Preparation of Infinitely Thin Plutonium Sources by Adsorption

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Infinitely thin sources have been prepared by adsorption of ^{239}Pu from hydrochloric acid solutions onto glass, aluminium and platinum surfaces. The validity of the method was studied by autoradiography. Results show that glass and aluminium are the best surfaces for such sources. The same method has been applied for the preparation of sources of ^{238}U on glass. Segregation of Pu and U has been observed on glass as well as on metallic surfaces.

Different methods exist for the preparation of α sources as described elsewhere¹. King has studied the adsorption of Pu(IV) at trace concentration on the internal surface of a glass pipet². He found that adsorption increases as the hydrogen-ion concentration is decreased. The purpose of the present work was to examine the possibility of using adsorption of Pu on glass and different metal surfaces as a method for the preparation of infinitely thin sources of α emitters as well as to determine the best conditions in which such preparation is to be carried out.

Haissinsky *et al.*³ have developed an autoradiographic method for the study of metallic surfaces by examining the distribution of ranges of α -particles of Po deposited spontaneously or electrolytically on these surfaces. They found that in all cases there is an apparent shortening of a certain number of ranges in the gelatine. This was explained by the penetration of a certain number of Po-atoms inside the metallic surface. This apparent shortening would be a disadvantage for the preparation of sources of α -emitters and may lead to wrong interpretations of the α -spectrum³.

In the present work we have applied the same method for the study of the adsorbed activity.

EXPERIMENTAL

The plutonium used in our experiments was separated from irradiated uranium by ion exchange technique⁴. Pu(IV) was prepared by dissolving a Pu-residue in 9 M hydrochloric acid containing 1 drop conc. nitric acid per ml⁵.

^{233}U was prepared by extraction from neutron irradiated ^{232}Th . The reagents used were of analytical grade and the water was bidistilled. The adsorption of the activity was performed by simple rotation of the glass or metal slide in the solution. The glass slides, before adsorption, were washed in *aqua regia*, sodium carbonate and finally in water. Two types of glass were used: ordinary soda-objectiveglass (0.15 mm thick) and pyrex-glass.

The aluminium plates were either rolled or electropolished and 0.1 mm thick. Before use they were washed with acetone and water.

The platinum (0.2 mm thick) was electropolished and before adsorption the slide was washed with boiling conc. nitric acid and finally in water. The different slides were, after adsorption, washed twice in water and dried by rotating in air.

The surface of the slide was then, for a certain time, brought into good contact with an α -nuclear plate, Ilford E. 1, 50 μ thick. The aluminium plates had to be kept in a desiccator with P_2O_5 during exposure to depress the chemical attack on aluminium by the photographic emulsion. The plates were developed by a conventional photographic technique⁴. Rubbing of the plates was avoided in order not to reduce the lengths of the α tracks.

The lengths of the α -tracks were measured with a microscope (type Laborlux III, Ernst Leitz). The magnification was 570: and 400 horizontal tracks for each plate were investigated (those which had an angle of less than 10° with the plane of the emulsion)³. In all cases corrections for the background of the slides used were applied.

RESULTS

In Table 1 are given the conditions under which the experiments were performed. The experiment with the pyrex glass gave results similar to those obtained with soda-objective glass, and is not therefore indicated here.

Table 1. Experimental conditions.

No.	Type of Slide	Acidity	Conc. in $\mu\text{g/ml}$	Ox. state added	Adsorption time in h.	Exposure time in h.
1.	Na-glass	0.01 M HCl	0.10	Pu(IV)	22	19
2.	Al-rolled	0.01 M HCl	0.10	Pu(IV)	4	42
3.	Electro-polished Al	0.01 M HCl	0.05	Pu(IV)	4	43
4.	Electro-polished Pt	3 M HCl	1.6	Pu(III) Pu(IV)	22	48
5.	"	0.01 M HCl	0.10	Pu(IV)	22	42
6.	Na-glass	0.1 M HNO_3	4.0	U(VI)	20	46

In Figs. 1—5 are plotted the histograms for ^{239}Pu of the number of α -tracks against their length, corresponding respectively to the experimental conditions indicated 1—5 in Table 1. Experiments were repeated several times, specially on glass (acidities from 0.01 to 3 M) and results were highly reproducible.

In Fig. 6 is given the histogram of the α -tracks of ^{233}U adsorbed onto glass (conditions 6 in Table 1).

Other experiments on Pu have shown that the shape of the histograms is independent of the lapse of time between the end of adsorption and the beginning of exposure, at least for 12 days. Again changing of acidity from 3 M hydrochloric acid (ionic species of Pu) to 0.01 M hydrochloric acid (colloidal Pu) similarly had no influence on the form of the range distribution curve.

