

Optimization of an Acid-Dissolution Front-End Process to Produce a Feed for the Current Mo-99 Recovery Processes

Chemical Science and Engineering Division

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Optimization of an Acid-Dissolution Front-End Process to Produce a Feed for the Current Mo-99 Recovery Processes

by

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OPTIMIZATION OF AN ACID-DISSOLUTION FRONT-END PROCESS TO PRODUCE A FEED FOR THE CURRENT Mo-99 RECOVERY PROCESSES

1 INTRODUCTION

The Global Threat Reduction Initiative (GTRI) Conversion Program develops technology necessary to convert civilian facilities that use high enriched uranium (HEU) to low enriched uranium (LEU) fuels and targets. The conversion from conventional HEU dispersion targets to LEU for ^{99}Mo production requires approximately five times the uranium in a target to maintain the ^{99}Mo yield per target. Under GTRI, Argonne National Laboratory (Argonne) is developing two frontend options for the current processes to allow the use of LEU-foil targets. In both processes, the aim is to produce a product after the frontend that will be compatible with current purification operations and that will, with the same number of targets irradiated, provide the same or a higher yield of ^{99}Mo at the end of processing.

The goal of the frontend process is to deliver a product solution that is of the same or higher Mo purity than the current solution and is of equal or better compatibility with current purification process. In the first frontend option, the LEU foil (contained in a thin [10–15 μm] Ni fission-recoil barrier) is removed from the annular target and dissolved in nitric acid. This report describes results from performance tests and design optimization of the nitric-acid-dissolver system that will be used for the acid frontend process. The design, fabrication, and performance test planning for this system are described in a fiscal year 2011 report (Jerden et al. 2011a, 2011b).

2 LOW-PRESSURE, NITRIC-ACID-DISSOLVER SYSTEM FOR LEU FOILS

A nitric-acid-dissolver system was designed to allow the dissolution of up to 250 g of irradiated LEU foil and associated fission recoil barrier metal (e.g., Ni) at near-ambient pressure. Components of the dissolver system were tested using a full-scale prototype so that the design could be optimized in preparation for a full-scale demonstration. The key design criteria that this dissolver system must incorporate are summarized below:

- All water vapor, reaction products, and fission gases must be contained within the dissolver system at a maximum temperature of 125°C and 2 atm of pressure (absolute) under both normal and off-normal (loss of cooling during reaction) conditions.
- The acid-feed system must be designed so that the thermally hot LEU foil (hot from decay heat) can be immersed in nitric acid without losing solution due to instantaneous boiling.
- All dissolver system components must be designed for remote operation in a hot cell facility.
- Gas-trap components must be designed to trap/neutralize all nitrogen oxide and acid gases (NO, NO₂, HNO₂, HNO₃) as well as trap iodine gas for possible extraction of economically important iodine isotopes (noble fission gases will be passively contained).

The dissolver system design basis requirements imposed by the thermal and chemical properties of the LEU-foil dissolution process as well as the dissolver system design and plan for performance testing are discussed in Jerden et al. (2011b). A schematic flow diagram showing the major components of the LEU–nitric acid frontend process is shown in Figure 1.

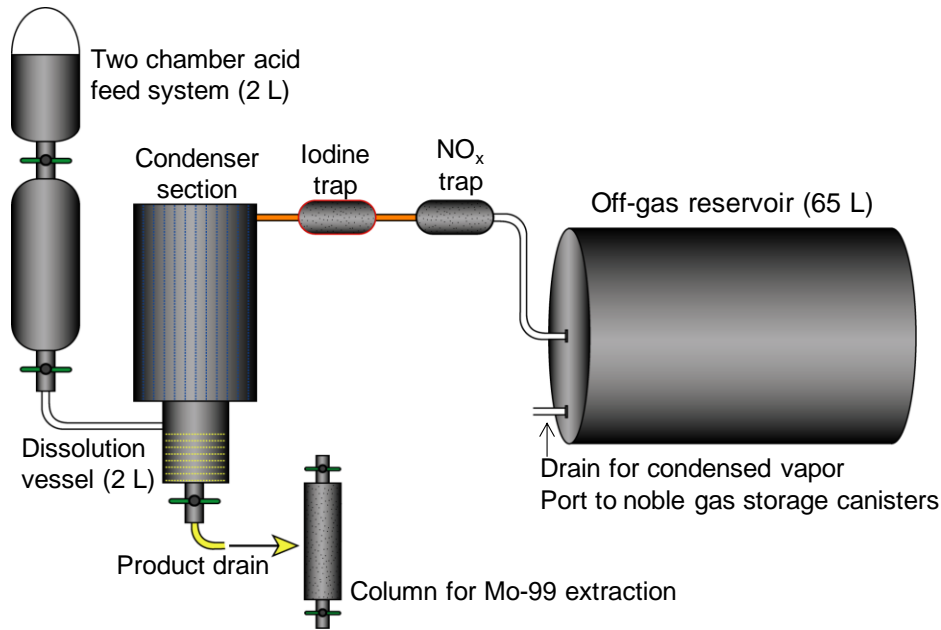
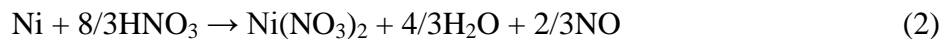
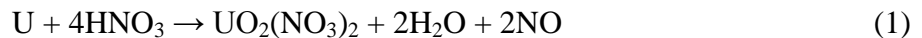


FIGURE 1 Schematic Flow Diagram Showing Components of the LEU Nitric-Acid-Dissolver System and the Mo-99 Extraction Column

In this report, we discuss results from the performance testing of the dissolver system and describe how the system is being optimized based on these experimental findings. Before discussing the test results, the design basis requirements for the dissolver system are summarized (for a more detailed discussion of design criteria, see Jerden et al. 2011b).

2.1 DISSOLUTION REACTION: REACTION HEAT, DECAY HEAT, AND OFF-GAS VOLUME

The volume and concentration of nitric acid for a given experiment will depend on the mass of the metal being dissolved, as well as the desired final acid concentration of the “product” solution (i.e., the solution produced by dissolution experiment). Controlling the final acid concentration is important for optimizing the Mo-99 extraction step that comes after dissolution. The volumes and concentrations of acid, as well as the amount of nitrogen oxide gas (NO_x: NO, NO₂, N₂O₄) that will be produced, are determined by the following general reactions:



In the presence of oxygen, the NO(g) produced in these dissolution reactions is rapidly converted to NO₂(g):



Using the kinetic rate law presented by Chilton (1968, page 29), one finds that the rate of reaction (3) is on the order of milliseconds to seconds, even at relatively low O₂ partial pressures (0.1–0.001 atm). When water vapor and oxygen are present, NO₂ can be converted to both nitrous and nitric acid vapors [HNO₂(g) and HNO₃(g)], which will dissolve in condensed water and flow back down into the dissolver.

In an effort to be more precise about the concentrations of off-gas species, relative amounts of the important nitrogen oxide gases produced by the dissolution of different amounts of LEU were calculated using the thermodynamic code OLI-ESP. These calculations predict that approximately 2.1 moles of NO_x + H₂O(g) will be present in the dissolver following the dissolution of 250 g of LEU foil. These results provide a design basis for the off-gas treatment components of the dissolver system (Jerden et al. 2011b).

Most of the off-gas from the dissolver will consist of NO_x (Jerden et al. 2011b); however, ORIGEN calculations show that iodine, xenon and krypton will also be present in the off-gas. The ORIGEN calculations, performed by Charlie Allen, University of Missouri, using ORIGEN2, Version 2.2, assume the following: irradiation of 1 g of uranium foil enriched to 19.75% ²³⁵U; power = 1.9×10^{-3} MW; burnup = 1.59×10^{-2} MW days; Flux = 2.1×10^{14} N/cm² sec; burnup is for 200 hr; foil composition is given for cooling times of 12, 24, 36, and 48 hr. The most abundant off-gas species will be iodine (4.2×10^4 Ci per 250 g LEU, after 12 hr cooling), xenon (2.6×10^4 Ci per 250 g LEU, after 12 hr cooling) and krypton (1.7×10^3 Ci per 250 g LEU, after 12 hr cooling). As part of this study, the feasibility of using copper metal to trap and extract iodine gas from the dissolver during the dissolution reaction has been assessed experimentally (Jerden et al. 2011b). The efficiency of NaOH to neutralize/trap the NO_x gases has also been investigated (Jerden et al. 2011b).

Thermodynamic calculations show that the dissolution of 250 g uranium metal (~1.05 moles) the total energy released may be up to 1600 kJ, but will probably be closer to 1000 kJ. Assuming adiabatic conditions and a 30-minute reaction time, this energy would correspond to a maximum thermal power output of approximately 890 W (Jerden et al. 2011b). The ORIGEN calculations show that a total thermal output for 250 g of irradiated LEU is around 1000 W for a 12-hr cooling (Figure 2).

Based on the enthalpy and decay heat calculations, the cooling system for the LEU nitric acid dissolver system must be able to sink out a maximum of 2000 W (thermal). Therefore, if it is assumed that the dissolution of 250 g of irradiated LEU foil takes 30 minutes, 2000 W of thermal power will be generated and will need to be removed from the system to ensure that water vapor (and acid) is not lost during the dissolution process. Experiments in which both the reaction and decay heats are simulated using heating coils wrapped around the dissolver vessel were performed to test the performance of the dissolver cooling system.

