Platinum-group mineralization in the Tertiary Igneous Province: new data from Mull and Skye, Scottish Inner Hebrides, UK

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Abstract – In recent years, platinum-group mineral deposits have been reported from several of the Tertiary igneous complexes in East Greenland. These intrusions form part of the same igneous province as the Tertiary igneous centres in northwest Scotland, and recent work confirmed the presence of abundant platinum-group minerals in the Rum Central Complex. In this paper we report for the first time the presence of abundant and diverse platinum-group minerals from the Tertiary Ben Buie intrusion on Mull and the Cuillin Complex of Skye, Inner Hebrides, Scotland. The platinum-group minerals in the Ben Buie intrusion are associated with chromitite layers in ultramafic cumulates. The grains are small (1–2 μm), locally very abundant, and dominated by Pd–Bi–Te–Sb phases. In the Peridotite Series of the Cuillin Complex, the platinum-group minerals are dominated by laurite commonly enclosed within chromite or silicate. A combined orthomagmatic–hydromagmatic origin is proposed for the mineralization. However, the style and type of mineralization differs between each of the igneous centres. Platinum-group element mineralization is a ubiquitous feature of the Tertiary Igneous Province in northwest Scotland and in Greenland and is intimately linked to the regional tectonic setting.

1. Introduction

Recently, numerous occurrences of platinum-group elements have been recognized in Tertiary intrusions in eastern Greenland, such as the Skaergaard intrusion (Bird *et al.* 1991; Andersen *et al.* 1998), the Kruuse Fjord complex (Arnason *et al.* 1997), the Nordre Aputitêq intrusion (Arnason & Bird, 1994) and the Kap Edvard Holm complex (Bird *et al.* 1995) (Fig. 1). These intrusions and associated flood basalts are part of the geographically extensive Tertiary Igneous Province, linked to the opening of the North Atlantic Ocean (Fig. 1). In northwest Scotland, this province is represented by the volcanic centres of Arran, Ardnamurchan, Mull, Skye and Rum along with extensive flood basalts. Butcher *et al.* (1999) documented the presence of abundant and diverse platinum-group minerals within the mafic–ultramafic complex on Rum. Given the regional tectonic setting, it was speculated that platinum-group element enrichment was the result of melting associated with proto-Icelandic 'hot-spot' activity linked to the opening of the North Atlantic Ocean (Butcher *et al.* 1999). Platinum-group element occurrences in a similar tectonic setting are seen at, for example, Noril'sk, Siberia and Duluth, Minnesota (Naldrett *et al.* 1992; Mainwaring & Naldrett, 1977; J. D. Miller, pers. comm., 1998, 1999; O. Simonov, pers. comm., 1999), both of which are associated with flood basalt volcanism. Butcher *et al.* (1999) speculated that if the platinum-group minerals on Rum are intimately linked to the regional tectonic setting, then the other mafic–ultramafic complexes elsewhere in northwest Scotland may host similar phases. The aim of this paper is to document for the first time the presence of platinum-group minerals from both Mull (within feldspathic peridotites in the Ben Buie intrusion) and Skye (the Peridotite Series of the Cuillin Complex). The mechanisms that led to the concentration of the platinum-group elements are briefly discussed, along with the implications of these new data.

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2. Geological setting

Chromite-bearing ultramafic lithologies host some of the largest platinum-group element resources in the world (e.g. Von Gruenewaldt, Hatton & Merkle, 1986). On Rum, platinum-group minerals are closely associated with sulphides and silicates within, and immediately adjacent to, chromitite layers and laminae. In this study we assumed a similar association for both the Ben Buie intrusion on Mull and the Peridotite Series of the Cuillin Complex on Skye. Sampling was limited to the chromitite layers and laminae that occur in the mafic–ultramafic cumulates of these complexes. The possibility that platinum-group elements are concentrated in different mineralogical settings, as seen in East Greenland, exists but is as yet untested.

