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THE PROSPECTS FOR APPLICATION OF NEUTRON-FRAGMENT RADIOGRAPHY IN STUDY OF LOCAL DISTRIBUTION OF URANIUM IN THE BAIKAL SEDIMENTS FOR PALEOCLIMATIC RECONSTRUCTIONS

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Data on the local distribution of uranium throughout the bottom sediments of Lake Baikal (Akademichesky Ridge, VER-95-2, St 3BC, 53°113′12′′N, 108°25′01′′E) have been first obtained using neutron-fragment radiography. Uranium is present in the sediments in various forms: evenly dispersed, in clays and diatoms; micro- and macroinclusions in highly radioactive minerals (uranium concentrators); and evenly dispersed, at microsites, with its contents 10–50 times exceeding those in clays and diatoms. Relative and absolute concentrations of uranium can be reliably determined in any mineral. The uranium distribution throughout the Baikal bottom sediments is regular. Using (*n*, *f*)-radiography for studying the sediments provides new information necessary for paleoclimatic reconstructions.

Bottom sediments, uranium, (n, f)-radiography, Baikal

International projects aimed at decoding the climatic past of the Earth [1] have recently contributed much to the complex study of Lake Baikal. It was established that one of the most sensitive indicators of changes in paleoclimate was distribution of trace elements, first of all U, in bottom deposits of Baikal. Uranium and molybdenum are positively correlated with amounts of diatoms (indicators of warm climate) as well as biogenic silica and organic carbon [2]. Direct relationship exists between U anomalies in Baikal deep-water deposits and diatom oozes enriched in humic acids [2, 3]. At the same time, analysis of the clay, diatom, and terrigenous fractions separated by an aerodynamic method indicates that the diatom component is almost sterile relative to U [4].

Though the study was carried out thoroughly, with the use of modern analytical techniques, it is evident that distribution and speciation of elements in the Baikal bottom deposits are required to be studied by local nondestructive methods. The most appropriate method is neutron-fragment (n, f) radiography. It determines U with high sensitivity (10^{-5}–10^{-6} wt.%) and locality and estimates a possible form of its occurrence and distribution pattern in the object of analysis [5, 6].

This work reports first results on U distribution in the Baikal deep-water sediments analyzed by (n, f)-radiography and considers prospects of the use of activation radiography for this purpose. For analysis, a core was taken from the borehole drilled in the Baikal sediments at the top of the Akademichesky Ridge, station VER-95-2, St 3BC, H = 265 m, 53°113′12′′ N, 108°25′01′′ E [1]. This interval of sediments demonstrates the transition from diatom ooze to clay sediment with rare diatoms. A block 35 mm wide and 15 mm thick was cut from the core and subjected to freezing vacuum drying and then saturated with an epoxide compound in a vacuum. Two polished preparations (sections) were made from the central portion of the bottom-sediment material cemented in this way. The support was made of quartz glass. Synthetic fluorphlogopite and lavsan served as solid track detectors (STD). The preparations were radiated by thermal neutrons on the reactor of the Tomsk Institute of Nuclear Physics (integral flux $1 \cdot 10^{16}$ neutron/cm^2). Etching of detectors was made by a known procedure [5, 6]: lavsan — in a 40% KOH solution, and fluorphlogopite — in a 20% HF solution.

