A REMOTELY OPERATED FACILITY FOR IN SITU SOLIDIFICATION OF FISSILE URANIUM

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ABSTRACT

A heavily shielded, remotely operated facility, located within the Radiochemical Processing Plant at Oak Ridge National Laboratory, has been designed and is being operated to convert ~1000 kg of fissile uranium (containing ~75% ^{235}U, ~10% ^{233}U, and ~140 ppm ^{232}U) from a nitrate solution (130 g of uranium per L) to a solid oxide form. This project, the Consolidated Edison Uranium Solidification Program (CEUSP), is being carried out in order to prepare a stable uranium form for long-term storage. The facility was constructed under restricted conditions, extensive process development studies and equipment modifications were made to enable successful operations, all startup requirements were met, and the equipment has been successfully operated through ~70% completion of the project.

This paper describes the solidification process selected, the equipment and facilities required, the experimental work performed to ensure successful operation, some problems that were solved, and the initial operations.

INTRODUCTION

A heavily shielded, remotely operated facility, located within the Radiochemical Processing Plant at Oak Ridge National Laboratory (ORNL), has been designed and is being operated to convert ~1000 kg of fissile uranium (containing ~75% ^{235}U, ~10% ^{233}U, and ~140 ppm ^{232}U) from a nitrate solution (130 g of uranium per L) to a solid oxide form. This project, the Consolidated Edison Uranium Solidification Program (CEUSP), is being carried out in order to prepare a stable uranium form for long-term storage. The uranium resulted from irradiation of a ThO_2-UO_2 fuel core in Consolidated Edison's Indian Point-1 Reactor during the early 1960s. The irradiated fuel was reprocessed in 1968 at the Nuclear Fuel Services plant in West Valley, New York, and the uranium nitrate product solution was sent to ORNL for storage. Soluble poisons (cadmium nitrate and gadolinium nitrate) were added to ensure subcriticality. After the solution had been stored for several years and no use for the uranium had become apparent, a decision was made to solidify the material for long-term, safe storage. This paper describes the solidification process selected, the equipment and facilities required, the experimental work performed to ensure successful operation, some problems that were solved, and the initial operations.

PROCESS DESCRIPTION

A simple process for the solidification via chemical and thermal decomposition of the nitrate was chosen, but the equipment and facilities were complicated by the following constraints:

1. The uranium contains 140 ppm ^{232}U. Its decay daughters are predominantly alpha emitters but one of them, ^{208}Tl, produces a 2.6 MeV gamma ray. Therefore, massive shielding and alpha containment were required.
2. The large amount of fissile uranium necessitated equipment designs that were geometrically favorable for subcriticality.
3. Available in-cell processing space was limited.
4. Intricate mechanical equipment was needed for remote operation and transport of the product storage cans.
5. The available facilities required direct, hands-on maintenance of the processing equipment.

A schematic diagram of the process flowsheet is shown in Fig. 1. The nitrate solution is processed in batches — each containing ~2.7 kg of uranium. The key process steps include (1) a batch evaporation to concentrate the uranium-cadmium-gadolinium nitrate solution to slightly less than the saturation concentration; (2) the use of formaldehyde in the evaporator to destroy nitric acid so that crystallization of the nitrate salts due to supersaturation will not occur; (3) thermal denitration to form the oxide in situ in the storage can; (4) remote welding of the can lid and installation of a crimp-sealed, secondary containment canister; and (5) transfer of the finished package to a shielded storage well.

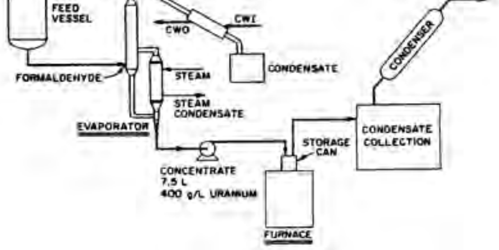

Fig. 1. Simplified Chemical CEUSP Flowsheet.

