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Establishment of a 3.83-Ga magmatic age for the Akilia tonalite (southern West Greenland)

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This paper is dedicated to the late Vic McGregor who first recognized, described, and mapped the rocks of the 'Akilia association' in southern West Greenland.

Abstract

A recurring challenge to ion microprobe U–Pb zircon geochronology has been to discriminate between preservation of original igneous zircon populations and inherited grains. This has proved particularly problematic in studying the polyphase metamorphic rocks that dominate Early Archean gneissic terranes. For example, differing interpretations exist for the origin of complex zircon populations investigated by U–Pb ion microprobe zircon geochronology from a granitoid (orthogneiss) body, previously used to establish a minimum age for a supracrustal enclave on Akilia (island) in southern West Greenland. We describe a method whereby the geochemistry of the Akilia orthogneiss, coupled with a U–Th–Pb vs. age depth profile in a zircon from the same rock, permits direct assessment of zircon inheritance. Results reveal evidence for three phases of concentric zircon growth at 3.83 Ga, ~3.6 Ga and 2.7–2.5 Ga; zircon growth at both ~3.6 Ga and 2.7–2.5 Ga is consistent with precipitation from a metamorphic fluid. Depth profile U–Th–Pb data demonstrate that only the >3.8-Ga zircon core could have crystallized from the host rock. We conclude that the magmatic age of this rock is 3.83 ± 0.01 Ga, not ~3.65 Ga as has been previously proposed. The U–Th–Pb zircon depth profile technique has wider applications to resolving other geochronological debates in high-grade metamorphic terranes where zircons populations are diverse and individual grains record complex overgrowth and dissolution/re-precipitation features.

Keywords: Archean; West Greenland; zircon; geochronology; life origin; ion probe data

1. Introduction

Early Archean supracrustal rocks, preserved within highly metamorphosed granitoid gneiss complexes, provide the only direct record of the state of the continental crust, hydrosphere and biosphere prior to ~3.5 Ga. These ancient volcano-sedimentary assemblages form the baseline for understanding early planetary processes, including the timing of the emergence of life on Earth and

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its potential to have appeared elsewhere in the solar system. In these terranes, sediments typically appear as highly metamorphosed and strongly deformed supracrustal enclaves, ranging in size from less than a meter wide to tens of kilometers in length. All are locked within voluminous granitoid gneisses [1]. The oldest recognized supracrustal enclave [2,3] is a ~5-m-thick layer of quartz–magnetite 'ironstone' interposed between 10–40-m-thick layers of amphibolite and ultramafic rocks on the southwestern point of Akilia (island) [4], Ameralik fjord region, West Greenland (Fig. 1). Tonalitic to granodioritic orthogneisses on Akilia found in contact with the northern and eastern margins of the supracrustal enclave yield complex U–Pb zircon ages as old as 3.87 Ga [3,5–7]. These results have been interpreted [3,5] as the age of the igneous protoliths to the orthogneisses and provide a minimum age of the associated supracrustal rocks. Carbon isotope data [2] have been interpreted to indicate bioorganic activity during deposition of the metasedimentary rocks on Akilia (cf. [35]). If these interpretations are correct, it would mean that life emerged on Earth significantly earlier than previously established [2,3,8–10]. Other carbon isotope evidence from the Isua supracrustal belt 150 km northeast of Akilia, places the emergence of life at least before 3.77 Ga [8,11], thereby raising the likelihood for an early and rapid emergence of the biosphere on Earth.

However, several authors [12,13] have argued based on whole-rock and mineral common Pb data that the geologically diverse 'Amîtsoq' orthogneisses of West Greenland, including those that host Akilia sedimentary enclaves, are the product of a single ~3.6-Ga crustal formational event. This interpretation is in conflict with the view that the at present large (3000 km^2) and geologically heterogeneous 'Itsaq Gneiss Complex' [5] formed in a protracted fashion from ~3.9 to 3.6 Ga. If the age of generically termed 'Amîtsoq' granitoid crystallization is actually ca. 3.6 Ga, this would suggest that all >3.8-Ga zircons contained in various orthogneisses of West Greenland thus far analyzed, including those on Akilia and nearby islands, are inherited.

Resolving the question of inheritance vs. protolith igneous origin of complex zircons, particularly with regard to the age of the orthogneisses on Akilia, has implications for the age of rocks hosting the oldest known sediments and reported evidence for life contained therein [2], as well as for the timing and nature of events at the surface during the emergence of the biosphere. Confirmation of a >3.8-Ga age for the Akilia orthogneisses and the associated metasediments and their biological signatures is significant because it would place their deposition prior to termination of an extended (and ill-defined) period of meteoritic and cometary bombardment of the inner solar system centered at ca. 3.85 Ga [14,15] and coeval with the presence of liquid water on early Mars [16,17].

2. Zircon growth events in the Archean (~3.8 Ga, 3.6 Ga and 2.7 Ga)

There is general agreement that the gneisses on Akilia contain at least three zircon age populations: ca. 3.85 Ga, ~3.6 Ga, and ~2.7 Ga [3,5–7]. A recently proposed explanation [6,7,13] for this age variation is that the ~2.7-Ga zircons are metamorphic overgrowths, the ~3.6-Ga grains are igneous and all of the oldest grains are inherited [12].

That the tonalitic protolith of the Akilia orthogneiss in question can only be ~3.6 Ga [6,7,12,13] is problematic in our view for the following reasons.

(1) The strongly zircon *undersaturated* [18] nature of the original magma (Table 1; Fig. 2) intruding at ~950°C [19] into zircon-poor supracrustal rocks such as amphibolite and hornblendite on Akilia is unlikely to preserve widespread inherited zircon. Tonalitic orthogneiss sample GR9716 [16] studied here (equivalent to G93/05 of Nutman et al. [3,5] and SM/GR/97/7 of Whitehouse et al. [6,7]) has a Zr abundance of 144 ppm (Table 1) and the observed whole rock composition is probably close to that of the protolith magma [19]. Thus, at temperatures ~950°C and $M=(Na+K+2Ca/Si \times Al)$ [18] of 1.54, the solubility of zircon in the tonalite melt would have been close to 900 ppm (Fig. 2) far below the 144 ppm Zr present in GR9716.

