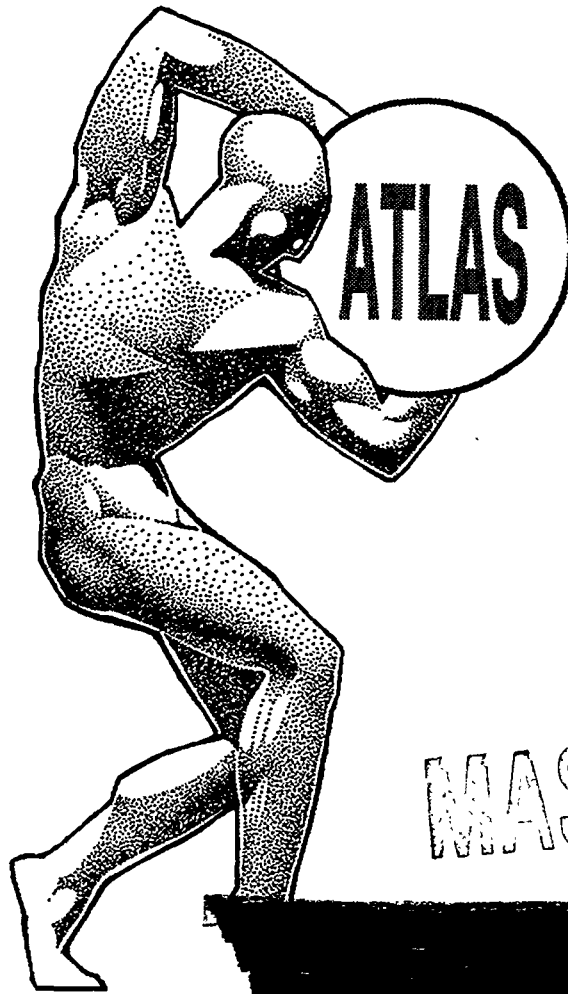




*An Introduction to the Advanced Testing
Line for Actinide Separations (ATLAS)*

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AN INTRODUCTION TO THE ADVANCED TESTING LINE FOR ACTINIDE SEPARATIONS (ATLAS)

by

Noah G. Pope, Stephen L. Yarbrow, Stephen B. Schreiber, and Roderick S. Day

ABSTRACT

The Advanced Testing Line for Actinide Separations (ATLAS) will evaluate promising plutonium recovery process modifications and new technologies. It combines advances in process chemistry, process control, process analytical chemistry, and process engineering. ATLAS has a processing capability equal to other recovery systems but without the pressure to achieve predetermined recovery quotas.

INTRODUCTION

The Advanced Testing Line for Actinide Separations (ATLAS) provides an opportunity to study and evaluate new processing techniques for recovering plutonium and other actinide materials. Research and production support are often incompatible. Research involves the evaluation of new and unproven ideas, many of which will be only moderately successful. But even an apparently unsuccessful idea can be valuable if it leads to a better understanding of the system. Full-scale evaluations interrupt normal recovery operations and result in a loss of throughput. There is constant pressure, therefore, to perform these evaluations as rapidly as possible to minimize the impact on process operations.

This conflict does not exist with ATLAS. Because it provides a flexible testing facility that is independent of recovery requirements, ATLAS will provide the opportunity to thoroughly evaluate promising process modifications and new technologies without interrupting regular processing functions. In addition, all of the major aqueous processing operations have been integrated in a modular fashion. Because any portion of the total system can be replaced or updated as better technology becomes available, ATLAS will represent the best technology for every aqueous process. For the first time a facility has been designed and constructed to promote the demonstration of new technology and to aid in technology transfer.

ATLAS focuses on three main areas of technology development: process chemistry, process control, and process analytical chemistry. These three areas, along with a description of the system's process engineering, are described in this document. ATLAS allows each technical requirement to be addressed by team members versed in that technical specialty. In general, the required chemical reactions are developed by process chemists, the required measurements are designed and performed by process analytical chemists, and then these measurements are used by control engineers to operate the processes. The accomplishments of these teams are orchestrated in ATLAS through the use of process engineering (Fig. 1). This document is appropriately divided so that each separate team addresses each operation with its own special perspective. Further information on each of these areas is available in the materials referenced throughout this document.

PROGRAMMATIC ROLE

ATLAS plays a dominant role in several current Department of Energy (DOE) programs including Waste Minimization, Complex Reconfiguration, and Radioisotope Recycle and Recovery. Technologies such as enhanced process control and on-line analytical chemistry will allow plutonium recovery processes to operate at their optimal

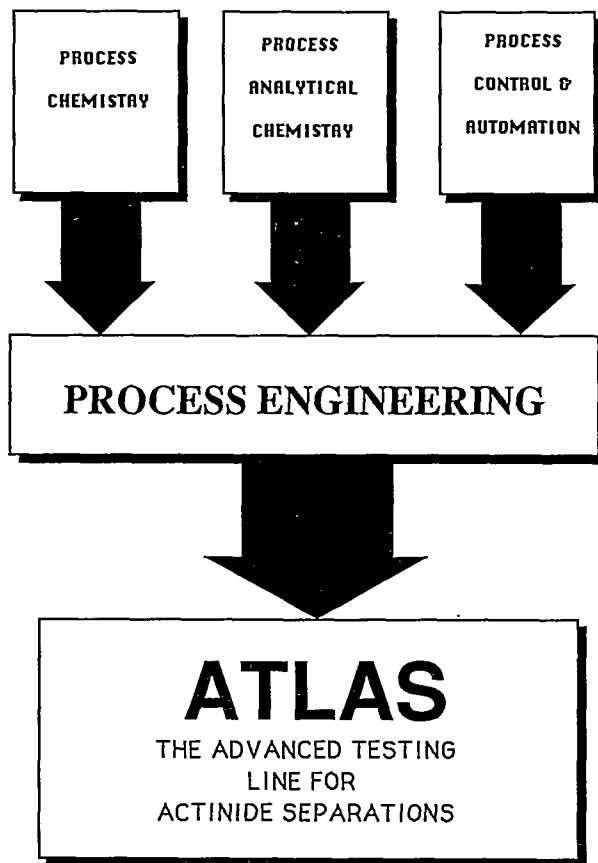


Fig. 1. ATLAS uses process engineering to bring together the contributions of other development teams.

level. This minimizes the waste produced at the source and reduces waste treatment and storage requirements. Coupling advanced analytical and control techniques with improved processing methods for dissolution, ion exchange, precipitation, waste polishing, and final treatment will also reduce waste. ATLAS will provide an integrated demonstration of all of these technologies, which can then be implemented in the current processing facilities at Los Alamos National Laboratory (LANL), Rocky Flats Plant, Hanford, and other DOE sites. Proper demonstration of these technologies will also provide reliable operating data for complex reconfiguration. In addition, ATLAS and the equipment at the Los Alamos Plutonium Facility will provide an opportunity for staff from Los Alamos and other sites to interact. This will make the smooth transfer of technology more likely.

PROCESS ENGINEERING

Process chemistry, process control, and process analytical chemistry are tied together through the use of proper

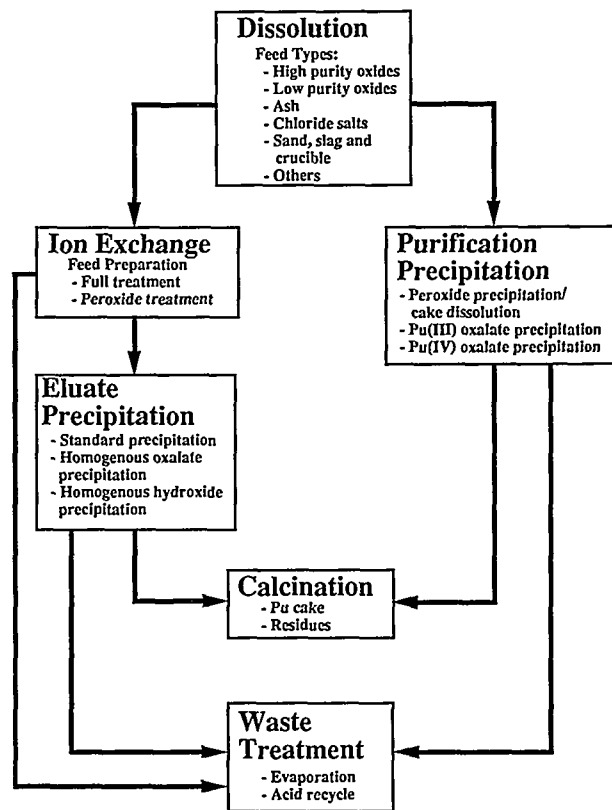


Fig. 2. ATLAS process flow diagram

process engineering. The goal of these interactions is to produce pure plutonium oxide for conversion to metal by the most effective means available. ATLAS's full-scale, modular design encompasses all of the major unit operations used in the nitrate aqueous recovery of plutonium (Fig. 2). This modular approach allows the interchange of new and original unit operations for the purpose of integrated and full-scale testing.

