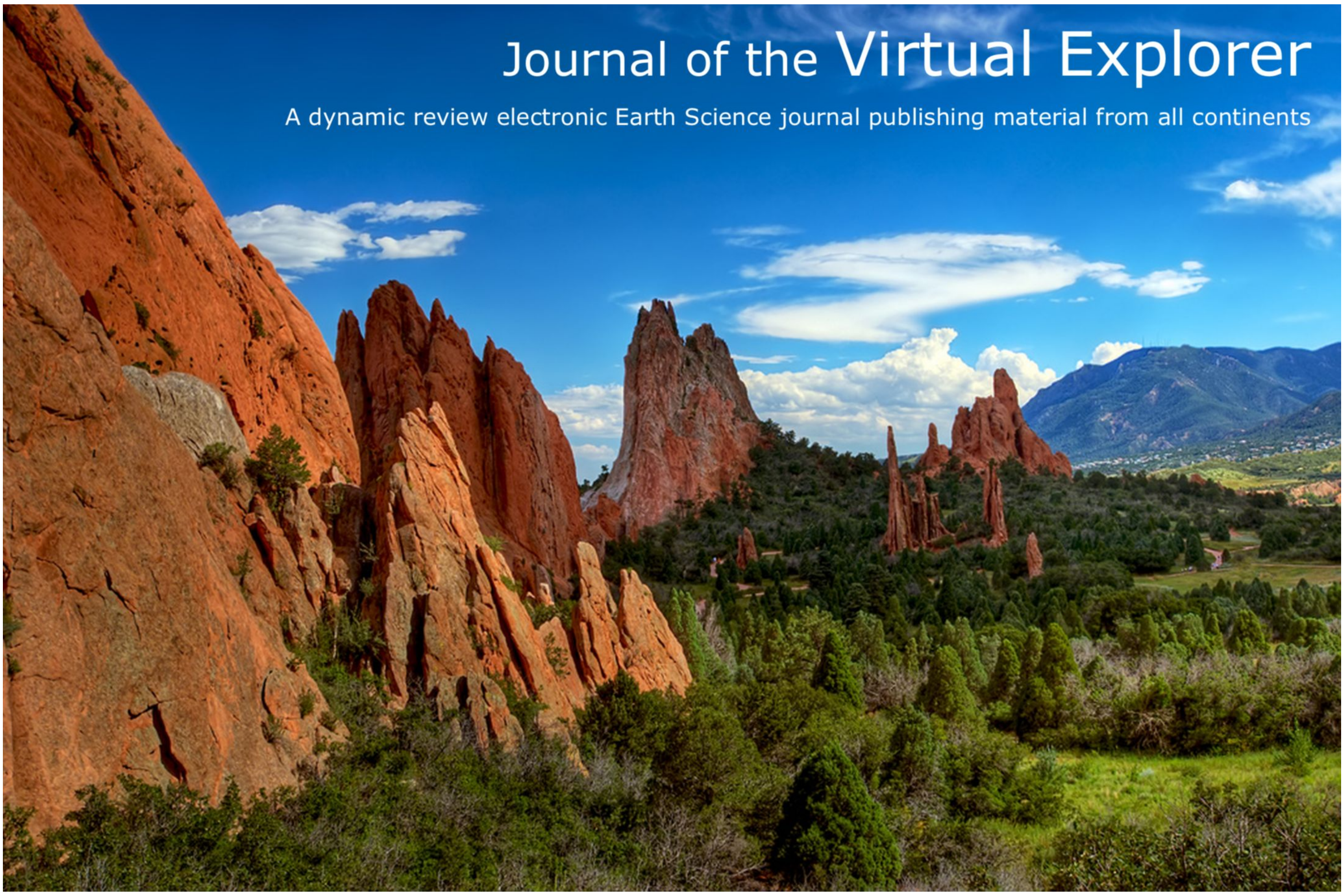

Ultrahigh-Pressure Minerals from Both Downwelling and Upwelling Environments: Snapshots of Mantle Convection on a Grand Scale