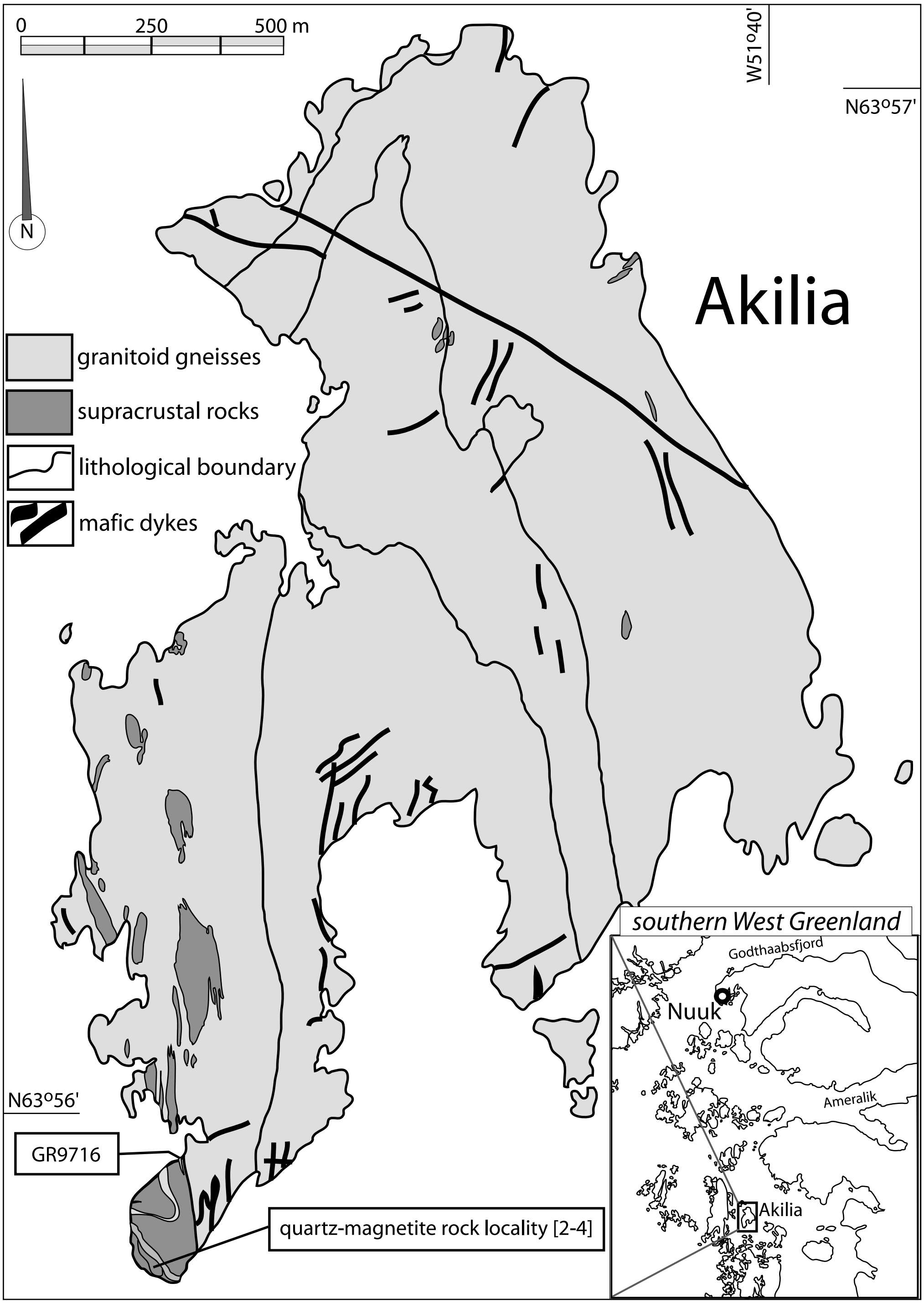

Fig. 1. Locality map showing Akilia (island) within the Early Archean 'Itsaq Gneiss Complex' in the vicinity of Ameralik (fjord) in southern West Greenland (inset). A complex association of supracrustal rocks and associated gneisses are found on Akilia (island). Adjacent islands and coastal areas contain similar relationships [3–5,22,26,27] that await further detailed study. Map modified after A.P. Nutman (unpublished manuscript).

(2) While Kamber and Moorbath [13] emphasized the lack of 3.8-Ga common Pb in feldspars from Akilia orthogneisses, metamorphism at ~3.6 Ga occurred under granulite grade conditions (>750°C and 7 kbar; [20]) that would readily cause exchange of primitive Pb isotopes in feldspar [21] with radiogenic Pb released from U-rich phases, dominantly zircon. Thus, their observation [13] is the expected result of a 3.6-Ga high-grade metamorphic event and does not preclude a 3.8-Ga protolith age.

It has been pointed out [22] that the diverse rocks arbitrarily assigned a common 'Amîtsoq gneiss' origin and analyzed together by Kamber and Moorbath [13] are in fact petrogenetically unrelated and therefore do not provide data relevant to the ages of the Akilia supracrustal enclaves or the orthogneisses that surround and in some cases, intrude them. Hence, the controversy we address here is based on zircon U–Pb isotope geochronology on an Akilia orthogneiss that was originally assigned an igneous protolith age of 3.85 Ga [3] but was subsequently reevaluated by Whitehouse et al. [6,7] who have argued for widespread zircon inheritance and a protolith age of ~3.65 Ga.

Table 1
Whole rock geochemistry of sample GR9716

Major elements	wt%
SiO_2	65.5
Al_2O_3	16.9
Fe_2O_3	3.91
MnO	0.06
MgO	2.26
CaO	4.65
Na_2O	3.75
K_2O	1.31
TiO_2	0.38
P_2O_5	0.29
LOI	0.54
Total	99.6
Trace elements	**ppm**
Zr	144
Th	1.8
U	0.5
Pb	8.0
$M = 1.54$[a]	

[a] [18].

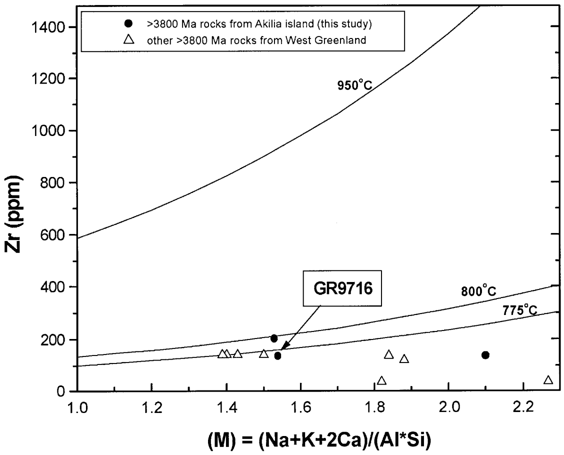

Fig. 2. Zircon saturation curves [18] plotted for a source temperature of 950°C and lower temperatures (800 °C, 775 °C) plotted near the freezing point of tonalitic melts [19]. We note that at the source temperature of 950°C all homogeneous orthogneisses identified from southern West Greenland that provide U–Pb zircon ages older than 3.8 Ga [4,5,16,22] are severely undersaturated in Zr and any zircon entrained in such melts would have a strong likelihood of dissolving. Hence, the chance for widespread zircon inheritance (cf. [6,7,12,13]) in any of these rocks is low.

In this paper, we undertake two new styles of analysis in order to evaluate the likelihood of zircon inheritance in the Akilia tonalitic orthogneisses, i.e. U–Th–Pb isotope depth profiling of individual zircons and in situ analysis of zircon in petrographic thin section.

