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Dedicated to Professor Atsuo Harumoto in Commemoration
of his Retirement on the 16th November of 1959

The Radioactivity of Rocks and Minerals Studied with Nuclear Emulsion VIII

The Radioactive Unequilibrium and the Radiocolloid of Ogamo and Ningyotoge

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Abstract

Radioactivity of the air in the Ogamo gallery was measured with three different procedures by means of nuclear emulsion, and was certified concerning radon only.

Radiocolloid of radium concentration was found in the same gallery and Ningyotoge. It is this radiocolloid that emanates radon into the air of the gallery.

Some radioactive minerals obtainable in those two mines have lost their radioactive equilibrium condition, and this seems to be in close relation with the air radioactivity and the radiocolloid.

Introduction

The Ogamo mine in Tohaku County, Tottori Pref., supplies coffinite and other uraniferous minerals, and Prof. KATAYAMA once reported the amount of radioactivity the air in this gallery has[1]. The author visited there on Nov. 21, 1956, to measure the radon content of the air with photo plates. It was found that the air in the gallery had anomalously high radon. In the same gallery, the radiocolloid with high concentration of radium was found in the fault clay, and the coffinite of this mine had radioactivity of unequilibrium condition which is deficient in radium.

Anomalously high content of radon in the gallery, presence of radiocolloid and radioactive unequilibrium condition of uranium mineral; all of these are very much closely associated genetically with each other. The radon content of Ningyotoge gallery was said to be high, though not yet measured by the author, but he found radiocolloid and radioactive unequilibrium condition of the minerals.

The present author is now inclined, in any way, to suppose that the radiocolloid and unequilibriate radioactive condition of uranium minerals of minute grains are not rare in the natural occurrences.

Radon content of the air in the Ogamo gallery

Three procedures were adopted to measure the radioactivity of the air in the Ogamo gallery.

A. The air in the gallery was introduced into emanation chambers, so that the photo plates might be exposed for 24 hours to it.

B. Nickel plates charged with about 400 volts and exposed in the gallery were brought into contact with photo plates.

C. Photo plates were exposed to the air in the gallery, with special care lest they should catch stray beams or get spoiled with water drops.

A. The emanation chambers are shown in Fig. 1. The air of the gallery was taken into them with a spray and through a tube filled with $CaCl_2$. The photo plates were developed after 24 hours' exposure to the air in the chambers. By this

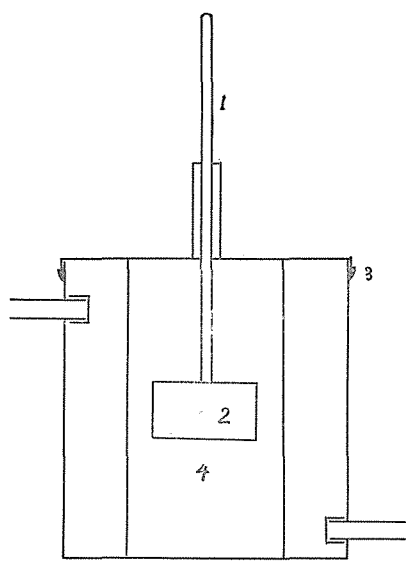


Fig. 1. Cross section of cylindrical emanation chamber.

procedure the radon contained in the air is obtainable at any spot in the gallery, but unfortunately radioactive gases of shorter half life like thoron and actinon remain undetectable, however much they may exist in it.

B. The second one is comparatively handy, if we are provided only with an adequate electric source. The active deposit (from radon, thoron and actinon) on the nickel plates consists mainly in RaB and ThB of longer half life. By this procedure our photo plates never fail to catch the ThC′ alpha tracks through the ThB deposit on the nickel plates wherever thoron is existent in the air.

C. Although the temperature and humidity in the gallery often disturb the third procedure, it enables us to catch the alpha tracks emitted from thoron and actinon, whose half lives are extremely short, if any small amount of them is contained in the air, for the photo plates are directly exposed to the air, though protected from light.

