Gallium in Weapons-Grade Plutonium and MOX Fuel Fabrication

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Gallium is used as an alloying element in the manufacture of plutonium pits in concentrations of up to 1 percent. (This information about gallium was declassified about one year ago.) Gallium at such concentrations presents various issues for MOX fuel fabrication and use. Therefore its concentration must be greatly reduced if the MOX option for plutonium disposition is pursued.

MOX fuel is essentially a ceramic material, prepared by sintering oxides of uranium and plutonium, which are initially both in the form of fine powders. At high concentrations, gallium affects the sintering behavior of the ceramic. Since plutonium pits do not all have the same concentration of gallium, the sintering process parameters would have to be adjusted as the gallium concentration changed (undesirable in an industrial-scale operation) unless the gallium was reduced to an acceptable level prior to fabrication.

In addition, there may be issues with using MOX fuel with excessive gallium concentration as a reactor fuel. While gallium would not interfere with the chain reaction (it is not a good neutron absorber), gallium metal chemically attacks zirconium. The tubes into which fuel is inserted are made of a zirconium alloy. The presence of excessive gallium in spent MOX fuel could therefore cause its deterioration and hence possibly cause waste management problems. There may also be other problems with the presence of large amounts of gallium, but it has not yet been established whether these are significant concerns. For instance, gallium may affect fission product migration in spent fuel.

Technologies for gallium removal and plutonium oxide production

The currently available fully developed technology for gallium removal and plutonium oxide production is an aqueous process which results in the generation of large quantities of liquid radioactive wastes. For this reason, it would be highly desirable to avoid using the aqueous process and in its stead use the dry processes being developed at the Los Alamos and Livermore National Laboratories.

Two approaches for converting plutonium pits into oxide are being investigated at Los Alamos and Lawrence Livermore National Laboratories. One is a process that converts plutonium metal into a hydride form, and then the hydride is oxidized. Another process would convert plutonium metal into a hydride, the hydride into a nitride (PuN), and thence to an oxide. After either process, gallium is also in oxide form, Ga2O3. (To remove plutonium and provide a metal product, the hydride is simply heated to drive off hydrogen, leaving a plutonium metal ingot.) These approaches are part of the “ARIES” process for pit disassembly and conversion.

In each case the gallium would be removed after plutonium oxide has been made. The gallium is driven out by reduction and conversion to a sub-oxide form (Ga2O) in an atmosphere of argon with 6 percent hydrogen. The sub-oxide of gallium rapidly volatilizes from the plutonium-gallium mix at 1100oC.

(Gallium volatilizes at lower temperatures, but at slower rates.) This process, which has been tested at the laboratory scale, gets gallium down from about 1 percent to ~200 parts per million. MOX fuel, which typically contains about 5 percent plutonium, would therefore have a gallium content of ~10 parts per million. Current thinking in the MOX fuel fabrication industry in Europe is that this level of gallium content is probably acceptable because it is comparable to or beneath the level of other contaminants, now present in MOX fuel used in European reactors, that also might interact with cladding.

The hydride process for converting plutonium to a metal ingot is now standard in plutonium processing at Los Alamos and Livermore. However, the conversion process to oxide and the gallium removal processes are not yet fully developed. The main problem with oxide conversion is managing the release of heat in converting the hydride to oxide, while maintaining the oxidation rate high enough to provide good production throughput. If the hydride becomes too hot, some hydride will be converted to plutonium metal, forming “clinkers.” Similarly, in conversion of the nitride to oxide, overheating can cause formation of large, hard clumps of oxide. Engineering is underway to remove the excess heat efficiently, allowing shorter oxidation times and greater throughput.

Los Alamos and Oak Ridge National Laboratories are doing additional experiments with migration and diffusion of gallium in MOX to see if there is a need to remove gallium more completely. In that case, a backup process involving an aqueous technology (using nitric acid dissolution, ion exchange or solvent extraction to remove the gallium, and oxalate precipitation, for instance) would be used to process the plutonium dioxide that comes from the ARIES process.

No problems involving gallium that would affect plutonium vitrification have been identified. The matter is under study at Livermore National Laboratory, but no problems are anticipated at this time.

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