3. Methods

We analyzed zircons from the same tonalitic orthogneiss locality collected from the northern margin of the southwestern peninsula of Akilia, West Greenland (Fig. 1), from which Nutman et al. [3], Whitehouse et al. [6,7] and Mojzsis and Harrison [16] (sample GR9716) obtained their results.

3.1. Conventional ion microprobe U–Pb geochronology

All U–Pb ion microprobe analyses of zircons, extracted by conventional heavy mineral separa-

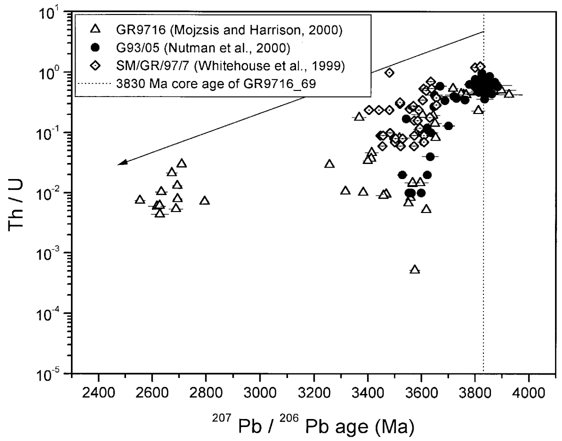

Fig. 3. Compilation of ^{207}Pb/^{206}Pb ion microprobe ages of zircons from orthogneiss sample GR9716 in contact with the supracrustal enclave on Akilia [3,5–7,22,27]. The zircon population at ~3.85 Ga, interpreted here to represent the magmatic age of the granitoid, has uniformly high Th/U values. Younger zircons generally exhibit lower Th/U values more characteristic of a metamorphic origin and correlate with well-documented metamorphic events in West Greenland at ~3.6 Ga and ~2.7 Ga [3–5,20,22].

tion of whole-rock powders or by in situ measurements in petrographic thin section, were determined using the UCLA CAMECA ims 1270 ion microprobe. Separated zircon grains were handpicked, mounted in epoxy and polished on 0.25 μm alumina; all samples were coated with ~100 Å of Au using a sputter-coater. The standard operating conditions for analysis presented here are a O_2^- primary beam focused to a ~20×25 μm spot; the ion microprobe was operated at a mass resolving power of 6000 with an energy window of 50 eV. An offset voltage of 5–15 eV was used for $^{238}U^+$ relative to Pb^+ and UO^+ to compensate for their contrasting energy distributions. Oxygen flooding to a pressure of 3.5×10^{-5} torr was employed to increase Pb^+ yields. U–Pb ages were determined by comparison with a working curve defined by multiple measurements of standard zircon AS-3 that yield concordant ^{206}Pb/^{238}U and ^{207}Pb/^{235}U ages of 1099.1±0.5 Ma [23].

All studies to date of the contested orthogneiss outcrop on Akilia using U–Pb zircon spot ages on separated zircons yield a similar age distribution and a general trend toward lower Th/U ratio with

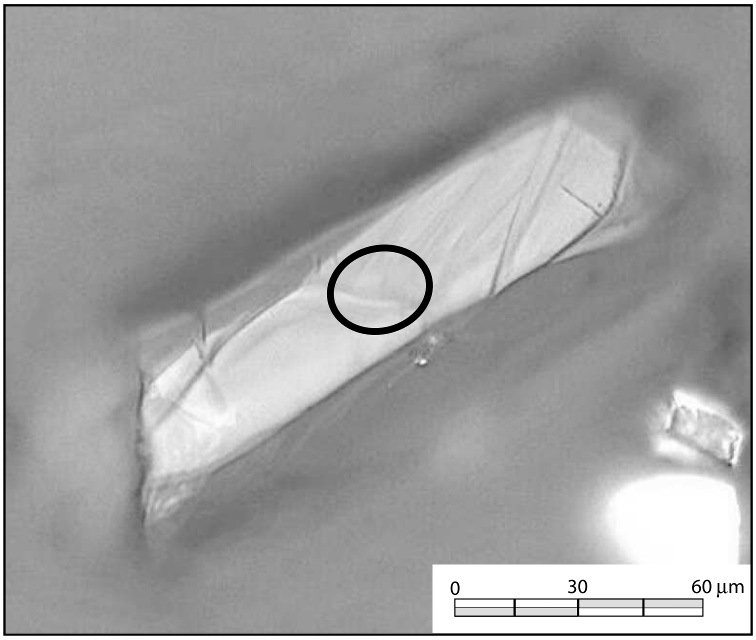

Fig. 4. Optical micrograph of unpolished euhedral zircon grain GR9716_69 used in U–Th–Pb depth profile analysis.

