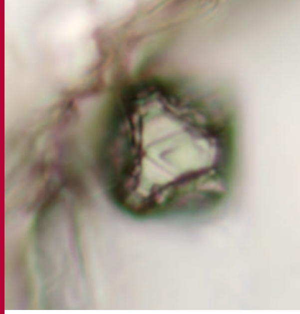
Microdiamonds in Ultrahigh-Pressure Metamorphic Rocks



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Since the first report of microdiamonds of metamorphic origin in crustal rocks of the Kokchetav Massif, northern Kazakhstan, diamonds have been described from several other ultrahigh-pressure (UHP) metamorphic terranes. In situ diamond is the best indicator of ultrahigh-pressure conditions (>4 GPa), and testifies to subduction of continental crust to depths within the diamond stability field followed by relatively rapid exhumation. In contrast to other UHP terranes, the Kokchetav Massif contains rocks with unusually abundant diamonds, particularly in the Kumdy-Kol region. Kumdy-Kol diamonds exhibit diverse morphologies, dependent upon the host rock. Raman and cathodoluminescence spectra and carbon isotope compositions differ between core and rim, indicating two distinct growth stages.

KEYWORDS: microdiamond, ultrahigh-pressure metamorphism, continental collision, Kokchetav

INTRODUCTION

After the discovery of coesite in metamorphic rocks from the Dora Maira Massif, western Alps (Chopin 1984), and the Western Gneiss Region, Norway (Smith 1984), many researchers were stimulated to look for coesite and other minerals indicative of extremely high-pressure formation in metamorphic rocks. What is so interesting here? Coesite, a dense form of SiO_2, forms at ~3 GPa (900°C), equivalent to a depth of 90 km under a continent, while diamond requires ~4 GPa (1000°C), or roughly 140 km depth. These minerals would be expected in the deep Earth where sufficient silica or carbon is present, so there is nothing revolutionary in their discovery. However, up to that time it was not thought that tectonic processes could send slabs of crust down to extreme depths and, more amazingly, bring them up again quickly enough to preserve these high-pressure minerals. As discussed by Stachel et al. in this issue, most diamond found on Earth is carried to the surface by deep-rooted volcanism. The collision of fragments of Earth's crust in the dance of plate tectonics was known to return rocks such as eclogites to the surface with minerals indicating pressures of >2 GPa, but these minerals are typically overprinted by reactions that returned most minerals to lower-pressure assemblages. Preservation of high-pressure coesite and even higher pressure diamond challenged existing models of Earth processes and required a whole new paradigm as more discoveries of ultrahigh-pressure metamorphism (UHPM) were reported. The subject has taken off in the last 20 years, and many reviews are available (e.g., Rumble et al. 2003; Roselle and Engi 2002). In the present article, some of the discoveries and implications will be reviewed, with attention to recent results from at least one occurrence pointing to multiple generations of diamond formation.

OCCURRENCES OF UHP METAMORPHIC DIAMOND

Although microdiamond was initially reported many years ago by Rozen et al. (1972) from the Kokchetav Massif, northern Kazakhstan, its metamorphic origin was accepted much later following a report by Sobolev and Shatsky (1990). Since then, additional occurrences have been discovered in the Dabie Mountains (Xu et al. 1992) and north Qaidam (Yang et al. 2003), China; the Western Gneiss Region, Norway (Dobrzhinetskaya et al. 1995; Van Roermund et al. 2002); Erzgebirge, Germany (Massonne 1999; Stöckhert et al. 2001); Sulawesi, Indonesia (Parkinson and Katayama 1999); and perhaps the Rhodope Massif, Greece (Mposkos and Kostopoulos 2001). The Dabie and north Qaidam rocks have been dated at 230–209 Ma (Ames et al. 1996) and 510–485 (Yang et al. 2001), respectively. Peak P–T conditions of the Dabie metamorphism are considered to be around 5–6 GPa and ca. 850°C (Zhang and Liou 1998). More than 10 diamond grains have been reported as inclusions in zircon from north Qaidam (Yang et al. 2003).