Fig. 2 is the outline of old Ogamo galleries where these experiments were performed.

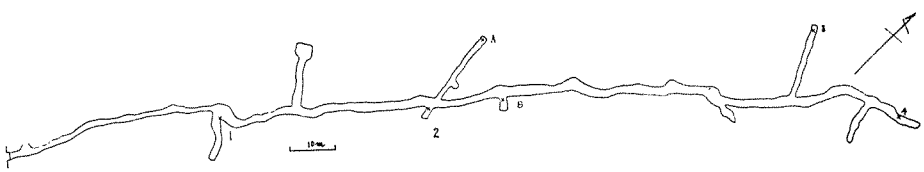


Fig. 2. The map of the drift of the Ogamo mine.

The results of the first procedure are given in terms of their radioactive comparison with the standard air whose radon content is already known[2]; and a photo plate exposed for 24 hours in an emanation chamber of the same size filled with an air containing 8.58 eman per litre, recorded 5.355/mm^2. Now, the track number was found 646.5/mm^2 in the spot 3 and 617/mm^2 in the spot 4 per day (see Fig. 2). Hence eman per litre must be 2.39, 10.10, 120.90 and 115.40 respectively in the spots 1, 2, 3 and 4. For a month or more nobody stirred the stillness of the air in this old gallery, and the author himself did his best not to stir it up by his own movement. The radon content is, despite of its richness in uranium minerals, rather little in the spot 1, for the gallery there is serving for the air circulation, while in the spots 3 and 4, though far from the minerals, the radon content is high because the air remains there still and unmoved. In the spot 2, notwithstanding its many coffinite bearing uranium veins, the radon content is merely 10.1 eman.

The apparent length of the 1112 alpha tracks which the air in the spot 4 (115.4 eman) recorded on the photo plate was measured under the microscope (see Fig. 3). But two very long tracks obviously of ThC′ origin are omitted, for they seem to have been due to some other origin than the radioactive gas in the gallery.

The three peaks prominent in Fig. 3 are of RaC′, RaA and Rn respectively, but the small one standing between the RaA and the RaC′ peaks still remains inexplicable, for, though ThA may be expected to form a peak hereabout, the short half life of thoron and the extreme scarcity of ThC′ tracks contradict this expectation.

So far as Fig. 3 is concerned, therefore, the alpha tracks are chiefly of radon and then of the radium series like RaA, RaC, and RaC′ alone. As the air must be shut up in emanation chambers, thoron, actinon and other gases of short half life remain indeterminable.

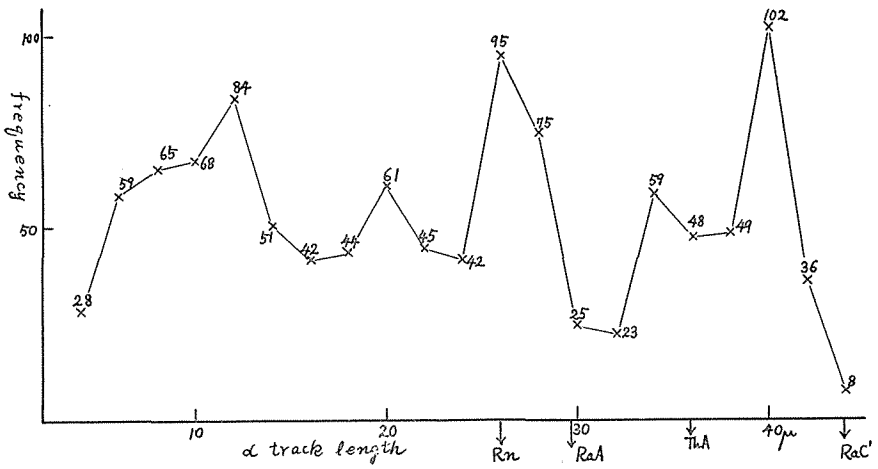


Fig. 3. The frequency of the track length ejected from the radioactive air (the spot 4 in Fig. 2).

