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The Relation between the Minute Content of Uranium in Zircon and its Mineralogical Characters

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Abstract

The uranium contents of representative zircon specimens collected from the weathered granite sands of the Tensei Mine were analysed by the microfluorimetric method. To eliminate the quenching effect of fluorescence intensity derived from the bulk zirconium, the actual contents were calibrated by multiplying the apparent values determined by the direct method by the coefficient 1.31.

The radioactivity of zircon was measured by the decathlon type countmeter, and it was generally proportional to the uranium content.

The natural fluorescence intensity of zircon activated by the ultraviolet ray was determined by means of the sensitive photomultiplier detector, and the fluorescence intensity of zircon containing a comparatively large amount of uranium was lower than that containing smaller amount.

An attempt to find some relation between the colour and uranium content of zircon was fruitless except for the fact that the colourless zircon was relatively low in its radioactivity, but the greenish brown member was moderately high.

The lattice defect of zircon was also investigated with the Geiger counter diffractometer and related to the uranium content.

Various minor elements contained in zircon were semi-quantitatively determined by the spectrographic method.

From the above experimental results, the intrusion of granite batholith was classified into three following stages, first the stage on which the first intrusion of granite represents the northern part of this district and the zircon content in granite is rich and the uranium content in zircon is small, and secondly the stage on which the second intrusion of granite represents the southern part and the zircon content is small and the uranium content is slightly richer than that of the granite found in the northern part, and finally the stage on which the third intrusion of granite represents the middle part and the zircon content is of moderate amount but the uranium content ranges from a small to a large amount.

Introduction

Zircon is one of the most common and familiar minerals which are distributed widely in various rocks. Therefore the mineralogical characters of zircon have attracted the interest of geologists, petrologists and mineralogists and have been

This rock consists mainly of quartz, albite, microcline and biotite, bearing amphibole, zircon, magnetite, ilmenite, anatase and allanite as the accessory minerals. There are found numerous dykes of lamprophyre in this region which strike north to south under the control of geological structure. Some zircons were also found accompanied by a large amount of anatase in the mineralized zone which was intimately related to the above lamprophyre dyke.

The rock mass collected from the appointed places of the geological and sampling map was ground in a crusher to 60 mesh and the heavy minerals were separated from this crushed sands by means of the panning and the heavy Toulet-solution. The above operations were repeated several times to reduce the loss of heavy mineral. The heavy minerals were next separated roughly into several fractions with the magnetic separator. The highest magnetic fraction obtained with a strong horseshoe magnet consisted chiefly of magnetite, and in the higher magnetic field of separator, ilmenite, biotite, hornblende, allanite and monazite were separated from the non-magnetic fraction which consisted of zircon and anatase. The crystal size of anatase was always larger than that of zircon, so they could be easily separated from each other by sieving through 60 mesh sieve.

Fluorimeter

In any fluorimeter which has been reported, there was no remarkable^{5,6)} difference in the principle. It was constructed with essential four parts, that is, emission part of ultraviolet ray, fluorescence detector, measuring device of the out-put current of detector and electric power source. Fig. 2 is the schematic fig. of fluorimeter, the upper part is the exciting ultraviolet source. The main body is the light-tight compartment into which the ultra-violet ray falls through the primary filter. The fluorescence light emitted from the uranium fluoride cake which is put on the bottom of compartment is introduced into the detector tube by which the light is converted into electric current through the secondary filter which is often combined with some other filters for the special requirements.

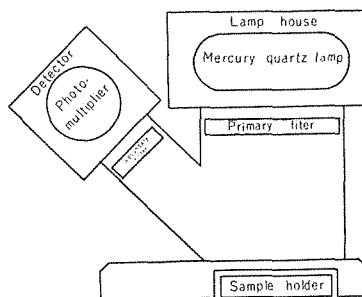


Fig. 2 Fluorimeter set

In this fluorimeter, photomultiplier was prepared as the detector tube because of its high sensibility, and the authors adopted the MS-6S tube (Matzuda). To supply the sufficient stable current to detector tube, an additional voltage stabilizer was provided, which is shown in Fig. 3, because the out-put current of this tube was proportional to the sixth power of dynode voltage.