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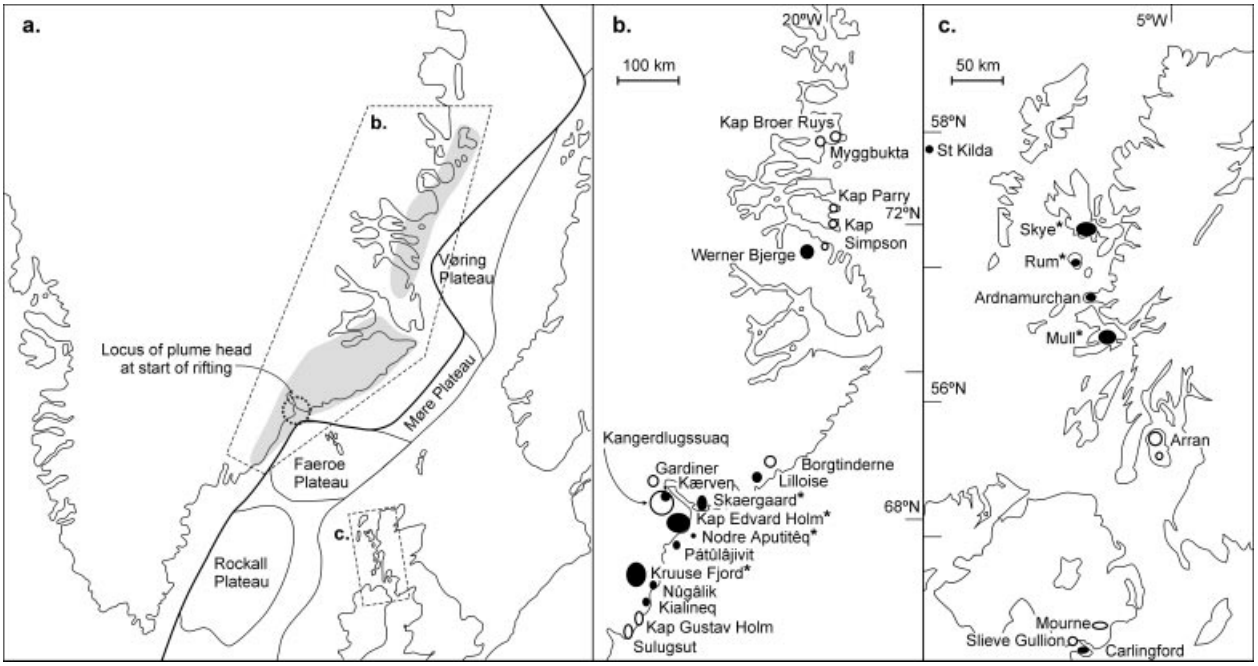


Figure 1. (a) Pre-rift plate tectonic setting of the North Atlantic region. The heavy line indicates the line of initial rifting. The Tertiary Igneous Province (igneous centres, flood basalts and dyke swarms) are shown in grey. (b) Map showing the East Greenland Tertiary Igneous Province. Unfilled symbols represent acid and alkaline intrusive centres. Dark shading indicates predominantly basic intrusive centres. Stars indicate intrusions known to host platinum-group minerals. (c) Map of the British Tertiary Igneous Province, symbols are as in (b). Platinum-group minerals are reported here for the first time from the Skye and Mull intrusive centres, whilst the platinum-group minerals on Rum were described by Butcher *et al*. (1999).

2.a. Mull

The Ben Buie intrusion occurs at the southwestern edge of the Mull central complex (Fig. 2). It predominantly comprises bytownite gabbro (eucrite) with subordinate allivalites (leuco-troctolites) and feldspathic peridotites (mela-troctolites) that were possibly part of a much larger layered intrusion, emplaced towards the end of the Mull Centre 1 activity at about 58 Ma (Lobjoit, 1959; Skelhorn, MacDougall & Longland, 1969; Emeleus & Gyopari, 1992). The gabbro has a high-Ca tholeiitic composition, similar to the early basic cone sheets on Mull, and is considered to belong to the Central Mull tholeiitic magma type (Skelhorn *et al*. 1979; Kerr *et al*. 1999).

Abundant chrome spinel (chromite for brevity) occurs in the feldspathic peridotites which crop out sporadically throughout the Ben Buie intrusion (e.g. W. M. Lobjoit, unpub. Ph.D. thesis, Univ. Manchester, 1957; Fig. 2a). Chromitite layers with distinctly different morphologies were identified at two stratigraphically discrete localities: on Caigeann Doire nan Cuilean [NM 5771 2924] and approximately 400 m northwest of Craig [NM 5807 2978] (Fig. 2a).

Shallow dipping (approximately 12°N) planar chromitites are well exposed on Caigeann Doire nan Cuilean. The layers are laterally persistent across the exposure (>10 m) and up to 2 cm in thickness (Fig. 3a). Little or no lithological variation is apparent above or below the chromitites and both upper and lower contacts are sharp. Small (<100 μm) sulphide minerals (predominantly pentlandite and chalcopyrite) are present but scarce (typically <<1 modal %). In addition to the planar chromitite layers a number of irregular chromitites also occur. These are laterally discontinuous (<2 m) seams, rarely more than 1 cm thick and may be highly convoluted (e.g. Fig. 3b). Alteration of silicate phases is common adjacent to the chromitite layers and seams.