Several grains of diamond were first described from residues separated from Western Gneiss Region gneisses by Dobrzhinetskaya et al. (1995) and confirmed in situ by Van Roermund et al. (2002). Peak P–T conditions for the Norwegian rocks were P >2.8 GPa and T >790°C (Carswell et al. 1999), and peak metamorphism occurred at 425–406 Ma (Griffin and Brueckner 1985). Metamorphic diamonds are far more abundant in the Erzgebirge than in other diamondiferous terranes, except for Kokchetav. Diamondiferous gneisses are restricted to a 1 km-long strip, and the diamonds are abundant as inclusions in garnet, kyanite, and zircon (Massonne 2001). Peak P–T conditions were >4.2 GPa and 900–1000°C (Massonne 2001) and occurred at 360–333 Ma (Schmadicke et al. 1995). A number of in situ diamond grains were identified by laser Raman spectroscopy within quartz pseudomorphs after coesite in garnet in jadeite quartzite of the Bantimala complex of Sulawesi, Indonesia. These rocks have yielded radiometric ages of 130–120 Ma and recrystallized at P >2.7 GPa at around 750°C (Parkinson et al. 1998).

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UHP rocks in these regions display some diversity in terms of tectonic setting. However, the commonalities are perhaps more significant—they are all restricted to collisional orogens and are predominantly of continental parentage. In general the distribution of diamond-grade rocks is restricted to considerably smaller areas than that of coesite-grade metamorphism, which is regional in extent in many UHPM terranes. Presumably, diamonds produced by subduction of carbon-bearing crust are not an unusual product of Earth processes, but their return to the planet's surface without an explosive kimberlite elevator requires special conditions. High-pressure metamorphic rocks are a characteristic feature of the "scars" of plate collisions, but UHP rocks appear to be limited to collisions of continental fragments, either full continent–continent collisions or ones involving microcontinental fragments.

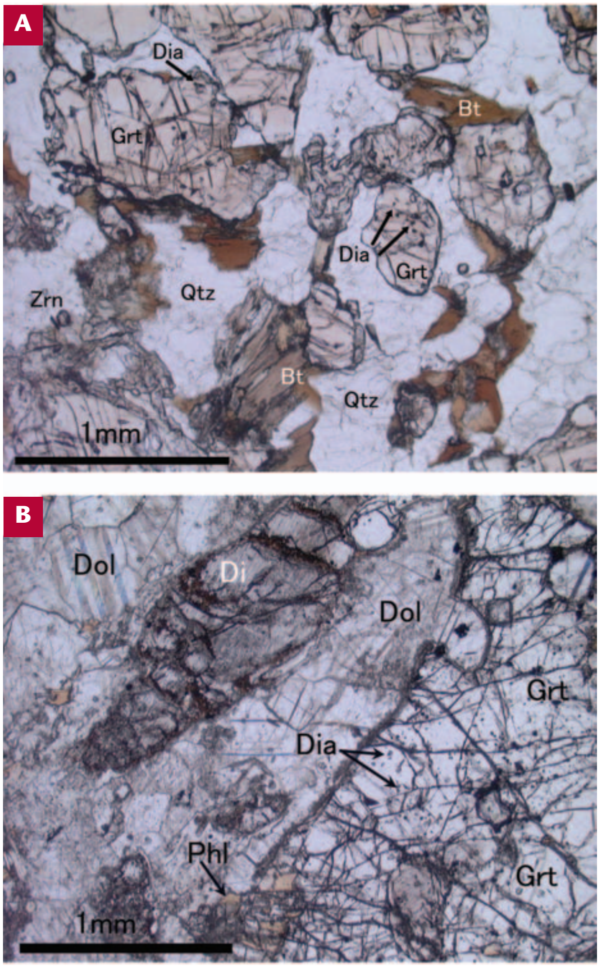

FIGURE 1 Photomicrographs (PPL) of (**A**) diamond-bearing garnet–biotite gneiss and (**B**) diamond-bearing dolomite marble at Kumdy-Kol. Qtz: quartz; Grt: garnet; Bt: biotite; Dia: diamond; Zrn: zircon; Di: diopside; Phl: phlogopite; Dol: dolomite.

