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U-Th-total Pb ages of Uraninite and Thorite from Granititc Rocks in the Japanese Islands

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Absract Dating based on total UO_2, ThO_2 and PbO contents of uraninite and anhydrous thorite from granitic rocks was performed using an electron microprobe analyser (EPMA) at the National Museum of Nature and Science, Tokyo. After careful comparison with the ages obtained by isotopic methods (SHRIMP and K-Ar), this study confirmed that the EPMA-derived ages of uraninites and thorites are consistent with ages obtained by isotopic methods. The ages of the uraninite samples in this study ranged from 1.5 to 240 Ma. Standard deviation of uraninite age was 0.6 at 2.4 Ma, 0.7 at 5.2 Ma, 1.1 at 66.5 Ma and around 4 at over 180 Ma. As the standard deviation of uraninite is expected to be less than 1 Ma from count statistics, the high deviation for the old uraninite may be due to partly metamicted conditions. Thorite is the most common radiogenic mineral in granitic rocks. It was totally hydrated in the granitic samples older than 150 Ma and highly hydrated even in Tertiary granite. The age obtained from thorite was determined to be up to 120 Ma.
Key words : uraninite, thorite, age, EPMA, granite Introduction

Holmes (1911) first established a method for dating minerals through normal wet chemical analyses on the basis of the assumption that inherited Pb was negligible at the time of crystallization of minerals. Holmes (1931) later dealt exhaustively with the application of radioactivity to the measurement of geologic time. The method was used by many researchers and was referred to as the chemical age. After the isotopic method had been widely accepted, the inherited lead in uraninite was confirmed to be less than 0.2 % (*cf.* Holmes, 1960). Although in his 1931 paper Holmes presented a correct calculation method which was very complex, a simpler and more approximate age calculation method was applied for the minerals. Mineral age was simply obtained by

Approximate age (Ma)$=$a*Pb/(U+bTh) (1)

where elemental concentrations are in wt%. The numerical values, a and b, were given as 7600 and 0.36, respectively, in Holmes (1931). Since then, many researchers have tried to modify the values of a and b. Bowles (1990) and Suzuki and Adachi (1990) have developed accurate age calculations for uraninite and monazite, respectively. Bowles (1990) assumed that the initial Pb concentration is negligible as did Holmes, whereas Suzuki and Adachi (1990) insisted that the initial Pb is obtained by an isochron line. Both the calculations yield essentially the same age results. From mineral composition analyzed by an electron microprobe analyser which is now a ubiquitous machine, many researchers have obtained mineral ages using the above calculation methods. In recent calculations of mineral age, the initial Pb has been treated as negligible in amount as was first done by Holmes (1911 & 1931).

In electron microprobe analyses (EPMA), monazite and zircon in young igneous or metamorphic rocks exhibit levels of radiogenic Pb that are too low to calculate an accurate age, due to low U and Th contents. For monazite, a number of papers have treated sufficiently old rocks, i.e. Paleozoic or older (*e.g*. Montel *et al*., 1996; Williams *et al*., 1999; Foster *et al*., 2000a; Santosh *et al*., 2006). On the other hand, uraninite, thorianite, thorite and huttonite contain high concentrations of radiogenic Pb even in Tertiary rocks. Uraninite ages were obtained by Bowles (1990) from various rocks ranging in age from 19 to 1800 Ma, by Santosh *et al*. (2005) for 500 Ma gneiss, and by Hurtado *et al*. (2007) for Tertiary granite. Yokoyama *et al*., (2010) obtained ages of thorite, uraninite and thorianite from samples of Vietnumese river sands. Naemura *et al*. (2008) obtained ages of thorianite in peridotite. Foster *et al*. (2000b) analyzed thorite and huttonite in samples of beach sands from New Zealand, and obtained ages from 2.3 Ma to 210 Ma. In most of these dating studies, the EPMA ages were consistent with those obtained by isotopic methods or the EPMA ages of monazite.

In the Japanese Islands, uraninites in granitic rocks have been analyzed using wet chemical methods and were calculated using the simple equation of Holmes (Iimori, 1941; Nagashima and Nagashima, 1960). The EPMA age method, i.e. U-Th-total Pb or chemical methods, has attracted attention for its quickness, high spatial resolution and relatively high precision as well as isotopic methods. Granitic rocks in the Japansese Islands generally vary in age from 1.5 to 280 Ma. In this paper, we attempted to confirm the calculated chemical ages of uraninite and thorite using an EPMA in the National Museum of Nature and Science, and demonstrate the validity of the method which yields age results comparable to those obtained by the isotopic methods such as SHRIMP and K-Ar method. A study of monazite ages comparing the results obtained by the same EPMA machine and isotopic ages has been previously reported by Santosh *et al*. (2006).

Samples and methods

a: Sample preparation and chemical analyses

Most of the granitic rocks in the Japansese Islands have been analyzed using the K-Ar method (*e.g*. Kawano and Ueda, 1966: Shibata *et al*., 1984: Uchiumi *et al*., 1990). Cretaceous granitic rocks are the most ubiquitous, whereas Tertiary granitic rocks occur mostly in Southwest Japan along the Pacific Ocean. Permian to Triassic granites occur only in the Hida Terrane. Ages of far older granitic rocks, Cambrian to Silurian, have been obtained using the SHRIMP method (Watanabe et al., 1995; Sakashima *et al*. 2003), but these samples were usually from small plutonic bodies. As such old granitic rocks do not preserve uraninite and anhydrous thorite, we analyse uraninites and thorites in eleven granitic rocks with ages from 1.5 Ma to 280 Ma (Fig. 1 and Table 1). Four samples were from the Hida Terrane. Three samples were from Cretaceous region, and the other three from Tertiary and Quaternary rocks.

All samples (except for the Quaternary aplite) have not been previously examined by isotopic methods. Six samples are newly analyzed by SHRIMP in this study. The other four rocks are

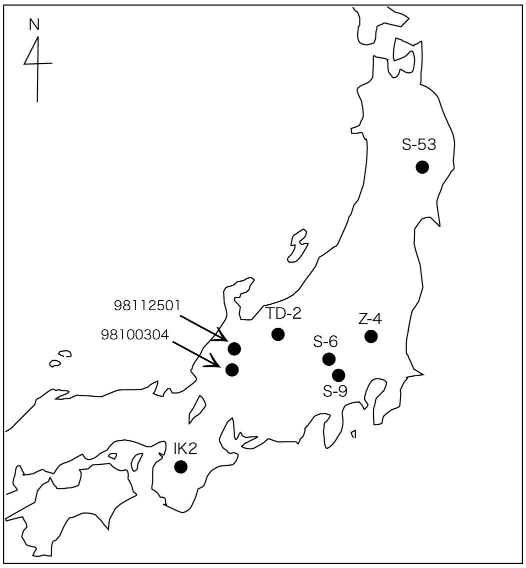

Fig. 1. Localities of the granitic rocks used for the EPMA age measurements.