The chromitites exposed to the northwest of Craig occur at the margins of small anorthosite pods (possibly xenoliths) enclosed within the feldspathic peridotites. These pods are rounded bodies, typically 40 cm in diameter (some reach 1 m), composed almost entirely of plagioclase, with thin (up to 1 cm thick), discontinuous chromitite seams developed at their margins (Fig. 3c).

2.b. Skye

Ultramafic lithologies of the Peridotite Series crop out in an arcuate band on the southwestern margin of the Outer Layered Series of the Cuillin Complex (Fig. 2b). The Peridotite Series comprises a layered succession of dunites, peridotites, feldspathic peridotites, allivalites and anorthosites (Claydon & Bell, 1992) believed to have crystallized from a Ca-tholeiitic melt which was emplaced with olivine primocrysts in suspension (Gibb, 1976). Olivine-dominant lithologies occur at the lowermost structural levels (units 1 to 4 of

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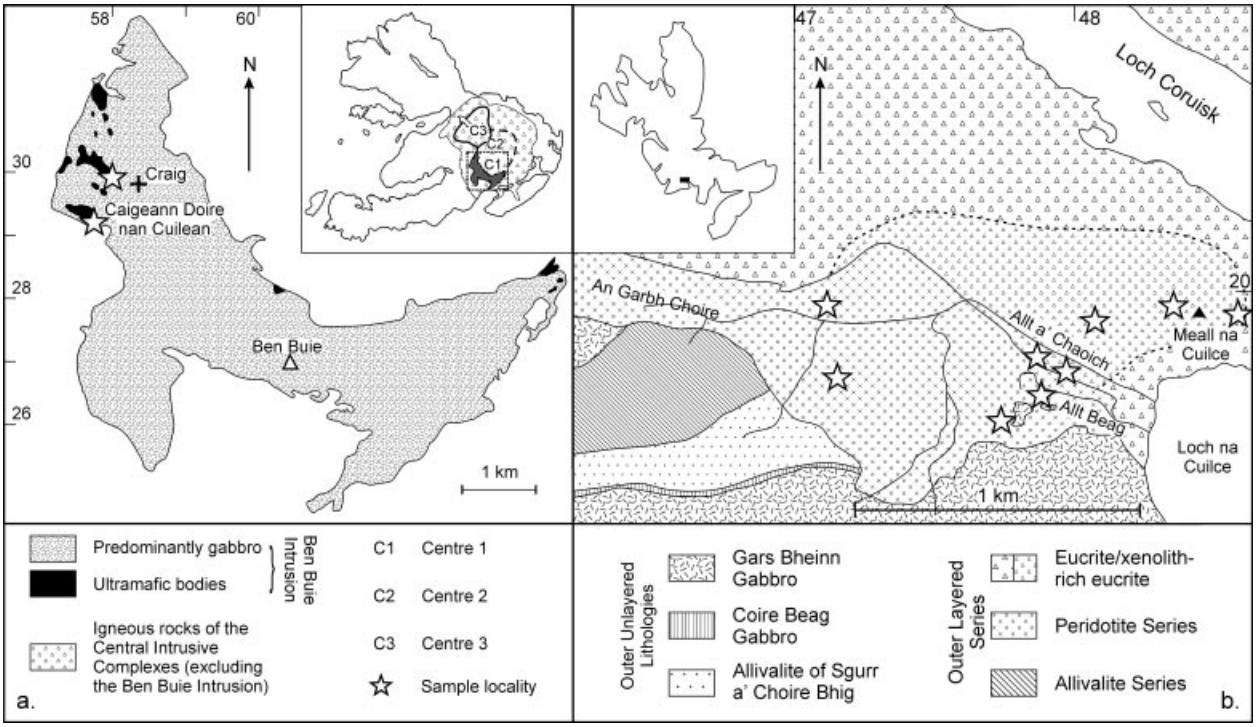


Figure 2. (a) Sketch geological map of the Ben Buie intrusion on Mull, showing the areas sampled in this study (modified from W. M. Lobjoit, unpub. Ph.D. thesis, Univ. Manchester, 1957; Emeleus & Gyopari, 1992). (b) Sketch map of the southern portions of the Outer Layered Series of the Cuillin Complex, Skye (after Claydon & Bell, 1992) showing location of samples.

