Study on Radioactivity of Hokutolite in Taiwan by Means of a Counter with Linear Amplifier

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I. Introduction

For the purpose of research in radioactivity of minerals, the intensity has generally hitherto been measured by using an electrometer, Geiger point counter or Geiger-Müller cylindrical counter. These all demand complicated procedures lasting for a number of days to decide the kind of rays (i. e., α-, β- or γ-rays) and to measure the contents of radioactive element in any sample. Practically, as they also suffer cosmic and local γ-ray effects, it is not easy to detect minute quantities of radioactive substance unless a chemical process is combined with them. These complexities are well circumvented by using a counting system with linear amplifier[1], which makes it possible to detect individual α-particles in the presence of β-and γ-rays. Thus accurate measurements may be carried out for such feeble α-emitter as igneous rocks. The author confirmed the superior efficiency of this counter in his study on α-radioactivity of Hokutolite and obtained a number of interesting results. Especially, the observation of the activity from part to part of a rock furnished us new knowledge concerning the essential properties of this mineral.

II. Historical

The radioactivity of Hokutolite was first discovered by Okamoto and Sunohara[2] on the river bed, within a limited range, of Hokuto-kei in Taiwan (Plate I). This is a spring-sediment which is deposited at most up to 2 cm. in thickness on the surface of andesite or sand-stone and is composed of an aggregate of granular rhombic crystals of white or light gray colour (Plate II a).

Among the results obtained by Okamoto and Sunohara[2] by means of prolonged photographic action, the following properties of the activity are of particular interest. i) The activity of the surface of any specimen is far stronger than that of its interior. ii) Regarding the surface activity, that which has distinct crystals is stronger than the

rest, and that of white colour is stronger than that of gray. iii) The activity of any surface does not decay during a period of several years.

Later, Hayakawa and Nakano[3] reported that they chemically separated ionium, radium and polonium from the sediment and observed the alpha-activity by a fontactoscope.

Then they considered the mechanism of the sedimentation of the mineral and concluded that the ionium only is the mother element from which all other radioactive materials are produced.

The results described in the present paper are derived by a new method different from that of preceding workers on the problem and are considered to be more reliable in some respects than their results.

III. Collections of Hokutolite

By permission of Taihoku Province collections of Hokutolite in Hokuto-kei were made several times between October 1935 and December 1938.

The main object of these collections at first was to provide a source of homogeneous alpha-rays, such as polonium if possible, which might be used for the standardization of a counting instrument, for we had none of the source for use at that time. For this purpose some twenty different sediments were collected, and their alpha-activities were detected by the counter with linear amplifier[1].

By this counting method it was easily found that the sample which was an aggregate of white colour crystals was most intense and one which was of distinct crystals was more intense than other samples. This result agrees with that of observations made earlier by Okamoto. These two kinds of aggregates of crystals were found, in general, dried on the dry stream-bed or in stagnant water; while others, such as some with a smooth surface, were under the flow of the spring. With this knowledge a great amount of specimens which had intense activity was collected, and polonium source of sufficient intensity was chemically prepared from them.

IV. The α-Activity of Natural Surface and that of Interior of Minerals

By preliminary experiments it was easily found that while the intensity of surface activity of various specimens showed distinct difference over a wide range, the activity of the interior of all of the samples was observed to be of almost equal intensity.

By bringing the sample up to 3 mm. before the window (1 cm. in diameter) of the ionization chamber[1] (5 mm. in depth), the aperture of which was covered by Al foil of 1 cm. equivalent of air, the number of α-particles emitted from the natural surfaces of various samples was counted. It was found to be 180/min. to 50/min., while that from interior of them was in all cases about 13/min. in contrast to the wide difference found in the activity on their natural surfaces. The same conditions were found in a sample* which is supposed to have been collected about 30 years ago. (This does not mean that the last sedimentation took place 30 years ago). The above results are summarised in Table I.

