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ULTRAHIGH-PRESSURE METAMORPHISM OF CRUSTAL ROCKS FROM THE RHODOPE METAMORPHIC PROVINCE: EVIDENCE FROM COESITE, DIAMOND AND MAJORITIC GARNET IN ECLOGITES AND METAPELITES

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ABSTRACT

The Rhodope Metamorphic Province represents an area of continental collision between the Balkan domain to the north and the Pangaeon domain to the south. Today, exposed astride the suture zone are Palaeozoic and Mesozoic protoliths of both continental and oceanic provenance that underwent Alpine deformation and metamorphism in a subduction zone setting. From petrostructural studies the picture that emerged is one of a central, structurally lower, marble-dominated terrain (i.e. a metamorphic core complex), and a surrounding, structurally higher, gneiss-dominated terrain.

Here, for the first time, we report the presence of ultrahigh-pressure metamorphic indicator minerals such as coesite, diamond and Si-Ti-Na-P-rich (i.e. majoritic) garnet in amphibolitized eclogites and garnet-biotite-kyanite gneisses from localities scattered throughout the structurally higher terrain. These findings, corroborated by optical microscopy, electron microprobe analyses and in situ laser Raman microspectroscopy, suggest that the protoliths of these rocks were dragged down to mantle depths exceeding 200 km. The individual pressure-temperature paths published before for various subunits of the structurally higher terrain should henceforth be regarded as peculiarities of the exhumation path followed by the subunits.

KEY WORDS: Rhodope Metamorphic Province (RMP), ultrahigh-pressure metamorphism (UHPM), coesite, rutile, diamond, majorite, laser Raman microspectroscopy.

1. INTRODUCTION

ULTRAHIGH-PRESSURE METAMORPHISM OF CRUSTAL ROCKS

To what depth can continental material be subducted? Up until the discovery of coesite in crustal lithologies from the Italian Alps (Dora Maira Massif; Chopin, 1984), the subduction of continental material to great depths, especially below the crust-mantle boundary, was considered impossible. The presence of coesite signalled the possibility of driving continental material to a depth of at least 85-95 km by placing a minimum pressure of about 2.6-3.0 GPa (Hemingway et al., 1998) at the inferred range of peak metamorphic temperatures (i.e. 600-1100°C). The documentation of diamond in crustal metamorphic rocks from continental plate collision zones such as the Kokchetav Massif, Kazakhstan (Sobolev and Shatsky, 1990), the Maksyutov Complex, Russia (Leech and Ernst, 1998), the Dabie Shan - Su Lu terrain, China (Xu et al., 1992), the Western Gneiss Region, Norway (Dobrzhinetskaya et al., 1995), and recently the Saxonian Erzgebirge, Germany (Massonne, 1998, Nasdala and Massonne, 2000) indicates continental subduction to even greater mantle depths. At the temperature range of 600-1100°C, diamond is stable only at pressures above 3.1-4.5 GPa (see Chatterjee, 1991, and Chatterjee et al., 1998, for a thermodynamic treatment of the Kennedy and Kennedy, 1976, high-temperature experiments and extrapolation to lower temperatures), bringing the depth of metamorphism to a minimum of 100-150 km. Moreover, Daniels et al. (1996) reported the existence of a staurolite inclusion in a demonstrably mantle diamond from the Dokolwayo kimberlite, Swaziland.

The staurolite had an iron-rich composition with a striking resemblance to that of staurolites from normal pelitic rocks, and these authors concluded that material of crustal provenance had been incorporated in the mantle.

In addition to the above, Huang et al. (2000) identified submicrometer-size inclusions of TiO_2 with an α-PbO_2-type structure in porphyroblastic garnet from the diamondiferous quartzofeldspathic rocks of the Saxonian

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Erzgebirge, Germany. Given Massonne's (1998) inference of temperatures in the vicinity of 1000°C (using the Ti content of Al-garnet coexisting with rutile and a SiO_2 phase) for the diamond-building stage and the U-shaped boundary between rutile and α-PbO_2-type TiO_2 (Olsen et al., 1999), minimum pressures of 4.5 and 6.5 GPa can be calculated for the peak of metamorphism, depending on whether nanophase or bulk α-PbO_2-type TiO_2 respectively is considered.

