# TECHNIQUES FOR HANDLING AND USING $Ba^{140}$ AS A TRACER

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Office of Naval Research

## TECHNICAL REPORT NO. 15

N 7 onr-292 T.O. 5
NR 074 081
October 15, 1953

*DEPARTMENT OF PHYSICS*
*UNIVERSITY OF MISSOURI*
*COLUMBIA*

# TECHNIQUES FOR HANDLING AND USING $Ba^{140}$

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# Abstract

Some experiments on oxide coated cathodes are described in which the radioisotope $Ba^{140}$ was used as a tracer. An oxide cathode of BaO containing $Ba^{140}$ was used as an evaporator in a tube which contained several movable receivers on which the evaporated BaO was deposited. The activity of the receivers indicated the amount of barium deposited. Using this method P. N. Russell had previously verified that the time rate of deposition is constant at a given temperature. The present tube design permits making the activity measurements through a thin glass bubble window in the side of the tube. Using it the rate was measured at which "free" barium, produced at the cathode-base metal interface, diffuses to the surface and is evaporated to the anode. A technique of spraying radioactive $BaCO_3$ was developed in order to produce cathodes of normal coating weight and density containing appreciable $Ba^{140}$ (.25mc/gm). These cathodes were operated under quiescent conditions for various periods of time. This activity is related to the interface thickness and its rate of formation could be determined as a function of life.

## INTRODUCTION

The use of $Ba^{140}$ as a tracer was first introduced in this research program by P.N. Russell[1] in 1952. Russell was interested in determining the rate of evaporation of BaO experimentally and in comparing it with the rate calculated from thermodynamic data. Since the quantity of BaO deposited by evaporation was small and since BaO is unstable in the presence of air and water vapor, direct chemical and physical procedures for measuring the amount present are not feasible. Thus the use of a radioactive tracer was considered. Isotopes of oxygen were immediately dismissed because of their short half lives of two minutes or less. This left an isotope of barium as the only other choice. Fortunately $Ba^{140}$ possesses the necessary qualifications, being a $\beta-\gamma$ emitter and having a half life of 13.4 days[2]. Being a product of the fission process, it was readily available.

In handling radioactive barium it was necessary to have a variety of instruments available for conducting the experiments and for radiological monitoring. Quantitative measurements of radioactivity were made using an end window Geiger tube and a conventional binary scaler. An ionization chamber and Geiger counter type survey meters monitored handling and decontamination procedures. These instruments

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[1] P.N. Russell, Technical Report No. 14, July 1, (1953).

[2] J.M. Cork, et al, Phys. Rev. 83, 856 (1951

were able to discriminate between $\beta$ and $\gamma$ radiation. All personnel were equipped with film badges and pocket dosimeters. Extreme care was taken to conform to the decontamination procedures and the standards for maximum permissible exposure prescribed by the AEC.

## PREPARATION AND HANDLING OF $Ba^{140}$

The $Ba^{140}$ isotope was obtained in 10 millicurie amounts from the Oak Ridge National Laboratory in the form of a solution of $Ba^{140}(NO_3)_2$ in dilute $HNO_3$. If the volume was much less than 10cc doubly distilled water was added by means of a calibrated pipette until the volume was 10cc.

For the preparation of radioactive $BaCO_3$ with high specific activity (2-30 millicuries) it was convenient to perform all operations in a centrifuge tube. The $BaCO_3$ was precipitated from a hot saturated solution of $Ba(NO_3)_2$ using $(NH_4)_2CO_3$. If it was desired to purify the radioactive material by recrystallization with the carrier the procedure described in Appendix I was used. For many applications this may not be necessary. In this case c.p. or purified $Ba(NO_3)_2$ may be used depending upon the purity desired.

Larger quantities of lower specific activity were made in beakers of appropriate size. Again c.p. $Ba(NO_3)_2$ is sufficiently pure for most work. The steps involved in preparation of the carbonate are similar to those in the micro-scale work and are described in Appendix II.

Special attention was given to all radioactive wastes to prevent contamination and health hazards. Wherever possible, the principal activity in the waste solution was removed and discarded* as active solid material. Active solutions were discharged to the public sewers after sufficient dilution.