Table I.

Relative number of α-particles/Min.							
Sample No.	1	2	3	4	5	6	
Surface	180	130	128	74	50	52	mean 13.5/min.
Interior	14	13.2	13 3	14	13.5	13	

Table I. No. 6 is the old sample in the Museum of Taiwan. No. 2, 4 and 5 are for smooth surfaces: others are for zig-zag surfaces.

From these results we may conclude that in the interior the activity for all different specimens is constant within statistical fluctuations. Thus the distribution of radio-elements seems to be uniform in the interior of all specimens, and certain radio-elements of long half period are probably in equilibrium with their products, while in the surface there may be some element of relatively short life deposited. Accordingly it seems wise to interpret the variety of intensity of surface activity as due to the age of the specimen rather than to its irregular contents. The justification of this interpretation will be made in a later chapter after the nature of the radio-elements included in the sediment has been studied.

V. Intensity Variation with Depth from Natural Surface

If an intensity variation of the activity corresponding to different depths under the surface is known, the above results will be further confirmed and then the speed of sedimentation can be roughly estimated.

* This had been exhibited in the Museum of the Governor-General of Taiwan, by whose kind loan these results were verified. The author here expresses his thanks to the staffs of the Museum.

For this purpose a sample, No. 4 in Table I, (collected in a fast stream) was chosen, as this surface was fortunately almost plane and smooth. The α-particles were counted for each successive new surface after it had been carefully cleaned with a hard brush in running water so as to remove any powder of the preceding surface. The results are shown in Table II and Fig. 1.

Fig. 1

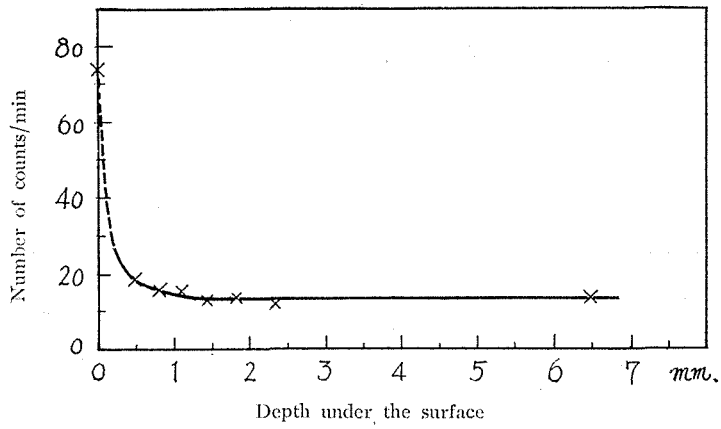


Table II.

Depth under the surface in mm.	0	0.5	0.8	1.1	1.43	1.83	2.33	6.5
Number of Counts/min.	74	19	16	16	13	14	12	14

Table II. The diameter of the window of the ionization chamber is 1 cm.

In this sample, the equilibrium comes at about 1.3 mm under the surface and then continues for further depth within the statistical fluctuations. As this result corresponds fairly well with that of Table I, it may be admitted to be true in general. Almost the same aspect has been obtained too, by the photographic action of α-particles (Plate II b). The black points which appear like sun-spots in the Plate II b are due to weak activities of the points which were probably worn down by some cause. It may be therefore conceivable that the growth of Hokutolite has been going on either exceedingly slowly, or intermittently in the course of a long time at the period when favourable conditions prevailed. But the latter explanation is more probable since

the temperature and chemical components of the spring suffer irregular variations from the meteorological conditions.

VI. Determination of the Kind of Radio-elements

Now, if we want a satisfactory explanation of the interesting aspects of radioactivity, it is necessary to make clear the kind of radio-elements

Table III.

