On the difficulty of assigning crustal residence, magmatic protolith and metamorphic ages to Lewisian granulites: constraints from combined in situ U–Pb and Lu–Hf isotopes

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Abstract: Zircons from two granulite facies gneisses from the central region of the Lewisian Complex have been investigated by high spatial resolution ion-microprobe U–Pb dating and laser ablation combined Pb–Hf isotope methods. The ion-microprobe data reveal a complex pattern of zircon ages distributed along the concordia curve between the time of granulite facies metamorphism at c. 2.5 Ga and the oldest zircon in each sample (respectively 2.89 Ga and 3.04 Ga). This Pb-loss pattern complicates assignment of an unambiguous magmatic protolith age to the zircon although cathodoluminescence (CL) imaging is used to suggest a preferred age of c. 2.85 Ga for both samples, with older grains being inherited. In situ Hf isotopes show a larger spread in the sample containing older grains which is also consistent with inheritance and further suggests that several crust extraction events are represented in the inherited population. Comparison of Hf isotope compositions with plausible model evolution curves suggests that crustal precursors to the Lewisian granulites were derived from their mantle source at c. 3.05–3.2 Ga.

Supplementary material: Analytical methods and data tables are available at: http://www.geolsoc.org.uk/SUP18395

The Lewisian Complex of NW Scotland (Fig. 1) has long played a key role in models for the evolution of Precambrian high-grade gneiss terranes. This is due in no small measure to the pioneering mapping and profound insights of the Geological Survey Highlands team at the turn of the 20th century culminating in the classic NW Highlands memoir (Peach et al. 1907). On the basis of a suite of dykes which were recognized as having intruded after formation of gneissic banding but prior to later movements, these workers established a tripartite division of the mainland outcrop of the Lewisian gneiss (as they then termed it) into northern (north of Loch Laxford), central (between Loch Laxford and Grui- nard Bay) and southern regions which is essentially still in existence, albeit somewhat modified. Their methodology was mirrored in the classic work of Sutton & Watson (1951) who further developed techniques of studying such complex rocks that have become commonplace. In a notably prescient passage, Peach et al. (1907, p. 44) stated...

...it is clear... that the Lewisian gneiss is not a geological formation in the ordinary sense of the word. Even if we exclude from it the later dykes and sills, there still remains a petrographical complex which future research will probably separate into its component parts.

Almost contemporaneous with Peach et al.’s geological mapping, Henri Bequerel’s discovery of radioactivity laid the foundation for modern methods of radiometric dating that would provide the means to test and develop their observations in a quantitative framework. The earliest radiometric dating studies using K–Ar (Holmes et al. 1955) provided minimum ages for the Lewisian of around 1 Ga but were hampered by inaccurate decay constants and large analytical uncertainties. Later investigations using the Rb–Sr system (Giletti 1959; Giletti et al. 1961) established a broad division into the late-Archaean (‘Scourian’) and mid-Proterozoic (‘Laxfordian’) metamorphic events that was not significantly different from our knowledge today. By the mid 1970s, a late-Archaean age for the earliest high grade metamorphism of the gneisses was well-established by whole-rock Pb/Pb (Chapman & Moorbath 1977) and zircon U–Pb methods (Pidgeon & Bowes 1972). Development of the Sm–Nd method in the late 1970s provided...
the first means to ‘see through’ early metamorphism and date the crust-mantle differentiation directly, testing the then prevailing model of ‘crustal accretion differentiation superevents’ (Moorbath & Taylor 1981) which predicted that separation of igneous precursors from a mantle source region should have occurred c. 100–200 Ma prior to the earliest metamorphic event. In its first application to the Lewisian gneisses, Sm–Nd systematics indicated a mantle separation age of 2920 ± 50 Ma.
(Hamilton et al. 1979). This Sm–Nd isochron, however, combined whole rock samples from the northern and central regions as well as felsic and mafic gneisses that were demonstrably non-cogenetic. Whitehouse (1988, 1989) applied the bulk-rock Sm–Nd method separately to suites of tonalite-trondhjemite-granodiorite (TTG) samples from the Scourie and Gruinard Bay parts of the central region and the northern region as well as a series of mafic-ultramafic enclaves. Despite large errors on regression ages from the TTG gneisses due to limited spread in Sm–Nd ratios, data from the granulate facies gneisses from Scourie were interpreted in terms of granulite facies Nd-isotope rehomogenization at 2660 ± 155 Ma (Whitehouse 1988), with the initial εNd value of −2.4 ± 1.9 pointing to a lengthy pre-metamorphic crustal residence. Using average depleted mantle model ages (tDM) as an indicator of regional crustal residence age, Whitehouse (1989) proposed that the precursors to the central region gneisses with an average tDM of 2.93 Ga separated from mantle almost 150 Ma earlier than the northern region gneisses which yielded an average tDM of 2.78 Ga, indicating significant differences in crustal history across the major boundary recognized to the south of Loch Laxford (the Laxford Shear Zone – see Goodenough et al. 2010) by Peach et al. (1907) and Sutton & Watson (1951).

Against this backdrop of whole-rock isotopic studies, the 1990s saw the application of increasingly sophisticated U–Pb dating methods to solve the problem of magmatic protolith age and metamorphic reworking in the Lewisian. High-precision isotope dilution thermal ionization mass spectrometry (TIMS) measurements on zircon revealed metamorphic episodes in the Scourie region at ≥2.71 Ga and 2.49 Ga (Corfu et al. 1994), the younger age also having been recorded by mineral Sm–Nd systematics (Humphries & Cliff 1982). The high spatial resolution ion microprobe (or secondary ion mass spectrometry, SIMS) method applied to zircon grains whose internal structures had been imaged by cathodoluminescence (CL) also revealed a 2.49 Ga age for a metamorphic event and, by targeting cores in polyphase zircon, was used to suggest magmatic protolith ages of c. 2.96–3.03 Ga for central region gneisses (Friend & Kinny 1995; Kinny & Friend 1997), at the same time revealing significantly younger protolith ages for the northern region gneisses (c. 2.84 Ga) consistent with the observations from bulk-rock Sm–Nd isotopes (Whitehouse 1989).

Derivation of 3.03–2.96 Ga protolith ages from central region granulate facies gneisses (Friend & Kinny 1995; Kinny & Friend 1997) was largely based on the principle of taking the oldest concordant 207Pb/206Pb age(s) as the age of the magmatic protolith and assuming that younger ages represent variable degrees of Pb loss during the later c. 2.5 Ga event and a possible unresolved intermediate age event (e.g. that of Corfu et al. 1994). This approach generally assumes that there are no inherited components in the protolith magma, although in the case of one sample examined by Kinny & Friend (1997), a single 207Pb/206Pb age of 3115 Ma from a rounded core in an otherwise 2.93–2.97 Ga grain was interpreted as inherited. Consideration of the SIMS U–Pb datasets presented in these earlier studies shows that in some cases, a cluster of younger ages occurs at c. 2.8 Ga, comprising the dominant pre-2.5 Ga population in two samples (GST9 and GST12) as well as a major component of a third (GST8). In another sample (GST10), the c. 2.8 Ga cluster is largely absent, with >2.9 Ga grains dominating the age spectrum, while the most complex sample (GST11) shows three age groups at c. 2.8, 2.87 and 2.95 Ga respectively. On the basis of these data, an alternative interpretation is that at least some of the samples have a magmatic protolith age of c. 2.8 Ga, with >2.8 Ga representing inheritance. Only the sample with dominant >2.9 Ga grains (GST10) appears to present a relatively strong case for a c. 2.95 Ga protolith. In all cases, however, the total number of grains analysed from each sample is too small (<20) to assign statistical significance to any of the age groups.

