Master's Thesis

Synthesis Of Thin Target Films for Nuclear Research and their Thickness Measurement using Alpha Source

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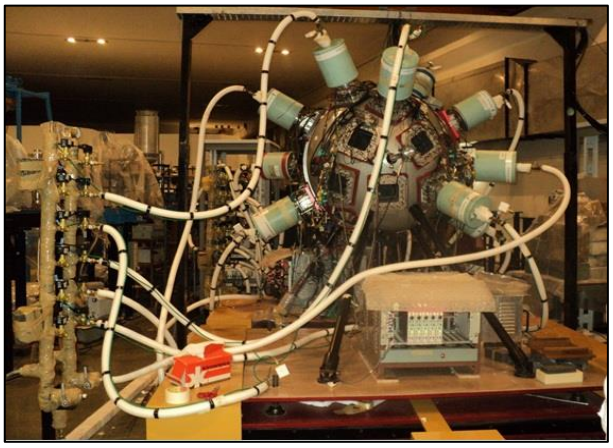
1. Introduction -

Thin films play a very important role in the field of Nuclear Research, without thin films almost no experimental research can be carried out in nuclear and atomic physics. Nuclear structure and various other properties of different elements can be obtained by experiments using thin films.

The thickness of such films varies in the range from fractions of a nano meter (monolayer) to several micrometres. These thin films are then carefully placed in the INGA setup where nuclear experiments take place.

2. Understanding Of the Setup



INGA (Indian National Gamma Array) is a collaborative research endeavor since 2001 By the nuclear physics groups from universities and Institutes across the country. It consists of an array of HPGe(High purity Germanium) clover Detectors. The response of fluid bodies to rotational stress provides a crucial framework to investigate widely different physical systems encountered in astrophysics, biophysics, nuclear physics, etc.

INGA is a powerful "femtoscope" for the study of structure of atomic nuclei at high spins. The nuclei are prepared in excited states (with 102l rotations per second) using energetic beams from the heavy ion accelerators. The fast-rotating state decays to the ground state, through the intermediate excited states, emitting copious gamma rays that are measured by the INGA detectors. By casting the nuclei to various shapes and studying their decays, the emergent properties of complex nuclear many-body system are elucidated.

3. Preparation Of thin Film (Chromium Thin Film)

For the purpose of research enriched chromium films of thickness in the range of 1-2 mg/cm^2 were required. There are various methods through which thin films can be prepared, a few of them are listed below:

a. Methods:

Physical Methods -

- **Thermal Evaporation**
- **Resistive Heating under High Vacuum**
- **Cold Rolling/Pressure Rolling**
- Electron-Bombardment Heating
- Flash Evaporation
- Arc Evaporation
- Exploding wire technique
- Laser Evaporation
- RF heating
- Cathodic Sputtering
- Sputtering
- Low pressure sputtering
- Reactive sputtering
- Glow-discharge Sputtering

Chemical Methods:

- Sol-gel technique
- Dip- Coating technique
- Spin-Coating technique
- Chemical Bath Deposition
- **Etching**

b. Substrate Preparation

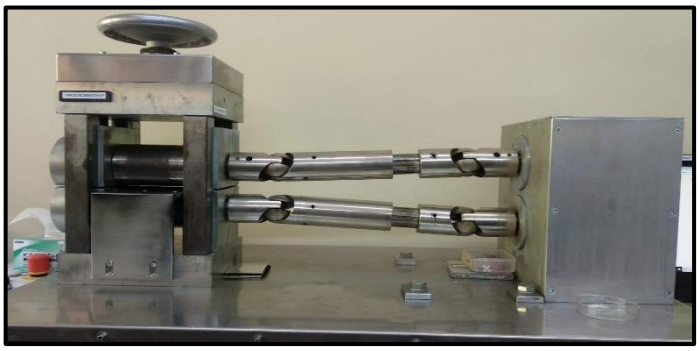
For The Preparation of chromium thin foils, we need a substrate that does not react with chromium and has good adhesion properties with chromium. Also, the substrate should be good enough under high temperatures, therefore we will need to go through three processes.
1. Cold Rolling/Pressure Rolling (Fig 2).
2. Resistive Heating under High Vacuum
3. Etching

