

Design, Fabrication, and Testing of an Acid-Dissolution Front-End Process for Current Mo-99 Recovery Processes: Optimization for Full-Scale Demonstration in Hot-Cell Facility

Chemical Science and Engineering Division

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Design, Fabrication, and Testing of an Acid-Dissolution Front-End Process for Current Mo-99 Recovery Processes: Optimization for Full-Scale Demonstration in Hot-Cell Facility

by

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DESIGN, FABRICATION, AND TESTING OF AN ACID-DISSOLUTION FRONT-END PROCESS FOR CURRENT Mo-99 RECOVERY PROCESSES: OPTIMIZATION FOR FULL-SCALE DEMONSTRATION IN HOT-CELL FACILITY

1 INTRODUCTION

The National Nuclear Security Administration's Global Threat Reduction Initiative Conversion Program (GTRI) develops technology necessary for converting civilian nuclear facilities that use high enriched uranium (HEU) to low enriched uranium (LEU) fuels and targets. The conversion of conventional HEU targets to LEU for the production of Mo-99 for medical use requires approximately five times the amount of uranium per target to maintain current Mo-99 yields. Under GTRI, Argonne National Laboratory (Argonne) is developing two frontend options for the current Mo-99 recovery processes to allow the use of LEU-foil targets: (1) an ambient pressure, nitric acid dissolution process; and (2) an electrochemical, basic dissolution process. The goal in developing both frontend options is to produce a product 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 Mo-99 at the end of processing. This work was done under the Mo-99 International Production Support, FY 2012 work package: 21.2.94.2.6 LEU Target – Foreign Production Development, ANL Activity 1.

In the first frontend option, the irradiated LEU foil and attached 10–15 micrometer Ni fission-recoil barrier is removed from the annular, aluminum clad target and dissolved in nitric acid. Results from performance tests using a full-scale experimental prototype of this nitric acid dissolver were presented in the March 2012 report entitled “Optimization of an acid-dissolution front-end process to produce a feed for the current Mo-99 recovery processes” by J. Jerden, J. Bailey, L. Hafenrichter, and G.F. Vandegrift. The March 2012 report also describes the baseline design for the nitric acid dissolver system. The recent work presented in this report involved refining the baseline design to facilitate full-scale testing of the system in a hot-cell facility. That is, the recent design work, discussed below, was done in preparation for a demonstration of the nitric acid dissolver technology using fully irradiated uranium foils within shielded cells that have specific footprint and operational height constraints.

2 FACILITY OPTIONS FOR HOT-CELL DEMONSTRATION OF NITRIC-ACID, LEU DISSOLVER SYSTEM

Two facility options are currently being pursued as locations for full-scale demonstration of the nitric-acid, LEU dissolver system: (1) the Institute for Nuclear Research (ICN) in Pitesti, Romania; and (2) the Radiochemical Engineering Development Center (REDC) and High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL). Argonne scientists have been in contact with facility personnel at ICN; however, no meetings have been held to explore the technical details of performing the frontend demonstrations in Romania.

An onsite meeting between Argonne scientists and ORNL hot-cell and reactor personnel was held in May 2012. The meeting included tours of the REDC and HFIR to determine the best cell locations for testing and the most feasible means of foil irradiation. A preliminary plan for the demonstration experiments was developed and some preliminary plans for addressing irradiation safety compliance issues were made. The ORNL personnel involved are currently developing an estimated schedule and cost matrix for performing the demonstrations.

From a technical standpoint, the most important issues discussed during the May 2012 Argonne–ORNL meetings were the maximum size limitations for getting the dissolver components into the hot cell, the in-cell foot-print dimensions, and the vertical manipulator working range required. It was determined that, for this project, the equipment and dissolver components going into the REDC hot-cell cubicles can be no larger than 45 cm wide by 91 cm long and 192 cm tall. The ideal in-cell footprint for the dissolver system is 76 cm by 51 cm. With these dimensions in mind, the dissolver system design was refined to minimize space requirements for the demonstration.

