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ELECTROLYTIC SEPARATION OF

bv Richard K. Money Melvin G. Bowman

POLONIUM FROM LEAD AND BISMUTH VERIFIED UNCLASSIFIED 6-18-79 Per Bv







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### ABSTRACT

A final step commonly used in the preparation of pure metallic polonium consists of the electrodeposition of the metal onto platinum from 1.5  $\underline{N}$  nitric acid at a cathode potential of +0.3 volt relative to a saturated calomel electrode.

To determine whether lead and bismuth co-deposit with polonium under these conditions, a plating was made from a solution containing radioactive isotopes of polonium, lead, and bismuth. Results indicate that the plating is effective in the separation of polonium from lead and bismuth since 87% of the polonium was plated from solution while less than 1% of either the lead or bismuth was co-deposited.



#### 1. Introduction

One method frequently used for the purification of polonium is the electrodeposition of the metal onto a platinum cathode.<sup>1,2</sup> In this laboratory, the plating of polonium from 1.5 N nitric acid at a cathode potential of +0.3 volt relative to the saturated calomel electrode has become rather routine for the purification of polonium as well as for the preparation of polonium metal for research and production use.<sup>3</sup>

At the time this procedure became semi-routine, there appeared to be a valid question as to whether the electroplating was really effective in the removal of bismuth and lead from the polonium. Of course, it was realized that critical cathode deposition potentials of lead and bismuth are more negative than the cathode potential used in the plating; even for concentrated lead or bismuth solutions. Also, the work of Hevesy and Paneth<sup>4</sup> and Haissinsky<sup>5</sup> had indicated that the deposition potential of bismuth onto gold or silver followed the Nernst equation over a concentration range from  $10^{-4}$  M to  $10^{-12}$  M. However, the possibility that lead or bismuth could co-deposit and alloy with polonium on a <u>polonium</u> cathode at more positive cathode potentials could not be excluded. In addition, the specific alpha activity of the plated polonium often appeared to be low. Therefore, since lead (from polonium decay) and bismuth (used in a process to purify polonium from other cations<sup>6</sup>) are the most probable impurities in the polonium plating solutions, an experiment was performed to determine the degree of separation of lead and bismuth under the conditions of the routine platings.

#### 2. Discussion of Radiochemical Procedure

A radiochemical procedure was used. Briefly, the procedure was to conduct an electroplating operation from a solution containing  $Po^{210}$ ,  $Pb^{210}$  (RaD),  $Bi^{210}$  (RaE), and inert lead and bismuth, followed by an analysis of the cathodic deposit for  $Po^{210}$ ,  $Pb^{210}$ ,  $Bi^{210}$  through a combination of alpha and beta counting.

 $Pb^{210}$  has a half-life of approximately 22 years and decays with the emission of a 0.025 Mev beta to Bi<sup>210</sup> (5.0 day half-life), which in turn decays with the emission of a 1.17 Mev beta to Po<sup>210</sup>. The decay of 138.4 day Po<sup>210</sup> to stable Pb<sup>206</sup> by alpha emission completes the chain. If a mixture of these isotopes is counted in a beta counter, any contribution to the count from the Pb<sup>210</sup> beta may be readily eliminated through the use of a relatively thin absorber between the sample and the counting tube. Under these conditions, the beta decay curve of the cathodic deposit would exhibit one of four characteristic forms, as follows:

a. If the cathodic deposit were polonium, uncontaminated with either lead or bismuth, the original beta count would be due only to the gammas from Po $^{210}$  and would decay with a

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half-life of 138.4 days. This  $Po^{210}$  background could be related to a definite quantity of polonium by alpha counting the sample.

b. If the cathodic deposit were a mixture of  $Po^{210}$  and  $Bi^{210}$ , the original beta count would be higher than the known polonium background (as determined by alpha counting) and this "excess" beta count would decay with a 5 day half-life.

c. If the cathodic deposit were a mixture of  $Po^{210}$  and  $Pb^{210}$ , the original beta count would result only from the  $Po^{210}$  gamma background. However, if the sample were counted over a period of time, the "excess" beta count (after subtracting the  $Po^{210}$  background) would exhibit the characteristic growth of a 5 day daughter from a long-lived parent.

d. If the cathodic deposit were a mixture of all three isotopes  $(Po^{210}, Pb^{210}, and Bi^{210})$  the original beta count, after subtracting the known  $Po^{210}$  background, would designate the total quantity of Bi<sup>210</sup> plated. Any "excess" beta count might exhibit either a decay or growth, depending on the relative quantities of Bi<sup>210</sup> and Pb<sup>210</sup> present in the deposit. How-ever, after a period of about 25 days, the entire "excess" beta count would designate the total quantity of Pb<sup>210</sup> plated.

