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The Microgranulary Uraninite from Iisaka, and Its Geologic Age.

BY

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The Iisaka pegmatite invades near the north end of the Abukuma granite region extending from north to south over a hundred kilometer in the north-eastern part of the Main Island of Japan. The pegmatite carries various sorts of rare minerals including thorogummite,(1) yttrialite,(2) fergusonite,(3) xenotime,(4) zircon(4) tengerite,(5) abukumalite,(6) allanites,(7) gadolinite,(8) etc. The peculiar mode of occurrence of uraninite in this pegmatite is rather noticeable as the mineral exists in a finely dispersed state through the whole rock body especially in the part of dusky greenish or reddish feldspar. Individual crystals ranging in diameter from about 0·01 mm to 0·2 mm are superficially almost invisible in the original rock, but elutriating the finely crushed pieces of the same rock mass a small quantity of minute granular black minerals which consist mostly of uraninite is always obtained as the final heaviest remnant.

The fine grains of the mineral, thus obtained, are sometimes still so intimately associated with fergusonite that the latter can only completely be removed by repetition of the panning process after the grains have been ground into a finer powder. In this manner, about two hundred grams of pure uraninite could be secured from about 2~3 tons of mother rock rich in biotite. This specimen of powdery uraninite was again purified by treating with concentrated hydrochloric acid to dissolve up any trace of adhering thorogummite, and subjected to the chemical analyses. Whenever the sample was dissolved in warm dilute nitric acid a very trifling amount of niobic and tantalic acids

(1) S. IIMORI and S. HATA: *Sc. Pap. I.P.C.R., Tokyo,* **34** (1938), 447.
(2) S. HATA: *ibid.*, **34**, 455.
(3) S. IIMORI and S. HATA: *ibid.*, **34**, 504.
(4) S. HATA; *ibid.*, **34**, 619.
(5) T. IIMORI: *ibid.*, **34**, 832.
(6) S. HATA: *ibid.*, **34**, 1018.
(7) T. IIMORI: *ibid.*, **36** (1939), 53; S. HATA: *Ibid.*, **36**, 112.
(8) c.f., S. Hata: *Bull. I.P.C.R.*, **19** (1940), 71.

was always liberated. These earth acids were, therefore, considered to be the proper minor constituents of this mineral as no trace of fergusonite could be decomposed by such a treatment. The amount of UO_2 was estimated by titrating the solution obtained upon heating the sample at 170°C in a sealed tube with a sufficient quantity of dilute sulphuric acid (1:6) by means of potassium permanganate solution. The result obtained is given in the following table:—

Constituents	Per cent.	Constituents	Per cent
UO_3	22·23	Fe_2O_3	0·17
UO_2	55·40	Al_2O_3	0·44
ThO_2	3·86	SiO_2	0·11
Y-earths	14·60	$(Nb, Ta)_2O_5$	0·74
Ce-earths	0·41	CO_2	0·28
PbO	1·01	H_2O	0·50
Bi_2O_3	0·00	Total	99·75

The analysis shows the mineral to be the cleveite type having a rather high amount of rare earths and lower percentage of thoria. In order to determine the ratio of the amount of radium to that of uranium the former was estimated by the emanation method using the Soddy's emanation-electroscope[9] whose constant was $45{\cdot}1\times10^{-12}$ g Ra per div. per min. The solution of the mineral in dilute nitric acid gave $2{\cdot}31\times10^{-7}$ g Ra per g, and the aqueous solution which was obtained by dissolving the fused mass of its niobic and tantalic acids fraction with sodium carbonate, $0{\cdot}7\times10^{-10}$ g Ra per g, hence showing the latter value to be quite negligible as against the former. The result thus obtained shows the ratio Ra: U to be $3{\cdot}43\times10^{-7}$, indicating that the radium exists in exact equilibrium with uranium. The mineral seems unaltered and to have been wellpreserved notwithstanding its powdery nature. Under the microscope the presence of octahedral crystalline grains is noticed together with fragmental particles. The approximate age was, therefore, calculated by the Holmes-Lawson's modified formula,[10] Age $= \dfrac{\text{Pb}}{\text{U}+0{\cdot}36\ \text{Th}}\times7600$ million years. The beforementioned analytical result gives thus

(9) F. Soddy: *Phil. Mag.*, [6] **18** (1909), 846.
(10) *vid.*, *Bull. Nat. Res. Council, Washington*, No. 8 (1931), 202.