## USES OF $Ba^{140}$

As mentioned previously the first use of $Ba^{140}$ in this research program was in Russell's determination of the evaporation rate of BaO. The results of this experiment, shown in Fig. 1, were found to agree within a factor of two with the rates calculated from the vapor pressure of BaO and geometrical considerations. The lower curve in the figure is corrected for the activity of the 40.2 hour half life daughter, $La^{140}$.

His measurements were made in a vacuum tube similar to the one shown in Fig. 2, but without a window. After a number of the nickel receivers had received, separately, different amounts of the evaporation product they were removed from the tube for the radioactivity determinations.

The new experiments which are being reported here were done in the tube shown in Fig. 2. This design allowed making the radioactivity measurements through a thin bubble window .005" thick in the side of the tube.

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* Waste solids were buried under a minimum of six feet of earth in a remote area.

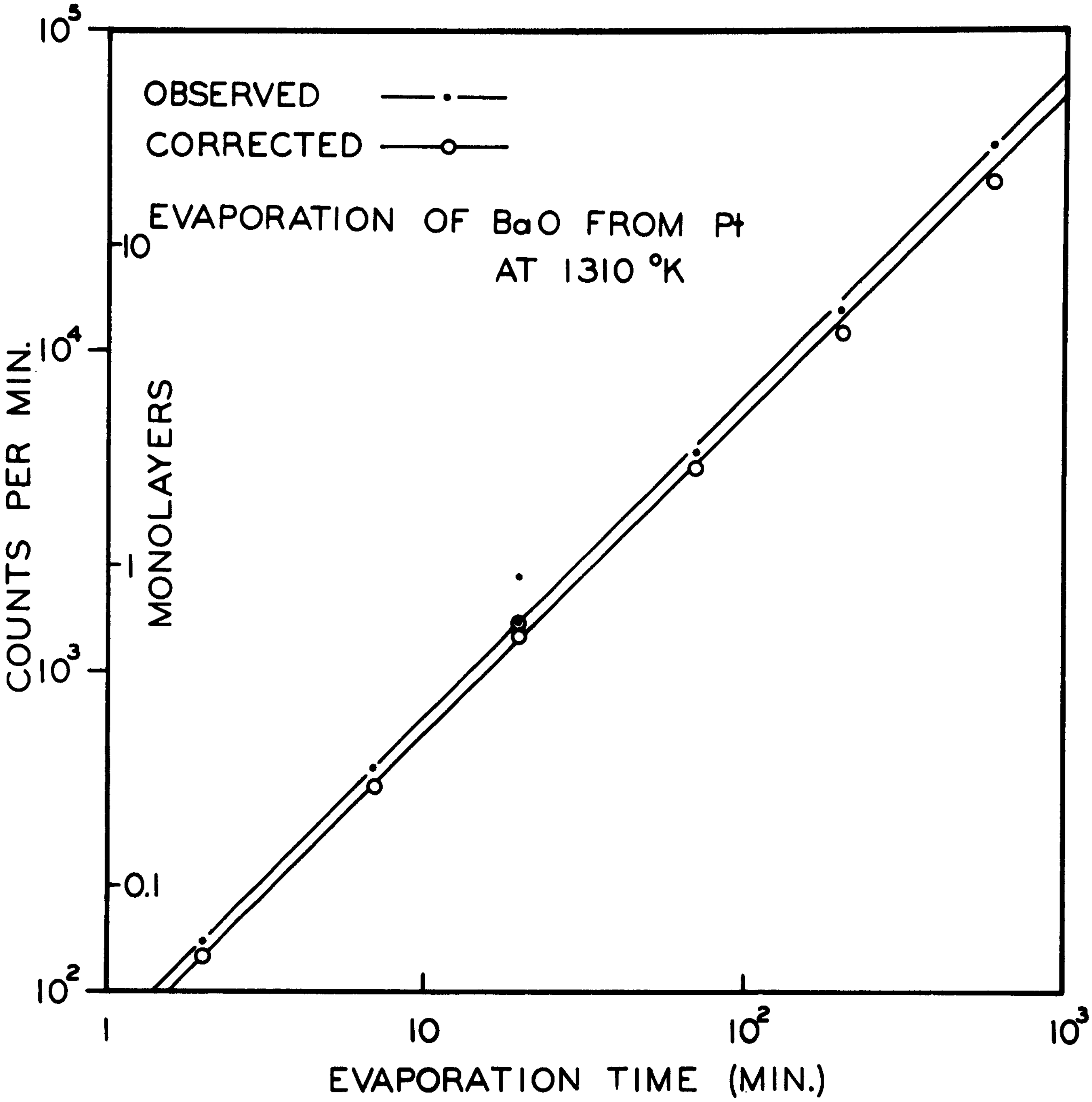

**Fig. 1** Counting rate of evaporated $Ba^{140}O$ as a function of evaporation time. Lower curve corrected for activity of daughter, $La^{140}$.

