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OF THIN URANIUM, PLUTONIUM  
AND BORON SAMPLES FOR NEUTRON  
CROSS SECTION MEASUREMENTS  
IN  $4\pi$  GEOMETRY

by

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Central Bureau for Nuclear Measurements  
Geel, Belgium

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## PREPARATION BY ELECTRO-SPRAYING OF THIN URANIUM, PLUTONIUM AND BORON SAMPLES FOR NEUTRON CROSS SECTION MEASUREMENTS IN $4\pi$ GEOMETRY

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The preparation of thin films is an important requirement for any measurements in nuclear physics. If the required surface is large, the problem is furthermore complicated. A machine

and a technique are described which allow to prepare samples with a diameter of up to 5 cm. The method has been applied to uranium, plutonium and boron.

### 1. Introduction

Precise measurements of nuclear properties often need  $4\pi$  counting. A typical sample for such work, which we had to prepare is the following: A foil of 100 - 500  $\mu\text{g}$  element/ $\text{cm}^2$  which is self-supporting over a surface of at least  $25\text{ cm}^2$ . The element of interest should be pure, that means it should be a metal rather than a compound. For most of the elements there are serious problems involved in the preparation, handling and storage of such thin films of metals.

There are also only very few methods that allow to prepare such a target. Most of the methods that could be used for preparing thin films require a thin backing.

This backing has to be so thin, that its effect is negligible on the particles to be counted.

This means that the backing would have to be as thin as possible and mostly thinner than  $50\ \mu\text{g}/\text{cm}^2$ .

The choice of the elements in the backing is also limited by the type of experiment to be performed. Substances especially suitable for e.g. neutron measurements are carbon, organic materials, alumina or silica.

### 2. Backings

If we use such a thin supporting foil, a ring is needed to support this thin foil itself. Fig. 1 shows the kind of arrangement we have chosen.

We tried the following supporting foils:

a) Carbon foils prepared by decomposition of methyl iodide on nickel foils<sup>1</sup>).

It proved to be extremely difficult to prepare thin carbon foils larger than 2 cm diameter, so that all preliminary experiments were made on smaller carbon foils

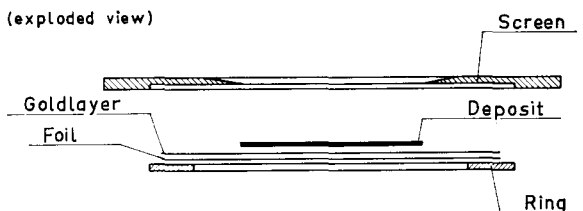


Fig. 1. Exploded view of backing and screen.

b) Thin nickel foils were prepared on copper substrates by electrolysis. Similar foils are available on the market.

Here it was possible to produce big foils and by electrolytic dissolution of the copper backing we were able to obtain self supporting nickel foils of 5 cm diameter, but these foils were rather inhomogeneous and unstable with time because of the rapid oxidation of these thin layers.

c) The vyns and cellulose foils were prepared in thicknesses from 10 to  $100\ \mu\text{g}/\text{cm}^2$ . The plastic foils were made conducting by metallizing with Al or Au by vacuum evaporation. Here again the thickness of the metal layer may vary from 10 to  $100\ \mu\text{g}/\text{cm}^2$ .

By varying the plastic and the metal thickness, foils with very different physical properties can be

<sup>1</sup>) E. Kashy, R. R. Perry and J. R. Risser, Nucl. Instr. and Meth. 4 (1959) 167.

obtained. In general: the thicker the plastic film the more mechanically strong is the foil. The thicker the metal foil becomes the more the optical transparency of the foil diminishes, whilst the electrical conductivity of the foil increases.

### 3. Choice of Method

#### 3.1. SOLUTION EVAPORATION AND PAINTING

Of the five methods taken into consideration we eliminated solution evaporation and painting because of the great difficulties encountered when using these methods for the preparation of homogeneous targets with large surfaces.

#### 3.2. VACUUM EVAPORATION

The thermal conditions during vacuum deposition are such that the extremely thin plastic films are easily damaged and so one has to place the target at a considerable distance from the heated substance. Therefore, the recovery factor for vacuum evaporation is extremely low, so that the vacuum evaporation of rare and  $\alpha$ -active isotopes becomes a quite problematic method. This especially as it is very difficult to obtain a homogeneous layer without losing considerable amounts of material. Besides of this, evaporation in glove boxes, which is necessary in the case of  $\alpha$ -active radio isotopes, is a difficult technique. There is also the danger re-

<sup>2)</sup> D. Carswell and J. Milstead, *J. Nucl. Eng.* **4** (1957) 51.

<sup>3)</sup> E. Bruninx and G. Rudstam, *Nucl. Instr. and Meth.* **13** (1961) 131.

sulting from the pyrophoric nature of evaporated Pu and U.

#### 3.3. ELECTROLYSIS

The next method we considered for the preparation of samples was electrolysis.

This method poses also a lot of practical problems and the electrolysis of a reasonable volume of solution on such an extremely fragile and thin foil again a very difficult technical procedure.

In table 1 the behaviour of different support materials is compared.

We can see that for the electrolytic method on two types of foils could be used and this only under very specific conditions. The results of these experiments are to be published later. Even though it is possible to electrolyse on such a thin foil, the technology becomes difficult with increasing size of foils. It is extremely difficult to obtain by electrolysis a layer which is larger than 3 cm in diameter.

#### 3.4. ELECTRO SPRAYING

One of the few methods remaining is the electro spraying method of Carswell and Milstead<sup>2)</sup>. The experimental conditions for preparing samples with small surface area for  $\beta$ -counting have been recently described by Bruninx and Rudstam<sup>3)</sup>.

## 4. Apparatus

#### 4.1. HIGH TENSION SUPPLY

The high tension supply is very simple and can be

TABLE 1  
Methods and supports used

	Solution evaporation	Painting	Electrolysis	Vacuum evaporation	Electro spraying
Carbon	(+)	(+)	(+)	+	+
Nickel	+	+	(+)	+	+
Vyns	(+)	+	—	+	(+)
Vyns + Al	+	+	—	+	+
Vyns + Au	+	+	(+)	+	+
Cellulose	+	+	—	+	(+)
Cellulose + Al	+	+	—	+	+
Cellulose + Au	+	+	(+)	+	+

— Not applicable.

+ Applicable.

(+) Only applicable under special conditions.

built easily from commercially available electronic components. The circuit diagram and components used are shown in fig. 2 and its legend. This installation allows us to work with up to 13 kV. One

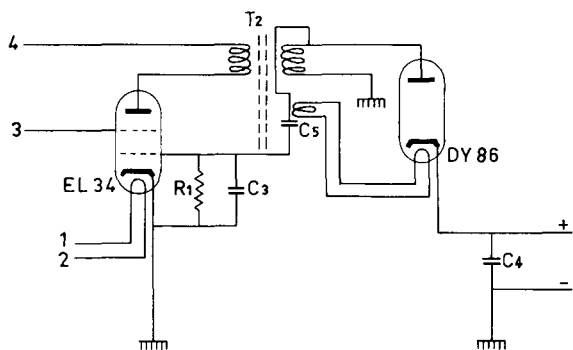


Fig. 2. Circuit diagram of the high tension supply.  
 high-tension transformer  
 100 k $\Omega$   
 50 pF  
 500 pF  
 condenser consisting of metallic plates at a distance of  $\pm 1$  cm from the secondary coil of the high tension transformer  
 } heating current for filament  
 } bias adjust 220-280 V.

advantage of the kind of generator we have used seems to be that the unit is not stabilized. If there is some secondary effect at the capillary, reducing the flow of liquid, the resulting decrease in current produces a rise in tension which again tends to balance the decreased flow rate. On the other hand, if the flow rate becomes too high, this means when it reaches the dropping point then the increasing current lowers at the same time the output voltage, which again balancing somewhat the overfeed of the capillary.