Natural Surface			
Aluminium in mm. air range	Number of counts per 10 min.	Aluminium in mm. air range	Number of counts per 10 min.
10	2310	45	201
15	1962	50	156
20	1603	52	126
25	1217	55	110
30	863	60	75
32	710	62	57
35	504	65	19
40	372	70	16
42	282	72	14

Table III. The diameter of the window is 1.5 cm. The distance between the source and the window is 2 mm. The depth of the chamber is 10 mm.

Fig. 2

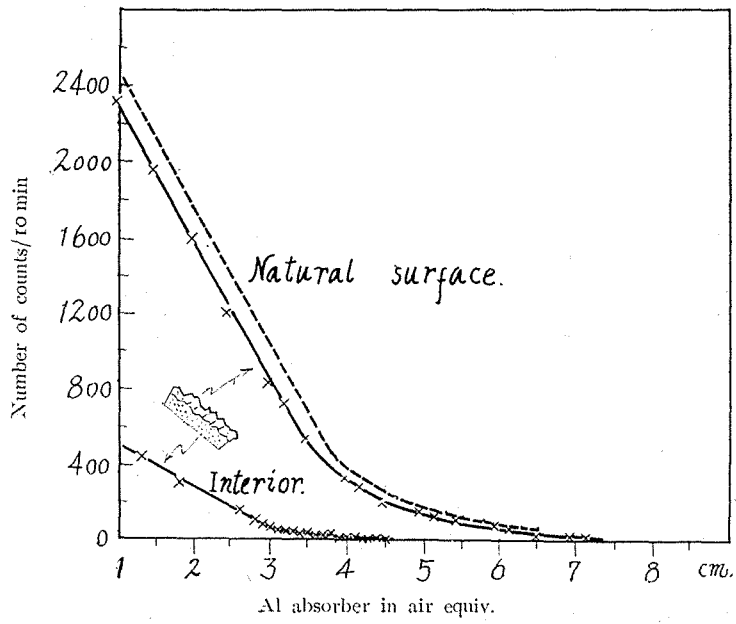
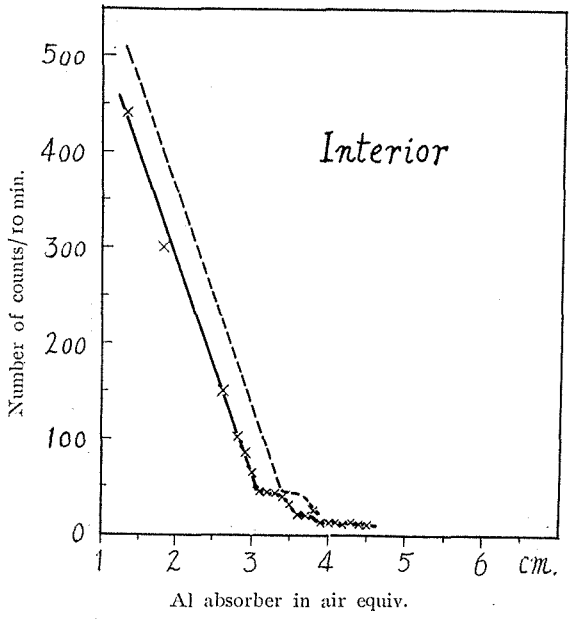


Table IV.

Interior			
Aluminium in mm. air range	Number of counts per 10 min.	Aluminium in mm. air range	Number of counts per 10 min.
13	438	35	35
18	308	36	20
26	150	37	24
28	106	38	32
29	86	39	14
30	66	40	16
31	53	41	14
32	46	42	10
33	46	43	14
34	40	44	8
		45	8

Table IV. Experimental conditions are quite the same as in Table III.

Fig. 3



contained in the sample.

For this purpose the number of α-particles from the natural surface of a sample and also from the interior was measured for various thicknesses of aluminium foil inserted between the ionization chamber and the sample.