Recently, Ye et al. (2000) have documented the exsolution of clinopyroxene, rutile, and apatite in porphyroblastic garnet from eclogites of the Su Lu region from within the Dabie Shan - Su Lu orogen, China. Their reconstructed pre-exsolution garnet composition matched that of experiments conducted at P>7 GPa and T>1000°C on basaltic systems and proposed the possible subduction of continental material to depths greater than 200 km.

2. GEOLOGICAL BACKGROUND

The Rhodope Metamorphic Province (RMP) is one of the major geotectonic units of northern Greece. It extends northwards into eastern former Yugoslavia and southern Bulgaria, and includes the Serbo-Macedonian (Kerdillion, Vertiskos and Ogra□den blocks) and the Rhodope Massifs. It mainly comprises amphibolites, often enclosing eclogite bodies, amphibolite-facies para- and orthogneisses and schists, in places migmatitic, and marbles, all invariably intruded by large granitic masses. The RMP has traditionally been viewed as a stable continental block, consolidated in Precambrian to Palaeozoic times. Recent structural and petrological work has nevertheless shown the RMP in fact to be a complex of Alpine synmetamorphic nappes characterized by south- to southwestward stacking and associated with both coeval and subsequent extension in an Alpine active margin setting (Ricou et al., 1998, Dinter, 1998, Barr et al., 1999).

Mposkos and Krohe (2000) have further subdivided the RMP into discrete entities on the basis of calculated metamorphic P-T paths and exhumation age criteria for the various metamorphic rocks. Thus, the earliest exhumed and structurally uppermost entity is the Kimi Complex (65-48 Ma), followed by the Sidironero (Central Rhodope) and Kechros Complexes (East Rhodope) (42-30 Ma), and then by the Pangaeon Complex (26-8 Ma), which also forms the well-defined Rhodope metamorphic core complex. In terms of metamorphic P-T paths the major difference between the structurally lower complexes (i.e. Pangaeon, lower Sidironero [Albite-Gneiss Series] and Kechros) and the overlying complexes (i.e. upper Sidironero and Kimi) is that in the latter, prograde assemblages developed at T>550°C, whereas in the former they developed at T<550°C. Available peak pressure estimates range from ca. 1.2 GPa for Pangaeon to about 1.4 for Kechros and 1.9 GPa for Sidironero (~1.6 GPa for Kimi; see Mposkos and Krohe, 2000).

3. ULTRAHIGH-PRESSURE METAMORPHISM INDICATORS

In this paper, for the first time, we report UHP metamorphic indicator minerals discovered in crustal rocks belonging to the Kimi and Sidironero Complexes as exemplified above (Fig. 1). With regard to the former, UHPM indicators include:

i) multicrystalline polygonal quartz (MPQ) aggregates included in garnet (Fig. 2A) from amphibolitized eclogites (Kimi-Smigada area),
ii) minute carbon cubes and octahedra included in garnet porphyroblasts (Figs. 2B, C) from grt-ky-st-chl schists and grt-bt-ky gneisses (Sidiro and Kimi-Smigada areas),
iii) rods (or needles) of silica, rutile, and apatite exsolved inside sodic garnet porphyroblasts (Figs. 3A, B, C, & D) in grt-bt-ky gneisses (Kimi-Smigada area)

As for the Sidironero Complex, UHPM indicators include:

i) MPQ aggregates included in garnet from garnet amphibolites (Pilima area) and, most importantly,
ii) minute carbon cubes and octahedra (Fig. 2D) included in garnet porphyroblasts in grt-bt-ky gneisses (Pilima area).

The preservation of coesite and microdiamond in UHP metamorphic crustal rocks is confined to rigid hosts such as garnet, clinopyroxene or kyanite and depends on various factors, the most important of which are the rate of exhumation, grain size of precursor coesite or diamond, fluid availability, and P-T conditions of superimposed retrograde metamorphism. Liou and Zhang (1996) have delineated six steps for the coesite to quartz conversion starting with the growth of fine-grained, thin, palisade quartz aggregates (within a single coesite grain), then proceeding to the development of polygonal coarser-grained quartz aggregates, and to final formation of a single quartz grain with either homogeneous or undulatory extinction. These authors have also stressed that positive identification of intergranular or matrix coesite in UHP metamorphic rocks strongly indicates the

