

Dec. 8, 1970

HIROKAZU UMEZAWA ET AL

3,545,925

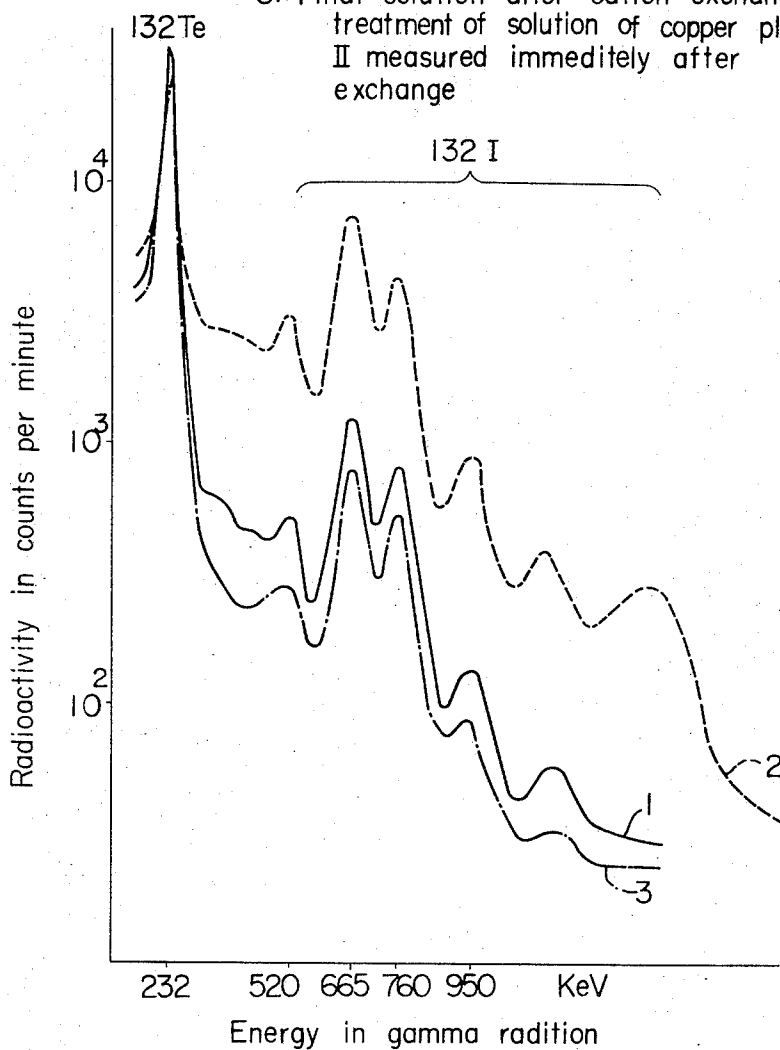
METHOD FOR PRODUCING RADIOACTIVE TELLURIUM

Filed Sept. 23, 1966

2 Sheets-Sheet 1

Fig. 1

1. Copper plate I
measured 25 minutes after separation
2. Copper plate I
measured 24 hours after separation
3. Final solution after cation exchange
treatment of solution of copper plate
II measured immediately after
exchange



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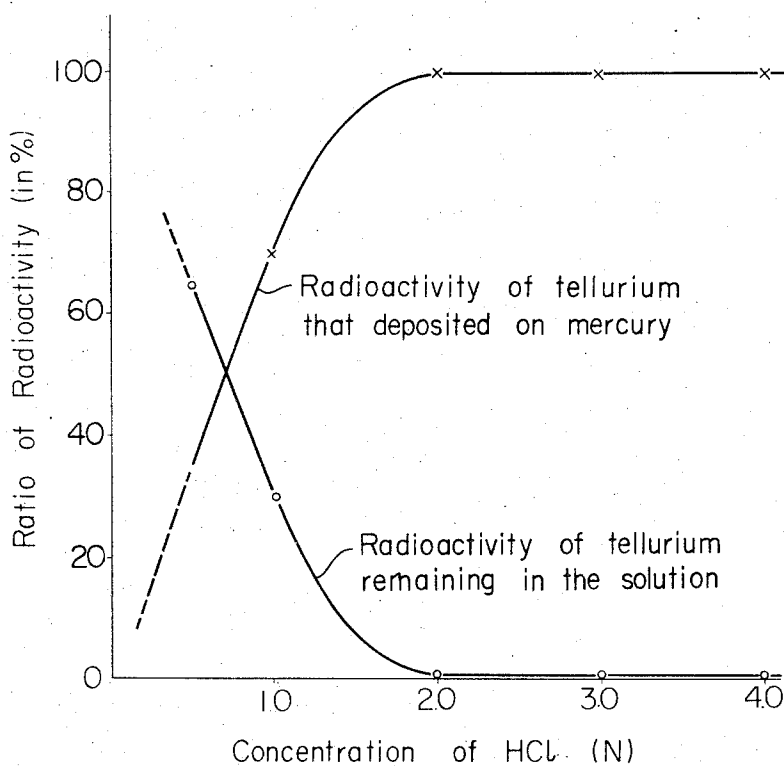
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Fig. 2