The out-put current from the detector tube was measured by the measuring device. The circuit of measuring device, which was a kind of bridge type D.C. amplifier, is shown in Fig. 4. In this circuit, as the more stabilized supply current was required at the first stage amplifier, the extra voltage stabilizer was prepared as is shown in Fig. 5.

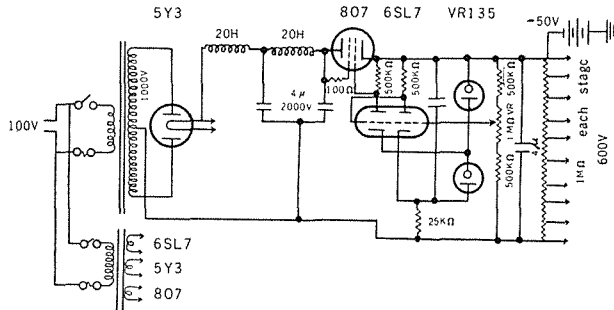


Fig. 3 Voltage stabilizer for detector

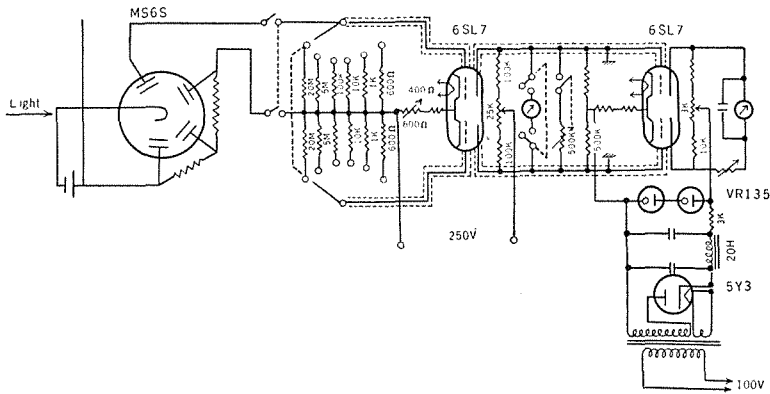


Fig. 4 Circuit of measuring device

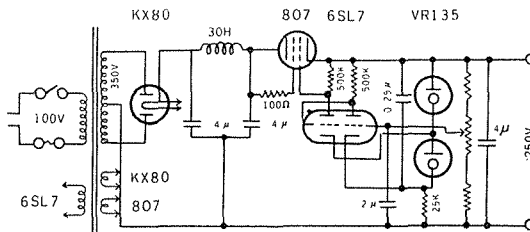


Fig. 5 Voltage stabilizer for measuring device

Procedure of fluorimetric analysis of uranium

The procedure of this method is classified into two methods, the indirect method when uranium is preliminarily extracted from the sample, and the direct^{7,8)} when not. By the latter method, the procedure is very simple, while the former obtains the more exact results in principle than the latter, excepting the errors derived from the relatively complex treatment in the chemical separation of uranium from the other component elements. However, even in the case of latter method, the considerably good results may be obtained if the quantity of the sample is restricted adequately to reduce the quenching effect of coexistent elements to a minimum.

In this paper, the direct method was applied and the quenching effects of Zr, Ce and Th to uranium were examined closely to obtain the accurate determination of uranium amount in zircon.

(a) Standard curve

To obtain the standard curve, two series of standard cakes which respectively contain the following amounts of uranium in 3 g. flux were prepared.

Table 1.

A-series		B-series	
added U in mg	scale reading	added U in mg	scale reading
0.00055	22.0	0.0055	11.0
0.00110	21.5	0.0110	22.5
0.00165	29.5	0.0165	29.5
0.00220	35.5	0.0220	41.0
0.00275	45.5	0.0275	50.5
0.00330	49.5	0.0330	53.0
0.00385	57.0	0.0385	62.0

Each cake was prepared by adding the calculated amount of standard uranium sulphate solution corresponding to the above described uranium amount. Thus the standard curves relating between uranium content and scale reading of each series are shown in Fig. 6 and 7.