Table 1. Summary of the analysed samples and ages obtained using EPMA and the other methods.

Locality	Sample No.	Rock type	EPMA age: Uraninite	error (1σ)	Thorite	error (1σ)	Age obtained by other methods: Isotope age	error (t-sigma)	method	Reference
Hida	98112501	granite	232.6	3.9			238.4*	4.6	SHRIMP	This paper
Hida	98092910	granite	204.4	4.3			200.6*	6.5	SHRIMP	This paper
Hida	99042103	granite	198.9	2.6			189.3*	4	SHRIMP	This paper
Hida	98100304	granite	189.6	4.0			192.5*	2.9	SHRIMP	This paper
Kitakami	SK53	granite	115.1	1.9	117	6.6	117*	2.7	SHRIMP	This paper
Ashio	Ashi-2	granite	99.2	1.7			103.8*	2.6	SHRIMP	This paper
Ikoma	IK-2	granodiorite	85.8	0.08			87**	4(2σ)	SHRIMP	Watanabe *et al*. (2000)
Tsukuba	Z-4	granite	66.5	1.1			63***		K-Ar(bi)	Kawano & Ueda (1966)
Yamanashi	S-9	granodiorite			12.1	1.5	12.1**	0.4	K-Ar(bi)	Uchiumi *et al*. (1990)
Yamanashi	S-6	granodiorite	5.2	0.7	4.8	1.2	4.4***	0.3	K-Ar(bi)	Shibata *et al*. (1984)
Hida	TD-2	aplite	2.4	0.6	3.4	1.1	1.4*	0.3	SHRIMP	Sano *et al*. (1999)

* same sample, ** sample is collected from the same outcrop, *** from the same granitoid body

collected from the same outcrops or igneous plutons as those analyzed by isotopic methods. The EPMA-derived ages of uraninites and thorites are compared with the ages as given by SHRIMP or the K-Ar methods (Table 1).

As uraninite and thorite are rare minerals in granitic rocks, the rock samples were crushed into fine fractions in a stainless-steel stamp mill. Powdered samples (<100 μm mesh size) were cleaned using water to remove the dust particles, and then dried in an oven. The magnetic minerals were then removed using a hand magnet. Zircon grains were hand picked for SHRIMP analyses. Other heavy fractions were separated using methylene iodide with specific gravity of 3.3, and mounted on glass slides using epoxy resin. Polished grain mounts were used for the study of grain characteristics and EPMA analyses.

The chemical analyses were carried out using a JEOL-8800 electron microprobe. Uraninite and thorite are at first fully analyzed to determine the presence of major and trace elements (P, Si, Ce, Pr, Nd, Sm, Gd, Dy, Er, Tm, Yb, Lu, Y, U, Th, Pb and Ca). The operating conditions of the microprobe were 15 kV accelerating voltage and 0.02 μA probe current. Counting time on each analytical point was 15 seconds at peak and 5 seconds at backgrounds for all the elements. PRZ corrections (modified ZAF) were applied for the analyses. Standards used were wollastonite for Si and Ca, synthetic (REE)P_5O_{14} for REE and P. The standards for U, Th and Pb were synthesized UO_2, ThO_2 and natural crocoite ($PbCrO_4$), respectively. Element uranium was calculated as UO_2. All the analytical points were selected under back-scattered images to avoid cracks and the metamicted parts of the sample. The analytical result is shown in Table 2.

In the age analyses, the elements were selected to minimize the damage to the minerals. They were P, Si, Y, U, Th, Pb and Ca. The probe operating conditions were 15 Kb and 0.1 μA. Counting times for Pb were 100 and 30 seconds at peak and backgrounds, respectively, whereas 50 seconds at peak and 20 seconds at backgrounds were used for U and Th analyses. Counting times for other elements were the same as those in the normal analyses. The U $M\alpha$, Th $M\alpha$ and Pb $M\alpha$ lines were used in the U, Th and Pb analyses, respectively, and the spectral interferences of the Th and Y lines with the Pb $M\alpha$ line, and the Th line with the U $M\alpha$ line were corrected (*cf*. Pyle *et al*., 2002).

b: Uraninite and thorite

In granitic rocks, uraninite (UO_2) and thorite ($ThSiO_4$) are usually rare minerals. Uraninite occurs mostly as a euhedral grain in the heavy fraction (Fig. 2), rarely surrounded by pyrite. Thorite is commonly altered into hydrous thorite. Anhy-

Table 2. Chemical compositions of uraninite and thorite in granitic rocks. Analytical conditions are different from age analyses. Current: 2×10^{-8} A, Measurement at peak and back ground are 15 and 5 seconds, respectively.

