Design of Acidic Dissolver for Processing of Irradiated LEU-Annular Targets

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by

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September 2021
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DESIGN OF ACIDIC DISSOLVER FOR PROCESSING OF IRRADIATED LEU-ANNULAR TARGETS

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September 2021

1. INTRODUCTION

Argonne National Laboratory, with support from The United States Department of Energy (DOE) National Nuclear Security Agency’s (NNSA’s) Office of Material Management & Minimization (M³) is developing technologies for a number of domestic entities in their pursuit of production of ⁹⁹Mo/⁹⁹mTc medical isotope without the use of highly enriched uranium (HEU). One of the potential domestic ⁹⁹Mo producers, Eden Radioisotopes (Eden), is developing a Triga-sized, all-target reactor system licensed from Sandia National Laboratories as a direct result of their involvement in ⁹⁹Mo in the 1990s. This reactor system uses up to 60 of the Argonne-designed annular Low Enriched Uranium (LEU)-foil target; each operates at up to 30 kW. The annular target contains approximately 137 g (±12 g) of LEU as a 250-µm (±25 µm) thick uranium foil covered on all sides by a thin Ni-foil fission barrier, and two aluminum tubes that form the outer and inner walls of the cladding cylinder. Eden expects to process between three and five targets per process batch, limiting the ⁹⁹Mo batch inventory to less than 5000 Ci, within a few hours of their discharge from the reactor, meeting U.S. ⁹⁹Mo demand with 10 process batches per week.¹ The cladding is mechanically removed before the uranium foil and Ni fission-recoil barrier are dissolved in nitric acid for molybdenum recovery.

Therefore, the goal for this project was to design an acidic dissolver that can handle up to 750 g of LEU metal foils and total mass of 16 g of Ni that is present as a fission fragment barrier to produce the feed solution with final desired concentration of 450 g-U/L in 1M HNO₃. The volumes and concentrations of acid as well as the amount of nitrogen oxide gas (NOₓ: NO, NO₂, N₂O₄) that is produced are determined by the following general reactions:

\[ U + 4\text{HNO}_3 \rightarrow U\text{O}_2(\text{NO}_3)_2 + 2\text{H}_2\text{O} + 2\text{NO} \] (1)

\[ \text{Ni} + 8/3\text{HNO}_3 \rightarrow \text{Ni}(\text{NO}_3)_2 + 4/3\text{H}_2\text{O} + 2/3\text{NO} \] (2)

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

\[ \text{NO} + 0.5\text{O}_2 \rightarrow \text{NO}_2 \] (3)

In the presence of water vapor, NO, and oxygen gas, NO₂ can be readily converted to nitrous and nitric acid vapors [HNO2(g) and HNO3(g)] (eq. 4 & 5), which will dissolve in condensed water and flow back down into the dissolver:

\[
3\text{NO}_2 + \text{H}_2\text{O} \rightarrow 2\text{HNO}_3 + \text{NO} \tag{4}
\]

\[
\text{NO} + \text{NO}_2 + \text{H}_2\text{O} \rightarrow 2\text{HNO}_2 \tag{5}
\]

If the NO is fully transferred back to acid, fumeless dissolution can be achieved. However, besides production of NO\(_x\) gasses, dissolution of irradiated U targets produces fission gasses, consisting mostly of iodine, xenon, and krypton isotopes. Fission iodine could be trapped using copper metal cold trap and any NO\(_x\) gasses that escape from the dissolver can be neutralized using NaOH trap. Xe and Kr gasses would be allowed to decay by exhausting through a decay line.

2. BACKGROUND: PREVIOUS ARGONNE DESIGNED ACIDIC URANIUM DISSOLVERS

2.1 HIGH-PRESSURE DISSOLVER

The high-pressure dissolver was developed by Argonne scientists in 2009\(^2\). The dissolver (Figure 1)\(^2\) was constructed of 304 Stainless Steel with a volume of 360 cm\(^3\). It consists of a cylindrical body and a separate lid assembly. The lid is a flanged lid with an O-ring seal, and a “cross” fitting with a pressure gauge, a plug valve attached to a quick-connect plug, a plug valve attached to a fitting for adding acid into dissolver, and a pressure relief valve calibrated to release at 700 psig. Hydrostatic pressure tests have verified that the dissolver can operate at 800 psig at 25\(^{\circ}\)C. Because the dissolver is supported within a cylindrical steel sleeve with an open base and a flange at the top, the dissolver remains vertical during dissolution. An electric heat gun blows hot air into the support sleeve to increase and maintain the temperature. The heat gun also provides the air for cooling (heater turned off) when the dissolution reaction is complete. Pressure changes during dissolution of 5.58 g of U metal in 40 mL of 3.3M HNO\(_3\) is shown in Figure 2.