0·0136 as the lead-ratio assuming the absence of ordinary lead from this material, and 103 million years of age. This is approximately in accord with the figure formerly obtained by S. Hata[11] on the yttrialite from the same pegmatite, being, however, somewhat lower than the latter.

The amount of UO_2 is rather considerably large in this Iisaka uraninite, *viz.* the ratio of UO_2 to UO_3 being 2·49. It is recently stated that this ratio decreases with increasing alteration as is seen in the Villeneuve uraninite,[12] owing to the gradual oxidation of UO_2. A. Holmes pointed out, however, that this ratio and also the lead-ratio increases as weathering progresses,[13] and this fact was ascribed to the nature of UO_3 which is liable to be leached out in relatively higher proportion than UO_2 and lead. In the Iisaka uraninite, the lead-ratio is not so particularly large as compared with those shown by the yttrialite and fergusonite from the same pegmatite,[14] and the good equilibrium existing between its radium and uranium indicates the mineral suffered no weathering at least in the last million years. The inferior amount of UO_3, in this mineral may, therefore, be attributed to its slight degree of alteration, and the Kirsch's opinion[15] that the crystalline uraninite occurring as an accessory constituent of granites is thought to consist initially of UO_2 containing together with some amounts of thoria and yttria earths seems to be compatible with this case of the Iisaka uraninite.

In conclusion I wish to express my sincere thanks to Dr. S. Iimori for his valuable advices, and to Mr. O. Nagasima for his help in preparing the sample used for analyses.

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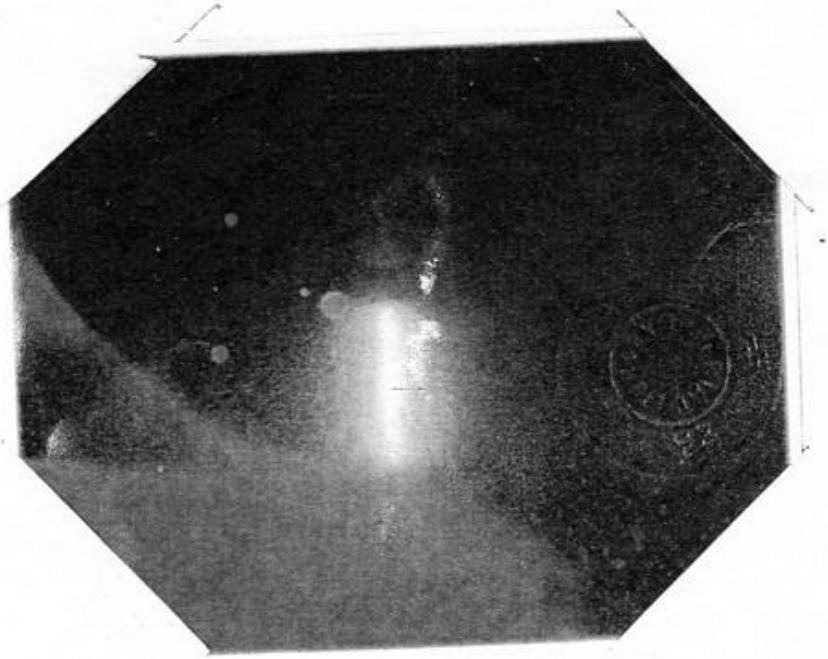
(11) S. Hata: *Sc. Pap. I.P.C.R.*, **34** (1938), 455.
(12) H. V. Ellsworth: *Am. Min.*, **15** (1930), 455.
(13) A. Holmes: *Am. J. Sci.*, **27** (1934), 343.
(14) S. Hata: *Sc. Pap. I.P.C.R.*, **34** (1938), 455; S. Iimori and S. Hata: *ibid.*, **34** (1938), 455.
(15) G. Kirsch: "*Geologie u. Radioaktivitit*" (1928), 140.

The microgranulary uraninite from Isaka in a cork-stoppered glass tube (Natural size).



The radiophotogram of the same.
(T. Iimori.)