Thoron is disintegrated, as shown in Table 1, and its short living daughter element ThB alone has a halfe life of 10.6 hours and, by gathering it on a nickel plate as an active deposit, the original thoron can be calculated indirectly. And this second procedure of ours was tried in the spots A and B in the gallery.

Among the alpha tracks recorded on the photo plates brought in direct contact with thus actively deposited nickel plates, 200 tracks apparently longer than 36 microns were taken up to show them in a histogram (see Fig. 4).

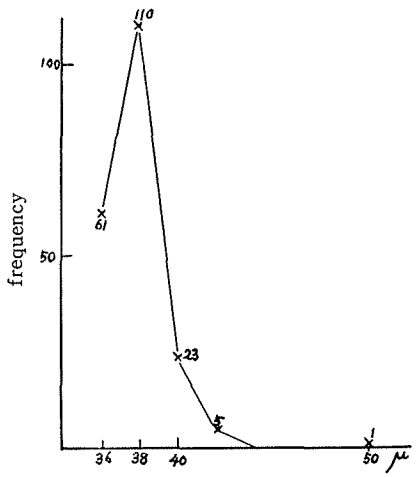
Had the air contained any little bit of thoron, it would have been deposited as ThB, and some ThC′ alpha tracks must have been found on the photo plates. But in Fig. 4 only one track is as long as 50 microns and this indicates that, thoron being almost absent, radon is the unique radioactivegas in the air of the gallery.

Table 1. Alpha track length.

Atom	Alpha track length	Mean range cm 15°C
UI	16.9 microns	2.65 cm
UII	20.4	3.21
Io	19.7	3.09
Ra	20.8	3.26
Rn	25.1	4.05
RaA	29.7	4.66
(RaC)	25.5	4.00
RaC′	44.1	6.91
RaF	24.5	3.84
AcU	19.1	3.00
Pa	22.7	3.57
RdAc	29.3	4.60
(Ac)	22.3	3.50
AcX	27.3	4.29
An	36.1	5.67
AcA	41.2	6.46
AcC	34.2	5.36
(AcC′)	41.6	6.52
Th	16.6	2.60
RdTh	25.5	4.00
ThX	27.5	4.32
Tn	31.8	5.00
ThA	35.9	5.64
ThC	30.1	4.73
ThC′	54.6	8.57

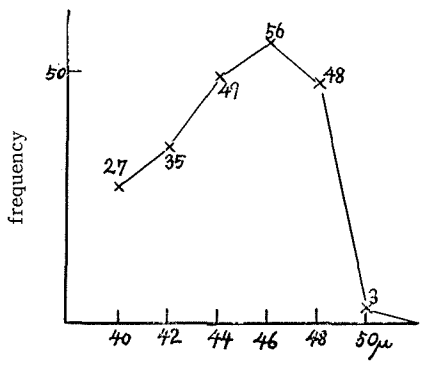
( ) show the small branching ratio.

The same experiment was performed with ThO_2 powder soon after it had just been baked. Had it contained any radon, this radioactive gas could no longer have stayed in it. Now, Fig. 5 shows the histogram of 218 alpha tracks apparently



α track length

Fig. 4. The frequency of the track length over 36 microns (the spot B in Fig. 2).

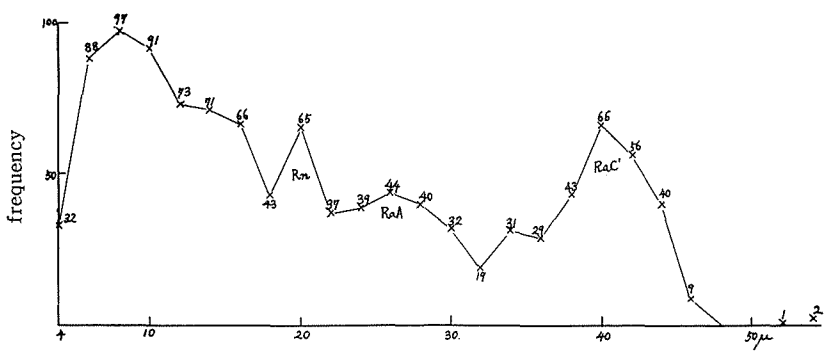


α track length

Fig. 5. The frequency of the track length over 40 microns (baked ThO_2 powder).

longer than 41 microns and, by comparing Fig. 4 with Fig. 5, the two peaks of different origin, namely, both of RaC′ and of ThC′, or of radon and of thoron, are sharply contrasted.

