

Experimental observation of the distortion of the uranium isotopic relationship and violation of the thorium- 234 secular equilibrium upon electric explosion

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ABSTRACT. Experiments dealing with a powerful electric explosion of titanium foil in a solution of Uranyl sulfate in distilled water are described. The experiments unambiguously show a distortion of the initial isotopic relationship of uranium and a violation of the secular equilibrium of thorium - 234.

1 Introduction

In the previous study [1], the results of tentative experiments on the electric explosion of metallic foils in liquids were reported. A noticeable distortion of the natural isotopic composition of titanium, in particular, a decrease in the ⁴⁸Ti proportion (to ~10%) was observed in experiments.

These results were so unexpected that the experimental facts reported in this publication [1] had to be verified. V. D. Kuznetsov’s group (Joint Institute for Nuclear Research, Dubna town) carried out verification experiments [2] and obtained results similar to [1]. In addition to the procedures used in [1] to determine the elemental composition and the isotopic relationships, they employed γ -activation analysis, which allowed them to elucidate a number of subtle regularities.

The effect of the electric explosion of titanium foils on a liquid dielectric was studied in a series of experiments in which a solution of uranyl sulfate in distilled water was used as the “dielectric”. The uranyl sulfate concentration varied from 10^{-2} to 10^{-1} mol/liter.

2 Experimental results

The setup used in the experiments was the same as described in the previous publication [1] with the only difference that the explosion chamber was sealed. The design of the explosion chamber is presented in Fig.1. The chamber was represented by a polyethylene unit placed in a hermetically sealed steel vessel. To ensure the possibility of comparing the results obtained with the results of previous publications [1,2], the current intensity through the load was the same as in [1]. The weight of the titanium foil (load) in the experiments was $m=0.09 \pm 0.01$ g.

As a current pulse is applied to the titanium foil, the foil explodes [3] and the pressure in the explosion chamber sharply increases. For this reason, most of the uranium solution is ejected into prechamber 6 through sealing 5. This gives two separate portions of the solution, which differ appreciably from each other, as follows from the measurements performed.

Below, the sample taken from the lower portion is conventionally called the precipitate (P), while the sample taken from the upper portion is called VGB (vapor gas blowout).

The samples were taken with a single-use syringe and placed in 1.5- cm^3 , 2- cm^3 , or 5- cm^3 standard plastic tubes. The tubes were sealed by PARAFILM “M”. An about 0.5- cm^3 portion of the sample was immediately dispatched to analysis on a mass spectrometer.

Laser mass spectrometry was chosen for determination of the elemental and isotopic compositions; having a relatively high sensitivity (10^{-4} – 10^{-5} at %), this method provides an error of determination of trace impurities at a level of 10-15% [4].

The water was evaporated and the dry residue was ground in a mortar with ethanol (the mortar material, either agate or leucosapphire, does not affect the results of analysis). The sample pressed under 200 atm was subjected to standard analysis. Primary attention was devoted to determination of the isotopic compositions of elements. The accuracy of determination of the elemental composition in model samples was 1.5 to 2.0% based on minor isotopes or less than 1% based on major isotopes, which is consistent with known published data [4].

Two identical samples (the test sample and the blank) were prepared for γ -ray spectrometric measurements. A 40- cm^3 germanium detector (made of

extra pure grade germanium), Canberra company, was used as the gamma-ray spectrometer. The energy resolution of the gamma-ray spectrometer determined based on the ^{137}Cs line was 1.8 keV.

In order to avoid possible errors, the test and blank samples were measured alternately. In order to eliminate the geometric factor, the samples were fastened tightly against the detector end wall using a special holder. The measurement geometry is shown in Fig. 2.