ATLAS consists of six interconnected gloveboxes and occupies a 400-sq-ft area (Fig. 3). It is connected to the rest of the Plutonium Facility through an overhead conveyor system. A chain-driven, automatic, floor-level trolley interconnects the ATLAS gloveboxes. This arrangement allows each glovebox to be independently ventilated, thus avoiding cross-contamination and potential corrosion. The processing line is made up of three double-width (4- × 8- × 10-ft) gloveboxes for general processing activities, one single-width glovebox for feed treatment and dissolution, and two single-width, standard-profile (2.5- × 3.75- × 5-ft) gloveboxes for analytical activities (Fig. 4). In addition, an external bank of 6-in.-diam "pencil" tanks provides the line with storage for various types of solution (Fig. 5). A small alcove houses computers that support the analytical instrumentation and the trolley controller (Fig 6). The personal computers that

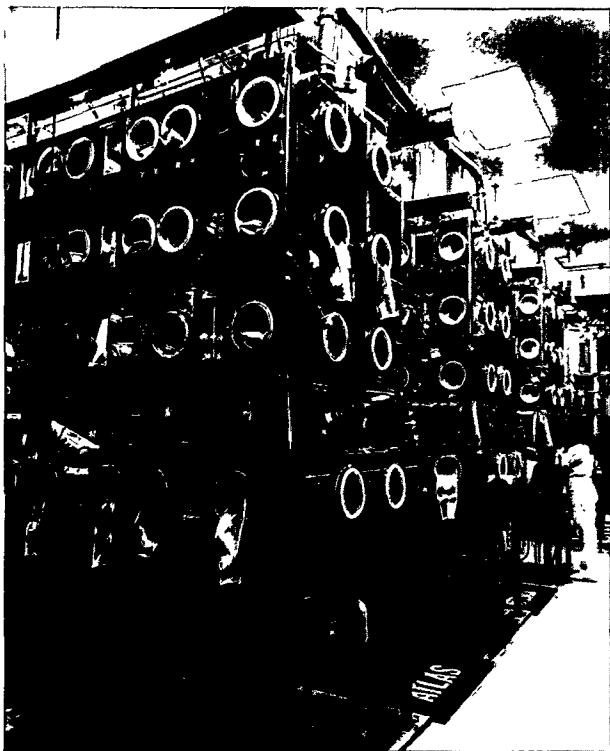


Fig 3. ATLAS gloveboxes.

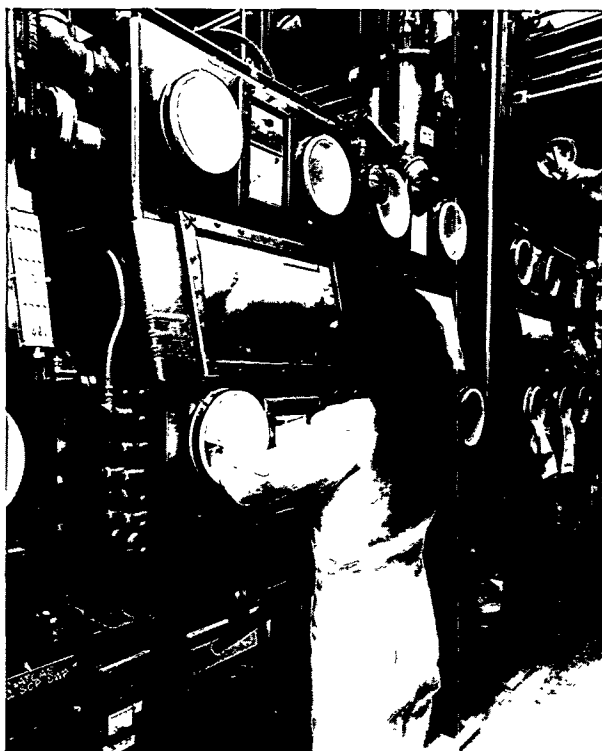


Fig. 4. ATLAS process analytical chemistry glovebox.

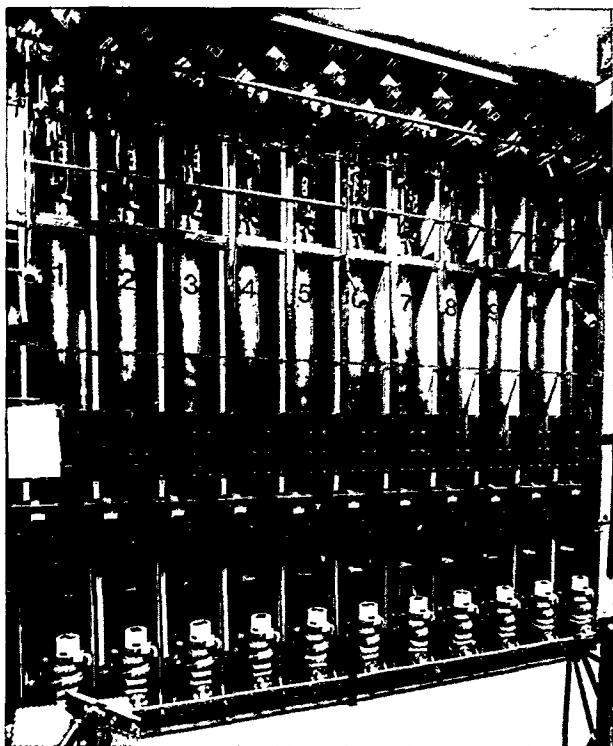


Fig. 5. ATLAS solution storage tanks.



Fig. 6. Several computers are networked and are used to control analytical instrumentation.

constitute the core of the process control network are housed in individual mobile cabinets located at various points around the line.

Each glovebox contains unique features for optimal flexibility and usefulness. Internal glass storage tanks are suspended from I-beams on the glovebox ceiling to provide open floor space (Fig. 7). Two of the processing gloveboxes are lined with fluoroplastic (polyvinylidene fluoride) to protect the stainless steel surfaces from corrosives such as hydrochloric acid. This allows ATLAS to run both nitrate- and chloride-based flow sheets. There are numerous extra, initially unused, service panels and utility penetrations to accommodate future process changes. All of the gloveboxes are built with 1/4-in lead shielding sandwiched inside the exterior walls to a height of 7 ft to minimize operator exposure.

ATLAS uses an evolutionary, rather than a revolutionary, approach to equipment and flow sheet development. Initially, ATLAS will predominantly use standard equipment that is currently in use at the Plutonium Facility. This will establish a baseline against which later equipment and flow

sheet changes will be compared. Many other pieces of equipment have been slightly modified to improve their overall performance. Changes, in both equipment and chemistry, can later be made to the unit operations so that any impact on the upstream and downstream operations can be characterized and quantified. Results from these studies will be used to help direct later flow sheets. In this manner, processes will "evolve" toward designs that will improve the consistency and purity of the product and reduce waste.

Unit Operations.

The startup flow sheet will consist of the following operations: batch dissolution, anion exchange, purification or eluate precipitation, calcination, and evaporation. Each of these is discussed below with regard to issues involving process engineering and then again in subsequent sections.

Dissolution.

Dissolution is the process by which the actinides in the initial scrap are dissolved into a nitric acid solution. The most common feed types used in this process are impure



Fig. 7. Glass storage tanks have been designed to increase the available floor space in ATLAS gloveboxes.

oxides, ashes, metal conversion residues, and pyrochemical salts.

The equipment normally used for the same process in other recovery areas at TA-55 consists of a round-bottom, 5-L glass flask set in an electric heating mantle. In ATLAS, two 3-L internally baffled glass resin kettles are used, and each is set in an electric heating mantle. The kettles are sealed with a ground-glass lid that has penetrations for a thermocouple, an offgas reflux condenser, a stirring shaft, and reagent addition (Fig. 8). The stirring shaft is run by a compressed-air-driven motor. The outlet of the condenser feeds the offgas into a 2-L caustic scrubber. For temperature control, a feedback controller located under the glovebox varies the amount of electrical current that is sent to the heating mantle. Dissolved feed is filtered through a polypropylene frit and stored in one of three 15-L glass columns that are mounted on the back wall of the glovebox.

Anion Exchange.

