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The Behavior of Polonium Evaporated from Fused Salt

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Studies were made of the evaporation of polonium from a fused potassium chloride - lithium chloride mixture and of the deposition of the evaporated polonium on several metal plates under different temperatures. Approximately 100% of the polonium evaporates, probably as polonium tetrachloride, from the fused salt at the fused salt temperature higher than 500°C when heated for 3 hr. in an argon atmosphere. The evaporated polonium deposited on copper, platinum, palladium and silver plates which were held over the fused salts. The relationship between the percentage of polonium evaporated from these metal plates and the temperature shows that the polonium deposits on these metal plates as elementary polonium or polonide.

Polonium-210 can easily be obtained by separating it from the neutron-irradiated bismuth, the reaction involved being $^{209}\text{Bi}\ (\text{n}, \gamma)^{210}\text{Bi} \xrightarrow{\beta^{-}\text{-decay}} {}^{210}\text{Po}$. The present authors previously studied the extraction of polonium from the irradiated bismuth, using a fused salt, and reported the distribution of polonium between the fused potassium chloride - lithium chloride (eutectic) mixture and the fused bismuth metal.1) It was found that the polonium in the fused salt was easily evaporated

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1) Y. Kamemoto and S. Yamagishi, *J. At. Energy Soc. Japan*, **6**, 500 (1964).

and was removed more rapidly from the fused bismuth metal in contact with fused salt than from that not in contact with fused salt. If the polonium evaporated from the fused salt - fused bismuth system could be collected, this system would be a useful method of separating polonium from the irradiated bismuth.

The evaporation of polonium from the fused potassium chloride - lithium chloride mixture and the deposition of the evaporated polonium on several metal plates were examined under different temperatures, using polonium-210 as the tracer.

Experimental

Reagents.—*Potassium Chloride - Lithium Chloride Eutectic Mixture.*[2]—Potassium chloride and lithium chloride were separately dried by slow heating at 120°C, cooled in a vacuum desiccator, and mixed in the eutectic ratio. The product was fused in an anhydrous hydrogen chloride atmosphere, and the hydrogen chloride was removed from the melt by passing through it purified argon gas. The melt was solidified by pouring it into carbon tetrachloride.

Polonium Tracer.—Polonium-210 was separated by the ion exchange method[3] from a solution of Ra-D, -E and -F extracted from a radon tube. This method involves the adsorption of Ra-D, -E and -F on the anion exchange resin, the elution of Ra-D with a 2 M hydrochloric acid solution, the elution of Ra-E with a 10 M hydrochloric acid solution, and the elution of Ra-F(Po) with a nitric acid solution. The final eluate was evaporated several times with hydrochloric acid, and the residue was dissolved in dilute hydrochloric acid.

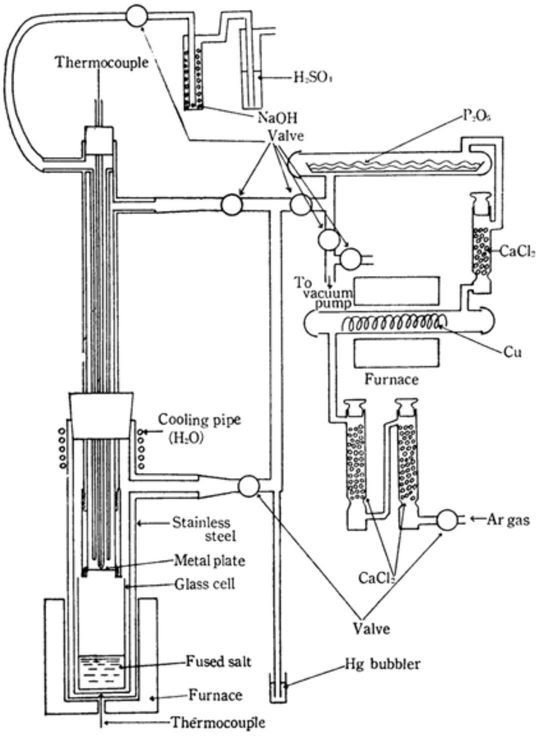

Fig. 1. Experimental apparatus.

