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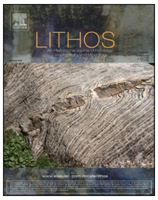

The significance of water in the genesis of ophiolitic chromitites

Hugh Rollinson [*]

School of Built and Natural Environment, University of Derby, UK

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ABSTRACT

Many ophiolitic chromitites contain hydrous silicate phases, implying that hydrothermal processes were involved in their genesis. This observation challenges the standard magmatic/ melt-rock reaction model for mantle chromitite genesis. This review explores three possible relationships between hydrous fluids and the genesis of chromitites formed in the mantle section of ophiolites. The models are described and critiqued in the light of our current experimental, geochemical and field-based understanding of ophiolitic chromitites, using in particular evidence from chromitites from the well-studied mantle section of the Oman ophiolite.

Matveev and Ballhaus (EPSL, 2002) and more recently Su et al. (Science China Earth Sci., 2021) have proposed an immiscibility model in which chromite is preferentially partitioned into a hydrous fluid and thus separates from the parental silicate melt. Here it is argued that there are problems with the scalability of this model. In addition there are petrographic and geochemical inconsistencies which do not conform to the proposed hydrothermal fluid-silicate melt partitioning.

Johan et al. (Eur. J. Mineral., 2017) and Arai and Akizawa, (Amer. Mineral., 2014) have championed the view that chromite can be precipitated directly from a hydrothermal fluid. This is evidenced by chromite-diopsidites in the Oman ophiolite. Recent experimental studies also support this view through the complexing of Cr^{2+} in a chlorine-rich fluid. However, this model is also an inadequate explanation for the majority of mantle chromitites because chromite-diopsidite occurrences are extremely rare in the mantle section of ophiolites, of small volume, and the chromitites formed have a different composition from typical ophiolitic mantle chromitites.

For these reasons, the preferred model and the one advocated in this paper derives from the work of Edwards et al. (GSA Spec Pap. 2000) in which water is present as a dissolved species in the parental melt from which the chromitites form. It is argued that mantle chromitites are associated with hydrous MORB and boninitic melts in which the presence of water modifies the structure of the melt to permit an increased solubility of Cr during partial melting. The model is supported with Cr solubility data from wet mantle melting experiments and from the modelling of Cr partitioning during mantle melting, using data from harzburgites from the mantle section of the Oman ophiolite. As the hydrous melts percolate through mantle harzburgites they become more siliceous through orthopyroxene dissolution triggering chromite crystallisation. The same melts also crystallise the hydrous phases sodic amphibole and micas.

1. Introduction

The standard model for the origin of ophiolitic chromitites is the magmatic model in which the chromitites are the product of a melt-rock reaction between a mafic melt percolating through depleted mantle harzburgite. However, recently it has become apparent that many ophiolitic chromitites contain hydrous phases leading to the possibility that hydrothermal processes were involved in their genesis and as a consequence a number of hydrothermal models for chromitite genesis have been proposed. This paper seeks to evaluate these competing claims and thereby establish a more integrated understanding of the role of water in the genesis of ophiolitic chromitites.

The standard magmatic model for the origin of ophiolitic chromitites builds upon the early work of Kelemen (1990) and Kelemen et al. (1995) on the origin of dunitic veins in depleted mantle peridotites and was developed more fully and applied to chromitite genesis by Zhou et al. (1996) and Arai and Miura (2016). Typically, podiform chromitites are enclosed in a sheath of dunite in a depleted mantle harzburgite host. This is explained in the standard model as the result of a primary mafic melt percolating along fractures through depleted mantle harzburgite.

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[*] Corresponding author.
E-mail address: h.rollinson@derby.ac.uk.

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At low pressures this melt is no longer in equilibrium with orthopyroxene which is consumed and replaced with olivine (Fig. 1a). As a result of this reaction the ensuing secondary melt is more siliceous than the primary melt. When viewed in the system olivine-chromite-silica the primary melt evolves along the olivine-chromite cotectic but on reaction with orthopyroxene in the harzburgitic host the more siliceous secondary melt migrates from the olivine-chromite cotectic into the chromite stability field, initiating the precipitation of chromite (Fig. 1b). In addition, it is possible that mixing takes place between a fresh batch of the primary melt with some earlier secondary melt (Fig. 1c) also leading to the primary melt composition shifting into the chromite stability field and initiating chromite crystallisation (Fig. 1d).

Advocates of an alternative hydrothermal model for ophiolitic chromitite genesis draw attention to recent observations which show that chromitites formed in the mantle section of ophiolites have formed in the presence of a hydrous melt or fluid phase (Matveev and Ballhaus (2002), Arai and Miura (2016); Johan et al. (2017); Su et al. (2020, 2021a)). Evidences include

- the presence of hydrous silicate phases such as micas and amphiboles co-crystallised with the chromite, both as inclusions and as interstitial grains (see for example from the Oman ophiolite - Ahmed and Arai, 2002; Lorand and Ceulneer, 1989; Rollinson, 2008, Rospabé et al., 2017, 2021);
- the presence of complex, multiphase inclusions containing hydrous phases and interpreted as former melt inclusions in chromitites (see Borisova et al., 2012; Rollinson et al., 2018);

