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[54] **METHOD FOR REMOVAL OF PLUTONIUM IMPURITY FROM AMERICIUM OXIDES AND FLUORIDES**

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[58] **Field of Search** .... 423/3, 19, 250, 251; 75/84.1 R

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[57] **ABSTRACT**

Method for removal of plutonium impurity from americium oxides and fluorides. AmF_4 is not further oxidized to AmF_6 by the application of O_2F at room temperature, while plutonium compounds present in the americium sample are fluorinated to volatile PuF_6, which can readily be separated therefrom, leaving the purified americium oxides and/or fluorides as the solid tetrafluoride.

5 Claims, No Drawings

METHOD FOR REMOVAL OF PLUTONIUM IMPURITY FROM AMERICIUM OXIDES AND FLUORIDES

This invention is the result of a contract with the Department of Energy (Contract No. W-7405-ENG-36).

BACKGROUND OF THE INVENTION

The present invention relates generally to a method of selective fluorination of actinide species, and more particularly to the removal of plutonium impurity from americium using fluorine gas followed by O_2F.

Americium is a by product of plutonium production, and is currently extracted from solutions of plutonium feedstock material by a peroxide precipitation process. However, the americium recovered by this process contains between approximately 1 and 20% of plutonium. Excess peroxide is subsequently neutralized by addition of caustic, forming thereby polyhydroxides of americium. Further purification is required to reduce the plutonium impurity to an acceptable 5000 ppm level. This is accomplished by dissolving the impure americium hydroxides in nitric acid an passing the resulting solution through ion-exchange material wherein the plutonium is preferentially fixed while the americium remains in solution, thereby passing through the exchange material. To produce metallic americium, the processed americium solution is mixed with oxalic acid to precipitate the americium as americium oxalate which is then calcined at about 450° C. to yield AmO_2. The oxide material may then be reduced to the metal. Of interest is a less complicated procedure for further reducing the plutonium content of americium.

Accordingly, it is an object of the present invention to provide a method for the reduction of the plutonium impurity content of americium.

Another object of the invention is to provide a nonaqueous procedure for reducing the plutonium impurity in americium.

Additional objects, advantages and novel features of the invention will be set forth in part in the description which follows, and in part will become apparent to those skilled in the art upon examination of the following or may be learned by practice of the invention. The objects and advantages of the invention may be realized and attained by means of the instrumentalities and combinations particularly pointed out in the appended claims.

SUMMARY OF THE INVENTION

To achieve the foregoing and other objects, and in accordance with the purposes of the present invention, as embodied and broadly described herein, the method of this invention may include reacting americium oxides containing plutonium impurity with fluorine gas to produce AmF_4, reacting the resulting impure AmF_4 with O_2F to produce PuF_6 while leaving the AmF_4 unreacted further, and separating the resulting PuF_6 from the AmF_4. Preferably, the step of reacting the impure americium oxides with fluorine gas is performed at substantially room temperature.

Benefits and advantages of the subject invention include the ability to reduce the plutonium impurity level in americium to acceptable concentrations in a single, simple nonaqueous step.

DETAILED DESCRIPTION OF THE INVENTION

Reference will now be made in detail to the present preferred embodiment of the invention. Briefly, our invention includes a method for removing the plutonium impurity in americium. The subject method takes advantage of the fact that AmF_4 is not further oxidized by O_2F to AmF_6, while plutonium compounds present (tetrafluorides, oxyfluorides and oxides) are converted to PuF_6. Theoretically, it should be possible to reduce the plutonium level to less than 100 ppm from about 1–20 impurity.

Having generally described the present invention, the following specific examples are given as a further illustration thereof.

EXAMPLE I

Fluorination of AmF_4 using both F_2 and O_2F:

A 0.558 g sample of pure americium oxide was placed in a passivated, sintered monel filter cup having a 60 micron pore size and designed such that all gases passed therethrough come into contact with the sample located therein. The conversion of AmO_2 to AmF_4 was effectuated using a mixture of 100 torr of fluorine and 100 torr of argon, which was permitted to flow over the sample for approximately 12 hours, followed by 600 torr of fluorine circulated over the sample for an additional approximately 5 hours. The reaction was followed using neutron counting techniques, the neutron count rate increasing as the quantity of AmF_4 increases. At the termination of the reaction, the neutron count rate was observed to have doubled and the sample weight increased by 35.4 mg, about 40% of the expected weight gain. Some impurities were observed in subsequent FTIR scans and are believed to arise from residual nitrates present in the AmO_2.