Experimental Apparatus.—The apparatus is shown in Fig. 1. The evaporation vessel consists of two cells, an outer, stainless steel cell and an inner, glass cell. The metal plate (24 mm. in diameter) is held over the inner cell with a stainless stell holder. The temperatures of the plate and the fused salt are measured by thermocouples and kept constant by a temperature controller.

Experimental Procedure.—A tenth of a ml. of a polonium tracer solution was added to 10 g. of the potassium chloride - lithium chloride mixture in the inner cell, which was dried by slow heating under reduced pressure. After this inner cell had been placed in the outer cell, as is shown in Fig. 1, the air in the outer and the inner cell was replaced with argon gas, which had been purified by calcium chloride, phosphorus pentoxide, and copper net heated at 350°C. The fused salt, containing polonium-210, was then heated at a certain temperature for 3 hr. under a pressure slightly higher than 1 atm. After cooling, the polonium remaining in the salt was determined and the percentage of polonium evaporated was calculated.

The amount of polonium deposited on the metal plate was also determined, and the percentage of polonium deposited was calculated. The metal plates were disks (24 mm. in diameter) of copper, platinum, palladium and silver.

The metal plate on which polonium had deposited upon the evaporation was held at the middle point in the outer cell with a quartz holder and heated at certain temperatures for 1 hr. in an argon atmosphere; the amount of polonium evaporated was then measured.

Procedure for the Determination of the Polonium.—The amount of polonium deposited on the disk was determined by α-ray counting with a proportional 2π counter.

The polonium remaining in the salt was determined by dissolving the salt in dilute nitric acid, pouring a 10-ml. portion into the cell shown in Fig. 2, depositing spontaneously the polonium on a silver plate by stirring the solution for 1 hr., and counting the alpha activity.

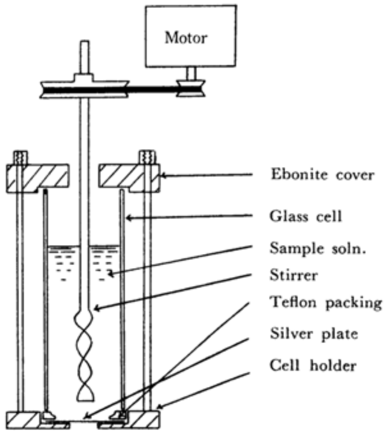

Fig. 2. Apparatus for spontaneous deposition of polonium.

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[2] S. Yamagishi and Y. Kamemoto, ibid., 5, 210 (1963).
[3] T. Ishimori, This Bulletin, 28, 432 (1955).

Results

The Evaporation of Polonium from the Fused Potassium Chloride - Lithium Chloride.—The relationship between the temperature of fused salt and the percentage of polonium evaporated is shown in Fig. 3. Approximately 100% of the polonium evaporated from the fused salt at a temperature higher than 500°C within 3 hr. in an argon atmospher.

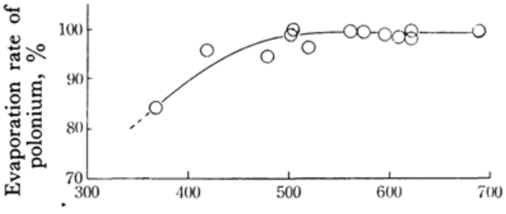

Fig. 3. Evaporation of ^{210}Po from fused salt in argon atmosphere.

The Deposition of Evaporated Polonium on Metal Plates.—The deposition of the polonium evaporated from the fused salt on copper, platinum, palladium and silver plates was determined at various temperatures. The results are shown in Figs. 4—7.