Nitrate anion exchange is the principal method of recovering, purifying, and concentrating plutonium from nitric acid. Feed is chemically adjusted to maximize the sorption of plutonium on the resin. The actual unit operation uses anion exchange resin loaded into 6-in.-diam glass columns of various heights to preferentially retain the nitrate com-

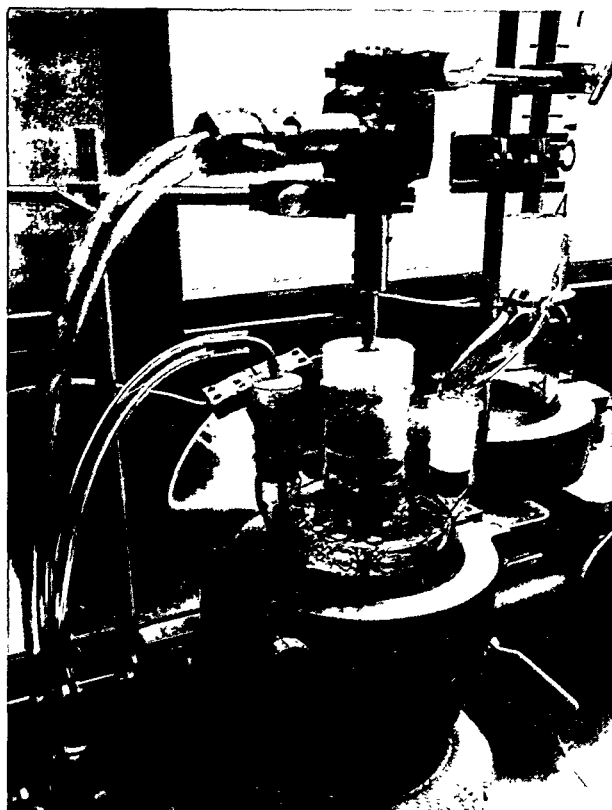


Fig. 8. ATLAS 3-L dissolution kettle.

plex of Pu(IV) over most other cationic impurities (e.g., americium and uranium) that may be present in the solution. After the impurities have been removed with the effluent, the column is washed, and the plutonium that was sorbed on the resin is eluted.

The feed is chemically adjusted in a single external 50-L stainless steel pencil tank. The volume in the tank is monitored with a magnetic-float level detector within a calibrated vessel. Fiber optics connect a spectrophotometer to a sampling loop on the recirculation line of the feed treatment tank (Fig. 9). This allows the feed solution to be continuously monitored to ensure proper chemical adjustment.

A normal anion exchange unit at the Plutonium Facility consists of one 2-ft primary column and a second 1-ft back-up column. In ATLAS, this configuration was changed to a pair of 3-ft columns. A series of ten solenoid valves connected by plastic tubing allows a variety of flow paths through the two columns to be controlled. This arrangement provides for maximum flexibility in developing optimal loading and stripping flow patterns. The feed, wash, and elution solutions are forced through the resin bed and associated cartridge filters by an air-driven, double-diaphragm pump. Six 25-L suspended glass columns are available to store solutions inside the glovebox.

Precipitation.

Selective precipitation based on the solubilities of plutonium complexes in nitric acid is another method of recovering, purifying, and concentrating plutonium. A peroxide precipitation is used to separate plutonium from americium and many other common cationic impurities. Oxalate precipitation, for either Pu(III) or Pu(IV), is used to separate plutonium from uranium, iron, and aluminum.

In ATLAS, this procedure is performed in a narrow-form, extra-high-profile glovebox containing specially designed suspended precipitators. Each precipitator is a 9-in.-diam glass column with an internal, air-driven, 2-in.-diam stirring shaft that is loaded with boron to ensure a criticality-safe geometry. The vessel is also equipped with two electric immersion heaters and a cooling coil. A feedback controller automatically maintains the temperature of the solution, thereby providing more consistent and complete precipitations. A pair of magnetically coupled gear pumps and two flowmeters are used to measure the flow of reagents from two suspended feed tanks into the vessel. The precipitates are collected on a polypropylene filter, washed, and dried. The filtrate is collected in a series of 25-L glass columns that are suspended inside the glovebox.

Two types of precipitation processes are performed in ATLAS: a concentration precipitation and a purification precipitation. The concentration precipitation converts concentrated plutonium nitrate solution from anion ex-

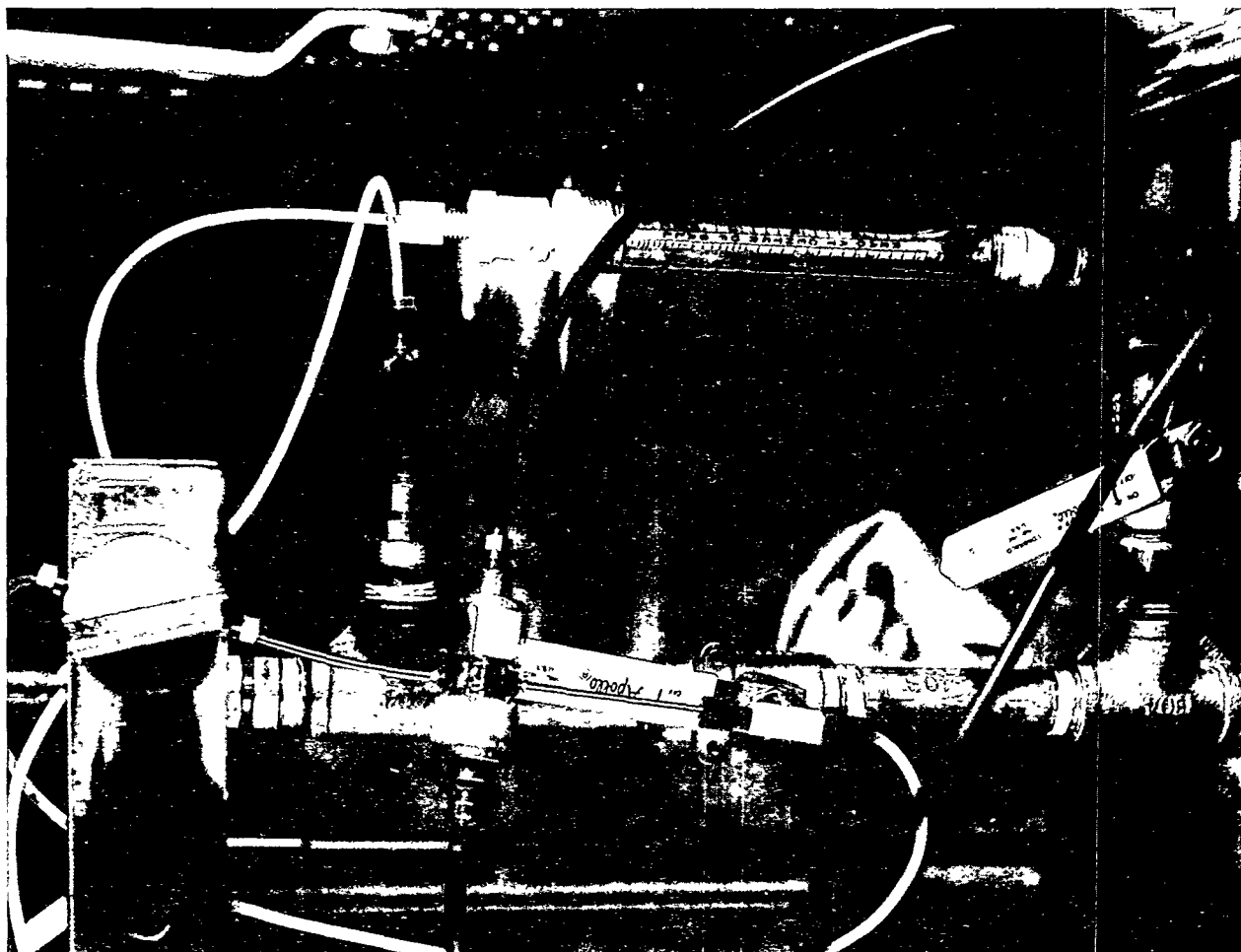


Fig. 9. Fiber optic solution-sampling loop.

change into a solid cake. The purification precipitation is used to purify dissolved plutonium-bearing scrap directly without using anion exchange. Traditionally, precipitation is performed by introducing solid oxalic acid directly into the solution. In ATLAS, a homogeneous precipitation can also be performed by the hydrolysis of diethyl oxalate.

Calcination.

The dried plutonium oxalate is calcined in a tube furnace to produce pure plutonium oxide. Similarly, other dry residues generated during the recovery operations are also calcined in preparation for further recovery or for long-term storage. These operations take place in two separate furnace enclosures, each having its own exhaust and prefilter systems.

The standard calcining operation uses an upright Lindberg tube furnace with the oxide contained in an Inconel can. In contrast, ATLAS uses a tilted can that rotates inside the furnace to mix the contents (Fig. 10). The

mixing reduces calcining time and generates a more uniform product.

Evaporation.