Presumably, the considerable momentum of colliding continents and the pull of an attached oceanic lithosphere 'anchor' buries a continental edge to diamond-producing pressures. Then exhumation lifts the buried rocks some 140 km, sufficiently fast that the UHP signature is not erased by thermal relaxation associated with slow ascent. Exhumation by regional uplift and erosion alone is not rapid enough, so several models of tectonic emplacement have been suggested: wedge extrusion, channel flow, and diapiric transport. Break-off of the subducting slab during collision is argued to cause extrusion of a buoyant narrow wedge of buried UHP material by normal and thrust faulting (e.g., Ernst and Liou 1995; Hacker et al. 2000). Although still somewhat controversial, wedge extrusion appears to have gained widespread acceptance as a general exhumation model. It can explain the regional nature of UHP metamorphism (at least for coesite-grade metamorphism), whereas channel flow and diapiric transport provide models for exhumation of discrete, essentially "exotic" blocks. In channel flow, the subducted rocks are considered to have sufficiently low viscosity that they can be squeezed back up the subduction channel in a reverse flow (e.g., Lardeaux et al. 2001). Diapiric emplacement relies on buoyancy forces of either low-density continental material or encapsulating hydrated peridotite (serpentinite) (e.g., Burov et al. 2001) to lift the UHP blocks. Each model has its own set of criteria to test its validity, and the jockeying of the models continues.

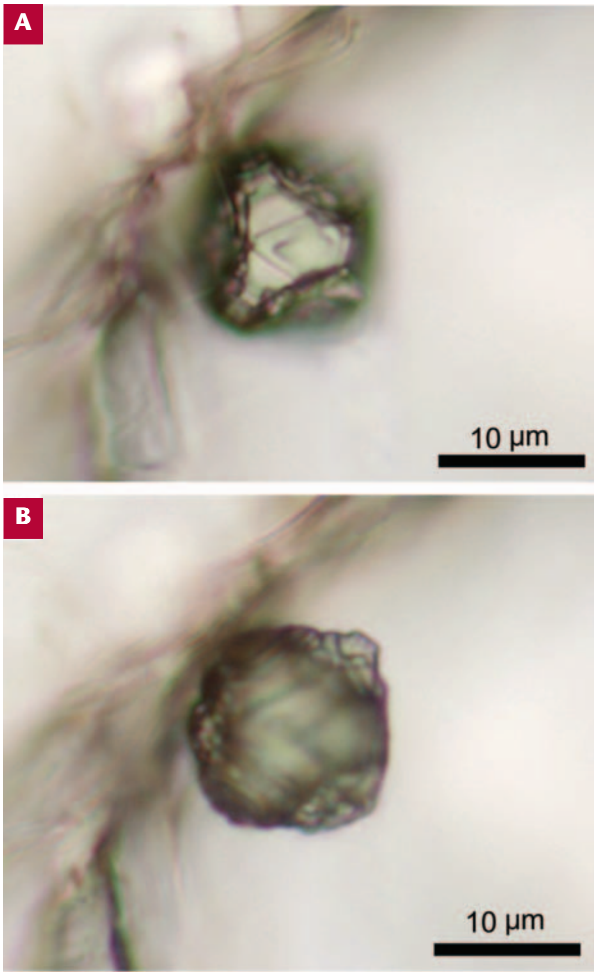

FIGURE 2 Photomicrographs (PPL) of a representative microdiamond in a garnet–biotite gneiss at different focus positions **A**, **B**; stepped octahedral faces are visible in (**A**).