3 COMPARISON OF NITRIC-ACID, LEU DISSOLVER SYSTEM BASELINE DESIGN WITH THE HOT-CELL SPACE OPTIMIZED DESIGN

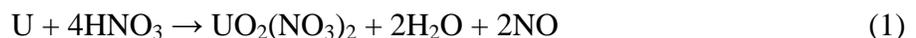
3.1 SUMMARY OF BASELINE DESIGN

As reported in Jerden et al. (2012), an ambient-pressure nitric acid dissolver system was designed to allow the dissolution of up to 250 g of irradiated LEU-foil and attached Ni fission recoil barrier. 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), and to trap iodine gas for possible extraction of economically important iodine isotopes (noble fission gases will be passively contained).

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

The volume and concentration of nitric acid used depends on the amount of LEU being dissolved and the desired final acid concentration of the “product” solution. Controlling the final acid concentration is important for optimizing the Mo-99 extraction step that follows dissolution. The volumes and concentrations of acid, as well as the amount of nitrogen oxide gas that will be produced, are determined by the following general reactions:



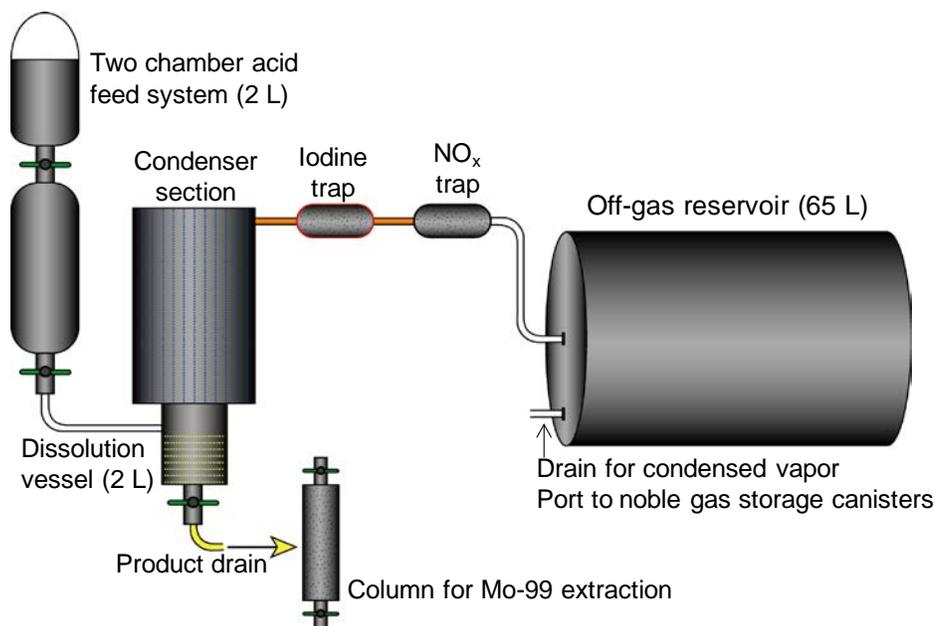
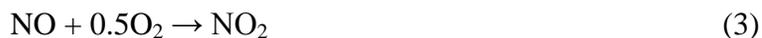


FIGURE 1 Schematic Flow Diagram Showing Components of the LEU Nitric Acid Dissolver System and the Mo-99 Extraction Column

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



When water vapor and oxygen are present, NO₂ is rapidly converted to both nitrous and nitric acid vapors [HNO₂(g) and HNO₃(g)], which dissolve in condensed water and flow back down into the dissolver vessel. An air-cooled steel condenser section with metal cooling fins condenses water and acid vapors so that they are not lost to the off-gas volume during the dissolution process. Figure 2 shows a schematic drawing of the condenser section and dissolver vessel that highlights the condensation process.

Most of the off-gas from the dissolver will consist of the NO_x and acid vapors (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% U-235, power = 1.9×10^{-3} MW, flux = 2.1×10^{14} N/cm² sec, and a burnup = 1.59×10^{-2} MW days for 200 hr. The most abundant off-gas species is predicted to 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. 2012).