The initial concentration of nitric acid was chosen so that the final concentration (after complete dissolution) would be 1 molar. The final acid concentration following the complete dissolution of uranium in experiment was 1.13 molar (as determined by titration). This indicates that using reaction (eq. 1) to determine the starting acid concentration is valid, although the actual stoichiometry of the dissolution is more complicated than is implied by the reaction (i.e., NO as the only gas product). The gas chromatography results confirmed the presence of N\(_2\) and NO\(_2\) and a small amount of O\(_2\), N\(_2\)O and CO\(_2\). The lack of NO is believed to be due to

presence of O₂ (air). Moreover, higher final HNO₃ concentration indicates that some of the NOₓ was recovered as either HNO₂ or HNO₃ acid.

FIGURE 1. Photograph of dissolver (left), dissolver with copper gas trap attached (middle), and schematic drawing of the dissolver set-up showing the cryogenic gas trap (right).

FIGURE 2. Pressure history for an experiment in which 5.58 grams of uranium metal were dissolved in 40 mL of 3.3 molar nitric acid using the dissolver shown in Figures 1 and 3 (below).
2.2 AMBIENT PRESSURE DISSOLVER

An ambient pressure, nitric-acid-dissolver system (Figure 3) was designed in 2013 for the dissolution of up to 250 g of irradiated LEU foil and associated fission recoil barrier metal (e.g., Ni), and to operate at pressures less than 2 atm (absolute) and at temperatures less than 125°C. The dissolver consists of a 304 stainless steel vessel (2-L volume) connected to an approximately 65-L (30 cm × 90 cm) off-gas reservoir. The dissolver vessel is open to the off-gas reservoir during the dissolution process. The volume of the reservoir and the sizing of the aluminum heat sink were 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.

![Dissolver test set-up in radiological hood](image)

**FIGURE 3.** Full-scale ambient pressure dissolver prototype for dissolution of up to 250 g-U foil.

Experimental results showed that the cooling system of the dissolver (reflux condenser) was sufficient to remove all heat produced by the exothermic dissolution reaction and continuous decay heat of the irradiated foil. The dissolution of 133 g of depleted uranium (DU) foil was completed successfully in approximately two hours. No measurable solution loss was detected during DU dissolution tests; however, NO\textsubscript{x} gas was observed escaping intermittently during the dissolution process. The dissolution time could be further minimized by optimizing the starting acid concentration to account for the effects of the loss of NO\textsubscript{x} and acid gases from the dissolver during dissolution\textsuperscript{3}.

### 2.3 FUMELESS DISSOLVER

Based on eq. 1, it is apparent that per each mole of dissolved U metal, two moles of NO gas can form. In addition, presence of Ni leads to additional formation of NO gas. If NO gas is not recovered to NO\textsubscript{2} according to eq. 3, and then to HNO\textsubscript{2} and HNO\textsubscript{3} according to eq. 4 & 5, the expected volume of NO\textsubscript{x} gas produced by dissolution of 750 g of U metal is \(\sim 180\) L at 212° F (100° C). If the dissolution process is to be performed at or under atmospheric pressure, the dissolution apparatus requires a large holding tank to contain all NO\textsubscript{x} gasses produced. If NO\textsubscript{x} gasses could be effectively recovered (eq. 3-5), requirement of a large holding tank to contain the NO\textsubscript{x} gasses could be essentially eliminated. Experimental work by Gresky et al.\textsuperscript{4} discusses several options. Addition of oxygen into the dissolution system could yield to formation of uranyl nitrate according to eq. 6:

\[
U + 2\text{HNO}_3 + 1.5\text{O}_2 \rightarrow \text{UO}_2(\text{NO}_3)_2 + \text{H}_2\text{O} \tag{6}
\]

\[
2\text{NO} + \text{O}_2 \rightarrow \text{N}_2\text{O}_4 \tag{7}
\]

\[
\text{N}_2\text{O}_4 + \text{H}_2\text{O} + 0.5\text{O}_2 \rightarrow 2\text{HNO}_3 \tag{8}
\]

However, it was observed that nitrogen was a product of reaction in the ratio of 0.04-0.05 moles N\textsubscript{2} per mole of U. Moreover, there is a potential for possible operational hazard due to exothermic reaction between U metal and oxygen in dry conditions, and oxygen would have to be added into nitric acid solution or headspace above the liquid. Efficiency of oxygen addition for both scenarios needs to be experimentally confirmed and will be part of experimentation plan for future work. Another alternative option for NO\textsubscript{x} removal is by their absorption in 70 % HNO\textsubscript{3} according to eq. 9-12:

\[
2\text{NO} + \text{HNO}_3 + \text{H}_2\text{O}, \text{ as } 70\% \text{ HNO}_3 \rightarrow 3\text{HNO}_2 \tag{9}
\]

\[
2\text{NO}_2 + \text{HNO}_3 + \text{H}_2\text{O}, \text{ as } 70\% \text{ HNO}_3 \rightarrow 2\text{HNO}_3 + \text{HNO}_2 \tag{10}
\]

---

\textsuperscript{4} Gresky, AT, Snyder, MD, Blair, JA, Lovelace, RC, Corbin, LT, Tyson, WR, Recovery of nitrogen Oxides and Rare Gas Fission Products from the Nitric Acid Dissolution of Irradiated Uranium, Oak Ridge National Laboratory Report 1208, April 1952.
According to these reactions, it would theoretically be possible to achieve absorption of ~700 mL of gas/mL of solution. Based on experiments performed, ~5 mL of 70 % HNO₃ was needed to capture 200 mL of gas/g-U. Therefore, it is of great practical importance to investigate the removal of NOₓ gasses from the system to eliminate high pressure buildup in the dissolver or necessity for a large holding tank.