Among the recognized microdiamond occurrences, the Kokchetav Massif is perhaps best suited to provide sufficient data to determine the local processes and environment of formation of metamorphic diamonds because of their extremely high concentration and unequivocal in situ occurrence in UHPM rocks. For these reasons, I focus on the diamonds of Kumdy-Kol. Studies of diamonds in the dolomite marble (dominant carbonate is dolomite) there provide a keystone for understanding their "metamorphic" origin and help to resolve the debate as to whether UHPM diamond is produced by a solid-state transformation or by crystallization from a fluid or a melt.

MICRODIAMOND IN THE KOKCHETAV UHPM TERRANE

Diamond in the Kokchetav Massif is best known from the Kumdy-Kol area where it was once mined. It occurs in carbonate rock, pyroxene–garnet rock, tourmaline-rich rock, and most abundantly in garnet–biotite gneiss (FIG. 1A) and dolomite marble (FIG. 1B) (see Parkinson et al. 2002 for a review). Diamonds are included in garnet and less frequently in phlogopite (pseudomorphic after garnet), zircon, tourmaline, and diopside in the silicate and carbonate rocks. All these diamonds are very fine grained, <25 µm. Diamond with an octahedral form in garnet in garnet–biotite gneiss is shown in FIG. 2.

Diamond occurs in greatest abundance in dolomite marble. Small amounts have been found in layers of calcite marble, but, for an as yet unknown reason, Ti-clinohumite-bearing dolomitic marbles (dominant carbonates are Mg-calcite and dolomite) contain no diamonds. Diamond abundance in the dolomite marble has been estimated by Yoshioka et al. (2001) as up to 2700 carat ton $^{-1}$, and Ishida et al. (2003) counted 4458 grains of diamond in four thin sections (a volume of ~20 x 160 mm by 30 µm in thickness) with grain sizes ranging from 5 to 25 µm. Some garnet grains contain unusually high concentrations of diamond, as shown in FIG. 3, but the distribution of diamonds in garnet is heterogeneous. Polycrystalline aggregates (5 to 30 µm) of graphite, probably after diamond, are included in garnet, diopside, phlogopite, and dolomite. Indeed, many microdiamonds are partially surrounded by graphite.