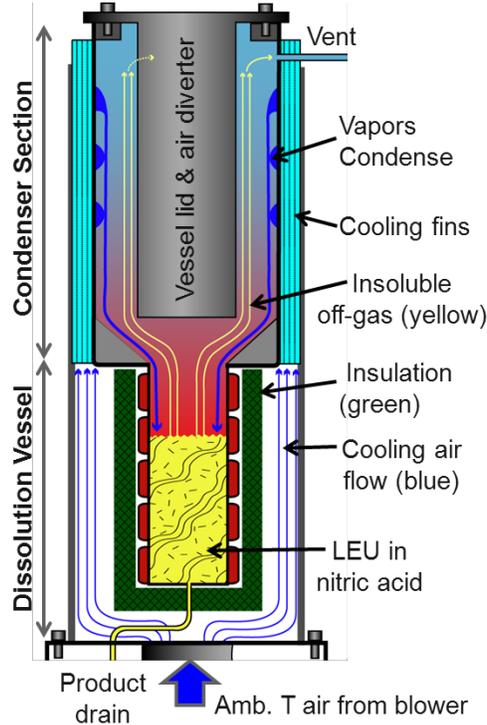


FIGURE 2 Schematic Cross-Section through the Condenser Section and Dissolution Vessel of the Dissolver System

Based on enthalpy of dissolution and decay heat calculations the cooling system for the LEU nitric acid dissolver must be able to remove a maximum of 2000 W. 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. Results from these tests are presented in Jerden et al. (2012).

Based on the experimental results and a mathematical model that was implemented in Mathcad, the following characteristics were determined for the baseline design of the dissolver system:

- Sizing and spacing of metal cooling fins for the condenser section of the dissolver—Calculated based on the required flow velocity as determined by the convective heat removal from the fins.

- Cooling air velocity and the pressure drop across the cooling fin section—The total air volume flow rate and temperature rise of the air across the fins was 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—It was assumed that all of the reaction heat and off-gas 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. Heat loss by radiation and heat loss by natural convection were 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 (Figure 2) 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; 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.

The baseline design for the nitric acid dissolver system is shown in Figure 3. The dimensions of the dissolver system based on the baseline design are shown in Figure 4. All components of the system are 304 stainless steel except for the heat sink rings attached to the off-gas reservoir, which are aluminum. For a more detailed description of dissolver components, see Jerden et al. (2011a,b).

3.2 SPACE-OPTIMIZED DISSOLVER DESIGN AND SUMMARY OF DISSOLVER SYSTEM OPERATION

The footprint of the space-optimized design for the dissolver system is shown in Figure 5. Note that in the refined design the length of the long dimension of the footprint has been decreased from 130 cm in the baseline design to 77 cm in the new design. The tallest part of the system in the new design is the off-gas reservoir (145 cm tall), which is placed in a vertical rather than horizontal position (compare Figures 3 and 4 to Figures 5 and 6). Figures 6 and 7 show conceptual cross-sections of the new design and summarize the steps involved in operation of the system. The space-optimized design is a refinement of the baseline design that minimizes the dissolver footprint and overall height so that the system can be used within the 76 cm by 51 cm hot cell space available at the REDC at ORNL.