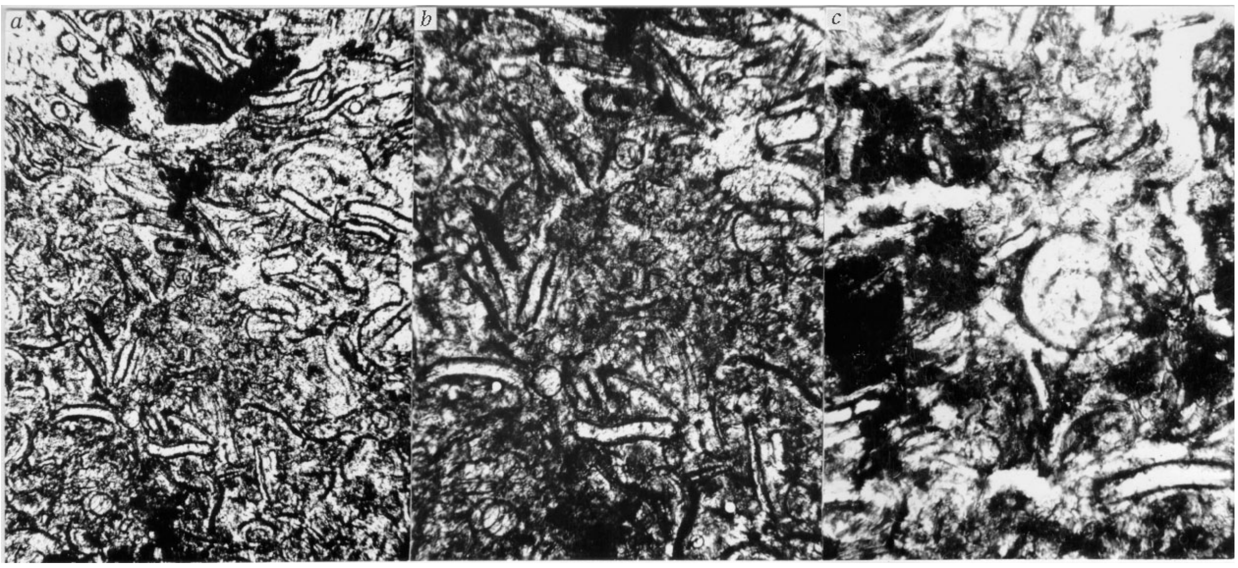

Fig. 1. Microphotographs of the material of column (VER-95-2, St 3BC) of bottom deposits in Lake Baikal. Sites with the highest concentrations of uniformly distributed ("dissipated") U are diatomites with an admixture of clay minerals and iron hydroxides. Relatively large "fragments" are terrigenous minerals — magnetite, quartz, feldspars, etc. *a* — × 250, *b* — × 500, *c* — × 1000.

Study of the distribution of tracks of ^{235}U fission fragments in STD and quantitative calculations were carried out with the use of the JENAVAL microscope. The best resolution and quality of tracks were obtained on a synthetic fluorphlogopite STD. To determine relative concentrations of U, the tracks were counted with a 1000-fold magnification over an area of 0.005 mm^2 from the central portion of 1 mm wide interval. When counting the tracks corresponding to a dissipated form of the occurrence of U, the sites corresponding to highly active inclusions of U-concentrating minerals and "clusters" (sites with elevated contents of dissipated U) were not taken into account. The number of these concentrating minerals and "clusters" was counted separately, plotting their distribution patterns in the column. The plots of distribution of the number of concentrators are of qualitative nature and do not reflect the U concentration in the column of bottom deposits as opposed to data on distribution of the dissipated form of U occurrence.

The material of the investigated interval of bottom deposits of the Akademichesky Ridge on Lake Baikal includes: upper 5 cm — diatom ooze, and lower 10 cm — clay sediment with an admixture of diatoms from a few to 10–20 vol.%. The diatom material makes up 85–95% of the diatom bed (Fig. 1), whereas the clay bed is made up chiefly of brown disperse clay minerals (probably, with iron hydroxides) measuring less than 50 μm. In places, quite large fragments of terrigenous minerals occur: quartz, feldspar, and ore minerals (magnetite); their amount is generally elevated in the clay part of the column.

Variations in relative concentrations of U (expressed in tracks/mm^2) in the bottom deposits from VER-95-2, St 3BC (1 mm spaced) are shown in Fig. 2. It follows from these data that the Baikalian sediments are characterized by variations in U concentration, owing to different factors whose nature is mostly veiled, though their relationship with changes in environmental conditions is evident. Evidence of the effect of different agencies on the pattern of U distribution in the Baikalian deposits comes from trend lines plotted by methods of smoothing (moving average), linear approximation, and polynomial approximation, based on data of U distribution in the column. Against the background of "high-frequency" variations in contents of U, its distribution shows cyclicity with periods of 6.5–7 ka and about 25 ka. The linear trend may be evidence of higher-order periodicity in U distribution.

Throughout the investigated interval (VER-95-2, St 3BC) of bottom deposits, U with different types of spatial distribution, possibly corresponding to different forms of occurrence, is documented in various quantitative ratios. The most typical pattern of U is uniform, without signs of layering. It was recorded both in diatom oozes and in diatom-clay (clay) material of sediments, with 5–10-fold variations in U contents (Fig. 3).