sample No.	98112501		98100304		SK53		SK53		IK2		S-9		S-6		S-6		TD-2		TD-2	
mineral	uraninite	n=27*	uraninite	n=14*	uraninite	n=12*	thorite	n=12*	uraninite	n=11*	thorite	n=14*	uraninite	n=7*	thorite	n=18*	uraninite	n=15*	thorite	n=15*
Comment	av. (wt%)	1σ	av. (wt%)	1σ	av. (wt%)	1σ	av. (wt%)	1σ	av. (wt%)	1σ	av. (wt%)	1σ	av. (wt%)	1σ	av. (wt%)	1σ	av. (wt%)	1σ	av. (wt%)	1σ
P_2O_5	0.01	0.01	0.13	0.29	0.01	0.02	1.24	0.37	0.01	0.01	0.24	0.09	0.02	0.01	0.43	0.33	0.02	0.01	1.62	0.78
SiO_2	0.07	0.13	0.11	0.18	0.08	0.06	17.98	0.13	0.05	0.04	18.32	0.18	0.04	0.02	18.21	0.25	0.03	0.02	17.67	0.33
UO_2	85.39	2.46	80.56	3.46	84.18	0.90	15.60	4.40	93.54	1.28	17.33	7.80	81.04	0.27	16.01	5.26	79.94	1.35	16.40	1.61
ThO_2	8.09	1.30	10.17	2.42	8.35	0.59	58.68	3.52	2.24	0.27	63.14	8.18	8.94	0.90	63.93	6.39	7.89	1.52	60.17	1.90
PbO	3.07	0.27	2.42	0.35	1.44	0.02	0.54	0.06	1.10	0.02	0.05	0.01	0.05	0.01	0.01	0.01	0.01	0.01	0.00	0.01
CaO	0.71	0.12	0.78	0.15	0.31	0.03	0.02	0.01	0.15	0.02	0.02	0.01	0.16	0.04	0.02	0.01	0.19	0.14	0.01	0.01
Y_2O_3	0.57	0.47	1.89	0.65	2.10	0.27	2.49	1.02	0.92	0.07	0.38	0.12	5.17	0.71	0.58	0.44	6.83	1.19	2.46	1.17
Ce_2O_3	0.30	0.10	0.60	0.08	0.45	0.09	0.35	0.16	0.20	0.05	0.11	0.07	0.74	0.06	0.12	0.05	0.75	0.27	0.18	0.05
Nd_2O_3	0.13	0.07	0.54	0.13	0.47	0.07	0.42	0.17	0.14	0.06	0.08	0.05	0.86	0.15	0.12	0.05	0.61	0.24	0.14	0.06
Sm_2O_3	0.07	0.06	0.23	0.08	0.27	0.03	0.18	0.09	0.11	0.05	0.04	0.04	0.51	0.07	0.05	0.05	0.41	0.09	0.11	0.05
Gd_2O_3	0.06	0.05	0.20	0.08	0.25	0.05	0.29	0.11	0.12	0.06	0.08	0.04	0.54	0.08	0.07	0.06	0.45	0.08	0.16	0.07
Dy_2O_3	0.09	0.06	0.28	0.09	0.37	0.07	0.34	0.15	0.18	0.05	0.05	0.05	0.89	0.07	0.09	0.08	1.10	0.21	0.29	0.14
Er_2O_3	0.04	0.04	0.21	0.10	0.24	0.08	0.22	0.09	0.10	0.05	0.04	0.05	0.59	0.10	0.06	0.05	0.86	0.19	0.21	0.13
Tm_2O_3	0.01	0.02	0.06	0.06	0.05	0.05	0.05	0.04	0.02	0.03	0.02	0.03	0.13	0.02	0.03	0.04	0.13	0.06	0.03	0.03
Yb_2O_3	0.04	0.05	0.19	0.12	0.17	0.05	0.13	0.08	0.05	0.06	0.03	0.03	0.40	0.12	0.04	0.04	0.75	0.29	0.15	0.10
Lu_2O_3	0.07	0.06	0.08	0.05	0.07	0.06	0.05	0.04	0.04	0.05	0.05	0.04	0.13	0.04	0.05	0.05	0.23	0.09	0.07	0.06
Total	98.73	0.83	98.46	1.16	98.79	0.43	98.57	0.51	98.95	0.97	99.95	0.34	100.20	0.44	99.79	0.49	100.20	0.35	99.66	0.44

*: number of analysed spot

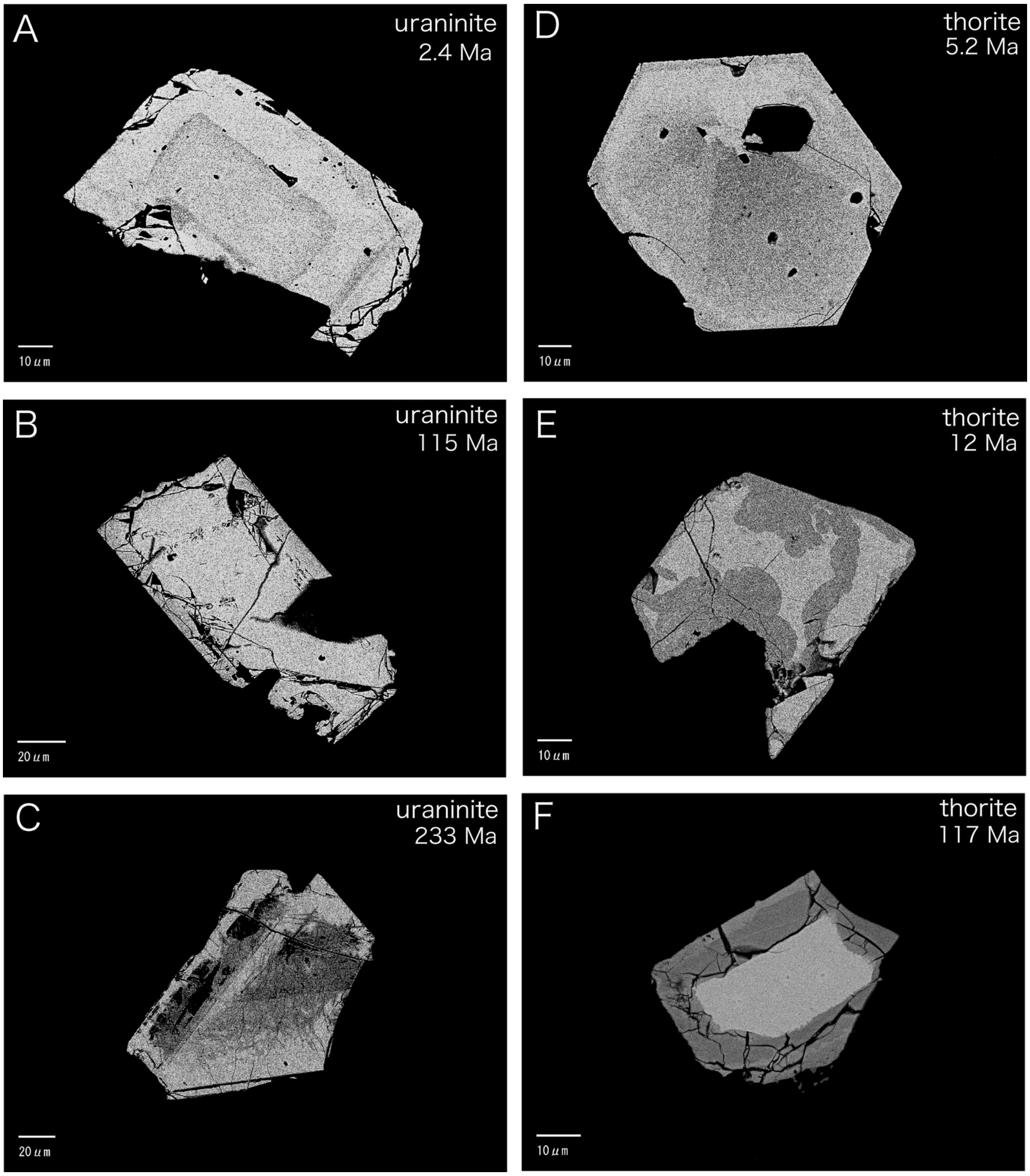

Fig. 2. BSE images of the uraninites and thorites. A: fresh uraninite in TD-2, B: fresh uraninite in SK53, C: highly metamicted uraninite in a sample 98112501. D: fresh thorite in TD-2, E: partly hydrated thorite in S-9 (dark parts are hydrous thorite), F: thorite surrounded by hydrous thorite in SK53.

drous thorite is mostly preserved partly in hydrated thorite grains (Fig. 2). In addition to the observation under BSE image, hydrous thorite is recognized by the presence of CaO content and low overall concentration.