Then a photo plate was exposed directly to the air (i.e. the third sort of pro-



α track length

Fig. 6. The frequency of track length (the spot B in Fig. 2).

cedure) for one hour and eighteen minutes in the spot B (see Fig. 2); out of the tracks recorded on the plate 1112 were taken up to be measured, and the result thereof is given in Fig. 6, from which it is evident that, as they are chiefly of RaC′, radon must be there the main radioactive element of the air, though thoron is not altogether absent.

Very high radon concentration, says Faul, is seen in unventilated mines[3]. How can this be possible? The emanating power of powdered rocks[2] as well as the extremely high emanating power that hot spring sinter deposit[4] shows, have already been studied and reported. Such a strikingly high concentration of radon inside the gallery cannot but be ascribed mostly to the extraordinary emanating power of rocks and minerals therein. Some unusual sources of radon must be found in the surrounding rocks in the gallery.

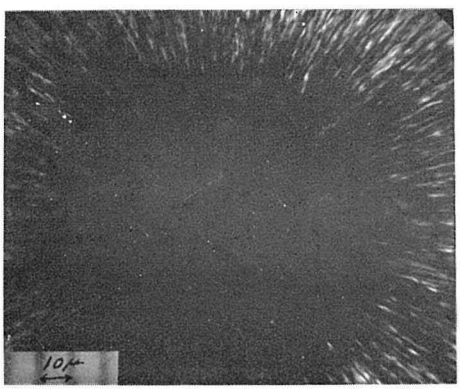
Radiocolloid

One of Ogamo fault clay samples given by Prof. KATAYAMA was remarkably radioactive, and its autoradiograph printed out a very strongly radioactive center (see Photo I), more strongly indeed than usual pitchblende, but the mineral constituting that radiation center is in no way discernible, only except that the radiating tracks printed on the photo plate localize it and determine vaguely its size. In such centers the $T\alpha$ was 3453 at the top, then 885, 344 and so on. Their size was about 30–80 microns in diameter and, generally, the smaller in size, the stronger in activity.

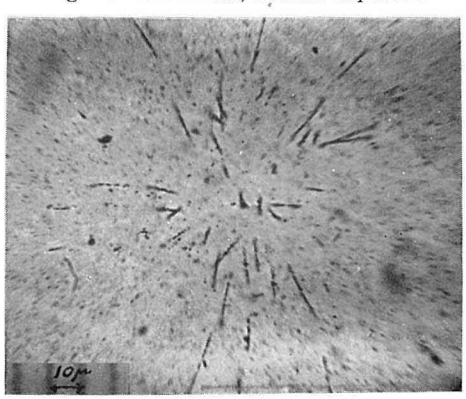
The present report is probably

Photo. I.

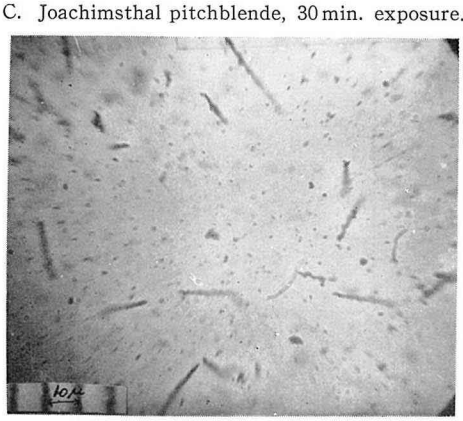
A. Ogamo radiocolloid, 7 days exposure.



