Electronic Supplementary Information (ESI)

In situ isotopic analysis of uranium using a new data acquisition protocol for 10^{13} ohm Faraday amplifiers

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Table S1 Uranium isotopic data for zircons, titanites and synthetic glass materials

Sample	Spot	$^{235}U/^{238}U$ (× 10^{-3})				$^{234}U/^{238}U$ (× 10^{-5})				Ablated area (μm²)
		Tau correction[a]		CIM[b]		*Tau* correction[a]		CIM[b]		
Nancy 91500 zircon	1	7.2612 ± 0.0836		7.2281 ± 0.0428		3.52 ± 6.63		5.80 ± 3.04		75 × 75
	2	7.2375 ± 0.0841		7.2243 ± 0.0397		3.40 ± 7.98		4.82 ± 2.81		75 × 75
	3	7.2496 ± 0.0687		7.2473 ± 0.0392		3.43 ± 7.82		4.06 ± 2.77		75 × 75
	4	7.2487 ± 0.1165		7.3030 ± 0.0393		3.69 ± 5.89		4.85 ± 2.78		75 × 75
	5	7.2506 ± 0.0915		7.2853 ± 0.0385		5.83 ± 10.32		6.08 ± 2.71		75 × 75
	6	7.2864 ± 0.0946		7.2572 ± 0.0388		9.15 ± 3.98		7.11 ± 2.74		75 × 75
	7	7.2717 ± 0.0624		7.2398 ± 0.0375		4.68 ± 7.31		5.19 ± 2.64		75 × 75
	8	7.2516 ± 0.0690		7.2651 ± 0.0370		3.23 ± 4.80		3.68 ± 2.60		75 × 75
	9	7.2813 ± 0.0895		7.2585 ± 0.0375		3.83 ± 4.19		6.03 ± 2.64		75 × 75
	10	7.2675 ± 0.0862		7.2556 ± 0.0374		4.77 ± 6.17		5.46 ± 2.63		75 × 75
AS3 zircon	1	7.2633 ± 0.0193		7.2577 ± 0.0083		5.16 ± 0.97		5.85 ± 0.44		75 × 75
	2	7.2426 ± 0.0235		7.2521 ± 0.0086		5.49 ± 1.32		5.84 ± 0.45		75 × 75
	3	7.2533 ± 0.0184		7.2506 ± 0.0054		5.52 ± 1.07		5.70 ± 0.24		75 × 75
	4	7.2499 ± 0.0243		7.2577 ± 0.0048		5.59 ± 0.72		5.69 ± 0.19		75 × 75
	5	7.2606 ± 0.0228		7.2564 ± 0.0094		5.55 ± 1.50		5.70 ± 0.52		75 × 75
	6	7.2532 ± 0.0254		7.2595 ± 0.0092		5.80 ± 1.04		6.06 ± 0.50		75 × 75
	7	7.2603 ± 0.0359		7.2599 ± 0.0088		6.05 ± 1.05		5.73 ± 0.47		75 × 75
	8	7.2569 ± 0.0271		7.2609 ± 0.0089		5.62 ± 1.25		5.86 ± 0.48		75 × 75
	9	7.2806 ± 0.0627		7.2755 ± 0.0212		7.04 ± 2.50		6.28 ± 1.39		75 × 75
	10	7.2663 ± 0.0224		7.2562 ± 0.0052		5.09 ± 0.99		5.53 ± 0.22		75 × 75
FCT zircon	1	7.1675 ± 0.0705		7.2462 ± 0.0284		9.39 ± 16.98		5.36 ± 1.94		30 × 30
	2	7.2073 ± 0.0701		7.2480 ± 0.0232		4.17 ± 9.93		5.52 ± 1.55		30 × 30
	3	7.1913 ± 0.0690		7.2586 ± 0.0326		-0.28 ± 15.54		5.03 ± 2.26		30 × 30
	4	7.2095 ± 0.0716		7.2527 ± 0.0198		6.90 ± 8.29		5.11 ± 1.29		30 × 30
	5	7.2046 ± 0.0640		7.2597 ± 0.0199		6.32 ± 10.43		5.79 ± 1.30		30 × 30
	6	7.2928 ± 0.2816		7.3961 ± 0.3204		8.79 ± 38.64		14.02 ± 24.38		30 × 30
	7	7.2010 ± 0.0725		7.2351 ± 0.0334		-5.08 ± 14.71		4.83 ± 2.32		30 × 30
	8	7.1943 ± 0.0635		7.2627 ± 0.0232		4.05 ± 14.11		5.32 ± 1.55		30 × 30
