REACTOR-BASED PLUTONIUM DISPOSITION: OPPORTUNITIES, OPTIONS, AND ISSUES

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Abstract

The end of the Cold War has created a legacy of surplus fissile materials (plutonium and highly enriched uranium) in the United States (U. S.) and the former Soviet Union. These materials pose a danger to national and international security. During the past few years, the U. S. and Russia have engaged in an ongoing dialog concerning the safe storage and disposition of surplus fissile material stockpiles. In January 1997, the Department of Energy (DOE) announced the U. S. would pursue a dual track approach to rendering approximately 50 metric tons of plutonium inaccessible for use in nuclear weapons. One track involves immobilizing the plutonium by combining it with high-level radioactive waste in glass or ceramic "logs". The other method, referred to as reactor-based disposition, converts plutonium into mixed oxide (MOX) fuel for nuclear reactors. The U. S. and Russia are moving ahead rapidly to develop and demonstrate the technology required to implement the MOX option in their respective countries.

U. S. MOX fuel research and development activities were started in the 1950s, with irradiation of MOX fuel rods in commercial light water reactors (LWR) from the 1960s – 1980s. In all, a few thousand MOX fuel rods were successfully irradiated. Though much of this work was performed with weapons-grade or “near” weapons-grade plutonium – and favorable fuel performance was observed – the applicability of this data for licensing and use of weapons-grade MOX fuel manufactured with modern fuel fabrication processes is somewhat limited.

The U. S. and Russia are currently engaged in an intensive research, development, and demonstration program to support implementation of the MOX option in our two countries. This paper focuses on work performed in the U. S. and provides a brief summary of joint U. S./Russian work currently underway.

1. Introduction

Over the last several years, the U. S. and Russia have engaged in an intensive dialog relating to the management and storage of surplus fissile materials in the two countries. In September 1993, the U. S. committed to eliminate surplus fissile materials. The following January, President Yeltsin and President Clinton issued a joint statement limiting the use of fissile materials and initiating joint studies regarding long-term disposition of plutonium. Several meetings and agreements have been made since that time with the latest, "Joint Statement on Principles for Management and Disposition of Plutonium", issued in September 1998. This statement declared the intent of Russia and the U. S. to each disposition 50 MT of military plutonium.

A decision has been made in the U. S. to pursue a dual approach to plutonium disposition, which utilizes immobilization and irradiating plutonium as mixed oxide (MOX) fuel in commercial nuclear power reactors. Several factors contributed to this dual-approach decision:

Two technologies provide some assurance that the mission will continue if any unforeseen technical, schedule, cost, or institutional obstacles are encountered. No domestic or international consensus exists concerning the best technical approach. Russia has expressed a concern that immobilization does not destroy plutonium and leaves it as weapons grade material that could be used for possible weapons reuse. Plutonium disposition as MOX fuel provides the best opportunity for the U. S. to work with Russia and other countries to reduce Russia's excess plutonium.

The goal of the U. S. plutonium disposition effort is to achieve the "Spent Fuel Standard". The Spent Fuel Standard states that surplus plutonium will be made as inaccessible and unattractive for retrieval and weapons use as the residual plutonium in spent fuel from commercial reactors. The primary characteristics associated with the Spent Fuel Standard are the material's radiation field, plutonium concentration, chemical form, size and weight, and storage location.

Currently, the U. S. plans to complete the 50 MT plutonium disposition campaign by the year 2022. Achievement of this goal will require the construction of a Pit Disassembly and Conversion Facility (to extract the plutonium from weapons components and convert it to plutonium oxide feed material), and a MOX Fuel Fabrication Facility. The U. S. Department of Energy (DOE) is currently negotiating with Raytheon Engineers & Constructor, Inc. for the preliminary and detailed design of the Pit Disassembly and Conversion Facility. Construction of the facility is expected to begin in 2001 with startup in 2006.