Relation between deposition yield of tellurium and concentration of HCl



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METHOD FOR PRODUCING RADIOACTIVE
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3 Claims

ABSTRACT OF THE DISCLOSURE

Radioactive tellurium is produced by dissolving a nuclear fission product containing radioactive tellurium in an aqueous hydrogen halide or thiocyanate solution, introducing Ni, Cu, Bi, Hg or Ag into the solution to deposit radioactive tellurium selectively thereon, separating the metal with it tellurium deposit from the solution, and recovering radioactive tellurium from the metal.

This invention relates to a process for producing radioactive tellurium, a mother nuclide of short half-life radioactive iodine which is in big demand for medical treatment and other purposes.

Radioactive tellurium is chiefly produced from nuclear fission products, and several processes, such as precipitation, distillation or separation by adsorption chromatography are in practical use. However, these methods have defects in that separation is incomplete and highly pure products or carrier-free products are not obtained, or remotely controlled operation is difficult.

The method of this invention comprises: dissolving in a hydrogen halide acid or a thiocyanate solution an actinide element or its compound that has been irradiated with neutrons or other accelerated particles such as protons, alpha-particles and photons, bringing said solution in contact with Ni, Bi, Cu, Ag, or Hg, letting radioactive tellurium found among the fission products of said element deposit spontaneously and selectively on said metal, and removing said metal together with the deposit from the solution, dissolving said metal in a mineral acid, and separating the radioactive tellurium from said mineral acid solution by a suitable method.

Each step of the above-mentioned process is quite simple and is easily carried out in a shielded chamber by means of remotely operated manipulators, and no fission product other than tellurium is found in the thus separated tellurium. These are remarkable advantages of this method.

By the term "radioactive tellurium," we mean ^{134}Te (42 min.), ^{133}Te (50 min.), ^{132}Te (78 hours), ^{131}Te (1.2 day), ^{129}Te (33 days), and ^{127}Te (105 days). (Half life is referred to in parentheses.) The species of isotope produced depends upon the duration of irradiation and cooling. This problem is discussed later.

The amount of non-radioactive tellurium in the fission products of actinide elements is negligible, though some may exist. Therefore, carrier-free radioactive tellurium is obtained by this method. The radioactive tellurium is obtained in the form of 0.5–1 N hydrochloric acid solution, which is a very suitable form for use or for further processing of the recovered radioactive tellurium. Tellurium in 0.1–0.5 N hydrochloric acid solution is adsorbed by a cation exchanger in hydrogen form, and the adsorbed tellurium is desorbed by washing with 1 N or more concentrated hydrochloric acid. The mechanism of the adsorption and desorption is not yet clearly known.

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However, tellurium must be quadrivalent in order to be adsorbed well by the cation exchanger.

Now referring to the attached figures we will illustrate our invention by typical examples of the experiments carried out by the inventors.

FIG. 1 shows the gamma ray spectrum of ^{132}Te separated by means of copper.

FIG. 2 shows the relation between the deposition yield of ^{132}Te and the HCl concentration in the deposition medium.

EXAMPLE 1

One-tenth gram (0.1 g.) of uranyl acetate was irradiated in the Japan Research Reactor No. 1 (JRR-1) at the neutron flux 10^{11} neutrons/cm² sec. for 10 hours (3.6×10^{15} neutrons/cm² in total flux). In this condition, the radioactive tellurium isotope to be produced is ^{132}Te . After being taken out of the reactor, the uranyl acetate was dissolved in a hydrochloric acid solution to make 5 ml. of 1 N hydrochloric acid solution.

Two pieces (denominated I and II) of 1 x 1 cm. thin copper plate (total surface area is 2 cm² for each piece) were placed in the solution, which was stirred for 10 minutes. The two pieces were taken out and washed with 1 N HCl and acetone. One piece (I) was used for determination of its radiation by means of a gamma ray spectrometer and a Geiger-Mueller counter. The other piece (II) was dissolved in nitric acid and the solution was evaporated; the solid remaining was dissolved in a concentrated hydrochloric acid solution, said solution was boiled for a few minutes so that the tellurium ions might be in quadrivalent state, and was diluted with water to a concentration of 0.1–0.5 N with respect to HCl.

