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LOCKHEED MARTIN

SURVEY OF WORLDWIDE LIGHT
WATER REACTOR EXPERIENCE
WITH
MIXED URANIUM-PLUTONIUM
OXIDE FUEL

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ABSTRACT

The United States (U.S.) and the Former Soviet Union (FSU) have recently declared quantities of weapons materials, including weapons-grade (WG) plutonium, excess to strategic requirements. One of the leading candidates for the disposition of excess WG plutonium is irradiation in light water reactors (LWRs) as mixed uranium-plutonium oxide (MOX) fuel. A description of the MOX fuel fabrication techniques in worldwide use is presented. A comprehensive examination of the domestic MOX experience in U.S. reactors obtained during the 1960s, 1970s, and early 1980s is also presented. This experience is described by manufacturer and is also categorized by the reactor facility that irradiated the MOX fuel. A limited summary of the international experience with MOX fuels is also presented.

A review of MOX fuel and its performance is conducted in view of the special considerations associated with the disposition of WG plutonium. Based on the available information, it appears that adoption of foreign commercial MOX technology from one of the successful MOX fuel vendors will minimize the technical risks to the overall mission. The conclusion is made that the existing MOX fuel experience base suggests that disposition of excess weapons plutonium through irradiation in LWRs is a technically attractive option.

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LIST OF ACRONYMS

Acronym	Definition
ABWR	Advanced Boiling Water Reactor
ADU	Ammonium Diuranate [$(NH_4)_2U_2O_7$]
AEC	U.S. Atomic Energy Commission
AECL	Atomic Energy of Canada, Limited
AGR	Advanced Gas Reactor
ALKEM	Alpha Kemistry and Metallurgy GmbH, now part of Siemens AG
ANRCP	Amarillo National Resource Center for Plutonium
ARHCO	Atlantic Richfield Hanford Company
ARIANE	Actinide Research in A Nuclear Element
ARIES	Advanced Recovery and Integrated Extraction System
ATR	1) Advanced Test Reactor (Idaho National Engineering and Environmental Laboratory) 2) Advanced Thermal Reactor (Japan)
AUC	Ammonium Uranyl Carbonate [$(NH_4)_4UO_2(CO_3)_3$]
AUPuC	Ammonium Uranyl-Plutonyl Carbonate [$(NH_4)_4(U,Pu)O_2(CO_3)_3$]
B&W	Babcock and Wilcox
BN	Belgonucléaire
BNFL	British Nuclear Fuels Limited, plc
BNWL	Battelle Pacific NorthWest Laboratories
BR2	Belgian Reactor 2
BR3	Belgium's first PWR, designed by Westinghouse
BRP	Big Rock Point BWR
BWR	Boiling Water Reactor
CANDU	Canadian Deuterium Uranium Reactor
CE	ABB Combustion Engineering
CEA	Commissariat à l'Énergie Atomique
CEC	Commission of European Communities
CFCa	le Complexe de Fabrication des combustibles au plutonium de Cadarache, a pilot MOX fuel facility at Cadarache, France

Acronym	Definition
CNA	Centrale Nucléaire des Ardennes (sometimes used to refer to Chooz A)
CNEN	Comitato Nazionale per l'Energia Nucleare
COCA	Cobroyage-Cadarache, a comilling fabrication procedure used in CFCa facility until recently
COGEMA	Compagnie Générale des Matières Nucléaires
COGEPEL	Coprecipitation process
COMMOX	Joint Belgonucléaire-COGEMA MOX marketing firm
COPRECAL	Coprecipitation and calcination process
CPS	Cold-pressed and sintered
DCA	Deuterium Critical Assembly
DEMOX	Dessel MOX plant at Mol, Belgium, more commonly known as the P0 plant.
DOE	Department of Energy
EBWR	Experimental Boiling Water Reactor
ECN	Energieonderzoek Centrum Nederland
EDF	Électricité de France
EEI	Edison Electric Institute
ENC	Exxon Nuclear Company, Inc.
ENEA	Ente per le Nuove tecnologie l'Energia e l'Ambiente
ENEL	Ente Nazionale per l'Energia Elettrica
EPRI	Electric Power Research Institute
ETR	Engineering Test Reactor
FBFC	Société Franco-Belge de fabrication de combustibles
FERTF	Fuel Element Rupture Test Facility
FFTF	Fast Flux Test Facility
FMDP	Fissile Materials Disposition Program
FRAGEMA	FRAMATOME Nuclear Fuel Division, a joint venture of FRAMATOME (50%) and COGEMA (50%)
FRAMATOME	French NSSS and fuel supplier
FSU	Former Soviet Union
GE	General Electric Company
GESMO	Generic Environmental Statement on the use of Recycle Plutonium in

Acronym	Definition
	Mixed Oxide in light water cooled reactors
GETR	General Electric Test Reactor
GKN	Gemeinschaftskernkraftwerk Nekar, also refers to Neckarwestheim-1
GUNC	Gulf United Nuclear Corporation
HFR	High Flux Reactor, Petten
HYDOX	Hydride-oxidation metal-to-oxide conversion
IFA	Italian fuel assembly
IFBA	Integral Fuel Burnable Absorber, a ZrB_2 coating developed by Westinghouse
INEEL	Idaho National Engineering and Environmental Laboratory
IS	Item Summary
JAERI	Japan Atomic Energy Research Institute
JAPCO	Japan Atomic Power Company
JNC	Japan Nuclear Cycle Development Institute
KBR	Kernkraftwerk Brokdorf GmbH
KKG	Kernkraftwerke Göesgen
KKP	Kernkraftwerk Philippsburg
KKU	Kernkraftwerk Unterweser
KRB-A	Kernkraftwerke Gundremmingen Betriebsgesellschaft mbH
KWL	Kernkraftwerk Lingen GmbH
KWO	Kernkraftwerk Obrigheim PWR
KWU	Kraftwerk Union AG
LANL	Los Alamos National Laboratory
LEU	Low enriched uranium
LMFBR	Liquid Metal Fast Breeder Reactor
LMR	Liquid metal reactor
LMR	Liquid Metal Reactor
LTA	Lead Test Assembly
LWR	Light Water Reactor
MAGNOX	Magnesium-alloy-clad, oxide-fueled, CO_2 cooled early UK reactor design
MDF	MOX Demonstration Facility
MELOX	COGEMA MOX plant at Marcoule

Acronym	Definition
MIMAS	Micronized Master blend
MIR	Materials Irradiation Reactor
MOX	Mixed uranium-plutonium oxide
MTR	Materials Test Reactor
MZFR	Mehrzweckforschungsreaktor
NMD	Nuclear Materials Division of The Babcock and Wilcox Company
NOK	Nordostschweizerische Kraftwerke
NPD	Nuclear Power Demonstration (reactor)
NRC	U.S. Nuclear Regulatory Commission
NSSS	Nuclear Steam Supply System
OCOM	Optimized Comilling (sometimes written as OKOM)
OD	Outside diameter
OKG	Oskarshamnsverkets Kraftgrupp Aktielbolag
OKG1	Refers to the first reactor built by Swiss Oskarshamnsverkets Kraftgrupp AB—Oskarshamn 1
ORNL	Oak Ridge National Laboratory
PDS	Plutonium Disposition Study
PFDF	Plutonium Fuel Development Facility
PFFF	Plutonium Fuel Fabrication Facility
PFPF	Plutonium Fuel Production Facility
PHWR	Pressurized heavy water reactor
PIE	Postirradiation Examination
PRDP	Plutonium Recycle Demonstration Program
PRTR	Plutonium Recycle Test Reactor
PUP	Plutonium Utilization Program
PUREX	Plutonium Uranium Recovery by Extraction
PVA	polyvinyl alcohol
PWR	Pressurized Water Reactor
RBG	Reaktor Beteilingungsgesellschaft, now RBU
RBU	Reaktor-Brennelement Union GmbH
RG	Reactor Grade

Acronym	Definition
RG&E	Rochester Gas and Electric
SBR	Short Binderless Route
SCK-CEN	Studiecentrum voor Kernenergie–Centre d'Étude de l'Énergie Nucléaire
SENA	Société d'Énergie Nucléaire Franco-Belge des Ardennes (utility operator for Chooz A, sometimes used as the reactor pseudonym)
SMP	Sellafield MOX Plant
SPERT	Transient Irradiation Test at Idaho National Engineering and Environmental Laboratory
SPHEREPAC	Sphere compaction
SS	stainless steel
T.D.	Theoretical Density
U.S.	United States
UK	United Kingdom
UKAEA	United Kingdom Atomic Energy Authority
UNH	Uranyl nitrate hexahydrate, $UO_2(NO_3)_2 \cdot 6H_2O$
VAK	Versuchsatom Kraftwerk
VENUS	SCK-CEN zero-power critical facility
VIPAC	Vibrationally compacted particle fuel
VVER	Soviet pressurized water reactor
W	Westinghouse
WG	Weapons-grade

Survey of Worldwide Light-Water Reactor Experience With Mixed Uranium-Plutonium Oxide Fuel

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1.   INTRODUCTION

The United States (U.S.) and the Former Soviet Union (FSU) have both declared significant quantities of weapons materials (highly enriched uranium and plutonium) surplus to strategic requirements. The U.S. Department of Energy (DOE) has completed the Plutonium Disposition Study (PDS) and initiated the follow-on Fissile Materials Disposition Program (FMDP) to address this material. While the excess uranium can be easily denatured (made unfit for weapons use) through blending with either depleted or natural uranium, the excess plutonium is not so easily dispositioned. One of the leading candidates for disposition of excess weapons-grade (WG) plutonium is irradiation in light water reactors (LWRs) as uranium-plutonium oxide (MOX) fuel. The nuclear fission process, often referred to as "burning" even though it is nuclear rather than chemical, converts the fissionable plutonium into nonfissile fission products. Plutonium has previously been used to fuel domestic reactors and currently fuels a number of foreign reactors.

Research, testing, and commercial utilization of plutonium have resulted in the existence of a vast experience base for MOX in general. This irradiation experience provides the foundation on which to build a disposition program. While the plutonium irradiation data base is extensive, the portion that is directly applicable to the FMDP is limited. Most of the plutonium that has been used as fuel is reactor grade (RG), which has a lower fissile isotope concentration than WG material. Furthermore, much of the irradiation experience is treated as proprietary information by the fuel vendors, reactor vendors, and/or utilities that sponsored the irradiations. Under these conditions, accurate projections of the additional irradiation experience that may be required to support the U.S. Nuclear Regulatory Commission (NRC) licensing of MOX fuel use in commercial reactors become more difficult.

Following standard nomenclature, MOX fuel refers only to LWR plutonium-uranium oxide fuel. Other plutonium-uranium oxide fuels are referenced in this report using alternative nomenclature such as plutonium fuels or fast reactor fuels.

From the early days of the nuclear era, plutonium has been recognized by many as a valuable reactor fuel. Although present in some uranium deposits in minute quantities, plutonium is generally viewed as a man-made element, created through neutron capture in uranium. Because LWRs use low-enriched fuel that contains a large fraction of ^{238}U, they breed a significant amount of plutonium during operation, part of which is fissioned in-situ. Early in the nuclear era, it was assumed that the plutonium bred in the LWRs would be used to fuel liquid metal fast breeder reactors (LMFBRs), which can breed additional plutonium more efficiently. Light water burner and converter reactors were viewed as interim options that would eventually be

supplanted by LMFBRs. Nevertheless, plutonium recycle into LWRs was also thought to be practical.

Several government- and industry-sponsored programs in the United States during the 1960s and 1970s demonstrated the feasibility of MOX fuels. Test irradiations of fuel specimens in research and test reactors led eventually to lead test assembly (LTA) irradiations in commercial reactors. However, President Carter's 1977 nuclear nonproliferation policy announcement, which called for indefinite deferral of domestic commercial reprocessing and recycling of plutonium, effectively ended all domestic recycling efforts (Ref. 1).

During the same period, many foreign countries began investigations into the use of MOX fuel. The Belgians irradiated their first MOX test assembly in 1963. Other European countries, including Germany, France, the United Kingdom (UK), Switzerland, Italy, Sweden, and the Netherlands eventually followed suit. A wealth of information about these programs exists in the open literature, and much more is known to exist in proprietary files. Other countries are known to have MOX irradiation experience (Japan, China, India, the FSU), but only limited open literature documentation on their programs has been located. While the United States' unilateral ban on reprocessing ended domestic MOX research in the late 1970s, several of these foreign countries continued to pursue plutonium recycle in LMFBRs and/or LWRs. MOX fuel is now widely used in several countries, and its use is expected to expand rapidly in both France and Japan in the near future.

This document summarizes the MOX fuel irradiation experience and, by necessity, is primarily limited to a review of information that is documented in the open literature. In this report, irradiation experience refers to the irradiation itself and any examinations performed during and after the irradiation. A comprehensive summary of MOX experience cannot be assembled because of the proprietary nature of much of the information. Most of the open literature information consists of descriptions of the material that was irradiated and the irradiation conditions. Results from the postirradiation examination (PIE) programs are in general unavailable.

Chapter 2 of this report summarizes MOX fuel manufacturing technology. Because the irradiation performance depends heavily on the particular manufacturing techniques employed, a basic understanding of the processes is needed to understand the subsequent sections of this report. Chapter 3 contains a description of the domestic irradiation experience, as reported in the open literature. Overseas MOX irradiation experience is then described, country by country, in Chapter 4. The irradiation experience in most cases consists of a description of the fuel that was tested and the test conditions without elaboration on the purpose of the test or the PIE results due to data unavailability. The lessons learned from all the irradiation experience are summarized in Chapter 5. Appendix A consists of a tabular summary of the domestic MOX experience. Appendix B contains a reprinted summary description of the U.S. experience from the Generic Environmental Statement on MOX (GESMO) report. In addition to the list of cited references, an extensive Bibliography is included in Appendix C.

CANDU (Canadian Deuterium Uranium Reactor) reactor experience with MOX fuel is not described in the body of this report. Differences between CANDU fuel and typical LWR fuel render the LWR MOX data base somewhat inapplicable to the CANDU MOX experience.

Atomic Energy of Canada, Limited (AECL), the CANDU vendor, has investigated a number of advanced fuel cycles, including MOX fuel use, for CANDU reactors. As is the case with the other vendors, most of the data resulting from the tests and examinations for CANDU reactors are proprietary.

2. MOX FUEL MANUFACTURING PROCESSES

As with standard urania (UO_2) fuels, the physical characteristics and irradiation performance of MOX fuels depend on the particular manufacturing process used to produce them. A number of processes have been developed for MOX fuels, and they are discussed in this chapter. Although some of the newer processes may be considered "advanced," no firm evidence was located in the open literature to prove that the irradiation performance of these advanced fuels is greatly superior to that of the fuels produced domestically in the 1970s. The advances have in fact focused more on simplification of the manufacturing process and suitability of the product for subsequent recycling than on in-reactor performance improvements. The suitability of the fuel depends on the program requirements. However, the "advanced" fuels in general seem to have lower fission gas release, better dissolution characteristics (important for reprocessing considerations), and more robust ability to withstand power changes.

This MOX fuel manufacturing discussion is not meant as an exhaustive review. It is included as an introduction to provide sufficient information to understand the issues raised in later sections of this report.

2.1 BASIC MANUFACTURING STEPS

The MOX fuel manufacturing process is similar to the UO_2 fuel manufacturing process, with a few additions. In fact, all of the specific MOX processes are adaptations of either standard UO_2 fuel processes, fast-reactor-fuel processes, or a combination of the two. According to ALKEM (Alpha Kemistry and Metallurgy GmbH), the German plutonium fuel manufacturer, the classic UO_2 production process was modified "only where dictated by the peculiar properties of plutonium. Major deviations occur in powder blending, sintering in diluted hydrogen to prevent explosions, outside contamination control of the fuel rods, and autogammagraphy of finished fuel rods" (Ref. 2).

The generic fabrication procedure has been broken down, for the purposes of this discussion, into three stages: oxide production, blending, and fuel consolidation. The first stage often takes place in a separate facility, but it directly affects the powder preparation required in the second stage. Not all of the integrated MOX manufacturing processes employ all three stages. Some, like the coprecipitation processes, combine oxide production and blending into a single step.

2.1.1 Oxide Production

In the existing commercial MOX fabrication plants, the plutonium that is utilized (and in some cases the uranium also) results from Plutonium Uranium Recovery by Extraction (PUREX) reprocessing operations and is in the form of an aqueous nitrate solution. The uranium streams contain uranyl nitrate hexahydrate, $UO_2(NO_3)_2 \cdot 6H_2O$, (UNH). The corresponding chemistry of the plutonium stream is not as simple.

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Multiple plutonium oxidation states are stable. The standard methods of conversion of UNH to oxide must, therefore, be modified for application to plutonium nitrate solutions. The commonly used methods of plutonium oxide production are discussed in this chapter. Four techniques (direct calcination, oxalate, peroxide, and microwave denitration) for converting plutonium nitrate to oxide and two techniques (coprecipitation and Ammonium Uranyl/Plutonyl Carbonate) for coconverting plutonium/uranium mixtures are described.

Most of the surplus plutonium is in the form of metallic weapons parts or pits. Two dry processes are under consideration for conversion of this material to PuO_2. The first is hydride oxidation (HYDOX) in which the metal is contacted with diluted hydrogen to form hydride that is subsequently nitrided and finally oxidized. The second is direct metal oxidation (DMO) in which metal is heated in an oxidizing atmosphere and allowed to burn. The key determinant in the choice of oxide production processes is the suitability of the final oxide powder to the fuel manufacturer. Subsequent powder conditioning may be used to obtain desired powder characteristics, but such conditioning requires additional time, expense, and facilities.

2.1.1.1 Direct Calcination

Calcination is a process in which a material, such as plutonium nitrate solution, is heated to a temperature below its melting point to effect a decomposition, such as oxidation. Direct calcination is the preferred method of conversion from UNH to UO_3 in the United States. It is also the simplest method of converting plutonium nitrate solution to oxide (Ref. 3, p. 443). This method is insensitive to the beginning oxidation state, which is important for plutonium nitrate solutions as several oxidation states, including Pu^{3+}, Pu^{4+}, PuO_2^+, PuO_2^{2+}, PuO_2^{3+}, can exist (Ref. 3, p. 436). Direct denitration of plutonium nitrate solution has been investigated at the lab scale using a continuous liquid phase screw-type calciner (Ref. 4). Other methods of direct calcination include flame calcination and fluidized-bed calcination.

One of the disadvantages of direct calcination is that it provides no appreciable decontamination of the plutonium. Any impurities present in the nitrate solution are carried through to the oxide, so clean feed is required. Also, the calcination temperature and time at temperature must be carefully controlled to obtain desirable oxide powder properties. A sufficiently low calcination temperature must be used to prevent sintering or fusing, which results in a product that is difficult to homogenize in the subsequent milling process.

2.1.1.2 Oxalate Process

The oxalate process is another process for conversion of plutonium nitrate to oxide. Depending on the oxidation state of the plutonium in the feed nitrate solution, one of two oxalate processes can be used (Ref. 3, p. 442–443). In both processes, oxalic acid ($H_2C_2O_4$) is added to the nitrate solution. Depending on the feed nitrate solution, either $Pu_2(C_2O_4)_3 \cdot 9H_2O$ (for Pu^{3+}) or $Pu(C_2O_4)_2 \cdot 6H_2O$ (for Pu^{4+}) is precipitated. The oxalate precipitate is filtered, dried, and calcined to form the oxide. This is considered by many to be the standard conversion process. It combines the advantages of high quality product and plutonium decontamination with formation of relatively safe intermediate compounds and solids. Unlike the direct calcination process, the oxalate process provides some separation between impurities in the nitrate solution and the final oxide product.

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2.1.1.3 Peroxide Process

The peroxide process is a third process for converting plutonium nitrate to oxide. By adding peroxide to plutonium nitrate at low temperature, large crystals of plutonium peroxide can be formed. These crystals are subsequently filtered from the solution, and the filter cake is dried in air and calcined to the oxide. The peroxide process yields better decontamination than the oxalate process, but it does so at the expense of safety. Impurities present in the nitrate feed can catalyze explosive decomposition of peroxide compounds (Ref. 5, p. 557). In its favor, the peroxide process results in excellent decontamination of cationic impurities. Nevertheless, because of the safety issues associated with the impurity-driven peroxide decomposition, the peroxide process has not been used commercially but remains a useful laboratory conversion process.