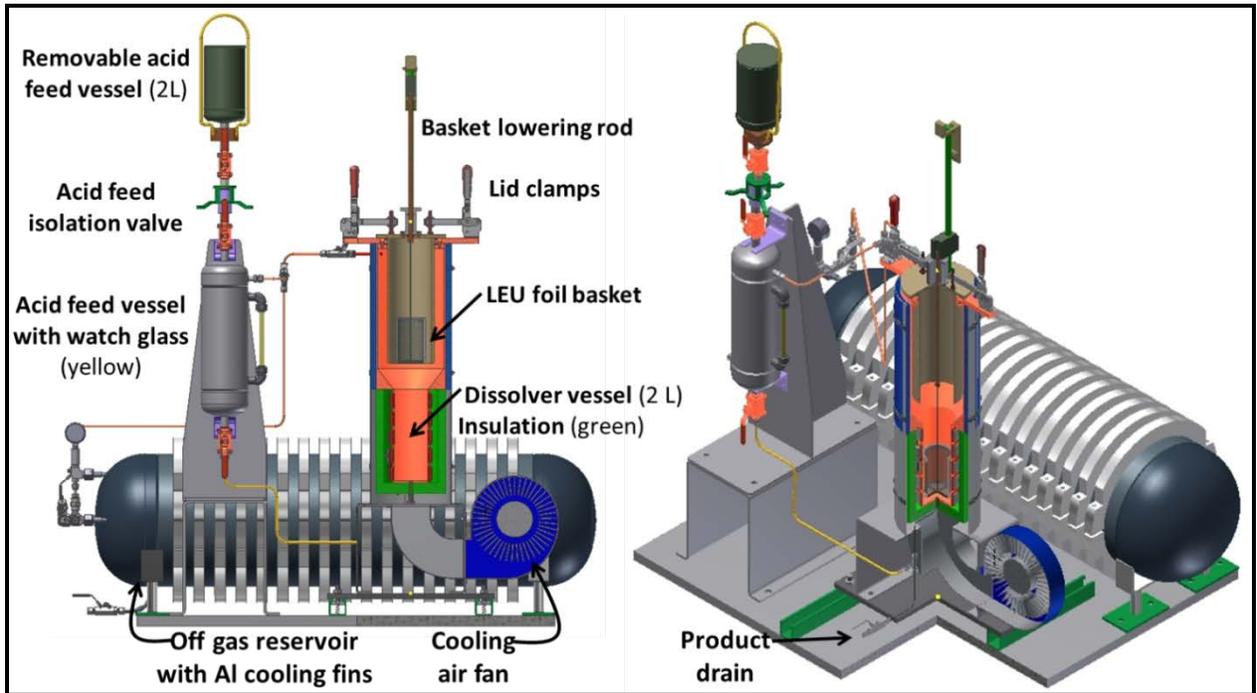


FIGURE 3 Conceptual Diagrams of Nitric Acid Dissolver System

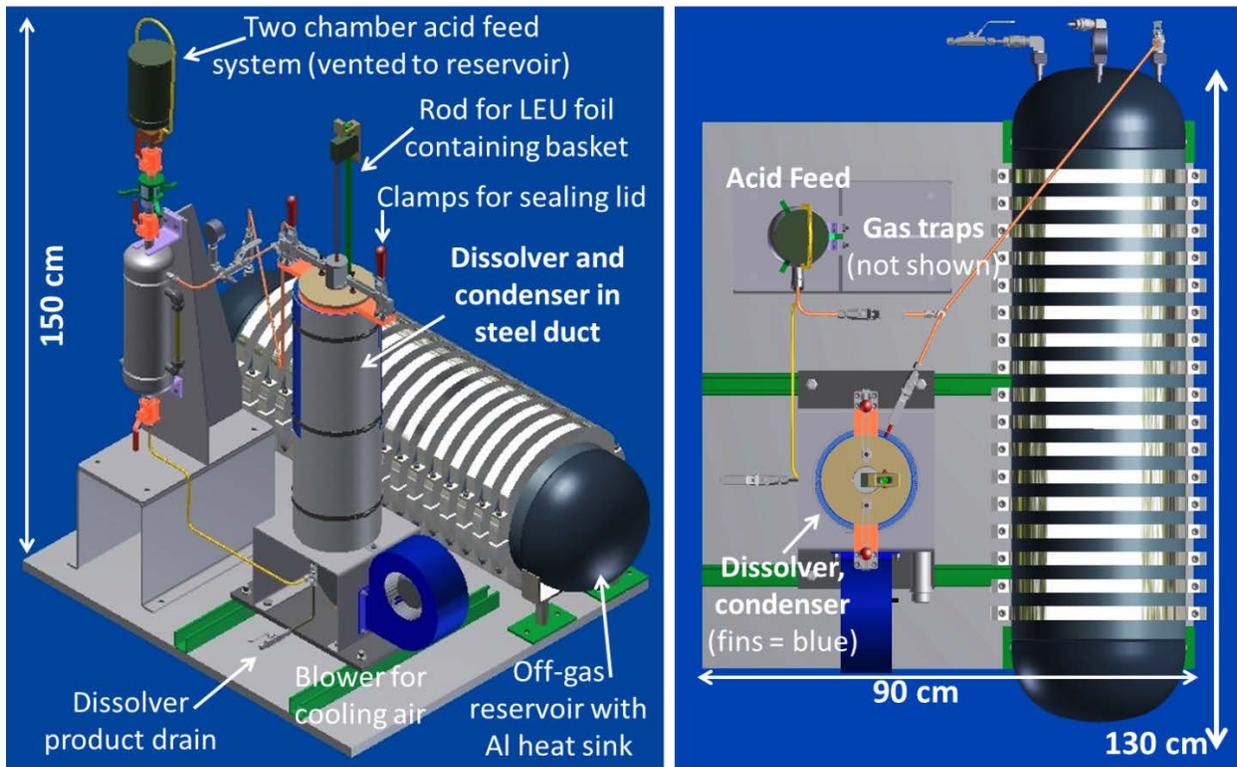


FIGURE 4 Conceptual Diagrams of Dissolver System Showing Dimensions of Baseline Design

