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Safety Issues in Fabricating Mixed Oxide Fuel Using Surplus Weapons Plutonium

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ABSTRACT

This paper presents an assessment of the safety issues and implications of fabricating mixed oxide (MOX) fuel using surplus weapons plutonium. The basis for this assessment is the research done at Los Alamos National Laboratory (LANL) in identifying and resolving the technical issues surrounding the production of PuO₂ feed, removal of gallium from the PuO₂ feed, the fabrication of test fuel, and the work done at the LANL plutonium processing facility. The use of plutonium in MOX fuel has been successfully demonstrated in Europe, where the experience has been almost exclusively with plutonium separated from commercial spent nuclear fuel. This experience in safely operating MOX fuel fabrication facilities directly applies to the fabrication and irradiation of MOX fuel made from surplus weapons plutonium. Consequently, this paper focuses on the technical difference between plutonium from surplus weapons, and light-water reactor recycled plutonium. Preliminary assessments and research lead to the conclusion that no new process or product safety concerns will arise from using surplus weapons plutonium in MOX fuel.
INTRODUCTION

The recent Record of Decision on the Storage and Disposition of Surplus Fissile Materials selects a hybrid approach in which relatively clean plutonium from disassembled pits would be irradiated in commercial light water reactors (LWRs) as mixed oxide (MOX) fuel, and relatively dirty plutonium would be immobilized and directly placed in the nation's permanent geologic repository. One basis for this "dual track" decision is the well demonstrated and industrialized fabrication and irradiation of MOX fuel in Europe. Although this experience base is almost exclusively with plutonium separated from commercial spent nuclear fuel, it directly applies to the fabrication and irradiation of MOX fuel made with surplus weapons plutonium. The most efficient way of adopting this technology is to focus primarily on the technical differences between these two fuels. In the fuel fabrication area, Los Alamos National Laboratory (LANL) is leading the effort to identify and resolve certain technical issues including the production of PuO₂ feed, removal of gallium from the PuO₂ feed, the fabrication of test fuel, and the conceptual layout of the domestic fuel production facility. LANL also operates a plutonium processing facility that performs both aqueous and dry processing of plutonium. Based on these experiences and other ongoing activities, this paper presents an assessment of the safety issues and implications of fabricating MOX fuel using surplus weapons plutonium.

The overall MOX fuel disposition process is depicted in Figure 1. Pits are disassembled, and the plutonium is removed and converted to oxide in the pit disassembly and conversion facility (PDCF). Gallium is removed from the oxide by either a dry process (Thermally Induced Gallium Removal or [TIGR]) or an aqueous process (ion exchange or solvent extraction), each of which can be located in either the PDCF or the MOX fuel fabrication facility (FFF). The fuel is fabricated in the MOX FFF and subsequently irradiated in a commercial LWR. The discussion here is necessarily limited to that identified in the figure and includes the gallium removal and fuel fabrication processes.
The primary difference between surplus weapons plutonium and plutonium arising from the reprocessing of spent LWR fuel is the presence of gallium in trace quantities. Although plutonium components from dismantled weapons may have up to 1 wt% gallium as an alloy, the current baseline pit disassembly and conversion process removes the gallium by the dry TIGR process to levels on the order of 100 ppm or less. This process is used to remove the gallium from the feed PuO$_2$ in the pit disassembly and conversion facility, and is similar to the MOX fuel sintering process in that the PuO$_2$ feed is subjected to a high-temperature (~1000°C) reducing atmosphere (argon, 6% hydrogen). This is an important consideration in the operation of the MOX FFF sintering furnaces, because any residual gallium in the feed PuO$_2$ will be further evolved (as Ga$_2$O) in its high-temperature (~1700°C) reducing atmosphere (argon, 6% hydrogen).

The impact of gallium impurities on fuel product quality is also important. Because gallium is a sintering aid, it can change the dynamics of the sintering process and, consequently, the fresh fuel’s microstructure. Such changes depend on numerous processing parameters and may have an impact on fission gas release fraction, pathways (grain boundaries) for migration of impurities to the clad, and the formation of additional phases. Furthermore, residual gallium may eventually evolve from the fuel body during the course of its irradiation. The mechanism for this release and its subsequent behavior are unknown and are very difficult to predict.

