RADIATION FROM PLUTONIUM-238 USED IN SPACE APPLICATIONS

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Plutonium-238 is an alpha emitter with an 87 year half-life that is produced by neutron irradiation of neptunium-237 in a reactor. Plutonium-238 has been used in all applications of space electric power using isotopes as the energy source. These applications include the SNAP-3, 9, 19, and 27 units. The SNAP-3 and SNAP-9 units were used in Navy TRANSIT satellites while the SNAP-19 was used for auxiliary power aboard a NASA NIMBUS and the SNAP-27 provided all the power for the ALSEP package left on the moon's surface by the Apollo 12 and planned subsequent Apollo landings.

The principal mode of the nuclear decay of plutonium-238 is by alpha particle emission at a rate of 17 curies per gram. Gamma radiation also present in the nuclear fuel arises primarily from the nuclear de-excitation of daughter nuclei as a result of the alpha decay of plutonium-238 and reactor produced impurities. The gamma radiation associated with the alpha decay of plutonium-238 ranges in energy from 0.044 to 1.085 MeV. Thallium-208, a subsequent decay product of the reactor produced impurity, plutonium-236, emits 0.583 and 2.6 MeV gamma-rays at a significant rate if the initial plutonium-236 content is of the order of one part per million of total plutonium.

Plutonium-238 has a spontaneous fission half-life of 4.8×10^{10} years. Neutrons associated with this spontaneous fission are emitted at a rate of 2.8×10^3 neutrons per second per gram. Since the space fuel form of plutonium-238 is the oxide pressed into a cermet with molybdenum, a contribution to the neutron emission rate arises from (α, n) reactions with 0-17 and 0-18 which occur in natural oxygen and contribute approximately 2×10^4 neutrons per second per gram of plutonium-238. This contribution can be controlled by 0-16 enrichment in the fuel. Other (α, n) reactions with low atomic number impurities (principally fluorine) bring the total neutron emission rate from the plutonia molybdenum cermet to near 3×10^4 neutrons per second per gram of plutonium-238. This contribution can, also, be reduced by proper fuel production techniques.

Plutonium-238 is an alpha emitter with a half life of 87.80 ± 0.02 years. [2] This isotope is also a neutron emitter due to a spontaneous fission half life of $4.8 \times 10^{10} \pm 0.2$ years. [3] There are gamma rays emitted that are associated primarily with the alpha particle decay. 99.9% of the gamma rays (5.5×10^8 photons per second per watt) are of less than 210 kev energy and rather easily shielded. [4] The more penetrating radiation is due to gamma rays of 766 kev present in an abundance of 3.7×10^5 photons per second per watt of Pu-238. [4] Other papers in this session deal with specific radiation properties of plutonium-238 as it is used in heat source applications. The purpose of this paper is to discuss the factors that affect and sometimes control the radiation characteristics of plutonium fuel used in space applications. It is intended to explain why it is difficult to produce a plutonium space fuel that has radiation characteristics of pure plutonium-238. In general, this difficulty stems from the fact that reactor production of Pu-238 and subsequent processing techniques to fuel form strongly influence the radiation properties of the space fuel form. The space heat source plutonium material [2] is 80% Pu-238, 16% Pu-239, 3% Pu-240, 1% Pu-241 and Pu-242 and 1×10^{-4}% (1 ppm) Pu-236. There is less than 0.5 percent total other actinides. This particular composition is obtained by mixing 84% Pu-238 from Np-237 neutron irradiation with 40% Pu-238 from reactor fuel reprocessing.

Reactor Production

Plutonium-238 is AEC reactor produced primarily by the neutron irradiation of Np-237 which in turn is produced in a reactor through the burnup of uranium-235. Plutonium-238 is also produced in the reactor fuel elements.

The reactor irradiation of neptunium results primarily in neutron capture by the Np-237. Subsequent beta particle decay of neptunium-238 produces 84% Pu-238. Neutron capture by Pu-238 while it is in the reactor produces the heavier plutonium isotopes. Forty percent Pu-238 with the remainder being primarily Pu-239 is obtained by periodic reprocessing of reactor fuel, a mixture of U-235 and U-238. Since the route to Pu-239 production is more straightforward than production of Pu-238, the percentage of Pu-239 is considerably higher.

To a much lesser but still significant extent, Np-237 undergoes nuclear reactions to produce Pu-236. These reactions are neutron capture with two neutron emissions to form Np-236 which beta decays into Pu-236 and gamma ray capture by Np-237 followed by a neutron emission and a beta decay to form Pu-236. Pu-236, with a half life of 2.8 years decays to U-232, an alpha emitter with a 72 year half life. U-232 then reaches secular equilibrium with its daughter products. One of these daughter products is the 2.6 MEV gamma emitter thallium-208 which grows in at a rate such that its maximum abundance is achieved in 18 years.

In the separation of plutonium from neptunium, Pu-236 will naturally follow the plutonium stream and become an undesirable contaminant in the plutonium product. The space fuel currently being produced contains on the order of 1 ppm Pu-236. As the U-232 and, hence, Tl-208 grow in, the gamma radiation from the fuel will increase so that after five years Tl-208 contributes equally to the total hard gamma radiation from Pu-238 space fuel. After five years, the hard gamma radiation is due primarily to the thallium-208.

