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On the Emanating Power of Powdered Rocks

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Abstract

In this paper are reported the measurements of radioactive gases like radon, thoron and actinon emitting from rocks and soil carried out by nuclear emulsion (ET-2E, Fuji Photo Co. Ltd.) exposed from one to seven days to powdered samples enclosed in an emanation chamber of about 300 ml. As already pointed out by some authors, the value of the emanating power seems to depend on whether the radioactive elements exist as primary minerals or as secondary ones.

The alpha track distribution obtained by the present authors of different lengths over 37.5 microns counted under a high power lens of microscope, clearly indicates RaC′ and ThC′ peaks in diagram, and this result seems applicable to the pursuit of thorium-uranium behavior in rocks.

Introduction

"Emanating power" was defined by R. D. Evans[1] as the fraction of radon lost from rocks and minerals to the sorrounding air. By emanating power we understand the radioactivity possessed by such gases as radon (Rn-222), thoron (Rn-220) and actinon (Rn-219) emitted from pulverized rocks and soil in their natural condition.[2][3] Many articles were written on this subject to discuss the geological age problem. When the rock radioactivity is measured, it is carried on generally on the supposition that the elements of thorium uranium series are in their radioactive equilibrium. But when radioactive gases are escaping from the rock, this equilibrium is lost.[4][5] This is of great importance concerning the geological age determination of rocks and other topics.

Two problems were treated in the present research: firstly, the emanating power of some pulverized rocks and soil, and secondly, which sort of gas (radon or thoron) is being issued from them. The value of emanating power and the radon leakage depend remarkably on the rock type and on whether they are metamorphosed or not, as pointed out already by Hurley[6] and Kulp.[4][5]

Generally, the origin of the measurable emanating radioactivity from rocks and minerals are twofold: 1) The emanating gas atoms are emitted, by the recoil force of radium alpha particles (Ra-226, Ra-224, Ra-223), from the rock surface and

minerals just under it into the air. 2) The floating radon atoms migrate, by diffusion, from cleavage or interstitial space between grains into the ionization chamber[7]. For the former, polished sections of hard crackless rocks are explicable only because their emanating power is presumed to eject from the recoil range of their alpha disintegration. The diffusion of the emanating gas into the air is much dependent upon the grain-size and other conditions of the sample.

Method of measurement

(1) Fifty grams of rock or soil sample ground to pass through 65-mesh screen was enclosed in emanation chamber, so that the surface of the powder might be flat, and besides great care was taken, so that no grains of the powder might remain fixed to the inner side of the chamber above the powdered surface. Lest the powder should be scattered over the emulsion, the sample surface was covered with four fold gauses, on which the photoplate was put with its emulsion upward (Fig. 1).

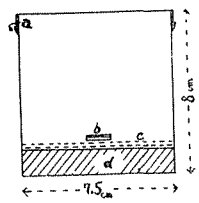


Fig. 1. Emanation chamber.
a) packing-clay
b) photo-plate (emulsion upward)
c) gause
d) powdered sample

The emanation chamber was tightly packed with packing clay and kept thus for a week—in winter, in any place where there is little variability of temperature and, in summer, in an ice box.* In this way the radioactive gases got diffused in the air inside the chamber till at last they attained to an equilibrium. To clarify the radon recovery, the exposure was shortened, in some specific cases, to one or three or five days. In a few other cases the chamber was opened and put upon a water bath for two hours,** to purge the old gases away from the chamber and, when the sample was cold again, the same experiment took place once more. In this manner it became clear that, after the purge of old gases, the recovery of thoron was so rapid, while that of radon was very gradual.

(2) Thus, due to the post emanation radioactive element, alpha tracks were left on the photoplates developed after certain days' exposure. Here RaF was put out of consideration, as the half life of RaD is 22 years long. Hence Rn, RaA′, RaC′, An, AcA, AcC′, Tn, ThA, ThC and ThC′ alone were dealt with. These plates were tested using high resolution cederwood oil immersion, microscopically at magnifications in excess 1000×, and it was observed that alpha tracks was almost homogeneously distributed all over the emulsion. The number per unit area and per day was easily obtainable, if the blank tracks due to the background were deducted beforehand.

