LIP-STUDENTS-22-22

Target preparation at FCUL

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Abstract. The aim of this work was to produce a thin calcium fluoride target with a formvar backing for an experiment to be carried out at Centre for Micro Analysis of Materials in Madrid, Spain. The films were produced at the target producing facility at FCUL. The films' thickness was determined using analytic methods. The target's thickness was measured using Rutherford Backscattering Spectrometry at the Van de Graaff CTN-IST facility with α particles. The measurements were validated measuring the energy loss of α particles through the target. The loss of energy was then used to compute the target's thickness using standard toolkits such as AlfaMC toolkit or the SimNRA software.

Keywords: Thin target, Thermal evaporation, Stopping power, Nuclear reactions, AlfaMC, calcium fluoride, formvar, RBS, SimNRA

1 Introduction

Targets are an essential part of nuclear physics experiments involving the study of nuclear reactions, and can range from a few nanometers to some micrometers depending on the requirement of the experimental campaign. During this internship, several targets were developed, with the main focus being on calcium fluoride (CaF_2) films.The mentioned target was requested by Prof. Dr. O. Tengblad, (*Instituto Estructura de la Materia*, CSIC Madrid, Spain) with a thickness of 120 μg/cm^2 for an experiment at Centre for Micro Analysis of Materials (CMAM). As calcium fluoride is not self supporting at the required thickness, a structural support, in other words a backing, was made from the thermoplastic resin formvar. The experiment was successfully carried out in September 2022.

Additionally, gold targets with and without formvar support were produced for an experiment at HIE-ISOLDE (CERN) facility in September 2022. Formvar supporting lead films for the same experiment were also produced.

2 Investigation into providing a support for the calcium fluoride targets

As calcium fluoride is not self-supporting, the thermoplastic formvar was chosen as a support. There are several reasons for this choice, namely it being thermal resistant, and having in its composition elements with low atomic number, so as not to cause an overload of the data acquisition system due to excessive signals. The first investigation was to produce different concentrations of formvar, 1.5 %, 2 %, 3 % and 6 %, by dissolving the formvar in chloroform. Two methods were employed to produce formvar films.

2.1 First method

For the first method, a pipette was used to drop formvar solution onto distilled water. The solution turned into a plastic film upon making contact with water, and it was possible to fish these films out of the water with the target frames. We observed that the higher concentrations of formvar solutions, the thicker the targets produced. Unfortunately, the films were probably too thin to withstand the evaporation process of the calcium fluoride or, if they survived, they would break whether in the thickness determination experiment or afterwards.

2.2 Second method

For the second method, a larger amount of formvar solution was poured into an 8 cm diameter Petri dish. The formvar solution solidified upon the evaporation of the chloroform. The resulting films' thickness depended on the quantity of solution used. Finally, 25 ml of 1.5 % formvar were used in the Petri dish to produce the backing films, which were able to withstand the entire production process of the calcium fluoride film on top of them using thermal evaporation and the characterization of the films. An example of a backing created using this method can be seen in figure 1.

Figure 1. Formvar backing made using the second method.

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3 Producing thin films using thermal evaporation process

After creating the backings, the thermal evaporation method was used to produce the layers of either of the aforementioned materials: calcium fluoride, gold or lead. In this method, the boats containing the evaporant and the target holders were placed inside the evaporation chamber. As illustrated in figure 2, a high current crosses the boat that's locked on the electrodes, heating it up by Joule effect and, due to thermal conduction, the boat will heat the material inside it until it evaporates. The atoms from the source travel to the substrate and condensate on the surface of our targets.

The boats used were made of tungsten or molybdenum, due to their melting points being higher than the source's, and because these materials are good current conductors.

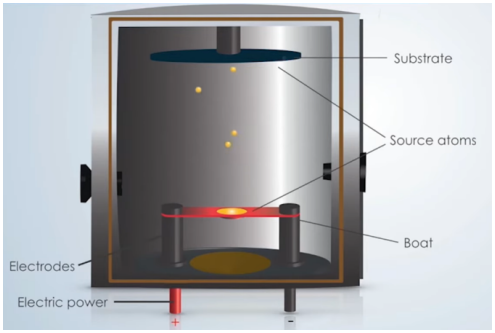

Figure 2. Schematic of a thermal evaporation chamber. The boat contains the evaporant. The evaporated material will travel to the substrate where it condenses, forming thin films [1].