Natural uraninite is composed mainly of UO_2 and ThO_2 with subordinate amounts of Y and heavy REEs. Anhydrous thorite is composed mainly of SiO_2, ThO_2 and UO_2 with subordinate amounts of Y and lantanoids. In the routine analyses for age determination, we analyzed only seven elements: P, Si, Y, U, Th, Pb and Ca. Under these conditions, total amounts of uraninite and thorite were found to be in a range from 90 to

100%. The lower values were usually Y-rich uraninites and thorite. The total amount of unanalyzed REE elements was roughly equivalent to Y_2O_3 contents (Fig. 3). The analyses of U, Th and Pb that yielded low totals were checked against Y_2O_3 contents.

c: Theoretical basis of chemical dating

Uraninite and thorite are abundant in U-Th system radioisotopes, which finally disintegrate into the stable Pb isotope through α and β decays. As the contents of nonradiogenic Pb in uraninite and thorite are negligible compared with the amount of radiogenic Pb, the age (t) of individual mineral was calculated using equation (2)

$$\frac{PbO}{W_{Pb}} = \frac{ThO_2}{W_{Th}} = \{\exp(^{232}Tht) - 1\} + \frac{UO_2}{W_U}\left[\frac{\exp(^{235}Ut) + 138\exp(^{235}Ut)}{139} - 1\right] \quad \cdots\cdots(2)$$

where W symbolizes the gram-molecular weight of each oxide (W_{Pb}=224, W_{Th}=264, W_U=270). λ is the decay constant of each isotope ($\lambda_{232}=4.9475\times10^{-11}\,yr^{-1}$, $\lambda_{235}=9.8485\times10^{-10}\,yr^{-1}$ and $\lambda_{238}=1.55125\times10^{-10}\,yr^{-1}$ according to Steiger and Jager, 1977). Although the solution of the complex equation for age (t) is given in different forms by each researcher (*e.g.* Bowles 1990: Suzuki and Adachi, 1990), the results are the same.

In younger granites the age is roughly calculated using the equation (1) of Holmes (1931) or, as far as uraninite is concerned, the age may be calculated as PbO×80 (Ma) as discussed later. This shows that the standard deviation of each analytical point depends on the count statistics of Pb.

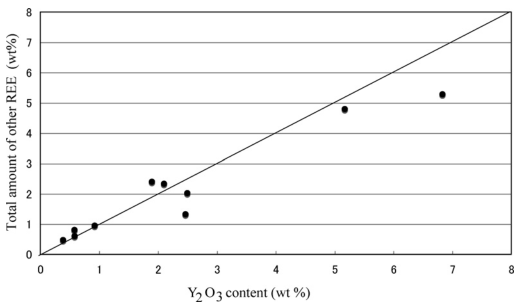

Fig. 3. Correlation of Y_2O_3 content and total other REE content in uraninite and thorite.

d: Zircon age (SHRIMP method)

Zircon grains from the samples and the zircon standard QGNG, with a TIMS U/Pb age of 1850±2 Ma (2σ), were cemented in an epoxy resin and polished until their centers were largely exposed on a flat surface. U–Pb dating was performed using a SHRIMP II installed at Hiroshima University, Japan. Instrumental conditions and measurement procedures are described in Sano et al. (2000). The spot size of the primary ion beam was about 20 μm. Both back-scattered images and cathodoluminescense images were used to select sites for SHRIMP analysis. The $^{206}Pb/^{238}U$ ratios of the samples were calibrated using the empirical relationship described by Claoue-Long et al. (1995). In this procedure, it is essential to subtract initial Pb from measured Pb to estimate the age accurately. The measured $^{206}Pb/^{204}Pb$ ratio was used for the correction of initial Pb, whose isotopic composition was assumed using a single-stage model (Compston et al., 1984).

Results

Data for U, Th and Pb in each analytical point for the age analyses are listed in Table 3. After age was obtained from each point, all the data in the granitic rock were summarized as an average and standard deviation. The standard deviation is usually 1 sigma calculated from a Gaussian distribution in probability theory. All the age data of eleven granitic rocks in the Japanese Islands are summarized in Table 1 as well as the results by SHRIMP analyses.

Thorite was found in most of the granitic rocks in the Japanese Islands. All the samples in Table 1 contained thorite but most of them were altered strongly into hydrous thorite, especially in the old rocks. Hence, thorite age was obtained only from the young granitic rocks. The standard de-

Table 3. UO_2, ThO_2, PbO contents and age for each analytical point in uraninites and thorites in the granitic rocks.

Uraninite in Hida 98112501

No.	UO_2	ThO_2	PbO	age (Ma)	
1	87.88	6.69	2.863	233.7	Average
2	89.11	6.87	2.923	235.2	232.6
3	88.65	6.69	2.943	238.1	error (1σ)
4	87.93	6.49	2.941	239.9	3.9
5	87.79	7.24	2.880	234.8	
6	87.89	6.58	2.934	239.4	
7	88.36	7.08	2.732	221.8	
8	87.00	8.63	2.830	231.8	
9	86.45	7.87	2.879	237.7	
10	89.13	7.54	2.820	226.5	
11	88.34	7.58	2.782	225.4	
12	87.06	8.51	2.820	230.9	
13	87.81	8.58	2.822	229.1	
14	86.89	8.62	2.892	237.0	
15	87.80	8.47	2.922	237.2	
16	84.03	11.70	2.800	234.5	
17	82.64	12.29	2.649	225.2	
18	83.62	11.24	2.770	233.5	
19	85.60	8.67	2.819	234.4	
20	85.54	8.77	2.777	231.1	
21	87.12	7.42	2.871	235.7	
22	83.66	8.98	2.805	238.1	
23	85.68	6.91	2.745	229.6	
24	85.42	7.87	2.720	227.4	
25	85.62	7.49	2.760	230.5	
26	84.65	8.11	2.741	230.9	
27	86.55	8.93	2.820	231.9	
28	82.41	8.07	2.614	226.1	
29	82.98	9.73	2.733	233.3	
30	89.70	6.26	2.893	231.8	
31	87.10	6.72	2.839	233.8	
32	88.58	6.87	2.863	231.8	
33	88.23	6.58	2.903	236.0	
34	88.54	7.04	2.880	233.1	
35	85.49	10.03	2.790	231.2	
36	88.01	6.77	2.873	234.1	
37	88.24	7.41	2.852	231.4	
38	83.67	10.52	2.727	230.4	
39	83.77	10.08	2.757	233.0	
40	87.26	6.41	2.812	231.4	
41	87.72	6.65	2.873	234.9	
42	88.03	8.55	2.883	233.5	
43	86.67	9.06	2.872	235.6	
44	88.26	6.13	2.873	234.0	
45	88.91	7.11	2.833	228.4	
46	88.86	6.96	2.895	233.5	

