Toward large-area targets for "TRAKULA"

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Introduction: TRAKULA (Transmutationsrelevante kernphysikalische Untersuchungen langlebiger Aktinide, i.e., nuclear physical investigations of long-lived actinides with relevance to transmutation) is a joint research project on nuclear physics investigations with modern scientific, technological and numerical methods, in which experiments concerning the transmutation of radioactive waste are a central topic. For this, large-area samples ($\ge 40 \text{ cm}^2$) of ${}^{235,238}\text{U}$ and ${}^{239,242}\text{Pu}$ are required for the calibration of fission chambers and for neutron-induced fission yield measurements. Another requires large-area topic targets for precise measurements of the half-life, $t_{1/2}$, of very long-lived α particle emitters like ¹⁴⁴Nd ($t_{1/2} \approx 2.10^{15}$ y).

Electrodeposition tests with Gd and Nd (used as chemical homologues of the actinides), were performed to find optimal deposition conditions for small-area targets that should be applicable to large-area targets.

Experimental: The layers were produced by Molecular Plating (MP) on 5 µm backings. The backing foils were pre-cleaned with 6 M HCl, water, and isopropanol. The lanthanide compound in the nitrate form was dissolved in 0.1 M HNO₃ to an elemental concentration of ≈ 25 mg/ml. Two different procedures were followed to produce targets: (i) an aliquot of 100 µl of the stocksolution was mixed with 1 ml isopropanol, and transferred into the electrochemical cell, which was then filled up with 24 ml isobutanol (hereafter referred to as inactive MP); (ii) an aliquot of 100 µl of the stock solution was irradiated in the TRIGA Mainz research reactor, in order to produce ¹⁵³Gd ($t_{1/2} = 239.47$ d) and ¹⁴⁷Nd ($t_{1/2} = 10.98$ d), respectively, as radioactive tracers. The irradiated solution was then used for the MP (hereafter referred to as active MP). The plating solution was stirred either with a magnetic stirrer (Variomag Compact) operated at 1000 rotations per minute (rpm) or a quartz tip ultrasonic stirrer (Bandelin Sonopuls HD 2070) operated at 30% power pulse. MP was carried out at 14°C by applying two different constant currents: 2 mA and 4.2 mA, yielding current densities, respectively, of 0.7 mA/cm² and 1.4 mA/cm².

The deposition yield was determined by an indirect and a direct method, respectively: Neutron Activation Analysis (NAA) and γ -spectroscopy. NAA was used after inactive MPs to determine the residual concentration of the lanthanide element in the supernatant solution via γ -spectroscopy of a standard sample and an aliquot of the supernatant solution irradiated simultaneously (indirect yield measurement). Gamma-spectroscopy was used after active MPs. This technique employed a high-purity germanium detector (HPGe) (GEM 23158 P-Plus, ORTEC Company) to measure the active samples (direct yield measurement). To obtain quantitative data, reference sources with known amounts of the tracer were prepared. They consisted of filter papers with the same geometry as the targets, soaked with the tracer-containing solution. To avoid any kind of contamination inside the γ -spectrometer, the active samples were sealed before being inserted into the sample holder.

The homogeneity of the radioactive targets was inspected by using Radiographic Imaging (RI), using a FUJIFILM FLA 7000 equipped with reusable imaging plates and a 650 nm laser for the reading process.

The morphology of the targets was studied by using a Scanning Electron Microscope (SEM) (Philips XL30), operated at 20 kV.

The surface roughness of the targets was investigated by using an Atomic Force Microscope (AFM) (MFP 3D, Asylum Research). Fig.1 shows a surface whose mean roughness is ~ 100 nm. The AFM used cantilevers with a resonance frequency of 283 kHz for imaging in the AC mode (tapping mode).



Figure 1. AFM picture of the surface of a Nd-target obtained at high current density (1.4 mA/cm²), applying ultrasonic stirring.

Results: The characterization of the layers with different analytical techniques (i.e. NAA, γ -spectroscopy, RI, SEM, AFM) proved that ultrasonic stirring is superior to mechanical stirring, especially for the production of large-area targets, and that the two different current densities applied, i.e. 0.7 mA/cm² and 1.4 mA/cm², are both suitable for target preparation.

According to the results obtained from these tests, a new electrochemical cell for the production of large-area targets was designed, built and recently tested with success.

References

[1] A.Vascon et al., Nucl. Instrum. Meth. A (submitted)

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