Table 2
Ion microprobe depth profile analyses of zircon GR9716_69

Depth (μm)	^{206}Pb*/^{238}U	^{207}Pb*/^{235}U	^{207}Pb*/^{206}Pb*	^{207}Pb*/^{206}Pb* age (Ma)	%^{206}Pb*	Th/U	^{208}Pb*/^{206}Pb*
0.00	0.455 ± 0.011	10.96 ± 0.35	0.1745 ± 0.0032	2602 ± 17	99.78	0.0134	0.0093
0.19	0.471 ± 0.011	11.46 ± 0.26	0.1764 ± 0.0020	2619 ± 10	99.73	0.0169	0.0068
0.37	0.479 ± 0.016	12.03 ± 0.37	0.1823 ± 0.0035	2674 ± 17	99.89	0.0151	0.0074
0.56	0.536 ± 0.012	12.33 ± 0.31	0.1669 ± 0.0022	2527 ± 12	99.65	0.004	0.0063
0.74	0.574 ± 0.025	13.62 ± 0.55	0.1721 ± 0.0022	2578 ± 12	99.93	0.0047	0.0029
0.93	0.582 ± 0.033	14.27 ± 0.79	0.1777 ± 0.0029	2632 ± 14	99.98	0.0042	0.0026
1.12	0.550 ± 0.011	13.34 ± 0.25	0.1759 ± 0.0016	2614 ± 8	99.95	0.0075	0.0045
1.30	0.546 ± 0.013	13.54 ± 0.31	0.1800 ± 0.0016	2653 ± 8	99.97	0.0136	0.0049
1.49	0.567 ± 0.015	14.10 ± 0.34	0.1806 ± 0.0013	2658 ± 7	100	0.0147	0.0055
1.67	0.551 ± 0.011	14.02 ± 0.29	0.1846 ± 0.0015	2695 ± 7	99.98	0.0212	0.0064
1.86	0.537 ± 0.018	13.79 ± 0.50	0.1861 ± 0.0024	2708 ± 11	99.98	0.0212	0.0064
2.05	0.522 ± 0.011	13.31 ± 0.30	0.1850 ± 0.0029	2698 ± 14	100	0.0174	0.007
2.23	0.439 ± 0.011	11.30 ± 0.27	0.1868 ± 0.0015	2714 ± 7	99.99	0.0226	0.0073
2.42	0.415 ± 0.015	10.54 ± 0.34	0.1843 ± 0.0013	2692 ± 6	100	0.0181	0.0061
2.60	0.358 ± 0.011	9.15 ± 0.24	0.1855 ± 0.0030	2703 ± 15	99.99	0.0113	0.0042
2.79	0.325 ± 0.009	8.33 ± 0.24	0.1859 ± 0.0031	2706 ± 15	99.99	0.0111	0.0035
2.98	0.310 ± 0.006	8.00 ± 0.16	0.1871 ± 0.0032	2717 ± 15	100	0.0084	0.0033
3.16	0.299 ± 0.005	7.69 ± 0.13	0.1864 ± 0.0018	2711 ± 9	100	0.0053	0.002
3.35	0.515 ± 0.028	12.01 ± 0.69	0.1690 ± 0.0028	2548 ± 15	99.81	0.0009	0.0054
3.53	0.538 ± 0.011	13.10 ± 0.29	0.1766 ± 0.0017	2621 ± 9	99.93	0.0014	0.0021
3.72	0.559 ± 0.017	13.73 ± 0.40	0.1782 ± 0.0023	2636 ± 11	99.97	0.0012	0.0015
3.91	0.717 ± 0.134	30.70 ± 5.57	0.3104 ± 0.0088	3523 ± 20	99.98	0.0149	0.0059
4.09	0.786 ± 0.094	33.83 ± 3.98	0.3120 ± 0.0039	3531 ± 9	99.99	0.0114	0.0048
4.28	0.705 ± 0.153	30.80 ± 6.49	0.3170 ± 0.0079	3555 ± 18	99.93	0.0165	0.0052
4.47	0.823 ± 0.069	37.66 ± 3.10	0.3320 ± 0.0045	3626 ± 9	100	0.0162	0.0037
4.65	0.782 ± 0.069	36.11 ± 3.19	0.3348 ± 0.0049	3639 ± 10	100	0.0155	0.0036
4.84	0.800 ± 0.120	36.20 ± 5.42	0.3281 ± 0.0061	3608 ± 12	99.95	0.0156	0.0037
5.02	0.638 ± 0.094	32.06 ± 5.07	0.3647 ± 0.0095	3769 ± 17	100	0.4487	0.1329
5.21	0.714 ± 0.229	36.24 ± 11.71	0.3680 ± 0.0151	3783 ± 27	99.86	0.4135	0.1392
5.40	0.600 ± 0.066	30.31 ± 3.33	0.3666 ± 0.0090	3777 ± 16	99.83	0.4521	0.1373
5.58	0.806 ± 0.105	35.79 ± 4.57	0.3218 ± 0.0039	3579 ± 8	100	0.0131	0.0039
5.77	0.850 ± 0.136	38.56 ± 6.30	0.3291 ± 0.0079	3613 ± 16	99.99	0.0103	0.0032
5.95	0.717 ± 0.082	31.45 ± 3.64	0.3182 ± 0.0045	3561 ± 9	99.97	0.0118	0.0035
6.14	0.782 ± 0.059	35.05 ± 2.64	0.3250 ± 0.0054	3593 ± 11	100	0.0081	0.0032
6.33	0.838 ± 0.059	37.29 ± 2.64	0.3227 ± 0.0046	3583 ± 9	100	0.0135	0.0029
6.51	0.840 ± 0.107	36.97 ± 4.99	0.3194 ± 0.0067	3567 ± 14	100	0.0102	0.0032
6.70	0.772 ± 0.082	34.97 ± 3.41	0.3284 ± 0.0066	3610 ± 13	99.96	0.0166	0.0062
6.88	0.848 ± 0.063	39.03 ± 2.86	0.3337 ± 0.0042	3634 ± 8	100	0.0431	0.0163
7.07	0.654 ± 0.074	29.81 ± 3.55	0.3304 ± 0.0064	3619 ± 13	100	0.0755	0.0223
7.26	0.607 ± 0.040	28.13 ± 1.92	0.3362 ± 0.0056	3646 ± 11	100	0.0784	0.0279
7.44	0.745 ± 0.095	34.89 ± 4.49	0.3395 ± 0.0060	3660 ± 12	99.89	0.1111	0.0417
7.63	0.730 ± 0.108	35.06 ± 5.49	0.3486 ± 0.0123	3701 ± 23	99.92	0.1421	0.0522
7.81	0.885 ± 0.177	43.50 ± 8.76	0.3563 ± 0.0071	3734 ± 13	99.92	0.1961	0.0687
8.00	0.717 ± 0.074	35.81 ± 3.70	0.3623 ± 0.0095	3760 ± 17	100	0.2606	0.095
8.19	0.846 ± 0.126	43.17 ± 6.48	0.3701 ± 0.0097	3792 ± 17	99.95	0.2754	0.1101
8.37	0.759 ± 0.099	38.17 ± 5.17	0.3647 ± 0.0072	3770 ± 13	100	0.3275	0.1164
8.56	0.818 ± 0.111	41.72 ± 5.38	0.3698 ± 0.0078	3790 ± 14	100	0.2991	0.1054
8.74	0.801 ± 0.116	40.85 ± 5.99	0.3697 ± 0.0133	3790 ± 23	99.8	0.4075	0.1182
8.93	0.626 ± 0.074	32.08 ± 3.95	0.3715 ± 0.0088	3798 ± 15	100	0.41	0.1313
9.12	0.778 ± 0.090	39.40 ± 4.26	0.3675 ± 0.0106	3781 ± 19	99.75	0.4266	0.1302
9.30	0.818 ± 0.182	41.04 ± 9.25	0.3637 ± 0.0142	3766 ± 25	100	0.3975	0.1277
9.49	0.744 ± 0.143	37.45 ± 7.14	0.3653 ± 0.0082	3772 ± 15	99.75	0.3961	0.1305
9.67	0.708 ± 0.168	36.74 ± 8.63	0.3762 ± 0.0126	3816 ± 20	99.87	0.4164	0.1335

Table 2 (*Continued*).