3. PROPOSED DESIGN OF AN APPARATUS FOR DISSOLVING IRRADIATED URANIUM METAL FOIL TARGETS

Initial discussions with EDEN and development of technical parameters were centered on a dissolver operating at below atmospheric pressure. This approach would make the dissolution process intrinsically safe as any breach of the primary containment fission gasses would not leak into the atmosphere of the hot cell. Extensive discussions of the dissolution parameters led to significant expansion of the scope of the planned experiments and dissolver specifications.

3.1 CHOOSING DISSOLVER DESIGN PARAMETERS

It should be noted that some of the design parameters need to be tested experimentally. For example, the major concern is dissolution time of up to 750 g of irradiated U-metal foils with foil thickness of 0.25 mm, which is significantly higher than thicknesses of the foils used in demonstrations for the previous dissolver designs [Ref and thickness]. It is expected that thicker foils would require longer time to dissolve. Moreover, if NOₓ gasses would be simply captured and not recovered (eq. 9-12), the concentration of nitric acid would drop significantly during the dissolution and lead to slower dissolution rates.

Therefore, the dissolver is designed in such a way that produced NOₓ gasses are intercepted by the water “raining” from the condenser. Another option is to re-introduce the NOₓ gasses back into solution or addition of O₂ to recombine the NOₓ gasses. For the testing purposes, not all hot cell friendly features like quick disconnects and tele-manipulator-friendly operations will be implemented; those would be implemented in later stages as needed. The set of system parameters that allows experimental flexibility is listed below:

Operating parameters

- Pressure: 14 - 300 psi
- Peak temperature: 100 - 180 C
- Uranium mass: 375 - 750 g
- Uranium enrichment: 19.8 %
- Uranium foil thickness: 250 ±25 µm
Number of targets: 3 - 5
Uranium mass per target: 137±12 g
Fission fragment barrier: Ni
Ni mass: 4 - 25 g
Initial nitric acid concentration: <68 %
Final desired HNO₃ concentration: 1 M
Final desired U concentration: 450 g-U/L
Nitric acid volume: 0.5 - 2.5 L
Total dissolution time: ~2.5 h

Other considerations

Basket size: 18” high, 3.15” diameter
Material: SS304L, SS316 (or other material compatible with nitric acid and elevated temperature)
Size: dissolver must fit into the frame hood and components must fit into 55 gallon drum for disposal
Connections: assembled with hands
Heating: electrical, 110 V, <2 kW
Cooling: water
Instrumentation: thermocouples (5), pressure transducers (2)
Procurement quality level: C - for prototype
Cooling coil above the solution for vapor condensing and rain down screen
Acid transfer via pump or syringe
Process solution transfer via pump (pull)
Welded construction except connections
NOₓ recirculating or trapping
Frit on the bottom of the dissolver to capture undissolved particles
Iodine trapping
Controls for pressure/flow between volumes
Safety relief valve
Dissolution vessel can be separated to add foils and can be connected back

The Piping and Instrumentation Diagram (P&ID) of the proposed setup is shown in Figure 4. In this design, we can replace or eliminate some of the traps, operate the dissolution process at significant pressure and elevated temperatures, and facilitate the dissolution process by introducing oxygen or recirculating the NOₓ gases produced in the dissolution process.
FIGURE 4. Piping and Instrumentation Diagram (P&ID) for the uranium foil targets dissolution test bench.

3.2 DESIGN OF THE PROTOTYPE

Drawings of the dissolver assembly prototype are shown in Figures 5 and 6. The assembly would be fabricated from off-shelf standard pipe and fittings. The assembly is designed to meet The American Society of Mechanical Engineers (ASME) pressure codes operating at maximum pressure of 300 psig at saturation temperature. Materials of construction are as noted on the drawings.

The target elements are inserted in the basket located in the bottom chamber of the dissolver. The chamber is evacuated and then filled to the top of the elements with acid. Band heaters, mounted on the outside of the lower chamber pipe, heat the solution in order to initiate the dissolving process. Once the exothermic reaction begins, the solution is heated to the vapor pressure (boiling point) and the water vapor (steam) rises to the cooling coils where it condenses. The water and acid vapor mixture is assumed to be all water vapor in order to simplify the thermal hydraulic calculations. This assumption is justified because the calculation result indicates that the condenser is significantly over designed and therefore is adequate for
mechanical design purposes. The liquid water then falls back into the solution. If the water vapor does not fully condense or the generated gas ($\text{NO}_x$) is not fully absorbed, the vapor/gas will flow through the holes in the top plate and through the $\frac{1}{2}$” tube to the I$_2$ and NO$_x$ traps to the reservoir tank.