A ≥2.71 Ga metamorphic event recognized in zircon U–Pb systematics by Corfu et al. (1994) is consistent with ion microprobe U–Pb studies of monazite (Zhu et al. 1997) as well as a number of earlier whole rock Sm–Nd (Whitehouse 1988, 1989), mineral Sm–Nd (Humphries & Cliff 1982) and Pb/Pb (Chapman & Moorbath 1977; Cohen et al. 1991) studies. On the basis of their ion-microprobe data set, however, Friend & Kinny (1995) considered it impossible to say whether a discrete event at c. 2.7 Ga event actually occurred in their investigated central region granulites, although they did not provide any alternative explanations for the aforementioned observations beyond vaguely assigning the monazite ages of Zhu et al. (1997) to the possible onset of granulite facies metamorphism. Subsequent development of the terrane model for the Lewisian (Love et al. 2004; Kinny et al. 2005), subdivided the central region into the Assynt (northern) and Gruinard (southern) terranes, in part by summarily dismissing evidence for a c. 2.7 Ga event in the former. In contrast to the general acceptance of broad differences between the central and northern region gneisses across the Laxford Shear Zone (Park 2005; Goodenough et al. 2010), vigorous debate continues on the existence and significance of an early c. 2.7 Ga metamorphism within the central region itself, as well as the re-assignment of previously recognized and
In this paper, we present U–Pb zircon age data from two granulite facies gneisses from the central region (Assynt terrane) that are considered to be close equivalents of two previously investigated samples. We adopt an identical methodology to Friend & Kinny (1995), namely CL-imaging guided SIMS, applied to a significantly greater number of grains in order to detect possible peaks in the pre-2.5 Ga spectrum that might relate to magmatic protolith age. The existence of a possible 2.7 Ga metamorphic event will also be addressed by these data. Additionally we present the first Hf isotope data obtained using laser ablation ICP–MS on the SIMS-dated zircons. The Hf-isotope data aid the interpretation of the complex zircon U–Pb systems and provide new constraints on the age and composition of the source components for the Lewisian tonalites.

**Sampling**

Two samples of granulite facies tonalitic gneiss were collected from similar localities to samples documented by Friend & Kinny (1995), Kinny & Friend (1997) and Corfu et al. (1994). However, since both samples were collected before the routine availability of precise GPS navigation, exact equivalence to previously described samples cannot be assumed.

Sample Lew99-Ky is a granulite facies tonalitic gneiss collected at Kylestrome from a small roadside outcrop (possibly previously quarried) approximately 100 m to the NW of the disused Loch Glencoul ferry ramp (Great Britain Ordnance Survey grid reference NC22863427). This is the assumed to be the same quarry sampled by Pidgeon & Bowes (1972, sample RC287) and Friend & Kinny (1995, sample GST12) although both of these papers quote an apparently erroneous grid reference that is c. 80 m to the west of the ferry ramp on the low water mark shoreline.

Sample Lew99-GE is a granulite facies tonalitic gneiss collected from Geodh nan Anruig (Scouriemore) in the vicinity of tonalitic gneiss samples previously described by Kinny & Friend (1997, sample GST10) and Cohen et al. (1991, sample 043) at grid reference NC142442. Based on an annotation in the 1996 Metamorphic Studies Group field guide, it is likely that these earlier sample sites are within a few metres of each other. The precise sample location is at the base of a small NW facing overhanging cliff c. 30 m to the SE and approximately 3 m higher than a prominent flat outcrop of pegmatite.

This same location showed signs of recent hammering and is probably one of the previous sample sites noted above.

**Results**

**Zircon characteristics and SIMS**

**U–Th–Pb results**

Lew99-Ky. Zircon separated from Kylestrome sample Lew99-Ky range in size from <100 to >300 μm, are equant to elongated with length:width ratios up to c. 3:1 and generally have a sub-rounded habit typical of high-grade metamorphic zircon. Cathodoluminescence imaging (Fig. 2a and Supplementary Material) reveals complex internal structures resulting from the polyphase history of the host rocks. Irregularly shaped zircon dark cores are found in most grains, some with oscillatory zoning (e.g. grains 24, 70). In some cases, possible polyphase cores are also evident (e.g. grains 12, 15). Cores are ubiquitously overgrown by a CL-medium-bright overgrowth which truncates zoning in the cores (e.g. grains 17, 24, 45). The rims generally lack internal growth zoning, instead exhibiting faint, irregular structures. Most grains also preserve an extreme outer rim just a few μm thick that is slightly darker in CL response and, in some cases (e.g. grain 34) is continuous with apparent annealed fractures. A number of grains are CL-medium (apparently) single phase, although it cannot be ruled out that these also contain cores below the level of polishing.

A total of 76 zircon grains were analysed by SIMS for U–Th–Pb, with 120 spot analyses performed in the distinct growth zones. Post-analytical imaging revealed that, in a few cases, the analytical spot sampled across two growth zones; these analyses are noted in the Supplementary Material and are not considered further. Cores range widely in U concentration from c. 15–800 ppm with Th/U mostly in the range 0.4–1.3, rims and single phase grains have a more restricted U concentration range from c. 10–30 ppm but with similar Th/U ratio to the cores (Fig. 3a). On a concordia diagram (Fig. 4), the data spread along concordia between a maximum core 206Pb/238U age of 2890 Ma and an age of c. 2500 Ma defined by the majority of the rim and single-phase analyses.

Considering first the core analyses (n = 62), the spread observed in these down to c. 2500 Ma (Figs 4a & 5a) most likely results from at least one episode of Pb-loss which, in some cores with ages as low as 2500 Ma, has been effectively complete. This extreme Pb-loss is consistent with the 2.5 Ga event being the main granulite facies metamorphic event in the region. There is, however, no
negative correlation of age with U concentration and, in fact, some of the youngest ages are recorded by cores with the lowest U content, while ages in excess of 2.8 Ga are preserved across almost the entire range of U content (Fig. 3b). There is also no obvious correlation between age and degree of zoning in the cores, although it is worth noting that two grains preserving fine-scale oscillatory zoning are among the oldest. Given the pervasive Pb-loss overprint, defining a single precise magmatic protolith age for this sample is complex and subject to various interpretations.

