A New Method for Radiochemical Prospecting of Uranium Ore. I. Estimation of the Amount of Uranium

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Introduction

The economic importance of uranium and thorium has been stimulating extensive investigations of terrestrial occurrence and distribution of these elements. One of the important parts of these investigations is prospecting for uranium and thorium ores.

For many years, radioactive survey with a Geiger-Müller counter has been extensively used for this purpose. Recently, the development of a scintillation counter has made it possible to prospect radioactive deposits by the airborne radioactive survey.

However, these methods are effective only for the cases in which a uranium deposit is cropping out on the surface of the ground, because gamma radiation from these elements can not penetrate thick layers of rocks and soils. Furthermore, it is considerably difficult to distinguish between uranium and thorium ores by these methods.

Therefore, practically there has been no method to prospect and evaluate an underground uranium deposit without a test boring of the ground. The aim in the present work is to establish a new radiochemical method in which naturally radioactive elements in natural waters are used as a kind of tracer for the purpose of estimating the amount and the depth of uranium layer under the ground.

It is well known that the uranium content of mineral waters is less than that of sea water, but radon and radium content of some mineral waters is much higher than that of sea water. (Table I)

TABLE I
RADIOACTIVE ELEMENTS IN NATURAL WATERS

	U (g./l.)	Ra (g./l.)	Rn (curie/l.)
Sea Water[1]	10^{-6}	10^{-13} to 10^{-14}	10^{-13}
Mineral Water[2]			
Hot Spring			
Arima	10^{-6} to 10^{-7}	10^{-10}	10^{-10}
Mineral Spring			
Masutomi	10^{-7} to 10^{-8}	10^{-11} to 10^{-12}	10^{-6} to 10^{-10}

The content of radon and radium in mineral waters could be explained as follows; mineral water passes through a uranium-rich layer under the ground, dissolving radon and radium which have greater solubility than their ancestors. Based on these facts, the author considered that the radiochemical studies of mineral waters can be used as a tool for prospecting a uranium-rich layer under the ground.

The Presence of Two Radioactive Strata

It has been suggested, in some of the previous reports[3], that there must be two radioactive strata under the ground of mineral springs. A deep stratum, which is probably related to the uranium or thorium ores, may be located at a depth of the order of several kilometers. This stratum is considered as a supplying zone of radium and its isotopes, such as thorium X, mesothorium 1 and actinium X, for mineral waters. The other radioactive stratum, which is considered to be the source of radon and thoron for mineral waters, is located very close to the surface of the earth at a depth from 10 to 50 meters below the ground.

When radium-containing mineral waters come close to the surface of the earth, deposition of the sinter deposits takes place, which is caused by the escape of carbon dioxide, contact with air, decrease in temperature etc. Then a part of radium and its isotopes is precipitated with sinter deposits.

The porous zones of the sinter deposits, which are as radioactive as ordinary radioactive minerals such as monazite, etc., are often found in the areas of radioactive springs. The radio activity of sinter deposits is mainly due to radium and its isotopes, and it is a very important fact that the uranium content of these deposits is much lower than the value expected from radium content. For example, the uranium content of sinter deposits from a mineral spring in Japan ranges from 0.004 to 0.05 p. p. m.[4] (both extremes for Masutomi Mineral Springs), whereas these sinter deposits contain about 10^{-10} g. Ra per gram.

The very low content of uranium in mineral waters and sinter deposits was apt to lead us to a faulty conclusion that there was probably no uranium-rich ore in the areas of radioactive springs of Japan. But the occurrence of a remarkably uraniumbearing layer is concluded by the author's present estimation.

Estimation of the Amount of Uranium Reserves

It is possible to estimate the amount of uranium, which occurs in a supplying zone of radium, by calculating the amount of supply of radium to mineral waters per second. The estimation may be carried out by two different ways.

In the first one, it is assumed that the atoms of radium, which are generated by the radioactive decay of uranium, are directly supplied into mineral waters.

The amount of radium, G_{Ra} which is generated from 1 g. of uranium per second, is calculated as follows.

$$G_{Ra}=\frac{226}{238}\lambda_U=4.6\times10^{-18}\ \text{g./sec.} \qquad (1)$$

where λ_U is a decay constant of uranium and 226 and 238 are mass number of radium and uranium respectively.