The other major differences between surplus weapons plutonium as produced in the current baseline PDCF process (TIGR gallium removal) and LWR recycle plutonium are the physical characteristics of the powder. Although the morphology of the particles are
different, it is the particle-size distribution that may have safety-driven, process-design implications. Current research indicates that under certain production conditions, a large fraction of the powder may be submicron in size. Small particle sizes may lead to handling, material accountability, and dose concerns. Fortunately, research results have also indicated that the TIGR process essentially presinters the smallest particles into larger ones, thus relieving the concerns associated with small particles. Consequently, the particle size distribution of the feed PuO$_2$ (after TIGR gallium removal) is expected to be very similar to that from oxalate precipitation and calcination processes. The primary difference will be in the morphology or shape of the particles. For fuel fabrication routes under consideration, the separate identity of the PuO$_2$ particles is destroyed by high-energy first-stage powder milling and sintering. This process essentially erases any history of the PuO$_2$ production route. Because the mean PuO$_2$ agglomerate size will be <40 μm, issues associated with very large PuO$_2$ agglomerates will be avoided (such as hot-particle ejection and overpower transient reactivity insertion).

The third major difference between surplus weapons plutonium and LWR recycle plutonium is the isotopic composition of the feed plutonium and its $^{241}$Am content. Weapons plutonium has about 6% higher plutonium isotopes (240, 241, 242) and low $^{241}$Am (<.5%) content, whereas reprocessed reactor plutonium has, for example, 43% higher plutonium isotopes, ~3% $^{238}$Pu, and relatively high $^{241}$Am (~1.5%) for plutonium separated from UO$_2$ with a burnup of 45 GWd/tU and aged two years. The processing implication is that for a given mass of plutonium, the fissile content is higher for weapons plutonium, which will lead to lower plutonium materials limits for process equipment because of criticality concerns. A benefit of weapons plutonium is the substantially lower dose rate due to the reduced levels of $^{241}$Am, the reduced levels of higher plutonium isotopes, and a lower heat load due to reduced $^{238}$Pu content.

SUMMARY OF FACILITY ACCIDENT ANALYSES

Two bodies of work were used to form the basis for the safety assessment reported here. The first are the hazards and safety analyses performed in support of obtaining the authorization basis for operating the Los Alamos plutonium facility. The second is the preliminary work done on a conceptual MOX FFF in support of the Surplus Plutonium Disposition Environmental Impact Statement (SPDEIS).
Los Alamos Plutonium Facility (TA-55/PF4)

The Los Alamos plutonium facility (PF4) at Technical Area 55 (TA-55) was approved for plutonium operation in April 1978 by the United States Department of Energy. TA-55/PF4 is currently operational with principal missions of basic special nuclear materials (SNM) research and technology development; processing a variety of actinide-containing materials; and preparing reactor fuels, heat sources, and other SNM devices.

A review of the TA-55/PF4 Final Safety Analysis Report (FSAR) and Hazards Analysis (HA) revealed several areas of concern for fuel fabrication and aqueous processing. The TIGR purification process is still in the development phase; however, a preliminary review of the process indicated few differences in hazards from a direct metal oxidation process recently analyzed for the LANL Advanced Recovery and Integrated Extraction System (ARIES) line. In brief, the most severe accident postulated for MOX fuel fabrication was a criticality accident. The use of a purified feed (high 239Pu content with few impurities) will typically decrease process or glovebox limits, which will affect equipment design, storage requirements, and throughput considerations. Administrative controls; operator training; critically safe, process-equipment design; moderator/reflector limits; and fissile material (glovebox) limits all serve to provide defense-in-depth against an accidental criticality accident. The most severe accident postulated for aqueous purification/processing was an anion exchange column thermal excursion. Should this event occur, the most severe realistic consequence would be a barely detectable release from the TA-55/PF-4 stack. Process controls, equipment design, and operator training all serve to minimize the possibility of such an accident occurring. The possibility of a nitric acid spill from a tank outside the facility was also evaluated. The environmental effects of this accident depend entirely on location of the nearest receptor, location of the tank, and the berm design.