Depth (μm)	^{206}Pb*/^{238}U	^{207}Pb*/^{235}U	^{207}Pb*/^{206}Pb*	^{207}Pb*/^{206}Pb* age (Ma)	%^{206}Pb*	Th/U	^{208}Pb*/^{206}Pb*
9.86	0.638 ± 0.140	33.09 ± 7.24	0.3761 ± 0.0167	3816 ± 27	100	0.4427	0.1367
10.05	0.970 ± 0.140	50.12 ± 7.60	0.3747 ± 0.0134	3810 ± 22	99.87	0.5511	0.1709
10.23	0.713 ± 0.109	37.81 ± 5.71	0.3846 ± 0.0088	3850 ± 14	99.8	0.5682	0.1856
10.42	0.841 ± 0.140	43.25 ± 7.23	0.3728 ± 0.0136	3803 ± 22	99.74	0.5551	0.1655
10.60	1.621 ± 0.547	86.47 ± 28.76	0.3869 ± 0.0103	3859 ± 16	100	0.5435	0.1481
10.79	0.735 ± 0.232	38.71 ± 12.30	0.3818 ± 0.0092	3839 ± 14	100	0.5837	0.1609
10.98	0.678 ± 0.232	34.49 ± 12.11	0.3691 ± 0.0198	3788 ± 35	96.62	0.6046	0.1735
11.16	1.206 ± 0.909	61.05 ± 46.28	0.3671 ± 0.0248	3780 ± 44	100	0.7226	0.2056
11.35	1.021 ± 0.289	53.21 ± 14.86	0.3780 ± 0.0098	3824 ± 16	100	0.7054	0.1954
11.53	1.224 ± 0.446	66.35 ± 24.01	0.3932 ± 0.0157	3883 ± 24	100	0.6005	0.2037
11.72	0.915 ± 0.196	48.08 ± 10.46	0.3813 ± 0.0092	3837 ± 15	100	0.5421	0.1912
11.91	0.802 ± 0.256	41.93 ± 13.07	0.3793 ± 0.0174	3829 ± 28	100	0.6326	0.1898
12.09	1.038 ± 0.223	53.96 ± 11.82	0.3772 ± 0.0129	3820 ± 21	99.91	0.5583	0.1744
12.28	0.837 ± 0.167	43.61 ± 8.81	0.3780 ± 0.0130	3824 ± 21	99.8	0.6451	0.2037
12.47	1.282 ± 0.556	67.58 ± 29.39	0.3824 ± 0.0168	3841 ± 27	100	0.5519	0.1953
12.65	0.857 ± 0.367	43.66 ± 18.61	0.3696 ± 0.0179	3790 ± 32	99.91	0.6612	0.1814
12.84	1.193 ± 0.301	62.12 ± 15.95	0.3778 ± 0.0144	3823 ± 23	99.86	0.5895	0.1916
13.02	0.948 ± 0.312	49.59 ± 16.01	0.3793 ± 0.0124	3829 ± 20	99.72	0.6256	0.1973
13.21	0.792 ± 0.299	42.50 ± 15.99	0.3892 ± 0.0292	3868 ± 45	100	0.6506	0.2049
13.40	0.923 ± 0.216	49.51 ± 11.63	0.3889 ± 0.0160	3867 ± 25	100	0.6443	0.1781
13.58	1.074 ± 0.207	57.28 ± 10.77	0.3868 ± 0.0112	3858 ± 18	99.57	0.5988	0.1752
13.77	0.826 ± 0.158	44.25 ± 8.36	0.3886 ± 0.0136	3866 ± 21	100	0.67	0.2014
13.95	0.897 ± 0.216	47.01 ± 11.06	0.3802 ± 0.0167	3833 ± 27	100	0.6721	0.1853
14.14	1.069 ± 0.212	57.66 ± 11.47	0.3911 ± 0.0093	3875 ± 14	99.7	0.6594	0.1984
14.33	1.737 ± 0.700	91.77 ± 37.30	0.3832 ± 0.0158	3845 ± 25	99.53	0.6534	0.1881
14.51	1.097 ± 0.234	56.91 ± 12.26	0.3762 ± 0.0162	3817 ± 26	99.7	0.6198	0.2051
14.70	1.207 ± 0.286	64.15 ± 14.42	0.3854 ± 0.0146	3853 ± 23	100	0.5865	0.1822
14.88	0.745 ± 0.150	38.92 ± 7.84	0.3789 ± 0.0154	3827 ± 24	100	0.6133	0.1879
15.07	0.790 ± 0.203	42.19 ± 10.80	0.3875 ± 0.0132	3861 ± 21	99.76	0.7056	0.2059
15.26	0.936 ± 0.341	49.44 ± 18.23	0.3830 ± 0.0131	3844 ± 21	99.3	0.6575	0.1986
15.44	0.729 ± 0.168	37.86 ± 8.86	0.3765 ± 0.0124	3818 ± 20	100	0.731	0.1896
15.63	1.187 ± 0.356	62.37 ± 18.54	0.3811 ± 0.0096	3836 ± 15	100	0.6226	0.1766
15.81	0.808 ± 0.149	42.20 ± 8.05	0.3789 ± 0.0110	3827 ± 18	100	0.6641	0.2166
16.00	0.963 ± 0.247	51.69 ± 13.16	0.3893 ± 0.0164	3868 ± 25	99.89	0.6962	0.2184

%^{206}Pb* is the percent of radiogenic ^{206}Pb. Isotopic ratios have been corrected for common Pb. U–Pb ages were determined by comparison with a working curve defined by measurement of standard zircon AS-3 that yields concordant ^{206}Pb/^{238}U and ^{207}Pb/^{235}U ages of 1099.1 ± 0.5 Ma by conventional methods [23]. Error demagnification was applied to the data; all uncertainties are reported at the 1σ level.

younger ages (Fig. 3). However, in spot analysis mode, the 2-D spatial selectivity of the ion microprobe is limited by the size of the primary beam, typically 10–30 μm. In the case where a crystal contains symmetrical overgrowths, it is possible to use the ion microprobe in depth profiling mode on an initially unpolished surface to increase spatial resolution by up to two orders of magnitude [24]. This approach, applied to zircons and described below, obtains a continuous age profile as well as provides geochemical signatures that could bear on the conditions of zircon growth coinciding with documented metamorphic episodes. The technique potentially transcends ambiguity of interpretations of zircon inheritance vs. protolith magmatic ages by linking the geochemistry of the host rock with the minute scale of growth and composition of an individual zircon at a spatial resolution unattainable in normal spot analysis of polished grains.

3.2. Zircon U–Th–Pb depth profile analysis

After evaluation by light microscopy, a separated euhedral zircon grain (GR9716_69) that yielded a concordant U–Pb age of ∼3.85 Ga [16] was plucked from its ion microprobe mount and an original crystal prism surface placed face down onto adhesive tape together with grains of standard zircon AS-3. The enclosing 1″ diameter mold was then recast with epoxy. When cured, the mount was removed from its adhesive backing, cleaned in distilled water and Au coated according to our usual procedures [25] but without initial polishing (Fig. 4). The internal distributions of U–Pb and Th/U in zircon GR9716_69 were then obtained by a high-resolution depth profiling technique [24] utilizing a ∼5-nA primary O^- beam focused to a 30×20 μm spot. Pre-sputtering times were 120 s and analysis comprised 15 cycles of 10-s measurements. Each data point listed in Table 2 is composed of one block of analyses made up of three cycles. Analytical sessions in depth profiling mode involved a prolonged and continuous collection time (∼30 min) from the same spot as the ion beam sputters into the unpolished zircon sample. The extended nature of this depth profile required us to develop an approach different from that of conventional ion microprobe zircon geochronology where spots are analyzed on minerals mounted and polished to their centers. After sputtering a ∼5 μm deep crater with the O^- ion beam, we measured the pit depth using a DekTak© surface profilometer and then repolished by hand the sample surface using a 0.25 μm diamond-impregnated lapping film under repeated supervision using reflected-light microscopy of the grain until the crater was ∼1 μm deep. By measuring the depth of the residual pit from the polishing, we could then accurately calculate the thickness of zircon removed from the polishing. Subsequently the zircon was reanalyzed in depth profile mode by ion microprobe after cleaning and Au coating. We repeated this sequence twice to obtain a ∼16-μm-long profile from the surface of the grain to the center.