The recorded spectra were processed in the energy regions of 92 keV (91.38 keV and 92.8 keV, the γ -line of ^{234}Th , which is the daughter product of ^{238}U) and 185.7 keV (the γ -line of ^{231}Th , the daughter product of ^{235}U). The areas of the peaks were found by approximating them by Gaussians. In the region of the 92.5 keV doublet, $K\alpha$ are the X-ray lines of actinides and the γ -lines of ^{231}Th , which form a complicated background, $K\alpha_1$ $K\alpha_2$ are the uranium lines due to uranium self-fluorescence, the $K\alpha_2$ peak giving a contribution to the doublet at 92.5 keV. The contribution of thorium $K\alpha_1$ $K\alpha_2$ lines related to the decay of ^{235}U is too low. Figure 3 shows the spectrum in the 86-102 keV range and the result of computer resolution of this spectrum into components.

To provide the possibility of comparing the results of γ and mass spectrometry, they are presented as the ratio $R = (^{235}\text{U} / ^{238}\text{U})_{\text{sample}} / (^{235}\text{U} / ^{238}\text{U})_{\text{blank}}$, which represents the separation factor of the isotopes.

The results of measurements of the time dependence of R_γ for sample No. 970-P with an uranyl sulfate concentration of 10-1 mol/liter are shown in Fig. 4. The value $t_0=0$ corresponds to the "shot" instant. The Figure shows the time variation for $T=24.5$ days, which corresponds to the periods of accumulation of ^{234}Th and ^{234m}Pa .

The results of measurements were approximated by the expression

$$R_\gamma = a / (b \cdot \exp(-t \ln 2 / 576) + c \cdot (1 - \exp(-t \ln 2 / 576))) \quad (1)$$

Here a is the parameter characterizing the change in the number of ^{235}U nuclei in the sample with respect to the blank ($a = (\text{the intensity of the } 186 \text{ keV line in the sample}) / (\text{the intensity of the } 186 \text{ keV line in the blank})$), b is the parameter characterizing the change in the number of ^{234}Th nuclei in the sample with respect to blank, c is the parameter characterizing the change in the number of ^{238}U nuclei in the sample with respect to blank. The parameter a in expression (1) is free and, as shown by the measurements, time-independent. The magnitude of a averaged over all measurements is 0.91 ± 0.03 . The electric explosion of the titanium foil was found to induce appre-

ciable changes in the number of the ^{234}Th nuclei in the sample with respect to the blank for the sample taken from the explosion chamber. At the initial point of time after the electric explosion of the titanium foil, a deficiency of the ^{234}Th ions (to 30%) is observed in the solution under study. The parameters a and c were determined by fitting, their ratio giving the value $R\gamma(t = \infty) = (0.98 \pm 0.03)$, which is in good agreement with the results of mass spectrometric analysis $RMS = (0.97 \pm 0.02)$.

Mass-spectrometric analysis of the solution ejected to the upper section of the steel vessel (VGB) averaged over a series of experiments gave $RMS.VGB = 1.18 \pm 0.07$. The average separation coefficient for the precipitate P over a series of nine electric explosions was $RMS.P = 0.942 \pm 0.012$. The weight of the titanium foil (load) was 0.09 g.

During conduction of the electric explosions, the neutron yield was monitored. To attain a sufficient efficiency of the fission neutron count, a special neutron detector was assembled from nine slow neutron counters (SNM-17). The whole assembly was surrounded by a 8 cm-thick polyethylene moderator. In order to evaluate the efficiency of neutron count, a calibrated neutron source was placed inside the explosion chamber. The efficiency of the fission neutron count was found to be 0.04%.

The protection from electromagnetic noises was attained by placing the whole detector unit and preamplifiers inside a copper screen. The possibility of excluding spurious counts (common-mode signals) was attained by dividing the slow neutron counters into two groups. Each counted pulse was identified as a neutron based on the pulse amplitude, time, and shape. It follows from the measurements that the flow of neutrons did not exceed $I < 103$ neutron/pulse.

3 Conclusions

The experiments carried out here are rather tentative. However, despite this fact, the experimental results provide a number of rather reliable and unambiguous conclusions.

