The enigma of crustal zircons in upper-mantle rocks: Clues from the Tumut ophiolite, southeast Australia

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ABSTRACT

We suggest a new explanation for the presence of crustally derived zircons in the upper-mantle rocks of ophiolitic complexes, as an alternative to subduction-related models. Integrated isotopic (U-Pb, Hf, and O isotopes) and trace-element data for zircons from the Tumut ophiolitic complex (southeast Australia) indicate that these grains are related to granitic magmatism and were introduced into the mantle rocks after their emplacement into the crust. These observations emphasize that a clear understanding of the origin of individual zircon populations and their relationship to the host rock is essential to interpretations of the tectonic history of upper-mantle rocks and the dynamics of crust-mantle interactions.

INTRODUCTION

Several studies have reported U-Pb ages on zircons recovered from upper-mantle rocks in ophiolitic complexes (e.g., Grieco et al., 2001; Saveleeva et al., 2007; Stern et al., 2010; Badanina et al., 2013; Yamamoto et al., 2013; González-Jiménez et al., 2013). Did these zircons crystallize in the mantle from metasomatic fluids (e.g., Grieco et al., 2001; Zaccarini et al., 2004; Zheng et al., 2006), or are they xenocrysts of crustal rock recycled during subduction (e.g., Yamamoto et al., 2013; Robinson et al., 2014)? Crucially, what is their significance in the framework of ophiolite evolution?

We have studied the geochemical and isotopic signatures of zircons recovered from mantle and crustal rocks of the Coolac serpentinite belt, part of the Tumut ophiolite complex in the Lachlan fold belt of southeastern Australia (Fig. 1). The integrated analysis of U-Pb, Lu-Hf, and O isotopes has provided critical information about the origin of zircons in both mantle and crustal rocks. The zircon data are complemented by field observations and in situ Re-Os isotopic information on laurite grains in the chromitites of the mantle section of this ophiolite.

This case study emphasizes that deciphering the origin of zircons requires integration of data sets beyond zircon U-Pb geochronology alone. It reveals that crustally derived zircons can be introduced into mantle rocks after their obduction via microscopic melt networks, and sends a cautionary message about the interpretation of zircon ages from ultramafic rocks in general.

GEOLOGICAL SETTING AND SAMPLE BACKGROUND

The Coolac serpentinite belt, Wambidgee serpentinite belt, Tumut Ponds serpentinite belt, and smaller bodies form the ~250-km-long Tumut ophiolitic complex in the Lachlan fold belt of southeastern Australia (Fig. 1; Graham et al., 1996a, 1996b; Spaggiari et al., 2003). The Coolac serpentinite belt is ~263 km long, up to 3.5 km wide, and ≥2 km thick. It was metamorphosed mostly to greenschist facies and consists of variably serpentined porphyroclastic meta-harzburgites with lenses of dunite that host chromitite bodies. The ultramafic rocks are cut by dikes of gabbro and plagiogranite, and dike-like bodies, veinlets, and pod-like masses of rodingite. On its eastern contact, it is either faulted against or intruded by the Silurian S-type Young Granodiorite; to the west, it is faulted against metavolcanic and metasedimentary units of the Silurian Tumut trough (Stuart-Smith, 1990; Graham et al., 1996b).

The age of formation of the ultramafic rocks from the Tumut region is unclear. Previous suggestions range from Cambrian-Ordovician (Stuart-Smith, 1990) to Devonian (Graham et al., 1996b). Graham et al. (1996b) suggested that a Devonian U-Pb age (ca. 400 Ma), obtained on zircons from plagiogranite dikes, indicated that the ophiolitic rocks and granitic magmas were generated during the same major tectono-thermal event in the Lachlan fold belt (430–390 Ma; Chappell, 1994). However, this age is considerably younger than those of the Young Granodiorite (zircon U-Pb age of 428 ± 1.9 Ma; Lyons and Percival, 2002) and the North Mooney Complex (horn-
blende K-Ar age 425 ± 6 Ma; Stuart-Smith, 1990), leaving the relationships of the ultramafic rocks to the rest of the Lachlan fold belt ambiguous.

We separated zircons from samples of two (high-Al, high-Cr) massive chromitites as well as from samples of leucogabbro, plagiogranite, and rodingite. We also sampled sediments in gullies draining outcrops of weakly serpentinized massive porphyroclastic harzburgite. Zircons from the Young Granodiorite sampled at the contact were analyzed to refine the timing of granitic magmatism and its relationship to the ultramafic-mafic rocks.

Several grains of laurite (RuS₂; up to 10–20 μm) were identified in polished thin sections of massive (high-Cr) chromitite from the Quilter’s open-cut mine in the Coolac serpentinite belt. Their predominantly euhedral shapes and occurrence within fresh chromite grains (Fig. DR1 in the GSA Data Repository) suggest a primary magmatic origin. Laurite grains were identified by their X-ray spectra, and their Os isotope compositions were analyzed in situ.