3.1 Evaporation procedure

The evaporation facility consists of a bell shaped steel thermal evaporation chamber, as can be seen in figure 3. The chamber is equipped with several windows, where it is possible to directly observe the boat holding the evaporant. With experience, the brightness of the boat, which reflects its temperature, is a good indication if the evaporation process has started, particularly when observing the quality of the vacuum at the same time.

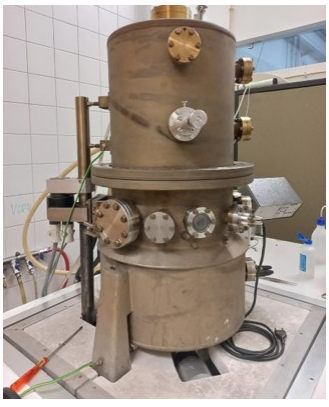

Figure 3. Evaporation chamber.

Before each evaporation, the chamber was cleaned to avoid cross contamination from previous evaporations. The boats are placed on either of the two available anode-cathode pairs, and the chamber is closed. The vacuum pumps are turned on, first the primary and then the turbo-molecular one. Once the pressure inside the chamber reaches approximately 10^{-2} mbar, the turbo pump is switched on to reach a working vacuum of about 10^{-6} mbar. The evaporation process has to be carried out in low pressure environments, as there are fewer contaminants and the evaporation points of the materials are lower. The evaporation is monitored using the pressure values as well as the brightness of the boat, which can be seen in figure 4.

Figure 4. Boat glowing through the glass window of the chamber.

4 Characterization of the films with alpha particle energy loss

It is possible to determine the thickness of a thin film from the measurement of an α particle's energy loss when passing through the sample. For the purpose of these measurements, the radioactive particle sources ^{232}U or ^{226}Ra were used.

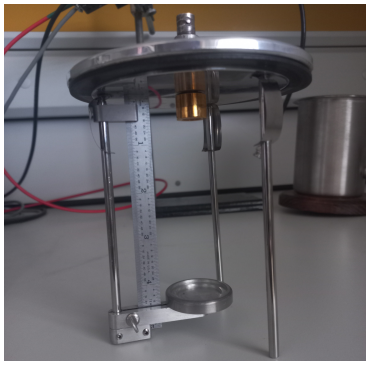

Figure 5. The picture shows, on top, the mounted detector, and on the bottom the holder for the α source with a ruler measuring the distance between the two components.

4.1 Detection and acquisition

The data acquisition system (DAQ) of α particles consists of a silicon surface barrier detector. The detector's pre-amplifier output signal is connected to an amplifier and the signal is sent to the multichannel analyzer. Figure 5 shows the experimental setup for the measurement using this method.

4.2 Calibration of the detector

The radioactive source, ^{232}U was used to calibrate the detector as the measured spectrum has well defined peaks. Additionally, this source has a relatively high activity so the acquisition time for each spectrum is approximately 30 min. From the ^{232}U decay chain, several of the elements corresponding to some of the peaks could be identified, afterwards making the relation between channel and energy (keV). The radioactive source faced directly the detector for the calibration runs. When measuring the thickness of the films, the targets were sandwiched between the source and the detector. Equation (1) shows the calibration obtained from figure 7.

$$E\ (\text{keV}) = (3.90 \pm 0.17) \times \text{Channel} + (108 \pm 276) \quad (1)$$

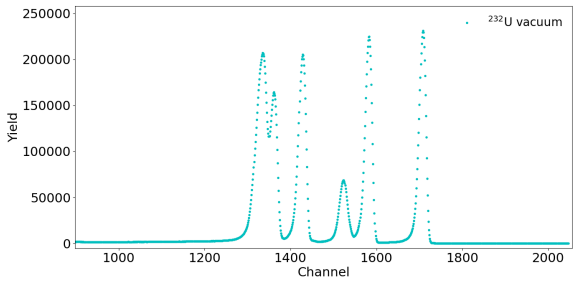

Figure 6. Spectrum of ^{232}U on vacuum, used for calibration of the detector.

