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by

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A NEW METHOD FOR PREPARING RADIOACTIVE SOURCES*

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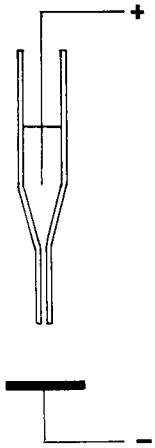
/e describe a new technique for preparing thin and uniform idioactive sources. The method is based: (1) on the spraying of a olution, containing the radioactive nuclide, by using a mechanal nebulizer; (2) on the use of a high electric field applied etween the nozzle of the nebulizer and the collecting surface.

This process is very simple and can be widely used. The yield of the deposition ranges between 50% and 70% but can even be higher than this last value. The deposition results uniform and compact.

Introduction

The preparation of uniform and thin sources is of reat interest for many researches in radioactivity. everal authors dealt with this problem and proposed arious and original solutions. A complete and detailed view of the different methods proposed was made by arker *et al.*[1]). These authors examined all the sugested and applied procedures up to 1959 and reported a ide bibliography of the published papers. Later, other searches were performed and interesting results apeared on this subject[2–8]).



g. 1. The principle of the original apparatus, proposed by Carswell and Milsted.

One of the more recently proposed method is Bruninx nd Rudstam's which improves the method firstly sugested by Carswell and Milsted[9]) in 1957. Essentially, the rocess consists of the electro-spraying of a solution ontaining the active nuclide. The principle of the ethod is shown in fig. 1. By applying a d.c. voltage of out 5000 to 10000 V, between the positive electrode mmersed into the liquid and the metallic foil, the liquid is forced out of the capillary and dispersed as extremely small droplets. The detailed study of this method, performed by Bruninx and Rudstam, has shown:

1. The salt containing the active nuclide must be soluble in a solvent of high vapor pressure and low surface tension;

2. The selection of the more convenient diameters of the capillaries and wires is difficult, it requires a large disponibility of samples among which to select, tentatively, the better ones; such tests are to made preliminarily with non active solutions;

3. The growing of air bubbles into the capillary damages the spraying; the quenching of the bubbles is not easy to obtain;

4. The deposit turns out very uniform and fine-grained (of the order of a micron);

5. By heating the metallic (aluminum) thin plate collector at 600° C, it is possible to obtain a very sticking deposit;

6. The spraying of 50 μliter, requires times ranging from some minutes to about half an hour; this fact implies fairly long times for spraying greater quantities when this is necessary.

We attempted this method for preparing a source of

This work was supported by Euratom–CNEN contract.

1) W. Parker, M. De Croës and K. Sevier, Nucl. Instr. and Meth. **7** (1960) 22.
2) E. Bruninx and G. Rudstam, Nucl. Instr. and Meth. **13** (1961) 131.
3) W. Parker, M. De Croës and K. Sevier, Nucl. Instr. and Meth. **7** (1960) 163.
4) L. B. Warner, Nucl. Instr. and Meth. **14** (1961) 315.
5) V. A. Gorodyskii, Y. F. Romanov, A. V. Sorokina and M. I. Yakunin, Prib. Tekh. Exp. **5** (1959) 128, (Quoted by 6).
6) D. Michelson and H. O. V. Richardson, Nucl. Instr. and Meth. **21** (1963) 355.
7) W. Parker and Y. Grunditz, Nucl. Instr. and Meth. **22** (1963) 73.
8) M. Mladenovic and Ali El Farash, Nucl. Instr. and Meth. **23** (1963) 175.
9) D. J. Carswell and J. Milsted, J. Nuclear Energy **4** (1957) 51.

Ca^{47} for an experiment on $\beta - \gamma - \gamma$ angular correlations[10]). Our tests evidenced two troubles, very hard to eliminate:

1. For selected wires and capillaries, in repeating the trials, sometimes took place an intermittent breakdown of corona discharge in air, without the deposition of the solution; the μA-meter, controlling the rate of deposition, indicated current flow but the spraying was absent;

2. In supposed good conditions, the behaviour of the apparatus was not always reliable; sometimes a spraying of large droplets occurred, which damaged the deposit already made.

The reason of these difficulties lies, probably, in the mechanism of the nebulization of the solution by the electric field.

2. The Present Method

We thought that these troubles could be avoided by dividing the electrospraying into two distinct steps: mechanical nebulization of the solution and use of an electric field for collecting the cloud on the deposition surface.

