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THE UNITED STATES PIT DISASSEMBLY AND CONVERSION PROJECT-MEETING THE MOX FUEL SPECIFICATION

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SUMMARY

The United States is actively involved in demonstrating the disassembly of nuclear weapon pits to an unclassified form readied for disposition. The United States issued a record of decision through the Department of Energy's (DOE) Office of Fissile Material Disposition (OFMD) in January, 1997 that will use a hybrid option for the disposition of plutonium. The hybrid option disposes of surplus weapon's plutonium by burning it as mixed oxide (MOX) fuel to meet the spent fuel standard or putting it into a ceramic form-immobilization. The MOX option is the most likely path forward for plutonium that originated from nuclear weapon pits. Pits are the cores of a nuclear weapon that contain the fissile material. The United States demonstration line for pit disassembly and conversion is known as ARIES, the advanced recovery and integrated extraction system. The ARIES demonstration line is located at the Los Alamos National Laboratory and is being used to gather data in an integrated fashion of the technologies needed for pit disassembly and conversion for the purpose of supporting the design of the larger pit disassembly and conversion facility.

These activities include the following modules: pit bisection, hydride-dehydride, oxide conversion, canning, electrolytic decontamination, and nondestructive assay (NDA). Pit

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bisection swages a pit in half, which allows the plutonium to be processed effectively into a metal or oxide form. As an example of metal conversion, hydride-dehydride converts the pit plutonium metal to an unclassified metal button, which can be stored or subsequently converted to plutonium oxide. To convert the plutonium metal to an oxide the United States is investigating a number of options. The primary oxide conversion approach involves variations of combining plutonium hydriding and subsequent oxidation. Another approach is to simply oxidize the metal under controlled conditions-direct metal oxidation (DMO). To remove the gallium from the plutonium oxide, a thermal distillation approach is being used. These pyrochemical approaches will substantially reduce the wastes produced for oxide conversion of weapon plutonium, compared to traditional aqueous processing. The packaging of either the plutonium metal or oxide to long term storage criteria involves the canning and electrolytic decontamination modules. The NDA suite of instruments is then used to assay the material in the containers, which enables international verification without the need to open the containers and repackage them. All of these processes are state of the art and incorporate waste minimization, reductions in personnel exposure, a relatively small footprint, etc.

BACKGROUND

In September 1993 President Clinton of the United States issued a *Nonproliferation and Export Control Policy* in response to the growing threat of nuclear proliferation. Four months later, in January 1994, President Clinton and Russia's President Yeltsin issued a *Joint Statement Between the United States and Russia on Nonproliferation of Weapons of Mass Destruction and the Means of Their Delivery.* A little more than a year later, 1 March 1995, President Clinton announced that approximately 200 metric tons of US-origin weapons-usable fissile materials had been declared surplus to US defense needs. The Advanced Recovery and Integrated Extraction System (ARIES) Demonstration Program is one part of the scientific response to President Clinton's promise to reduce the nuclear weapons stockpile.

The goal of the disposition program is to remove and dispose of surplus fissile weapon material simultaneously from both the United States and Russian inventories, never again to be used in nuclear weapons. (This paper addresses only the disposition of plutonium as the fissile nuclear weapon material that is removed from a pit, the core of a nuclear weapon that contains the nuclear material.) Pit disassembly and conversion refers to removal of plutonium from the nuclear weapon pit and conversion to an unclassified form. This unclassified form enables verification by inspectors of other nations that results in irreversibility of the host country to reuse the nuclear material or for terrorist threats to gain access to the nuclear material. The subsequent plutonium disposition process, immobilization or MOX fuel, must allow for inspection all the way through its processing, use, and final end state. These international inspectors could be bilateral inspectors from Russia and the United States or others, such as the International Atomic Energy Agency (IAEA). ARIES is the United States pit disassembly and conversion demonstration line that converts the nuclear weapon pit to an unclassified form that can be inspected and used in subsequent disposition processes. As part of the ARIES Demonstration Line activities, meeting the MOX fuel feed specification for plutonium oxide is a critical part of the activity. ARIES is sponsored by the DOE OFMD.