2.1.1.4 Microwave Denitration

The Japanese Power Reactor and Nuclear Fuel Development Corporation (PNC) and its successor Japan Nuclear Cycle Development Institute (JNC) have developed the microwave denitration process for application to plutonium nitrate or mixed-nitrate feed streams (Ref. 6). Because it was developed primarily to enhance the diversion resistance of the PUREX process, it is most often discussed for mixed-nitrate feed. Microwave denitration is similar in many respects to direct denitration/calcination. The principal difference is the use of microwave heating.

2.1.1.5 Coprecipitation

Coprecipitation is a method for converting mixtures of plutonium nitrate and uranyl nitrate to MOX. Coprecipitation has been pursued for two reasons: the homogeneity of the resulting MOX powder and the potential for use in reprocessing plants in which no separated plutonium stream exists. The principal coprecipitation process is an adaptation of the ammonium diuranate (ADU) process that is commonly used for conversion of uranyl nitrate to oxide. (A similar aqueous process known by the same name is used to convert UF_6 to UO_2.)

In the ADU process, ammonia is added to uranyl nitrate, forming the ammonium diuranate precipitate, $(NH_4)_2U_2O_7$. The precipitate is then filtered, dried, and calcined. In the coprecipitation process, ammonia is added to a dilute solution of plutonium and uranyl nitrates. The ammonia precipitates out both heavy metals simultaneously—the plutonium as plutonium hydroxide and the uranium as ammonium diuranate. The precipitates are filtered from the solution together, and the resulting filter cake is dried and calcined. An alternative is concentration and direct calcination of the slurry in a fluidized-bed reactor (Ref. 7, p. 132).

The resulting powder is a uniform blend of the two oxides, although some powder milling may be required to deliver the required fuel fabrication feed specifications. With coprecipitation, agglomerates of plutonium hydroxide can form, which must be broken up during the milling stage to prevent their occurrence in the finished MOX product. Coprecipitated material may be used directly if the concentrations of plutonium and uranyl nitrate are controlled such that the final plutonium concentration in the oxide is correct. Alternatively, the coprecipitated material

may be blended with additional uranium oxide powder to obtain the desired plutonium concentration.

A coprecipitation process known as COGEPEL has been investigated by Belgonucléaire (BN), although the details of the process are not described in the available reference (Ref. 8). The General Electric Company (GE) also investigated the coprecipitation and calcination (COPRECAL) process (Ref. 9).

2.1.1.6   AUPuC Process

The Ammonium Uranyl-Plutonyl Carbonate (AUPuC) process is another method of coconverting mixed-plutonium nitrate, uranyl nitrate solutions to MOX. The AUPuC process, like coprecipitation, is an adaptation of a uranyl nitrate conversion process. In the ammonium uranyl carbonate (AUC) process, ammonia and carbon dioxide are added to uranyl nitrate to precipitate ammonium uranyl carbonate. (A similar aqueous conversion process known by the same name is used to convert UF_6 to UO_2.) Filtration, drying, and calcination of the AUC precipitate produce oxide powder.

Because of the good physical properties of UO_2 derived from the AUC process (sometimes referred to as ex-AUC material), a similar process that would coprecipitate both uranium and plutonium was sought. The difficulty with such a process is the different oxidation states of the two metals. In mixed plutonium/uranium nitrate solutions, the metals usually exist in the Pu^{4+} and U^{6+} oxidation states. Researchers at ALKEM developed a successful process by oxidizing the Pu^{4+} to Pu^{6+}, then adding ammonia and CO_2 (Ref. 10). Ammonium uranyl/plutonyl carbonate is formed. The crystalline precipitate, $(NH_4)_4(U,Pu)O_2(CO_3)_3$, is filtered and calcined to form the oxide.

The resulting oxide powder has an average particle size of 40 μm. The Germans have used this process to a limited extent to produce feed for their MOX fabrication. However, use of the AUPuC process is dependent on collocated reprocessing and oxide conversion facilities because of restrictions on shipment of plutonium solutions.

2.1.1.7   Hydride Oxidation

Hydride Oxidation (HYDOX) is a dry process under development by Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory as part of the Advanced Recovery and Integrated Extraction System (ARIES). The ARIES system integrates a number of processes (cutting the pits apart, separating the plutonium from other components with gas, converting the plutonium to an oxide form, packaging it in sealed containers, decontaminating and determining the characteristics of the resulting product) necessary to remove plutonium from the cores of surplus nuclear weapons and converts the plutonium into an unclassified form that is suitable for international inspection, long-term storage, and disposition.

The reference HYDOX process is commonly referred to as the 3-step HYDOX process. Plutonium metal is subjected to hydrogen gas that is diluted in an inert carrier gas. Plutonium hydride spalls from the metal surface, exposing unreacted metal. The plutonium hydride is collected and subsequently exposed to nitrogen gas. The hydride is thus converted to plutonium

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nitride. The plutonium nitride is then oxidized through exposure to oxygen gas diluted in an inert carrier gas. The three step HYDOX process avoids the simultaneous use of hydrogen and oxygen gases. PuO_2 produced through HYDOX retains a crystalline structure from the hydride that is slate-like in appearance.

2.1.1.8   Direct Metal Oxidation

Direct metal oxidation (DMO) is the second dry process under consideration for application to the surplus plutonium mission. DMO has been utilized previously for production of feed material for MOX fuel fabrication. It has also been utilized to convert pyrophoric plutonium metal into stable oxide for storage and/or dissolution for subsequent purification.

DMO may be performed in any oxidizing atmosphere. The complexity of a DMO system can vary from a heated reaction vessel to a sealed vessel with temperature and atmospheric control. Unalloyed plutonium metal will oxidize slowly even at room temperature, especially in the presence of moisture. More rapid oxidation occurs above about 300°C.

It has been demonstrated that moisture enhances the oxidation process, so the reactant gas in a DMO system is usually bubbled through water. The water is best described as a catalyst. It reacts with the surface plutonium metal to release hydrogen that is retained in the metal as plutonium hydride. As additional oxygen is made available, this hydrogen is released for reaction with additional metal. A hydride reaction front thus moves through the metal, followed by an oxide front. As a result, PuO_2 produced through DMO is similar in its crystalline makeup to PuO_2 produced through the HYDOX process.

DMO has also been tried in dry air. However, ignition under dry conditions requires higher temperatures. Some experimenters also describe the reaction as more difficult to control under dry conditions.

Most of the experiments performed with DMO suggest that relatively high surface area is required in the feed to obtain reasonable reaction rates at the low temperatures (< 500°C) that are desirable. The chemical reactivity of the product is affected by the reaction temperature. At low reaction temperatures, residual metal, hydride, or hydroxide can be found in the product. At higher reaction temperatures, large sintered particles known as clinkers can be produced.

2.1.2   Oxide Blending and Milling

In most reprocessing plants, plutonium nitrate is converted to oxide powder. This PuO_2 must be subsequently blended with UO_2 for fabrication into MOX fuel. The blending process is very important because the presence of large particles or agglomerates of PuO_2 (especially those $\geq 100\,\mu m$) has been found to be detrimental to both performance of the fuel under irradiation and subsequent solubility of the irradiated fuel in nitric acid. Even those oxide-production processes that combine the plutonium and uranium nitrate solutions (coprecipitation, AUPuC) often require some blending and/or milling to produce MOX that meets the particle size specifications. Several methods of blending/milling have been used for MOX production, and each is discussed in this chapter.

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2.1.2.1 Comilling

Comilling is the "reference" process for blending PuO_2 and UO_2 powder and for adjusting the size of the particles and/or agglomerates in the MOX powder. The PuO_2 powder produced using one of the methods discussed in Section 2.1.1 is placed in a mill along with UO_2 powder. The UO_2 powder is typically produced from either the ADU, AUC, or direct dry-conversion process, depending on the fuel manufacturer. The two powders are milled together for up to 72 hours to ensure good homogeneity and to reduce the average particle size.

Although comilling was used until recently (with satisfactory results according to the manufacturers), fuel produced using this technique is often not as homogeneous as that produced by other methods. The milling time required to obtain a homogeneous product in a ball mill may be as long as tens of hours. This milling time and the associated pickup of metallic impurities from the milling media places a practical limitation on the achievable homogeneity. Based on the limited open literature information, the irradiation performance of this comilled fuel has not differed greatly from that produced from other methods.

One potential problem with comilled fuel is low solubility in nitric acid. Low solubility can cause large losses in subsequent reprocessing, although this is not detrimental for the WG plutonium disposition mission. Another difficulty with comilling is the large energy requirement that results from the need to mill all the MOX powder and from the inefficiency of the ball mill. Advances in comilling have focused on different types of mills that achieve better homogeneity in a shorter time. The hammer mill, jet mill, and high energy attritor mill have been investigated as potential improvements over the ball mill.

The ability of a mill to comminute MOX powder is limited by the buildup of electrostatic charges in the powder. These charges in effect create a practical limit of a few microns on the achievable particle size. Addition of a milling aid is necessary to comminute below this size. Zinc stearate $[Zn(CH_2=(CH_2)_5COO)_2]$ is commonly utilized as a milling aid. A better milling aid is 1,3-propane diol $[HOCH_2-CH_2CH_2OH]$, which is distributed better than zinc stearate because of its higher vapor pressure. Addition of milling aids allows comminution to proceed to submicron particle sizes.

2.1.2.2 Preparation of a Master Mix or Blend

As discussed above, one of the disadvantages of comilling is an intensive milling requirement. One method of oxide blending developed to help alleviate this concern is known as either master mix or optimized comilling. In this process, approximately 20% of the final MOX powder, including all the plutonium, is micronized in a mill. This creates a homogeneous master mix that is approximately 30% plutonium. The milling effort is thus reduced approximately by a factor of five. The homogeneous MOX powder is subsequently mixed with additional UO_2 powder to generate the final MOX blend. Large agglomerates of the master blend are not as detrimental to fuel performance as are large agglomerates (or particles) of PuO_2, because the master blend agglomerates are 70% UO_2.

An additional benefit of master-mix preparation is the ability to utilize the free-flowing properties of the diluent UO_2 to avoid granulation. UO_2 produced via the AUC process is free-

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flowing. If the quantity of master mix is limited to 20% or so of the final blend, the free-flowing properties of the UO_2 dominate the behavior of the final blend such that it is also free-flowing. The optimized comilling (OCOM) and AUPuC processes developed by ALKEM (now Siemens) are based on this principle.

A similar process can be used with either coprecipitated or AUPuC-derived MOX powder. The coprecipitated or AUPuC material is homogeneous but often requires some milling to achieve the desired particle sizes. The high-plutonium-concentration powder can be milled and then later blended with free-flowing UO_2 powder to achieve the final desired plutonium assay.

2.1.3   Fuel Constitution

Fuel constitution comprises a series of steps in which MOX powder is conditioned, formed into cylindrical pellets, sintered, ground to size, clad, and compiled into completed fuel assemblies. Most of these MOX operations are based on the corresponding operations utilized for production of LEU (low enriched uranium) fuel.

Once a homogeneous mixture of MOX powder with the desired plutonium assay has been obtained through milling and blending, the MOX powder must be conditioned for pellet pressing. Press feed must be free-flowing to ensure complete, uniform, and repeatable filling of the die cavity. Most MOX feed powders must be conditioned because the milling step for homogenization renders them too fluffy to flow. The flowability is readily achieved through granulation of the fine powder into larger agglomerates. Three techniques for granulation have been utilized: wet binder addition, mechanical granulation, and self-agglomeration.

Wet binder addition, which is no longer commonly practiced with MOX, is described because it was utilized for fabrication of some of the early MOX fuels. In wet binder addition, an organic binder suspended in a solvent is mixed with the MOX powder forming a slurry. The slurry is then spray-dried or evaporated into a paste and finally into coarse granules.

In an effort to streamline the MOX fabrication process, alternatives to wet binder addition were developed. One of the alternatives, mechanical granulation, may be accomplished in one of several ways. In one granulation technique referred to as slugging, the powder blend is pressed at a low pressure (to 30%–50% theoretical density) into large compacts or slugs. These are then crushed and screened to form granules. The slugs can be produced in a high speed pellet press or even in a roll compactor (Ref. 11). In some processes the slugging is eliminated through use of forced sieving. Forced sieving uses a blade to force the powder through the screen, in effect pressing and sizing it simultaneously. An organic binder such as polyvinyl alcohol (PVA) is commonly utilized to increase the strength of the granules regardless of their method of fabrication.

A further refinement to the fabrication process is elimination of mechanical granulation in lieu of self-agglomeration. As described in Section 2.1.2.1, finely ground MOX powder will self-agglomerate due to the buildup of electrostatic charge in the powder. This is the root cause of the lower limit on achievable particle sizes in a mill without the addition of a milling aid. Self-agglomeration is enhanced through tumbling of the powder. Tumbling can be accomplished either in specialized equipment such as the British Nuclear Fuel Limited (BNFL) spheroidizer, or

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through judicious design of the mill itself such that milling and agglomeration are possible in a single vessel. If a volatile milling aid is utilized, one can thus comminute the MOX powder below 1 μm for good homogenization, and then eliminate the milling aid through volatilization resulting from the powder heating caused by the input of milling energy.

Following production of a free-flowing MOX powder, die lubricant may be added to the press feed or added directly to the die. The powder is then pressed to > 50% theoretical density (TD) to form green pellets. If a binder has been added, the green pellets are subjected to a binder removal treatment as part of the sintering cycle. Sintering to temperatures as high as 1750°C increases the pellet density to between 93%–96% TD.

Incomplete die filling and wall friction result in nonuniformity in the density distribution in green pellets. These nonuniformities are removed during sintering, but necessarily result in geometric distortion of the cylindrical pellets. Sintered pellets take on the shape of an hourglass. This geometric distortion is removed through centerless grinding. Criticality concerns usually restrict the use of water as a grinding coolant/lubricant, so most MOX pellets are dry ground. Cleaned pellets are then formed into fuel column stacks and loaded into fuel cladding tubes. Fuel pin assembly is completed through attachment of the upper end cap, pressurization with helium, and seal welding.

2.2 INTEGRATED COMMERCIAL PROCESSES

The leading commercial MOX-fabrication processes are described in greater detail in this chapter. These processes are (1) Comilling, (2) Micronized Master Blend (MIMAS), (3) Optimized Comilling (OCOM), (4) AUPuC, (5) Short Binderless Route (SBR), and (6) particle compaction. While the basic flowsheets for the processes are available in open literature, details about individual process steps are proprietary and unavailable.

It is believed that MOX fuel manufactured by any of the leading pellet processes could be irradiated satisfactorily in U.S. LWRs, although the effort required to license the fuel would depend on the particular process and on the willingness of the fuel manufacturer to share (or sell) the corresponding irradiation data base for similar fuels.

2.2.1 Reference Comilling

This is the integrated MOX-production process that was used previously by the domestic fuel manufacturers. It was also used previously by several foreign fuel manufacturers, especially for LMFBR fuel. The term comilling derives from the method of oxide blending/milling, described in Section 2.1.2, that is utilized. PuO_2 and UO_2 powders are added to a mill in the correct concentrations to yield the desired fissile concentration. Either natural, depleted, or recycled UO_2 may be used. The powders are milled together for several hours—typically in a ball, jet, or hammer mill—to reduce the average particle size and to ensure uniformity in the MOX powder. It is important to reduce the maximum plutonium particle or agglomeration to less than 100 μm to prevent hot spots in the fuel. The blended powder is usually mixed with an organic binder. Slugging and granulation are then used to form the desired free-flowing MOX granules, suitable for introduction to the pellet press. The green pellets are sintered and centerless ground to the

final dimensions. All of the other pellet fabrication processes may be considered variations of comilling. One set of improvements is based on use of advanced powder mills. Hammer mills, vibromills, jet mills, and attritor mills have been used to reduce the required milling time.

BN produced several thousand fuel rods using variations of the comilling process. The earliest variation is a textbook example of comilling in which fine, comilled MOX powder is granulated into press feed. Subsequent attempts were made to simplify the fabrication process. To reduce the amount of handling of fine plutonium-bearing powders, PuO_2 was blended (not milled) into granulated UO_2. This simplified the fabrication process, but the microstructure produced was unsatisfactory. In the final evolution of comilling at BN known as the "Reference Process," PuO_2 was blended into free flowing UO_2 produced via the AUC process.

ALKEM also utilized variations of the comilling process during early development. Over 9000 MOX fuel rods were fabricated using their "Former Standard" process. ALKEM's experience was similar to BN's in that the fuel performance was adequate while the powder handling aspects of the fabrication and the solubility of the product were not. Two advanced processes, OCOM and AUPuC, were developed to overcome these difficulties.

The French used a variation of the Reference Comilling process in COGEMA's (Compagnie Générale des Matières Nucléaires) CFCa (le Complexe de Fabrication des combustibles au plutonium de Cadarache) MOX fabrication facility located at the Cadarache Nuclear Center until quite recently. One of the fabrication lines at the plant was converted from production of liquid metal reactor (LMR) fuel to MOX fuel in 1989. Due to its LMR heritage, the plant utilized a variation of the reference comilling process that is known as the COCA (Cobroyage-Cadarache) process. The flow sheet for this process is given in Figure 1 (Ref. 12, p. 71). During 1995, the CFCa facility converted to the MIMAS process described in the following section.

2.2.2   Micronized Master Blend (MIMAS) and Advanced MIMAS

The MIMAS process is the latest integrated MOX production process developed and used by BN. BN's MOX-manufacturing development is summarized in Table 1 (Ref. 13, p. 14). Early fuel was produced using one of several variations of the reference comilling process described in Section 2.2.1. As indicated in the table, BN used their "reference" process for MOX manufacture in their Dessel fabrication plant, P0, from 1973 through 1984. While the irradiation performance of this fuel was claimed to be satisfactory by BN, the fuel was unsatisfactory in that it lacked similarity to UO_2 fuel, and in that the plutonium it contained was not sufficiently soluble during reprocessing. Up to 5% of the plutonium remained undissolved, even in boiling nitric acid. It was recognized at the time that large plutonium particles might be responsible for this behavior, because pure PuO_2 is difficult to dissolve in nitric acid, while the solid solution of UO_2 and PuO_2 is not. The MIMAS process, developed in 1984, is an attempt to remedy difficulties with the "reference" fuel.