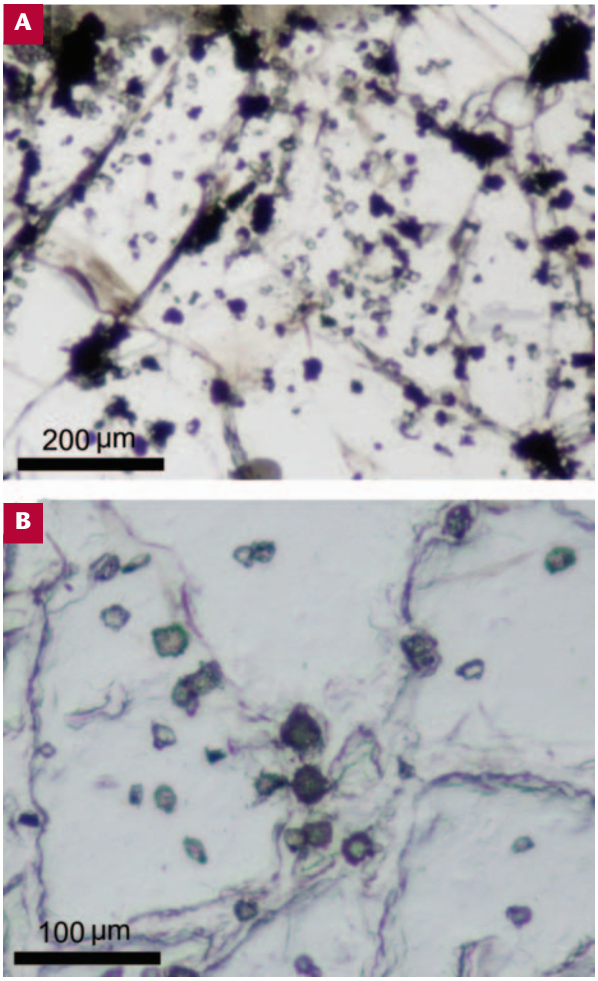

FIGURE 3 Photomicrographs (PPL) of the highest concentration domain of microdiamond in Kumdy-Kol dolomite marble.

Ishida et al. (2003) classified microdiamonds, according to their morphology and other characteristics, into three types: (1) S-type—"star-shaped" diamond consisting of a translucent core and transparent, subhedral to euhedral, very fine-grained, polycrystalline rims (FIG. 4); (2) R-type—translucent crystals with "rugged" surfaces; (3) T-type—transparent, very fine-grained crystals. S-type diamonds predominate (85%), the R-type is minor (~10%), and the T-type is rare (<5%). In a given rock, the spatial distribution of these types with respect to one another appears to be random. The authors proposed a two-stage growth mechanism for diamonds in the dolomite marble based on morphology and other characteristic features. Micro-Laue diffraction using a finely collimated synchrotron X-ray beam (1.6 to 50 µm in diameter) demonstrated that the rims of S-type diamonds have crystallographic orientations that are different from those of the cores, which are single crystals, and that R-type diamonds are single crystals.

Yoshioka and Ogasawara (in press) reported strong broad bands in the cathodoluminescence (CL) spectra at 514 to 537 nm from the rims of all S-type diamonds. This band is a green color that is not common among mantle-derived diamonds (FIG. 5). The cores also have the same broad band, but its intensity is very weak. Moreover, the main CL band of S-type diamonds is very similar to the 520 nm CL band of carbonado (Magee and Taylor 1999 and see Heaney et al. in this issue). These data may indicate different geochemical environments for the growth of cores and rims of S-type diamonds and support evidence for two-stage growth.

Imamura et al. (2004) conducted SIMS (ion-probe) carbon isotope analyses of S- and R-type diamonds in a Kokchetav dolomite marble. Rims of S-type diamonds have light isotopic compositions with $\delta^{13}C$ values ranging from –17.2 to –26.9‰, whereas cores have isotopically heavier carbon, with $\delta^{13}C$ values ranging from –9.3 to –13.0‰. The $\delta^{13}C$ values for two R-type grains fall between –8.3 and –15.3‰ and are similar to those for cores of S-type diamond. Thus, R-type grains may form at the same stage of growth and from the same carbon source as the cores of S-types. These data also suggest two stages of growth for S-type diamonds. The extremely light carbon isotopic compositions of rims of S-type diamond may be explained by a light organic source or fractionation (see Cartigny article in this issue).

CONDITIONS OF DIAMOND GROWTH IN KUMDY-KOL CARBONATE ROCKS

The two-stage growth model of Ishida et al. (2003) is strongly supported by the morphology of S-type diamonds, the micro-Laue diffraction data, CL interpretations, and carbon isotope compositions. The first stage corresponds to the growth of R-type diamond and the cores of S-types. The second stage represents the growth of rims on S-type and of T-type diamonds. The origin of the microdiamond cores is still unclear, but one possibility is prograde (i.e., increasing T and P) transformation of graphite. Katayama et al. (2000) interpreted composite graphite–diamond inclusions in zircon in this way. Whatever the origin, mineral textures indicate that first-stage diamond growth was coeval with garnet and diopside crystallization. Because both S-type and T-type diamonds often occur in the same garnet grain, the second stage of growth is either not homogeneous, even on a µm scale, or some factor, like fast growth rate, controls the second stage, which is microcrystalline. Moreover, second-stage growth for the rims of S-types and T-types involves a source of carbon different from that of the cores, such as a fluid or melt rather than graphite.