2. CAPILLARIES

Normally the capillaries used for spraying are simple straight capillary tubes holding only limited amounts of solution (fig. 3). In our experiments we had to prepare samples, containing much larger amounts of isotopes. A greater volume of solution has to be used. Because of the change in height of the liquid column it was impossible to obtain a constant liquid pressure and hence constant spraying conditions. It, therefore, proved to be

necessary to construct a container which allowed a free flow of liquid for a considerable length of time. In case of an appreciable evaporation from the upper or the lower part of the capillary one of the well-

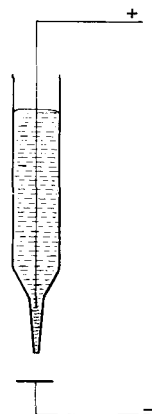


Fig. 3. Normal straight capillary.

known troubles with normal thin capillaries is that they tend to clog. This is mainly due to crystals forming within and even more at the surface of the solution and then lodging themselves in the finest part of the capillary. We have developed different types of capillaries with a small reservoir for the solution.

Because of the above mentioned clogging effect the first type of capillary we tried (fig. 4) proved to be not very useful. The next type we constructed was a slight modification and the addition of the

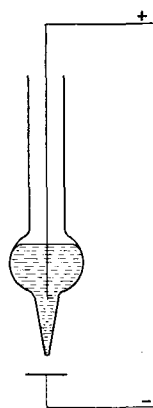


Fig. 4. Straight capillary with liquid container.

A swans neck improved the performance of the capillary considerably (fig. 5). The introduction of the fine wire into the tip of the capillary was very tedious and the position was not easily reproduc-

ible. The capillary we are using presently is shown in fig. 6. The hypodermic needle that is cemented to the glass part has two advantages. First the bore of the needles we are using is known with a good

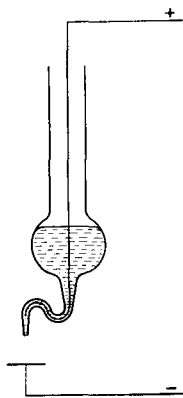


Fig. 5. Capillary with swans neck.

precision. The ideal opening in our case proved to be 0.05 – 0.08 mm. As can be seen from figs. 3 – 6 the electrical contact with the solution is made via a wire. The thickness of this wire has to be adjusted in such a way that the final opening of the capillary is just in the above mentioned optimum region. In the case one does not dispose of the appropriate Pt-wire, it is often possible to use a tungsten wire that has been reduced to the desired thickness by heating it in a flame and stripping off the formed

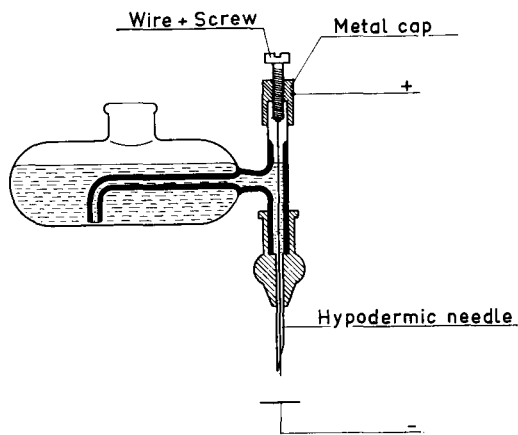


Fig. 6. Capillary with hypodermic needle tip.

oxide. In this way we have also produced conical wires that, used together with a self-drawn glass capillary, allows to have a very reproducible and

<sup>4</sup> J. C. Charlton, RCC-R119 (1961).

small opening as the wire functions like a needle valve.

The second advantage of the metallic capillary we are now using is that the spraying is very steady and the standing drop formed at the tip is not disturbing the even spraying over a considerable length of time. This is not the case when glass capillaries are used.

The wire is fixed to a screw and can be easily lowered and raised, thus allowing to adjust the flow rate. One is also able to clean the capillary itself during spraying by turning the screw several times up and down. In view of very high purity solutions, that should not be contaminated, it would be preferable to replace the hypodermic needle by a small piece of Pt-capillary that could be sealed directly into the glass or quartz container.

## 5. Preparation of Solutions

We prepared foils with uranium, plutonium and boron deposits.