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┌─────────────┐      ┌─────────────┐      ┌──────────────────┐
│    PuO₂     │      │    UO₂      │      │ Dry recycled scrap│
└──────┬──────┘      └──────┬──────┘      └─────────┬────────┘
       │                    │                       │
       └────────────────────┼───────────────────────┘
                            │
                  ┌─────────▼──────────────┐
                  │ Weighing and lot       │
                  │ preparation            │
                  └─────────┬──────────────┘
                            │
                  ┌─────────▼──────────────┐
                  │     Ball milling       │
                  └─────────┬──────────────┘
                            │
                  ┌─────────▼──────────────┐
                  │    Forced sieving      │
                  └─────────┬──────────────┘
                            │
                  ┌─────────▼──────────────┐
                  │ Lubricant and porogen  │
                  │ addition               │
                  └─────────┬──────────────┘
                            │
                  ┌─────────▼──────────────┐
                  │    Pellet pressing     │
                  └─────────┬──────────────┘
                            │
                  ┌─────────▼──────────────┐
                  │       Sintering        │
                  └─────────┬──────────────┘
                            │
                  ┌─────────▼──────────────┐
                  │       Grinding         │
                  └─────────┬──────────────┘
                            │
                  ┌─────────▼──────────────────┐
                  │ Pellet visual inspection    │
                  │ and selection               │
                  └─────────┬──────────────────┘
                            │
                  ┌─────────▼──────────────┐
                  │    Rod fabrication     │
                  └────────────────────────┘
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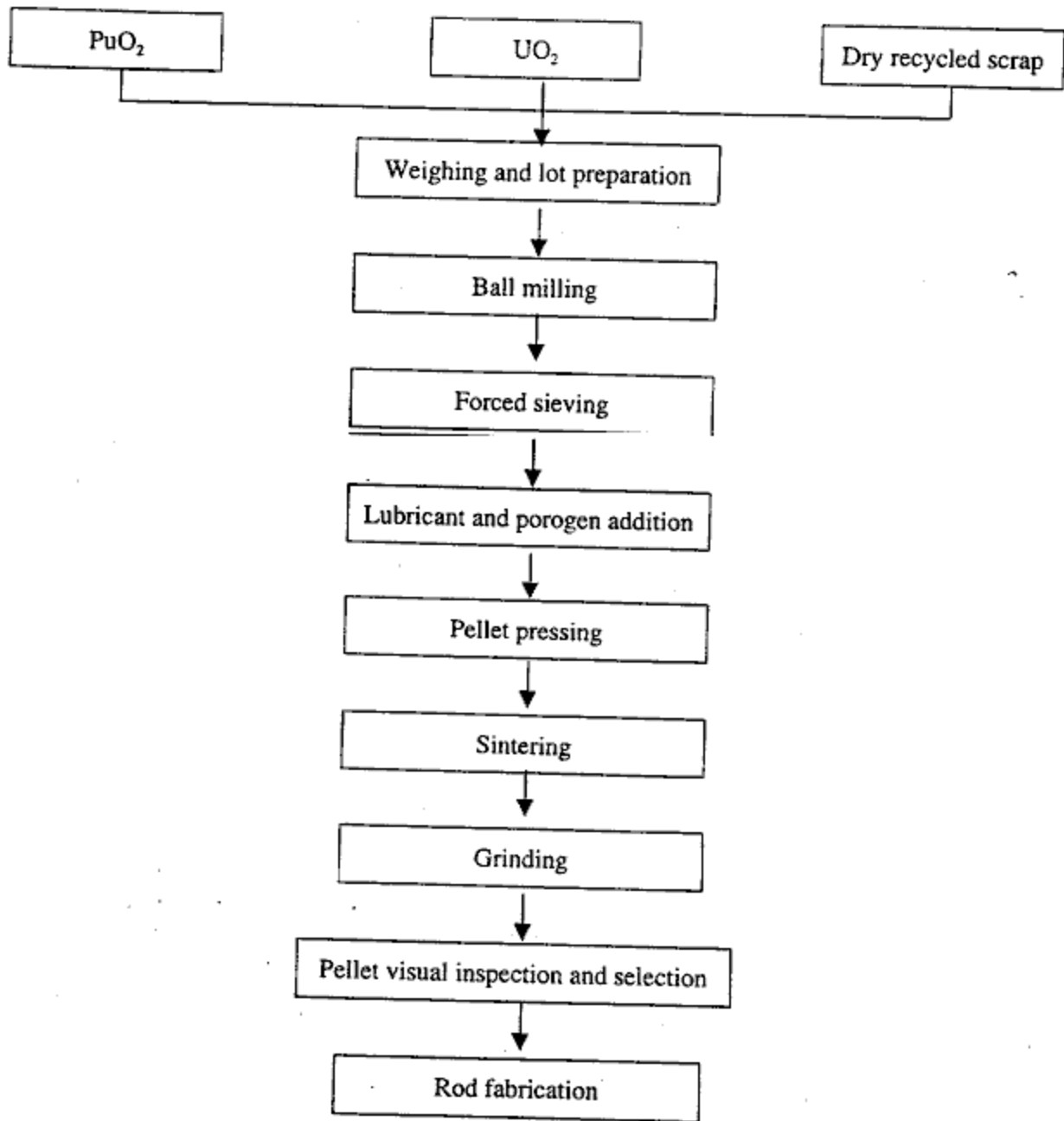
Figure 1. COCA MOX fabrication flow sheet.

Table 1. Evolution of the MOX fuel fabrication techniques of BN

Fuel Type	Period	Advantages	Disadvantages
Granulated $(UO_2 + PuO_2)$ blend	1960–1962 + 1965–1969 (laboratory)	- Assumed best similarity to UO_2 fuel	- Contamination levels, personnel exposure, and waste resulting from complex handling of fine powder
PuO_2 blended into granulated UO_2	1967–1975 pilot facility	- Simplified handling of fine powder	- Significant departures from UO_2 fuel behavior - Unfavorable thermal conductivity - High fission gas release - Large Pu-rich agglomerates
"Reference," i.e., PuO_2 blended into free-flowing UO_2	1973–1984 (fabrication plant)	- Fuel microstructure governed by UO_2 matrix microstructure	- Occurrence of Pu-rich agglomerates - Too large proportion of the Pu in insoluble residues (reprocessing problems)
"MIMAS," i.e., mixing of free-flowing UO_2 and a micronized $(UO_2 + PuO_2)$ primary blend	1983–present (fabrication plant)	- Same advantages as the "reference" MOX and applicability of its data base - Disappearance of the Pu-rich agglomerates issue due to dilution - Resolution of the reprocessing issue	

In the MIMAS process, a PuO_2 - UO_2 mixture of approximately 20%–30% plutonium is prepared. This mixture is milled or micronized, forming a fine uniform powder that is not free-flowing. This master mix is then diluted and mixed with free-flowing ex-AUC UO_2 to the final plutonium concentration. The overall flow sheet is shown in Figure 2 (Ref. 8, p. 66). By micronizing only the master mix, the milling energy requirements may be reduced by a factor of five or more, depending on the final plutonium concentration of the fuel. Also, the addition of a large fraction of free-flowing ex-AUC UO_2 results in a free-flowing MOX powder that is suitable for direct pellet pressing without binder addition or granulation. The resulting pellets consist of small plutonium-rich regions interspersed throughout a UO_2 matrix. This distribution more closely resembles that found in irradiated UO_2 fuel. Thus, the irradiation behavior is quite similar to that of UO_2 fuel. Furthermore, the dissolution characteristics of MIMAS fuel are
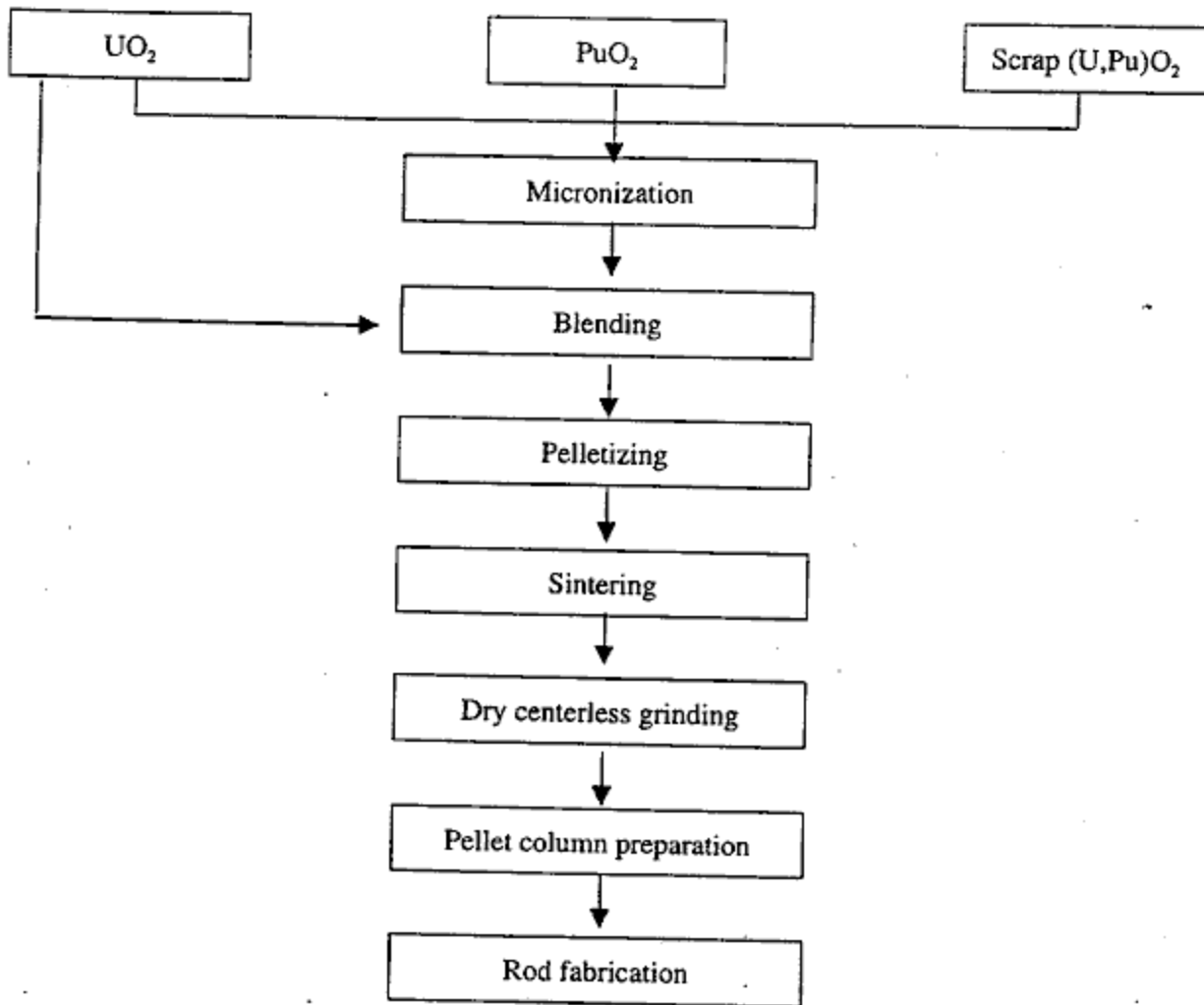
Figure 2. MIMAS flow sheet.

satisfactory to fuel reprocessors. While agglomerates of the master mix are encountered due to the self-agglomeration tendency of the micronized material, these agglomerates are more easily accommodated than pure plutonium agglomerates because of the diluting UO_2 in the master-mix particles.

COGEMA has adopted BN's MIMAS process for production of MOX fuel in France. The process as implemented by COGEMA is known as the advanced MIMAS process but is in most respects identical to BN's original. Advanced MIMAS has been implemented in the new MELOX plant and has been retrofitted into the CFCa plant at Cadarache. One important distinction is the use of UO_2 powder from the TU2 plant rather than the ex-AUC UO_2 utilized originally by BN (Ref. 14, p. 50). TU2 UO_2 is described by the developers as a modified ADU process that produces free-flowing product. Another difference is the use of forced screening of the MOX blend to produce press feed. In addition to allowing use of nonflowable UO_2, inclusion of forced screening allows inclusion of more primary blend in the final blend. Although forced

screening is not considered granulation by COGEMA, the net effect of forced screening is production of loosely held agglomerates. The agglomerates thus produced are harder than those produced by self-agglomeration, but softer than those produced through pressing and crushing.

2.2.3 Optimized Comilling (OCOM)

MOX research in Germany began in 1965. Early fabrication efforts utilized variations of Reference Comilling for pellet production. This "Former Standard" process produced satisfactory fuel that exhibited irradiation performance and reliability roughly equivalent to that of contemporaneous UO_2 fuel. However, as with BN's "Reference Process," fuel produced through the "Former Standard" process was found to be somewhat insoluble in nitric acid. The influence of reprocessors forced evolution of the MOX fabrication process.

The OCOM (or OKOM) process was developed by ALKEM as an improvement to the "Former Standard" process. It is similar in many respects to the MIMAS process. A primary blend is prepared by dry ball milling UO_2, PuO_2, and dry recycle MOX. The plutonium content of this primary blend is maintained at the upper end of the range in which uranium and plutonium form a solid solution ($\geq 30\%$). This primary blend is then diluted with an eight to ten fold larger quantity of free-flowing ex-AUC UO_2. The primary blend is tumbled prior to its dilution to produce free-flowing granulate via self-agglomeration. The agglomerates are allowed to grow to a size that roughly corresponds to that of the diluent ex-AUC UO_2 to enhance the dry blending. By restricting the primary blend to <15% of the secondary blend, the ceramic properties of the ex-AUC UO_2 are retained. This allows direct pressing, with direct die lubrication, without prior granulation. The OCOM flowsheet is shown in Figure 3 (Ref. 15, p. 178).

The key to OCOM is the reliance on the ceramic properties of ex-AUC UO_2. This same material has been utilized to produce many tons of LEU fuel. OCOM MOX fuel is similar in microstructure to this LEU, and has similar behavior under irradiation. The primary blend agglomerates form a solid solution during sintering so that the fuel mass itself has acceptable solubility in nitric acid. These agglomerates are also sufficiently diluted with UO_2 that larger particles of primary blend are acceptable than are acceptable for pure PuO_2 particles.

2.2.4 AUPuC-Integrated Process

The AUPuC process, named after the particular method of nitrate-to-oxide conversion utilized, helps to address reprocessor concerns of MOX solubility.

Because of the favorable experience with UO_2 derived from the AUC process, development of a similar process that would coprecipitate both uranium and plutonium was begun. One of the difficulties with such a coprecipitation is the different oxidation states of uranium and plutonium in nitrate solution. The uranium usually exists in the U^{+6} state, while plutonium is usually in the Pu^{+4} state. The approach was to adjust the oxidation state of either the uranium or plutonium prior to precipitation. Difficulties were encountered with reducing uranium from U^{+6} to U^{+4}, because U^{+4} in turn reduces Pu^{+4} to Pu^{+3}. Therefore, the plutonium is oxidized from Pu^{+4} to Pu^{+6}. This difficult valence adjustment must be complete to produce a filterable product. Residual Pu^{+4} causes precipitation of fines that cannot be filtered easily.

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          AUPuC Process                              OCOM Process

  ┌──────────────────────────────┐    ┌────────┐ ┌──────────┐ ┌──────────────┐
  │Uranium nitrate (60%) Plutonium│    │  UO₂   │ │PuO₂ (30%)│ │ Dry MOX scrap│
  │       nitrate (40%)          │    └────┬───┘ └────┬─────┘ └──────┬───────┘
  └──────────────┬───────────────┘         │          │              │
                 │                          └──────────┼──────────────┘
       ┌─────────┴─────────┐                           │
       │  Coprecipitation  │                            │
       └─────────┬─────────┘                ┌───────────┴────────────┐
                 │                           │ Comilling (micronization)│
  ┌──────────────┴───────────────┐          └───────────┬────────────┘
  │Calcination to (U,Pu)O₂ powder │                      │
  └──────────────┬───────────────┘          ┌────────────┴───────────┐
                 │                           │ Blending 1:8 with ex-AUC UO₂│
  ┌──────────────┴───────────────┐          └────────────┬───────────┘
  │  Blending 1:10 with ex-AUC   │                       │
  │           UO₂                │                        │
  └──────────────┬───────────────┘                       │
                 └────────────────────┬──────────────────┘
                            ┌──────────┴──────────┐
                            │      Pressing       │
                            └──────────┬──────────┘
                            ┌──────────┴──────────┐
                            │      Sintering      │
                            └──────────┬──────────┘
                            ┌──────────┴──────────┐
                            │      Grinding       │
                            └──────────┬──────────┘
                            ┌──────────┴──────────┐
                            │   Rod fabrication   │
                            └─────────────────────┘
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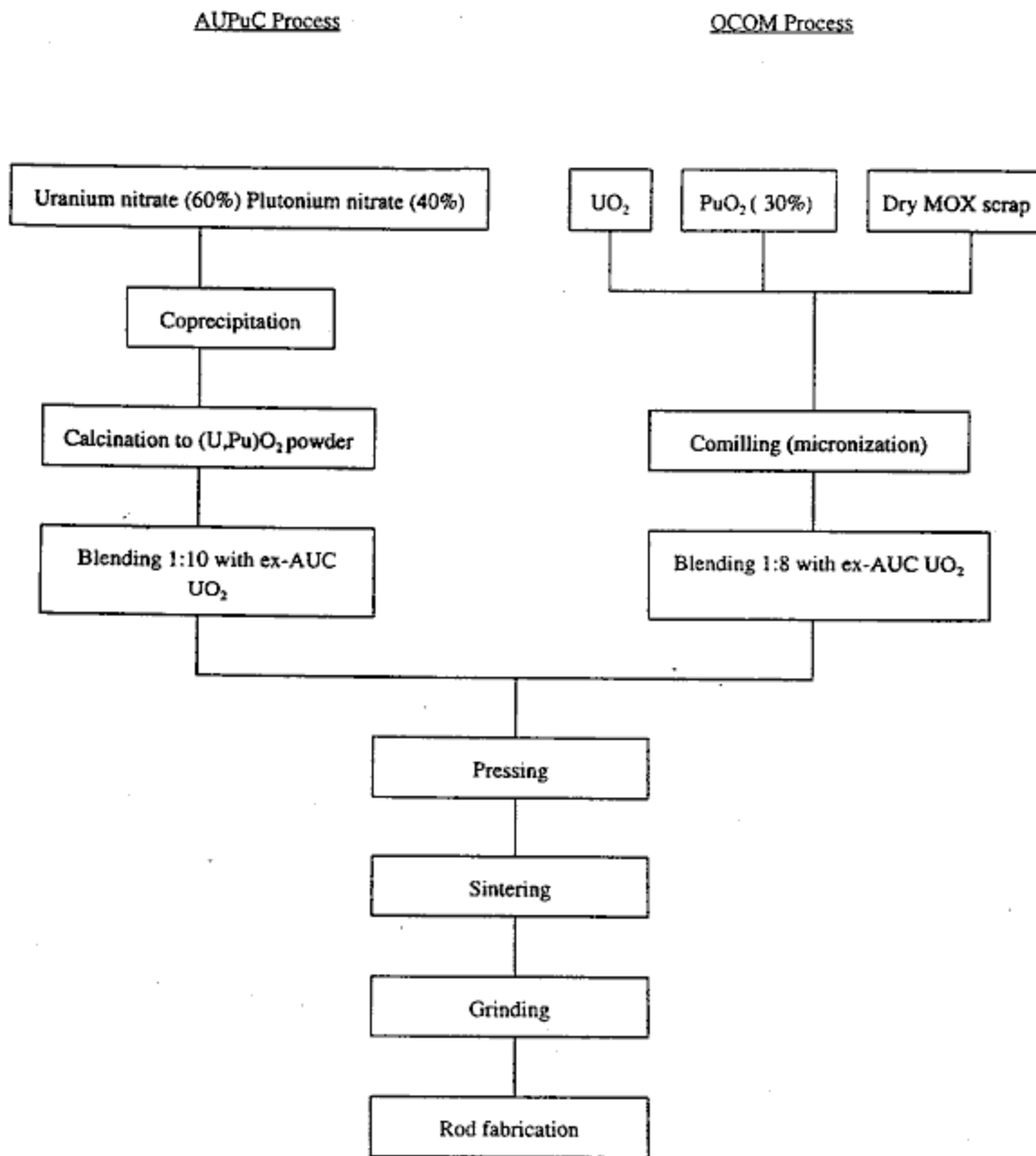
Figure 3. Flow sheets for AUPuC and OCOM MOX fabrication processes.

With both uranium and plutonium in the $(U,Pu)^{+6}$ state, ammonia and CO_2 are added to the solution. A mixed uranium and plutonium complex, ammonium uranyl/plutonyl carbonate or $(NH_4)_4(U,Pu)O_2(CO_3)_3$, is formed. This crystalline precipitate is filtered, dried, calcined, and reduced (Ref 10). The resulting MOX powder is treated in much the same way as the master

mix in the OCOM flow sheet. Free-flowing ex-AUC UO_2 powder is added to the MOX powder to dilute it to the desired plutonium concentration. The resulting free-flowing powder is sent directly to the pellet press. The AUPuC-process flowsheet is shown in Fig. 3 (Ref. 15, p. 178).

Fuels were first produced using the AUPuC process in 1981. According to ALKEM, the irradiation performance of these fuels was satisfactory, and the solubility in nitric acid was much better than that of the "Former Standard" fuel. However, use of the AUPuC process requires the availability of plutonium nitrate solution, and the lack of collocated reprocessing and MOX plants has prevented widespread use of AUPuC.

2.2.5 Short Binderless Route (SBR)

The SBR is the integrated MOX-manufacturing process used by BNFL in their MOX Demonstration Facility (MDF), which has a nominal capacity of 8 MT/yr. This demonstration plant and the large-scale follow-on Sellafield MOX Plant (SMP), expected to come on-line in mid-1999 with a nominal capacity of 120 MT/yr, use a modified version of the comilling process described in Section 2.2.1 that is known as the SBR. The United Kingdom Atomic Energy Authority (UKAEA) and its successor, BNFL, have manufactured MOX since the early 1960s. However, a large portion of this MOX, especially that produced during the 1970s and 1980s, was fuel for the Dounreay fast breeder reactor. As the prospects for a fast breeder reactor economy dimmed in the mid- to late 1980s, interest in LWR MOX use expanded. BNFL and the UKAEA combined their MOX expertise and developed the SBR process as a collaborative effort.

Short in SBR refers to the time required for comilling. All the MOX powder, including all the UO_2 required to adjust the plutonium concentration to its final value, is milled together. However, instead of a conventional ball mill, a high energy attritor mill is used. The attritor mill is more efficient, allowing the entire powder charge to be milled to a suitably small size (maximum plutonium agglomerate size <100 µm with the average size less than 30 µm) while ensuring homogeneity of the MOX powder. Milling times may be reduced from 3–4 hours to approximately 30 minutes. Although details about the mill and spheroidizer are proprietary, they are shown diagrammatically in Figure 4 (Ref. 16, p. 175). The mill consists of a fixed outer casing, a rotating paddle assembly, and a ball charge.