On the other hand, the amount of radium,

1) F. Herneggar, *Anz. Akad. Wis. Wien*, **1**, 19 (1933), R.D. Evans, A.F. Kip and E.G. Moberg, *Am. J. Sci.*, **36**, 241 (1938), I. Iwasaki and T. Ishimori, *J. Chem. Soc. Japan*, **72**, 14 (1951), M. Nakanishi, This Bulletin, **24**, 36 (1951), E. Rona and W.D. Urry, *Am. J. Sci.*, **250**, 241 (1952).

2) T. Nakai, This Bulletin, **15**, 333 (1940), P.K. Kuroda and Y. Yokoyama, ibid., **22**, 34, 43 (1949), M. Nakanishi, ibid., **24**, 33 (1951), Y. Yokoyama, *J. Chem. Soc. Japan*, **76**, 558 (1955).

3) P.K. Kuroda and Y. Yokoyama, This Bulletin, **21**, 52, 58 (1948); **22**, 34, 43 (1949); *Anal. Chem.*, **26**, 1509 (1954); Y. Yokoyama, *J. Chem. Soc. Japan*, **70**, 399 (1949).

4) M. Nakanishi, This Bulletin, **24**, 33 (1951).

which is supplied into mineral waters per second, is $C_{Ra} \cdot F$, where C_{Ra} is the radium content of mineral waters (g./l.), and F is the amount of flow (l./sec.).

From the supplying rate of radium, therefore, one can expect that the following amount of uranium, W, is present in supply zone.

$$W=\frac{C_{Ra} \cdot F}{G_{Ra} \cdot K} \text{ g.} \qquad (2)$$

where K is a dissolving coefficient of radium into mineral waters from uranium ore. In the case of perfect dissolving, the value of K is 1.00.

For example, in Hot Spring Arima, which has the highest radium content in Japan, C_{Ra} is 2×10^{-10} g./l. and F is 5 l./sec. Then one can obtain the following equation.

$$W=\frac{2\times10^{-10}\times5}{4.6\times10^{-18}\times K}=\frac{2\times10^{8}\text{g.}}{K}=\frac{200\text{ ton}}{K} \qquad (3)$$

From the foregoing calculation, one might expect that 200 ton or 2,000 ton of uranium is present under the ground of Hot Spring Arima, on the assumption that K=1.00 or 0.10 respectively.

The exact value of K is unknown, but it may be not unreasonable to suppose that the value of K is of the order of 0.1. It was found by the present author that mineral waters contain usually a comparable amount of thorium X with mesothorium 1[5].

Table II

Isotopes of radium in mineral waters

Name of Spring		^{226}Ra 10^{-12} c./l.	^{224}Ra(Th X) 10^{-12} c./l.	^{228}Ra(MsTh I) 10^{-12} c./l.	^{225}Ra(Ac X) 10^{-12} c./l.	Th X/MsTh I
Arima	Shin-onsen	170±5	560±20	580±10	9±0.5	0.97±0.04
〃	Ariake-no-yu	170±7	310±10	380±20	9±0.9	0.82±0.05
Ikeda	No. 1	33±1	100±2	85±8	——	1.18±0.07
〃	No. 2	36±3	113±12	53±7	——	2.12±0.36
Masutomi	A 5	66±3	200±7	230±10	——	0.87±0.05
〃	B 7	24±1	66±2	240±1	1.3±0.1	3.6 ±0.1
Misasa	Misasa-kan	14±1	34±2	38±4	——	0.90±0.11
〃	Ōhashi-sotoyu	9±2	95±6	30±6	——	3.2 ±0.7
Tamagawa	Ōbuki	16±1	144±2	109±3	——	1.32±0.04
〃	Kobuki	24±0.5	160±6	131±2	——	1.22±0.05

Table III

Amount of uranium estimated by radiochemical method

Name of Spring	Pref.	Ra Content 10^{-12} g./l.	Amount of flow l./sec.	Ra Discharge 10^{-12} g./sec.	Amount of Uranium K=1.00 ton	Amount of Uranium K=0.10 ton
Springs with Ra Content $>10^{-11}$ g./l.						
Tamagawa	Akita	20	150	3,000	700	7,000
Arima	Hyōgo	300	5	1,000	200	2,000
Masutomi	Yamanashi	30	2	60	10	100
Matsunoyama	Niigata	15	4	60	10	100
Misasa	Tottori	15	3	45	10	100
Yugakae	Shimane	30	1	30	7	70
Ikeda	Shimane	40	0.25	10	2	20
Takarazuka	Hyōgo	25	0.4	10	2	20
Isobe	Gumma	15	0.3	5	1	10
Springs with Ra Content $\leq 10^{-11}$ g./l.						
Onogawa	Yamagata	8	600	4,800	1,000	10,000
Toyotomi	Hokkaido	10	300	3,000	700	7,000
Yuzaki	Wakayama	8	15	120	30	300
Kawatana	Yamaguchi	10	8	80	20	200
Wakura	Ishikawa	5	10	50	10	100
Ōshio	Fukushima	4	3	12	3	30

5) Y Yokoyama, *J. Chem. Soc. Japan*, **76**, 558 (1955).