The deposition of polonium on the copper plate decreases with an increase in the temperature of the plate. However, polonium was not deposited on the platinum, palladium and silver plates at temperatures lower than about 290, 300 and 250°C respectively. The amounts of polonium deposited on platinum, palladium and silver plates are constant in the regions between 290—480, 300—400 and 250—425°C respectively, but decrease sharply at higher temperatures.

The Evaporation of the Polonium Deposited on the metal Plates.—The relationship between the amounts of polonium evaporated from metal plates and their temperatures is given in Fig. 8. The evaporation originates at the critical deposition temperatures of polonium (Figs. 4—7). This appears to indicate that the decrease in the percentage of polonium deposited is due to the partial evaporation of polonium from the metal plates.

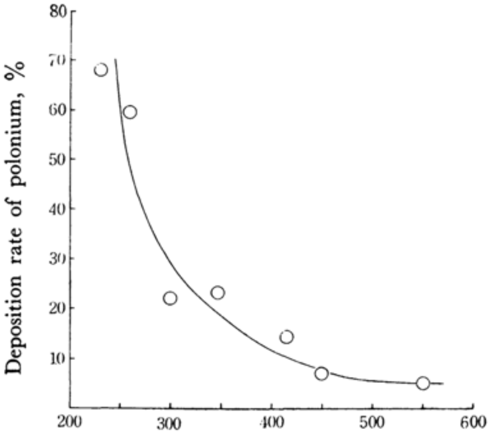

Fig. 4. Deposition of ^{210}Po on copper metal.

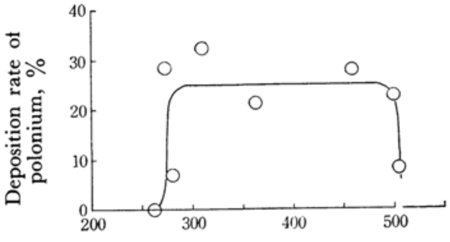

Fig. 5 Deposition of ^{210}Po on platinum metal.

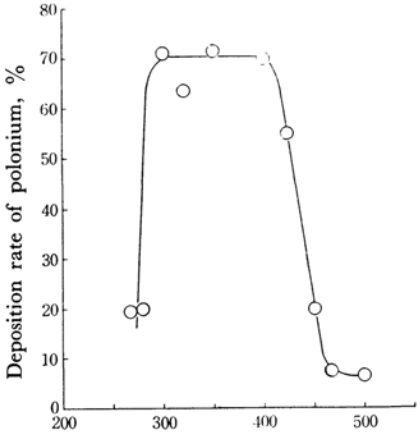

Fig. 6. Deposition of ^{210}Po on palladium metal.

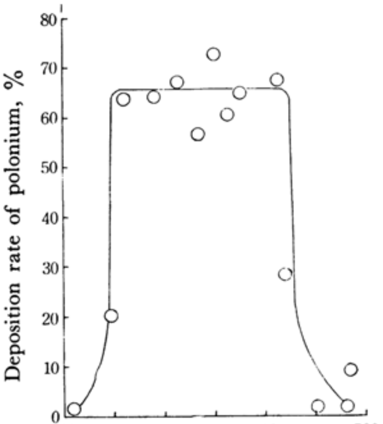

Fig. 7. Deposition of ^{210}Po on silver metal.

Discussion

In the polonium solution extracted from a radon tube, polonium probably exists as tetrachloride, because polonium tetrachloride[4-6] is produced by evaporating a solution obtained by dissolving metallic polonium in hydrochloric acid. Thus the fused potassium chloride - lithium chloride mixture, to which this tracer solution has been added, seems to contain polonium in the same form at a low temperature. From this fused salt, polonium is completely evaporated at 500°C in an argon atmosphere under our experimental conditions. The evaporated polonium can not be in the form of polonium dioxide or polonium metal, because neither compound seems to evaporate at this temperature (polonium dioxide sublimes at 885°C and the polonium metal boils at 962°C). The vaporized polonium may be polonium tetrachloride, as its boiling point is 390°C, near to the vaporization temperature experimented with.