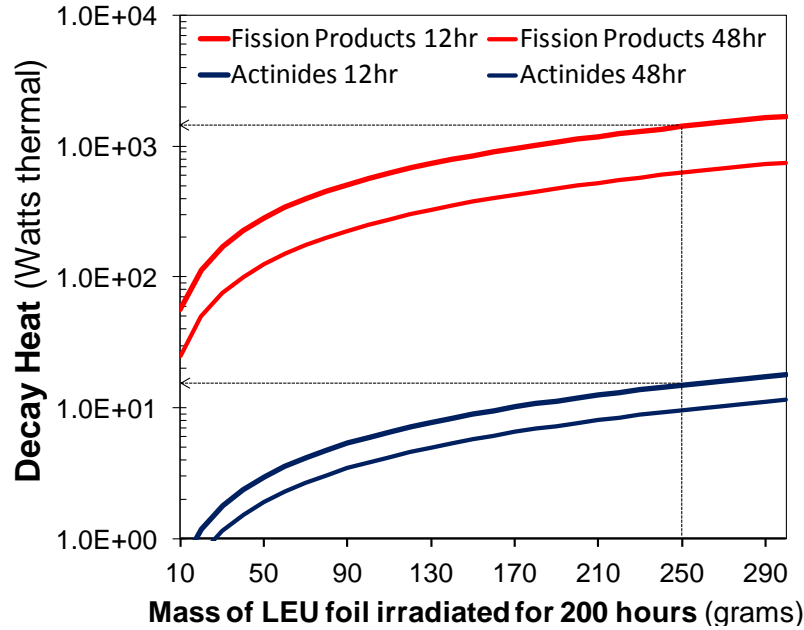


FIGURE 2 Cumulative Decay Heat for Fission Products and Actinides for a Range of LEU Foil Masses after 200-hr Irradiation (Curves are for 12 and 48 hr of cooling [time out of reactor]. Calculated using ORIGEN2, assuming the following: irradiation of 1 g of uranium foil enriched to 19.75% U-235, power = 1.9×10^{-3} MW; burnup = 1.59×10^{-2} MW days; flux = 2.1×10^{14} N/cm² sec.)

2.2 SUMMARY OF DISSOLVER SYSTEM DESIGN CALCULATIONS

A mathematical model was implemented in Mathcad for the design optimization of the nitric-acid LEU-foil dissolver system. The key design criteria targeted in these calculations are listed in Section 2. The current dissolver design was based on the Mathcad calculations. By comparing the model calculations with test results (discussed below), the system design has been optimized: specifically the cooling system fan size and off-gas reservoir design and dimensions. The Mathcad design model was used for the following design aspects (details of dissolver system design are described in Section 2.3 below):

- Sizing of the cooling fins and air flow requirements from the cooling fan in order to remove both the reaction heat and decay heat from the dissolver: with a reaction heat of approximately 1600 kJ over 30 minutes and a constant decay heat of approximately 1500 W, the average required heat removal rate is 10181 kJ/hr. This value determined the original cooling system design.
- The pressure drop across the cooling fin section was calculated based on the required flow velocity as determined by the convective heat removal from the fins. The total air volume flow rate and temperature rise of the air across the

fins were also calculated. These results are used to determine the cooling fan size from fan performance curves.

- The required gas volume and heat sink capacity of the off-gas reservoir was calculated. It was assumed that all the reaction heat and off-gases are transferred to the reservoir (loss of cooling scenario). In addition, the steady state requirement for the decay heat removal is assumed to be dissipated by the reservoir. A thermal analysis was performed to determine an approximate value for the heat loss from the off-gas reservoir. The reservoir tank was assumed to be maintained at a uniform temperature of 100°C with an ambient air temperature of 32°C. Heat loss by radiation and heat loss by natural convection are calculated separately and then added together to determine the total heat flow from the outside of the reservoir to the air in the hot cell. Results from this calculation were used to size and design the aluminum heat sink rings that encase the off-gas reservoir tank.
- The heat-removal capacity due to condensing of the process vapor on the inside surface of the dissolver condenser section was calculated and accounted for in the cooling system design. Boiling of the dissolver solution occurs in the vessel at the bottom of the dissolver, and the vapor moves upward to the condenser section where it condenses on the cooler walls that have been cooled by the air flow over the fins on the outer surface. The wall temperature is assumed to be below 100°C.

2.3 DISSOLVER DESIGN OVERVIEW

The nitric-acid LEU-foil dissolver is designed to operate at pressures less than 2 atm (absolute) and at temperatures less than 125°C. The design and footprint of the dissolver system are shown in Figures 3 and 4. The full-scale dissolver prototype used in the performance tests is shown in Figure 5.

The dissolver system consists of a 304 stainless steel vessel (2-L volume) connected to an approximately 65-L (30 cm × 90 cm) off-gas reservoir (Figure 5). Figure 6 shows the full-scale, prototype dissolver system setup (off-gas reservoir not used) during one of the dissolution experiments discussed in this report (Section 3). The dissolver vessel is open to the off-gas reservoir during the dissolution process. The volume of the reservoir was chosen to provide passive containment of all water vapor and reaction products at a pressure less than 2 atm (absolute), during both normal and off-normal (loss of cooling during reaction) conditions. The role that the condenser section of the dissolver plays during a typical dissolution run is shown schematically in Figure 7. In order to keep the temperature of the gas within the reservoir to below 100°C during a potential loss of cooling, the reservoir is clad in an aluminum heat sink (Al fin rings) (see Figures 3 and 4).

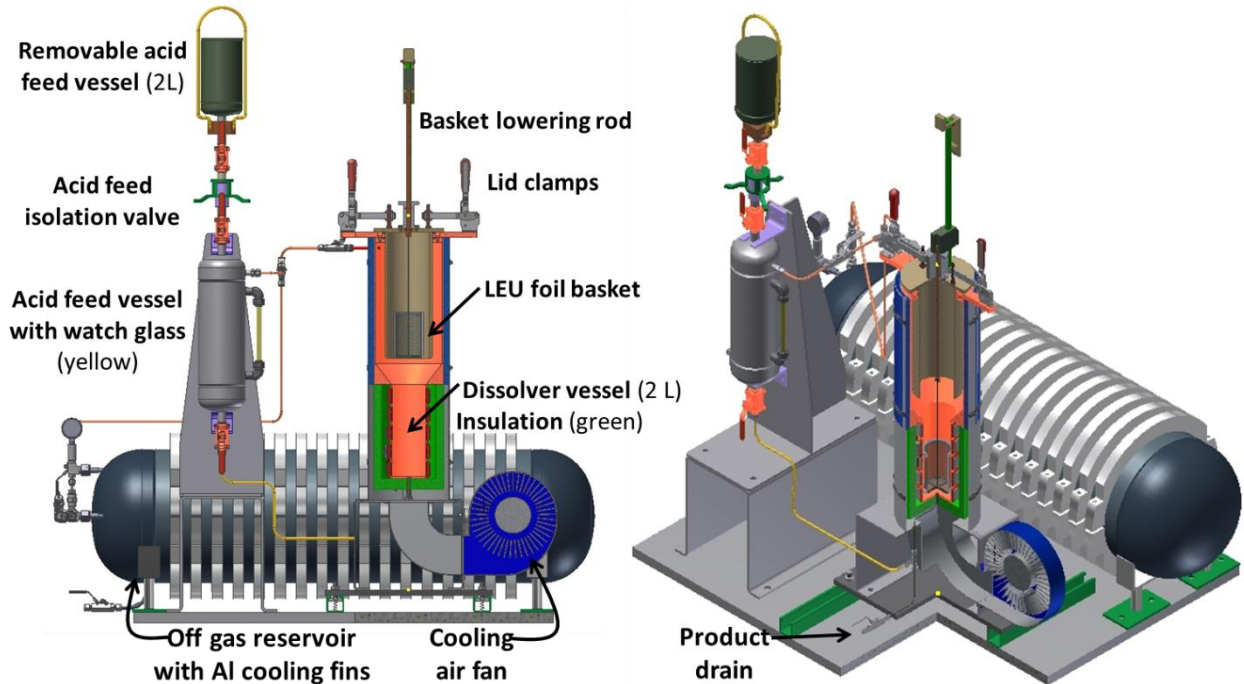


FIGURE 3 Conceptual Diagrams of Nitric-Acid-Dissolver System

The dissolution process is started by first lowering the uranium foil (contained within a steel mesh basket) into the dissolver vessel and then sealing the vessel with a metal cap. Pre-heated acid ($\sim 100^{\circ}\text{C}$) is then added to the vessel using a two-chamber acid feed system that is designed to avoid pressurization of the acid bottle in the event that the dissolution reaction begins instantaneously when the acid addition step is started. The dissolver vessel is cooled by forced air blown from the base of the unit. The temperature of the dissolver solution is monitored by a thermocouple. The dissolver vessel is insulated so that the top of the vessel is cooled continuously during the reaction. Heat loss from the top of the vessel is optimized by the presence of steel cooling fins attached to the condenser part of the dissolver system (Figure 5). This design causes the water vapor to condense along the walls at the top of the vessel during the dissolution reaction (as acid is boiling). This process is shown schematically in Figure 7.