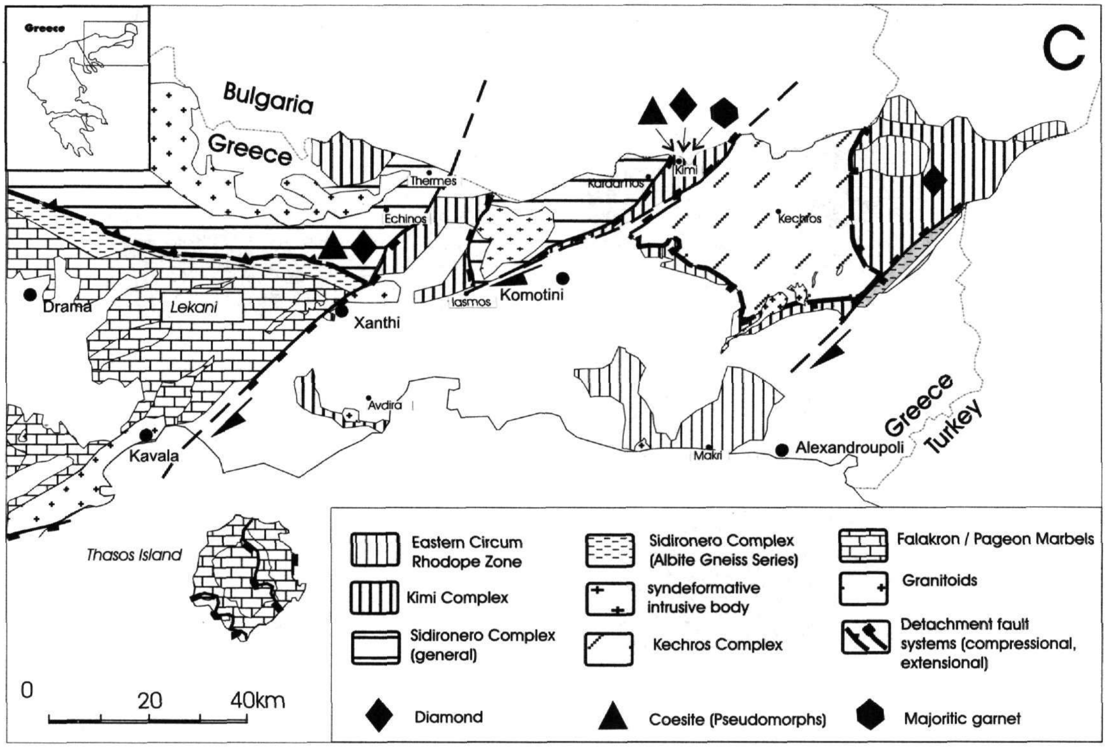

Figure 1: Geological map of central and eastern Rhodope (after Mposkos and Krohe, 2000, simplified) with locations of samples containing diamond, coesite, and majoritic garnet.

lack of fluids during rapid exhumation (see also Mosenfelder and Bohlen, 1997). Wain et al. (2000) have further elaborated on the morphology and significance of quartz aggregates resulting from coesite transformation. They emphasized that only polycrystalline radial quartz (PRQ) aggregates, which develop prior to radial fracturing and stress release, may be regarded as conclusive pseudomorphs after coesite, whereas polycrystalline polygonal quartz (PPQ; 10-50 grains per inclusion), multicrystalline polygonal quartz (MPQ; 2-10 grains per inclusion), and finally monocrystalline quartz develop by continued recovery post-dating radial fracturing and should be treated with caution. We have optically identified and subsequently verified by electron microprobe (EMP) analyses (pure SiO_2) MPQ aggregates as inclusions in garnet porphyroblasts in amphibolitized eclogites from the Pilima and Kimi- Smigada areas. Strong sub-radial cracks emanate from the inclusions (Fig. 2A).

As was the case with coesite, diamonds in RMP rocks were also initially identified optically and subsequently verified by both EMP analyses (pure carbon) and in situ laser Raman microspectroscopy. They appear as minute(average size ´10 μm) cubo-octahedral inclusions in garnet porphyroblasts in grt-bt-ky gneisses from the Kimi-Smigada, Sidiro and Pilima areas (Fig. 2B, C, D). Their mode of occurrence highly resembles that of diamonds in very similar rocks from the Kokchetav Massif, Kazakhstan (grt-bt±ky±cpx gneisses; Sobolev and Shatsky, 1990) and the Saxonian Erzgebirge, Germany (grt-phe-ky±bt gneisses; Massonne, 1998). Occasionally, the garnets are literally teeming with microdiamonds (~30 per 1.5 cm^2), a feature that has also been observed for the Erzgebirge garnets (Massonne, 1998). As mentioned above, in situ diagnosis of the physical state of carbon in selected samples was achieved through laser Raman microspectroscopy. First-order spectra were obtained at room temperature from 1000 to 1800 cm^{-1}. The Raman spectrum of diamond is dominated by a narrow, intense band observed at 1331-1337 cm^{-1}, which represents the main C-C bond vibration (Nasdala and Massonne, 2000). Despite consisting of virtually only one main band this Raman spectrum is highly typical of diamond and sufficient for its identification.

