvironment was proposed by Jacob et al. (2004) to explain their finding of FeC and native Fe inclusions in diamonds. They suggested that local and transient reducing conditions result from production of hydrogen when diamond grows from a methane-dominated fluid (see also Thomassot et al., 2007). This explanation encounters problems when considering the distinctly different C-isotope compositions of moissanite and diamonds as described in the following section. A possibility suggested by Mathez et al. (1995) is that subducted carbonaceous sediment from the surface could create a reducing microenvironment in the mantle if it could remain intact. This may be unlikely, but it has the attraction of also explaining the distinctive C-isotope composition of moissanite (see below).

5.2 The carbon isotope perspective

Reviews of the isotopic composition of carbon in mantle xenoliths, magmatic rocks and related fluids by Deines (2002), and of diamonds by Cartigny (2005) establish that the dominant mantle carbon reservoir sampled by these materials has a $\delta^{13}C$ value near -5 ‰. Our new SIMS results from moissanite in ophiolites, and the compilation of published C-isotope compositions of moissanite from other settings (Fig. 3) show that all terrestrial moissanites analyzed thus far are strongly $^{13}C$-depleted relative to this value. Mathez et al. (1995) pointed out this isotopic feature of moissanite based on the then-available 30
analyses, and it remains valid now with more than twice as much data. Considerably more data on the
stable isotope composition of diamonds are also now available and we briefly summarize what is known
about $^{13}$C-depleted carbon in the mantle from the diamond perspective. Of the more than 4400 C-isotope
analyses compiled in the review by Cartigny (2005), 72% have a narrow range of $\delta^{13}$C values from -8 and
-2 ‰, and these are attributed to the main C reservoir in the mantle. From the remainder, a few analyses
are enriched in $^{13}$C (to a maximum of +5 ‰), but the great majority are $^{13}$C-depleted, with a minimum
value of -38.5 ‰. Several features of these $^{13}$C-depleted diamonds (based on Galimov, 1991; Kirkley et
al., 1991; Deines, 2002, Cartigny, 2005 and Heaney et al., 2005) are worth emphasizing:

1. Most crustal-derived microdiamonds from deeply-subducted ultra-high pressure (UHP)
metamorphic terranes have $^{13}$C-depleted compositions. A compilation of 120 $\delta^{13}$C values from UHP
metamorphic diamonds by Cartigny (2005), shows a range from -4 to -30 ‰ with a broad peak around
-10 ‰. Microdiamonds from the Erzgebirge UHP terrane in Germany have more restricted $\delta^{13}$C range
of -17 to -29 ‰ (Dobrzhinetskaya et al., 2007). The $^{13}$C-depleted composition of these diamonds is
logically attributed to biogenic carbon in the protoliths, although as Horita (2005) discuss, abiogenic
processes can produce the same degree of isotopic fractionation as biogenic ones (see also discussion
of $^{13}$C-depleted Hadean microdiamond and graphite from Jack Hills, Australia, by Nemchin et al.,
2008).

2. All grains of the enigmatic, polycrystalline variety of diamond called carbonado have $^{13}$C-depleted
compositions in the $\delta^{13}$C range of -23 to -30 ‰. The origin of carbonado diamonds is a major puzzle
but several features suggest a crustal setting (Heaney et al., 2005). Interestingly, moissanite is among
the inclusion phases reported from carbonados (Heaney et al., 2005).

3. Among kimberlitic diamonds, $^{13}$C-depleted compositions ($\delta^{13}$C < -10 ‰) are proportionally much
more common in eclogite-types (34%) than in peridotite-type diamonds (2%).

4. Kimberlitic diamonds with $^{13}$C-depleted compositions ($\delta^{13}$C < -10 ‰) have a wide range of $\delta^{13}$C
values, and compositions from individual localities can be distinctly different (Galimov, 1991).
The reason(s) for C-isotope heterogeneity and $^{13}$C-depleted composition of diamonds is the subject of lengthy discussion (see Galimov, 1991; Kirkley et al., 1991; Deines, 2002; Cartigny, 2005), which centers around three options: i) introduction of surface-modified, biogenic carbon into the mantle by subduction, ii) intrinsic heterogeneity of mantle carbon, and iii) isotopic fractionation of C from a once-homogeneous reservoir. Different authors have favored one scenario over another but most conclude that multiple carbon sources are required to explain the compositional range of all $^{13}$C-depleted diamonds.