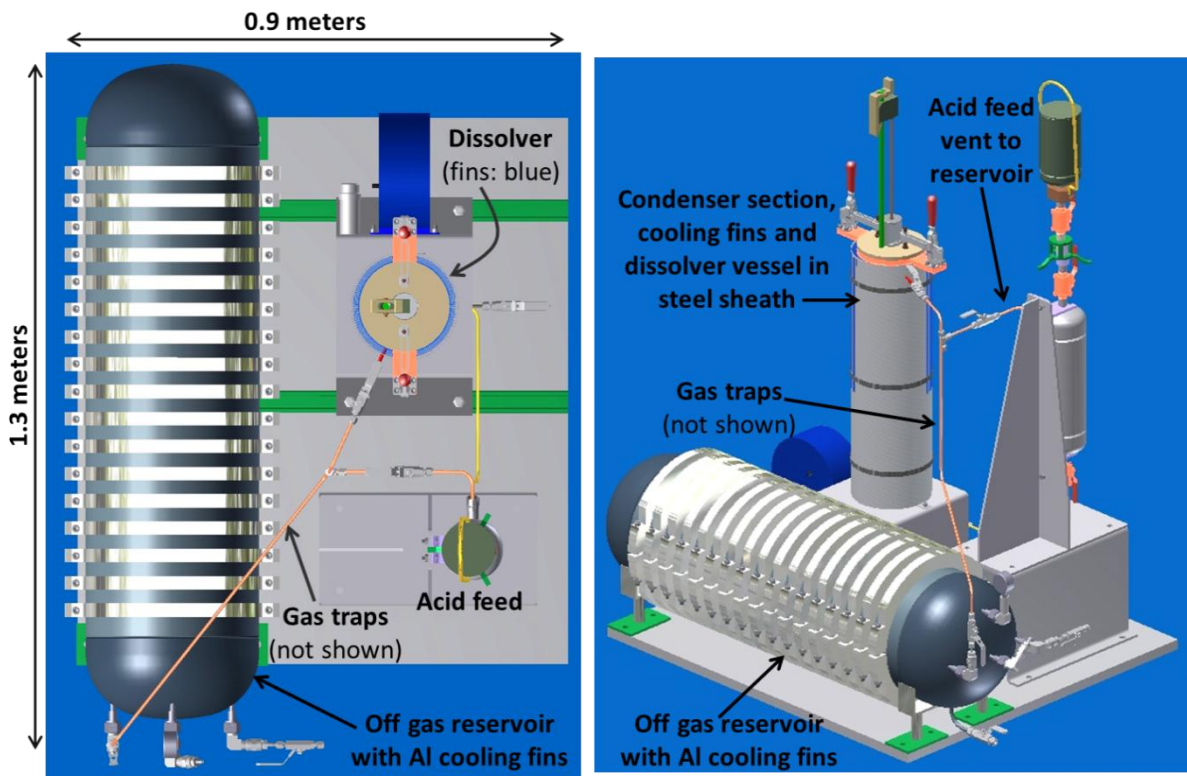


FIGURE 4 Conceptual Diagrams of Dissolver System Showing Plan View with Approximate Footprint Dimensions (left) and an Isometric View Showing the Gas Vents from the Dissolver and Acid Feed Components to the Off-Gas Reservoir (right)

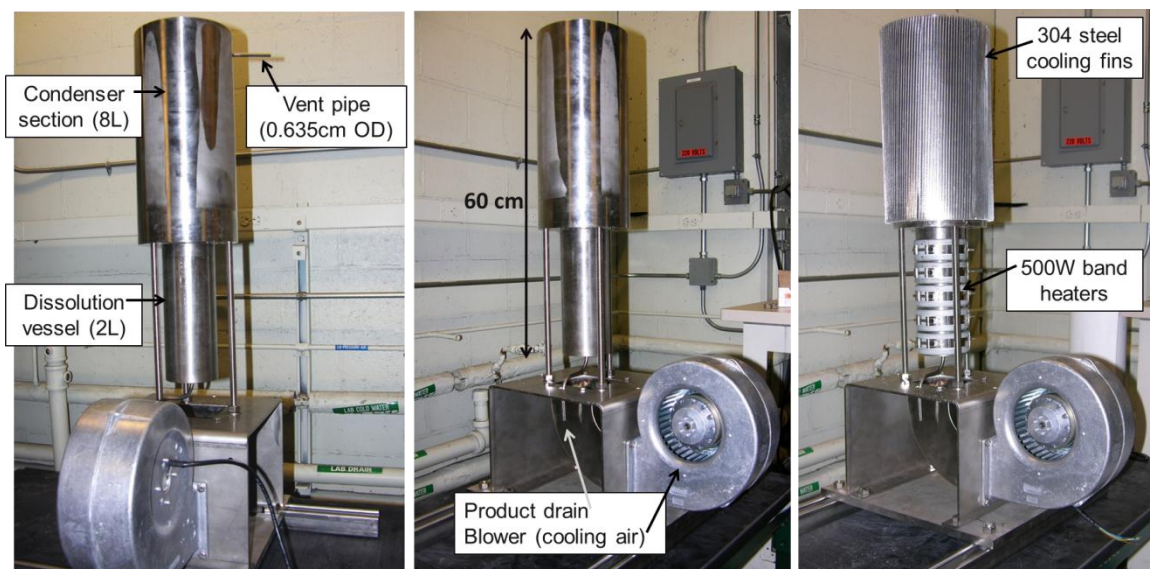


FIGURE 5 Full-Scale Dissolver Prototype Used in Performance Tests

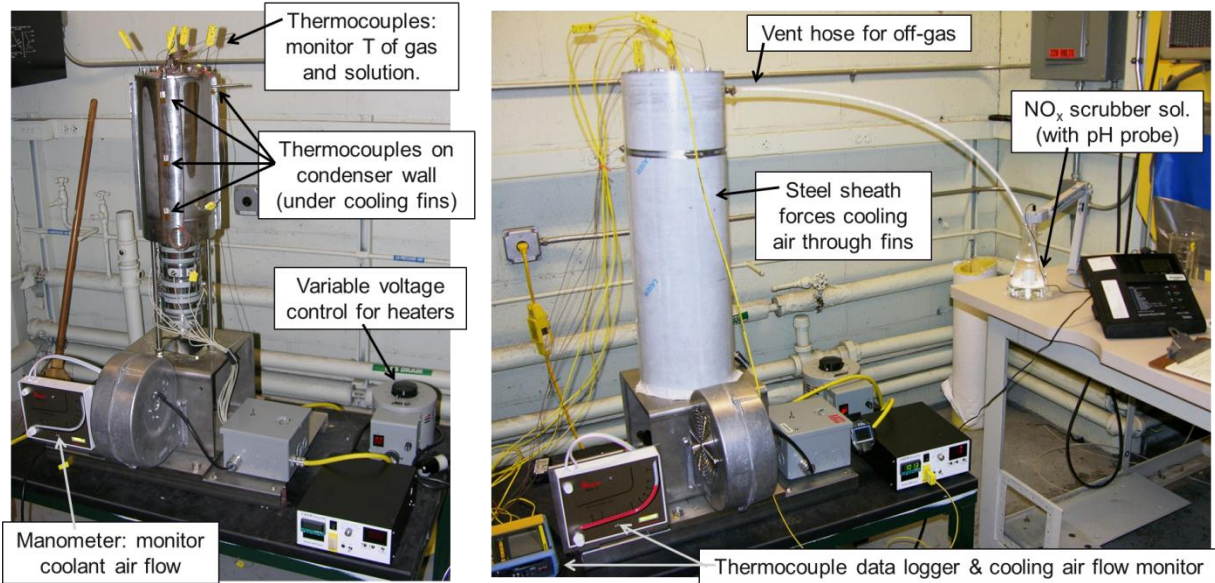


FIGURE 6 Full-Scale Dissolver Prototype Used in Performance Tests (Photo on right was taken during a dissolution experiment.)

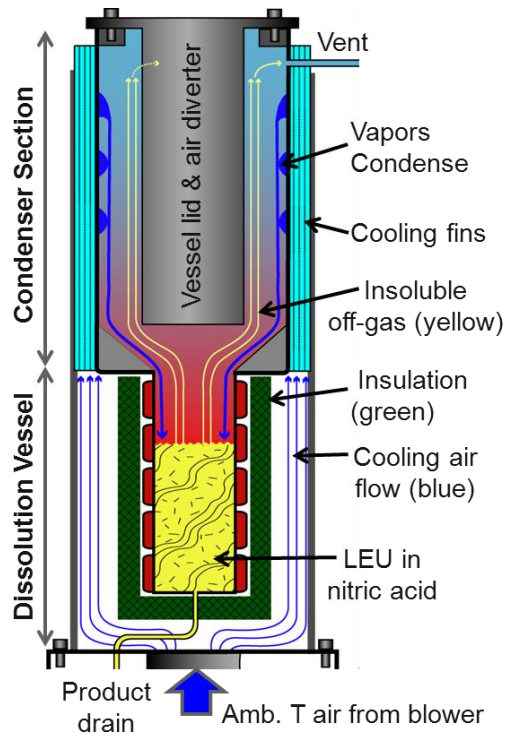


FIGURE 7 Schematic Cross-Section through the Condenser and Dissolution Vessel Sections of the Dissolver System

3 DISSOLVER SYSTEM PERFORMANCE TESTS

As stated above, all water vapor, reaction product, and fission gases must be contained within the dissolver system at a maximum temperature of 125°C and 2 atm (absolute) under both normal and off-normal (loss of cooling during reaction) conditions. A key technical challenge for the dissolver system design was making sure that water vapor and acid gases did not escape from dissolver vessel condenser section. If these gases escaped during the dissolution reaction, the reaction would slow significantly or even halt, thus delaying the overall Mo-99 production process.

