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Conceptual Design of a Pilot-Scale Pyroprocessing Facility

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Abstract — Argonne National Laboratory and Merrick & Company developed the conceptual design of a pilot-scale (100 T/year) pyroprocessing facility for the treatment of used fuel generated by commercial light water reactors and subsequent treatment of waste streams generated during the process. The primary purpose of this study was to perform sufficient engineering for the pilot facility conceptual design so that credible capital and operating cost estimates could be developed. Initial safety, safeguards, and security assessments were also completed to provide a detailed evaluation in these areas that can significantly affect both capital and operating costs. Electrorefining-based pyroprocessing resulted in a compact hot-cell facility with few process equipment systems. The process equipment and support systems were estimated to cost \$93 million and the facility \$305 million for a project total cost of \$398 million. The annual operating cost was estimated at \$53 million/year. Scaling up to a commercial-scale (400 T/year) was also evaluated and the capital cost was estimated at \$911 million with annual operating cost of \$90 million/year.

Keywords — Pyroprocessing, nuclear fuel cycle, spent fuel management.

Note — Some figures may be in color only in the electronic version.

I. INTRODUCTION

An early version of pyroprocessing based on melt-refining was employed for the fuel cycle closure demonstration in the Experimental Breeder Reactor-II (EBR-II). About 35 000 fuel pins were recycled based on melt-refining and injection-casting fabrication in the adjacent Fuel Cycle Facility (FCF) with a typical turnaround time of 45 days from 1965 through 1969.¹ However, melt-refining could not remove noble metal fission products and separate higher actinides from uranium. When the Integral Fast Reactor (IFR) Program² was initiated in 1983, an electrorefining process was adopted in place of melt-refining. The pyroprocessing technology was further developed during the IFR Program, and the original EBR-II FCF was refurbished using the new electrorefining-based

equipment to demonstrate pyroprocessing at the engineering scale.

The IFR Program and EBR-II operation ended in 1994, however the refurbished FCF has been in operation since 1996 treating the EBR-II used fuel for disposal purposes.^{3–5} Hence, the application of pyroprocessing for metal-fueled fast reactor fuel has successfully been demonstrated at the engineering scale. Pyroprocessing development continued as part of the Nuclear Technology Research and Development Program and its predecessor programs, which were funded by the U.S. Department of Energy (DOE), Office of Nuclear Energy. Although the sodium-cooled fast reactor (SFR) with metal fuel and pyroprocessing will remain as a most promising long-term next-generation reactor/fuel cycle system, in the near term, pyroprocessing can be applied for treatment of the current generation commercial light water reactor (LWR) used fuel to make its ultimate disposal simpler.^{6–8}

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The radiological toxicity of typical LWR used fuel over the years is shown in Fig. 1 (Ref. 2). The radiological toxicity due to the fission product portion of the waste decays below the natural uranium ore level in about 300 years and becomes relatively harmless. On the other hand, the toxicity level associated with the actinide portion stays far above uranium ore and remains at least three orders of magnitude greater than fission products for hundreds of thousands of years. If 99.9% of actinides are removed from the waste form, then the radiological toxicity of the remaining 0.1% of actinides stays below the natural uranium ore at all times and the effective lifetime of the waste is reduced from about 300 000 years to about 300 years. A geologic repository is still required but any regulatory requirements or performance requirements placed on the repository can be met on an a priori basis, and hence, the repository siting and the repository design will become easier tasks. The recovered actinides need to be transmuted, which is feasible only in fast spectrum reactors.²

The FCF demonstration for EBR-II metal fuel is not directly applicable for treatment of the LWR used fuel. First, a front-end step of oxide-to-metal conversion is required to use the demonstrated metal fuel electrorefining process. In FCF, the equipment size was constrained by criticality considerations. Since the fissile content of the LWR used fuel is about an order of magnitude smaller than the fast reactor fuel, scale-up of unit operations is required for economic viability within criticality constraints. Pyroprocessing was originally developed for recycling fast reactor metal fuel, but Japanese utilities were interested in the feasibility for the LWR used fuel application and provided the necessary funding for Argonne to conduct research on the oxide-to-metal reduction processes. Initial efforts were focused on a

lithium reduction step, which was determined not to be effective for commercial application. In later years, electrolytic reduction was proven most effective. On the scale-up potential, parallel plate electrodes were developed successfully to facilitate scale-up of the technology.

Previous conceptual design efforts demonstrated the potential of pyroprocessing very well.⁶⁻⁸ In this study, a joint team from Argonne and Merrick & Company performed sufficient engineering for the conceptual design so that credible capital and operating costs could be developed. The Argonne team developed a process flowsheet amenable for commercial-scale operation, the process equipment designs, and their cost estimates. The Merrick & Company team developed the facility design and provided the construction cost and schedule estimates. Areas (i.e., safety, safeguards, and security) that can significantly affect designs and costs were assessed on the completed design and updates were incorporated in the designs and cost estimates.

II. PYROPROCESSING FLOWSHEET

The process flowsheet adopted in our facility design for the treatment of LWR used fuel is shown in Fig. 2. The treatment process can be divided into five functional areas: fuel preparation, oxide-to-metal conversion, actinide and fission product separations, material recycle, and waste treatment.

In the fuel preparation step, fuel assemblies are mechanically disassembled and cladding removed. This approach reduces the volume of materials processed and eliminates some front-end chemistry issues. However, a drawback is that the cladding may contain some actinides. The actinide loss is minimal for low-burnup fuel but could increase up to a fraction of a percent for burnups greater than 50 000 MWd/T.

Electrolytic reduction converts the fuel oxides into metals⁹⁻¹² for subsequent treatment in the electrorefining process. The oxide fuel, contained in basket modules, serves as the cathode of the electrolytic cell where the oxide-to-metal conversion occurs. Oxide ions released from the fuel during the conversion process are transported in the molten salt electrolyte (i.e., lithium chloride with approximately 1 wt% lithium oxide maintained at 650°C) to the carbon anode where they react to produce a mixture of CO and CO₂ gas. Active metal (e.g., Cs, Sr, Rb, Ba), halide (e.g., I), and chalcogenide (e.g., Se, Te) fission products partition to the salt phase during the process. The bulk of the fission product gases (e.g., Xe, Kr) in the oxide fuel are released from the fuel matrix and sent to

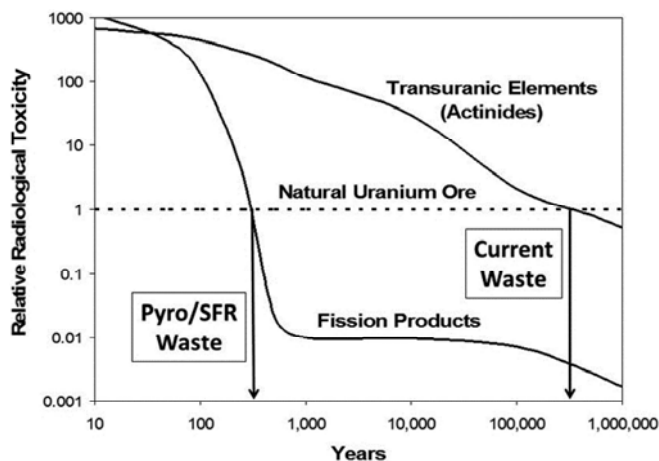


Fig. 1. Relative radiological toxicity of used fuel constituents (from Ref. 2).

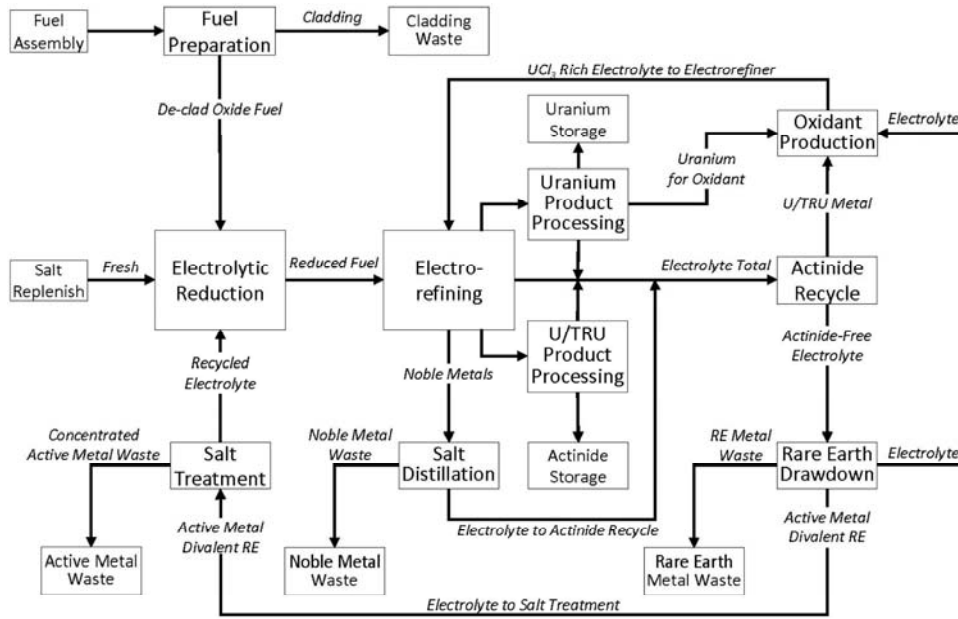


Fig. 2. Pyroprocessing flowsheet adopted for the conceptual design.

the off-gas treatment system along with the CO and CO₂ gas stream.