* It was found that any radioactivity inside the chamber was made to escape by washing its inner wall with alcohol before the powdered sample was put into the chamber.

** It should be noticed that the heating of the samples with water bath makes our experiment favourable in putting them in a uniform initial condition.

Track counting and its interpretation

Forty-five samples collected from a few localities in Tertiary shale in Nagano Pref. recorded on our photoplates per day 0～5 tracks per mm^2, and there was no remarkable exception to this value. The following histograms indicate the classification of the samples according to their different emanating powers (Fig. 2). Tertiary shale and sandstone of Ningyotoge and the gallery clay of Ogamo, both

Table 1.

Rock type	Localities	Alpha/mm^2/d	Remarks
granite	Gyojayama I	2.28	fresh
〃	〃 II	4.56	〃
〃	Kitashirakawa I	5.55	〃
〃	〃 II	5.96	〃
〃	〃 (K-27) III	5.26	〃
〃	Miyoshi	7.36	weathered
〃	Suzuka	2.84	〃
〃	Yokorodani	14.7	〃 (1)**
〃	Hirose	78.7	〃 (7)**
aplite	Miyoshi	82.4	〃
〃	Mikumo	52.9	〃
〃	Kitashirakawa I	52.2	〃
〃	〃 II	14.2	〃
liparite	Nishinomiya	2.93	
quartzite	Kitashirakawa	2.35	
fault clay	〃 I	8.00	
〃	〃 II	15.8	
kokaseki	Niijima	0.00	
limonite	Ningyotoge	149.7	(2)**
sandstone	〃	361.5 (126.0)*	lens in shale (3)**
shale	〃	211.0	(4)**
gallery clay	Ogamo	260.0 (99.1)*	(5)**
manganic material	〃	408.0 (193.7)*	weathered, black torbernite outcrop (6)**
〃	Miyoshi	100.0	black, found in granite
black shale	Kyoto	1.57	Palaeozoic sediment
hornfels	Suzuka	4.21	〃
black shale	Ashi dam	9.56	〃

* Value of 7-days exposure after water bath treatment.
** The numbers correspond to those in Fig. 3.

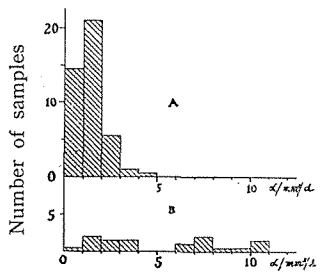


Fig. 2. Emanating power of rocks.
A) Tertiary shales in Nagano Pref.
B) Some weathered aplitic rocks.

in Tottori Pref. were issuing incomparably more radioactive gases (Table 1). No diffinite trend was to be seen among some samples from Palaeozoic black shales (Table 1). Ordinary aplitic dikes of some localities show lower values (Fig. 2) but some weathered aplite of Suzuka, Mie Pref., Mikumo, Shiga Pref., Kitashirakawa, Kyoto Pref. gave considerably high values. Some these severely decomposed samples seem to have undergone much secondary radioactive deposition.

Granite samples of certain localities such as Gyoja-yama, Kitashirakawa, Kyoto Pref., Graniteville and Milford, America* showed unexpectedly low values, and the same result was obtained from some basic rocks of foreign localities* (Table 2). These tendency are owing to the deficient emanation from the fresh rock sample. As for some aplitic dikes of Mikumo district in Shiga Pref., Dr. ASAYAMA's measurements[12] with Lauritsen radioscope and our own result well coincide with each other (Table 2). Seven days exposure was repeated on a few

Table 2.

Rock type	Localities	α/mm²/d	Ra (10^{-12}g/g)	div/min	Remarks
aplite (a)	Mikumo	8.22	0.82	0.081	weathered
" (b)	"	17.0	2.88	0.179	"
" (c)	"	45.4	1.54	0.409	"
" (m)	"	24.1	1.64	0.250	"
" (d)	"	30.6	—	—	"
granite	Tanakamiyama (466)	3.40	0.68		
"	" (800)	6.65	0.92		(48 g)
"	" (550)	7.04	1.87		
"	" (665)	13.7	1.72		(49 g)
"	Graniteville, Missouri*	5.32	3.3 ±0.2		
"	Milford, Massachusetts*	2.70	0.23±0.02		
gabblo diorite	Woburn, Mass.*	1.65	0.18±0.02		
Deccan trap	Bombay, India*	1.10	0.21±0.04		
basalt	Columbia river U. S. A.*	1.61	0.33±0.03		

* The radioactive standard rocks of Bureau of Standard, America.

