

# ON THE DEVELOPMENT OF A DISTILLATION PROCESS FOR THE ELECTROMETALLURGICAL TREATMENT OF IRRADIATED SPENT NUCLEAR FUEL

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As part of the spent fuel treatment program at the Idaho National Laboratory, a vacuum distillation process is being employed for the recovery of actinide products following an electrorefining process. Separation of the actinide products from a molten salt electrolyte and cadmium is achieved by a batch operation called cathode processing. A cathode processor has been designed and developed to efficiently remove the process chemicals and consolidate the actinide products for further processing. This paper describes the fundamentals of cathode processing, the evolution of the equipment design, the operation and efficiency of the equipment, and recent developments at the cathode processor. In addition, challenges encountered during the processing of irradiated spent nuclear fuel in the cathode processor will be discussed.

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**KEYWORDS :** Vacuum Distillation, Actinide Recovery, Electrometallurgical Treatment, Pyroprocessing

## 1. INTRODUCTION

Actinide products are being recovered from Experimental Breeder Reactor-II (EBR-II) spent nuclear fuel by an electrometallurgical treatment process at the Idaho National Laboratory (INL). The treatment process comprises a set of operations designed to recover actinides from radioactive fission products and place the fission products into acceptable waste forms [1-3]. Two distinct products are recovered during spent fuel treatment; a low-enriched uranium product and an actinide product. The low enriched uranium products are currently being stored pending a disposition decision while the actinide products are returned to the process to enable further testing [4-5]. Treatment of the EBR-II spent fuel was initiated in June 1996 and continues currently on the balance of the spent fuels.

Batch operations performed on the spent fuel in the Fuel Conditioning Facility (FCF) include chopping, electrorefining, cathode processing, and casting. The FCF is an atmosphere-controlled, shielded hot cell facility where all operations are performed remotely by overhead cranes, electromechanical manipulators, or manual manipulators. Spent fuel is either dismantled into elements from assemblies or retrieved directly as elements from storage containers. Initial fuel element handling operations are performed in the air cell of FCF prior to their transfer to the argon cell for element chopping. Elements are chopped into segments by either a solenoid-driven or pneumatic press depending on the

fuel type, driver or blanket, respectively. Table 1 contrasts the two EBR-II fuel types. Driver fuel was utilized in the core region of the reactor and contains a highly enriched uranium-zirconium alloy. Blanket fuel, consisting of depleted uranium, encircled the driver-core.

The segments are then loaded into an anode assembly which contains up to nine perforated baskets depending on the fuel type. The anode assembly is installed into the electrorefiner commensurate with the fuel type; Mk-IV for driver and Mk-V for blanket. Although the two electrorefiner vessels are identical in design, the electrode configurations, liquid contents, and process conditions are different. During electrorefining [5-7] in both units, fuel is anodically dissolved in a LiCl-KCl eutectic salt such that the transport of actinides and deposition as a cathode is feasible. In addition to the LiCl-KCl, other chloride species are present in the electrorefiner salt due to the oxidation of fission products, bond sodium, and actinides from the spent fuel. For solid cathodes, uranium is transported, deposited on a steel cathode, and collected for further processing in a vacuum distillation furnace. The uranium or solid cathodes contain uranium, chloride salt, and a small amount of cadmium if the cathode products are from the Mk-IV electrorefiner. A cadmium pool is present in the Mk-IV electrorefiner to recover cathodic material via a deposition operation. The Mk-V electrorefiner does not have cadmium due to a different anode to cathode arrangement [2]. Actinide products consisting primarily of plutonium and uranium

**Table 1.** EBR-II Spent Fuel Compositions (wt. %)

Constituent	Driver	Blanket
Uranium	81.3	98.4
Zirconium	9.7	0.0
Sodium	2.1	0.6
Fission Products	6.4	0.1
Transuranics	0.5	0.9

are produced by utilizing a liquid cadmium cathode for electrorefining and then removing cadmium by distillation [5, 8]. Distillation operations for both solid and liquid cathodes are performed in the same piece of equipment, the cathode processor, to produce consolidated metal ingots. A casting step following distillation allows for isotopic dilution, if necessary, and sampling of the ingots for chemical analyses.

The context of this paper is to provide a compilation of the research and development activities involving the separation of actinide products from both salts and cadmium during the processing of cathodes following electrorefining. Initially, a brief explanation on the fundamentals of distillation will be presented followed by a description of

the cathode processing equipment and operations. The degree of salt separation from uranium, via the use of representative isotopic tracers of the fission product chlorides, and cadmium from actinides will be specified for cathode processor operations to date. In addition, recent research on the development of crucible materials for cathode processing will be presented along with other challenges encountered.

## 2. DISTILLATION

Shown in Fig. 1 are the vapor pressure curves for cadmium, the chloride salts, plutonium, and uranium [9]. Given the relevant differences in vapor pressures, the separation of salts and cadmium from actinides by distillation is easily affected. A breakdown of the major chloride salt species involved with cathode processing is given in Fig. 2 [9]. Note that barium and samarium chloride have the lowest vapor pressures and are the most difficult chloride species to remove while cesium chloride has the highest vapor pressure and thus, would be easily removed. It should also be noted that Fig. 2 represents the vapor pressures of pure substances. Multi-component chloride systems, e.g. binary, ternary, and possibly quaternary, would be considerably more complex and have distinctly different vapor pressures due to miscibility gaps.

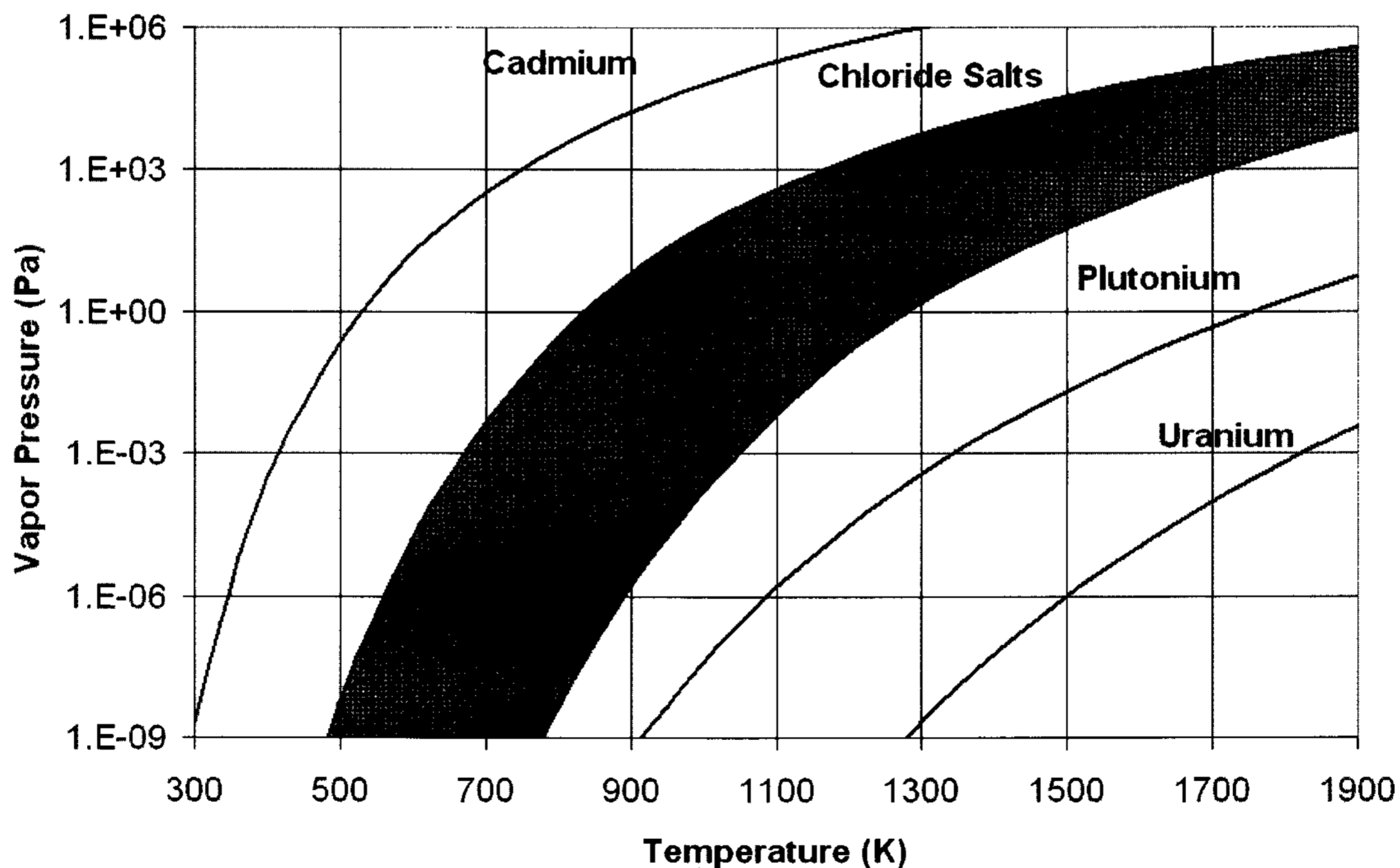


Fig. 1. Vapor Pressure Curves for Cadmium, Chloride Salts, Plutonium, and Uranium

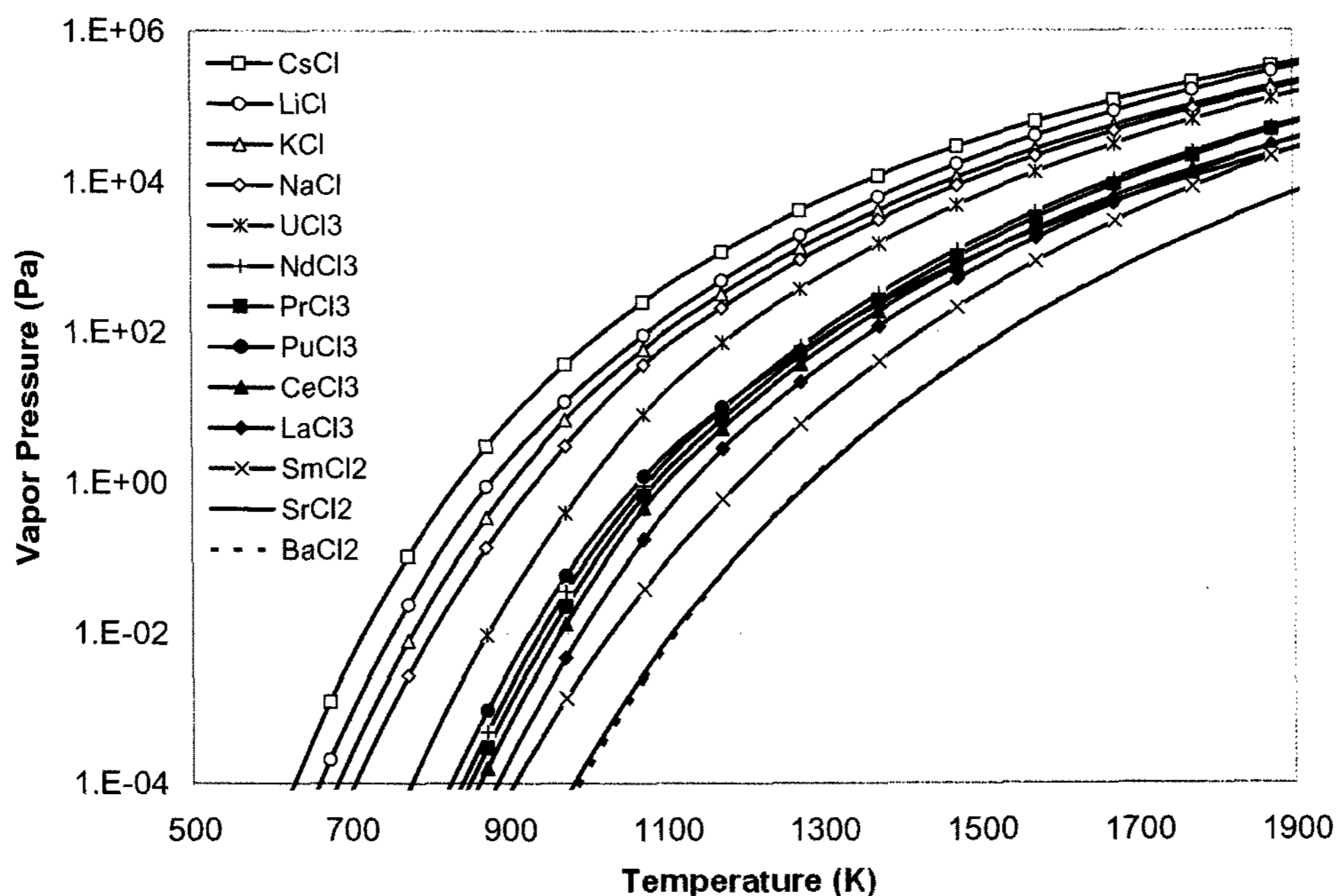


Fig. 2. Vapor Pressure Curves for Chloride Species Involved with Cathode Processing

### 3. EQUIPMENT

#### 3.1 Description

Development of a distillation process for spent fuel treatment can be traced to efforts at the Argonne National Laboratory (ANL) associated with the original EBR-II fuel cycle [10-11]. For this process, the uranium was separated from a zinc-magnesium solution by a vacuum retort furnace. The evolution of the current design of the cathode processor is based on this research. Two identical cathode processor units were originally put into operation; a prototype unit for development activities at the Illinois site of ANL and a production unit in the FCF at the Idaho site of then ANL, now INL.

Fig. 3 shows a layout of the cathode processor equipment. The crucible assembly (splatter shield, process crucible, radiation shield, condenser, and receiver crucible) is bottom loaded into the cathode processor vessel by translating the assembly to below the vessel and raising the assembly until seated. For ease of operation, the cathode processor vessel is elevated above the operating corridor. Horizontal motion of the crucible assembly is controlled by means of a trolley and rail assembly. With this configuration, the crucible assembly can be removed, dismantled, loaded or unloaded, and cleaned without disturbing the furnace components.

The furnace region of the cathode processor is heated by a passively cooled induction coil that suscept to the

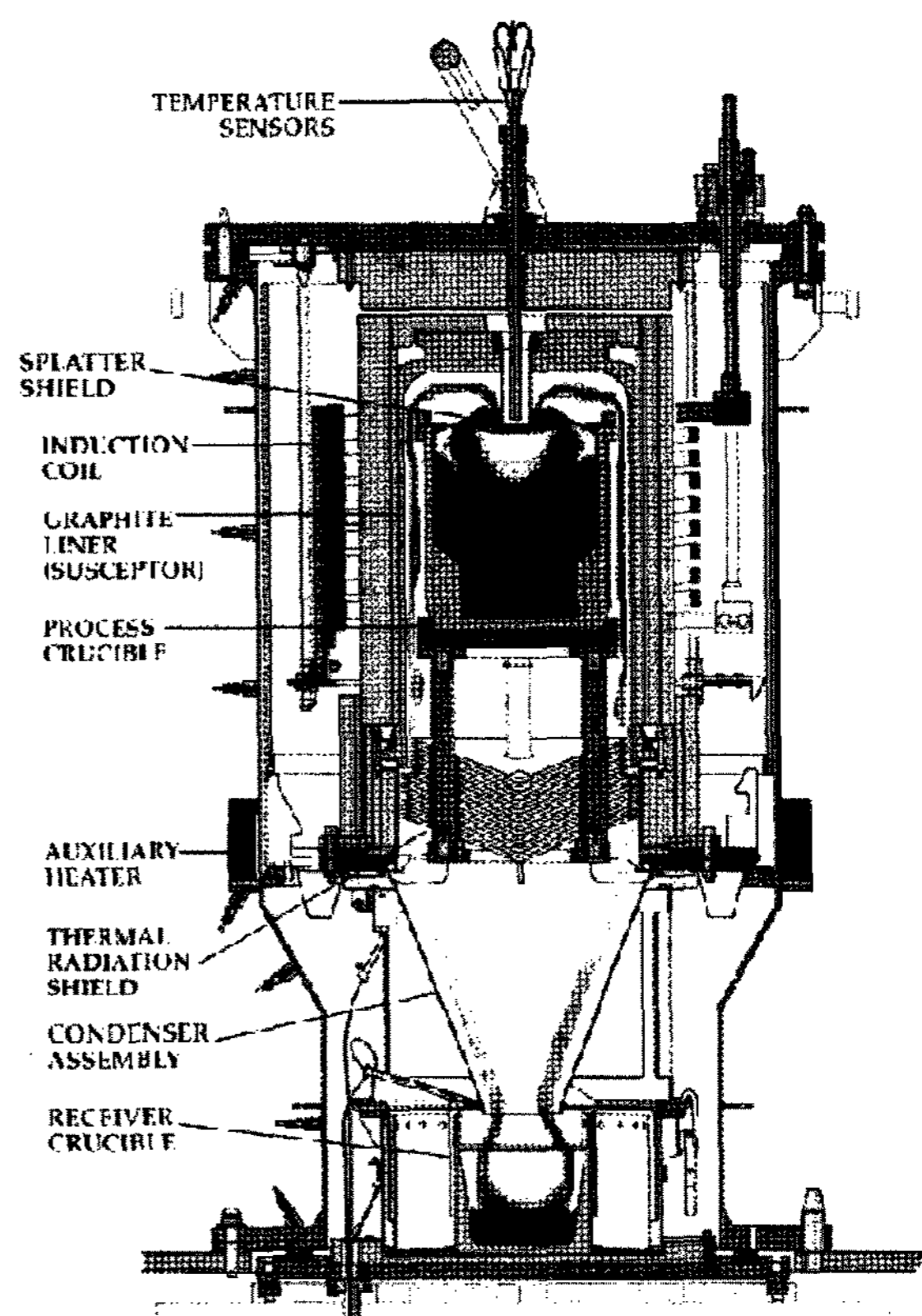


Fig. 3. Schematic of Cathode Processor Equipment

graphite furnace liner which, in turn, heats the process crucible by radiation. The process crucible consists of a graphite mold coated by a zirconia wash that prevents contact of the process materials with the graphite and insures ingot release. The induction heating power supply is rated to 30 kW output at 2500 Hz, although only 10-15 kW is utilized for normal operations. Isolation of the induction coil from the crucible assembly is necessary to prevent the migration of vapor species to the coil during a run. Distillate material is transferred to the condenser region from the process crucible in the vapor phase and collected in a receiver crucible. The crucible assembly constitutes the inner confinement chamber for volatile species. A vacuum system is connected to the condenser region and consists of two oil-free mechanical pumps, either operated in parallel or independently, that are connected to the vessel through an assembly of piping, manifolds, and valves. A thorough description of the cathode processor equipment is given elsewhere [12].

### 3.2. Operations

Operational testing of the prototype cathode processor in an atmosphere-controlled glovebox enclosure commenced in January 1995, slightly in advance of the production unit to allow for potential design modifications. Since one of the key design requirements of the cathode processor was to process both solid and liquid cathodes, initial development activities in the prototype focused on optimizing the process cycle for both cathodes. Slight modifications were necessary initially for the condenser and induction heating regions

to achieve the desired process goals [13]. The condenser assembly was redesigned to include new seals and more efficient collection of distillate material. For the induction region, additional insulation and larger radiation shields were incorporated to improve the thermal performance. Subsequent research endeavors in the prototype unit have included crucible material testing and other high-temperature support activities [12-13]. The prototype cathode processor is currently on standby status at ANL.

Testing with the production unit in FCF was initiated in April 1995 following development of process cycles in the prototype. Initially, operations in the FCF cathode processor were performed with simulated materials consisting of depleted uranium since the processing of spent EBR-II fuel did not commence until June 1996 [13]. In August 1996, the first batch of irradiated solid cathodes from the Mk-IV electrorefiner was processed in the production unit [14]. To date, over 250 heats have been performed in the FCF cathode processor on solid and liquid cathode products from the Mk-IV and Mk-V electrorefiners.

As designed, the cathode processor is capable of temperatures as high as 1673°K and pressures less than 10 Pa. For typical distillations, it is only heated to 1473°K at less than 100 Pa to achieve sufficient separation of distillate from the actinide products. Shown in Figs. 4 and 5 are typical temperature versus time plots for both the crucible and condenser regions during solid and liquid cathode operations, respectively. A more detailed discussion on the derivation of the operating parameters and the specific phases involved for both solid and liquid cathode processing has been

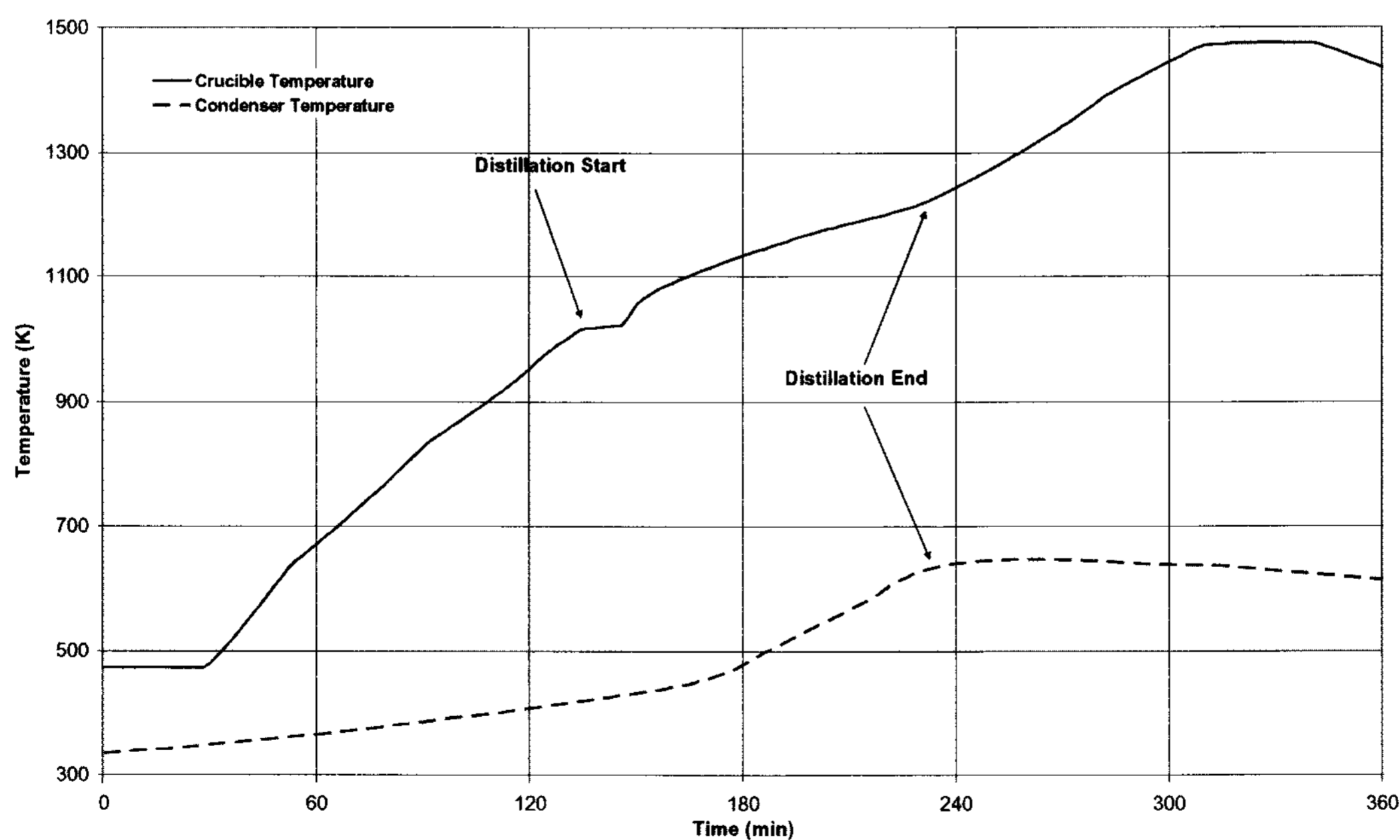


Fig. 4. Typical Crucible and Condenser Temperatures for Solid Cathode Operations

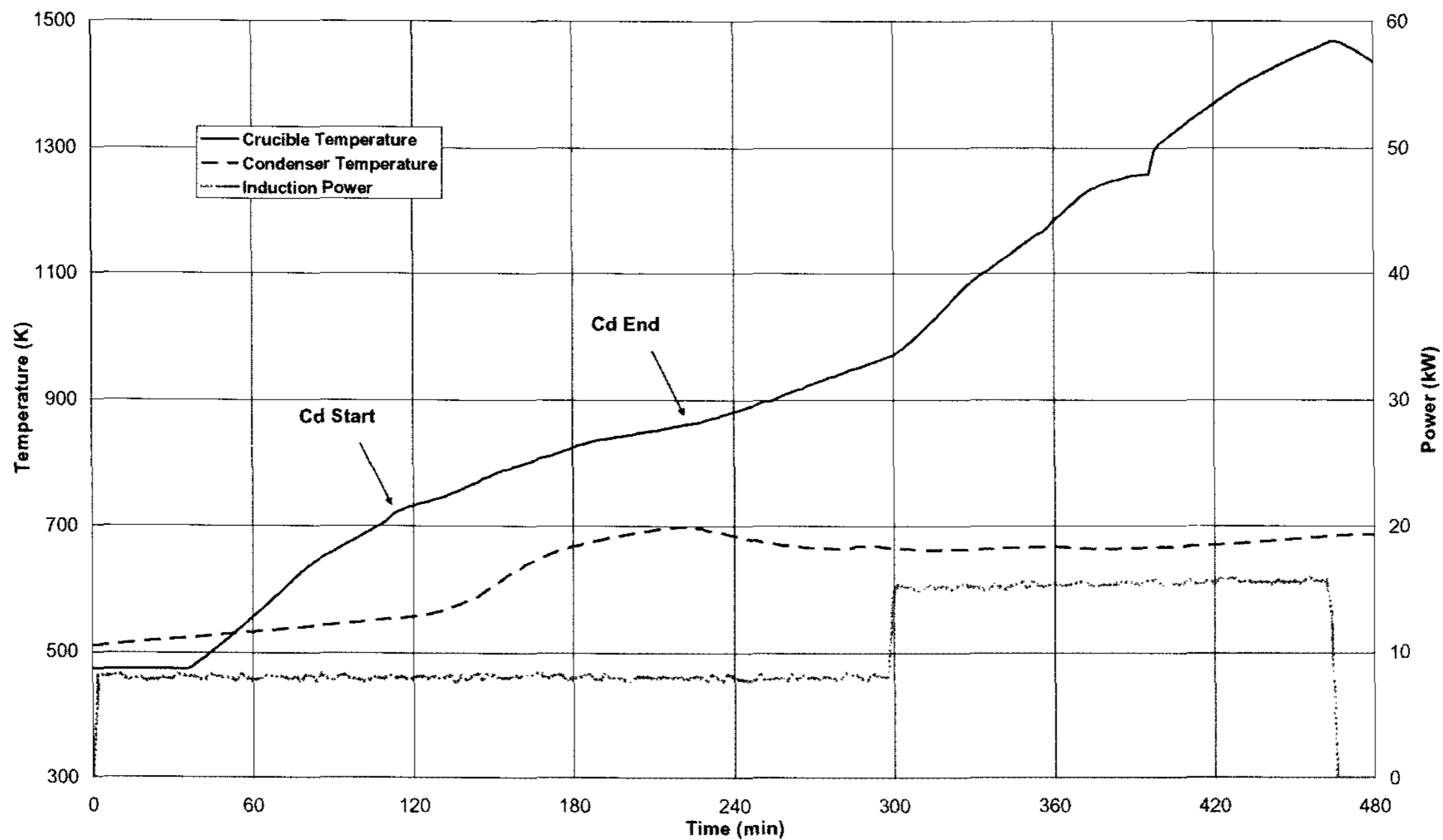


Fig. 5. Typical Crucible and Condenser Temperatures for Liquid Cathode Operations

documented [8, 12, 15].

Standard solid cathode batch sizes in the cathode processor range from 20-50 kg depending on the fuel type, driver or blanket, respectively. The driver batch sizes are limited by criticality concerns due to the higher enriched uranium while the blanket batch sizes are constrained by crucible volume. Modifications to the cathode processor equipment for increased throughput, particularly for blanket solid cathodes, have been implemented to accommodate larger batches [16]. The process crucible volume was increased 50% to give a batch size of approximately 80 kg for blanket products. For reference, the typical salt fraction for solid cathode products from both electrorefiners has been 20 wt.%. Liquid cathodes have only been produced in the Mk-V electrorefiner and their batch sizes consist typically of 26 kg cadmium and approximately 1-2 kg

actinide metal [5, 8].

As part of the modifications to the cathode processor equipment for increased throughput, a new induction heating coil was designed and installed in both cathode processor units to increase the heating performance. The redesign of the induction coil included three additional coil turns and a reduced insulation package allowing for a smaller coil diameter, see Table 2. As expected, the power output for the new coil increased by approximately 25% to 15 kW resulting in the batch run times decreasing by at least 20%.

Another benefit of the new coil has been a significant increase in distillation rates for salt due to the increased power output [17]. The quantity of salt distilled is calculated from the change in mass of the process crucible before and after a cathode processor run. Distillation time is determined

Table 2. Comparison of Old and New Induction Coil

Parameter	New Induction Coil	Old Induction Coil
Number of Turns	12	9
Coil ID (cm)	53	58
Power Output (kW)	15	12
Batch Run Time (hr)	7	9
Salt Distillation Rate (g/min)	120	73
Cadmium Distillation Rate (g/min)	243	-

from inflections on the crucible and condenser temperature curves (Fig. 4). Shown in Table 2 are averaged salt distillation rates for the new and old induction coils. For cadmium (Fig. 5), the averaged rate data from four experiments is shown in Table 2 for the new coil since liquid cathodes have only recently been processed [8]. Compared to salt distillation, the operating pressure for cadmium was slightly higher (400 Pa) in order to control the rate.

### 3.3 Efficiencies

The degree of salt and cadmium separation in the cathode processor has been determined for both solid and liquid cathodes. For salt, representative isotopic tracers of the fission product chlorides have been utilized to quantify the efficiency of salt separation [18]. Cadmium separation from the actinide products has been established by sampling of the molten actinide products and subsequent chemical analyses [8].

Since the concentrations of salt constituents in the uranium product are below the detection capabilities of standard analytical techniques, gamma-ray spectroscopy has been employed for the detection of concentrations into the trace quantity regime. Gamma-ray spectroscopy not only has the advantage of being able to detect several radioactive species simultaneously but also of detecting those species to extremely sensitive levels. It is assumed that the chemical behavior of the radioactive tracer is the same as that of the element or compound it represents. Of the radioactive nuclides routinely detected by gamma spectroscopy in the uranium product samples, the isotopes of cesium (Cs-137), cerium (Ce-144), and europium (Eu-155) are of particular interest since they represent the salt species of cesium chloride (CsCl), cerium trichloride (CeCl<sub>3</sub>), and europium dichloride (EuCl<sub>2</sub>) and are consistently detected in the uranium products.

Distillation efficiency is defined as the degree of salt separation from uranium in the cathode processor. Knowing the amount of radioactive isotope or tracer in both the product and feed of the cathode processor, the distillation efficiency can be calculated according to:

$$\text{Distillation Efficiency (\%)} = [1 - (\text{Product/Feed})] * 100 \quad (1)$$

Measurement of the tracer in the feed is performed by multiplying the amount of salt adhering to the cathode product, found after cathode processing, by the concentration of the tracer in the Mk-IV electrorefiner salt sample taken after a cathode product. The activity of the tracer in the product is quantified from the sample taken after casting. Both are adjusted for radioactive decay of the isotopes based on the cathode processor operating date.

The distillation efficiencies for CsCl (Cs-137), CeCl<sub>3</sub> (Ce-144), and EuCl<sub>2</sub> (Eu-155) have been calculated and averaged for over twenty batches of solid cathode products

from the Mk-IV electrorefiner. The averaged distillation efficiencies for CsCl, CeCl<sub>3</sub>, and EuCl<sub>2</sub> are 99.96%, 99.86%, and 98.61%, respectively. Assuming that the distillation efficiencies are related to the vapor pressures of the representative chloride species, Henry's law was applied since the concentrations of the chloride species in the Mk-IV electrorefiner were all less than 1 wt. %. Henry's law states that the partial pressure or vapor pressure of the species is proportional to its mole fraction for dilute solutions. Shown in Fig. 6 are the distillation efficiencies against mole fraction of the chloride species. A correlation is evident and represented by the regressed line.

Other minor chloride species of the system have been modeled based on their concentrations in the Mk-IV electrorefiner and are shown in Fig. 7. Applying the distillation efficiencies of the minor chloride species in Fig. 7, the concentrations of the elements in the uranium product can be calculated. Currently, less than 10 ppm total of chloride species would be expected in the uranium products based on the distillation efficiencies shown in Fig. 7.

To date, four cadmium distillation experiments have been performed with irradiated liquid cathode products from the Mk-V electrorefiner. Following these tests, the consolidated actinide ingots were taken to the casting furnace where molten samples were cast and sectioned for chemical analyses. For all four tests, the amount of cadmium detected by ICP-OES was less than the detection limits of the technique, typically ~35 ppm.

### 3.4 Maintenance

As mentioned, over 250 heats have been performed in the FCF cathode processor over the past 12 years. During this time, several maintenance periods have been necessary to keep the equipment operational. The primary cause for these outages has been the migration of distillate material outside the inner containment chamber. Although unintended, this loss of containment is not surprising considering the sealing requirements at high temperatures for the internal components and the nature of the distilling vapor species. Two distinct events will be discussed to highlight the challenges encountered with distillate migration.

The first event occurred during a pressure transient in an attempt to selectively distill certain chloride species. During the run, the operating pressure of the cathode processor was taken from ambient to vacuum conditions in a short period of time causing the flash distillation of salt. Considering the quantity (>9 kg) of salt involved, a significant amount of salt was forced through the vapor seals of the inner containment boundary causing instantaneous shorting of the induction coil. Ceramic components of the crucible assembly were also damaged by the sudden thermal gradient caused by the salt distillation. Shown in Fig. 8 is a photograph of the induction coil and support structure following the experiment. Note the partially melted bolt heads and

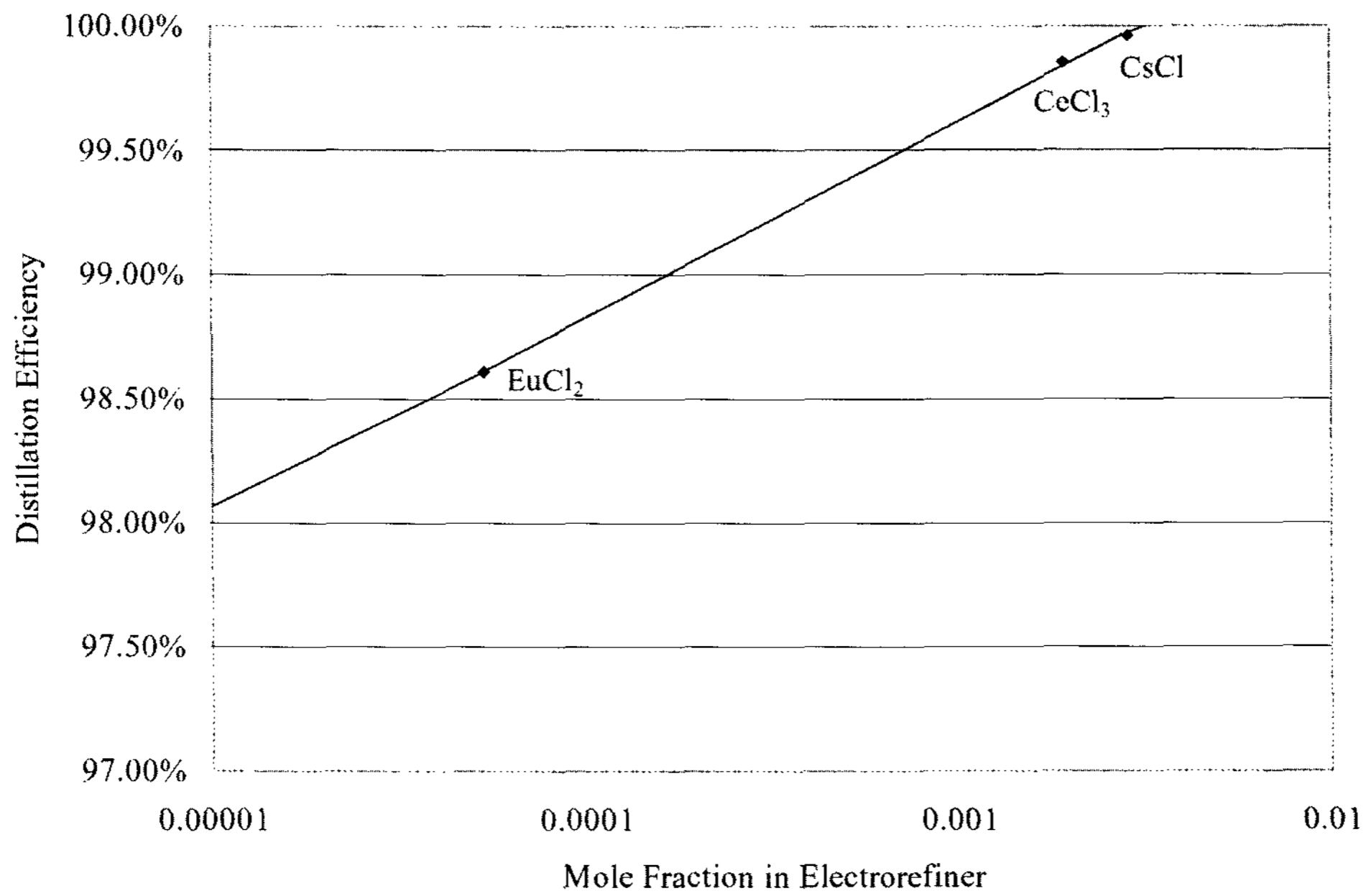


Fig. 6. Distillation Efficiencies Versus Mole Fraction of EuCl<sub>2</sub>, CeCl<sub>3</sub>, and CsCl

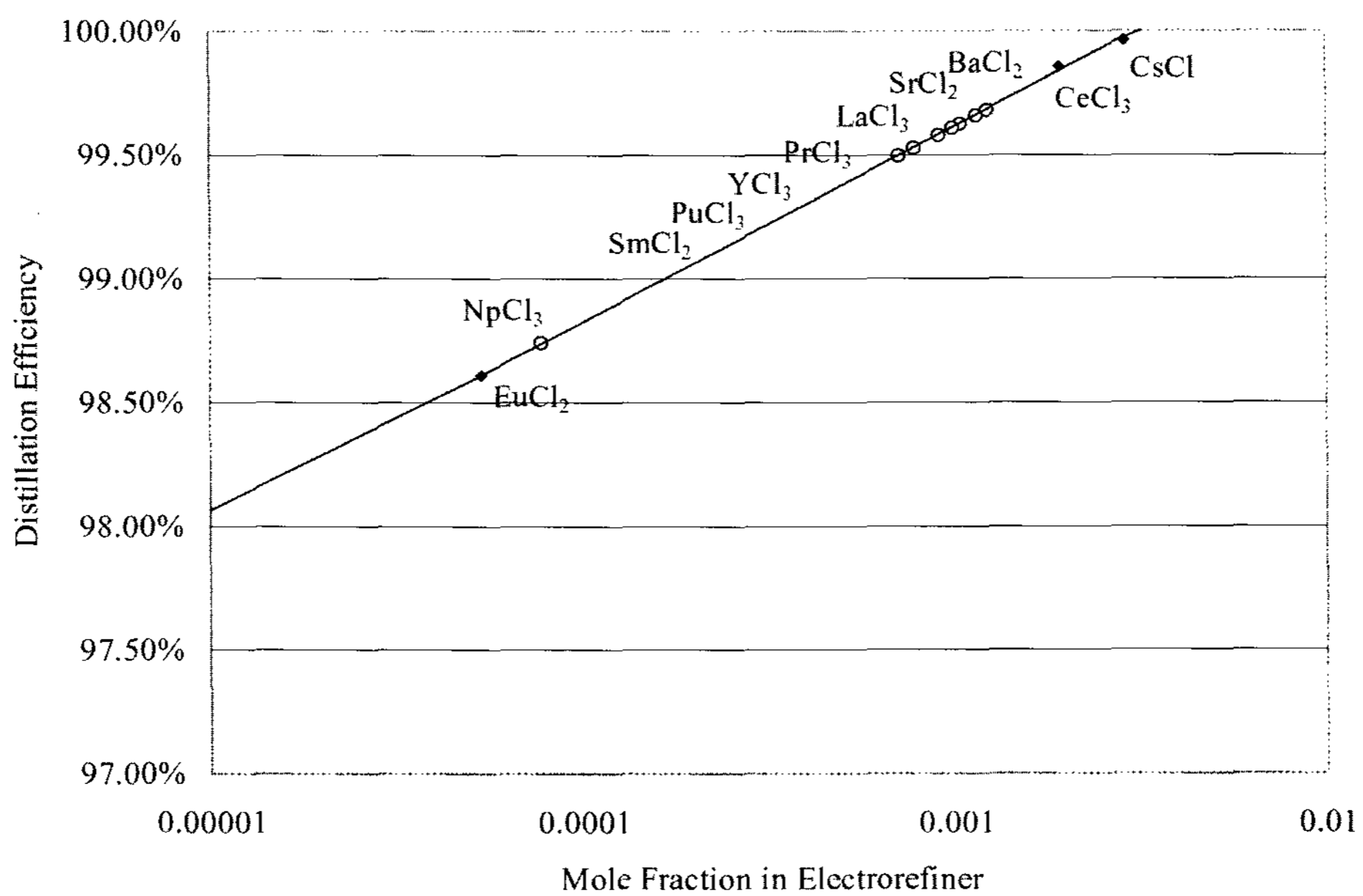


Fig. 7. Distillation Efficiencies Versus Mole Fraction of Minor Chloride Species

broken ceramic support pieces of the coil. Although unanticipated, the fragility of the cathode processor was recognized, particularly with respect to uncontrolled distillation.

More recently, the intermittent shorting of the coil has been causing problems in the completion of normal operations. During the latter portions of the process

cycle, power failures occur which make it difficult to maintain sufficient temperatures for the distillation and consolidation portions of the run. Several inspections have been performed including dismantling of the equipment for the detection of salt in the coil region, electrical troubleshooting from the power supply to the in-cell equipment, and salt cleanout operations to reduce

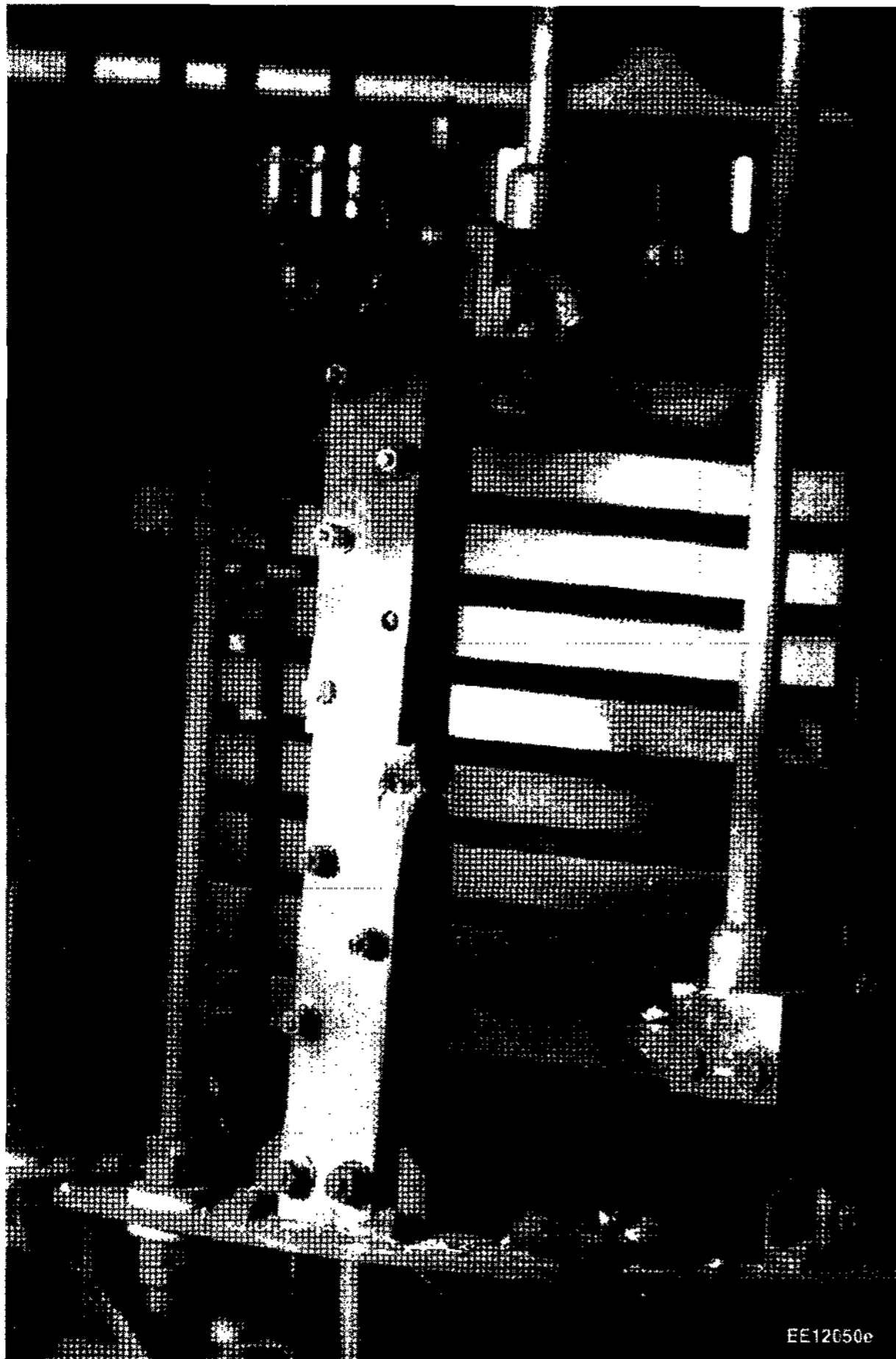


Fig. 8. FCF Cathode Processor Induction Coil Following Pressure Transient Experiment

the amount of salt held-up on the components of the crucible assembly. A certain amount of salt is held-up every run as detected by mass balance between the process and receiver crucibles. Periodic cleanouts are performed to retrieve this salt, although a complete balance is still not achieved. None of these activities resulted in a conclusive cause for the intermittent power failures, so the induction coil, the insulation package, and the graphite liner were all replaced. Only minimal testing has been performed since these changes for which shorting has not recurred. Two possible explanations exist for the shorting; a buildup of salt in the graphite furnace liner and/or cadmium migration due to the recent processing of liquid cathodes. Intercalation of chloride species into graphite is a well known phenomenon [19-20] and has been confirmed by sampling and chemical analyses of the graphite components. Speculatively, the salts would then be released as the liner reaches a certain temperature, i.e. towards the end of the run, and short the

coil. On the other hand, the accumulation of cadmium, with its higher vapor pressure, outside the inner boundary may be the cause.

Other issues with the FCF cathode processor equipment to date have included broken thermocouple wells and the inability to achieve design temperatures. For the thermocouple wells which are also graphite, several failures have occurred due to either dimensional tolerancing or perhaps degradation from chloride intercalation. The gap between the process crucible splatter shield and the graphite well has been adjusted but may still cause interference if the splatter shield is not in place correctly. Additionally, materials other than graphite will be tested in place of the current thermocouple wells. Early in the development program, it was recognized that the old coil was undersized for the anticipated high-throughput requirements [12]. As such, difficulties were gradually encountered with the old coil in achieving the normal operating temperature of 1200°C. It was hypothesized at the time that thermal degradation of the insulation and/or the graphite liner was causing the difficulties in heating [21]. Based on the recent shorting problems and salt misbalance, it would appear that chloride intercalation was a factor in the earlier heating problems. No problems have been experienced to date with the new coil achieving the design temperatures, perhaps due to its ability to overcome the degradation.

## 4. RECENT DEVELOPMENTS

### 4.1 Plutonium Reduction

Disposition of the blanket uranium products will most likely be dependent on the plutonium concentration levels associated with the final ingots. At present, the blanket products would not meet the Low Level Waste (LLW) requirements with respect to their plutonium content. In an effort to provide more options for disposition, an understanding of the plutonium levels in the blanket products was desired so that the plutonium levels could either be reduced or controlled.

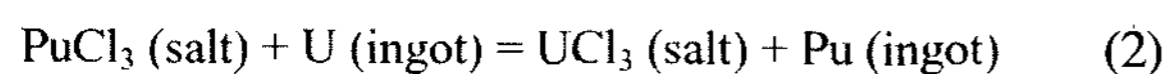
Since two primary pieces of equipment (electrorefiner and cathode processor) are being utilized for the treatment of spent fuel, the first step in the study was to distinguish where the plutonium was originating [22]. Two experiments were performed in the cathode processor with salt containing plutonium, and uranium that was plutonium-free. The results of these tests indicated that the electrorefiner was not the source, but rather the cathode processor.

The next step was to establish the chemical state of plutonium in the cathode processor product. Various experiments were performed in the cathode processor to determine if the plutonium contamination of the uranium products was as a chloride, an oxide, or a metal. Although other species are possible, their presence was not considered likely. For the chloride species, tests were performed at higher than normal temperatures and longer



processing times to assure distillation of the plutonium trichloride species. For the oxide species, tests were performed with different crucible coating materials as well as a sacrificial liner to prevent the reaction of plutonium trichloride with zirconia. No significant changes in the plutonium levels were recognized for any of these tests.

The results of these tests point to metallic plutonium via the following reaction:



The reaction is driven to the right by the distillation of uranium trichloride with respect to plutonium trichloride leaving metallic plutonium in the uranium ingot. Plutonium trichloride is approximately a factor of ten less volatile than uranium trichloride at distillation temperatures in the cathode processor [9].

In order to control and/or reduce the plutonium in the blanket uranium products, the molar ratio of uranium trichloride to plutonium trichloride must be influenced during the cathode processor operation as shown by the following thermodynamic relationship:

$$e^{(\Delta G^\circ / -RT)} = \frac{A_{\text{UCl}_3} A_{\text{Pu}}}{A_{\text{PuCl}_3} A_{\text{U}}} \quad (3)$$

where  $A_i$  are molar activities of species  $i$ ,  $\Delta G^\circ$  is the standard free energy at temperature  $T$ , and  $R$  is the gas constant. Since spent fuel is continuously being processed in the electrorefiner, reducing the amount of accumulated plutonium trichloride in the salt is not currently feasible. Alternatively, increasing the amount of uranium trichloride is possible and has shown promise [23-24]. The addition of uranium trichloride, either directly or indirectly via the oxidation of uranium metal, should decrease the plutonium content of the final uranium product. Testing in the cathode processor with the addition of cupric chloride to oxidize uranium has demonstrated that the plutonium levels of blanket uranium products can be reduced by at least 90%.

## 4.2 Process Crucible

As background, zirconia coated graphite crucibles have been the standard for cathode processing due to their ability to withstand the high temperatures, release consolidated products, and react minimally with the process charge. Although many different crucible materials and coatings have been tested in the prototype cathode processor, zirconia on graphite remains the standard [12]. The zirconia mold wash is applied to the graphite crucibles as a slurry and baked out to 873°K to form a semi-hard layer that provides durability for the loading of electrorefiner products. Following

a cathode processor run, the crucible is cleaned by removing the excess zirconia coating via wire brush or damp rags and then allowing a small amount of water to react in a closed environment with the adhering coating. The entire cleaning and coating operation may span several days due to the time required for separate tasks, such as the bake-out and water reaction. The removed zirconia coating contains significant amounts of uranium as a result of the reaction of metallic uranium with the coating. Currently, the removed coating or dross stream accounts for between 2-4 wt. % of the uranium metal from the electrorefiners.

Obviously, facility throughput could be increased dramatically if cleaning and coating were eliminated. This goal is the basis for the development of a zirconia castable crucible. The zirconia-based castable material is composed of approximately 78 wt. % zirconia, 20 wt. % alumina, and 2 wt. % carbon. It is vibrocast and vacuum fired to produce a hard, sintered liner. A graphite cathode processor crucible has been modified to accept the zirconia castable liner, thus allowing the graphite to act as an outer mechanical shell for handling operations. The zirconia lined graphite crucible has a high resistance to fracture due to the combination of intentional porosity and a dense fused aggregate. Although the formulation for the zirconia castable liner has been modified for uranium processing, the base materials are commercially available as refractory linings, very inexpensive, and easily scalable. The resulting composite crucible utilizes the graphite for handling and mechanical strength, and the castable liner for reuse without coating and cleaning.

Two different sized composite crucibles have been tested in the FCF cathode processor with irradiated cathode products. The first crucible is 8.8 liter in volume and has been tested a total of six times. Due to its successful performance without recoating, a larger (14.9 liter) crucible was fabricated and has been tested a total of four times. Pre- and post-test photographs of the larger crucible have been taken and are shown in Fig. 9. No difficulties have been encountered on ingot dumping and the appearance of the castable liner remains relatively unchanged. Uranium losses have been slightly higher for the smaller crucible (2-3 wt. %) than for the larger crucible (0-1 wt. %), most likely due to fabrication improvements in the surface density and firing. The developmental plan for the composite crucibles is to continue testing with electrorefiner products in the FCF cathode processor to confirm their compatibility and life-cycle.

Another alternative crucible material is being developed and has been tested in the FCF cathode processor. The crucible material is niobium coated with a plasma-sprayed hafnium nitride coating [25-26]. Prior to testing in FCF, the hafnium nitride coated crucible (6.3 liter) was previously tested for multiple runs (15) in the prototype cathode processor with uranium and salt (no fission products or plutonium chlorides). No apparent degradation to the crucible or its hafnium nitride coating was observed during testing in the prototype cathode processor. The purpose of testing

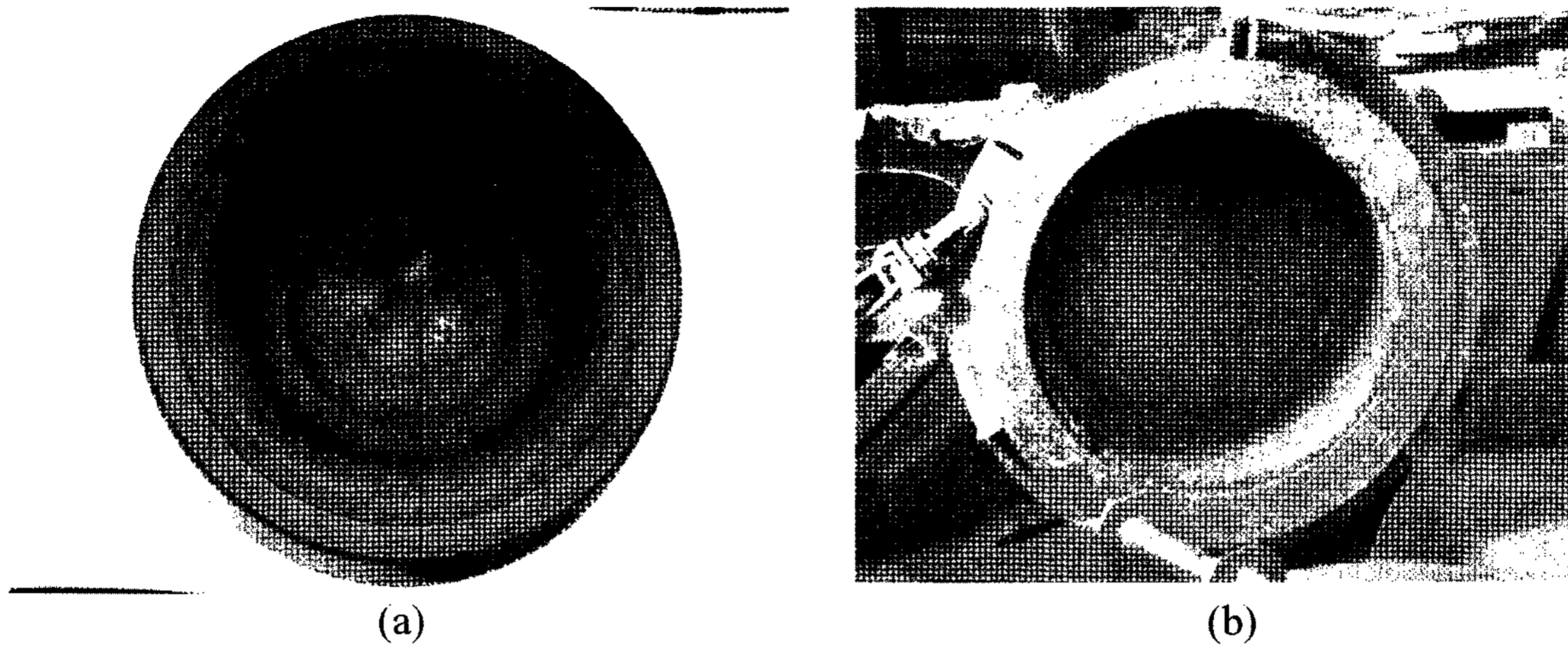


Fig. 9. (a) Zirconia-Based Castable Crucible Prior to (b) and After the FCF Cathode Processor Test

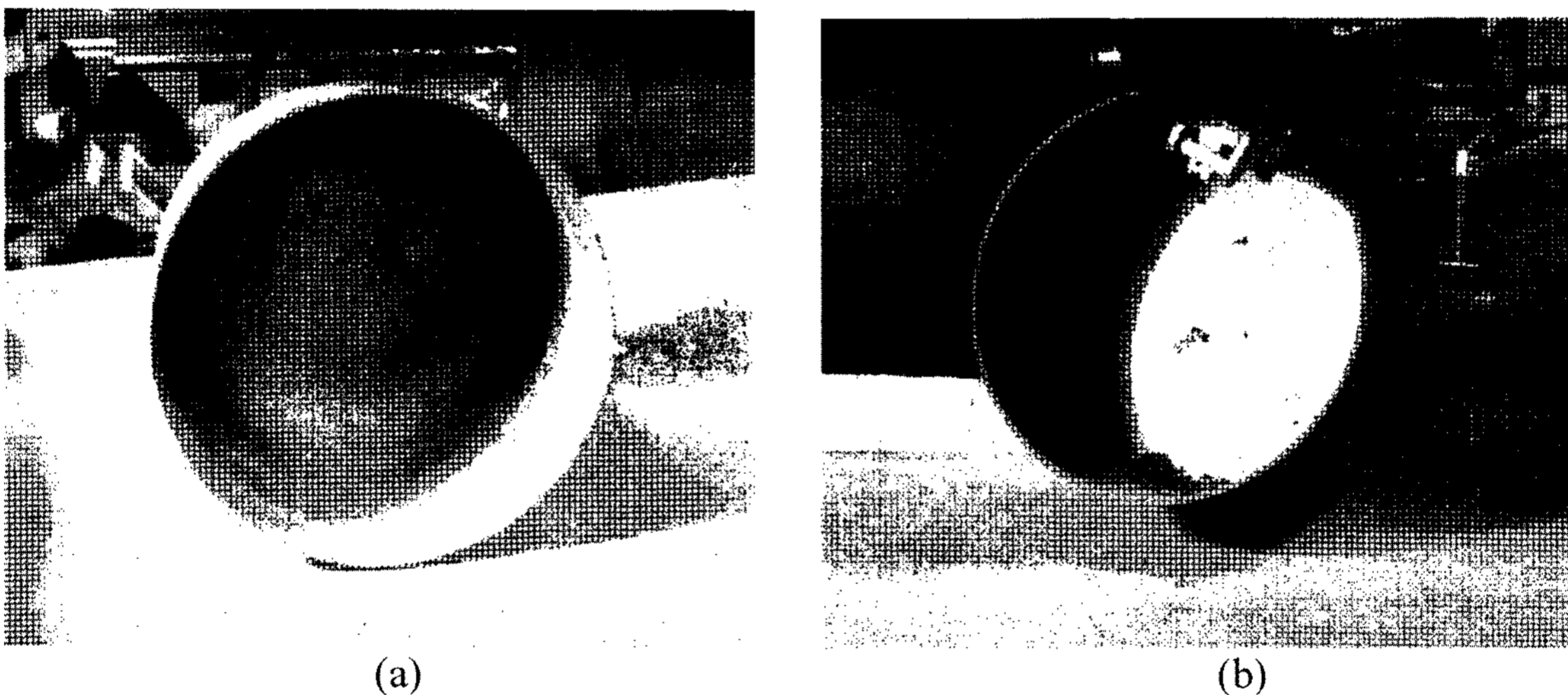


Fig. 10. (a) Hafnium Nitride Coated Crucible Prior to (b) and After the First FCF Cathode Processor Test

in the FCF cathode processor is to verify the crucible's compatibility with salts containing fission products and plutonium.

The hafnium nitride coated crucible has been tested a total of five times to date in the FCF cathode processor with products from the Mk-IV electrorefiner. In these tests, the niobium crucible was placed into the standard graphite process crucible for handling purposes and containment of the melt should the niobium crucible fail. Following the first test, no difficulties were encountered on ingot dumping and the appearance of the crucible was unchanged (Fig. 10). The crucible gained a total of only

7 g for the first test and no cleaning was required for its reuse. The next three tests utilized the same original batch but were rerun due to consolidation and holdup problems caused by a high zirconium content of the cathode products. A fifth test was performed with new cathode product and the ingot released successfully despite residual holdup material from the previous three tests. The developmental plan for the niobium crucible is to continue testing with electrorefiner products in the FCF cathode processor to confirm its compatibility and longevity. A full-scale (~17 liter) hafnium nitride coated crucible is being procured and is currently in the fabrication process.

## 5. CONCLUSIONS

The development of a distillation process for the separation of salt and cadmium from actinide products has been achieved for a variety of electrorefining products. The equipment designed for cathode processing has performed exceptionally under extraordinary conditions over many years of operation. Several notable outcomes have been recognized during cathode processing with respect to operating conditions, enhancements, and maintenance. The dual testing capabilities provided by two distillation furnaces have been an invaluable resource during the development of the distillation process. Specific operating parameters have been distinguished to effectively distill salt and cadmium from actinide products. Significant improvements in equipment performance have been realized by the redesign of the induction coil for high-throughput operations. As anticipated, periodic maintenance has been necessary to keep the equipment functional in light of the challenges presented by the hostile, high temperature environment. Most recently, alternative crucible materials have been successfully tested in the cathode processor and are being developed for future applications.

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