We first need to prepare Copper thin films on which we will deposit chromium using resistive heating method since chromium is brittle and can't be directly rolled. As we know copper is a soft metal which is malleable and ductile therefore it is easy to roll copper into thin films using the cold rolling method.
Now a small copper piece was first taken then repeatedly rolled between two SS (Stainless steel) plates to make it thin to a certain level then it was cut in proper square shape so that it

can be further rolled into desired thickness. After every roll we have to clean the film with isopropyl alcohol. We need to make sure that we don't roll copper too thin as during deposition the chances of curling or melting of copper might take place. Therefore, copper foils of thickness ranging from 7 mg/cm^2 - 15 mg/cm^2 are required.

The process of rolling is very simple and the steps are as follows

SS plates (Fig 1)

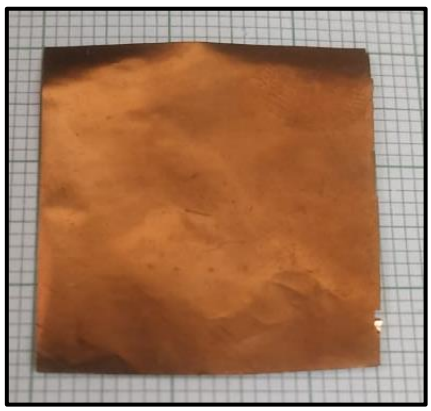


Rolling Machine (Fig 2)

- First place the material between the SS plates *(Fig 1)*.
- Now set the gap between the roller using the lever provided to roughly the thickness of both the plates sandwiched.
- Now press the forward movement button of the roller and allow the plates to expand. As the plates expand the material inside also starts expanding.
- Repeat the above step and clean the foil with isopropyl alcohol after every roll.
- After every roll remove the foil and place the foil in a different position the SS plates to avoid stamping on Foil.

In the process of rolling 4 different copper thin films were prepared in various thicknesses

1. Area - 3 x 2.8 cm / weight - 0.0656 gm
   Thickness - 7.809 mg/cm^2
2. Area - 3.2 x 2.8 cm / weight - 0.0731 gm
   Thickness - 8.15 mg/cm^2
3. Area - 2.5 x 2.5 cm / weight - 0.0853 gm
   Thickness - 13.64 mg/cm^2
4. Area - 2.4 x 2.4 cm / weight - 0.0802 gm
   Thickness - 13.92 mg/cm^2



Copper Film (Fig 3)

Once copper foils (fig 3) were prepared, they were carefully dipped in dilute nitric acid solution to make it rust and containment free and then kept in butter paper for further deposition of chromium

c. Deposition of chromium:

For the experimental purpose most, thin films prepared are enriched isotopes of the same element. Therefore, for the experiment **enriched isotope** of Chromium thin film was required. As Enriched isotopes are very expensive and cannot be mishandled or wasted, first optimizing of parameters was required. Therefore, for optimizing parameters first we have to use naturally occurring isotope of Chromium which is not very expensive.

It is utmost important to have a very clean deposition chamber as we are optimizing parameters for deposition of enriched material which will used later.

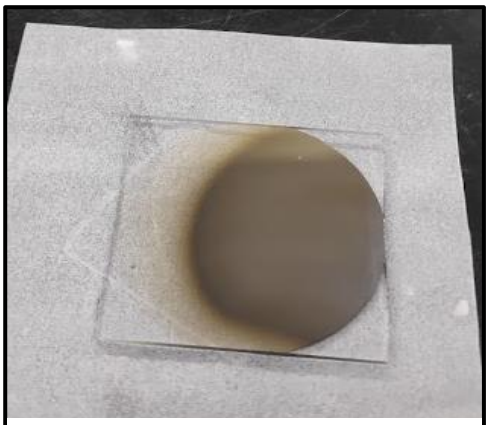
The parameters that need to be optimized are:

- Substrate height from boat
- Shape of the Boat (fig 4)
- Temperature at which Cr Evaporates
- Vacuum extent
- Amount of material required to achieve desired thickness
- Time needed for evaporation
- Speed at which current needs to be increased so as to increase temp.
- Deposition rate

To deposit natural chromium[4], we first take chunks of chromium and then use a hand ceramic grinder to grind to a coarse powder. The powder is then taken inside a tantalum boat and carefully mounted on the setup making sure that both the ends of the boat are properly attached to the electrodes of the heating setup. The shape of the boat is flat bottom cone. The height of the boat is 16mm and the opening diameter is 7mm.

Tantalum Boat (Fig 4)

First a blank run is taken in which instead of copper film we use glass slide(fig 5) for deposition. This is done to get the correct estimate of the knob position at which chromium starts to evaporate and deposit on to the film. Once this is known then we replace the glass slides with copper film and then we make sure to clean the chamber with isopropyl alcohol. The vacuum needed for our evaporation is in the range of 10^5 - 10^6 mbar. To get the vacuum inside the chamber we use a rotary pump in combination with a diffusion pump to achieve such a high



Blank Deposition(Fig 5)

vacuum. Once the vacuum level is reached, we slowly start increasing the temperature.
The reason for slow increase is that if there any vapour inside our material or boat it needs to be taken out before deposition happens. Chromium starts to evaporate at around 1600 ^{0}C in vacuum. Since we can't use the thickness monitor to get the amount of deposition, we have to evaporate all the material that we take in our boat so that we reach our required thickness of the chromium film (therefore adequate amount of powdered material should be taken so as to reach an optimum thickness). In our case roughly 11-14 mg of chromium material was taken to reach to a thickness in between 1-2 mg/cm^2.

Copper film Inside chamber (Fig 6)

Once chromium starts to evaporate it can be seen in the mirrored SS plate(fig 6) that was placed just below the boat to get view of the deposition on the copper film. Another important parameter is the time for deposition, we cannot keep the deposition time for a very long as the adhesion of chromium to copper is affected if the copper film is heated too much. Therefore, from the moment when desired temperature is reached and chromium starts to evaporate timer is started for 5 min after which we have stop the deposition.

Now the process of deposition is completed and the prepared film of chromium on copper is carefully taken(fig 7) out of the chamber and is sandwiched between two tantalum holders for further removal of copper as the requirement is of a self-supporting chromium film.

Final Film after deposition (Fig 7)

d. Extraction of self-supporting Chromium Film

It is rather a very simple process as copper dissolves in concentrated Nitric Acid solution.
But since the films are very thin, we have to keep in mind a lot of factors (vibrations when dipped in solution, heat, etc). Therefore, the concentration of solution needs to adjusted accordingly. (In our case 40% conc.)

Now the prepared foil is carefully attached to a tantalum holder which will be its final resting place and then it dipped

Self-supporting Chromium film on holder (Fig 7.1)

into the nitric acid solution (HNO_3). After a few hours all the copper will be dissolved in the solution releasing a brown gas (nitrogen dioxide).
The concentration is kept low so as to decrease the vibrations caused due to chemical reaction and therefore a longer duration of dipping is needed. The prepared film was dipped for about 6 hours.

Once we are able to get a perfectly fine self-supporting chromium (Cr_{52}) film (fig7.1) without any pinholes and deformations we can say that all are parameters are now set and we can now use the more expensive enriched isotope of Chromium to make thin film using the found parameters and follow exactly the same process.