The above scenarios for explaining $^{13}$C-depleted diamonds are also relevant for the interpretation of moissanite compositions, but the moissanite situation is special in several ways. One distinction is the extremely low oxygen fugacity required for moissanite stability and for its inclusions of Si and Fe-Si alloy. Another important distinction is the relatively narrow range and low values of $\delta^{13}$C from moissanites (-18 to -35 ‰) compared with those of $^{13}$C-depleted diamonds (Fig. 3), excepting carbonado (-23 to -30 ‰; Heaney et al., 2005). One possible explanation for the distinctive and rather homogeneous $^{13}$C-depleted composition of natural moissanite is a systematic isotopic fractionation from an equally homogeneous carbon source, i.e., the mantle reservoir at -5 ‰. The fractionation hypothesis has been widely discussed with respect to $^{13}$C-depleted diamonds and found to be lacking (Cartigny, 2005), and the same problems apply for SiC. Equilibrium C-isotope fractionation between diamond and CO$_2$ or CH$_4$ at high temperatures are about 4 ‰ and 1 ‰, much too small to explain the $\delta^{13}$C variations from a single starting composition of -5 ‰. A greater shift in isotopic composition could be achieved by open-system fractionation where one C component is continuously removed. However, modeling of this process (Deines, 1980) shows that it would result in a skewed distribution of $\delta^{13}$C values quite unlike the narrow range in moissanite, and nowhere near the observed low values of around -30 ‰. Experimental fractionation studies involving SiC are lacking, but we can still expect low fractionation factors at high temperature and a continuous distribution of $\delta^{13}$C values resulting from open-system processes.
If the $^{13}$C-depleted composition of moissanite cannot be produced by fractionation from the carbon reservoir in the mantle with -5 ‰, then there must be at least one other, isotopically distinct source of carbon, which is either introduced from the crust via subduction or is inherent to the mantle. The C-isotopic composition of moissanite overlaps closely with the range of biogenic carbon and as noted above, the subduction of carbonaceous material could satisfy both the isotopic and the redox constraints for moissanite formation. As Mathez et al. (1995) pointed out, it would be difficult to avoid dilution and oxidation of the carbonaceous component during subduction-zone metamorphism and deformation before reaching the high-temperature conditions (>1000° C) needed for moissanite formation. Subduction of biogenic carbon cannot be completely ruled out, but it is not an easy explanation because of the need to chemically isolate the special microenvironment through the subduction process.

The alternative hypothesis to subduction of an appropriately light carbon source is that the mantle inherently contains more than one isotopically distinct carbon reservoir. This is a view widely held among diamond researchers (e.g., Deines, 2002; Cartigny, 2005), and the best evidence for indigenous $^{13}$C depleted carbon comes from studies of extraterrestrial carbon. The C-isotope variability in meteorites is often cited as evidence that the accreted earth did not have a homogeneous bulk composition of -5 ‰ (e.g., Kirkley et al., 1991; Deines, 2002; Rollinson, 2007). An important piece of evidence in this regard comes from a study of carbon in Mars meteorites by Grady et al. (2004). They reported $\delta^{13}$C values of -20 ±4 ‰ for 12 Mars meteorites, suggesting the Martian mantle may have been uniformly $^{13}$C-depleted. Furthermore, Kashizume et al. (2004) suggested that solar carbon must be isotopically light ($\delta^{13}$C < 100 ± 20‰) based on SIMS analysis of solar wind-implanted C in the lunar regolith.

There is abundant, good evidence for a major carbon reservoir in the Earth’s mantle with $\delta^{13}$C = -5 as mentioned above, but that is based on materials from intermediate (rarely) to upper mantle (mostly) depths. If we accept the extraterrestrial evidence that the bulk Earth, like Mars, had a more $^{13}$C-depleted composition, then the prevalence of C-isotope values around -5 ‰ in the upper mantle implies a deeper reservoir of lighter carbon. It is speculative but certainly possible, that the moissanite C-isotope
composition reflects a deep-mantle origin. The source region for moissanite has the additional constraint of very low oxygen fugacity (although the two conditions need not be directly related). As discussed in the previous section, robust estimates of oxygen fugacity in the mantle are limited to the relatively shallow depths from which there is observational evidence, and it is certain that moissanite cannot stably form there. The lower mantle redox conditions are not known to be appropriate for moissanite formation but they are model-dependent and much less certain.

