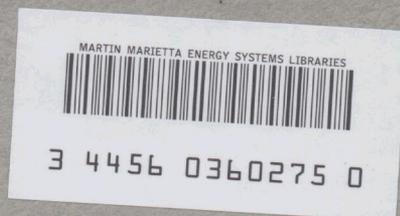


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FROM RADIO-IODINE

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A Method for the Separation of Radio-Tellurium from
Radio-Iodine

A. D. Bogard and J. E. Hudgens, Jr.

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February 10, 1948

Date Received: 7/29/48

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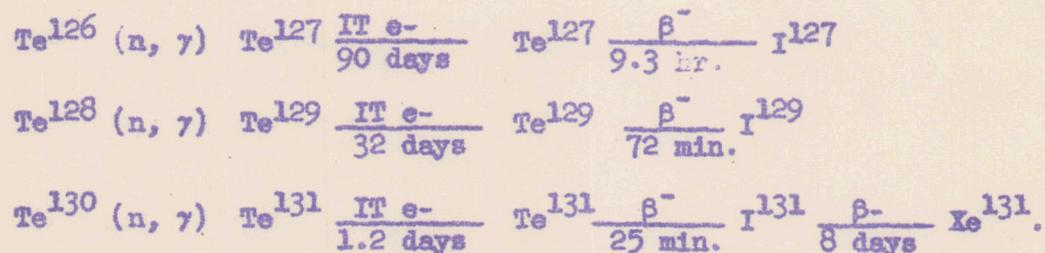
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Introduction

The determination of small amounts of radioactive tellurium in the presence of relatively large amounts of radioactive iodine is necessary in the production of 8 day iodine 131 in the Oak Ridge National Laboratories radioisotope program. The tellurium activity is produced by the (n, γ) reaction on tellurium 126, 128, and 130. These transformations may be expressed graphically by the following scheme (6).



The concentration of tellurium normally present in the iodine preparations necessitates the determination of less than microcurie (3.7×10^4 disintegrations per second) amounts of tellurium activity in the presence of millicurie (3.7×10^7 disintegrations per second) amounts of iodine activity. It was desirable to minimize the quantity of iodine activity carried through and also the time required for each analysis.

Since the ratio of the iodine concentration to the tellurium concentration in the samples on which tellurium analyses were required was very high, few methods applicable to this problem are found in the literature and no method capable of isolating the tellurium activity free of iodine activity was discovered. Abelson (1) separated tellurium activity from other fission element activities by precipitating elemental tellurium (and selenium) from a weak hydrochloric acid solution with sulfur dioxide and separating the selenium from the tellurium by precipitation of the selenium from a 12 N hydrochloric acid solution with sulfur dioxide as before. Tellurium is not precipitated under these conditions. Iodine was removed by precipitation of silver iodide from a 6 N sulfuric acid solution with silver sulfate. Seaborg, Livingood, and Kennedy (6) separated tellurium activity from

iodine activity by reducing sodium iodate with an equivalent amount of iodide and extracting with chloroform. A separation of tellurium activity from other fission element activities was accomplished by Glendenin (4) by using a method similar to the one reported here; however, volatilizations of elemental bromine or holdback carriers were not used to aid the separation of tellurium activity from iodine activity. These improvements have been incorporated in the method given in this paper with the result that a cleaner separation is accomplished. Some improvement in technique has also been realized.

Preparation of Carrier Solutions

Tellurium carrier (10 mg. of elemental tellurium per milliliter) was prepared and standardized according to the method of Hume (4). Briefly, this method consisted of dissolving 10 to 11 grams of analytical reagent grade tellurium in approximately 10 ml. of C. P. concentrated nitric acid (an insoluble residue remained; however this residue was ignored since it was dissolved by the hydrochloric acid treatment.) The resulting solution was evaporated to approximately 2 ml., 10 ml. of C. P. concentrated hydrochloric acid added and this solution again evaporated to approximately 2 ml. The evaporation with hydrochloric acid was repeated two more times to remove traces of nitric acid and the resulting solution diluted to one liter with 3 M hydrochloric acid.

Standardization of Tellurium Carrier Solution

The carrier solution was standardized by precipitating elementary tellurium with sulfur dioxide. An aliquot of the solution (5 ml.) was pipetted into a 125 ml. Erlenmeyer flask, approximately 40 ml. of 3 M hydrochloric acid added, the solution heated nearly to boiling, and the tellurium precipitated by saturating the solution with sulfur dioxide. The precipitate was then quantitatively transferred to a weighed sintered glass crucible to which suction was applied, washed three times with 5 ml. portions of 95% ethanol, and three times with 5 ml. portions of ether, and dried at ~ 110° C. to constant weight. The precipitate was weighed as elemental tellurium.

Iodine carrier 10 mg. of iodine per milliliter of solution was prepared by dissolving 13 gms. of reagent grade potassium iodide and 0.1 g. C. P. grade sodium bicarbonate in one liter of distilled water.