e. Final thickness measurement

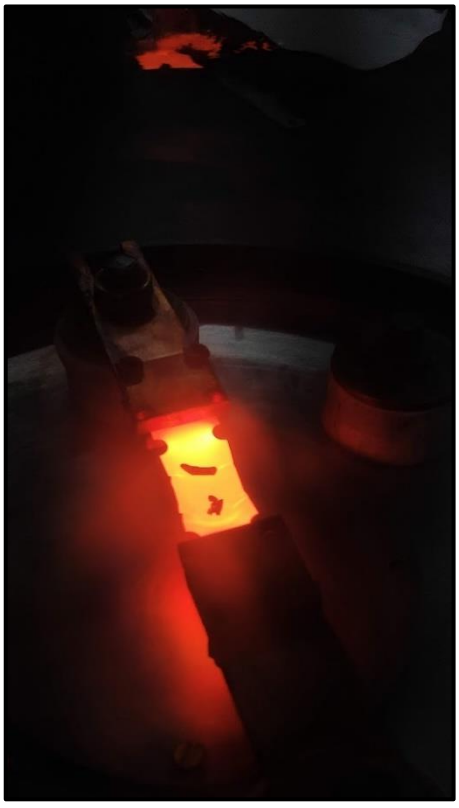
For the measurement of thickness one method that is easy to use is by first weighing the copper film and then weighing the same film after deposition and then difference between the weight measurement can be used to calculate the thickness using the below formula

Thickness – weight / area

The thickness was found to be around 1.5 mg/cm^2 which is approx. 2 microns

f. Preparation of Holmium on Gold film Using Rolling method

For preparation of Holmium on Gold the most suitable and easy method is to roll both the foil under pressure similar to the synthesis of copper but the difference here is that both the metals are very expensive and there many difficulties that we will encounter while rolling. Holmium is rare lanthanide therefore even a milligram costs too much not just that but also the abundance of holmium is very less on our planet therefore care needs to be taken while rolling so that we don't waste too much. Due to this reason rolling is the choice of method for the preparation of holmium thin film. Our backing material is gold.



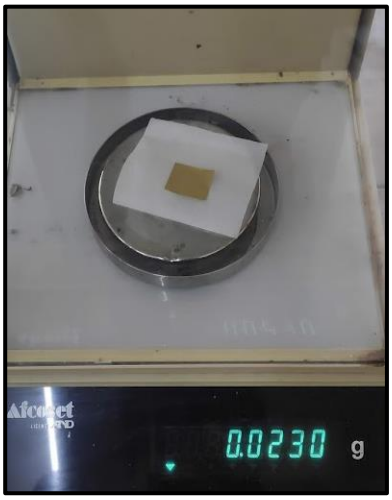
Heating of holmium (Fig 8)

As both the metals are malleable and soft it is possible to roll both the metals using a high-pressure roller. We needed a backing of 6 mg/cm^2 thickness of gold film [2]. A thin film of gold of thickness 14 mg/cm^2 was readily available therefore it was taken and then repeatedly rolled keeping them

sandwiched between two SS plates. After every roll gold was cleaned with isopropyl alcohol to keep contaminant out of the film. During rolling of gold one thing that needs to kept in mind is that it is very soft therefore pressure should be increased very slowly after every roll as too pressure might lead in tear of the film while we are peeling it from SS plates for change in position and cleaning. Gold was rolled to a thickness of around 6.5 mg/cm^2 and kept aside to be further rolled together with holmium.

The process of synthesizing Holmium thin film is bit different from other films as holmium easily oxidizes if kept open or alcohol is used to clean. Therefore, this makes preparation of holmium thin film very challenging. During my try one film turned into net (fig 10) as a little bit of alcohol was used to stick it to the SS plates since the film was curling.

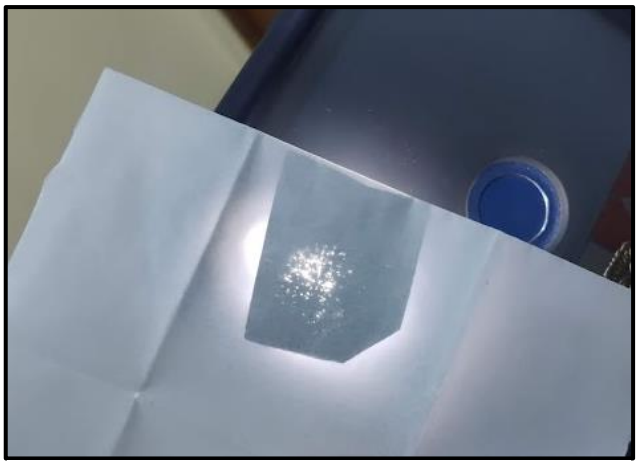
Holmium film (Fig 9)