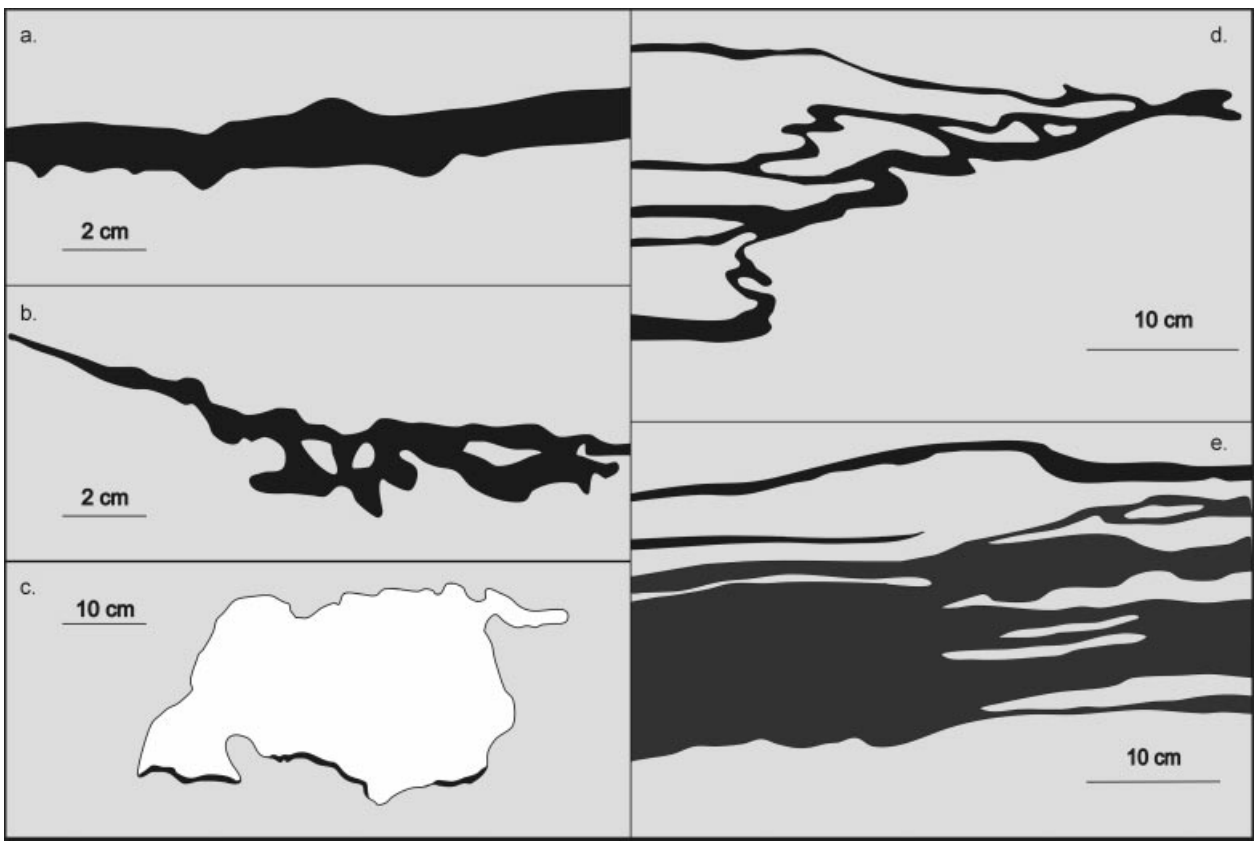


Figure 3. Sketch of some typical chromitite morphologies for Mull: (a) planar chromitite layer, Caigeann Doire nan Cuilean; (b) irregular chromitite seam, Caigeann Doire nan Cuilean; (c) chromitite at margin of anorthosite pod, Craig. Typical chromitite morphologies for Skye: (d) irregular chromitite seam; (e) concentration of planar chromitites. Note that the chromitite layers from Skye are substantially thicker than those from Mull. Grey shading indicates olivine cumulates whilst white signifies plagioclase cumulates.