The Department has contracted with Duke Engineering & Services, COGEMA, Inc., and Stone & Webster (collectively known as "DCS") to provide mixed oxide fuel fabrication and reactor irradiation services in support of the department's mission to dispose of surplus weapons plutonium. DCS will design, provide construction management services, operate and deactivate a weapons-grade mixed oxide fuel fabrication facility in the U. S. The team will also modify six existing U. S. commercial light water reactors at three sites to irradiate mixed oxide fuel assemblies. These reactors sites are Catawba in York, South Carolina; McGuire in Huntersville, North Carolina; and North Anna in Mineral, Virginia. The consortium will be responsible for obtaining a license to operate the fuel fabrication facility and the license modifications for the reactors from the Nuclear Regulatory Commission (NRC). DOE is selecting a contractor to provide these fabrication and irradiation services in parallel with determining the location for the fuel fabrication facility.

The department is preparing a Surplus Plutonium Disposition Environmental Impact Statement that analyzes the potential environmental impacts associated with establishing plutonium disposition facilities at DOE sites. Those sites are: the Hanford Reservation near Richland, WA; the Idaho National Engineering and Environmental Laboratory near Idaho Falls, ID; the Pantex Plant near Amarillo, TX; and the Savannah River Site near Aiken, SC. In June 1998, DOE announced that the Savannah River Site was the preferred site for the mixed oxide fuel fabrication facility. The Record of Decision on this environmental review is expected this summer.

2. Unique Characteristics of Weapons-Grade Plutonium

The technical issues relating to reactor-based plutonium disposition stem directly from the differences between reactor-grade (RG-) plutonium and weapons-grade (WG-) plutonium. Weapons-grade plutonium contains more Pu-239, less Pu-240, and less Am-241 than reactor-grade plutonium. While neither the U. S. or Russia has made a final decision regarding the weapons disassembly and plutonium conversion and purification processes to be employed in the disposition mission, the U. S. has not ruled out the use of new dry Pu conversion and purification processes. These processes could lead to differences in weapons-grade plutonium oxide powder morphology and impurities (relative to commercial reactor-grade plutonium). All the conversion processes under consideration in the U. S. are dry processes. The U. S. will employ a hydride-dehydride process for disassembly of the weapons components and extraction of the plutonium metal.

Metal-to-oxide conversion processes under consideration at the present time include both direct metal oxidation and a multi-step process in which the plutonium metal is first converted to a nitride and then to an oxide. The U. S. has evaluated the use of both dry thermal plutonium purification processes, and traditional aqueous polishing techniques for removal of trace elements such as gallium. The best available data in the U. S. suggests that the use of dry processes for conversion and purification will yield gallium concentrations in the fabricated MOX fuel of approximately 0.1–1.0 parts per million. (For purposes of comparison, the fission yield of gallium in existing LEU and RG– MOX fuel is on the order of 0.1 parts per billion.)

3. U. S. LWR MOX Fuel Experience

U. S. MOX fuel experience is substantial but dated, because LWR MOX fuel development work was halted in the late 1970s following a Presidential Executive Order banning fuel recycle. U. S. MOX fuel research and development activities were started in the 1950s, with irradiation of MOX fuel rods in commercial reactors from the 1960s – 1980s. Table 1 provides details of the irradiations that were conducted on the MOX rods.

Table 1. U. S. LWR MOX Irradiation History

Reactor	**Dates of Irradiation**	**No. of MOX Assays (rods)**	**Burnup (MWd/Mt) Max Avg. Assay (Peak MOX Pellet)**	**Examin-ations**	**Comments**	**Data Utilization**
Ginna (PWR)	1980-1985	4 (716)	39,800 (?)	None	Assemblies intact (82% fissile Pu)	FY99 spent fuel exam plan
Quad Cities–1 (BWR)	1975-1980s	5 (48)	39,900 (57,000)	D & ND core phys	Well documented EPRI (80 & 90% fissile Pu)	Neu benchmarks constructed and analyzed
Big Rock Point (BWR)	1969-late 1970s	53 (1248)	~20,000 est (30,200)	D & ND	Little documentation	No current plans
San Onofre–1 (PWR)	1970-1972	4 (720)	19,000 (23,500)	Some D	PIE documents found	FY99 neu analysis plan
Dresden–1 (BWR)	1968-early 1970s	15 (103)	~19,000 (~14,000)	–	Little documentation	No current plans
Saxton (PWR research reactor)	1965-1972	9 (638)	Many reconstitu-tions (51,000)	Fuel perf D and physics tests	Relatively well documented fuel perf data (91.4% fissile Pu)	Critical experiments analyzed and reported
Misc. Test Rx (Exp BWRs) (Pu Re Test Rx) (Mat Test Rx) (Eng Test Rx)	1960s-1970s	1000s of rods	?	Variety of D	Capsules and rods irradiated. Little historical research conducted	No current plans

