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METHOD OF DISSOLVING URANIUM METAL

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This invention relates to a process of efficiently recovering radioactive fission products having short half-lives. This invention is also concerned with an economical means of dissolving metallic uranium.

An object of the invention is to provide an improved method for dissolving metallic uranium. Other and further objects will become apparent from the accompanying disclosure.

It is known, as a result of recent developments in nuclear physics, that if uranium is subjected to bombardment with thermal or slow neutrons originating in a cyclotron or similar device, uranium is converted to other products including the transuranic element 93^{239} and fission products having atomic numbers 35 to 46 and 51 to 60 inclusive. Many of these products produced as a result of nuclear fission in the uranium are radioactive elements having short half-lives. These half-lives vary for different elements some being as short as 30 seconds to about 30 minutes.

In the study of such radioactive fission products, it is important that the products be recovered in condition for study or use as radiation sources in as short a time as possible after the neutron bombardment has ceased. Accordingly, the ordinary chemical laboratory methods of recovery, such as by dissolution of irradiated material in nitric acid, may take such a long time that important short-lived radioactive fission products may be entirely lost.

Although uranous oxide (UO_2) can be rapidly dissolved in nitric acid, metallic uranium will not dissolve in nitric acid with sufficient speed to make certain that all radioactive elements with short half-lives are recovered. For example, radioactive elements with a half-life of less than about 30 minutes could not be recovered by the ordinary procedure of dissolving uranium metal in nitric acid.

In addition to the need for dissolving uranium containing radioactive elements having short half-lives rapidly enough so that these fission products can be recovered, dissolution of scrap uranium metal for purposes of reworking is an important problem. As ordinarily carried out, concentrated nitric acid is used, which involves the well-known difficulties of a highly corrosive material.

In accordance with the present invention, it has been found that, in the removal of radioactive fission products from neutron-irradiated uranium metal containing such products, the addition of a small amount of perchloric acid ($HClO_4$) to the concentrated nitric acid in which the uranium is being dissolved greatly shortens the time necessary for dissolution of the metal. As a result, the short-lived radioactive elements can quickly be separated from the resultant solution by precipitation and separation processes well-known to the art.

It has also been found that by adding a small amount of perchloric acid ($HClO_4$) to the nitric acid used to dissolve scrap uranium metal, a much more dilute nitric acid can be used and the metal will dissolve in approximately

the same length of time as it would in concentrated nitric acid which is advantageous. When a large amount of valuable scrap uranium is being dissolved this factor is of importance.

In view of the fact that concentrated nitric acid is itself a strong oxidizing agent, the fact that the addition of a small quantity of another oxidizing agent, perchloric acid, brings about a disproportionately large increase in speed of dissolution of uranium metal is an unexpected result. Thus it has been found that the addition to the nitric acid of a substantial quantity, usually not less than about 1 or 2 percent of perchloric acid based on the weight of the nitric acid used, reduces the time of dissolution of uranium by a factor of about 100.

Specific embodiments of the present invention are given in the following examples:

Example I

A cylinder of metallic uranium 2¼ inches in diameter and 2¼ inches high was provided with a hole ½ inch in diameter bored along its cylindrical axis to a distance equal to ⅔ of its height. The innermost half of the hole was filled with discs of uranium metal approximately 1/16 of an inch thick and the remaining half of the hole was filled with a plug of solid uranium metal. The whole assembly was placed in a thin iron can and subjected to bombardment by thermal neutrons produced in a self-sustaining neutron chain reacting system, capable of establishing a neutron intensity of about 10^{15} to 10^{20} neutrons per second, for a period of 90 days to produce substantial quantities of various products including transuranic elements and also radioactive fission products formed by nuclear fission of uranium atoms. These radioactive fission products are valuable as radiation sources, tracers, and for study of half-lives.

After the neutron bombardment, the discs containing about 0.02 percent by weight of fission products and 0.02 percent by weight of element 94^{239} having a total weight of 28 grams being located in the middle of the uranium cylinder were dissolved in 60 cc. of 16 N nitric acid to which was added 4 cc. of concentrated perchloric acid ($HClO_4$). The addition of the perchloric acid shortened the time of dissolution of the uranium metal discs by a factor of about 100 compared to a similar dissolving procedure performed without the use of perchloric acid.

Example II

40 grams of uranium metal were treated with 60 cc. of 65 percent (by weight) nitric acid. After 1.5 hours only about three-fourths of the metal had gone into solution.

The same experiment was repeated using the same quantities of material with the addition of 1.5 cc. of 60 percent (by weight) perchloric acid. The uranium entirely dissolved in 0.5 minute.

The addition of perchloric acid has such a large effect on the speed of reaction that it is generally necessary to control the concentration of perchloric acid to avoid having the reaction occur with explosive violence. Practically any concentration of nitric acid can be used, but it is desirable to avoid any concentration of perchloric acid greater than 10 percent by weight of the total quantity of acid in order to avoid danger of explosion.

It will be understood that whenever the term perchloric acid is used, it includes the use of an equivalent quantity of its salts that are metathesized in the presence of nitric acid, so as to form perchloric acid in situ. Thus for example: sodium perchlorate, $NaClO_4$; potassium perchlorate, $KClO_4$, or barium perchlorate, $Ba(ClO_4)_2$ may be used.

Although the present invention has been described with particular reference to the specific details of certain em-

bodiments thereof, it is not intended that such details shall be regarded as limitations upon the scope of the invention except insofar as included in the accompanying claims.

I claim:

1. A process of dissolving uranium metal which comprises treating uranium metal with a mixture of nitric acid and one of the group consisting of perchloric acid and salts that react with nitric acid to form perchloric acid.

2. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and one of the group consisting of perchloric acid and salts that react with nitric acid to form perchloric acid.

3. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and perchloric acid.

4. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and sodium perchlorate.

5. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and potassium perchlorate.

6. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and barium perchlorate.

7. A process of dissolving uranium metal which comprises treating uranium metal with a mixture of nitric acid and an amount of one of the group consisting of perchloric acid and salts that react with nitric acid to form perchloric acid equivalent on a perchloric acid basis to not more than 10 percent by weight of the total acid present.

8. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and an amount of one of the group consisting of perchloric acid and salts that react with nitric acid to form perchloric acid equivalent on a perchloric acid basis to not more than 10 percent by weight of total acid present.

9. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and not more than 10 percent by weight of total acid present of perchloric acid.

10. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and an amount of sodium perchlorate equivalent on a perchloric acid basis to not more than 10 percent by weight of total acid present.

11. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and an amount of potassium perchlorate equivalent on a perchloric acid basis to not more than 10 percent by weight of total acid present.

12. A process of dissolving uranium metal containing products of neutron bombardment which comprises treating said uranium metal with a mixture of nitric acid and an amount of barium perchlorate equivalent on a perchloric acid basis to not more than 10 percent by weight of total acid present.

13. The process which comprises dissolving uranium metal that has been subjected to neutron bombardment in a mixture of nitric acid and not more than about 10 percent of perchloric acid and then precipitating and recovering the short-lived radioactive fission products.

14. A process of dissolving uranium metal which comprises treating metallic uranium with a mixture of nitric and perchloric acids.

No references cited.