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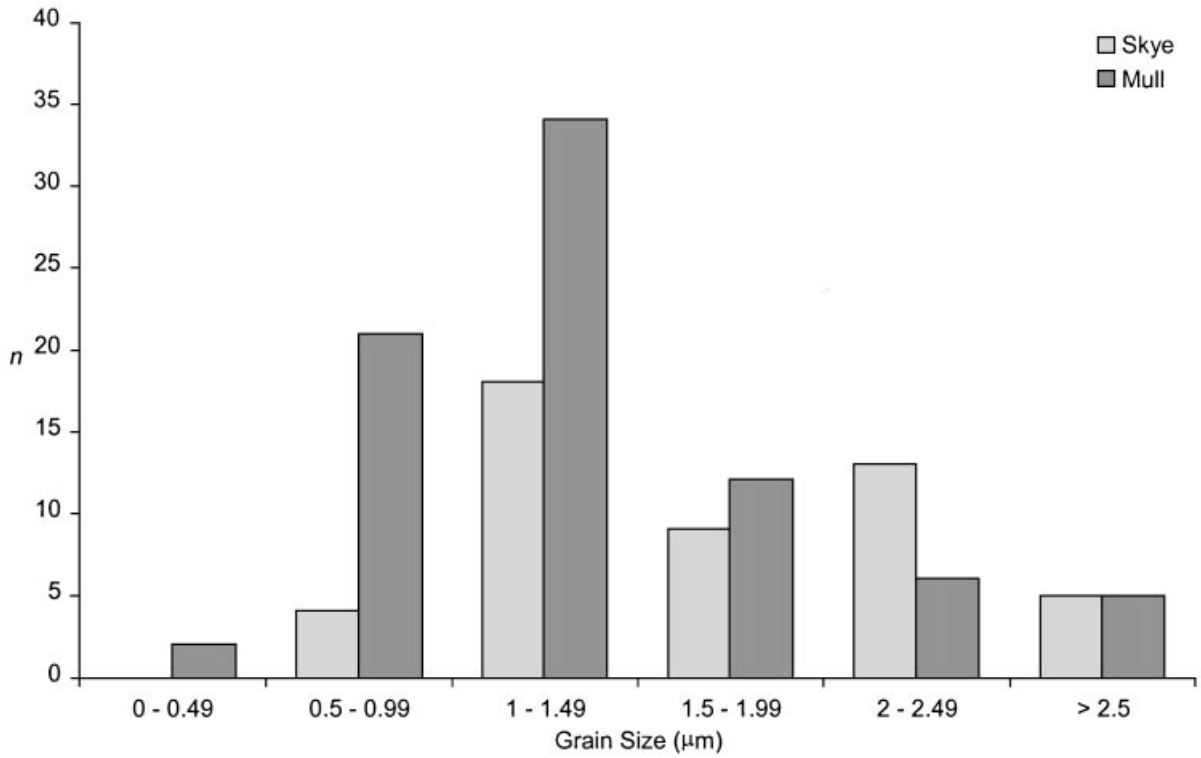


Figure 4. Histogram showing grain size distribution of platinum-group minerals from (a) Mull (n = 81) and (b) Skye (n = 49). Note the very small modal platinum-group mineral grain size for both Mull and Skye.

Claydon & Bell, 1992) which are well exposed in the southeast, whilst plagioclase-dominant lithologies occur in the higher structural levels (units 5 and 6 of Claydon & Bell, 1992) which crop out in the northwest. A broad zone of ultramafic breccia (Unit 4 of Claydon & Bell, 1992) crops out below An Garbh Choire and comprises peridotite clasts within a plagioclase-dominant matrix. Younger bytownite gabbro intrusions of the Outer Layered Series truncate the northern margin of the Peridotite Series and contain numerous ultramafic xenoliths (up to 20 m in diameter) (Fig. 2b; Weedon, 1961). The Peridotite Series is further cut by numerous basalt and gabbro intrusions particularly at lower structural levels and at its eastern margin.

Chromitites are abundant throughout the lowermost stratigraphic levels of the Peridotite Series. In particular, steeply dipping (approximately 80°N) chromitite layers are well exposed along Allt Beag (e.g. Weedon, 1965; Bell & Claydon, 1992). Sampling focused on the lower stratigraphic levels in an area to the west of Loch Coruisk between Meall na Cuilce [NG 4833 1990] and An Garbh Choire [NG 4680 2010] (Fig. 2b). The chromitites have variable morphologies from relatively uniform, planar layers through to highly convoluted irregular seams (Fig. 3d,e). Individual layers can be traced laterally for over 50 m (Weedon, 1965), although they commonly pinch (to under 2 mm) and swell (to over 2 cm). In a few places, thin chromitite layers converge to form pods >20 cm thick (e.g. Fig. 3e). Chromitites are also well developed in many of the ultramafic xenoliths in the Eucrite Series, commonly preserving the regional dip (Weedon, 1965).

The chromite grains within the chromitites exhibit a variety of colours (and compositions) from olive green, through reddish brown to opaque. The green spinels appear to be restricted to the lowermost structural levels and have compositions approaching spinel (*sensu stricto*) with little Cr. A trend of increasing Cr/Cr+Al ratio and TiO_2 content with higher structural levels has been identified (Bell & Claydon, 1992). Sulphide minerals, typically chalcopyrite and pentlandite, are present as accessory phases within the chromitites but are not abundant (<<1 modal %; Bell & Claydon, 1992). Chalcopyrite is, however, locally abundant along the margins of some of the small basic intrusions.