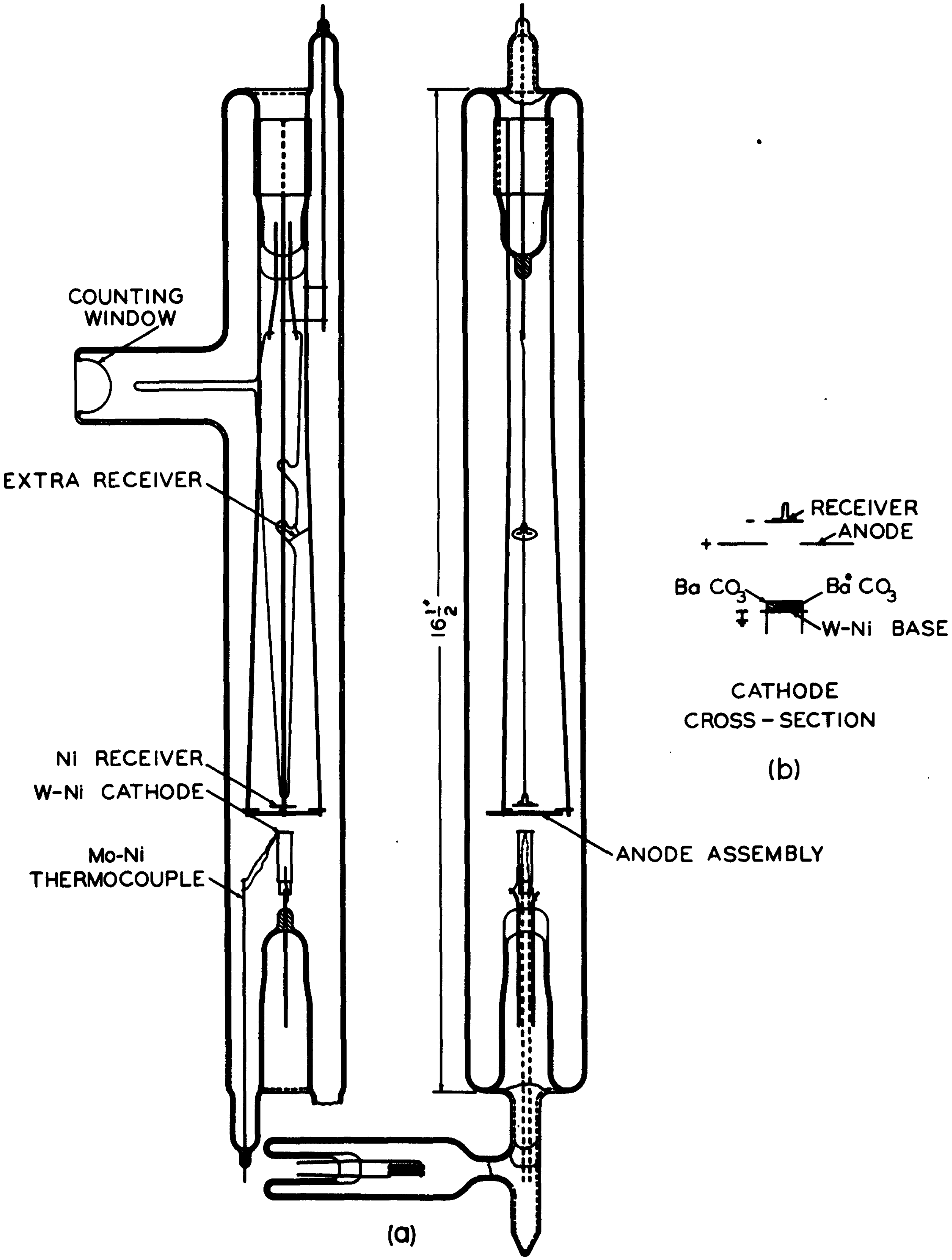

Fig. 2 **Evaporation tube for measuring diffusion rate of $Ba^{140}$ from metal-oxide interface. Window permits radioactivity measurement on evaporation receivers.**

This eliminated the necessity of opening the tube to measure the activity of the receivers. This type of tube was also used to study the rate at which free barium, which is produced at the cathode-base metal interface, diffuses to the cathode surface and is evaporated to the anode.

