SEARCHES FOR DISTANT TRANSURANIUM ELEMENTS IN FERROMANGANESE NODULES

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Abstract

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SEARCHES FOR DISTANT TRANSURANIUM ELEMENTS IN FERROMANGANESE NODULES

In recent years a considerable number of theoretical and experimental works have appeared devoted to the problem of the existence of very stable distant transuranium elements ([1–10]).

However, theoretical studies, in which the region of existence of such elements is predicted with a high degree of reliability, obviously cannot give a sufficiently precise value of the half-life, nor of the atomic number Z for the longest-lived nuclide. On the other hand, experimental studies of natural minerals, carried out by Schintlmeister ([7]), Cherdyntsev ([8]), and also by Jerry and Adams ([9]), led to the detection of an unknown α-emitter with an energy of about 4.6 MeV, which, according to the data of the authors of the first two papers, has chemical properties close to osmium. However, the conclusion of the authors of work ([10]) that the observed α-activity belongs to a transuranium element—the chemical analogue of osmium ($Z = 108$)—does not appear sufficiently reliable. Apparently, other possible explanations of the observed effect have not yet been excluded.

In the work of P. Fowler and co-workers on the investigation of the heavy component of cosmic radiation, performed by the nuclear-emulsion method, the identification of very heavy particles with charge $Z = 104$–108 was reported ([11]). The charge of the particles was determined on the assumption that the track blackening density in the nuclear emulsion is proportional to the square of the particle charge. The results of work ([11]) were not confirmed in simultaneous experiments by Fleischer et al. ([12]) on recording the heavy component of cosmic radiation with plastic detectors.

Thus, the question of the existence of distant transuranium elements in cosmic radiation remains open; on the basis of ([11]) one can only indicate an upper limit for the flux of such nuclei, $\leq 5 \cdot 10^{-10}$ nuclei/cm^2, for the upper layers of the Earth' s atmosphere. Nevertheless, despite the absence of an unambiguous result from these works, it was impossible completely to exclude the possibility of the existence of primordial transuranium elements in terrestrial minerals and rocks ([13]).

According to detailed theoretical calculations carried out by Nilsson ([6]), the most stable nuclei should be in the region $Z = 110–111$. However, searches undertaken in the USA in 1968–1969 for transuranium elements–chemical analogues of platinum and gold–gave a negative result ([14,15]).

In setting up analogous experiments at the Joint Institute for Nuclear Research ([16,17]), the sensitivity of the recording methods was substantially increased–by 10^3–10^4 times compared with ([14,15]). For a number of reasons, other objects of investigation were chosen. In experiments to search for the effect of spontaneous fission in lead and its compounds ([16]), tracks of nuclear fission fragments were recorded with the aid of dielectric detectors.

The observed effect corresponds to an apparent period of spontaneous fission, referred to lead, of $(2–4) \cdot 10^{20}$ years. Control experiments showed···

established that this effect cannot be due to known sources of background. One of the possible explanations of this effect is the spontaneous fission of a heavy element–a chemical analogue of lead or of an accompanying element–with a half-life of more than 10^8 years, whose concentration in the samples is 10^{-12}–10^{-13} g/g ([16]). The results of work ([16]) received independent confirmation in experiments on the study of the effect of spontaneous fission in lead glasses and lead minerals, carried out by the proportional-counter method ([17]).

Despite the high sensitivity of the methods described above ([16,17]), the search for distant transuranium elements requires the examination of a large number of samples and of substantial quantities of the substances under study. In addition, with such an experimental arrangement it does not appear possible to register the spontaneous fission of transuranium elements of cosmic origin because of their extremely low concentration in terrestrial minerals and rocks ($\leq 10^{-16}$ g/g). It was necessary to find samples in which, over very long intervals of time, both distant transuranium elements of terrestrial origin and transuranium elements from the primary component of cosmic radiation could have become concentrated. A. P. Vinogradov drew our attention to the possibility of using ferromanganese concretions as such samples.

Table 1

Sample No.	Area, mm^2	Number of tracks	Track density per 1 mm^2
1	0.075	9	120
2	0.24	21	87
3	0.55	32	58
4	0.38	18	47
5	0.76	31	41
6	0.47	14	30
7	0.78	10	13
8	1.20	9	7

Such formations have a number of advantages in comparison with all other samples.

1. Concretions are a selective adsorbent for a number of heavy elements, including Pb, Tl, Hg, W; the enrichment coefficient of such elements is 10^5–10^6 in comparison with ocean water.
2. The age of concretions ranges from several hundred thousand to millions of years.
3. Concretions are usually found at depths of several kilometers under water, which practically completely excludes background from the fission, by cosmic rays, of heavy-element nuclei contained in them.
4. Concretions and the inclusions of organic and mineral origin contained in them can in principle serve as detectors of nuclear-fission fragments.