The results are shown in Table III and Fig. 2, and Table IV and Fig. 3 for surface and interior respectively. In this case the diameter of window was 1.5 cm., the distance between source and window was 2 mm. (earthen cap was excluded), and the depth of the ionization chamber was 10 mm. For the purpose of comparing the activity of the surface to that of the interior, the number of counts at

the interior is given annexed to that of the surface in Fig. 2. We see a) that the activity of the surface is far stronger than that of the interior and b) that the range of α-particles i. e. the kind of radio-elements is quite different in the two cases.

As the abscissa of Fig. 2 (and 3) gives only the thickness (in air range) of Al foil inserted between the source and the window of the detector, the true curve should be displaced some 2 mm. (air part) to the right as shown in the broken curve. The end of each straight curve represents the range of α-particles, i. e. *ca.* 3.8 cm. and *ca.* 3.3 cm. in Fig. 2 and Fig. 3 respectively. The former is very near to the range of Po-α-particle and the latter to that of Ra-α-particle.

It is natural to conclude that the activity of the young natural surface is due to mainly polonium and that of the interior to radium and its products. This explanation accounts for the various facts we have mentioned concerning the activity of Hokutolite. Moreover, by the curve in Fig. 2 the presence of some long range particles in small amount is recognized. These are probably some members of the thorium series, although Hayakawa and Nakano[3] were convinced that none of these were present.

This method is certainly not a refined one for analysing radio-elements, but it is a rather convenient one for our purpose.

The ordinary accurate method is to measure the range-number curve by using the source of fine powder deposited on a glass plate by floating method, but experiment in this line has not yet been carried to a conclusion.

VII. Estimation of the Contents of Both Radium and Polonium

It is now possible to make a rough estimate of the radium and polonium content, though there can be no accurate measure of the polonium content because its value diminishes as a function of time.

Thin sources of fine powder granulated from the surface and the interior respectively of a sample, the activity of which was relatively strong, were prepared on glass plates by the floating method with ethyl alcohol. The thickness of each was 5.5 mg/cm^2. Bringing this source up to 3 mm. from the window of the ionization chamber, the depth of which was 10 mm., we counted the number of particles by insertion of an aluminium sheet of 1 cm. air equivalent. In this case the activity of U_3O_8 was used as standard. The numbers of counts per 1 mg per 10 minutes are as follows ;—

$(U_{I,II})_3O_8$～847, hence for 1 mg $U_{I,II}$
$U_{I,II}$～1016/mg/10 min. (1),
Simultaneously for Hokutolite { interior～8 (2), surface～35.4 (3).

By Geiger and Lord Rutherford[4], the total number of α-particles emitted by $U_{I,II}$ was found to be 2.37×10^4/g/sec. i. e. 14220/mg/10 min. The present values should also be multiplied by about 14 so as to estimate roughly the total number of α-particles emitted,

that is, for Hokutolite { interior～112/mg/10 min. (4), surface～496/mg/10 min. (5).

As in the interior the α-activities are mainly due to Ra and its four products, at least, up to RaC′ in equilibrium (see VI), 28 in 112 are due to Ra. Then we have radium contents of 1.3×10^{-9} g per 1 g of Hokutolite, by using 3.72×10^{10} particles per 1 g of Ra per second. On the surface $496 - 112 = 384$ particles per mg per 10 min. are mainly due to Po and the upper limit of concentration of this is 3.8×10^{-12} g per 1 g of the surface material, as 1 g of Po emits 10×10^{17} particles per 10 minutes.

Since 1.3×10^{-9} g Ra equilibrate with 2.6×10^{-13} g Po, the content of Po on the surface of Hokutolite is more than 15 times that of the direct derivatives of Ra.