Uraninite in Hida 98092910

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	76.01	8.62	2.244	209.9	Average
2	80.97	6.46	2.358	209.1	204.4
3	81.90	5.29	2.336	205.8	error
4	78.16	9.83	2.276	206.3	4.3
5	83.02	5.98	2.333	202.5	
6	81.47	7.77	2.340	205.4	
7	82.26	7.02	2.398	209.0	
8	79.36	7.57	2.326	209.5	
9	83.85	6.33	2.348	201.5	
10	86.70	5.39	2.445	203.7	
11	84.44	5.37	2.320	198.5	
12	82.77	6.32	2.424	210.5	
13	84.65	5.56	2.417	206.0	
14	85.97	5.54	2.407	202.1	
15	83.47	5.79	2.332	201.5	
16	84.09	5.37	2.380	204.3	
17	81.94	6.87	2.286	200.3	
18	88.07	4.96	2.347	193.1	
19	86.74	6.18	2.448	203.4	
20	85.84	5.80	2.378	199.9	
21	83.54	5.76	2.381	205.5	
22	85.04	6.07	2.404	203.6	

Uraninite in Hida 98092910

spot No.	UO_2	ThO_2	PbO	age (Ma)
23	83.42	5.19	2.422	209.6

Uraninite in Hida 99042103

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	81.42	11.95	2.267	196.3	Average
2	81.35	11.88	2.288	198.2	198.9
3	80.56	12.01	2.237	195.6	error (1σ)
4	81.38	11.28	2.299	199.5	2.6
5	82.15	10.58	2.338	201.6	
6	82.77	8.27	2.289	197.7	
7	83.55	8.41	2.301	196.8	
8	83.97	8.73	2.380	202.2	
9	82.60	9.07	2.349	202.5	

Uraninite in Hida 98100304

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	86.24	7.38	2.236	186.3	Average
2	85.46	7.86	2.267	190.2	189.6
3	85.14	7.94	2.246	189.1	error (1σ)
4	83.35	8.49	2.256	193.4	4.0
5	84.09	7.97	2.285	194.6	
6	81.67	8.30	2.206	193.0	
7	82.06	8.37	2.084	181.7	
8	83.99	8.56	2.205	187.7	

Thorite in Kitakami SK53

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	14.98	59.76	0.531	116.9	Average
2	15.60	59.97	0.524	113.2	117.0
3	16.29	57.94	0.576	124.0	error (1σ)
4	16.56	59.68	0.565	118.8	6.6
5	16.60	59.66	0.572	120.1	
6	21.79	52.76	0.636	123.0	
7	22.68	54.24	0.593	110.9	
8	27.22	48.38	0.594	104.0	
9	28.09	46.99	0.707	122.3	

Uraninite in Kitakami SK53

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	88.25	7.47	1.392	113.7	Average
2	88.47	7.68	1.424	115.9	115.1
3	88.50	7.54	1.385	112.8	error (1σ)
4	88.75	7.68	1.444	117.1	1.9
5	88.76	8.19	1.404	113.7	
6	90.21	6.90	1.436	115.0	
7	90.48	7.15	1.477	117.8	

Uraninite in Ashio Ashi-2

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	91.92	4.82	1.245	98.7	Average
2	92.37	4.67	1.222	96.5	99.2
3	92.78	4.25	1.235	97.2	error (1σ)
4	92.76	4.25	1.245	98.0	1.7
5	92.66	2.94	1.262	99.9	
6	91.64	2.91	1.258	100.7	
7	91.27	3.74	1.270	101.7	
8	91.25	3.65	1.209	97.0	
9	91.42	4.24	1.242	99.2	
10	91.96	3.88	1.254	99.7	
11	91.62	4.39	1.213	96.6	
12	91.04	4.34	1.234	98.9	
13	90.16	2.44	1.225	99.8	
14	90.30	3.16	1.246	101.0	
15	90.50	2.43	1.223	99.3	
16	92.88	3.57	1.236	97.4	
17	93.86	3.40	1.297	101.2	
18	91.62	4.98	1.212	96.4	
19	91.57	4.77	1.254	99.8	
20	93.27	3.40	1.255	98.6	
21	92.57	3.49	1.257	99.5	

Uraninite in Ashio Ashi-2

spot No.	UO_2	ThO_2	PbO	age (Ma)
22	91.90	3.70	1.264	100.6
23	91.16	4.20	1.221	97.8
24	93.07	3.57	1.266	99.6
25	93.18	3.83	1.236	97.0
26	91.14	3.40	1.230	98.9
27	91.21	2.68	1.239	99.7
28	91.38	3.67	1.250	100.1
29	90.86	2.81	1.258	101.6
30	90.51	4.76	1.241	99.9
31	89.53	5.13	1.272	103.4

Uraninite in Yanai 92072403

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	87.09	9.84	1.127	93.0	Average
2	88.55	9.77	1.148	93.3	92.9
3	88.45	10.11	1.126	91.5	error (1σ)
4	89.82	9.45	1.157	92.9	1.2
5	90.14	7.60	1.191	95.8	
6	92.58	7.68	1.171	91.8	
7	92.13	7.73	1.142	89.9	
8	92.44	6.93	1.172	92.2	
9	90.52	8.35	1.179	94.3	
10	90.82	8.05	1.171	93.4	
11	90.95	7.84	1.150	91.7	
12	92.68	7.21	1.171	91.8	
13	90.84	7.50	1.161	92.8	
14	91.85	7.73	1.160	91.6	
15	92.38	7.86	1.171	91.9	
16	90.98	8.34	1.170	93.0	
17	92.78	7.29	1.172	91.8	
18	88.34	10.12	1.126	91.6	
19	89.89	9.24	1.159	93.0	
20	86.43	10.40	1.137	94.3	
21	89.25	9.52	1.167	94.2	
22	86.96	10.20	1.146	94.6	
23	88.01	10.15	1.147	93.6	
24	87.93	10.06	1.136	92.9	
25	88.28	9.78	1.146	93.4	
26	89.39	9.26	1.158	93.4	
27	88.39	9.24	1.128	92.0	
28	89.81	9.45	1.149	92.2	
29	88.61	9.65	1.138	92.5	
30	87.80	10.59	1.146	93.7	
31	87.02	10.33	1.135	93.6	
32	87.98	10.09	1.146	93.6	
33	86.81	10.35	1.134	93.8	

Uraninite in Ikoma IK2

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	96.11	2.17	1.093	84.1	Average
2	95.73	2.19	1.103	85.2	85.8
3	95.51	2.38	1.103	85.3	error (1σ)
4	94.96	2.63	1.122	87.3	0.8
5	96.20	2.22	1.124	86.4	
6	96.38	2.12	1.124	86.3	
7	96.48	1.89	1.113	85.4	
8	96.56	1.98	1.123	86.1	
9	96.74	2.10	1.123	85.9	