To retain water vapor and acid gases within the dissolver, we designed a forced-air cooling system to keep the condenser section cool enough to efficiently retain water vapor and nitric acid within the dissolver vessel. To test the condensation process, only the condenser section and reaction vessel were being used in our first set of heat flow experiments. The full-scale prototype dissolver shown in Figures 5 and 6 was used for these tests.

Two types of experiments were performed: (1) Condensation performance tests were run using boiling water to confirm and quantify the performance of the condenser section of the dissolver system. An array of thermocouples was used in the initial testing to measure all relevant thermal gradients during the dissolution reaction process (inside and outside the dissolver vessel and condenser section). (2) Dissolution tests were also performed to measure the loss of NO_x from the dissolver and to test the performance of the NaOH-based NO_x scrubber. The general procedures used for the experiments are summarized as follows:

Condenser section performance tests: The purpose of these experiments was to determine the heat-exchange capacity between the coolant air flow on the outside of the condenser section and the condensing vapor to liquid on the inside of the vessel. Band heaters attached to the base of the dissolver vessel (Figures 5) were used to simulate the exothermic heat from the LEU dissolution reaction, as well as the decay heat from the irradiated foil. The condenser section performance tests were performed as follows:

- 600 mL of deionized water was fed into the dissolver vessel. The dissolver was sealed using bolted lid on the top of the condenser section. At this point, the heaters were off, and the vessel was at ambient temperature (~22°C).
- The blower was turned on, and the monitoring and recording of temperatures and cooling air flow velocity were initiated. The temperatures were allowed to stabilize before starting the next step.
- The heaters were turned on at low power, and the transient temperature increase was monitored and recorded.
- Once the system reached steady state (water boiling), temperatures within and outside the dissolver were continuously monitored using a computer data-

logger. The cooling-air flow velocity was measured, and the electrical power to the heaters was measured.

- The experiments were run at this steady-state condition for approximately 1 hr. The vent tube was constantly monitored to see if vapor was escaping from the condenser section.
- After 1 hr of running the experiment at steady state, the heaters were turned off, and the blower was kept on until the vessel returned to ambient temperature.
- When the vessel was at ambient temperature, the water was drained out (using the product drain valve), and its mass/volume was measured to determine how much vapor was lost from the condenser section during the experiment.

Copper metal dissolution tests: Similar to the “water” tests described above, the purpose of these experiments was to determine the heat-exchange capacity between the coolant-air flow on the outside of the condenser section and the vapor condensing to liquid on the inside of the vessel. Band heaters attached to the base of the dissolver vessel (Figures 5) were used to simulate the exothermic heat from the LEU dissolution reaction as well as the decay heat from the irradiated foil. The difference between these tests and the “water” tests was that the copper metal was dissolved in nitric acid during the experiment so that the behavior of the NO_x and acid gases could be observed and the performance of a simple NaOH NO_x trap tested. These tests were performed as follows:

- 100 g of copper metal foil (surrogate for LEU foil) was lowered into the base of the dissolver vessel.
- 600 mL of 7 molar nitric acid was fed into the bottom of the vessel. In some experiments, nitric acid was fed into the dissolver prior to the metal foil being added, while in other tests the acid was added after the metal foil had been lowered into the vessel. The dissolver vessel was then sealed. At this point the heaters were off and the vessel was at ambient temperature.
- The blower was then turned on, and monitoring and recording of the temperatures and air flow velocity was initiated. The temperatures were allowed to stabilize before initiating the next step.
- The heaters were turned on at low power, and the transient temperature increase was measured.
- The temperature of the acid was monitored and recorded continuously during the exothermic dissolution reaction.

- Once the acid was boiling, the heaters were turned off, and the blower was kept on. The temperature within the dissolver vessel was monitored and recorded continuously until it reached ambient temperature.
- The dissolver solution (copper nitrate in ~1 molar nitric acid) was removed using the product drain valve, and its mass was measured to determine the loss of acid from the dissolver during the experiment.
- The dissolver lid was removed and the interior of the dissolution vessel was examined using a borescope (1-m long flexible fiber optic camera) to determine whether any undissolved solids remained in the dissolver.

3.1 DISCUSSION OF DISSOLVER PERFORMANCE TEST RESULTS

Results from the condenser section performance tests and the copper metal dissolutions are presented in Figures 8–17. Figures 8, 9, and 10 show the locations of thermocouples (identified as data logger channels 1–8 [CH1–CH8]). Figure 9 also shows the locations where the

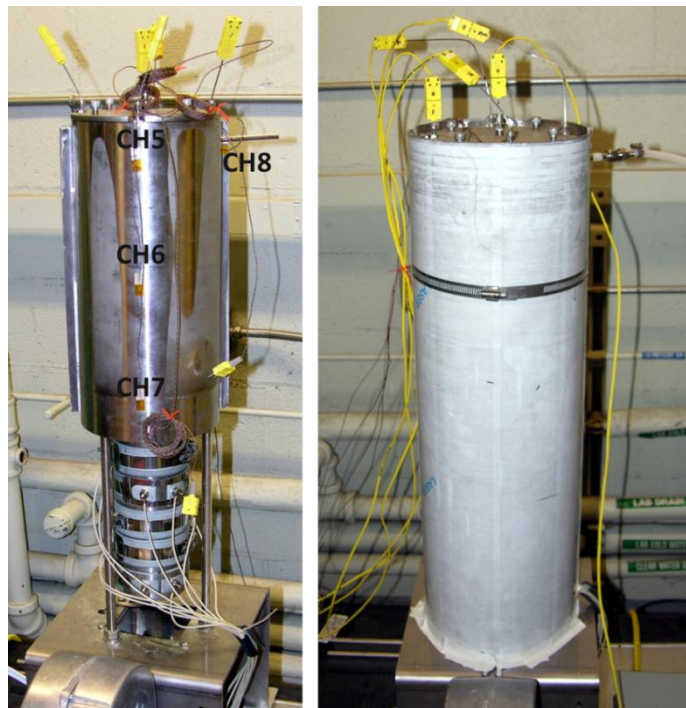


FIGURE 8 Full-Scale Dissolver Prototype Showing Locations of Thermocouples Used to Monitor Temperature within and along the Wall of the Condenser Section of the Dissolver (“CH” refers to data-logger channel. In the temperature profile plots shown below, the thermocouple location is identified as “CH1–CH8.”)

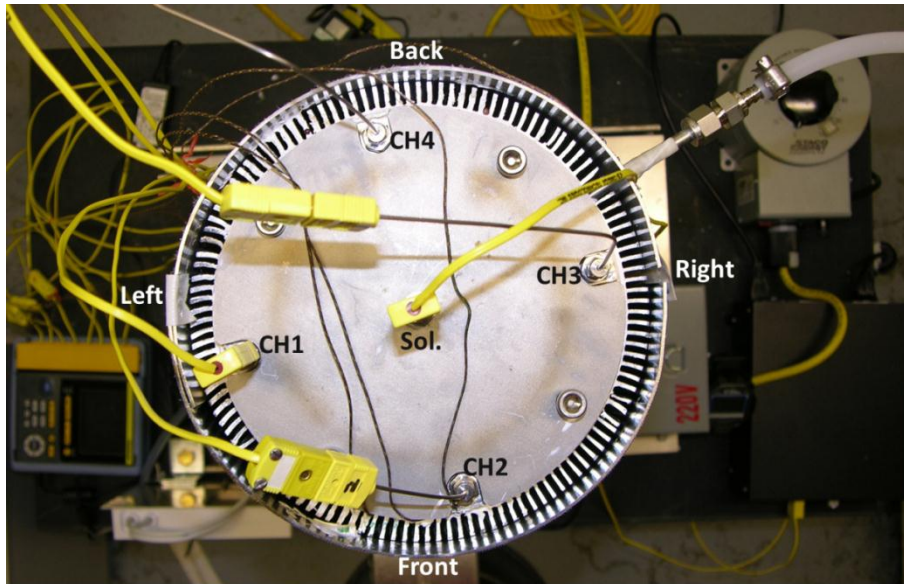


FIGURE 9 Top of the Full-Scale Dissolver Prototype Showing Locations of Thermocouples Used to Monitor Temperature within and along the Wall of the Condenser Section of the Dissolver (“CH” refers to data-logger channel. In the temperature profile plots shown below, the thermocouple location is identified as “CH1–CH8.”)

temperature and velocity of cooling air exiting the steel fins were taken during each experiment (locations labeled “front,” “back,” “left,” and “right”).

The linear flow velocity of the cooling fan was measured throughout each test. The cooling air flow remained constant at 26.42 m/s where it entered the cooling fin section at the base of the dissolver. The cooling air flow measured at the top of the dissolver, where it exits through the cooling fins was consistently 20.32 m/s. The loss of flow velocity is due to turbulence as the cooling air travels up through the dissolver ductwork and is channeled into the cooling fins and perhaps by small leaks in the steel sheath that contains the fins.

Figures 11–15 show thermal profiles within the dissolver during the experiments. To the right of the thermal profile plots are two cross-sections of the dissolver identifying the locations of the thermocouples, which are identified as “Sol.” and “CH1–CH8.” Results from six condenser tests are shown. The first five were performed at different heater powers (simulating decay heat and heat of LEU foil dissolution) that range from 1250 to 2250 W. The results shown in the bottom plot of Figure 13 are from a loss of cooling test that involved first establishing a steady state thermal profile for about 40 minutes and then abruptly cutting power to the cooling fan. Power to the cooling fan was cycled to show the sudden increase in temperature with loss of cooling and the rapid reestablishment of the steady-state (controlled) temperature profile when cooling was restored.