The solution was passed through a column of cation exchange resin in hydrogen form so that the tellurium might be adsorbed by the resin. For this purpose, for the above-mentioned reason, the concentration of hydrochloric acid in the solution must be 0.1–0.5 N. As the cation exchanger, Diaion SK-1 manufactured by Mitsubishi Chemical Industries, Ltd., which is equivalent to Dowex 50 X-8, was employed. The exchanger column was washed with a hydrochloric acid solution, the concentration of which is 0.1–0.5 N. Then the adsorbed tellurium was eluted selectively by 1 N hydrochloric acid solution.

The collected solution was checked by gamma ray spectrometry. The gamma ray spectra obtained are shown in FIG. 1, and they are the same as those of the known specimen of ^{132}Te . By beta decay, ^{132}Te incessantly produces ^{132}I , which accompanies the former all the time. In FIG. 1, no gamma radiation caused by nuclides other than ^{132}Te – ^{132}I were detected. The amount of ^{132}Te produced in the irradiated uranyl acetate was about 10 microcuries immediately after the irradiation was finished; the amount of ^{132}Te in the collected final solution was 2 microcuries which corresponds to 8 microcuries as of immediately after the irradiation.

In the following experiments, uranyl acetate irradiated in the same condition as above was used.

EXAMPLE 2

To the same solution (2 ml.) of irradiated uranyl acetate as prepared in Example 1, 60 mg. of mercury was added, and the solution was agitated by means of supersonic waves for 1 minute. The dispersed mercury was coagulated by centrifuging, and was taken out of the solution. The radioactivity of the collected mercury was determined. Then the collected mercury was dissolved in nitric acid, and the tellurium therein was separated by cation exchange resin as described in Example 1.

The result was as follows:

Initial amount of ^{132}Te in the irradiated uranyl acetate:
4 μCi

Amount of ^{132}Te finally collected in hydrochloric acid solution: 3 μCi

EXAMPLE 3

An experiment closely paralleling Example 1 was carried out using hydrobromic acid as the deposition medium instead of hydrochloric acid and nickel pieces instead of copper pieces as the depositing agent.

That is, 0.1 g. of irradiated uranyl acetate was dissolved in 5 ml. of 1 N hydrobromic acid and two pieces of thin nickel plate 1 x 1 cm., were added. The nickel pieces upon which tellurium deposited were treated as in Example 1, and a quite similar result was obtained.

The initial amount of ^{132}Te in the irradiated uranyl acetate: 10 microcuries.

The amount of ^{132}Te finally collected in hydrochloric acid solution: 8 microcuries.

EXAMPLE 4

One-tenth gram (0.1 g.) of uranyl acetate irradiated under the same conditions as in Example 1 was dissolved in 2 M/l potassium thiocyanate solution to finally make 2 ml. of solution.

From this solution, ^{132}Te was deposited by means of 60 mg. of mercury in the same way as described in Examples 1 and 2.

The result was as follows:

Initial amount of ^{132}Te in the irradiated uranyl acetate: 10 microcuries.

Amount of ^{132}Te finally collected in hydrochloric acid solution. 10 microcuries.

EXAMPLE 5

The same experiment was carried out with respect to 1 mol/ μ sodium thiocyanate solution and 5 mol/l ammonium thiocyanate solution, 1 x 1 cm. copper plates being used as depositing agent.

The results were as follows:

	NaSCN	NH ₄ SCN
Initial amount of ^{132}Te in the irradiated uranyl acetate.	10 μCi	10 μCi
Amount of ^{132}Te finally collected in hydrochloric acid solution.	6 μCi	6 μCi

EXAMPLE 6

One-tenth gram (0.1 g.) of the irradiated uranyl acetate was dissolved in 1 N hydrofluoric acid to finally make 2 ml. of solution.

From this solution, ^{132}Te was deposited by means of 60 mg. of mercury in the same way as described in Examples 1 and 2.

The results were as follows:

Initial amount of ^{132}Te in the irradiated uranyl acetate: 10 μCi

Amount of ^{132}Te finally collected in hydrofluoric acid solution: 1 μCi

EXAMPLE 7

Two milliliter batches of the solutions of various concentrations of hydrochloric acid and potassium thiocyanate each containing 0.1 microcuries of ^{132}Te were prepared. To each solution, 60 mg. of mercury was added and the same operation as in Example 2 was repeated, and radioactivity of the tellurium deposited on the mercury and that remaining in the solution was measured, and thus the yields of tellurium under various conditions were determined. The results with respect to variety in concentration of hydrochloric acid are shown in FIG. 2. When the concentration of HCl was 1 N, the yield was 70% by radioactivity. When it was more than 2 N, the yield was more than 99% by radioactivity. In the case in which potassium thiocyanate was used, the yield was 90% by radioactivity for 1 M/l, and more than 99% by radioactivity for more than 2 M/l concentration. Even

if uranium existed in the solution, the yield did not decrease.