Gold film (Fig 10)

A small piece of holmium was first taken and then heated (fig 8) at high temperature to get rid of the oxidation layer. Then the piece was rolled carefully without using alcohol. The thickness requirement for holmium was around 2 mg/cm^2. Therefore, the film was rolled to thickness of about 2.5 mg/cm^2.

Now since we needed holmium on gold backing, therefore both gold as well as holmium was stacked together on top of each other and then placed between the plates so that they can be rolled together. It was later encountered that gold does not stick well with holmium even after putting a lot of pressure and rolling which further lead to the thickness of the films decreasing beyond our requirements. Therefore, instead of gold we have choose any other material which has good adhesion properties with holmium. One such suitable metal could be lead.



Net effect Holmium (Fig 10)

Thickness Measurement Using Alpha Emitting Source

g. Introduction and Understanding of the Setup

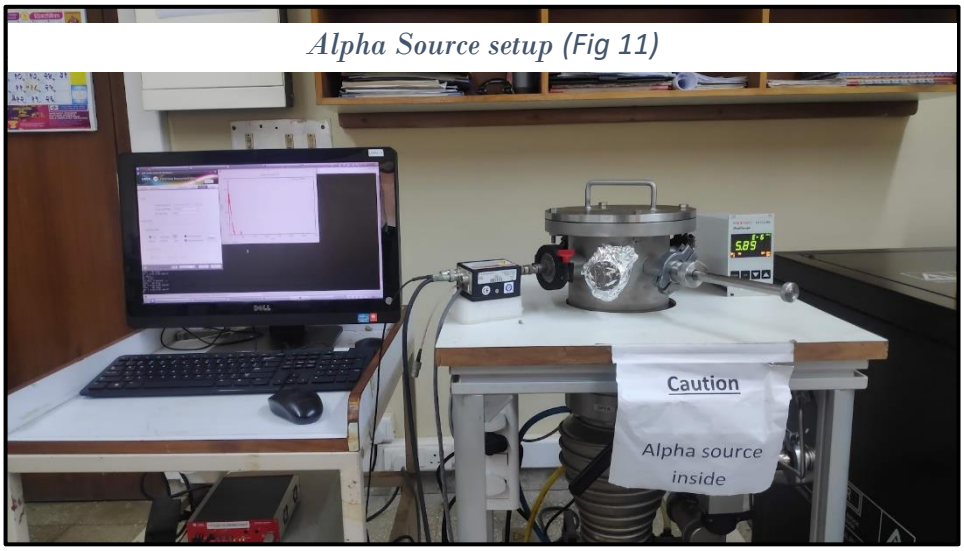
One method which is generally used in measuring the thickness is by weighing the foil on a microbalance scale and then finding its area using a graph paper. The ratio between the weight and the area will give us the final thickness. Since this method has a lot of errors such as

1. while weighing the microbalance scale was not tared properly
2. if the area of the foil is irregular it becomes difficult to take an estimate,

it is not reliable to get the thickness using this method if our experiment demands accurate results. Therefore, a method which is more accurate and will give results without errors needs to used.

Methods based on charged particles energy loss could be used to measure the thickness of the foil. Here we have a setup which has elements who emit alpha particles and could be detected easily on a detector[3].

