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ELECTRODEPOSITION OF PLUTONIUM FOR ALPHA PULSE HEIGHT ANALYSIS OF PLUTONIUM-236 IN PLUTONIUM-238

W. R. Amos and R. H. Lambek

AEC Research and Development REPORT

MONSANTO RESEARCH CORPORATION

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MOUND LABORATORY

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

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ELECTRODEPOSITION OF PLUTONIUM FOR ALPHA PULSE HEIGHT ANALYSIS OF PLUTONIUM-236 IN PLUTONIUM-238

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ABSTRACT

An apparatus was constructed and a method was developed for the electrodeposition of nanogram quantities of plutonium onto a stainless steel slide. The deposit was thin, uniform, and alpha-wipe free, and the plutonium was quantitatively transferred from the plating solution. Because of the characteristics of the deposit, accurate alpha pulse height analysis of small quantities of ^{236}Pu in ^{238}Pu was possible.

INTRODUCTION

During neutron irradiation of ^{237}Np for the production of ^{238}Pu, small amounts of ^{236}Pu, the decay product of ^{236}Np, are formed by (n, 2n) or (γ, n) reactions in the ^{237}Np. Because ^{208}Th, which emits a 2.6 MeV gamma ray, is a member of the ^{236}Pu decay chain, the quantitative determination of the ^{236}Pu content of the ^{238}Pu is of interest, especially in biomedical programs such as the Cardiac Pacemaker. Therefore, a method was developed to determine small quantities of ^{236}Pu (0.1-3 ppm) in ^{238}Pu. In samples that typically contain approximately 80% of the plutonium as the 238 isotope, this determination can be accomplished by alpha pulse height analysis in which the ratio of the concentration of ^{236}Pu to ^{238}Pu is measured.[1,2] With the plutonium electrodeposited on a thin metal slide, better resolution of the ^{236}Pu and ^{238}Pu alpha energy levels are possible, and more accurate data are obtained than with slides prepared by other techniques. This report describes the electroplating apparatus and the method employed for the plutonium deposition.

EXPERIMENTAL

Apparatus The components of the electrodeposition apparatus, which is similar in design to that of Donnan and Dukes,[3] are shown in Figure 1, and the assembled apparatus is shown in Figure 2. A 0.64 cm i.d. by 7.62 cm long Pyrex cylinder (A) contains the plutonium solution. The bottom of the cylinder is set on a 0.64 cm i.d. vinyl O-ring (B) which is placed on a 2.54 cm square stainless steel slide, 0.47 mm thick (C). The seal for containment of the solution is attained by four rubber bands between glass hooks on the cylinder and metal hooks (D) on the stainless steel base plate (E). The combination stirrer-anode (F) is a strip of heavy gage platinum 10.16 cm long by 1.27 cm wide, which is modified on the bottom in a paddle-type design. As shown in Figure 2, the top of the platinum strip is connected to a 0.64 cm diameter stainless steel rod (G) which fits into the shaft of a stirrer (H), operated at approximately 100 rpm. The power supply (I) (Model LH 125 FM, Lambda Electronics Corporation, Huntington, L. I., N. Y.) is operated at 5 V with variable current controlled by a Leeds and Northrup decade resistance box (J). The current is maintained at 2 A. The platinum stirrer with electrical brush contact (K) serves as the anode, and the stainless steel slide serves as the cathode.