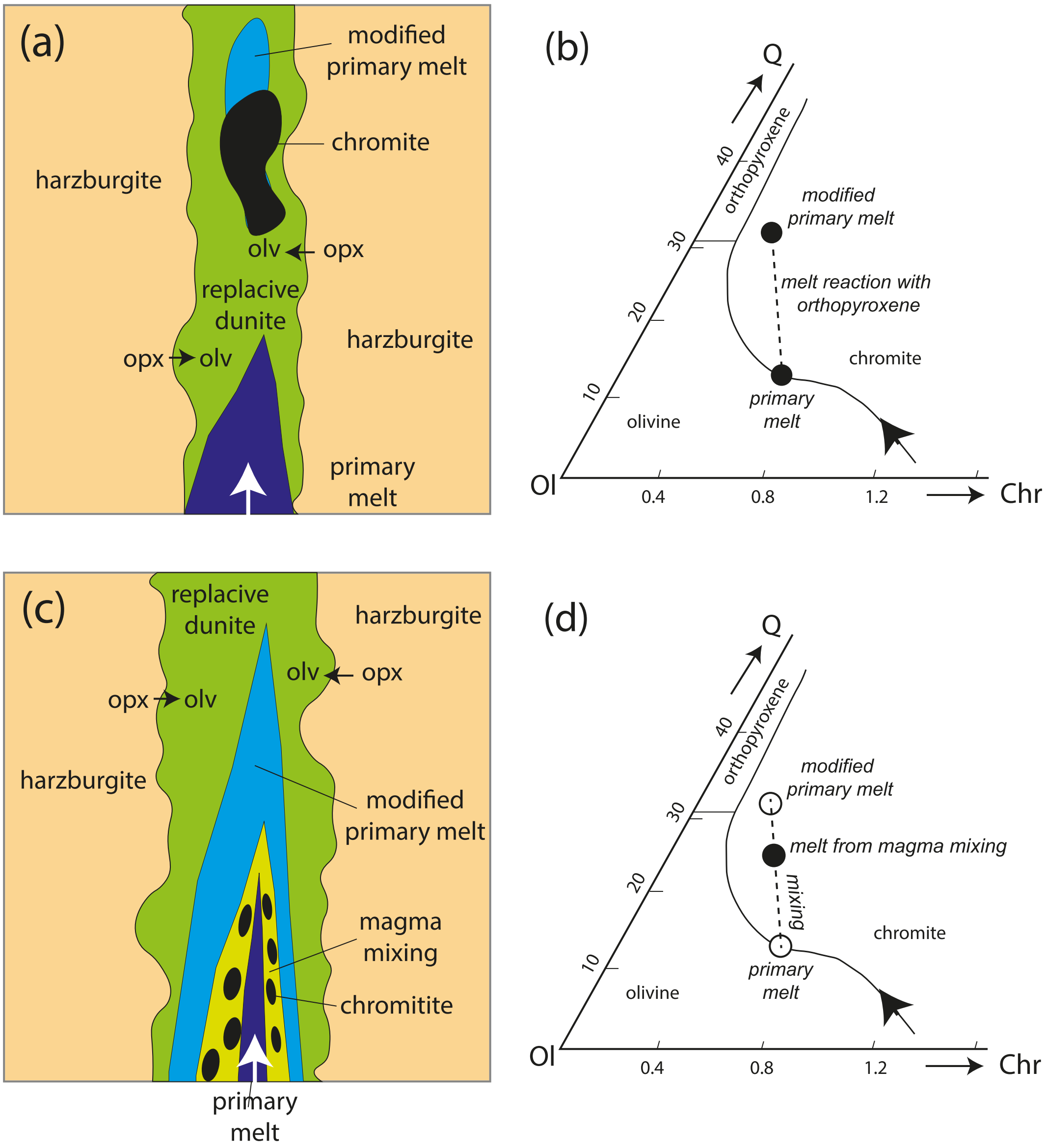

Fig. 1. Models for the genesis of podiform chromitites in the mantle section of ophiolites according to the standard model, adapted from Arai and Miura (2016). (a) The migration of a mafic melt (primary melt) along a fracture through mantle harzburgite. A reaction takes place in which the mafic melt consumes orthopyroxene and precipitates olivine creating the replacive dunite. The net effect of this reaction is to modify the composition of the primary melt and make it more siliceous, leading to chromite precipitation. This is explained in (b) a part-schematic olivine (Ol)-silica (Q)-chromite (Chr) phase diagram based upon the petrographic and experimental studies of Irvine (1975, 1977) modified from Zhou et al. (1996). The diagram shows the stability fields of olivine, chromite and orthopyroxene. The values on the axes are cation percent. A primary melt evolves along the olivine-chromite cotectic but as it absorbs orthopyroxene it becomes siliceous (the modified primary melt) and because of the curvature on the cotectic moves from the cotectic into the chromite stability field, initiating the precipitation of chromite. (c) In addition the mixing between the primary melt and the modified primary melt also creates melts which are more siliceous than the primary melt leading to chromite crystallisation. (d) The mixing process illustrated in panel (c) depicted on the olivine-silica-chromite diagram.

- the rare occurrence of saline fluid inclusions in chromite (Johan et al., 2017; Melcher et al., 1997);
- the relatively high water content of nominally anhydrous minerals associated with massive chromitites (Su et al., 2020);
- the association in ophiolitic sequences of chromitites with boninites – the product of the hydrous melting of harzburgite, and 'moist MORB' – basalts which have a higher water content than that expected in normal MORB (Falloon and Danyushevsky, 2000; MacLeod et al., 2013).

Currently there are three different approaches which seeks to offer a primary reason why water is important in chromite genesis. Matveev and Ballhaus (2002) pioneered the *immiscibility model* in which they argued that the presence of water provides a physical mechanism for the nucleation and growth of chromite grains in ophiolitic chromitites. This model was adopted by Zhou et al. (2014) and recently extended by Su et al. (2020, 2021a) to argue that melt-fluid immiscibility is a key process in the transportation and aggregation of chromite to form podiform chromitites. A second approach advocated by Arai and Miura (2016) and Johan et al. (2017) is that chromite crystallisation takes place by means of *hydrothermal processes* as evidenced by field-based and experimental studies. A third approach, discussed by Edwards et al. (2000) is that the presence of dissolved water in a mafic melt *influences the structure of the melt* and thereby provides a mechanism for the increased solubility of Cr in a mafic melt and the subsequent crystallisation of chromitites.

Clearly, whilst there is strong evidence that water has a role to play in the genesis of ophiolitic chromitite deposits, there is no consensus on what that role is. The purpose of this paper therefore is to briefly review current hydrothermal models and to critique them in the light of current experimental, geochemical and field-based understanding of ophiolitic chromitites. In particular, evidence is cited from the well-studied Oman ophiolite where the lava sequence contains evidence of hydrous melts, the 'moist MORB' of MacLeod et al. (2013) and relatively abundant boninites (Belgrano et al., 2019) together with chromitites in the mantle section of the ophiolite.

2. The role of liquid immiscibility in the genesis of ophiolitic chromitites

In an important and much cited paper Matveev and Ballhaus (2002) describe a series of experiments in the basalt-water system in which excess water (>4 %) and chromite were added to a picritic melt which at 1150–1200 °C and 0.4 GPa was shown to contain an immiscible, water-rich fluid phase. Under these conditions chromite and olivine were partitioned between the two phases with chromite grains favouring the exsolved fluid pools whereas olivine remained in the silicate glass. In experiments of longer duration the fluid pools coalesced leading to concentrations, about 200 μm in diameter, of chromite micro-phenocrysts – each grain being only a few microns across (Fig. 2a).