Uraninite in Tsukuba Z4

spot No.	UO_2	ThO_2	PbO	age (Ma)	
10	96.06	1.84	1.114	85.8	
1	87.53	8.50	0.792	65.2	Average
2	84.15	5.17	0.758	65.6	66.5
3	88.82	6.85	0.810	66.2	error (1σ)
4	87.47	6.85	0.798	66.2	1.1
5	86.03	7.81	0.804	67.5	
6	88.14	8.15	0.836	68.5	
7	88.05	8.04	0.813	66.7	
8	87.83	8.08	0.800	65.8	

Table 3. (Continued)

Thorite in Yamanashi S-9

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	10.84	70.43	0.058	13.1	Average
2	17.65	63.55	0.050	10.0	12.1
3	27.87	53.24	0.069	11.6	error (1σ)
4	11.56	69.40	0.052	11.7	1.5
5	17.11	63.72	0.067	13.6	
6	12.12	69.06	0.045	10.1	
7	11.73	69.98	0.052	11.6	
8	10.95	69.68	0.049	11.1	
9	9.61	72.15	0.050	11.6	
10	12.52	68.31	0.048	10.5	
11	27.87	52.93	0.067	11.4	
12	28.03	52.17	0.075	12.6	
13	28.30	52.30	0.070	11.8	
14	8.98	71.92	0.040	9.5	
15	15.43	65.66	0.067	13.9	
16	8.80	72.73	0.047	11.1	
17	16.01	64.78	0.051	10.6	
18	5.38	76.15	0.041	10.4	
19	5.54	74.83	0.054	13.8	
20	27.83	52.42	0.074	12.6	
21	28.87	51.73	0.082	13.7	
22	28.69	51.86	0.089	14.9	
23	13.21	67.65	0.056	12.3	
24	10.89	69.68	0.057	12.9	
25	10.75	69.84	0.060	13.7	
26	10.89	70.05	0.050	11.4	
27	10.98	69.55	0.053	12.2	
28	9.17	71.92	0.064	15.2	
29	20.47	60.15	0.070	13.3	

Thorite in Yamanashi S-6

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	21.83	58.34	0.032	6.0	Average
2	22.75	58.02	0.022	4.1	4.8
3	12.42	69.77	0.036	7.9	error (1σ)
4	15.39	66.84	0.019	3.9	1.1
5	15.70	65.82	0.012	2.4	
6	18.29	63.44	0.019	3.7	
7	9.44	72.89	0.019	4.4	
8	12.37	70.09	0.026	5.7	
9	21.81	57.07	0.038	7.1	
10	23.38	56.77	0.033	6.1	
11	13.47	69.83	0.020	4.2	
12	13.14	69.14	0.012	2.5	
13	11.26	71.48	0.017	3.7	
14	17.40	63.68	0.027	5.4	
15	10.47	71.63	0.024	5.4	
16	14.55	65.29	0.022	4.8	
17	9.78	73.22	0.008	1.9	
18	13.02	69.80	0.027	5.8	
19	12.62	64.87	0.021	4.8	
20	20.94	53.85	0.028	5.5	
21	10.84	72.06	0.021	4.6	
22	10.66	72.27	0.020	4.6	
23	18.38	43.85	0.011	2.7	
24	15.47	66.30	0.025	5.1	
25	12.83	69.63	0.022	4.7	
26	15.79	66.67	0.029	5.9	
27	21.63	59.01	0.029	5.5	
28	22.05	56.18	0.019	3.5	
29	21.00	59.87	0.027	5.1	
30	21.21	57.65	0.031	5.8	
31	16.62	65.54	0.029	5.8	
32	11.88	70.57	0.019	4.2	
33	10.31	72.60	0.010	2.2	
34	15.63	66.63	0.021	4.3	
35	15.90	65.96	0.033	6.8	
36	17.20	65.34	0.021	4.1	
37	13.03	69.54	0.023	5.0	
38	22.72	58.04	0.037	6.8	
39	17.95	64.00	0.023	4.4	
40	16.81	64.49	0.015	3.1	
41	15.11	66.98	0.024	4.9	
42	12.32	64.59	0.015	3.4	
43	23.32	57.37	0.030	5.4	
44	22.75	57.78	0.025	4.6	
45	22.66	57.47	0.027	4.9	
46	20.20	60.94	0.024	4.5	
47	21.14	60.85	0.031	5.8	
48	16.70	63.87	0.033	6.8	
49	14.43	67.85	0.022	4.6	
50	15.83	66.38	0.028	5.7	
51	15.62	66.72	0.031	6.3	
52	13.45	69.52	0.017	3.6	
53	20.52	60.65	0.027	5.0	
56	10.03	73.13	0.024	5.3	
57	17.63	66.01	0.018	3.6	
58	11.53	71.45	0.029	6.4	
59	22.69	58.43	0.021	3.8	
60	14.66	67.86	0.022	4.5	
61	18.17	63.86	0.024	4.7	
62	16.40	65.46	0.033	6.7	
63	16.10	65.74	0.019	3.8	
64	22.81	56.77	0.027	4.9	
65	15.74	66.14	0.025	5.0	
66	16.22	65.98	0.020	4.1	
67	12.49	70.23	0.023	5.1	
68	14.06	68.31	0.031	6.6	
69	14.88	67.92	0.026	5.3	
70	13.16	68.88	0.024	5.1	
71	18.14	64.47	0.022	4.3	
72	12.41	70.40	0.020	4.2	
73	18.70	62.95	0.022	4.2	
74	14.82	67.16	0.020	4.2	
75	12.12	70.04	0.021	4.5	
76	23.06	57.56	0.016	2.9	
77	20.35	60.39	0.029	5.5	
78	23.86	56.35	0.027	4.9	
79	18.73	63.08	0.024	4.6	
80	23.30	58.02	0.022	3.9	
81	9.80	72.49	0.020	4.6	
82	16.77	65.39	0.016	3.3	
83	11.65	71.59	0.022	4.8	
84	12.76	69.78	0.025	5.4	
85	16.62	64.62	0.018	3.7	
86	13.19	69.07	0.030	6.5	
87	11.27	71.35	0.026	5.8	
88	13.90	68.46	0.023	4.9	
89	21.57	59.10	0.027	5.0	
90	14.72	67.04	0.022	4.6	
91	11.94	70.20	0.025	5.4	
92	12.90	69.29	0.016	3.4	
93	12.36	69.93	0.015	3.2	
94	22.28	58.74	0.020	3.7	
95	24.41	56.60	0.036	6.3	
96	12.56	69.76	0.025	5.4	
97	15.92	66.33	0.029	5.9	

Uraninite in Yamanashi S-6

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	83.28	8.21	0.066	5.7	Average
2	83.84	9.30	0.061	5.3	5.2
3	85.16	7.90	0.061	5.2	error (1σ)
4	85.69	9.57	0.062	5.2	0.7
5	86.25	9.21	0.070	5.9	
6	90.18	9.81	0.050	4.0	