1. The electric explosion of the titanium foil induces distortion of the initial isotopic relationship of uranium in a solution of uranyl sulfate. The most pronounced distortion of the isotopic relationship is found in the vapor gas blowout VGB, which contributes to the ^{235}U enrichment. $RMS.VGB = 1.18 \pm 0.07$ for VGB and $RMS.P = 0.942 \pm 0.012$ for the precipitate.
2. A substantial violation of the ^{234}Th secular equilibrium occurs. Immediately after the electric explosion, a deficiency of ^{234}Th up to 30 % is observed. Analysis of the curve shown in Fig. 4 shows that the amount of ^{235}U decreases by approximately 10 %, while the

amount of ^{235}U decreases by approximately 10 %, while the amount of ^{234}Th decreases to even a greater extent, the content of ^{234}Th being restored over a period of 24 days.

3. The neutron flow did not exceed $I < 10^3$ neutron/electric explosion. Therefore, the change in the uranium isotopic composition cannot be attributed to the induced fission.
4. To elucidate the mechanism of the observed effects, further research is needed. It is noteworthy that the effects are observed only upon electric explosion in a liquid (a solution of uranyl sulfate) in which high pressures are generated. Thus, the phenomenon observed might be of the same nature as that described in publication [5] for the collapse of artificially generated cavitations in a liquid.

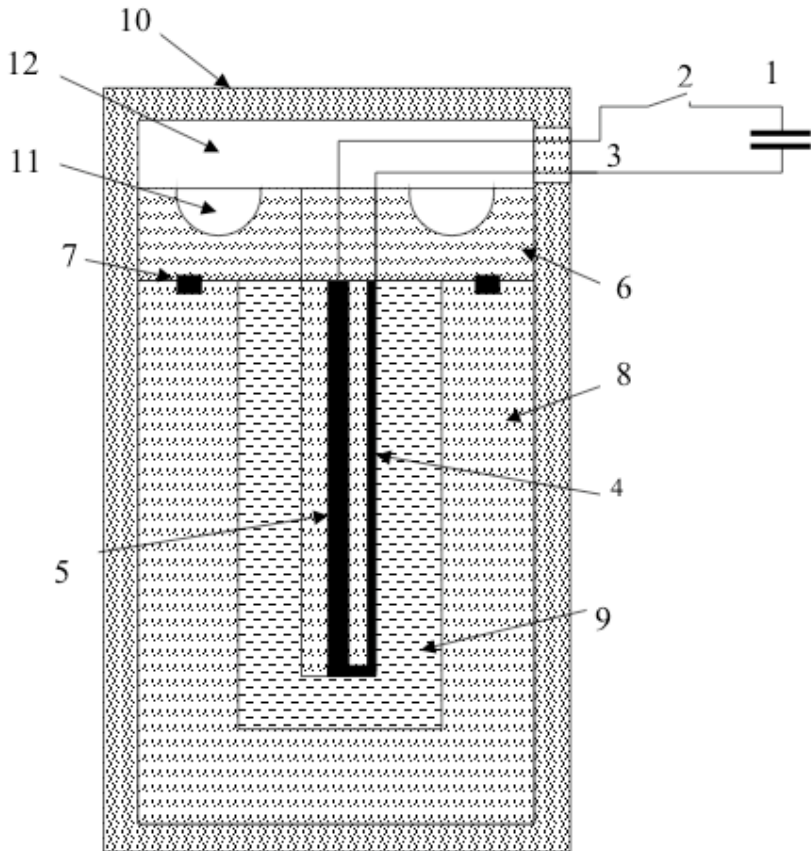


Fig. 1. (1) capacitor bank; (2) discharger; (3) cable; (4) foil; (5) electrode; (6) polyethylene cover; (7) sealing; (8) explosive chamber; (9) a solution of UO_2SO_4 ; (10) stainless-steel casing; (11) a cavity for the VGB; (12) pre-chamber.