### 5.1. URANIUM AND PLUTONIUM

The U and the Pu were sprayed in the acetate form as experiments had shown that these compounds are practically not hygroscopic and are soluble in methyl alcohol.

The preparation was started with the acidic nitrate solution.

This nitrate solution is evaporated in a platinum dish and the residue then calcined for 4 hours at 250° to 300° C. The so formed mixed oxide is then easily dissolved on heating in aqueous acetic acid. The solution is very carefully concentrated using the apparatus described by Charlton<sup>4</sup>), then carefully recrystallized and the acetate is dissolved directly in methyl alcohol.

### 5.2. BORON

In case of the boron samples a direct dissolution of boric acid ( $H_3BO_3$ ) in methyl alcohol or acetate is possible. The product sprayed from this solution corresponds exactly to the above formula.

## 6. Spraying Machine

The spraying machine has been constructed in our workshop and gives us the possibility to co-



with our capillary a surface of up to  $8 \times 9 \text{ cm}^2$ . A schematic view is given in fig. 7. The type of coverage obtained can be seen from fig. 8 which gives sweep diagrams of the machine. It can be seen

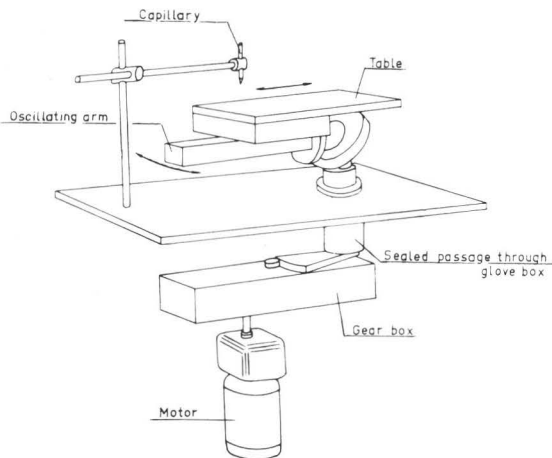


Fig. 7. Schematic view of the spraying machine.

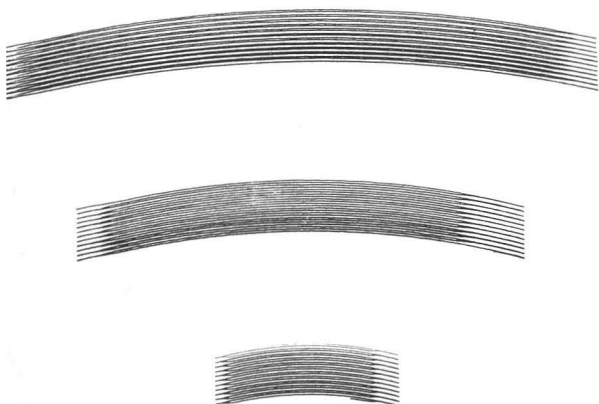


Fig. 8. Sweep diagram of the machine.

From this, that coupled with each back and forward sweep there is a small perpendicular movement of the carriage. The complete installation is fixed under a glove box in such a way that the major parts of the mechanical drive, including the motor, are outside of the active area. Fig. 9 shows the installation before mounting in the glove box.

**Description of Layers Obtainable**

**1. MACROSTRUCTURE**

Fig. 10 shows the type of layer obtained with uranylacetate.

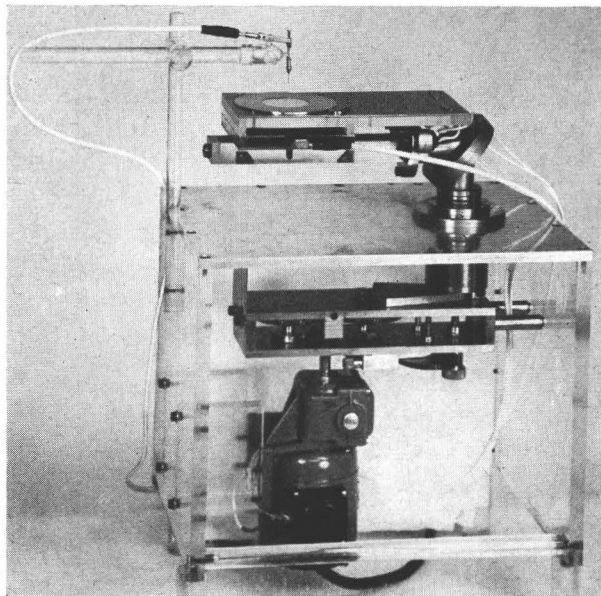


Fig. 9. Complete installation before mounting in the glove box.