Alpha Source setup (Fig 11)



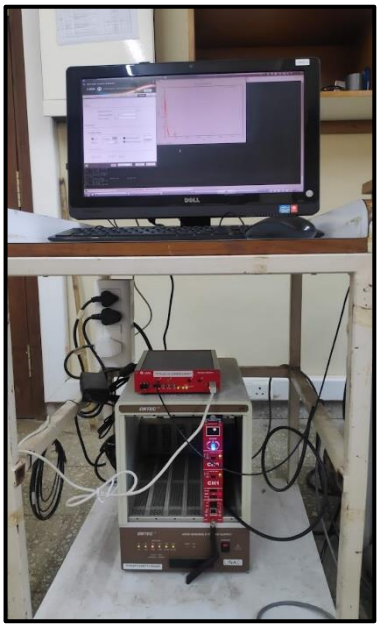
The picture of the setup is shown beside. It consists of a vacuum chamber. Inside the chamber at one end there is an alpha emitting source and exactly to the opposite end of the source there is a detector. The foils can be mounted on a retractable rod which will then be pushed in between the source and the detector. The source (fig 12) is a mixture of Americium (241) and Plutonium (239) while the detector is silicon surface barrier detector. The chamber needs to be in vacuum (below 10^3 mbar) for the measurement to take place since alphas lose energy even while traveling through air. Vacuum is attained by pumping the air out using a rotary and diffusion pump.

h. Mounting of the Film

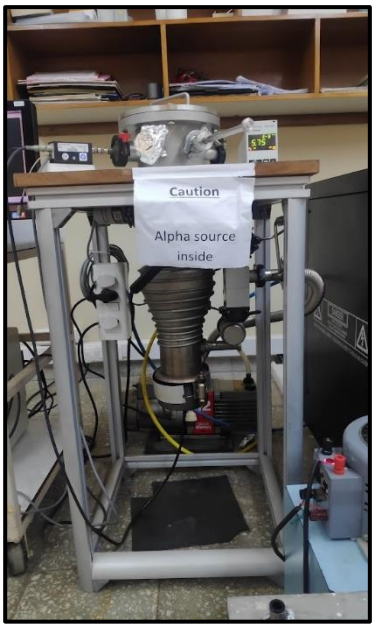
It is utmost important to mount the film on the rod very carefully as even a slight vibration might lead to crack and tear in the film (fig 13). Therefore, very carefully using forceps the film was mounted on the sliding rod of the setup.

Mounting of film (Fig 13)

Source (Fig 12)



Computer and bias provider



Pumps(Fig 14)

i. Readings Acquisition

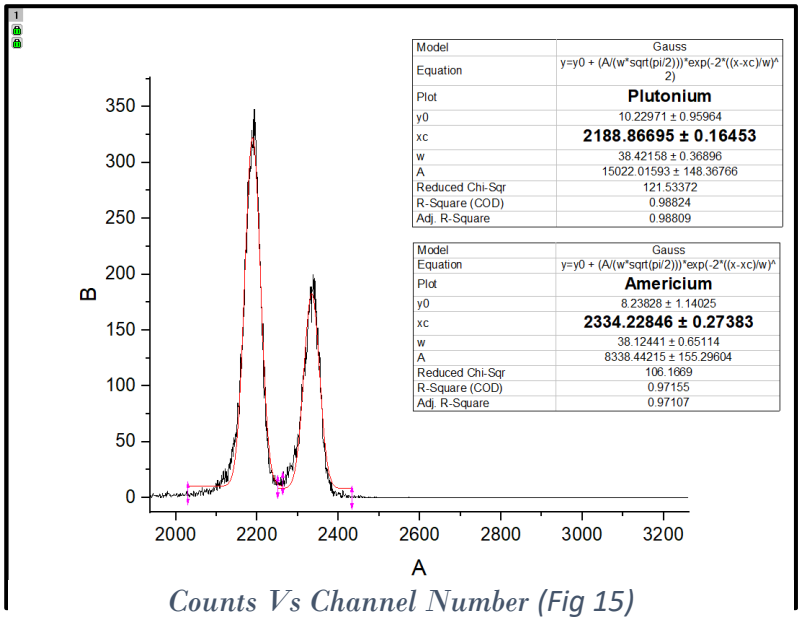
Once the film is mounted properly and chamber is bought into vacuum the next step is to start giving bias to the detector. Once 100V of bias is reached it's the right time to start the acquisition of data. Using software provided by the detector's manufacturer the reading of first the blank (no film between detector and source) is taken for 30 min. This data is then saved onto the computer. Now we need to the push the rod on which our film is mounted make sure that the source, the film and the detector are in a same line. Now we again start to take readings for 30 min. This data is again saved onto the computer.

j. Change of scale from channel number to energy

The data that we get on the graph are in terms of channel number vs counts, since we need the energy of the alpha particles, we have to convert from channel number to energy on x-axis. Therefore, for conversion we use two-point calibration.

