Mantle eclogites: evidence of igneous fractionation in the mantle

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Despite recent geochemical studies advocating an origin of eclogite xenoliths in kimberlite by subduction of altered sea-floor basalts and crustal cumulates, there are numerous examples showing abundant exsolution of garnet and kyanite from clinopyroxene that could only have occurred on cooling from near-solidus temperatures at pressures in excess of 3 GPa. Samples showing extensive exsolution tend to have more calcic garnets and sodic pyroxenes than the average eclogite, but do not form a group that is geochemically distinct in major, minor, or trace elements. Also, composite samples containing both kyanite and biminaric eclogites demonstrate the co-genetic relation of extremes of composition of the eclogite suite. Accordingly, we present an igneous fractionation model to account for the major and trace element variations observed in these rocks: grosspydites and kyanite eclogites are early pyroxene, or pyroxene-plus-corundum, cumulates that have undergone extensive subsolidus exsolution of kyanite and garnet. These were then followed by more Fe-rich eclogites which had pyroxene plus garnet as solidus phases and fractionated towards more magnesian biminaric eclogites.

1. Introduction

Together with alpine-type peridotite and eclogite bodies and abyssal peridotites, xenoliths in kimberlites and alkali basalts provide the only natural samples of rock-types from the upper mantle (30–200 km). Of these, the xenolith suite in diamondiferous kimberlites, especially those of Southern Africa, appears to have equilibrated at the greatest depths (pressures up to 6.0 Gpa and temperatures of 1050–1250 °C), and to have been exhumed with the minimum of retrograde reactions. This suite of xenoliths is dominated by olivine-rich rocks (depleted garnet peridotites, lherzolites, and harzburgites) which account for 90% or more of all inclusions. Less than 10% of the suite is made up of eclogites and related kyanite eclogites and grosspydites (grossular-pyroxene-kyanite rocks), and there are no samples intermediate in composition between the eclogites and the olivine-bearing suite. However, if the inclusions are representative of the region of the mantle from which they were derived, then most of the “fertile” component, that from which basaltic melts could be derived, is in eclogites and not in fertile lherzolites or harzburgites. The eclogites display a range of major, minor, and trace element and isotopic compositions, and an understanding of the causes of these variations is essential for a comprehensive view of the origin and evolution of silicate liquids in the Earth.

The general features of mantle eclogites have been reviewed [1,2]. Two major hypotheses have been advanced to account for chemical variation in eclogite samples that appear to have equilibrated within the upper mantle (temperatures ~ 1050 °C and pressures > 3 GPa): (1) a high-pressure cumulate magmatic origin, and (2) subduction of crustal cumulates and oceanic basalts that had been affected by varying degrees of near-surface alteration prior to subduction. Several recent studies [3,4] have favored the latter interpretation citing geochemical evidence that the samples may be subducted Archean oceanic crust.

Whatever the ultimate origin, the samples have undergone substantial subsolidus re-equilibration and recrystallization which obscures many lines of petrographic evidence. Nevertheless, in many samples significant textural evidence in the form of garnet and kyanite exsolution from pyroxene is

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preserved which strongly indicates a high-pressure igneous origin at least for the more calcic compositions. Further, composite samples spanning large compositional ranges demonstrate the co-genetic relationship of the varieties of eclogite. The objectives of this paper are to describe the petrographic, mineralogical, and geochemical characteristics of the eclogite suite, to review the evidence for a high pressure igneous origin, and to present a model for an igneous fractionation process to account for the major and minor element variations observed.