A radically different form of occurrence is ubiquitous. It is related to U-concentrating minerals, such as zircon, apatite, ilmenite, and others, and is detected as fields with high density of tracks or specific "stars"

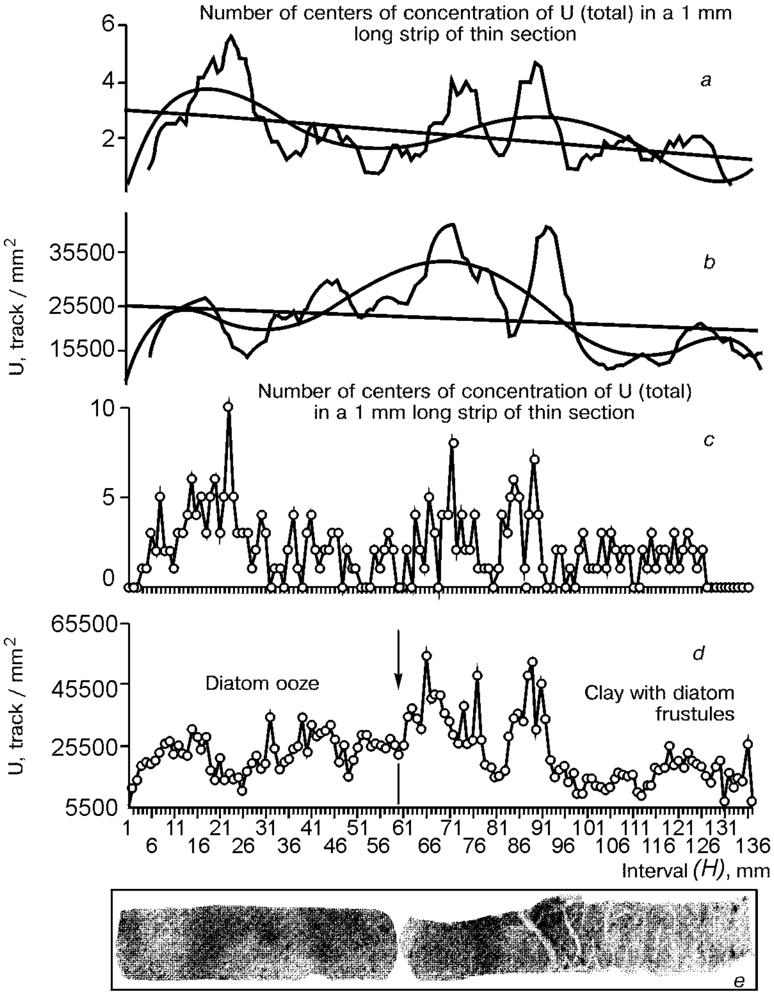

Fig. 2. Variation in concentrations of "dissipated" U (*d*) and the number of centers of U concentration (sum of accumulated large and small "stars") (*c*), within the 150 mm interval of the core (st. VER-95-2, St 3BC. St. 16; see Fig. 4 in [1]) of bottom deposits of the Akademichesky Ridge of Lake Baikal (1 mm spaced), according to (*n,f*)-radiography data. *a*, *b* — Trend lines constructed by methods of smoothing (moving average), linear approximation, and polynomial approximation for data presented on plots *c* and *d*. Against the background of variations in U contents, cyclicity in U distribution with a period of about 6.5–7 and 25 ka is observed, yielding 4 cm thick sediment for 1000 years. The linear trend can be evidence of the higher-order periodicity. Arrow shows the transition zone between diatom ooze and diatom-admixed clay (diatom-free ooze according to [1]), represented by the lithified Fe-Mn crust. *e* — (*n*, β)-Radiograph (beta-radiograph) recording a spatial distribution of Eu, Co, and, to a lesser extent, Ce and Sc in the investigated column of bottom deposits of Lake Baikal (two sections). Variations in blackening density on the radiograph reflect variations in concentration of the above-mentioned elements in the sediment. Small dark points are accessory minerals concentrating Eu, Co, Ce, and Sc. Cracks of clay shrinkage are well visible in the right part of the radiograph (*e*). Composition of radionuclides in the irradiated section was determined at the Analytical Center of UIGGM, Novosibirsk, on the Intertechnique equipment for neutron-activation analysis. Analyzed by V. S. Parkhomenko and S. T. Shestel'. Emulsion AF, exposure — 240 hours.

resulting from an imperfect contact between the substance and the detector (Fig. 4). This group may also include nonidentifiable microinclusions of U and U-bearing minerals of extremely minute sizes (less than