The first purpose was carried out by using commercial nebulizers, commonly employed in the aerosol therapy. The air jet produces a cloud of very fine droplets and transports them.

The electric field was applied between the nozzle of the nebulizer and the thin metallic plate, heated at a temperature of the order of 500° C.

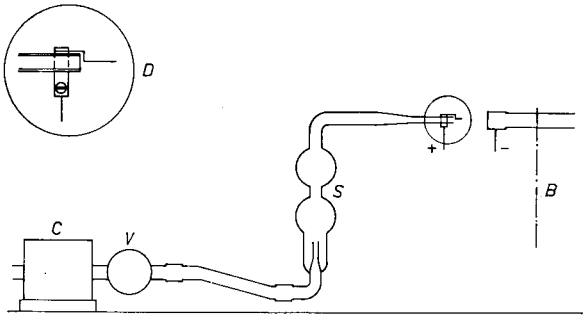


Fig. 2. The principle of the present method: C, pulsed air compressor; V, pin valve; S, nebulizer (very enlarged in the sketch); B, metallic rod on which is set up a cover of aluminum working as collecting sheet; D, detail of the positive electrode.

The principle of the set up is sketched in fig. 2. C is a pulsed air compressor, V a pin valve, S the nebulizer (very enlarged in the sketch) and B a metallic rod heated by a flame. On the rod there is set up, very sticking, a little cover of aluminum, which works as collectin surface. D shows, enlarged, the positive electrode. Th is formed by a nickel-chromium wire of 0.3 mm in dia meter placed at the nozzle of the nebulizer. Our resul have shown that the best working conditions are th following:

1. Diameter of the efflux tube: 5 mm;
2. Length of the positive electrode wire: 1 cm;
3. Distance wire-collecting sheet: 2 cm;
4. Electric field: 10 kV.

In these conditions, the absolute yield is higher tha 50% on a collecting surface of 1 cm^2 and higher tha 70% on a surface of 2 cm^2. The measurements have bee made with solutions having a concentration of 3 mg/cm^3 the yields were determined by weighing the deposite salt with a sensitive balance (0.1 mg). The precision of ou measurements was better than 10% for high yields o deposition and of the order of 50% for lower yields.

For the sake of good deposition, the followin cautions are to be held in account: the sheet must b heated at about 500° C; the solution of the active salt sprayed in a volume of $2 \div 4$ cm^3; for the purpose o washing the nebulizer, sprayings of ethanol are to b made; it is convenient to repeat at least four times ever spraying lasting $6 \div 8$ minutes; preliminary trials wit non active solutions are to be performed; this caution of course banal, but undoubtedly useful.

At the end we call attention to the necessity of workin in conditions of safety in order to protect ourselves fro the radioactive cloud which is not collected.

3. Influence of the Different Parameters

We studied the influence of the different parameters o the processes of nebulization and deposition; in th present paragraph we refer on the experiments perform ed and on the results obtained.

3.1. Nebulizer

In general the process does not depend upon th nebulizer; the only condition that must be satisfied that the cloud results "dry", that is made by very fin droplets. We performed numerous experiments with th more different types of "dry" nebulizer, either found i the market or built "ad hoc". The results were satis factory.

3.2. Solvent

The method is fit for whatever solvent. Water, eth alcohol, ether, mixtures alcohol-water and so on, gav good results. With the water, the nebulization is mor difficult and it needs a lot of washings. When possible, is convenient to use ethanol.

10) E. Fuschini, V. Gadjokov, C. Maroni and P. Veronesi, Nuovo Cimento **29** (1963) 310.

.3. Efflux Velocity and Distance Between wire and Collecting Surface

These two parameters strictly depend on the optimum f the nebulization. In other words, the amount of air in- ected into the nebulizer, by regulating valve V con- eniently, must ensure the making of a very fine and uni- orm cloud. However this amount should not be much igher than the quantity needed to have this situation. uch a condition is to be fixed disregarding the other arameters. As a consequence, the efflux velocity is egulated by the diameter of the nozzle. We made several sts to study the influence of the efflux diameter on the roplets stream. The results we obtained, with efflux dia- eters of 3, 4, 5, 6, 8 and 10 mm, do not suggest to use ozzles of diameter smaller than 4 mm. In fact, as a good ebulization requires a consistent injection of air into the ebulizer, the stream gains velocity if the efflux dia- eter is reduced. Small diameters, that is high efflux elocity, allow a satisfactory uniform deposition only if ne distance between wire and collecting plate is in- reased. This would reduce the effect of the electric field nd very high tensions (> 10kV) would be necessary. ummarizing the distance appears to be a parameter rictly dependent on the efflux velocity, that is on the ozzle diameter.