Following the approach of Friend & Kinny (1995) by assuming that the observed data derive from zircon formed during a single magmatic event, the oldest $^{207}\text{Pb}/^{206}\text{Pb}$ age of 2890 ± 36 Ma may be taken to represent this, although this particular analysis is slightly reverse discordant. If this reverse discordance is due to an analytical artefact affecting the U/Pb ratio calibration, the $^{207}\text{Pb}/^{206}\text{Pb}$ age will remain accurate. If however, the reverse discordance is a real phenomenon, reflecting ancient localized redistribution of radiogenic Pb formed over a short interval, a process proposed by Hinton & Long (1979) but to date undemonstrated, the $^{207}\text{Pb}/^{206}\text{Pb}$ age could be overestimated. Two other concordant grains have similar $^{207}\text{Pb}/^{206}\text{Pb}$ ages and together, the three oldest ages yield a weighted average age of 2886 ± 15 Ma [mean square of weighted deviates (MSWD) value = 0.08].

An alternative approach commonly used in the analysis of SIMS data from polyphase zircon again assumes that a single magmatic protolith age component is present and has been subject to later Pb-loss resulting in a spread of ages. Successive rejection of the youngest ages from the weighted average is performed until its MSWD reaches a threshold value at which assigned analytical errors alone can account for the observed scatter of
data. Accepting an MSWD value of 1.0 as the threshold value results in a weighted average $^{207}\text{Pb} / ^{206}\text{Pb}$ age of $2872 \pm 10$ Ma (MSWD = 0.94, probability = 0.46) based on the oldest seven analyses. Grouping of the oldest 13 analyses results in a weighted average age of $2861 \pm 9$ Ma with only a modest increase in MSWD to 1.3 (probability = 0.19), while inclusion of the next youngest analysis causes the MSWD value to increase significantly to 2.1. From a purely objective statistical point of view, an age in the range 2860–2870 Ma is therefore supported by the oldest 13 out of 62 core analyses, the remainder reflecting variable degrees of Pb loss during later metamorphic events.

Applying a more subjective interpretation to the cumulative age plot for Lew99-Ky cores, the three oldest cores could be assigned to a possible inherited component. Disregarding these and grouping the next 13 youngest analyses yields a weighted average $^{207}\text{Pb} / ^{206}\text{Pb}$ age of $2848 \pm 6$ Ma with a still acceptable MSWD value of 1.1, while addition of another younger analysis expands MSWD to $>2$. Cathodoluminescence images and Th–U chemistry do not obviously support such an interpretation however, since the three oldest cores are not distinctive (Fig. 3b, c).

Interpretation of the younger rim ($n = 37$) and single phase ($n = 11$) data (Fig. 4b) as metamorphic accords with previous interpretations from central region Lewisian granulites (Corfu et al. 1994; Friend & Kinny 1995; Kinny & Friend 1997). On a probability density plot (Fig. 4b) most of these analyses define a slightly skewed to younger age peak around 2500 Ma, together with a handful of older analyses. A coherent group of 30 analyses defines a weighted average $^{207}\text{Pb} / ^{206}\text{Pb}$ age of $2494 \pm 10$ Ma (MSWD = 1.2). Skewing of the peak to younger ages that are still concordant within analytical error might be related to further post 2495 Ma disturbance of the system during formation of the thin outer rim and annealed cracks although no age can confidently be assigned to this event using these data. Older analyses range in age up to $c. 2830$ Ma and in most cases, probably reflect incomplete resetting of U–Pb systematics in older zircon during formation of the low-U, structureless rims. Depth profiling into older cores buried just below the CL-imaged surface cannot be entirely ruled out although only one analysis showed a marked increase in radiogenic Pb counts during analysis and a change in age of the sampled zircon can explain the substantially larger analytical error in this case.

Lew99-GE. Zircon from the Geodh Eanruig sample are similar in size, external appearance and internal structure as revealed by CL imaging (Fig. 2b and Supplementary Material) to those from Lew99-Ky.
and the same tripartite division of growth phases into irregularly shaped, mostly CL-dark cores, CL-bright rims and single phase grains can be made. In contrast to Lew99-Ky, however, more cores may be classified as polyphase (e.g. grains 38, 43) and some cores preserve distinct fine-scale oscillatory growth zoning (e.g. grain 71). Rims and single phase grains vary in internal structure from grains that are relatively homogeneous (e.g. grain 35) to grains that preserve coarse zoning (e.g. grain 33). The thin outer rim seen in zircon from sample Lew99-Ky is absent in grains from this sample.

Guided by internal growth zoning, 155 analyses were performed on 86 grains. Post-analytical imaging revealed that seven of these analyses straddled two growth zones, while a further three analyses had excessive levels of common Pb either because they were placed on a crack or hit an inclusion – these analyses are omitted from

Fig. 4. Concordia diagrams showing ion microprobe data from sample Lew99-Ky: (a) cores only; (b) rims and single phase grains. Colour coding follows that used in Figure 3. Error ellipses are plotted at 2σ.
consideration of the data set. Cores range widely in U concentration from c. 10–400 ppm with Th/U mostly in the range 0.3–1.2 with the exception of a few outliers with very low Th/U. Rims and single phase grains have a U concentration range from c. 10–200 ppm with a restricted Th/U ratio range of c. 0.9–1.1 (Fig. 6a). On a concordia diagram (Fig. 7), analyses are mostly concordant within analytical error and define a near-continuous spread between a maximum $^{207}\text{Pb}/^{206}\text{Pb}$ age of 3043 ± 28 Ma and a significant group of analyses (mostly rims) at c. 2500 Ma.

A total of 97 core analyses were performed (Fig. 7a) of which two were highly discordant (one reversely) and are not considered further. Based on the CL imaging, 25 of these are oscillatory zoned cores (designated ‘cz’, see Supplementary Material) and 27 are simple, mostly CL dark cores. The remainder are polyphase cores which are further divided into relatively the oldest group (designated ‘c1’, $n = 25$) and apparent younger overgrowths on this group (‘c2’, $n = 21$), with a

Fig. 5. Cumulative age plot of individual zircon analyses (2σ error bars) with superimposed relative probability plot (grey line) for sample Lew99-Ky. (a) cores; (b) rims (red) and single phase grains (green).

Fig. 6. Th–U chemistry of zircon from sample Lew99-GE plotted as (a) Th concentration (ppm) v. U concentration (ppm) contoured for Th/U ratio; (b) U concentration v. $^{207}\text{Pb}/^{206}\text{Pb}$ age (Ma); (c) Th/U ratio v. $^{207}\text{Pb}/^{206}\text{Pb}$ age (Ma). Growth phases recognized from CL are colour-coded blue for cores, red for rims, and green for single phase grains.
single analysis (‘c3’) in a grain with a possible three-phase core. Within these groups, the cores may or may not exhibit oscillatory zoning.

An interpretation in which the oldest core ages represent the magmatic protolith age would place this at $3043 \pm 28$ Ma based on the single oldest grain or, accepting that the reverse discordant analysis of grain 24 has an accurate $207\text{Pb}/206\text{Pb}$ age, $3037 \pm 18$ Ma based on an average of these two oldest analyses. This age accords with the interpretation of data from the likely equivalent sample GST10 by Kinny & Friend (1997) who, additionally, found an older grain in excess of 3.1 Ga which they interpreted as inherited.

An alternative interpretation of the core analyses is that the two oldest grains are themselves inherited into a younger magmatic rock. While indistinguishable in terms of U concentration from most of the

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**Fig. 7.** Concordia diagrams showing ion microprobe data from sample Lew99-GE: (a) cores only; (b) rims and single phase grains. Colour coding follows that used in Figure 6. Error ellipses are plotted at $2\sigma$. 

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**Constraints from Combined In Situ U–Pb and Lu–Hf**
core population (Fig. 6b), these two oldest grains exhibit relatively low Th/U ratios (c. 0.3, Fig. 6c). One of these analyses is from grain 71 which in contrast to the other cores in this sample shows well-defined, fine-scale oscillatory zoning in CL imaging. On a cumulative age plot (Fig. 8a), the two >3 Ga analyses are noticeably older than the next youngest grains that cluster at around 2950 Ma. Applying the technique of including successively younger grains into a weighted average pushes the MSWD value to 1.5 with only three grains and to 2.3 if the four oldest grains are grouped. By comparison, if these two oldest grains are set aside as possible inherited components, the next six oldest grains yield a weighted average $^{207}\text{Pb}/^{206}\text{Pb}$ age of 2956 ± 19 Ma with a statistically significant MSWD value of 0.86 (which increases to 2.7 with inclusion of the next youngest grain). It might therefore be concluded that the magmatic protolith age is 2956 Ma, an age similar to many of the oldest components observed in other samples from the central region granulites (Friend & Kinny 1995; Kinny & Friend 1997).

Further complexities in the internal structure of the cores suggest, however, that even the 2956 ± 19 Ma age might represent an inherited component. As mentioned previously, a number of the cores are classified as polyphase, in which an older central region (c1) is clearly corroded, embayed and has its zoning truncated by an outer region (c2). In some cases the c2 core regions display oscillatory growth zoning which would be consistent with a magmatic origin (e.g. grain 43, which has a $^{207}\text{Pb}/^{206}\text{Pb}$ age of 2851 ± 14 Ma). Comparing cumulative age probability plots for the c1 and c2 cores (Fig. 8a) shows that the latter do not preserve $^{207}\text{Pb}/^{206}\text{Pb}$ ages in excess of c. 2850 Ma, the three oldest c2 analyses combining to give a weighted average of 2850 ± 11 Ma (MSWD = 0.061). If the dataset is expanded to include single phase cores on the basis that some of these may have concealed older regions, nine analyses define a coherent group with a weighted average age of 2843 ± 9 Ma (MSWD = 0.63) although this age should be treated with additional caution given the possibility that some of the included ages may be from older grains that have lost Pb so that they fall coincidentally into this group.

The younger rims (n = 38) and single phase grains (n = 10) show strong similarities with those from Lew99-Ky. Concordant $^{207}\text{Pb}/^{206}\text{Pb}$ ages (Fig. 7b) mostly cluster around a peak in the age distribution at 2500 Ma (Fig. 8b). Omitting a few outliers with slightly younger ages, which may reflect post-2500 Ma disturbance, a coherent group of 28 concordant analyses yields a weighted average $^{207}\text{Pb}/^{206}\text{Pb}$ age of 2501 ± 6 Ma (MSWD = 1.4). A few ages in the rim and single-phase categories extend up to c. 2840 Ma and are interpreted as reflecting incomplete resetting during the c. 2500 Ma event.

**LA–ICP–MCMS Hf isotope results**

Recent advances in the measurement of the Hf isotopic composition of zircon in situ using laser ablation ICP–MS has opened the possibility to investigate the nature of the source region for a magmatic rock using its zircon. This is analogous to the use of whole-rock Sm–Nd and Lu–Hf
isotope systematics to derive model ages but with the distinct advantages that zircon has an extremely low Lu–Hf ratio, hence its in-growth of radiogenic $^{176}$Hf is much slower than a corresponding whole rock and, unlike whole-rocks, it is less susceptible to disturbance during metamorphic events since Hf is a major element (commonly c. 1–2%) in the zircon structure (Patchett et al. 1981). Increasingly, geochronological studies utilizing high spatial resolution ion microprobe analysis are being complemented by LA–ICP–MS Hf isotope analysis not just on the same zircon but in some cases on the same growth phase within a zircon that has been independently dated (e.g. Kemp et al. 2006). In order to obtain adequate precision in the Hf isotope measurement however, a considerably larger volume of material is sampled compared to ion microprobe and care is required to decipher time-resolved data signals to ensure that data do not represent mixed regions of a zircon. In this study, concurrent measurement of Pb and Hf isotopes for some of the analysed zircons provides an additional age constraint to that obtained from the ion microprobe.

**Lew99-Ky.** Twenty-one analyses (see Supplementary Material) were performed on zircon grains from sample Lew99-Ky, 13 of which were located in cores of polypehase grains and the remainder in single phase grains, one analysis was discarded on the grounds that the laser sampled across a boundary during the analysis. Concurrently determined $^{207}$Pb/$^{206}$Pb ages performed on 11 grains range from 2680 to 2794 Ma, a smaller range than SIMS $^{207}$Pb/$^{206}$Pb ages, which for these same grains ranges from 2520 to 2850 Ma. In some cases the LA–ICP ages exceed those obtained by SIMS and in other cases are younger which probably reflects the considerable difference in sampled volume, with SIMS being more sensitive to localized Pb-loss (e.g. along annealed cracks) due to its smaller sampling volume, while the LA–ICP analyses are more likely to average extreme variations. On a plot of age corrected $^{176}$Hf/$^{177}$Hf against the concurrently determined LA–ICP $^{207}$Pb/$^{206}$Pb age or SIMS $^{207}$Pb/$^{206}$Pb age when only Hf isotopes were determined (Fig. 9a), data from Lew99-Ky all lie within error of a sub-horizontal line suggesting that they had a similar initial Hf-isotopic composition that has remained unaffected by metamorphic disturbance(s) that results in the apparently too young U–Pb ages. This interpretation is supported by the time resolved Hf–Pb isotope signals from individual analyses (Fig. 10), which reveal that the $^{176}$Hf/$^{177}$Hf ratio remains unchanged during the run despite pronounced shifts in $^{207}$Pb/$^{206}$Pb age (this is the general case for zircons of Lew99-Ky). As a result of this, $\varepsilon_{Hf}$ values calculated using the measured $^{207}$Pb/$^{206}$Pb ages define a steep trend on