Iron Carrier 10 mg. of ferric iron per milliliter was prepared by dissolving 72.5 gms. of reagent grade ferric nitrate, monohydrate, and 1 ml. of concentrated nitric acid in one liter of distilled water.

Details of the Analytical Procedure

The size of the sample which can be used for a tellurium activity determination is limited by the counting rate that the counting arrangement is able to determine accurately, and by health considerations. The optimum counting rate for the counting apparatus which has been previously described (3) is approximately 6000 counts per minute. A sample size should be chosen to give this counting rate if (1) the sample contains sufficient tellurium activity per unit volume and (2) the concentration of radioactivity attributable to elements other than tellurium is not sufficient to make the solution hazardous from a health standpoint. (A radiation dosage rate above 12 milliroentgens per hour is considered hazardous in this laboratory, and efforts are made to minimize the exposure of personnel to radioactivity by shielding the sample from the operator, performing the operations from a distance and by limiting the time the operator is permitted to remain in a region of high radiation intensity.) The sample is pipetted into a 50 ml. centrifuge tube, and 2 ml. each of iodine and tellurium carriers added. The solution is then neutralized by adding concentrated ammonium hydroxide dropwise until a white precipitate of tellurous acid mixed with ammonium tellurite just remains. The tellurium is precipitated by adjusting the volume of the solution to approximately 15 ml. with distilled water, adding approximately 15 ml. of 6 M hydrochloric acid, heating nearly to boiling and rapidly bubbling in sulfur dioxide. After precipitation is complete, as indicated by the absence of a finely divided precipitate, the elemental tellurium is removed by centrifuging and the supernatant solution is discarded. The precipitate is washed with approximately 20 ml. of distilled water.

The tellurium residue is suspended in 3 ml. of concentrated hydrobromic acid and the mixture slowly heated until the boiling point of the hydrobromic acid is reached. A further separation of tellurium from iodine and selenium activities is then accomplished by removing the mixture from the

flame and adding ~5 drops of 6 M nitric acid to volatilize elemental bromine. Caution must be exercised since this reaction may proceed with the violent evolution of gas if the hydrobromic acid is too hot. After the initial rapid evolution of bromine subsides, the solution is heated gently to drive off additional bromine and the addition of 6 M nitric acid in 2 to 4 drop portions continued until no more bromine is evolved and the solution is water white. The solution is evaporated to dryness, and the residue dissolved in 4 to 8 drops of 6 M hydrochloric acid.

The tellurium activity is separated from rhodium and rare earth activities by the precipitation of ferric hydroxide as a scavenging precipitation. Although a step whose purpose is to separate tellurium from these activities may not always be necessary when the iodine is prepared by the bombardment of tellurium with neutrons, the step has been incorporated in order to make the procedure applicable to a wider variety of solutions and in particular to the determination of tellurium in the presence of iodine separated from fission element mixtures. Neutralization of the solution is accomplished by adding C. P. concentrated ammonium hydroxide dropwise until the white precipitate of tellurous acid mixed with ammonium tellurite first formed just dissolves then 4 drops of ammonium hydroxide are added in excess. The solution is heated nearly to boiling and 1 mg. of ferric iron (0.1 ml. of iron carrier) is added while stirring vigorously. The solution is then allowed to stand a few minutes before removal of the ferric hydroxide by centrifugation. The supernatant solution is removed to a second 50 ml. centrifuge tube and the ferric hydroxide precipitate discarded.

Precipitation of elemental tellurium is accomplished by adding to the supernatant solution from the ferric hydroxide precipitation an equal volume of 6 M hydrochloric acid and 2 ml. of iodine holdback carrier, heating nearly to boiling, and rapidly bubbling sulfur dioxide through the solution as before. The precipitate is centrifuged from solution and the supernatant discarded.

The tellurium is dissolved by warming with approximately 5 drops of 6 M nitric acid, this solution evaporated to dryness and the residue dissolved in 4 to 8 drops of 6 M hydrochloric acid. A reprecipitation is then performed by diluting to 10 ml. with distilled water, adding 10 ml. of 6 M hydrochloric acid and 2 ml. of iodine holdback carrier, heating to boiling and saturating the solution with sulfur dioxide.

The elemental tellurium precipitate is filtered on a tared filter paper (1.4 cm. in diameter) in a size 0000 hirsch funnel. The filter paper is prepared by washing with hot distilled water, ethanol and ether, drying at $\sim 105^{\circ}$ C. for five minutes and weighing on a one inch watch glass. After filtration, the precipitate is washed three times with 5 ml. volumes of hot distilled water, absolute ethanol and finally anhydrous ether. The filter paper containing the tellurium precipitate is then dried at $\sim 105^{\circ}$ C. for five minutes and weighed as before. A chemical yield can then be calculated from the weight of the tellurium added and the weight of this tellurium precipitate.