**7.2. HOMOGENEITY**

The homogeneity was measured in the case of U and Pu samples by low geometry counting of the layer using collimating screens for picking out the wanted part of the surface to be measured.

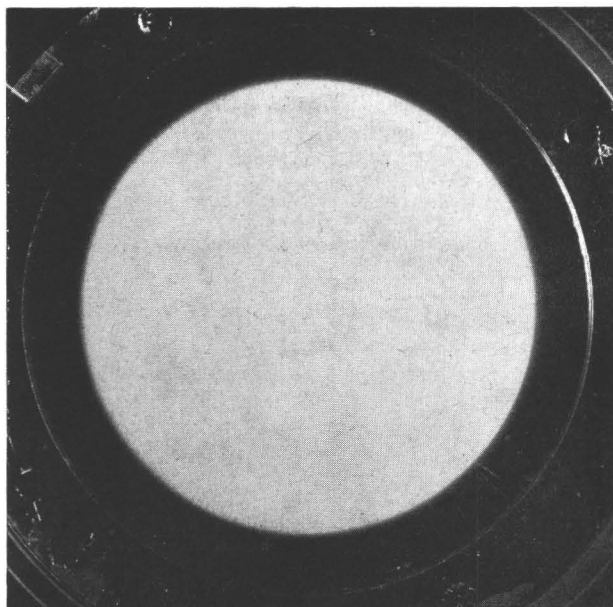


Fig. 10. Aspect of an uranium acetate layer.

### 7.3. STABILITY

#### *Uranium acetate*

Uranium acetate can be kept under normal laboratory conditions without any measurable change in aspect and weight occurring. Under vacuum there is no weightloss.

The method has been also used to prepare layers of uranium oxides on thick metallic supports (Pt, etc.). By applying successive coats and intermittent calcination of the acetate to the oxide, layers of several mg uranium oxide/cm<sup>2</sup> have been prepared. These relatively thick layers are perfectly stable, adherent and have the same homogeneity as the acetate layers they were prepared from.

#### *Plutonium acetate*

The behaviour of the plutonium acetate is identical with the one of the uranium acetate.

#### *Boric acid*

The boric acid is stable under normal conditions. Under vacuum the boric acid has such an appreciable vapour pressure that it slowly evaporates. It is, therefore, only possible to use such boric acid layers under atmospheric or higher pressures.

#### *Efficiency*

The efficiency of the method depends on the ratio of the area of the layer to be prepared and the area covered by the machine used. We are using metallic diaphragms and the product sprayed onto these

diaphragms is easily recovered. The loss occurring at the tip of the capillary by cleaning away crystals is also very small and all the material lost in such a way during one preparation is recovered chemically and can be reused after retreatment. Considering the above mentioned points we can say that our efficiency is better than 95%. In case of extremely diluted solutions the losses by adsorption are becoming important and there the efficiency is of course, much less.

### 8. Conclusions

The described method allows us to prepare layers of chemical compounds on any kind of conducting surface. The thickness of these layers is not limited to any special value and the surface area of such a layer is only limited by the machine used for the preparation. The method has been applied to uranium, plutonium and boron compounds.

The layers prepared varied from zero to over several milligrams of compound per square centimeter.

The homogeneity of the prepared layers was about 1%. The method is applicable to any compound that can be dissolved in an appropriate solvent.

### Acknowledgements

We are indebted to Mr. Moret and Mr. Van Saer for the design of the spraying apparatus. The plastic foils have been prepared using a technique developed by Mr. Spennol.



