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Developing of a method of determination of concentration and isotopic composition of thorium in natural waters in radiation monitoring

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Determination of the isotopic composition of natural radionuclides in natural waters is an integral part of radioecological monitoring. A full analysis of thorium radionuclides should solve two problems: determination of analytical concentration and isotopic composition. The main problem in the analysis of fresh water with low thorium content is preconcentration step; for samples with a high salt content and the complex composition it is removal of the most non-radioactive contaminants and alpha-emitters, e.g. uranium, preventing the preparation of high-quality source for alpha spectrometry. The aim of this work is to develop a thorium preconcentration step for large volumes of natural water. Th-234 was used as tracer in all experiments.
Coprecipitation with iron hydroxide (III) followed by precipitation on cellulose in frontal chromatography condition was choosed for preconcentration of thorium. Theoretical and experimental modeling of thorium coprecipitation with iron hydroxide was performed. Calculations of solubility of thorium hydroxide have shown that its own hydroxide phase is not formed in range of concentrations typical for natural waters. Therefore, coprecipitation with carrier (iron hydroxide) is the only possible way for thorium preconcentration. Processes of iron hydroxide precipitation and thorium coprecipitation with iron hydroxide from freshwater were studied by ultrafiltration method in dependence on pH, concentration of the thorium and iron (III). To isolate the precipitate of iron hydroxide, which contains thorium, we proposed filtration through a nozzle in frontal chromatography condition. Cellulose was used as a nozzle.
The study allowed to determine conditions thorium preconcentration that provide thorium yield (98 ± 8) %: the concentration of the iron (III) is 100 mg / l; pH 6.5-7.5; sediment formation time - 1 hour , the flow rate through column with filtration nozzle ~ 130 ml/min*cm2 . Weight of nozzle depends on volume of the sample. The ratio of height to diameter of the column is not less than 1:10. Extraction of thorium from filter nozzle by ammonium oxalate solution was performed under dynamic conditions.
The method of coprecipitation with cerium fluoride was used for preparation of samples for alpha-spectrometric determination of thorium isotopes. Coprecipitation method with further separation of the precipitate by ultrafiltration was selected due to the fact that uranium(VI) remains in solution under these conditions. By fluoride precipitation the quality of the sample depends on presence of calcium in the sample, forming a crystalline precipitate. Therefore, solubility of cerium, thorium and calcium fluorides with taking into account hydroxycomplexes was calculated to optimize the conditions for sample preparation for α-spectrometric determination of thorium isotopes. The results have shown that precipitation of calcium fluoride does not occur, while thorium and cerium fluoride will be precipitated when concentration of fluoride ions was 1,8·10^-5 mol/l ≤ CF- ≤ 5,0·10^-3 mol/l. Optimal concentrations of cerium nitrate and hydrofluoric acid, providing a quantitative release of thorium and good quality of sample for α-spectrometry were determined. Thorium yield in fluoride precipitation step was (98 ± 9)%.
Total yield of thorium in the whole method, defined by thorium-234 was (98 ± 11)%. Decontamination factor of uranium for samples containing up to 500 g/l of uranium according to the methods was ~ 10^2 ÷ 10^3.
Thus, the proposed method make it possible to perform a thorium preconcentration from large volumes of natural waters, providing its quantitative extraction and good separation from uranium. This methom can be recommended for determination of total analytical concentration of thorium and its isotopic composition by alpha-spectrometry.

Primary authors: PALIGA, Anna (ФГАОУ ВПО «УрФУ имени первого Президента России Б.Н.Ельцина»); NEDOBUH, Tatyana (ФГАОУ ВПО «УрФУ имени первого Президента России Б.Н.Ельцина»)

Presenter: PALIGA, Anna (ФГАОУ ВПО «УрФУ имени первого Президента России Б.Н.Ельцина»)

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