The *binderless* term refers to the absence of organic binder addition and slugging/granulation. The milled powder coming from the attritor mill is not free-flowing and has a strong tendency towards self-agglomeration. It is, therefore, sent to a spheroidizer, the details of which are proprietary to BNFL. As suggested in Figure 4, the spheroidizer apparently tumbles the fine MOX powder to enhance self-agglomeration. Through control of milling aids and moisture content, similar processes have been used to produce soft agglomerates that are free-flowing and suitable for direct compaction.

The attritor mill and spheroidizer are vertically oriented. They both utilize fixed, static cases. This arrangement greatly reduces the risk of alpha contamination during transfer of the MOX powder from the mill through the powder conditioner to the pellet press.
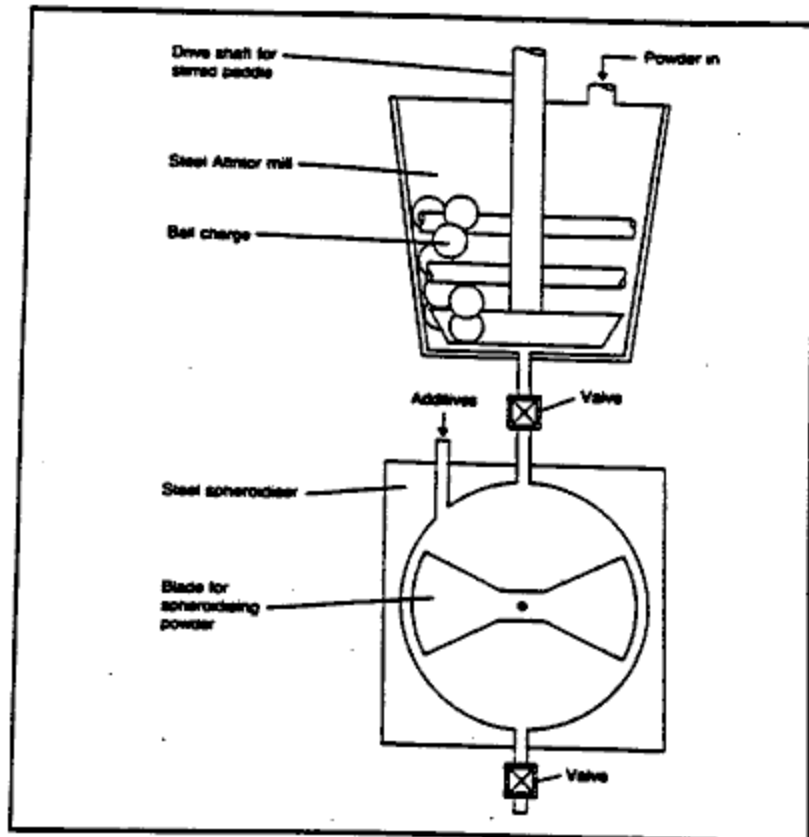
Figure 4. Attritor mill and spheroidizer used in BNFL's SBR.

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BNFL claims extremely good homogeneity with their SBR. The autoradiographs used in their public relations and sales brochures support this claim. Although their fuel specifications are proprietary, the maximum plutonium particle (or agglomerate) size allowed is on the order of 100 μm. The average sizes experienced have been less than 20–30 μm (Ref. 17). This is better than the reported experience with Master-Mix-type fuels. However, this comparison may not be appropriate. SBR agglomerates may consist of pure PuO_2, whereas master-mix agglomerates include only 20%–40% plutonium.

2.2.6   Particle Fuel Technology

Although the vast majority of MOX fuel has been produced in pellet form, some alternative fabrication technologies utilizing particle fuel have been attempted. Many particle fuel fabrication processes have been investigated. The more important of these are described in this section. These processes all differ from those described in Sections 2.2.1–2.2.5 in that fuel pellets are not produced. Rather, fuel particles are loaded directly into the cladding tube prior to compaction. These particles are then compacted through one of several techniques directly in the cladding tube. Particle fuel technologies have been investigated because of their promise of greatly simplified fuel fabrication, which translates to lower cost and adaptability to remote operation. As pellet fuels gained dominance during the 1960s, interest in particle fuel technology waned. Nevertheless, limited particle fuel research continues to this day.

One of the most studied of these particle fuel fabrication processes is vibratory compaction (VIPAC). Fully dense feed material (i.e., produced through arc melting and crushing) is separated into 3–5 size ranges by screening. Precise amounts of each size fraction are then mixed to form a feed of known particle size distribution. This feed is then poured into a cladding tube. Vibration, often with the help of a static load placed atop the powder, is then utilized to compact the powder. Smear densities as high as 80% TD are readily achievable. An improved VIPAC process developed by the Research Institute of Atomic Reactors in Dimitrovgrad, Russia, reportedly produces smear densities as high as 88% TD. This is nearly as high as the smear density of a pellet fuel rod, taking into account the dish, chamfer, and fuel/clad gap.

Another particle fuel fabrication type often referenced in early MOX fuel literature is swage compaction. In swage compaction, particles are loaded into an oversize cladding tube, which is then sealed with an upper end cap. The entire fuel rod is then swaged (mechanical reduction in diameter) to the desired fuel pin diameter. In the process, the particle fuel is compacted. Development of this process was abandoned based on both fabrication and irradiation difficulties.

One other important particle fuel technology is sphere compaction (SPHEREPAC). Microspheres produced by sol-gel techniques are used as feed to a VIPAC process. The primary advantage of SPHEREPAC is its elimination of dusting processes. Limited research on SPHEREPAC continues to this day.

3. DOMESTIC MOX IRRADIATION EXPERIENCE

One of the primary difficulties with assessing the state of the irradiation experience base in the United States is that the information is somewhat dated, sometimes incomplete, and spread among a variety of historical documents. A number of different fuel manufacturing processes were tried by U.S. fabricators in test reactors, ultimately leading to the selection of oxide pellets for use in MOX LTAs and even in partial core loads in commercial reactors. Many of the deficiencies that were identified by earlier irradiation tests have since been resolved. The advantage of the U.S. MOX documentation is that much of it is publicly available.

This chapter attempts to provide as complete a summary of the domestic MOX-irradiation experience base as can be developed from the open literature. The experience can be categorized by fuel vendor or by the reactor that irradiated the fuel. In this chapter, a historical description of the irradiation experience is given by fuel vendor. This is somewhat confusing in several cases, because while one vendor designed the fuel, a second vendor fabricated the fuel. For example, some of the MOX fuel irradiated in the Garigliano boiling water reactor (BWR) was designed by GE, but manufactured by BN. This experience is primarily attributed to BN, but is also described under GE's experience.

Appendix A has the U.S. MOX experience categorized by the reactor that irradiated the MOX. This appendix contains a description of the methodology that was used and the table that contains detailed data for the irradiations. In this approach, an emphasis was placed on the rods that were nondestructively and/or destructively examined. The table identifies approximately fifty characteristics of the irradiation. The major areas covered are reactor specifics, MOX assembly design and fuel isotopics, fabrication techniques, a summary of the examinations conducted, and miscellaneous fuel performance observations. The table also contains references that support the data so that the reader can find additional information. The advantage of categorizing irradiations by plant is that a uniform cross-comparison of the characteristics of the irradiation in different U.S. reactors can be made. The table also contains some detail that is not provided in the discussion below. The table does not include the Plutonium Recycle Test Reactor (PRTR), Materials Test Reactor (MTR), Engineering Test Reactor (ETR), or Experimental Boiling Water Reactor (EBWR) irradiations, but these are discussed below.

3.1 U.S. GOVERNMENT DEVELOPMENT

The United States Atomic Energy Commission (AEC) began plutonium recycling studies in 1956. Its efforts were concentrated in two programs, the Plutonium Utilization Program (PUP) managed by the Battelle Pacific Northwest Laboratories (BNWL) and the Saxton Program managed by Westinghouse (W). Each of these programs is described in the following two subsections.

In the early 1970s, the AEC decided that additional government support of plutonium recycling was not justified because private industry was fully capable of commercializing the technology. One of the best summaries of the AEC work, and other early plutonium recycle work, is located

in the *Draft Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in LWRs, WASH 1327* (Ref. 18, Vol. 2, pp. II-34 through II-60) and the follow-on *Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors, NUREG-0002* (Ref. 19, Vol. 2, pp. II-11 through II-40). The latter summary is reproduced for the convenience of the reader in Appendix B. In addition to the PUP and Saxton programs, the AEC participated to a limited extent in commercial and international plutonium recycle programs.

3.1.1   Plutonium Utilization Program (PUP)—Early Test Reactor Irradiations

The PUP began in 1956 at the BNWL and was funded by the AEC. The purpose of the program was to develop the necessary technology to implement plutonium recycle in thermal reactors. The time frame of the program must be kept in mind during review of the program results, because the nuclear industry was in its infancy during the first half of the program. Tests were performed in the MTR, the ETR, the EBWR, and the PRTR. Some of the capsule tests performed in the MTR, ETR, and PRTR are described in Table 2 (Ref. 20, p. 673).

The bulk of the testing under the PUP was performed in the PRTR, which was built specifically for this purpose. This reactor is heavy-water moderated and cooled, with a thermal power of 70 MW (Ref. 21, p. 126). The reactor fuel elements, which consisted of groups of rods strapped together, were contained in vertical pressure tubes. The reactor design is similar in many respects to the modern CANDU design. The reactor also included the Fuel Element Rupture Test Facility (FERTF), in which "high-risk" experiments could be performed without risking contamination of the entire system. The FERTF included its own coolant system and was used for tests such as the intentionally defected fuel tests.

At the time of PRTR construction and during the early years of its operation, it was not clear that the now common, cold-pressed and sintered (CPS) pellet fuels would dominate the nuclear industry. Thus, testing under the PUP did not focus on CPS fuel. In fact, most of the testing was for other fuel types including hot-pressed pellets, swage-compacted, and VIPAC. The nonpellet fuels were believed to hold great promise for lowering fuel production costs without reducing fuel performance. As the nuclear industry expanded, however, and pellet fuels began to dominate, research into these alternative processes dwindled and finally ceased in the United States.

In the Saxton program (discussed in the following section), approximately 23% of the fuels tested were VIPAC, with the remainder being CPS fuels. This seems to reflect the general shift in MOX fuel manufacturing philosophy towards the CPS fuels. The fuel types are too different to extrapolate performance data from one to another. In fact, extrapolation of even the CPS data to modern fuel is questionable due to the great advances in MOX fuel production that have been made in the interim.

The irradiation experience resulting from the PRTR MOX fuel tests are summarized in Table 2 (Ref. 20, p. 127). One of the CPS rods did develop a defect during irradiation due to internal hydriding, which was a common cause of failure in the early MOX development programs. Additional tests on metallic plutonium fuels and on UO_2 fuels were also performed in the PRTR.

Table 2. Summary of the U.S. PUP early test reactor experience

Reactor	Fuel Type	Number of Elements	Pu Concentration (wt. %)	Peak Linear Heat Rating (W/cm)	Peak Burnup (GWd/MTM) [1E20 fissions/cm³]
PRTR	VIPAC	20	0.5	525	18.5
PRTR	Swaged	61	0.5	425	12.5
PRTR	VIPAC	16	1.0	445	11.5
PRTR	Swaged	33	1.0	510	13.5
PRTR	?	1	1.5	145	13.0
PRTR	VIPAC	79	2.0	655	7.8
PRTR	Swaged	2	2.0	655	8.1
PRTR	Hot and Cold Press Pellet	2	2.0	705	1.8
PRTR	VIPAC	1	2.0	560	1.2
PRTR	?	1	4.0	885	3.5
MTR	MCO*	2	0.03	496 & 560	[0.33 and 0.93]
MTR	MM**	10	0.03 to 5.7	220 to 1250	[0.3 to 2.6]
MTR	MCO	2	0.03	624 and 786	[0.47 and 1.0]
MTR	MM	10	0.2 to 7.5	108 to 885	[0.1 to 4.2]
MTR	High-energy-impacted fuel	2	2.5	1290	[0.3]
ETR	MM and high-energy-impacted fuel	4	2.5	460	[0.37]
MTR	High-energy-impacted fuel	32		1035	[6.8]
MTR	MCO	1	1.0	570	[0.2]
ETR	MM	1	0.6	460	[0.8]
ETR	MM	1	0.5	404	[0.2]
ETR	MM	1	0.8 to 3.0	440	[0.3]
ETR	?	1	?	550	[0.2]

* Mixture of UO_2 and $(U,Pu)O_2$ coprecipitate
** Mechanically mixed UO_2 and PuO_2

As shown in Table 2, most of the pellet irradiation experience from the early test reactors involved very low burnup as compared to the high burnup obtained with more recent fuels. In addition, the majority of the early experience is with alternative fabrication techniques that are not relevant to the FMDP. PIEs were performed on several irradiated rods in the PRTR.

It is concluded that the experience from the very early test reactors, while interesting in the historical context of MOX fuel development, is for the most part irrelevant to the domestic plutonium disposition program.

3.1.2   Saxton Program

Based on the early success of MOX irradiation experiments described in the previous section, the Saxton Program was initiated. The Saxton program was an AEC program that was subcontracted to Westinghouse. Westinghouse provided nine LTAs for irradiation in the second core of the Saxton pressurized water reactor (PWR). The Saxton reactor was rated at 23.5 MWth with an active fuel length of approximately one meter. The MOX assemblies, first loaded in 1965, contained 638 MOX rods with a plutonium concentration of 6.6%, such that a total of 23 kg of plutonium was loaded. Most of the rods were clad with Zr-4, but 30 rods were clad with 304 SS (stainless steel). The majority of rods contained CPS pellets, but a sizable fraction were VIPAC (~23%). One additional distinction about the Saxton fuel is that the plutonium contained in both the pellet and VIPAC fuels was converted from metal reduction buttons to oxide through DMO.

These rods were irradiated for nearly three years (known as Saxton Core II) at peak linear heat rates of 18.7 kW/ft to a peak pellet burnup of 28 GWd/MT. Of the 638 rods initially inserted into Saxton, 250 were reconstituted into a looser lattice and reintroduced for additional irradiation. These 250 rods (known as Saxton Core III) remained in-core for an additional 2.5 years, and Saxton ceased operation on May 1, 1972. The peak linear heat rate achieved during the second irradiation was 21.1 kW/ft. The peak pellet burnup achieved was 51 GWd/MT.

No fuel failures occurred during the initial irradiation during Core II. PIE of several of the rods indicated satisfactory performance. However, during the second irradiation, there were 37 identified failures out of the 250 MOX rods initially placed in Core III. All the failures occurred in high-power rods between the middle and end of Core III. Accelerated oxidation was seen on the defected rods, and hydriding was noted in the metallography of several rods. In any event, none of the 37 rod failures were attributed to any generic fuel problem intrinsic to the use of MOX. Westinghouse obtained a great deal of MOX experience from the Saxton irradiations (Ref. 22 and Ref. 23). More information on the extensive PIE program at Saxton is presented in the tables contained in Appendix A.

In addition to the large amount of fuel performance information generated in the Saxton program, a significant amount of overall operating experience was obtained with a partial MOX core (9 out of 21 assemblies were MOX). In addition, significant core physics measurements such as control rod worth and temperature coefficients were measured and compared to calculations. The plutonium used in the Saxton program was 91.4% fissile.

With respect to the FMDP, specifically PWR irradiations, the Saxton experience is relevant. The fuel performance information gathered on pellet fuels was rather extensive. This experience was the springboard into the manufacture of LTA's for insertion in commercial reactors. It is true, however, that the techniques used to make the fuel are somewhat outdated as compared to modern fabrication techniques, expertise, and fabrication equipment. However, the fact that 91.4% fissile plutonium was used may help provide answers to questions concerning the differences in physics models for RG plutonium versus WG plutonium.

3.2 WESTINGHOUSE MOX DEVELOPMENT

At the conclusion of the Saxton Program and the PUP, the AEC decided that additional government support of plutonium recycle was not justified because the utilities, reactor vendors, and fuel manufacturers were fully capable of commercializing the technology. Westinghouse, having been the AEC's subcontractor for the Saxton Program, was in a position to take the lead in these commercialization efforts. Westinghouse had also been involved in a parallel development program, a joint program between the Edison Electric Institute (EEI), a consortium of utilities, and Westinghouse, known as the EEI/Westinghouse Plutonium Recycle Demonstration Program (PRDP).

3.2.1 EEI/Westinghouse Plutonium Recycle Demonstration Program

The PRDP was conceived to study and demonstrate the economical use of MOX in PWRs. It consisted of two phases—an initial analytical phase and a follow-on MOX irradiation demonstration. The initial phase, which began in 1967, studied the technical issues and economics of MOX use in PWRs. Many of the facets of MOX use were explored using analytical techniques and critical experiments. Among other issues, plutonium's influence on control rod and soluble-boron worth were studied. Computer codes were modified to accommodate the characteristics of MOX fuel, bringing the modeling capabilities for MOX cores to the same level as those for UO_2 cores.

During the second phase of the PRDP, a reference MOX core design was developed. The core, based on the concept of self-generated recycle, was fueled with 1/3 MOX assemblies. (Self-generated recycle refers to the situation in which plutonium recovered from previous core discharges is reinserted as MOX fuel. No additional plutonium is placed in the core.) With all the MOX located in discrete assemblies, the 1/3-core fraction could be accommodated within the existing design limits without changes to the control system because none of the MOX bundles were located in control positions.

Another portion of Phase II of the PRDP consisted of manufacture and irradiation of MOX LTAs in the San Onofre Unit-1 reactor. San Onofre 1 is an early three-loop Westinghouse PWR with an electric capacity of 456 MW. A total of 720 MOX rods were placed in four LTAs (the assemblies consisted of all MOX rods). These four LTA's were the first PWR MOX assemblies irradiated in a U.S. commercial reactor. The plutonium (approximately 86% fissile) was supplied by the AEC at a reduced cost, as partial sponsorship of the study. Figure 5 shows the assembly design.
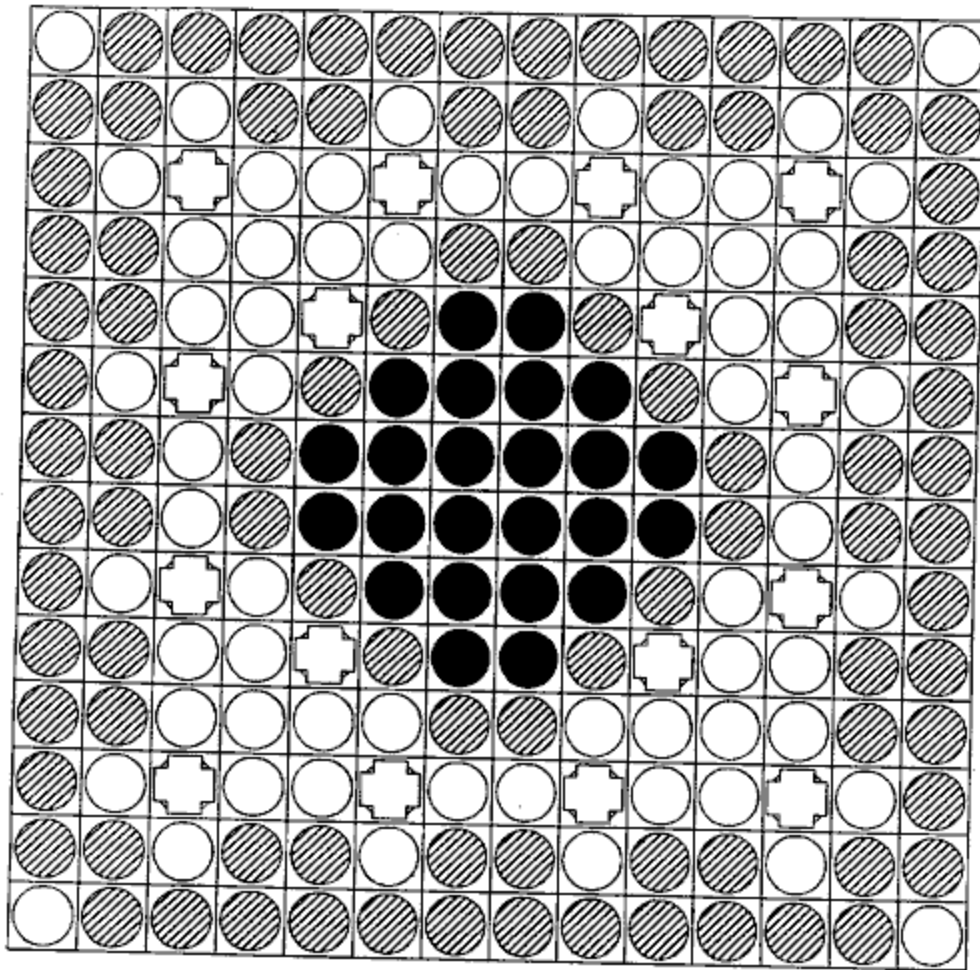
Center Rods: 3.85 wt% total plutonium

Edge Rods: 3.65 wt% total plutonium

Corner Rods: 3.30 wt% total plutonium

Figure 5. MOX assembly design for San Onofre 1.