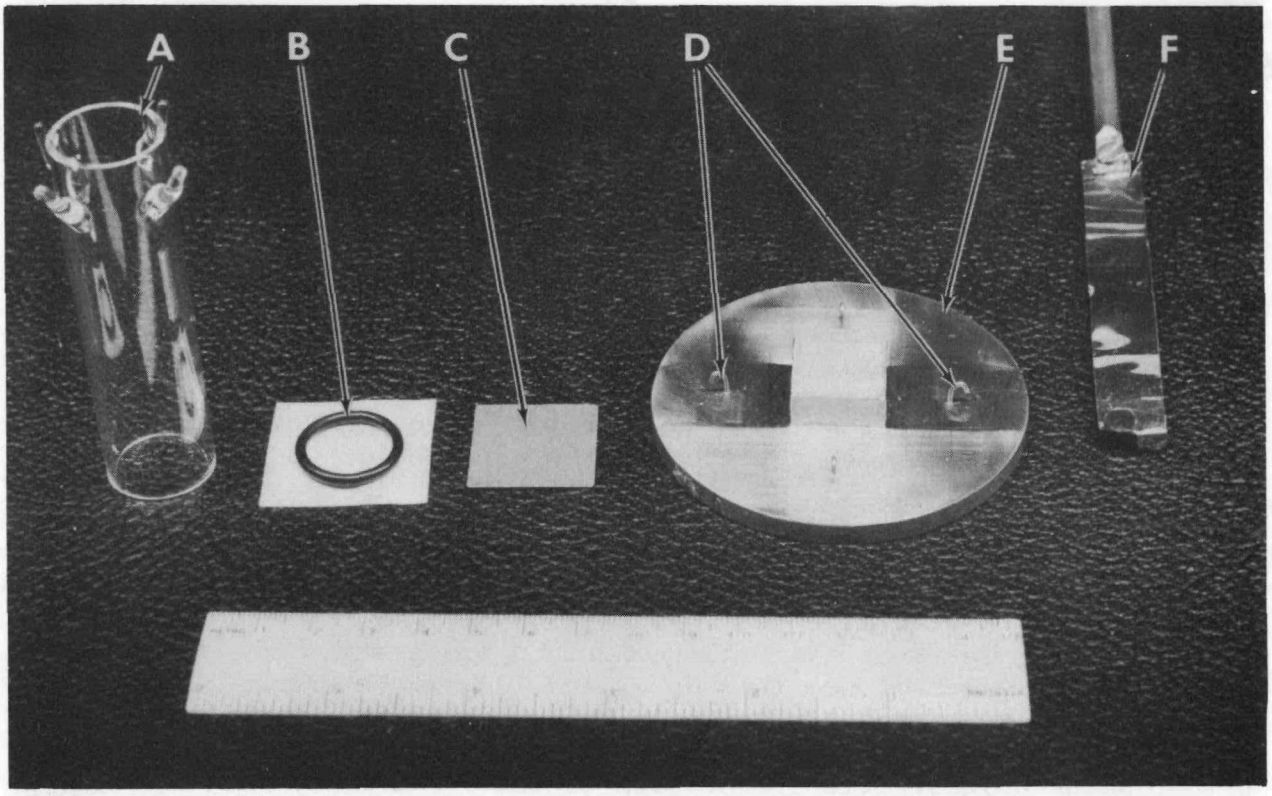

FIGURE 1 - Dissassembled electroplating cell.

Procedure If the sample is plutonium metal, a weighed quantity (<0.5 g) is dissolved in concentrated hydrochloric acid and the solution evaporated several times after addition of concentrated nitric acid to remove chloride ion. If the sample is plutonium dioxide, a weighed quantity (<0.5 g) is dissolved in a Teflon or platinum beaker with concentrated nitric acid and several drops of hydrofluoric acid. After dissolution of the dioxide the solution is evaporated to remove fluoride ion. From the sample weight and the isotopic ratio of plutonium, calculations can be made to permit dilutions with 0.35 N nitric acid so that 10 microliters of the plutonium solution will have approximately 10^5 alpha dis/min (approximately 3 nanograms of plutonium). To the electroplating cell, the following are added: a) 4 ml saturated ammonium nitrate solution which has been adjusted to a pH of 1-2 with ammonium hydroxide and hydrochloric acid, b) 0.5 ml concentrated formic acid, c) 10 microliters of ^{238}U containing 10 μg of uranium in nitric acid and d) 10 microliters of the plutonium nitrate solution.

The stirrer is positioned approximately 5 mm above the stainless steel slide and the solution is stirred. The power supply is then connected and the current is adjusted to 2 A. After 5 min, an additional 10 microliters of uranium solution is added and the electroplating is continued for 10 min. Then the stirrer and power supply are turned off, and the stirrer and walls of the cylinder are washed with 4 ml of the saturated

