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Actinide target preparation at IRMM—then and now

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ABSTRACT

Since the beginning of its activity IRMM (originally CBNM, Central Bureau for Nuclear Measurements) was engaged in measurements of parameters relevant to nuclear energy. High-purity samples and targets of radioactive materials required for measurements of cross-sections and studies of fission fragments or fuel elements were prepared by the IRMM's target group by means of various techniques reviewed in this presentation. Applying these techniques the target group had been preparing targets of U, Pu, Np, Am, Th for in-house physicists and for external customers.

Recently, after a long process of decontamination and refurbishment of the old equipment, custom-made ^{233}U and ^{235}U targets were prepared by electro-deposition and by high-vacuum evaporation. Other actinide targets (U, Th) have been prepared using mechanical reshaping techniques.

1. Introduction

The first actinide, uranium, was discovered in 1789 (by M.H. Klaproth) but it took over 100 years until its radioactive properties were found by H. Becquerel, and another 40 years until it was used for the first time in nuclear studies. In 1934, E. Fermi and others tried to produce elements heavier than U by bombardment with neutrons from a Ra–Be source. As it is known today, the experimentalists had problems with the proper interpretation of the observed reaction, as the obtained products were not actually heavier than the uranium starting material. A few years later this reaction was explained as a fission process induced by neutrons, and the fission products were properly identified by Lise Meitner, Otto Hahn and Fritz Strassmann with the help of Otto Robert Frisch. Describing their studies in Ber. Dtsch. Chem. Ges. 69 (1936) 905 they reported use of "A suitable amount of uranium purified as quantitatively as possible from its ordinary decay products, precipitated by ammonia as uranate and, after drying, exposed to bombardment with neutrons" (translation of the original text is taken from contribution of G. Hermann to the 14th INTDS meeting, 1988 [1]). This sample of ammonium diuranate $(NH_4)_2U_2O_7$ can be considered as the first uranium/actinide target ever used in nuclear studies.

The number of nuclear studies using different kinds of radioactive materials was growing very quickly, and so was the demand for samples/targets of different thicknesses, different radioactive elements and their isotopes.

2. Actinide targets at IRMM

IRMM (originally CBNM, Central Bureau for Nuclear Measurements) since its establishment in 1960 was engaged in measurements of parameters relevant and important for nuclear energy. Such measurements require well-defined samples of high quality and thus it was decided to bring a group engaged in their preparation into service.

Samples were and are prepared for measurements of cross-sections, studies of fission fragments, neutron beam filters, neutron flux mapping and dosimetry, and studies of fuel elements, to mention only a few. All those studies require pure metals or metals doped with certified amounts of radioactive elements, alloys, compounds or solutions of certified composition. For decades the target preparation group at IRMM (CBNM) has produced and characterised samples for those experiments both for its own studies and, in the past, for external research groups, being a world-wide recognised supplier of targets. Due to the possibilities of preparation of the targets in individual glove boxes (each isotope had its own preparation stand) the final products were of high purity. Through decades hundreds of samples of different actinides (Table 1) were prepared applying various techniques.

3. Target preparation

3.1. Methods used in the past

Depending on the required target material, its thickness and thickness homogeneity, type of backing, and the amount of available material, the targets were prepared using the following methods (Table 2).

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Table 1
List of targets prepared at CBNM (IRMM).

89 *Actinium Ac*	96 *Curium Cm*
90 Thorium Th: 230	97 *Berkelium Bk*
91 *Protactinium Pa*	98 *Californium Cf*
92 Uranium U: 233, 234, 235, 236, 238	99 *Einsteinium Es*
93 Neptunium Np: 237	100 *Fermium Fm*
94 Plutonium Pu: 238, 239, 240, 242	101 *Mendelevium Md*
95 Americium Am: 241, 243	102 *Nobelium No*
	103 *Lawrencium Lr*

The elements of which targets were prepared over decades are listed in normal font.

Table 2
Pros and cons of each method; samples prepared on metallic or metalised backings.