Figure 11a (green arrow) that is an artefact of age resetting (i.e. the use of ages that are increasingly too young in the $\varepsilon_{Hf}$ formulation), rather than representing the evolution of a plausible source reservoir. For the purpose of more accurately calculating initial $^{176}$Hf/$^{177}$Hf and $\varepsilon_{Hf}$ values, a ‘preferred’ age is used based on the analysis of the entire population dated by SIMS, which suggests an age of c. 2850 Ma for this sample. Using this approach, individual $\varepsilon_{Hf}(2850)$ values range from −0.93 to +1.67. The two lowest $\varepsilon_{Hf}(2850)$ values are from grains (41a and 71a) with relatively high Lu/Hf ratios which result in a large age correction. Omission of these two analyses yields a weighted average $^{176}$Hf/$^{177}$Hf(2850) of 0.280982 ± 0.000008 (2σ, MSWD = 2.1), corresponding to an $\varepsilon_{Hf}(2850)$ of 0.87 ± 0.28 (2σ, MSWD = 2.1, Fig. 9b). The slightly elevated MSWD value indicates a possible component of scatter in the data which cannot be explained by assigned analytical errors alone. This may reflect some Hf-isotope heterogeneity in the protolith magma and will be discussed in more detail below.

**Lew99-GE.** Consistent with the greater complexity of its zircon growth structures and U–Pb systematics, Hf isotope data from sample Lew99-GE are considerably more scattered than those from Lew99-Ky. Concurrently determined $^{207}$Pb/$^{206}$Pb ages range from 2541 to 2894 Ma, which is slightly smaller than the SIMS range of 2501 to 2977 Ma. As with sample Lew99-Ky, the unsystematic relative age difference between the two methods probably reflects the difference in sampled volume. Within-run Hf–Pb isotope data for individual zircons also show no shifts in $^{176}$Hf/$^{177}$Hf attending abrupt changes in age (Fig. 10), suggesting that U–Pb disturbance had negligible effects on the Hf isotopes. The plot of age corrected $^{176}$Hf/$^{177}$Hf against the concurrently determined LA–ICP $^{207}$Pb/$^{206}$Pb age or SIMS $^{207}$Pb/$^{206}$Pb age when only Hf isotopes were determined, (Fig. 9a), however, reveals considerably greater complexity in Hf-isotope systematics in Lew99-GE. The age-corrected $^{176}$Hf/$^{177}$Hf ratios are consistently lower than those of Lew99-Ky and while a sub-horizontal trend is defined by the most radiogenic samples over a wide age range, a distinct trend to less radiogenic Hf is present in the analyses yielding the oldest ages. For this sample, assignment of a preferred age in order to calculate initial $\varepsilon_{Hf}$ values is complicated by the possibility that more than one age of magmatic zircon may be represented in the Lew99-GE cores and the extensive c. 2500 Ma Pb-loss overprint means that for example, a 2850 Ma age might actually represent a partially disturbed 2960 Ma (or even 3030 Ma) zircon. One approach, following the reasoning for Lew99-Ky,
is to group those analyses falling along a broadly horizontal line on Figure 8a as a single population treating less radiogenic analyses as older inherited components, since these plot along a separate Pb-loss trajectory on Figure 9a. Considering only grains for which a minimum age of 2850 Ma can be assigned \( n = 9 \), see Supplementary Material) and omitting other analyses where an age of \( 2850 \) Ma is indicated, a weighted average \( ^{176}\text{Hf}/^{177}\text{Hf}(2850) \) of \( 0.280957 \pm 0.000013 \) (2\( \sigma \), MSWD = 2.7) is obtained by further rejecting the least radiogenic outlier, corresponding to an \( ^{176}\text{Hf}(2850) \) of \( 0.22 \pm 0.45 \) (2\( \sigma \), MSWD = 2.7, Fig. 8c). The higher MSWD relative to that for Lew99-Ky could reflect a greater degree of Hf-isotope heterogeneity in the protolith magma from which the zircons precipitated as well as likely inclusion of grains of different primary crystallization age in the sub-population (i.e. older grains with different \( ^{176}\text{Hf}/^{177}\text{Hf} \) that have lost Pb).

**Fig. 9.** Zircon Hf isotope data plotted as (a) initial \( ^{176}\text{Hf}/^{177}\text{Hf} \) ratio against age, using \(^{207}\text{Pb}/^{206}\text{Pb} \) ages derived concurrently with Hf isotope data (circles) or separately by SIMS (squares). Horizontal arrow indicates schematic Pb-loss/age-resetting vector for zircon. Panels (b) and (c) plot initial \( ^{176}\text{Hf}/^{177}\text{Hf} \) ratios (calculated at 2850 Ma) as a histogram and superimposed relatively probability curve for Lew99-Ky and Lew99-GE respectively. Vertical scale bars show the evolution of \( ^{176}\text{Hf}/^{177}\text{Hf} \) over a 100 Ma period with a depleted mantle Lu/Hf ratio (black) and typical mafic crust ratio, Lu/Hf = 0.021 (grey).
Fig. 10. Time-resolved variations in $^{176}\text{Hf}/^{177}\text{Hf}$ ratios and $^{207}\text{Pb}/^{206}\text{Pb}$ ages obtained from zircon grains of sample 99-Lew-GE (a) and 99-Lew-Ky (b). Analysis time, and thus depth of penetration of the laser into the zircon, increases from left to right. Although both analyses are sited in CL-defined single-phase zircon, marked age complexities beneath the polished surface are evident, with apparent ages either increasing (GE) or decreasing (Ky) during the run; Hf isotope ratios are unchanged in both cases.

Discussion

Protolith ages

Geochronological data presented from two granulite facies tonalitic gneisses show the extreme difficulty in assigning a magmatic protolith age when the zircons have experienced incomplete, but in many cases extensive, Pb-loss during at least one later event. In part, this is a function of the SIMS method utilized in which relatively large (typically c. 1–2%) errors on U–Pb ratios preclude recognition of true concordance with a precision
Fig. 11. Hf-isotope evolution diagrams for Lewisian granulite data. Data plotted in (a) assume a minimum age of the analysed growth zone based on whichever is the highest of the ages from the two applied methods (squares, SIMS; circles, LA–ICPMS; symbol colours as in Fig. 9). Three depleted (relative to CHUR) mantle reservoir evolution lines
necessary to decipher an original magmatic age lying on concordia from one which has suffered later Pb-loss within just a few hundred Ma. For example, 2.95 Ga zircon experiencing variable amounts of Pb-loss at 2.5 Ga will deviate from concordia by a maximum of 1.7% in their $^{206}\text{Pb}/^{238}\text{U}$ ratio. This deviation will be even smaller close to the starting age, such that distinguishing, for example, a 2.85 Ga magmatic component from a 2.95 Ga inherited component is effectively impossible on the basis of concordance. In effect, the problem is reduced to a rather unsatisfactory one-dimensional interpretation of $^{207}\text{Pb}/^{206}\text{Pb}$ age.