Abbreviations:
D – destructive, ND – non-destructive, Neu – neutronics, est – estimated, perf – performance, Rx – reactor, Exp – experimental, Re – recycle, Mat – material, Eng, – engineering

A few thousand MOX fuel rods were successfully irradiated in the U. S. Plutonium isotopics used for the tests included near weapons-grade compositions. The fuel performance was found to be equivalent to contemporary LEU fuel performance. Reviews conducted by the NRC, and documented in NUREG-0002, concluded there were no significant health and safety impacts to the public of MOX fuel fabrication or reactor operations.

Critical experiments were conducted at the Critical Reactor Experiment facility at the Westinghouse Reactor Evaluation Center. These experiments included 44 lattice configurations of MOX and LEU rods involving both single and multiple regions. MOX lattice pitches ranged from 1.32 to 2.64. The MOX rods contained 6.6 wt % PuO_2 with 90% ^{239}Pu in the plutonium. The plutonium rods were then irradiated in the Saxton reactor.

As an extension of the Saxton critical experiments, critical experiments were conducted at ESADA to consider variations in ^{240}Pu. Variations included 2 wt % PuO_2 in natural uranium with 8% ^{240}Pu and 24% ^{240}Pu. Single region experiments were conducted to evaluate buckling, reactivity worth, and power distribution. Multi-region experiments were conducted to evaluate reactivity worth, power distribution, and lattice pitches. In addition concentric regions and "salt and pepper" arrangements were evaluated.

Additional critical experiments were conducted at Pacific Northwest National Laboratory (Battelle). Six lattices of MOX rods with 2 wt % PuO_2 with 8% ^{240}Pu in water, and six lattices of UO_2 were constructed. Results of the study are documented by EPRI in *Clean Critical Experiment Benchmarks for Plutonium Recycle in LWRs*, report number EPRI NP-196. The ANS Cross Section Evaluation Working Group has adopted this work as a benchmark.

4. U. S. MOX Licensing Considerations

The U. S. regulatory review focus and process is dictated by 10CFR50, and is facilitated by U. S. NRC Regulatory Guidelines, NUREG-0800 (NRC's Standard Review Plan), and NRC Generic Letter 88-20 (which relates to the execution of Probabilistic Risk Assessments and Individual Plant Risk Examinations). The focus of the U. S. regulatory review process is to ensure:

1. no fuel damage can occur during normal operations and anticipated transients;
2. fuel damage during a design basis accident cannot proceed to the point of preventing control rod insertion;
3. a coolable core geometry is maintained during design basis accidents; and
4. the radioactivity release during a design basis accident is not underestimated.

Three basic questions will need to be answered as a part of the U. S. regulatory review of WG-MOX use in commercial reactors:

- Are safety margins significantly reduced?
- Is the probability or consequence of any previously analyzed accident increased?
- Does the use of MOX fuel create the possibility of new types of accidents?

In order to address these questions, the U. S. licensee will need to demonstrate that the thermal, mechanical, and physics performance of weapons-grade MOX fuel is equivalent to LEU fuel, and that analytical methods used to demonstrate fuel performance are as accurate as current methods used for LEU fuel.