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Ultrahigh-Pressure Minerals from Both Downwelling and Upwelling Environments: Snapshots of Mantle Convection on a Grand Scale

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Abstract: The concept of Ultra-High-Pressure Metamorphism (UHPM) grew out of the discovery that blueschist minerals in metasediments required simultaneous high pressure and low temperatures that, in turn, required rapid travel to several 10s of km to grow the minerals in the first place, and similarly rapid return to the surface to avoid their reaction to greenschist facies. Both the trip down and the trip back had to occur faster than any known mechanism could provide. The birth of plate tectonics resolved this conundrum. Subsequent discovery of coesite and diamond in shallow protoliths extended the depth of implied subduction to > 150 km but the context remained transport of surface rocks on a scale small compared to mantle convection. Indeed, the presence of coesite and/or diamond or implication of their former presence became the working definition of UHPM. However, mantle rocks caught up in UHPM terranes also can carry evidence of very deep origin. Prior to that discovery, metamorphic petrology implicitly assumed that travels of rocks begin at or near the surface; solid-state transport upward of dense rock from significant depth was implicitly thought to be impossible. Over geological time, peridotite behaves as a fluid and is the medium by which the mantle of Earth convects, with the unavoidable implication that large volumes of such rock have “seen” the deep upper mantle and some must have circulated deep into the lower mantle. We now understand that these rocks can carry records of their travels in their microstructures. A simple example of this is the ubiquitous presence of pyroxene-spinel symplectites in spinel peridotites of ophiolites and mantle xenoliths, recording the former presence of garnet. A more complex example is exsolution of high-pressure pigeonite as lamellae at high angles to [001] in diopside, recording depths of ~400 km in the Alpe Arami peridotite in the Swiss Alps. UHPM terranes now have yielded “memory” in continental lithologies of almost that deep. Even more surprising, minerals recording comparable depths and a very reducing environment are now known from inside massive chromite of an ophiolite and the presence of diamonds in another demonstrates that this deep environment is not unique. The evidence that ophiolites form at Earth’s surface is overwhelming. Nevertheless, chromites within them show unambiguous evidence of a highly-reduced environment of great depth and high temperature. One of them contains coesite-after-stishovite and TiO_2 (II), as well as titanium and boron nitrides, with no evidence of down-pressure reaction. Lastly, we now know that some diamonds contain inclusions from the mantle transition zone and the lower mantle. What more can these rocks tell us? We discuss three things that point to much-needed new work in a variety of directions: (i) the harzburgites of ophiolites must be at least in part solid material that rose from great depth (carrying chromite) rather than the residue of melting from the basalts above them; (ii) continental material has been subducted to the base of the mantle transition zone and we suggest that ocean island basalts are generated there; (iii) Massive chromite carrying a mineralogical record of very high pressure either has a very deep origin or represents material from a previous Wilson Cycle that has been recycled.

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Introduction

Many aspects of metamorphic petrology had a delayed response to the plate tectonic revolution. The concept of isostasy was so powerful that the possibility of deep subduction of continental material (100s of km) was not considered seriously in the early years. A major exception was the interpretation of blueschists. W.G. Ernst (1963) in field and experimental studies of the Franciscan Terrane of California determined that glaucophane was stable only at low temperatures and “high” pressures, implying 10s of km depth. This discovery led him to surmise correctly that the rocks had to go down fast and back up equally fast and he tied the implied dynamics to the new concept of subduction (e.g. Ernst, 1971). This connection immediately solved the blue-schist conundrum – explaining how sediments could be subducted so quickly and coldly that glaucophane could become stable and return of this material to the surface in mélanges like the Franciscan Formation so quickly that glaucophane could be preserved. Nevertheless, there was a great resistance to acceptance that rocks could be transported to even these depths where such mineral assemblages could grow under equilibrium conditions and then be returned to the surface preserving those assemblages. As a consequence, the faulty concept of tectonic overpressure was created and continues to be appealed to today (see Green, 2005). Moreover, the possibility of much greater subduction of such rocks – or even exhumation of mantle rocks sufficiently fast that “memory” of very high pressure events could be preserved – apparently was little considered. There were exceptions but even eclogites and garnet peridotites were generally considered to represent depths of considerably less than 100 km (see the extensive review by Spalla *et al.*, 2010, and references therein).