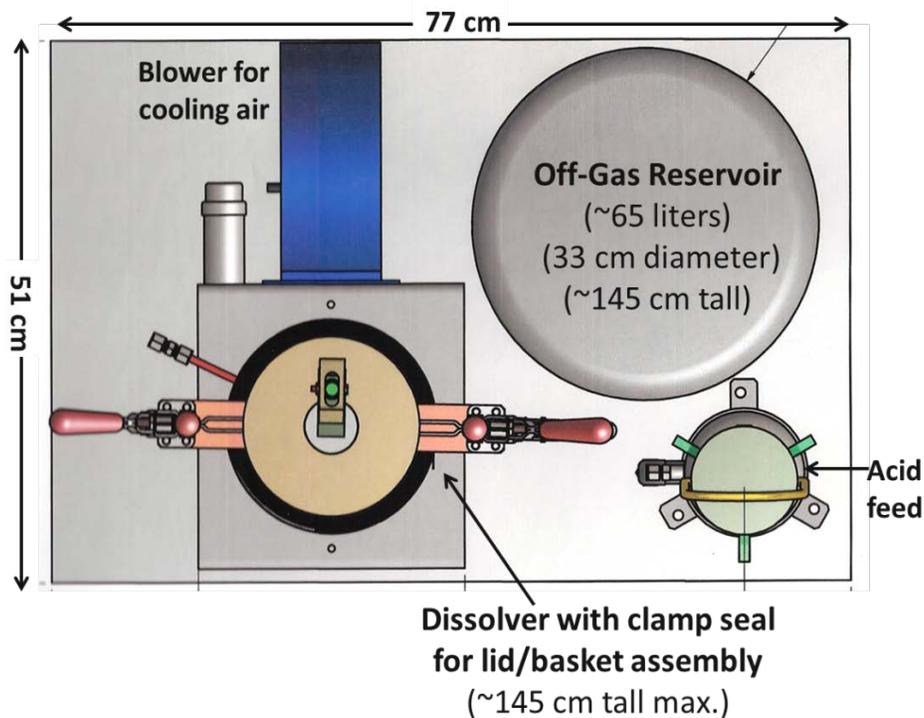


FIGURE 5 Footprint of the Hot Cell–Space Optimized Nitric Acid Dissolver System Design

Operation of the dissolver system (summarized in Figure 7) involves the following steps:

- 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.
- Nitric acid is added to the vessel using the two-chamber acid feed system designed to avoid pressurization of the acid bottle in the event that the dissolution reaction begins instantaneously when the acid addition step is started.
- Heating elements attached to the base of the dissolver vessel are energized to bring acid to boiling temperature (if acid does not boil due to the decay heat of the irradiated LEU foil). The acid temperature is measured using a standard thermocouple (not shown in the figures).
- The dissolver vessel is cooled during the reaction by forced air blown from the base of the unit. 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 the steel cooling fins attached to the condenser part of the dissolver system.