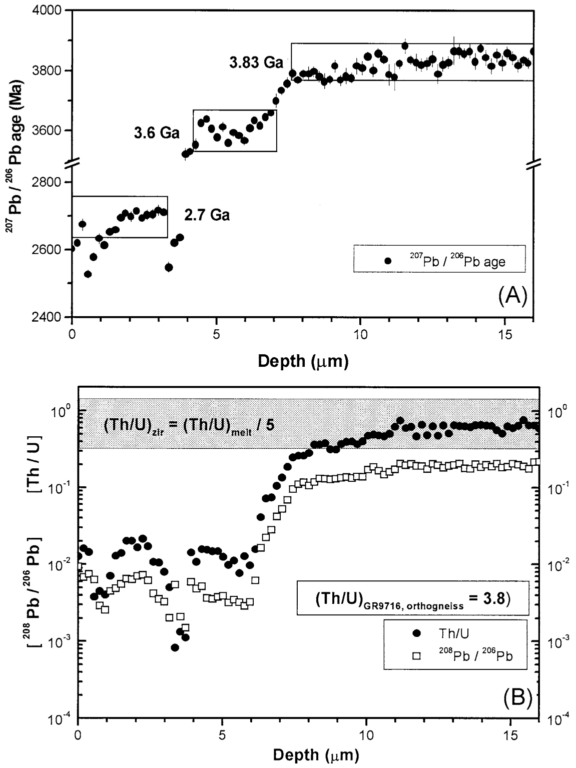

Fig. 5. (a) U–Pb age vs. depth profile in zircon GR9716_69. The spectrum of ages is interpreted to be due to a magmatic core with an age of 3828 ± 8 Ma and metamorphic overgrowths at ∼3.6 Ga and 2.5–2.7 Ga. (b) Depth profiles of measured Th/U and ^{208}Pb/^{206}Pb. Note the correlation between Th/U increasing from $\sim 10^{-2}$ to 0.8 and increasing age from ∼3.6 Ga to 3.83 Ga. Grey region shows predicted zircon Th/U crystallizing from a melt of Th/U = 3.8 assuming $K_{Th/U}^{liq/zir} = 5$ (± factor of two) [30].

4. Results

Our depth profile data (Table 2; Fig. 5) reveal a concentrically zoned crystal containing the previously recognized age components in the Akilia orthogneisses [3,5–7,16,22,26,27]: between 0–3 μm depth, concordant ^{207}Pb/^{206}Pb ages increase essentially monotonically from 2.5 to 2.7 Ga; ages then rise abruptly to a ∼3.6-Ga plateau between 3 and 5 μm; between ∼8 and 16 μm ages are consistent with a single population. The weighted mean of all 43 ^{207}Pb/^{206}Pb ages between ∼8 and 16 μm depth (Table 2) is 3828 ± 8 Ma (2 σ; MSWD = 1.2) (Fig. 5A).

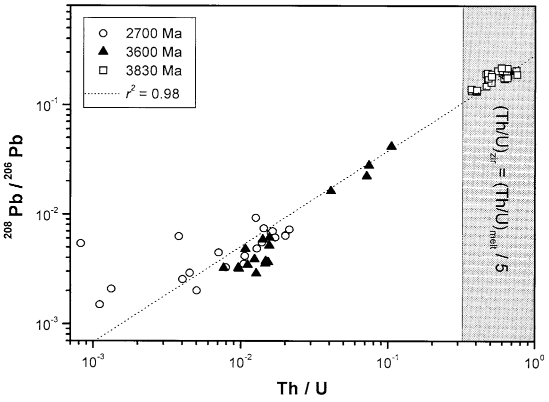

Fig. 6. Correlation diagram of ^{208}Pb/^{206}Pb vs. Th/U for ion microprobe depth profile data of zircon GR9716_69. The variability of data for the ~3.6-Ga and ~2.7-Ga zones of the zircon are in contrast to the relatively small spread of values for the 3830-Ga component.

4.1. Whole-rock and zircon Th/U chemistry

These depth profile data also show a clear correlation between both the measured Th/U and time-integrated Th/U (^{208}Pb/^{206}Pb) with apparent U–Pb age. Because zircon is the only magmatic phase in this rock that could significantly fractionate Th from U, the crystallizing tonalitic melt would maintain a Th/U ratio similar to the whole rock value of 3.8 (typical of continental crust [28]) until zircon became saturated (Table 1; Fig. 2) [29]. A zircon grown from a melt with Th/U = 3.8 is expected to have a Th/U ratio of ~0.8 (i.e. $D_{Th/U}^{zircon/melt} = 0.2$) [30]. The only portion of zircon GR9716_69 characterized by a similar value is the 3.83-Ga component, which closely matches the predicted Th/U (Fig. 5B). In contrast, Th/U for the ~3.6-Ga and ~2.5–2.7-Ga zones are indistinguishable from one another and are nearly two orders of magnitude lower than the 3.83-Ga portion (Fig. 6).

Because there is agreement that zircon growth at ca. 2.7 Ga is metamorphic [3,5–7], the simplest explanation for the ~3.6-Ga portion is that it too precipitated from an aqueous metamorphic fluid. During prograde metamorphism, Th/U generally increases in orthogneisses from an average crustal value of ~4 to as high as 50, implying that U is preferentially leached relative to Th and that metamorphic fluids are characterized by low Th/U [31,32]. Thus our expectation is that the Th/U ratio of zircon precipitated from a metamorphic fluid would be substantially lower than the magmatic value, both because of the low Th/U of the intergranular fluid and the preferential partitioning of U with respect to Th into the precipitating zircon. The combination of these two effects could explain the decrease in Th/U by two orders of magnitude in the 2.5–2.7-Ga and ~3.6-Ga overgrowths relative to the 3.83-Ga core. We interpret these data to indicate that the only primary magmatic component of this zircon is associated with an age of 3828 ± 8 Ma.