	9	7.2005 ± 0.0695		7.2486 ± 0.0189		5.40 ± 8.23		5.73 ± 1.22		30 × 30
	10	7.2068 ± 0.0753		7.2526 ± 0.0204		5.92 ± 8.23		5.15 ± 1.33		30 × 30
	11	7.2220 ± 0.0947		7.2799 ± 0.0277		2.17 ± 18.64		6.22 ± 1.89		30 × 30
	12	7.1807 ± 0.0645		7.2560 ± 0.0226		3.54 ± 5.56		6.12 ± 1.50		30 × 30
	13	7.1949 ± 0.0761		7.2454 ± 0.0248		10.77 ± 26.43		5.35 ± 1.67		30 × 30
	14	7.1950 ± 0.0832		7.2732 ± 0.0315		11.36 ± 13.02		5.55 ± 2.18		30 × 30
OD-3 zircon	1	7.2696 ± 0.0240		7.2660 ± 0.0103		6.03 ± 2.42		5.42 ± 0.57		75 × 75
	2	7.2627 ± 0.0425		7.2548 ± 0.0157		5.61 ± 2.74		5.75 ± 0.98		75 × 75
	3	7.2348 ± 0.0214		7.2529 ± 0.0078		4.91 ± 0.95		5.17 ± 0.40		75 × 75
	4	7.2606 ± 0.0276		7.2572 ± 0.0125		5.25 ± 2.05		5.22 ± 0.74		75 × 75
	5	7.2549 ± 0.0159		7.2529 ± 0.0078		5.57 ± 0.77		5.39 ± 0.39		75 × 75
	6	7.2500 ± 0.0224		7.2565 ± 0.0072		5.70 ± 1.16		5.61 ± 0.35		75 × 75
	7	7.2513 ± 0.0299		7.2604 ± 0.0114		5.51 ± 1.71		5.23 ± 0.66		75 × 75
	8	7.2608 ± 0.0403		7.2536 ± 0.0147		6.33 ± 1.30		6.01 ± 0.90		60 × 60
	9	7.2819 ± 0.0658		7.2680 ± 0.0280		5.06 ± 2.71		5.86 ± 1.91		60 × 60
	10	7.2403 ± 0.0360		7.2518 ± 0.0075		5.58 ± 0.80		5.32 ± 0.37		60 × 60
	11	7.2534 ± 0.0293		7.3086 ± 0.0128		5.99 ± 1.17		7.50 ± 0.76		60 × 60
	12	7.2480 ± 0.0242		7.2606 ± 0.0098		5.55 ± 1.09		5.48 ± 0.54		60 × 60
	13	7.2660 ± 0.0468		7.2700 ± 0.0160		6.35 ± 3.02		5.92 ± 1.00		60 × 60
PSV zircon	1	7.2590 ± 0.0146		7.2584 ± 0.0060		5.74 ± 0.78		5.38 ± 0.27		75 × 75
	2	7.2557 ± 0.0292		7.2567 ± 0.0063		5.42 ± 0.70		5.60 ± 0.29		75 × 75
	3	7.2586 ± 0.0213		7.2575 ± 0.0063		5.59 ± 0.79		5.48 ± 0.30		75 × 75
	4	7.2566 ± 0.0192		7.2632 ± 0.0064		5.63 ± 1.03		5.70 ± 0.30		75 × 75
	5	7.2522 ± 0.0234		7.2595 ± 0.0068		5.37 ± 0.83		5.32 ± 0.33		75 × 75
	6	7.2679 ± 0.0244		7.2545 ± 0.0060		5.59 ± 0.60		5.44 ± 0.28		75 × 75
	7	7.2457 ± 0.0266		7.2539 ± 0.0066		5.56 ± 1.08		5.42 ± 0.31		75 × 75
	8	7.2523 ± 0.0242		7.2597 ± 0.0066		5.51 ± 0.83		5.42 ± 0.32		75 × 75
	9	7.2449 ± 0.0227		7.2524 ± 0.0069		5.60 ± 0.73		5.31 ± 0.33		75 × 75
	10	7.2544 ± 0.0181		7.2598 ± 0.0064		5.50 ± 0.74		5.37 ± 0.30		75 × 75
	11	7.2547 ± 0.0232		7.2614 ± 0.0058		5.60 ± 0.84		5.28 ± 0.26		75 × 75
	12	7.2474 ± 0.0146		7.2586 ± 0.0052		5.35 ± 0.36		5.46 ± 0.22		75 × 75
	13	7.2666 ± 0.0274		7.2558 ± 0.0055		5.47 ± 0.69		5.43 ± 0.24		75 × 75
	14	7.2554 ± 0.0102		7.2551 ± 0.0054		5.57 ± 0.66		5.30 ± 0.23		75 × 75
	15	7.2536 ± 0.0199		7.2588 ± 0.0054		5.41 ± 0.43		5.50 ± 0.24		75 × 75