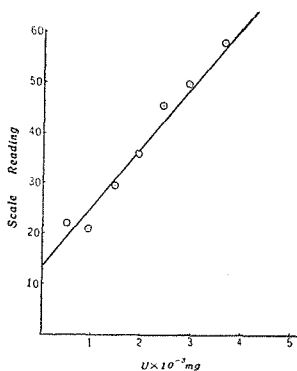


Fig. 6. Standard curve of A series

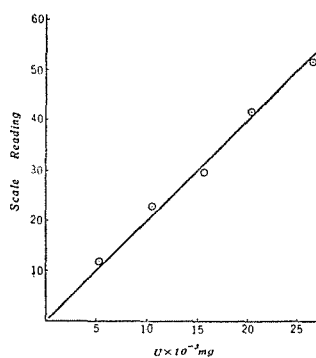


Fig. 7. Standard curve of B series

(b) Sensibility of fluorimeter

The sensibility of the fluorimeter is variable by changing the dynode voltage of photomultiplier or load register of grid of the bridge. Besides, the sensibility is also variable by interposing the filter in the optical pass. However, the high sensibility, by changing the register or dynode voltage disturbs the stability of the out-put current of photomultiplier. In the case of the most favorable sensibility, one scale division is equivalent to 0.00014 mg. of U, ie 0.05%. In this condition, the observational error is about one scale division, that is, 0.00014 mg. U.

(c) Quenching effect of coexisting elements in zircon

It is well known that when the direct method is applied there are many quenchers of coexistent elements which reduce the fluorescence intensity and the quenching degree depends only on the concentration of the quencher in the flux and not on the ratio of concentration of quencher to that of uranium. Based on this account, by using the sufficiently small amounts of samples, it is possible to reduce the quenching effect to a negligible low degree.

However, in the case of zircon, zirconium is the bulk component and we can not neglect the quenching effect of zirconium itself, even if zirconium is a relatively mild quencher. Furthermore, the effects of Ce and Th were also examined.

(1) Effect of a definite amount of Zr to various contents of U

Quenching effect of a definite amount of Zr (1.48 mg.) to the fluorescence intensity emitted from the various concentration cake of U were tested. Each sample contains following amounts of U and additional Zr amounting to 1.48 mg.

Table 2. Quenching effect of Zr to various U contents.

added U in mg	apparent U in mg	coefficient
0.1100	0.0880	1.25
0.0550	—	—
0.0110	0.0083	1.32
0.0055	0.0042	1.31
0.0028	0.0021	1.31
0.0011	0.00083	1.32

mean 1.31

From these experimental results, it was ascertained that the average value of coefficient was 1.31 and each value did not deviate so much from the above value.

(2) Effects of various amounts of Zr, Ce and Th to a definite amount of U

Each cake was prepared by evaporating the standard sulphate solutions of Zr, Ce and Th corresponding to those amounts in Table 3 and adding it to the flux which contains 0.0275 mg. of U.

As is indicated in Fig. 8, the quenching effects to the fluorescence intensity increased exponentially with the increase of these elements and Th-effect was most

Table 3. Quenching effects of Zr, Ce and Th.

added Zr in mg.	intensity	added Ce in mg.	intensity	added Th in mg.	intensity
0.0222	48.8	0.0205	47.5	0.0277	47.0
0.0444	46.7	0.0409	45.2	0.0555	41.5
0.0740	41.6	0.0382	42.5	0.0925	36.0
0.1480	—	0.1365	38.5	0.1850	30.7
0.3700	33.8	0.3410	27.5	0.4625	20.0
0.7400	33.6	0.6820	26.5	0.9250	12.0
1.1100	31.0	1.0230	21.2	1.3875	10.0
1.4800	30.6	1.3650	21.3	1.8500	6.9