B. Ogamo radiocolloid, 30 min. exposure.



C. Joachimsthal pitchblende, 30 min. exposure.



open to the blame that the author's measurement of their size was not sufficiently strict. Still it is certain that something now in measurement, which is much stronger than uraninite ($T\alpha=180$), the strongest mineral in radioactivity, is radiocolloid derived from radium concentration[5]. Is not this radiocolloid, a material both highly radioactive and easily emanating Rn, rather the main source of the rich Rn in the air in the mine?

A similar mineral is often existent also in Ningyotoge mine; for example, in the third gallery some 60 metres far from the entrance, the porphyritic feldspar alone contains radiocolloid, among whose 35 total tracks 5 are over 20 microns, the ratio being 14.3%.

The black mineral in the second gallery is not devoid of it and, though invisible under the microscope, its mineral size well discernible from its radiating tracks, eject 135 tracks among which 25 or 18.5 percent of the total surpass 20 microns.

In these minerals thorium is lacking, for, among their numerous tracks, none is of ThC′ origin, while many are of RaC′. Like radiocolloid of Ogamo mine, therefore, they may be identified as radium concentrations.

Here, however, as the size of their radioactive source is indeterminable, their unit area activity remains also unknown. As a whole, nevertheless, not necessarily higher than uraninite or pitchblende in radioactive order which declines, in turn, as the radium decay goes farther.

Radioactive Minerals out of radioactive equilibrium

Now let us turn to autoradiography with nuclear plates whose emulsion is 15 microns thick.

A Korean monazite specimen, thorium being its almost sole radioactive source, ejected 444 alpha tracks, among which 57 (namely, 12.8 percent of the total) were longer than 20 microns. Likewise, an Ogamo (Tottori Pref.) coffinite sample, whose radioactive material is entirely uraniferous, gave 619 tracks, among which only 13 (namely, 1.88 percent) were over 20 microns. As for uraniferous minerals, see Table 2.

Table 2. The track length ratios of radioactive minerals.

Radioactive mineral	Exposure time	Number of total alpha Tracks	Number of alpha tracks over 20 microns	Ratio
Ningyo-toge autunite	1.1 days	448	16	3.57%
Arukidani autunite	0.63 days	285	9	3.16%
Ningyo-toge ningyoite	2 days	286	8	2.80%
Ningyo-toge ningyoite	0.78 days	381	11	2.88%

Are these uranium containing minerals, however, really in perfect radioactive equilibrium?

To make sure if they are or not, the theoretically trustworthy standard of track length ratio must in the first place be persued the ratio, among the total alpha tracks, how many (or rather how few) of them must be apparently longer than 20 microns, so that their radioactive source may be identified as an element belonging to the uranium series, say, RaC′, when the alpha particle comes from within a thick source layer upon an emulsion 15 microns in thickness.

As the matter is quite complicated, and to make long story short, let us assume for a while that the alpha permeability remains always the same—both in rocks and in emulsion.

Then, as shown in Fig. 7, if the ejector is RaC′ and the ejection comes directly from the mineral surface, the track is over 20 microns upon the 15 microns emulsion, whenever the injecting angle is sharper than 36° 53′.

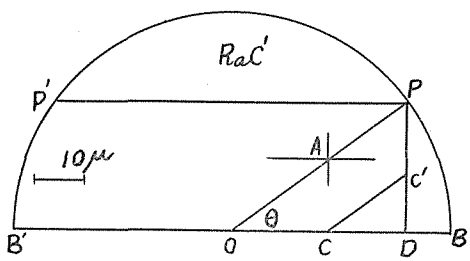


Fig. 7. Geometrical relation of the direction and the apparently length of the alpha tracks.

On the $\angle$AOB $\tan\theta = 15/20$ $\therefore$ $\theta = 36° 53'$

Hence, nearly 60 percent of the total are longer than that. Now draw a line from point P (see Fig. 7) perpendicularly to the line OB, and another one from a point C parallelly to the OP line, and let their cross point be C′. Then, when the line C′D is the deepest, it informs us how thick the radioactive mineral must be in the rock, so that over 20 microns alpha tracks may be left upon the 15 microns emulsion.