Figs. 1-6. Histograms of number of tracks vs. length.

Fig. 7. Segregate on aluminium.

Fig. 8. Segregate on glass.

Fig. 9. Random distribution of tracks.

In all the plates examined segregates of ^{239}Pu or ^{233}U were observed on certain points. In Figs. 7 and 8 are given photographs of segregates of ^{239}Pu on aluminium and on glass, respectively, and in Fig. 9 a photograph from a region with random distribution of α -tracks.

DISCUSSION

The experiments show that the most probable range of the α -particles of ^{239}Pu (α : 5.10, 5.15, 5.17 MeV) in the Ilford E. 1 plate is $21 \pm 1 \mu$ which is in good agreement with the α -energy-range curve⁷. For that of ^{233}U (α : 4.80 MeV) we found $18.5 \pm 1 \mu$.

Apparent shortening has been observed in the case of platinum (14 tracks per 100 have ranges shorter than 18μ). This phenomenon is far less pronounced on aluminium and glass (only 3 tracks per 100 have ranges shorter than 18μ).

Apparent shortening may be due to:

- the penetration of plutonium atoms into the surface, phenomenon comparable to that found previously for polonium³.
- possible backscattering of the α -particles from the sample plate.

The backscattering of ^{239}Pu α -particles from platinum has been studied by Cunningham *et al*⁸. They found that identical quantities of essentially weight-

less samples of ^{239}Pu show a 3 % lower counting rate when mounted on quartz in a 50 % geometry counter than similar samples mounted on platinum and concluded that this difference is possibly due to small angle scattering in the latter case. This backscattering would be more manifest for the horizontal or nearly horizontal tracks on which we concentrated our studies. Backscattering would lead to a loss of a part of the energy of the scattered particles and hence to shorter ranges in the gelatin. In fact our experiments show that the histograms for electropolished and rolled aluminium are similar, in spite of the relatively rougher surface of the latter. This could be understood if we assume that backscattering plays an appreciable role for apparent shortening. Due to the low atomic number of aluminium, backscattering from it should be far less pronounced than from platinum. Therefore the absence practically of apparent shortening on glass and aluminium may be explained by the highly plane surface of glass objective slides and negligible backscattering from both materials. The presence of a certain number of shorter ranges for ^{233}U on glass is probably due to a trace of thorium impurity.

The study of the adsorbed phase represented certain peculiarities. In all conditions using ^{239}Pu or ^{233}U on glass or metallic surfaces, and irrespective of the acidity, part of the adsorbed atoms was distributed at random while the rest was in the form of segregates. The measurements of the length of the tracks originating from the segregates were often very difficult, due to the high activity in these regions. No conclusions regarding shortening may then be drawn from these cases. The numbers of Pu atoms accumulated in the segregate may be calculated from the number of tracks emanating from it. Assuming 2 π -geometry the number of ^{239}Pu atoms, N , corresponding to n tracks in the emulsion after d days exposure is:

$$N = \frac{24\,300 \cdot 2 \cdot n \cdot 365}{0.69 \cdot d} = (2.57 \, n/d) \times 10^7 \text{ atoms} \quad (1)$$

Using this formula we have found segregates with 10^8 up to $\gg 10^{10}$ atoms. The number of atoms in the biggest segregates, as for example that of Fig. 7, was impossible to calculate due to the high density of tracks in their regions.

The maximum activity in 2 α -geometry of the adsorbed ^{239}Pu was 135 c/m/cm² which corresponds to 5×10^{12} atoms/cm². Assuming a homogeneous surface and taking the value of 1 Å for the radius of the Pu atom, the maximum covering of the surface in our experiments did not exceed 0.2 % of a "mono-atomic" layer. Under such conditions the distribution of the energy of adsorption on the surface should play an important role in the distribution of the adsorbed atoms. The segregation phenomenon may be attributed to the presence of particularly privileged microscopic regions of the surface possessing a high energy of adsorption. Further study of this phenomenon is necessary to elucidate clearly its nature.

CONCLUSION

The experiments show that the adsorption method can be used for the preparation of extremely thin sources of plutonium. Glass and aluminium offer particularly good conditions. A relatively high pH would be preferable owing

to the higher adsorbability, probably due to the formation of radiocolloids. The adsorption method presents the advantages of simplicity and the possibility of having sources without self-absorption. The limit of the method is that a relatively high specific activity of the α -emitter is to be accounted for, but the method could be applied with good results even for ^{233}U ($T_{1/2} = 1.6 \times 10^5 \text{y}$).

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