FIGURE 5. Cross section of the assembled dissolver.
3.2.1 Condenser Coil Sizing

The assumed cooling coil configuration is shown in Figure 7. There are 3 columns with 20 coils per column. A manual calculation is performed to determine the effective heat removal due to condensing of the saturated water vapor from the process chemical reaction. The average evaporation rate of the water is determined from the given total heat generation, due to the reaction, divided by the total time of reaction. This rate is multiplied by 2 in order to account for the non-uniform rate of reaction. In general, the rate of reaction initially increases to a maximum and then decreases towards the end of reaction. Doubling the average rate provides a conservative estimate for the maximum heat generation rate (1.1 kW) based on this rate of reaction behavior. Calculations are based on the Wigner-Way formula (13)

$$P_d(t) = 0.0622 \cdot P_0 \left[ t^{-0.2} - (t_0 + t)^{-0.2} \right]$$  \hspace{1cm} (13)

where

- $P_d(t)$ = thermal power generation due to beta and gamma rays
- $P_0$ = thermal power before shutdown
- $t_0$ = time, in seconds, of thermal power level before shutdown
- $t$ = time, in seconds, elapsed since shutdown.
The decay heat for the 750 g LEU is calculated to be a constant 0.75 kW. In these calculations, we assume five 150 g targets irradiated at 30 kW power per target for 21 days and cooled down for 6 hours prior to dissolution. The total peak heat generation in the dissolver is 1.85 kW. This heat value is what is required before removal by the condenser.

The calculations assume that the condenser is near atmospheric pressure and at the saturated temperature of 212°F (100°C), assuming 100% water vapor. It can be shown, by inspection of the equations, that at higher pressure and temperature conditions the condenser efficiency improves and the atmospheric condition is the worst case when considering condensing rate.
First, a classic correlation for condensing heat transfer (eq. 14) is used to determine the effective convective thermal coefficient for condensing on vertical stacked coils at a given surface temperature on the coils. All variables are functions of condenser geometry and water (liquid and vapor) properties at the operating temperature and pressure.

\[
hc_{fg} := 0.728 \left( \frac{\rho_1 \left( \rho_1 - \rho_v \right) g \Delta_{fg} h_{fg}}{\mu_1 N_{tube} D_{tube} (T_{sv} - T_s)} \right)^3 \left[ \frac{1}{4} \left( \frac{0.2 \cdot C_p_T (T_{sv} - T_s)}{h_{fg}} \right) \left( N_{tube} - 1 \right) \right]
\]

(14)

The average surface temperature of the coils, 69° F (21° C), is determined by considering a heat balance between the maximum heat generation rate of 1.8 kW as required above and the coolant water flow in the tubes. The water coolant flow is assumed to enter the coil at 60° F (16° C) and 0.5 gpm (gallons per minute) (1.9 L/min). The effective thermal heat transfer coefficient, the temperature difference between the tube surface and vapor and the tube surface, is then used to determine the heat transfer from the vapor to the tube. The heat transfer rate is calculated to 18.9 kW per coil or 56.7 kW total heat removal, which provides a factor of over 30 to the required rate for cooling of 1.85 kW. A significant overdesign is necessary in order to provide a large surface area for NO\textsubscript{X} adsorption. Detailed calculations are shown in the appendix.

### 3.2.2 Connection Tube and Valve Sizing

The maximum flow to the reservoir from the condenser is determined by considering the total saturated water vapor and gas generation at peak heat generation during the dissolution process. Further, it is assumed there is no coolant flow through the coils, which provides the worst-case scenario. At the maximum heat generation of (1.85 kW), the vapor and gas flow rate are calculated to be 0.103 lb/min (48 g/min). Detailed calculations are shown in the appendix. An AFT ARROW\textsuperscript{5} computer model is used to determine the compressible steam flow behavior through the baffle plate and downstream tubing and valves. The model is shown in Figure 8. The maximum pressure in the condenser was found to be less than 1 psig with a flow velocity of 46 ft/sec (14 m/s).

\textsuperscript{5} AFT Arrow Version 4.0 (2012.05.04), Applied Flow Technology Corporation, Colorado Springs, Colorado.
A second hydraulic calculation was performed considering the incompressible flow of the coolant water through the condenser tube coils. The AFT FATHOM⁶ computer model was used to determine the pressure loss at the design flow. The design flow was determined by the coolant temperature in and an allowable temperature out. Using these temperatures and the maximum process heat generation, a heat balance calculation was determined a design flow of 0.5 gpm. This flow rate was then input into the computer model. The model and results are shown in Figure 9. The required pump pressure at the design flow was found to be 8.6 psid.