The ultimate origin of moissanite found in our ophiolite samples and in kimberlites remains unanswered, however we argue that there is a good possibility that moissanite in mantle-derived rocks originates in the lower mantle. For this reason alone, natural miossanites are worth further critical study. More insights may from application of other isotopic systems (N, Si) to moissanite collections already available. For example, Mathez et al. (1995) reported two N-isotope determinations from kimberlite-hosted moissanite ($\delta^{15}$N = 9.7 ‰ and 5.6 ‰). These values are at the heavy end of the upper mantle range (-25 to +15 ‰) and overlap with metamorphic diamonds and N from metasediments (Cartigny, 2005).

More studies of this kind would be helpful but it is also important that further occurrences of moissanite be actively explored and investigated.

6 Conclusions

We have shown that moissanite is a relatively common mineral in podiform chromitites and/or host peridotite from four ophiolites of different age and geographic location (Luobusa and Dongqiao in Tibet, Ray-Iz in the Polar Urals and Semail in the Arabian peninsula). Our results, combined with previously published data firmly demonstrate the existence of natural SiC as a rare but rather widespread mineral in Earth’s mantle. In the chromitites, moissanite is associated with a wide variety of unusual minerals including diamonds, coesite, kyanite, Fe-Si alloys and native elements. Many moissanite grains from both ophiolites and kimberlites contain inclusions of native Si and Fe-silicides. The rounded form of these inclusions suggests formation as trapped liquids. The redox and temperature conditions implied by these inclusions are compatible with the stability conditions for moissanite, but are not compatible with the
conditions expected either in the ophiolite or in the kimberlite source regions of the upper mantle. We conclude that the ophiolitic moissanite grains are xenocrysts that have been incorporated into the chromitites, though the possibility of deep origin for some of the chromitite is indicated by a recent finding of coesite lamellae in chromite grains from the Luobusa ophiolite (Yamamoto, et al., 2009).

All of the ophiolite-hosted moissanite grains have a distinctive and narrow range of $^{13}$C-depleted isotopic compositions (-18 to -35 ‰), and this same range applies to all natural moissanite studied thus far. The C-isotope composition of moissanite is thereby distinct from that of kimberlitic diamonds, even those of the ecologite-type. It is not possible to explain the $^{13}$C-depleted range of moissanite by isotopic fractionation from the mantle carbon reservoir with a $\delta^{13}$C value of -5 ‰, with or without Rayleigh distillation. Moissanite, like some $^{13}$C-depleted diamonds, must tap a separate carbon reservoir in the mantle. There is good evidence from studies of extraterrestrial materials that $^{13}$C-depleted carbon was incorporated in the early Earth, so the observed prevalence of upper mantle $\delta^{13}$C values near -5 ‰, implies the existence of isotopically lighter carbon in a deeper reservoir. The ultimate source of moissanite from the ophiolite and kimberlite associations is still unknown but a lower mantle origin is one of the better possibilities and it should be further explored.

Acknowledgements

We thank E. Mathez, AMNH for providing a sample of ANMH 100578 reference SiC. D. Rhede, R. Wirth and M. Gottschalk, GFZ, are gratefully acknowledged for their assistance with electron microprobe, TEM and XRD analyses, respectively. Thanks also go to A. Shiryaev, I. Veksler and R. Wirth for their very helpful discussions. Journal reviews by L. Dobrzhinetskaya and H. Rollinson were most helpful. Hugh Rollinson pointed out the possible significance of C-isotopes in the Mars meteorites.
References


of the Ray-Iz Ultramafic Massif. The Ural Branch of the Academy of Sciences of the USSR, Sverdlovsk, Russia, p. 149-194 (in Russian)


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

Figure 2. Photomicrographs of moissanite from ophiolites. (a) loose moissanite grains from Luobusa orebody 31 (PPL). Note the euhedral outline of several grains, the round to ovoid inclusions of Si (circled) and optical zoning in one grain (lower center). The dark grains at the top of the image are graphite; (b) moissanite grain from the Semail ophiolite (PPL) with small ovoid inclusion of Si (circled); (c) angular moissanite fragments from Ray-Iz ophiolite with large, rounded Si inclusions (reflected light); (d) back-scattered electron image of the inclusion in (b) showing grains of Fe-Si alloy at the margin of the Si inclusion in moissanite.