It has been known that various radioactive tellurium isotopes exist among fission products of any actinide element irradiated with neutrons and other accelerated particles such as protons, alpha-particles and photons. The species of tellurium isotope produced depends upon the duration of irradiation and cooling. When uranium is irradiated by thermal neutrons (about 10^{11} neutrons/cm.² sec.), ^{134}Te or ^{133}Te is produced by irradiation of 10-60 minutes, ^{132}Te or ^{131}Te by irradiation of 10-60 hours, and ^{129}Te or ^{127}Te by irradiation of 10-100 days. (Refer, e.g., to Nucleonics, vol. 18, No. 11, p. 201 (1960), by S. Katcoff.) Therefore, by choosing a suitable irradiation time, a desired isotope can be produced. Chemical behavior is common to all the isotopes, so the method of this invention is applicable to production of any radioactive tellurium isotope. When a long life isotope is desired, irradiated uranium (or any other actinide element) is left standing for short life isotopes to decay out.

In all the examples tellurium was separated from the metal on which it was deposited by means of an ion exchanger after the metal had been dissolved in nitric acid, the solution evaporated, and the remaining solid dissolved in HCl. However, this separation can be carried out by any known method, for instance, electrolytic deposition, liquid-liquid extraction by tributyl phosphate, or precipitation by addition of carrier tellurium. In the last method, radioactive tellurium with a non-radioactive tellurium carrier is obtained.

The advantages of this invention are as follows:

(1) Deposition of tellurium from the solution of a starting material is highly selective and rapid.

(2) Addition of a special carrier (vehicle) for tellurium is unnecessary, and thus carrier-free radioactive tellurium can be obtained in very high yield.

(3) Any material containing radioactive tellurium that can be made into a solution with hydrogen halide acid or thiocyanate solution can be used as a starting material.

(4) Radioactive tellurium is obtained in a solution in 0.5-1 N hydrochloric acid, which is a very convenient form for later treatment or use.

We claim:

1. A method of producing radioactive tellurium, comprising

(1) dissolving a nuclear fission product containing radioactive tellurium in an aqueous solution of a hydrogen halide acid having a concentration of at least 1 N to obtain an aqueous solution of the fission product,

(2) introducing Ni, Cu, Bi, Hg, or Ag metal into said aqueous solution of the fission product and selectively depositing radioactive tellurium on the metal,

(3) separating the metal with the radioactive tellurium deposit from the solution,

(4) dissolving the metal with the radioactive tellurium deposit in aqueous nitric acid,

(5) evaporating said aqueous nitric acid solution to dryness,

(6) dissolving the dried substance in a concentrated hydrochloric acid solution,

(7) boiling said hydrochloride acid solution and diluting the same to 0.1-0.5 N in respect to hydrochloric acid,

(8) passing the said hydrochloric acid solution through a cation exchange resin of hydrogen form,

(9) then washing the resin with hydrochloric acid of a concentration of 0.1-0.5 N, and

(10) recovering radioactive tellurium by eluting the adsorbed tellurium from the resin with 1 N hydrochloric acid.

2. A method of producing radioactive tellurium, comprising

(1) dissolving a nuclear fission product containing radioactive tellurium in an aqueous solution of a

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thiocyanate having a concentration of at least 1M/liter to obtain an aqueous solution of the fission product,

(2) introducing Ni, Cu, Bi, Hg, or Ag metal into said aqueous solution of the fission product and selectively depositing radioactive tellurium on the metal,

(3) separating the metal with the radioactive tellurium deposit from the solution, and

(4) recovering radioactive tellurium from the separated metal.

3. A method as specified in claim 2, in which the metal onto which the radioactive tellurium deposits is separated from said solution and is dissolved in aqueous nitric acid, said nitric acid solution is evaporated to dryness, the remaining solid is dissolved in a concentrated hydrochloric acid solution, said hydrochloric acid solution is boiled and is diluted to 0.1–0.5 N with respect to hydrochloric acid, and said hydrochloric acid solution is

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passed through a cation exchange resin of hydrogen form, and then said resin is washed with hydrochloric acid, the concentration of which is 0.1–0.5 N, and the adsorbed radioactive tellurium is eluted from said resin with 1 N hydrochloric acid.

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23—87; 252—301.1

UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,545,925 Dated Dec. 8, 1970

Inventor(s) Hirokazu Umezawa et al

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

┌ In the heading, "Oct. 25" should read --Oct. 12--

Signed and sealed this 11th day of May 1971.

(SEAL)
Attest:

EDWARD M. FLETCHER, JR.
Attesting Officer

WILLIAM E. SCHUYLER,
Commissioner of Patent

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