Lanthanide and americium separation with DGA resin

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In the nuclear fuel cycle, several lanthanide and trivalent actinide isotopes are produced by fission or transmutation of the uranium fuel. As these nuclides are usually present in small quantities and are often pure alpha/beta emitters (e.g. Am-241, Pm-147 and Sm-151) [1], they are classified as difficult-to-measure nuclides (DMNs). In order to analyze these DMNs successfully, a separation procedure using extraction chromatography with DGA resin (diglycolamide derivative) was developed. The lanthanides and/or Am separated were detected by nuclear measuring techniques.

Measurements and Methodology

The N,N,N',N' tetraoctyl-diglycolamide resin (TODGA, (commercialized as DGA resin by Triskem International) has extremely high capacity factors ($k'>10^4$) for trivalent lanthanides and actinides in concentrated mineral acids (e.g. 3M HNO_3 used in this study). The components retained can be sequentially eluted with dilute acids [2, 3].

A jacketed glass column was filled with about 0.6 g DGA resin. A peristaltic pump was used to pass the solutions through the resin bed with a flow rate of 0.1-0.25 cm^3min^{-1}. The column can be heated with water flow (typically 40°C) from an external thermostat to keep constant and well-regulated temperature. During the elution, 0.5-2.0 cm^3 fractions were collected.

To optimize the separation parameters (temperature, flow-rate, acid dilution) quickly and efficiently, gamma-emitting isotopes of the elements of interest were applied instead of their corresponding DMNs. These tracers were produced by thermal-neutron capture from stable elements with neutron bombardment at well-thermalized vertical channels of the Budapest Research Reactor. These easy-to-measure radioactive tracers (e.g. Ce-141, Nd-147, Sm-153, Eu-152) were detected with a well-shielded gamma-spectrometer, equipped with a 36% relative efficiency, n-type HPGe detector and Ortec DSPEC 502 digital electronics [4].

From liquid nuclear waste the lanthanides and minor-actinides were co-precipitated with LaF_3 in acidic environment [5]. The recovery of these elements was determined with a Tb-160 tracer. After dissolution of the precipitate in 20 ml 3 M HNO_3 and 0.5 g H_3BO_3, separation of lanthanides and americium was performed by the optimized chromatographic procedure.

Liquid scintillation counting (LSC) [1, 6] and gamma-spectrometry were applied to detect the appropriate nuclides.

Results and Discussion

An example for the optimized separation of lanthanides is given in Fig.1.

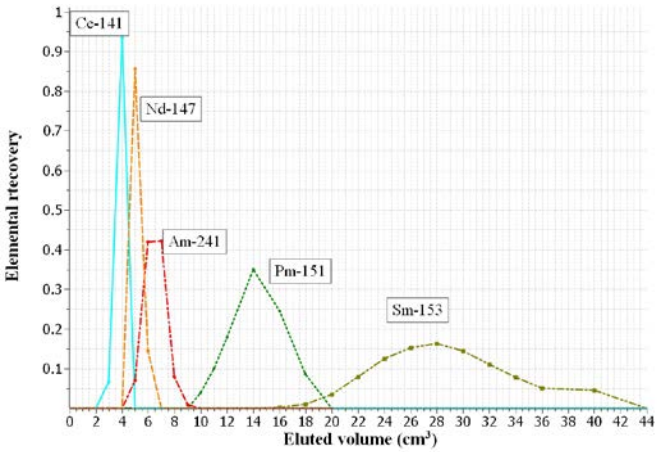

Figure 1: Chromatogram of lanthanides. Sample was loaded on DGA column from 3 M HNO_3. Lanthanides were eluted with 0.03 M HNO_3.

Lanthanides were eluted in order of increasing atomic number. The chromatographic peaks of the neighbouring elements were slightly overlapping, but fairly pure fractions were collected by properly choosing the peak boundaries. Application of two eluents, i.e. 0.03 M HNO_3 and 1.5 M HCl was tested extensively.

Activities of the DMNs, Am-241 and Pm-147 in radioactive waste samples were determined by LSC of the appropriate fractions.

Whenever gamma spectroscopy was relevant, e.g. in case of Am-241, Eu-154 and Eu-155, three different detectors (coaxial-, low-energy-, well type- HPGe) were used and compared.

References

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