2. Major element trends

The composition of coexisting garnet and clinopyroxene from 65 eclogites, kyanite eclogites, and grospydites from Bellsbank, South Africa are displayed in Fig. 1. The figure shows that there is a continuous compositional trend from the grospydites to kyanite eclogites, to bimineralic eclogites, and that the grospydites, either with accessory corundum or with accessory coesite and sanidine, are compositionally continuous with the rest of the group in major elements. The crossing tie line is likely due to the elevated alumina content of the pyroxene equilibrated with kyanite. The eclogites that plot to the right in the figure have low-Ca garnets, and fairly high Mg(Mg + Fe) ratios in both garnet and clinopyroxene. Moving to the left, the garnets increase in Ca and generally also in Fe. The pyroxenes also increase in Ca, but generally also in Na, and lift off the bottom of the ACF diagram with increasing grossular content of the coexisting garnet. It is CaEs (Ca-Eskola, CaAl$_2$Si$_4$O$_{12}$) and CaTs (Ca-Tschermaks, CaAl$_2$SiO$_6$) components of the pyroxene that causes them to plot off the bottom of the diagram. Kyanite coexists only with garnets that exceed about 35 mol% grossular and pyroxenes with considerable CaTs and CaEs components. The grospydites typically have fairly high-Na pyroxenes (jadeite > 50 mol%) and commonly have accessory corundum or coesite and occasionally sandine. Among the South African eclogites, coesite, corundum, and sandine are common only in grospydites, and do not generally occur with low-Ca garnets in these high-pressure eclogites. However, coesite-, quartz- and sandine-bearing eclogites of broad range of compositions have been reported from other localities [6].

In major element composition, the bimineralic eclogites are similar to oceanic tholeiites and high-alumina basalts, and typically contain small amounts of olivine in the (CIPW) norm (but, no modal olivine). A coesite grospydite [7] contains about 70 mol% normative plagioclase with about 15% each of olivine and diopside. MacGregor and Manton [3] have postulated that such rocks (i.e. kyanite eclogites and grospydites) may be subducted feldspathic cumulates from an Archean oceanic crust. Whereas major element compositions of coesite grospydites may resemble anorthosites, the corundum grospydites do not correspond to any known crustal rock type, being much too low in K and high in Na to correspond to pelitic sediments. Bulk compositions exceed 28% Al$_2$O$_3$, well outside the range for altered lavas (spilites) [8]. Also grospydites have been reported with both high (> 0.07 wt%) [7] and low (< 0.07 wt%) [9] K$_2$O contents of pyroxenes.

3. Exsolution

There is substantial evidence of abundant exsolution of garnet from clinopyroxene in these rocks (Fig. 2), and recent experimental studies discussed below suggest that this took place on a decrease in temperature rather than an increase in
Fig. 2. A photomicrograph of exsolution texture of garnet from clinopyroxene in an eclogite from Roberts Victor showing approximately 40% volume exsolution.

Fig. 3. A photomicrograph of exsolution of both garnet and kyanite from clinopyroxene in a corundum grosspydite from Bellsbank (Bobbejaan Mine).
pressure. Harte and Gurney [10] discuss extensive garnet exsolution from pyroxene as evidence of a high-pressure igneous origin. The exsolution is significant not only to the petrogenesis of these rocks, but also because it has a first-order effect on the densities of the rocks, so that it will likely control the convection dynamics of an eclogite-rich region of the mantle [5,9].

Exsolution of both kyanite and garnet (Fig. 3) has been noted by several authors [9,11,12]. X-ray precession studies have shown that the structures are epitaxial [9], and the boundaries have been shown to be coherent with high-resolution transmission electron microscopy (HRTEM) [13]. This is strong evidence that the garnet and kyanite exsolved from a precursor pyroxene and that all three phases are not the breakdown products of some other precursor phase such as feldspar. Kyanite has a cation/oxygen ratio of 0.60, whereas pyroxene and garnet both have ratios of 0.67. Therefore kyanite exsolution from pyroxene is evidence that the precursor pyroxene contained substantial Ca-Eskola end-member. This end-member is unstable below about 3 GPa [14] and is stabilized by high temperature (i.e. near solidus), and anomalously high-Al pyroxenes have been noted as liquidus phases in some experimental studies [14,15]. Because equilibration conditions of these rocks are 3–6 GPa and 1050–1250 °C, based on Fe-Mg and Al distributions between garnet and clinopyroxene [16,17], there is not sufficient pressure increase between the minimum stability of the GaEs end-member and the final equilibration pressure of the rock to account for the large volume of garnet and kyanite exsolution observed. The exsolution must therefore be primarily due to a decrease in temperature rather than an increase in pressure. The exsolution is, therefore, strong evidence that these kyanite eclogites and grossopydites showing exsolution were once near liquidus temperatures at pressures in excess of 3 GPa. The precursor pyroxene has very nearly the same composition as the entire grosspydite [9,18]. The crystal chemistry of these pyroxenes has been explored by McCormick [19].