The U. S. is currently transitioning to "risk-informed regulation." In July 1998, the U. S. Nuclear Regulatory Commission issued Regulatory Guide 1.174, "An Approach for Using Probabilistic Risk Assessment in Risk-Informed Decision on Plant-Specific Changes to the Licensing Bases." As recently stated by Shirley Ann Jackson, Chairman of the Commission [Nuclear News, January 1999],

> "This Regulatory Guide provides a general framework for plant-specific NRC decisions that have been requested and initiated by licensees. It sets forth the Commission-approved principles for NRC staff evaluation of such proposals, including expectations for application of the Commission Safety Goal Policy,reliance on traditional defense-in-depth approaches, and maintenance of sufficient safety margins when initiating changes to the licensing bases. In addition it provides criteria for the scope, level of detail, and quality the PRA supporting the licensee submittal."

In addition to the traditional deterministic safety case reflected by 10CFR50, it is clear that reactor licensees who intend to employ WG-MOX fuel will need to demonstrate that the impacts of WG-MOX fuel use are acceptable from the overall risk standpoint, including beyond design basis accidents.

Thus the U. S. reactor owner will be required to demonstrate a thorough understanding of the impacts of WG-MOX substitution on overall plant performance and safety. Demonstration of the safety case will rest upon the availability of data and validated computational tools. Where reliance is made on commercial (reactor-grade) MOX fuel data, it will, of course, be necessary to show that the data is both applicable to weapons-grade MOX fuel and sufficient to support the safety case. However, based upon the established track record of favorable fuel performance in the commercial MOX industry, there is high confidence that WG-MOX fuel can be successfully licensed in the U. S.

5. U. S. R&D

Given the abundance of favorable international commercial MOX experience, the U. S. reactor-based plutonium disposition research and development program has focused on issues that derive from the unique characteristics of weapons-grade MOX: (1) isotopics, (2) dry-processed oxide powder morphology, and (3) impurities.

Reactor physics studies have provided a better understanding of the impact of weapons-grade plutonium on MOX fuel performance. An integral part of the current work is the international ARIANE project, which includes irradiation of RG–MOX fuel samples at Dodewaard in Holland, and at Beznau and Goesgen in Switzerland; with examination of the irradiated samples at CEN in Belgium, PSI in Switzerland, and TUI in Germany. Both BWR and PWR LEU and MOX data are being obtained at multiple burnups, with the intent of expanding the database of on fission products and actinides yields during irradiation. Potential applications of this data include reactor physics, accident analysis, reactor operations, spent fuel management, and spent fuel shipping.

Figure 1 provides a comparison of the predicted actinide inventories (as calculated by SCALE and HELIOS-2) of the Dodewaard BWR RG–MOX fuel. The uncertainty in the burnup of the test fuel is estimated to be up to 4%. The sensitivity of the U-235 concentration to burnup is 2.5; (i.e. an uncertainty of 1% in burnup translates to an uncertainty of 2.5% in the U-235 concentration). For Pu-239 the sensitivity to burnup is about 2.0. Pu-239 is also sensitive to the large concentration of U-238 and thus any uncertainty in the U-238 capture cross section is magnified. Am-241 and Am-242m agreements are poor. The latter nuclide, however, has a very low concentration (high measurement uncertainties) and the former is complicated by the Pu-241/Am-241 ratio in the fresh fuel. The calculated-to-measured agreements for the curium isotopes are considered reasonable. While the HELIOS results compare much more favorably than the SCALE calculations, the *trends within* the HELIOS

results are similar to those in SCALE and suggest that uncertainties in cross section data are driving the major observed discrepancies between computed and measured data. It is not surprising that the HELIOS results compare more favorably with the data than the SCALE results. SCALE is designed to provide assembly-average information. However, the MOX fuel samples analyzed in this program were extracted from fuel assemblies in which a few MOX fuel rods were surrounded by LEU rods. Additional data on PWR MOX fuel will be forthcoming from the ARIANE project in the near future.