Uraninite in Hida T-2

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	81.56	7.13	0.029	2.6	Average
2	84.56	7.34	0.032	2.7	2.4
3	81.89	7.06	0.027	2.4	error (1σ)
4	80.88	10.54	0.017	1.5	0.6
5	84.58	7.33	0.040	3.5	
6	82.97	7.49	0.034	3.0	
7	81.23	7.63	0.040	3.6	
8	83.72	7.13	0.026	2.3	
9	84.76	6.60	0.024	2.1	
10	82.20	9.68	0.035	3.1	
11	82.68	10.11	0.029	2.5	
12	82.48	9.84	0.020	1.8	
13	83.46	8.19	0.030	2.6	
14	83.93	7.17	0.022	1.9	
15	84.12	7.54	0.023	2.0	
16	81.15	7.41	0.026	2.3	
17	80.66	8.41	0.021	1.9	
18	83.21	6.72	0.024	2.1	
19	82.96	7.14	0.026	2.3	
20	82.81	7.31	0.015	1.3	
21	80.34	10.47	0.029	2.6	
22	80.45	8.69	0.032	2.9	
23	80.22	10.18	0.015	1.3	
24	79.97	9.36	0.028	2.6	
25	83.49	6.57	0.038	3.3	
26	82.96	5.31	0.033	2.9	
27	82.39	7.71	0.027	2.4	

Thorite in Hida T-2

spot No.	UO_2	ThO_2	PbO	age (Ma)	
1	11.55	63.24	0.015	3.6	Average
2	14.91	62.00	0.005	1.0	3.4
3	16.12	62.50	0.013	2.7	error (1σ)
4	15.34	57.16	0.015	3.3	1.1
5	13.73	63.36	0.016	3.5	
6	21.01	53.58	0.019	3.9	
7	16.07	60.75	0.014	3.1	
8	15.28	61.93	0.021	4.5	
9	77.73	7.02	0.034	3.2	
10	18.73	60.40	0.014	2.7	
11	12.80	67.40	0.009	2.1	
12	26.26	51.00	0.017	3.1	
13	15.86	60.43	0.018	3.9	
14	13.41	65.32	0.020	4.3	
15	11.55	64.82	0.025	5.9	
16	20.71	56.36	0.017	3.4	
17	17.12	59.57	0.015	3.1	
18	18.28	58.67	0.015	3.1	
19	18.00	60.65	0.021	4.3	
20	15.67	57.43	0.023	5.2	
21	20.76	55.06	0.014	2.7	
22	15.86	61.19	0.009	2.0	
23	23.34	53.84	0.025	4.7	
24	20.18	57.47	0.009	1.7	
25	11.63	63.03	0.015	3.6	

viation for uraninite was much lower than that of thorite, simply due to the very high U content which produced radiogenic Pb. Hurtado et al. (2007) obtained 0.04 Ma as 2σ for 18 Ma uraninite. Numerically, the error depends on the probe current, measurement time and number of analyses. In addition, error may depend on machine drift, standard material conditions and surface conditions. In the present study, the standard deviations of uraninites were found to be around 4 Ma at around 200 Ma, 1 Ma at 66 Ma and less than 1 Ma at 2.4 Ma and 5.2 Ma, comparable to those obtained by the isotopic methods.

Correlation of the EPMA and isotopic ages is presented in Fig. 4, showing a concordant relationship between them. The coincidence of the ages is evidence that EPMA age is comparable to those yielded by the isotopic methods. Using the same machine in the National Museum of Nature and Science and the same analytical conditions,

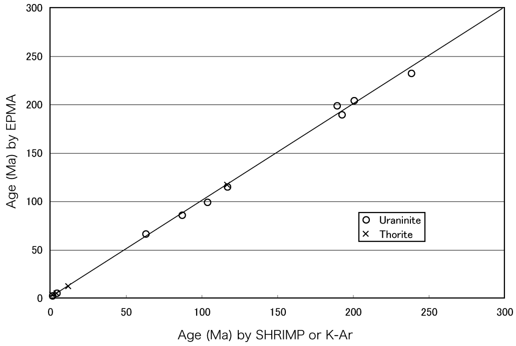

Fig. 4. Correlation of ages obtained by EPMA and isotopic methods.

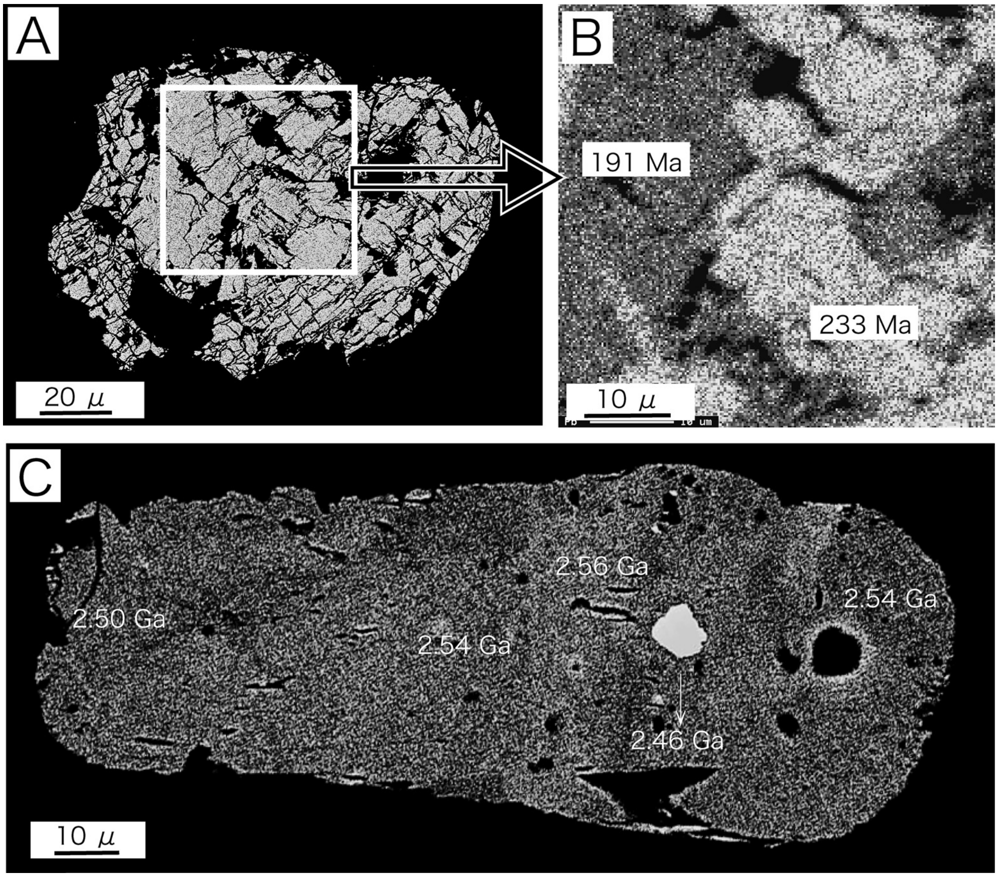

Fig. 5. A & B: Partly recrystallized uraninite in the grey granite from the Hida Terrane (KM1331-1). C: the oldest uraninite, 2.46 Ga, included in a monazite grain from the Yellow River.