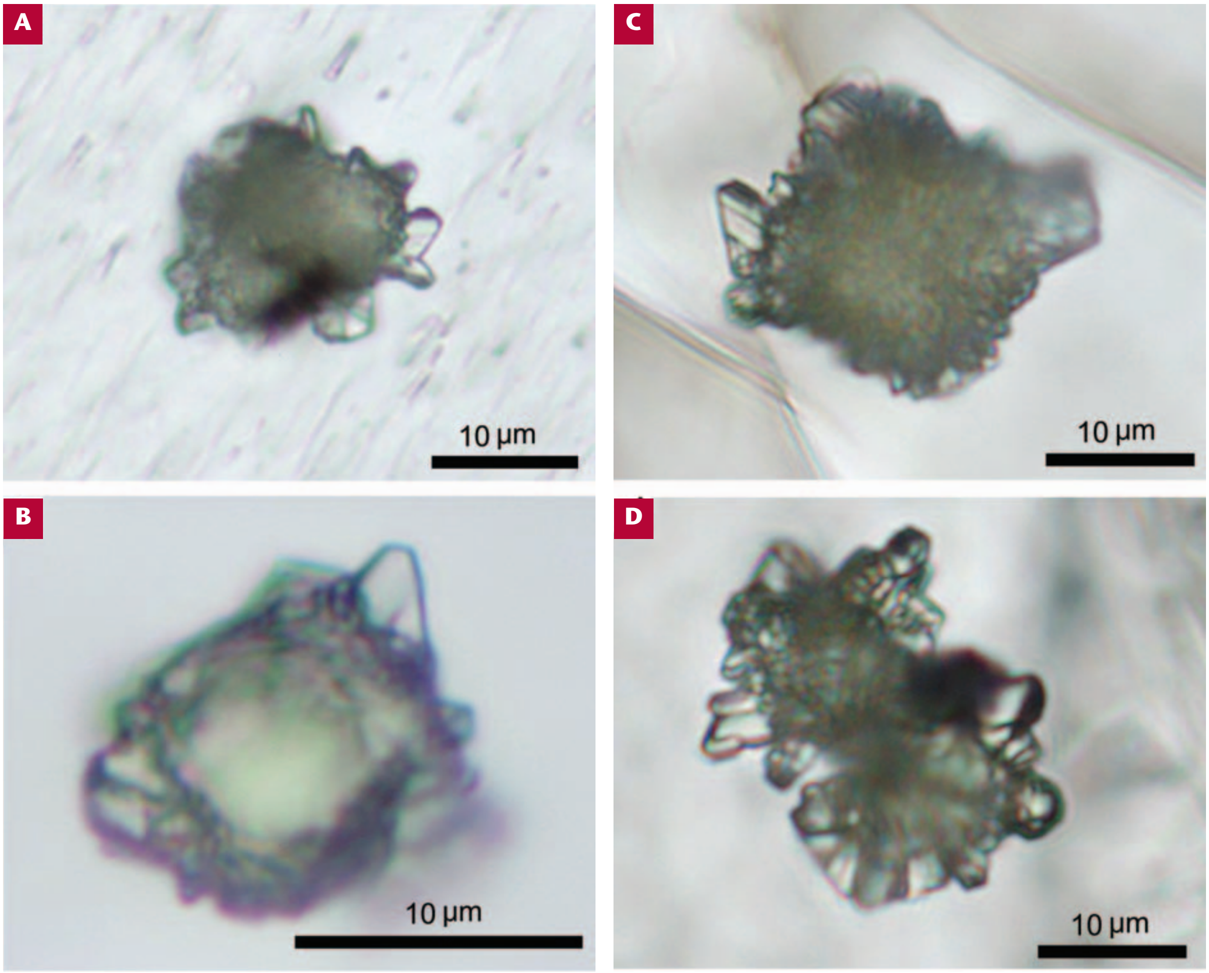

FIGURE 4 Photomicrographs (PPL) of representative S-type microdiamonds in dolomite marble. **A**: diamond inclusion in diopside. **B**, **C**, **D**: diamond inclusions in garnet.