The thermal profiles shown in Figures 11–15 indicate that the maximum air temperatures within the condenser section of the dissolver (CH1, CH2, CH3, CH4) at steady state vary from

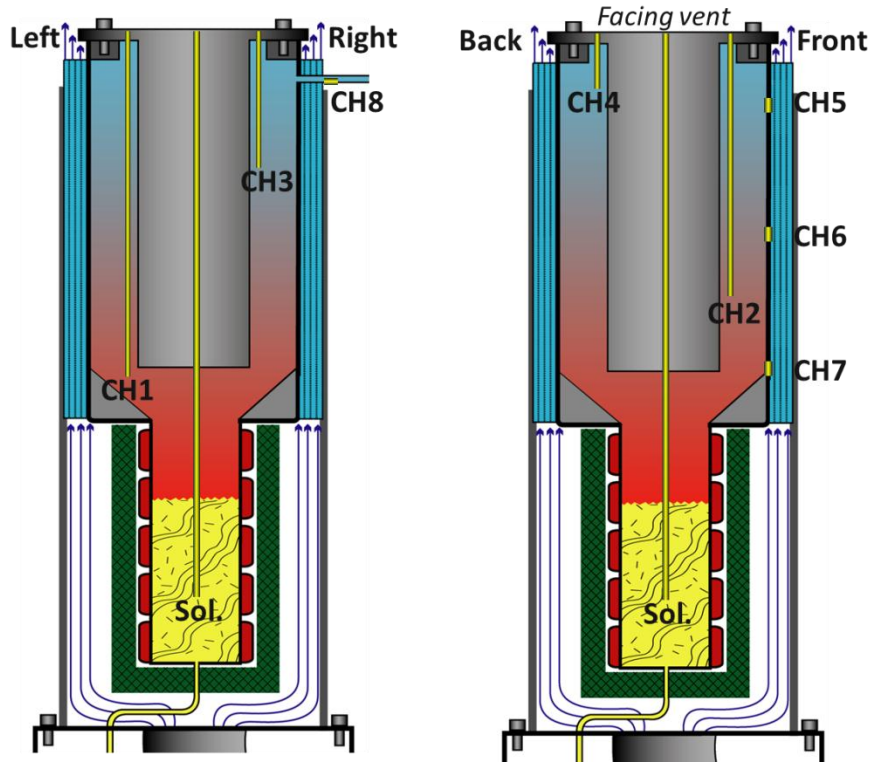


FIGURE 10 Schematic Cross-Sections through the Test Dissolver Showing the Locations of Thermocouples within and along the Walls of the Condenser Section (“CH” refers to data-logger channel. In the temperature profile plots shown below, the thermocouple location is identified as “CH1–CH8.”)

around 70°C for the 1250-W tests to as high as 110°C for the 2250-W tests. The minimum air temperatures in the condenser section range from approximately 55°C for the 1250-W tests to around 90°C for the 2250-W tests. The hottest air temperatures within the condenser section are closest to the dissolution vessel; however, the range of intermediate temperatures does not correlate with thermocouple positions. This observation is believed to be due to a non-uniform heat distribution azimuthally around the dissolver condenser section. The non-uniform heat distribution may be caused by a combination of uneven heating associated with the non-uniform positions of the dissolution vessel heaters and the fact that the dissolution vessel is not completely filled with water (the heaters cover the entire dissolution vessel, but the vessel is only half filled with solution). Specifically, the uneven heating causes the “back” of the dissolver vessel and condenser section (see Figures 9 and 10) to reach a higher temperature than other parts of the dissolver. This observation is demonstrated by the distribution of the air temperatures exiting the cooling fins (Figure 15). For example, at steady state, the air exiting the fins from the “front” of the dissolver ranges from around 31°C for the 1250-W tests to 48°C for the 2250-W tests; the air exiting the fins from the “back” of the dissolver ranges from approximately 44°C for the 1250-W tests to 75°C for the 2250-W tests (Figure 15). Despite the uneven heating during these tests, the results still provide a direct measure of the performance of the condenser section that can be used to optimize the design.

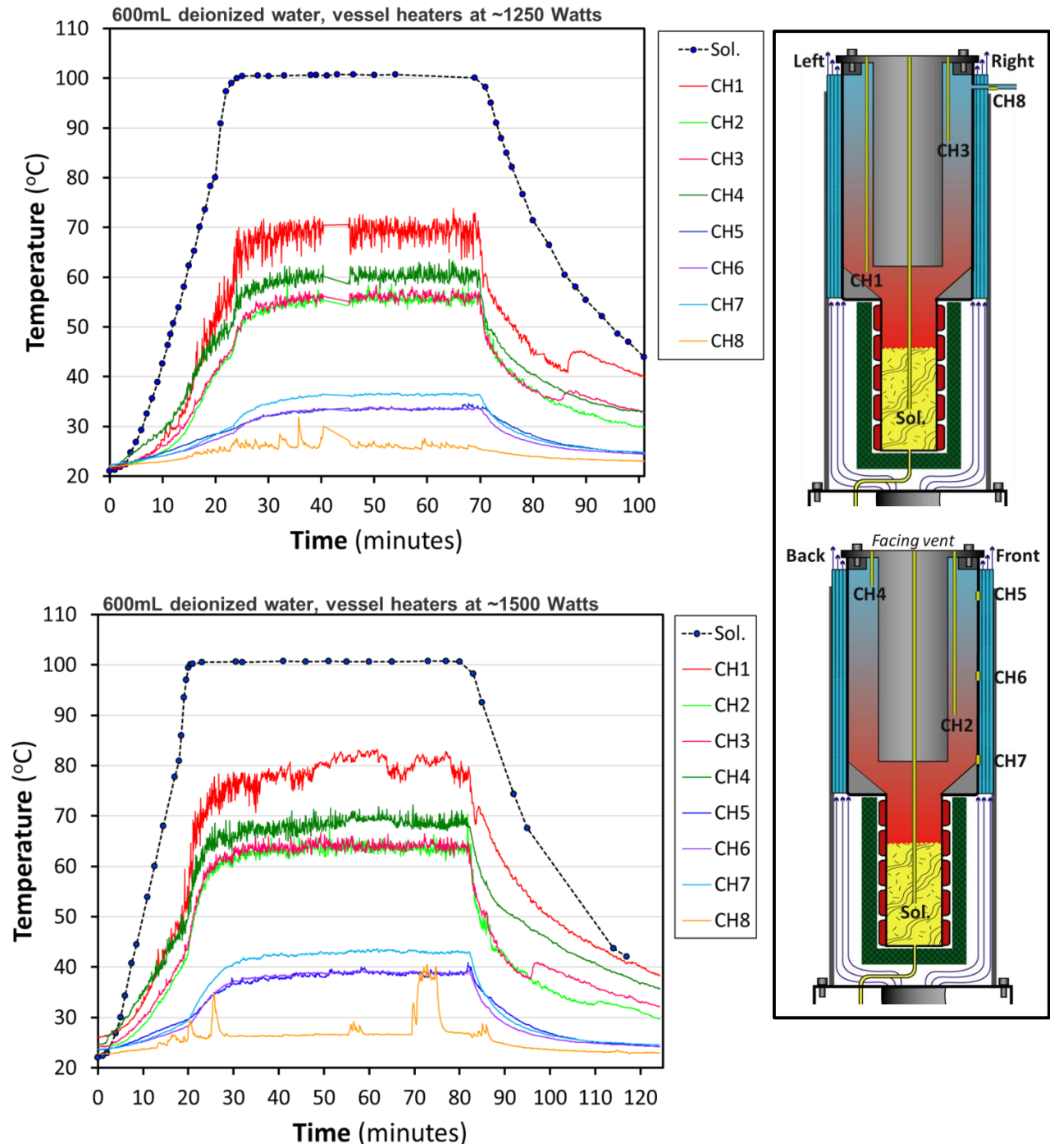


FIGURE 11 Data from Heat Flow Tests (1250 and 1500 W) Using Boiling Water to Study Thermal Gradients within the Dissolver