ARIES PROCESS DESCRIPTIONS

The ARIES demonstration line for pit disassembly and conversion is modular in design to offer credible scaling, incorporation of modifications or new concepts, and future transportability and exportability capabilities. ARIES will develop and demonstrate the following modular elements:

- A pit bisection module used to cut open and physically separate the various components of the weapon pits;

- A metal to metal conversion module, hydride-dehydride recycle, to remove the plutonium from the weapon component substrate surfaces and cast it as a metal ingot, and also a recasting module;

- A parallel metal to oxide conversion module, hydride-oxide (HYDOX) and DMO, to process plutonium metal from pits or from metal ingots into a form suitable for the fabrication of mixed oxide fuel;

- A canning module that results in a double containment, long term storage package of either the plutonium metal or oxide that can be inspected by international safeguards;

- An electrolytic decontamination module to decontaminate the outside surfaces of the material containers for removal from the plutonium glovebox environment; and

- A NDA/instrument support module for analyzing the sealed storage cans to provide accountability and process control of all the special nuclear material and waste items produced by ARIES.

In addition to these modules, other tasks are important to the demonstration for meeting the MOX fuel feed specification. These activities include a process to remove gallium, particle size analysis, and analytic measurements to determine that impurities in the plutonium oxide are acceptable.

The objective of ARIES is to demonstrate feasibility of a process for disassembly, extraction, and conversion of plutonium from weapons components into forms suitable for disposition and /or storage while emphasizing pollution prevention. Currently, no integrated system exists for the complete dismantlement and recovery of nuclear material from different weapons' designs. A second objective is to develop and demonstrate feasibility of the ARIES technologies that could be used by both the US and Russia.

PROCESSES TO MEET THE MOX FUEL FEED SPECIFICATION

A MOX fuel feed specification was released as a guideline to potential bidders for the United States MOX fabrication request for proposals, June 1998. This specification was based on the ASTM specification and a general commercial specification. This specification includes particle size (95% < 44 microns and 100% < 100 microns), batch size (50 kg preferred), containerization of the oxide (stainless steel-hermetically sealed), impurities (See Table 1), etc. However, the main concern for fuel fabricators has been expressed to be consistent feed; thus, as an example the particle size and surface area are to fall within a certain range. A goal of the ARIES demonstration is to verify that pyrochemically produced plutonium oxide will meet this specification.

OXIDATION PROCESS

The oxidation process converts plutonium metal to plutonium oxide, which is used in the disposition options immobilization and MOX fuel. It has three criteria for becoming a successful process. The first involves instituting a process that will have an acceptable safety margin. The second is a process that will convert the metal to oxide in a timely fashion, adequate throughput rate. And the third criteria, is the production of acceptable feed for the use of the oxide in mixed oxide fuel. To reiterate, the preferred disposition option for clean weapons grade plutonium is to burn it as MOX fuel. For MOX fuel, the particle size and surface area are two of the most stringent characteristics. Currently, the pyrochemical process being studied most intensely for ARIES is the conversion of metal to oxide by hydriding, followed by nitriding, followed by oxidation, HYDOX. This process has shown promise from experiments done at LLNL, but further studies are required.

An alternative process that is also being studied to make plutonium oxide is to do DMO. This process has been used traditionally throughout the DOE complex. However, for the application of producing suitable MOX fuel, process control of the oxidation process to achieve a specific particle size is paramount. More demonstrations of this process are planned.

GALLIUM REMOVAL

Gallium was a necessary additive to weapon plutonium during fabrication of pits. However, its presence in plutonium oxide bound for MOX fuel is a topic of much debate and concern. There is little doubt that gallium levels in weapon derived plutonium oxide are unacceptable for fabrication of MOX fuel. As a result, a pyrochemical process for the removal of gallium is under development at Los Alamos. This process, known as TIGR or Thermally-Induced Gallium Removal, exploits the volatility of gallium sub-oxide, Ga_2O. TIGR is a simple chemical process whereby plutonium/gallium oxide is heated in a flow of reducing gas, H_2. During this process, the gallium oxide is reduced to gallium sub-oxide, which volatilizes into the gas stream and is carried away.

$$Ga_2O_3(s) + 2H_2(g) \rightarrow Ga_2O(g) + 2H_2O(g)$$

Initially, gallium levels of about 200 ppm in the PuO_2 were achieved using this process. However, additional research has resulted in gallium levels as low as 30 ppm. This results in a gallium concentration of 1.5 ppm in MOX fuel because PuO_2 typically comprises only 5% of the MOX fuel. At this level, it is unlikely that gallium will prove to be a problem in thermal reactor MOX fuel. Ongoing development activities will strive to lower the gallium content even further. In addition, materials compatibility studies are being conducted in support of TIGR design efforts. The goal of this work is to determine and /or develop materials of construction which can withstand the very aggressive TIGR environment (> 1000 °C, H_2, Ga_2O). Research directed towards complete and efficient collection of gallium is ongoing as well.