The Raman spectrum of graphitic material is a sensitive function of its degree of crystallinity. The latter is judged from the length of the crystallite along the α crystallographic direction, designated as L_α. Well-crystallized graphite is characterized by a single band at about 1580 cm^{-1} (order-peak [O]); disorder in graphite brings

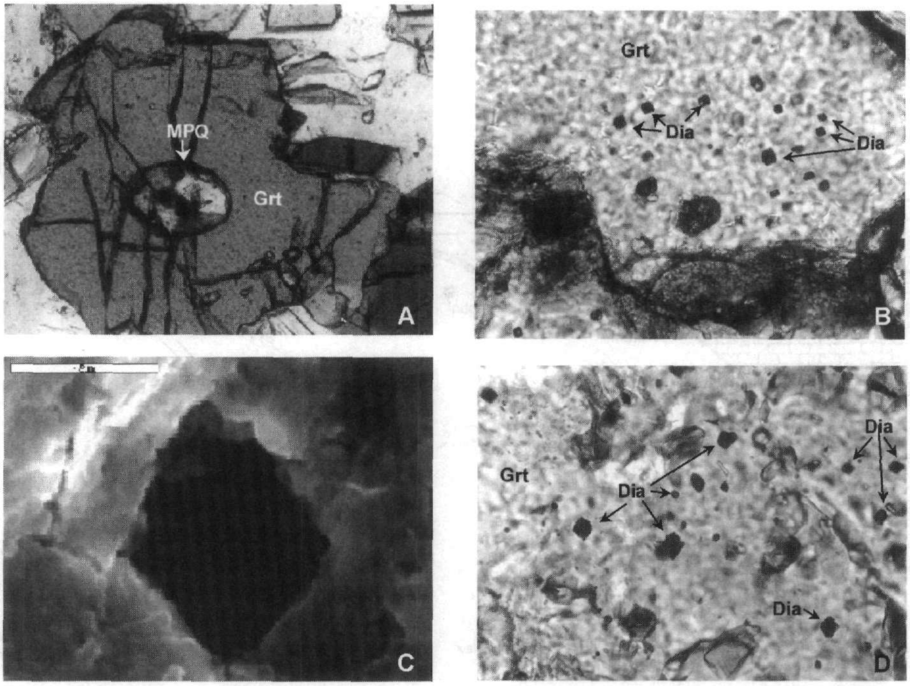

Figure 2

A: Photomicrograph of multicrystalline polygonal quartz (MPQ) aggregate pseudomorphs after coesite, included in garnet from amphibolitized eclogite of the Kimi area, Kimi Complex. Note the radial fracturing of the garnet host. Field of view: 0.9 mm; crossed polars.

B: Photomicrograph of inclusions of microdiamonds in garnet porphyroblast. Some of the diamonds show well-developed octahedral crystal faces. Grt-Ky-St-Chl-schist (Sample PS-11) Sidiro area, Kimi Complex. Field of view: 0.3 mm; polarizer only.

C: SEM image of idiomorphic diamond included in garnet porphyroblast. Grt-Ky-St-Chl-schist (Sample PS-11). Sidiro area, Kimi Complex.

D: Photomicrograph of inclusions of microdiamond in garnet porphyroblast. Some of the diamonds show well developed-octahedral crystal faces. Grt-Bt-Ky gneiss (sample XTH-1). Pilima area, Sidironero Complex. Field of view: 0.3 mm; polarizer only.

about broadening of the 1580 cm^{-1} band and shifts it towards higher wavenumbers because of the development of an additional band near 1360 cm^{-1} (disorder-peak [D]; Pasteris and Wopenka, 1991; Wopenka and Pasteris, 1993; Pasteris and Chou, 1998). The D:O ratio of both the maximum intensities and the peak areas of these two bands are sensitive functions of the L_a value of the graphitic material, i.e., the larger the D:O intensity or area ratio, the smaller the L_a and the more disordered the graphite.