4. Composite samples

Banded eclogites, samples containing two or more chemically distinct eclogites, have been described by several authors [13,20,21]. The significance of these samples is that they firmly establish the co-genetic relationship between grossopydites and bimineralic eclogites. McCormick [13] has described two composite eclogites that are compositionally banded samples in which grossopydites abut bimineralic eclogites. An ACF plot of the coexisting phases is shown in Fig. 4. This figure shows the chemical continuity of grossopydites with other eclogites. The range in pyroxene compositions spans the gap between the Group B and C eclogites defined by Shervais and co-workers and thought to be compositionally distinct [4,22]. The compositional gap seen by these authors may possibly be the result of slightly different pyroxene compositions for those equilibrated with kyanite and those not.

5. Trace element trends

Rare earth analyses of whole rocks and mineral separates from these rocks confirm the chemical continuity of the sequence. Representative whole-rock analyses either obtained directly or by reconstruction from cleaned mineral separates are shown in Fig. 5. The lack of LREE enrichment in the grosspydite is evidence that there is no contamination of the samples by kimberlite fluids. The bimineralic samples are generally flat, at 5–15 ×
Fig. 5. A plot of the rare earth concentration relative to chondrites for a grospydite (\(\times\)) [23], a bimineralic eclogite (\(\bullet\)) [23], and a kimberlite (\(\ast\)) [24]. Also shown is the rare earth concentration of a natural clinopyroxene relative to a lava (+) [24]. The similarity of the latter pattern with that of the grospydite is consistent with the proposed model for the origin of grospydites as igneous accumulations of aluminous pyroxene at high pressures.

chondritic and show little or no Eu anomaly. These rocks resemble MORB in trace as well as major elements. The grospydites, however, are distinctive in that they are enriched (about 10–20 \(\times\) chondritic) in the middle rare earths, Sm, Eu, and Gd, but depleted in both light and heavy REE. The corundum grospydites are similar to the coesite grospydite which suggests that they result from a similar process. Exsolution features discussed above suggest that this process is accumulation of an anomalous, liquidus, hyperaluminous pyroxene, with or without corundum, followed by equilibration to lower temperatures with exsolution of kyanite, garnet, and possibly silica. Middle rare earth enrichments have been noted by several authors in both experimental and natural clinopyroxenes, and one such determination for a natural clinopyroxene relative to host liquid is plotted in Fig. 5.

6. Oxygen isotope studies

Bimineralic mantle eclogite \(\delta^{18}O\) values show a range of +2.2 to +8.5 [3,25] which is much greater than that shown by either unaltered MORB or mantle-derived peridotites [26,27], but smaller than that shown by ophiolites. Mantle igneous processes are generally believed to be too high in temperature to give rise to significant fractionation of oxygen isotopes [28]. This has prompted several authors [3,29,30] to suggest that the range of \(\delta^{18}O\) values requires a low-temperature process so that at least some of these xenoliths represent metamorphosed, hydrothermally altered, seafloor basalts which show a range of +2.0 to +20 [29]. The kimberlites, themselves, show a range in \(\delta^{18}O\) values of +2 to +12 [31], but this may be due to post-emplacement deuteric alteration. Many eclogites show disequilibrium in oxygen isotopes between primary and apparently high-pressure secondary phases, so there may be some degree of oxygen isotope heterogeneity in the subcontinental upper mantle.

There is an insufficient number of oxygen isotope data from grospydites and kyanite eclogites to base any firm conclusions as yet, however it appears that the spread in oxygen isotopic ratios may be too great to be the result of mantle igneous processes alone, and a more complex history involving low temperature processes is suggested. Such low temperature processes may be either in the derivation of the metasomatic fluids that have affected the eclogites within the mantle or in post-emplacement alteration. Also, oxygen isotopic data on grospydites for both whole-rocks and clean mineral separates, particularly of secondary phases if possible, would help to clarify this situation. We are in the process of obtaining these latter data and some preliminary results have been reported [32]. Recent results of Smyth and Clayton [33] in prediction of oxygen isotope fractionation based on electrostatic site potentials computed from crystal structure offer considerable hope for clarification of oxygen isotope fractionation in the mantle.