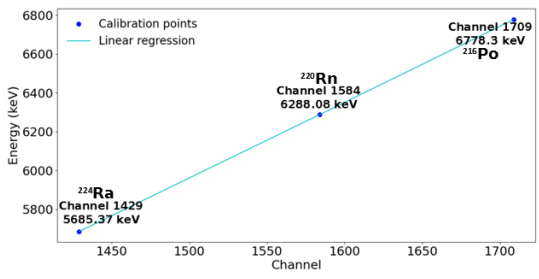

Figure 7. The energy calibration was obtained using a linear regression analysis.

In addition, a radioactive radium source was available for detector calibration. The radium source creates a spectrum with narrower and more isolated peaks, unlike the uranium source, where some of the peaks lie very close together. Despite this, the uranium source was used due to its higher activity, and consequently, the acquisition with ^{232}U is faster than with ^{226}Ra. To better understand the difference between the two sources, figures 6 and 8 show the spectra of ^{232}U and ^{226}Ra, respectively, both obtained with very similar acquisition times.

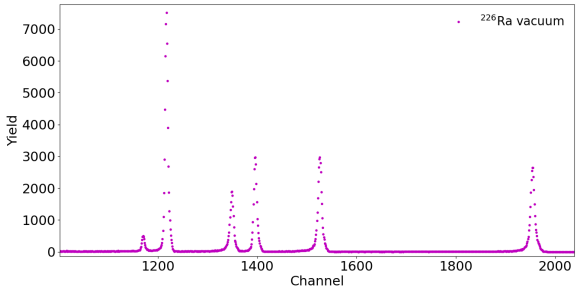

Figure 8. Energy spectrum of ^{226}Ra in vacuum. The observed peaks are 4601 keV (^{226}Ra), 4784.3 keV (^{226}Ra), 5304.4 keV (^{210}Po), 5489.5 keV (^{222}Rn), 6002.4 keV (^{218}Po) and 7686.8 keV (^{214}Po). This is a typical spectrum, used to convert channels to energy values.

4.3 Analytical method

The thickness Δx corresponding to the energy difference ΔE between the situation with and without the sample (the sample is positioned between the α particle source and the detector), is given by the following equation [2]:

$$\Delta x = \int_{E_i}^{E_f} -\left(\frac{dE}{dx}\right)^{-1} dE \quad (2)$$

Where $\frac{dE}{dx}$ is the stopping power. These values can be found on the NIST website [3] for different energies and many materials.

4.3.1 *Determination of the thickness of the formvar backing*

It is possible to see in figure 9 the energy lost in the peaks due to the α particles crossing the formvar film. To apply the equation (2), it is necessary to know the stopping power of formvar for different energies. Unfortunately, these values are not included in NIST.

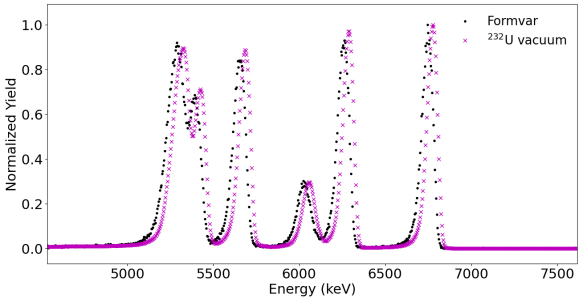

Figure 9. ^{232}U spectrum before and after crossing a formvar film, the loss of energy can be seen by the shift of the peaks.

The problem was overcome by an approximation approach, where these values were represented by PMMA (Polymethyl methacrylate), another plastic containing the same elements.

Table 1. Energy losses on formvar using the α particles' energy loss method and the estimated thickness of the formvar.