Figure 4B shows the Raman spectrum obtained for a carbon octahedron enclosed by porphyroblastic garnet from a grt-bt-ky gneiss of the Pilima area, Sidironero Complex. Two narrow, intense bands become immediately apparent, one at 1334 cm^{-1}, the other at 1581 cm^{-1}, which are diagnostic of diamond and ordered graphite respectively. The total absence of a graphite disorder-peak suggests a high degree of crystallinity for the graphitized part of the diamond.

But perhaps the most important of our discoveries is the identification (by means of optical microscopy, EMP analyses and in situ laser Raman of quartz, rutile and apatite exsolution rods (or needles) in porphyroblastic garnet from grt-bt-ky gneisses of the Kimi-Smigada area, Kimi Complex. Similar exsolution textures (i.e., clinopyroxene, rutile, and apatite exsolution rods in garnet) have been reported only once in the literature (Ye et al., 2000) for eclogites - not metapelites – from Yangkou, Su Lu UHP metamorphic province, China. As is the case for the rods in the Yangkou eclogitic garnet, the rods in the Rhodope metapelitic garnet also occur in groups of parallel rods along crystallographically controlled planes of the host garnet, thus displaying an orthogonal or equilateral triangular pattern in cross-section (Figs. 3A, B, C); oriented apatite rods are also observed (Fig.

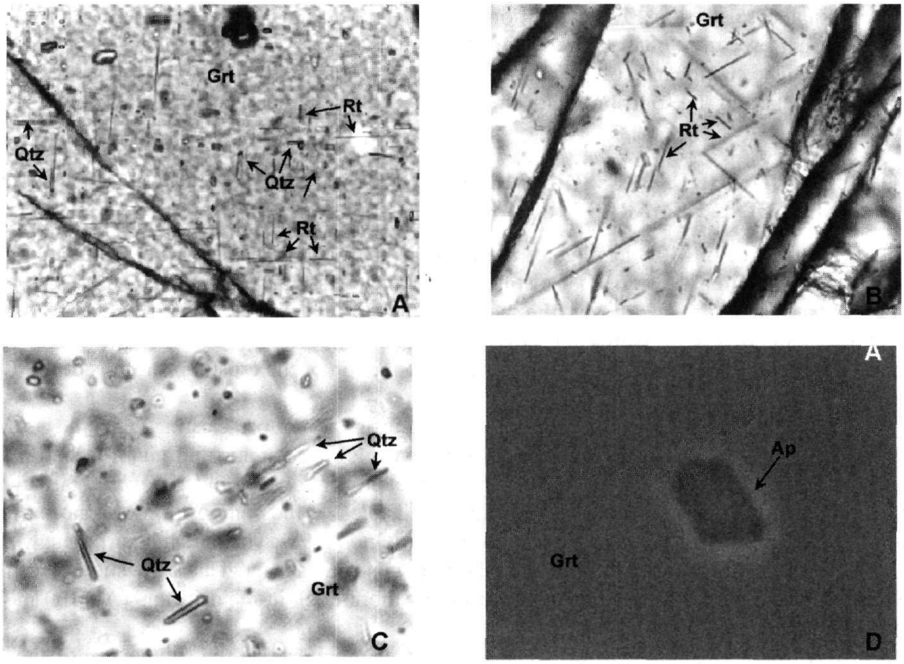

Figure 3

A: Photomicrograph of rutile needles and quartz rods exsolved from majoritic garnet. Note the preferred orientation of the exsolved phases in the host garnet. Grt-Bi-Ky-gneiss (Sample OS-3). Kimi area, Kimi Complex. Field of view: 0.4mm; polarizer only.

B: Photomicrograph of rutile needles exsolved from a majoritic garnet. The needles are oriented parallel to the octahedral faces of the host garnet. Grt-Bi-Ky-gneiss (Sample OS-4A). Kimi area, Kimi Complex. Field of view: 0.3 mm; polarizer + condenser.

C: Photomicrograph of quartz rods exsolved from majoritic garnet. Grt-Bi-Ky-gneiss (Sample OS-4A). Kimi area, Kimi Complex. Field of view: 0.4mm; polarizer + condenser.

D: SEM image of apatite inclusion in majoritic garnet, verified by electron microprobe analysis. Field of view: 0.023mm.