3.2.3 Reservoir

The required gas volume of the reservoir was calculated. It was assumed that all 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. 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 212°F (100°C) with an ambient air temperature of 90°F (32°C). Heat loss by radiation and heat loss by natural convection are calculated separately 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 the reservoir tank. An acceptable reservoir size was found to be 21” diameter by 31.5” high, which will fit into a standard 55-gallon drum (internal diameter of the drum is 22.5” x 33.5” high) for disposal at the end of lifetime. Detailed calculations are shown in the appendix.

3.3 OPERATION OF PROPOSED FULL-SCALE DISSOLVER

Figures 4, 10 and 11 help illustrate an outline of the proposed general operating procedure for the dissolver. A detailed test plan is to be developed in conjunction with this procedure that will incorporate measurements to determine the performance of the dissolver.
3.3.1 General Operating Procedure

1. Dissolver is closed, clean and with empty basket installed (Figure 10)
2. Valves are closed to the acid feed, O₂ feeds, product drain, de-gas and vent lines (Figure 4)
3. Back pressure valve is set to operating pressure
4. Tank reservoir is pumped down to vacuum pressure
5. Establish water coolant flow through the condenser coils
6. Acid feed syringe is filled
7. Rotate condenser section of dissolver to open loading position (Figure 10)
8. Remove basket (Figure 11)
9. Load targets and spacer rods (as required) into basket (Figure 11)
FIGURE 11. Basket removed from dissolver and basket with target foil and spacers installed.

10. Insert loaded basket into dissolver
11. Rotate condenser section of dissolver to closed operating position
12. Open de-gas valve to evacuate dissolver and condenser chambers
13. Close de-gas valve
14. Open acid feed valve and use syringe to inject acid into dissolver
15. Close acid feed valve
16. Turn on band heaters to initiate dissolution process
17. Keep heaters at desired power to achieve planned solution temperature during dissolution
18. Monitor parameters of dissolver system throughout dissolution process

Upon completion of the dissolution process,

1. Open de-gas valve to de-gas solution
2. Close de-gas valve
3. Turn off back pressure regulating valve
4. Open vent valve to bring dissolver chamber to atmospheric pressure
5. Open product drain valve in order to drain/pump solution to recovery column
6. Close product drain valve
7. Close vent valve
8. Evacuate reservoir through NO\textsubscript{x} and Iodine traps
9. Vent reservoir to bring tank to atmospheric pressure
10. Drain reservoir
11. Close reservoir valves
12. Un-bolt flanges and rotate condenser to open position
13. Remove basket and confirm complete dissolution
14. Install clean basket

The system is now ready to perform subsequent tests.

4. SUMMARY AND FUTURE WORK

A full-scale prototype of fumeless nitric acid dissolver for dissolution of up to 750 g of LEU target foils was designed. The purpose of the prototype dissolver is to test experimental conditions, such as starting concentration of nitric acid and addition and rate of oxygen or NO\textsubscript{x} to recombine NO\textsubscript{x} gasses back to nitric acid, to determine the dissolution time needed for dissolution of 750 g of LEU. Because of unknown NO\textsubscript{x} recombination efficiency under operating conditions, the dissolver contains a large reservoir tank to collect all off-gas and water vapor produced during dissolution. The reservoir tank (21” diameter by 31.5” high) keeps the dissolution vessel under atmospheric pressure and would fit into a standard 55-gallon drum (ID of drum is 22.5” x 33.5” high) for disposal at the end of lifetime.

In the next stage of this project, a prototype dissolver will be built with experiments planned to test performance. The focus of the experimental tests will be to determine optimum operational conditions for fast dissolution and to test if the dissolver can operate under atmospheric pressure. The effectiveness of NO\textsubscript{x} recombination and the optimum rate of oxygen gas will also be determined. Once the dissolution parameters are identified, we will identify modifications necessary for the hot cell operations for later implementation.
5. APPENDIX

5.1 CONDENSER THERMAL HYDRAULIC PERFORMANCE CALCULATIONS

This calculation is performed to determine the heat removal capacity due to condensing of the process vapor on the tube surfaces inside the dissolver condenser section. Boiling of the process solution occurs in the cup at the bottom of the dissolver and the vapor moves upward to the condenser where it condenses on the tube surfaces that have been cooled by the water flow through the tubes. The wall temperature is assumed to be below 212 F. The dissolver pressure is assumed to be atmospheric.