Matveev and Ballhaus (2002) proposed a mechanism for the chromite-olivine separation whereby in 'silicate melt-water systems, crystal faces of oxides are wetted better by water-rich fluid than by silicate melt' and the converse for olivine. The rounded, 'nodular' form of the immiscible fluid pools led them to suggest that they represent a mechanism whereby nodular chromitites might form. Nodular chromitites are a particular, but relatively rare, expression of ophiolitic chromitites in which aggregates of chromite grains are bound together into nodules, normally 1–2 cm across, in a silicate matrix. They are restricted in occurrence to ophiolitic chromitites (Pritchard et al., 2015). [In subsequent uses of the Matveev and Ballhaus, 2002 study some confusion has arisen over the terms 'nodular' and 'podiform', as applied to chromitites. In this study nodular describes a *texture* in which abundant rounded chromite aggregates 1–2 cm across are found in a silicate matrix. Podiform, on the other hand, is a more general term applied to ophiolitic chromitites which describes the *overall shape of the chromitite/dunite body* – often many tens of metres in width and length, and different from the more extensive chromite seams found in layered intrusions.]

The Matveev and Ballhaus (2002) model has recently been adapted by Su et al. (2020) to explain the relatively high levels of water in clinopyroxene associated with podiform chromitites in the Kızıldağ ophiolite, Turkey. They argue that because of the superior wetting properties of chromite, hydrous fluids from a melt aggregate on the surfaces of chromite grains. These fluids are then available, through diffusion, to enrich surrounding silicate minerals such as olivine or clinopyroxene in water, mix with evolving melts to generate

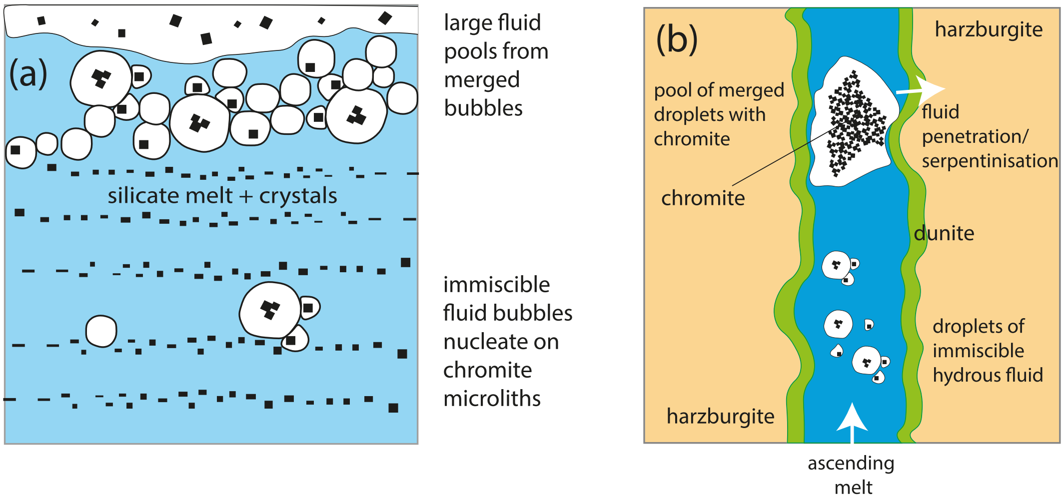

Fig. 2. The immiscibility model of Matveev and Ballhaus (2002) adapted by Su et al. (2021a). (a) The results of the Matveev and Ballhaus (2002) experiment showing immiscible fluid bubbles in silicate melt. Chromite microphenocrysts are preferentially partitioned into the immiscible fluid bubbles and on ascent the fluid bubble merge to form pools enriched in chromite grains. This mechanism is thought to explain the occurrence of chromitite nodules. (b) The model of Su et al. (2021a) in which the mechanism of Matveev and Ballhaus (2002) is generalised to explain the origin of ophiolitic podiform chromitites. Droplets of immiscible hydrous fluid exsolve from a mafic melt into which chromite nuclei are preferentially partitioned. The droplets merge to form fluid pools and concentrations of chromitite. Some of the fluid escapes into the dunite and harzburgite creating serpentinite.

hydrothermal-like clinopyroxene or assist in the formation of solid and fluid inclusions in the chromite. In a later paper, as an extension to this argument, Su et al. (2021a) propose that water rich fluids associated with chromite grains collect to form pockets of chromite together with a water-rich fluid, which is immiscible in the silicate melt. This water is subsequently expelled from the silicate melt facilitating the serpentinisation of the enclosing dunites and harzburgites (Fig. 2b).

3. The role of hydrothermal processes in the genesis of ophiolitic chromitites

Cr^{3+} has a very low solubility in water of near neutral pH (Rai et al., 1987). Nevertheless, a number of different workers, influenced by the field and petrographic evidence for hydrothermal activity in the mantle section of ophiolites have argued for a hydrothermal origin for ophiolitic chromitites. These arguments are based upon experimental studies, field-based observations and fluid inclusion studies.

3.1. *Experimental*

Johan et al. (2017), in a paper with the bold title 'Fluids are bound to be involved in the formation of ophiolitic chromite deposits' proposed that the network of dunite veins in harzburgitic mantle found at the base of ophiolitic sequences is the product of hydrothermal alteration and that Cr is mobile in high temperature hydrothermal fluids. They tentatively 'extend the model of Matveev and Ballhaus (2002) to propose that chromite particles are preferentially wetted by an aqueous fluid, and could thus be concentrated in a fluid medium and transported as a high-temperature slurry along a fissure'. In this model the highly magnesian phases found as inclusions in chromite are explained as the product of hydrothermal Fe—Mg exchange, rather than subsolidus magmatic processes and the presence of metallic alloy inclusions as an indication that the chromite crystallised in a highly reducing environment.

In order to validate these ideas Johan et al. (2017) conducted a series of experiments at 1000–1300 °C and 2 kb, in a reducing environment in an attempt to synthesise chromite from a hydrothermal fluid. These experiments involved the dissolution of oxide or silicate phases in a Cl-rich hydrothermal fluid in the presence of carbon and produced small (up to 100 μm) grains of Al-free chromite or magnesio-chromite. Johan et al. (2017) concluded that the precipitation of chromite from a fluid phase is realistic in the temperature range of 1000–1050 °C and that the process requires a reducing environment.