The discovery of coesite (Chopin, 1984; Smith, 1984) and diamond (Sobelev and Shatsky, 1990) in continental collision terranes ushered in the new discipline of Ultra-High-Pressure Metamorphism (UHPM). Nevertheless, these rocks were considered to be rare outliers that probably represented depths just beyond the onset of stability of these pressure indicators. Indeed, discovery of microstructural evidence of exhumation from hundreds of kilometers (Dobrzhinetskaya *et al.*,1996; van Rohrmund and Drury, 1998; Bozhilov *et al.*, 1999) once again evoked disbelief. This Ultra-High-Pressure saga continues to evolve with numerous observations of coesite and diamond across the globe, including evidence of the former presence of stishovite in pelitic gneiss (Liu *et al.*, 2007b) that requires subduction of shale to more than 350km and their return to the surface carrying evidence of the voyage.

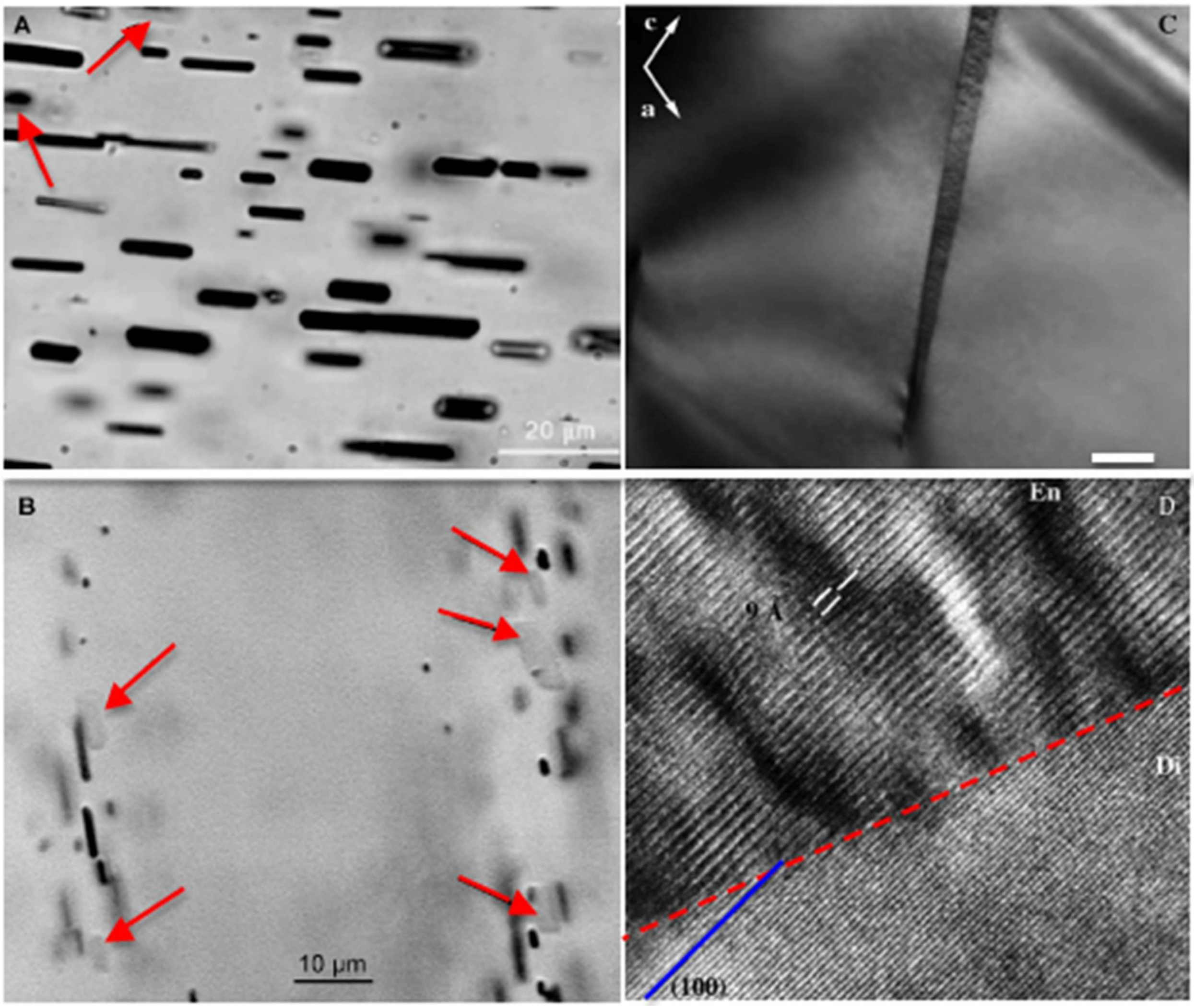
The latest turn of events in this field is verification of phases and microstructures indicating depths greater than 300 km in the mantle portion of ophiolites (Yang *et al.*, 2007; Dobrzhinetskaya *et al.*, 2009). Thus, to a degree never imagined until very recently, we know that rocks have been exhumed from more than 300km from parts of both the downwelling and upwelling arms of mantle convection. This paper briefly recapitulates a few of these discoveries, evaluates their implications, and looks toward the future.