The electrorefining process^{13–16} separates a purified uranium product and a codeposited transuranic (TRU)/uranium product from the fission products. The reduced metal contained in the basket module from the electrolytic reducer functions as the anode. As electric current is passed between the anode(s) and cathode(s), actinides are anodically dissolved into the molten salt and transported through the electrolyte, which is comprised of lithium chloride plus actinide and fission product chlorides at 650°C, and deposited onto two types of cathodes that differ in design and electric potential. One set of cathodes is for uranium deposition and the other set is for codeposition of TRU elements and uranium.

Rare earth (e.g., Gd, Nd, La) fission products partition to the salt phase during the refining process and are recovered in subsequent operations. Active metal, halide, and chalcogenide fission products that enter the electrorefining process in the salt adhering to the basket module from the electrolytic reduction process remain in the salt phase during the process. Noble metals (e.g., Zr, Tc, Mo) that remain in the basket module after electrorefining are recovered and treated in a subsequent salt distillation operation that also reclaims any adhering electrolyte salt, which is recycled. The noble metals are converted to an engineered waste form comprised of the noble metals, iron, and zirconium.¹⁷

The U and TRU/U products from the electrorefiner are transferred to their respective product processing operations to recover the adhering electrolyte salt and create consolidated U or TRU/U ingots. In the case of uranium product processing, the salt is removed via vaporization prior to consolidation of the metal into an ingot. Salt removal from the lower-melting TRU/U alloy is accomplished by melting the product, allowing the metal and salt phases to separate due to the large density difference between the salt and metal phases, and casting the liquid TRU/U metal into an ingot.

Salt recovered from the U and TRU/U processing and the salt distillation operation is treated in the actinide drawdown operation (i.e., electrolysis process¹⁸) to recycle the residual actinides to the electrorefiner via the oxidant production process. The drawdown operations ensure that the concentration of fission products in the overall fuel treatment system remains low enough to allow recovery of uranium and TRU at the desired quality, to maximize recycle of process salt, and to minimize loss of TRU to the waste streams. After the actinide drawdown operation, the actinide-free salt is treated in a follow-on drawdown operation (i.e., electrolysis process) to remove the rare earths, except for samarium and europium that remain in the salt as divalent chlorides.

The salt then moves forward to the salt treatment process where the active metal and divalent rare earth fission products are concentrated by fractional crystallization^{19–21} into a concentrated liquid phase that is combined with the rare earth metal waste collected in the drawdown process to

produce an engineered ceramic waste form. The process also yields an electrolyte salt that can be recycled to the electrolytic reduction or electrorefining operation.

After the drawdown processes, a fraction of the electrolyte salt is not transferred to the salt treatment process. This salt is used in the oxidant production operation to generate the electrotransport species required for electrorefining (e.g., UCl_3) by chlorinating the uranium and TRU elements recovered during the actinide drawdown process plus a quantity of additional uranium metal. This salt maintains the desired concentration of actinide chlorides in the electrorefiner salt.

The output from the above flowsheet consists of uranium and TRU/U ingots, cladding waste, hardware waste, noble metal waste, ceramic waste,²² and off-gas waste. The bulk of the molten salt used in the process is recycled to minimize waste production. Additional lithium chloride can be added as needed to the electrolytic reduction operation to maintain the salt balance in the system. Impurity buildup in the operations is monitored and controlled, through the salt recycle process, to the levels required to meet product and waste specifications.

III. FACILITY DESIGN

The overall site plan is presented in Fig. 3. The site covers about 21 ha (52 acres), and the facilities and

buildings are located inside the secure area but the Processing Facility and Generator Building are located in the inner protected area. The other structures outside the inner protected area include the Protected Area Receiving Building, Waste Storage Facility, Laboratory and Operations Building, Maintenance and Mockup Building, Utility Building, Warehouse and Staging Building, Fire Station, and Utility Electrical Substation. The Emergency Operations Center is located offsite and not shown in Fig. 3.

The Processing Facility shown in Fig. 4 has a complex of primary buildings and ancillary systems that provide the necessary services to receive and store used fuel, operate the process equipment, store waste and products, and ship waste to disposal sites. The fuel is first received in the Protected Area Receiving Building (far right in Fig. 4) where the fuel cask is unloaded from the truck and the cask is lowered to the cask transfer tunnel. At the center of the Processing Facility, four hot cells are arranged as shown in Fig. 5. The hot-cell area is approximately 930 m². Below the operating floor of the hot cells is the service floor (approximately 1200 m² including the TRU vault). The vital area (far left in Fig. 4) is three floors of 1200 m² each and houses the control room, refrigeration system, ventilation system, exhaust filtration system, and other hot-cell support systems.

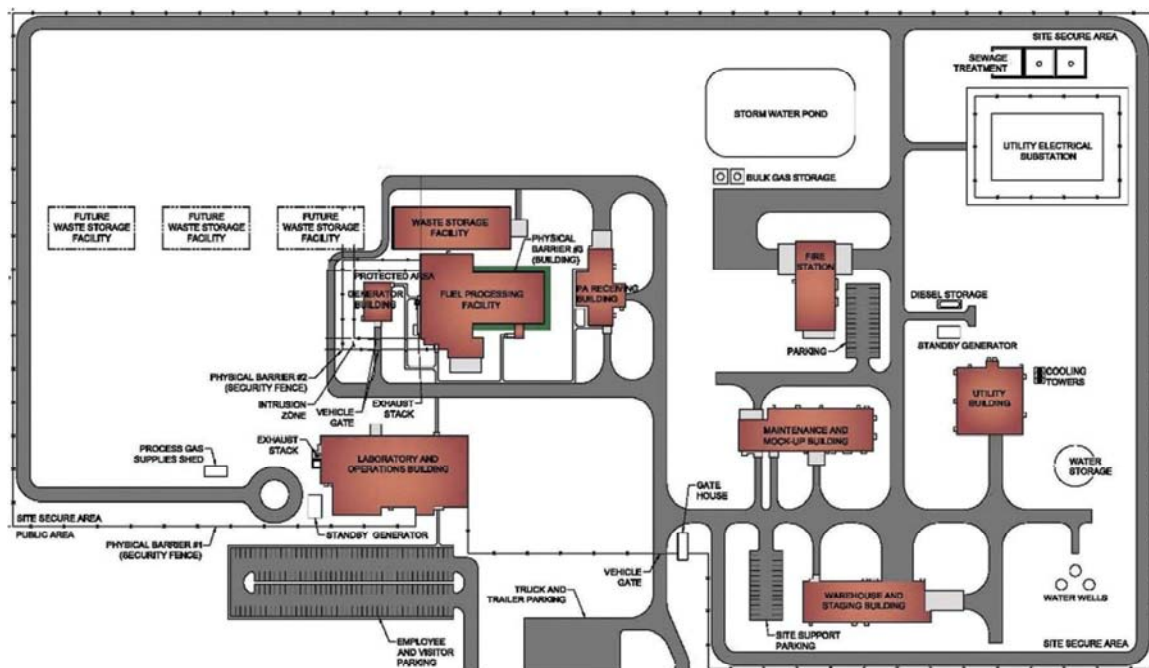


Fig. 3. Site plan.



Fig. 4. Processing Facility.