In a more recent experimental study Huang et al. (2019) investigated the mobility of Cr in upper mantle fluids using the thermodynamic properties of a range of Cr mineral and aqueous species. Using the Deep Earth Water Model of Sverjensky et al. (2014) they predicted the solubility of Cr oxide minerals and identified the importance of Cr^{2+}. On this basis they proposed a new Cr^{2+} complex $CrCl(OH)^0$ formed via the reduction of Cr_2O_3. Their predicted range of solubilities, relevant to the formation of ophiolitic chromitites, indicate very low solubilities in MORB fluids (0.1 mg kg^{-1} H_2O) whereas in reducing, Cl-bearing, subduction zone fluids Cr can be highly soluble with solubilities of 0.5, 2, and 7 mg kg^{-1} H_2O in ultramafic, mafic, and pelitic fluids, respectively. They show that the strongest influences on Cr solubility are oxygen fugacity and Cl concentration - suggesting that mafic fluids could transport larger concentrations of Cr in Cl-rich, reducing conditions.

3.2. *Field based*

In a series of field-based studies in the Oman ophiolite Akizawa and Arai (2014), Arai and Akizawa (2014), Arai and Miura (2016) and Arai et al. (2020) have also argued for a hydrothermal origin for some chromitites. Arai and Akizawa (2014) and Akizawa and Arai (2014) describe two occurrences of hydrothermal diopsidites from the Wadi Fizh area, Oman. Above the MOHO in the crustal gabbros they describe chromite-bearing diopsidites in which there are micro-pods of chromitite less than 1 cm across. Below the MOHO in the mantle peridotite-diopsidites contain thin segregations or pods of chromitite which are up to several centimeters in thickness. Chromite in the mantle diopsidites is similar in composition to other mantle podiform chromitites in the area (cr# ~ 0.6) although the rims of these grains are altered to more chrome- (cr# ~ 0.8) and iron-rich compositions. Chromite compositions in the crustal diopsidites are similar to those found in the altered rims of the mantle diopsidite chromites and are relatively poor in Mg, Al and Cr but enriched in Fe^{2+}, Fe^{3+} and Mn. Here the chromite is typically enclosed within Cr-rich garnet. Arai and Akizawa (2014) proposed that the small chromitite pods in the mantle section are relict primary igneous chromitites which have in part been altered by hydrothermal fluids during the diopsidite metsomatism. The crustal chromitite micropods however are hydrothermal in origin and represent the mobilisation of mantle Cr during the formation of the crustal diopsidites. They suggest that the Cr transport occurred via high-temperature hydrothermal solutions that contain Cl^-, SO_4^{2-} and CO_3^{2-} and are thought to be derived from seawater (Fig. 3). The high concentrations of Fe^{3+} suggests that these fluids were oxidising.

3.3. *Fluid inclusion studies*

Evidence for the existence of a free hydrous fluid during the crystallisation of ophiolitic chromitites comes from the study of fluid inclusions. Melcher et al. (1997) describe fluid inclusions from the giant chromitite deposits of the Kempirsai ophiolite in Kazakhstan. Relative to the occurrence of hydrous mineral inclusions in chromite, fluid inclusions are very rare. They occur in massive ores and in amphibole-chromite-rich veins. Typically, they are hydrous with a range of salinities (2–15 wt% NaCl equivalent) and a low CO_2 content. They are up to 50 μm in length and texturally form primary and pseudo-secondary inclusions implying trapping during chromite growth. Homogenisation

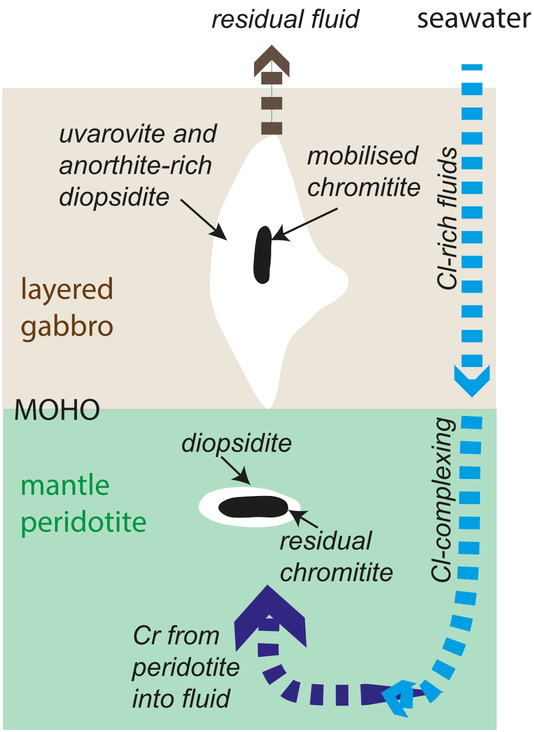

Fig. 3. Model for the formation of hydrothermal chromitites (modified after Arai and Miura, 2016). Cl-rich seawater penetrates the ocean crust and in the mantle section of the ophiolite dissolves Cr from Cr-bearing minerals in peridotites, creating diopsidite rims to former chromitite pods. Cooling fluids rise through the gabbro section creating complex diopsidites and small regions of mobilised chromite.

temperatures are 305 ± 10 °C into the liquid phase.

Johan et al. (2017) also report fluid inclusions in massive chromitites from the Al Ays ophiolite in Saudi Arabia and from ophiolites in New Caledonia. These inclusions tend to be up to 10 μm across and form primary and pseudo-secondary inclusions. The fluid is an aqueous saline solution containing 5 wt% equivalent of NaCl. The gas phase also includes a CO_2-CH_4 mixture, with very light carbon isotope ratios (−22 to −28 ‰). Carbon isotope fractionation between the two gaseous phases implies a closure temperature of at least 800 °C.

4. The influence of water on melt structure in the genesis of ophiolitic chromitites

Edwards et al. (2000) proposed a very different role for water in the genesis of ophiolitic chromitites. They suggested that the presence of water in a mafic melt enhances the solubility of chromium in the melt. Their argument is developed in the following steps:

- water is incompatible during mantle melting and so will preferentially partition into the melt;
- the water is dissolved in the melt, rather than existing as a separate phase;
- dissolved, dissociated water in a silicate melt has the effect of depolymerising the silica network;
- Cr^{3+} has very high octahedral site preference energy and so will be concentrated in melts where there is a high abundance of octahedral sites, ie, melts with a low degree of polymerization of the silica network;
- hydrous silicate phases will co-crystallise with the chromite.