The 84% plutonium-238 from reactor irradiation of neptunium contains slightly less than 1 ppm Pu-236. The 40% Pu-238 material contains more than 1 ppm Pu-236. The space fuel obtained from mixing these two products results in 80% Pu-238 and approximately 1 ppm Pu-236.

There are several actions that could be taken to reduce the level of the Pu-236 impurity. Each of these affects availability or cost or else requires time. Allowing the Pu-236 to decay, followed by chemical processing to remove the daughter products is an approach but since the half life of Pu-236 is 2.8 years, this method requires the storage of material for several years. This may be feasible at some future time when there is an inventory of plutonium material, but at this time it is used almost as fast as it is produced. Another approach would be to use only the plutonium material made from irradiation of Np-237. However, 84% Pu-238 is mixed with 40% Pu-238 in order to increase the availability of Pu-238 as an 80% material. Again, at some future date when the availability of Pu-238 is considerably greater than it is now, it should be possible to use only the higher percentage plutonium-238.

Fuel Form and Chemical Processing

The current space fuel form is a cermet consisting of 83% plutonium oxide and 17% molybdenum. This fuel form will be the third form of plutonium used on space missions. Plutonium metal was the first form, used in the early 1960's when the means of disposing of the plutonium was high altitude burnup when the heat source reentered the earth's atmosphere. This fuel form did not have extraneous radiation unless light element impurities were present, in which case neutron emission could increase due to alpha capture-neutron emission (α, n) reactions between the alpha particles from Pu-238 and the impurities. The melting point of plutonium metal and the need to have a more chemically inert form of plutonium limited the use of the metal which was replaced by plutonium oxide in the form of small spherical particles (microspheres). These microspheres were made by passing plutonium oxide powder through a plasma torch, a process which produced extremely inert particles. However, these oxide particles increased the fuel neutron radiation from approximately 4000 neutrons per second per gram of Pu-238 in the metal fuel, to over 20,000 neutrons per second per gram of Pu-238. The increase in neutron activity was due to (α, n) reactions on natural oxygen. Natural oxygen consists of the O-16, O-17, and O-18 isotopes. Only the O-17 and O-18 undergo (α, n) reactions at the alpha particle energies involved. Separation of O-16 from natural oxygen have been successful to a degree and oxygen enriched in O-16 has been used to exchange the natural oxygen in Pu oxide. Attempts were made to exchange the oxygen in microspheres with oxygen-16 but these attempts were not successful, quite possibly due to the impervious,

glassy surface of the microsphere.

Microspheres are being used in the SNAP-27 (ALSEP) missions but temperatures to which the fuel was exposed under normal generator operating conditions has continued to rise. Further, it was determined that the heat source should be designed to reenter from space and impact the earth's surface without breaking open. Through this, the fuel from should remain relatively unchanged. From these criteria evolved the solid fuel, plutonia molybdenum cermet. Molybdenum was chosen since it was found to be the most compatible metal with plutonium oxide at temperatures above $1000^\circ C$. Fabrication procedures for this cermet consist of essentially two steps, coating of oxide particles with molybdenum and hot pressing the coated particles into a disc shape of greater than ninety percent theoretical density. The oxide particles are coated by the chemical vapor deposition of MoF_6 in a fluidized bed system. MoF_6 was chosen because of the control it gives over the quantity of molybdenum coated onto the particles. This fuel form has the highest neutron radiation of those discussed here. This is due to (α, n) reactions with fluorine. Fluorine has a large cross section for this reaction. Steps have been taken to limit the neutron radiation to less than 30,000 neutrons per second per gram. This level is achieved by a combination of exchanging the PuO_2 particles with 0-16 prior to their being coated and bake-out of the discs following fabrication. Without these steps, the neutron radiation would be at least ten to fifteen thousand neutrons per second per gram greater.

The fluorine impurity could be eliminated by using another method for introducing the molybdenum into the fuel form. There are alternative methods for carrying out this step.

Coating with molybdenum chloride is one method that is being developed. So far this method has been more time consuming and the molybdenum coat thickness more difficult to control. However, this method is being used to produce a limited quantity of fuel for thermoelectric generators to be used on a NASA Pioneer mission.

It should become increasingly less difficult to obtain plutonium fuel that is very low in or completely free from radiation producing impurities. As commercial power reactors become more plentiful, more Np-237 becomes available and the production of Pu-238 without blending with lower Pu-238 isotopic ratio material becomes more feasible. Curium-242, another by-product of commerical power reactor operation, decays into Pu-238. Recovery of Cm-242, storing it, and periodic extraction of the Pu-238 should produce essentially 100% Pu-238. Unfortunately Cm-242 will be in very short supply until the late 1970's or early 1980's and Pu-238 from this source will be coveted by those developing nuclear power for medical applications. In addition to the chloride process, there are other possible ways to reduce extraneous neutron levels. These include powder metallurgy which involves mixing very fine molybdenum powder with 0-16 enriched PuO_2 followed by hot pressing to the final fuel form.

In terms of timing, it is difficult to predict the exact point in time when radiation will be at a given level. However, there are solutions for ridding the space fuel of both the gamma and neutron emitting impurities and certainly within the next few years a fuel form should be available that will satisfy both the generator designers and those who are responsible for determining the radiation levels in space.

REFERENCES

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