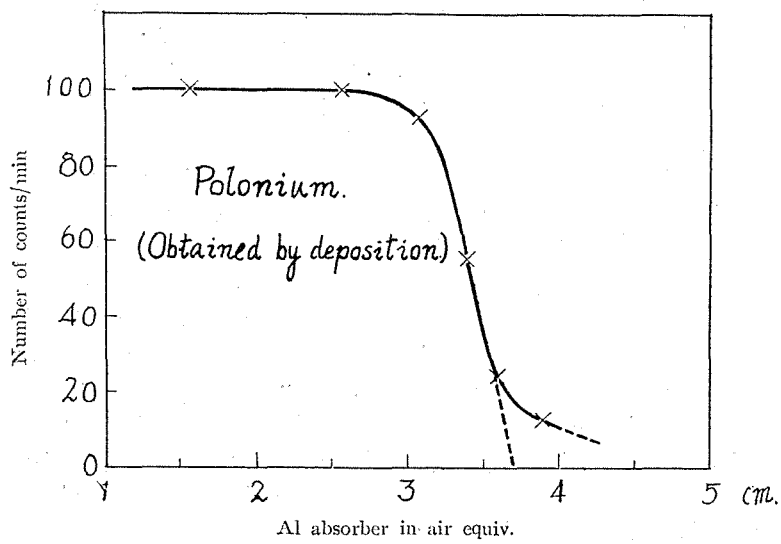
This result is very significant and shows that the activity of Hokutolite comes from two mother elements Ra and probably RaD which are thought to be deposited as sulphates in crystals of the mineral with the elements Ba and Pb,

VIII. Chemical Separation of Polonium

As the content of Po on the natural surface seemed to be large enough to provide the source of homogeneous α-particles, chemical separation was performed as follows:

After boiling about 100 g of fine powders of mineral with sodium-carbonate, the melt was thoroughly washed with water and dissolved in dilute hydrochloric acid. Immersing copper (or nickel or silver in some cases) plate in this solution for about 20 hours, we found Po—a strong α-active substance—deposited on the plate. The number of counts was 1958/min. By carefully adjusting the counting system, it was found that the α-particles emitted by the deposit are homogeneous as shown in Plate III a, in which the deflections are uniform, while those by the α-particles from natural surface are irregular (Plate III b). The range curve is given in Fig. 4.

Fig. 4



IX. Discussion (Process of Sedimentation)

From the above experimental results we see that the alpha-activity of Hokutolite is mainly due to polonium and radium with radium products. The radioactive equilibrium from radium up to radium C′ takes place after about 40 days starting from pure radium. Further most of the polonium, especially on the natural surface, is not a direct derivative of radium, because in respect to intensity the former is far stronger than would be expected from the equilibrium relation with the latter. Since an equilibrium from radium to radium D and consequently to polonium comes after about 110 years,* it is difficult to consider such intense activity of polonium on the young surface as due to radium. If we now take radium D which has a half period of 16 years* and equilibrate after 800 days* with polonium as mother element, the activity of the latter should then reach maximum and slowly decay keeping equilibrium with radium D. This character can not be proved by taking up real examples one by one, but may be theoretically argued from the following facts: (1) The activity on the surface does not seem to decay markedly for a number of years and (2) that of an old sample probably collected 30 years ago has diminished to less than half

* St. Meyer and Schweidler, Radioaktivität, p. 456.

that of a young one. Therefore samples of opaque and white colour, which show stronger activity than those tnat are gray and somewhat transparent (which is taken to be young), may probably be in a state of maximum intensity being in equilibrium with radium D. But on this point more samples and a longer period of observation are necessary to give complete explanation.

Now with this knowledge and the results of chemical analysis of both specimens and the spring water we can picture to ourselves the process of sedimentation of Hokutolite.

By chemical analysis the spring water was found to be strongly acid and to contain much sulphate with a very small amount of lead and barium. As Ishitani[5] pointed out, the spring water contains a large amount of radium emanation and so it may be inferred that radium is deposited with barium as sulphate on the surface of mother stone (i. e. andesite or sand stone), and radium D simultaneously with lead as sulphate, since all the daughter elements of radon soon change to radium D for their short half periods.