Recent UHPM research has focused on determining whether microdiamonds formed from a fluid or a melt. Diamond has been synthesized stably from a CO_2–H_2O fluid (Kumar et al. 2000) and from dolomite + carbon and dolomite + fluid + carbon systems (Sokol et al. 2001). Diamond has also been synthesized in the dolomite–Si (or SiC) system from a carbonatitic melt (Kozai et al. 2000). One of the possible media for second-stage growth of microdiamonds is an aqueous fluid. An important question is why dolomite marble contains diamond while some other types of marble do not. Ogasawara et al. (2000) argued that the CO_2 concentration (X_{CO_2}) in the metamorphic fluid could explain this distribution. Ogasawara and Aoki (in press) suggest that in diamond-bearing dolomite marble, $0.01 < X_{CO_2} < 0.1$, whereas in diamond-free dolomitic marble, $X_{CO_2} < 0.01$.

Numerous observations point to crystallization of microdiamond from a fluid, but a melt origin has been suggested as well. Dobrzhinetskaya et al. (2001) concluded that composite, nanometer-sized mineral inclusions (Cr_2O_3, TiO_2, $MgCO_3$) and presence of cavities in diamond from a Kokchetav felsic gneiss suggested that diamond had crystallized from a fluid; also, H_2O-bearing fluid inclusions in diamond from a garnet–pyroxene rock were confirmed by Fourier-transform infrared spectroscopy (De Corte at al. 1998). The study of fluid inclusions in diamond from dolomite marble indicates that microdiamonds crystallized in the presence of fluid and possibly from it (Dobrzhinetskaya et al. 2004). Diamond-bearing metal-sulfide inclusions in garnet in a garnet–clinopyroxene–quartz rock have been interpreted as evidence of diamond growth from a fluid (Hwang et al. 2003). Similar composite inclusions (phlogopite, quartz, paragonite, phengite, apatite, and rutile) were also reported in UHP gneiss from Erzgebirge, Germany, and suggest a dense C–O–H fluid rich in K, Na, and SiO_2 under UHP conditions (Stöckhert et al. 2001). Conversely, Massonne (2003) proposed that diamond in silicate rocks of the Kokchetav and Erzgebirge crystallized from a silicate melt, and Herman and Korsakov (2003) and Korsakov and Herman (2004) proposed that diamond in Kokchetav dolomite marble crystallized from carbonate melt, based on textures and trace-element analyses. However, it is difficult to explain the extremely negative $\delta^{13}C$ values (Imamura et al. 2004) by diamond crystallization from carbonate melt, because strong isotopic fractionation is not expected between diamond and carbonate melt at such high temperatures.

Constraining P-T conditions for diamond formation in the dolomite marble is difficult. Two-stage growth, by itself, does not provide such information. If the carbon source was graphite for the first stage, diamond, in principle, may have started to crystallize near the minimum pressure for diamond stability, or slightly above. However, rim growth requires an additional source of carbon, such as fluid or melt. The release of H_2O by dehydration reactions from surrounding rocks may be one of the fluid sources, and growth of rim diamond could be near peak P–T conditions, which for the Kokchetav UHP marble are 6 to 9 GPa (Ogasawara et al. 2002) and 980 to 1250°C. (Ogasawara et al. 2000).