7. Radiogenic isotope studies

There have been several studies of radiogenic isotopes in mantle eclogites, but none so far has included kyanite eclogites and grospydites. Manaton and Tatsumoto [34] investigated the U-Pb and Rb-Sr of Roberts-Victor eclogite whole rocks and concluded that the model ages of the rocks were Archean, but that secondary phases might have affected the results. Kramers [35] also studied these isotope systematics from whole-rocks and carefully prepared mineral separates and con-
cluded that the ages were Archean, but that the mineral systems had remained open to these isotopes during the histories of the rocks. This is not surprising for rocks equilibrated at 1050–1250 °C. Jagoutz et al. [30] and McCullough [36] investigated the Sm-Nd systematics of several Roberts-Victor eclogites and also concluded that the ages were Archean at 2.5–3.5 Gy, but that the systems had remained open to these isotopes during their histories. MacGregor and Manton [3] have used the Archean ages to infer that the Roberts-Victor eclogites are subducted Archean oceanic crust. Shervais et al. [4] have found unusual Nd isotope ratios (εNd > 200) in some bimodalic samples which they infer to be the result of metasomatic alteration at low pressures.

8. Discussion

Two major hypotheses have been advanced to account for the chemical and mineralogical variations observed in these rocks: (1) high-pressure igneous fractionation, and (2) subduction of low pressure cumulates that have been affected by varying degrees of near-surface alteration.

Citing predominantly petrographic and major element compositional evidence, several authors have advocated a high-pressure igneous origin for these samples. MacGregor and Carter [37] and Hatton and Gurney [21] describe textures suggesting a high-pressure cumulate origin consistent with high-pressure experimental studies [11]. Harte and Gurney [10], Smyth et al. [9], and Lappin [18] cite exsolution phenomena in clinopyroxenes to suggest that some of the more calcic eclogites have recrystallized from near solidus temperatures at pressures in excess of 3.0 GPa. These studies have considered the grospydites and kyanite eclogites to be late stage differentiates of this high-pressure igneous fractionation. If derived by igneous processes at depth, the eclogite series would be evidence of basaltic melt at depths greater than 200 km and would provide clues to the igneous fraction of these melts at upper mantle pressures.

The principal shortcomings of this hypothesis are that it fails to account for the lack of trend in Fe enrichment in the grospydites, and it fails to account for the wide variations in oxygen isotopic ratios which appear to require low-temperature processes.

Alternatively, citing predominantly geochemical evidence [30,38,39] or studies of North American kimberlite nodules [40–42], other authors have interpreted some of these rocks as possible subducted relicts of an Archean oceanic crust. MacGregor and Manton [3] have further postulated that their group I and group II eclogites from Roberts Victor correspond to subducted oceanic crustal cumulates and altered oceanic basalts respectively. This interpretation is based on a review of major and trace element compositions of whole-rocks and some mineral separates plus oxygen, Sm-Nd, and Rb-Sr isotope studies. The group I and II classification was originally proposed on the basis of texture with group I being high pressure cumulates and group II being fractionated liquids. Revising the interpretation [3] the two types were found to be geochemically distinct, with group I (subducted crustal cumulates) having potassic pyroxenes (> 0.07 wt% K2O) and δ18O values slightly above the mantle average, whereas group II (subducted altered basalts) have low-K pyroxenes and δ18O values below the mantle average. By this classification the grospydites and kyanite eclogites would correspond to subducted feldspathic cumulates (group I).

Some of the gropsydites, especially corundum-bearing varieties, do not readily fit into this classification. Helmstaedt et al. [40] suggested that the grospydites may be subducted equivalents of rodingites which are Ca- and Al-rich rocks resulting from near-surface metasomatic alteration of ultramafic and mafic rocks. However, the major element trends observed with rodingitization appear to be quite distinct from those in going from bimodalic eclogites to gropsydites. Rodingitization, as observed in Alpine mafic and ultramafic rocks [43], results in strong enrichment of CaO (up to 28 wt.%) with depletion in alkalies, particularly Na, and reduction of silica (down to 36–40 wt.%). Gropsydites, however, typically have CaO contents of 12–14 wt.%, are not depleted in Na having pyroxenes with more than 50 mol% jadeite plus occasional primary sanidine, and silica contents of 45–52 wt.%.