Figure 3. Frequency histograms showing the C-isotope composition of moissanites from ophiolites determined in this study (upper panels separately for each locality) and a stacked histogram (lower panel) with all moissanite δ\textsuperscript{13}C values compiled from our study and the literature. Key for references to moissanite data: (1) this study; plus two analyses from Armenia in Marshintsev, (1990), (2) Di Pierro et al., 2003; (3) Mathez et al., 1995; (4) Marschintsev, 1990; (5) Leung et al., 1990; (6) Wagner et al. 1988. Also shown in gray shading on the lower panel is the relative frequency distribution of δ\textsuperscript{13}C values from 997 eclogite-type diamonds (modified from Cartigny, 2005).
The carbon isotope ratio of synthetic SiC reference material used for SIMS analyses was measured after sample combustion in a Carlo Erba EA-1500 reactor connected to a ThermoFisher Delta V Plus mass spectrometer. The sample was individually wrapped in tin foil cups and dropped into the reactor with a small dose of oxygen gas mixed into the He stream. The sample was then oxidized in the reactor held at 1050°C and filled with cobaltous oxide as a catalyst for the oxidation. Excess oxygen from the He-stream was adsorbed in another reactor column filled with metallic Cu held at 500°C. The residual gases of CO₂, (N₂), and H₂O were then passed over a water trap of magnesium perchlorate (Mg(ClO₄)₂) and into a gas chromatograph filled with a molecular sieve (5A) to separate N₂ and CO₂. The CO₂ was transported by the He-stream via a CONFLO III (open split) system into the mass spectrometer for isotopic analyses. The sample yields using this procedure were between 90 and 100%. Reproducibility of the internal graphite standard used was better than 0.10‰ and USGS-24 graphite (−16.0 per mil VPDB) and NBS-22 oil (−29.6 per mil VPDB) were used as calibration standards. The results for duplicate analyses of the reference SiC GFZ-S113 are δ¹³C values of −28.71 and −28.79‰.
Figure 2
Figure 3
### Table 1. Results of SIMS measurements on SiC reference materials

<table>
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<tr>
<th>Analysis</th>
<th>Sample</th>
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<th>$^{13}\text{C}/^{12}\text{C}$ (meas.)</th>
<th>precision$^a$</th>
<th>IMF$^b$</th>
<th>$\delta^{13}\text{C}^c$</th>
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<tr>
<td>Mean</td>
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<td></td>
<td></td>
<td>-28.8</td>
</tr>
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<td>Repeatability in permil$^d$</td>
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<td></td>
<td></td>
<td>1.98</td>
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| AMNH-100578 | S100578 | G1    | 0.01061                           | 1.10         | 0.0969  | -24.6          |
| PRmar047 | AMNH 100578 | G1    | 0.01061                           | 0.96         | 0.0969  | -24.6          |
| PRmar057 | AMNH 100578 | G2    | 0.01058                           | 1.06         | 0.0967  | -27.4          |
| PRmar058 | AMNH 100578 | G3    | 0.01061                           | 1.14         | 0.0969  | -24.6          |
| PRmar059 | AMNH 100578 | G4    | 0.01059                           | 1.11         | 0.0968  | -26.5          |
| PRmar066 | AMNH 100578 | G6    | 0.01061                           | 1.01         | 0.0969  | -24.6          |
| Mean     | AMNH 100578 |       | 0.01060                           |              |         | -25.4          |
| External precision in permil$^d$ | | | | 1.25 | |

Values for S113 from this study (see text), for AMNH 100578 from Mathez et al. (1995)

**a)** Analytical uncertainty in permil (1σ) for each analysis of 80 cycles

**b)** Instrumental mass fractionation ($^{13}\text{C}/^{12}\text{C}_{\text{measured}} / ^{13}\text{C}/^{12}\text{C}_{\text{reference material}}$)

**c)** Values in permil relative to the zero-value reference PDB ($^{13}\text{C}/^{12}\text{C} = 0.112372$)

**d)** Repeatability based on multiple analyses of reference SiC ($1\sigma / \text{mean} \times 1000$)
Table 2. Results of in-situ C-isotope composition of moissanites by SIMS

<table>
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<th>Locality</th>
<th>Sample-Grain</th>
<th>$^{13}$C/$^{12}$C (meas.)</th>
<th>Precision</th>
<th>$^{13}$C/$^{12}$C (corr.)</th>
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<td>Turkish beach</td>
<td>G4</td>
<td>0.01063</td>
<td>0.148</td>
<td>0.10981</td>
<td>-22.8</td>
</tr>
<tr>
<td>Turkish beach</td>
<td>G5</td>
<td>0.01058</td>
<td>0.119</td>
<td>0.10930</td>
<td>-27.4</td>
</tr>
</tbody>
</table>

** Moissanite-rich ultramafic volcanic sample found on Turkish beach (di Pierro et al., 2003)