The anion-exchange effluents and all precipitation filtrates are collected and processed by a steam-heated, natural circulation evaporator that reduces the volume of solution by a factor of 24:1. The overhead distillate meets the Facility's economic discard limits for acid waste. The concentrated bottoms solution is transferred to the Plutonium Facility's waste-processing operation for disposal as a transuranic waste.

The thermosyphon evaporator is located within another narrow-form, extra-high-profile glovebox. The glass-and-stainless-steel unit uses two control loops to monitor liquid level and steam flow. Seven 25-L glass columns are suspended inside the glovebox to store feed and distillate. Once the baseline processing parameters have been established, one of the first flow sheet modifications will be to



Fig. 10. Rotating calcination furnace.

recycle the overhead acid distillate to minimize waste. This will require running the distillate through the evaporator a second time to obtain an ~10-M acid that will be used in the dissolution and anion-exchange pretreatment operations.

This is the industry's first opportunity to integrate all of the major aqueous unit operations for recovering plutonium from various scrap with process control and near-real-time analytical support. ATLAS will provide an environment for testing new equipment and unit operations using actual process solutions. The overall goals of ATLAS are to develop the optimal processing flow sheet and to minimize waste generation in a controlled, evolutionary manner.

PROCESS CHEMISTRY

Since the late 1950s, Los Alamos has primarily used anion exchange in nitric acid as the primary aqueous process for recovering and purifying plutonium from a wide variety of impure nuclear scrap materials.

In 1984, LANL initiated a major development program to upgrade and improve this process. Recent efforts have focused on (1) dissolution of impure nuclear feed materials, (2) pretreatment of the resulting feed solutions, (3) retention of plutonium and rejection of impurities by the ion-exchange column, and (4) treatment of waste from processing operations. As a result, DOE has asked the Los Alamos Plutonium Facility to be the lead laboratory for plutonium processing technology for the entire complex.

ATLAS technology will include the latest developments in plutonium processing. These baseline technologies, which are described below, will serve as a starting point from which new and improved processes will be implemented.

Dissolution.

All dissolution of plutonium-containing scrap materials are dissolved in a batch operation in 5-L flasks. In ATLAS, a more efficient design, which improves mixing and reduces corrosive fumes, will be installed and tested as a part of the optimized baseline. Even more advanced technolo-

gies, such as Catalyzed Electrochemical Plutonium Oxide Dissolution (CEPOD) and molten salt, are being studied for use with materials that resist conventional dissolution techniques.

Anion-exchange Pretreatment.

The pretreatment converts all dissolved plutonium to the strongly retained valence state of Pu(IV), adjusts the total nitrate concentration to 7 M to maximize the sorption of Pu(IV), and adds appropriate masking agents that will minimize the interference of complexing anions (fluoride, phosphate, sulfate, etc.).^{1,2} The formerly common "full treatment" involved a fourfold increase in feed solution volume and the addition of kilogram quantities of reagents; these resulted in longer processing times and unnecessary liquid and solid waste. New techniques that involve little or no volume increase and add essentially no solid residue to the feed³ have been implemented recently. Increased product purity and decreased waste generation are the results. ATLAS will evaluate other pretreatment possibilities without interrupting normal processing operations.

Anion Exchange.

The most significant gains in aqueous process chemistry at TA-55 have been with anion-exchange resin. As recently as 1984, the anion-exchange procedure used at TA-55 was virtually identical to the original process developed in the late 1950s. Since 1984,⁴ development efforts at TA-55 have resulted in an improved anion-exchange resin that has faster sorption kinetics for Pu(IV), more than double the plutonium capacity, increased resistance to chemical attack and radiation,⁵⁻⁸ and improved rejection of impurities.

Waste Treatment.

As the dissolution, pretreatment, and anion-exchange portions of the overall process have become more efficient, the demands on waste treatment facilities have decreased proportionately. However, increasingly stringent regulations and a desire to protect the environment have stimulated efforts to quantitatively recover actinides from waste solutions. Consequently, the evaluation of several highly selective solid and liquid sorbents is being continued. If these processes are successful, they could allow us to selectively remove the actinides from the bulk of nonradioactive impurities, which could then be disposed of as low-level industrial waste. The recovered, highly concentrated actinides could be recycled or stored far more economically.

Process chemistry research efforts have traditionally had to compete with recovery needs. As a result, the need to develop a testing and evaluation facility is often secondary to recovery operation commitments. ATLAS eliminates this conflict by providing an ideal facility in which

promising new process chemistry options can properly be assessed. ATLAS also provides an ideal demonstration opportunity to facilitate the transfer of proven new technology to DOE production sites.

PROCESS CONTROL

The ATLAS control system is structured to provide a safe, efficient, and reliable operating environment. The following goals provide the foundation for the ATLAS control system. First, information should be shared between processes so that the total operation flows smoothly from start to finish. Second, the hardware should be decentralized to avoid reliability and maintenance problems that are common to large single processors. Third, the control system should serve as an extension of the process operators, increasing their capabilities rather than replacing them. Fourth, control should be layered so that each process has at least two back-up modes of operation, including a manual override capability. Fifth, the control system must be flexible enough to accommodate unforeseen aspects of current projects or new projects. Sixth, the scientists, operators, and engineers on the project should avoid writing custom software and hardware drivers when possible. Finally, the system must be able to effectively integrate the information that is supplied by the process analytical chemistry efforts.

ATLAS will accomplish these goals by using a combination of three personal computers including both 80386- and 80486-based machines to run a commercially available control software package (Fig. 11). Each computer will be located near a different unit operation and will allow multiple operations to be displayed and controlled concurrently (Fig. 12). Software drivers for each piece of input/output hardware will be supplied commercially. The system will allow all operations to be monitored from remote stations that may be located in supervisors' offices. In addition, remote terminals that will allow ATLAS to be shutdown in an emergency can be purchased.

All operators of ATLAS will be trained to create their own control screens (Fig. 13) and design their own experiments. A complete data-logging capability will allow large quantities of information to be automatically collected and readily analyzed (Fig. 14). ATLAS will include both batch and recipe control capabilities as well as an on-line statistical product-quality-control package. An artificial intelligence software package will eventually be included to assist operators in interpreting alarms and problems. All of these special capabilities are available commercially, which will significantly reduce development time.

Operation of the ATLAS control system is an ambitious effort that will require skills and training beyond those common to most technicians. However, preparations are

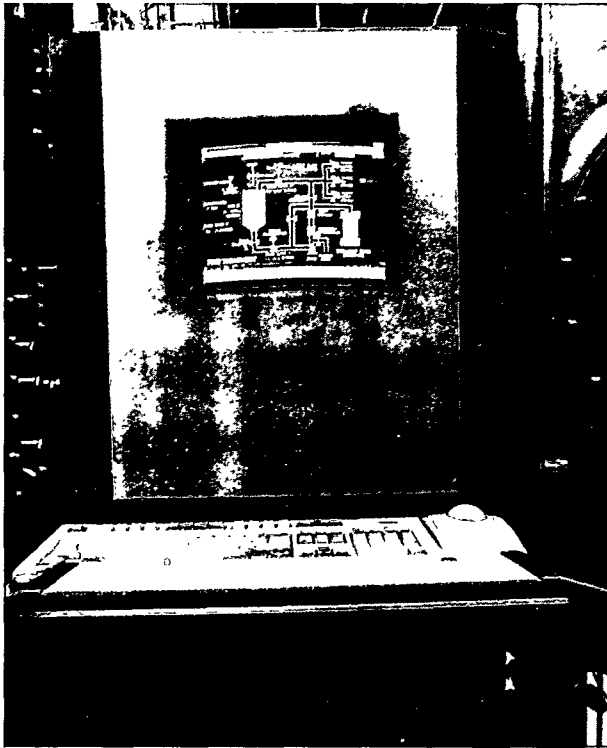


Fig. 11. Process control computer.

under way with over a dozen workers already trained to use the control software package. A similar system is already operational on an existing process at TA-55, and these control efforts have already helped to set new processing records. ATLAS's process control system will help to make ATLAS the safest and most technically advanced plutonium recovery operation in the complex.

ATLAS will incorporate such unit operations as feed preparation, anion exchange, evaporation, eluate precipitation, calcination, and purification precipitation.⁹ Other operations include solution transfers to and from a tank farm and acid preparation. All of these operations will involve automation and computer control. The preliminary designs for these control systems are outlined separately below.

Feed Preparation.

Feed materials for anion exchange will be dissolved in nitric and hydrofluoric acid solutions and then pretreated before being loaded onto anion-exchange resin. The pretreatment step will adjust the acid concentration and the plutonium valence of the feed solution to optimal conditions. The solution will first be pumped through a flowmeter and then analyzed with a spectrophotometer and acid sensor. Additional reagents including water, nitric acid, peroxide, hydroxylamine nitrate, and ferrous ammonium sulfamate will then be added until optimal chemical condi-



Fig. 12. The process control computers are used by process operators and are stationed adjacent to the process gloveboxes.

tions are reached. The solution would then be transferred to anion exchange for further processing.