Continental Collision Zones

Alpe Arami, Switzerland

There is a large and sometimes acrimonious literature concerning the first discovery of rocks exhumed from great depth. Microstructures in the Alpe Arami peridotite of southern Switzerland (Figure 1) reflect events that were originally proposed to have occurred at depths exceeding 300 km (Dobrzhinetskaya *et al.*, 1996; 2000; Bozhilov *et al.*, 1999; 2003) but that eventually could be ascribed to depths approaching 400 km (Liu *et al.*, 2007a). The critical observations are exsolution of ilmenite rods plus spinel flakes in olivine (Figure 1a,b) and exsolution of high-pressure clinoenstatite/pigeonite in diopside (Figure 1c) in the same rocks, in crystals separated only by 10s to 100s of microns. Although alternative interpretations of these observations were cited in rebuttal to the original publications, no alternative interpretations have been published that can offer a viable interpretation of both observations with a single alternative model. Although the olivine observations were center stage during most of the discussions in the literature, the pyroxene observations turned out to be definitive because a second instance of high-pressure clinoenstatite/pigeonite exsolution from diopsidic pyroxene in the Bixiling complex of the Dabie Mountains UHPM terrane in eastern China (Liu *et al.*, 2007a), coupled with high-pressure experimental studies (Tribaudino *et al.*, 2000; 2002; Nestola *et al.*, 2004), allowed unambiguous identification of the clinoenstatite lamellae in diopside as having been precipitated as the high-pressure polymorph.