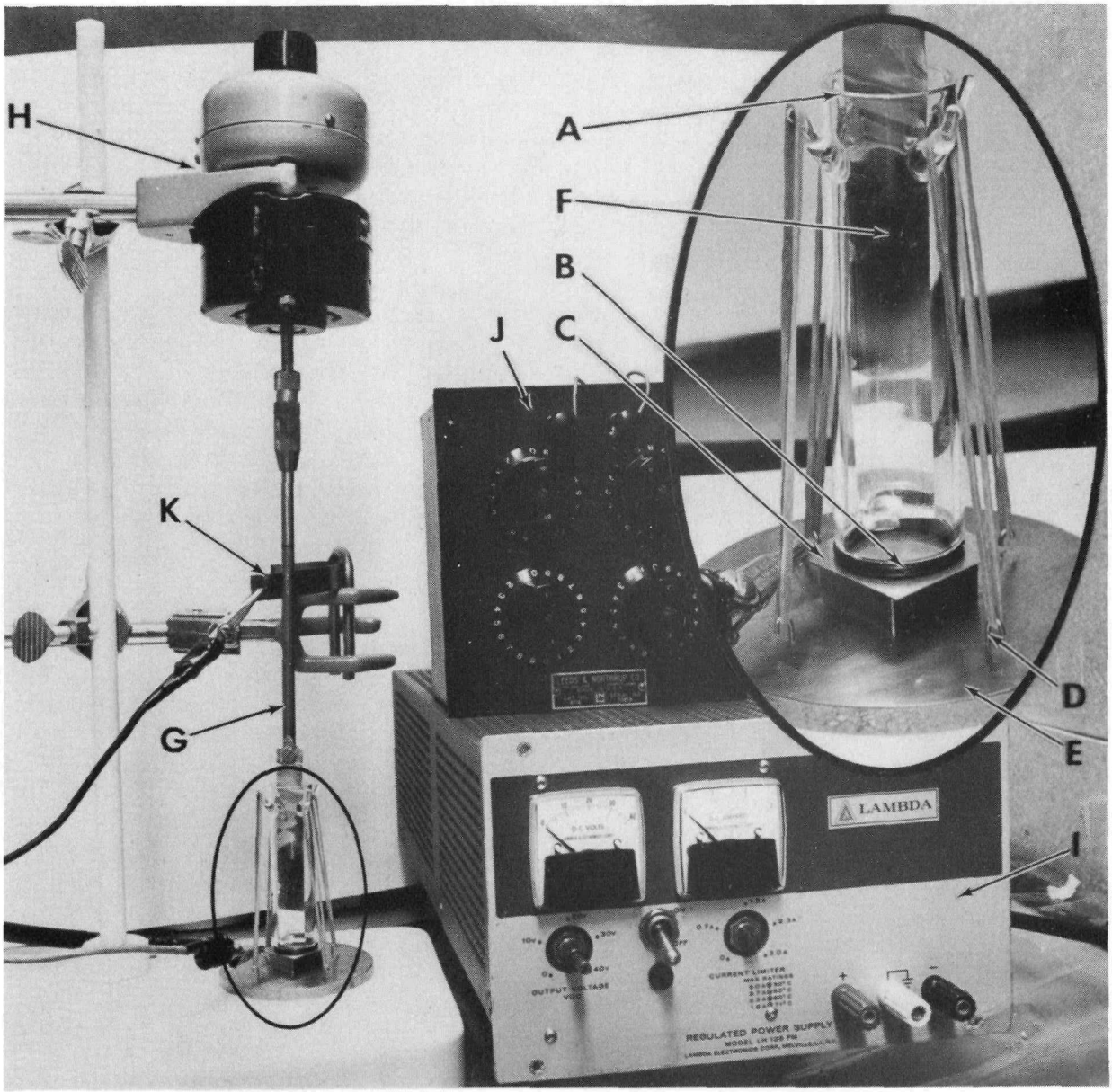

FIGURE 2 - Electroplating equipment.

ammonium nitrate solution. Another 10 microliters of the uranium solution and 0.5 ml of formic acid are added, and the electroplating is continued for an additional 10 min. The slide is removed, washed with water and ethanol, and dried on a hot plate.

RESULTS AND DISCUSSION

The quantity of ^{238}Pu before and after electroplating was compared to determine whether quantitative deposition of the plutonium was

attained. Plutonium-238 analyses were performed by conventional alpha counting techniques with an NMC-2 π proportional alpha counter. In one sample 99.4% of the plutonium was deposited; in five other samples there was quantitative deposition of the plutonium. The slides prepared by the electrodeposition technique had a thin, uniform deposit with a surface that was alpha-wipe free. Because of these characteristics, there was little alpha self-absorption, a problem usually encountered with other slide mounting techniques. Sufficient resolution was obtained between the ^{236}Pu and ^{238}Pu peaks to enable accurate alpha pulse height analysis data in the concentration range of 0.1-3 ppm ^{236}Pu.

Other mounting techniques were unsuccessful in attaining the proper sample for accurate alpha pulse height analysis.[2] Non-electroplating techniques resulted in the sample being confined to such a small area that relatively thick deposits caused pulse pile-up or self-absorption. Another electroplating technique (platinum gauze anode and ammonium chloride as the electrolyte) did not plate a uniform area on the slide.[3] Also, the hydrogen evolution at the anode caused spattering on the container walls which prevented quantitative deposition of the plutonium. The best results were obtained with saturated ammonium nitrate as the electrolyte and a flat paddle-type platinum anode-stirrer, which produced a uniform deposit over the plating area.

The presence of formic acid causes an increase in pH during electroplating, since oxidation of formate at the anode results in fewer hydrogen ions being formed than are lost at the cathode.[4,5] This increase in pH enables quantitative precipitation of plutonium hydroxide at the cathode. Uranium-238, deposited as the hydrated oxide, serves as a carrier for the plutonium and increases the plutonium recovery values.[3]

Other actinides or metals that form insoluble hydroxides would be codeposited with the plutonium.[4] If these actinides had alpha energies in the same range as ^{236}Pu and/or ^{238}Pu and were present in sufficient quantities, the ^{236}Pu results would be in error. Either chemical separation prior to electrodeposition would be necessary or the quantity of interfering element would have to be determined.

In the material of interest, ^{241}Am (5.477 and 5.435 MeV) interferes with the ^{238}Pu alpha energy (5.495 and 5.452 MeV), necessitating gamma pulse height analysis of ^{241}Am and correction of the ^{238}Pu content for the ^{241}Am contribution. This is performed on a conventionally mounted slide from each sample, even though the americium concentration ($<0.05\%$) is usually not sufficient to cause a significant error.

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