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The evaporation step was included in the process design to reduce the overall time requirement for the solidification process. In the evaporation, the feed solution is concentrated by a factor of 2 to 3. Evaporation can be done in a 2- to 3-h time period and is much faster than the thermal denitration that requires ~24 h. Thus, the facility equipment includes three thermal denitration systems, which are operated in a parallel mode, but only one evaporation system.

By including the evaporation step in the process design, the need for a destruction process for part of the nitric acid was incorporated. This destruction process was necessary because, with the desired concentration factor of 2 to 3 and without destroying some of the acid, a supersaturated concentrate solution would be produced.

EQUIPMENT AND PROCESS DEVELOPMENT

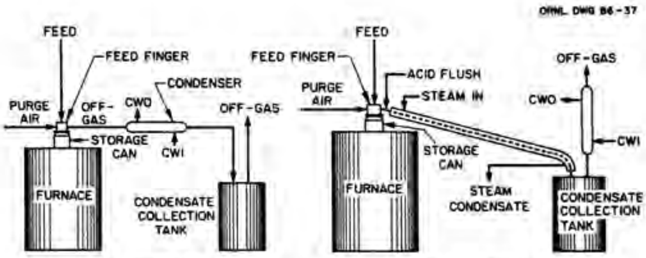
The original process flowsheet and the equipment designed for the thermal denitration step were based on a laboratory-scale study. In early tests of the full-scale equipment, numerous difficulties were encountered. A significant process development study was conducted to determine appropriate equipment modifications, to elucidate the process characteristics, and to select acceptable operating conditions. In addition, many of the mechanical components were given extensive reliability tests.

Evaporation/Acid Destruction Process Study

The evaporation/acid destruction step is carried out in a thermosyphon-type evaporator vessel that is operated semicontinuously to concentrate ~21-L batches of feed solution to a ~8-L volume. This operation is complex and difficult to control because of several factors which include (1) the turbulence of the recirculating solution in the thermosyphon leg, (2) the continuous and simultaneous addition of feed solution and formaldehyde solution, (3) the changing nitrate concentration in the evaporator and thus, the changing boiling temperature, (4) the exothermic reaction of formaldehyde and nitric acid, and (5) the continuous evolution of steam, CO_2, and nitrogen oxide gases.[1] In addition, the quantity of formaldehyde used must be related to the amount of nitric acid present. If insufficient formaldehyde is fed, then the destruction of nitric acid is incomplete, and a supersaturated concentrate will be produced; if excess formaldehyde is added, uranyl formate will precipitate. Thus, a one-quarter scale, glass evaporator was built and operated experimentally to observe the hydraulic characteristics of the process, to determine satisfactory operating conditions, and to develop correlations of operating parameters. A synthetic Consolidated Edison Uranium (CEU) solution, in which depleted uranium was used as a stand-in for the fissile uranium, was used in the experimental tests.

Denitration Equipment Modification and Process Study

The thermal denitration step is a semicontinuous process in which a batch of the concentrated CEU solution is fed into a can (which eventually becomes the primary storage container) and, as it is fed in, the solution is evaporated to dryness and the nitrate is decomposed, leaving a solid oxide cake in the can. This process minimizes solids handling problems. However, several serious processing problems, such as foaming and splattering inside the can and plugging of the off-gas line, can be encountered. These problems were observed in early tests of the full-scale equipment (illustrated in Fig. 2a) and required a significant process development study and equipment improvement program to circumvent difficulties.



(a) ORIGINAL DENITRATION DESIGN (b) CURRENT DENITRATION DESIGN

Fig. 2. Evolution of Denitration Equipment.

Equipment modifications (illustrated in Fig. 2b) that were made to minimize plugging problems at the feed entrance and in the off-gas line included redesign of (1) the nozzle that connects the feed and off-gas lines to the can, (2) the off-gas line, and (3) the condensate collection system. The process study described below included (1) a determination of the effects of the feed components on the behavior of the process, (2) a theoretical and experimental examination of conditions inside the can as it is filled, and (3) an interpretation of the effects of process variables (e.g., such as, feed method, feed rate, and time/temperature profiles in the furnace).[2]

Denitration Feed Pump Selection

The originally specified pump was a precision, close-tolerance gear pump. The gear pump failed to operate successfully because the gears seized due to a lack of lubricity in the nitrate solution. When this problem was discovered, a study was undertaken to find a suitable replacement pump. Based on literature searches and discussions with vendors and users, several types of pumps were obtained and tested using synthetic nitrate feed solution. Results of these studies are summarized in Table I.

The "Ramoy" progressive cavity pump, manufactured by the Robbins and Myers Pump Company, was judged to be the most acceptable for use in the CEUSP Facility. Although the pump is not a low-flow-rate metering pump, it was found to be reliable for rates as low as 5 mL/min. The pump has a stainless steel rotor and a Viton stator. Chemical attack of the Viton by either nitric acid or nitrites (which are produced in the evaporation/acid destruction step) and radiation

TABLE I

Comparison of Pumps Tested

Pump type	Advantages	Disadvantages
Zenith gear pump	• Accurate metering • Compact	• Gears seized in CEU solution • Very close tolerances
Lapp pulsafeeder Hydraulic check valve pump	• Frequently used in nuclear processes • Able to handle solids	• Sticking ball check valves • Size was too large for allowed equipment space
FMI rotating plunger pump	• Accurate metering • Compact, able to handle solids	• Seals leaked • Fragile
Ramoy progressive cavity pump	• Compact • Rugged, able to handle solids • Acceptable metering	• Rubber stator eventually fails in radiation and acid environment • Requires extensive testing to find matched rotor-stator combinations

exposure were found to cause the Viton to swell, loosening the fit between the rotor and the stator. Therefore, failure of the pumps during the course of the project was anticipated, and preparations for their replacement were made.

Piping Connector Corrosion

The stainless tubing and valves used in the CEUSP Facility were joined by means of compression tubing fittings because they allowed the replacement of faulty components and were not difficult to install during construction. Over 1000 compression fitting joints were installed behind the massive shielding in the CEUSP Facility. Most of these fittings were single-ferrule type which contained a nitrided sealing surface. These fittings developed leaks during preoperational testing of the facility.[3] The leaks were found to be due to acidic attack on the ferrule nitride surface. Figure 3 illustrates where the leaks occurred. Repair required disassembly of all shielding and lines, followed by replacement with either nonnitrided ferrule fittings or specially designed welded connectors. After the repair was completed, no further problems occurred.

Equipment Testing

Reliability of mechanical components was an important consideration because the constraints of the operating space were not conducive to easy equipment repair or replacement. Although the CEUSP Facility was scheduled to operate for only 1 year, the mechanical components were tested for a 10-year duty life. Several of the components did not withstand this severe testing; some examples and the corrective actions taken are described in Table II. More than 50 test procedures were prepared to test the fabricated equipment components: first, as individual pieces, then as subassemblies, and finally, in integrated preoperational tests.

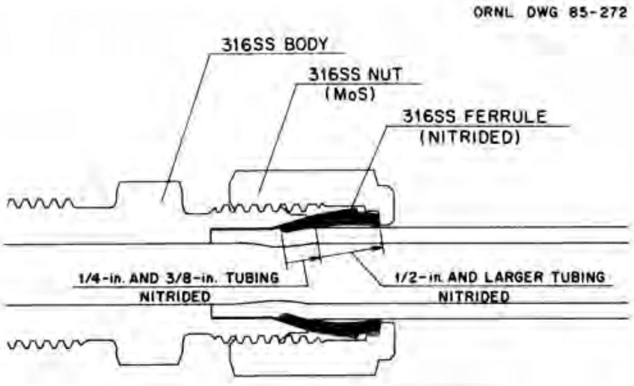


Fig. 3. CEUSP Parker Fittings.

STARTUP REQUIREMENTS

Preparations for startup of the CEUSP Facility were complicated by the extensive training efforts and the safety and quality assurance (QA) approvals that were required. Operating and maintenance procedures were developed, tested, and approved. Preoperational testing with synthetic feed provided final equipment checkout and hands-on training for the operating crew. Each three-shift operating crew consists of an operations supervisor and two technicians. These personnel underwent classroom training that involved certification testing. The extensive training required a full-time specialist to coordinate the training program.

Safety analysis and QA documents were prepared and reviewed by committees from ORNL, DOE-Oak Ridge, the DOE Program Office, and the Los Alamos National Laboratory. The latter group was a Safety Analysis Review Team, under contract to the DOE Program Office. Operational readiness reviews included observation of operations by the reviewers and a thorough audit of training records and procedures. Final approval to operate was given in April 1985.

OPERATION

Approximately 70% of the feed material has been successfully processed during the first 9 months of operation, as illustrated in Fig. 4. The downtime periods in July, October, and December were required to change failed feed pumps. As indicated in Fig. 4, the designed processing rate has been maintained. No significant difficulties have been encountered. Uranium losses in the waste condensate solutions have been <0.3%. Operations should be completed before the scheduled date (July 1986).

TABLE II

EQUIPMENT FAILURES DUE TO LIFE CYCLE TESTING

Item	Purpose	Failure mode	Corrective action
Ball bearings	Allow remotely operated carriage to move	Decontamination solution induced corrosion	Replaced open, carbon steel with sealed, stainless steel bearings
Vacuum hoses	Provide pick-up force to handle remote cans	Stainless steel metal bellows hose cracked due to work hardening	Replaced with reinforced rubber hose
Remote valves	Seal off system and provide utilities	Leak through seats	Investigated several types before selection
Gear pump	Meter concentrate at 9 mL/min	Gear seized due to improper lubrication	Piloted several pumps, selected progressive cavity
Compression fittings	Couple tubing to allow good seal and easy equipment replacement	Corrosion on nitride surface of ferrule	Replaced with another brand using non-nitrided ferrules

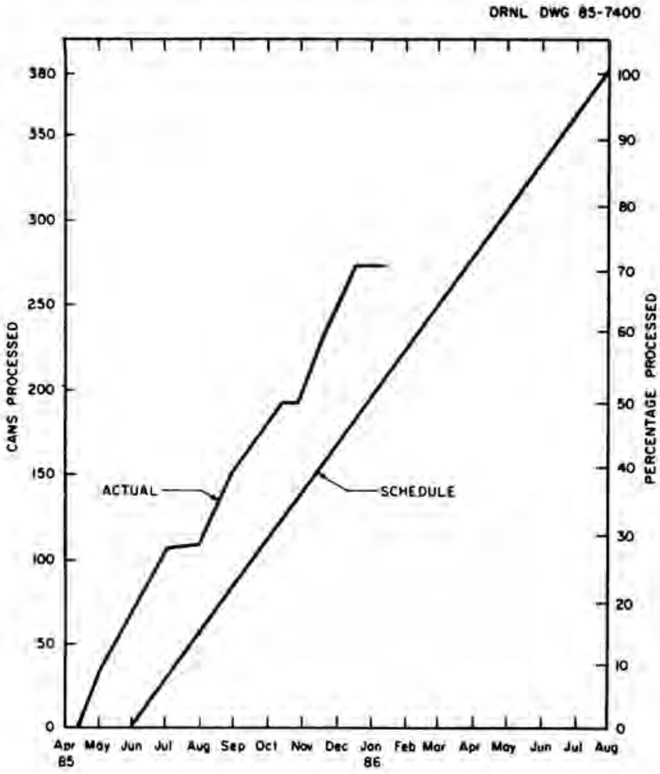


Fig. 4. CEUSP Production Rate Over the Life of the Project.

SUMMARY

In summary, a remotely operated facility has been built and operated at Oak Ridge National Laboratory to convert a highly fissile and radioactive solution to a stable solid. The facility was constructed under restricted conditions, extensive process development studies and equipment modifications were made to enable successful operations, all startup requirements were met, and the equipment has been successfully operated through ~70% completion of the project.

REFERENCES

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