Pros	Cons
Electro-spraying	
High efficiency	Thickness limitation, generally to few mg/cm^2
Simple equipment	Impurities from solvent
Homogeneity ~5% (in some cases <2%)	Deposition of compounds only
Electro-deposition	
Process fast and simple	Thickness homogeneity lower than by vac. evaporation
No cross contamination	Sometimes deposits composition is unknown/uncertain
High efficiency	Impurities from solvent
Very good adherence	
Big range of the thickness, up to mg/cm^2	
Sedimentation	
Very high efficiency	Limited only to thick deposits (g/cm^2)
Good for thick deposits	Impurities from solvent
Electrophoresis	
Quick	Preparation of the colloidal suspension requiring grinding the powder (potential source of contamination)
Suitable for thick targets	Upper thickness limitation (due to peeling off of the deposit)
Deposits made of very fine particulates (bigger ones precipitate out, significantly influencing the thickness homogeneity)	Problems caused by gas generated during the process
High-vaccum evaporation	
High purity of the final product	Very low efficiency
The thickness homogeneity is very high	Expensive equipment
Suitable for very thin targets	Time-taking preparation of the material for evaporation
Melting by levitation is very suitable for materials with big melting point. This melting technique is very suitable for alloys preparation	*Levitation melting* requires big amount of the evaporated material
	Method requires additional expensive equipment

3.1.1. Electro-spraying

In this technique a solution (actinides acetates) or suspension (actinides oxides suspended in e.g. acetone) of the target material forced by a high voltage (3–20 keV) to pass through a narrow capillary is deposited on a conductive substrate (carbon or metallic/metalised foil) [2–4]. Fig. 1 shows the electro-spraying set-up used at IRMM.

In this method the solvent had to evaporate before the deposited material touched the backing. Targets of 15 mg/cm^2 for U or even of 30 mg/cm^2 for other materials were prepared using this technique, all with good adherence to the baking assured by material acceleration in the electrostatic field.

3.1.2. Spray painting

An alternative to the electro-spraying was spray painting using an air brush (Fig. 2) for depositing actinides on metallic backings

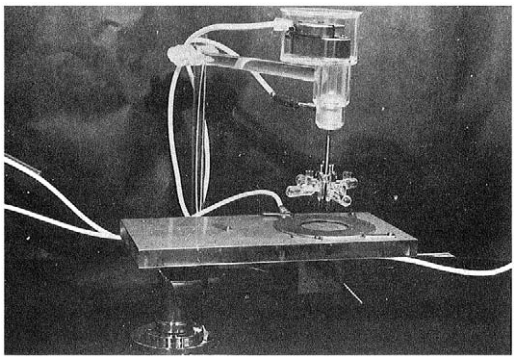

Fig. 1. Electro-spraying facility.

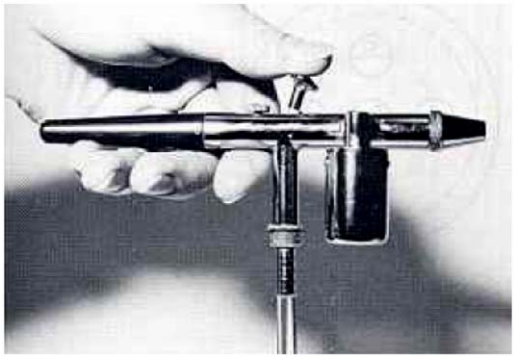

Fig. 2. GRAFO retouching air brush.

[5]. Usually, the solution (uranyl nitrate dissolved in ethanol) used for painting was mixed into a carrier solution of cellulose nitrate dissolved in isoamylacetate.

Solutions were sprayed on backings pre-heated to 393 K (120 °C), which were further heated up to 773–823 K (500–550 °C) to stabilize that layer. The deposits prepared this way had excellent adherence to the backing and good thickness homogeneity. The targets prepared had no border effect at the mask edge.

3.1.3. Electro-deposition

Electric current passing through inorganic or organic solutions of actinides causes deposition of the layers on, mainly, metallic backings. Tests with backing made of polyimide foils covered with Pt or Au to assure the conductivity were done as well but the results were not very satisfactory [6] as metallic layers tend to peel off during the electro-deposition process. Nevertheless, thin targets of U and Pu [7] were reported as being prepared by electro-deposition on plastic substrates. Although, some positive results were reported for polyimide (PI) foils covered with carbon as a backing [6] but unexpected problems with mechanical stability of such PI foils require further developments to assure repeatability of the deposition process. Electro-deposition is a very effective method (efficiency between 80% and 95%) resulting in final products of relatively high purity as each requested target type is prepared using deposition cells constructed for processing this particular element/isotope (Fig. 3).

3.1.4. Powder sedimentation

Spontaneous/gravimetrical Applied to prepare targets of actinide oxides with thickness up to several g/cm^2. Often the layer of powder was canned under vacuum or inert gas after suspension. This technique was used for preparation of such targets as ^{241}Am