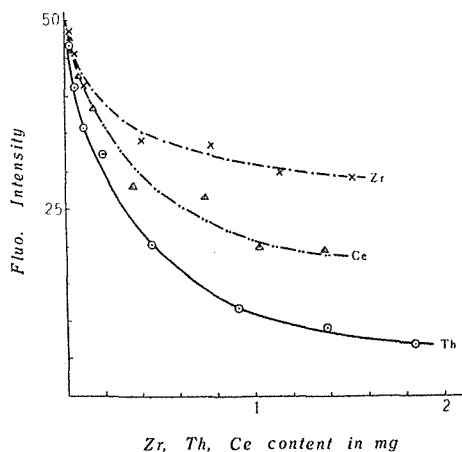


Fig. 8. Zr, Th and Ce quenching effect

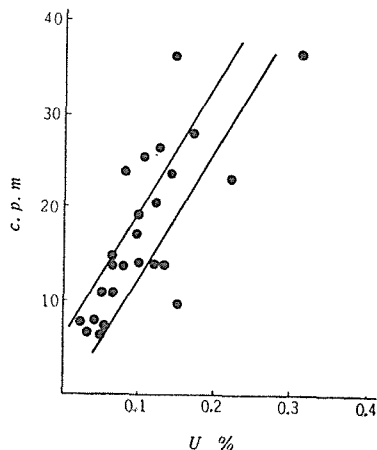


Fig. 9 Relation between the C.P.M. and U-content

intense. Therefore in the case of malacon, the effects of these elements must be considered but in the case of zircon from granite, Zr-effect only must be taken into account.

The radioactive^{9,10,11} intensity and uranium content of zircon

The uranium contents of 28 representative zircons were determined by micro-fluorimetric method. The actual uranium content was calibrated by multiplying the apparent value determined by direct method by the coefficient 1.31.

The radioactive intensity was measured strictly in the sealed chamber protected with lead tube by the decahlon type counter for 10 minutes. The measured counts were converted into the counts per 0.1 g. sample per minute deducting the natural count. Fig. 9 shows a general proportionality between the radioactivity and uranium content. The cause of broad distribution of dots along the linear band in this figure may be attributed to the following facts ;

Table 4. Summary of experimental results.

sample No.	zircon content in 0.1%	C.P.M.	U content in zircon 0.1%	natural fluorescence	color	sample No.	zircon content in 0.1%	C.P.M.	U content in zircon 0.1%	natural fluorescence	color
1-1	0.077					6	0.082	14	0.60		R
2	0.077	8	0.97		R	7	0.045			5.9	
3	0.226	24	1.38	6.5	G1	8	0.096	11		1.6	W
4	0.178			6.5		9	0.088	27		7.7	G
5	rock					10	0.140	36	3.18		G
6	0.357	18		6.2	R1	11	0.240			3.2	
7	rock					12	0.150			2.7	
8	0.270	14	1.31	4.8	R1	13	0.060	27	1.25		R
9	0.180		0.45		G	14	0.127	11	0.50	4.5	W
10	rock					15	0.165				
11	0.093			3.6		16	0.075			6.0	
12	0.135			4.9		17	0.119	13	0.86		W
13	rock					18	0.130			3.3	
14	0.084	14	1.17	3.0	R	19	rock				
15	rock					20	0.140	23	2.18	8.5	G
16	rock					21	0.198	7	0.48	5.0	W
17	0.107	26	1.01		R	22	rock				
18	0.373			5.3		23	0.255	7	0.24	4.0	R1
19	rock					24	0.110	6	0.51		W
20	0.160					25	0.135			5.2	
21	0.122	29	1.90	5.2	G	26	rock				
22	rock					27	0.217	8	0.37		G
23	0.130	22		4.0	G1	28	0.094			8.2	
24	rock					29	rock				
25	0.160	20	1.16	4.1	R1	30	rock				
26	rock					31	0.204	24	0.77	3.0	
27	1.353	15	0.58	6.2	G	32	0.557	38	1.45	0.4	R
28	0.193			3.1		34	0.231	10	1.44	3.3	R1
29	1.374			1.6		35	0.121	17	0.98	2.0	R
30	0.601			1.8		36	0.035			3.7	
31	0.223					37	0.178	14	0.66	4.5	G
32	0.090	38	1.13	2.2		38	rock				
33	0.104			3.0		39	0.304			6.7	
34	0.715			2.2		40	rock				
35	0.051			2.0		41	rock				
2-1	0.130			11.2		42	0.157			6.1	
2	0.242			9.6		43	0.179	19	0.98	4.5	R1
3	0.218	11	0.54	3.3	R1	44	0.237				
4	0.141			8.0		45	0.123	14	0.94	5.0	R1
5	rock					46	0.111			4.4	

R : reddish brown G : greenish brown
R1 : R member G1 : G member

- (A) for the radioactivity
 - a) effect of thorium impurity
 - b) grain size and amount of sample
 - c) abundance of contaminated substance
- (B) for the fluorimetric analysis
 - a) quenching effect of coexistent elements
 - b) unstability of exciting ultra-violet ray intensity
 - c) melting condition of cakes

Superposition of the above mentioned and other disturbing effects may introduce some irregular distribution of dots.