Table S1 (Continued)

Sample	Spot	$^{235}U/^{238}U$ ($\times 10^{-3}$)						$^{234}U/^{238}U$ ($\times 10^{-5}$)						Ablated area (μm^2)
		Tau correction[a]			CIM[b]			*Tau* correction[a]			CIM[b]			
BLR-1 titanite	1	7.2565	±	0.0214	7.2580	±	0.0051	5.67	±	0.66	5.51	±	0.21	75 × 75
	2	7.2644	±	0.0133	7.2633	±	0.0052	5.48	±	0.50	5.57	±	0.22	75 × 75
	3	7.2637	±	0.0237	7.2641	±	0.0051	5.64	±	0.49	5.58	±	0.22	75 × 75
	4	7.2673	±	0.0218	7.2666	±	0.0052	5.69	±	0.50	5.30	±	0.22	75 × 75
	5	7.2698	±	0.0199	7.2633	±	0.0052	5.84	±	1.00	5.45	±	0.22	75 × 75
	6	7.2522	±	0.0162	7.2583	±	0.0053	5.62	±	0.42	5.55	±	0.22	75 × 75
	7	7.2571	±	0.0155	7.2613	±	0.0052	5.67	±	0.59	5.37	±	0.22	75 × 75
	8	7.2680	±	0.0152	7.2641	±	0.0052	5.64	±	0.41	5.53	±	0.22	75 × 75
	9	7.2683	±	0.0123	7.2621	±	0.0052	5.51	±	0.40	5.53	±	0.22	75 × 75
	10	7.2666	±	0.0121	7.2648	±	0.0052	5.44	±	0.56	5.61	±	0.22	75 × 75
FCT titanite	1	7.2746	±	0.0350	7.2564	±	0.0159	5.53	±	1.68	5.40	±	0.99	75 × 75
	2	7.2752	±	0.0391	7.2807	±	0.0162	7.69	±	2.36	6.93	±	1.02	75 × 75
	3	7.2984	±	0.0347	7.2769	±	0.0153	6.80	±	1.87	7.09	±	0.95	75 × 75
	4	7.2876	±	0.0304	7.2672	±	0.0195	7.30	±	2.49	7.01	±	1.27	75 × 75
	5	7.2796	±	0.0330	7.2681	±	0.0189	5.71	±	3.44	6.94	±	1.22	75 × 75
	6	7.2832	±	0.0343	7.2745	±	0.0146	7.39	±	2.66	5.65	±	0.90	75 × 75
	7	7.2853	±	0.0340	7.2700	±	0.0127	5.39	±	1.94	5.81	±	0.75	75 × 75
	8	7.2799	±	0.0277	7.2650	±	0.0127	6.96	±	2.21	6.46	±	0.75	75 × 75
	9	7.2585	±	0.0372	7.2632	±	0.0159	7.04	±	3.59	6.08	±	0.99	75 × 75
	10	7.2230	±	0.0342	7.2043	±	0.0139	6.68	±	2.52	6.86	±	0.85	75 × 75
	11	7.2286	±	0.0408	7.2303	±	0.0146	6.81	±	2.26	6.30	±	0.90	75 × 75
	12	7.2273	±	0.0585	7.1621	±	0.0245	9.28	±	5.15	8.44	±	1.65	75 × 75
	13	7.3015	±	0.0321	7.2653	±	0.0219	8.34	±	3.32	6.66	±	1.44	75 × 75