Element	ΔE (keV)	Δx (nm)
^{224}Ra	28.02	299.3
^{220}Rn	25.87	299.5
^{216}Po	32.19	386.3

The approximated thickness of the formvar backing was calculated by using the thickness measurements from the three peaks (third column) and computing the mean. The major difference on the last value, relatively to the other two, may be justified as a result of the uncertainties associated to the parameters of calibration. Therefore, the formvar film described in 4.3.1 has an estimated thickness of (328±58) nm.

4.3.2 *Thickness of the calcium fluoride film*

To estimate the thickness of the calcium fluoride, the difference between the completed target and the backing has to be evaluated. This difference can be seen in figure 10.

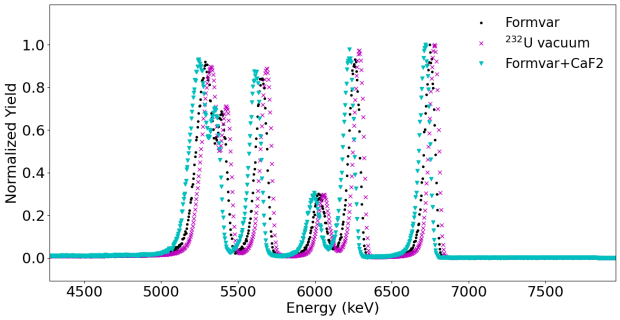

Figure 10. Spectrum before and after the evaporation.

The CaF_2 stopping powers were obtained from the NIST website, and the AlfaMC code was expanded to include this compound.

Table 2. Thicknesses and energy losses for the calcium fluoride film.

Element	ΔE (keV)	Δx (nm)
^{224}Ra	52.47	289.6
^{220}Rn	38.67	238.9
^{216}Po	28.42	182.2

Δx (CaF_2)= (75.4±17.5) μg/cm^2.
We conclude that Δx (CaF_2+Formvar)≈565 nm.

4.4 AlfaMC toolkit

AlfaMC toolkit is a Monte Carlo simulation that simulates the alpha's energy loss spectra in defined conditions [2]. With this simulation toolkit it is possible, in combination with the results from the analytical method, to predict more accurate values for each of the films´ thickness.

After setting parameters such as the stopping power, thickness of the sample and some of the detectors´ characteristics, it is possible to simulate the spectra and determine if they match the experimental data. The thickness used for the input is the first approximation, obtained from the aforementioned analytical method.

In the first approach, the input of the thickness was the value obtained in the analytic method, for both films. For the formvar backing, this value was initially 328 nm and, for the calcium fluoride film, 237 nm.

Overall, the value obtained for the thickness of the formvar backing looked correct. However, for the calcium fluoride film, a simulated spectrum of 210 nm provided a better fit. This value is consistent with the previously calculated value of (237±55) nm.

So, in conclusion the films' total thickness should be Δx (CaF_2 (237 nm) + Formvar (328 nm)) = (565±80) nm. Figures 11 and 12 show some of the simulations made using AlfaMC.

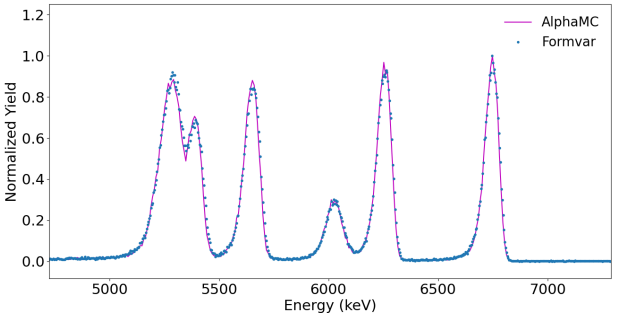

Figure 11. Simulated with AlfaMC and experimental values for a target consisting of a 328 nm formvar film.