The first tube in this study had a 4.6% W-Ni base cathode to promote the production of free barium at the interface. A small amount of radioactive $BaCO_3$ was placed in the center of the cathode base over which was sprayed a coating of stable $BaCO_3$ (Fig. 2b). With the receiver mounted on the tracks, it was possible to move it about the tube to the side arm which had the bubble window. This window was 90% transparent to the 1 Mev $\beta$ - rays for activity measurements. The side arm and the counter were enclosed by a 100 pound lead "pig" during the measurements to minimize the background produced by the high activity of the evaporator. Two receivers were used, one to receive material deposited during conversion and the other during activation. The following processing cycle was used: The cathode was held at 1225°K under quiescent conditions for a given period of time (30 to 60 min.) and then emission current was drawn from the cathode for the same time interval. It should be noted that during depostion the receiver was biased at a negative potential with respect to the cathode to prevent possible stripping of the deposited material by electron bombardment. After each step in the cycle, the receiver was moved to the counting window and the activity was measured. During the cycling process the activation was carried out until the emission current density reached 72 ma/cm$^2$ with 250 volts applied between the anode and the cathode.

A second tube with a cathode of pure nickel also was processed and operated in the same manner. The maximum emission observed from this cathode was lower by a factor of 50 than that of the most active BaO cathodes prepared in this laboratory.

These studies of the traced barium, which was originally located next to the base metal and had diffused through the coating and evaporated to the receivers, gave the results shown in Fig. 3. The initial rate of transfer of barium in the case of the W-Ni cathode is about five times greater than in the case of the pure nickel cathode. As much barium was transferred from the pure nickel surface during conversion as during the first 45 hours of operation. In the W-Ni base cathode the same amount was transferred in 8 hours of operation as was released during the conversion.

Since it is difficult to make absolute thickness measurements of the cathode interface by x-ray diffraction techniques [3], a new method has been developed using Ba$^{140}$. The interface formed on a cathode will be radioactive if radio active BaCO$_3$ is used for the cathode coating.

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[3] J.H. Affleck, Quarterly Progress Report-December 15, 1951