CANNING MODULE

The canning system receives the plutonium oxide and places it into a convenience container, which enables transport of the oxide through the glovebox line. The limits for metal and oxide are 4.4 kg and 5.0 kg, respectively, which were set in the DOE's Long Term Storage Standard (DOE-STD-3013-96). The convenience container, a food pack can, is placed into a material container. With the convenience container inside, the material container is hermetically sealed by welding and tested for leaks based on the 3013 standard. Each container is labeled for identification. The material container is removed from the plutonium glovebox line by the use of electrolytic decontamination. Once outside the glovebox, the material container is placed into the boundary container, which is also hermetically sealed by welding and also leak tested. This two-container package meets the criteria for long term storage of metal or oxide as described under the constraints of the 3013 standard. Packaging to the 3013-96 standard has been done at Los Alamos National Laboratory since the standard was released. The ARIES long term storage package has completed the required testing and qualifications. There is a large number of engineering design requirements that went into this package's development including the following: cost, ease of manufacture, ease of welding, weight, ergonomics, potential container reuse, height, diameter, robotics handling ability, strength, etc. The electrolytic decontamination of material containers was demonstrated in December 1995.

The DOE 3013 standard for long term storage has a loss on ignition (LOI) < 0.5 % of hydrogenous materials. As part of the demonstration of the LOI, the plutonium oxide is calcined at 950 °C for two hours. The ARIES oxidation furnaces are capable of performing this calcination step. However, the plutonium oxide product is sintered at these temperatures and studies are being done to determine the effect of meeting the LOI criteria on the grinding of the plutonium oxide.

RESULTS

The ARIES demonstration line integrates many new technologies to reduce the waste stream from the activities needed for pit disassembly and conversion and will be in full operation during the summer of 1998. The data gathered from the demonstration line will enable proper design of the United States' full scale pit disassembly and conversion facility. Currently, the Title II design of this facility is scheduled to begin in October 1999. Moreover, as the United States and Russia engage in the disposition of the plutonium from nuclear weapons, activities with our Russian counterparts may also include the use of some of the ARIES technologies, which will also reduce the waste streams generated in Russia.

As part of the complete disposition of plutonium, plutonium oxide is to be burned in nuclear reactors as MOX. The ability to meet the MOX fuel specification has been demonstrated by the oxide conversion processes, gallium removal, and container packaging. Plutonium oxide produced by the ARIES processes has already been used to fabricate fuel for reactors. This fuel is now being tested. Further demonstrations of these processes and manufacture of the oxide into fuel will continue as part of the United States effort for plutonium disposition.

Table 1. Various impurity specifications in mg/gm Pu for MOX Fuel.

Impurities	ASTM	Commercial Range	Draft Specification
Ag		50 - 100	100
Al		100 - 150	150
B	3	1 - 3	3 -10
Be		100	100
Bi		10 - 100	100
C	200	150 - 500	200-500
Ca		150 - 250	250-500
Cd	3	3 - 100	10
Cl	300	100 - (+Fl < 250)	(+Fl < 250)
Co		50 - 100	100
Cr	200	200	100
Cu		100	100
Dy		0.5	0.5 - 1.0
Eu		0.5	0.5 - 1.0
F	200	100 - (+Cl < 250)	(+Cl < 250)
Fe		300 - 500	500
Ga			100
Gd	3	0.5 - 3	3
In		20	20
K		100	100
Li		100	100
Mg		100 - 200	200 - 500
Mn		100	100
Mo		100	100
N	200	150 - 300	300
Na		100 - 300	300
Nb		100	100
Ni	100	100 - 200	200
P		100	100
Pb		100 - 200	100
S		250	250
Si		150 - 200	200
Sm		2	2
Sn		100	100
Ti		100 - 300	100
Th	200	50 - 200	100
U		1000 - 5000	100 - 5000
V		5 - 300	300
W		100 - 150	150 - 200
Zn		100 - 200	100
Zr		50	50
Boron Equivalent		10	10
Total Impurities	6000	4000 - 5000	5300