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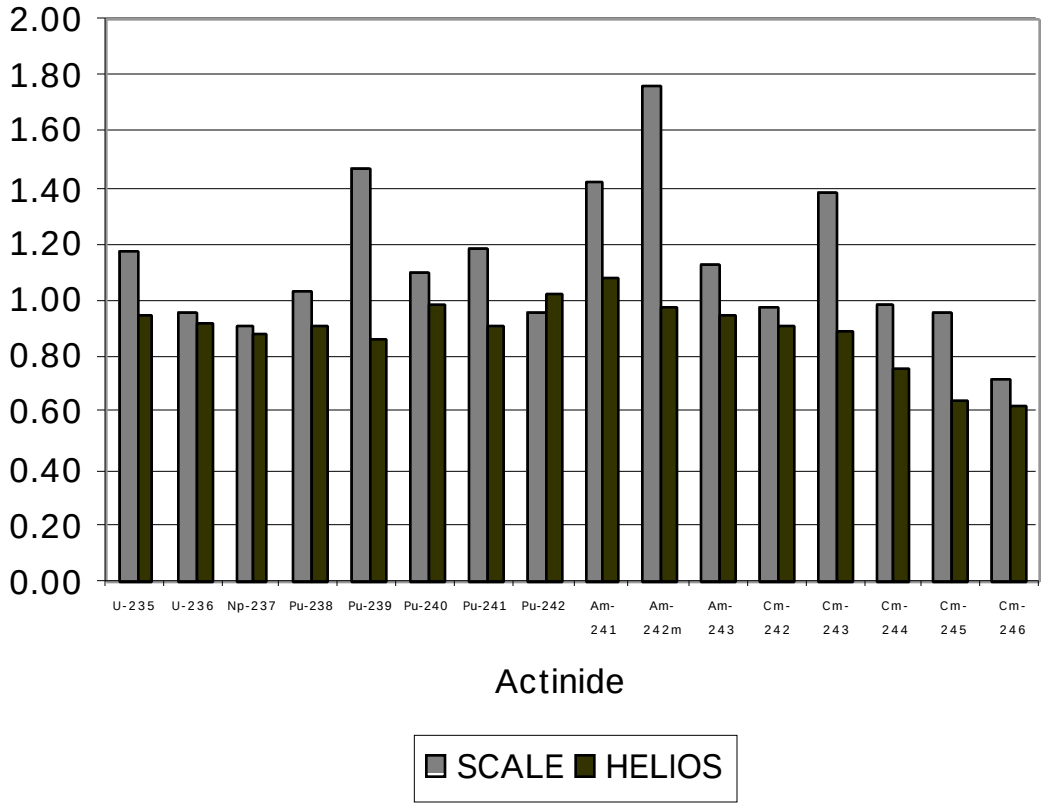