Other series, presumably a faint amount of thorium series, may possibly be contained in the surface as Fig. 2 suggests; but if so, since the half life of surface activity is short, they must be mesothorium (half period is about 6 years) and its products.

X. Summary

In the present work the author observed the radio-activity of α-particles of Hokutolite by a counter with linear amplifier and obtained the following interesting results:

1) The activity of the natural surface of young mineral is far stronger than that of the interior; for example, we have a sample in which the former is more than 10 times the latter.

2) The wide variation of the activity of surfaces in various samples, in spite of their being almost constant in the interior, is due not to difference in constituents of the specimens of Hokutolite, but to the lapse of time after the last sedimentation took place.

3) α-activity of the surface is mainly due to polonium (3.8×10^{-12} g) and that of the inside to radium (1.3×10^{-9} g) with its products being in equilibrium. The content of Po is more than 15 times that of direct derivatives of Ra.

4) Considering (2), it seems a large time-lag exists between the period of sedimentation of surface and that of interior at most 1 mm.

beneath it. Consequently, the sedimentation of this mineral must have been going on very slowly, or intermittently when favourable conditions prevailed.

5) Finally, the process of sedimentation has been found to be as follows ;—

i) The direct mother element of polonium in Hokutolite is RaD- the derivative of radon considerably contained in the spring.

ii) While $RaSO_4$ in the spring is deposited withB aSO_4, RaD SO_4 or $RaBSO_4$, directly derived from radon, crystalises out with $PbSO_4$.

Acknowledgement

In conclusion the writer wishes to express his sincere thanks to Prof. Dr. B. Arakatsu for his many helpful suggestions and valuable advice so generously given throughout the course of his work ; and to Prof. Dr. I. Hayasaka of Taihoku Imperial University for his kind support in collecting samples of Hokutolite. He is also indebted to Mr. M. Naito and Mr. Y. Uemura for their help in this work. Lastly his thanks are due to Hattori Hokokwai for financial support which rendered the study possible.

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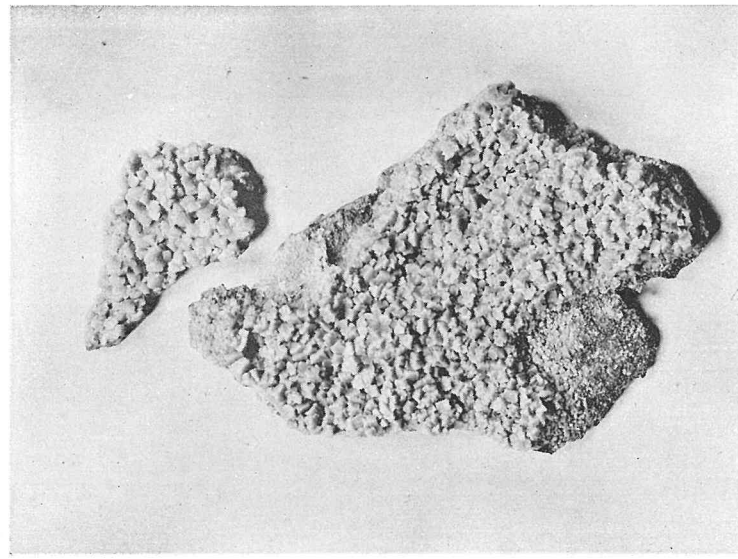
Plate I