Anion Exchange.

The solution from the feed treatment tank will be transferred through a flowmeter to the anion-exchange system and into feed-holding tanks. As the treated feed solution passes through two resin-filled anion-exchange columns, the plutonium product will be loaded onto the resin while impurities pass through. A Proportional, Integral, and Derivative (PID) controller will maintain a constant flow rate through the columns. A gamma-ray-energy monitoring system that is adjacent to the process piping will monitor the gamma signal strength of various plutonium, americium, and uranium isotopes to determine the effectiveness of this separation.¹⁰⁻¹² On the basis of these signals, the solution will be automatically directed to a cold tank for additional processing or to a hot tank for recycling. The solution will also pass through an X-Ray Fluorescence (XRF) instrument that will analyze it for various actinide concentrations, trace metals, and other impurities. A third column that is loaded with a chelating resin may be installed on the effluent line to remove any remaining low levels of plutonium and americium that may have passed through the regular anion-exchange column. A gamma monitor downstream of this column will detect actinides and log the information. An alpha-monitoring¹³ system has also been installed on this line to allow a comparison of the two monitors' sensitivity levels (Fig. 15).

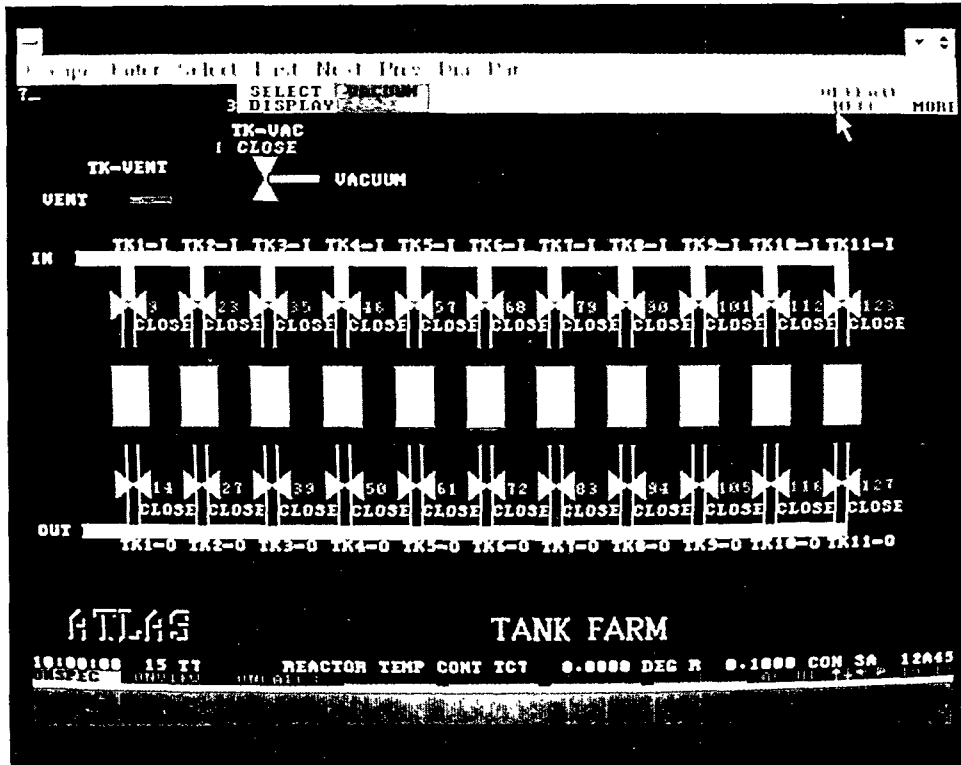


Fig. 13. The ATLAS tank farm control screen.

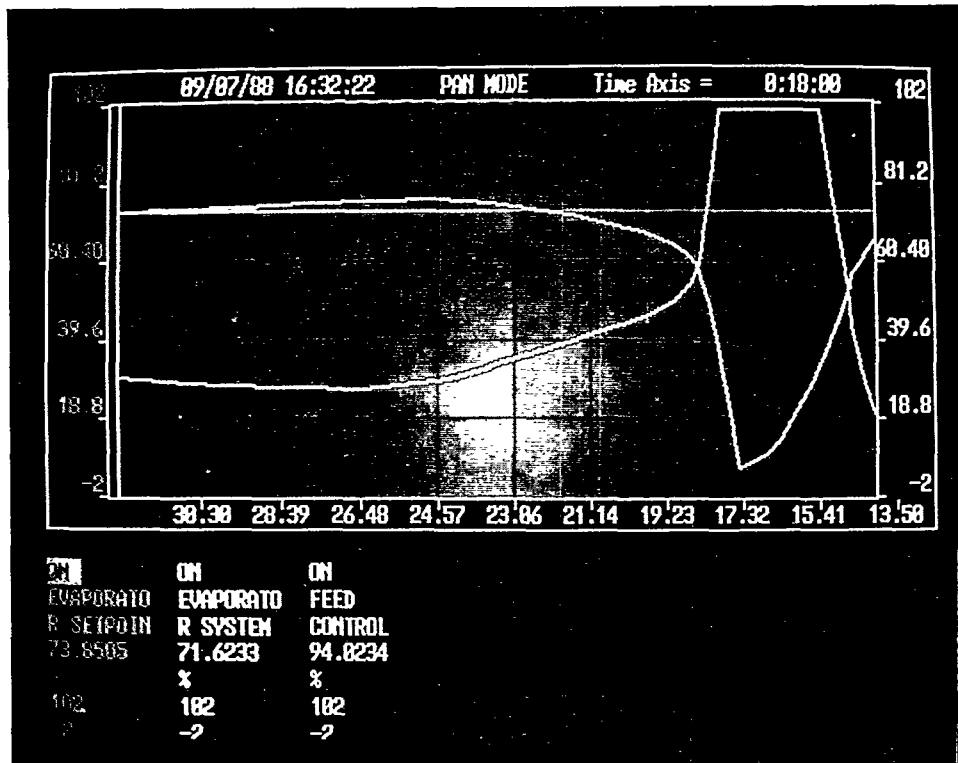


Fig. 14. ATLAS data display screen.

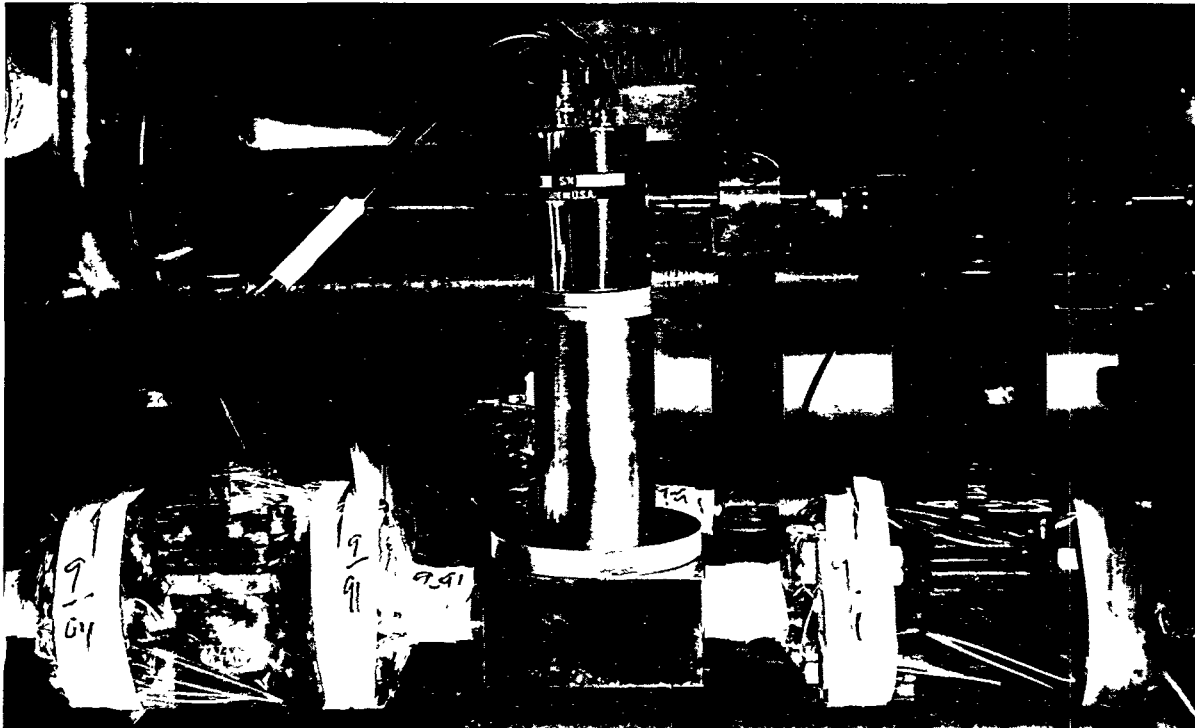


Fig. 15. Continuous on-line alpha monitor prototype.