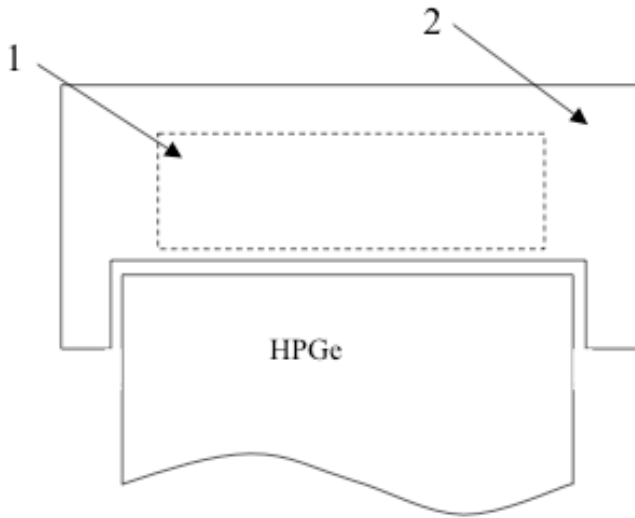


Fig. 2. Geometry of measurements. (1) container with the sample; (2) Co-axial holder.

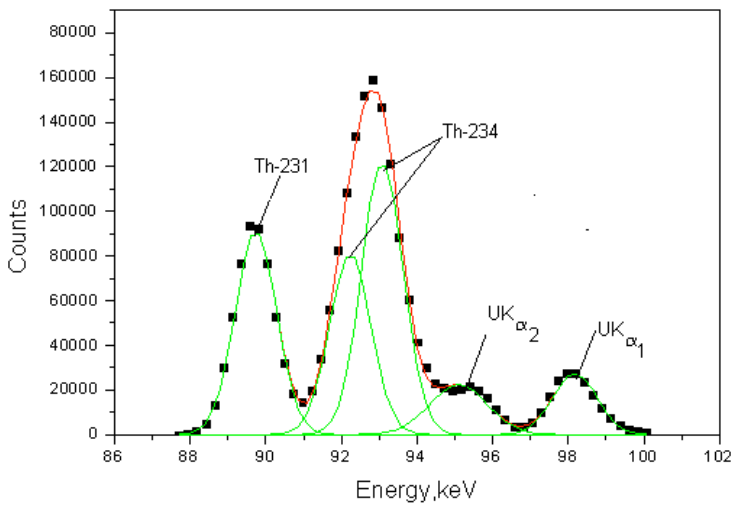


Fig.3. Deconvolution of the spectrum of the test sample in the energy region of 86-102 keV.

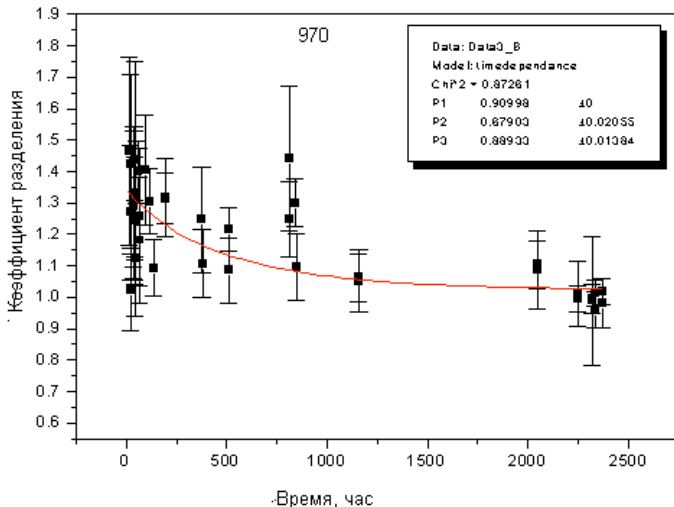


Fig.4. Separation factor vs. time

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