The approach of Edwards et al. (2000) also sheds light on a related problem in the genesis of ophiolitic chromitites. Experimental studies show that chromite is often the first crystallising phase from a mafic melt and forms prior to the onset of silicate fractionation. And yet the solubility of Cr in a basaltic melt is low (max ca 340 ppm at 1400 °C, low oxygen fugacity and 1 atmos; Roeder and Reynolds, 1991) and the Cr-content is only enriched as those silicates phases in which Cr is incompatible begin to crystallise. How then can such high volumes of chromite be produced from what should be relatively low-Cr mafic melts? It is suggested that the presence of dissolved water in the melt explains this anomaly.

5. Discussion

In this section each of the three different approaches to understanding the role of water in the genesis of ophiolitic chromitites described above is examined and critiqued more fully. The aim of this discussion is to argue that the only viable role for water is as a dissolved species in the mafic melt as suggested by Edwards et al. (2000) and that its presence in the melt enhances the standard model of ophiolitic chromitite formation.

5.1. *The liquid immiscibility model of Matveev and Balhaus (2002)*

Four particular concerns arise directly from the study of Matveev and Ballhaus (2002) in which they propose a model in which there is the physical separation of chromite and olivine between an immiscible hydrous fluid and a silicate melt.

1. Is the proposed mechanism scalable to the size of nodules found in ophiolitic chromitites? In the study of Matveev and Ballhaus (2002) the hydrous fluid pools have a maximum diameter of 200 μm, whereas chromitite nodules observed in the field are much larger and typically 1–2 cm in diameter.
2. They propose a high percentage of dissolved water in their experiment (ca 4 %). Is this realistic, for it is much higher than that estimated for basaltic melts in ophiolites (MacLeod et al., 2013)?
3. Further, as noted by Matveev and Ballhaus (2002) themselves, how might the occurrence of antinodular textures in disseminated chromitites in the associated dunites also be explained (Rollinson and Adetunji, 2013)?
4. The original model of Matveev and Ballhaus (2002) applied strictly only to nodular chromitites. These are a relatively rare form of chromitite in the mantle.

5.1.1. *Geochemical and petrographic objections*

In addition there are geochemical and petrographic observations on nodular chromitites which raise questions about the suitability of the immiscibility model. The Maqsad chromitite dyke in the Oman ophiolite (Leblanc and Ceuleneer, 1992; Rollinson and Adetunji, 2013; Zagrtdenov et al., 2018; Henry et al., 2024) contains rounded, elliptical and angular chromitite nodules which are between 1 and 3 cm across and are enclosed in a matrix of olivine forming large single grains. At a macro scale the nodules exhibit a layered structure in some parts of the dyke (Leblanc and Ceuleneer, 1992). In detail the nodules comprise a mixed silicate-chromite core, up to 3–4 mm across, and have a chromite rim. The silicates present in the core include clinopyroxene and amphibole. Close to the rim of the nodule there are abundant rounded melt inclusions some of which contain hydrous phases. The individual chromite grains which make up the nodules are irregular and 0.5–1.0 mm in diameter. There is some chemical variation within and across the nodules. In the core, individual chromite grains may be zoned with core-rim compositions cr# = 0.59–0.52. In addition a traverse across the nodule shows a low-cr# core (cr# ~ 0.53), a higher cr# shoulder region (cr# ~ 0.57) and a lower cr# rim (cr# ~ 0.515).

Although there is petrological evidence for the presence of water in the melt during the crystallisation of the Maqsad chromitite dyke, the structural, textural and chemical variability in the chromites observed do not conform to the model proposed by Matveev and Ballhaus (2002). For example, there is no evidence for the clear separation between the crystallisation of silicate phases and chromite. Further, the variations in chemistry imply that the chromitite nodules evolved in equilibrium with an evolving silicate melt, rather than in a separate hydrous-fluid domain.

A similar view is taken by Pritchard et al. (2015) who described nodular chromitites from the Troodos ophiolite in Cyprus. In this instance the nodular chromitites contain chromite grains showing a rare skeletal macro-texture, which is thought to represent rapid crystal growth from a super-saturated magma. Using high-resolution X-ray computed tomography they show that some nodules also have small skeletal chromite grains in their cores overgrown by ‘polycrystalline equant chromite aggregates’. They interpret the rounded nature of the nodules as indicative of late chromite dissolution in the melt. Taken together the textural changes represent a change from super-saturation to equilibrium crystallisation to dissolution. On this basis Pritchard et al. (2015) argue that this supports a model of sequential, accretionary crystal growth of the nodules rather than the bubble-collection model of Matveev and Ballhaus (2002).

5.1.2. *Objections to the generalisation of the Matveev and Balhaus model*

In their extension of the Matveev and Ballhaus (2002) immiscibility model into a more general mechanism for chromitite genesis in ophiolites Su et al. (2020, 2021a, 2021b) make the additional observations:

- the water contents in the nominally anhydrous minerals (NAMs) in the Kızıldağ ophiolite in Turkey are in the range olivine 0–53 ppm; orthopyroxene 59–113 ppm; clinopyroxene 366–801 ppm (Su et al., 2020);

- in the Kızıldağ ophiolite there is a positive correlation between the water content of nominally anhydrous silicate minerals and the modal abundance of chromite implying that water in the silicate minerals was derived from hydrous fluids associated with chromite;
- ‘most chromitites are more highly serpentinized than the surrounding dunites and harzburgites’ implying a process of ‘self-alteration’; in the Kızıldağ ophiolite there is an increasing degree of alteration/serpentinisation outwards from chromitite through the dunite envelope to harzburgite suggesting that the hydrous fluids originated from the chromitite;
- anomalous ranges of stable isotope ratios for the elements Li, O, Mg and Fe in olivine and Mg and Fe in chromite in chromitites from the Pozantı-Karsantı ophiolite in Turkey suggest that these are the product of fluid mediated diffusion processes (Su et al., 2021b).