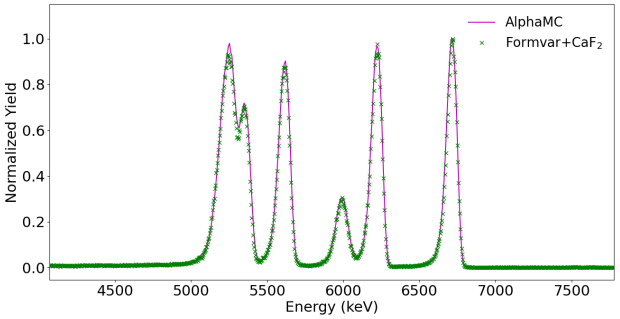

Figure 12. AlfaMC simulation and experimental values for a target consisting of CaF_2 (210 nm) + Formvar (328 nm).

5 Characterization of the films with RBS at CTN

The second method used for the films' characterization was the Rutherford Backscattering Spectrometry method (RBS). To do this, the 3 MV Van de Graaff accelerator at

CTN - IST (Centro Tecnológico Nuclear - Instituto Superior Técnico) shown in figure 13 was used, which produces beams of protons or α particles with energies in the order of some MeV.

Figure 13. Photograph of the Van de Graaff accelerator at the CTN-IST facility.

The beam will impinge on the targets in the chamber with the reaction to be measured in backwards angle by several detectors: RBS1, RBS2 and ERD. A scheme of these detectors' positions can be seen in figure 14. For analysis, the spectrum of the data has to be evaluated. The RBS provides information regarding - the thickness, the composition (including possible contamination) and the surface characteristics.

During this process, the samples are kept inside a chamber under a pressure of around 10^{-6} mbar and, with a goniometer, it is possible to ensure the position and relative angle of the target. Energy calibration is carried out using standards.

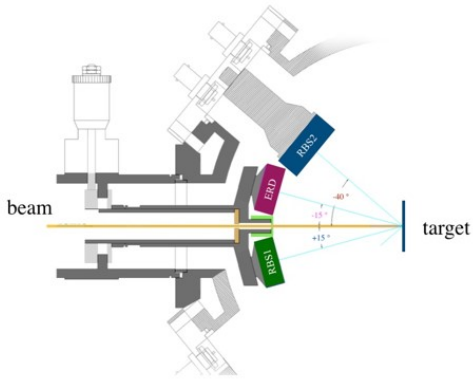

Figure 14. Detector scheme inside the RBS chamber at the CTN/IST Van de Graaff laboratory. Two detectors (RBS1 and ERD) are located under 165 °, and one detector (RBS2) is positioned under 140 ° with respect to the beam direction.

5.1 Characteristics of the RBS spectrum

The RBS spectrum is different for thinner and thicker films. For very thin films, each element has a well defined peak, separated from the others, which allows an analytical determination of each target's thickness. However, for the thicker films a surface barrier was observed, and thus the determination of the thickness is not possible.

5.2 Analytical Calculations

The thicknesses of the film can be calculated, as mentioned in section 5.1, from the backscattering spectrum using the element's kinematic factor (K) given by equation (3) [4]. The kinematic factor is proportional to the mass of the element (M), the mass of the beam's particles (M_b) and the angles involved (θ). Instead of making these calculations, the specialized SimNRA [5] software was used to deduce the thickness of each sample.

$$K = \left(\frac{\sqrt{1 - (M_b/M)\sin^2(\theta)} + (M_b/M)\cos(\theta)}{1 + (M_b/M)} \right)^2 \quad (3)$$

5.3 Simulations with SimNRA

To simulate the spectrum for the thin targets, the SimNRA software was used, where the layers present in the target and the experimental conditions, such as the beam's energy, are defined. As said previously, instead of making analytical calculations, a set of adjustments was made to the different layers' characteristics in order to try and fit the experimental spectrum to the simulated spectrum. Figures 15 and 16 show simulations made using SimNRA.

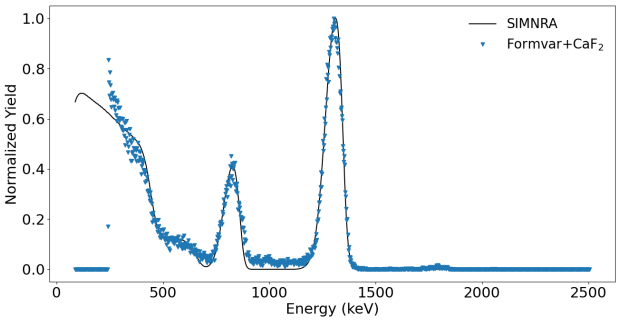

Figure 15. Simulation using SIMNRA and experimental values for one of the targets with CaF_2 (134 μg/cm^2) + Formvar (15.2 μm).