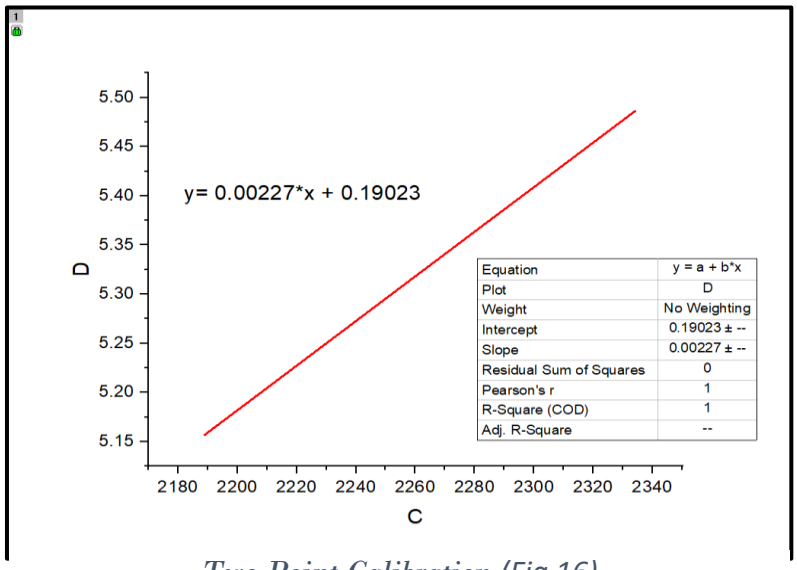
The steps for conversion are given below-

1. We first need to import all the data into Origin Software and then plot a graph of counts vs channel number (fig 15).



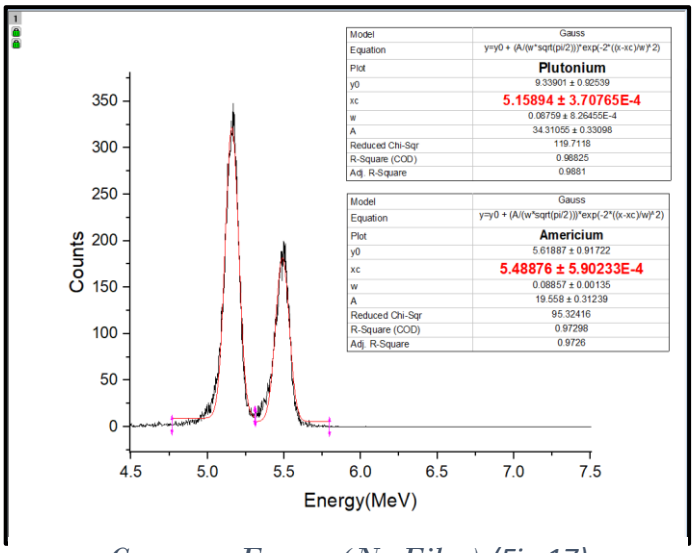
Counts Vs Channel Number (Fig 15)

2. Once this is done, we have to make curve fitting as the graph is not smooth and getting the point corresponding to maximum energy and count is difficult.
3. The choice of fitting is Gaussian as the graph is non linear and has normal distribution curve.
4. Once we fit the data, we get the peak points of both the elements emitting alpha.