Saturated Steam Properties at 212F and 1 Atm.

\[ \rho_v = 0.0372 \frac{\text{lb}}{\text{ft}^3} \]

\[ C_p_v = 0.451 \frac{\text{Btu}}{\text{lb} \cdot \Delta^\circ \text{F}} \]

\[ \mu_v = 3600 \cdot 870 \cdot 10^{-5} \frac{\text{lb}}{\text{ft} \cdot \text{hr}} = 0.031 \frac{\text{lb}}{\text{hr} \cdot \text{ft}} \]

\[ k_v = 0.0145 \frac{\text{Btu}}{\text{hr} \cdot \text{ft} \cdot \Delta^\circ \text{F}} \]

\[ Pr_v = 0.96 \]

Liquid Water Properties at 212F and 1 Atm.

\[ \rho_l = 60.1 \frac{\text{lb}}{\text{ft}^3} \]

\[ C_p_l = 1.00 \frac{\text{Btu}}{\text{lb} \cdot \Delta^\circ \text{F}} \]

\[ \mu_l = 0.205 \cdot 10^{-3} \cdot 3600 \frac{\text{lb}}{\text{ft} \cdot \text{hr}} = 0.738 \frac{\text{lb}}{\text{ft} \cdot \text{hr}} \]

\[ k_l = 0.394 \frac{\text{Btu}}{\text{hr} \cdot \text{ft} \cdot \Delta^\circ \text{F}} \]

\[ Pr_l = 1.88 \]

\[ h_{fg} = 970 \frac{\text{Btu}}{\text{lb}} \]
Effective Convective Coefficient for Condensing

\( T_{sv} := 212^\circ F \)  
Saturated vapor temperature

\( T_s := 69^\circ F \)  
Surface temperature of tubes

\( D_{\text{tube}} := \frac{0.25}{12} \text{ ft} = 0.021 \text{ ft} \)  
Suggested tube diameter

\( N_{\text{tube}} := 20 \)  
Suggested number of horizontal tubes stacked vertically

\[
h_{fg} := h_{fg} + \frac{3}{8} \cdot C_{p1} \left( T_{sv} - T_s \right) = 1.024 \times 10^3 \frac{\text{Btu}}{\text{lb}} \\
\text{Reference 3rd Ed. Kreith. Pp 528}
\]

\[
h_{fg} = 0.728 \left[ \frac{\rho_1 (\rho_1 - \rho_v) g h_{fg} k_1 \cdot 1}{\mu_1 N_{\text{tube}} D_{\text{tube}} (T_{sv} - T_s)} \right]^{1/4} + \frac{0.2 C_{p1} (T_{sv} - T_s)}{h_{fg}} (N_{\text{tube}} - 1)
\]

\[
h_{fg} = 1.374 \times 10^3 \frac{\text{Btu}}{\text{hr} \cdot \text{ft}^2 \cdot \Delta^\circ F} \]  
Effective convective coefficient for condensing

\( D_{\text{coil}} := 3 \text{ in} \)  
Suggested coil diameter

\( L_{\text{tube}} := N_{\text{tube}} D_{\text{coil}} = 60 \text{ in} \)  
Total tube length per coil

\( A_s := \pi D_{\text{tube}} L_{\text{tube}} = 47.124 \text{ in}^2 \)  
Total surface area of tube in coil

\( q_{\text{cond}} := A_s h_{fg} (T_{sv} - T_s) = 1.885 \times 10^4 \text{ W} \)

\( q_{\text{cond}} = 6.431 \times 10^4 \frac{\text{Btu}}{\text{hr}} \)  
Total heat transfer from condensing
\[ R_m := \frac{750}{250} = 3 \quad \text{Mass ratio} \]

Heat generated from reaction.
Scaled from 250g report

\[ q_{\text{reaction}} := \frac{R_m \times 1600 \times 10^3 \text{J}}{2.5 \text{hr}} = 533.333 \text{ W} \]

\[ q_{\text{decay}} := 750 \text{W} \quad \text{Heat generated from decay heat.} \]

\[ q_{\text{total}} := q_{\text{reaction}} + q_{\text{decay}} = 1.283 \times 10^3 \text{ W} \]

\[ q_{\text{total}} = 4.379 \times 10^3 \cdot \frac{\text{Btu}}{\text{hr}} \quad \text{Total heat required to be removed} \]

\[ F_{h2o} := 0.5 \text{gpm} \quad \text{Suggested water flow through tube} \]

\[ \Delta T_{h2o} := \frac{q_{\text{total}}}{\rho_1 C_{p1} F_{h2o}} = 18.168 \cdot \Delta^\circ \text{F} \quad \text{Temperature rise at outlet from coil} \]

\[ T_{\text{amb}} := 60^\circ \text{F} \quad \text{Assumed coil inlet temperature of water} \]

\[ T_{h2o} := T_{\text{amb}} + \frac{\Delta T_{h2o}}{2} = 69.084^\circ \text{F} \quad \text{Average temperature rise of water through coil} \]

---

**Coil size**

\[ D_{\text{tube}} = 0.25 \text{-in} \]

\[ H_{\text{coil}} := 2 \cdot D_{\text{tube}} \cdot N_{\text{tube}} = 10 \text{-in} \]
5.2 VAPOR AND GAS FLOW FROM THE CONDENSER CALCULATIONS

The purpose of this calculation is to determine the maximum flow rate of the vapor and the reaction gas as it enters the condenser section of the dissolver. The vapor generation is due to the boiling of the solution and the gas is a product of the reaction between the U and the solution.