	14	7.2875	±	0.0295	7.2671	±	0.0161	6.90	±	2.79	6.96	±	1.01	75 × 75
	15	7.2777	±	0.0315	7.2400	±	0.0145	6.92	±	1.54	6.05	±	0.89	75 × 75
	16	7.2591	±	0.0417	7.2507	±	0.0170	6.74	±	3.37	7.40	±	1.08	75 × 75
	17	7.2640	±	0.0309	7.2425	±	0.0140	6.52	±	2.29	6.14	±	0.85	75 × 75
	18	7.2667	±	0.0317	7.2606	±	0.0146	5.68	±	2.51	6.19	±	0.90	75 × 75
MKED1 titanite	1	7.2842	±	0.0259	7.2613	±	0.0101	6.12	±	1.41	5.74	±	0.56	75 × 75
	2	7.2646	±	0.0100	7.2700	±	0.0103	5.93	±	0.98	5.50	±	0.57	75 × 75
	3	7.2536	±	0.0297	7.2700	±	0.0102	4.97	±	0.98	5.54	±	0.57	75 × 75
	4	7.2787	±	0.0233	7.2690	±	0.0098	5.96	±	1.46	5.82	±	0.54	75 × 75
	5	7.2670	±	0.0305	7.2649	±	0.0099	5.87	±	1.23	5.35	±	0.55	75 × 75
	6	7.2727	±	0.0324	7.2664	±	0.0100	5.78	±	1.09	5.26	±	0.56	75 × 75
	7	7.2711	±	0.0241	7.2599	±	0.0097	5.76	±	0.83	5.53	±	0.54	75 × 75
	8	7.2708	±	0.0320	7.2635	±	0.0099	5.44	±	2.09	5.38	±	0.54	75 × 75
	9	7.2788	±	0.0322	7.2703	±	0.0098	6.40	±	1.22	5.47	±	0.54	75 × 75
	10	7.2674	±	0.0291	7.2729	±	0.0097	5.72	±	1.23	5.53	±	0.53	75 × 75
OLT1 titanite	1	7.2599	±	0.0262	7.2636	±	0.0052	5.45	±	0.37	5.56	±	0.22	75 × 75
	2	7.2592	±	0.0183	7.2654	±	0.0053	5.35	±	0.68	5.50	±	0.23	75 × 75
	3	7.2616	±	0.0281	7.2637	±	0.0054	5.66	±	0.25	5.61	±	0.23	75 × 75
	4	7.2620	±	0.0157	7.2619	±	0.0054	5.76	±	0.32	5.50	±	0.23	75 × 75
	5	7.2643	±	0.0201	7.2568	±	0.0054	5.54	±	0.53	5.52	±	0.23	75 × 75
	6	7.2513	±	0.0259	7.2589	±	0.0054	5.59	±	0.58	5.43	±	0.23	75 × 75
	7	7.2538	±	0.0151	7.2613	±	0.0054	5.50	±	0.39	5.51	±	0.23	75 × 75
	8	7.2585	±	0.0234	7.2595	±	0.0055	5.40	±	0.66	5.40	±	0.24	75 × 75
	9	7.2615	±	0.0189	7.2606	±	0.0053	5.64	±	0.56	5.53	±	0.23	75 × 75
	10	7.2553	±	0.0268	7.2639	±	0.0055	5.67	±	0.51	5.72	±	0.24	75 × 75

