ESTIMATE OF THE $^{242m}$Am CONTENT IN FUEL OF THE FOURTH POWER UNIT OF THE CHERNOBYL NUCLEAR POWER STATION

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It is well known that the even—-even curium isotopes $^{242}$Cm (half-life $T_{1/2} = 162.9$ days) and $^{244}$Cm (18.1 yr) form spontaneous fission neutron sources in irradiated nuclear fuel. In addition $^{242}$Cm appears when $^{242m}$Am decays (141 yr). A fragment of its decay scheme is given in Fig. 1. There is undoubted interest in estimating the content of $^{242m}$Am and that of the daughter $^{242}$Cm in the fuel of the fourth power unit of the Chernobyl nuclear power station, and also the specific activity of the spontaneous decay neutron source formed by the $^{242}$Cm.

The question of the content of transuranic elements in the fuel of the fourth power unit was most fully considered in [1, 2]. These publications give calculated data on the buildup of almost all the fission products and transuranic elements up to the time of the accident, but there are no calculations of the amount of $^{242m}$Am produced. The methodical approach presented in the present paper is based on an analysis of the measured radionuclide content of the fallout and on a calculation of the content of radionuclides in the fuel in the reactor before the accident.

The physicochemical properties of the radioactive contamination of the territory caused by the accident at the Chernobyl nuclear power station are a consequence of the action of a complex aerodisperse system consisting of aerosols of various kinds [3]:

- fuel particles of dispersed fuel (uranium dioxide);
- hot particles of dispersed substance from a mixture of isotopes of ruthenium, rhodium, silver, and technetium which have emerged from the uranium dioxide at high temperature;
- aerosols formed in an ejected jet by vapor of the radioactive elements condensing on the surfaces of aerosol particles and on atmospheric condensation nuclei;
- fractal structures in the form of condensation aerosols and conglomerates based on soot particles whose volume density equals that of air;
- radioactive noble gases and different physicochemical forms of iodine isotopes.

In contrast to the situation for the radionuclides of the volatile elements, iodine, tellurium, and cesium, the natural environment in the near zone of the Chernobyl nuclear power station is contaminated with transuranic and transplutonic elements in the form of fuel particles. A characteristic feature of samples of soil or other objects of the external environment that contain several tens of fuel particles is the closeness of the measured radionuclide ratios of the nonvolatile radionuclides to the corresponding ratios calculated for the fourth unit prior to the accident. In other words, such samples can be considered to be "representative" of the fuel prior to the accident.

In the period following the accident, extensive investigations were performed of the distribution of transuranic elements in environmental objects. In [4] the statistical characteristics were given of the ratios of the content of $^{239+240}$Pu, $^{241}$Am, and $^{242,244}$Cm to the total content of $^{239+240}$Pu in the soil, as calculated from the results of investigations of the content of the radionuclides in the soil in the near zone of the Chernobyl nuclear power station (13 areas, more than 200 samples). Data were also given concerning the activity of these radionuclides in units of the summed activity of $^{239+240}$Pu in the active zone of the accident reactor. A comparative analysis of the data confirms that in the near zone of the Chernobyl nuclear power station the ratio between the radionuclides in the soil corresponds to the corresponding ratio in the active zone of the reactor.

The ratios of the activities of $^{241}\text{Am}$, $^{242}\text{Cm}$, and $^{244}\text{Cm}$ were determined by radiochemical analysis of soil samples taken in 1994 and 1995 at test site 34-02-03 (6 km from the nuclear power station at azimuth 340°).

With the passage of time the fuel particles undergo dissociation, under the action of geochemical and meteorological factors, and this is accompanied by the release of radionuclides and their inclusion in migration processes. It is therefore reasonable that it is impossible correctly to predict the subsequent redistribution of the radionuclides without a reliable estimate of the degree of solution (transformation) of the uranium matrix of the fuel particles. We have studied this question in detail. An investigation was made of the layer-by-layer distribution of $^{137}\text{Cs}$, $^{90}\text{Sr}$, $^{239,240}\text{Pu}$, $^{241}\text{Am}$, and also $^{244}\text{Cm}$ in sampled columns of soil. The ratios of the activities of the radionuclides indicate that the radioactive fallout substances are of the fuel type [5].

The concentration and separation of $^{241}\text{Am}$ and $^{242,244}\text{Cm}$ without carriers and soil samples were performed using the method shown schematically in Fig. 2 [6].

Microscopic quantities of americium and curium were separated from the main soil-forming elements by precipitating their fluorides with the fluoride of stable lanthanum LaF$_3$, which was added as a carrier. The americium and curium without carriers were separated from the rare-earth elements using chromatographic separation in a 0.2-cm diameter 10-cm high ion-exchange column filled with KU-2-8 sulfonic acid cationite (State All-Union Standard GOST 20298-74) in the form of NH$_4^+$. Gradient elution [7] of the americium and curium was carried out with a buffer solution of α-oxyisobutyric acid (α-OIBA) with a pH of 4.75. The concentration of the eluting solution was chosen in such a way that the americium and curium were washed out from the ion-exchange column together with 0.3-0.4 mliter of eluent under conditions of complete purification from rare-earth elements.