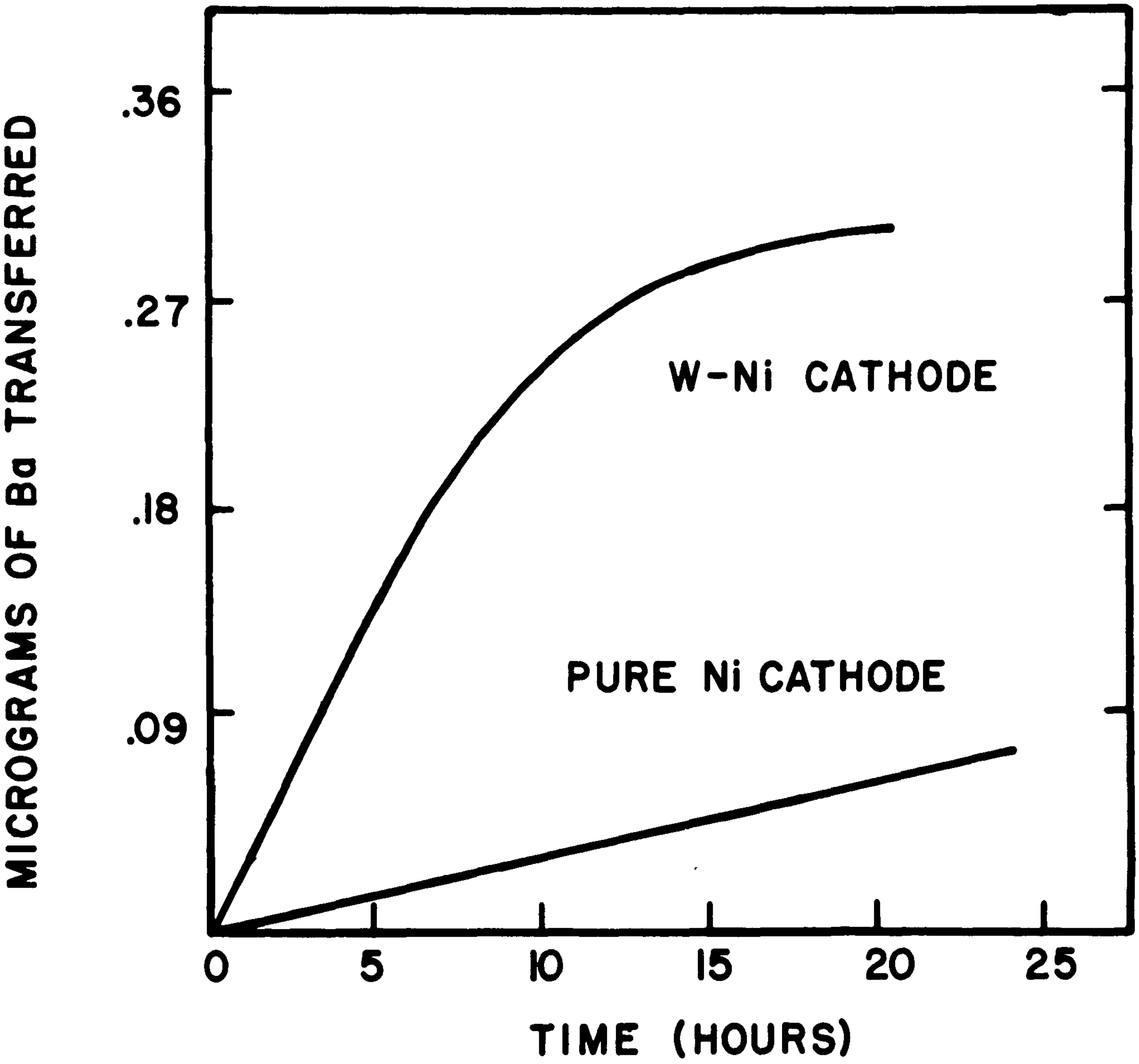

Fig. 3 Amount of $Ba^{140}$, originally at cathode base, transferred to the exaporation receiver after diffusion through oxide coating.

For example, on a cathode base containing silicon as the major impurity and having a radioactive coating of $BaCO_3$, radioactive $Ba_2SiO_4$ will be formed. If the oxide coating is then removed the activity of the interface can be compared with a given amount of radioactive $BaCO_3$ of known activity. From the area of the cathode and the density of $Ba_2SiO_4$ the thickness of the interface can be calculated. When a number of cathodes have been operated for various periods of time the rate of formation of the interface can be obtained.

In order to produce a radioactive coating of normal weight and density it was necessary to spray the coating onto the cathodes. Since the $BaCO_3$ contained appreciable $Ba^{140}$ (.25 mc/gm), special techniques were required. An inexpensive plywood chamber was constructed for spraying the radioactive carbonate. This chamber prevented contamination of the working area and minimized health hazards. The spray box, (see Fig. 4) was equipped with a glass fiber filter and was attached to an exhaust hood. A spray gun held in a lead shield was placed inside the chamber and all manipulations were carried out with the aid of rubber gloves. Extreme care was taken at all times to avoid overexposure of personnel to the radiation present.

Preliminary investigations have been made using this method. Nine cathodes were made on a nickel base metal containing 0.2%