Detailed information on sample locations, sample processing, and analytical techniques are provided in Appendix DR1 and Tables DR1–DR5 in the Data Repository.

RESULTS

Analytical data are given in Tables DR2–DR5. Table 1 provides a summary of the information collected on zircons.

Zircon U-Pb Age Dating

The most important observation from the U-Pb data is that the main zircon populations from the Coolac belt rocks (430.5 ± 4.5 Ma for plagiogranite, 429.4 ± 4.7 Ma for leucogabbro; Figs. DR2 and DR3) are indistinguishable from the age defined for the Young Granodiorite during this study (428 ± 3 Ma; Fig. DR4) and previously (428 ± 1.9 Ma; Lyons and Percival, 2002). Detrital zircons from the stream samples are also dominated by Early Silurian age populations (432.9 ± 3.1 and 426.6 ± 6.3 Ma; Figs. DR5–DR7), indistinguishable from the ages defined for the Coolac and Young Granodiorite samples. The inherited populations in both Coolac and Young Granodiorite samples are also similar: ages range from Ordovician to Paleoproterozoic and Archean with clusters around 442–470 Ma, 1.0–1.3 Ga, 1.7–1.8 Ga, and 2.2–2.4 Ga (Table 1; Figs. 2 and 3; Fig. DR8). Moreover, the youngest (ca. 370–385 Ma) populations of zircons from the Coolac samples have ages identical to the rims on zircons from the Young Granodiorite.

Sixteen (16) U-Pb analyses on 12 zircon grains recovered from the chromitite samples also show ages ranging from Ordovician (442 ± 5 Ma, 1σ) to Paleoproterozoic (2289 ± 25 Ma), with a small cluster (n = 3) around 595 Ma (Figs. 2 and 3; Fig. DR9); grain morphology and the scatter in age support their xenocrystic origin. Zircons from the rodingite show ages from Devonian (376 ± 9 Ma) to Paleoproterozoic (1718 ± 22 Ma) with a population at 390 ± 5 Ma (Figs. 2 and 3; Fig. DR10); this younger age is indistinguishable from the age of the younger overprint observed in zircons from the other Coolac samples and the Young Granodiorite (Table 1).

Zircon O- and Hf-Isotope Data

Most of the Coolac belt zircons show δ18O ranging from 6.5‰ to almost 10‰ (Fig. 2; Table 1), clearly outside the typical mantle range (5.3‰ ± 0.3‰; Valley, 2003) and much higher than the range defined by zircons from plagiogranites of other ophiolitic complexes (3.9‰–5.6‰; Grimes et al., 2013). By and large, the Coolac zircons are dominated by crustal O isotope compositions, typical of zircons from granitoids of the Lachlan fold belt (Kemp et al., 2009). They overlap the main magmatic population from the Young Granodiorite (7.7‰–10‰; Fig. 2). A more restricted range of δ18O (7.5‰–8.4‰; Fig. 2B) is observed in the younger population of zircons from the rodingite sample; this coincides with values in the rims of the granodiorite zircons (8.0‰ and 8.2‰).

The crustal O-isotope signatures of the studied zircons are consistent with their non-radiogenic Hf isotope compositions; most grains show negative εHf (Fig. 3; Fig. DR11; Table 1), mainly from 0 to –16 for the Coolac samples and from –2 to –9.5 for the Young Granodiorite. The mean Hf model ages (TDM) of the main magmatic populations range from 1.6 to 1.7 Ga and from 1.5 to 1.8 Ga for the Coolac rocks and granodiorite, respectively. However, in grains from the plagiogranite sample, crustal δ18O values contrast with a distinctive mantle Hf isotope signature (εHf ranging from +9 to +13, and mean TDM of 0.76 Ga).

Several (mainly inherited) grains from the Coolac samples plot within or close to the mantle field in Figure 2. However, a few zircons from the main Early Silurian population, including one from the plagiogranite, and two detrital zircons, have both juvenile εHf and mantle δ18O values. These two detrital zircons (grains 24 and 41; Tables DR2–DR4) have εHf of +15.5 and +14.8 and are dated at 439 ± 10 Ma and 442 ± 12 Ma, respectively.

Zircon Trace-Element Composition

Zircons from all the Coolac belt samples have trace-element compositions typical of continental-crust zircons (Belousova et al., 2002); none plot within the U/Yb–Y field that is unique to oceanic-crust zircons (Fig. DR12; Grimes et al., 2007). Both magmatic and inherited populations show nega-

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**TABLE 1. SUMMARY OF THE U-Pb AGE AND HI- AND O-ISOTOPE INFORMATION FOR ZIRCONS FROM THE COOLAC SERPENTINITE BELT, SOUTHEAST AUSTRALIA**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Rock type</th>
<th>Main population/ inferred crystallization age (Ma)</th>
<th>Younger overprint (Ma)</th>
<th>Inheritance (Ma)</th>
<th>Hf model ages*</th>
<th>O isotope signature*</th>
</tr>
</thead>
<tbody>
<tr>
<td>M104-M13</td>
<td>Chromitite (high-Cr)</td>
<td>–</td>
<td>–</td>
<td>442–2289</td>
<td>TDM ** 1.18–1.64</td>
<td>Mainly crustal</td>
</tr>
<tr>
<td>ML101A</td>
<td>Chromitite (high-Al)</td>
<td>–</td>
<td>–</td>
<td>470–1953</td>
<td>–</td>
<td>Mainly crustal</td>
</tr>
<tr>
<td>RH106</td>
<td>Rodingite</td>
<td>390 ± 5 (?) (1)</td>
<td>376–390 (?)</td>
<td>424 (?)–1718</td>
<td>1.24 &amp; 1.59 (?)</td>
<td>Mainly crustal</td>
</tr>
<tr>
<td>#298</td>
<td>Leucogabbro</td>
<td>429.4 ± 4.7 (15)</td>
<td>361–416</td>
<td>447–408 (3412?)</td>
<td>1.26 &amp; 1.63</td>
<td>Mainly crustal</td>
</tr>
<tr>
<td>#20</td>
<td>Plagiogranite</td>
<td>430.5 ± 4.5 (11) / 439 ± 10 (1)</td>
<td>375–420</td>
<td>458 ± 6</td>
<td>0.66 &amp; 0.76</td>
<td>Strongly juvenile</td>
</tr>
<tr>
<td>TC12-ML01</td>
<td>Stream sample</td>
<td>432.9 ± 3.1 (15)</td>
<td>385 ± 6</td>
<td>456–1877</td>
<td>1.30 &amp; 1.68</td>
<td>Mainly crustal</td>
</tr>
<tr>
<td>TC12-BG01</td>
<td>Stream sample</td>
<td>426.6 ± 6.3 (4)</td>
<td>388 ± 5</td>
<td>471–2257</td>
<td>1.27 &amp; 1.66</td>
<td>Mainly crustal</td>
</tr>
<tr>
<td>Young Granodiorite</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>YG202</td>
<td>Granodiorite</td>
<td>428 ± 3 (11)</td>
<td>367–385</td>
<td>450–3053</td>
<td>1.25 &amp; 1.62</td>
<td>Crustal</td>
</tr>
</tbody>
</table>

*Note: TDM—depleted mantle model age; TDM—mean Hf crustal model age.

*For main magmatic population (excluding inherited grains) or youngest populations.

**TDM (Re-depletion model age); Re-Os model age from laurite.

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1GSA Data Repository item 2015049, Appendix DR1, Tables DR1–DR5, and Figures DR1–DR17, is available online at www.geosociety.org/pubs/ft2015.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.
In contrast, the trace-element signatures and REE distribution patterns of zircons from the plagiogranite are clearly distinct from those of the rest of the zircons of the Coolac belt and the Young Granodiorite: they have lower Hf contents (<1 wt%, common in zircons from mafic rocks), and lower Th/U. Apart from a single grain, they lack Eu anomalies (Fig. DR14A), suggesting that they crystallized before plagioclase. The only feature in common with the rest of the samples is variable LREE enrichment. The grain (15) that appears to be least affected by alteration (no LREE enrichment) is the only one with a mantle O-isotope composition (\( \delta^{18}O = 5.61\% \)) and that also yielded a slightly older U-Pb age (439 ± 10 Ma) than the other zircons in the main population from this plagiogranite. Furthermore, \( \delta^{18}O \) in the plagiogranite sample increases with decreasing U-Pb age (Fig. 2C), suggesting that zircons from this sample have experienced progressive alteration by \(^{18}O\)-enriched supracrustal fluids soon after their crystallization. Thus, the lone zircon “survivor”, with no LREE enrichment and a mantle O-isotope composition, suggests that the plagiogranite is at least 439 Ma old. It is important that the two detrital grains (grains 24 and 41) with mantle isotopic signatures also have “undisturbed” REE patterns (Fig. DR14A).

**Laurite Re-Os Data**

The Os-isotope data for 10 laurite grains from the chromitites (Table DR5) show a range of unradiogenic \(^{187}Os/^{188}Os\) (0.1146–0.1220) and low \(^{187}Re/^{188}Os\) ratios. Their \(T_{RD} \) (Re-depletion) model ages (relative to Enstatite Chondrite Reservoir; Walker et al., 2002) vary between 0.84 Ga and 1.89 Ga, with a mean peak at around 1.18 Ga and a shoulder at ca. 1.64 Ga (Fig. DR15). The older \(T_{RD} \) model ages overlap the range of zircon Hf crystall model ages (ca. 1.62–1.68 Ga), except for the plagiogranite sample (mean \(T_{RD} \) of 0.77 Ga). The youngest “event” (\(T_{RD} = 0.84 \) Ga) recorded by the Re-Os system is very close to the age of the protolith reflected in the Lu-Hf systematics of the plagiogranite zircons.

**DISCUSSION**

**Origin of Crustal Zircons in Upper-Mantle Rocks of the Coolac Belt**

The combined isotopic and trace-element data indicate that most zircons are of crustal origin. Previous studies of crustal zircons in upper-mantle rocks (including chromitites) have interpreted such grains as remnants of crustal rocks subducted into the mantle or as material picked up by ascending melts and/or fluids released during dehydration of subducting slabs containing crustal sedimentary rocks (e.g., Stern et al., 2010; Yamamoto et al., 2013; Robinson et al., 2014; Zhou et al., 2014). However, in this case the striking resemblance to zircon populations from the adjacent Young Granodiorite suggests that the grains recovered from the Coolac belt rocks are related to the granitic magmatism. Indeed, the isotopic and trace-element signatures recorded in the Coolac zircons are consistent with the timing and the nature of granitic magmatism within the larger Lachlan fold belt (420–390 Ma; Chappell, 1994). This similarity includes not only the grains of the main magmatic populations, but the inherited populations and the younger overprint (Table 1); this could hardly be a coincidence.

The presence of similar populations of zircons in ultramafic rocks, chromitites, and granitic rocks suggests that late melts from the granite batholith penetrated the ultramafic rocks. Granodioritic and aplite dikes from the Young Granodiorite intrude the Coolac belt peridotites (Stuart-Smith, 1990; Graham et al., 1996b), and zircon could have been more widely introduced via intergranular networks percolating from the larger fluid channels (Fig. DR16). These fluids precipitated zircons of the latest generations (e.g., 375–390 Ma) and transported grains of the main magmatic and inherited populations. Such intergranular networks could have healed later, leaving little visible record under conventional microscopy.

These crustally derived fluids were most likely responsible for the alteration of primary magmatic zircons in the plagiogranite, where enrich-
ment in LREE and increase in δ18O correlates with a progressive resetting of the U-Pb isotope system. The fact that the Hf-isotope system in these zircons retained the original mantle signature emphasizes the robustness of this system.

**Implication for the Local and Regional Geology**

The Re-Os data from the laurites indicate that the mantle section in the Coolac serpentinite belt is at least Mesoproterozoic in age, with a main tectonomagmatic event recorded at ca 1.2 Ga. The coincidence of Os model ages on laurite and HF crustal model ages on zircon at 1.64–1.68 Ga implies that Proterozoic lithosphere (or at least ribbons of it) extends as far as the Tumut region of the Lachlan fold belt and supports existing models (Betts et al., 2002, and references therein) allowing for incorporation of an older continental ribbon into the Paleozoic crust during back-arc inversions.

Our observations suggest that the zircon U-Pb age information reported here and in previous studies (e.g., Graham et al., 1996b) cannot be used directly to constrain the timing of magmatic events that have produced the upper-mantle rocks of the Coolac belt. The U-Pb age of the zircon “survivor” from the plagiogranite suggests that the ultramafic rocks were part of an oceanic lithosphere ≥439 m.y. ago. This is supported by the ages (439 ± 10 Ma and 442 ± 12 Ma) of two detrital zircons with mantle isotopic compositions, recovered from the Mount Lightning-stream sample TC12-ML01 (Fig. 1; Table DR1). Even if we consider a subduction-related model to explain the presence of crustal zircons in upper-mantle rocks, then the youngest xenocrystic grain would provide us with a maximum age estimate for subduction. The youngest zircon from the chromitites that has a supracrustal O-isotope composition (δ18O = 7.9 ± 0.4, 2ε) was dated at 442 ± 10 Ma, which is consistent with this timing.

**Broader Implications**

We have presented a new explanation for the presence of crustal zircons in the upper-mantle sections of ophiolitic complexes, as an alternative to the existing subduction-related models. In this case, the granitoid-related melts and/or fluids, injected into already-emplaced mafic-ultramafic rocks, apparently carried existing zircons (including inherited ones) and possibly crystallized new grains. This study emphasizes that integrated information on zircons (not U-Pb data alone) is critical to the interpretation of events recorded by upper-mantle rocks, including their emplacement and subsequent tectonic history. A clear understanding of the origin of xenocrystic zircons and their relationship with the host rock increases the probability of correct conclusions about the history of uppermantle rocks (e.g., the upper-mantle sequences of ophiolitic complexes) and interpretations of the dynamics of crust-mantle interaction.

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