The sample is then mounted by placing the watch glass on a flat surface, covering it with a cellophane sheet (usually ~ 2 inches square), and pressing a stiff card (2 1/2 inches by 3 1/4 inches with a 1 1/16 inch hole in the center) down from above so that the watch glass now covered with the tightly stretched cellophane rises through the hole. A strip of gummed paper or scotch tape is then placed on the bottom of the card, sealing the watch glass into place. This method of mounting supplies firm support and good centering for the sample and prevents contamination of the counting equipment by loss of sample.

The counting apparatus consists of a thin window Geiger tube with scaling circuit and recording system. This apparatus has been used successfully for the analysis of protoactinium 233 and is described in detail in a previous report (3).

The observed counting rate is corrected for chemical yield through the separations and for the coincidence losses and for the background of the counting

arrangement. The usual chemical yield is greater than 80% and the error introduced by this correction is small. Since these corrections have been adequately discussed previously (3, 4, 5) a detailed discussion will not be included in this paper.

Decontamination Studies

A sample containing 1.3×10^9 disintegrations per minute of iodine¹³¹ activity was analyzed for tellurium according to the methods of Glendenin (4). Beta activity was present in the recovered tellurium, and decay and adsorption curves indicated this activity was from iodine¹³¹ (5). There was poor correlation between activity present in the isolated tellurium and the activity initially present. The elemental tellurium precipitate was contaminated with greater than 0.004% of the iodine activity initially present.

Experiments were performed to determine the specificity of the method of this report. A sample of the most radiochemically pure iodine activity available from the radioisotope group was used as a starting solution and the separations performed in the usual manner. After the tellurium analysis had been completed and the activity of the final tellurium precipitate determined, the tellurium was dissolved and the concentration of radioactive iodine in this solution determined using the method of Hume (4). The results of these experiments are shown in Table I.

Table ISeparation of Tellurium Activity from Large Amounts of Iodine Activity(c/m Are Given for a Geometry of ~33%^{**})

<u>c/m Iodine Introduced in Raw Sample</u>	<u>c/m Recovered on Te ppt.</u>	<u>c/m I-Isolated With Te ppt.</u>	<u>Decontamination Factor*</u>
9.7 x 10 ⁸	3.6 x 10 ³	7.7 x 10 ¹	7.9 x 10 ⁻⁸
9.7 x 10 ⁸	4.2 x 10 ³	9.2 x 10 ¹	9.5 x 10 ⁻⁸
9.7 x 10 ⁸	3.9 x 10 ³	6.8 x 10 ¹	7.0 x 10 ⁻⁸
9.7 x 10 ⁸	3.1 x 10 ³	6.7 x 10 ¹	6.9 x 10 ⁻⁸

*Decontamination factor = $\frac{\text{Final foreign activity}}{\text{Initial foreign activity}}$

**This geometry includes a correction (~30% of the observed counting rate) for backscattering from the one inch watch glasses on which the samples were counted. In accurate radiometric analyses the magnitude of this effect must be evaluated accurately or efforts made to minimize the effect of mounting samples on more nearly weightless mounting materials. Since the mounting procedure was rigidly standardized for these experiments the accuracy is satisfactory on a comparative basis.

Discussion

The tellurium method outlined in this paper is well adapted to remote operation. The sample may be taken with one of the several types of remote samplers, the reagents added with little or no exposure of personnel and the first precipitation of elemental tellurium performed at a distance by the operator. The agitation necessary may be performed by using motor or air-driven agitators which can be operated from a distance. The concentration of foreign activity is usually reduced to a safe working level by the first precipitation and further manipulations may be made using only those precautions necessary in ordinary radiochemical work.

When iodine carrier is added to the tellurium carrier a black precipitate is formed. This precipitate is either tellurium tetraiodide or a compound of

the type Me_2TeI_6 (2). This precipitate may be ignored since it is decomposed upon heating and is soluble in ammonium hydroxide.

The major portion of the radioactive iodine is removed in the supernatant solution from the first precipitation of tellurium. There is some removal of iodine activity in connection with the nitric and hydrobromic acid treatment, however, the primary function of this step is to remove selenium activity. Rhodium and rare earth activities are removed most efficiently with the ferric hydroxide scavenging.

In addition to the usual analytical errors, there are additional errors peculiar to radiochemistry. These include contamination from outside sources such as glassware used in the analysis, contamination from large amounts of other fission elements present in the sample but not satisfactorily removed by the chemical manipulations, and counting equipment defects. Counters must be kept at constant temperature and operating voltage. Electrical elements within the unit must operate at constant output values. A small variation in input voltage, temperature changes of the counter tube, or a breakdown in any part of the counter circuit may make restandardization necessary.

Summary

A method capable of separating trace concentrations of tellurium activity from large concentrations of foreign activities (especially from iodine activity) is given. Data are given to show the specificity of the method and the decontamination which can be obtained. The method is well adapted to remote manipulations.

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