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Multi-layer $^{235}UF_4$–^{6}LiF–Au targets for high-resolution fission fragment measurements

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HIGHLIGHTS

- Multi-layer $^{235}UF_4$–^{6}LiF–Au targets were produced on thin polyimide foils.
- Prepared for future high-resolution measurements of fission-fragment properties.
- Different layers were prepared by physical vapour deposition.
- The foil and each layer were characterized separately for their areal density.

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ABSTRACT

Multi-layer $^{235}UF_4$–^{6}LiF–Au targets have been produced by vacuum deposition on thin polyimide foils with an areal density, measured by spectrophotometry, of about 33 μg cm^{-2}. The foils were first covered with an Au-layer and then, with a second layer of ^{6}LiF, both by vapour deposition. The $^{235}UF_4$ layer was prepared by fluoride sublimation. Each deposited mass was characterized separately by means of differential weighing for the Au and ^{6}LiF layers and by low-geometry alpha-particle counting for the $^{235}UF_4$ layer. The atomic abundances of the uranium base material have been measured by thermal ionization mass spectrometry. The targets were prepared for measuring fission-fragment emission yields with high mass-resolution.

1. Introduction

Thin homogeneous and well-characterized targets are required for accurate nuclear data measurements (Schillebeeckx et al., 2010). The energy loss of the emitted particles by passing through the target material has to be as small as possible and constant over the entire target area. These thin layers are produced at the Joint Research Centre (JRC) IRMM by molecular plating or vacuum deposition and characterized for quantities (Sibbens et al., 2010; Sapundjiev et al., 2010). If in addition the particles to be measured are emitted in opposite direction, also the backing material has to be as thin as possible to minimize energy loss. In that case, thin polyimide foils seem to be most suitable amongst several other thin plastic foils, because of its high resistance to damage from charged particles and common chemicals. The polyimide foils are produced at the JRC-IRMM by in-situ polymerization (Pauwels et al., 1979).

For future high-resolution fission fragment experiments at the JRC-IRMM with the double time-of-flight (TOF) spectrometer VERDI (Oberstedt et al., 2010, 2013) special multilayer $^{235}UF_4$–^{6}LiF–Au targets are requested. VERDI measures fission fragment masses and kinetic energies prior to prompt neutron emission by employing the double TOF technique. A subsequent measurement of the particles' kinetic energy provides mass information after evaporation of prompt neutrons from the highly excited fragments. Striving for an unambiguous mass identification requires a minimum of energy loss and straggling in the target and backing material. Additionally, interruption for changing targets for absolute time calibration is not desirable, because it may lead to a deterioration of signal stability and spectrometer resolution. Therefore, it is tempting to combine the measurements of fission-fragment emission yields and the absolute time-of-flight calibration in one simultaneous run. The development of multi-layer targets, which contain both active target (e.g. ^{235}U) and calibration source on the same backing, is the obvious choice. The multilayer target has to consist of three thin layers of Au, ^{6}LiF and $^{235}UF_4$, deposited respectively on a thin polyimide foil. The ^{6}LiF layer is thus "sandwiched" between a thin Au layer deposited on a polyimide foil and the thin $^{235}UF_4$ layer. This arrangement assures minimum fission-fragment energy loss and straggling on the backing (polyimide) side and no additional energy loss on the active target side.

As all three layers will be produced by vacuum deposition, care has to be taken to ensure that the thin polyimide foil can survive the air pressure differences, increased temperatures and manipulations during the evaporation processes in the three different evaporation

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set-ups. In addition the multilayer target needs to be stable with good adherence of the different layers on the backing (Maier-Komor and Speidel, 2002; Vanleeuw et al., 2013).

Besides the production of the multilayer targets, the areal density of the material of interest of each layer and the backing should be known. In this paper the preparation and the characterization of the 'sandwich' targets, are described.

2. Experimental

2.1. Preparation and characterization of the backings

Thin polyimide foils with an areal density of about 33 μg cm^{-2} were prepared as backing in order to minimize energy loss of the fission fragments. Polyimide foils are mechanically strong plastic foils with a good resistance to temperature, irradiation with charged particles and common chemicals. The foils were prepared by in-situ polymerization as described by Sibbens et al. (2011). The first part of the procedure was done in a dry atmosphere and consisted of the preparation of the polycondensate by dissolving dianhydride and diamino acid in dimethylformamide and spreading the solution on a glass plate to form the foil. The glass plates were then heated in an oven for 12 min at 350 °C for completing the polymerization process. The foil, still on the glass plate, was cut with a fine sharp knife into a circular shape in order to cover the 0.3 mm thick support Al ring with outer diameter of 50 mm and inner diameter of 40 mm. To release the foils from the glass plate, the plate was immersed vertically in a bath with de-ionized water until the foils could be floated from the glass plate onto the water surface and transferred onto the Al ring. The rings with foil (Fig. 1) were dried in vertical position in a laminar flow cabinet.

The polyimide foils were characterized for their areal density by spectrophotometry. This is a quick and simple non-destructive characterization technique, which allows estimation of the local thickness of the film deposit. The technique is based on the acquisition of an UV/Vis spectrum of light reflected or transmitted by the thin film deposit. The resulting interference spectrum is then used to calculate the areal density of the film (Pauwels et al., 1982).