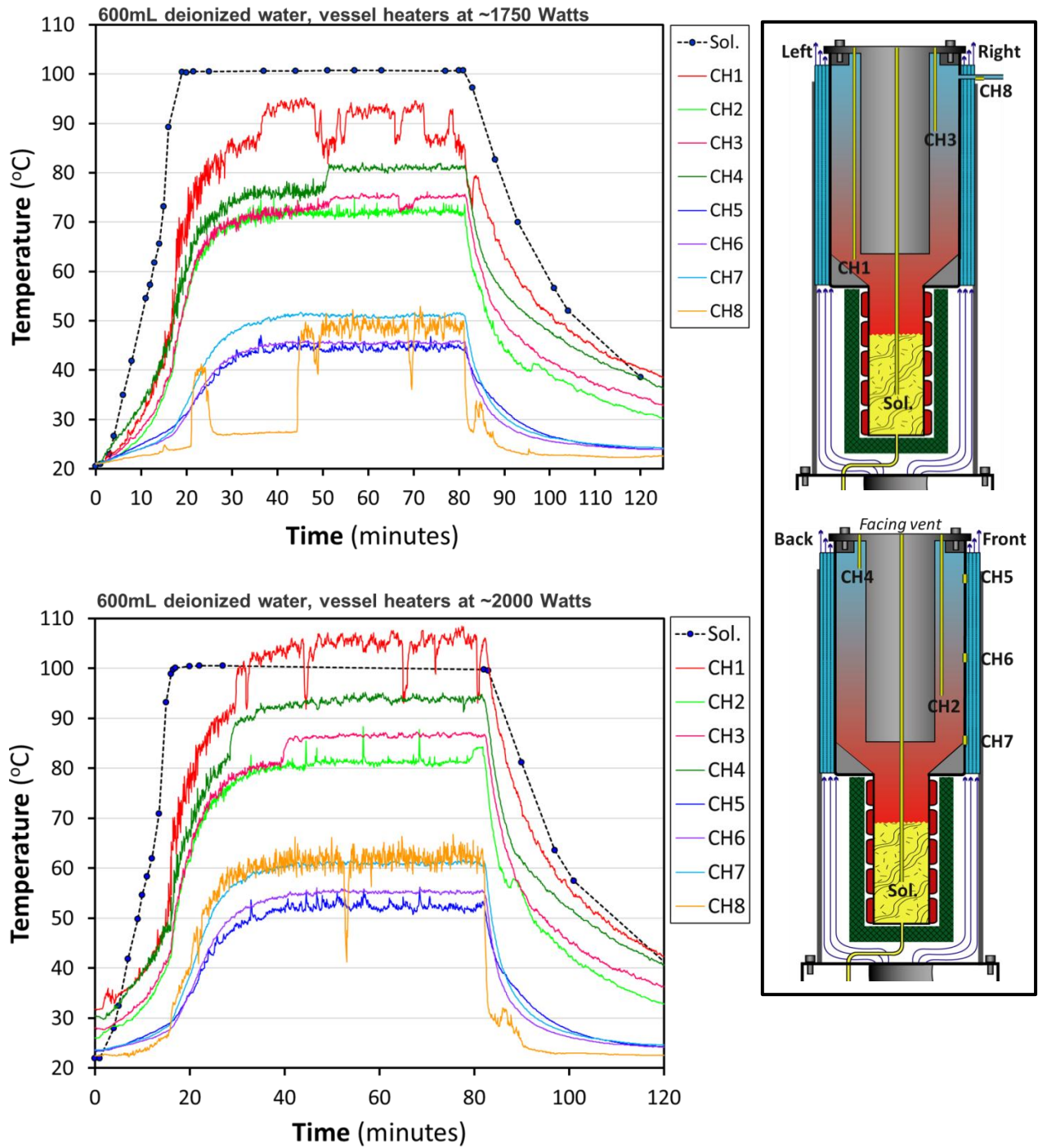


FIGURE 12 Data from Heat Flow Tests (1750 and 2000 W) Using Boiling Water to Study Thermal Gradients within the Dissolver

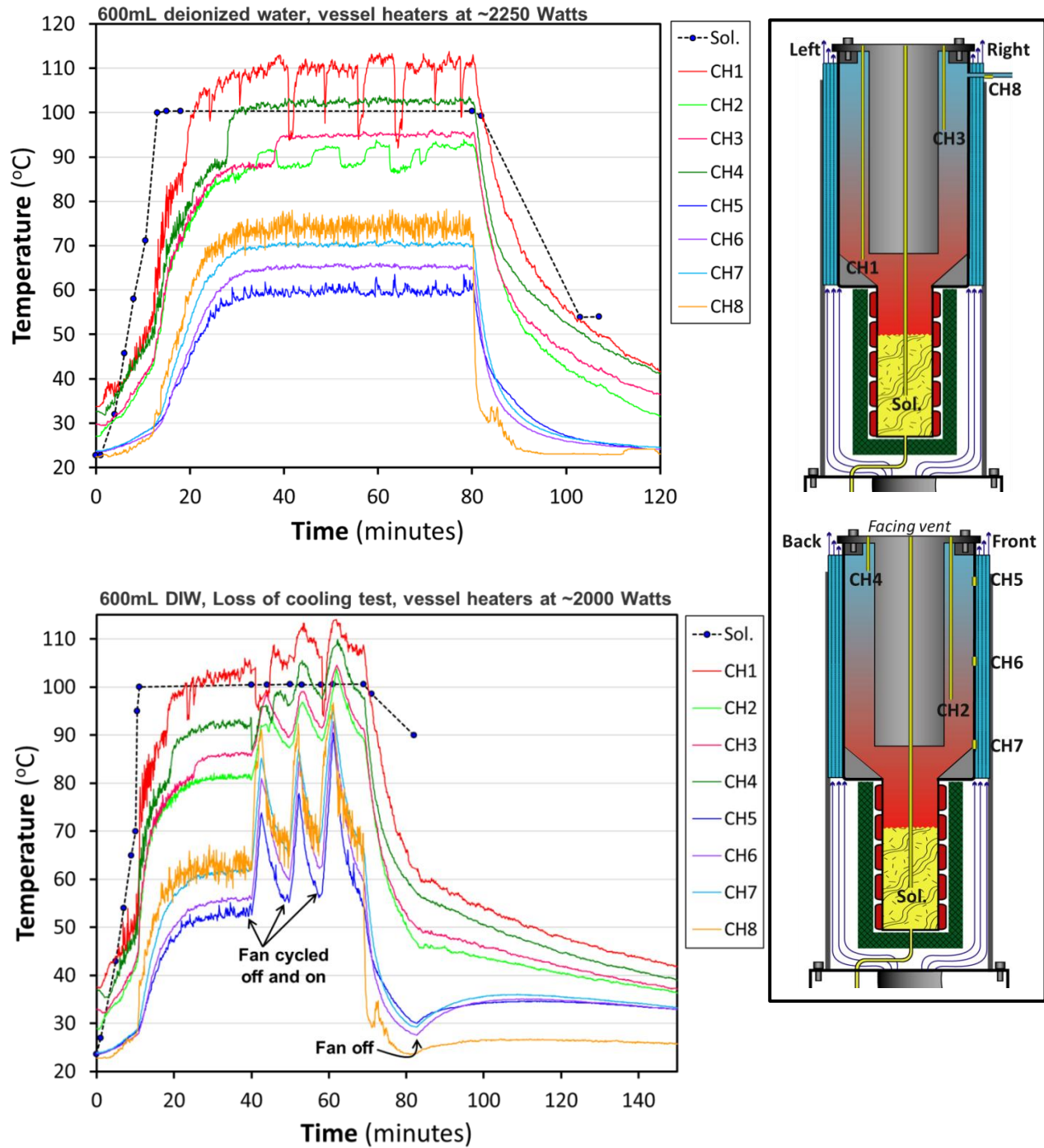


FIGURE 13 Data from Heat Flow Tests Using Boiling Water to Study Thermal Gradients within the Dissolver; Bottom: Data from a Loss-of-Cooling Experiment Where the Fan Was Cycled on and off to Measure the Rate at which Thermal Gradients Change and are Reestablished

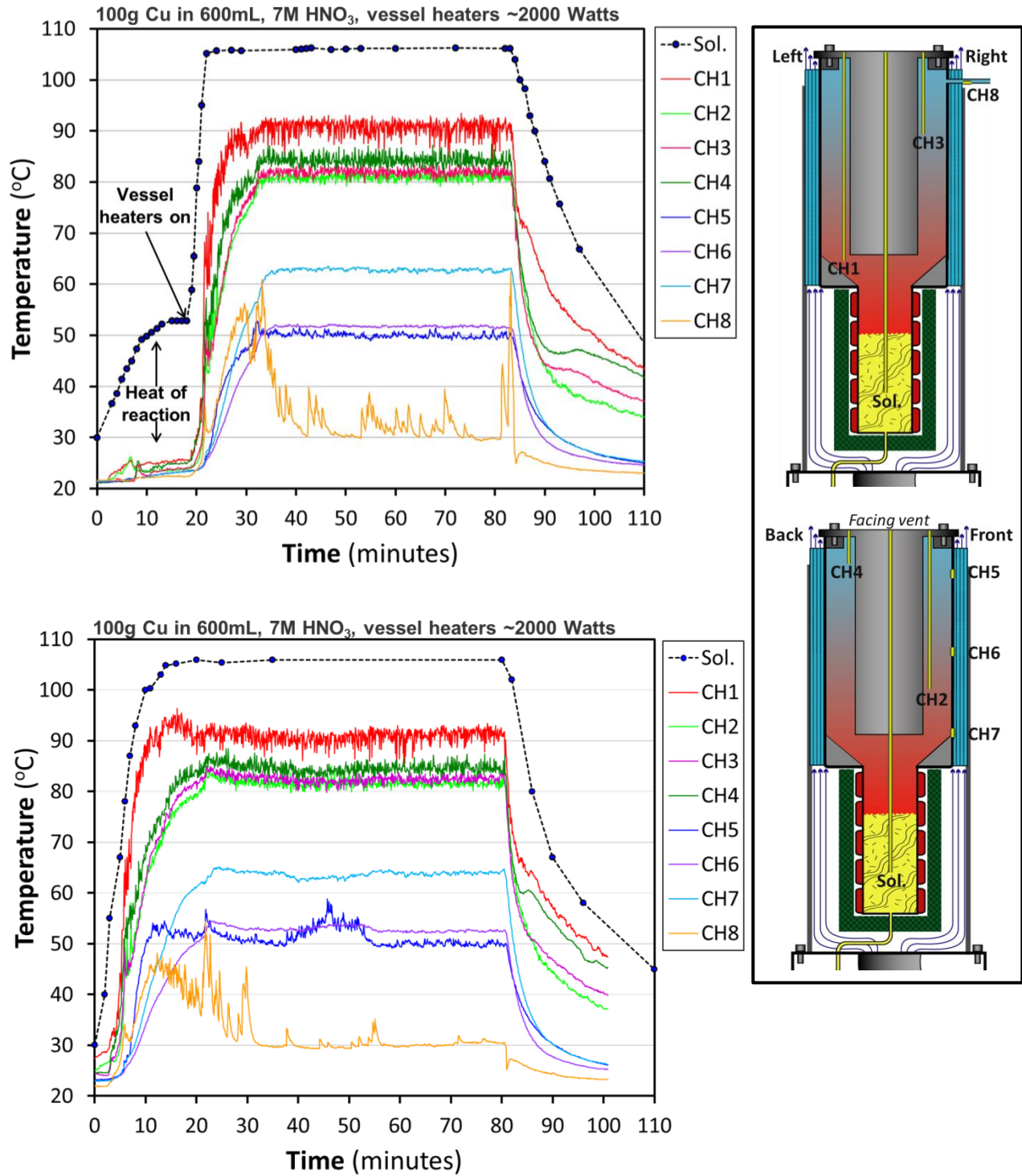


FIGURE 14 Data from Metal Dissolution Tests in which 100-g Copper Metal was Dissolved to Simulate the Dissolution of Uranium Metal (These experiments allowed us to observe the input of heat from the exothermic dissolution reaction and account for the NO_x and acid gases that are generated.)