4.2. In situ measurements of zircon in petrographic thin sections

We gained additional insight into the zircon age distribution by U–Pb dating zircons in petrographic thin section of GR9716. Zircons are the dominant Th/U carrying phase and occur in three textural relationships in GR9716, i.e. (1) small ($<$ 50 μm) zircons hosted entirely within minimum-melt leucosomes; (2) similarly small zircons hosted by plagioclase; and (3) zircons ranging in size from 50-μm to rarely 250-μm grains, hosted by mafic phases. Eighty-eight U–Pb ages from 71 grains were measured in situ by this technique. In general, ages span the range from ca. 3.8 to 2.5 Ga (Table 3; Fig. 7) with the exception of one zircon encompassed with in biotite that yielded an anomalous age of 4.1 Ga. Zircons in leucosomes yielded only ~3.6-Ga ages. This latter observation is consistent with the earlier inference [20] that partial melting accompanied granulite facies metamorphism at that time [5] and potentially explains the variation of Th/U in ~3.6-Ga zircons (Fig. 3). Both values consistent with primary magmas (~0.5) and those expected for precipitation from a metamorphic fluid ($\leq 10^{-1}$) are represented in our data (Fig. 7). We propose that the dissolution/re-precipitation of a significant fraction ($>$ 50%?) of the protolith zircon at ~3.6 Ga via partial melting, solution in an aqueous fluid, or Ostwald ripening [33,34], provides a source of radiogenic Pb at that time

Table 3
Ion microprobe analyses of zircons in situ from orthogneiss sample GR9716

Grain spot	^{206}Pb*/^{238}U age	^{207}Pb*/^{235}U age	^{207}Pb*/^{206}Pb* age	Th/U
g1-1	3738 ± 173	3963 ± 66	4079 ± 18	0.0740
g11-1	3444 ± 105	3440 ± 40	3437 ± 14	0.1480
z11-1	3249 ± 58	3496 ± 21	3641 ± 4	0.0700
z14-1	3452 ± 49	3529 ± 18	3574 ± 6	0.0910
g6-1	3491 ± 54	3631 ± 20	3709 ± 6	0.4070
g17-1	3475 ± 62	3617 ± 22	3696 ± 8	0.3640
g17-2	3383 ± 86	3570 ± 33	3677 ± 10	0.6030
z1-1	3295 ± 69	3472 ± 26	3576 ± 7	0.3960
z1-2	2335 ± 95	3002 ± 46	3485 ± 19	0.4480
z2-1	3773 ± 64	3746 ± 22	3732 ± 7	0.4560
g4-2	3793 ± 54	3755 ± 18	3735 ± 5	0.7630
g4-1	3376 ± 47	3554 ± 17	3656 ± 6	0.7570
g16-1	3108 ± 78	3438 ± 31	3636 ± 4	0.3320
g16-2	1690 ± 40	2706 ± 27	3586 ± 14	0.7210
z13-1	1213 ± 40	2368 ± 34	3598 ± 14	0.3610
g5-1	3801 ± 96	3756 ± 33	3732 ± 11	0.4050
g2-1	2791 ± 139	3053 ± 60	3231 ± 12	0.1040
z3-1	3614 ± 71	3624 ± 25	3629 ± 7	0.2250
z4-1	3627 ± 65	3606 ± 25	3594 ± 7	0.9690
z6-1	3349 ± 82	3514 ± 30	3610 ± 10	0.5920
z8-1	3573 ± 46	3612 ± 16	3633 ± 5	0.1720
z10-1	3201 ± 88	3222 ± 35	3236 ± 14	0.4660
g3-2	3281 ± 158	3480 ± 60	3597 ± 5	0.0930
g3-1	2702 ± 65	2896 ± 31	3033 ± 20	0.1470
g9-1	1768 ± 66	2850 ± 44	3741 ± 14	0.2780
g18-1	3083 ± 81	3428 ± 34	3636 ± 11	0.3310
g14-1	2434 ± 80	2556 ± 38	2654 ± 16	0.0440
z5-1	2811 ± 77	3002 ± 34	3133 ± 15	0.2760
z7-1	3343 ± 82	3465 ± 32	3536 ± 8	0.1490
g8-1	3129 ± 99	3468 ± 40	3670 ± 5	0.6620
g12-1	3115 ± 87	3333 ± 36	3466 ± 13	0.1190
z9-1	3464 ± 43	3623 ± 16	3713 ± 3	0.4430
z12-1	3495 ± 52	3625 ± 20	3697 ± 7	0.0930
z15-1	2935 ± 62	3307 ± 27	3540 ± 9	0.1560
z16-1	3397 ± 46	3517 ± 17	3587 ± 6	0.1630
z17-1	3174 ± 33	3413 ± 14	3556 ± 7	0.2310
z18-1	3314 ± 41	3545 ± 15	3678 ± 4	0.3080
15_2_29_1	3577 ± 47	3606 ± 24	3623 ± 14	0.2160
15_2_29_2	3546 ± 58	3535 ± 42	3528 ± 28	0.2660
15_2_33_1	3550 ± 34	3569 ± 17	3580 ± 10	0.5920
15_2_38_1	3773 ± 107	3597 ± 45	3500 ± 20	0.1570
15_2_38_2	3730 ± 68	3650 ± 38	3607 ± 24	0.2360
15_2_39_1	3538 ± 25	3545 ± 10	3548 ± 4	0.1200
15_2_40_1	3313 ± 18	3457 ± 8	3542 ± 6	0.0778
15_2_42_1	2855 ± 51	3021 ± 39	3133 ± 28	0.2520
15_2_5_1	3316 ± 29	3457 ± 17	3540 ± 11	0.2150
15_2_6_1	2957 ± 37	3072 ± 32	3148 ± 25	0.0808
16_2_10_1	2817 ± 18	2840 ± 14	2857 ± 11	0.0848
16_2_11_1	3606 ± 32	3611 ± 17	3614 ± 11	0.2190
16_2_1_1	3693 ± 21	3699 ± 10	3702 ± 7	0.3950
16_2_21_1	3337 ± 37	3379 ± 18	3404 ± 11	0.3140
16_2_23_1	3197 ± 47	3303 ± 24	3369 ± 13	0.1060
16_2_24_1	3312 ± 29	3417 ± 18	3478 ± 11	0.0802
16_2_2_1	3389 ± 26	3513 ± 12	3584 ± 5	0.1210

Table 3 (*Continued*).