Table S1 (Continued)

Sample	Spot	$^{235}U/^{238}U$ ($\times 10^{-3}$)		$^{234}U/^{238}U$ ($\times 10^{-5}$)		Ablated area (μm^2)
		Tau correction[a]	CIM[b]	*Tau* correction[a]	CIM[b]	
NIST SRM 610	1	2.3822 ± 0.0146	2.3901 ± 0.0036	1.15 ± 0.53	1.44 ± 0.20	75 × 75
	2	2.3874 ± 0.0118	2.3908 ± 0.0037	1.00 ± 0.60	1.29 ± 0.21	75 × 75
	3	2.3865 ± 0.0104	2.3860 ± 0.0036	1.58 ± 0.97	1.27 ± 0.21	75 × 75
	4	2.3914 ± 0.0093	2.3897 ± 0.0036	1.26 ± 0.62	1.24 ± 0.21	75 × 75
	5	2.3856 ± 0.0059	2.3906 ± 0.0036	1.16 ± 0.93	1.37 ± 0.21	75 × 75
	6	2.3878 ± 0.0073	2.3879 ± 0.0037	1.38 ± 0.58	1.40 ± 0.21	75 × 75
	7	2.3814 ± 0.0099	2.3852 ± 0.0037	1.18 ± 0.40	1.36 ± 0.21	75 × 75
	8	2.3847 ± 0.0101	2.3858 ± 0.0036	1.38 ± 0.67	1.31 ± 0.21	75 × 75
	9	2.3891 ± 0.0047	2.3857 ± 0.0037	1.51 ± 0.77	1.35 ± 0.21	75 × 75
	10	2.3858 ± 0.0086	2.3868 ± 0.0036	1.06 ± 0.50	1.18 ± 0.21	75 × 75
NBS SRM 610	1	2.3893 ± 0.0061	2.3868 ± 0.0036	1.23 ± 0.68	1.26 ± 0.21	75 × 75
	2	2.3890 ± 0.0089	2.3882 ± 0.0035	1.20 ± 0.51	1.30 ± 0.20	75 × 75
	3	2.3916 ± 0.0093	2.3912 ± 0.0036	1.27 ± 0.47	1.43 ± 0.20	75 × 75
	4	2.3934 ± 0.0072	2.3916 ± 0.0036	1.23 ± 0.60	1.21 ± 0.20	75 × 75
	5	2.3937 ± 0.0157	2.3905 ± 0.0036	1.17 ± 0.47	1.34 ± 0.21	75 × 75
	6	2.3920 ± 0.0083	2.3888 ± 0.0035	1.28 ± 0.51	1.14 ± 0.20	75 × 75
	7	2.3931 ± 0.0132	2.3926 ± 0.0034	1.14 ± 0.70	1.25 ± 0.19	75 × 75
	8	2.3916 ± 0.0122	2.3897 ± 0.0035	1.32 ± 0.52	1.31 ± 0.19	75 × 75
	9	2.3946 ± 0.0138	2.3901 ± 0.0035	1.37 ± 0.52	1.32 ± 0.20	75 × 75
