ELECTROLYTIC PURIFICATION OF CARRIER FREE TELLURIUM TRACER

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Carrier free tellurium tracer is generally prepared from fission products in accordance with the following procedure[1, 2]: preliminary purification in presence of a copper salt used as a carrier; co-precipitation of the tellurium activity with iron hydroxide in presence of dilute ammonia, which keeps the copper in solution; and final extraction of iron with isopropyl ether in 9 M hydrochloric acid. The preparation thus obtained is very often contaminated with traces of iron which is difficult to eliminate by the solvent extraction as above, and which if present even in very minute traces, is not desirable for certain investigations. A good yield of tellurium activity is also difficult to be realised by the extraction operation in presence of strongly acidic solutions. Apart from these difficulties, the purification step based on co-precipitation of tellurium activity with iron hydroxide should be avoided as far as possible since the latter can carry with it the activities due to other impurities,[3] which might escape the preliminary step. These are briefly the considerations which led us to search for a modified method for the purification of tellurium tracer obtained from fission products, a method in which the addition of iron carrier would be avoided. Applications of paper chromatography to the above problem, as well as to the problem of separation of Te^{IV} and Te^{VI} have already been described.[4] The conditions of a suitable electrolytic method of purification could also be arrived at as a result of our electrochemical studies on tellurium tracer in the concentration range of 10^{-3} to 10^{-11} N.[5] In this article the details of this electrolytic method are described.

It is known since long that Te^{IV} is reduced to the metallic state by electrolysis of acidic solutions, whereas Te^{VI} is not.[6, 7] That radio-tellurium behaves similarly at tracer concentration has been verified.[5] Moreover, hydrochloric or hydrobromic acid solutions, preferably normal hydrobromic acid,

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have been found convenient as supporting media for the electrolytic deposition with a small cathodic current density. A smooth well adhering macro-deposit of tellurium metal can be obtained in these conditions by electrolysis of 10^{-3} N solution of Te^{IV}.[5]

Since copper is used as a carrier in the preliminary separation of tellurium tracer from fission products, our objective was to develop an ultra-micro method of purification which would work in presence of milligram quantities of copper. In this connection a reference to Norwitz's[8] work on the macro-estimation of tellurium in a mixture containing copper may be relevant. According to this author copper telluride can be electrolytically deposited from nitric acid solutions of the elements, the tellurium in solution being in tetravalent state. Cu can be separated from Te by electrolysis after having oxidised the tellurium to the hexavalent state. By difference the amount of Te can be estimated.

EXPERIMENTAL

Materials.—For this work, a source containing initially 10 millicuries of ^{132}Te (β emitter) was used. Since this was obtained from fission products, it also contained a few hundred microcuries of ^{129}Te and ^{127}Te (β emitters), the amount of ^{129}Te being about ten times that of ^{127}Te. Now, ^{132}Te (half-life: 77·7 hours), being not always conveninet for use as tracer, because of its daughter ^{132}I (half-life: 2·4 hours), was eliminated by decay for about one month (*i.e.*, ten times its half-life). The radioactive characteristics of the product after decay, however, indicated that it was significantly contaminated with some activity other than those of ^{129}Te and ^{127}Te.

The usual counting techniques with a Geiger Muller counter were adopted for measurements involving radioactivity.

Procedure.—A few milligrams of a copper salt were mixed up with a portion of the above-mentioned active source. The method which we have established for the purification of tellurium tracer is obviously applicable in case when the solution does not contain weighable quantities of copper. The details of the method are given below:

The solution was evaporated to dryness. The residue was heated with perchloric acid, and the excess of the latter eliminated by distillation.† The

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† In the method of Jacobson and Overstreet described at reference 1, the solution just before treatment with iron carrier contained besides tellurium tracer, copper in perchloric acid medium.

Radio-ruthenium is a possible contaminant in fission products. Since Rd-Ru is volatile with boiling perchloric acid, distillation is recommended in order to avoid a possible contamination of the laboratory.

residue was taken in a 3% solution of HNO_3, and the solution thus obtained was electrolysed with a rotating cathode of platinum, the current density being 20 mA/sq.cm. In these conditions not only the copper but other impurities depositable with a realtively high current were eliminated, leaving the tellurium tracer in the solution. It was, however, noticed that with a cathode of about 2 sq. cm. surface, the copper deposit began to turn blackish in about a quarter of an hour. It was, therefore, preferred to interrupt the electrolysis every quarter of an hour, to clean the cathode with nitric acid (1 : 1), and to continue in this way the electrolysis for a total period of about two hours.

The solution was then evaporated to dryness in a tared beaker. The residue was taken with a few c.c. of concentrated hydrobromic acid and again evaporated to dryness on a water-bath. In this operation the tellurium tracer was reduced in accordance with the following reaction:

$$Te^{6+} + 2\,Br^- = Te^{4+} + Br_2{}^{\nearrow}.$$

So long as the temperature of heating did not exceed that of a water-bath, there was no fear of any loss of Te as the volatile bromo-complex. A few c.c. of concentrated hydrobromic acid were again taken in the beaker and concentrated to a small volume. The beaker and its contents were allowed to cool and weighed, whereby the quantity of the constant boiling hydrobromic acid remaining in the beaker was known. This was diluted with calculated amount of water so that the concentration of HBr was approximately normal. The solution thus prepared was electrolysed for a second time with a current density of 0·05 mA/cm.^{2}, and with a separated anodic compartment. Separation of the anode was necessary in order to avoid partial oxidation of Te^{IV} to Te^{VI}. In our experiments a very simple arrangement consisting of a glass tube drawn into a capillary and plugged at the capillary end with filter-paper was used as the anodic compartment. The plugged end dipped in the main electrolyte. The anolyte consisted of $\overline{N}H_2SO_4$ and the anode a small piece of plantinum wire.