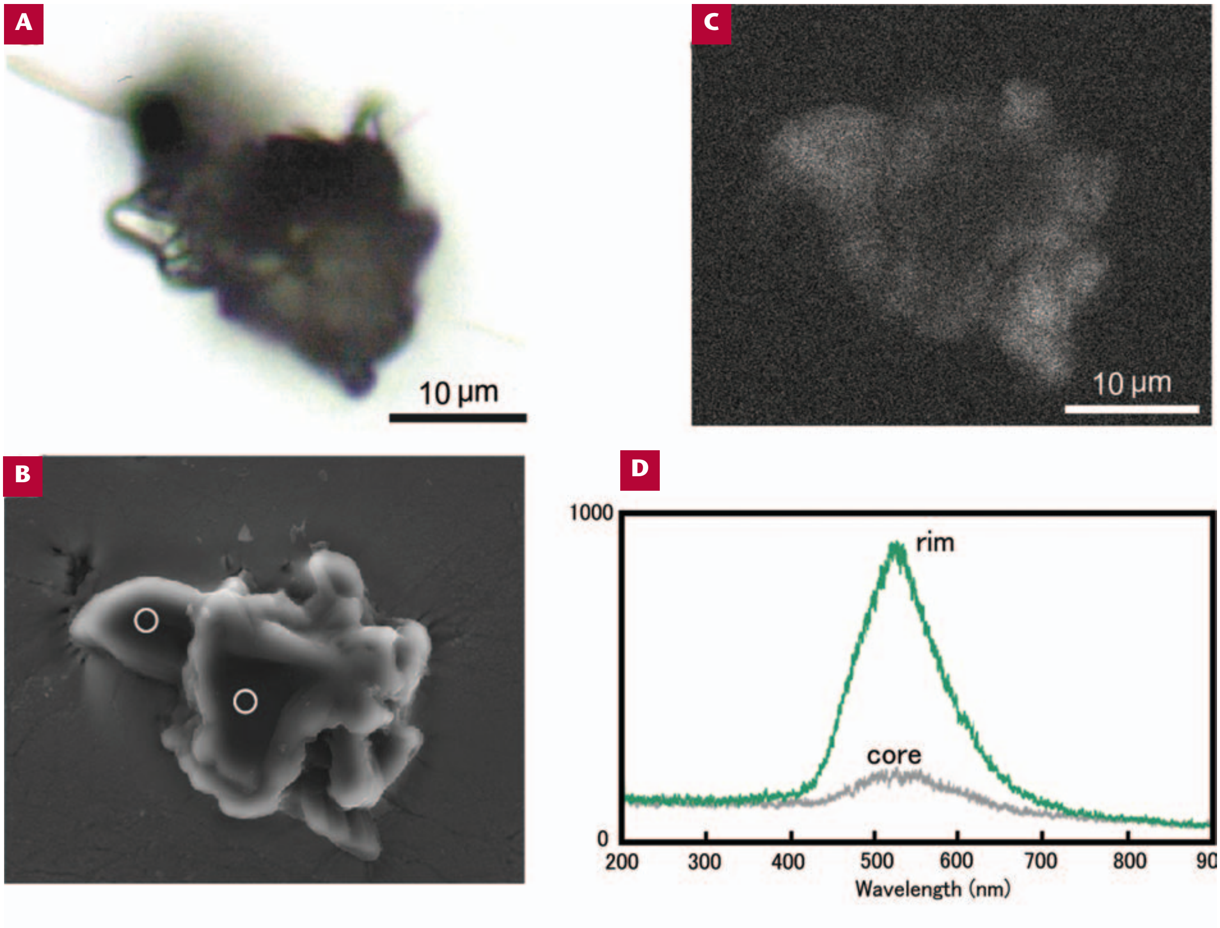

FIGURE 5 Images and cathodoluminescence (CL) spectra of an S-type microdiamond in dolomite marble (after Yoshioka and Ogasawara in press). **A**: Photomicrograph of analyzed grain. **B**: Secondary electron SEM image of analyzed grain. Small circles are locations from which CL spectra were obtained. **C**: CL image at the peak wavelength (523 nm) of the strongest CL band. **D**: CL spectra of core and rim.

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