Despite this apparently gloomy prospect, a number of additional constraints mean that it is not necessary to rely entirely on the SIMS U–Pb data as the basis for interpretation. First, detailed CL imaging indicates that in sample Lew99-Ky cores are relatively simple compared to those in sample Lew99-GE. Thus, inferring a single magmatic protolith age is more reasonable for Lew99-Ky than for Lew99-GE. The latter sample may well contain inherited zircon rendering interpretation of the older $^{207}\text{Pb}/^{206}\text{Pb}$ ages as magmatic protolith insecure. Second, by analysing a large number of grains, peaks in the age distribution are more obvious and statistically more significant. Thirdly, application of in situ Hf isotope analysis of zircon reveals complicated isotope systematics in Lew99-GE that are best explained in terms of the zircon population representing more than one magmatic event, the older ages, present as an inherited component, having lower $^{176}\text{Hf}/^{177}\text{Hf}$ ratios.

**Source materials of the protolith magma and crustal residence history of the Lewisian**

A significant problem in inferring crustal residence ages from TTG gneisses from either bulk-rock Sm–Nd or zircon Lu–Hf isotope data is that the crystallization age of the rock and/or zircon does not record the crust-mantle differentiation event itself but merely the later extraction of TTG melts from an intermediate, probably mafic, precursor (Rollinson & Windley 1980; Smithies et al. 2009). Back-extrapolation from initial ratios calculated at the age of TTG crystallization must therefore be made using assumed Sm/Nd and Lu/Hf ratios for this source that are difficult to constrain. Additionally, the Hf-isotope evolution curve of the ultimate mantle source is poorly known, introducing a further uncertainty in the calculated crustal residence age.

Using bulk-rock Sm–Nd isochrons obtained from a suite of mafic/ultramafic intrusions, Whitehouse (1989) argued that their depleted mantle source had an $\epsilon_{\text{Nd}}$ value of c. +2.5 at 2.95 Ga, similar to that predicted by the DePaolo et al. (1981) model. This model was then applied to various TTG gneiss suites from three regions in the Lewisian complex in order to obtain ‘average’ $t_{\text{DM}}$ model ages, calculated using the average Sm/Nd ratio from each suite of samples but acknowledging that such estimates would inevitably be approximations due to the likelihood of an interim crustal precursor to the TTG rocks. For the amphibolite facies northern region gneisses, the average $t_{\text{DM}}$ of c. 2.8 Ga was similar to magmatic protolith ages subsequently inferred from ion microprobe U–Pb zircon data (Kinny & Friend 1997). Applying this method to the central region gneisses is, however, complicated by disturbance of the whole-rock Sm–Nd system during granulite facies metamorphism (Whitehouse 1988), although the average $t_{\text{DM}}$ of 2.93 Ga, calculated assuming homogenization of isotopic composition with no significant fractionation of Sm/Nd ratio, is similar to protolith ages inferred from zircon U–Pb data (Friend & Kinny 1995; Kinny & Friend 1997).

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**Fig. 11.** (Continued) are indicated on the plot: DM (MORB source depleted mantle, Griffin et al. 2000); Neo-Mesoarchaean mantle (Shirey et al. 2008) and model Slave craton mantle (Pietranik et al. 2008). The green arrow labelled ‘zircon/Pb-loss’) represents evolution with very low Lu/Hf typical of that found in zircon and approximates a trend that would be defined by Pb-loss in which the age would become younger with no change in Hf-isotope ratio. Inset plot (b) schematically illustrates how a range of crustal extraction ages may be derived from a single zircon U–Pb and Hf-isotope analysis (see text for discussion). For the investigated samples, the range of Hf-isotope evolution possibilities is evaluated in panel (c). In this plot, data from both samples are shown at their preferred 2.85 Ga age based on evaluation of overall data set as discussed in text (note that data have been plotted 10 Ma further apart so that data symbols do not overlap). For Lew99-GE, the broad green polygon extending from the array of data at the preferred magmatic crystallization age of c. 2.86 Ga represents the evolution of Hf-isotopes in zircon back to c. 2.95 Ga, the maximum plausible magmatic crystallization age. Dashed lines show the evolution trajectory of individual zircon analyses where these provide a limiting constraint on model ages: green lines represent the low Lu/Hf zircon/Pb-loss trajectory while purple lines represent typical mafic crust with $^{176}\text{Hf}/^{177}\text{Hf}$ ratio of 0.021. The orange arrow labelled ‘GE source?’ and identically coloured dashed lines is a Lu/Hf trajectory derived by regression of the Lew99-GE points shown with solid blue fill (Lu/Hf = 0.018). Vertical dashed lines show the average Sm–Nd depleted mantle model age inferred from central region Lewisian gneisses (Whitehouse 1989) at c. 2.93 Ga and the oldest ages obtained from TTG gneisses and/or zircons in the Lewisian at 3.13 Ga (Kinny & Friend 2005). Typical reproducibility of ± 0.85 e units (2σ) is shown by error bar in bottom right, based on analysis of five different standards (Kemp et al. 2008). $\epsilon$ values were calculated relative to the chondritic parameters of Blichert-Toft & Albarède (1997).
Zircon Lu–Hf data are used here to provide an alternative and potentially more robust constraint on crustal residence age, the Lewesian example highlighting both the benefits as well as the difficulties inherent in using this approach. The combined Pb–Hf isotope method has particular utility for the polyphase zircons of this study. Time-resolved isotope data (Fig. 9), suggest that modification of CL-defined zoning and resetting of U–Pb isotopes by younger thermal events was not accompanied by a significant incorporation of externally-derived Hf into the zircon (cf. Kemp et al. 2009), either from the rock matrix or via metamorphic fluids. The discussion below therefore assumes that the Hf-isotope systematics of the zircons largely reflect that of the magma from which these grains crystallized, rather than being due to secondary processes.

Because zircon has a very low Lu/Hf ratio, and thus low ingrowth of radiogenic Hf, its trajectory on a Hf isotope evolution diagram is generally quite predictable, the steep negative trend highlighted in Figure 11a (which corresponds to the essentially horizontal trend on Fig. 8a) reflecting the evolution of the chondritic uniform reservoir (CHUR) away from the zircon composition. Compared to zircon, all bulk rock reservoirs will have shallower (i.e. higher Lu/Hf) trends on the Hf-isotope evolution diagram. In cases where the zircon has experienced simple Pb-loss, this process is not commonly accompanied by any change in Lu/Hf ratio, thus enabling a relatively unambiguous estimate to be made of \( \varepsilon_{\text{Hf}}(t) \) at a measured and/or assumed time of crystallization (Figs 9a & 11a).