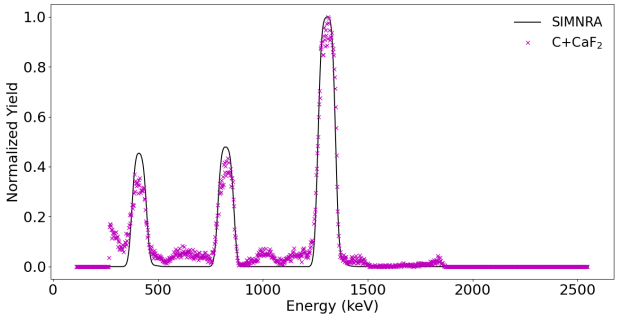

Figure 16. Simulation using SimNRA and experimental values for a target consisting of CaF_2 (126 $\mu g/cm^2$) + Carbon (83 nm).

6 Other targets produced

As mentioned in section 1, lead and gold targets were also produced during this internship. The process to make these was significantly easier than with the calcium fluoride films.

For both of these targets, the gold and lead was either evaporated onto a thick formvar film or onto a glass slide with a releasing agent. When evaporated onto the formvar, it is possible to dissolve the backing with chloroform, leaving only the evaporated metal. When evaporated onto a glass slide, it is possible to remove the film by dipping the glass-slide into distilled water at a 35 ° angle. The metal film floats in the water and can be fished onto the target frames. As an example, one of the gold targets produced can be seen in figure 17.

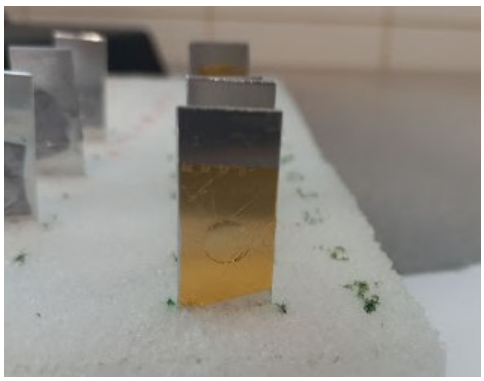

Figure 17. Self-supporting gold target with an approximate 100-150 nm thickness.

7 Conclusion

During this internship, a specific CaF_2 target with a formvar backing was developed, which has been successfully used in a dedicated experimental campaign at CMAM. Prior to sending the targets to Madrid, they were characterized using RBS, and alpha particles' energy loss through the target. For the energy loss through the target, the thicknesses were estimated using either the AlfaMC toolkit or SimNRA, which were in good agreement with the results from the RBS. This would indicate that the energy loss method can be used as a first approximation of the thickness. In addition, the RBS method has the advantage that it makes it possible to determine whether there are any contaminants within the film or not. From the stand point of a nuclear physics experiment this is very important to know, as it influences the quality of the experimental campaign.

The characterization of thin targets is very successful using the energy loss through the target methods. However, if the target is thicker, the method can not be used due to the fact that the peaks cannot be separated, and thus the thickness can not be estimated. On the other hand, this method has a clear advantage in that it is faster to obtain results, and that it is inexpensive since the particles originated from a source instead of a Van de Graaff accelerator.

The SimNRA code is a very easy manageable toolkit and allowed for the analysis of all the targets produced. The AlfaMC code uses as input the tabulated data from the NIST databank, and it is possible to expand this code manually giving complete control to the user to develop it as required. Future plans include the setup to be simulated using Geant4, creating an alternative method for confirming the thickness of the targets.

Overall, several targets were developed throughout the time of this internship, which were then sent to international facilities, where they formed an important part of the experimental campaigns.

Acknowledgements

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We also would like to thank the whole NUC-RIA group for receiving us.

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