Table 3.

Sample	I	II
Ningyotoge limonite	149.7	118.4
" " black shale	211.0	194.8
Hirose weathered granite	78.7	68.5
Graniteville granite	5.32	5.51
Milford granite	2.70	3.02

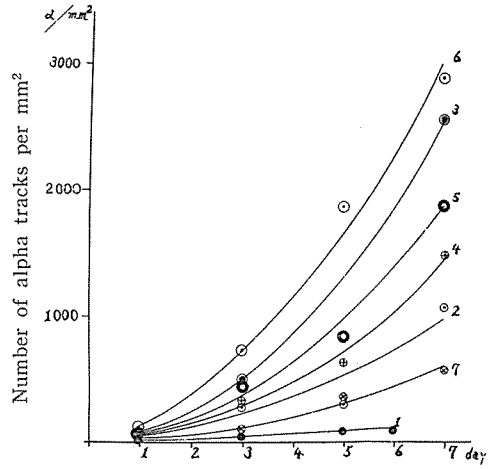


Fig. 3. Variation of emanating power on the different exposure.
The numbers of the curves correspond to those of Table 1.

samples and the result was found with fairly good reproducivility as follows (Table 3). According to our measurement, one eman of radon gas confined in a chamber of about 300 ml. was leaving on our emulsion 5.4 alpha tracks per day per mm^2. Radon is not of course the only radioactive gas that rocks and soil are issuing. However diverse the radioactive gases issued therefrom may be, we are still inclined to believe that the above given value of 5.4 can well serve as our provisional unit.

The exposure lasted 1 day, 3 days or 5 days to see if the increase of the track number was linear or not; and it was found linear in samples of Yokorodani and Hirose granite in Tottori Pref. but accelerative in such samples as gallery clay in Ogamo mine, black shale of Ningyo-toge, both in the same Pref. (Fig. 3).

Water bath operation of samples diminishes the track number into about less than one half of the non-operated (Table 1)

Radon whose half life is 3.825 days reaches to its radioactive equilibrium in about one month, while thoron whose half life is but 54.5 seconds in some 10 minutes. It results therefrom that the linear increase of track number with prolonged exposure is expected where thoron is comparatively dominant, whereas the abrupt increase of beginning radioactivity is liable where radon is abundant.

After water bath operation the radioactive gases are left to go out from the emanation chamber into the air, then, while the recovery of radon takes much time, the effect of thoron appeares rather swift. This is evident also from another method soon to be mentioned.

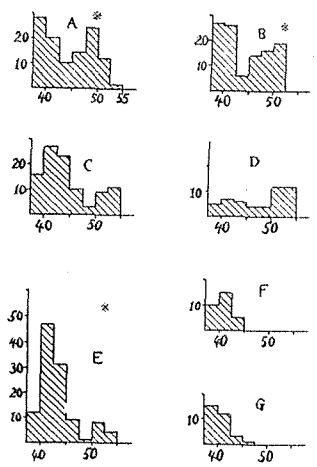
Discrimination of radon from thoron by track length.[7),8),9)]

To study the distribution of RaC′ and ThC′ tracks, our present method of affecting our emulsion with radioactive gases (Rn-222, Rn-220 and Rn-219) and their daughter elements is more favourable with greater accuracy than ordinary

Table 4.

Radioactive elements	ThC′	RaC′	ThA	Tn	ThC	RaA	Rn
Range in air (cm)	8.57	6.91	5.64	5.00	4.73	4.66	4.05
Range in emulsion (μ)	54.6	44.0	36.0	32.2	30.1	29.7	25.8

contact autoradiographic method. The alpha track length of these radioactive gases and their daughter elements differ from one another on our developed emulsion (Table 4).