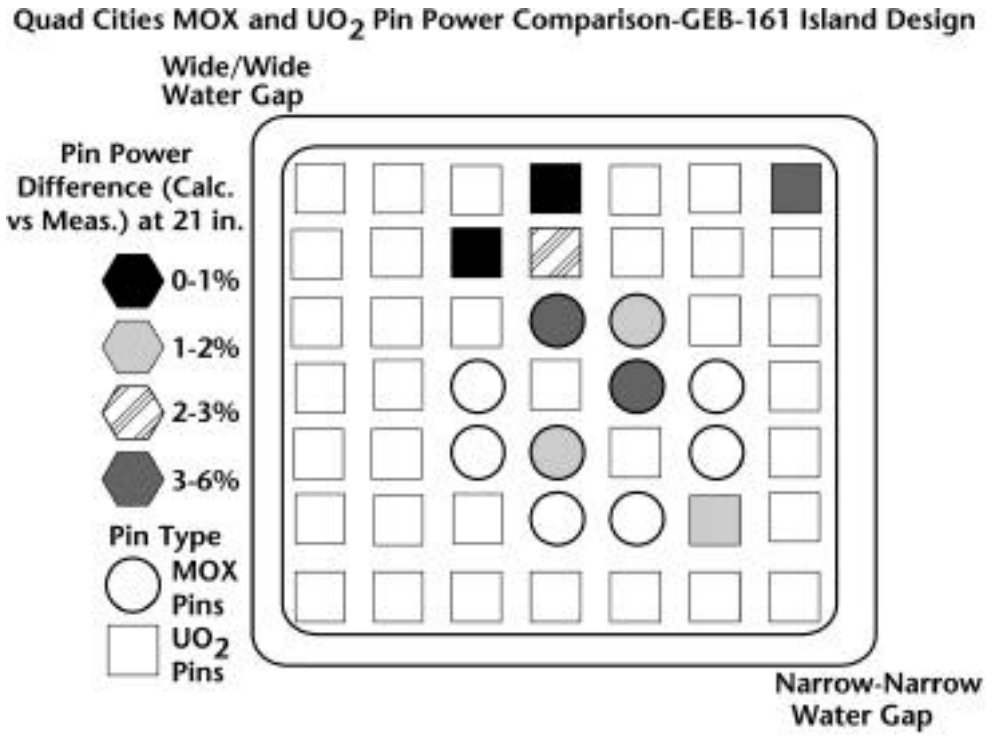

Figure 2. Comparison of HELIOS-2 calculations for Quad Cities BWR MOX fuel bundle pin powers to measured data.

A coordinated test program is underway to characterize the impact of residual gallium in MOX fuel. The intent is to gain an understanding of the gallium transport mechanisms, reactions, kinetics, and damage mechanisms. A two-phase out-of-reactor test program was conducted to determine general and separate effects. Table 2 summarizes the test parameters for the study. The results of the limited testing performed to date indicate that intermetallic compound formation is the only potential gallium/cladding corrosion mechanism. No evidence of either grain boundary corrosion or liquid metal embrittlement has been observed. This intermetallic compound formation has been observed to occur at temperatures above 300 C in liquid gallium and at temperatures above 500 C in gallium oxide. No structural deformation of cladding specimens has been observed to occur at gallium concentrations 10000 – 100000 times greater than that expected in WG-MOX fuel.

-Table 2. Out-of-reactor gallium/clad compatibility tests for weapons-derived MOX fuel.

Variable	Phase I Static	Stressed	Phase II Static	Stressed
Gallium form	Metal	Metal	Ga oxide Ga/Ce oxide	Ga oxide Ga/Ce oxide
Concentration (%)	100	100	100, 1, 0.2, 0.1	100, 1, 0.2, 0.1
Cladding	Zr, Zircaloy-2, Zircaloy-4, Zirlo	Zr, Zircaloy-2, Zircaloy-4, Zirlo	Zr, Zircaloy-2, Zircaloy-4, Zirlo	Zr, Zircaloy-2, Zircaloy-4, Zirlo
Temperature (°C)	30, 300, 500[a,b]	30, 300, 500[a,b]	300, 500, 700	300, 500, 700
Time	200 h, 700 h	200 h, TTF	6 wk, 12 wk	6 wk
a - without cadmium, b – with cadmium, TTF – time to failure				

A complementary materials irradiation testing program has been initiated to extend the out-of-reactor tests to in-reactor conditions. These capsule tests employ two different WG-MOX test fuels fabricated at Los Alamos National Laboratory. Both fuels were fabricated with dry-processed 5% WG-Pu. One of the fuels was manufactured with plutonium that had received no additional purification (yielding a final gallium concentration of approximately 2 parts per million in the fuel), while the other fuel was manufactured from plutonium that had been thermally treated to remove gallium (resulting in a final gallium concentration of approximately 0.7 parts per million in the fuel). The fuel is being irradiated in the Advanced Test Reactor at Idaho National Engineering and Environmental Laboratory at liner heat generation rates of approximately 18-30 kW/m, and will be burned to approximately 30 GWd/MT burnup. The goal of this test is to develop a better understanding of gallium migration mechanisms within the fuel pellet and gallium/clad reaction mechanisms at the pellet/clad interface. The first irradiated fuel specimens (irradiated to about 8 GWd/MT) are currently undergoing post-irradiation examination at Oak Ridge National Laboratory.

6. Cooperative R&D With Russia

In keeping with the declarations made as a part of the "Joint Statement on Principles for Management and Disposition of Plutonium" in 1998, the U. S. and Russia are cooperating to accelerate the pace of the plutonium disposition activities. The focus of current cooperation is on the use of existing Russian reactors (VVER-1000s and the BN-600) for plutonium disposition activities. The U. S. is working with Russia to develop and test VVER-1000, BN-600 (fast reactor) WG-MOX fuels, and to validate reactor physics, fuel cycle criticality, and safety codes as they apply to Russian reactors, Pu conversion, and fuel fabrication facilities. Based on analyses performed to date, it appears that the VVER-1000 reactors would be capable of disposition approximately 425 Kg of plutonium per year. The BN-600 could burn approximately 300 Kg of Pu per year with a "hybrid core" option in which the reactor's current radial breeding blankets are removed, and up to 1.3 MT of plutonium per year if fully converted (by removal of the axial breeding blankets) for the plutonium consumption mission.