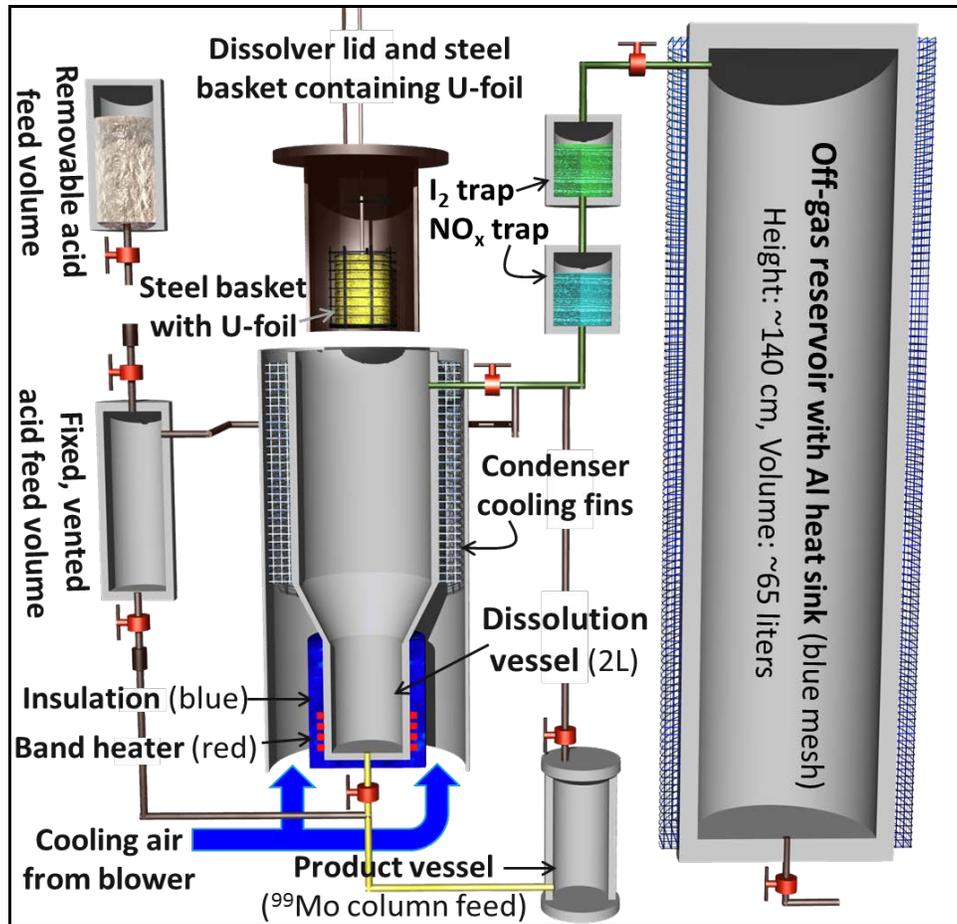


FIGURE 6 Conceptual Diagram of the Components of the Hot Cell-Space Optimized Nitric Acid Dissolver System Design

- Following approximately 1 hr of dissolution in boiling acid (enough time to completely dissolve 250 g of uranium foil) the product solution is drained into a vessel that is vented to the off-gas reservoir. This product vessel will serve as the feed for the Mo-99 extraction step.
- The dissolver lid is removed and the basket is examined (using remote camera if necessary) to ensure that dissolution was complete.

The operation of the dissolver has been tested during the heat flow and dissolution experiments described in Jerden et al. (2012). Operation of the acid feed system and use of the uranium foil basket assembly were tested separately by hand to ensure ease of remote operation.

As stated above, 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 (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