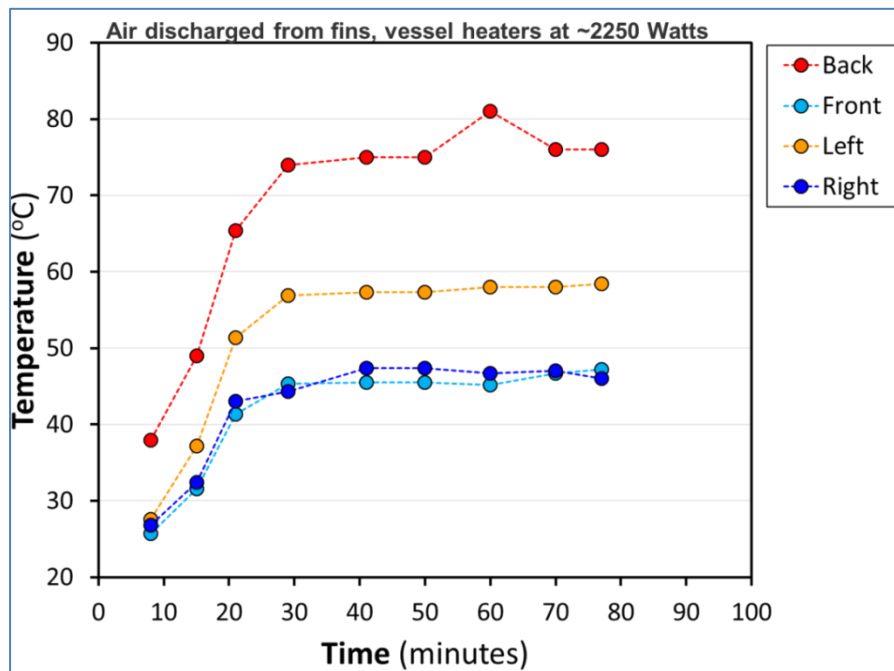
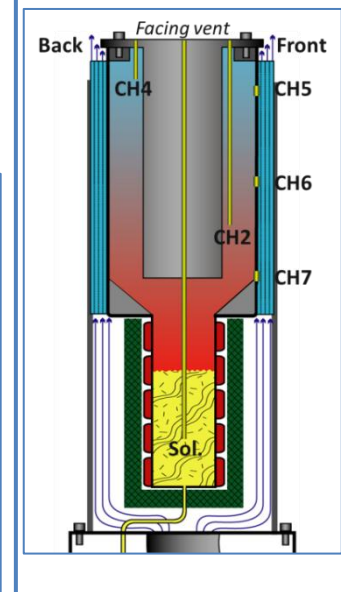
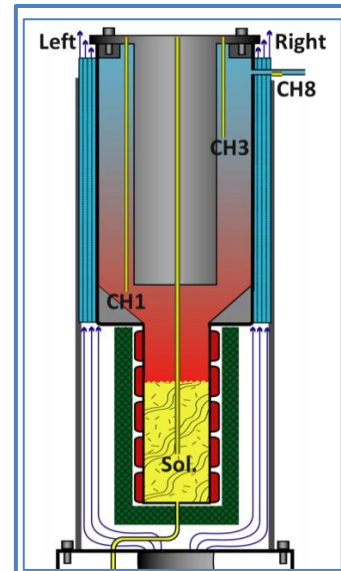
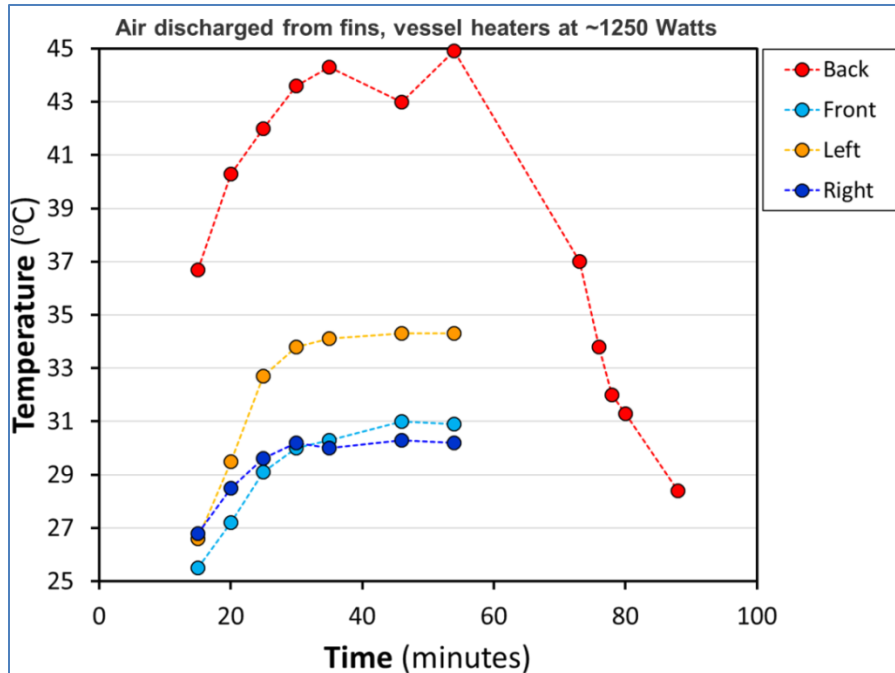


FIGURE 15 Temperature Profiles of Cooling Air as it Exits from the Cooling Fins (The data show that the heating of the cooling air is non-uniform around the dissolver. This non-uniform heat distribution does not significantly affect the efficiency of the condenser system [see text].)

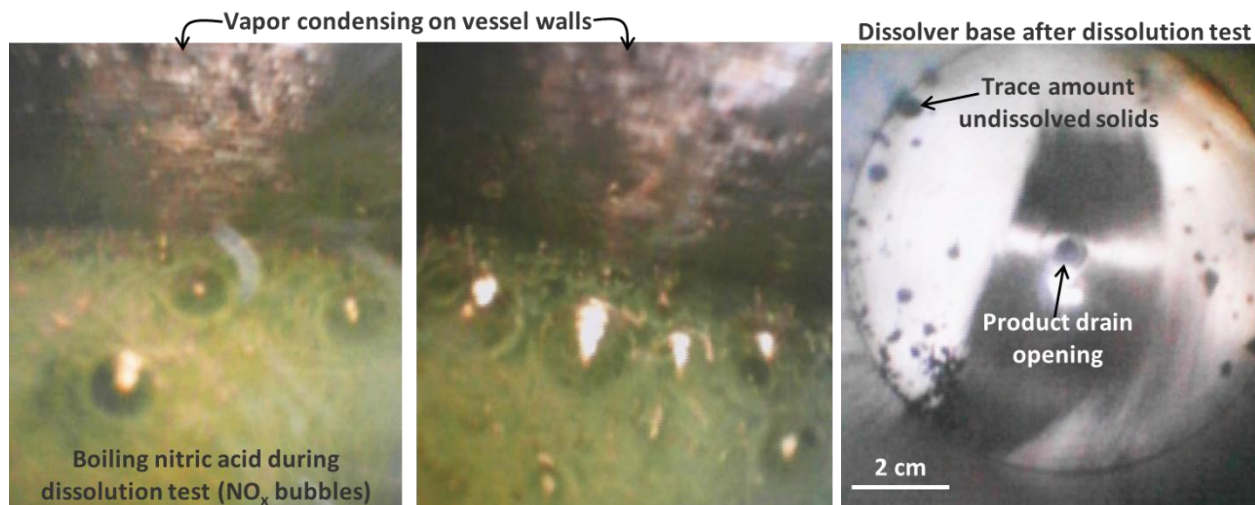


FIGURE 16 Photographs of the Dissolver Interior Taken with a 1-m-long, Fiber-Optic Camera with an LED Light Source (borescope) (The photos on the left were taken during a copper dissolution experiment and the one on the right was taken after the dissolution experiment to check the base of the dissolution vessel for undissolved solids.)