such a concordant relationship had previously been reported for uraninite and huttonite in India (Santosh *et al*. 2003) and thorianite (ThO_2) in Bohemian Massif (Naemura *et al*. 2008). Huttonite is a monoclinic polymorph of thorite. In India, uraninite and huttonite grains in gneiss, granulite and granite are zoned in the same range from 460 to 580 Ma as the coexisting monazite. In Bohemian Massif, the EPMA age of thorianite in peridotite was 333.8±4.5 Ma, quite comparable with the values of 338±1 Ma and 339.8±2.6 Ma obtained by isotopic methods for the associated granulites. As a rare case, we found uraninite included in monazite grain from the river sands (Fig. 5). The uraninite age was determined to be 2.46 Ga, slightly lower than the monazite age from 2.5 to 2.56 Ga.

During study of the other granitic rocks in the Japanese Islands, bimodal age distributions of uraninite have been occasionally found. Age data of grey granite from the Hida Terrane are shown in Fig. 5 and 6. One uraninite grain exhibited ages of 191±1.8 Ma and 233±3.4 Ma. The zoning texture of this sample has not been recognized, but high and low Pb areas are distributed in a patchy fashion. Any micro-scale diffusion between these areas has not been observed. The older age is similar to that of coexisting monazite age at 237±6.8 Ma (Fig. 6).

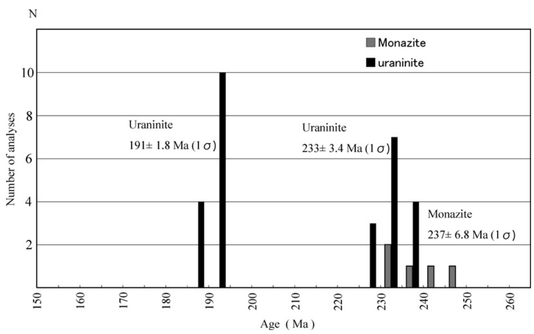

Fig. 6. Bimodal age distribution of uraninite in the grey granite from the Hida Terrane (KM1331-1). Associated monazite is similar in age to the older uraninite.

Discussion

Age data obtained by EPMA analyses are consistent with those of the isotopic methods. Standard deviation of the derived ages is around 1Ma in the range from 1.5 to 100 Ma, comparable with that of SHRIMP dating. The EPMA system is the most commonly used machine for mineral analyses and has high spatial resolution, i.e. 1–2 μ, compared with 10–20 μ of SHRIMP. Although uraninite and thorite are trace minerals and occur in selected rocks, it is more preferable to use young granite with uraninite or thorite. The coincidence also supports the assumption that the initial Pb content is negligible in the minerals at the time of the mineral crystallization, as stated first by Holmes (1911). As reported in previous studies using the same machine, this method is also applicable to thorianite and huttonite (Naemura *et al*., 2007: Santosh *et al*., 2003).

Granitic rocks have a thermal history, i.e. cooling or overprinting by later stage granite. Hence, the age result cannot be accepted simply as the representative age of the igneous stage. Since in general the closure temperature of zircon is around 900°C, it has been considered that the SHRIMP age of zircon in granitic rock shows the age of an early plutonic event. Monazite has also high closure temperature, i.e. around 800°C. On the other hand, there is no experimental datum about the closure temperature of uraninite and thorite. In the grey granite from the Hida Terrane (Fig. 6), uraninite has bimodal age distribution and monazite is consistent with the older of the two ages. These facts apparently show that uraninite is lower in closure temperature than monazite. However, uraninite grains in gneiss and granulite from India are similar in range to the associated monazite (Santosh *et al*., 2003). Although a few problems including closure temperature and meaning of the obtained age have not solved, the age data by EPMA are reliable.

Many researchers have reported the ages of minerals such as monazite and uraninite analyzed using EPMA, assuming that their initial Pb contents were negligible. It is amazing that the present method is essentially the same as that developed first by Holmes (1911). The ages obtained by the simplified calculation of Holmes (1931) are similar for thorite and higher for uraninite than those given by the present complex calculation (Fig. 7). The difference is mainly due to the effect of ^{235}U. In 1931, ^{235}U content in total U was estimated to be less than 0.28%, far less than the present accepted value, 0.72%. In uraninite, a more simplified equation from equation (1) is

Age (Ma) $=80\times$PbO (wt%)

This is only applicable to young uraninite (Fig. 8). This simplified equation is also similar to that presented by Holmes (1911), i.e. age $=8200\times$ Pb/U (Ma) where Pb and U are wt%. This simple equation means that the standard deviation is totally dependent on the error of Pb analyses. In our normal analytical conditions for uraninite and thorite, the standard deviation is around 1 Ma for the samples dating from less than 100 Ma. High probe current and measurement time will reduce the error.

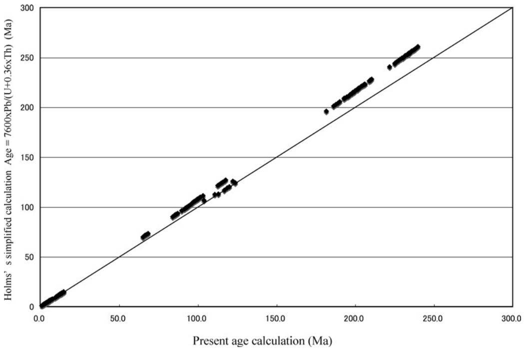

Fig. 7. Relationships between present age calculation and simplified age of Holmes (1931) for uraninite and thorite. Pb, U, and Th as wt%.

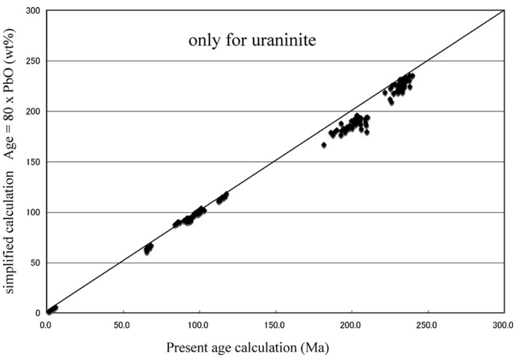

Fig. 8. Relationships between present age calculation and simplified age, $80\times$PbO, for uraninite. PbO as wt%.

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