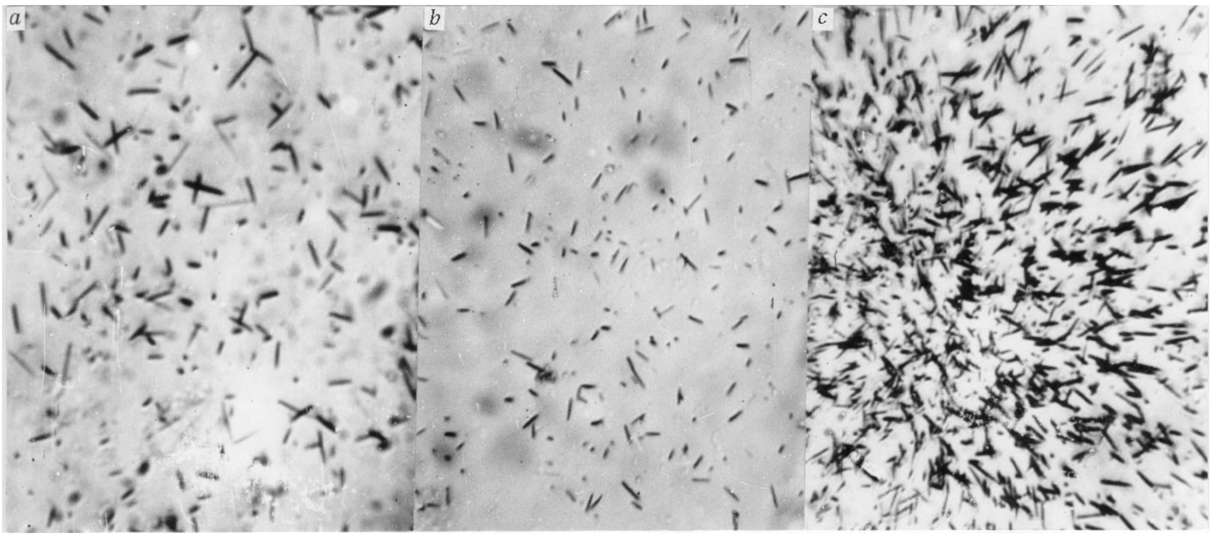

Fig. 3. Neutron-fragment radiographs recording the spatial distribution of U in Baikal deposits. *a* — In clay-diatom, *b* — in clay; *c* — (*n*,*f*)-radiographs of a site with increased concentrations of uniformly dissipated U among clay-diatom material. *a*, *b* — × 450, *c* — × 400. Integral flux is $S = 1 \cdot 10^{16}$ neutron/cm^2. Number of tracks per unit area of detector reflects the U concentration in the substance analyzed.

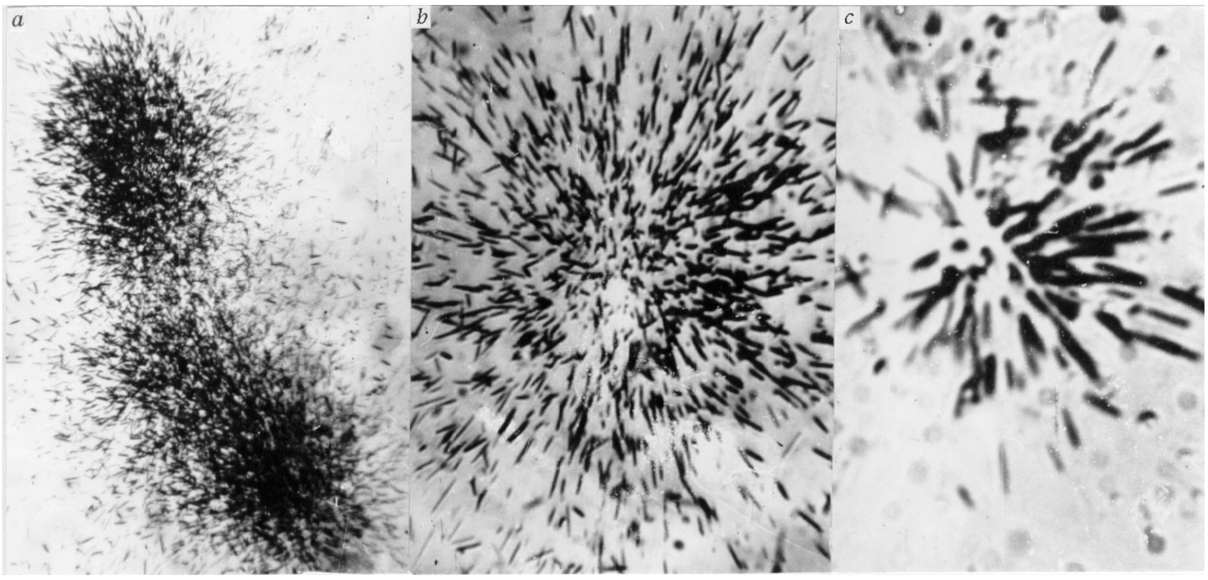

Fig. 4. Neutron-fragment radiographs recording spatial distribution of U in terrigenous U-concentrating minerals from separate portions of the column of diatom and clay deposits of Lake Baikal. *a* — Ilmenite, × 250; *b* — micro-inclusions of U-bearing minerals, × 450; *c* — ultramicroinclusion, × 1000. Detector is synthetic fluorphlogopite. Integral flux is $S = 1 \cdot 10^{16}$ neutron/cm^2.