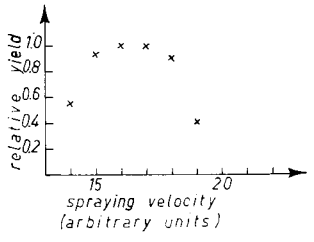


Fig. 3. Yield as a function of the spraying velocity.

Fig. 3 shows the influence of the efflux velocity on the ield. In ordinate are reported the relative yields and in bscissae the graduations of the regulating valve; they re, therefore, arbitrary units of efflux velocity. The gure clearly shows the existence of a range of velocities here the yield reaches an optimum. For small velocities ne yield is low because of the poor nebulization, for igh velocities the strong conveying movement desperses ne cloud, forbidding a good deposition.

4. Electric Field

It has a very important role in this method. The higher ne field, the higher the yield; the field, therefore, is to be laced at the highest value. If a sufficiently high voltage enerator is not available, it is necessary to decrease the istance between the end of the wire and the collecting neet. To keep the optimum distance between nozzle and

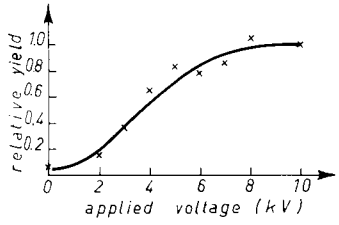


Fig. 4. Yield as a function of the applied voltage between wire and collecting sheet.

sheet, the wire must be lengthened. However, its length should not be more than 2 cm.

Fig. 4 shows the behaviour of the relative yield as a function of the field. The curve was obtained in the conditions 1, 2 and 3 of the previous section, changing only the voltage applied to the electrodes. The strong influence of the electric field on the yield clearly appears.

3.5. Collecting Surface

In the conditions 1 to 4 above stated, the yield depends upon the area of the collecting surface as fig. 5 shows.

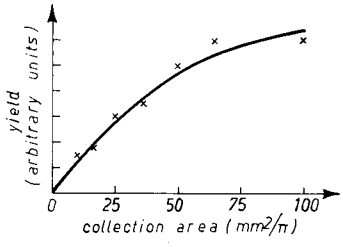


Fig. 5. Yield as a function of the collecting area.

4. Uniformity of Deposition

Our results have shown a good uniformity of deposition, very good in the central part of the deposit; fig. 6 shows the behaviour of the thickness as a function of the distance from the center. The tests have been performed by spraying Sr^{90} ($\beta_{max} \sim 545$ keV) and Y^{90} ($\beta_{max} \sim 2270$ keV) on a thin circular aluminum sheet of 20 mm

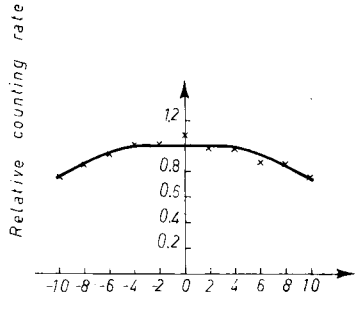
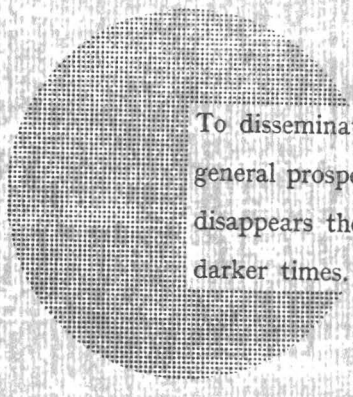


Fig. 6. Thickness of the deposit measured by β counting.

in diameter. The sheet did not turn during the deposition. We have assumed that the thickness of the deposit was proportional to the number of electrons emitted per unit area. The counting has been made with an anthracene scintillation counter, shielded by a lead collimator having a hole of 1 mm in diameter, and an electronic chain consisting of a linear amplifier, an integral discriminator and a scaler. The measurements were performed on various diameters of the deposit. As fig. 6 shows, w have a very satisfactory uniformity in a central region o about 4 mm in radius; a thinning of the order of 25% appears at the extreme edges. The results reported ar averaged on measurements performed on several dia meters which resulted in good agreement among them selves. It is evident, thus, a quite marked circular sym metry in the deposition.



To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel