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Urán és transzurán elemek meghatározása kis és közepes aktivitású atomerőművi radioaktív hulladékmintákból

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Új analitikai módszert dolgoztam ki a paksi atomerőmű VVER-440-es reaktora üzemelése során keletkezett kis és közepes aktivitású folyékony radioaktív hulladékait alkotó kimerült ioncserélő gyanták, iszapok, bepárlási maradékok, valamint a technológiai szándék szerint inaktív kibocsátásra kerülő vizek, urán (^{238}U , ^{235}U , ^{234}U) és transzurán ($^{239+240}\text{Pu}$, ^{244}Cm .) tartalmának alfa-spektrometriás meghatározására.

A kidolgozott kémiai elválasztások és analitikai módszerek alkalmazásával kapott mérési eredmények pontosságát az U.S. Department of Energy Environmental Measurements Laboratory (U.S. DOE EML) által szervezett környezetvédelmi radiológiai mérési eredmények értékelését végző nemzetközi minőségértékelő program (QUALITY ASSESSMENT PROG-RAM (QAP)) keretében teszteltem.

Témavezetőmnek, Dr. Gresits Ivánnak ezúton fejezem ki köszönetemet.

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Determination of uranium and transuranium elements in radioactive wastes of low and medium activity from a nuclear power plant

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I developed a new analytical procedure for the alpha-spectrometric determination of uranium (^{238}U , ^{235}U , ^{234}U) and transuranium isotopes ($^{239+240}\text{Pu}$, ^{244}Cm) in low- and medium-activity liquid radioactive wastes, including exhausted ion-exchange resins, sludges and evaporation residues formed during the operation of the VVER-440 reactor of the nuclear power plant in Paks, and in waters to be released into the environment, which should be, according to the technology, inactive.

The main steps of the procedure developed are as follows:

- sample preparation based on chemical separation,
- production of a thin-layer prepate,
- analysis by alpha spectrometry.

Analysis of α -emitting isotopes in radioactive wastes from nuclear power plants is not a simple task technically, since in some cases due to the low radioactive concentration (in waste waters for ^{238}U 0.5-1.0 $\mu\text{g/l}$), in others to the difficult matrix environment (for ion-exchange resins and sludges) a highly efficient enrichment or complicated chemical separation become inevitable. Hence, the heel of Achilles of the analytical procedure here is sample preparation.

The other crucial point of transuranium analysis is the production of a thin-layer prepate for energy-selective α -spectrometric measurement. The small sample obtained after enrichment is to be spread on a highly polished stainless steel disc in a well-adhering form in a uniform layer of a thickness which is infinitely small with respect to the transmission of α -rays in a way that the radionuclide to be measured is situated at the surface of the prepate.

The results of the work aimed at the development of new methods developed for the sample preparation to the alpha-spectrometric analysis of liquid radioactive wastes from nuclear power plants can be summarised as follows:

- Ion-exchange resins were decomposed in an oxygen atmosphere at overpressure in a closed calorimeter bomb. I have found no paper in the literature on the use of such technique for the sample preparation of radioactive wastes.
- The solid fraction of radioactive sludge samples was treated, following decomposition, by extraction and re-extraction, using an extractant with apolar solvent base and containing tributyl phosphate. Although tributyl phosphate is widely applied as an extractant, this type of procedure was first developed and applied as a reactor-specific method by myself to the determination of uranium and plutonium in low- and medium-activity radioactive liquid waste (sludge) samples from a nuclear power plant.
- I worked out a new selective method for the determination of the alpha-emitting constituents in evaporation residues containing both organic and inorganic compounds and saturated with boric acid.
- The organic compounds present in radioactive sludges, evaporation residues and waters to be released into the environment were decomposed in an Erdey-Jankovits digester modified by myself for my specific purposes. The modified digester, in which the entire sample preparation procedure involving chemical separation can be carried out, is universally applicable for sample preparation for the determination of alpha-emitting isotopes in any type of low- and medium-activity liquid radioactive waste occurring in a nuclear power plant.
- I developed and was the first to apply an electrodeposition technique for the production of thin-layer prepreates to alpha-spectrometric analysis of radioactive waste streams in the nuclear power plant in Paks. Uranium and transuranium isotopes were deposited by electrolysis in a device designed and

constructed in our laboratory with thermostation and controlled cathode potential adjusted at different values for each element to be deposited.

The accuracy of the results of determinations carried out by means of the sample preparation and analytical methods developed was tested in the framework of the international QUALITY ASSESSMENT PROGRAM (QAP) organized by the U.S. Department of Energy Environmental Measurements Laboratory (U.S. DOE EML) for evaluating the results of environmental radiological measurements.

- The test results show that the accuracy of the results of measurement made by means of the methods developed meet the requirements set by QAP.
- The test results prove that the methods are reliable and can be applied for the routine determination of the activity concentrations of alpha-emitting isotopes in low- and medium-activity radioactive wastes formed in the VVER-440 type reactor and in waste waters released into the environment.
- Using the new analytical methods developed, we regularly measure the uranium and transuranium radionuclides in the radioactive liquid wastes, released water, and in the water in test wells bored around the nuclear power plant in Paks.

Publications

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