	10	2.3899 ± 0.0127	2.3929 ± 0.0036	1.25 ± 0.74	1.22 ± 0.20	75 × 75
NIST SRM 610	1	2.3777 ± 0.0146	2.3855 ± 0.0036	0.70 ± 0.53	1.00 ± 0.20	75 × 75
(MS interference corrected)	2	2.3828 ± 0.0118	2.3863 ± 0.0037	0.56 ± 0.60	0.85 ± 0.21	75 × 75
	3	2.3820 ± 0.0104	2.3814 ± 0.0036	1.14 ± 0.97	0.83 ± 0.21	75 × 75
	4	2.3868 ± 0.0093	2.3852 ± 0.0036	0.82 ± 0.62	0.81 ± 0.21	75 × 75
	5	2.3811 ± 0.0059	2.3860 ± 0.0036	0.72 ± 0.93	0.93 ± 0.21	75 × 75
	6	2.3833 ± 0.0073	2.3834 ± 0.0037	0.94 ± 0.58	0.96 ± 0.21	75 × 75
	7	2.3768 ± 0.0099	2.3806 ± 0.0037	0.73 ± 0.40	0.93 ± 0.21	75 × 75
	8	2.3801 ± 0.0101	2.3812 ± 0.0036	0.93 ± 0.67	0.88 ± 0.21	75 × 75
	9	2.3846 ± 0.0047	2.3811 ± 0.0037	1.06 ± 0.77	0.92 ± 0.21	75 × 75
	10	2.3812 ± 0.0086	2.3823 ± 0.0036	0.61 ± 0.50	0.74 ± 0.21	75 × 75
NBS SRM 610	1	2.3843 ± 0.0061	2.3818 ± 0.0036	0.75 ± 0.68	0.79 ± 0.21	75 × 75
(MS interference corrected)	2	2.3839 ± 0.0089	2.3831 ± 0.0035	0.71 ± 0.51	0.83 ± 0.20	75 × 75
	3	2.3866 ± 0.0093	2.3862 ± 0.0036	0.78 ± 0.47	0.96 ± 0.20	75 × 75
	4	2.3883 ± 0.0072	2.3866 ± 0.0036	0.74 ± 0.60	0.74 ± 0.20	75 × 75
	5	2.3887 ± 0.0157	2.3854 ± 0.0036	0.68 ± 0.47	0.87 ± 0.21	75 × 75
	6	2.3869 ± 0.0083	2.3838 ± 0.0035	0.80 ± 0.51	0.67 ± 0.20	75 × 75
	7	2.3881 ± 0.0132	2.3875 ± 0.0034	0.65 ± 0.70	0.78 ± 0.19	75 × 75
	8	2.3865 ± 0.0122	2.3847 ± 0.0035	0.83 ± 0.52	0.83 ± 0.19	75 × 75
	9	2.3896 ± 0.0138	2.3851 ± 0.0035	0.88 ± 0.52	0.85 ± 0.20	75 × 75
	10	2.3849 ± 0.0127	2.3878 ± 0.0036	0.76 ± 0.74	0.75 ± 0.20	75 × 75