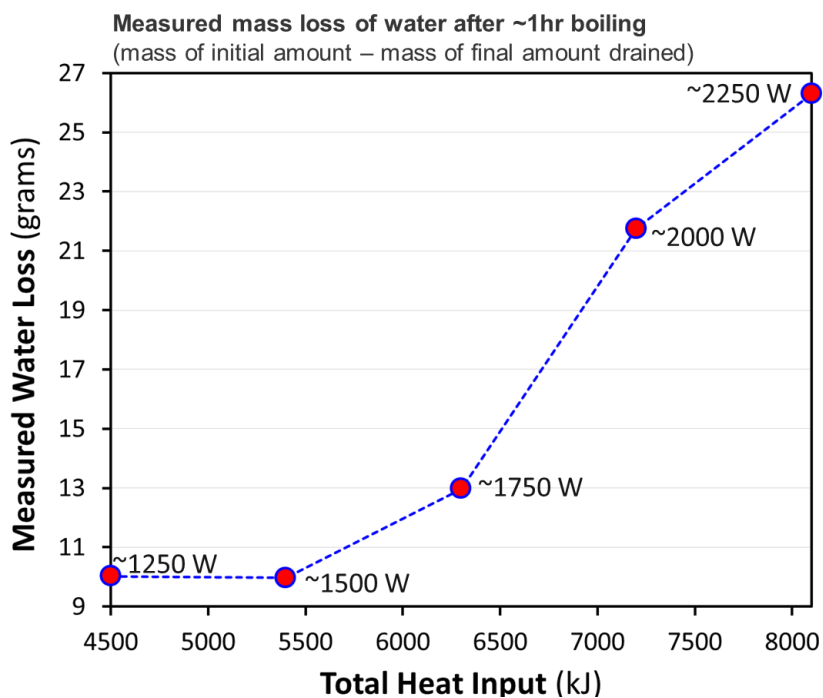


FIGURE 17 Compiled Water Loss Data from the Boiling Water Experiments

The temperature of the vent tube (CH8: orange line in Figures 11–14) is strongly affected by the presence or absence of a vent hose attachment. For example, when a hose is attached (results shown in Figure 11) the heat dissipation is rapid and the temperature of the vent opening is usually less than 30°C. However, when a vent hose is not attached the steady-state temperature of the vent opening (CH8) varies from 50°C for the 1750-W test up to 75°C for the 2250-W test. These observations suggest that the vent tube opening is heated by vapor “pumped” from the relatively higher temperature/pressure dissolver vessel to the lower temperature/pressure region around the vent opening. The condenser section wall near the vent opening (CH5) is 5°C to 15°C cooler than the vent tube thermocouple itself (Figures 12 and 13). Therefore, it is plausible that much of the vapor that is transported to the vent tube will condense and run back down into the dissolver vessel.

Results from loss of cooling experiments (example shown in bottom plot of Figure 13) demonstrate how sensitive the condenser/dissolution vessel system is to a sudden loss of air flow through the fins. After the system was allowed to establish a steady-state thermal profile at 2000 W, the cooling fan was abruptly turned off (around 40 minutes bottom plot, Figure 13). This resulted in a rapid increase in temperature of the condenser section wall (e.g., CH7) from 60°C to 85°C in approximately 4 minutes. When the fan was powered on again, the condenser wall temperature (CH7) decreased to 65°C in approximately 8 minutes. These results demonstrate the efficiency of the forced air/steel fin cooling system design. In the event that power to the cooling fan was permanently lost during a dissolution, all of the solution would vaporize and be passively contained (along with fission gases) at <125°C and <2 atm absolute within the off-gas reservoir (see Figures 3 and 4).

The data shown in Figures 11–15 also show that the walls of the condenser section (under the cooling fins: CH5, CH6, CH7) have a maximum steady-state temperature that varies from around 35°C for the 1250-W tests to approximately 70°C for the 2250-W tests. That is, for the experiments performed at the highest power (2250 W, Figure 13), there is a thermal gradient of approximately 40°C between the hottest part of the condenser walls and the hottest vapor temperature within the condenser. This thermal gradient is sufficient to cause efficient condensation and refluxing of the dissolver solution during a typical LEU foil dissolution run. However, due to the dynamic and turbulent nature of the convection cells that develop within the condenser section, some solution is lost to the vent tube as vapor.

Results from the copper dissolution tests (Figure 14) differ from those run with water at the same power (2000 W; bottom plot of Figure 12). As expected, the steady-state temperature of the dissolver solution is higher for the copper tests (106°C vs. 100°C) due to the presence of the non-volatile solute copper nitrate. However, the steady-state temperature of the vapor phase within the condenser section (CH1, CH2, CH3, CH4) is lower for the 2000-W copper dissolution test (Figure 14) relative to the vapor in the 2000-W water test (bottom plot of Figure 12). For example, the gas nearest the dissolution vessel (CH1) is approximately 104°C for the water test at 2000 W, but only 90°C for the copper dissolution test at 2000 W. This is believed to be due to the different thermal properties (heat capacity, thermal conductivity, etc.) of pure water vapor compared to those of nitrogen oxide and acid gases present in the dissolution tests. The temperatures of the condenser wall in the copper dissolution tests do not vary significantly from those measured in the corresponding boiling water tests.

A 1-m-long, fiber optic camera with an LED (light-emitting diode) light source (borescope) was used to examine the interior of the dissolver vessel during and after the dissolution tests (Figure 16). The examinations made while the dissolution was in progress were done to observe the nature of boiling dissolver solution (bubble size, vigorousness of reaction, degree of splashing, etc.) and to note the reflux of vapor as it condensed on the vessel walls. The examinations done after the dissolutions allowed us to observe the amount of undissolved solids so that we could confirm that the initial acid concentration and test duration were sufficient to completely dissolve the metal sample. Trace amounts of undissolved solids were observed following the dissolution tests which could be from impurities in the copper or strictly undissolved copper metal. (Figure 16).

The solution loss during these experiments is shown in Figure 17. The results show that approximately 10 mL of solution is lost from the dissolution vessel after 1 hour of boiling at powers < 1500 W. Approximately 26 mL of solution was lost after 1 hour of boiling for the 2250-W test. These results indicate that the forced air cooling system for the dissolver is efficient and could be used for a full-scale dissolution of irradiated foil, as long as the dissolver solution loss was accounted for. However, the results also indicate that the cooling system is not as efficient as it could be. The goal is to lose less than 10 mL of dissolver solution as vapor even for the highest wattage tests. Therefore, a second set of heat flow/dissolution experiments is planned to test the performance of an optimized cooling component for this ambient pressure dissolver system. Optimization involves fitting the dissolver with a more powerful cooling fan and optimizing the flow path of the cooling air into the fins (i.e., sealing any leaks in the steel sheath that forces the cooling air into the fins).

3.2 COMPARISON OF MATHCAD DISSOLVER SYSTEM DESIGN MODEL WITH TEST RESULTS

The purpose of Mathcad analysis is to compare thermal hydraulic theory calculated results to the results obtained from prototype tests.

Using the measured air flow velocity at the cooling fins discharge, the calculated pressure drop from the fan discharge to the fins discharge is in good agreement with the measured pressure drop. This result can be used to validate the pressure drop calculations for other flow rates. In addition, the measured flow rate and pressure drop across the fan is in reasonable agreement with the fan manufacturer's fan performance curve, which provides validation for future fan selection from these curves. The calculated temperature rise of the air across the fins is not in good agreement with that measured. This difference is probably due to the electrical power not being accurately set by the variable voltage controller. Ongoing performance tests include accurate electrical power measurement to the heaters to resolve this apparent discrepancy between calculated and measured temperature rise of air across the cooling fins. In addition, the variation in the measured azimuthal air temperatures at the fin discharge was not expected based on the original design calculations. This discrepancy is believed to be due to the non-uniform heating of the dissolver vessel by the band heaters. The setup for the ongoing tests has been altered to address this apparent discrepancy.

4 SUMMARY OF RESULTS AND DESIGN OPTIMIZATION

- A full-scale prototype of the ambient pressure, nitric acid LEU-foil dissolver system (capable of dissolving 250 g of irradiated LEU for Mo-99 extraction) was designed and tested using H₂O and Cu.
- Test results show that the cooling system of the dissolver (reflux condenser) is sufficient to remove all heat produced by the exothermic dissolution reaction and continuous decay heat of the irradiated foil.
- Simulated LEU-foil dissolutions at a maximum power of 2250 W (~8000 kJ heat input) showed that less than 5% of the dissolver solution was lost as vapor to the condenser vent (a maximum loss of 26 mL of a 600 mL dissolver solution). During actual operation of the dissolver, this vapor loss could be accounted for by altering the starting volume and concentration of the initial nitric acid solution to ensure complete dissolution of the LEU foil within a 30- to 40-minute timeframe.
- The vapor loss observations and some of the discrepancies between the Mathcad design model and the test results do, however, indicate that the cooling system for the condenser section of the dissolver can be further optimized. The optimization work that is underway involves simply fitting the cooling system with a higher pressure (max velocity) blower (cooling fan) and sealing any leaks in the steel sheath that forces the cooling air through the fins. It is anticipated that this relatively simple change will greatly increase the efficiency of the condenser section. A second set of performance tests with the improved cooling system are currently being set up.

5 FUTURE WORK IN PREPARATION FOR FULL-SCALE DEMONSTRATION

- The optimization changes to the dissolver system that were made based on the first set of performance tests (cooling system improvements) will be tested in a second set of experiments with the full-scale dissolver prototype. For these experiments, depleted uranium foils will be dissolved to more accurately simulate the LEU-foil dissolution.
- The performance tests of the off-gas traps for NO_x (NaOH trap) and iodine (copper metal or silver loaded zeolite) will be completed and these gas traps will be added to the dissolver system design.
- The complete dissolver system (including gas traps and off-gas reservoir) will be tested in a manipulator mock-up facility to ensure that the dissolver system can be easily operated at a production scale in a hot cell facility.

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