Here it is noted that the range of water contents observed in the NAMs reported by Su et al. (2020) is within or close to the normal range found in the upper mantle (olivine 0–200 ppm, enstatite 5–400 ppm, clinopyroxene 10–600 ppm, Novella et al., 2024) and so does not require anomalous fluid concentrations. Further, the clinopyroxenes with their relatively high water content have very different REE concentrations from those known to be of hydrothermal origin (Su et al., 2020) and so are unlikely to be hydrothermal in origin. In addition, whilst there may be an association between the degree of serpentinisation and chromitites in the Kızıldağ ophiolite this is not universally observed and not seen in the Oman ophiolite. More generally, some degree of serpentinisation is present in most ultramafic rocks even when there is no chromite present, suggesting that the observed association between hydrous fluids and chromitite mineralisation cannot support the genetic model. The variability in δ^{56}Fe and δ^{26}Mg in olivine and chromite observed in the Pozantı-Karsantı ophiolite in Turkey is also seen in other ophiolites such as Luobusa in Tibet (Xiao et al., 2016). In both localities the range of observed isotopic variations is very similar. Whereas Su et al. (2021b) interpret this variability as a function of fluid-mediated Fe—Mg diffusion, previous studies have proposed that it is as result of magmatic processes. Given that the magmatic model for chromitite genesis requires melt-rock interaction between a mafic melt and mantle harzburgite it would seem equally plausible that the observed isotopic variations are the product of diffusion processes related to melt-rock reactions and that a hydrous fluid is not required to explain these data.

It can also be argued from the experiments of Parman and Grove (2004), that under mantle melting conditions (1.2–2.2 GPa and 1175–1500 °C) water is present in the melt as structurally bound water, rather than free molecular water, and therefore unlikely to be present as an immiscible phase.

For these reasons it is argued here that Matveev and Ballhaus (2002) immiscibility model for the genesis of nodular ophiolitic chromitites is inadequate to explain their genesis. Further, the arguments used by Su et al. (2020, 2021a, 2021b) to extend this model into a generic model for all ophiolitic podiform chromitites are also thought to be implausible.

5.2. Hydrothermal Models

In their 2014 paper on the ‘Precipitation and dissolution of chromite by hydrothermal solutions’ Arai and Akizawa suggested that ‘the idea of a hydrothermal origin for chromitite can now be revived’. This work was based on their study of chromitiferous pods in hydrothermal diopsidites close to the Moho in Wadi Fizh in the Oman ophiolite, as discussed above. More recently however, Arai et al. (2020) point out that this process may have a limited application and may not apply to podiform chromitites in general. They observe that the hydrothermal diopsidic chromites that they describe are (a) relatively rare and of small volume, and (b) have a different composition from typical ophiolitic chromites inasmuch as they are enriched in Fe and Cr.

Nevertheless, the recent experimental work by Huang et al. (2019) on Cr complexing in a reducing environment to produce $CrCl(OH)^0$ makes the mobility of Cr in a hydrothermal environment plausible. This is particularly important given that Melcher et al. (1997) and Johan et al. (2017) both describe saline fluid inclusions in podiform chromitites. Whilst reports of fluid inclusions in ophiolitic chromitites are very rare, the presence of chlorine-bearing fluids is significant.

It is concluded therefore, that on rare occasions, in the present of Cl-bearing hydrothermal fluids, Cr can be mobilised from mantle ultramafic rocks and redeposited as chromitiferous diopsidites.

5.3. The role of dissolved water in the melt

The argument being advanced in this review is that the association between hydrous silicate mineral phases and chromitites in the mantle sequence of ophiolites is because they are derived from hydrous mafic melts. That is, the chromitite and the hydrous silicate phases have crystallised from mafic melts in which water is dissolved in the melt, not from a separate immiscible hydrous fluid, nor from an associated hydrothermal fluid.

5.3.1. The presence of water enhances Cr solubility in a mafic melt

Edwards et al. (2000) suggested that the presence of water in a mafic melt has the effect of depolymerising the silica network, creating more octahedral sites in the melt, thus enhancing the solubility of chromium. There is strong theoretical support for this approach as is evidenced in the early work of Kushiro (1972). More recently Mysen (2014) quantified the degree of polymerization in terms of the proportion of non-bridging oxygen (NBO) atoms relative to that of tetrahedrally (T) coordinated cations using the term NBO/T (Mysen, 2014). So for example, a typical tholeiite has a NBO/T ~ 0.8 whereas a more polymerised rhyolite has a value of about 0.1. Further, melt depolymerisation increases with increased water content of a melt as Si-OH bonds replace Si-O-Si bonds (Mysen, 2014). Experimental support for this view comes from the work of Parman and Grove (2004) who had shown that, during the hydrous melting of mantle harzburgite, water is predominantly present in its dissociated rather than molecular state, that is, it is dissolved as OH and serves as a network modifier. Indirect evidence for the role of water in melt polymerization also comes from trace element partitioning experiments. Gaetani et al. (2003) showed that during the hydrous partial melting of mantle peridotite the partition coefficients for the rare earth elements between clinopyroxene and melt are lower than those reported for dry melts. The discrepancy was attributed to the presence of water in the melt reducing the degree of polymerization and thus influencing the trace element partitioning.

5.3.2. Ophiolitic mantle chromitites are derived from wet melts

In the case of ophiolitic chromitites, the likely parental melts are tholeiitic basalt and boninite, as inferred from chromite compositions. In the Oman ophiolite Rollinson (2008) argued that chromites with cr# ~ 0.6 formed from basaltic melts whereas chromites with a higher cr# (cr# ~ 0.75) formed from boninitic melts. Similar values are found in other ophiolites (Zhang et al., 2023). In both cases these melts contain appreciable water, consistent with the argument developed here that water dissolved in the melt is the important control on chromitite formation. MacLeod et al. (2013) showed that basaltic melts in the Oman ophiolite had the composition of ‘moist MORB’ with initial water contents in the range 0.2–1 wt% H_2O, ‘significantly higher than for the MORB suite’. In addition, boninites are widely regarded as the product of the hydrous melting of mantle harzburgite (Falloon and Danyushevsky, 2000; Parman and Grove, 2004) and typically have high LOI values implying up to 6–8 wt% H_2O (Falloon et al., 2008).