0.1 μm) (see Fig. 4, *d*). Uranium-concentrating minerals and microinclusions are most frequently met in diatom-clay and clay intervals of the columns. They are much more rare in diatom oozes.

In addition, "fields" of isometrical form, up to 0.001 mm^2 in area, have been recognized in clay-diatom deposits. Here, concentrations of uniformly distributed U are many factors of ten higher than its contents

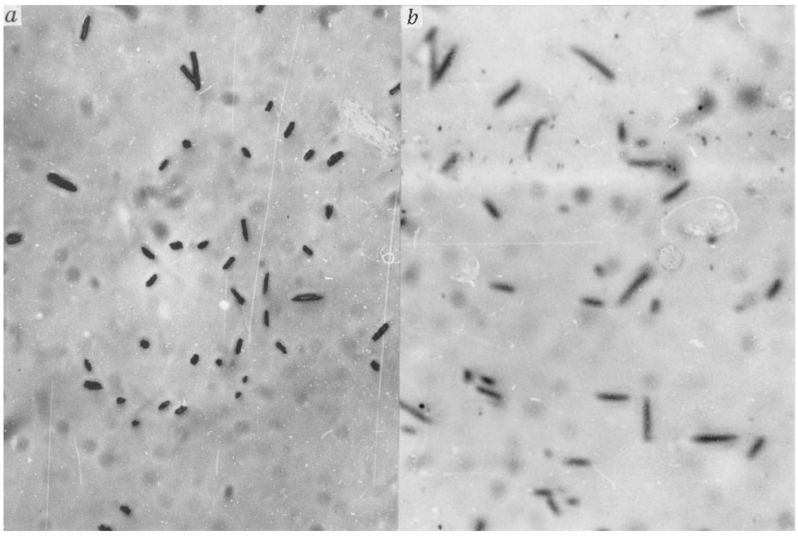

Fig. 5. Neutron-fragment radiographs (*a*, *b*) recording the spatial distribution of U in diatom oozes. The photograph shows orientation in distribution of U atoms in connection with diatoms. × 1200. Detector is synthetic fluorphlogopite. Integral flux is $S = 1 \cdot 10^{16}$ neutron/cm^2.

recorded in the clay and diatom components (see Fig. 3, *c*). It is now determined that these "fields" are situated above the sites that abound in diatom frustules; it is likely that they had appeared during sedimentation.

Analysis of solid-state track detectors with a 1000× magnification above the column intervals with abundant diatoms often demonstrates an oriented distribution of U decay track, whose pattern corresponds to morphology of diatom frustules in the sediment (Fig. 5). This fact is required to be thoroughly checked, with study of U distribution in thin sections of diatoms in both nondestructured sediment and separated fractions in order to establish forms of U occurrence in diatoms.

Thus, the results of study evidence that qualitatively new information on distribution and forms of occurrence of U in the bottom deposits of Lake Baikal can be obtained by neutron-fragment radiography. The terrigenous component of U in the bottom deposits is recognized safely and can be taken into account. Changes in U content in the process of sedimentation can be recorded within years and months, taking into account that the rate of sedimentation in the explored part of the Akademichesky Ridge in Lake Baikal is 4–6 cm/ka [1, 7] and the locality and resolution of (n, f)-radiography of U are about a few microns. It is quite possible that the revealed signs of "multilevel" periodicity in U are caused by different agencies, and additional detailed studies are required to recognize them. It is necessary to test and comprehensively study the factors permitting us to recognize the oriented distribution of U in diatoms and sites ("fields") with high contents of uniformly distributed U in clay-diatom formations. This makes possible to determine forms of association of U with diatoms and, therefore, the role of diatoms in accumulation of U by Baikalian sediments and its distribution in the waters of Baikal.

The method of activation track radiography is also appropriate for studying the local distribution not only of U but also of B and, with the use of beta-radiography, some other elements. Thus, Fig. 2 shows a beta-radiographic record of distribution of Eu and Co and, to a lesser extent, Cs and Sc in the investigated column of the Baikal bottom deposits. It demonstrates the alternation of sites ("horizons" in column) enriched and depleted in accessory minerals. In general, U-enriched sites also contain microinclusions of Eu, Co, Cs, and Sc.

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