At the conclusion of San Onofre's first cycle, fifty-two of the 157 original UO_2 assemblies were off-loaded and replaced with fresh fuel. The four MOX LTAs were included in this new fuel and were placed on the core periphery in nonpeaking positions such that core symmetry was maintained. At the end of Cycle 2, the LTAs had reached an average assembly burnup of 6,450 MWd/MT, and a peak linear heat rate of 6.8 kW/ft. The LTAs were visually inspected. Two of the four LTAs contained 52 removable fuel rods each. At the end of Cycle 2, four rods were removed and subjected to nondestructive examinations. Two of these rods were subjected to destructive examinations.

The assembly was reconstituted using UO_2 rods and then reloaded in the core for an additional cycle. At the conclusion of Cycle 3 (after two irradiation cycles), the four LTAs were again removed. It was originally intended that they be irradiated for three cycles, but contemporaneous failures in unpressurized UO_2 rods led to the conservative decision to remove them. Fuel densification in UO_2 fuels had, in some cases, led to cladding collapse onto the fuel. Rod pressurization eventually solved this problem. While the MOX rods were not pressurized, they showed no indications of cladding collapse at the end of Cycle 3.

At the end of the third cycle, six rods were nondestructively examined, and two rods were destructively examined. These rods were from the same assembly used in Cycle 2, but ended the third cycle with an average assembly burnup of 19,000 MWd/MT and a peak MOX pellet burnup of 23,500 MWd/MT. The results of these examinations are especially useful because of the comparison they provide between two competing fuel manufacturing processes. Of the two rods destructively examined at the end of each cycle, one contained pellets produced through comilling, and the other contained pellets produced via a coprecipitation process. The PIE results identified some advantages of coprecipitated fuel. Appendix A provides further details of this irradiation.

The results of the San Onofre irradiations provide a comparison between the performance of comilled MOX and coprecipitated MOX. Although neither of these fuels is typical of the high quality, high density fuels produced today, the relative performance is important nonetheless. One of the differences in the fuel behavior was densification, which is of much less interest today as a result of improved fuels. Another difference, which is closely related to the densification behavior, is fission gas release.

In MOX fuel, the majority of the power production takes place in the plutonium. In comilled fuels, the plutonium exists as discrete particles surrounded by a UO_2 matrix. Power production is highly localized in the plutonium particles. In coprecipitated fuel, the plutonium is distributed into plutonium-enriched regions surrounded by a UO_2 matrix. The power production is, therefore, more diffuse. The more homogeneous coprecipitated fuel more closely resembles the plutonium distribution seen in irradiated UO_2 fuel, and its behavior is (not surprisingly) more similar to that of UO_2 fuel.

In comilled MOX pellets, the fission products tend to coalesce around the plutonium particles in which most fissions occur. In the tested coprecipitated fuel, the fission products are distributed through a much larger fraction of the surrounding UO_2 matrix due to the uniformity of the coprecipitated master mix. Therefore, fission gas bubbles form more readily in the comilled

fuel. It is the formation of bubbles that eventually leads to fission gas release from the fuel matrix. Fission gas release from the comilled San Onofre MOX fuel was nearly twice as high as that from the coprecipitated fuel. The San Onofre data provided early evidence of the desirability of a high degree of homogeneity in MOX fuel. Reference 24 provides the original documentation for the San Onofre irradiation.

3.2.2 Overseas Westinghouse LTA Programs

Following the successful irradiations in Saxton and San Onofre, Westinghouse participated in a number of collaborative research efforts with foreign participants. Westinghouse produced a total of sixteen LTAs for irradiation in the Italian Trino reactor, the Swiss Beznau-1 reactor, and the Japanese Mihama-1 reactor. Each of these reactors had a Westinghouse-supplied nuclear steam supply system (NSSS). The only additional domestic MOX programs in which Westinghouse participated are the Electric Power Research Institute (EPRI) Plutonia Fuel Study (Ref. 25) and the Ginna MOX irradiations.

3.2.3 Ginna LTAs

The Ginna irradiations are believed to be a continuation of the EEI/W - EPRI program described in Section 3.2.1. MOX rods sufficient to fill four assemblies were manufactured by Westinghouse in 1974 for irradiation in Rochester Gas and Electric (RG&E) Company's Ginna nuclear power station. Ginna is an early-design Westinghouse PWR with an electric output of 498 MW. The assemblies were originally to be loaded in 1975, but other issues facing RG&E forced a delay in the MOX licensing activities. These rods were manufactured when utilities viewed MOX use as an eventual certainty. However, by the time the rods were ready for insertion, domestic MOX use had essentially been abandoned as a result of the Executive Order on nonproliferation issued by President Carter. The licensing revision to permit insertion of the LTAs, in fact, faced the issue of whether or not their irradiation was permitted under the associated NRC policies. The unassembled rods were stored in Westinghouse's Cheswick facility until 1979, when they were transferred to Exxon Nuclear Company, Inc. (ENC) for assembly. It was determined by RG&E management that irradiation of the MOX rods was the best method of dispositioning the rods, and this argument was made to the NRC. They were eventually loaded into the reactor for Cycle 10 in 1980.

The four 14 × 14 LTAs contained a total of 736 MOX rods utilizing approximately 83% fissile plutonium. Figure 6 shows the assembly design for these LTAs. The LTAs were irradiated until 1985 to an average assembly burnup of approximately 40,000 MWd/MT. No operational problems were reported for the LTAs. In fact, these LTAs operated successfully through Ginna's steam generator tube rupture in 1982. No PIE work was done on the assemblies, and the assemblies are currently stored intact in the Ginna fuel pool. These assemblies were the last commercial MOX assemblies irradiated in the United States. Information on the Ginna irradiation is taken from Refs. 26 and 27.
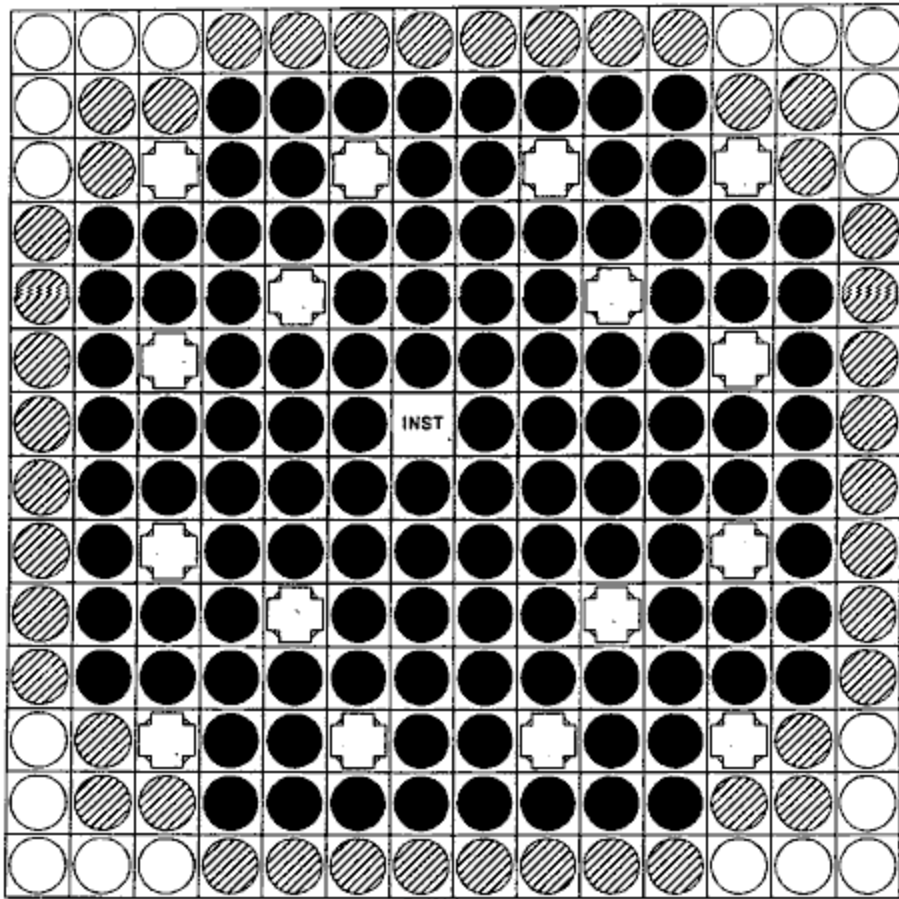
○ Center Rods: 3.20 wt% total plutonium

○ Edge Rods: 3.00 wt% total plutonium

○ Corner Rods: 2.60 wt% total plutonium

INST: Instrumentation tube

Crosses: represent rod cluster control guide tubes

Figure 6. The Ginna LTA MOX assembly.

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3.3 GENERAL ELECTRIC (GE) MOX EXPERIENCE

The other domestic reactor and fuel vendors also had active MOX research programs during the 1960s and 1970s. In particular, GE joined the EEI in a joint program to pursue MOX use in BWRs. This program was parallel to the EEI/W PWR program described in Section 3.2.1. GE also has some MOX experience from capsule tests. Eventually, GE assisted in a number of international MOX studies and in EPRI-MOX work. All these activities are described in the following sections. Open literature documentation is available for the EEI/GE Big Rock Point (BRP) tests and the EPRI Quad Cities 1 irradiations.

3.3.1 GE's Early MOX Testing

General Electric, the U.S. AEC, and Euratom jointly conducted a BWR MOX program in the early 1960s. The program consisted of irradiation of a number of MOX rods in the Vallecitos BWR. Vallecitos was an early BWR prototype reactor, which had a thermal output of 30 MW. The reactor was fueled with plate-type fuel, but was capable of operating with rod bundles for testing (Ref. 28). The AEC-GE-Euratom program focused upon obtaining basic operating data for MOX fuel in thermal reactor environments. The data were used to develop a model for predicting the isotopic and reactivity behaviors of MOX fuel.

Sixteen MOX test rods were inserted in the Vallecitos reactor. The rods were clad with Zircaloy and measured 1.07 cm OD (outside diameter). The MOX, which contained 1.5% plutonium of unreported isotopic composition, was irradiated to a maximum burnup of 1.4E20 fissions/cm^3 (Ref. 20, p. 669). Some PIE, including measurement of the plutonium distribution in the irradiated fuel, was performed. Detailed results have not been located in the open literature, however.

GE also participated in irradiations of LMR fuel (20% plutonium) in a number of reactors, including their General Electric Test Reactor (GETR). The extent of GE participation in the PRTR tests is not clear. However, according to Ref. 29, the 80 rods containing hot- and cold-pressed pellets that were irradiated to ~9 GWd/MT, at up to 21.5 kW/ft, were manufactured by GE. This indicates that the PRTR pellet irradiations are more applicable to BWRs than to PWRs.

Ref. 29 and Ref. 30 list GE as the manufacturer of four rods that were tested in the Dresden-1 nuclear power station. Four fuel bundles, with a single MOX rod per bundle, were inserted in 1967. The rods contained hot-pressed pellet fuel and utilized Dresden self-generated plutonium that was ~80% fissile. GE is also known to have been a participant in EPRI's Plutonia Fuel Study (Ref. 25) described in Section 3.8.

3.3.2 EEI/GE Plutonium Utilization in BWRs Program

As with the parallel EEI/W PWR program, the initial stage of the EEI/GE program was analytical in nature. The promising results of the analytical work led rapidly to plans for irradiation testing in the Consumers Power Company BRP BWR. Four types of fuel consisting of annular, solid, and dished pellets with different plutonium loadings were tested. The tests

focused on the behavior of annular fuel, with several solid and solid-dished pellet rods included for comparison. All of the test rods contained cold-pressed and sintered MOX pellets produced from mechanically blended powder in a process similar to that described in Section 2.2.1. The sixteen bundles were loaded for Cycle 7 in May 1969.
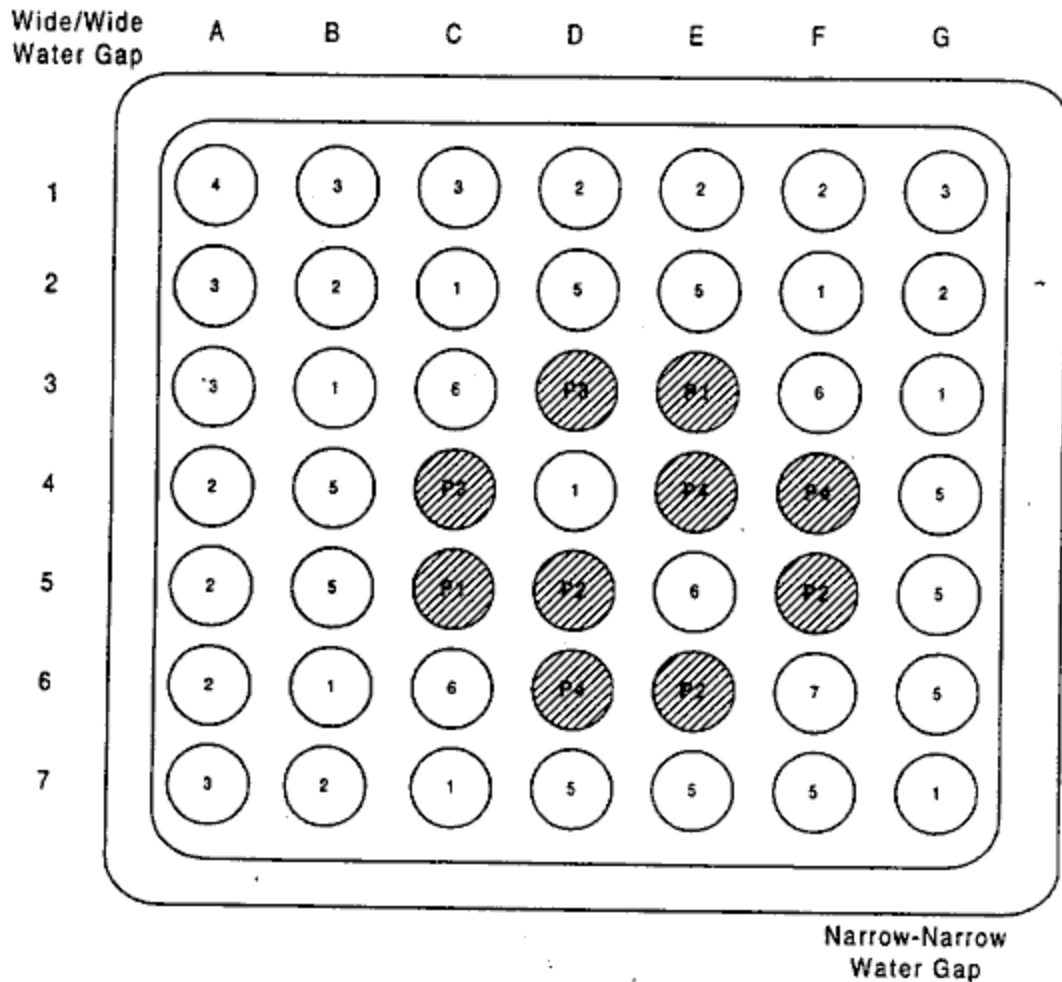
At the conclusion of BRP Cycle 7, the MOX bundles were removed from the core and sipped. One of bundles was suspected of containing a leaking rod based on the results of the sipping tests. Four rods were permanently removed and sent to the Vallecitos Nuclear Center for PIE (Ref. 31). The remaining 28 rods (including six of the ones on which profilometry was performed) were returned to the reactor for additional irradiation and eventually removed in the mid-1970s. Appendix A shows some information regarding the 32-rod irradiation. However, as shown in this appendix, detailed rod-by-rod information on many rods could not be located.

Three reload MOX bundles were introduced into BRP for Cycle 8. Each of the 9×9 bundles contained 68 MOX rods with a maximum plutonium loading of 9.1 wt%. Rod descriptions for the three reload bundles are given in Ref. 32, p. 10. All of the fuel was manufactured via cold-pressing and sintering of comilled powder. Natural uranium was used as the MOX diluent, and both 80% and 90% fissile plutonium was utilized. The 90% fissile plutonium used in 180 of the 204 rods was obtained from the AEC. The 80% fissile plutonium was obtained from Dresden-1 recycled fuel.

The MOX reload bundles operated during Cycles 8, 9, and 10. At the conclusion of Cycle 10, sipping tests indicated leaking rods in two of the three bundles. Due to a nonnuclear primary system problem, high rates of crud formation were experienced on all fuel in the reactor during this period. Two rods in the third MOX bundle failed during operations to remove the adherent crud during the shutdown between Cycles 10 and 11. The two failed rods were replaced, and this single MOX bundle was reinstalled in the reactor for Cycle 11.

All of the GE MOX rods were removed from BRP at the end of Cycle 11. As with the two-rod-per-bundle tests, only limited data is available for the PIE that is known to have occurred on the BRP MOX fuel. Some of the data is believed to be documented in EPRI reports, with the remainder in proprietary GE documents. EPRI took over sponsorship of the EEI/GE MOX program in 1974, and it is likely that all the EPRI reports from this program have not been located. It is important to note that most of the BRP data are for annular fuel.

Following the initial success of the BRP reload bundles, GE proceeded to prepare MOX rods for the initial core of the Vermont Yankee reactor. Due to some licensing difficulties, these rods were eventually constituted into five bundles (four center and one periphery) that were loaded into Quad Cities 1 with the first core reload in 1974. Each of the four central bundles contained ten MOX rods in an island arrangement. The MOX island was surrounded by UO$_2$ rods and was slightly offset towards the water gap corner to isolate the MOX from the control blade. The peripheral bundle contained eight MOX rods scattered near the reflector corner of the bundle. As with the BRP MOX, two plutonium assays were used: Dresden recycle (~ 80% fissile) and AEC plutonium (~ 90% fissile). The isotopic makeup of the plutonium was similar to, but not identical to, that used in the BRP LTAs. All of the pellets were cold-pressed and sintered from comilled powder, with natural uranium used as the diluent. Figure 7 below shows the assembly design for a central MOX bundle at Quad Cities.

Wide/Wide Water Gap

	A	B	C	D	E	F	G
1	4	3	3	2	2	2	3
2	3	2	1	5	5	1	2
3	3	1	6	P3	P1	6	1
4	2	5	P3	1	P4	P4	5
5	2	5	P1	P2	6	P2	5
6	2	1	6	P4	P2	7	5
7	3	2	1	5	5	5	1

Narrow-Narrow Water Gap

Numbers: UO$_2$ or UO$_2$-Gd pins of various enrichments

P1: solid 80% fissile MOX pin, 2.14 wt% fissile plutonium

P2: solid 90% fissile MOX pin, 3.52 wt% fissile plutonium

P3: annular 80% fissile MOX pin, 2.34 wt% fissile plutonium

P4: annular 90% fissile MOX pin, 3.62 wt% fissile plutonium

Figure 7. Quad Cities central MOX bundle.

The MOX bundles were irradiated for Cycles 2, 3, 4, and 5 with various reconstitutions taking place. EPRI sponsorship of the program ended at the conclusion of Cycle 5. Nevertheless, GE and Commonwealth Edison continued the program, irradiating a reconstituted central assembly and the peripheral assembly for an additional cycle. The extent of GE proprietary information on the fuel examinations is not known but is likely to be substantial.

A wealth of information was obtained as a result of the Quad Cities' irradiations and the PIEs. The Quad Cities' program demonstrated that extended burnup of BWR MOX fuel was feasible. The bundles were irradiated for up to eight years, to maximum burnups of 39.9 GWd/MT bundle average and 57 GWd/MT peak pellet. The MOX fuel operated without failure at peak linear heat rates up to 15.5 kW/ft. Appendix A shows the examinations that were conducted and the associated EPRI reference documents. Ref. 33 provides a good overall summary of the program and the fuel performance results.

