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Semiannual

Report for March-December 1991

Volume 1, Number 1

Defense-Related Technologies

Transition from the SIS Program to the APT Program

LLNL Plutonium Processing Technologies for Complex 21

Automated Glovebox Processing

Process Modeling, Flowsheet Simulation and Analysis

The APT Semiannual Report

is published twice each year by the Advanced Processing Technology Program at the Lawrence Livermore National Laboratory. The journal reports on current applied research and development activities within the APT Program. The APT Program develops advanced material processing technologies and equipment in the areas of environmental restoration, waste management, and plutonium processing.

Questions and comments relating to the technical content of this journal should be addressed to the APT Program Office, Lawrence Livermore National Laboratory, P.O. Box 808, L-436, Livermore, CA 94551. (510) 423-6629.

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Advanced PROCESSING Technology

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On the Cover

Inside an atmosphere-controlled glovebox, an automated IBM gantry robot lowers a crucible into the advanced pyrochemical furnace (APF). APT's automated glovebox processing technology is faster, safer, better, and more cost-effective than manual methods. See "Automated Glovebox Processing" on page 16. (Cover photo: James E. Stoots Jr.)

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Foreword

With this first issue, the Advanced Processing Technology (APT) Program at Lawrence Livermore National Laboratory (LLNL) launches a new publication, the *Advanced Processing Technology (APT) Program Semiannual Report*. Our goal for this *Report* is to showcase our applied research and development activities. Our work concentrates on two major areas: (1) environmental restoration and waste management; and (2) plutonium processing technology for the reconfigured Nuclear Weapons Complex, Complex 21—including waste minimization and the treatment of plutonium-containing, transuranic (TRU), low-level (LLW), and mixed wastes.

The *APT Semiannual Report* will highlight APT technologies that optimize solutions to problems of health and safety, waste and plutonium processing, waste characterization, and waste minimization within the Department of Energy (DOE) complex and industrywide. Each issue of the APT Report will focus on a different segment of our development activities. The focus of our first issue is on defense-related technologies. In subsequent issues, we will describe activities in the area of environmental restoration and waste management.

For our colleagues at LLNL, those in the DOE community, and key players in the waste treatment industry, we believe the results reported in this publication will not only provide information, but encourage new partnerships. Several of the APT technology development programs are multidisciplinary team efforts with industrial partners that focus on transferring advanced processing technologies into the U.S. marketplace. APT technologies, outlined in this new publication, will continue to provide many opportunities for government and industry to work together to strengthen our position in a competitive world marketplace.

Martyn Adamson
Scientific Editor

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Overview of the APT Program

LLNL's Advanced Processing Technology (APT) Program develops innovative and revolutionary advanced material processing technologies and equipment in support of DOE programs in two major areas:

- *Environmental Technologies*—waste detection, characterization, and restoration technologies needed for the environmental cleanup of the DOE complex. APT's environmental program researches, develops, demonstrates, tests, and evaluates innovative technologies for reducing the costs and increasing the effectiveness of environmental restoration and waste management. The main areas of research and development are risk assessment and characterization, environmental restoration, and waste treatment and minimization. Future issues of the *APT Report* will highlight environmental technologies.
- *Defense-Related Technologies*—advanced technologies for the planned reconfiguration of the nuclear weapons production complex, Complex 21. The APT staff is contributing its expertise in this field to the DOE Technology Assessment and Selection Panel (TASP), a panel of representatives of all major DOE laboratories and production sites who are evaluating and selecting the best technologies for Complex 21.

The theme of this issue, APT defense-related technologies (see box) focus on molten salt and aqueous chemical processing, equipment development, plutonium waste processing, process modeling, mechanization, automation and robotics. Key defense-related process and equipment technologies include:

- *Incinerator-Alternative Technologies* replace nonpermissible incineration as a process for treating or destroying hazardous and mixed wastes. (Mixed wastes are radioactive wastes that also contain chemically toxic materials.) Incinerator-alternative technologies under development will avoid the potential release of hazardous chemicals into the environment and the generation of corrosive gases (e.g., HCl and SO₂) and other toxic gases released by incinerators. Process technologies being developed include mediated electrochemical oxidation (MEO), molten salt destruction (MSD), and photolytic destruction.
- *Robotic Handling Systems* reduce worker exposure to both radiation and hazardous materials, and minimize the waste streams of used gloves, wipes, etc., generated by “hands-on” glovebox operations. An extremely reliable robotic system, developed by IBM, has been adapted at LLNL to replace manual hazardous materials processing and treatment operations.
- *Plutonium Processing Technologies* being developed by APT meet modern DOE requirements for safety, waste minimization, reduction of worker radiation exposure, and reliability, availability, and maintainability (RAM) of equipment. These process and equipment technologies, such as the hydride/dehydride pyrochemical process, the tilt-pour furnace, and MEO, will be used in the ongoing cleanup of existing sites and incorporated into Complex 21. APT technologies will maximize the flexibility of Complex 21 while reducing its physical size and infrastructure requirements.

- *Systems Analysis and Integration* APT is developing flowsheet modeling and systems analysis tools to assess process technologies for use in integrated systems. Factors of health and safety, residue and waste generation, waste minimization, and waste processing will have major impacts on future processing and manufacturing technology developments. Modeling and systems analysis tools are being used to assist the TASP Plutonium Working groups in their task of evaluating candidate technologies and process flowsheet options.

APT Defense-Related Programs

Complex 21 Technologies

- *Plutonium Processing Technology Development*
 - Electrochemical Dissolution and Residue Recovery
 - Electrochemical Processing of Organic Waste
 - Mixed Waste Processing
 - Photolytic Processing
 - Molten Salt Processing
 - Evaporative Purification
 - Liquid-Liquid Separation Processing
 - Tilt-Pour Furnace Development
 - Plutonium Purification by Hydride Chlorination
 - Direct Chloride Reduction
 - Chloride Volatility
 - Pressure-Cycled Calcination
 - Reusable Crucibles
 - Oxide Reduction Salt Regeneration
 - Scrubber Development
 - Recycle of Electrorefining Salt
 - Bottom-Pour Casting
 - Continuous Oxide Reduction
 - Electrowinning
 - Hydride/Dehydride
 - Flowsheet Development and Systems Analysis

- Automated Weapons Assembly
- Calcining/Dust Control
- Automated Waste Sorting
- Leaching and Dissolver Development
- Material Accountability Technology
- Weapons Assembly
- Bagless Transfer

- *Supporting Research*
 - Catalyzed Plutonium Dissolution
 - Chloride Volatility
 - Continuous Oxide Reduction
 - Evaporative Purification
 - Intelligent Robotics

Process Development for Rocky Flats Plant

- Alternate Alloy Salt Scrub
- Dicesiumhexachloro-plutonate Reduction
- Incinerator Off-Gas Treatment Technology
- Electrochemical Processing of Organic Waste
- Photolytic Processing of Organic Waste

Booth R. Myers

*Deputy Associate Director (Acting)
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Close Out of the SIS Program

The Special Isotope Separation (SIS) Program was completed in September 1991. The APT Program is working to transfer SIS technology to Complex 21.

The goals of the Special Isotope Separation (SIS) Program were to develop and demonstrate the technology of atomic vapor laser isotope separation (AVLIS) for transforming fuel-grade plutonium into weapon-grade material, and to identify and develop the chemical processing technologies necessary to support the AVLIS process.

The AVLIS process (Fig. 1) is a cost-effective method of providing special materials, such as plutonium, with individual isotopic tailoring. The vapor stream is produced by electron beam evaporation of the feed metal. Copper laser-pumped dye lasers generate the proper frequencies to photoionize selected isotopes from the vapor stream. The ionized species are then extracted by an electric field, collected, and processed or stored. The enriched vapor stream condenses on the product collector and is carried to further processing.

The Laboratory has been working for a decade to develop the plant prototypes of the isotope separation and chemical processing equipment. The SIS program planned to integrate major subsystems such as the Laser Demonstration Facility (LDF), the Engineering Demonstration System (EDS), and the Materials Processing Laboratory (MPL) facilities in order to verify process performance for isotope photoionization and separation. Single and multiple EDS separator unit enrichment demonstrations were conducted with run durations ranging from approximately 10 h to approximately 100 h. Activities included the preparation of feed for the AVLIS process, separator handling, isotope separation, product/by-product processing, recycle of residues, and waste processing. These activities represented the end-to-end processing of plutonium from oxide purification and metal conversion to product collection as a metal, by-product stabilization as oxide, and packaging and certification of waste.

In early January 1989, the Department of Energy (DOE) selected the Idaho National Engineering Laboratory as the site for the proposed SIS plant. This plant was designed to derive weapon-grade material from DOE-owned reactor-fuel-grade plutonium and to recover plutonium from existing residues. Had the plant been built, it would have provided a contingent supply of weapon-grade plutonium, thereby reducing dependence on DOE's reactors and permitting flexibility in response to changing plutonium supply requirements.

In January 1990, the Secretary of Energy announced the cancellation of the proposed SIS plant, given existing budget problems and funding priorities within the DOE. With emphasis on the importance of developing a thorough understanding of the SIS technology for national security and other purposes, he proposed to complete research and development experiments for SIS—except for plutonium testing in the EDS—and to end the research program in an orderly fashion.

E. Ng

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M. DeMicco

Figure 1. Essential elements of the AVLIS process for separating isotopes. A copper vapor laser excites tunable dye lasers, providing selected frequencies to photoionize the by-products of the plutonium vapor stream. The by-products are separated by an electric field while the enriched stream condenses on the product collector.

Figure 2. LLNL Integrated Plutonium Processing Facility

Integrated Laser Operation

At LLNL, demonstration of integrated laser operation was performed in tests using the LDF and the EDS. Laser light from the LDF was sent 2 km to the Plutonium Facility (Fig. 2) through an underground optical system.

The laser hardware in LDF (Fig. 3) comprises five subsystems: the copper laser system, the dye laser system, the auxiliary laser system, the beam combination and optical transport system, and the instrumentation and control system. Highly efficient and tunable to precise frequencies, dye lasers generate the multiple process wavelengths. Copper lasers convert electrical energy into fixed-wavelength green and yellow light beams, which then excite (pump) the dye lasers. Dye lasers and copper lasers are both arranged in chains. The first laser (called an oscillator) in each chain produces a laser signal with relatively low power but with controlled spatial, temporal, and spectral characteristics. These low-power

signals are then boosted to high average power by a series of laser amplifiers, which constitute the rest of the chain.

By improving copper laser performance and by constructing a second corridor of copper lasers [each corridor contains six master oscillator/power amplifier (MOPA) chains], current average performance of the copper lasers is 85% of SIS production plant requirements per corridor. Individual MOPA chains have demonstrated power levels greater than 100% of plant requirements. The dye laser systems have also been upgraded to provide the entire suite of colors necessary to fully demonstrate plutonium AVLIS.

EDS

The EDS, a plutonium-capable separator, was designed to assess the full-scale separator run cycle for the SIS Plant. This run cycle included the enrichment performance of the AVLIS process at production scale, and post-run processing and handling of the separator components. The EDS (Fig. 4) is a plant prototype,

Figure 3. Laser Demonstration Facility (LDF). LDF provided the required laser light to selectively ionize the vapor stream.

designed to gather important plant performance data related to the isotope-separation process and to the reliability, availability, and maintainability (RAM) of the integrated system. In accordance with DOE policy concerning radiological exposure, important design features were incorporated into the EDS to reduce worker exposure.

The EDS was not approved for full plutonium operations, but extensive full-scale and fully functional testing of the equipment has been accomplished. Nearly 1000 unit-hours of vaporization of plutonium-surrogate (nonradioactive) materials, and approximately 500 unit-hours of photoionization and isotopic enrichment, at and above production requirements, were completed. Particularly significant is the long duration, multichamber, integrated enrichment demonstration accomplished in November 1989. In this demonstration, the separator and laser systems were operated at full capacity for more than 100 hours, validating the performance of the equipment at full scale. Good agreement was achieved between the measured change in isotopic composition and the projected performance. In addition to demonstrating the integration of the laser and separator systems and verifying the physics models, these experiments tested many of the operating and maintenance procedures important for efficient and safe operations with plutonium.

Figure 4. Engineering Demonstration System (EDS), a plutonium-capable separator, demonstrated full-scale and fully functional plutonium plant equipment testing.

MPL

The Materials Processing Laboratory (MPL), shown in Fig. 5, was designed to assess full-scale pyrochemical processing for the SIS plant. This included conversion of oxide feed to metal, removal of daughter product ^{241}Am , casting the required shapes to feed the AVLIS separators, recovering the product by hydride/dehydride operations, purifying the recovered weapon-grade metal by electrorefining, recovering the ^{238}Pu – ^{240}Pu by-product as oxide, and recovering, purifying, and recycling metal residues generated throughout the SIS plant. The sequencing of these processes is illustrated in Fig. 6, a simplified SIS plutonium-processing flow diagram.

The MPL is a plant-prototype design. Its full-scale equipment gathers plant performance data related to the chemical processing necessary for the SIS plant and to the RAM of the integrated system. Significant full-scale equipment testing included the hydride/dehydride unit operation for recovery of plutonium from the 1984 AVLIS Demonstration Runs, the hydride/dehydride chlorination unit operation for preparation of PuCl_3 for use in molten salt extraction, and electrorefining.

PPVS

During 1990, LLNL completed an extensive series of Plant-Performance Verification Series (PPVS) isotopic-tailoring demonstrations, which provided significant verification of the SIS plant design. The SIS completion plan governed work through September 1991. Key activities were the completion of PPVS, including documentation of the results; laser, separator, and plutonium process development to close out the SIS technical basis; extensive cold testing of the tilt-pour furnace; cold development and demonstration of the mediated electrochemical oxidation (MEO) system; and dose-reduction evaluations, including initial tests of the handling system.

Process Demonstrations

During the final year of the development program, several key demonstrations were completed and documents describing the performance and equipment were prepared and

Figure 5. Materials Processing Laboratory (MPL). The pyrochemical processing of plutonium is conducted in inert atmosphere gloveboxes.

archived. The PPVS multi-isotope integration tests were completed with surrogates, showing that the integrated system performed as predicted. The separator development activities that formed the technical basis of the plant separator were documented and archived. The process laser development and operation activities were completed and documented. Significant demonstrations verified the readiness to deploy plant-scale pyrochemical processing equipment as part of the balance-of-plant processes. The pyrochemical processes for plutonium processing were demonstrated in the MPL. Also demonstrated and documented was the residue and recycle process using MEO.

Documentation

The SIS records close-out process involved an intensive effort by program personnel over a period of weeks. An integral part of the close out of the SIS Program, the documentation process involved collecting, cataloging, and storing

Figure 6. SIS process flow diagram indicating existing NWC production process technologies.

program records for future retrieval. To manage this activity, a records close-out procedure was written. Personnel participating in the records close-out process were formally trained. Documents were collected into record packages consisting of a Records Close-Out Traveler, Document Close-Out Inventory Sheet(s), and any Incomplete/Interim Document Status Reports. Records packages were then reviewed for completeness by the SIS Document Control Center and Quality Assurance (QA). Records packages were duplicated and stored.

A total of 3,292 documents are separately listed in the SIS Project computerized data base, exclusive of records contained in the EDS QA Records File. The EDS QA records contain specifications, as-built drawings, purchase orders, design analyses, reports, procedures, test records, and safety and engineering notes.

The final report on plutonium process development and operation was completed. This report formed the technical basis of the balance-of-plant processes. The final physics point

design and process flowsheets were updated to supply the integrated basis for plant criteria and requirements. Significant headway was made in validating the technical basis for predicting a tenfold reduction in plant worker exposure through remote operation of key, high-exposure glovebox processes. All development activities, along with their operation procedures, are documented in reports including the *Process Laser System*, *Separator Technical Data and Design Package*, *Separator Performance Report*, *Tilt-Pour Furnace Activation and Operation*, *Pyrochemical Processing Technical Data Package*, *Pyrochemical Performance Report*, *Aqueous Chemical Processing Technical Data Package*, and the *Plant Performance Verification Reports*.

From SIS to APT

In response to the National Defense Authorization Act for FY 1988/89, the Secretary of Energy was directed to develop an integrated plan to reconfigure the Nuclear Weapons

Complex (NWC). Underlying the reconfiguration of the NWC is the assumption that nuclear deterrence will remain a principal element in the security of the United States for the foreseeable future. The primary objectives of the reconfigured complex—Complex 21—are that it will:

- Support the U.S. nuclear deterrence stockpile objectives set by the President and funded by Congress.
- Be smaller, less diverse, and less costly to operate than the current NWC.
- Be constructed and operated to comply with all applicable federal, state, and local laws and regulations.

To meet the objectives of Complex 21, LLNL organized the Advanced Processing Technology (APT) Program in June 1991 from elements of the canceled SIS development program.

Acknowledgments

We acknowledge the accomplishments of the hundreds of LLNL contributors whose efforts and energies over the 12-year life of the SIS Program developed and demonstrated AVLIS as a cost-effective and low-risk solution to isotopic separation of actinides. Key SIS partners included Bechtel, WINCO and MK-Fergusson, whose teams admirably carried out the architect engineer, integrator, and construction manager roles. Major industrial contributors to the development program included SAIC, IBM Federal Sectors Division, KMI, Schott, HP, and Martin Marietta. Idaho National Engineering Laboratory also contributed to the effort.

Plutonium Processing Technology for Complex 21

For Complex 21, the APT technology focus includes chemical processing, equipment development, process modeling, and mechanization, automation and robotics.

To select and develop technologies for the reconfigured Nuclear Weapons Complex (NWC)—Complex 21—the Department of Energy (DOE) Weapons Complex Reconfiguration Office established a Technology Assessment and Selection Panel (TASP) to develop and implement a Technology Assessment and Development Plan (TADP). The primary objective of TASP is to ensure that technologies, equipment, and processes used in Complex 21 represent the best achievable balance among cost, performance, schedule, and risk.

Because the TASP deals with the entire NWC manufacturing operations, working teams were established to deal with each of the following manufacturing areas:

- Plutonium manufacturing operations.
- Uranium manufacturing operations.
- Tritium operations.
- Salt manufacturing operations.
- Assembly and high-explosives operations.
- Non-nuclear manufacturing operations.

The Advanced Processing Technology Program (APT) is a key player in the plutonium working team (PuWT) and its subgroups. The mission of the PuWT is to develop and implement a TADP specifically for the plutonium manufacturing operations, including site-return disassembly, metal purification, plutonium recovery, and waste management. The PuWT and its six processing working groups have defined two flowsheets:

- The reference flowsheet, based on the wrought manufacturing process used at Rocky Flats during the 1980s.
- The baseline flowsheet, based on technology projected to be ready for production operations by 1993.

In addition, APT and other programs have proposed a number of alternative primary technologies, which could have even greater

impact on waste generation and dose reduction than the baseline technologies. As these technologies mature, many will be inserted into the baseline flowsheet.

To achieve the best balance among cost, performance, schedule, and risk, the PuWT has determined that both processes and equipment must be developed that enhance worker safety, minimize operator dose—without transferring the dose to maintenance personnel, maximize plutonium utilization, minimize waste at the point of generation, and provide for built-in recycle of residues, reagents, and solvents. To comply with present and anticipated federal, state, and local regulations, a number of changes are needed in plutonium-processing operations, chemistry, and equipment (Table 1).

The APT Program, capitalizing on the research, development, and engineering work already done by the Special Isotope Separation (SIS) Program, is developing plutonium processing technologies to meet the recommendations of the PuWT for Complex 21. Many of the unit operations being developed under the SIS program have been included in the TADP baseline flowsheet; the remainder are included by the PuWT as alternatives.

APT Plutonium Processing Technologies

The APT technology focus includes chemical processing, equipment development, process modeling, and the mechanization, automation, and robotics required to achieve:

- Compliance with laws, regulations, orders, and accepted practices regarding environmental, safety, and health protection of workers and the public
- Waste minimization at the point of generation.

L. W. Gray

- As-low-as-reasonably-achievable (ALARA) radiation dose.
- Improved safeguards and nuclear materials accountability.
- Reduced operating cost.
- Improved reliability, maintainability, operability, and availability.
- Waste treatment to assure minimum volume/mass and certifiable packaging.

The following sections describe a number of processing projects underway at LLNL.

Site-Return Processing

Metal Purification

The baseline flowsheet, defined by the PuWT and described in the TADP, assumes mechanical disassembly of the site-return components—nuclear weapon components that have been

returned from the deployment site for recycling or decommissioning. Plutonium is removed from these components by a number of separate operations (Fig. 1). Once plutonium components have been separated from other materials, americium and plutonium are separated from one another by use of a molten salt extraction (MSE) unit operation. If plutonium requires further purification, it is electrorefined (ER) using a molten CaCl_2 solvent and PuCl_3 as the seed salt. The purified plutonium metal is transferred to the pit-fabrication area. Residues requiring chemical processing for recovery and purification include spent chloride salts, ceramic crucibles, electrorefining anode heels, and glovebox floor sweepings. These residues are transferred to chloride-recovery operations for dissolution and purification. Waste materials go to the waste operations and management area.

Table 1. Comparison of current NWC plutonium operations, chemistry, and equipment with those needed for Complex 21

Present	Complex 21
Operations	
Labor intensive, hands on	Automated, mechanically assisted
High-maintenance equipment	Simplified equipment, fewer moving parts
High intrinsic waste generation (e.g., gloves, wipes)	More efficient processes that eliminate multiple recycling, decrease glove box requirements, and lower intrinsic waste generation per unit throughput
Inadequately designed processes	
Aging, obsolete equipment	Modern equipment with low corrosion, low maintenance requirements, and reduced operator attendance
Chemistry	
Solids-producing reagents	Gas-producing reagents, reagents that can be regenerated <i>in situ</i>
Single-pass reagents and solvents	Regenerable/recyclable reagents and solvents
Side reactions poorly understood	Chemistry understood, parasitic-reaction chemistry controlled
Equipment	
Batch operations, long cycle times	Multiple-unit operations per workstation
Single-use items	Multiple-use items
Labor-intensive accountability, hand breakdown and cleaning	On-line accountability (via nondestructive analysis), equipment designed to satisfy safeguards and security requirements
Poor corrosion resistance	Process chemicals and equipment matched to each other to reduce or eliminate corrosion

Molten Salt Chemistries

Molten salt chemistries are the key to high-density, rapid plutonium throughput and low waste generation. For these molten salt processes, APT is developing two new, advanced pyrochemical-reaction furnaces (APFs). One of these is based on the standard stationary furnace, modified with thermal valves that allow the transfer, on demand, of either molten salt or molten metal; the other furnace is based on the tilt-pour concept (Fig. 2). Both of these furnaces use thermal-shock-resistant metal crucibles and computer-controlled process automation. Both furnaces can be serviced by hand or by robotics.

In traditional, stationary furnace operations, the products of the reactions solidify in place. The solidified products are then manually removed from the furnace and broken out of the crucible. In the breakout process, the crucible itself is destroyed and the operator is exposed to a high radiation dose. APFs, which allow movement of molten metal, offer several advantages over stationary furnaces in a plutonium processing plant. First, the movement of molten metal eliminates the manual breakout step, thereby eliminating both the broken ceramic crucible waste stream and operator exposure. Computer modeling suggests that APFs will reduce operator radiation dose by up to 50% per unit throughput when compared to a standard stationary furnace in the PuWT reference and baseline flowsheets. The tilt-pour furnace also has a more rapid cycle time (2 h for MSE vs. 8–16 h for the standard stationary furnace; 24 h for ER vs. 120 h for the standard stationary furnace). Several third-generation prototype furnaces are currently operational at LLNL, with more than 400 cycles accumulated on one of the machines.

Purification of plutonium metal can be accomplished by a combination of molten salt extraction and ER. Both of these unit operations require a salt oxidant to oxidize either the americium or plutonium metal. As the best oxidant for these two unit operations is PuCl_3 , the flowsheet must provide for its preparation. The baseline flowsheet calls for the preparation of plutonium hydride followed by metathesis with HCl gas to prepare PuCl_3 . The PuCl_3 is then used as seed salt for electrorefining and as the ^{241}Am oxidant for MSE. Spent salts and ER

Figure 1. A simplified Complex 21 Pit Manufacturing Flowsheet showing the processing technologies under development at LLNL.

anode heels are transferred to aqueous chloride recovery to reclaim and purify the plutonium. (Each of these unit operations was part of the SIS reference flowsheet.)

APT is developing three alternative approaches for processing the metal purification residues. One method is to strip spent CaCl_2 salts with calcium metal, eliminating the aqueous dissolution of CaCl_2 salts. Another method is to hydride spent anode heels and metathesize this product by reaction with HCl , to PuCl_3 . Volatile chlorides (e.g., GaCl_3 , FeCl_3) are vaporized from the PuCl_3 , leaving behind a sufficiently pure product to be used as either a seed salt for ER or as an oxidant for MSE. This eliminates the need to dissolve spent anode heels in aqueous chloride recovery (see discussion of the chloride recovery below). The third approach for purification of the anode heels is to evaporate the impurities (Ga, Fe, etc.) from the anode heel using electron-beam evaporation.

Figure 2. LLNL's tilt-pour furnace for molten salt processing of plutonium eliminates the broken ceramic crucible waste stream and reduces operator radiation dose by up to 50%.

Manufacturing

According to the baseline flowsheet, purified plutonium metal is blended and cast into near-net-shape parts. After heat treatment for phase stabilization, parts are machined to final dimensions using dry machining under a high-purity argon atmosphere. Parts are inspected and assembled with non-Special Nuclear Materials (SNM) materials into the final pit. Residues are generated in the form of casting sprues, casting oxide, and machining chips. Many of the metal forms can be recycled directly to a casting furnace within the foundry; others are oxidized to PuO_2 . Oxide residues are transferred to aqueous nitrate recovery for dissolution, purification, and recycle.

The use of precision casting instead of near-net-shape casting is being studied by the Defense Systems Program. Computer modeling, conducted jointly by Defense Systems, APT, and A-Division at Los Alamos National Laboratory (LANL) suggests a possible waste-savings of up to 80% compared to the reference process (wrought processing). A large portion of this saving is the result of fewer residues being generated that require recycling through the aqueous nitrate recovery processes.

Aqueous Nitrate Recovery

All nonchloride-contaminated process residues and analytical samples are transferred to aqueous nitrate recovery for processing. First, residues are pretreated as necessary (e.g., organic destruction, passivation, size or mass reduction) prior to being either leached or dissolved in HNO_3 . Anion exchange is used to purify the solution, followed by oxalate precipitation and calcination to the oxide. PuO_2 is transferred to the chloride recovery operation where it is converted to metal by direct oxide reduction (DOR) using calcium metal as the reductant and CaCl_2 as the solvent molten salt. Solutions from both anion exchange and precipitation are fed to an evaporator-acid recycle system to oxidize residual hydroxylamine to N_2 and H_2O , convert residual $\text{C}_2\text{O}_4^{2-}$ ions to CO_2 and H_2O , purify recovered HNO_3 for recycle, and convert excess HNO_3 to environmentally benign N_2 gas. Residual salts are transferred to the waste-processing area for treatment and disposal.

The baseline flowsheet lists mass reduction of combustible materials in nitrate head-end operations as a potential show stopper. At present, permissible methods to oxidize combustible materials in plutonium-contaminated residues do not exist. Also, baseline methods for both leaching and dissolution use glass-pipe cascade dissolvers with a mixture of HNO_3 and HF (added as CaF_2) as the dissolution media. This method of PuO_2 dissolution typically dissolves from 10–95% of the plutonium in the feed, depending upon the feed stock, and a portion of the glass dissolver. As a rule, the residues produced are not discardable to waste. The baseline

flowsheet includes a pot-type dissolver at the end of the cascade dissolver chain to repeatedly leach residual PuO_2 from these residues.

APT is developing alternative processes in both areas. For mass reduction of combustible materials, APT is developing a molten salt oxidation (MSO) technique. This method uses a Na_2CO_3 -based melt for both mass and volume reduction without the use of an open flame. For leaching and dissolution, APT is engineering a mediated electrochemical oxidation (MEO) technique to totally dissolve the PuO_2 in one pass. Using high-efficiency dissolution avoids the high amount of waste produced and the operator radiation dose incurred from multiple leaching operations.

A single vessel/single stage unit operation, MSO was originally developed by Rockwell International as part of a coal-gasification project. APT is exploring a two vessel/two stage approach (Fig. 3). Stage one operates under

oxygen-deficient (pyrolysis) conditions; stage two completes the oxidation of the evolved gases. Wastes, together with oxidant gas such as air, O_2 , or a mixture of O_2 and CO_2 , are injected into a bed of molten salt (sodium, potassium, and lithium carbonates) that has a melting point of approximately 580°C . Organic constituents (cellulose, plastic, rubber, etc.) are decomposed by pyrolysis. Heteroatoms, such as chlorine and sulfur, are metathesized into stable salts (e.g., NaCl and Na_2SO_4), and are retained by the melt. Ash, including the actinides in the waste, is also retained within the first stage melt as a result of both wetting and partial dissolution. Therefore, first stage salts must be replaced, either continuously or in batches. Spent salt is contacted with H_2O to dissolve the soluble salts, chiefly NaCl and residual carbonates. Fractional crystallization allows the carbonates to be recovered and recycled. Other salts are discharged as low-level waste. The overall effect is about the same in

Figure 3.
A two-vessel/two-stage
Molten Salt Oxidation
(MSO) operation.

volume and mass reduction as incineration. The insoluble ash, containing all the actinides, can then be treated by MEO to dissolve the actinides.

Residue leaching and dissolution operations are crucial to high utilization of plutonium. APT is developing an MEO system for use on high-value feed stocks. The heart of this system is an electrolytic cell (Fig. 4) that continuously regenerates a strong oxidizing, mediating charge-transfer ion [e.g., Ag(II), Co(III), Ce(IV)]. Mediated electrochemical dissolution offers a number of important advantages over the standard cascade dissolvers. Depending on scrap type, these advantages include a two- to tenfold increase in dissolution rate, an increase in dissolution completeness (greater than 95–99.9% for MEO, and much less than 40% to approximately 90% for standard HNO_3 -HF dissolution), and the elimination of secondary residues that require vault storage and reprocessing. Computer modeling has indicated that implementation of MEO will result in up to a twofold reduction in operator radiation exposure and up to a fourfold reduction in waste. The waste savings is highly dependent on the feedstock—the more difficult to dissolve by HNO_3 -HF, the greater the waste savings by MEO.

Chloride Recovery

In the baseline flowsheet, spent MSE and ER salt residues, spent anode heels, and other chloride-contaminated residues (e.g. crucible shards) will be dissolved in HCl. Iron-contaminated materials are first partially purified by sorbing FeCl_4^- using anion exchange resin. Otherwise, iron would follow plutonium in chloride solvent-extraction systems. Solutions containing large amounts of ^{241}Am are fed to one solvent-extraction system, other solutions are fed to a second solvent-extraction system. In the solvent-extraction separation of plutonium and americium, calcium follows the americium through to precipitation and calcination. As a result, the americium oxide product contains approximately 70% CaO, making it very difficult to purify should a need for this product arise. After being separated in this way, plutonium and americium are separately precipitated as oxalates, filtered, and calcined to their respective oxides. Oxides from both

nitrate and chloride recovery are then converted to plutonium metal, using calcium metal as the reductant and molten CaCl_2 as the solvent.

Use of chloride volatility (discussed above under “Site-Return Processing, Metal Purification”) would have major benefits in the chloride recovery area. Using chloride volatility to remove iron and gallium from the spent anode heels would eliminate the need for an anion exchange step in the chloride-recovery system, by eliminating this feed stock from the aqueous chloride recovery system.

The process of stripping the actinides from spent MSE and ER CaCl_2 salts would avoid the need to dissolve the bulk of the spent CaCl_2 salt. The two main advantages of this process are (1) the spent salts could be converted back to required feed reagents (calcium metal and Cl_2 gas) using an electrowinning process that APT is developing; and (2) the excess calcium would not follow americium through to precipitation and calcination. As a result, the americium product would be in a form very easy to purify.

Incorporating these technologies into the baseline flowsheet could remove up to 90% of the anticipated feed stock for the chloride recovery line. Eliminating one of the solvent extraction lines could cut the volume of highly corrosive chloride waste by as much as 90%.

Waste Management

At the site of generation, waste is sorted into characteristic types: transuranic (TRU), low-level (LLW), hazardous, mixed (hazardous plus radioactive), and sanitary. Classified shapes are smashed, or otherwise declassified, and solids are segregated into separate streams for processing. Solids are treated to ensure that they are in compliance with Environmental Protection Agency (EPA) and Resource Conservation and Recovery Act (RCRA) requirements. The baseline flowsheet assumes incineration for combustible solids and passivation for reactive metals. Resultant waste products are immobilized by cementation and packaged to satisfy Department of Transportation (DOT) requirements prior to disposal at LLW or TRU waste disposal sites (assumed to be the Nevada Test Site and the Waste Isolation Pilot Plant, respectively). The baseline flowsheet also assumes

that, whenever possible, liquid organic wastes are separated and destroyed by incineration along with solid organic wastes. Acids are neutralized, evaporated to near dryness, and finally immobilized by cementation to generate EPA- and RCRA-compliant forms.

Mass reduction of combustible materials in waste management operations is recognized as a critical point in the baseline flowsheet. At present, permissible methods are not available for oxidizing alpha-contaminated combustible materials. However, APT is developing processes for nitrate recovery, MSO, and MEO, which are applicable to waste treatment. Also, MSO and MEO are likely to meet with public acceptance. With MSO, the size of the units may be different for waste treatment, but the principle is the same. One advantage of MSO is that nonincinerable items, such as the lead-lined rubber gloves used in the glovebox, can be decomposed, with the lead being converted to lead metal instead of PbO_2 —an EPA-listed item. Decomposition of plutonium-contaminated rubber gloves with an incinerator is impractical due to the amount of soot produced.

The MEO system, shown in Fig. 5, which converts organic materials to CO_2 and H_2O , can be used for the destruction of hazardous organic materials such as energetic materials (e.g., propellants and high explosives), chemical pesticides, chemical warfare agents, and infectious biological materials (e.g., tissue or blood samples). MEO can be used to convert TRU and LL mixed waste to TRU and LL waste, and possibly convert TRU waste to LLW. MEO can also be used to decontaminate to LLW levels many TRU items (e.g., plastics or metals) that are resistant to direct oxidation or dissolution. Combustible materials not dissolved by MEO (e.g., chlorinated plastics) could then be fed to MSO for volume/mass reduction.

Robotics and Automation

The key to operator dose reduction is to reduce the time an operator is exposed to the radiation. Developing robust equipment that requires little maintenance reduces the exposure of maintenance personnel. Development of automation techniques reduces operator radiation dose by removing the operators' hands from the gloveboxes. The robotics and automation effort within the APT program is detailed in the "Automated Glovebox Processing" article in this report.

Conclusion

Most of the plutonium processing technologies and related hardware originally developed for the SIS Program can be applied directly to Complex 21; others require only minor modification. Development of both the technology and the hardware is well-advanced, and demonstrations of hardware reliability and durability are underway for some of the critical equipment. The projected advantages of these processes over both the reference flowsheet and the baseline flowsheets include:

- Elimination of second-generation process residues requiring storage and reprocessing.
- Reduction of process waste by up to 90% for the reference flowsheet and 50% for the baseline flowsheet.
- Reduction of cycle times, ranging from 10% to factors of ten or more.
- Decrease in the plutonium going to waste by up to 95% for the reference flowsheet and up to 80% for the baseline flowsheet.
- Reduction in operator exposure by approximately 50–80%.

Incorporating these processing technologies into Complex 21 will enhance the probability that we can design and operate the new plant in full compliance with all applicable laws and regulations.

Figure 5. A prototype MEO organic destruction system, incorporating a commercial ICI electrochemical cell, is shown in the photograph (above) and the schematic (below).

Automated Glovebox Processing

APT's glovebox automation activities focus on robotics technology and automation of the Advanced Pyrochemical Furnace (APF). Development efforts concentrate on integrating manual, remotely controlled, and automated processes to minimize operator radiation exposure and operator-associated wastes.

S. A. Couture
M.C. Bronson

The Advanced Processing Technology (APT) Program has demonstrated state-of-the-art automated systems for servicing process equipment in a glovebox environment for remote chemical and metallurgical processing of plutonium and other hazardous materials. Because of LLNL's role in developing automated glovebox technology and the potential for applying this technology in other areas, LLNL was selected as the lead laboratory by the DOE Office of Technology Development's Robotics Waste Minimization Program. In its leadership role, LLNL has responsibility for coordinating efforts aimed at using automation to reduce waste generation in the Nuclear Weapons Complex (NWC) laboratories and production facilities. The APT program, an outgrowth of the Special Isotope Separation (SIS) Program, is a sign of DOE's commitment to applying technologies developed for the SIS Program to a broad range of reconfigured NWC (Complex 21) needs.

Minimization of both operator radiation exposure and operator-associated waste, which accounts for approximately 70% of the waste generated in existing plutonium processing facilities, is the primary objective of automated glovebox material handling. DOE-sponsored robotics activities emphasize practical solutions that allow remediation to be accomplished faster, safer, better, and cheaper than using manual methods. To accomplish these objectives, technology development concentrates on glovebox robotics. These technologies focus on high-priority applications that the NWC must demonstrate prior to final design and implementation decisions. The following sections provide a snapshot of APT glovebox automation activities

including glovebox robotics technology, supporting technology developments, and automation of the Advanced Pyrochemical Furnace (APF).

Glovebox Robotics

Glovebox robotics activities focus on improvements to the IBM Electric Cantilevered Robot (ECR). Because of its excellent record of reliability, the IBM ECR has been used extensively in industry for high-speed assembly applications. In a team effort, LLNL, IBM, SAIC, and other participants adapted it for use as an integral part of the glovebox automation project. Now dubbed the IBM Glovebox Robot Gantry (GR-G), it was first installed in February 1991 in the Automated Testbed at LLNL.

The Automated Testbed (ATB), shown in Fig. 1, is an atmosphere-controlled glovebox. The primary mission of the ATB is surrogate testing of automated plutonium processing systems and equipment, including the robot. Operation with surrogates allows continual process improvement and refinement without the attendant problems of operator radiation exposure, radioactive waste, and plutonium-residue generation. Because the ATB is only used with surrogate materials, removing and installing automated equipment for new demonstrations is straightforward. The ATB is being used in a pseudoproduction mode to gather reliability, availability, maintainability (RAM), and production data. Operating in this mode requires configuration control. So, although installing and reconfiguring glovebox equipment may be easy, it is not always convenient. To accommodate the rapid development of new equipment and procedures while maintaining

Figure 1. The Automated Test Bed (ATB), an atmosphere-controlled glovebox, performs surrogate testing of automated plutonium processing systems and equipment.

Figure 2. The Process Demonstration Testbed facilitates innovations in equipment and procedures.

ATB configuration control, a Process Demonstration Testbed (PDTB) has recently been installed alongside the ATB.

The first demonstration of the ATB performed the material handling required for automated calcining of plutonium oxide powder. Since that time, a more complex system has been developed that integrates material handling with operation of the APF to reduce plutonium oxide to plutonium metal.

Information learned during the first year of ATB operation has led to improvements in the second-generation Glovebox Robot Gantry installed in the PDTB. Primary improvements address ease of disassembly for maintenance activities, improved access to components, and replacement of wear surfaces on the vertical mast with precision linear bearings. The PDTB (Fig. 2) is essentially a second-generation ATB with modular, removable floor panels but

without the windows. Sliding panels serve double duty as (1) safety interlocks to prevent operator access while the robot is in operation and (2) glovebox windows. This open-frame design allows equipment to be installed and removed easily for quick-turnaround demonstrations. Existing automated calcining equipment is being installed for initial system tests, but will soon be replaced by automated equipment for weapons assembly and disassembly demonstrations.

The PDTB is the primary vehicle for incorporating advanced technologies developed in the lab into demonstrations of production feasibility. IBM and LLNL are jointly developing a state-of-the-art controller to replace the original IBM ECR robot controller still in use in both testbeds. The original controller, based on the IBM Series I computer, has neither the computing power nor the advanced software features needed for real-time, sensor-based trajectory control; model-based path planning; or improved operator interfaces requiring high speed communications, such as force-reflecting hand controllers. All of these technologies are under development in labs at LLNL and across the nation and will be incorporated when the IBM RS/6000-based controller arrives.

To accommodate final testing of new plutonium-processing technology, a plutonium-capable glovebox is in the early stages of design. The plutonium-capable glovebox will accommodate a radiation-hardened version of the GR-G, incorporating any improvements suggested during operation of the ATB and PDTB.

Supporting Technology Development

Automation of glovebox operations has been successful in addressing a class of processing tasks that are both well-defined and within the manipulation capabilities of the robot arm. However, a much larger set of more complex tasks exceeds the current capabilities of existing robots and robot controllers. This set of tasks includes most complex tasks involving object manipulation, tasks that must be performed in a cluttered environment, tasks requiring sensory input for control, and error-recovery tasks. At present, the performance of these tasks in a hazardous environment is relegated to teleoper-

ators with human-in-the-loop controllers. The amount of sophistication required to handle most of these tasks is beyond the capabilities of the most advanced robotics research controllers and will remain so throughout the initial design phase of the Complex 21 facilities.

To perform these complex robotic tasks, advanced control system, teleoperation, and telepresence technologies are being developed and integrated in LLNL's Interactive Controls Laboratory for incorporation into the GR-G suite of tools. By integrating autonomous and teleoperation capabilities into the robot control system, operators will be able to switch seamlessly between both modes of operation to perform maintenance activities, error recovery operations, and process intervention without suiting up to manually perform these tasks. This capability is essential to successful application of automated glovebox manufacturing systems in hazardous environments.

Technologies being developed in the Interactive Controls Lab include:

- Advanced robot controller technology for sensor-based control and real-time collision avoidance.
- Force-sensitive robot control to accommodate variations in expected and actual environments.
- Improved man-machine interface technology including force-reflecting hand controllers, 3-D vision systems, and context-sensitive control systems.
- Machine-vision and object-recognition systems to minimize hard-tooling requirements and manual robot programming.
- Fiducial target matching for system calibration and off-line programming.
- Dexterous end-effectors for improved flexibility.
- Part scanning and digitizing systems for real-time CAD model revisions.

These technologies must be integrated with advanced controller technologies to provide the flexibility needed to accommodate differences between theoretically perfect computer models and the relatively indeterminant world of manufacturing. This indeterminance arises from the inherent inaccuracy of robot arms and is independent of repeatability issues. Conventional robot teach-pendants are ineffective tools for manual on-line programming,

while off-line programming packages treat robots as six-axis numerical control machines with inherent accuracy. This approach has limited the successful application of robots to many manufacturing processes.

To ease integration, recognized and emerging standards-based protocols are being used wherever possible. The Department of Defense Next Generation Controller architecture is being used as a reference model to simplify transfer of developed application code to vendor-supported platforms in development. The Government Open Systems Interconnect Profile (GOSIP) is being adopted as the communication protocol of choice for integrating manufacturing subsystems whenever practical.

Teleoperation provides an attractive alternative to manual process intervention when variations in process parameters exceed the capability of the automated system's preprogrammed response. Teleoperation eliminates the need for the operator to suit up and perform manual operations, thereby reducing hazards to the operator, minimizing operator-associated waste generation, and reducing special nuclear materials (SNM) accountability concerns. This flexibility is necessary for the timely demonstration of the diverse automated processes needed for Complex 21.

The APF Automation Demonstration

The first real test of the Automated Testbed and the Glovebox Robot Gantry occurred with the integration and testing of the APF for direct oxide reduction of plutonium. Direct oxide reduction (DOR) is a pyrochemical operation that converts plutonium oxide directly to metal in the presence of a molten salt. Current DOR processing is performed in stationary furnaces. Operational characteristics of these furnaces include relatively long cycle times, waste-generating ceramic crucibles, and manual operations. By automating pyrochemical furnace operation, the APF eliminates ceramic crucibles, significantly decreases cycle time, and decreases manual operations—the primary goal. Complete process automation reduces operator radiation dose and minimizes operator-associated tertiary wastes.

The APF is an improved furnace design based on earlier work at the Argonne National Laboratory, the Rocky Flats Plant, and other DOE sites. The APF is a tilt-pour furnace that allows the melt to be poured into a chilled mold at the appropriate time in the processing cycle. Using chilled refractory metal molds and crucibles eliminates the ceramic crucible waste generated in the stationary furnace and improves process efficiency. The APF's shorter furnace cycle time permits a 75% decrease in the pyrochemical-facility space requirements. This size reduction has a significant impact on plant capital costs and tertiary wastes—factors that are dependent on the square footage of the facility.

Automation activities related to tending the APF include feed charge makeup, mold and charge insertion into the furnace, automation of the furnace operation, product removal, and separation of the product from the salt cake—an operation known as button breakout. Figure 3 shows a diagram of these operations. Automated button breakout has yet to be demonstrated with surrogate materials from the APF. However, teleoperated button breakout has been developed and demonstrated in the lab to accommodate uncertainties in the button position and location that occur due to process variations.

Automation of the APF operation involves coordinated control of four motion axes, a rate-controlled stirrer, the temperature profile control system, and a large number of device and safety interlocks. The APF controller software is a Paragon system resident on an Intel 80286-based personal computer platform. Due to limitations in the Paragon controller, data acquisition was off-loaded to an Apple Macintosh system running Labview software. The APF control system was quickly integrated with the IBM GR-G control system for a semi-automated system operation demonstration.

The unique sensing capabilities of the IBM GR-G were used during all of the material handling steps to ensure a known operating state prior to product movement. Multiaxis strain gauges in the fingers measured contact-sensing tasks (e.g., the presence or height of product to be removed from the load-out can). An optical transmitter/receiver pair incorporated into the finger tips allowed the robot to

Figure 3. Robotic operation of the tilt-pour furnace. The automated furnace-tending process uses robotics to (a) load the crucible and the furnace, (b) melt and stir, (c) pour the melt into the chilled mold, (d) cast, (e) remove the charge from the mold, and (f) separate the button from the salt.

detect the presence of objects between its fingers and determine placement locations in storage racks, after checking for the presence of existing containers. The robot's ability to detect objects quickly and easily provides a layer of redundancy in case fixed sensors fail and reduces the number sensors required to be wired into the glovebox system. The ability to respond to changes in the expected environment adds a measure of robustness to the application that will be useful when handling radioactive and hazardous materials.

Future Work

Development of an advanced, sensor-based robot controller for the GR-G is underway in a collaborative effort with IBM. When the controller is completed later this year, it will be the most advanced robot controller commercially available. The installation of the new controller will allow all of the interactive control technologies to be used on the robot in the PDTB.

Calibration systems, now in development, will accommodate robot errors and define positions of workcell components. These calibration systems, which facilitate off-line programming of robot motions, will then be transferred to the PDTB to support rapid application programming.

Advanced technologies developed in the Interactive Controls Lab will be ported to, and integrated with, the GR-G's own capabilities. Porting these capabilities to the GR-G robot will allow remote human process intervention during development of automated pyrochemical processes and will facilitate changes in process configuration, allowing the PDTB to quickly change from one application to another.

Conclusion

Development of automated technology and applications for Complex 21 is a complicated task involving intelligent integration of manual, remotely controlled, and automated processes. Automated glovebox processing technologies under development in LLNL's APT program are addressing a wide range of manufacturing needs. Wherever possible, technology developments are linked to existing or emerging standards activities. This approach will facilitate concurrent development and implementation of advanced technologies required to minimize waste, reduce hazards to operators, and reduce criticality and SNM accountability concerns during Complex 21 reconfiguration efforts. Automated glovebox technology developed with these issues in mind is already providing, and will continue to provide, many opportunities for government and industry to work together to strengthen our position in a competitive world marketplace.

Systems Analysis for the Optimization of Complex 21

Computer simulations will provide tools to optimize important parameters such as dose, waste generation, and cost in the design of the Complex 21 weapons plant.

The Special Isotope Separation (SIS) Program developed a modeling and systems analysis program as part of the design work for the SIS Plant. This analysis effort was used to optimize the SIS flowsheet for plutonium processing and to analyze and reduce both radiation dose and process waste. The eventual plant design was a result of this analytical effort.

Since the close out of the SIS Program and the transition to APT, the Program's modeling and systems analysis expertise has supported the Department of Energy (DOE) Nuclear Weapons Complex (NWC) reconfiguration effort—Complex 21. As key players in the Plutonium working group of the Technology Assessment and Selection Panel (TASP), APT's systems analysis group assists in the evaluation of candidate technologies and process-chemistry flowsheet options for Complex 21.

The objective of the TASP-supported systems analysis effort is to develop and apply modeling tools to optimize overall facility parameters such as plutonium utilization, waste generation, worker radiation dose, and capital and operating costs. The TASP has developed a baseline flowsheet for the proposed plant. This flowsheet is being used by the DOE architect engineer in the Programmatic Environmental Impact Study (PEIS) and for preliminary conceptual design studies.

This report presents the analytical tools, approaches to flowsheet development, modeling options, and computational results for the TASP baseline flowsheet development effort.

Analytical Tools for Flowsheet Development

G.A. Armantrout

The three components needed for systematic analysis of the flowsheet and technology options for Complex 21 are :

1. Chemical process flowsheets representing alternative processing strategies.
2. Accurate data bases that describe the chemical and mechanical performance of technology options. Parameters include yield, processing parameters, chemical usage, waste generation, and worker time and dose.
3. A computational framework that accurately describes the movement of materials within the flowsheet and calculates the flowsheet performance parameters of interest, based on a given data base input.

Developing the Chemical Process Flowsheet

The key factor in developing a flowsheet format that allows systematic analyses is to define a unit process operation as a "black box" with defined terminal characteristics, as shown in Fig. 1. These unit process operations can be of any arbitrary size and complexity, but most conveniently match a given piece or grouping of equipment. A flowsheet is simply the wiring diagram for these unit process operations.

Model Parameters

Process Equipment Characteristics

Batch size, kg
 Cycle time, hour
 Man-hours/kg Pu
 Number of processing stations
 Number of pyrochemical boxes
 Number of pyrochemical boxes with vacuum pump
 Number of aqueous boxes
 Whole body dose, mrem/kg Pu
 Extremity dose, mrem/kg Pu
 Processing floor space, ft²

Process Wastes

Spent Pyrochemical salts
 Excess acid
 Silver compounds
 Other chemicals
 Solvents
 Nonreactive metals
 Ceramics
 Sludge
 Paper
 Water

Secondary Transuranic (TRU) Waste

Tools
 Refractory metals
 Nonreactive metals
 Heavy metals
 Ceramics
 Graphite
 Plastics
 Lead gloves
 Rubber
 Filters
 Failed equipment (metal)
 Oils/solvents
 Wipes
 Glass
 Sweeps

Low-Level Waste

Scrubber solution
 Surgeon's gloves
 Laundry sludge
 Paper trash

The Data Base

The data base is composed of the various parameters (Fig. 1) that describe a unit process operation. One basic category of data is all product and residue streams containing plutonium that are generated by a given plutonium input. The branching yields and destinations are part of the data base for all defined plutonium inputs. Unless the unit operation is a storage site, the sum of all streams is zero.

Figure. 1 Unit Process Operation. The unit process operations are the building blocks of the process flowsheet. Each unit process operation, usually representing a piece or group of equipment, is a "black box" with a set of terminals indicating the inputs and outputs of the operation.

All plutonium inputs are defined by the output specifications of other flowsheet unit process operations. Various reagents and materials are specified as inputs for the given unit process operation and may be plutonium throughput or time-dependent parameters. Similarly, all wastes that are generated during process operations are specified inputs in the data base. These wastes may be either throughput-dependent, time-dependent, or both.

Certain process parameters, such as dose and man-hours, can be specified for each unit operation. Other data, such as the number of processing stations and process cycle time, can also be defined. These parameters (see box) then become part of the overall data base used to evaluate selected process operations.

Determining what data to specify, and the appropriate values for that data, is a monumental task. Some process data can be determined from historical operations. Unfortunately, most historical data are incomplete or were not kept in a way that permits individual process data to be extracted. As a result, most data gathering is based on expert judgment and involves interviewing operating personnel. The Rocky Flats Plant (RFP) and Los Alamos National Laboratory (LANL) have been actively developing such a data base for a number of years.

In January 1992 the TASP convened experts from DOE sites [LANL, RFP, Savannah River Site (SRS), and LLNL] to develop a baseline data base for use in the TASP studies, to support the architect engineer in the PEIS evaluation, and to develop a design guidance document. At that meeting, participants developed a data set to determine the envelope of operations for the PEIS. The APT staff, actively involved in developing the data set, have used this preliminary data base for initial studies. A revised version, refined in subsequent meetings, will be used in future studies.

Computational Framework

The computational framework will provide the bookkeeping system that simulates the performance of the chemical process flowsheet. This flowsheet is based on the process input and output data base. A range of approaches is possible: from a standard spreadsheet program that provides a steady-state balance and output calculation, to a direct simulation of actual process operations and material flows as they may occur in an operating plant. We have used both ends of this range for our analysis of various options.

The more detailed computational approach, designated by TASP as the official one, was developed by the LANL A-7 Group. This approach attempts to simulate plant operations, in step-time, by calculating plant operations in the appropriate sequences whenever a unit process input reaches a predetermined batch size. Processing times, batch capacities, and simulated random failure rates determine actual unit process throughput and allow material to build up in a processing queue. Using the supporting data base, the actual process operation

(Fig. 1) computes the splits and additions for a given batch size. The resulting outputs then show up as inputs for follow-on unit process operations. Various waste residues, operating parameters, and product streams are appropriately summed as a function of time, stepped in two-hour increments.

This computational approach, which demonstrates the dynamic response of the simulated flowsheet, is particularly useful for understanding upset conditions and varying production and site-return rates. Static material-balance conditions are achieved either by running until steady-state conditions are achieved or by inputting a batch of site returns and then exercising the plant until more than 99% of the plutonium appears in an output stream.

Due to the large number of parameters being tracked, this model requires a microcomputer with sufficient memory and processing speed to support the simulation. Typical run times are 2–3 h. Additional codes, now in development, will facilitate the laborious tasks of data input and the interpretation of calculational results.

Because many of the performance parameters of interest are based on annualized or steady-state operations, a second computational framework, developed at LLNL, uses a PC platform with a much shorter computational time. This computational approach uses the same chemical process flowsheets and input data bases, and calculates all of the stream splits, wastes, and parameter-generation rates as indicated in Fig. 1. However, rather than simultaneously time-sequencing and tracking all material flows as in the A-7 computational approach, this code obtains only a plutonium stream flow-rate balance for the flowsheet. This stream flow balance is achieved by inputting a batch of plutonium and then exercising the process operations (Fig. 1) for the sequence of operations defined for the flowsheet, until all plutonium appears at the product and waste destinations. This sequence takes a few seconds to ten minutes, depending on the complexity of operations and the processing speed of the PC. The various waste- and process-parameter values are then calculated for each unit process operation, based on the appropriate data base and the determined plutonium throughput for that unit operation.

The results for each unit process operation are summed for the portion of the flowsheet being evaluated (e.g., site-return/metal purification processing, manufacturing, and residue recovery) or for the total flowsheet. These summed values make it possible to predict total waste-stream generation rates, worker dose, man-hours of processing time, number of gloveboxes, etc.

Both models yield the same basic steady-state results. By applying the appropriate cost-assumption algorithms, overall values (e.g., average worker dose, plant size, and plant capital and operating costs) can be calculated based on these results.

The PC model, which performs a large number of simulation computations quickly and simply, is used to prescreen results and to perform various parameter-sensitivity studies. The optimum cases are input to the TASP model for the same flowsheet and data base to obtain verification and to produce an output which is consistent with TASP requirements. This model can also perform other calculations, such as the effect of random equipment failure and equipment utilization rates.

Studies To Date

Attaining a fully reviewed and accepted data base for use by TASP is a continuing activity. To begin analysis of higher level options, the PEIS data base was modified and expanded where key data were missing. Input values were calculated and prepared for use by the code. With this data base, a number of technology and flowsheet options were studied using the PC computational framework. The results of the PC code were subsequently confirmed (using the TASP computational code).

Baseline Study

The TASP baseline flowsheet (January 1992 version) was modeled, and performance parameters consistent with the January PEIS input were used. Plutonium yield, plutonium streams, and various waste production rates were calculated.

LLNL Processing Technology Impacts

A number of LLNL technologies are under development including precision casting of plutonium parts, an advanced pyrochemical furnace, mediated electrochemical plutonium dissolution, and automation for plutonium handling. Although the basic TASP baseline chemistry flowsheet was used, the data base was modified to reflect the enhanced performance parameters that have been measured, or which are expected when development is complete for the above technologies. The same output parameters were calculated.

Flowsheet Variations

The impact of implementing several flowsheet variations was studied. These variations included the substitution of new pyrochemical process operations in place of some of the aqueous operations for residue recovery and the elimination of certain chemical processing operations. Flowsheet model changes were developed for these processing assumptions, and the updated data bases for the new processes were used. Existing processes operated with the original data base. Plutonium yield, plutonium streams, and the various waste streams were calculated.

Computational Results

Analyses and verification of these computational results are ongoing. Figures 2 and 3 indicate preliminary results of these studies and show the expected order-of-magnitude improvement, if such technology options were fully developed and deployed.

Figure 2(a) shows the impacts of various technology improvements or flowsheet options on the radiation dose received by the production workers. Each of the three main flowsheet areas was considered separately: (1) site-return/metal purification operations, (2) manufacturing, and (3) residue recovery. The RFP assumptions are for the TASP baseline flowsheet, based on the lower yields experienced at RFP for the various chemical process operations and casting. Precision casting is shown to have a major impact on the residue recovery dose by greatly

reducing the amount of plutonium that must be recovered. Site-returns are virtually unchanged. The notable improvement in manufacturing operations is due to reduced handling during machining operations.

The application of additional LLNL technologies (advanced pyrochemical furnaces, mediated electrochemical oxidation, automation, and precision casting) has a major impact on all areas when compared to the TASP baseline. A further reduction in chemical processing activities drives site-return and residue-recovery operations dose values almost to zero, leaving only manufacturing as a source of radiation dose in the plant.

Figure 2(b) shows the impact of various processing technologies and processing options on the total waste generated in the plant. Assuming RFP yields for the TASP baseline flowsheet causes a major increase in the waste generated by the residue-recovery operations. This is due to the much larger plutonium streams which must be processed. The use of precision casting substantially reduces waste generation, particularly in the residue area. Further application of LLNL technologies results in reduction in the manufacturing and site-return/metal purification areas. As expected, minimal chemical processing yields the biggest decrease in waste reduction from the site-return/metal purification and residue recovery areas.

Figure 3 shows the impact of the various options on the total cost of plant waste, dose, and life cycle. Costs for the total plant, which include construction, operating, and decommissioning, decrease as plant complexity and size decrease. Likewise, dose drops as technological improvements are added, until the manufacturing area contributes the dominant share of the total dose. Waste is reduced as each succeeding level of options is added. Advanced technologies and reduced chemical processing reduce the important parameters of cost, dose, and waste by a factor of five.

Future

The data base is continually being updated with ongoing refinements in the calculations of such factors as worker dose, plant size, and costs. Dose is a particularly important consideration in the design of Complex 21. Detailed dose

Figure 2. The relative impact of five processing technology options on (a) dose and (b) total waste is shown for each of the three processing areas in the plant: manufacturing, site-return/metal purification, and residue recovery.

Figure 3. The relative impact of the five processing technology options on the total plant of waste, dose, and life cycle costs.

analyses are underway for each unit process operation in the model. Dose analyses being developed use detailed time-motion studies and calculated source-term radiation fields. On the basis of these calculations, projected worker radiation dose levels per kilogram of plutonium processed are evaluated and readied for input to the plant-simulation code.

Cost algorithms are also being developed, based on experiences in the design of the SIS plant and in the design and construction of nuclear facilities. Cost factors that have been calculated include capital construction costs, operating costs, and decommissioning costs.

These cost projections are very important in the selection of technology options for further development.

At present, modeling has been applied only to the primary flowsheet operations of site-return/metal purification processing, manufacturing, and residue recovery. Waste processing operations and the analytical laboratory have not yet been included, but will be added as part of future modeling efforts.

Because many Complex 21 flowsheet options involve new technologies now in development, the evolution of the data base is an essential tool for evaluating these options. As active participants in the TASP process, the APT team is working to develop a unified data base for establishing the TASP baseline.

Publications*

A

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About This Issue

This first issue of the *APT Semiannual Report* focuses on APT's defense-related technologies. These technologies are a continuation of the research, development, and engineering work performed by LLNL's Special Isotope Separation (SIS) Program. SIS was the first large-scale DOE venture that had environmentally conscious manufacturing processes and facilities as its deliverables. The objectives were to create a facility where the only outputs were either usable products or disposable wastes, and to comply with existing and anticipated federal, state, and local regulations related to safeguards, security, health and safety.

To meet these objectives, revolutionary changes were needed in plutonium processing operations, chemistry, and equipment. New processes had to be developed that enhanced worker safety, minimized operator radiation dose, minimized waste at the point of generation, and provided for built-in recycling of residues. The SIS Program developed and demonstrated the technology (both chemistry and physics) necessary to provide plutonium with individual isotopic tailoring. This process made it possible to transform fuel-grade plutonium into weapon-grade material. However, due to the changing world political climate, the country's need for plutonium to make new weapons has decreased dramatically. As a result, the planned SIS plutonium-separation plant will not be built.

After the SIS Program was canceled in 1991, Congress directed that the plutonium processing technologies under development for the SIS Program be redirected to the weapons program. APT took over the development of the innovative SIS technologies and is applying them to the development of a new, reconfigured Nuclear Weapons Complex—Complex 21. "Close Out of the SIS Program" describes the completion of the SIS research and development work, and the transfer of key technologies to support this reconfiguration effort.

APT's contribution to the development of Complex 21 centers on plutonium metal purification, manufacturing, residue recovery, and waste management operations. "Plutonium Processing Technology for Complex 21" provides an overview of the role of significant APT contributions to Complex 21 including site-return processing/metal purification, aqueous nitrate recovery, aqueous chloride recovery, waste management, and robotics and automation. The article on "Automated Glovebox Processing" gives an in-depth look at the APT state-of-the-art automated robotic systems for glovebox material handling.

The APT Program's expertise in process modeling and computer simulations provides sophisticated tools for optimizing Complex 21 operations. The article "Systems Analysis for the Optimization of Complex 21" describes modeling and analysis of primary flowsheet operations of site-return processing, manufacturing, and residue recovery.