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Preparation of thick uranium layers on aluminium and stainless steel backings

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HIGHLIGHTS

- Target preparation procedures included electrodeposition and molecular plating.
- Organic and inorganic solutions used as electrolytes.
- Characterisation of layers for activity, thickness, homogeneity and morphology.

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ABSTRACT

The methods of electrodeposition and "molecular plating" were studied for the production of uranium targets with an areal density up to 0.6 mg cm^{-2} on aluminium and up to 1.5 mg cm^{-2} on stainless steel backings from aqueous and organic electrolytes. For characterisation of the deposited material, gamma-ray spectrometry, alpha-particle spectrometry, X-ray fluorescence, X-ray powder diffraction, scanning electron microscopy and autoradiography were applied.

1. Introduction

Uniform layers of actinides are required in experimental nuclear physics to measure accurate nuclear data. The production of these layers on different metallic backings is very difficult due to the fact that their stability depends on the chemical composition, as well as on the thickness of the deposit, the mode of preparation and the surface of the metallic backing. If the deposited material after drying does not adhere to the backing material, the layer can contain cracks and damage which is problematic for experiments in nuclear laboratories, research reactors and accelerators.

Numerous methods for radioactive source preparation have been published (Sibbens and Altzitzoglou, 2007): electrolytic deposition, spontaneous deposition, micro-precipitation, direct evaporation, vacuum sublimation, drying of liquid drops directly deposited on the substrate, electrospray, electrostatic deposition etc.

The techniques available for preparation of layers of actinides on metallic backings involving electroplating from organic and aqueous media and the characterisation of such targets have been described in numerous papers (Mirashi et al., 1986; Ingelbrecht et al., 1997; Lobanov et al., 1997; Eberhardt et al., 2004; Liebe et al., 2008; Jobbágy et al., 2013; Sadi et al., 2011; Vascon et al., 2012, 2013; He et al., 2013).

The objective of this work was to prepare uranium layers on aluminium and stainless steel backings with an areal density up to 1.5 mg cm^{-2} and to characterise the deposited material by gamma-ray spectrometry, alpha-particle spectrometry, X-ray fluorescence (XRF), X-ray powder diffraction (XRDF), scanning electron microscopy and autoradiography.

2. Experimental

2.1. Target preparation

Targets of uranium were prepared by electrodeposition in the Department of Environmental Sciences at the Jožef Stefan Institute (JSI) in Ljubljana, Slovenia and in the Target Preparation Laboratory at the Institute for Reference Materials and Measurements of the Joint Research Centre (JRC-IRMM) in Geel, Belgium.

Two different types of backing material, mirror polished stainless steel (SS) and commercially available aluminium (Al), both with a diameter of 25 mm and a thickness of 0.4 mm, were used. As anode a flat spiral platinum wire was used at the JSI (Klemenčič and Benedik, 2010) and a vertical rotating rectangular platinum grid at the JRC-IRMM (Ingelbrecht et al., 1997). The active area of

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the deposited material was about 3.4 cm^2. As material stock solutions of uranium with natural isotopic abundances were prepared at the JRC-IRMM and at the JSI in the form of $UO_2(NO_3)_2$. Targets were prepared by electrodeposition in isopropanol (IP), isobutanol and ammonium oxalate (Ox) (Ingelbrecht et al., 1997; Puphal and Olsen, 1972; Mirashi et al., 1986). Approximately 10 cm^3 of the solution was transferred to the cell and an aliquot of the uranium solution was added. The cathode–anode distance was set between 8 mm and 10 mm. When the targets were prepared by electrodeposition using an aqueous electrolyte, the targets were rinsed with water and ethanol containing a few drops of an ammonia solution. The discs with deposited material were dried in air and then heated at 100 °C to fix the uranium.

2.2. Target characterisation

The uranium sources were characterised for their activity, mass and areal density. A first approximate assay of the deposited uranium was done by gamma-ray spectrometry. Therefore the γ line of ^{235}U at 185.72 keV was measured.

At the JRC-IRMM the activity was determined by low solid angle α-particle counting (Heyse et al., in press). The solid angle subtended by the 20 mm diameter diaphragm in front of the silicon surface barrier detector was 1.4% of 4π sr.

Table 1
U activity in Bq, mass in μg and areal density in μg cm^{-2} of four typical uranium targets. The uncertainties, unc, are combined standard uncertainties.

Target no.	Backing	Electrolyte[a]	Activity (Bq)	unc (Bq)	Mass U (μg)	unc (μg)	Areal density (μg cm^{-2})
17	SS	Ox	133.1	1.4	5453	57	1574
22	Al	Ox	38.0	0.4	1558	18	450
23	SS	IP	62.3	0.7	2554	28	737
27	Al	IP	51.4	0.6	2105	24	608

[a] Ox=5.7% $(NH_4)_2C_2O_4$ in 0.3 M HCl; IP=$(CH_3)_2CHOH$.