3. Samples and methodology

Chromite-bearing mafic and ultramafic rock samples have been examined from Mull (15 samples) and Skye (25 samples). Platinum-group minerals were located and identified using a JEOL 840 scanning electron microscope (SEM) with an Oxford Instruments (Link System) AN 10000 EDS analyser. The grains were initially located using backscatter electron imaging using a beam current of 3×10^{-9} Å and an accelerating voltage of 20 kV. Due to the small size of the grains, a

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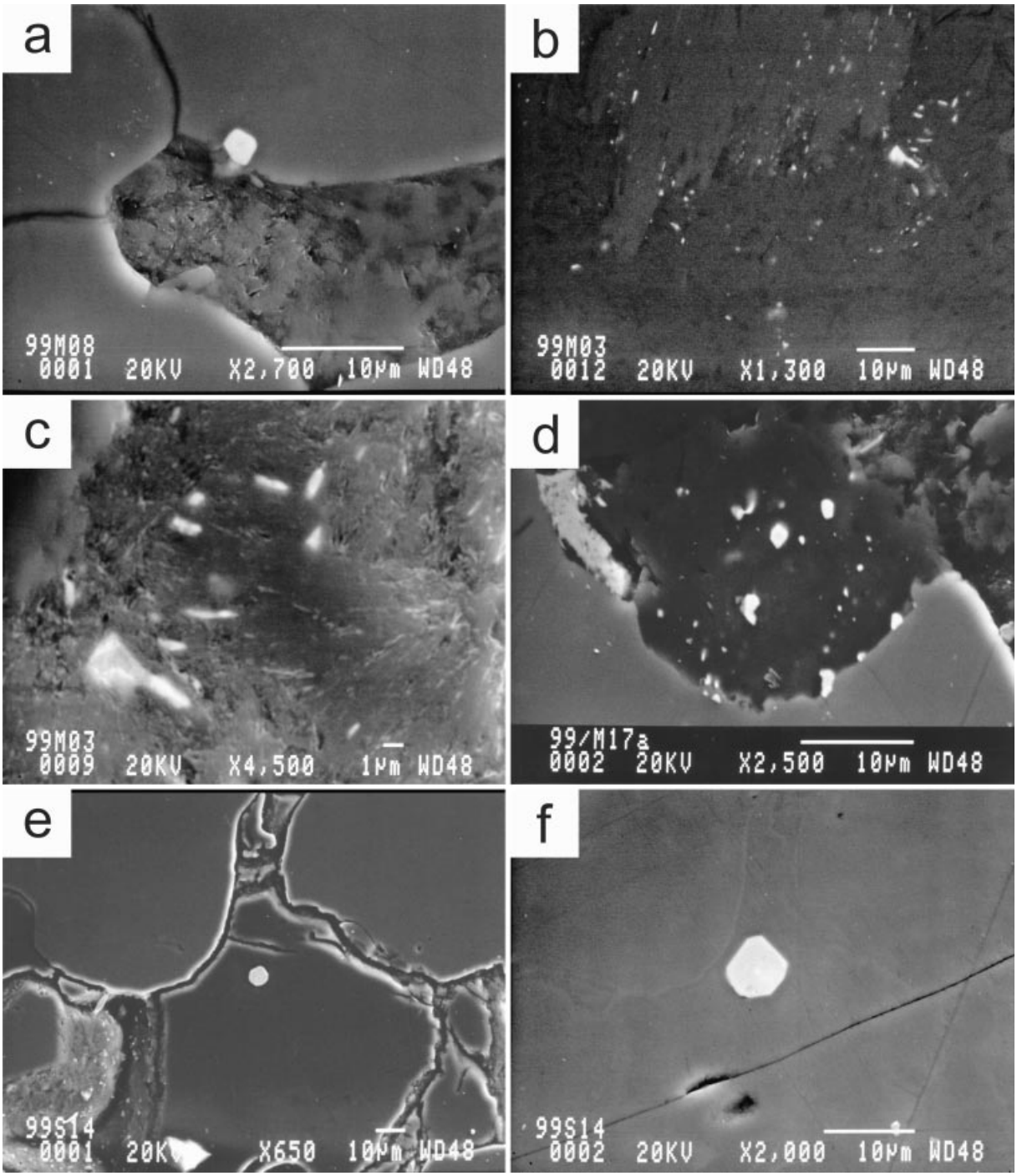