In these experimental conditions, practically 90% of the tellurium tracer was found to be deposited in 2 to 3 hours on a rotating gold cathode. A gold cathode was preferred to a platinum one, since electro-deposited tellurium tracer can be removed by dissolution in hydrobromic acid from gold much more easily than from platinum.

Purity of the Deposit.—It was found that with the radioactive source at our disposal, only a very small fraction of activity was deposited along with the copper in course of the first electrolysis, whereas the major part of the

activity was shared between the electrode and the solution in course of the second electrolysis. The radioactive property of the two portions thus separated was then examined. It was found that the half life of decay for the substance deposited cathodically was 34 days, whereas that for the non-depositable substance was 65 days. Moreover, the absorption curves of the radiations were also traced as a function of thickness of aluminium absorbers for estimating the energy of radiations: the curve B of Fig. 1 corresponds to the depositable activity and the Curve A to the non-depositable one (*see* also Fig. 2 for the analysed component of the latter curve).‡ These
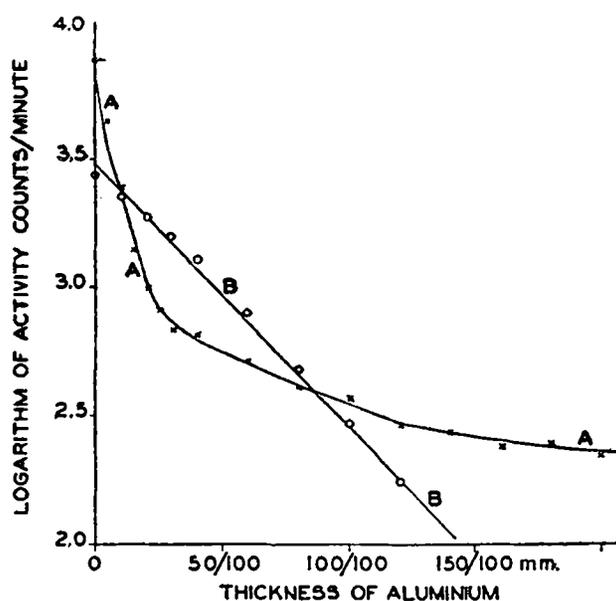

FIG. 1. Absorption curves of radiations.

A. Non-depositable substance.   B. Purified source of Rd-Te (Max. energy of β: 1·8 Mev.).

examinations showed that the activity of the cathodic deposition corresponded mainly to ^{129m}Te metastable in equilibrium with ^{129}Te (half-life: 34 days; β rays of 1·8 Mev. maximum energy); whereas the activity of the non-depositable substance might have arisen out of the presence of ^{95}Zr (half-life: 65 days; β particles: 0·4 Mev. max. energy).

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‡ Slope of these curves is a measure of the energy of the β particles.[9] A curve of the type B is usually obtained in the case of an uncontaminated β emitter. On analysis of the absorption curve pertaining to the non-depositable substance, one would obtain the curve B of Fig. 2. The slope of the latter curve would represent the energy characteristic of the radiations emitted by a major component of the non-depositable substance.
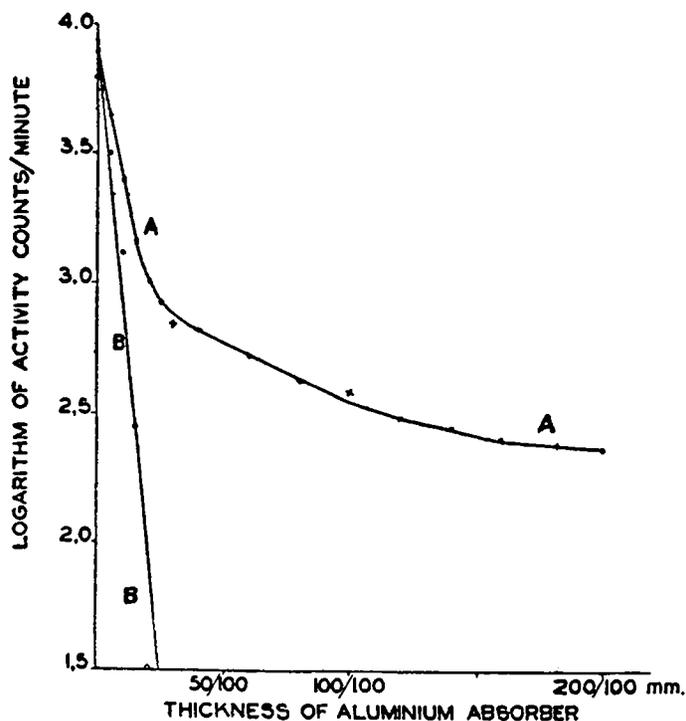
Fig. 2. Absorption curves of radiations.

A. Absorption curve of the non-depositable substance. B. Analysed component of curve A (Max. energy of β: ~0·4 Mev.).

The method described above, therefore, seems to be quite suitable for the purification of carrier free tellurium tracer and for the preparation of its sources exempt from other foreign matter in weighable quantities.

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