Proposed MOX FFF

In the process of developing supporting documents for the SPDEIS, preliminary hazards and accident analysis was performed for both the MOX FFF and contingent aqueous purification process. To effect this evaluation, bounding accidents of various types were postulated to occur. These hypothetical accidents were evaluated with realistic assumptions to estimate the consequence to the health and safety of the public, workers, and
environment. The spectrum of accidents analyzed in the FFF are criticality, fire, and explosion.

A postulated explosion in the sintering furnace, which is the most serious incident internal to the facility, was analyzed. Sintering gas, a nonexplosive mixture of 6% hydrogen in argon or nitrogen, is supplied to the sintering furnace. The accident is caused by the introduction of an explosive mixture of hydrogen and air due to equipment failure and/or operator error. Even though the introduction of hydrogen into the facility at a concentration higher than the planned 6% is highly unlikely, the event is assumed to occur. The potential consequences of an accident of this type will be reduced to very low values because (1) of the inherently safe plant design, (2) the possibility of an occurrence of such an accident being very remote, and (3) because the facility design reflects the potential seriousness of this type of accident. If such an event were to occur, the consequence would be very low and much less than what was reported in the SPDEIS analysis. This event will generally result in the temporary shutdown of the affected portion of the facility for decontamination and repair.

Another postulated event is a criticality accident involving pure PuO₂ that releases gaseous iodine and noble gases to the facility, and then to the atmosphere through the ventilation system. Accidental moderation of insoluble powder plutonium or uranium do not lead to fission yields greater than 2E+18 fissions. The energy released by such an excursion is minimal, and although the container in which the excursion occurs may be damaged, the building and filtration system are assumed to remain operational. The consequences from this very remote event are low and will be reduced further because of the inherently safe design of the facility. The potential for an accident of this type in a facility containing PuO₂ in only powder form is extremely unlikely. If such an event were to occur, the consequence would be much lower than analyzed in the SPDEIS because of criticality safety features and administrative controls that would be incorporated in the design and operation of the facility.

Another postulated event is a facility fire where burning of all combustibles occurs. The majority of combustible materials in the fabrication facility are not readily ignitable. The most likely sources are overheated motors, electrical fires, etc. The effects of a postulated fire are (1) the effects of the evolution of gases and heat on the ventilation system and (2) the release of these gases from the facility. The consequence of an accident of this type is low and will be reduced further because of the safe design of the facility. The possibility of
an occurrence of such accident is highly unlikely. If such an event were to occur, the consequence would be much lower than analyzed in the SPDEIS because of the fire safety features that would be incorporated in the facility’s design. This event would generally result in the temporary shutdown of the affected portion of the facility for decontamination and repair.

The analyses of these accidents clearly showed that the maximum offsite dose, which could result from a criticality accident, is comparable to the dose from a criticality accident at a UO$_2$ fuel fabrication facility. The slightly different fission product yield and the presence of a small amount of plutonium did not significantly alter the effect of a PuO$_2$ criticality accident relative to those of a UO$_2$ criticality accident. The analyses also showed that the consequences of an explosion are similar to those of a fire and have relatively minor offsite consequences.

If aqueous purification of the feed PuO$_2$ is implemented, then an additional postulated event is a fire in a glove box where a liquid solvent leak occurs. The leakage would occur as a pressurized spray into the glovebox and is assumed to build up to a flammable concentration, which would then be ignited by an ignition source. The consequences of such an accident are low and would be reduced further because of the design safety features of the facility. The possibility of such an accident is extremely unlikely; however, if such an event were to occur, the consequence would be much lower than the consequence from the analysis in the SPDEIS because of the design safety features that would be incorporated.