III.A. Fuel Preparation/Storage Cell

The fuel preparation/storage cell is an air-atmosphere cell that receives the used fuel assembly in a shipping cask. The cell has two large vertical transfer locks, two small horizontal transfer locks, a fuel assembly nondestructive assay station, two fuel rod slitters, and an assembly hardware waste crusher. One vertical equipment transfer lock is used to bring in new equipment and remove equipment for repairs. The other large lock mates with a shipping cask containing up to four fuel assemblies at the hot-cell boundary. A plastic transfer sleeve bagging system, which has been successfully used at the Hot Fuel Examination Facility²³ (HFEF) and FCF, is used to contain radiological contamination so the commercial cask and other parts of the facility can be kept radiologically clean.

III.B. Fuel Processing Cell

The fuel processing cell is a dry argon inert atmosphere cell and contains the majority of the pyrochemical separation processes and the appropriate storage areas between process steps to provide sufficient buffer for efficient operations. The high purity argon is inert to the exposed metal fuel and the lithium chloride, lithium oxide, and other materials in the electrolyte salt, at the same time preventing pyrophoric reactions. The fuel processing cell also contains three specialty work areas: basket loading area, TRU packaging area, and remote repair area. The basket loading area operates at a lower pressure than the rest of the cell to minimize radioactive dust contamination of other parts of the cell. The TRU packaging area operates at a positive pressure relative to the main cell so loose contamination on TRU ingot cans is minimized when they are transferred to the TRU vault. The important cell features include an overhead crane, two overhead robotic cranes, space for in-process storage, and four transfer locks.

III.C. Waste Treatment Cell

The waste treatment cell contains the process equipment to encapsulate the radioactive elements into a stable ceramic waste form for final disposal, plus a waste

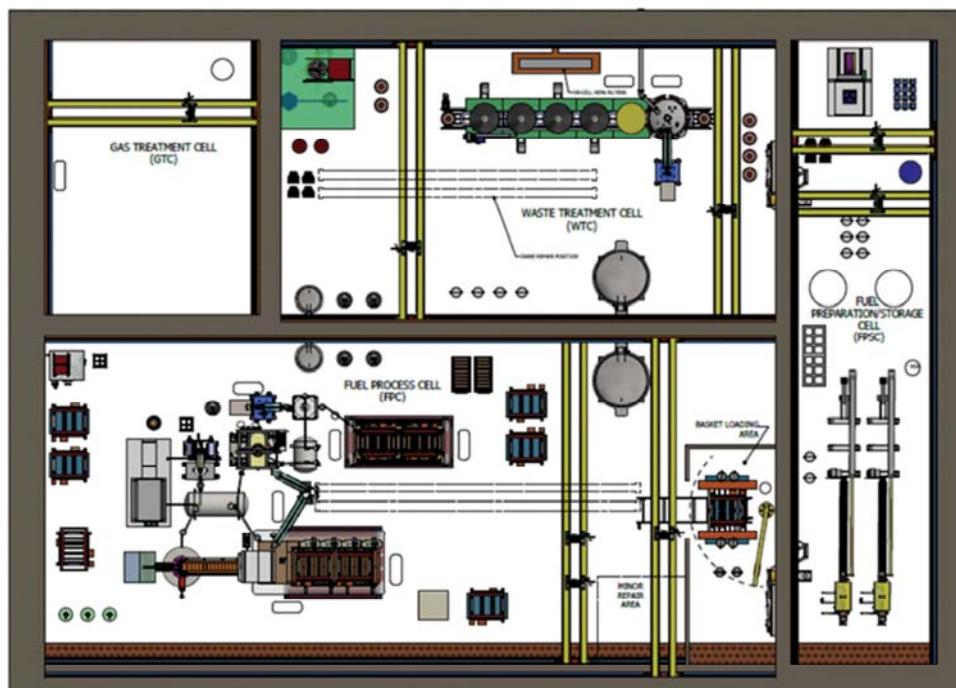


Fig. 5. Operating floor of the four hot cells with process equipment.

packaging area and temporary in-process storage spaces. The primary features include an overhead crane, a robotic crane, and four transfer locks: a horizontal transfer lock from the fuel preparation/storage cell, a vertical lock and tunnel from the fuel processing cell and decontamination cell, a vertical material transfer lock from the material transfer tunnel, and a vertical waste transfer lock to the waste transfer tunnel. This hot cell has a pass-through air atmosphere and several storage areas for waste cans.

III.D. Gas Treatment Cell

The gas treatment cell receives gases directly from the electrolytic reduction vessel and a side stream of the argon gas recirculation loop that maintains the fuel processing cell atmosphere. These gases go through a system that converts oxygen to water and carbon monoxide to carbon dioxide using catalytic beds with the addition of hydrogen and oxygen. After removing the water and carbon dioxide in molecular sieves, the gas stream is sent to the cryogenic system that removes argon, xenon, and krypton. The argon is recycled to the fuel processing cell while the xenon is vented to the facility exhaust stack. The krypton is concentrated in standard pressurized gas bottles that are packaged in waste canisters and transferred to the Waste Storage Facility.

III.E. TRU Vault

The TRU/U metal ingot products have a high security attractiveness level and require reliable heat removal for long-term storage. These products are stored in a special vault located in the lower level of the Processing Facility with a direct transfer lock from the fuel processing cell. The storage cans are lowered through the TRU transfer lock to the material transfer room and placed in a storage container for transfer into the vault.

III.F. Waste Storage Facility

The Waste Storage Facility (Fig. 6) stores high-level waste streams, assembly hardware waste, and radioactive krypton gas. The decay heat is passively removed. This facility initially stores 10 years of waste production and is designed to accommodate expansion with additional modules to hold up to 40 years of waste production. The initial module also provides a transfer path for uranium product and low-level waste boxes that will be temporarily stored in the Warehouse and Staging Building for off-site shipment. The exterior waste

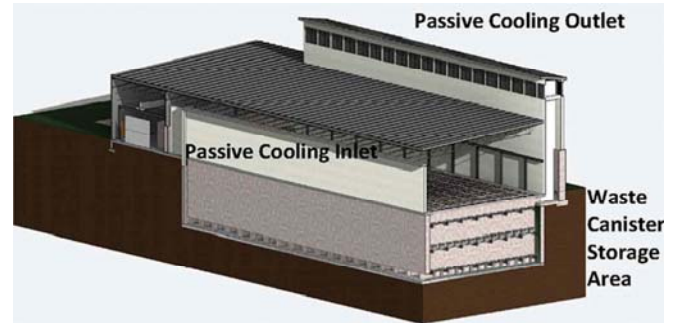


Fig. 6. Waste Storage Facility.

canister has been designed to provide an interface similar to the DOE high-level waste canister. The waste canisters contain either two inner ceramic waste cans, 12 compacted fuel assembly hardware cans, three noble metal waste cans, or two krypton gas cylinders.

IV. PROCESS EQUIPMENT DESIGN

The process equipment layout in the Processing Facility is illustrated in Fig. 5. Process equipment includes the disassembler, fuel rod slitter, basket module loader, electrolytic reducer, electrorefiner, TRU processor, TRU down-blending furnace, uranium processor, drawdown vessel, noble metal processor, salt crystallization vessel, salt storage tank, and ceramic waste processor.

IV.A. Disassembler and Fuel Rod Slitter

The fuel assembly introduced into the fuel preparation/storage cell is nondestructively assayed to establish a shipper/receiver difference and placed into a storage rack, which is passively cooled. From the storage rack, the fuel assembly is moved to a fuel disassembly machine where the assembly is disassembled, removing the fuel rods from the fuel assembly hardware and passing them to the fuel rod slitter. The end caps are mechanically sheared, cladding is removed from each rod, and the oxide fuel meat is collected in a transport can. The cladding slitter design adopted here was developed by the Korea Atomic Energy Research Institute²⁴ and demonstrated in the DUPIC fuel cycle application.²⁵

IV.B. Basket Module Loader

The fuel transport cans are transferred into the fuel processing cell and the basket module loader dispenses fuel powders into four thin rectangular baskets of the basket module, shown in Fig. 7. The basket module

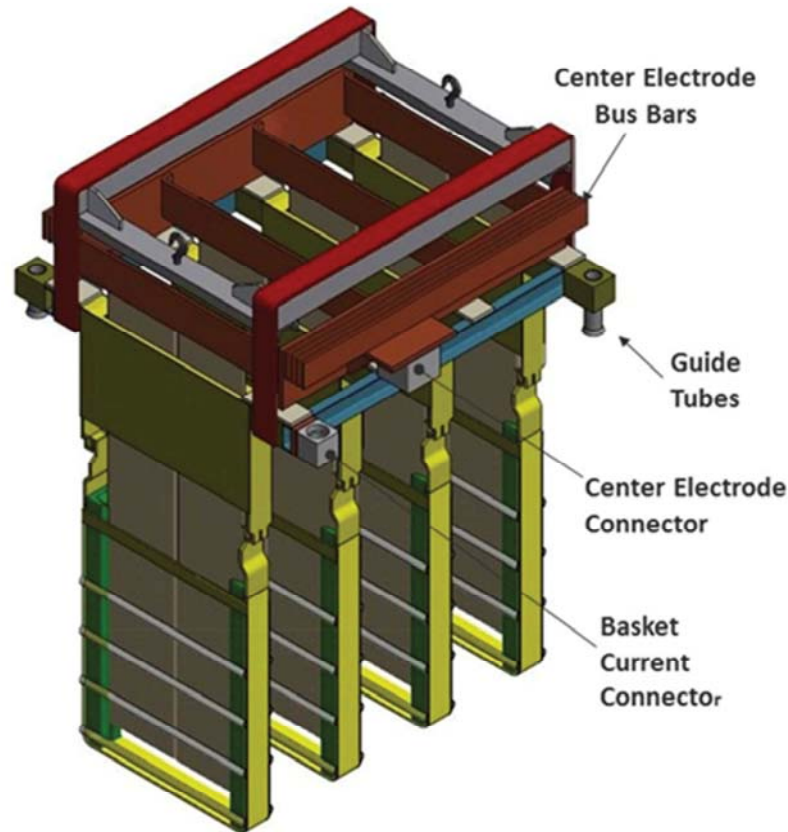


Fig. 7. Basket module.

loader also dispenses the fresh makeup salt in granular form into the basket module.

IV.C. Electrolytic Reducer

The electrolytic reducer consists of an inner crucible in direct contact with the salt and an outer containment vessel, which serves as a guard vessel that will contain the molten salt should any leakage occur from the crucible in an off-normal situation. The latter also serves as the structural support for the crucible, the heaters used to initially bring the salt up to operating temperature, and the components inserted into the crucible. Two basket modules with oxide fuel are loaded into the reducer, which operate as cathodes. Each basket module, shown in Fig. 7, is operated independently. Between the eight rectangular baskets (four per basket module), nine anode assemblies are installed with consumable graphite blocks.

IV.D. Electrorefiner

The electrorefiner, shown in Fig. 8, is designed to be compatible with the basket module from the electrolytic reducer. The basket modules used in the electrolytic

reducer, now containing reduced metals, are directly transferred to the electrorefiner and operate as anodes. Nine cathode assemblies are semipermanent fixtures (i.e., removed only as necessary for maintenance): seven between the eight fuel baskets and one on each end. The uranium product removal system is located inside the crucible at the bottom and the sloped end of the vessel. The metallic uranium dendrites are scraped from the cathode rods by the harvester frames, fall onto the conveyor trays, and are carried across the bottom of the electrorefiner and up the sloped wall. The TRU/U collection system consists of a cabinet assembly semipermanently mounted on the electrorefiner and four TRU/U cathode modules. The cathode module is built around an annular recovery cup that collects the codeposited TRU and uranium metals. The annular design of the TRU/U collection system allows fissile material to be collected while maintaining criticality safety requirements.

IV.E. Uranium Processor

This system accepts the uranium cathode product—metal and adhering salt—delivered by the electrorefiner

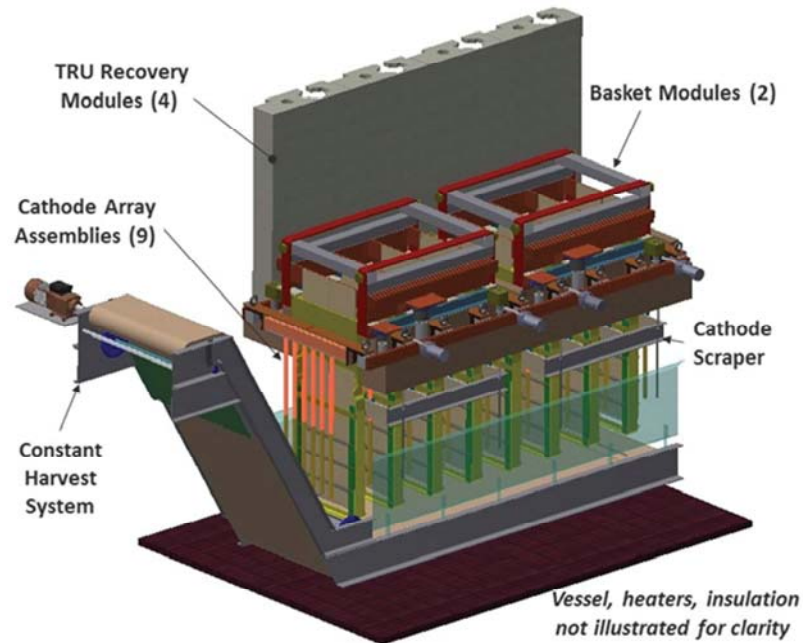


Fig. 8. Electrorefiner.

conveyor. The metal dendrites typically contain about 25 wt% salt when removed from the electrorefiner. Prior to entering the processor, the dendrites are shredded and then fed into the processing chamber where the remaining salt is separated by vaporization. The salt vapor condenses and flows to the salt collection tank where it eventually returns to the process.

IV.F. TRU Processor

The TRU processor accepts the TRU/U cathode cups from the electrorefiner and collects their content for separation of the salt and metal via bottom-pour technique. The recovered salt is transferred to the salt collection tank and the metal is cast into TRU/U ingots for transfer to storage. The apparatus comprises a heated vessel and end cap. The top cover has penetrations for instrumentation, bottom-plug activation, and a heated cup receiver. The bottom of the vessel includes a tapered plug valve, capable of gravity draining the metal and then resealing.

IV.G. Drawdown Vessel

The drawdown vessel combines three operations in one apparatus. The first operation is the actinide drawdown where actinides are removed from the waste salt and stored in situ. Next is the rare earth drawdown operation, where this material is separated from the waste salt and removed from the vessel. After a portion of the

salt is transferred to the waste salt treatment system, uranium is added to the vessel and chlorinated along with the in-vessel actinides into the remaining salt, which is re-introduced to the electrorefiner.

IV.H. Salt Treatment System

When the process has approached steady state, salt in the drawdown vessel, after the actinides and rare earths have been deposited on cathodes, contains other fission product chlorides. A portion of this salt is discharged to the salt treatment system (Fig. 9). This system consists of the salt storage tank, salt crystallization vessel, waste salt transport station, and salt transport tank. Salt from the drawdown vessel is first transferred to the storage tank from where it is transferred to the salt crystallization tank. The salt crystallization vessel separates salt with concentrated fission products from the bulk salt that is returned to the electrolytic reducer or electrorefiner. In the salt transport station, the salt with concentrated fission products is combined with rare earth metals and salt collected on rare earth cathodes from the drawdown operation. The combined materials are transferred to the waste treatment cell for ceramic waste form production.

IV.I. Noble Metal Processor

After electrorefining, the fuel basket module is transported and placed in the basket disassembly rack

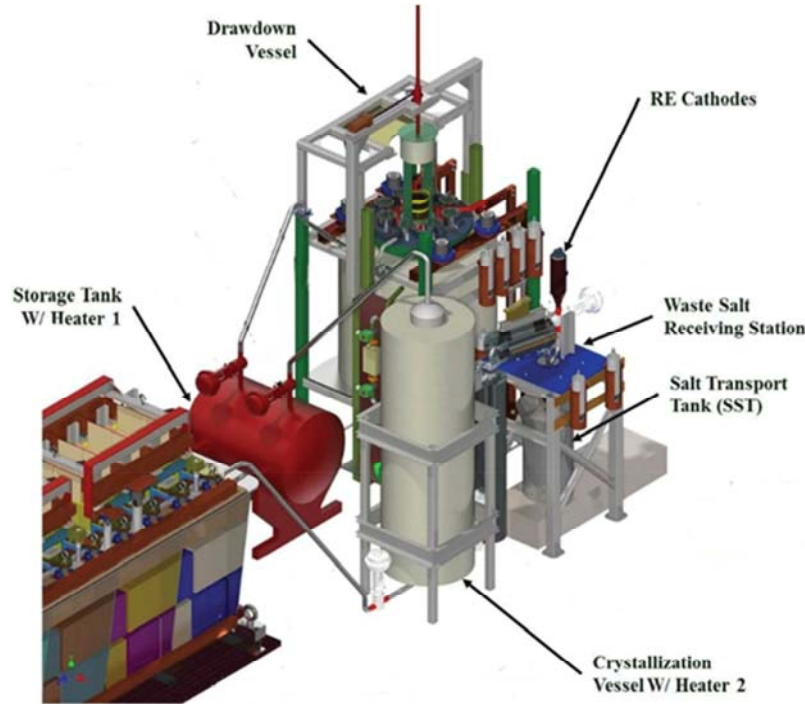


Fig. 9. Salt treatment system.

where the four individual baskets are detached from the module. The detached baskets are placed in the fuel basket inverter to pour residues into the hopper. The inverter and hopper are heated to 650°C to melt the salt. The semifluid residues are fed to a vertical screw feeder that periodically injects them into the iron crucible in the furnace that maintains 1200°C. After salt vaporizes from the noble metals, the furnace temperature rises to 1600°C and the noble waste metal form consolidates.

IV.J. Ceramic Waste Processor

This system located in the waste treatment cell consists of a conveyor, a high temperature furnace, several cooling zones, and a station for feeding cladding shards into an inner waste can as illustrated in Fig. 10. The conveyor carries an empty inner waste can under the ceramic waste blender to be filled with the 500°C zeolite/glass/salt mixture and then transferred to the furnace. The inner waste can is lifted into

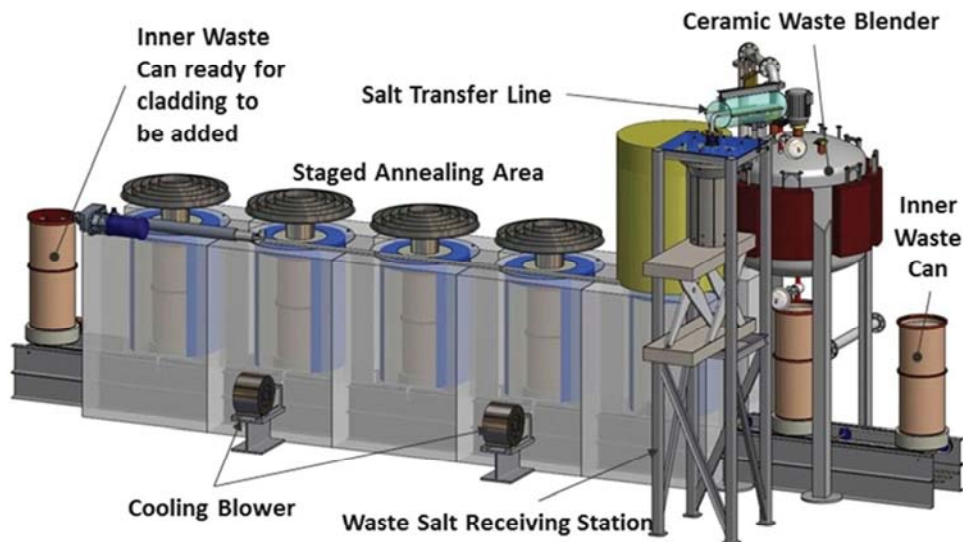


Fig. 10. Waste treatment cell operations to make ceramic waste form.

the furnace where it is heated to at least 900°C and held for about 24 h to convert the zeolite to sodalite, melt the glass encapsulating the sodalite particles, and produce the ceramic waste form. Then the inner waste can is lowered from the furnace onto the conveyor and moved to the cooling zones prior to packaging.

V. OPERATIONS AND MAINTENANCE

Integrated material-handling systems move materials and equipment between process steps or to storage areas. These and other in-cell material-handling systems also install process equipment systems into the hot cells, support maintenance operations on process equipment, and transfer equipment from the hot cells

to the decontamination and hot repair area. The various types of in-cell remote-handling systems are illustrated in Fig. 11. Operations and maintenance in the hot cells are performed using remotely controlled equipment, which dictates that active components or modules have to be remotely repairable in place, remotely removable for repair at a dedicated repair station, remotely removable for remote decontamination followed by hands-on repair, or remotely replaceable. This requirement applies to both process and hot-cell systems, including the remotely operated handling systems (e.g., overhead cranes and electromechanical manipulators). Although the basic overhead handling system design varies for different hot cells, material-handling systems have a modular design with interchangeable parts and remote maintainability.

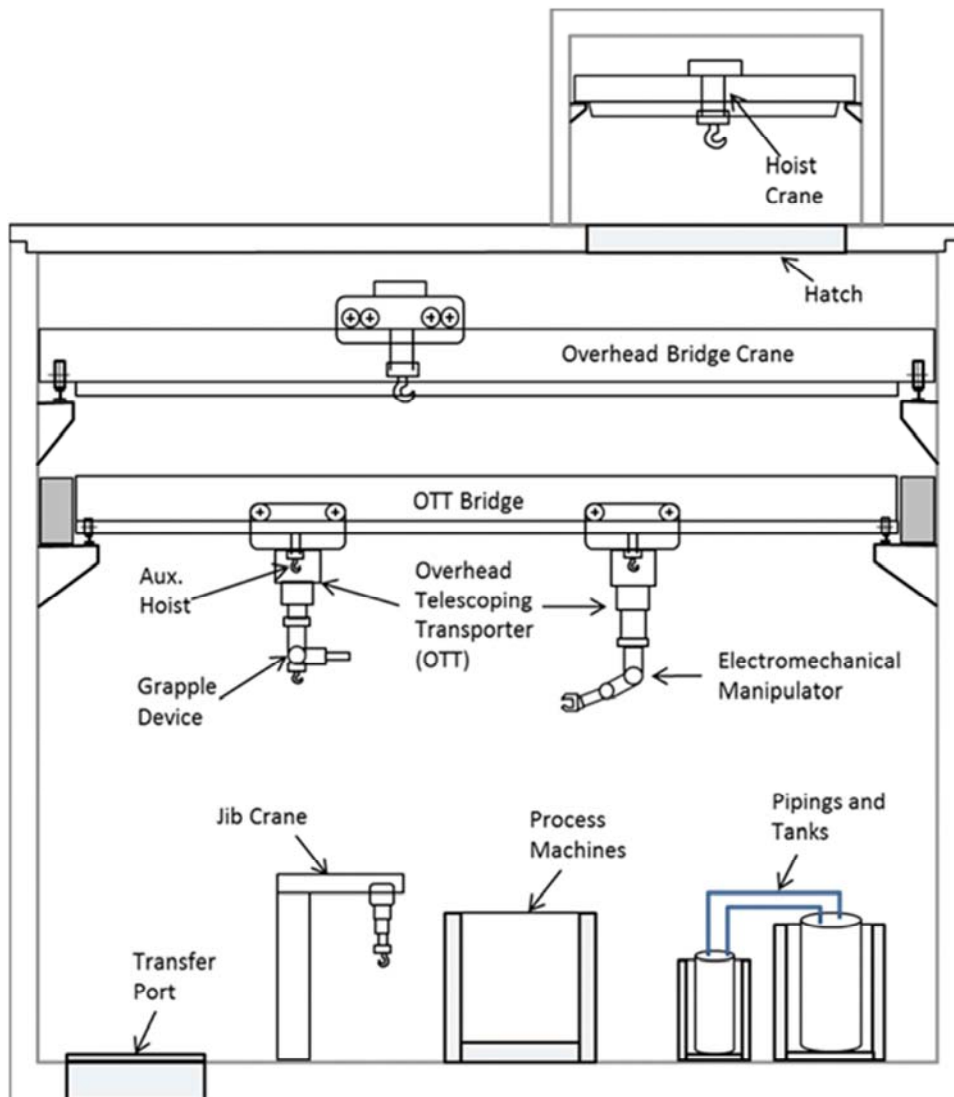


Fig. 11. In-cell remote-handling systems configuration.

The Processing Facility has a central control room used to monitor and control process equipment in the hot cells, facility systems, and material-handling equipment. A distributed control system is used, so data from different systems can be shared where necessary. The overall system is divided into seven areas: operation and control of remotely operated processes, material tracking, in-cell material-handling and robotics control systems, facility monitoring, facility support system monitoring and control, out-of-cell material-handling monitoring and control systems, and security systems. These seven systems are integrated and consist of plant operation computers, programmable logic controllers, user interfaces, and control and operational logic software.

VI. SAFETY ASSESSMENT

VI.A. Safety Approach

There are gaps in existing regulations for a pyroprocessing facility, but existing regulations are a starting point for safety purposes. New U.S. Nuclear Regulatory Commission (NRC) regulations may change specifics of the safety approach and the nature of required documents, but any approach and any regulatory framework requires systematic methods for identifying potential abnormal events and determining their safety significance. Likewise, any approach requires systematic methods for determining the importance of various structures, systems and components (SSCs), and administrative controls for safety purposes.

A full safety analysis was not performed, but selected high-profile scenarios were analyzed to develop an understanding of the primary safety concerns. The safety analysis was based on a risk matrix strategy consistent with Appendix A of NUREG-1520 (Ref. 26). Accident consequences were divided into high consequence, intermediate consequence, and low consequence categories. Accident likelihoods were divided into not unlikely, unlikely, and highly unlikely categories. Each risk index value in the risk matrix is the product of the integer value of the likelihood category and the integer value of the consequence category. Risk indices above 4 are unacceptable.

Sets of accident scenarios were developed for the four hot cells, the service floor, and the TRU vault based on credible accident initiators, preventative design and operational features, and mitigating factors. Unmitigated release doses were computed at various

distances from the facility to determine the dose to the public resulting from various types of limiting accident scenarios and the associated risk index. The set of computed risk indices can be used to select the SSCs and administrative controls that need a higher safety category to satisfy regulatory requirements.

VI.B. Metal Fire in Fuel Processing Cell

The most severe accident scenario in the fuel processing cell is the breach of the cell boundary, leading to loss of inert argon atmosphere and ingress of air, possibly causing a metal fire. The total mass of potentially pyrophoric material in the fuel processing cell at any time is approximately 1000 to 2000 kg. If the atmospheric integrity in the fuel processing cell is lost, air will enter the cell. If the atmospheric oxygen content exceeds the 4% ignition threshold, pyrophoric materials oxidize rapidly, releasing heat and radioactive particles into the cell atmosphere. The heat raises the cell pressure, which may force radioactive particles into the ventilation system or into adjacent areas.

To evaluate the consequences of loss of the fuel processing cell atmospheric integrity, computational tools developed to analyze cell breach scenarios at FCF and HFEF were obtained from Idaho National Laboratory.²⁷ These tools were modified to evaluate the current design of the fuel processing cell. Various breach sizes were assumed, ranging from 1.9 to 213 cm in diameter. During each scenario, it was assumed that power was lost to the equipment and ventilation system. The only method of control over the cell atmospheric temperature and pressure was through the cell pressure relief system.

An example of cell temperature results from one analyzed scenario is presented in Fig. 12. In this scenario, the passive pressure relief system maintains a slightly negative relative (to adjacent cells) cell pressure allowing air ingress into the cell. This condition results in an increase in the oxygen content in the cell, which exceeds a value of 4% in approximately 125 min. At this point, the metal-oxygen reaction occurs, which rapidly increases the relative cell pressure and temperature. The metal inventory of the cell is consumed over a 30-min time period. After the metal-oxygen reaction ends, the relative cell pressure/temperature rapidly decreases due to the flow of cooler air into the cell through the breach. Shortly after this negative relative pressure spike, the cell temperatures and relative pressure level off to a constant value. During the transient, the maximum temperatures of the cell atmosphere, steel liner, and

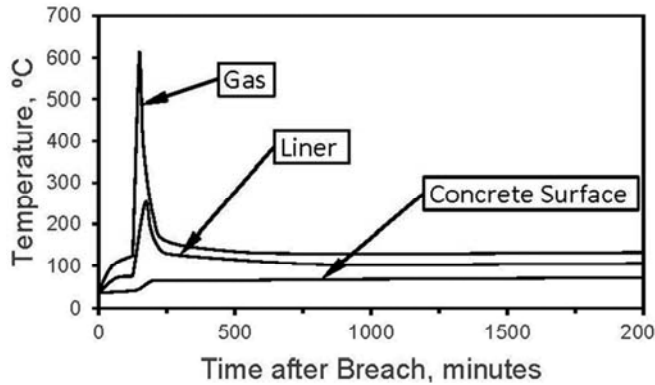


Fig. 12. Cell temperatures during and after the metal fire.

concrete wall surface are approximately 620°C, 250°C, and 60°C, respectively. Based on the preliminary analyses, a safety-grade cell exhaust system would be required to minimize unfiltered release to the atmosphere during these postulated scenarios.

VI.C. Active and Passive Cooling in the TRU Vault

During storage in the TRU vault, the TRU/U ingots generate heat from radioactive decay that must be removed in a controlled manner to ensure safe storage conditions. During normal operation, an active cooling system removes heat generated by the ingots and maintains air temperatures in the 21°C to 38°C range. During certain accident scenarios, the active system is nonfunctional or unavailable. In these scenarios, the heat generated by the TRU/U ingots would be passively removed via natural convection and rejected to the environment.

Active cooling of the TRU vault is maintained by a combination of two refrigeration units and three blowers. Each refrigeration unit can handle the entire heat load of the TRU vault, and only two of the three blowers are required to provide the necessary air flow rate. Each blower provides a cooling air flow rate of 900 m³/min at 13°C, and each refrigeration unit can remove up to 600 kW of heat load.

The current design of the TRU vault (at the rated capacity of 430 kW) contains 1865 storage modules in vertical stacks with each stack consisting of 5 storage modules (373 total stacks). Each storage module has a heat load of 230 W. At rated flow (1800 m³/min with two of the three blowers operating) and assuming the flow is distributed equally to each stack, the velocity of air through a single stack would be 3.7 m/s with an outlet

air temperature of 25°C. The heat transfer analysis for the active cooling scenario uses these values.

Heat can also be removed from the TRU vault via a passive heat removal system, which includes a system of chimneys, headers, ductwork, and dampers that provide a flow path for air from outside the facility to the TRU vault before being exhausted back outside the facility. The flow path of the air includes the storage module stacks in the TRU vault. The passive air flow rate through the storage module stacks is calculated by solving the one-dimensional Bernoulli equation.

The results for both the active and passive cooling scenarios are provided in Table I. As expected, due to the significantly higher cooling air velocity and lower inlet air temperature of the active cooling scenario, the steady-state storage can and ingot temperatures are significantly lower than in the passive cooling scenario (147°C versus 415°C and 320°C versus 496°C, respectively). The higher temperatures of the passive cooling scenario are still below thresholds where eutectic formation between the TRU/U ingot and the storage can become a concern (approximately 700°C). The temperature results provided in Table I are maximum values calculated at the top of the storage module stack.

VI.D. Criticality Safety

Used LWR fuel has a very low effective ²³⁵U enrichment and reactivity worth. In the absence of significant internal moderation, a critical configuration with the used LWR fuel processed in the facility for any process step prior to electrorefining is not possible due to the composition of the used LWR fuel and the nature of process steps prior to electrorefining. This result was confirmed by inspection of general criticality safety references such as TID-7016 (Ref. 28), by specific criticality calculations for relevant process steps and

TABLE I

Steady-State Results for Active and Passive Cooling Analyses

Parameter	Active Cooling	Passive Cooling
Air inlet temperature	13°C	20°C
Air outlet temperature	25°C	67°C
Maximum storage can temperature	147°C	415°C
Ingot surface temperature	313°C	489°C
Maximum ingot temperature	320°C	496°C

configurations, and by calculations for steel-reflected spheres of UO_2 with masses of 10 000 kg for ^{235}U enrichment of 5% and 4000 kg for ^{235}U enrichment of 10%. For any step prior to electrorefining, strict control of moderators provides sufficient criticality control for effective ^{235}U enrichments less than 5% in the used LWR fuel.

Electrorefining separates high reactivity worth TRU elements from uranium, which has low reactivity worth. Since the process salts are known to be hygroscopic, one possible upset event is the loss of the dry, inert atmosphere, as salts absorb water from a humid atmosphere (deliquescence). The limiting criticality safety configurations for the electrorefiner result from deliquescence and precipitation, which may also be a consequence of deliquescence. Criticality calculations show the electrorefiner remains subcritical following credible deliquescence and precipitation events. The uranium product has very low reactivity worth, and the uranium product ingots cannot be arranged in a critical configuration under any credible circumstances. Criticality calculations were performed for the TRU processor, down-blending furnace, and TRU product transfer process. These operations remain subcritical for credible abnormal events. The TRU vault was analyzed for various accident scenarios including flooding. Calculations show the vault remains subcritical for all accident scenarios provided active and passive cooling are adequate to maintain confinement of the TRU/U product ingots in the storage cans. Criticality safety constrains the designs of the electrorefiner, TRU processor, down-blending furnace, and all operations with consolidated TRU product.

VII. SAFEGUARDS ASSESSMENT

Sandia National Laboratories (SNL) modified a safeguards model for pyroprocessing to reflect the new process flowsheet.^{29,30} SNL and Argonne personnel used this new safeguards model to evaluate the process, and adopted the use of nondestructive analysis rather than the full suite of traditional sampling and destructive analysis for the facility. Similar to other facilities, this conceptual design incorporates nondestructive analysis and a full analytical laboratory for destructive analysis (and process operations). The conceptual design safeguards plan recognizes that any nondestructive analysis techniques would have to be shown to give comparable and reliable results, especially for formal material balances as those determined by destructive analysis. Locations, conceptual designs, and costs for nondestructive methods are incorporated into the facility design because adequate shielding, facility space, and

remote-operations capability are important to identify early in the design process since they are difficult to add in later design and construction phases. Many improvements are still necessary in nondestructive equipment; however, enough different systems have been proposed for the conceptual layout to provide for both passive and active interrogation equipment.

The in-process inventory is dominated by the inventory in the electrorefiner, and different measurement methods and their limitations were evaluated. For the other pieces of equipment, the special nuclear material inventory is minimized and does not influence the material balance. Specific equipment configurations for formal material balances and necessary steps to resume processing operations were evaluated, such as adjusting equipment sizes so they hold integer quantities of fuel assemblies. For example, the oxide transport can and basket modules could be modified to hold the entire contents of one pressurized water reactor assembly or two boiling water reactor assemblies. Also, the TRU/U cathodes could be adjusted in size so two cathode cups would contain the TRU product from one assembly, and possibly, one cathode cup would produce one TRU/U ingot. These changes would help reduce the uncertainties that are found during a formal material balance and help to assign any process equipment hold-up to the appropriate piece of equipment.

Although NRC regulations would need to be rewritten for reprocessing, SNL considered 10CFR Part 74 (Ref. 31) which specifically excludes reprocessing but can be used for general terms and expectations for smaller throughput facilities. The requirement for a shutdown and flush-out plus strict inventory difference goals and detection requirements would be unrealistic and likely difficult to achieve. These challenges support the need to rewrite the regulations for a large throughput facility. International Atomic Energy Agency (IAEA) requirements are written for large reprocessing facilities and provide a better basis to design and evaluate the Pyroprocessing Facility proposed safeguards approach. The standard error of the inventory difference should be at or below 2.42 kg plutonium. The IAEA goal of expected accountancy capability should be 1% throughput. The differences in regulatory requirements are summarized in Table II.

VIII. SECURITY ASSESSMENT

Since security systems are an important part of the overall facility costs and impacts, SNL evaluated the conceptual design from the security standpoint and made the following recommendations:

TABLE II
Comparison of Existing Regulatory Requirements

Description	NRC	IAEA
Standard error of inventory difference	<0.1% active inventory and plant flush-out required	<1.0% mass throughput and 2.4 kg plutonium per balance period
Timeliness detection goal for abrupt loss	2 kg plutonium within 3 days	8 kg nonpure plutonium < 3 months or 8 kg pure plutonium < 1 month
Physical inventory	Every 6 months	Once per year

1. Minimize the footprint of the inner protected area to minimize locations to respond to and the amount of real estate (doors, surfaces, etc.) that must have sensors and assessment equipment. It is especially important to minimize the length of the perimeter intrusion detection and assessment system (PIDAS) since it is difficult and expensive to install, operate, and maintain.

2. Minimize entry control points and locate them inside the area that they protect so that they are less vulnerable to threats.

3. Locate the central alarm station (CAS) and secondary alarm station (SAS) inside the area that they protect.

4. Provide emergency electrical power to the CAS and SAS which are an important part of the physical security system’s ability to respond to an attack by the design basis threat (DBT).

5. Protect the response force from a pre-emptive attack by the DBT.

The elimination of the primary armed responders at the start of their attack is an advantageous tactic. The ready responders should be located as close as practical to the target material (inside the Processing Facility) and it is important to protect them from an initial attack.

Sandia National Laboratory’s approach takes advantage of the delay and protection built into the very robust construction of the Processing Facility and Waste Storage Facility. Siting both underground is a significant security advantage. The very thick concrete walls around the hot cells and the nearly self-protecting nature of the fuel processing cell’s argon atmosphere, high temperature, and limited accessibility support this approach. These characteristics suggest an approach to replace the PIDAS around the entire area with intrusion detection on the Processing Facility exterior walls.

The evaluation of an existing or proposed physical security system requires a methodical approach to measure

the ability of the security system to meet defined protection objectives. Without this kind of careful assessment, valuable resources might be wasted on unnecessary protection or, worse yet, fail to provide adequate protection of materials against a theft attack by the defined threat. The vulnerability assessment methodology evaluates performance-based physical security concepts at nuclear sites and facilities.

Virtual simulation software models were used for the facility and its physical security system under adversary attack. The model uses artificial intelligence along with detailed information about a facility’s buildings, utilities, and landscape; physical security system performance characteristics; and tactics and weapons for both the adversary and response force. For the parameters modeled, the security system simulations showed the response force protected the targeted material 314 of 314 runs. The security system contains adequate detection and delay and features a sizable onsite response force to provide interruption and neutralization.

The most significant facility change from the security assessment minimizes the inner protected area by moving the Protected Area Receiving Building and Waste Storage Facility outside of the inner protected area. The Generator Building is also a candidate to move outside of the inner protected area. This change will be determined as additional safety/security assessments are completed. The analyses should specify the emergency power safety function and length of time that the facility can be operated without emergency power.

IX. CAPITAL AND OPERATING COST ESTIMATES

The facility capital cost estimate by Merrick & Company is based on the floor plans, roof plans, sections, and elevations for all buildings plus the SSC list for the primary support systems. Detailed measurements of quantities of materials are used, where possible. The detailed estimates are divided into the following categories for each building: foundations, vertical

structure, floor and roof structures, exterior cladding, roofing, interior construction, conveying, mechanical systems, electrical systems, and equipment. Each of these categories is divided into elements where the estimates are based on square footage, cubic yards, or linear feet with applicable costs for these units of measure.

The ten different categories sum to give direct building costs, and 10% general conditions and construction management costs are added to the direct costs. In addition, a contingency is added to this total based on level of design maturity and type of construction. For standard buildings with equivalent industrial examples, the contingency is 15% whereas the site security system (20%) and Processing Facility (25%) contingencies reflect their complex design requirements and specialized systems.

The costs for the process equipment and support system as well as the operating cost are estimated by the Argonne

design team. Costs for each equipment system or component (if not a part of a specific system) cover four general categories: engineering design and analysis, fabrication, purchased parts, and qualification. Qualification consists of three distinct phases: Phase 1—Fit-up, integration, and outfitting, Phase 2—Out-of-cell qualification including remote operations, and Phase 3—Final in-cell installation and remote qualification. The same methodology applies to the process support systems. However, the overhead lifting and robotics cost estimates use vendor quotations.

The project total capital cost estimate is summarized in Table III. The cost estimate is in 2015 dollars and is “overnight cost” with no escalations. The contingency factors range from 10% to 25% depending on the building type and the maturity of technology and design. The land acquisition cost, if needed, is not included. The annual operating cost is summarized in Table IV.

TABLE III
Project Total Capital Cost*

Category	Cost (\$K)	Contingency Factor (%)	Cost with Contingency
Facility			
Processing Facility	84 543	25	105 679
Protected Area Receiving Building	3 982	15	4 580
Waste Storage Facility	7 957	15	9 150
Laboratory and Operations Building	19 468	15	22 388
Maintenance and Mockup Building	9 815	15	11 288
Utility Building	13 585	15	15 623
Warehouse and Staging Building	4 302	15	4 948
Generator Building	2 776	15	3 192
Emergency Operations Center	6 775	15	7 791
Fire Station	3 438	15	3 954
Site Work	29 593	15	34 032
Site Physical Protection System	25 100	20	30 120
Design Engineering	43 253	20	51 904
Subtotal	254 587		304 649
Process equipment and support			
Process equipment	21 905	25	27 381
Transfer locks and feed-throughs	10 109	25	12 636
Overhead lifting and robotics	26 599	10	29 259
Electrical systems	2 251	25	2 814
Monitoring and control systems	4 480	25	5 600
Materials control and accountancy	1 030	25	1 288
Safeguards and security equipment	4 645	25	5 806
Analytical laboratory equipment	3 800	25	4 750
Start-up materials	2 859		3 431
Subtotal	77 678		92 965
Total	332 265		397 614

*In thousands of dollars.

TABLE IV
Annual Operating Cost*

Category	Cost (\$K)
Staffing	41 790
Materials and services	4 180
Process chemicals	579
Spare parts	120
Product and waste containers	5 472
Utilities	984
Total	53 125

*In thousands of dollars.

X. POTENTIAL FOR SCALING UP TO COMMERCIAL-SCALE FACILITY

In order to quantify the economies of scale potential, the project has evaluated a potential approach for scaling the current pilot-scale design up to a 400 T/year facility. A similar approach would apply to scale up to other throughput rates as needed.

In this example, for the main processing equipment, namely, the electrorefiner, electrolytic reducer, and some supporting equipment, such as the TRU processor, drawdown vessel, etc., constrained by the criticality safety or materials control and accountancy consideration, the equipment unit size is kept at the original size and the number of units is duplicated four times. For the front-end operations in the fuel preparation/storage cell, the equipment has flexibility in utilization, and hence, the fuel disassembly station, fuel rod slitter, and hardware waste packaging are replicated three times. Other equipment that can be scaled up easily is duplicated twice. This category includes the basket module loader, salt tank, noble metal processor, salt crystallization vessel, waste salt transfer system, and waste packaging.

Having decided on the number of equipment systems, various ways of configuring the processing cells were evaluated. The layout in Fig. 13 shows the most promising approach. This layout option has another advantage in that further scale-up can be achieved by simply adding additional north-south-oriented processing lines, both in the fuel processing cell and the waste treatment cell. As compared to the 100 T/year facility,

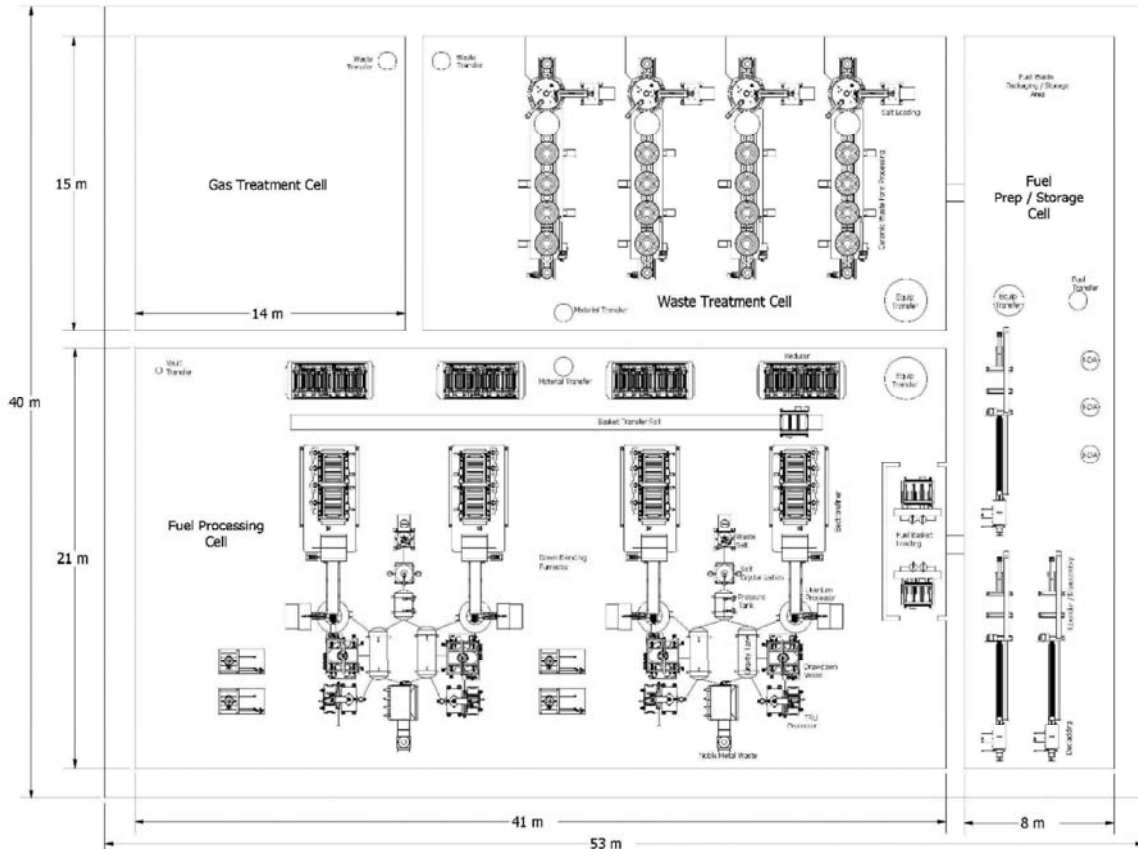


Fig. 13. Layout of pyroprocessing cell for a 400 T/year facility.

the hot-cell floor area is 1.8 times larger, hence significant economies of scale are achieved and further economies of scale are possible as scale-up continues.

The resulting project total capital cost with contingencies is \$911 million and the annual operating cost is \$90 million for the 400 T/year facility. The basis for this cost estimate is for a first-of-a-kind and not a follow-on to the 100 T/year pilot-scale facility. In the latter case, the cost would be further reduced without the first-of-a-kind costs.

XI. IMPLICATIONS ON USED FUEL MANAGEMENT

Ultimately the TRU products recovered in pyroprocessing must be transmuted through fission in a reactor to provide a solution to used fuel management. Neutron capture in the TRU elements without fission results only in their evolution to other TRU elements of higher mass, which are generally more radioactive. To fission TRU elements effectively, high-energy neutrons are needed. There is a tremendous difference between the transmutation reactions possible from the low-energy neutrons of thermal spectrum and the high energies of fast spectrum. The transmutation probability and the percentage of neutrons absorbed that cause fission of typical thermal and fast spectra for the TRU isotopes are compared in Table V (Ref. 2).

In a thermal spectrum only a limited number of isotopes fission effectively. If fuel is recycled continuously, higher TRU isotopes will continue to build up until they approximately equal the amount of plutonium in the fuel. In a fast spectrum all the isotopes fission substantially and the

equilibrium composition is reached with relatively small, quite normal, amounts of higher TRU isotopes. Therefore, the TRU products from pyroprocessing are suitable only for fast reactors, particularly metal-fueled SFRs.³²

XII. SUMMARY

The conceptual design of a pilot-scale (100 T/year) pyroprocessing facility for treatment of LWR used fuel and high-level waste production incorporates an advanced process flowsheet designed for commercial scale-up. The detailed engineering design of the process equipment systems and the hot-cell facility is based on lessons learned from past operations. The current design only requires one train of the main process equipment, such as electrolytic reducer, electrorefiner, salt treatment system, and ceramic waste processor, to achieve the design throughput of 100 T/year. The resulting hot-cell facility containing the equipment is very compact, only about 930 m². The safety assessment identified the breach of the argon atmosphere hot cell, leading to air ingress and metal fire, as the most severe accident scenario. Even if the metal fire consumed the entire inventory of metal in the cell, the maximum cell temperatures reach manageable levels, and the safety-grade cell exhaust system can minimize unfiltered radioactivity release to the atmosphere. The safeguards and security assessments showed the design can meet the applicable regulations.

The project total capital cost estimate for the 100 T/year facility is \$324 million without contingency and \$398 million with contingency factors ranging from 10% to 25% depending on technology maturity. The annual operating cost is estimated at \$53 million/year. For a 400 T/year commercial-scale facility, the capital cost is estimated at \$911 million with contingency factors, and the annual operating cost at \$90 million/year.

We should hasten to point out that the TRU products from pyroprocessing are not suitable for recycle to LWRs. The TRU products are suitable only as fuel for fast reactors, particularly metal-fueled SFRs. Nevertheless, a near-term deployment of a pilot-scale pyroprocessing facility will demonstrate a technological solution to the legacy LWR used fuel management, at the same time bridging to longer-term complete fuel cycle closure based on a fast reactor economy.

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TABLE V
Transmutation Probabilities*

Isotope	Thermal Spectrum (%)	Fast Spectrum (%)
²³⁷ Np	3	27
²³⁸ Pu	7	70
²³⁹ Pu	63	85
²⁴⁰ Pu	1	55
²⁴¹ Pu	75	87
²⁴² Pu	1	53
²⁴¹ Am	1	21
^{242m} Am	75	94
²⁴³ Am	1	23
²⁴² Cm	1	10
²⁴³ Cm	78	94
²⁴⁴ Cm	4	33

*From Ref. 2.

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