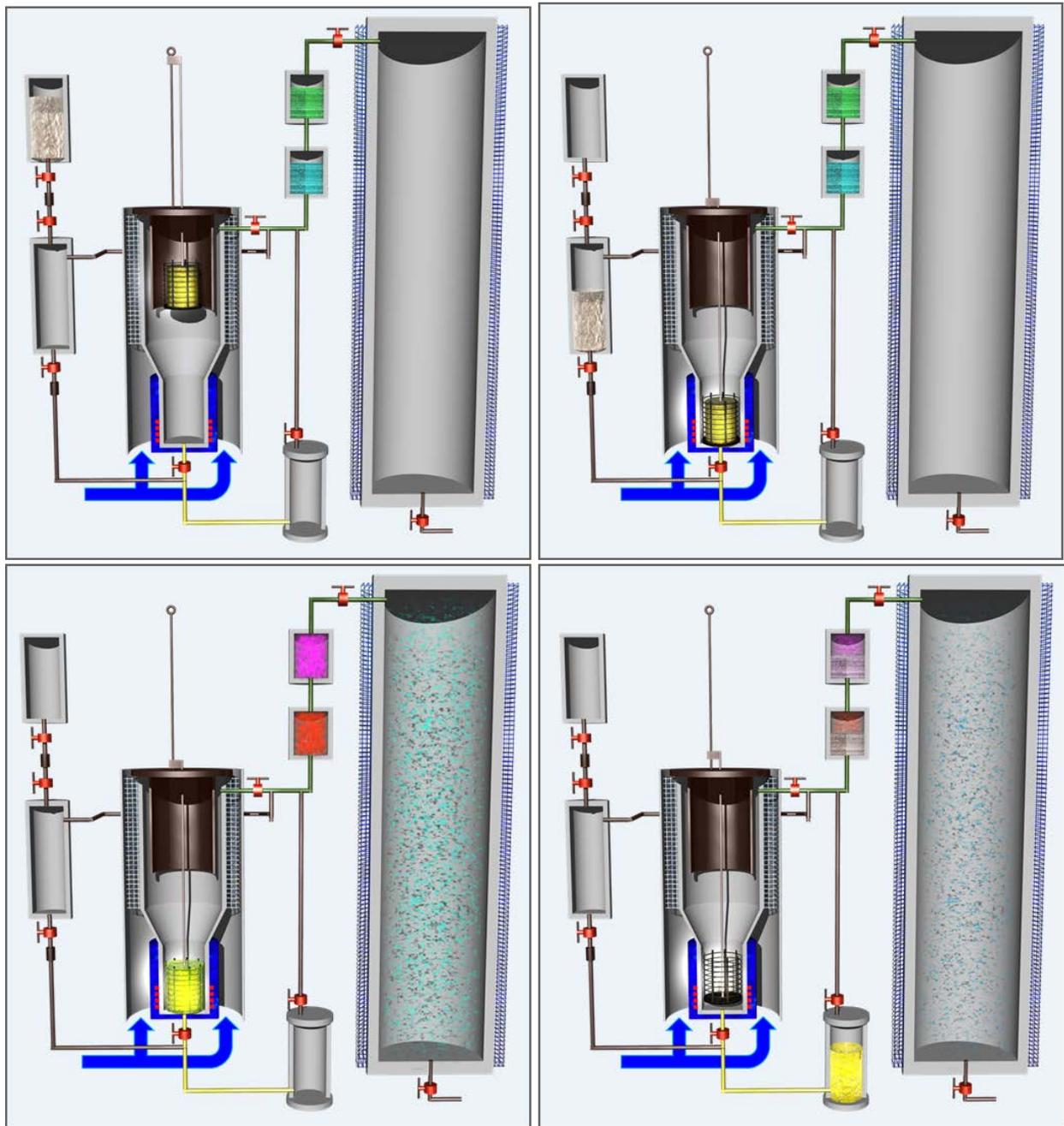


FIGURE 7 Conceptual Diagrams of the Hot Cell-Space Optimized Nitric Acid Dissolver System Showing the Four Main Operation Steps (see Figure 6 for component labels) ([1] fill removable acid feed vessel and load irradiated foil into steel basket [upper left]; [2] drain acid into permanent, vented acid feed volume [close valves] and lower irradiated foil basket into dissolution vessel [upper right]; [3] drain acid into dissolution vessel to start foil digestion: off-gas reacts with gas sorbents [NO_x shown in orange; I_2 as magenta; Xe, Kr as blue] [lower right diagram]; [4] product of foil digestion is drained into vented vessel that will serve as the feed for the Mo-99 extraction columns.)

not escape from dissolver vessel condenser section. If these gases escaped during the dissolution reaction the reaction would slow significantly or event 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 used in our first set of heat flow experiments. A full-scale prototype dissolver 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 experimental setup and results from these experiments are presented in Jerden et al. (2012).

4 SUMMARY OF RESULTS AND DESIGN OPTIMIZATION

- The baseline design for the full-scale ambient pressure, nitric acid dissolver system for LEU foils was refined to facilitate a demonstration of this technology within an existing hot cell facility.
- Two hot cell facility options are currently being pursued as locations for full-scale demonstration: (1) ICN in Pitesti, Romania; and (2) REDC and HFIR at ORNL.
- An onsite meeting between Argonne scientists and ORNL hot cell and reactor personnel was held in May 2012. The meeting included tours of the REDC and HFIR to determine the best cell locations for testing and the most feasible means of foil irradiation.
- A preliminary plan for the demonstration experiments was developed and some preliminary plans for addressing irradiation safety compliance issues were made. The ORNL personnel involved are currently developing an estimated schedule and cost matrix for performing the demonstrations.
- The most important issues discussed during the May 2012 Argonne–ORNL meetings were the maximum size limitations for getting the dissolver components into the hot-cell, the in-cell foot-print dimensions, and vertical manipulator working range required. With these dimensions in mind, the dissolver system design was successfully refined to minimize space requirements for the demonstration.

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