3.3.3 Other GE MOX Testing

GE has been involved in a number of additional MOX programs. In their recent study on plutonium disposition (Ref. 34), GE lists two foreign MOX programs: the Halden Plutonium Program and the Kritz Program. Only brief descriptions are given. Furthermore, GE performed a number of transient tests on MOX fuel in the SPERT [transient irradiation test reactor at Idaho National Engineering and Environmental Laboratory (INEEL)] facility. They also designed some of the MOX fuel (manufactured by BN) that was irradiated in the Italian Garigliano BWR. Finally, GE contributed to EPRI's MOX fuel densification study (Ref. 25).

GE's involvement in the Halden Plutonium Program, as described in Ref. 34 consisted of ramp testing of MOX fuel rods containing annular pellet fuel. The tests were performed at linear heat rates of 17 to 22 kW/ft. The results are described as "the effects of pellet-clad interaction showed acceptable behavior of the MO_2 rods." According to Ref. 29, two rods were tested in Halden. Both solid and annular pellets were tested to peak pellet burnup of 16 GWd/MT.

The Kritz Program is described in the vendor report (Ref. 34) as the preirradiation testing of the Garigliano MOX reload bundles, which showed that the existing analytical models were adequate for prediction of MOX neutronic performance. The available references suggest that these bundles were manufactured by BN to GE's specifications.

Additional testing occurred in the GETR, which was a test reactor that was used for fuel and materials development (Ref. 35). Two rods containing annular MOX fuel were irradiated to ~22 GWd/MT at up to 18 kW/ft (Ref. 29). It is probable that a number of additional tests were performed by GE in support of MOX development. Irradiation of MOX fuel samples was performed in the GETR as part of the EPRI Plutonia Fuel Study as described in Section 3.8.

The available information on the Garigliano irradiations is incomplete. The available open literature information indicates that GE fabricated 96 MOX fuel rods contained in four demonstration island-type assemblies. These assemblies were introduced into the reactor in 1968. This fuel included 12 VIPAC rods, 24 hot-pressed pellet rods, and 60 CPS rods. An assembly average burnup of ~25 GWd/MT at a design peak linear heat rate of 15 kW/ft (Ref. 29,

p. XV-2) was achieved. Two plutonium concentrations were used in the fuel: 2.0% and 3.2%. Selected rods were removed after one, two, and three irradiation cycles corresponding to 15, 21, and 25 GWd/MT. PIE was performed under the auspices of a CNEN/ENEL (Comitato Nazionale per l'Energia Nucleare/Ente Nazionale per l'Energia Elettrica) program (described further in Section 4.2 below) at the Risø Laboratory in Denmark (Ref. 36). It is not known whether GE was granted access to any or all of the PIE results, but some of the preliminary results have been reported openly. BN produced 204 MOX rods, contained in four assemblies designed by GE, that were loaded at the same time (Ref. 37).

In 1975, a full reload of MOX island assemblies was introduced into the Garigliano core. This reload consisted of 46 assemblies, each of which contained 32 MOX rods. The MOX rods were manufactured by BN and designed by GE. The bundles remained in the core through 1981, to a peak burnup greater than 25 GWd/MT (Ref. 34). Details about the irradiation performance and/or PIE of this fuel have not been located; however, it is likely that GE has access to at least a portion of this information.

GE is currently involved with the Japanese in a program to load full-MOX cores into advanced BWRs. No information on this work is available. However, it is thought that this work would have direct applicability to the FMDP.

3.4   ABB COMBUSTION ENGINEERING (CE) MOX EXPERIENCE

In their Plutonium Disposition Study report (Ref. 38), CE does not reference any original MOX experience. They provide a brief summary of the domestic and foreign MOX programs, but none are CE programs. Based on this information and on the lack of MOX fabrication facilities at the height of the domestic MOX development (Ref. 22, p. 5–2), it seems that CE has no MOX experience of its own. However, CE reportedly participated in joint experiments and testing with ALKEM and KWU (Kraftwerk Union AG) during the 1970s (Ref. 39).

3.5   EXXON NUCLEAR MOX EXPERIENCE

Exxon Nuclear Company, Inc. (ENC) had an extensive MOX development program during the 1970s. Available reports of Exxon's activity are specific to BWR fuel. No reports of irradiation of ENC PWR MOX have been found. Two 9 x 9 MOX bundles were introduced into the BRP core in 1972. These bundles were tested at the same time as the GE BRP bundles. BRP's owner, Consumers Power Co., considered the plant a fuel test bed during this period. Two additional bundles were introduced at the next refueling in 1973. Based on the early success of these test bundles, partial-core MOX reloads were initiated.

Eighteen 11 x 11 bundles, each containing 24 MOX rods, were introduced in 1974. Eight additional bundles of the same design were introduced in 1976. This experience is the closest to commercial recycling of MOX that has occurred domestically. The license for MOX use in BRP limited the plutonium content of the MOX to 50 kg total. It was expected at the time that with the successful completion of the GESMO proceedings, this limit would be raised (Ref. 19, p. II-32).

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In addition to the BRP experience, ENC manufactured eight MOX LTAs for the German Kahl BWR. No detailed descriptions of the LTAs, their irradiation performance, or PIE have been found for the ENC MOX fuel. It is likely that this information remains proprietary. ENC is not currently involved in the FMDP. However, Siemens Power Corp., a wholly owned subsidiary of Siemens AG, now owns what was ENC. Unfortunately, the purchase of ENC specifically excluded the mixed-oxide fuel data.

ENC participated in EPRI's Plutonia Fuel Study (Ref. 25). The extent of their participation is not clear, however, as described in Section 3.8 below. Further information can be found in Appendix A.

3.6   GULF UNITED NUCLEAR CORPORATION (GUNC) MOX EXPERIENCE

From the limited information available, it appears that GUNC's MOX experience is limited to that obtained from the joint GUNC/Commonwealth Edison program in Dresden 1. Self-generated plutonium was reintroduced into Dresden 1 in 1971. Starting with Cycle 7, eleven MOX bundles (each containing ~77% fissile plutonium in nine MOX rods) were introduced into the core. All the MOX rods contained 2.3% plutonium in natural uranium (Ref. 40). Reports of the remainder of the irradiation (Cycle 10) and the expected PIE on the fuel have not been found. Commonwealth Edison may have access to some of this information as a party to the original research. Further information on this irradiation can be found in Appendix A.

3.7   BABCOCK AND WILCOX (B&W) MOX EXPERIENCE

B&W manufactured fuel for the Fast Flux Test Facility (FFTF). They also provided fuel pellets for EPRI's densification and homogenization experiments as part of the Plutonia Fuel Study (Ref. 25). The MOX pellets were manufactured using a process similar to that used for the FFTF fuel. Ross and Benson (Ref. 41) summarize the process:

> "Pellets for the EPRI program were fabricated using the same process and equipment used to fabricate the FFTF fuel except that the lot size was 5 kg (instead of 50 kg). For the EPRI program only centerless ground fuel pellets were to be supplied by NMD. Encapsulation for irradiation testing and other physical and chemical tests are to be performed by BNWL.

> "The PuO_2 as received from ARHCO was used without any calcining treatment. The UO_2 used to manufacture the EPRI pellets was depleted UO_2 as prepared by the ADU process at NMD's Uranium Fuel Fabrication Plant.

> "After V-blending the PuO_2 and UO_2 powders in a P-K blender using an intensifier bar, the material was jetmilled using a Troust Fluid Energy Mill and then screened through a 60 mesh screen. No recycle material was added to this powder as would be done during commercial operation.

> "As a result of the jet-milling process, the mixed oxide becomes very active [meaning it sinters to a very high density], especially when there is no

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recycle present, and it is necessary to add a burnable organic pore former such as carbowax to the powder to lower the density. The sintered density of jet-milled material, with no organic added to the powder, was 97–97.5% T.D.

"Carbowax pore former was added as -120/+200 mesh granules to the powder and the powder is slugged to a nominal density of 46% T.D., granulated and screened to -20 mesh granules. A die lubricant of -200 mesh sterotex (0.3%) is added prior to pelletizing. The pellets were pressed using a Hydramet hydraulic press with a single cavity punch and die set. The green pellets were 0.440" diameter × 0.480" long and were compacted at 5.7 tons to 55% T.D.

"The pellets were...sintered...and dry centerless ground using a Royal Master grinder to 0.3640 ± 0.0005" diameter.... The average density of the pellets was 94.315% T.D. and the one-sigma standard deviation is 0.188% T.D.... The sintered pellets were evaluated for plutonium homogeneity using standard alpha-radiographic techniques. The pellets do meet the B&W specification for MOX zones, which for 25% PuO_2-UO_2 particles, can be as large as 849 microns."

Reported irradiation experience with this B&W MOX fuel is limited to that available in the Plutonia Fuel Study reports. However, the fuels are intentionally referred to as fuel Type 11, 12, etc., such that one cannot readily extract the behavior of a single manufacturer's fuel. No additional reports of production or irradiation of B&W MOX have been found.

3.8 OTHER DOMESTIC MOX EXPERIENCE

In addition to the domestic programs described in Sections 3.1–3.7, Nuclear Fuel Services manufactured four MOX demonstration assemblies that were irradiated in the BRP BWR in the early 1970s (Ref. 19, p. II-32). Detailed descriptions of the assemblies, irradiation experience, and PIE have not been located. No additional references to Nuclear Fuel Services MOX research have been found.

EPRI instituted the Plutonia Fuel Study in 1975 under sponsorship by B&W, BNFL, Central Research Institute of Electric Power Industry (Japan), CE, ENC, GE, and Westinghouse. The program was meant to reproduce the UO_2 densification study (Ref. 42). Fuel was manufactured by a number of the participants, and possibly by all. The fuels are not labeled as to their manufacturer in the program write-ups. It may be possible, however, to reconstruct this information from other sources. The fuels underwent extensive preirradiation examination and then were irradiated in the GETR. PIEs were performed. The purpose of these studies was to examine the thermal- and irradiation-induced densification of MOX fuels for comparison with the behavior of UO_2 fuels. If a correlation between the fuels and their manufacturer/method can be deduced from other data or obtained from project reports, the data may provide a useful comparison of the behavior of the various fuels.

4. FOREIGN MOX IRRADIATION EXPERIENCE

MOX research in Europe, as with nuclear power research in general, followed developments in the United States closely during the 1960s and 1970s. For many European countries, MOX use promised energy independence—a long-sought goal. The Belgians irradiated their first MOX assembly in 1963, and they were soon followed by a number of other countries.

In December 1974, the Council of Ministers of the European Communities initiated the Research and Development Pluriannual Programme on Plutonium Recycling in Light Water Reactors. Ten of the forty-eight contracts issued to carry out the program investigated MOX fuel performance through irradiation and PIE (Ref. 43). This program brought together the numerous domestic programs throughout Europe and led to additional MOX irradiations and reloads. The ten programs are summarized in Table 3, taken from Ref. 43.

Table 3. CEC plutonium recycling program PIEs

Contract Number	Contractor	Reactor	Purpose	Assembly Burnup (GWd/MT)	Laboratory
1	BN - SCK-CEN	BR3/PWR	MOX rods in standard assembly (Z0–100)	32	SCK-CEN
2	INTERFUEL/ECN	HFR/PWR	Three MOX rod bundles in rig (vibrasol)	4–9	ECN/Petten
3	BN/GKN	Dodewaard/BWR	MOX rods in standard assembly (B201)	20	SCK-CEN
4	ENEA/ENEL	Garigliano/BWR	MOX rods in standard assembly	14, 21	RISØ
5	KWU	Lingen/BWR	Thorium-plutonium rods in standard assembly	4–20	TUI
6	ENEA/ENEL	Garigliano/BWR	MOX rods in standard assembly	7, 25	RISØ
7	BN - SCK-CEN	BR3/PWR	MOX rods in standard assembly	20–43	SCK-CEN
8	BN/GKN	Dodewaard/BWR	MOX rods in standard assembly	28	ECN/Petten
9	BCR	CNA/PWR	MOX rods in standard assembly	27	SCK-CEN and CEA/Saclay
10	FRAGEMA	CNA/PWR	MOX rods in standard assembly	27	CEA/Saclay

The Commission of the European Communities (CEC) program continued throughout the 1970s. However, when the program came up for renewal in 1980, both France and the United Kingdom believed that rapid deployment of LMRs would, in the very near future, eliminate all plutonium surpluses. The inaction of these two key countries kept the program from being continued, and shortly thereafter many of the national programs ended. Only two of the countries involved in the CEC program, Belgium and the Federal Republic of Germany, continued research into LWR MOX use.

As the 1980s progressed, the LMR revolution did not materialize as expected. In addition, success with reprocessing plants led to large and growing plutonium surpluses. France decided to pursue plutonium recycle in their existing LWRs. In the United Kingdom, BNFL reinstituted MOX research and planned to enter the commercial MOX fabrication arena. By the end of the 1980s, MOX use was expanding rapidly in Europe. Large MOX fabrication facilities were being planned and constructed. At the present time, the MOX industry in Europe has become a fully developed, commercialized enterprise.

The Europeans continued MOX development efforts after the 1977 Executive Order postponing reprocessing indefinitely in the United States. This Order, which effectively ended U.S. MOX research, was meant to influence the other nuclear powers to halt their plutonium recycle programs. However, despite the Executive Order and U.S. policy, other countries proceeded with the technical development and commercial use of RG MOX fuel. This continued development has led to advancements that partially eclipse the earlier domestic experience. Not only does the foreign experience base comprise more irradiated rods, but also most of the foreign experience is for modern high-density, homogeneous fuel.

Due to the widespread use of commercial MOX overseas, the foreign MOX irradiation experience with RG material is extensive. However, much of the data is proprietary, and a central table summarizing the examinations conducted on MOX fuel in foreign countries was not feasible to produce. Chapter 4 does, however, contain a general discussion of the foreign MOX experience.

Not surprisingly, extensive utilization overseas has resulted in improvements in MOX fuel performance. An example is fission gas release, which has been shown to be highly dependent on the fuel microstructure. Early fuels were rather inhomogeneous, containing relatively large plutonium particles and/or agglomerates. Also, the pellet density associated with earlier fuels was not nearly as high as that currently obtained. The net result was higher fission gas release from MOX than from UO_2. MOX produced with the current processes that result in more homogeneous fuel with smaller plutonium particles and higher density has shown much lower fission gas release rates, although these rates remain somewhat higher than those of UO_2 fuels.

4.1   BELGIAN MOX EXPERIENCE

The Belgians entered the commercial nuclear power arena early, as evidenced by the presence of a Belgian team at the startup of the Shippingport PWR (Ref. 44, p. 13). They became interested in LWR recycle of MOX shortly after their domestic nuclear research began. In a cooperative effort between the fuel vendor BN and the Belgian Nuclear Research Centre at Mol—Studiecentrum voor Kernenergie-Center d'étude de l'Énergie Nucléaire (SCK-CEN)—the Belgians introduced their first MOX assembly into the BR3 reactor in 1963. In the years since, they have become one of the major players in MOX fuel development and manufacture.

4.1.1   Early Experience in BR3 Reactor

BR3 was the first PWR built outside the United States. It was basically an early Westinghouse design with a thermal output of 41 MW. The MOX bundle introduced into BR3 in 1963 was the

world's first MOX assembly introduced into a commercial reactor (Ref. 45). The island-type assembly utilized 12 stainless steel-clad VIPAC rods. The MOX rods reached burnups of only 3 GWd/MT assembly average and 6 GWd/MT peak pellet (Ref. 37, p. 184).

In 1965, the BR3 reactor core was redesigned with a triangular lattice to accommodate additional experimental and instrumentation capabilities. Included in the updated BR3/Vulcain core was a single MOX bundle containing 18 pins filled with pellet fuel, and 19 pins filled with vibrocompacted MOX powder (Ref. 46). The assembly reached an average burnup of 25 GWd/MT. The pellet-filled rods reached a peak pellet burnup of 44 GWd/MT (Ref. 37). The vibrocompacted rods were placed in lower power positions and reached only 26 GWd/MT peak burnup.

Two additional MOX assemblies, each containing 18 MOX rods, were loaded into the BR3 core in 1969. Half of these rods contained VIPAC fuel, and the other half contained pellet fuel. In subsequent reloads, the MOX core fraction increased steadily, reaching almost 50% with the final reload, which was 70% MOX. The BR3 reactor was shut down in 1987 after 25 years of MOX research. Although it is probable that substantial PIE was performed on the BR3 fuel, only limited documentation of these efforts has been found. The BR3 MOX data provided BN a firm foundation from which to launch their commercial MOX venture.

4.1.2   Other Fuel Development Irradiations

In addition to the BR3 MOX fuel irradiations, BN either led or participated in a number of additional MOX fuel development irradiations that eventually led to commercial use of their MOX fuel. These programs inserted MOX fuel into the Garigliano and Dodewaard BWRs, the Chooz A PWR, and the NPD (nuclear power demonstration) heavy water reactor. The fuel development and demonstration program irradiations are summarized in Table 4 taken from Ref. 47.

The fuels manufactured by BN during this period, from the mid-1960s to the mid-1970s, were produced using one of several experimental processes. Two types of vibrocompacted fuel were tested: homogeneous and heterogeneous. In the homogeneous VIPAC fuel, particles of homogeneous $(U,Pu)O_2$ were loaded into the cladding. In the heterogeneous fuel, the two larger particle fractions were UO_2, and the finest particle fraction consisted of PuO_2. The heterogeneous VIPAC fuel offered the promise of minimal plutonium handling.

The early pellet MOX fuels were manufactured by one or more processes referenced in more recent literature as "previous" techniques. These may also be categorized as homogeneous and heterogeneous. The earliest technique, which is a variation of comilling, pressed a homogeneous blend of granulated $(U,Pu)O_2$. A later technique, which was used in the pilot facility operated from 1967 to 1975, blended PuO_2 into granulated UO_2. The later tests used fuel from the commercial fabrication plant, which used a process now referred to as the "Reference" process, from 1973 to 1984. The "Reference" process blended PuO_2 powder into free-flowing UO_2 powder, avoiding the granulation step used in earlier processes.

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Table 4. BN fuel irradiation

Reactor Type	Reactor	Years Operation	Assemblies	MOX Rods					Peak pellet burnup (GWd/MT)	Actual Programs
				BN* previous	BN* reference	BN MIMAS	CPCs	Others		
PWR	BR3	1963–87	129	512	1092	-	-	25	82	
		1984–87	24	-	224	178	./	10	42	PRIMO
	CAP	1985–87	2	-	48	-	40	-	24	
	CNA	1974–78	4	-	144	-	16	-	39	
	Beznau 1	1990-present	2	-	-	22 segments	-	2 segments	55	FIGARO/NOK PIE program
	BR2	1992–94	-	-	-	2	-	-	45–50	CALLISTO
BWR	Dodewaard	1971–87	7	84	-	-	-	-	47	
		1988–93	5	-	-	25 segments	-	15 segments	58	DOMO
	Garigliano	1970–84	51	204	1426	-	-	48	26	
	Oskarshamn 1	1974–79	3	-	51	-	-	-	19	
	Total		227	800	2985	225	56	100		

* See discussion in Section 4.1.2.

4.1.2.1 Garigliano Irradiations

Garigliano is an early-generation GE BWR fueled with standard 8×8 fuel bundles. BN produced 204 rods of MOX pellet fuel for the Garigliano core (Ref. 37). These rods, contained in four reload assemblies, were part of a larger group (total of 600 rods contained in 12 bundles) introduced into the core in 1968 (Ref. 37). The successful irradiation of these prototype bundles led eventually to commercial recycle of MOX in the Garigliano reactor in 1975.

4.1.2.2 Dodewaard Irradiations

A second demonstration of BN MOX fuel took place in the Dodewaard BWR. Like Garigliano, Dodewaard is an early GE BWR with a thermal output of 183 MW. A total of seven LTAs containing BN fuel were irradiated in Dodewaard from 1971 through 1987. The first two assemblies contained a total of 30 MOX rods; 27 rods containing pellet fuel and 3 rods containing VIPAC fuel. The VIPAC fuel was used in the three highest power rods in an effort to reduce the peak power in those rods through lower fuel density (Ref. 48). Both types contained 2.7% plutonium (2.5 wt % fissile) in natural uranium. These assemblies were irradiated at linear heat generation rates up to 440 W/cm (13.4 kW/ft) during Cycles 2–5 to assembly average burnups of 20 GWd/MT. PIE was performed on selected rods from these assemblies as part of the CEC plutonium investigations at SCK-CEN (Ref. 43).