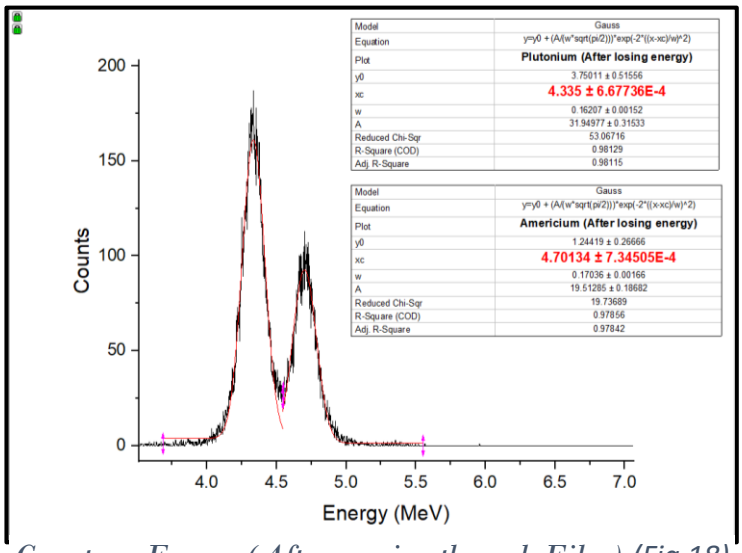


Two Point Calibration (Fig 16)

5. Since we know the energies of both alpha particles, we can use them to plot a graph of channel number vs energy which will be a straight-line graph (Fig16).
6. Using the equation of the line plotted we can convert the all the channel number into energies.
7. Now we can plot the graph for counts vs energy and again repeat the Gaussian fitting process to get the peak points.



Counts vs Energy (No Film) (Fig 17)



Counts vs Energy (After passing through Film) (Fig 18)

8. Now we will have two points for both blank as well as for when the film was in between source and detector (fig 17,18).
9. Using these points, we can determine the thickness of the film by stopping power data.

To determine thickness, we use the values given below in the table:

Element	E_α (MeV)	E' (After passing through Aluminium) MeV	ΔE MeV	dE/dx (Stopping power) MeV [1]	Thickness mg/cm^2
Am	5.48876	4.70134	0.78742	0.4590	1.715
Pu	5.15894	4.33500	0.82394	0.4782	1.723

The stopping power values are found from SRIM software.

k. Accuracy with respect to Direct weighing method

As we can see from the above table that the calculated thickness using alpha source is almost near to our value which was calculated using weight by area method.
Let's now estimate the error that we encountered while measuring thickness by weighing method

$$\text{Error (\%)} = ((1.7190 - 1.5)/1.5) * 100$$
$$= 0.1460 * 100$$
$$= 14.6\%$$

Therefore, there is an error of about 14% in the weight measurement compared to alpha thickness measurement

Reasons for behind the error:

1. The area taken while we calculate the thickness could have a lot errors as we are estimating the area considering all the sides are uniform.
2. The weight measurement could have a lot of error as the weighing scale will have instrument errors
3. Also, the thickness is considered uniform across the area which is not true as some portions of the area could have more deposition while some portions have less deposition.

Conclusion:

During the course, foils of different metals were prepared with different areal density which will further be used in research.

Copper foils of thicknesses 7 mg/cm^2, 8 mg/cm^2, 13.6 mg/cm^2, 13.9 mg/cm^2, Gold film of thickness 6.5 mg/cm^2 and Holmium film of thickness 2.3 mg/cm^2 was prepared.

Thickness measurement of chromium thin film using alpha source was done which gave accurate and corrected thickness of the prepared film. Also, the working and principle of the electron gun setup for target preparation was studied during which Boron thin films on backing of tantalum were prepared.

References :

[1] Stopping power data http://www.srim.org/

[2] C.K. Gupta, Proceedings of the DAE Symp. On Nucl. Phys. 59 (2014)

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[4] Chromium target preparation for the measurement of nuclear data for fusion technology (2014), Bhawna Pandey, P.M. Prajapati, Ajit Mahadkar , Journal of Materials Science & Surface Engineering Vol. 1 (3), 2014, pp 78-80

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