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REPORT OF INVENTION

INCREASING AMOUNTS OF Pu-241 ISOTOPE

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4 pages.

REPORT OF INVENTION
INCREASING AMOUNTS OF Pu-241 ISOTOPE

by

L. W. Lang

IRRADIATION PROCESSING DEPARTMENT

January 28, 1964

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REPORT OF INVENTION

INCREASING AMOUNTS OF Pu-241 ISOTOPE

Uranium fueled reactors produce plutonium-239 as a product of the irradiation. Depending on the length of irradiation and the fuel arrangement, the plutonium-239 itself is either fissioned by thermal neutrons or absorbs thermal neutrons to produce Pu-240. With increasing exposures, the Pu-240, in turn, absorbs neutrons to form Pu-241. Pu-241 is a uniquely useful isotope from several aspects. It is a good nuclear fuel having a smaller absorption-to-fission cross section ratio than Pu-239. Pu-241 is unstable and decays with a 13-year half-life to americium-241. Americium-241 is a valuable isotope that can be readily converted by thermal reactor irradiation to curium-242 and/or its decay product, plutonium-238.

Americium-241 and the products that can be obtained from its irradiations are more valuable than the neutrons secured from the atoms by using Pu-241 as a reactor fuel. However, because of the large thermal neutron cross section of the Pu-241 isotope, it is difficult to obtain appreciable percentages in a Pu isotopic mixture. Despite the fact that nearly one-third of the atoms of Pu-239 are ultimately converted to Pu-241 with continued irradiation, only 5 to 10 percent of them will exist in the fuel at any specific time. The balance will have fissioned or have been converted into Pu-242.

This invention report suggests a method for drastically increasing the amount of Pu-241 in isotopic mixtures of plutonium. It is felt that with process experience, as much as 70 percent or more of the potential Pu-241 atoms can be concentrated in the fuel at one time. Such a concentration step would double the amount of production obtainable from Pu-241 by nuclear decay.

The process to concentrate the Pu-241 consists of two basic steps:

1. Irradiate plutonium consisting largely of Pu-239 isotope until the Pu-239 has largely been converted to Pu-240 and 241 by thermal neutron fission and absorption events. Most of the fuel value has been taken advantage of at this point. The isotopic mixture will be largely Pu-240 but contain smaller percentages of 241 and 239. The irradiation is terminated at this point and the products are separated.
2. The depleted plutonium isotopes from the first irradiation are refabricated for a second irradiation with a thermal neutron absorber surrounding the depleted plutonium isotope. This element is again irradiated, preferably in an epithermal neutron flux with a peak energy slightly above 1 ev. The Pu-240 has a huge resonant cross section at the 1 ev level and will be most rapidly converted to Pu-241 by neutrons of this energy. The Pu-241 has a minimum reactor cross section for neutrons of 1 ev and thus will not be destroyed faster than created if the atoms are protected from the thermal neutrons by a cutoff absorber.

Specific Recommendations for Thermal Neutron Cutoff

Cadmium appears to be the most efficient absorber in that its cross section rapidly rises with neutron energies below 1 ev. The Pu-239 cross section, likewise, increases

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rapidly just below the 1 ev range. The fact that this cross section is complimentary to a 1 ev cutoff suggests that possibly the first and second irradiation could be carried on in the same fuel element. Thus, if the two grades of plutonium could be segregated, the Pu-239 could be used on the outside of the fuel element to absorb the majority of neutrons below 1 ev and the Pu-239 on the inside would see only epithermal flux of 1 ev or higher.

Xenon-135 could conceivably serve as an extremely efficient absorber if an exotic design could be achieved in which the xenon were allowed to build up in a cavity between the highly depleted plutonium and a thermal flux.

Flux Spectrum

The initial irradiation of the plutonium elements would be accomplished in a thermal neutron reactor in which the rate of heat generation would be controlling as to the amount of higher isotope produced. A net surplus of neutrons would accrue to the irradiation. Thus, the plutonium will have a tangible fuel value. The isotopic composition at the conclusion of the irradiation (after most of the Pu-239 has been burned out) will be high enough to warrant setting aside and milking it for americium-241 as it decays from the Pu-241. However, a second irradiation could be undertaken at any time to transform the bulk of the remaining Pu to Pu-241.

The second irradiation would ideally be accomplished in an epithermal reactor or in the blanket of a fast reactor in which the neutron energy peak occurs slightly above 1 ev energy. The cadmium cutoff would appear to be most appropriate if such a flux were available for the irradiation. If, however, epithermal or fast reactor fluxes are not available to carry out the irradiation, it could be done in a thermal reactor but at some loss of neutrons. This loss of neutrons would be minimized in a thermal reactor if the combination plutonium element described above were considered. In such an element, the Pu-239 would act at least as a partial filter for the epithermal cadmium cutoff case. Some minor losses of Pu-241 will accrue to resonant peaks above 1 ev neutron energy levels, but this loss should be small as compared to the Pu-241 produced by the large resonant cross section of Pu-240.

SUMMARY

Americium-241 secured from the decay of Pu-241 is worth considerably more than its fuel value since it can be readily converted to curium-242 or Pu-238--desired thermal isotope power sources. The amount of Pu-241 can be maximized by the process described in this document at the same time that it is being used as the reactor fuel. Up to 70 percent of its ultimate fuel value may be obtained. The Pu-241 that can be produced from a given amount of plutonium can be increased by a factor of two or three by using a thermal neutron cutoff in a flux peaking slightly above 1 ev energy levels.

Claims

1. A method of converting plutonium of lower isotope content to a mixture of higher isotope that maximizes the amount of Pu-241 at a level two to three times that presently obtainable by thermal neutron irradiations.

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2. A neutron cutoff can be used with highly depleted plutonium isotopes to prevent destruction of the Pu-241 by thermal neutron absorption.
3. The use of cadmium and Pu-239 as thermal neutron shields for plutonium isotopes.
4. The irradiation of plutonium isotopes in an epithermal flux or in the flux of a fast reactor blanket to maximize Pu-241.
5. The use of reactor-grade Pu as an external cylinder for a core of highly depleted plutonium to maximize the amount of Pu-241 obtainable in a thermal reactor.
6. The use of a combination cutoff, such as an outer cylinder containing Pu-239, an inner cadmium cylinder and a core of highly depleted plutonium isotopes, to maximize the amount of Pu-241 obtainable in a thermal reactor.
7. The use of cutoffs in an epithermal flux to prevent degradation of Pu-241 built into highly depleted plutonium isotopes.

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