Four additional MOX assemblies, containing a total of 56 MOX rods (again both VIPAC and pellet), were loaded in 1973. These assemblies, as with the first two, were of the island type. They contained 2.9% plutonium in natural uranium. After the first cycle of irradiation, sipping indicated a possible failure in one of the MOX assemblies. Two of the four were reloaded. One was irradiated three additional cycles, reaching an assembly average burnup of 28 GWd/MT. It subsequently underwent destructive examination in Petten. The other intact MOX bundle was irradiated four additional cycles to 33 GWd/MT assembly average burnup. After replacement of the leaking rod(s), the other two MOX assemblies were reintroduced to the core. They both operated for four additional cycles to assembly average burnups of 32 and 33 GWd/MT.

The seventh bundle, which contained 12 MOX rods (again VIPAC and pellet), was loaded at the beginning of Cycle 6 in 1974. It operated for a total of six cycles with two interruptions, with final discharge in 1987, reaching an assembly average burnup of 42 GWd/MT (Ref. 13).

4.1.2.3 Chooz A Irradiations

Chooz A is an early-design Westinghouse PWR located in France. It is often referred to in the relevant literature as the SENA (Société d'Énergie Nucléaire Franco-Belge des Ardennes) or CNA (Centrale Nucléaire des Ardennes) reactor. As part of a joint program between BN, the Commissariat à l'Énergie Atomique (CEA), and Reaktor Beteilingungsgesellschaft [RBG, now Reaktor-Brennelement Union GmbH (RBU)], four MOX demonstration assemblies were loaded in the reactor in 1974 (Ref. 49). The four island assemblies contained a total of 160 MOX rods containing 5% plutonium in natural uranium (Ref. 43). However, only 144 of these rods contained fuel produced by BN (Ref. 37). The remainder were produced by the CEA at Cadarache. The rods were all clad with 304 SS, as were all fuel rods in the CNA reactor. The

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assemblies were irradiated for three cycles to an average discharge burnup of 27 GWd/MT. PIE was performed on the rods at both SCK-CEN and CEA/Saclay.

A fifth MOX assembly manufactured by FRAMATOME was irradiated in Chooz A simultaneously. These irradiations were meant to lead to commercial self-generated recycle in Chooz A. In fact, an order for 112 MOX assemblies, sufficient for three successive reloads, was placed at about the time the five MOX bundles were originally loaded into the reactor (Ref. 49). However, no mention of this subsequent commercial use has been found. According to Ref. 13, this failure to proceed with commercial reloads was a result of the political decision by the United States to defer reprocessing indefinitely, although other sources have attributed the temporary decline of European MOX research to the rise of breeder reactor programs.

4.1.2.4   Oskarshamn LTAs

Three LTAs containing MOX fuel produced by BN were irradiated in the Swedish Oskarshamn 1 reactor. This 1375-MWt Asea-Atom BWR is also referred to in BN literature as the OKG1 reactor, for the owning utility (OKG Aktiebolag). Three island assemblies containing a total of 51 MOX rods were irradiated from 1975 to 1979.

4.1.2.5   Test Reactor and Non-LWR Irradiations

In addition to the demonstrations performed in commercial reactors, BN conducted a number of irradiations in various test reactors including the High Flux Reactor (HFR) at Petten, the Halden BWR, and the Belgian Reactor 2 (BR2). The VENUS critical facility was used to validate calculational methods. BN provided some fuel to Atomic Energy of Canada, Limited (AECL) for irradiation in the NPD-2 reactor. The available data do not indicate whether this was prototype fuel for CANDU reactors, but the pellet dimensions suggest that this is the case. The preliminary results reported indicate that an assembly average burnup of 16 MWd/MT had been reached, with the irradiation continuing (Ref. 37).

4.1.3   Commercial BN MOX Experience

Based on the successful irradiation of the MOX demonstration assemblies, a full quarter-core reload of MOX island assemblies was introduced into the Garigliano core in 1975. This reload consisted of 46 assemblies, each of which contained 32 MOX rods. The MOX rods were manufactured by BN to GE specifications. The bundles remained in the core through 1981, to a peak burnup greater than 25 GWd/MT (Ref. 34). Details about the irradiation performance and/or PIE of this fuel have not been located; however, it is likely that GE has access to at least a portion of this information.

By the early 1980s, BN had accumulated an appreciable experience base with their "reference" fuel. However, the solubility of the "reference" fuel was unsatisfactory, as up to 5% of the contained plutonium could not be dissolved in nitric acid during the head-end reprocessing operations. The MIMAS process was therefore developed by BN. Tests on MIMAS fuel expanded BN's MOX experience rapidly. Development of the MIMAS process happened to coincide with Électricité de France's (EDF's) decision to pursue commercial MOX use. A joint company, COMMOX, was formed by COGEMA and BN to market MOX production from BN's

P0 plant, COGEMA's CFCa plant, and the planned MELOX plant (Ref. 50). As a result, a large portion of P0's production went to French PWRs including Saint-Laurent B1, Saint-Laurent B2, Gravelines 3 and 4, and Dampierre 1 and 2. These irradiations are discussed further in Section 4.4.

In addition to the Franco-Belgian COMMOX venture, BN has supplied MIMAS fuel to a number of German reactors including Unterweser, Grafenrheinfeld, Philippsburg, Brokdorf, and Gundremmingen. BN has also provided MOX fuel for the Swiss Beznau 1 reactor, described more fully in Section 4.5 below. A summary of BN's commercial MOX deliveries with MIMAS fuel is contained in Table 5 taken from Ref. 8.

Table 5. BN's MIMAS commercial MOX deliveries

Delivery Year	Reactor	MOX Reloads
1987	EDF	1
	CNA	2
1988	EDF	2
	Beznau 1	1
1989	EDF	4
	Beznau 1	1
1990	EDF	4
	Beznau 1	1
1991	EDF	4
1992	EDF	2
	Beznau 1	2
	Unterweser	1
1993	EDF	2
	Grafenrheinfeld	1
	Philippsburg	1
1994	EDF	4
	Brokdorf	1

According to BN, irradiation experience with their MOX fuel has been quite good, with only ten recorded rod failures, only one of which, a hydride failure, can be attributed to the fuel itself. These failures are described in Ref. 51:

"1. A single end plug weld leak was observed in 1972 on a BR3 MOX fuel assembly after 1243 days at power and a peak pellet burnup of 59000 MWd/tonne HM. It was also a common type of defect in UO_2 fuel in these early days. This deficiency has been corrected by improving the welding technology and revising the welding specifications.

"2. A single hydride failure occurred in 1973 in a Dodewaard assembly. It was also a common mode of failure of UO_2 BWR fuel at that time. A

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revision of the pellet specification and of the fabrication technology has taken care of this problem.

"3. Coolant/cladding interaction failure was experienced in 1980 on six MOX rods in BR3 as a result of excessive crud deposition. This failure mode has been recognized to be due to inadequate primary water chemistry in conjunction with the average primary coolant temperature, which is lower than in modern PWRs, and to the surface heat flux level as a consequence of the irradiation conditions of those rods just below the maximum acceptable limits. It has no relation to the type of fuel inside the rod and disappeared with the adoption of a more restrictive permissible heat flux level in the core design criteria.

"4. Two failed MIMAS fuel rods were identified in 1990 in one MOX fuel assembly in Beznau-1. The two failed rods were located in two positions close to each other, and the investigations indicate a failure due to fretting by debris carried around by the coolant." [These failures are independent of the use of MOX, as similar failures were experienced in neighboring UO_2 assemblies.]

An interesting observation is that with the exception of the experimental BR3 reactor, no Belgian reactors were fueled with MOX until 1995. MOX fuel was introduced into two commercial Belgian reactors, Doel 3 and Tihange 2, in 1995.

4.2 ITALIAN MOX EXPERIENCE

The Italians undertook a substantial MOX development program in 1966. Two Italian organizations, Comitato Nazionale per l'Energia Nucleare (CNEN) and Ente Nazionale per l'Energia Elettrica (ENEL), pursued reprocessing and MOX recycle efforts simultaneously. Under this program, numerous test rod irradiations were performed across Europe. These early test irradiations, including the UO_2 tests used to baseline the MOX performance, are summarized in Table 6 (Ref. 52). The data obtained provided sufficient experience to undertake commercial MOX irradiations. ENEL operated two BWRs based on the GE NSSS, a PWR based on the Westinghouse NSSS, and a gas-cooled reactor during this period. Two of these reactors, the Garigliano BWR and the Trino PWR, were used for MOX development and demonstration.

The CNEN/ENEL program was one of the most comprehensive MOX research programs of its time. All facets of plutonium recycle were investigated. A pilot MOX fuel fabrication facility was constructed at the Cassacia Center in which research on pellet, powder VIPAC, and microsphere VIPAC fuel was conducted (Ref. 52, p. 364).

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Table 6. CNEN/ENEL test reactor irradiation experiments

Test	Reactor	Number of Rods	Fuel	Linear Power (kW/ft)	Burnup (GWd/MT) (as of 4/72)
IFA-130	Halden	12	UO_2	16.0	1.5
IFA-131	Halden	12	UO_2	16.0	31.5
IFA-124	Halden	2	UO_2-PuO_2	13.0	0.5
IFA-132	Halden	2	UO_2	27.5	11
IFA-133	Halden	2	UO_2	27.5	11
IFA-136	Halden	8	UO_2	16.0	15
IFA-170	Halden	8	UO_2-PuO_2	16.0	18
IFA-178	Halden	8	UO_2	16.0	20
SK-1	R-2	42	UO_2	8–40	not complete
SK-2	R-2	30	UO_2-PuO_2	8–40	not complete
AP-1	Ågesta	12/4	UO_2/UO_2-PuO_2	16.0	11
P-22	Kahl	36	UO_2-PuO_2	13.0	7
P-41	Kahl	36	UO_2-PuO_2	13.0	not complete
HFR-Gd	HFR	12	UO_2/UO_2-PuO_2	16.0	not complete

The test fuel manufactured in the Cassacia pilot plant represents the only known experience with burnable poison in MOX fuel. An all-plutonium 6 x 6 assembly manufactured for irradiation in the Kahl BWR contained two burnable poison rods: one enriched UO_2 rod and one $(U,Pu)O_2$ rod. The only reference to this assembly indicates that a burnup of 3500 MWd/MT had been reached with irradiation continuing (Ref. 36). An additional test in CEA's SILOE reactor discussed in the same reference contained four "rods containing either 1% or 2% Gd_2O_3 in either enriched uranium-oxide or MOX matrices, totaling four types of rods. For each rod type, irradiations are performed up to three different levels to check the gadolinium evolution at different stages." (Ref. 36, p. 270)

4.2.1 Garigliano MOX Irradiations

Four fuel fabricators were contracted by ENEL to manufacture a total of 16 fuel bundles containing 860 MOX fuel rods for the Garigliano BWR (Ref. 53). The plutonium was recovered from spent fuel from the Latina gas reactor. BN and ALKEM jointly supplied four bundles, GE supplied four bundles, and the United Kingdom Atomic Energy Authority supplied eight bundles (Ref. 52, p.363). The bundles were designed to have the same performance parameters as the UO_2 assemblies that they replaced. Twelve of the sixteen bundles were inserted into the reactor in 1968. In 1970, two of these twelve were removed for inspection, and the remaining four (of the original 16 bundles) were loaded. The four GE fuel assemblies included 12 VIPAC rods, 24 hot-pressed pellet rods, and 60 cold-pressed and sintered rods. The GE fuel was irradiated to ~25 GWd/MT assembly average burnup at a design peak linear heat rate of 15 kW/ft (Ref. 29, p. XV-2). All of the 204 rods produced by BN and ALKEM contained pellet fuel.

In 1975, a full reload of MOX-island assemblies was introduced into the Garigliano core. This reload consisted of 46 assemblies, each of which contained 32 MOX rods. GE prepared the fuel design, and contracted with BN for the actual MOX fuel fabrication. The bundles remained in the core through 1981 and were irradiated to a peak burnup greater than 25 GWd/MT (Ref. 34). Details about the irradiation performance and/or PIE of this fuel have not been located.

4.2.2 Trino MOX Irradiations

Westinghouse manufactured eight LTAs for irradiation in the Trino reactor. Trino is a small (270 MWe) PWR with a Westinghouse-supplied NSSS. The MOX assemblies were irradiated from 1975 to 1978 to a peak pellet burnup of approximately 35 GWd/MT (Ref. 22).

4.3 GERMAN (FORMER FEDERAL REPUBLIC OF GERMANY) MOX EXPERIENCE

MOX fuel development in Germany was conducted originally by Alpha Kemistry and Metallurgy GmbH (ALKEM). ALKEM was eventually purchased by Kraftwerk Union Aktiengesellschaft (KWU). Siemens Aktiengesellschaft eventually purchased KWU in 1988. Thus, references to MOX fuel development by the same group over a period of years can be attributed to either ALKEM, KWU, or Siemens. For simplicity, most of the work described in this section is attributed to ALKEM because of the historical nature of this summary.

ALKEM's original investigations into MOX fuel fabrication and utilization began in the 1960s. A single MOX fuel assembly was inserted into the Versuchsatomkraftwerk (VAK) Kahl BWR in 1966. Additional MOX insertions into VAK were made through the 1960s and early 1970s. ALKEM's first PWR MOX fuel, designed by KWU, was inserted into the Kernkraftwerk Obrigheim (KWO) in 1972. MOX fuel was also supplied by ALKEM for the Mehrzweckforschungsreaktor (MZFR), a prototype pressurized heavy water reactor. Commercial MOX use was expanded to German BWRs in 1974 with the insertion of 16 MOX assemblies into the Kernkraftwerk Gundremmingen. MOX utilization in these reactors slowly expanded during the 1970s and eventually expanded to additional German reactors.

ALKEM utilized their "Former Standard" pellet fabrication process for all the fuel fabricated through 1981. Table 7, reproduced from Ref. 54, summarizes ALKEM's experience with this early MOX fuel. Fuel performance with "Former Standard" fuel was adequate according to the manufacturer. Some early fuel failures were attributed to local hydriding that resulted from excessive moisture in the fuel. Improvements to the pellet-drying process subsequently eliminated this type of failure (Ref. 39). The generally good MOX fuel behavior is consistent with the experience of the Belgians from irradiation of fuel produced using their similar "Reference" process. However, as with the early Belgian fuel, the irradiated MOX was not sufficiently soluble in nitric acid to satisfy reprocessing requirements. The AUPuC and OCOM processes were, therefore, developed to address this insolubility.

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Table 7. Siemens/KWU-ALKEM experience with "Former Standard" MOX fuel

Reactor (Type)	Year of First Insertion	Number of All-MOX Fuel Assemblies	Number of MOX Island Assemblies	Total Number of Rods	Maximum Assembly Burnup Achieved (GWd/MT)
VAK, Kahl (BWR)	1966	7	88	972	21
KWL, Lingen (BWR)	1970		1	15	26
KRB-A, Gundremmingen (BWR)	1974	64		2240	20
MZFR, Karlsruhe (PHWR)	1972	8		296	14
KWO, Obrigheim (PWR)	1972	33		5940	35
Total		112	89	9463	

ALKEM produced MOX fuel using the OCOM and AUPuC processes from 1981 to 1991. The 35-MT/yr pilot fabrication line was then shut down in anticipation of the opening of the 120-MT/yr second generation MOX plant that has since been abandoned. ALKEM's experience base with OCOM and AUPuC fuel is summarized in Table 8, taken from Ref. 54. The performance of the two fuels is quite similar, as one would expect from their very similar microstructures, and is basically equivalent to that of modern UO_2 fuel.

Table 8. Siemens/KWU-ALKEM experience with OCOM and AUPuC MOX fuel

Reactor (Type)	Year of First Insertion	Number of All-MOX Assemblies	Total Number of Rods	Fuel Type	Maximum Assembly Burnup Achieved (GWd/MT)
KWO, Obrigheim (PWR)	1981	29	5220	OCOM/AUPuC	35
GKN-1, Neckarwestheim (PWR)	1982	32	6560	OCOM	42
KKU, Unterweser (PWR)	1984	20	4720	OCOM/AUPuC	37
	1987	28	6496	OCOM/AUPuC	25
BZN-2, Beznau-2 (PWR)	1984	52	9308	OCOM/AUPuC	36
KKG/BAG, Grafenrheinfeld (PWR)	1985	16	3776	OCOM/AUPuC	34
		16	3712	OCOM/AUPuC	34
KKP-2, Philippsburg (PWR)	1988	12	2784	OCOM/AUPuC	18
KWG, Grohnde (PWR)	1988	20	4640	OCOM/AUPuC	27
KBR, Brokdorf (PWR)	1989	20	4640	OCOM/AUPuC	13
Total		245	51856		

Although Siemens no longer has MOX fabrication capabilities, MOX utilization continues in a number of German reactors. The MOX fuel is still fabricated to specifications provided by Siemens, but the fuel itself is fabricated by BN, COGEMA, and BNFL. None of these

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fabricators produces OCOM or AUPuC fuel. In fact, MIMAS fuel supplied by COMMOX is currently loaded in several German reactors.

In addition to the standard irradiation tests described above, ALKEM performed PIEs on numerous irradiated MOX rods (Ref. 15). These examinations on thirteen "Former Standard" rods, six AUPuC rods, and four OCOM rods (as of 1987) indicate the performance similarities between MOX and UO_2 fuels. The dimensional changes as a result of irradiation (both densification and swelling) have been less with MOX. The fission gas release is higher, although the modern OCOM and AUPuC fuels have lower fission gas release rates than the earlier "Former Standard."

Siemens also performed ramp testing on MOX fuel in the KWO and HFR Petten reactors. The tests in general indicate that MOX fuel is actually superior to UO_2 fuel with respect to response to rapid power increases.

"1. In KWO, 14 test rods with former standard MOX fuel, preirradiated at KWO to a burnup range of 9.0 to 21.8 MWd/kg (m), were ramp tested starting at 160 to 360 W/cm and leading to 270 to 420 W/cm. Thereafter, two of these ramp-tested rods were ramp tested a second time after a further preirradiation period at 165 to 230 W/cm and a burnup of 17 to 27 MWd/kg (M) to 260 W/cm.

"2. In the High Flux Reactor (HFR), Petten, The Netherlands, 10 test rods with former standard MOX fuel, preirradiated at KWO to a burnup range of 9.3 to 32.1 MWd/kg (M), were ramp tested between 290 and 480 to 560 W/cm.

"3. In the HFR Petten, three test rods with AUPuC-MOX fuel, preirradiated to ~35 MWd/kg (M), were ramped from 250 W/cm to 420 to 490 W/cm" (Ref. 15).

No defects resulted from these ramp tests. At the time of the publication (1987), additional ramp tests with both AUPuC and OCOM fuels were planned. Although Siemens' experience, like that of BN, is well documented in open literature, the company undoubtedly maintains a much more complete, albeit proprietary, data base.

4.4 FRENCH MOX FUEL EXPERIENCE

The French nuclear industry is quite complex to the outside observer. A brief explanation of the relationships between the different companies is therefore useful for understanding the follow-on discussion. Development through the early 1980s was concentrated within the Commissariat à l'Énergie Atomique (CEA). As part of a 1983 reorganization, CEA-Industrie was formed as a holding company, which owns CEA's industrial interests. As part of this reorganization, COGEMA was created for fuel cycle activities. FRAMATOME is the French NSSS designer, originally as a Westinghouse licensee but independent since 1981. FRAMATOME and COGEMA jointly fabricate LEU fuel and fuel assembly hardware through their subsidiary

Société Franco-Belge de fabrication de combustibles (FBFC). Marketing of LEU fuel assemblies is also controlled through a joint subsidiary, FRAGEMA. COGEMA is solely responsible for the fabrication of MOX fuel, but design and fabrication of the fuel assembly hardware is performed by FRAMATOME and FBFC. COGEMA and BN jointly market the MOX fuel production from BN's P0 and from COGEMA's CFCa and MELOX MOX plants through COMMOX. EDF is the French national utility.

CEA and its successor COGEMA have been involved in MOX fuel research for many years. However, the early focus in France on plutonium utilization focused on in-situ utilization through extended burnup in their gas-cooled reactors. It was also planned that plutonium produced in these gas-cooled reactors would be utilized as fuel for fast reactors. Plutonium utilization in thermal reactors was investigated as a contingency. CEA collaborated with BN and RBG for insertion of four MOX LTAs into the CNA reactor in 1974. These four island assemblies contained a total of 160 MOX fuel rods, 16 of which were fabricated by CEA at Cadarache. This project was conducted under one of the 48 contracts among the Commission of the European Communities (CEC) and various research and industrial organizations. FRAMATOME supplied two full-MOX LTAs containing rods produced by CEA at Cadarache to CNA for irradiation starting in 1975.