Mass biases on the $^{235}U/^{238}U$ and $^{234}U/^{238}U$ ratios were corrected by those of the GJ-1 zircon (see main text for details). These uncertainties mean two times standard deviation of the median.

[a] Calculated from data sets at plateaus in the time profile of $^{235}U/^{238}U$ ratio based on the conventional *tau* correction method

[b] Calculated by the present CIM method, where the uncertainty was estimated based on propagation of counting statistics on both signal and background

Table S2 Effect of *tau* factor onto the measured $^{235}U/^{238}U$ ratio

Tau factor	$^{235}U/^{238}U$ ($\times 10^{-3}$)	
	Tau correction[a]	CIM[b]
0.25	1.9500 ± 0.0113	1.9635 ± 0.0030
0.45	1.9591 ± 0.0080	1.9635 ± 0.0030
0.65[†]	**1.9682 ± 0.0071**	**1.9635 ± 0.0030**
0.85	1.9773 ± 0.0092	1.9635 ± 0.0030
1.05	1.9864 ± 0.0130	1.9635 ± 0.0030

[†] Optimum *tau* factor
Uncertainty is two times standard deviation of the median.

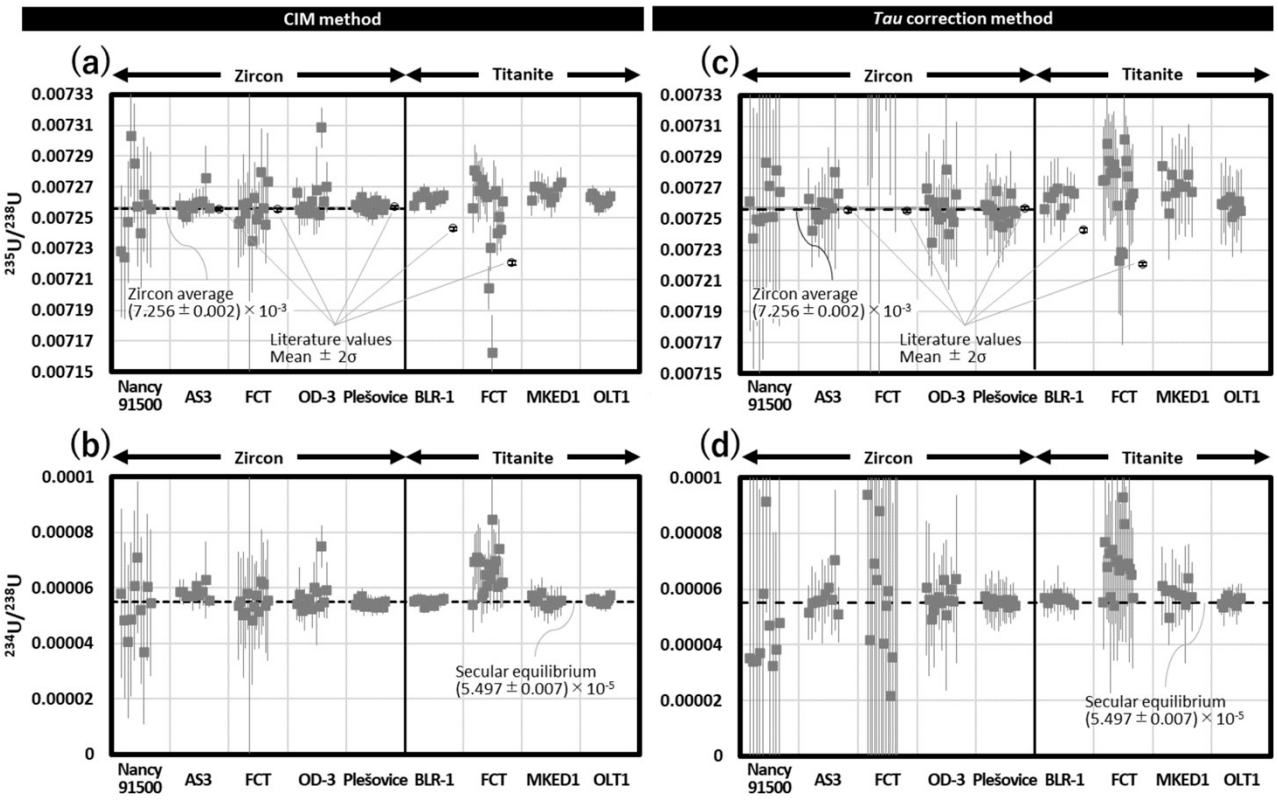

Fig. S1 Comparison of the resulting $^{235}U/^{238}U$ and $^{234}U/^{238}U$ ratios of zircons and titanites obtained by the CIM (a, b) and conventional *tau* correction methods (c, d). Note that the precision of the $^{235}U/^{238}U$ and $^{234}U/^{238}U$ ratios with the CIM method were 2 to 10 times better than those achieved by the tau correction method. Isotopic ratio data from individual signal events are shown as grey squares. The dashed lines are the averaged $^{235}U/^{238}U$ ratio for zircons reported by Hiess *et al.*,[1] and an expected $^{234}U/^{238}U$ ratio of materials in secular equilibrium. The $^{234}U/^{238}U$ ratio of minerals in a secular equilibrium is calculated from the decay constants of $\lambda_{234} = (2.82206 \pm 0.00080) \times 10^{-6}$ and $\lambda_{238} = (1.55125 \pm 0.00083) \times 10^{-10}$.[2,3] Mass bias was corrected using the GJ-1 zircon as a bracketing standard. The error bars were calculated based on propagation of counting statistics and baseline noise.

Supplementary references

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