The major shortcomings of this second hypothesis are that it requires a different origin for corundum gropsydites whereas these appear to be genetically related to the sequence, and it fails to
account for the evidence that many of the samples have re-equilibrated from near-solidus temperatures at pressures in excess of 3 GPa.

Previous authors advocating a high-pressure igneous origin have suggested that the grospydites result from evolved eclogitic liquids. We are here postulating that these rocks may be the result of an early accumulation of liquidus pyroxenes followed by re-equilibration to lower temperatures at pressures in excess of 3 GPa. This is based on several lines of evidence. First, the reconstructed pyroxene compositions approach the whole-rock grospydites in major element composition. Second, experimental studies [15] have found that garnet does not readily nucleate in experimental runs, so that a hyperaluminous pyroxene is the liquidus phase. Third, the high Mg/(Mg + Fe) ratios of the grospydites favor an early, rather than a late, formation of these pyroxenes. Fourth, this model would restrict grospydite compositions to the mantle which would be consistent with there being no crustal equivalents to most of these compositions. Fifth, the peculiar rare earth patterns of the grospydites are consistent with an igneous accumulation of pyroxene, and do not require plagioclase accumulation to explain the slight positive Eu anomalies.

The igneous process involved appears to be accumulation of a liquidus aluminous pyroxene with or without corundum. Subsequent re-equilibrium results in exsolution or evolution of substantial amounts of garnet, kyanite, and silica. If corundum was present, the evolved silica reacts with it to form additional kyanite resulting in the residual primary corundum being restricted to inclusions in kyanite or garnet, but never in grain boundary contact with pyroxene. If corundum were not present, the evolved silica may form coesite, although coesite has not been seen in exsolution relationship to the pyroxene. The early pyroxenes have very high Mg/(Mg + Fe) ratios and the peculiar middle-rare-earth enrichment observed in both the corundum and coesite grospydites. The liquids fractionate and accumulate magnesian, high-alumina pyroxenes until garnet nucleates as a liquidus phase with its much lower Mg/(Mg + Fe) ratio. Thereafter, the pyroxenes become much less aluminous, and subsequent fractionation is much more difficult to define. Because of the large radius of the eight-fold site in garnet, it is likely that the garnet would be more iron rich than the liquid so no clear iron enrichment pattern, such as results from removal of olivine or orthopyroxene, would be expected.

The spread in oxygen isotopic ratios appears to require that some low-temperature processes be involved, so it may be that the silicate liquids are the result of melting of hydrous subducted oceanic crust. However, our preliminary results suggest that the primary phases are not in oxygen isotopic equilibrium with the secondary phases that appear to be due to metasomatism in the mantle. We believe that further study of stable isotope fractionation and heterogeneity in mantle metasomatic systems is required before the origin of the liquids can be constrained.

9. Conclusions

Crystal-chemical evidence of extensive exsolution of garnet and kyanite from clinopyroxene together with experimental evidence of liquidus pyroxene compositions indicate that many grospydites and kyanite eclogites are likely to be the result of igneous accumulations of aluminous pyroxenes at pressures in excess of 3 GPa followed by equilibration to lower temperatures. Composite samples containing both kyanite eclogites and bimineralic eclogites confirm the chemical continuity of the fractionation trend and show that these rocks are not genetically distinct. Rare earth data on grospydites are consistent with experimental measurements of liquidus pyroxene-liquid distributions and are consistent with a high-pressure pyroxene cumulate origin for these rocks. Oxygen isotopic studies, however, suggest that low-temperature processes may have been involved in the evolution of the liquids, although additional stable and radiogenic isotopic data on grospydites and kyanite eclogites would help to clarify these preliminary conclusions.

The exsolution and geochemical evidence for a high pressure igneous origin for many of the eclogites from South African kimberlites indicates that a simple subduction of Archean oceanic crust followed by solid-state metamorphism is not responsible for much of the chemical variation observed in these rocks. For eclogite liquids to have existed at depths of 120–150 km, they must have been derived from much greater depths, or geo-
thermal gradients must have been much higher in the past. This implies that eclogite (i.e. basaltic) compositions may be a significant and geochemically distinct component throughout the upper mantle. The spread in oxygen isotopic data, however, suggests that low temperature processes may have played a role in the geochemical evolution of this component of the mantle, possibly through low-temperature alteration and subsequent subduction and melting of oceanic crust.

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