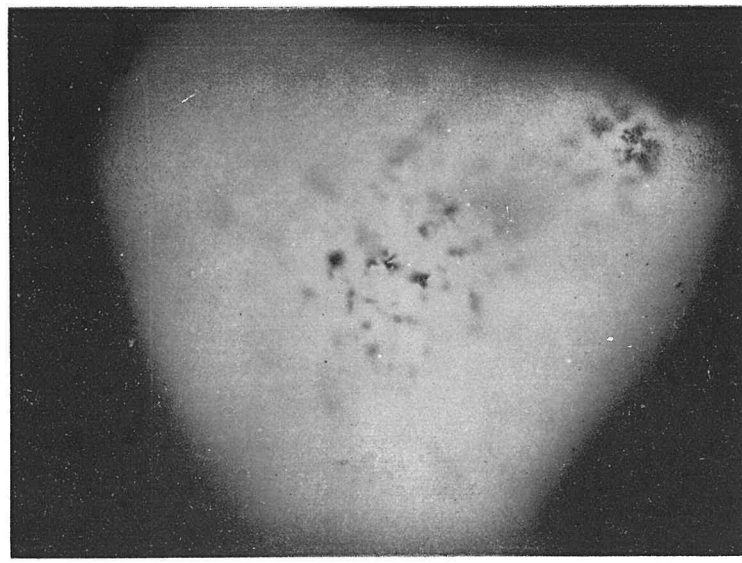
a——A part of Hokuto-kei

b——A large stone covered by Hokutolite of strong activity.

Plate II

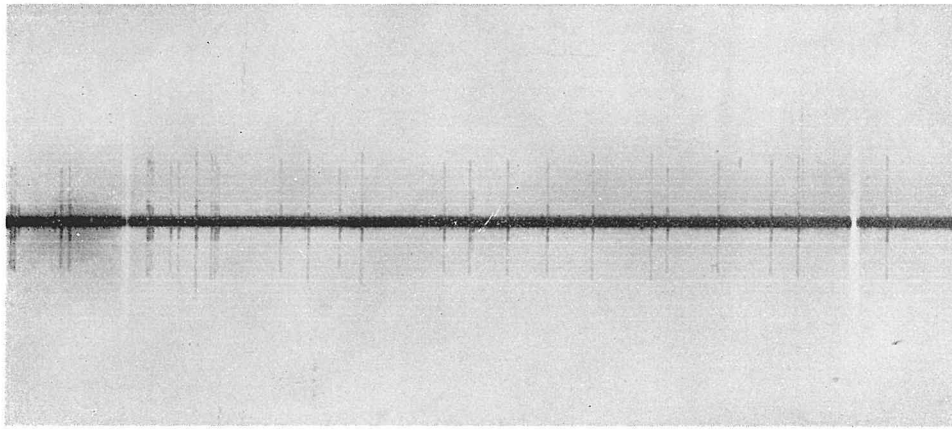


a——Hokutolite

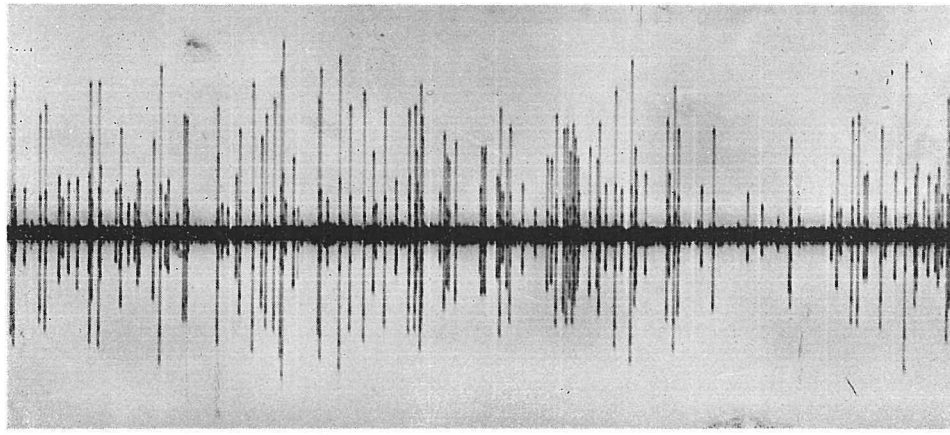


b——Blackening of photographic plate by Hokutolite through Al sheet of 1/100 mm. Exposure is 50 days.

Plate III



a——Polonium



b——Hokutolite