Figure 5. Representative scanning electron microscope photomicrographs of platinum-group mineral grains from Mull and Skye; (a) euhedral laurite grain enclosed within chromite, mottled phase is chlorite, Caigeann Doire nan Cuilean, Mull; (b) backscatter scanning electron image showing a concentration of small platinum-group mineral grains (mostly Pd–Sb–Bi phases) within clinopyroxene (light phase) and chlorite alteration (darker phase), Craig, Mull; (c) magnified image of top right hand field of image (b), showing platinum-group mineral grains (Pd–Sb–Bi and Pd–Te–Bi phases) enclosed within chlorite, Mull; (d) numerous platinum-group mineral grains of varying composition within plagioclase. Larger grain at centre of image is a Pd–Bi–Sb–Te phase whilst the majority of smaller grains are Pd–Sb–Pb phases, Craig, Mull; (e) euhedral laurite grain within cumulus olivine, Skye; (f) euhedral laurite within chromite, Skye.

lower beam current of 6×10^{-10} Å was used whilst undertaking EDS analysis to reduce the X-Ray excitation volume (nominally 2 μm in diameter). None of the platinum-group minerals have been analysed quantitatively due to their small size.

4. Platinum-group mineralization on Mull

Of the 15 samples analysed, 12 yielded platinum-group minerals with >80 discrete grains recognized. The platinum-group minerals are small, ranging in size

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from <1 to 3 μm (mode 1 μm, mean 1.1 μm) (Fig. 4). Few of the platinum-group minerals show clear crystal form although a small number of laurites, especially those enclosed within chromite, are euhedral (Fig. 5a). The composition and association of the platinum-group minerals is different at the two localities; this may be due to stratigraphic variation in the platinum-group mineral assemblage. However, the poor exposure of the ultramafic lithologies and their sporadic outcrop pattern does not allow accurate correlation.

The samples collected near Craig are dominated by Pd-rich phases and in particular Pd–Sb–Bi and Pd–Te–Bi minerals. Other, less common phases include grains tentatively identified as laurite $(Ru,Ir,Os)S_2$, sperrylite ($PtAs_2$), froodite? ($PdBi_2$) and minerals of the platirasite–hollingworthite–irarsite (PtAsS–RhAsS–IrAsS) series amongst others. In all of these samples there are concentrations of minute platinum-group mineral grains, typically occurring in restricted areas (up to 100×100 μm) of silicate (commonly intercumulus plagioclase or clinopyroxene) where numerous (sometimes >50) small (typically <<1 μm) grains of similar composition (commonly Pd–Sb–Bi and Pd–Te–Bi) occur together (Fig. 5b).

In contrast, the platinum-group minerals within the samples from Caigeann Doire nan Cuilean are widely disseminated and dominated by Pt-rich phases (most commonly sperrylite) and laurite. Other phases include Pd–Pb sulphides, minerals of the platirasite–hollingworthite–irarsite series, Pd–Bi tellurides and Pd-arsenides. The platinum-group minerals typically occur at the grain boundaries between plagioclase, olivine, clinopyroxene, chlorite and chromite (Fig. 5), or rarely enclosed within one of these phases. Sulphides are rare and there is no clear correlation between the platinum-group minerals and the sulphides. Only three platinum-group mineral grains (of Pt–As sulphides) are clearly associated with sulphides (in all cases, pentlandite).

5. Platinum-group mineralization on Skye

Platinum-group minerals are recognized in 14 of the 25 samples collected from the Peridotite Series and from xenoliths within the Outer Layered Eucrite. However, the total number of grains is relatively low (<50), significantly less than Mull, with up to a maximum of 10 grains in a single polished section. Platinum-group minerals occur in most of the layers examined, including the Cr-poor spinel layers. In addition, they also occur within chromitites preserved in xenoliths, and from chromitite clasts within the Unit 4 breccia. No platinum-group minerals have been found in association with the sulphide-rich zones adjacent to the small basic intrusions.

The grains range in size from <1 μm to 7 μm and have a broadly uniform grain size distribution centred about a mode of 1 μm (Fig. 4). No stratigraphical variation in the distribution or composition of the platinum-group minerals within the Peridotite Series has been identified, although this may be a function of the limited number of samples collected and the relative rarity of the minerals.

The platinum-group mineral assemblage is fairly diverse, although over half (57%) of the grains identified are laurite. Other, less common, phases include sperrylite, ruarsite (RuAsS), Pt–Rh–Fe alloy, Pd arsenides, Pd–Bi tellurides, and minerals of the platirasite–hollingworthite–irarsite series. The grains occur in a range of different mineralogical associations. They are most commonly found enclosed within chromite, or at chromite–sulphide (pentlandite, chalcocite) or chromite–silicate grain boundaries. A small number of grains are enclosed within silicate minerals, typically intercumulus clinopyroxene, or alteration phases (chlorite), although one euhedral laurite grain is enclosed within cumulus olivine (Fig. 5). Very few of the grains are enclosed within sulphides, and those that do represent a subset of the overall platinum-group mineral assemblage, hence there is no clear association between the platinum-group mineral and sulphide phases on Skye. The majority of the platinum-group minerals enclosed within chromite are euhedral laurite; those in other associations show a variety of compositions and are typically anhedral.