Evaporation.

A steam-heated thermosiphon evaporator system will be installed in ATLAS to concentrate anion-exchange effluents and precipitation filtrates. The evaporator's process control system (similar to, but more sophisticated than, one recently installed on another evaporator process at TA-55) will consist of two computer-monitored PID controllers that maintain the solution level within the evaporator and optimize the steam flow (Fig. 16). Flow rates and temperatures will also be monitored and recorded.

Eluate Precipitation.

Eluate from the anion-exchange columns will pass through the precipitation process to convert the aqueous plutonium to a solid cake. The temperature will be closely controlled using a PID controller, immersion heaters, flow meters and a thermocouple (Fig. 17).

Calcination.

A rotary calciner will be used to convert plutonium oxalate cake to oxide. A container within the calciner will rotate at an angle to thoroughly mix the cake. The process will be regulated by a furnace controller system that contains a modular ramp-soak controller, a strip chart, and an automatic-shutdown safety mechanism (Fig. 18). The computer system will collect and monitor temperature information from these instruments.

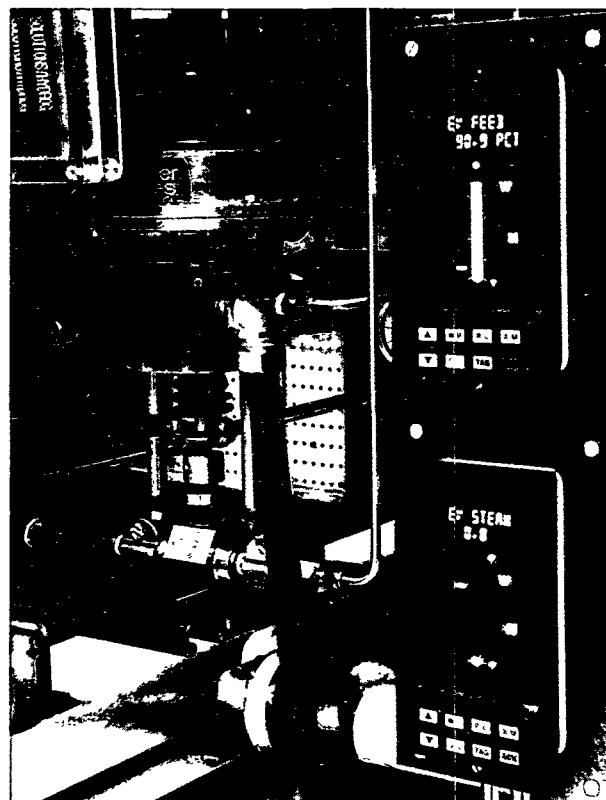


Fig. 16. ATLAS evaporator PID controllers.

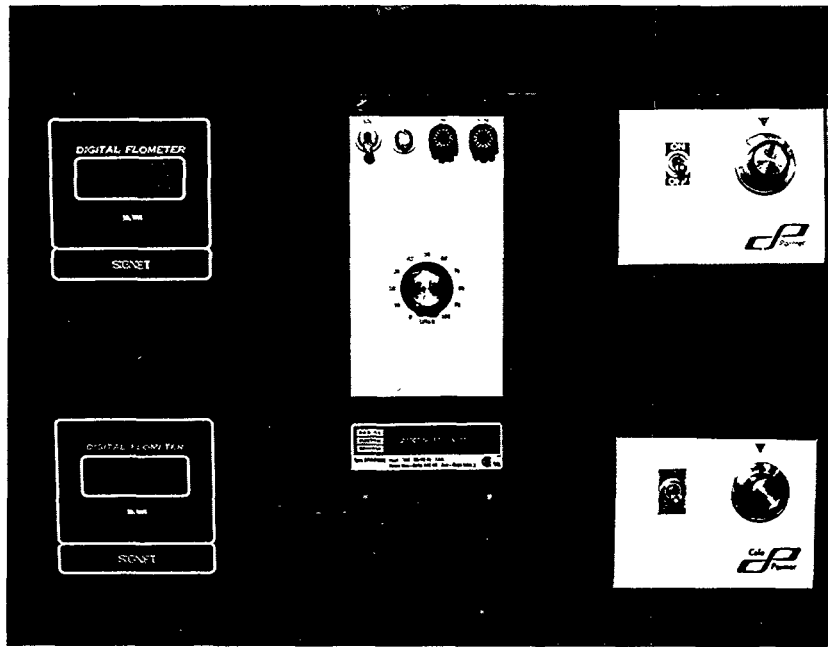


Fig. 17. Precipitator instrumentation.

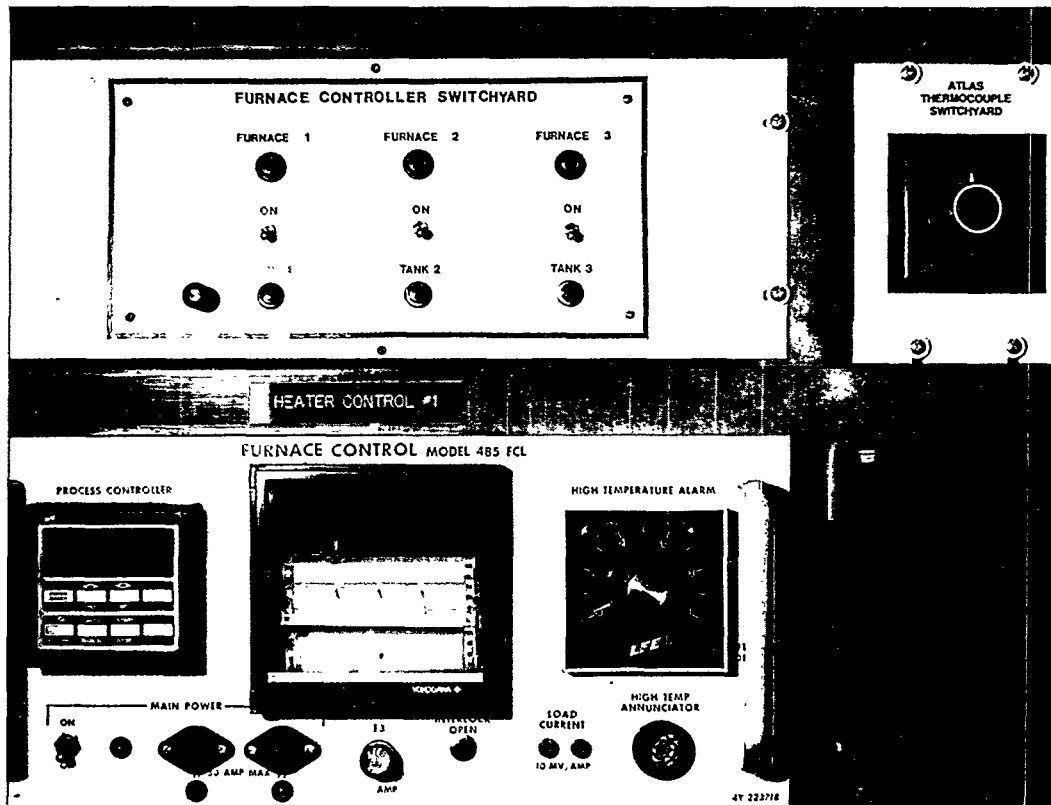


Fig. 18. Furnace and solution heater controller instrumentation.

Purification Precipitation.

Similar in function to the eluate precipitation process, the peroxide/oxalate precipitator process is an alternative to anion exchange that will allow different kinds of precipitations. Flowmeters and a PID controller that regulate the pump rate will control the ratio of plutonium feed to reagents. The temperature of the reaction will be closely controlled using a thermocouple, heaters, and a PID controller.

Tank Farm.