Following this early investigation, French interest in thermal recycle dropped. The lack of French interest in thermal recycle was instrumental in stopping the renewal of the CEC program on plutonium recycle in LWRs. Plutonium fuel work in France continued, but was focused exclusively on fast-reactor applications.

By 1985, the expected fast-reactor fuel cycle had not materialized as expected. Simultaneous success with reprocessing plants had created a substantial and growing surplus of separated plutonium. EDF responded to this situation by announcing its decision to pursue MOX utilization in the 900 MWe series of PWRs. Because of the historical cooperation between the French fuel suppliers and BN through FBFC, the adoption of BN's MIMAS process for production of the necessary MOX fuel was straightforward. The first reload quantity of MOX fuel was delivered to St. Laurent B1 in 1987. Additional reloads have followed in many of EDF's other 900-MWe class reactors.

MOX fuel assemblies for the French reactors have been produced in the three facilities that supply fuel to COMMOX: P0, CFCa, and MELOX. Some of the early fuel produced in CFCa was not MIMAS, because the plant was only recently converted to this process. CFCa formerly utilized a comilling process known as COCA that was developed for fast-reactor fuel fabrication. No breakdown in the type of fuel utilized in EDF's reactors has been located, but the vast majority has clearly been MIMAS. The early fuel was fabricated with ex-AUC UO_2, but more recent fuel has been produced with TU2 UO_2.

French operational experience with MOX fuel has been reported more frequently in the last few years as postirradiation results from the early MOX fuel assemblies have become available. However, it is clear that EDF and COGEMA have relied extensively on BN's experience base. In addition to participation in many of BN's international programs, FRAMATOME, EDF, and CEA have conducted a joint analytical and experimental program to investigate MOX fuel performance (Ref. 55). The results of this study were consistent with BN's findings that

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MIMAS MOX fuel performance is roughly equivalent to that of similar LEU fuel and adequate for the 1/3 core refueling strategy being utilized. Fission gas release late in fuel life was found to be the only topic warranting additional intensive study. Continued investigation under the FRAMATOME-EDF-CEA program focused on fuel performance under both steady-state and transient conditions (Ref. 56). The experimental results from the various experimental projects conducted under the auspices of the FRAMATOME-EDF-CEA program have been utilized to improve, extend, and validate the computational fuel performance codes. The studies have investigated fission gas release, thermal stability, irradiation-induced densification, pellet-clad mechanical interaction, and reactivity insertion accidents (Ref. 57).

The future plans for MOX utilization in France are quite ambitious. EDF plans to obtain licensing authority to load MOX fuel in any or all of the 28 existing 900 MWe class PWRs in France. The recent approval for MOX introduction and utilization in the four Chinon units brings the total number of French reactors that are licensed to burn MOX fuel to 20. This number is able to absorb the licensed production from MELOX, currently 100 MTHM/yr. An extension to MELOX, the MELOX West Fitting Building, is expected to be commissioned during 1999. The claimed full capacity of MELOX including this extension is 250 MTHM/yr.

4.5   SWISS MOX EXPERIENCE

Nordostschweizerischen Kraftwerke (NOK) operates a total of five reactors including the two Westinghouse-designed Beznau units. Four MOX LTAs were loaded into Beznau-1 in 1978 (Ref. 22). Routine MOX utilization began in 1984 with the insertion of a MOX reload into Beznau-2. This fuel and subsequent reloads were fabricated by ALKEM using the OCOM and AUPuC processes (Ref. 54). MOX fuel was also utilized in Beznau-1, starting in 1988 with the insertion of a reload of MIMAS MOX fuel supplied by COMMOX (Ref. 51). One noteworthy aspect of this MIMAS fuel is that some of it is in the form of segmented rods; these rod segments have been utilized in several international programs conducted by BN. Beznau-1 has also irradiated SBR MOX LTAs fuel fabricated by BNFL in the MOX Demonstration Facility at Sellafield. NOK plans to continue MOX utilization in Beznau through the end of their operating lives. In addition, MOX fuel use was expanded to the Göesgen reactor in 1997 (Ref. 58).

4.6   UNITED KINGDOM MOX EXPERIENCE

In the early 1960s, limited plutonium fuel fabrication capacity was installed in a laboratory at the UKAEA's Windscale complex (now Sellafield). This laboratory produced limited quantities of fuel for a variety of reactors including BR3 and Garigliano. The emphasis was on LMR fuel production, however, because of faith in the rapid implementation of an LMR fleet. The UKAEA did participate in the CEC program investigating plutonium recycling in thermal reactors. However, the UK's emphasis on LMR development helped prevent the extension of the CEC program when it came up for renewal in 1988.

Throughout the 1970s and early 1980s, various LMR fuel fabrication processes were researched at Windscale. High-density annular pellet fuel was chosen over low-density solid fuel, VIPAC, and SPHEREPAC. Additional fabrication capacity was required to supply the fuel required for

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the planned irradiation program. Design and construction of a new facility for enhanced throughput (FACET) proceeded from the mid-1980s.

FACET was ready for commissioning in 1988. Changes in energy policy in the United Kingdom were reflected through reduced LMR budgets. With this change, the need for FACET evaporated. The facility was never fully commissioned. Concurrent developments in the rest of Europe resulted in the decision by the UKAEA and BNFL to convert FACET to the MDF. This conversion was completed in 1993.

One of the key aspects of MDF's development was the adaptation of the lessons learned in LMR fuel development to LWR MOX applications. The wet binder granulation process was abandoned in favor of the SBR because of the simplifications it entailed (Ref. 59).

Although a detailed accounting for MDF's production has not been found, it is known that both LTAs and reload assemblies have been supplied to Beznau from MDF. Fuel has also been produced for irradiation in test reactors as part of the qualification of SBR fuel. Furthermore, the extensive LMR fuel development that led to development of the SBR provides additional irradiation performance information.

A large second-generation SBR fabrication plant is under construction at Sellafield. This 120 MT/yr SMP is based on the technology proven in MDF. SMP is nearly complete and is awaiting authorization to start uranium commissioning.

Because BNFL is a relative newcomer to the LWR MOX market, their experience base with SBR fuel is limited compared to that of COMMOX and Siemens. Nevertheless, SBR test fuel has been successfully irradiated, and the associated irradiation results reported (Ref. 60). Additional information will become available when the Beznau-1 LTAs are subjected to PIE over the next 1–2 years (Ref. 61). One of the four MDF LTAs supplied to Beznau-1 included a group of 8 specially characterized rods, which will be the focus of the PIE.

BNFL has claimed that the SBR process produces a more homogeneous fuel than the master mix processes. The fabrication experience obtained in MDF has demonstrated that good homogeneity is obtained (Ref. 61). Electron probe microanalysis has been utilized to confirm the results of alpha autoradiography. The highest reported plutonium concentration in the plutonium-rich zones is nearly that of master mix, about 30% using a 1 μm spot size for analysis. The plutonium-rich zones in SBR fuel are typically less than 30 μm. This homogeneity is roughly equivalent to that found in master-mix fuel. MIMAS fuel typically contains plutonium-rich zones that are 50 μm or less in equivalent diameter, and these plutonium-rich zones consist of master-mix containing 30%–40% plutonium. The issue of whether SBR or master-mix processes produce better homogeneity cannot be resolved until statistically significant quantities of SBR fuel have been irradiated and subjected to PIE. The fact remains that both the SBR and master-mix processes produce fuel containing no plutonium particles large enough to cause problems during power transients.

Although BNFL and the UKAEA have extensive LMR fuel experience and experience with thermal plutonium recycle, plutonium is not utilized in Sizewell B, the only PWR in the United Kingdom.

4.7 JAPANESE MOX EXPERIENCE

Plutonium fuel development work in Japan has historically been performed under the auspices of the Power Reactor and Nuclear Fuel Development Corporation (PNC), now the Japan Nuclear Cycle Development Institute (JNC). A small laboratory, the Plutonium Fuel Development Facility (PFDF), was constructed in 1965 by PNC at the Tokai Works to study the basic properties of MOX fuel, to develop fabrication processes, and to produce fuel for irradiation testing. Both LMR and thermal MOX fuels were fabricated in this laboratory. Several fabrication processes were investigated in the PFDF including comilling, master-mix, coprecipitation, VIPAC, and SPHEREPAC. Fuel pin tests with these fuels were conducted in both the GETR and the Japan Research Reactor. Fuel assembly tests were conducted in Saxton, the Halden BWR, Mihama-1, and Tsuruga-1.

In cooperation with the Japan Atomic Energy Research Institute (JAERI), PNC fabricated two 3 x 3 assemblies for irradiation in the Halden BWR. One of the assemblies contained SPHEREPAC fuel and the other contained comilled fuel (Ref. 62). This test demonstrated no safety-significant differences between the fuel behaviors. PNC also fabricated one complete fuel assembly plus 4 additional fuel rods for irradiation in Saxton Core III. All of the Saxton fuel rods contained comilled pellet fuel. PNC also fabricated 2 rods incorporated into 2 of the 4 MOX fuel LTAs fabricated by Westinghouse and inserted into Mihama-1 (Ref. 63). PNC produced 48 MOX fuel rods for incorporation into 2 island-type MOX fuel LTAs inserted into Tsuruga-1 under a joint program between PNC and Japan Atomic Power Company (JAPCO). Additional MOX fuel rods were produced in PFDF for fast reactor irradiation tests and for critical experiments.

A larger facility, the Plutonium Fuel Fabrication Facility (PFFF), was completed in 1972. This facility was constructed near the PFDF at Tokai Works to produce larger quantities of fuel for both fast and thermal reactors. Two separate lines were installed: one for production of Advanced Thermal Reactor (ATR) fuel and a second for production of LMR fuel. The ATR line was utilized to produce over 9 MT of MOX fuel for use in the Deuterium Critical Assembly (DCA). It has since been utilized to produce MOX fuel for the Fugen ATR. The LMR line has produced test fuel for irradiation in test and prototype LMRs including GETR, Japan Materials Test Reactor, Rapsodie, Joyo, and Dounreay (Ref. 64).

In 1988, the Plutonium Fuel Production Facility (PFPF) was completed for supplying Monju fuel. PFPF is a large pilot facility complete with a pusher furnace. The plant is highly automated and experienced some operational difficulties during early operation as a result. These problems have been resolved, but Monju's sodium leak in December 1995 and the political fallout resulting from that leak have drastically reduced the need for fuel from PFPF.

Plutonium utilization policy in Japan is currently under governmental review. In February 1997, the Japanese Cabinet approved a nuclear policy on "Utilization of Plutonium in LWRs" that called for the introduction of MOX fuel into a single BWR and PWR during 1999. The subsequent accident in the Bituminization Facility in PNC's Tokai reprocessing plant resulted in a postponement in this schedule.

Tokyo Electric Power Co. and Kansai Electric Power Co. originally planned to load a BWR and PWR, respectively, during 1999 and to add one additional unit each during 2000. Longer term policy is for all utilities to implement MOX utilization by 2010. The political uncertainty resulting from the Monju leak and the Tokai fire will likely delay this implementation schedule. Japanese utilities currently store more than 10 MT of separated plutonium at Sellafield and La Hague. The MELOX extension and SMP are both being constructed in part to produce MOX fuel from this plutonium for utilization in Japanese reactors. Most plans for MOX utilization in Japanese reactors call for 30% MOX in the cores, but plans also exist for construction of one or more GE Advanced BWRs at Ohma that will burn full MOX cores.

4.8 MOX EXPERIENCE IN THE RUSSIAN FEDERATION AND FORMER SOVIET UNION

Plutonium utilization in the Russian Federation has been focused on LMR applications. Plutonium fuel has been tested in four LMRs: the BR-10 at Obninsk, the BOR-60 at Dimitrovgrad, the BN-350 at Aktau, and the BN-600 at Beloyarsk (Ref. 65). Four fabrication technologies have been investigated: comilling, VIPAC, sol-gel, and coprecipitation. The total quantity of fuel produced is over 1 MTHM. Pellet technology has been developed by researchers at the A. A. Bochvar All-Russian Research Institute of Inorganic Materials (VNIINM) and implemented at the large laboratory scale in the Paket laboratory at the Production Association Mayak. VIPAC technology and the associated pyroelectrochemical reprocessing technology used to produce high density VIPAC feed have been developed by researchers at the All-Russian Research Institute of Atomic Reactors (NIIAR). The VIPAC technology has been demonstrated extensively in the BOR-60 LMR at NIIAR.

MOX fuel has never been utilized in Russian PWRs. Less than 10 rods containing MOX test fuel have been irradiated in the Materials Irradiation Reactor (MIR) at NIIAR. The fabrication processes utilized have been taken from LMR development work.

Ambitious plans for implementation of a plutonium economy have been delayed and modified as a result of economic difficulties. The second generation reprocessing plant at Krasnoyarsk (known as RT-2), capable of reprocessing PWR fuel, has not progressed beyond the conceptual design stage. Limited construction work has been performed, but the design has never even been finalized. Some recent reports by officials of the Russian Ministry of Atomic Energy (MINATOM) indicate that RT-2 has been canceled.

The existing reprocessing plant at Chelyabinsk has separated more than 30 MT of plutonium. The large MOX fuel fabrication plant planned to convert this plutonium into MOX fuel has only been partially completed (<20%). No credible schedule for completion of this Complex-300 plant has been located. Therefore, MOX fabrication capacity in Russia is limited to the existing laboratories at PO Mayak and at NIIAR. Neither lab is equipped for production of MOX fuel meeting PWR specifications.

5. SUMMARY AND CONCLUSIONS

The worldwide MOX experience constitutes a vast body of evidence suggesting that irradiation of surplus WG plutonium as MOX fuel in commercial nuclear reactors is a technically viable disposition option.

The most important goal of the effort documented in this report is to determine what is known worldwide about MOX fuel, and what additional information is needed to irradiate the excess weapons plutonium safely in domestic commercial reactors.

5.1 SUMMARY OF FINDINGS

MOX fuel has been investigated for almost forty years in the United States and abroad. Domestic MOX fuel development, by 1975, had progressed to the commercial demonstration stage. The plutonium used in much of the domestic research and development had a high fissile content, similar to that of surplus WG plutonium. Commercial use of MOX fuel in the United States was delayed by completion of the *Final Generic Environmental Statement on the Use of Recycle Plutonium in MOX Fuel in Light Water Cooled Reactors*, commonly known as GESMO, and the availability of plutonium from commercial reprocessing plants. Domestic MOX research ended by 1980 as a result of President Carter's Executive Order on nonproliferation that indefinitely postponed commercial reprocessing. At the time, overseas MOX research for the most part lagged behind domestic efforts. In the intervening years, however, continued development and commercial utilization overseas has eclipsed the domestic experience.

Overseas development of MOX did not end in the 1970s as hoped by domestic policy makers. However, it received less attention than did the utilization of plutonium fuels in LMRs. Several European countries researched MOX during this period, but Belgium and Germany continued appreciable MOX programs despite the expected emergence of a plutonium-fueled LMR economy. The Belgian and German programs led to the development and commercial utilization of state-of-the-art MOX fuels by the mid-1980s.

Later in the 1980s, as it became apparent that the expected LMR deployment schedule would either be delayed or canceled, France and then the United Kingdom renewed their respective MOX programs. The French licensed BN's MIMAS process and planned for rapid expansion of their utilization of MOX fabricated by this process. In the United Kingdom, the UKAEA and BNFL combined their efforts to develop the SBR process for MOX manufacture.

Because of various developments and programs in Europe, commercial MOX use is a reality in a number of European countries. The experience base resulting from such commercial use dwarfs the previous U.S. MOX experience in terms of both breadth and depth. The gap between domestic and overseas knowledge continues to grow.

In addition to the European efforts, the Japanese have actively pursued MOX continuously since the 1960s. One of the latest developments in the Japanese MOX program is the plan to build

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new Advanced Boiling Water Reactors fueled entirely with MOX. A Japanese MOX fuel manufacturing plant is also planned.

It is important to remember, when reviewing the worldwide experience, that the goals and requirements of the FMDP are different from those driving the commercial MOX programs throughout the world. Commercial MOX programs are meant to close the nuclear fuel cycle by providing economical fuel from reprocessed nuclear materials. In the commercial context, plutonium is a valuable resource, not a troublesome legacy. The goal of commercial MOX programs is therefore to maximize the energy obtained from it.

In the FMDP, plutonium disposition—not economic energy production—is the goal. Irradiation is meant to make the plutonium self-protecting through the generation of an intense radiation field. Because the goals of the FMDP differ from those of commercial plutonium recycle, optimization results in distinct approaches. For example, in existing commercial MOX programs, MOX fuel is limited to a fraction of the core load—typically 30% to 50%. In the FMDP, because high-plutonium throughput rate is one of the most important requirements, full-core loads of MOX have been proposed. If the FMDP requirements fall outside that covered by existing data, the merits and costs of required development programs will have to be weighed against alternatives that closely mimic existing commercial MOX programs. These issues are more fully explored in the FMDP report on fuel qualification issues (Ref. 66).

5.2   CONCLUSIONS

One of the most important lessons learned during this review is that MOX fuel performance information is specific to a particular fuel fabrication process. Thus, to apply one of the existing experience bases to licensing efforts, the corresponding production process would have to be utilized for manufacturing the mission fuel. For a number of production processes, the existing performance data are sufficient to justify overseas commercial utilization of the MOX fuel. This is the case with the Belgian (and now French) MIMAS fuel, the German OCOM and AUPuC fuel, and BNFL's SBR fuel. Although licensing regimes overseas are not exactly compatible with NRC regulations, it is possible that if the data are sufficient for licensing in these European countries, they are sufficient to support the majority of NRC licensing concerns.

Another important conclusion drawn from this work is that development (and qualification) of a new domestic MOX manufacturing process would likely entail a lengthy and expensive program. Without assistance from one of the overseas MOX manufacturers, many years and hundreds of millions of dollars would likely be required to recover the previous experience base, adapt the production procedures to meet modern requirements (in terms of worker safety, environmental compliance, and product quality), and satisfy both licensing and end-user requirements.

Full MOX usage in a reactor has never been demonstrated. In cores limited to less than 50% MOX, the excess reactivity can be compensated through alternative means, including incorporation of burnable poison into the UO$_2$ rods. No technical impediments to incorporation of burnable poison in MOX have been found, and indeed some preliminary studies of such incorporation have been performed overseas. However, the fuel development program necessary to include burnable poison in MOX would likely require many years and tens of millions of

dollars. Foreign research (either ongoing or planned for the near term) could help accelerate the U.S. program, if cooperation were to be arranged.

Gallium is present in weapons plutonium and will have to be dealt with. Gallium removal from the MOX is desirable in terms of overall fuel irradiation performance because gallium behavior under irradiation is unknown. ORNL, LANL, and INEEL are currently investigating the array of issues associated with gallium.

Americium impurities, caused by decay of ^{241}Pu, have also been identified as a potential source of irradiation behavior difficulties. However, upon closer inspection, it was determined that americium is of more concern during fuel fabrication than during irradiation. Commercial MOX often contains much more americium than does weapons plutonium. Finally, research performed under this program suggests that the americium ceases to represent an exposure problem once it is blended with UO_2.

Two other potential concerns, the proposed use of higher plutonium concentrations than those used in commercial MOX and the use of depleted rather than natural uranium as the diluent in MOX, have been identified. Plutonium concentrations in the range proposed for the FMDP mission will almost certainly be within the span of the foreign experience base. Also, depleted uranium has been used previously as the diluent in MOX and is not expected to present any difficulties for the FMDP. In the commercial arena, the choice of diluent material has been driven primarily by availability (and cost) of the material. Thus, these two issues are not of major significance to the FMDP. The concerns identified do not present insurmountable obstacles to the use of WG plutonium in LWRs.

The current direction of the FMDP program is (1) rapid initiation of plutonium disposition, (2) low technical risk associated with initial plutonium disposition, (3) decoupling of fuel development success from mission success, (4) maximum use of commercial technology, and (5) gradual movement from known parameter space (partial core loads/no burnable poison) into a more desirable range (full core loads/burnable poison) or the use of more reactors. A detailed fuel development program plan may be developed when the necessary decisions regarding program goals and fuel performance requirements have been made by the DOE.

In conclusion, the existing MOX fuel experience base suggests that disposition of excess weapons plutonium through irradiation as MOX in LWRs is a technically viable option. This conclusion was reached through a review of the open foreign literature and a categorization and reorganization of the domestic experience base. Most of the detailed irradiation performance data remains proprietary and is unavailable. Based on the available information, it appears that adoption of foreign fuel technology and MOX fuel use patterns from one of the successful MOX fuel vendors will minimize the risks to the overall mission.

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