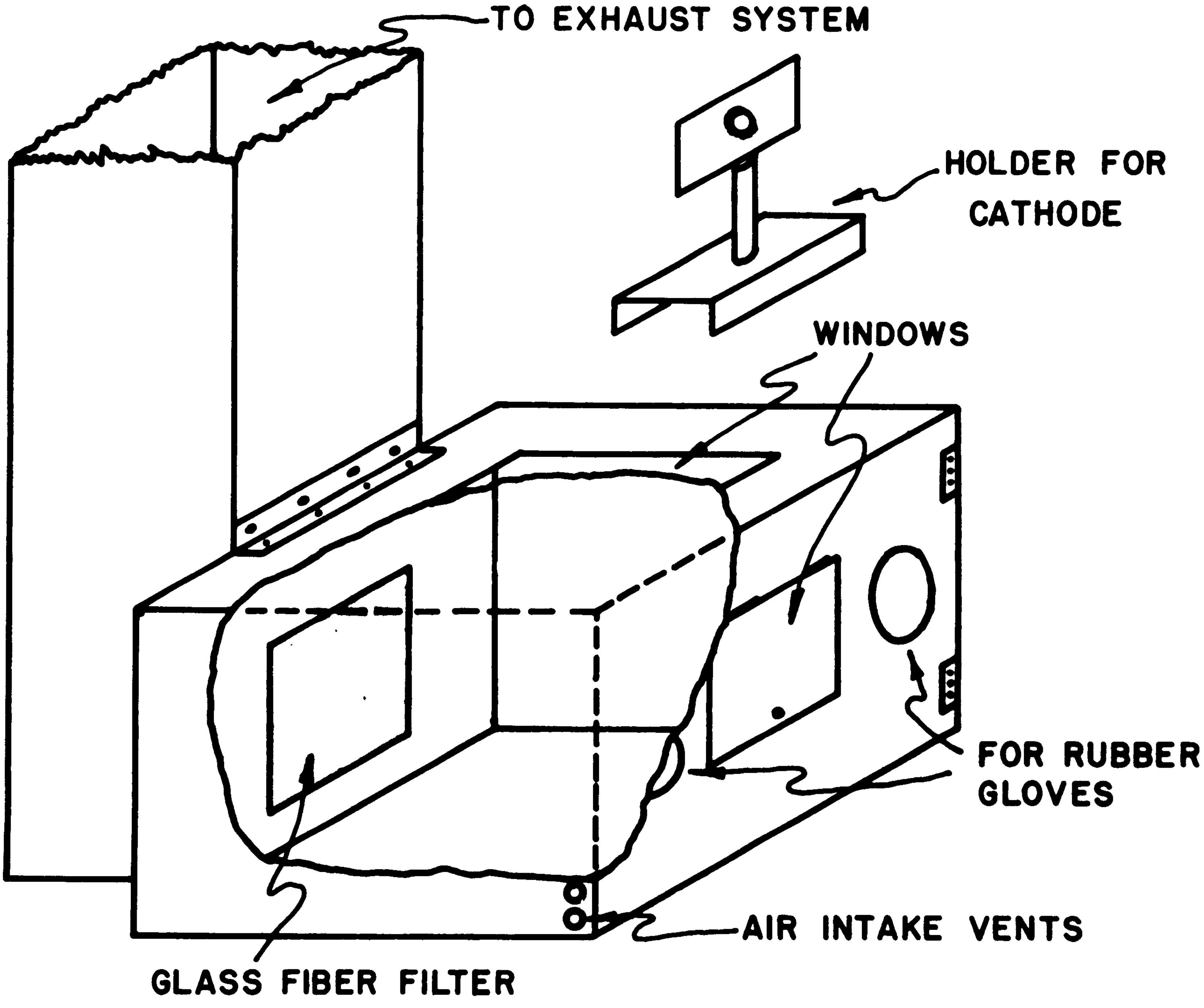

Fig. 4 Spray box for spraying radioactive oxide coatings.

silicon by spraying to a coating weight of 10 $mg/cm^2$ of radioactive $BaCO_3$. Of these nine cathodes eight were selected for life test at 850°C. In order to determine the accuracy of the method, three cathodes were operated for 0 hours, three for about 100 hours, and two for 580 hours after seal off.

After this operation, the tubes were cracked open and the coating removed by boiling in methyl alchol and gently brushing off bits of coating that remained. The activity of each cathode base was then measured and compared with a known amount of the original $BaCO_3$. The possibility remained that radioactive barium was diffusing into the base metal. As a check a cathode with a pure nickel base was prepared and tested. No activity was detected from barium which had diffused into the base metal. A small amount of diffusion might not have been detected because of the low specific activity of the oxide. Therefore, the activity present was due to the radioactive $Ba_2SiO_4$. This permitted the determination of the interface thickness. The results are shown in Fig. 5.

More experiments are needed using oxide coatings with better physical properties, but this appears to be a reliable method for determining interface thicknesses.