ATLAS has a closely packed bank of 11 vertical pencil tanks for the storage of solutions. The tanks have 24 remotely controlled digital valves that are air actuated by electronic solenoids (Fig. 19), which are controlled by either computer or manual switches (Fig. 20). The state of each valve will be confirmed by an electronic position sensor, which will be connected to the computer. Solutions will be transferred automatically when the starting and ending points in an operation are defined. The computer will determine the

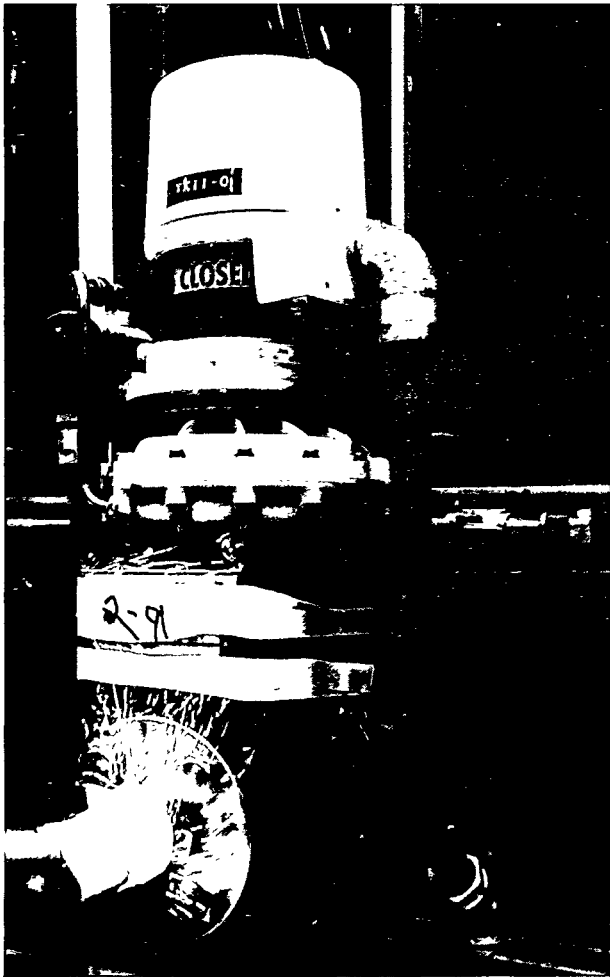


Fig. 19. Computer-controlled valve.

proper routing and will confirm this with the position sensors. Nonintrusive point-level sensors are attached to the outside of the tanks to monitor solution levels.

PROCESS ANALYTICAL CHEMISTRY

Although the ATLAS line is capable of testing a variety of Special Nuclear Materials (SNM) separation processes, it will initially be focused on the existing nitrate anion-exchange technology. One of ATLAS's first tasks will be to explore the extent to which this technology can be improved through the use of process analytical support.

ATLAS's analytical systems will provide analytical support on demand, with a maximum turnaround time of 2 hours. Other analytical systems located in PF-4 should provide a maximum turnaround time of 1 day.

The following analytical methods will be used at various points in the nitrate anion-exchange¹⁴ process: free-acid determination by acid-base titration and acid sensing; oxidation state determination by spectrophotometry; anion analysis by ion chromatography and spectrophotometry; cation analysis by emission spectroscopy, XRF, and mass spectroscopy; and radiochemical analysis by gamma spectroscopy.

Feed and Feed Treatment Acidity.

An appropriate method of titration will be automated and implemented (Fig. 21) to determine the total free-acid content. The method will be based on either the standard LANL free-acid method or the standard addition method developed at Savannah River Laboratory.¹⁵ The standard

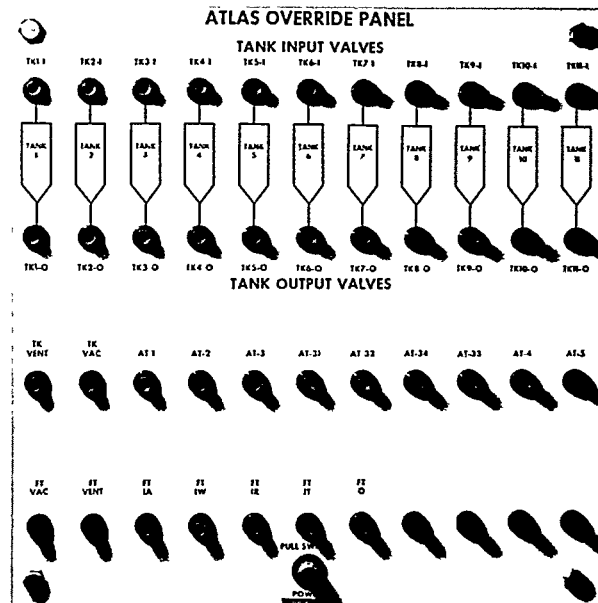


Fig. 20. ATLAS tank farm switch panel.



Fig. 21. Acid titrator.

addition method will analyze only the strong acids that are present and would have a wider range of applications (e.g., in determining the acidity of the eluates, which contain the weak acid hydroxylamine nitrate).

In addition to performing titration, ATLAS will be a testing ground for a new optical high-acidity sensor. This recently developed sensor will determine nitric acid concentrations up to 12 M. The sensor will be used to measure concentrations of recycled, relatively clean evaporator nitric acid distillates and to prepare acid solutions in user-defined concentrations and quantities (Fig. 22).

Feed and Feed Treatment Anion Concentrations.

Ion chromatography¹⁴ (IC) (Fig. 23a and 23b) will be used to analyze for inorganic anions including F^- , Cl^- , SO_4^{-2} , $C_2O_4^{-2}$, and PO_4^{-3} . An Ion Selective Electrode (ISE) will also measure F^- . A method using UV spectrophotometry will be used to measure NO_3^- .

Feed and Feed Treatment Metal Impurities.

Inductively coupled plasma-emission spectroscopy (ICP-ES) will be used to analyze for the presence of aluminum, silicon, calcium, magnesium, potassium, sodium, uranium, and thorium. Other metal impurities will also be analyzed, however these will not be specified until after ATLAS operational experience is gained. In addition to performing the ICP-ES analysis, XRF will provide a survey of other impurities.¹⁴

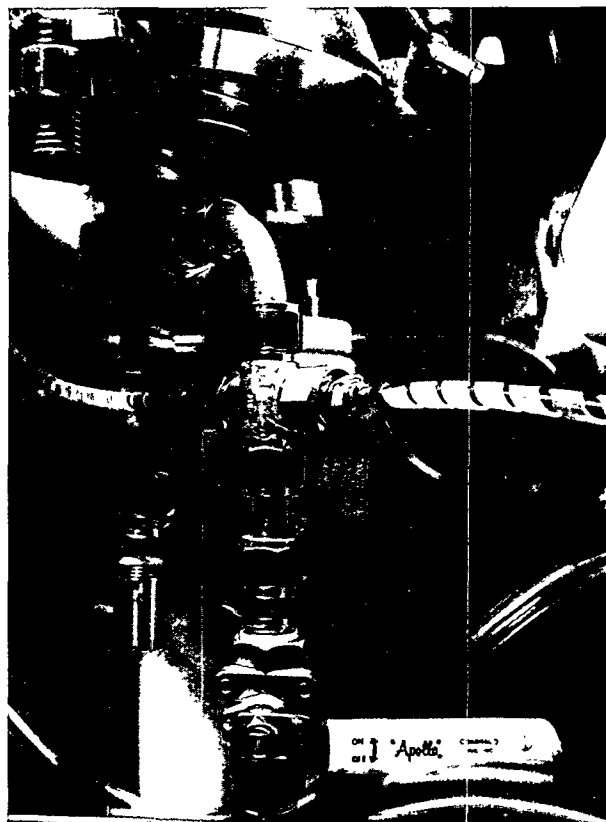


Fig. 22. In-line fiber-optic spectrophotometer sample cell.

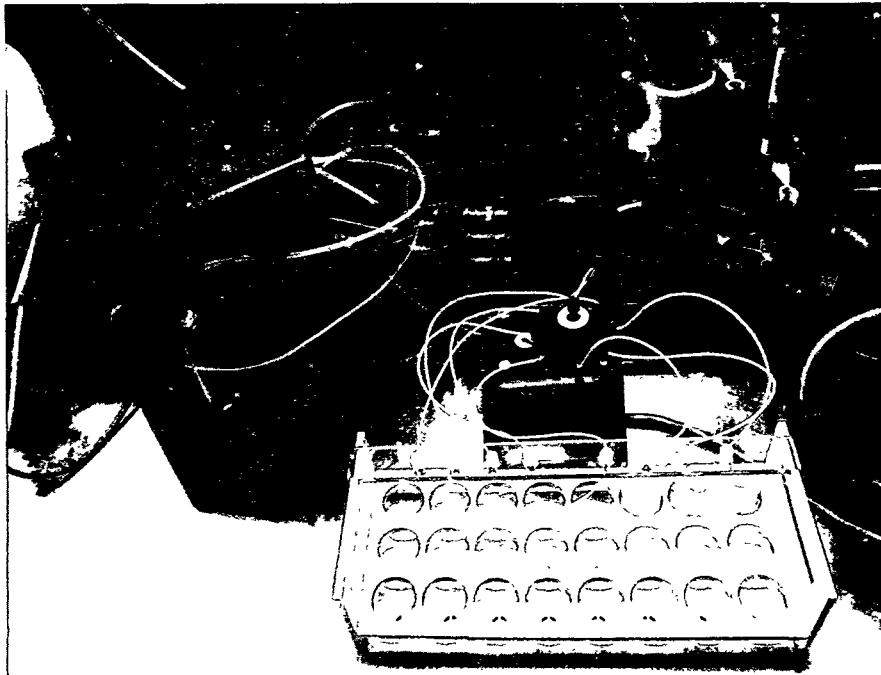


Fig. 23a. Ion chromatograph located inside glovebox.