Fig. 8 illustrates the track lengths impressed upon the emulsions by the elements belonging to the uranium series. The UI, UII, Io and Ra were omitted because of the scarcity of longer tracks.

The track length ratio calculated by means of similar diagrams was found to be 4.37 percent in uranium series and 12.88 percent in thorium series, but 6.94 percent in actinium series is rather of little concern because of its comparative rarity.

The radioactive equilibrium of uranium and thorium series in rocks and minerals was the author's first premise throughout all his previous papers. But during his studies the real ratio was often showing much deviations, namely, smaller than 4.37 percent in genuine uranium series, or far bigger than 12.88

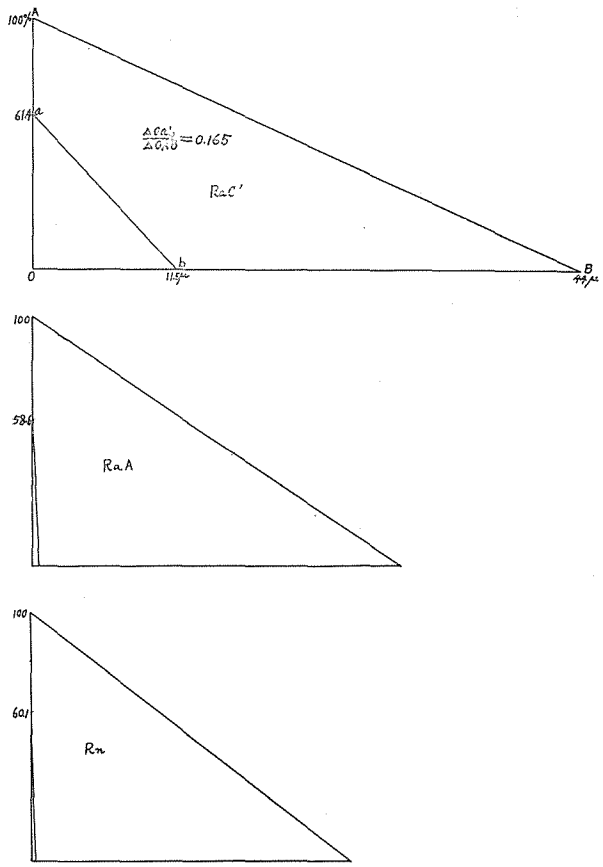


Fig. 8. The diagrammatic figure of (track number of over 20 microns)/(total track number) of *RaC'*, *RaA* and *Rn*.

percent in pure thorium series. Surely the reason of the deviation is not single.

1) If the radioactive source is but a thin layer, from the view-point of alpha activity, and in radioactive equilibrium, the reason of the deviation is discernible to an elaborate microscopic observation. If all the radioactive sources lie, either on a thin section or a polished surface, but few microns deep and none deeper than that, the ratio gets much bigger, for instance, about 30 percent in uranium and some 50 percent in thorium series. If on the contrary, something is inserted between the active source and the emulsion, the ratio is diminished.

2) If the radioactive equilibrium has been broken, the ratio varies greatly: it is smaller, for example, in uraniferous minerals, if radon is going out or the radium amount is under equlibrium.

And, if this be taken into consideration, the ratio (or the percentages given

in Table 1 concerning Ogamo coffinite, autunite and ningyoite) is *too small*—too small for radium to be in radioactive equilibrium in the surrounding rocks.

It is noteworthy, on the contrary, that near the occurrences of these minerals there lies radiocolloid!

How can this be explained? The fact that radium, leached away from coffinite or ningyoite, is being concentrated to build radiocolloid, is perhaps one of the possible explanations thereof. And, to sum up, the circumstances in the mines seem to suggest that the radioactive gases come out not directly from the minerals, but rather from this radiocolloid.

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