This fact indicates that the dissolving coefficient of radium and its isotopes is not very large. The reason can be explained as follows. If mesothorium 1 dissolves prefectly into mineral waters, it would be impossible to supply thorium X in mineral waters from supply zone. Because, in such case, no mesothorium 1 and its descendants (radiothorium and thorium X etc.) are present in supply zone.

Another method of calculation for the amount of uranium is as follows.

It is assumed that at first, uranium of supply zone is in radioactive equilibrium with radium, then mineral waters dissolve radium. The amount of radium, which is in equilibrium with 1 g. of uranium, is

$$E_{Ra} = \frac{\lambda_U \cdot 226}{\lambda_{Ra} \cdot 238} = 3.4 \times 10^{-7}\ \text{g}. \qquad (4)$$

where λ_U and λ_{Ra} are decay constants of uranium and radium respectively.

During the time interval T second, the amount of radium brought to the surface of the earth by mineral waters is $C_{Ra} \cdot F \cdot T$(g.). From the total supplying amount of radium, therefore, one can expect that the following amount of uranium, W', is present in the supply zone.

$$W' = \frac{C_{Ra} \cdot F \cdot T}{E_{Ra} \cdot K'} \qquad (5)$$

where K' is a portion of radium, which is soluble in mineral waters, to total radium.

For example, on Hot Spring Arima, assuming $T=100$ year (3×10^9 sec.) and $T=1{,}000$ year (3×10^{10} sec.) we get

$$W' = \frac{2\times10^{-10}\times5\times3\times10^{9}\text{g}.}{3.4\times10^{-7}\times K'} = \frac{10\ \text{ton}}{K'}, \text{ for } T=100\ \text{year},$$

$$W' = \frac{2\times10^{-10}\times5\times3\times10^{10}\text{g}.}{3.4\times10^{-7}\times K'} = \frac{100\ \text{ton}}{K'}, \text{ for } T=1{,}000\ \text{year} \qquad (6)$$

The foregoing calculation shows that the value of W' in Eq. (6) becomes close to the value of W in Eq. (3) when the time interval T is as long as the order of the half life of radium (1622 years). Therefore these two calculating methods will give practically the same value, if issue of mineral waters continues during some long time in which a kind of equilibrium state takes place between leaching and supplying of radium. More general discussions about these points will be reported in future in detail. But it may be supposed that the times required for equilibrium will be shorter than the values expected from the half life of radium, because those times are determined not with physical half life but with actual half life in which geochemical half life by leaching effect is also considered.

Using Eq. (2), the author estimates the amount of uranium in the supplying zone of the main mineral springs, containing radium, in Japan. The results are shown in Table III. It must be noticed that the amount of uranium, calculated in this way, is not the weight of minerals but the weight of uranium metal only. In Table III, two values are shown for the amount of uranium, where K is supposed to be 1.00 and 0.10.

The largest amount of uranium ore in Japan, discovered in the past, is Suishō-yama at Ishikawa, Fukushima Pref. The amount of uranium reserves in Suishō-yama is estimated as about 1 to 2 ton as uranium metals, or 2,000 to 4,000 ton as minerals. The mean uranium content of ore is estimated as 0.05% in Suishō-yama.

It is found in Table III that the amounts of uranium in some mineral-spring areas estimated by the author, are very much higher than that of Suishō-yama.

Recently a new uranium deposit has been discovered at Ogamo Mine near the Hot Spring Misasa, which is one of the largest radioactive springs in Japan. This fact may support the author's opinion.

Summary

A radiochemical method for the prospecting and the estimation of the amount of uranium layer under the ground based upon the determination of radium in mineral waters, is described. Results show, 700 to 7,000 ton of uranium may occur in Hot Spring Tamagawa and 200 to 2,000 ton of uranium in Hot Spring Arima. The natural radioactive elements in mineral waters will give valuable information on the geological structures, especially the nature of the underground radioactive strata.

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