6. Discussion

Platinum-group minerals from the Eastern Layered Series on Rum are much more diverse, widespread and extensive than on Mull or Skye (Butcher *et al*. 1999; Power *et al*. in press). Within the Eastern Layered Series they are associated with sulphides within chromitite layers (Butcher *et al*. 1999; Power *et al.* in press). This suggests that the platinum-group elements were included in small amounts of sulphide liquid, which exsolved in response to magma mixing (cf. Butcher *et al*. 1999), probably in a similar way as argued for the Stillwater J-M reef by Irvine, Keith & Todd (1983). The textural setting of the platinum-group minerals on Mull and Skye differs from that on Rum, and even Mull and Skye are distinctly different from each other. On Mull the platinum-group minerals typically occur at silicate or silicate–chromite grain boundaries; very few grains are enclosed within sulphides. In contrast the platinum-group minerals on Skye are most commonly found enclosed within chromite, or at chromite–sulphide (pentlandite, chalcocite) or chromite–silicate grain boundaries; the most common phase, laurite, is typically enclosed within chromite. Thus the mechanism for the local concentration of the platinum-group elements on Mull and Skye is different to that on Rum, despite the similar tectonic setting; this is directly analogous to the diverse platinum-group element occurrences in East Greenland.

The platinum-group mineral assemblage on Mull

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includes various platinum-group element complexes of the semi-metals Bi, Te and Sb, indicative of lower temperature, hydromagmatic processes (Coghill & Wilson, 1993). The majority of the platinum-group minerals are enclosed within, or occur at the boundaries between, primary intercumulus silicate phases supporting this interpretation. However, there is also evidence for alteration within the chromitites on Mull, suggesting that hydrothermal activity may have altered (oxidized) any sulphides originally present, potentially remobilizing associated platinum-group minerals. Secondary sulphides, although rare, are mainly subordinate quantities of chalcocite (cf. Maier & Barnes, 1996), but texturally there is little evidence that there has been significant remobilization of the platinum-group minerals. Although a small number of grains occur in association with chlorite they appear to be residual platinum-group minerals rather than secondary phases.

Relatively little alteration is apparent in the samples from Skye and the platinum-group minerals predominantly occur in association with unaltered cumulus and intercumulus phases. Laurite is commonly associated with cumulus olivine and chromite (as are other orthomagmatic phases: see Coghill & Wilson, 1993). Once again, as on Mull, lower temperature hydromagmatic platinum-group minerals such as Pd–Bi tellurides occur in association with the intercumulus phases (clinopyroxene and plagioclase). Laurite may therefore have crystallized at the early magmatic stage together with cumulus olivine and spinel, whilst other platinum-group minerals crystallized at lower temperature from residual interstitial liquid which had become enriched in platinum-group elements (e.g. Boudreau & McCallum, 1992).

This paper shows that the widespread platinum-group element mineralization recognized in the layered intrusions of the Tertiary Igneous Province in East Greenland (Bird *et al*. 1991, 1995; Arnason *et al.* 1997; Andersen *et al*. 1998) also extends throughout northwest Scotland although in all cases is subeconomic. Indeed, the deposits in northwest Scotland are directly analogous to those in East Greenland. In both areas the initial magmatism is clearly linked to the proto-Icelandic 'hot-spot' activity, but the local mechanism for the concentration of platinum-group elements and formation of the platinum-group minerals differs between each of the recognized deposits. The distribution of mafic–ultramafic igneous centres associated with this magmatic episode is shown in Figure 1; those that have already been proven to host platinum-group mineralization include the Skaergaard, Kap Edvard Holm, Nordre Aputitêq and Kruuse Fjord complexes in East Greenland, and the Rum, Skye and Mull central complexes in Scotland. It is interesting to note that the focus of mineralization in East Greenland is spatially related to the location of the mantle plume head at the time of magmatism. The mineralization in northwest Scotland follows an earlier failed arm of this rifting event. We believe that the addition of plume-type mantle material was a critical factor leading to platinum-group element mineralization. The possibility exists that other comparable intrusions in this province may also host similar mineralization. Thus the North Atlantic Tertiary Igneous Province can now be recognized as one of the most extensive platinum-group mineral provinces in Europe and represents an ideal natural laboratory to study the stages of plume-related platinum-group element enrichment processes.

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