Derivation of crustal residence age from initial \( \varepsilon_{\text{Hf}}(t) \) in zircon remains subject to uncertainties in both the Lu/Hf characteristics of the crustal precursor to the TTG gneiss and its ultimate mantle source. Figure 11b illustrates schematically how a range of possible model ages may be derived from a single zircon analysed for its U–Pb and Lu–Hf systematics. The open circle representing measured age and Hf-isotope data yields two extreme crustal residence ages via extrapolation of Hf-isotope evolution trajectories from the measured zircon composition to their intersection with the assumed mantle evolution curve. The oldest crustal residence age \( t_{\text{DM(max)}} \) assumes that the measured zircon age represents TTG crystallization from a crustal precursor with a Lu/Hf ratio higher than zircon but lower than a typical mantle source. Younger crustal residence ages \( t_{\text{DM(min)}} \) result if the measured zircon age reflects Pb-loss from an older zircon which has a true crystallization age lying along the zircon/Pb-loss trend. For the limiting case of a very short residence time in the crustal precursor, the zircon crystallization age and the apparent crustal residence age \( t_{\text{DM(min)}} \) will be the same. An intermediate crustal residence age will be derived by evolution along a combination of the zircon/Pb-loss and TTG-source evolution lines if the zircon crystallization age (represented by the filled circle in Fig. 10b) is older than the measured age but younger than \( t_{\text{DM(min)}} \).

With regard to mantle evolution models, a major uncertainty in calculation of model ages arises from the assignment of isotope parameters for the depleted mantle reservoir. For Hf isotopes, a commonly used depleted mantle trend is derived by simply back-calculating the mean isotope composition of present day MORB \( (176\text{Hf}/177\text{Hf} = 0.28325) \) and using \( 176\text{Lu}/177\text{Hf} \) of 0.0384 (from Griffin et al. 2000). However, there is growing evidence from bulk rocks and zircon studies that the assumption of quasi-linear evolution from 4.5 Ga to the present day is unjustified and that the Lu–Hf system in the ancient terrestrial mantle may not have been as depleted as such an evolutionary trend may imply (e.g. Vervoort & Blichert-Toft 1999; Griffin et al. 2000; see also discussion in Shirey et al. 2008). Alternative Hf isotope depleted mantle evolution curves have been proposed recently by Shirey et al. (2008) for Neo/Mesoarchaean mantle in general and Pietranik et al. (2008) for the specific case of the Slave Province. Both of these models lie several epsilon units below the ‘model’ depleted mantle over the age range shown in Figure 11.

The possible range of crustal residence ages for the Lewesian granulites is investigated in Figure 11c with specific regard to constraints imposed by the data obtained from sample Lew99-GE. Regardless of which mantle Hf-evolution model is chosen, the minimum crustal extraction age is obtained from the most radiogenic zircon at 2850 Ma (analysis 1) by assuming that its zircon age represents Pb-loss from an older grain and that there is a short crustal precursor. Projecting back along a zircon/Pb-loss Lu/Hf trajectory yields a MORB depleted mantle model age of c. 3.06 Ga. If the less depleted Neo/Mesoarchaean mantle model (Shirey et al. 2008) applies, this minimum model age is c. 2.93 Ga, while a mantle resembling that proposed for the Slave Province (Pietranik et al. 2008) yields a minimum model age of c. 2.98 Ga (Fig. 12). The maximum crustal extraction age is obtained from the two least radiogenic >2850 Ma zircon grains by assuming that their measured U–Pb age represents their true crystallization age (i.e. unaffected by later Pb-loss) from a mafic crustal precursor with Lu/Hf = 0.021 (based on volcanic rocks with between 45 and 52% SiO2 from the GEOROC database, http://georoc.mpch-mainz.gwdg.de). In this case, crustal extraction from a MORB depleted mantle at c. 3.52 Ga is implied, while for Neo/Mesoarchaean and Slave mantle models, this event is constrained to c. 3.20 Ga and c. 3.34 Ga respectively (Figs 11c & 12). For each potential crustal
precursor and mantle source, Figure 12 summarizes the complete range of model ages obtained from the entire Lew99-GE data set. Using this approach, it could be inferred that the total range of crustal extraction ages for the Lew99-GE precursor extends over nearly 600 Ma starting at 3.52 Ga, a range that is even greater when analytical uncertainties on the Hf-isotope analyses are propagated onto model ages (c. ±30 Ma for the zircon/Pb-loss trajectory, c. ±70 Ma for the mafic crust trajectory).

An alternative and probably more realistic approach considering that these are meta-igneous rocks, is to attempt to include as many analyses as possible into a model with a single crustal precursor. Treating the Lew99-GE data set in this way, the five most radiogenic of the c. 2.85 Ga zircons may be combined with the three most radiogenic c. 2.85 Ga zircons to yield a regression line corresponding to a Lu/Hf ratio of 0.018 ± 0.004 (MSWD = 1.2, Fig. 11c). The minimum crustal extraction ages obtained from this regression line range from c. 2.96 Ga for Neo/Mesoarchaean mantle model to c. 3.16 Ga for MORB depleted mantle, with the Slave Province type mantle intermediate at c. 3.06 Ga. It is implicit in this model that >2.85 Ga analyses lying on the regression represent magmatic sampling of this same source reservoir at the specific times indicated by their U–Pb age. It is also noted that a similar (or the same) crustal precursor is also appropriate for the most radiogenic zircons in the Lew99-Ky dataset.

Less radiogenic analyses not included in the regression line can be derived in a number of different ways. If their measured U–Pb ages reflect Pb-loss instead of actual crystallization, then they project back along a steep zircon/Pb-loss trajectory and can be derived from the same crustal precursor inferred from the regression line. For the least radiogenic >2.85 Ga analyses however, the intersection between the zircon/Pb-loss trajectory and the regression-derived crustal precursor occurs at a very elevated εHf(t). In the most extreme case represented by analysis 71a (Fig. 11c), this is +8.5 at 3.34 Ga, a value which is not only incompatible with the less depleted mantle models but even exceeds contemporaneous MORB depleted mantle by over 3σ units. A more likely scenario is that some of the inherited grains were derived from magmas melted from a crustal precursor that separated from the mantle prior to the regression-constrained source. This would imply mixed-aged sources for the TTG magmas and a slightly more complex crustal growth history for the Lewisian. A variety of Hf-isotope evolution pathways can be constructed for individual zircon analyses which would still permit derivation from any of the plausible mantle models, although the present data do not provide additional constraints on such models. There are, however, potential constraints available from some of the oldest dated zircons and/or rocks in the Lewisian complex as a whole. The oldest zircon age recorded from the central region is a c. 3.12 Ga inherited grain in Lew99-GE equivalent sample GST-10 (Kinny & Friend 1997). Magmatic protolith ages of c. 3.13 Ga have been inferred for TTG gneisses from North Harris, Outer Hebrides (Friend & Kinny 2001) and the Loch Torridon area (Love 2004; Kinny et al. 2005) on the basis of U–Pb zircon data. If these represent the earliest magmatic rocks in the Lewisian complex, their relatively young age would lend...
support to derivation of crustal precursors from the less depleted mantle reservoirs indicated in Figures 11c and 12.