## APPENDIX I

Procedure for the preparation of radioactive $BaCO_3$ with high specific activity:

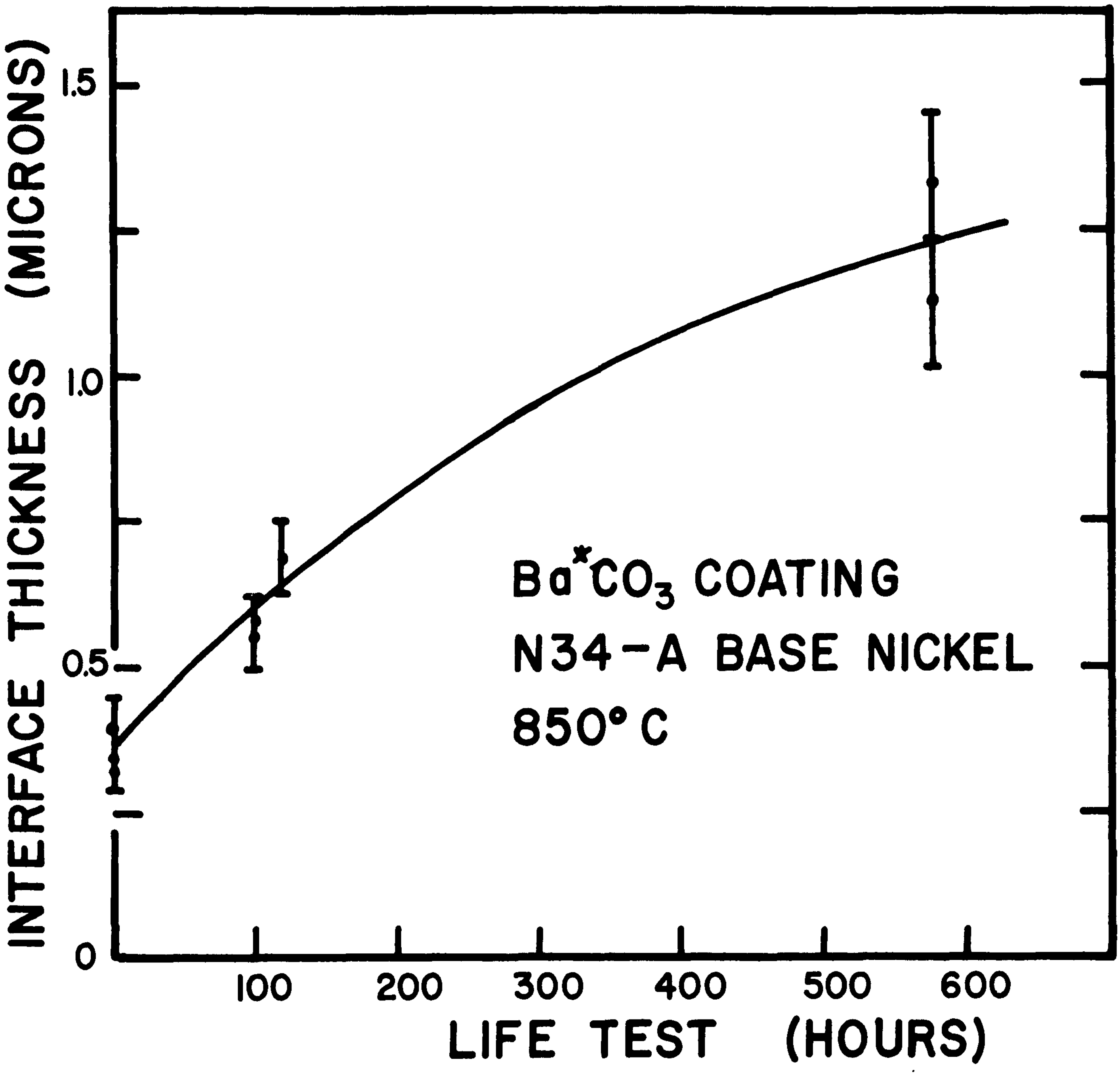

Fig. 5 Interface thickness vs. length of time of cathode operation at 850 °C.