The uranium content and fluorescence intensity of zircon

There are found numerous minerals¹²⁾ which show the fluorescence emission by ultra-violet ray excitation and zircon is one of them. The authors investigated that the fluorescence intensity of zircon including more uranium was inferior than that of less uraniferous zircon and the zircon which contained uranium more than 2% showed almost no fluorescence emission. This relationship was shown in Fig. 10.

These facts may be attributed to the lattice defect of zircon, caused by the metamictization of radioactive elements such as uranium. As is shown in Fig. 10, the general tendency which indicated the above relation was observed, but somewhat broad distribution of dots along the inversed proportional line was inevitable for the following reasons. If the intensity of fluorescence depends upon the degree of metamictization, the crystal defect may depend not only upon the content of radioactive element but also upon the time elapse of radiogenic damage. Other reasons of this irregularity may be attributed to the different absorption degree of fluorescence by the different colour of zircon or the contamination of other impurities.

The colour¹³⁾ and uranium content of zircon

Zircons show various colors according to the discrepancy of their localities and occurrences. It has long been accepted that the variety of colour is due to the minor impure elements such as Fe, Cu, Cr and etc. However since it has been ascertained that the coloured zircon loses its colour only by heating and is recoloured by radiogenic irradiation, it has been accepted that the colouration should be ascribed to the radiogenic colouration¹⁴⁾ which makes the crystal defects into colour centers. From the above facts, we can expect some relationships between the colour and radioactivity or uranium content. In this experiment, an attempt to find any relations between the colour and uranium content brought nothing excepting the fact that the colourless zircon showed generally low radioactivity. Respecting the localities of zircons, as will be discussed later, there were found some relationships

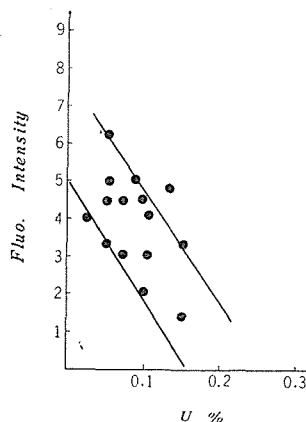


Fig. 10 Relation between the fluorescence intensity and U-content

that the greenish colour zircons had high radioactivity, colourless zircons had low radioactivity and reddish brown zircons had moderate intensity.

The crystal^{15,16,17)} defect and uranium content of zircon

Generally some minerals which have any radioactive elements undergo more or less crystal damage owing to the radiogenic irradiation, therefore it is impossible

Table 5.

index	(213) (400)	(312)	(220)	(112)	(200 α)	(200 β)	(101)	(321)	U 0.1%
sample No.									
2-23	12.4	20.2	15.5	16.0	33.6	9.8	17.6	9.2	0.24
2-21	21.9	30.0	25.0	26.0	47.5	26.0	26.3	11.3	0.48
2-37	32.0	41.3	17.0	28.1	94.8	15.6	48.0	16.1	0.66
2-17	26.0	30.4	22.0	31.3	58.7	8.3	30.4	21.8	0.86
1-8	25.9	31.6	20.6	32.6	63.0	13.7	27.0	15.2	1.31
2-31	19.5	20.4	15.0	16.3	78.4	32.7	28.0	11.5	1.60
2-9	14.3	26.3	19.3	35.9	50.7	8.2	24.4	10.4	1.75

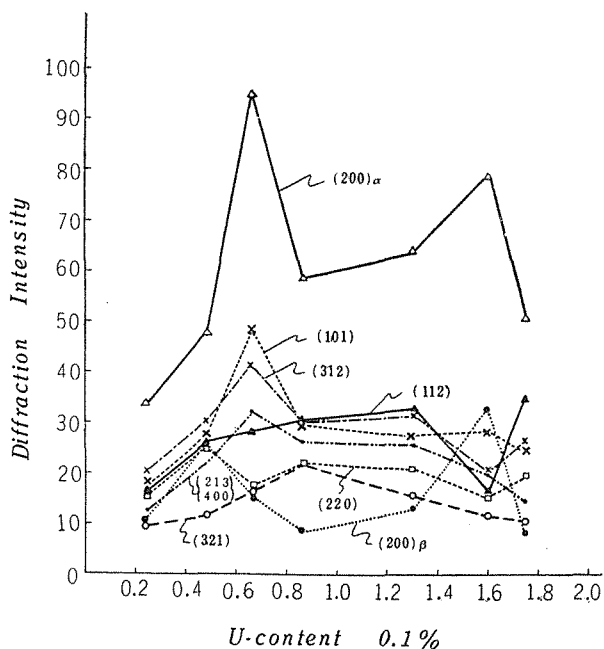


Fig. 11. Relation between the diffraction intensity and U-content

to analyse the crystal structure by the X-ray diffraction method. To investigate these relations, the relative intensities of diffraction patterns of some principal gitter planes were investigated by means of X-ray geiger counter spectrometer. The so-called malacon which contained a few % of uranium showed almost no diffraction pattern, but members from this locality contained very small amount of uranium ranging from 0.02 to 0.20% and had high crystallinity. The relation between the crystallinity and uranium content is shown in Table 5 and Fig. 11.

The members which contained above 0.066% of uranium decreased the diffraction intensity of each gitter plane

according to the amount of uranium but the others which contained below 0.066% of uranium also weakened their intensities. The relative intensities were recalculated by inserting the quartz powder.

The above experimental results show that the increase of uranium content promotes the metamictization, but there remain certain problems on the behaviour of remarkably low uraniferous zircon less than 0.066%.

The uranium and hafnium¹⁸⁾ content

The semi-quantitative ratio of hafnium to zirconium was measured by the spectrographic method. This experiment was indebted to the workers of the Nagoya Industrial and Engineering Institute. As the number of experiment was not so many, the conclusion derived from these results was not quite sure. The direct relationship between the content of Hf and U was not recognized but by classifying these samples into three regions: northern part, southern part and middle part, it was clarified that the higher ratio of Hf to Zr corresponds to higher content of uranium in each region.

Table 6. Hf-content.

sample No.	Hf %
1-9	1.12
2-27	1.75
2-13	1.85
2-6	1.10
2-24	2.25
2-21	1.80

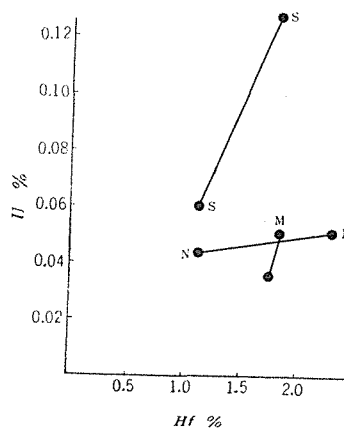


Fig. 12. Relation between U- and Hf-content, N-Northern part, S-Southern part and M-Middle part.

The minor elements in zircon

Thirty-three minor elements in zircon were semi-quantitatively analysed by the spectrographic method under the following conditions, primary voltage 35 volt, 3 amp., secondary voltage 220 volt, 7 amp., intermittent charge speed 1r/min., intermittent ratio 1/20, exposed 3 min. The content of each element was determined by referring to more than three ultimate lines. The results were tabulated in Table 7.

Table 7. Minor elements in Zircon.

sample	Pb	Mg	Ca	Si	Al	Mn	Ni	Zn	Cu	V	Be	Cb	Sn	Ti	Yt	Tl		
Kotei	±	≡	+	≡	+	+	+	≡	±	+	±	+	+	≡	+	±		
Hagata	≡	≡	+	≡	+	+	+	≡	±	±	—	≡	≡	≡	+	≡		
Henderson	≡	≡	+	≡	+	≡	+	+	+	±	—	≡	≡	+	≡	±		
Australia	≡	≡	+	≡	+	+	±	+	+	±	—	≡	≡	+	+	±		
Malay	±	≡	+	≡	+	≡	+	≡	+	+	—	+	≡	+	+	≡		
Zircon of the Tensei mine																		
G(109)	≡	≡	+	≡	≡	≡	+	≡	≡	+	+	±	≡	≡	+	+		
G(2027)	≡	≡	+	≡	≡	≡	+	≡	≡	+	±	—	≡	≡	+	+		
R(20B)	+	≡	+	≡	≡	+	+	≡	≡	+	+	±	≡	≡	±	±		
R(206)	+	≡	+	≡	≡	+	+	≡	+	+	±	±	≡	≡	±	±		
W(2024)	+	≡	+	≡	≡	+	+	≡	≡	+	±	±	≡	≡	+	+		
W(2021)	+	≡	+	≡	≡	+	+	≡	≡	+	±	+	≡	≡	+	+		
sample	Os	Yb	Ta	Rh	B	Co	Sr	Th	Ru	U	Nd	Ce	W	Sc	Lu	Cr	As	Fe
Kotei	—	+	≡	≡	≡	—	—	≡	+	+	+	≡	+	+	—	+	±	≡
Hagata	+	+	≡	≡	+	+	—	+	≡	+	+	+	±	+	+	—	≡	≡
Henderson	+	+	≡	≡	≡	—	—	+	≡	+	±	≡	±	+	±	—	±	≡
Australia	+	±	≡	≡	+	—	—	+	+	±	—	≡	—	+	—	—	±	≡
Malay	—	+	≡	≡	+	—	—	+	≡	+	—	≡	+	+	+	+	±	≡
G(109)	+	+	≡	≡	+	+	—	+	≡	+	+	≡	+	+	+	≡	+	≡
G(2027)	+	+	≡	≡	+	—	+	+	≡	+	+	≡	±	+	+	±	≡	≡
R(20B)	+	+	≡	≡	+	—	+	+	≡	+	±	≡	+	+	—	±	—	≡
R(206)	+	+	≡	≡	+	—	+	+	≡	±	±	≡	+	+	—	+	—	≡
W(2024)	+	+	≡	≡	±	—	+	+	≡	±	±	≡	±	+	—	+	—	≡
W(2021)	+	+	≡	≡	±	—	+	+	≡	±	±	≡	±	+	—	+	—	≡

relative intensity of ultimate spectral line ≡ > ≡ > ≡ > +, ± trace, — none

From the above experiments, following items may be summarized on the behaviour of minor elements in zircon,

(A) elements contained in nearly equal amounts in zircons

a) in large amount

Mg(≡), Al(≡), Zn(≡), Cu(≡), Ti(≡)

Ta(≡), Rh(≡), Ce(≡ or ≡), Fe(≡)

b) in small amount

Pb(≡ or +), Ca(+), Ni(+), V(+), Os(+), Yb(+), Th(+), Sc(+), Sr(+)

(B) elements contained in various amounts in each zircon

Pb(≡, +), Mn(≡, +), Sn(≡, ≡, ≡), Yt(≡, ±), Ru(≡, ≡, ≡), U(+, ±), Nd(+, ±), Ce(≡, ≡), W(+, ±), Cr(≡, +, ±), As(≡, +, —), Lu(+, —), B(+, ±), Tl(+, ±), Be(+, ±)

(C) elements not contained or very rare

Cb(+, ±), Co(+, -), Lu(+, -), As(±, +, -)

In the case of (B), greenish brown coloured zircons contained impurity in large amount and reddish brown members were next to the above zircons with some exceptions of Ce, W, Ru and the impurity of non-coloured zircon was relatively little.

As was stated before, zircons under investigation were selected from the fine powdered rocks, therefore the contamination of anatase, ilmenite, allanite and silicate minerals was unavoidable and it should not be expected that the above elements were included in zircon. Nevertheless, comparing these impurities with those of the pure single crystal from foreign locality, they had much in common with each other in the variety and amount of minor elements. The contents of Pb, Cb, Lu, As of zircons from this locality were smaller than those of other localities and on the contrary, the contents of Al, Zn, Cu, Be, Ti, Sr, Ru, Cr and etc. were larger than those of others; of these Al and Ti may be attributed to the contamination of the above described minerals. It is an interesting fact that there were intimate similarity with those of Korea and Malay which occurred under same geological conditions.

The uranium content in zircon and zircon content in weathered granite

This relation was plotted in Fig. 13, in which the distribution of plotted points may be classified into three regions as described in the foregoing paragraph.

a) Northern region: the samples represented the northern part of mine showed

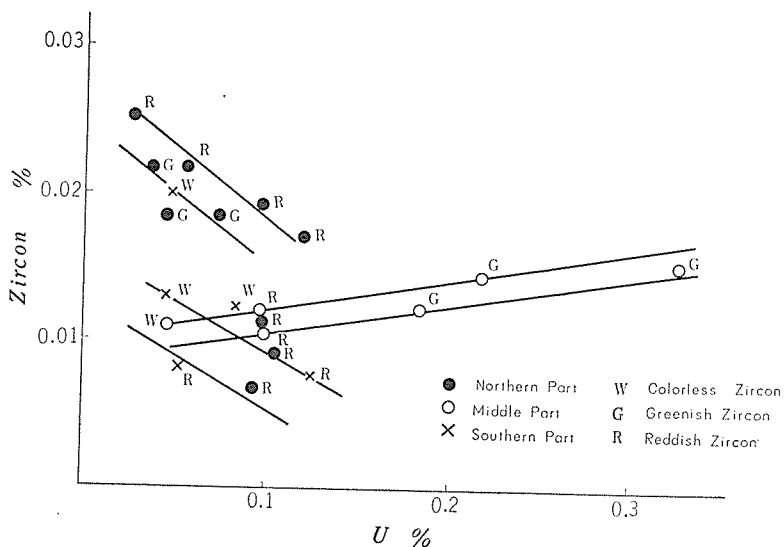


Fig. 13 Regional characters of zircons

generally the higher zircon content and lower uranium content, and a decreasing tendency of the zircon content with increasing of uranium content. In colour, these samples belonged to the reddish brown to greenish red group.

- b) Southern region: the samples represented the southern part of mine showed generally the lower zircon content and somewhat higher uranium content, and in colour they belonged to the colourless or reddish brown group.
- c) Middle region: the samples represented the middle part of mine showed the intermediate zircon content, but the uranium contents were distributed in wide range from lower to the highest content. The colours of these samples varied from colourless to greenish members with increasing of the uranium contents.

Evolution of granitic magma and mineralogical character of zircon

From the mineralogical investigation of uraniferous zircon, the evolution process of granitic magma could be divided into three consolidation stages. Accepting the general principle that uranium and other minor elements were enriched in the later stage of magmatic evolution, it should be concluded that the northern granite consolidated in the earlier stage, the southern granite succeeded to the northern granite and that of the middle region consolidated in the latest stage.

Many tectonic fissures striking to N-S direction were accompanied in this district by the last intrusion of granite and numerous lamprophyre dykes intruding along these geotectonic fissures are attended with the mineralizations of anatase and zircon. The amount of heavy mineral in these mineralized zones was relatively large, but anatase predominated. The zircon was greenish coloured and had high radioactivity.

Summary

- (1) The authors constructed a sensitive fluorimeter by using a photomultiplier tube as the fluorescence detector. The sensibility of this fluorimeter could be varied over a wide range and under the most sensitive condition, one scale division corresponded to 0.00014 mg. of uranium, i.e. 0.05 γ .
- (2) This fluorimeter was applied for the microanalysis of uranium in zircon. In this case, the quenching effects of Zr, Ce and Th to the fluorescence intensity of uranium fluoride cake were respectively investigated. It was clarified that the quenching effects increased exponentially with the increase of these elements and Th effect was most intense.
- (3) Thus the uranium contents of zircons collected from the weathered granite of the Tensei Mine in Osaka Prefecture were analysed. The actual uranium content in zircon was calibrated by multiplying the measured value determined by the direct fluorimetric method by the coefficient 1.31.
- (4) Various mineralogical characters of zircon such as radioactivity, natural fluorescence, colour, crystal defect, Hf and other minor elements were investigated in detail and intimately related to the uranium content.

- (5) From these mineralogical studies on the uraniferous zircon, the evolution process of granitic magma distributed in the Tensei Mine could be divided into three consolidation stages.

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