The distribution and homogeneity of the deposited uranium layer were checked by phosphor imaging autoradiography using Fuji Photo Film BAS-IP TR 8 and BAS-IP MS 2025 imaging plates at the JSI, while at the JRC-IRMM the Cyclone™ Storage Phosphor Imaging System (Packard) was employed (Sibbens et al., 2003).

The uranium samples were analysed non-destructively for their elemental composition by energy dispersive X-ray fluorescence spectrometry (EDXRF). A ^{109}Cd disc radioisotope excitation source was used to irradiate the samples. Analysis of the complex X-ray spectra was performed by the AXIL spectral analysis program (Van Espen and Janssens, 1993). Quantification of the metal content after the necessary calibration of the XRF system was then performed by the QAES (Quantitative Analysis of Environmental Samples) software developed by P. Kump (Nečemer et al., 2011).

Surface characterizations of the deposited material to visualise the layer structure were performed with a field emission scanning electron microscope (FE-SEM, Zeiss ULTRA plus).

3. Results and discussion

At the JRC-IRMM the experimental work was focused on the preparation of the targets containing thick layers of deposited uranium from aqueous and organic electrolytes onto SS and Al backings and the characterisation by low solid angle alpha-particle counting. The measured activity and calculated mass and areal density are presented in Table 1 for four typical targets out of thirty prepared.

Fig. 1 shows pictures of the prepared targets and the corresponding alpha-particle spectra obtained by low solid angle alpha-particle counting. Samples no. 17 and no. 22 were prepared from 5.7% ammonium oxalate electrolyte on SS and on Al, respectively, while samples no. 27 and no. 23 were prepared from isopropanol on SS and on Al, respectively. The spectrum of sample no. 22 demonstrates its low resolution and long tailing in the low-energy region because of self-absorption. And from Fig. 1, it is evident that the aluminium disc (no. 22) during electrodeposition of uranium

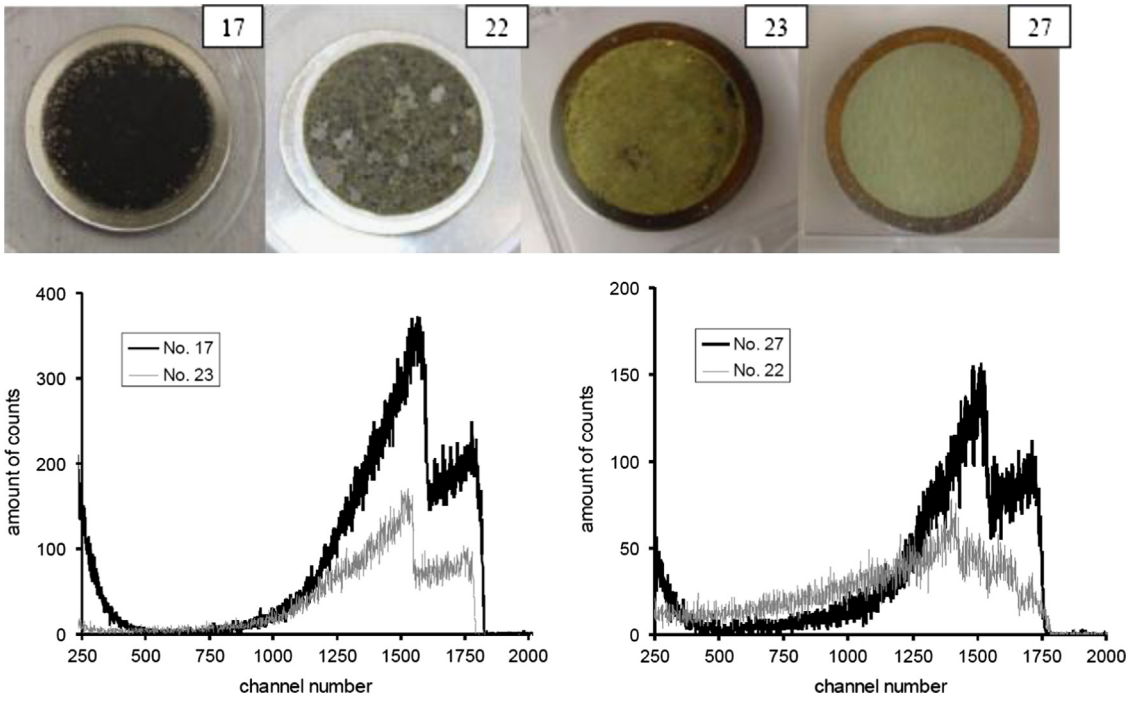

Fig. 1. Pictures of uranium layers on SS and Al backings as listed in Table 1 prepared by electrodeposition and molecular plating using various electrolytes and corresponding alpha-particle spectra obtained by low-solid angle counting.