It should be noted, that hydrous phases and melt inclusions have also been reported from chromitite micropods in modern oceanic basalts, typically regarded as having a very low water content (eg. Abe, 2011; Arai and Matsukage, 1998; Tamura et al., 2014). Thus apparently challenging the claim that the presence of hydrous phases and hydrous melt inclusions in chromite implies a wet basaltic melt. However, it must be recognised that these dunite hosted chromitite pods are rare and small (max 2 × 10 cm). In the context of this study it is argued that the relatively small volume of chromitite and rarity of hydrous primary phases associated with nominally dry melts in ocean basins supports the argument presented here. Namely, that when mafic melts are wet, as in the case of some ophiolites, then chromite precipitation is enhanced, whereas in dry mafic melts chromite formation is rare.

5.3.3. Wet mantle melting leads to higher melt yields with higher Cr concentrations

Experimental studies by Hirose and Kawamoto (1995) show that during the partial melting of spinel lherzolite the presence of water leads to (a) a lowering of the solidus and (b) an increased volume of melt at a given temperature relative to dry melting. Experimental studies also show that the Cr-concentration of a melt rises with increased melt-fraction, contrary to what might be expected of a compatible element with a large D-value (see Fig. 4, data from Hirose and Kawamoto, 1995; Johnston and Schwab, 2004). A similar feature is noted in the production of boninitic melts during the wet partial melting of harzburgitic mantle (Falloon and Danyushevsky, 2000; Parman and Grove, 2004). The implications of these experimental studies are that the presence of water during mantle melting leads to a larger melt fraction, which in turn has higher concentrations of Cr.

5.3.4. Trace element modelling

Johnston and Schwab (2004) show that the increase in Cr with increasing melt fraction in wet and dry melts of lherzolite is the result of decreasing D values for spinel and clinopyroxene with increasing melt fraction. A similar feature is evident for orthopyroxene in the data of Parman and Grove (2004). The partial melting of depleted harzburgite and lherzolite has been modelled here using partition coefficients derived from the studies of Johnston and Schwab (2004) and Parman and Grove (2004). The starting materials used here are from depleted harzburgites in the Oman ophiolite (Cr = 1860–3350 ppm, data from Hanghøj et al., 2010; Kanke and Takazawa, 2014) and lherzolite KLB1, (Cr = 2210 ppm). The results of harzburgite melting are given in Fig. 4 and show that at 20 % melting Cr concentrations are ~1400 ppm and at 30 % melting are ~2500 ppm, very similar to those reported in the Hirose and Kawamoto (1995) wet lherzolite experiments. It is also clear in Fig. 4 that the concentration of Cr in the starting material also strongly influences the amount of Cr in the melt.

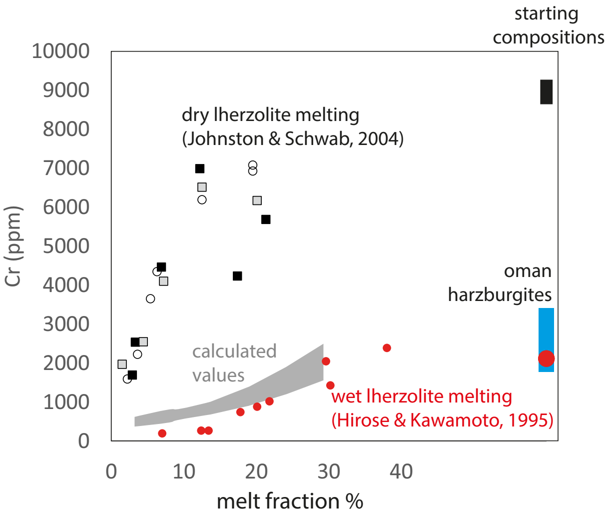

Fig. 4. Cr concentrations vs melt fraction in melting experiments on dry lherzolite (data from three different starting materials shown as black squares, grey squares and open circles after Johnston and Schwab, 2004) and on wet lherzolite melting (red circles, after Hirose and Kawamoto, 1995). The composition of the starting materials are colour coded and shown on the right of the diagram. Also shown in the grey shaded area are the compositions of melts from an Oman harzburgite (50 % olivine, 49 % orthopyroxene, 1 % spinel) and lherzolite KLB1 (48 % olivine, 47 % orthopyroxene, 7 % clinopyroxene 1 % spinel), calculated for batch melting using the starting compositions given in the text and using partition coefficients derived from the experiments of Johnston and Schwab (2004) – chromite, olivine and clinopyroxene, and Parman and Grove (2004) – orthopyroxene. From 5 % to 30 % mineral-melt melting partition coefficients decrease from 70 to 25 (chromite), 3.8 to 1.0 (clinopyroxene), 11 to 2 (orthopyroxene; olivine was set at 0.5. Orthopyroxene exerts a major control on the Cr concentrations in these models. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

In the Oman ophiolite tholeiites (n = 45) have Cr concentrations between 20 and 874 ppm (average 203 ppm) and low-silica boninites (n = 35, max. MgO =14.4 wt%) between 236 and 956 ppm (average 560 ppm), (data from Kusano et al., 2014; Belgrano et al., 2019, and see Fig. 5). The measured values shown in Fig. 5b are lower than the calculated values illustrated in Fig. 4 for 20–30 % melting, indicating that both basaltic and boninitic melts originally had excess Cr and the capacity to precipitate chromite.

5.4. Origins of the water

In an ophiolite water may become incorporated into a mafic melt either at the site of melting, through for example, the dewatering of a subducting slab triggering melting in the mantle wedge, or at the site of emplacement, through interaction with deep circulating fluids derived from seawater. Both types of fluid will be sodic and chlorine-rich but can be differentiated on the basis of the higher F and fluid mobile trace element concentrations in slab derived fluids (Ribeiro et al., 2015). In the Oman ophiolite amphibole and mica inclusions have high F, up to 0.57 wt%, and 0.31 wt% respectively (Borisova et al., 2012). In addition, melt inclusions in chromite are enriched in the fluid-mobile elements Cs, Rb, Ba and Sr (Rollinson et al., 2018) similar to those from the Mariana arc (Ribeiro et al., 2015), see Fig. 6. This suggests that the hydrous melts associated with ophiolitic chromitites in Oman have an affinity with fluids derived from a subducting slab. This is not to negate the observations of other authors who have shown that there were also deep, seawater-derived, hydrothermal fluids circulating in the ophiolite during its emplacement and cooling (see Section 5.2 above and Bosch et al., 2004; Dygert et al., 2017; Rospabé et al., 2017, 2019).