2700 Ma metamorphism?

Friend & Kinny (1995) noted that within analytical error, their SIMS data from central region granulite facies gneisses lie on a discordia line from a ≥2.96 Ga protolith to c. 2.5 Ga metamorphism and they were thus unable to recognize a 2.7–2.8 Ga metamorphic event. Their preferred interpretation envisaged a single Pb-loss event at 2.5 Ga affecting zircon which had crystallized from their igneous parent magma at ≥2.96 Ga. At its furthest deviation from the concordia curve, which occurs at 2.73 Ga, a 2.96–2.5 Ga discordia line will have a $^{238}\text{U}/^{206}\text{Pb}$ ratio that is c. 1.66% higher than that of concordia, while a 3.03 Ga starting point will deviate by up to 2.26% (at 2.78 Ga). The average 2σ error on the $^{238}\text{U}/^{206}\text{Pb}$ ratio of 5.2 ± 1.2% in the studies of Friend & Kinny (1995) and Kinny & Friend (1997) thus clearly precludes resolution between analyses lying on a simple two-stage discordia and those affected by an intermediate event which would therefore lie closer to the concordia line; for a single intermediate event data would lie in a discordia triangle defined by the three ages (Fig. 13). In the present study, however, considerably smaller uncertainties attach to $^{238}\text{U}/^{206}\text{Pb}$ ratio, averaging 1.8 ± 0.3% (2σ) for Lew99-Ge and 1.2 ± 0.3% (2σ) for Lew99-Ky. It is therefore possible, at least for Lew99-GE with the older grains and hence larger deviation of a single stage Pb-loss line from concordia, to investigate whether the distribution of analyses relative to concordia approximates a simple single stage

Fig. 13. Inverse concordia diagram showing the range of possible zircon U–Pb compositions arising from modelling of single- and two-stage evolution between hypothetical events at $t_1$ (magmatic crystallization), $t_2$ and $t_3$ (both metamorphism). If there is no event at $t_2$, all data will lie along the chord joining $t_1$ and $t_3$. The simulations involving an event at $t_2$ have been generated in two ways; in the first model (red points labelled ‘random’) there is a random relationship between the degree of Pb-loss occurring in each of the events at $t_2$ and $t_3$; in the second model (blue points labelled ‘equal’), the amount of Pb-loss in each of these events is set to be the same, a scenario that might arise for grains with similar U-content which will have accumulated a similar degree of radiation damage over similar length intervals $t_1–t_2$ and $t_2–t_3$.

Fig. 14. Combined probability density plot and histogram of the deviation of measured $^{238}\text{U}/^{206}\text{Pb}$ ratios for zircon from Lew99-GE relative to (a) the concordia curve; (b) a discordia line defined by a single-stage Pb-loss event at 2.5 Ga affecting 2.95 Ga zircon; and (c) a discordia line defined by a single stage Pb-loss event at 2.5 Ga affecting 3.03 Ga zircon.
Pb-loss event or plots more closely to concordia consistent with an intermediate age event having occurred.

Figure 14 shows probability density plots of the deviation (in %) of the $^{238}\text{U}/^{206}\text{Pb}$ ratio from cores in Lew99-GE relative to concordia, and two single
stage 2.5 Ga Pb-loss discordia lines starting at 2.95 Ga and 3.03 Ga. In the two models based on discordia lines, the peak of the distribution is clearly shifted towards the negative side, implying that the data plot between concordia and the discordia line as would be expected for a two-stage Pb-loss involving an event occurring between the assumed protolith age and the metamorphism at 2.5 Ga. Thus, while the new data are unable to resolve the timing of this event, their distribution suggests at least that such an event (or multiple events) is likely to have occurred.

Conclusion

Two granulite facies gneisses from the central region of the Lewisian complex contain complex polyphase zircon yielding ion microprobe U–Pb ages that spread along the concordia curve to a common metamorphic event at c. 2.5 Ga. This event has resulted in profound and variable Pb-loss severely complicating identification of magmatic protolith age and earlier metamorphic events.

Cathodoluminescence guided ion microprobe analyses of zircon from the Kylestrome sample suggests that a single magmatic protolith age of c. 2.85 Ga is represented. In accord with the relatively simple geochronology, in situ Hf-isotope analysis of zircons from this sample yields a small range of $\varepsilon_{\text{Hf}}$ (2850) values averaging $0.87 \pm 0.28$, omitting two unradiogenic outliers that may result from inheritance or a heterogeneous protolith magma. The second sample from Geodh Eanruig contains zircon with more complex cores which yield ages up to 3.03 Ga with distinct groupings at both c. 2.96 Ga and c. 2.85 Ga. An unambiguous magmatic protolith age cannot be assigned to this sample on the basis of the age data alone although c. 2.85 Ga is preferred on the basis of internal zircon structure. In situ Hf-isotope data also reveal a greater scatter in initial $\varepsilon_{\text{Hf}}$ values, which is best explained if the older ages represent inherited zircon that have suffered varying degrees of Pb-loss. Setting these analyses aside, a similar mafic source to that indicated by the Kylestrome sample is indicated, whether the main protolith age is 2.85 Ga or 2.95 Ga.

The time of separation of the crustal source region for the magmatic precursors to these gneisses from the mantle is strongly dependent on the chosen Hf-isotope mantle evolution model as well as the Lu/Hf ratio of the crustal source region itself.

Regression of the most radiogenic zircon analyses (both protolith magmatic and inherited) from sample Lew99-GE suggests a common crustal source region with a Lu/Hf ratio of 0.018, somewhat more enriched than typical mafic crust. However, the least radiogenic zircons of the inherited population cannot be derived from this crustal precursor and require additional, earlier crustal extraction event(s). Accepting recently proposed models for late-Archaean mantle that are several k units lower than contemporaneous MORB depleted mantle as applicable, a range of crustal extraction ages from c. 3.05 to 3.2 Ga may be inferred for the central region of the Lewisian. This age range is similar to the age of the oldest zircon ages reported from the Lewisian.

The spread of data along concordia do not reveal any specific metamorphic events prior to 2.5 Ga. However, data plots closer to concordia than to model single-stage Pb-loss lines from $\geq 2.95$ Ga suggesting that an earlier metamorphism, possibly at c. 2.7 Ga, has occurred in the northern part of the central region and is recorded in these data.

In summary, the data presented here highlight the difficulties with unambiguously establishing magmatic protolith crystallization and crustal residence ages from zircons of Precambrian gneisses with complex metamorphic histories. However, these difficulties can be minimized by the coupled application of CL-guided, high resolution SIMS U–Pb dating and concurrent laser ablation Pb–Hf isotope analysis, which enable a more confident linkage between age and isotope tracer information and provide some insight into the response of these isotope systems to younger thermal events. Such techniques can contribute towards a more elaborate reconstruction of the crustal evolutionary history and prehistory of ancient gneiss terranes, such as represented by the Lewisian complex.

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