2.2. Preparation and characterization of Au and ^{6}LiF layers

Before depositing uranium, the polyimide foils were covered with a first layer of about 43 μg cm^{-2} Au to provide electric conductivity followed by a second layer of 95.65% enriched ^{6}LiF with an areal density of about 13 μg cm^{-2} to make use of the reaction ^{6}Li(n, α)t for absolute time-of-flight calibration online. The two layers were prepared by physical vapour deposition in different set-ups, dedicated to each material.

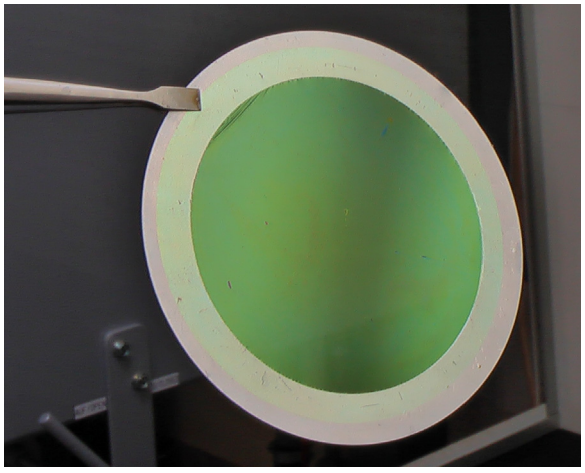

Fig. 1. Polyimide foil with an areal density of 32 μg cm^{-2} mounted on an Al ring.

Both the Au and the Li evaporator consisted of a Ta crucible connected to a power supply in a closed chamber, connected to a pump system in order to work under vacuum conditions. The polyimide backings were mounted in a holder on a carrousel on top in the chamber. During the evaporation process the holder was rotating to achieve the optimum uniformity of the deposited material. Masks with an inner diameter of 49 mm for the Au deposit and 30 mm for the ^{6}LiF layer , were positioned in front of the backings and determined the spot size of the deposited material. The Au was heated up in the Ta crucible to about 1100 °C and the ^{6}LiF material to about 900 °C for evaporation. By varying the current through the Ta crucible the evaporation rate was controlled. The deposition rates for the Au layer production were between 0.01 nm s^{-1} and 0.04 nm s^{-1} and for the ^{6}LiF between 0.1 nm s^{-1} and 0.2 nm s^{-1}. The film thickness during the deposition process was monitored with a Thickness-Shear-Mode (TSM) Acoustic Wave Sensor with quartz crystal. Equipment and process are described by Sapundjiev (2011). Each layer was characterized separately for its mass by differential weighing. Therefore, the backing was positioned on a Mettler-Toledo XP56 microbalance before and after each deposition and the weighing was done applying the substitution method.

The areal densities of the polyimide backings, the first layer of Au and the second layer of ^{6}LiF are presented in Table 1. For the Au and ^{6}LiF layers the areal densities were calculated from their mass determined by weighing and the diameter of the deposit. The latter was not directly measured. Instead, the inner diameter of the masks positioned in front of the backing during the physical vapour deposition was measured.

Fig. 2 shows a polyimide foil on an Al ring with Au and ^{6}LiF layer.

2.3. Uranium material

Highly enriched ^{235}U (lot 680, 99.934% mol mol^{-1}) oxide was selected from the available uranium materials at IRMM. The $^{235}U_3O_8$ was first converted to $^{235}UF_4$ by a "wet chemical precipitation" method (Pauwels et al., 1984). The fluoride has the advantage that it sublimates between 1000 °C and 1500 °C, so that heat sensitive substrates like plastic foils can be used and that there is no interaction with the resistance heated crucible. Oxide sublimation would require a temperature above 2500 °C. The original triuranium octoxide was first converted into uranylchloride by dissolving in hydrogen chloride (1). In the second step the uranium was reduced by adding tin chloride at a temperature of about 40 °C (2). Finally the reduced uranium chloride was converted into a fluoride in reaction with hydrogen fluoride and a residue was formed that consisted of uranium fluoride (3).

$$U_3O_8 + 6HCl \leftrightarrow 3^{6+}UO_2Cl_2 + 2H_2O + H_2\uparrow \quad (1)$$

$$^{6+}UO_2Cl_2 + SnCl_2 \leftrightarrow {}^{4+}UCl_4 + SnO_2 \quad (2)$$

$$UCl_4 + 4HF \rightarrow UF_4\downarrow + 4HCl \quad (3)$$

Table 1
Areal densities in μg cm^{-2} of polyimide backing (PI), the first layer of Au and the second layer of ^{6}LiF; the combined standard uncertainties are given in parentheses and apply to the last digit(s) of the value.

Target ID	PI areal density [μg cm^{-2}]	Au areal density [μg cm^{-2}]	^{6}LiF areal density [μg cm^{-2}]
TP2010-008-01	33(3)	44.5(8)	12.1(21)
TP2010-008-02	30(3)	43.4(8)	13.8(21)
TP2010-008-03	32(3)	42.8(8)	12.9(21)
TP2010-008-05	33(3)	44.7(8)	13.9(21)