5.5. A model for the genesis of ophiolitic chromitites from hydrous melts

The following steps are envisaged in the genesis of ophiolitic chromitites from hydrous mafic melts:

- Hydrous fluids, probably derived from a subducting slab, initiate melting in the depleted harzburgites of a mantle wedge;
- The presence of water facilitates harzburgite melting and causes a rapid increase in the melt fraction;
- As the melt fraction increases, so does the solubility of Cr due to the influence of water on the structure of the melt leading the Cr-content of the melt to increase;
- The presence of chlorine in the hydrous melt may also facilitate Cr-complexing and dissolution;
- Reaction between the percolating, Cr-rich melt and the host harzburgite, through the dissolution of orthopyroxene and the crystallisation of olivine creates a dunitic sheath to the chromitite; it also leads to an increase in the silica content of the melt (and a consequent loss of octahedral sites), shifting the melt composition into the chromite field;
- The high Cr-content of the melt leads to extensive chromite crystallisation;
- Water in the melt becomes locked into sodic amphiboles and micas during the crystallisation of the melt.

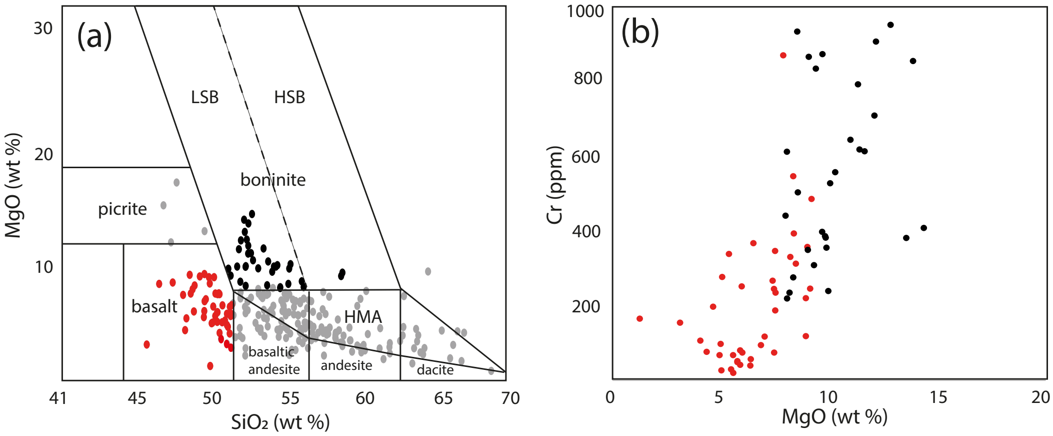

Fig. 5. (a) Boninite classification diagram after Pearce and Reagan (2019) showing boninites and tholeiites from the Oman ophiolite (data from Kusano et al. (2014) and Belgrano et al. (2019). LSB – low silica boninite, HSB – high silica boninite, HMA high magnesium andesite. The grey symbols represent the compositions of other volcanic rocks from the Oman ophiolite. (b) Plot of Cr (ppm) vs MgO (wt%) for tholeiites (red symbols) and boninites (black symbols) from the Oman ophiolite, (data from Kusano et al. (2014) and Belgrano et al. (2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

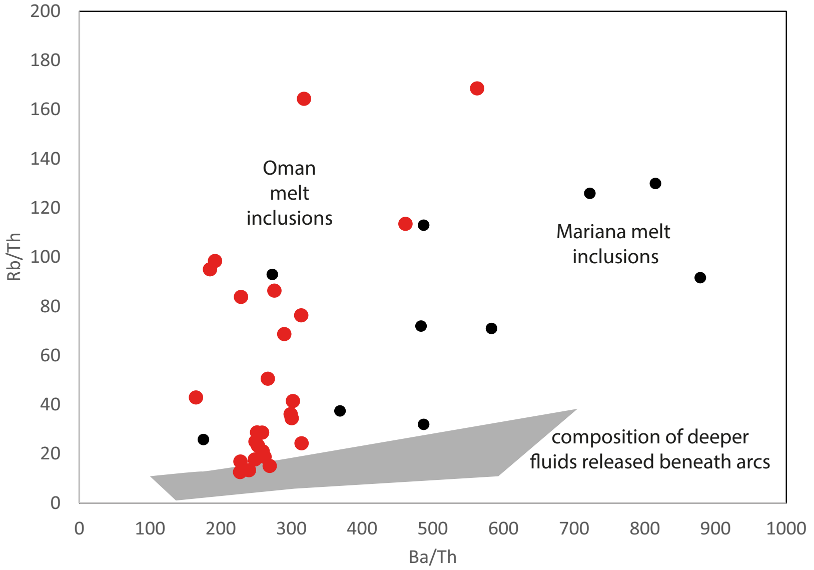

Fig. 6. Plot of Ba/Th vs Rb/Th for melt inclusions in lavas from the Mariana arc (black symbols) and the Oman ophiolite (red symbols) relative to the composition of fluids released from arcs where the subduction is deeper (grey field). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

6. Conclusions

There is good petrological evidence to show that water was involved in the genesis of chromitites found in the mantle section of ophiolites. This review has examined three different possible roles of the water in chromitite genesis, that of

- liquid immiscibility resulting in the partitioning of chromite between the silicate melt and a hydrous fluid,
- hydrothermally generated chromitite as a result of the mobilisation of Cr during deep seawater circulation in the ophiolite,
- dissolved water in the parental melt of the chromitites leading to enhanced Cr solubility during partial melting.

It is concluded that

1. The liquid immiscibility model does not satisfy the constraints imposed by petrographic and geochemical criteria for both the original concept, that of nodular chromitites, nor in the more generalised model applied to the broader suite of ophiolitic chromitites.
2. There is evidence that some chromitites have formed by hydrothermal processes, through the circulation of Cl-rich fluids, most probably seawater-derived; however, they are very rare, low in volume and geochemically different from most ophiolitic chromitites.
3. The presence of water during mantle melting leads to dissolved water in the melt, enhanced melting and increased Cr solubility. It is these Cr-enriched hydrous melts of basaltic and boninitic composition that are the likely parent of ophiolitic chromitites. Chromite is precipitated in the mantle harzburgite host through a process of melt percolation and melt-rock reaction. The hydrous melts also crystallise the phases sodic amphibole and micas.

4. There is geochemical evidence to suggest that in Oman the hydrous fluids were derived from the dewatering of a subducting slab.

CRediT authorship contribution statement

Hugh Rollinson: Writing – review & editing, Writing – original draft, Visualization, Validation, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The author declares that he has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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