\[ V_a = 2.51 \quad \text{Acid Volume} \]
\[ V_a = 152.559 \text{-in}^3 \]

Uranium Volume
\[ W_u = 750 \text{g} \]
\[ \rho_u = 19.1 \frac{\text{g}}{\text{cm}^3} \]
\[ V_u = \frac{W_u}{\rho_u} = 2.396 \text{-in}^3 \]

\[ V_t = V_a + V_u = 154.956 \text{-in}^3 \]

\[ C_{id} = 3.5 \text{in} \]

\[ C_h = \frac{4 \cdot V_t}{\pi \cdot C_{id}^2} = 16.106 \text{-in} \]

Flow rate of reaction gas
\[ V_m = 22.4 \frac{1}{\text{mol}} \]
\[ M_g = 6.3 \text{mol} \]
\[ V_{gstp} = V_m \cdot M_g = 8.612 \times 10^3 \text{-in}^3 \quad \text{Total volume of reaction gas @ STP} \]

\[ V_g = V_{gstp} \cdot \frac{460 + 212}{460 + 70} = 178.93 \text{L} \quad \text{Volume of reaction gas at 212F} \]

\[ t_r = 2.5 \text{hr} \quad \text{Assumed reaction time of 2.5hr.} \]

\[ F_g = \frac{V_g}{t_r} = 1.213 \text{-in}^3/\text{s} \quad \text{Flow rate of reaction gas @ 212F atm. pres.} \]
Rate of vapor generation

\[ Q_r = 3.1600 \times 10^3 \text{ J} \]

Total reaction heat

\[ q_r = \frac{2Q_r}{t_r} = 1.067 \times 10^3 \text{ W} \]

Peak reaction heat rate assumed 2x average

\[ q_r = 3.64 \times 10^3 \frac{\text{ Btu}}{\text{ hr}} \]

Decay heat rate continuous

\[ q_{rd} = q_r + q_d = 6.199 \times 10^3 \frac{\text{ Btu}}{\text{ hr}} \]

\[ q_{rd} = 1.817 \times 10^3 \text{ W} \]

\[ h_{fg} = 1000 \frac{\text{ Btu}}{\text{ lb}} \]

Enthalpy per lb to vaporize acid (approx.)

\[ \rho_a = 62.4 \frac{\text{ lb}}{\text{ ft}^3} \]

\[ W_a := V_a \rho_a = 5.509 \text{ lb} \]

Approx. weight of acid

\[ Q_a := W_a h_{fg} = 5.509 \times 10^3 \text{ Btu} \]

Heat required to vaporize the acid @1 atm.

\[ t_v := \frac{Q_a}{q_{rd}} = 3.199 \times 10^3 \text{ s} \]

\[ t_v = 53.325 \text{ min} \]

Time to vaporize acid

Flow rate of vapor

\[ F_{vm} := \frac{W_a}{t_v} = 0.103 \frac{\text{ lb}}{\text{ min}} \]

Peak total mass flow rate (from the reaction cup) of gas and vapor

\[ \rho_V := 0.0372 \frac{\text{ lb}}{\text{ ft}^3} \]

\[ V_V := \frac{W_a}{\rho_V} = 2.559 \times 10^5 \text{ in}^3 \]

Total volume of vapor
Flow rate of vapor

\[ F_V = \frac{V_V}{t_V} = 79.984 \text{ in}^3 \text{s} \]

Flow rate of vapor

\[ F_V = 2.777 \cdot \text{ft}^3 \text{min} \]

Peak total volume flow rate
(from the reaction cup) of gas and vapor

\[ F_{gv} = F_g + F_V = 81.197 \text{ in}^3 \text{s} \]

Peak total volume flow rate
(from the reaction cup) of gas and vapor

\[ F_{gv} = 0.047 \cdot \text{ft}^3 \text{s} \]

Velocity in condenser

\[ D_o := 6.357 \text{ in} \quad \text{Assume 6" Pipe, Sch10} \]

\[ D_i := 4.5 \text{ in} \quad \text{Assume 4" Pipe, Sch10} \]

\[ A_a := \pi \cdot \frac{D_o^2}{4} - \frac{\pi \cdot D_i^2}{4} = 15.835 \text{ in}^2 \quad \text{Annular xsection area} \]

\[ V_{gv} := \frac{F_{gv}}{A_a} = 5.128 \text{ in} \text{s} \quad \text{This resultant flow velocity is reasonable} \]
5.3 RESERVOIR CALCULATIONS