1. Add to a 5 cc centrifuge tube a weighed quantity of purified $Ba(NO_3)_2$ such that not more than 3 cc of the solution containing the radioactive $Ba(NO_3)_2$ will give the desired specific activity. The upper limit of the amount of $BaCO_3$ which can be processed in a 5 cc centrifuge tube is about 0.5 gm.

2. Add the desired amount of radioactive solution by means of a calibrated pipette to the centrifuge tube containing the $Ba(NO_3)_2$*. The centrifuge tube should be clamped to a ring stand and all of these operations performed in an exhaust hood. A short stem micro-funnel should be set on the top of the tube to aid in the transfer of the solution from the pipette. After the solution has been transferred the funnel is washed with a little doubly distilled water.

3. Add doubly distilled water until the volume is about 4 cc. Heat the tube containing the solution in a beaker of boiling water until all the $Ba(NO_3)_2$ has dissolved. Add 2-3 drops of concentrated $HNO_3$.

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* If purified $Ba(NO_3)_2$ is not available for use as a carrier, pure $BaCO_3$ (e.g. Mallinckrodt's Ultra Pure) can be used. It will be necessary to add the radioactive solution cautiously since it contains $HNO_3$ and will cause the liberation of $CO_2$. The solution may contain enough $HNO_3$ to dissolve all the $BaCO_3$; if not, more should be added.

4. Put the centrifuge tube in the drying oven and evaporate at 80-90°C until crystallization begins. This will reduce the volume to about 2 cc.

5. Remove the tube containing crystals and allow to cool overnight. Remove the solution above the crystals by means of a glass tube, the end of which has been drawn to a diameter of 1-2 mm, fitted with an eye dropper squeeze bulb. This solution will be quite radicactive, so it should be transferred to a bottle and stored until disposed.

6. Dissolve the $Ba(NO_3)_2$ crystals in about 2 cc of doubly distilled water. It will be necessary to heat the centrifuge tube in a boiling water in order to dissolve the $Ba(NO_3)_2$.

7. While the solution is still hot, (60-70°C) add $(NH_4)_2CO_3$ by drops. The concentration of the $(NH_4)_2CO_3$ solution should be such that 2 cc will be more than sufficient to precipitate all of the barium.

8. Digest the solution by heating the centrifuge tube in a beaker of boiling water for 3-5 hours. Increasing the digestion time increases the particle size. It is necessary for good adhesion of the oxide coating that the particle size be large.

9. Cool the tube and centrifuge. Remove the supernatant liquor with a micro-pipette.

10. Wash the precipitate with doubly distilled water, i. e. add doubly distilled water to the centrifuge tube and stir throughly with a small stirring rod and centrifuge again. Wash twice more with doubly distilled water, three times with acetone and three times with amyl acetate.

11. After removing the last amyl acetate wash, add a few drops of binder.

## APPENDIX II

Preparation of radioactive $BaCO_3$ on a large scale with lower specific activity.

1. Dissolve the $Ba(NO_3)_2$ in doubly distilled water.
2 Add a measured volume of the radioactive solution to give the desired activity.
3. Heat the solution to 80-90°C.
4. Precipitate by adding a $(NH_4)_2CO_3$ solution at 60-70°C.
5. Digest at 80-100°C for 3-5 hours.
6. Filter. The filtration should be performed with care in order to avoid splashing. The precipitate cannot be washed in the usual manner because of contamination hazards. The doubly distilled water, acetone and amyl acetate rinses should be performed as well as possible while the precipitate is on the filter. If it is thought that this type of washing is

insufficient, the precipitate may be dried in the oven and then mixed with the binder. In this case glass beads will probably have to be rolled with the $BaCO_3$ and the binder in order to obtain a good suspension.

## APPENDIX III

The following general precautions should be observed in preparing and handling $Ba^{140}$.

1. Rubber gloves, respirator and laboratory coat should be worn at all times during the operations.
2. All operations should be performed under an exhaust hood.
3. A glass shield should be placed in front of the hood as a guard against splattering.
4. The bottle containing the $Ba(NO_3)_2$ solution should not be opened, nor should anything be added or taken from this bottle unless it is clamped to a ring stand.
5. For any transfer involving a pipette, a rubber syringe should be used to draw the liquid into the pipette.
6. All containers of radioactive material should be handled carefully and with the aid of tongs.
7. Radiological monitoring equipment should be available for use.