Figure 1. Optical micrographs of ilmenite and chrome spinel precipitates



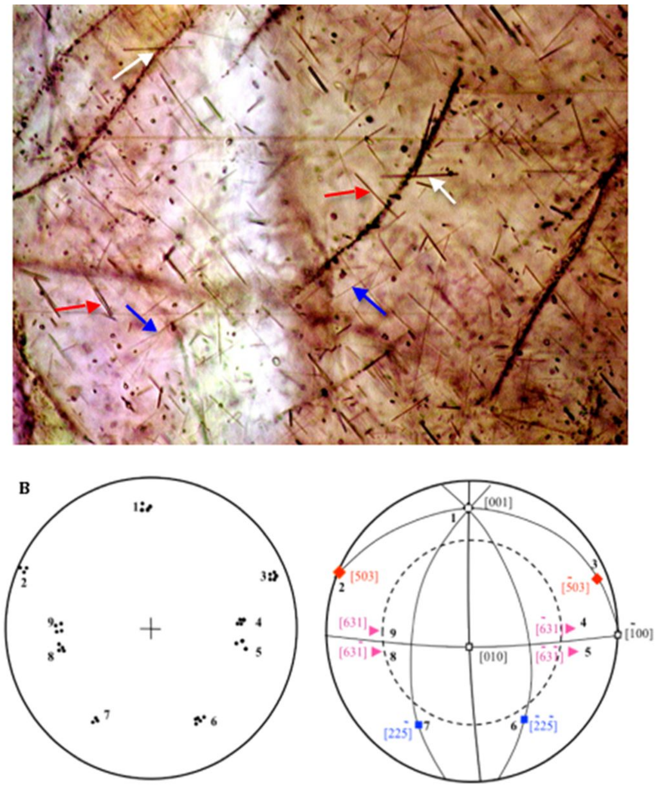
Optical micrographs of ilmenite and chrome spinel precipitates in first-generation Alpe Arami olivine (A&B) and Transmission electron micrographs of precipitates of high-pressure clinoenstatite/pigeonite in diopside (C&D) A. Ilmenite rods (black) are oriented parallel to [010] of olivine; spinel platelets (gray; difficult to see in this image) are oriented parallel to (100) of olivine. Two spinels that are in focus and attached to ilmenite rods are indicated by red arrows (see Dobrzhinetskaya *et al.*, 1996; Green *et al.*, 1997). B. Precipitates are in a few cases concentrated on (001) planes, but exhibit the same topotaxy as in A. In this image, virtually all ilmenite rods have a spinel platelet attached; arrows point out spinels without ilmenite (see Bozhilov *et al.*, 2003). C. Precipitate of clinoenstatite in diopside. Note large angle between c-axis of diopside and the lamella. D. High resolution image viewed down the b-axis showing (100) lattice fringes in both host (Di) and precipitate (En) and the boundary between them (outlined by red dashed line). Angle between blue and red lines is the angle between the lamella boundary and the c-axis of diopside. Short white lines show the ½ unit cell offset across antiphase boundaries in the lamella. The antiphase domains and the angle between the c-axis and the lamella unambiguously demonstrate that the originally precipitating phase was high-pressure clinoenstatite/pigeonite and that it precipitated at approximately 400 km (see Bozhilov *et al.*, 1999 and Liu *et al.*, 2007a for discussion). Panel A is modified after Figure 1 of Green *et al.* (1997); Panel B is modified after Figure 3C of Bozhilov *et al.* (2003); panels C&D are modified after Figure 1C and Figure 3 of Bozhilov *et al.*, 1999, respectively.

Realization that the pressure of exsolution could be estimated from the orientation of the lamellae then allowed deduction that the Bixiling complex (commonly interpreted to have originated as shallow ultramafic cumulates) was subducted to more than 300 km and returned to the surface and that the Alpe Arami peridotite was exhumed from perhaps as great as 400 km. In the case of Bixiling, the accompanying quartzofeldspathic host rocks have been determined to have been similarly exhumed from great depth based on abundant coesite inclusions in zircons. For Alpe Arami, the accompanying eclogites also have been shown to have a UHPM history (Dobrzhinetskaya *et al.*, 2002) but the surrounding Lepontine gneisses have not been systematically examined for zircon inclusions. Nevertheless, there is no alternative carrier for the garnet peridotites and eclogites included within the Lepontine gneisses.

Alten Tagh, China

Although it may be ambiguous whether or not the Bixiling ultramafic rocks have a low-pressure history that preceded their subduction to great depths, such is not the case for pelitic gneisses from the Alten Tagh UHPM terrane of western China in which rods of kyanite and spinel were precipitated in stishovite (Liu *et al.*, 2007b). In those rocks, abundant groups of rods of kyanite and spinel in polycrystalline quartz domains exhibit exsolution morphology but cross high-angle boundaries without deflection (Fig. 2a). The abundance of the rods is much more than could be dissolved in quartz or coesite but consistent with stishovite. Moreover, and most importantly, the rods in each polycrystalline quartz domain have orientations requiring that the host from which they precipitated had tetragonal symmetry (4/m 2/m 2/m) consistent with stishovite (Fig. 2b). The inferred topotaxy between the precipitates and host is also consistent with stishovite structure. The deduction that the rods precipitated within stishovite requires that the rocks began their return to the surface sufficiently deep that the solubility of kyanite and spinel components could exsolve at the amounts observed before leaving the stishovite stability field. That, in turn, requires that the upward journey began at 350km or more.

Figure 2. Optical micrograph of kyanite and spinel needles



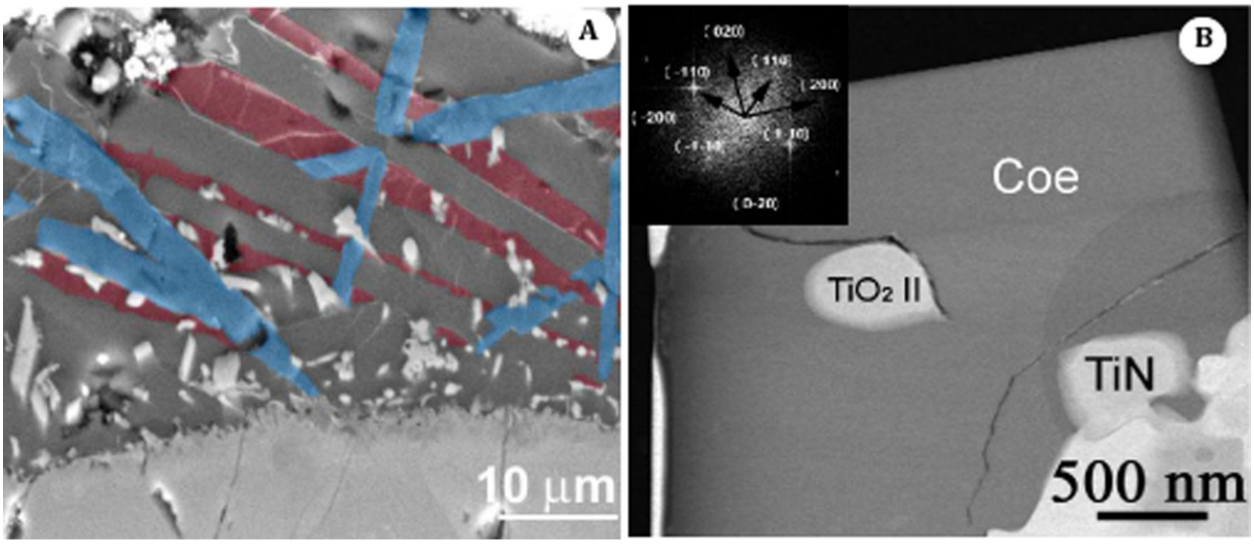
Optical micrograph of kyanite and spinel needles crossing high-angle quartz grain boundaries (A) and stereographic projections of needle orientations (B). (A) Bright band is a left-dipping high-angle grain boundary viewed between crossed polarizer's (see Liu *et al.*, 2007b for the EBSD-measured orientations of these two crystals) showing multiple sets of needles crossing the boundary undisturbed (most abundant group trends NE (red arrows); two other groups shown by white and blue arrows). There are more groups; can you find them? (B) Left diagram shows that needle orientations form tight clumps. The most abundant orientation (#1) is assumed to be the c-axis because that is the only unique direction in tetragonal crystals. Right diagram shows that the 9 groups imply 4 mirror planes through the c-axis and one normal to the c-axis, establishing that the rods exsolved from a host with tetragonal symmetry. Dotted circle marks 30° to the thin section, beyond which Universal stage measurements cannot establish orientations (See Liu *et al.*, 2007b for further explanation). Figures modified after Liu *et al.* (2007b).

Ophiolites

The Luobusa ophiolite, Tibet, lies along the suture between Asia and India. It was emplaced early in the Tertiary epoch (Aitchison *et al.*, 2002). Although faulted and moderately serpentinized, it consists primarily of harzburgite and dunite and exhibits minor pillow basalts and cherts (Zhou *et al.*, 1996). It has been interpreted as having formed at a mid-ocean ridge and subsequently modified in the mantle wedge above a subduction zone (Malpas *et al.*, 2003). It shows no evidence of either subduction-zone metamorphism or meteoritic impact.

Since the 1980s there have been periodic publications reporting diamonds and a bewildering array of other phases including silicon carbide and exotic metals (see Yang *et al.*, 2003 and Robinson *et al.*, 2004 and references therein) extracted from a massive chromite deposit found at an altitude of about 5,000 meters. The overwhelming evidence that ophiolites form at oceanic spreading centers has led to extreme reluctance of the community to accept that the reported materials, not found in place but only in heavy mineral separates from massive chromite, are real, indigenous, minerals of the ophiolite. However, Yang *et al.* (2007) described a diamond inclusion within a grain of OsIr alloy and a pellet of Fe-Ti metal to which is attached a small fragment of silicate rock consisting primarily of coesite and kyanite (Figure 3a). Loose diamonds previously reported from Luobusa chromite had essentially been rejected as probable contamination. The presence of a diamond included within a grain of OsIr alloy was more convincing as indigenous to the rock because many OsIr pellets have been identified in intact specimens of the massive chromite ore. Most importantly, however, was the identification of coesite by Raman spectroscopy that finally presented an observation from these remarkable rocks that is impossible to reject as contamination. The presence of coesite requires that the chromite crystallized at a depth in excess of 90 km and diamond extends that depth to a minimum of ~120 km. Further, the coesite exists as elongated prisms that strongly suggested pseudomorphic replacement of stishovite, which would imply much greater depth; demonstration that the elongated prisms of coesite are polycrystalline, greatly enhanced the stishovite interpretation.

Figure 3. Ultra-High-Pressure minerals found in massive chromite of Luobusa Ophiolite, Tibet.



(A) SEM false-color image of prisms of coesite (gray) and kyanite (blue) with interstitial unknown amorphous phase (red) attached to pellet of Ti-Fe alloy (light gray). (Modified after Yang *et al.*, 2007). (B) TEM bright field image of FIB-prepared foil of material in (A): A single crystal of TiO_2 II is included in coesite (Coe); at the lower right corner are inclusions of TiN (osbornite). The insert is an electron diffraction pattern of the crystal of TiO_2 II calculated from a high-resolution image by Fast Fourier Transform (Modified after Dobrzhenitskaya *et al.*, 2009).

Additional investigation of this specimen by transmission electron microscopy (TEM) by Dobrzhinetskaya *et al.* (2009) confirmed the presence of coesite. Furthermore, included within the coesite is abundant titanium nitride (osbornite) and the cubic (high-pressure) form of boron nitride (BN). This was the first finding of nitrides in mantle rocks. Osbornite has been found only once previously on Earth but is common in meteorites, especially iron meteorites; BN had not been found in nature previously. Lastly, and perhaps most importantly, two observations point to a very high pressure history of the chromite. The coesite also contains inclusions of: (i) TiO_2 II, the high-pressure polymorph of rutile with the *alpha* lead oxide (αPbO) structure (Figure 3b); (ii) native iron.

The presence of TiO_2 II confirms the earlier implication that the coesite in this rock originally crystallized as stishovite. That is because the phase boundary between rutile and its high-pressure polymorph has a very steep positive slope; the intersection of that boundary with that for coesite-stishovite occurs at ~9.2 GPa, 1220˚C (Withers *et al.*, 2003). A likely minimum temperature for upwelling upper mantle material beneath an ocean ridge is 1300˚C, which would require a depth in excess of 300 km to stabilize TiO_2 II, well into stishovite stability. The presence of native iron is also critically important because it requires very low oxygen fugacity. Nevertheless, the ferric/ferrous ratio in the host massive chromite is elevated whereas the nodular and disseminated chromite of the ophiolite have normal Fe^{3+} content (Ruskov *et al.*, 2010). This paradox is resolved by the hypothesis that this is the first naturally-occurring example of disproportionation of ferrous ion into ferric ion plus native iron at very high pressures induced by the mineral assemblage being dominated by phases that have no place in their structures for the ferrous ion (see extended discussions in Dobrzhinetskaya *et al.*, 2009 and Ruskov *et al.*, 2010).

A critical question arises following the remarkable observations described in the four previous paragraphs: “Is the Luobusa ophiolite unique in displaying a high-pressure highly reduced mineral assemblage in Fe^{3+}-rich massive chromite or is there a subset of ophiolites that carries this very high-pressure signal?” The data to answer this question remain few but they are sufficient to confirm that the Luobusa ophiolite is not unique. Diamonds and silicon carbide have been reported from an ophiolite in the polar Ural mountains (Trumbell *et al.*, 2009) and several ophiolites display elevated ferric/ferrous ratios in massive chromite (see references and discussion in Ruskov *et al.*, 2010).

Discussion

Recent studies of Ultra-High-Pressure Metamorphism have replaced xenoliths from kimberlites as the source of the deepest rocks brought to Earth’s surface. Here we have briefly described both a mantle peridotite that apparently has experienced a one-way trip to the surface from about 400 km and a shale that was subducted to more than 350km and returned. Depths at least as great have been suggested in the form of pyroxene exsolution rods in previously majoritic (super-silicic) garnets from the Western Gneiss region of Norway (Spengler *et al.*, 2005). To that is now added potentially equal depths sampled in the upwelling of mantle beneath ocean spreading centers. The only material that has come to the surface with evidence of deeper origin than these examples of the mantle convection system are inclusions in diamonds from the mantle transition zone and lower mantle (Hayman *et al.*, 2005; Wirth *et al.*, 2007; Kaminsky, 2012 and reference therein).

What can we infer from this new direct evidence of mantle convection? The first is that at least some of the harzburgites in ophiolites do not represent the residue of melting left over from generation of the basalts that overlie them. Chromite is a very dense material. Thus, the only way the massive chromites carrying a very deep signal can have risen from great depth is if they were carried by a larger volume of less dense but strong rocks. The only possibility is the harzburgite in which the chromite is embedded (exactly analogous to the UHPM eclogites and peridotites being carried by the quartzofeldspathic rocks that surround them). Indeed, diamonds have recently been found in the harzburgites of Luobusa (J. Yang, personal communication, 2010). This change in understanding of the mantle portions of ophiolites opens a window for further prospecting for other deep mantle mineralogy.

The second implication we can draw is that if continental material, even sediments, can reach depths in excess of 350 km and return, there must have been other cases in which the “point of no return” was reached when upper continental crust becomes more dense than ambient mantle and never returns (Irifune *et al.*, 1994; Dobrzhinetskaya and Green, 2007; Liu *et al.*, 2007b; Wu *et al.*, 2009). In particular, Irifune *et al.* (1994) and Wu *et al.* (2009) showed that if the point of no return is reached by such lithologies, they would fall to the base of the mantle transition zone (MTZ) but not into the lower mantle. Evidence of chemical components suggestive of continental sediments and crust is recorded in ocean island basalts (e.g. Rapp *et al.*, 2008). The materials from Luobusa may be evidence of return of “partially digested” upper mantle material. The dominant chemical components of the high-pressure material from Luobusa discussed here are SiO_2 and Al_2O_3 and surprisingly include boron. These major components strongly suggest an upper crustal origin and the presence of boron implicates sediments. However, the Nitrogen isotopes of this rock are incompatible with a crustal origin (Dobrzhinetskaya *et al.*, 2009) and therefore most likely represent a component added from the lower mantle. Thus, the simplest explanation of all of these observations would appear to be that some continental material gets subducted to the base of the MTZ where it can be contaminated by at least volatiles from the deeper mantle. Furthermore, ocean island basalts probably are either generated at that depth or they interact with such contaminants on their way to the surface.

Lastly, the question arises as to whether chromitites, which are only found in opholites, are generated at great depth rather than at shallow depth as is currently understood. Or could chromitites like those in Luobusa be, like some of their included constitutents, relics of a previous Wilson Cycle? In either case, they provide immediately a point for prospecting for deep materials. The role of ophiolites in providing information about the deep mantle has only begun to be investigated. Modeling studies are proving productive in creating viable scenarios of subduction and exhumation guided by these mineralogical discoveries in continental collision terranes (see for example Gerya *et al.*, 2002; Roda *et al.*, 2010) and very likely will provide additional insight into the dynamics of mantle upwelling as well.

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