We studied concretions collected on the floor of the southern part of the Pacific Ocean. Experiments were undertaken in order to reveal traces of fission fragments in the surface layer of shark teeth contained in the concretions. These experiments, however, were not crowned with success, which is possibly connected with the relatively loose, finely crystalline structure of the dental tissue of these samples. In some of the concretions studied, small transparent glassy bodies with volumes up to several cubic millimeters were found. Subsequent analysis showed that these inclusions are minerals of the feldspar group.

To develop the etching procedure, these minerals were irradiated with a flux of 10^5 fragments/cm^2 of Cm^{244}, then treated for 5-30 min in 3.5% hydrofluoric acid at room temperature and examined under a microscope at a magnification of 450–900×. The traces of fission fragments had the form of rhombic hollow channels up to 3-4 μ wide and up to 7-8 μ long. Minerals extracted from the concretions were treated under the conditions indicated above and examined under a microscope. Tracks with a length of not less than 2 μ were recorded.

The results of examination of the samples are given in Table 1.

The observed effect could have been caused by various reasons:

1. Spontaneous fission of uranium contained in feldspar crystals.
2. Spontaneous fission of nuclei adsorbed on the surface of feldspar crystals.
3. Spontaneous fission of uranium contained in a thin layer of concretion material (1—1.5 mg/cm^2) adjacent to the surface of the mineral.
4. Spontaneous fission of nuclei of an unknown transuranium element contained in the layer of concretion material adjoining the minerals.

To test the first assumption, measurements were made of the density of fission-fragment tracks inside some of these samples. Over an area of 2.8 mm^2, not a

single fission-fragment track was found, which corresponds to an upper limit for the effect inside the minerals of 0.4 tracks/mm^2. This is tens and hundreds of times smaller than the track density on the surface of the minerals.

To test the assumption concerning adsorption of spontaneously fissioning elements on the surface of the minerals, the lengths of 40 fission-fragment tracks were measured. It was found that the fission-fragment tracks have a continuous distribution of lengths from 2 to 11 μ, which contradicts the assumption of adsorption of a thin layer of fissioning elements on the surface of the crystals.

For a final conclusion about the nature of the observed effect, data on the uranium content in the concretions and on their age are of decisive importance. The uranium content in the concretion material in which feldspar minerals were found was determined by the activation-analysis method ([18]). It turned out that in different parts of the concretions the uranium content is approximately the same and amounts to $(2—3) \cdot 10^6$ g/g. Thus, in order to explain the observed effect by spontaneous fission of uranium, it is necessary to assume that the age of the concretions is from 10—15 to 25—30 million years for the majority of the samples studied.

Sufficiently reliable measurements of the age of ferromanganese concretions have not yet been carried out. Indirect conclusions about the upper limit of the age of concretions may be obtained from the rate of deposition of bottom sediments in the World Ocean ([19]). According to these data, the rate of deposition of silts in the southern part of the Pacific Ocean is 0.3—0.5 mm per 1000 years. Since the concretions we studied were collected by surface dredging of the ocean floor, it may be concluded that their age does not exceed 1 million years.

Other information on the age of the concretions we studied was obtained by the paleontological method at the Institute of Geology and Geochronology of the Precambrian of the USSR Academy of Sciences. Specimens of shark teeth contained in the central parts of the concretions were assigned to the Quaternary period (0—1 million years) and the Pliocene epoch (1—9 million years) ([20]), i.e., the age of the concretions is apparently from several hundred thousand to several million years.

Measurements of the growth rate and growth time of concretions, carried out by radium and ionium methods ([21—23]), give different results. On the basis of these measurements, it was possible only to conclude that the age of the concretions is from hundreds of thousands to 1—2 million years.

We had the opportunity to make an independent determination of the upper limit of the age of the concretions, based on the density of fission-fragment tracks measured by us in minerals from the central regions of the concretions, 0.4 tracks/cm^2, and on the uranium content in these inclusions, $(6—7) \cdot 10^{-8}$ g/g. Hence the upper limit of the age of the concretions studied by us is 2—2.5 million years.

On the basis of the upper age limit and the measured uranium content in the

concretions, it follows that, for the majority of samples, spontaneous fission of uranium can account for no more than 10—20% of the observed effect (Table 1). One of the possible explanations of the observed excess-

···the traces of fission fragments is the hypothesis of spontaneous fission of distant transuranium elements of terrestrial or cosmic origin, present as a minor admixture in the material of the concretions.

The difference in the effect observed on different samples of concretions (Table 1) may be due to different concentrations of spontaneously fissioning elements in the concretions, owing to differences in the adsorption of heavy elements and in the growth rate of the adjoining layer.

In conclusion, the authors consider it their duty to express their deep gratitude to Prof. P. L. Bezrukov for placing at our disposal numerous samples of iron-manganese concretions and for very useful discussions. The authors are also grateful to L. S. Glikman for his determinations of the ages of teeth from concretions, and to Yu. A. Vinogradov, S. P. Tretyakova, and K. I. Merkina for assistance in carrying out these experiments.

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Note: Figure translations are in progress. See original paper for figures.