Grain spot	^{206}Pb*/^{238}U age	^{207}Pb*/^{235}U age	^{207}Pb*/^{206}Pb* age	Th/U
16_2_35_1	2729 ± 18	2755 ± 25	2775 ± 23	0.0166
16_2_43_1	2826 ± 22	2915 ± 13	2977 ± 10	0.0436
16_2_47_1	3714 ± 42	3619 ± 21	3567 ± 13	0.3480
16_2_4_1	3170 ± 30	3270 ± 15	3332 ± 8	0.1680
16_2_6_1	3571 ± 42	3578 ± 21	3582 ± 12	0.4350
16_2_7_1	2523 ± 18	2571 ± 11	2609 ± 8	0.0123
15_10_1_1	3890 ± 211	3715 ± 74	3622 ± 17	0.9770
15_10_1_2	4078 ± 265	3833 ± 91	3707 ± 23	0.2770
15_10_1_3	3432 ± 224	3494 ± 87	3529 ± 20	0.8750
15_10_3_1	3222 ± 251	3314 ± 105	3370 ± 28	0.1840
15_10_3_3	3174 ± 160	3381 ± 72	3507 ± 25	0.9940
15_6_1_1	2893 ± 270	2741 ± 111	2631 ± 13	0.0767
15_6_2_2	2587 ± 63	2601 ± 29	2612 ± 8	0.0539
15_6_3_1	2569 ± 89	3055 ± 41	3392 ± 8	0.0765
15_6_4_1	3524 ± 127	3516 ± 47	3512 ± 6	0.1230
15_6_5_1	3656 ± 77	3611 ± 28	3586 ± 5	0.1690
15_6_6_1	2621 ± 93	3061 ± 43	3364 ± 13	0.2340
15_6_7_1	3266 ± 121	3257 ± 48	3251 ± 12	0.1540
15_7_10_1	3374 ± 198	3429 ± 76	3461 ± 12	0.1220
15_7_11_1	3395 ± 218	3386 ± 84	3380 ± 22	0.3850
15_7_11_2	2985 ± 129	3227 ± 56	3381 ± 21	0.1590
15_7_1_1	3308 ± 160	3444 ± 64	3524 ± 11	0.6300
15_7_2_1	3477 ± 128	3555 ± 48	3599 ± 7	0.6490
15_7_2_2	3574 ± 176	3530 ± 64	3505 ± 13	0.2740
15_7_2_3	3204 ± 190	3349 ± 75	3437 ± 16	0.1120
15_7_3_1	3550 ± 81	3554 ± 30	3557 ± 4	0.0688
15_7_4_1	3196 ± 100	3356 ± 40	3454 ± 12	0.1510
15_7_5_1	3225 ± 97	3399 ± 40	3502 ± 9	0.1070
15_7_6_1	3060 ± 145	3322 ± 58	3484 ± 11	0.3080
15_7_7_1	3065 ± 120	3341 ± 51	3511 ± 13	0.6420
15_7_7_2	3859 ± 267	3612 ± 93	3478 ± 12	0.1090
15_7_8_1	2778 ± 148	3025 ± 67	3193 ± 22	0.2840
15_7_9_1	2905 ± 130	3245 ± 56	3461 ± 14	1.3700
15_7_9_2	2740 ± 123	2956 ± 56	3106 ± 21	0.0888

Ages are expressed in millions of yr (Ma). Nomenclature is sample+grain number+ion microprobe spot number. ‘g’, ‘z’, ‘15’ and ‘16’ analyses were done during separate sessions; ‘15_’ and ‘16_’ are aliquots of the same sample GR9716. Isotopic ratios have been corrected for common Pb. U–Pb ages were determined by comparison with a working curve defined by measurement of standard zircon AS-3 that yields concordant ^{206}Pb/^{238}U and ^{207}Pb/^{235}U ages of 1099.1 ± 0.5 Ma by conventional methods [23]. Error demagnification was applied to the data; all uncertainties are reported at the 1σ level.

to reset the common Pb isotopic composition of the coexisting feldspars.

An anomalous ~4.1-Ga zircon age we observed for one grain in thin section is unusual in both its surprising antiquity (no U–Pb zircon age this old has been reported for the ‘Itsaq Gneiss Complex’) and low Th/U value (0.074) relative to the cluster of ages at ~3.8 Ga (Table 3). While we noted earlier the unlikelihood of the parent tonalite melt containing a high fraction of inherited zircons (see [6]), this specific and isolated occurrence can be explained by the fact that the zircon is included within biotite. This association suggests that zircon survived the magmatic episode because it was incorporated into the magma late in its crystallization history and was temporarily armored against dissolution by the encompassing stable phase. The apparent rarity of this relationship suggests that the postulated armoring mechanism is not robust and thus the inherited

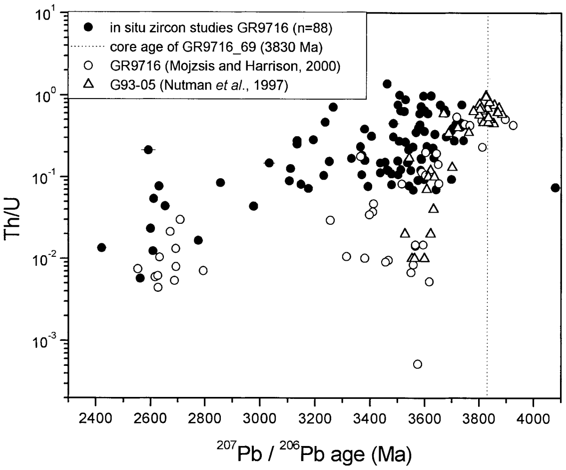

Fig. 7. Results of zircon U–Th–Pb in situ studies of orthogneiss GR9716 plotted with data from [3,6,16] and listed on Table 3.

zircon is unlikely to have been transported far from its source, otherwise the grain and its host biotite would have been resorbed by the tonalite. Indeed, based on this finding it is of interest whether there may be rocks even older than the 3.83-Ga Akilia orthogneiss present somewhere in the ‘Itsaq Gneiss Complex’ of West Greenland and perhaps in the vicinity of Akilia.

5. Conclusions

Our conclusions regarding the age and origin of the contested orthogneiss on Akilia are as follows.

(1) Age profiles from near surface (≤ 5 μm) regions of individual zircons can be obtained using the depth profiling mode of the ion microprobe. The increased spatial resolution of this zircon geochronological technique permits detection of age variations at the ~200-nm scale. By employing sequential repolishing, depth–age-composition profiles of zircons of essentially unlimited length can be obtained.

(2) U–Pb age depth profiling of a complex tonalitic orthogneiss from Akilia, West Greenland, reveals evidence for three phases of concentric zircon growth at 3.83 Ga, ~3.6 Ma and 2.7–2.5 Ga.

(3) Zircon growth at both ~3.6 Ga and 2.7–2.5 Ga is consistent with precipitation from a metamorphic fluid. Only the zircon core is consistent with crystallization from magma of the composition of the host rock.

(4) The crystallization age of the previously contested Southwest Akilia tonalitic orthogneiss is 3828 ± 8 Ma. Hence, sediments [3] and evidence for life therein [2] on Akilia are > 3.83 Ga in age.

(5) That these data do not agree with common-Pb compositions of the Akilia orthogneiss [12] calls for quantitative studies of open-system behavior of Pb during multiple periods of high-grade (granulite) metamorphism. An important aspect of future work would be the evaluation of fluid transport of U and Th that proceeded during granulitization of, for example, West Greenland orthogneisses.

(6) An anomalously old (~4.1 Ga) zircon (Table 3; g1-1) measured in situ in thin section may point to the unforeseen existence of primordial crust in the vicinity of the ‘Itsaq Gneiss Complex’.

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