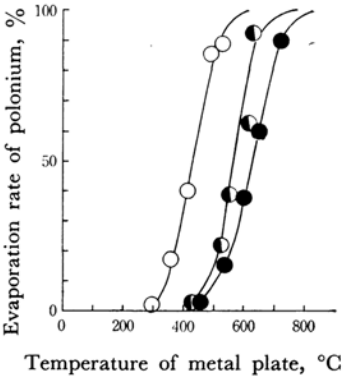

Fig. 8. Evaporation of ^{210}Po from metal plate in argon atmosphere.
○ Cu ● Ag ◐ Pt

Paneth and Hevesy[7-8] reported that almost all the polonium was condensed on a palladium or platinum foil when trace amounts of polonium were distilled in a quartz tube in the flow of a gas and the foil was placed in the cold region of the tube. However, in our experiment, the polonium evaporated from the fused salt was not deposited at a low temperature, but it was deposited at higher temperatures. These results suggest that the polonium evaporated from fused potassium chloride - lithium chloride is not metallic polonium but some polonium compound (probably polonium tetrachloride). This compound seems to be deposited on the metal plate by some reaction and not by adsorption.

Platinum polonide and silver polonide decompose at 450—600°C and approximately 559°C respectively.[7,9] These decomposition temperatures agree with the respective evaporation temperatures of the polonium deposited on platinum and silver metal by our technique using the fused salt. On the other hand, Abe et al. studied the evaporation of polonium-210, at several temperatures and in an argon atmosphere, from certain metal plates on which polonium had spontaneously deposited from a dilute nitric acid solution.[10] Their findings on the relationship between the percentage of polonium evaporated and the temperature of the metal plate are shown in Fig. 9. The evaporation curves of polonium from copper and silver plates, in Figs. 8 and 9, are similar to each other. This shows that the polonium evaporated from the fused salt deposits on the copper and the silver plates in the same form as that spontaneously deposited from a dilute nitric acid solution. It has been thought that the chemical form of the spontaneously-deposited polonium is elementary. From these observations, it seems that the polonium evaporated from the fused potassium chloride - lithium chloride mixture deposits on the metals as elementary polonium or polonide.

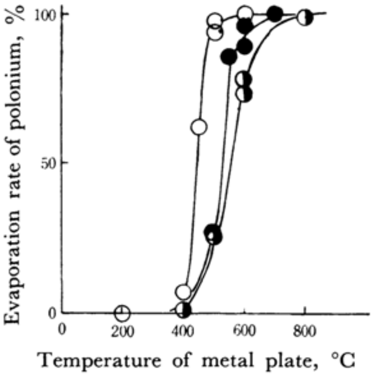

Fig. 9. Evaporation of ^{210}Po deposited spontaneously on metal plate.
○ Cu ● Ag ◑ Ni

On the basis of the arguments presented above, the following explanation can reasonably be made of our results. Polonium is evaporated from the fused potassium chloride - lithium chloride mixture as polonium tetrachloride and deposits on several metals as elementary polonium or polonide.

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4) K. W. Bagnall, R. W. H. D'Eye and J. H. Freeman, *J. Chem. Soc.*, **1955**, 2320.

5) J. J. Burbage, *Rec. Chem. Prog.*, **14**, 157 (1953).

6) E. F. Joy, *Amer. Rep.*, M-4123 (1947).

7) K. W. Bagnall, "Chemistry of the Rare Radioelement," Butterworths Scientific Publications, London (1957).

8) F. Paneth and G. Hevesy, *Sitzungsber. Akad. Wiss. Wien. Abt.*, **IIa**, **122**, 1049 (1913).

9) J. M. Goode, *Amer. Rep.*, HLM 677 (1952).

10) S. Abe, S. Yajima and M. Handa, *The 6th Symposium on Radiochemistry, Preprint*, 3B30 (1962).