**FACILITY SAFETY ISSUES**

**TIGR**

It is currently anticipated that the TIGR process will be used to remove the gallium from the feed PuO$_2$ in the PDCF and is similar to the fuel sintering process in that the PuO$_2$ feed is subjected to a high-temperature-reducing (-1000°C) atmosphere (argon, 6% hydrogen). The hazards produced by this operation involve the potential for fire and/or explosion due to the thermal energy and to the presence of hydrogen, the dispersion of PuO$_2$ powder, and the evolution of Ga$_2$O as a vapor that can alloy with metals as it cools.
The selection of a 6% concentration of hydrogen for this operation was made to limit the possibility of approaching an explosive concentration, even in a situation where air instead of argon was introduced into the furnace. This safety approach has proven effective because it is the same approach used for sintering MOX fuel at facilities operating around the world. The thermal treating and processing of plutonium materials has been conducted at the LANL TA-55/PF4 facility for many years, and the safety evaluation of that facility shows that with appropriate safety design considerations, these operations can be accomplished with limited worker exposure and no environmental consequence.

The TIGR development effort at LANL has shown that the process of combining small particles into larger agglomerates will reduce the possible dispersion of the powder in the event of an accident. Through further development of the TIGR process, an optimal particle-size distribution for fuel production and safety considerations may be obtained. Research results at LANL during the development of the TIGR process have indicated that the volatile Ga₂O condenses in a predictable way so that minor modifications to the gas flow and collection system of the furnace can be designed and implemented to collect the gallium.

**Aqueous purification**

The aqueous process analysis of a criticality explosion in the solution tank clearly indicated that the largest dose to an individual in the vicinity resulted from an accident involving plutonium in a soluble form. Therefore, special emphasis must be directed toward mitigating such a release and should provide, as necessary, additional engineered safety features and administrative control for operation in this aqueous process.

**MOX FFF**

Gallium is an important consideration in the operation of the MOX plant’s sintering furnaces because any residual gallium in the feed PuO₂ will be further evolved (as Ga₂O) in its high-temperature-reducing (~1700°C) atmosphere (argon, 6% hydrogen). Safe and reliable operation of the furnace is a key requirement, and research results at LANL have indicated that the volatile Ga₂O condenses in a predictable way so that minor modifications to the gas flow and collection system of the furnace can be designed and implemented to
collect the gallium. These changes should not present any new hazards or safety problems for the operation of the MOX FFF.

Other safety issues at the MOX FFF are criticality safety and worker dose. The higher fissile content of the weapons grade plutonium means that the control on the inventory in any confined area will be more stringent, and that the amount of plutonium will be reduced compared with MOX fuel production from recycled plutonium. Concurrently, the concentration of the plutonium in the final fuel will be reduced for a given fuel type compared with MOX manufactured with recycled plutonium so that there is a compensating effect. In the event of an accident, the doses to workers and the public should be reduced because of the reduced material at each location and because the isotopes that produced the highest radiological effects are reduced compared with recycled plutonium. The heating, ventilation, and air conditioning (HVAC) system in the plant areas where plutonium powder is present will have a multizoned air circulation system that will provide confinement of any material that escapes the glovebox. This confinement system includes two components. The first is a pressure gradient between zones that would sweep any material back into the contaminated area. The second consists of multiple high efficiency particulate air (HEPA) filter stages, which are protected from a fire or explosion.

Explosion and fires are another important safety concern in the MOX plant. The material at risk in any particular location will be limited by criticality and dose concerns; however, in the event of a fire or explosion, the dispersion of this material may have a high consequence for the worker or the environment. Therefore, the initiators and fuels for fires or explosions must be minimized. This involves limiting combustibles in gloveboxes where plutonium is present. One possibility is locating equipment (which might cause fires such as electric motors, etc.) outside the glove box and using a coupling to transmit the force into the glovebox. Another possibility is to inert the atmosphere in the glovebox to preclude any fire or explosion.

**CONCLUSION**

The preliminary assessments presented here, coupled with research currently under way, are leading to the conclusion that no new process or product safety concerns will arise as a consequence of using surplus weapons plutonium in MOX fuel.
References

1. Record of Decision for the Storage and Disposition of Weapons-Usable Fissile Material Final Programmatic Environmental Impact Statement, found at http://www.doe-md.com