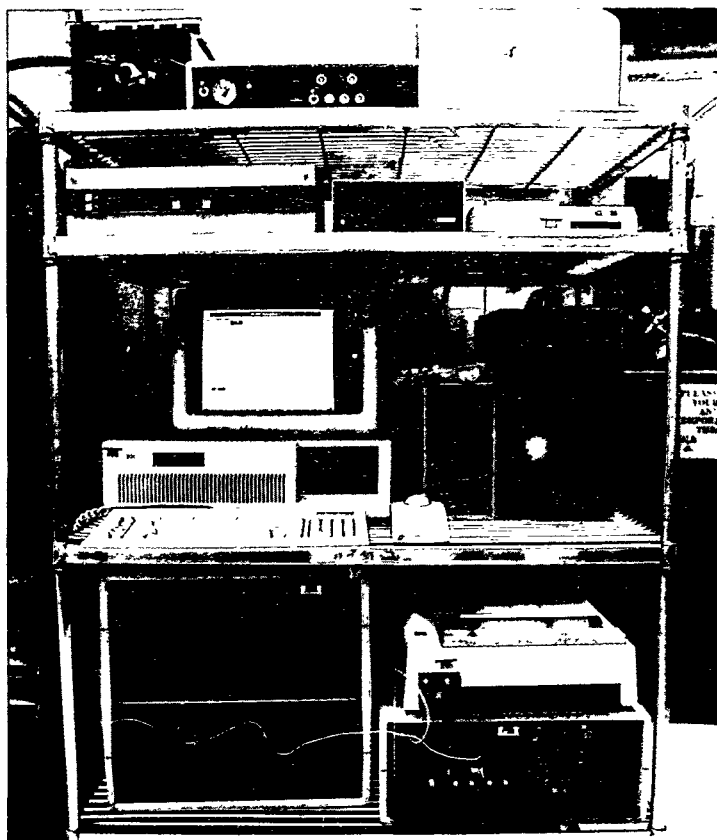


Fig. 23b. Computer-controlled ion chromatography instrumentation.

During the feed treatment step, visible spectroscopy will be used to determine the levels of Pu(VI). As working methods are developed, visible spectroscopy will also predict such values as the K_d (equilibrium distribution coefficient), the HNO_3 , the NO_3 , and the anion impurities.^{2,16,17,18} The visible spectra (Fig. 24), which are rich in information, can be used for these predictions.

Wash and Eluate Solution Preparation.

The reagents used for the wash and elution steps will need to meet operational requirements. Initially, the following parameters will be checked: acidity (using titration and/or acid sensor),¹⁷ fluoride (using ISE or IC), and hydroxylamine nitrate (using titration). These analytical methods will be available in ATLAS on-demand.

Effluent.

The effluent, while being the target of several development concepts, will not be heavily analyzed during the startup of ATLAS. Existing gamma monitor technology will be used to follow plutonium, uranium, and americium.¹¹ Recent developments in effluent analysis include using visible spectroscopy for plutonium analysis, on-line XRF

and/or off-line ICP-ES for impure metal analysis,¹⁴ and acid sensors.

Visible spectroscopy will provide information in real time on the plutonium that passes through the anion-exchange column in the effluent. From the resulting spectra, the oxidation state of the plutonium in solution can be measured to better understand plutonium losses in the effluent. The integrated quantities of the various forms of plutonium during the loading and washing steps will approximate the total plutonium in the effluent.

Trace metal analysis, such as ICP-ES or XRF, may help follow the cleanup of the plutonium by tracking the wash-off of the trace impurities. This analysis could lead to improved product purity and reduced wash volumes.

Eluate and Filtrate.

The eluate will be the target for both existing process analysis and process control development projects. Methods of controlling the major parameters (i. e., Pu[IV], Pu[III], and acidity) that are involved in the batch precipitation of Pu(III) oxalate are being developed.¹⁸ ATLAS will be one of the targets for the demonstration of these on-line analytical methods.

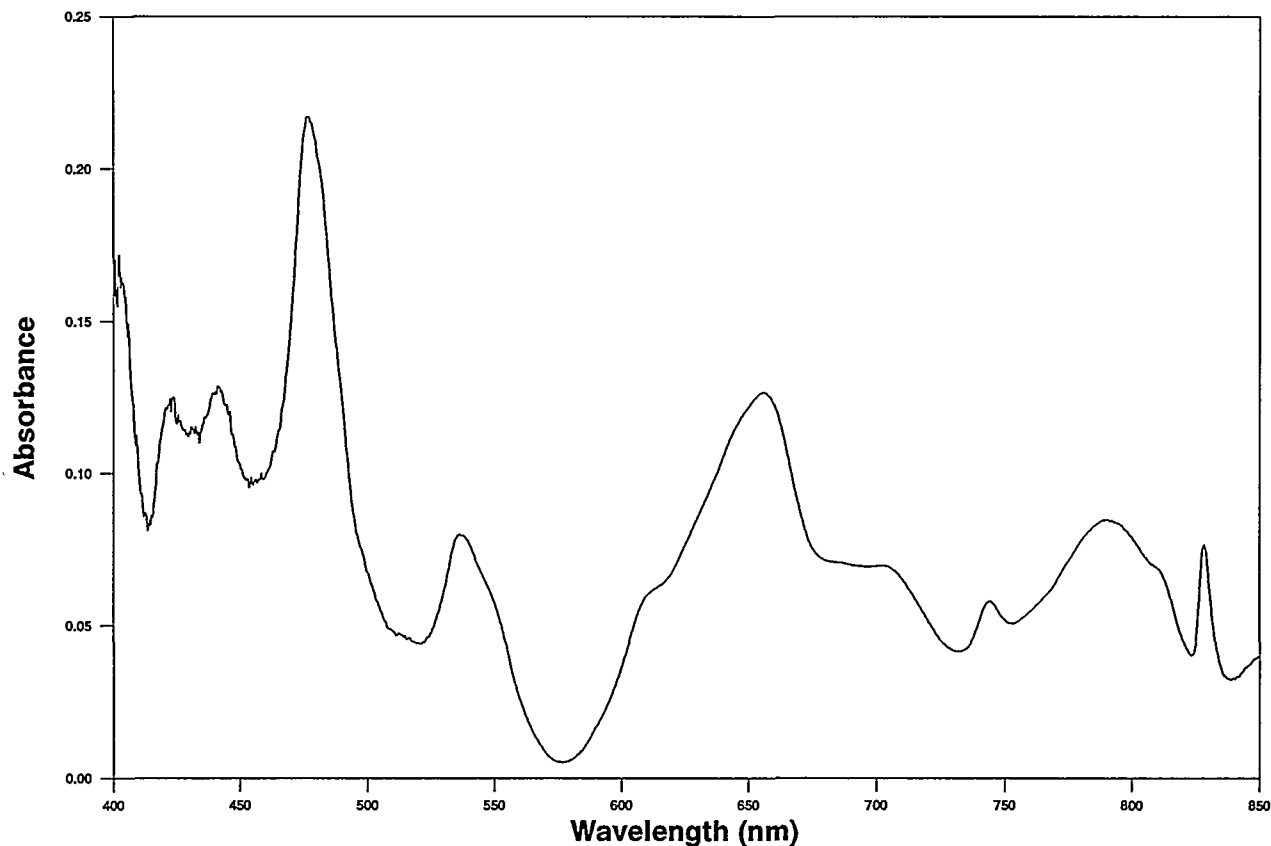


Fig. 24. Visible spectrum for plutonium nitrate solution.

When inductively coupled plasma mass spectroscopy capabilities are available at TA-55, the impurities in the eluate solutions will be able to be analyzed. This will be useful in predicting whether the plutonium oxide that results from the precipitation and calcination of the eluate will meet impurity specifications. This would reduce the effort that is currently expended in "reworking" impure oxide. Unfortunately, the detection limits of the currently available ICP-ES are not low enough to generate confidence in a prediction of the final oxide's purity. An impurity level of 1 ppm in solution could result in a level of ~30 ppm in the final oxide. This is too close to many of the specification limits to allow accurate prediction.

Finally, examination of the filtrates for oxalate (using IC) and for acid (using titration) will yield information about the oxalate precipitation process.

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