

1

2,938,768

**METHOD OF SEPARATING Pu FROM
METATHESIZED BIPO₄ CARRIER**

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5 Claims. (Cl. 23—14.5)

This invention relates to a process for the separation of actinide rare earth elements from contaminating elements and more specifically is concerned with a process for separating such actinide rare earth elements as plutonium and neptunium from a bismuth hydroxide carrier precipitate.

Plutonium and neptunium are usually formed by the neutron-irradiation of uranium. Neutron-irradiation of a uranium mass to produce such transuranic elements as plutonium and neptunium is ordinarily stopped when the transuranic elements are present in the mass in very small concentration. One of the primary reasons for this is the concurrent production of radioactive fission products in the mass in such concentration that the radioactivity produced thereby in the mass rapidly rises to such large levels that the mass becomes very hazardous to human beings. The separation of transuranic elements from a neutron-irradiated uranium mass requires special techniques because of this radioactivity and because of the extremely small concentration of these elements in relation to the mass.

One method of separating transuranic elements from a neutron-irradiated mass which has been found to be most successful is by alternate oxidation and reduction steps with intervening carrier precipitations whereby the plutonium and fission products are carried from the process solutions with a carrier precipitate. One such process in which bismuth phosphate is employed as the carrier is set forth in copending application S.N. 478,570, filed in the U.S. Patent Office March 9, 1943, and entitled "Phosphate Method for the Separation of Radioactive Elements"; it was patented on July 16, 1957, as U.S. Patent No. 2,799,553. By means of this process a final bismuth phosphate carrier precipitate containing plutonium substantially freed of fission products and the uranium may be obtained. The bismuth phosphate carrier containing plutonium may then be converted into a more easily soluble carrier precipitate by a metathesis step to form a bismuth hydroxide carrier precipitate containing plutonium. A process for the carrying out of this metathesis is described in copending application S.N. 745,108, filed in the U.S. Patent Office April 30, 1947, and entitled "Metathesis of Bismuth Phosphate Plutonium Carrier Precipitate With an Alkali."

A conventional method of recovering the plutonium from such a bismuth hydroxide carrier precipitate comprises the dissolution of the precipitate in a strong mineral acid followed by the precipitation of the plutonium with a carrier such as lanthanum fluoride under such conditions that the bismuth ions do not precipitate. While this method of recovery is effective, it does require fairly large quantities of acid to effect the dissolution and requires the further separation of the plutonium from the lanthanum fluoride carrier before the plutonium can be obtained in a pure state.

It is an object of the present method to provide a simple and direct process for the recovery of actinide rare earth

2

elements from a bismuth hydroxide carrier precipitate containing said elements.

Additional objects of the present invention will be apparent from the following detailed description thereof.

We have discovered that an actinide rare earth such as plutonium or neptunium may be recovered from a bismuth hydroxide carrier precipitate containing said element by treating the bismuth hydroxide carrier precipitate with a dilute solution of nitric acid. This treatment of the bismuth hydroxide carrier precipitate with dilute nitric acid will result in the actinide rare earth dissolving in the dilute acid, whereas the bismuth hydroxide precipitate does not dissolve therein. Although we do not wish to be bound by any theory advanced, it is believed that the bismuth hydroxide is converted to the bismuth oxynitrate salt and that it is this salt which is insoluble in the dilute nitric acid.

The concentration of the acid used as the wash is important, since a concentrated nitric acid will dissolve not only the actinide rare earth hydroxides, for example plutonium hydroxide, but also the bismuth hydroxide. It is therefore essential that the aqueous solution used to dissolve the plutonium be less than about 0.5 N in nitric acid. A preferable range of acid concentration has been found to be about 0.05–0.5 N with solutions of about 0.1 normality in HNO₃ giving the best results. Higher acid concentrations give somewhat increased reaction speeds, but at the expense of a lowered separation factor, whereas lower concentrations are in general too slow for plant operations. The amount of acid used will in general vary with the conditions of use, but 10 to 15 ml. of 0.1 M HNO₃ per gram of bismuth will in general give satisfactory solution in a reasonable time.

The temperature of the reaction vessel is not critical and dissolution at room temperature may be effected in a reasonable time. Various methods of contacting the bismuth hydroxide carrier precipitate with the acid solution may be used, and the process of this invention is extremely flexible as to the equipment in which it may be carried out; thus it may be carried out in a centrifuge bowl, a tank reactor, or in any other suitable apparatus. It is desirable that the reaction be carried out with agitation. Thus, for large scale operations, a very convenient method is to carry out the metathesis of bismuth phosphate plutonium-containing precipitate in a centrifuge bowl and, after washing the carrier precipitate in the bowl, the nitric acid solution may be added directly to the centrifuge bowl and slurried in the bowl. Upon completion of the dissolution reaction the plutonium-containing solution may be separated from the bismuth oxynitrate cake by centrifugation.

Now that the process of this invention has been described it may be illustrated by the following example.

Example

A bismuth hydroxide carrier precipitate cake containing 1250 mg. of bismuth hydroxide and 12 mg. of plutonium hydroxide was slurried with 15 ml. of 0.1 N HNO₃ solution for 30 minutes. The slurry was then subjected to centrifugation whereby the plutonium-containing liquid medium was separated from the bismuth-containing solid. Two more washes each with 15 ml. of 0.1 N HNO₃ were then carried out and the three supernatants were combined. Analysis disclosed that 97% of the plutonium present in the original bismuth hydroxide cake was contained in the supernatant solution and only 3% of the bismuth had dissolved in the nitric acid washes.

While recovery of plutonium from a bismuth hydroxide carrier has been described, it is to be understood that such other actinide rare earths as uranium, neptunium or mixtures of actinide rare earths such as plutonium and

3

neptunium may be recovered by the process of this invention. Other modifications will also be apparent to those skilled in the art and it is the intention to claim all novelty inherent in the invention as broadly as possible in view of the prior art.

What is claimed is:

1. A process of recovering an actinide rare earth from a bismuth hydroxide carrier precipitate which comprises washing said precipitate with an aqueous solution 0.05-0.5 N in HNO_3 , whereby the transuranic polyvalent actinide rare earth is dissolved, and separating the aqueous solution from the insoluble bismuth precipitate.

2. The process of claim 1 wherein the actinide rare earth is uranium.

3. The process of claim 1 wherein the actinide rare earth is neptunium.

4. The process of claim 1 wherein the actinide rare earth is plutonium.

4

5. The process of recovering plutonium from a bismuth hydroxide carrier precipitate which comprises washing said precipitate with 10-15 ml. 0.1 M HNO_3 per gram of bismuth, whereby the plutonium is dissolved in said solution, and separating said solution from the insoluble bismuth precipitate.

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