A 0.2988 g sample ofAmF_4, prepared as detailed in the preceding paragraph, was placed in the filter cup. A flowing (18 std-1/min), room temperature mixture of 300 torr of oxygen and 300 torr of fluorine was irradiated with a XeCl laser (308 nm, 20 Hz, 3–4 watts, 175–200 mJ/pulse) and the resulting mixture passed through the filter cup for approximately 1 hour and then into a cold trap. The residence time between the photolysis cell and the sample was between 500 and 700 ms. Uncondensed fluorine and oxygen were recirculated through the region of irradiation and the irradiated mixture continuously passed through the filter cup. No AmF_6 could be detected by FTIR measurements. An attempt to fluorinate a 0.5 g sample of AmF_4 in a sintered nickel filter cup by exposing the sample to 600 torr of F_2 irradiated for about 1 hour under the conditions set forth hereinabove did not produce detectable quantities of AmF_6.

EXAMPLE II

Fluorination of PuF_4 using both F_2 and O_2F:

Pu_4 was generated by laser photolysis of 1–2 torr of PuF_6 in 250 torr of argon. The PuF_4 was collected on a sintered nickel filter. Typical sample sizes were between 80 and 120 mg. Using similar conditions to that of the unsuccessful fluorination of AmF_4 described hereinabove, a 100 mg sample of PuF_4 was fluorinated using O_2F in 5000 to 7000 laser pulses. Similar fluorinations were performed in a sintered monel filter cup using PuF_4 derived from hydrofluorination of PuO_2 using HF at 600° C., and PuO_2 derived from a burned anode heel, both generating PuF_6.

EXAMPLE III

Fluorination of a mixture of AmO_2 and PuO_2:

A 0.5 g sample of a mixture of plutonium and americium oxides (0.386 g of AmO_2 and 0.114 g of PuO_2) was pretreated with 300 torr of F_2 to convert the AmO_2 to AmF_4. Ater 1 hour of treatment with flowing 0_2F generated as described hereinabove, 0.038 g of PuF_6 was produced. After another hour, 0.040 g additional PuF_6 was generated. This latter quantity represented 46.5% removal of the plutonium present in the mixture.

EXAMPLE IV

Fluorination of a mixture of AmO_2 and PuO_2:

A 0.5 g sample of a mixture of plutonium and americium oxides (0.454 g of AmO_2 and 0.046 g of PuO_2) was pretreated with 300 torr of F_2 to convert the AmO_2 to AmF_4. After 1 hour of treatment with flowing O_2F generated as described hereinabove, 0.011 g of PuF_6 was produced. After another hour, 0.006 g additional PuF_6 was generated. This latter quantity represented 23.9% removal of the plutonium present in the mixture.

In summary, AmF_4 does not oxidize to AmF_6 under the conditions described, while PuF_4 and PuO_2 oxidize readily thereto.

The foregoing description of a preferred embodiment of the present invention has been presented for purposes of illustration and description. It is not intended to be exhaustive or to limit the invention to the precise form disclosed, and obviously many modifications and variations are possible in light of the above teaching. The embodiment was chosen and described in order to best explain the principles of the invention and its practical application to thereby enable others skilled in the art to best utilize the invention in various embodiments and with various modifications as are suited to the particular use contemplated. It is intended that the scope of the invention be defined by the claims appended hereto.

What is claimed is:

1. A method for the removal of plutonium impurity from oxides and/or fluorides of americium which comprises the steps of:
   a. reacting impure americium oxides containing plutonium impurity with fluorine gas to produce impure AmF_4;
   b. reacting the resulting impure AmF_4 with O_2F to produce PuF_6; and
   c. separating the PuF_6 from the AmF_4.

2. The method as described in claim 1, wherein said step of reacting said impure americium oxides with fluorine gas is performed at substantially room temperature and wherein about 300 torr of fluorine gas is employed.

3. The method as described in claim 1, wherein said step of reacting said impure AmF_4 with O_2F includes flowing O_2F prepared by irradiating a flowing mixture of oxygen gas and fluorine gas with ultraviolet radiation in the vicinity of the impure AmF_4 and wherein the O_2F prepared thereby is flowed over the impure AmF_4.

4. The method as described in claim 1, further comprising pyrohydrolyzing said AmF_4 to produce AmO_2 after said step of separating the PuF_6 therefrom.

5. The method as described in claim 1, further comprising reducing said AmF_4 to produce metallic americium after said step of separating PuF_6 therefrom.

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