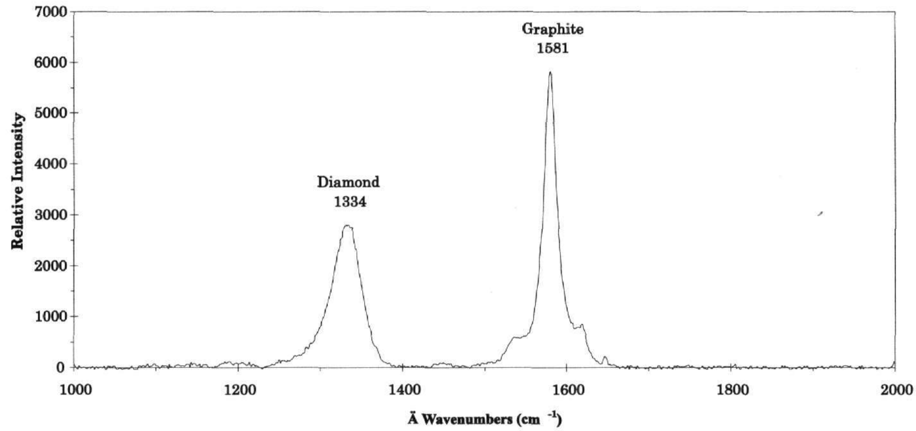

Figure 4: Laser Raman spectrum of octahedral carbon inclusions in garnet from Grt-Bt-Ky gneiss, Pilima area, Sidironero Complex (Sample XTH-1).

3D) as well as hexagonal basal sections. The exsolution of quartz, rutile, and apatite is taken here as indicative of the existence of a garnet precursor phase richer than normal in Si, Ti and P, that is, of a majoritic garnet (see Collerson et al., 2000). The significance of the presence of coesite, diamond, and majoritic garnet in Rhodope crustal metamorphic rocks and the contribution of these phases to evaluating peak pressures of metamorphism will now be addressed.

4. CONDITIONS OF ULTRAHIGH-PRESSURE METAMORPHISM IN THE RHODOPE METAMORPHIC PROVINCE – CONCLUDING REMARKS

In the RMP, the presence of MPQ aggregates as inclusions in porphyroblastic garnet from amphibolitized eclogites is attributed to the breakdown of pre-existing coesite and constrains metamorphic pressures to a minimum of 2.6-2.9 GPa at 600-900°C. Moreover, the mere presence of diamonds in porphyroblastic garnet from metapelites dictates metamorphic pressures in excess of 3.1-3.9 GPa at 600-900°C for the host rocks. Thus, at a first approximation, minimum depths of 80-140 km for the subduction of the crustal protoliths of the RMP can be inferred by using only mineralogical criteria.

An estimate as to the P-T conditions that prevailed during formation of pre-existing majoritic garnet in the RMP grt-bt-ky gneisses may be obtained from experiments on relevant systems and majorite barometry. We have conducted detailed EMP traverses of the rod-bearing garnet porphyroblasts and discovered rare unexsolved domains of the composition shown in Table 1. Also shown in Table 1 is the composition of a garnet produced experimentally at 7 GPa and 1100°C on a pelite composition by Ono (1998). The match is surprisingly good and suggests similar P-T conditions of formation for the Rhodope majoritic garnet. This suggestion is further reinforced by the results obtained (7 GPa) by applying the recently calibrated Si-in-majorite and (Al+Cr)-in-majorite barometers of Collerson et al. (2000).

Taken collectively, the above pieces of evidence conclusively demonstrate the subduction of the RMP to depths of the order of 220 km and establish the province as one of the most important UHP metamorphic belts in the world.

	Rhodope majoritic garnet	Experimental majoritic garnet
SiO_2	38.52	39.89
TiO_2	1.03	1.02
Al_2O_3	19.58	20.38
FeO	31.65	24.92
MnO	2.06	–
MgO	3.87	5.04
CaO	2.80	8.47
Na_2O	0.60	0.62
K_2O	–	0.11
P_2O_5	0.13	–
Total	100.25	100.45

Table 1: Electron microprobe analyses (wt.%) of majoritic garnet from grt-bt-ky gneiss, Kimi area, Kimi Complex (Sample OS-3), and of that produced experimentally at 7 Gpa / 1100 0C on a pelitic composition (Ono, 1998; run # sh23).

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