U. S. LWRs employ solid fuel pellets very similar in thermomechanical/physical design to European reactor-grade MOX fuel pellets. However, VVER-1000 reactors employ annular pelletized fuel with a central hole in the fuel pellet and a Zr-1%Nb alloy for cladding. Thus there is more uncertainty regarding the applicability of European reactor-grade MOX fuel performance data to the VVERs than there is with regard to the applicability of this data to the

U. S. LWRs. Plans are being made to conduct VVER WG-MOX irradiation tests in the MIR test reactor at the Research Institute of Atomic Reactors at Dimitrovgrad, and for irradiation of lead assemblies in a commercial VVER. The most likely candidate for irradiation of lead assemblies appears to be the Balakovo-4 plant. These tests should provide a good basis for judging the applicability of European commercial MOX fuel performance data to the VVERs.

Work is also underway to plan and implement conversion of Russia's BN-600 fast reactor to a plutonium-burning reactor. Russia has proposed to use their one BN-600 reactor at Beloyarsk to disposition 20 Mt of plutonium from the year 2005 to 2020. Reviews are currently being conducted of the safety and licensing requirements and economics.

As stated above, the U. S. is also collaborating with Russia to benchmark and validate the computational physics and criticality safety computer codes required for analysis and licensing of reactor and fuel cycle facilities. These codes include VENTURE, SCALE, HELIOS, MCNP, MCU, TVS-M, and TRIANG-PIN. A suite of over 100 benchmark calculations have been performed with aqueous WG-Pu solutions, Pu and MOX powders, fuel assemblies, and reactor lattices. The results of these analysis indicated that calculated k-effectives agree with experimental values to within 1%. Calculated power distributions within lattice experiments generally agree with measurements to within 2% although values as high as 6.5% were seen. These variations are larger than those found for LEU calculation-to-experiment comparisons. However, since MOX assemblies are expected to be placed in non-limiting positions in the reactor core host reactors should be able to operate at currently rated powers. Based on these results, it would appear that no additional critical experiments are required prior to insertion of the lead test assemblies into a VVER-1000. Furthermore, our Russian counterparts have judged that no additional critical experiments are necessary to support nuclear safety analyses for the out-of-reactor portion of the MOX fuel cycle.

In addition to these activities, the U. S. is cooperating with Russian to evaluate the feasibility of using reactors outside of Russia (CANDU reactors in Canada) or new/advanced reactors within Russia (gas turbine modular helium reactors) to disposition plutonium to supplement the disposition capacity provided by the current VVER-1000s and the BN-600. One CANDU test fuel bundle, comprised of fuel manufactured in the U. S. at Los Alamos National Laboratory and in Russian at the A. A. Bochvar All-Russian Research Institute of Inorganic Materials is scheduled to be loaded into the NRU reactor at Chalk River, Canada later this year. This test program will provide valuable information on the performance of WG-MOX fuel in CANDU reactor environments. Preliminary design work is just beginning for a plutonium consumption GT-MHR that could disposition 250 Kg of plutonium per year per module.

7. Summary

The end of the Cold War has created a legacy of surplus fissile materials (plutonium and highly enriched uranium) in the U. S. and the former Soviet Union. During the past five years, the U. S. has devoted significant effort to the development of a practical, safe, and robust plutonium disposition option based on the use of existing commercial light water reactors and immobilization. The work conducted to date, along with international commercial MOX experience provides a strong technical basis for optimism that the use of weapons-grade MOX fuel can be successfully licensed and implemented in the U. S. The U. S. and Russia are cooperating to develop and demonstrate the technologies required for successful implementation of the reactor-based plutonium disposition option in Russia.