Volume of gas generated from reaction

\[ M_{250} = 2.1 \text{ mol} \quad \text{From 250g report moles of gas generated} \]

\[ R_{\text{mass}} = \frac{750\text{gm}}{250\text{gm}} = 3 \quad \text{Ratio of U masses} \]

\[ M_{750} = R_{\text{mass}} \cdot M_{250} = 6.3 \text{ mol} \quad \text{From 750g U moles of gas generated} \]

\[ V_{\text{m}} = 22.4 \frac{1}{\text{mol}} \quad \text{Volume of 1 mole of gas at STP} \]

\[ V_{ga} = M_{750} \cdot V_{\text{m}} = 8.612 \times 10^3 \text{ in}^3 \quad \text{Gas @STP Volume of gas at STP} \]

\[ V_{g} = 460 + \frac{212}{460 + 70} \cdot V_{ga} = 1.092 \times 10^4 \text{ in}^3 \]

\[ V_{g} = 178.93 \text{ L} \quad \text{Gas at 1 atm and 212F From ideal gas law} \]

\[ V_{g} = 47.268 \text{-gal} \]

Reservoir Sizing

\[ D_{\text{res}} = 21 \text{ in} \quad \text{Reservoir diameter} \]

\[ H_{\text{res}} = \frac{V_{g}}{\frac{D_{\text{res}}}{\pi}} = 31.525 \text{ in} \quad \text{Reservoir height} \]
A thermal analysis is performed to determine an approximate value for the heat loss from the reservoir. The reservoir tank is assumed to be maintained at a uniform temperature of 100°C (212°F) with an ambient air temperature of 32°C (90°F). 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 tank to the air in the hot cell. All correlations are referenced from Keith 3rd Ed.

\[ T_t := 373K \quad T_t = \text{assumed temperature of the reservoir tank} \]

\[ T_{am} := 305K \quad T_{am} = \text{assumed ambient temperature} \]

\[ \varepsilon_t := 0.6 \quad \varepsilon_t = \text{assumed emittance of all radiating surfaces} \]

\[ f_s := 1 \quad f_s = \text{radiation shape factor} \]

\[ C_0 := \frac{5.669}{m^2 \cdot K^4} \quad C_0 = \text{Stefan-Boltzman constant for the radiation equation} \]

\[ q_{2rad} := \varepsilon_t f_s C_0 \left[ \left( \frac{T_t}{100} \right)^4 - \left( \frac{T_{am}}{100} \right)^4 \right] = 364.06 \frac{W}{m^2} \quad q_{2rad} = \text{heat flux from radiation} \]

\[ L_r := 48\text{in} = 1.219\text{m} \quad L_r = \text{length of reservoir tank} \]

\[ D_r := 12\text{in} = 0.305\text{m} \quad D_r = \text{dia. of reservoir tank} \]

\[ Q_{trad} := q_{2rad} \left( D_r \frac{\pi}{2} + D_r \cdot \pi \cdot L_r \right) = 478.151 \cdot W \quad Q_{trad} = \text{total heat loss by radiation from the reservoir tank} \]

\[ \rho_a := 1.177 \frac{\text{kg}}{\text{m}^3} \quad \rho_a = \text{Air density at standard conditions} \]

\[ \beta_g := \frac{1}{293K} \quad \beta_g = \text{coefficient of thermal expansion for an ideal gas} \]

\[ \mu_a := 1.846 \cdot 10^{-5} \frac{\text{kg}}{\text{m} \cdot \text{s}} \quad \mu_a = \text{dynamic viscosity of air at standard conditions} \]
\[ C_1 := \left( g \cdot \beta \cdot \rho_a \cdot \frac{2}{\mu_a} \right)^2 = 1.361 \times 10^8 \cdot \frac{1}{K \cdot m^3} \quad C_1 = \text{constant used in the Grashof Number} \]

\[ \delta_t := L_r = 0.8 \text{ m} \quad \delta_t = \text{characteristic length of the tank} \]

\[ Gr_a := C_1 \cdot (T_t - T_{am}) \cdot \delta_t^3 = 4.739 \times 10^9 \quad Gr_a = \text{Grashof Number for natural convection correlation} \]

\[ Pr_a := 0.71 \quad Pr_a = \text{Prandtl for air at standard conditions} \]

\[ Nu_{an} := 0.53 \cdot (Gr_a \cdot Pr_a)^{\frac{4}{3}} = 127.647 \quad Nu_{an} = \text{Nusselt Number for natural convection} \]

\[ k_a := 2.62 \cdot 10^{-5} \frac{kW}{m \cdot \Delta^\circ C} \quad k_a = \text{thermal conductivity of air at standard conditions} \]

\[ h_{an} := Nu_{an} \cdot \frac{k_a}{\delta_t} = 4.18 \frac{W}{m^2 \cdot \Delta^\circ C} \quad h_{an} = \text{natural convection coefficient} \]

\[ Q_{tn} := D_r \cdot \pi \cdot L_r \cdot h_{an} \cdot (T_t - T_{am}) = 381.087 \text{ W} \quad \text{Total heat loss by natural convection from the tank} \]

\[ Q_{tt} := Q_{trad} + Q_{tn} = 1.14 \times 10^3 \text{ W} \quad Q_{tt} = \text{total heat loss from the tank} \]

Approximately equal to average heat generation in the dissolver.
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