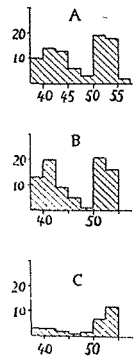
Ordinate: Number of alpha tracks
Abscissa: Track length in microns

Fig. 4. Distribution of RaC′ and ThC′ tracks from the emanations.
(A) Gyojayama granite, Kyoto Pref.·····4.56 alphas/mm²/day
(B) Kitashirakawa granite IV, Kyoto Pref.····· 8.10
(C) Graniteville granite 5.32
(D) Milford granite ····· 2.70
(E) Hirose granite, Tottori Pref.····· 78.7
(F) Ogamo torbernite outcrop, Tottori Pref.·····408.0
(G) Miyoshi (Hiroshima Pref.) black material in granite ·····100.0
* emulson of 50 microns in thickness

Now, for the comparison of alpha tracks of different length, a certain number, say 100, of tracks over 37.5 microns in length were selected from each developed emulsion, and the track distributions of different length were illustrated in diagrams. Tracks over 44 microns, originating from ThC′, proves at least the existence of thoron in the emanation chamber, and the peak representing in histograms the tracks of nearly 40 microns in length (RaC′) assures us the presence of radon in it. The advantage of this method is that RaC′ tracks and ThC′ ones can easily be distinguished from each other with comparatively small mechanical errors.

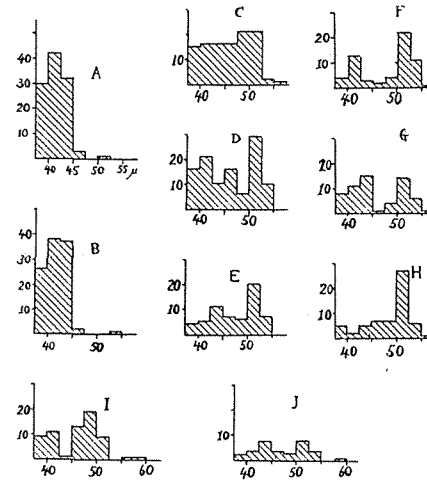
On granitic rocks

As shown in Fig. 4 (A), (B), in fresh Kitashirakawa and Gyojayama granitic samples above already mentioned, the tracks about 40 and of some 50 microns long formed two peaks of the same height in histogram indicating the same amount of radon and thoron issued from these samples, and we call here this type to be "Radioactive normal granite". In emanating power, Graniteville granite is only twice strong as Milford specimen in spite of their radium contents (Table 2), and RaC′, ThC′ tracks of these two samples show us the opposite peak height. (Fig. 4, C. D.) Samples of the following three localities were ejecting much more RaC′ tracks than ThC′ ones: fine brownish black and severely weathered Hirose granite, weathered one near the torbernite outcrop at Ogamo both in Tottori Pref., and black



Ordinate: Number of alpha tracks
Abscissa: Track length in microns

Fig. 5. Distribution of RaC′ and ThC′ tracks from the emanations.
Yokorodani weathered granite
[(Table 1 (1)]
(A) 6 days exposure
(B) 5 days exposure
(C) 1 day exposure



Ordinate: Number of alpha tracks
Abscissa: Track length in microns

Fig. 6. Distribution of RaC′ and ThC′ tracks from the emanations.
(A) Ningyotoge sandstone in black shale 361.5/mm²/d
(B) Ogamo gallery clay ..... 260.0 "
(C) Kitashirakawa fault clay II ..... 15.8 "
(D) Mikumo aplite (10) ..... 52.2 "
(E) " " (c) ..... 45.4 "
(F) " " (m) ..... 24.1 "
(G) " " (b) ..... 17.0 "
(H) " " (d) ..... 30.6 "
(I) Shinano-Ikeda Tertiary shale, Nagano Pref. ..... 1.41 " (S5616)
(J) Iiyama, Tertiary shale, Nagano Pref. ..... 2.50 " (H61)

manganic materials in fine Miyoshi* granite, Hiroshima Pref. (Fig. 4 E.F.G.)