### 3. Apparatus and Procedure

# 3.1 Alpha and Beta Counting

Counting samples (or foils) were prepared by evaporating a known volume of solution onto thin mica disks (about 1" diameter). Alpha counting was done in a low-geometry methane proportional counter. A conventional Geiger counter was used for the beta counting and (except for the absorption curve) all counting was made with an aluminum absorber ( $100 \text{ mg/cm}^2$ ) placed between the sample and the counter tube. This absorber not only prevented counting of the Pb<sup>210</sup> beta but also greatly reduced the high Po<sup>210</sup> background.

# 3.2 Plating Bath

The plating solution consisted of a 200 ml volume of 1.5 N nitric acid containing 0.5 curie (0.1 mg) of polonium, 0.2 mg each of inactive lead and bismuth ions, and  $1.81 \times 10^6$  beta counts per minute (for the counting conditions described in Section 3.1) of a RaD-RaE equilibrium mixture.

#### 3.3 Electroplating Procedure

The electroplating was conducted at a temperature of about  $25^{\circ}$ C for a period of 4 hours. During this time, the cathode potential was maintained constant at +0.3 volt with respect to a saturated calomel electrode by the Model 101 Plating Control described by Bowman and Krikorian.<sup>3</sup> Both electrodes were made from platinum foil.



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The platinum cathode was leached with hot concentrated nitric acid and the leachings diluted to a volume of 10 ml. At this time, it was noted that the platinum cathode was still discolored, so it was dissolved completely in aqua regia. This solution was also diluted to a volume of 10 ml. A 0.01 ml aliquot was taken from each solution and evaporated onto a mica counting disk.

#### 4. Counting Data

The counting sample from the nitric acid leach contained 0.088 millicurie of polonium, as determined by alpha counting. The sample from the dissolved cathode contained 0.346 millicurie of polonium. Therefore, a total of 434 millicuries of polonium, or 87% of the quantity in the plating bath, was plated onto the cathode.

The sum of the beta counts of the two samples totaled 60 counts per minute, or 14 counts per minute over the expected background from the  $Po^{210}$  (106 c/m per millicurie). Apparently, therefore, the cathode contained  $1.4 \times 10^4$  "excess" beta counts per minute.

An absorption curve, using aluminum absorbers, was obtained in the beta counter for a sample of the plated material. Figure 1 shows this absorption curve superimposed on one obtained for a sample of pure polonium. Both curves were corrected to unit polonium activity.

The decay of the plated material was followed by counting a sample of the material in a beta counter (100 mg/cm<sup>2</sup> aluminum absorber) at intervals of 1 to 2 weeks over a period of 4 months. Within statistical counting errors, the sample decayed with the 138.4 day half-life of  $Po^{210}$ , with no evidence of either  $Bi^{210}$  decay or  $Bi^{210}$  growth from  $Pb^{210}$ .

#### 5. Discussion of Results

From a consideration of the decay data alone, one would conclude that measurable quantities of  $Pb^{210}$  or of  $Bi^{210}$  were not co-deposited with polonium during the electroplating. However, the counting rates were quite low and some decay of  $Bi^{210}$  or growth of  $Bi^{210}$  from  $Pb^{210}$  could have been masked by the relatively high statistical counting errors.

From the first beta counting, which indicated that an "excess" (over the polonium background) beta count of  $1.4 \times 10^4$  counts per minute was deposited on the cathode, and from the procedural discussion, one would conclude that about 0.8% of the Bi<sup>210</sup> originally present in the plating bath was cathodically deposited.

The aluminum absorption curves of Fig. 1 indicate the presence of some additional substance in the plated material, since for absorber thicknesses greater than 20  $mg/cm^2$  the counting rate (per unit of polonium) is appreciably higher for the plated material than for

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pure polonium. Of course, one cannot say that the excess count is due to  $Bi^{--}$ , since the absorption curves of Fig. 1 are also compatible with the possibility that the plated polonium contained a smaller proportion of some low energy beta or gamma emitter than was present in the supposedly pure polonium sample. Although the possibility of some undetected con-taminant in polonium cannot be excluded, it must be noted that the decay curve gives no more evidence for such a contaminant than is given for the presence of  $Bi^{210}$ . Therefore, it seems reasonable to attribute the <u>entire</u>  $Po^{210}$  background (in the low-energy region) to  $L\alpha$  and  $L\beta$  X rays\* (and possibly X rays derived from the M shell) from the  $Pb^{206}$  daughter of  $Po^{210}$ , and to accept the absorption curves as evidence that a small quantity of  $Bi^{210}$  was deposited on the cathode.

One may conclude with certainty that the electroplating procedure used in this laboratory is quite effective in the separation of polonium from lead and bismuth, since 87% of the polonium was deposited from the experimental plating solution and <u>less</u> than 1% of either lead or bismuth was co-deposited.

### 6. <u>References</u>

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\*These X rays have been described by Rubinson and Bernstein."

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