One day's exposure after water bath treatment showed the same trend concerning the last two samples of Japan. (Fig. 4 F. G.). If thoron had been present in this case ThC′ would have formed a distinct peak in diagrams. But the absence of thoron is evident from the reversed result that we obtained. Moreover, as Fig. 5 shows us, in a Yokorodani example in Tottori Pref. the ThC′ and RaC′ peaks are of equal height when the exposure lasts 5 or 6 days, but in one day's exposure** RaC′ is fairy feeble because of the much slower recovery of radon.

In amount, say Keevil et al., the Th/U ratio is 3~4 in igneous rocks,[7),10)] but in alpha activity thorium is 4 times as feeble as uranium, while the proportion of RaC′ to the total alpha activity of uranium is $1/8=0.125$, and in the case of ThC′ to thorium is $(1/6)\times(66.3/100)=0.11$ and these values are nearly the same. Hence

* Not Okayama Pref.

** Only 30 tracks longer than 37.5 microns were obtained in spite of examinating of 1,292 cm².

it is natural that these two elements (RaC′ and ThC′) are showing the same radioactive order in "Radioactive normal granite".

In Hirose the "Radioactive normal granite" seems to have been influenced by radium or uranium solution, and this effect seems, as shown in diagrams, to have been still more conspicuous in Miyoshi and Ogamo.

On other rocks

The above described trend of radon dominance is striking also in Ningyotoge shales (Fig. 6 A) and Ogamo mine gallery clay (Fig. 6 B). These must be a natural outcome that follows every uranium deposite. Despite the feebleness in their total activity, the fault clay in Kitashirakawa granite (Fig. 6. C), the above mentioned Yokorodani decomposed granite, (Fig. 5) and the weathered aplitic rocks of Mikumo district, Shiga Pref., were all issuing much thoron, and so was the Tertiary shales in Nagano Pref. (Fig. 6 D~J).

One hundred tracks longer than 37.5 microns could be selected easily in samples of strong emanating power, but in specimens of feeble activity they must be searched for in wider areas of the plate or even in vain. Still the RaC′ and ThC′ peaks in diagrams may denote at least the ratio between these two radioactive elements that each sample contains.

It is supposed that, as dissolution and migration had taken place in igneous rocks to alter the Th/U ratio, some samples are issuing more radon, while others more thoron. This accounts for the fact that the secondary altered or weathered rocks issue much more radon or thoron than the fresh ones (only minute radioactive minerals), because the radioactive substances in the altered rocks had scattered and deposited in the microfissure of crystal or in the grain boundary.

Conclusion

(1) The radioactivities of emanation from the powdered rock were measured by the aid of nuclear emulsion.

(2) Thoron and radon are detectable by different alpha track length.

(3) Emanating power is relatively high in the altered or weathered rock, and not in the fresh rock.

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References

1) EVANS, R. D.: Phys. Rev., **45**, 38, 1943.
2) HAYASE, I.: Kagaku, **24**, 365, 1954.
3) BEHOUNEK, F. & KLUMPAR, J.: Nature, **160**, 641, 1947.
4) BATE, G. L., GILETTI, B. J. and KULP, J. R.: Bull. Geol. Soc. Amer. **63**, 1233, 1952.
5) ditto Amer. Min. **38**, 329, 1953.
6) HURLEY, P. M.: Bull. Geol. Soc. Amer., **61**, 1, 1950.
7) YAGODA, H.: Radioactive measurements with nuclear emulsion John Wiley, 1949.
8) HAYASE, I.: Reports of the Balneological Labor., Okayama Univ., No. **18**, 1957.
9) TOKUNAGA, S.: Mem. Col. Sci. Univ. Kyoto, Ser. A, **XXVI**, 109, 1950.
10) FAUL, H.: Nuclear Geology John Wiley, 1954.
11) BROWN, H., BLACK, W. J. and CHODOS, A. A. Bull. Geol. Soc. Amer. **64**, 1400, 1953.
12) ASAYAMA, T., HAYASE, I. and YOSHIDA, N.: Jour. Geol. Soc. Jap., **63**, 422, 1957.