DEVELOPMENT OF A PRODUCTION-SCALE DISSOLVER FOR NITRIC-ACID DISSOLUTION OF LEU FOILS

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ABSTRACT

In the LEU-Modified Cintichem process, the first processing step in extracting ^{99}Mo is dissolving the irradiated uranium foil in nitric acid. A pilot-scale dissolver system, designed at Argonne was used to collect data needed to design a production-scale dissolver capable of dissolving up to 250 g of irradiated LEU and the accompanying fission-recoil barrier. This paper presents the conceptual design for the production scale dissolver as well as the experimental and modeling results that were used for the design. The key information collected to ensure a sound design for the scaled-up dissolver included: the stoichiometry and chemical nature (exothermic, endothermic) of the dissolution reaction, the phenomena that control the rate of the reaction, the chemistry of the dissolver solution during and after dissolution, the chemistry of the off-gas, and the pressure and temperature profiles produced when a specific mass of uranium metal is dissolved in heated nitric acid in a closed vessel.

1. Introduction: key reaction and fission gases

The dissolution of uranium metal in nitric acid can be represented by the following reaction:

$$U_{metal} + 4HNO_3 \rightarrow UO_2(NO_3)_2(aq) + 2H_2O + 2NO(g) \tag{1}$$
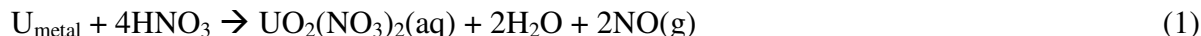
The stoichiometry of this reaction is consistent with our experimental results (discussed below). To predict these speciation of nitrogen in the system of interest thermodynamic calculations were performed using the code "The Geochemist's Workbench®" Release 8.0 (GWB) and an adapted version of the thermodynamic database "thermo.com.V8.R6.full" [1]. The calculations show that the species $NO_3^-(aq)$, $NO_2(g)$, $NO(g)$, $HNO_2(g)$ and $HNO_3(g)$ are stable depending on the temperature and oxidation/reduction potential.

Researchers at the University of Missouri Research Reactor (MURR) performed a calculation to

determine the total inventory of all fission gases generated during the irradiation of an LEU-foil target (19.75% U-235) for the following conditions: 150 days irradiation time, 8.0E-10 neutrons/cm^2 seconds (personal communication with Charles Allen (MURR), August, 2007 [2]. The calculation was performed using the ORIGEN 2.0 neutronics code. The key conclusions of the MURR calculations are that: the yield of xenon is around six times more than krypton and iodine (individually). Xenon makes up around 75% of the total fission gas generated while krypton makes up around 12%, and iodine makes up approximately 13%. The stable xenon isotopes make up around 64% of the total fission gas inventory, while the stable krypton isotopes make up around 10% and stable iodine (I-127) makes up approximately 0.12%. [2]

The experimental results presented in this paper do not deal with xenon, krypton or iodine; however, the MURR calculations [2] shows the importance of accounting for fission gases in the scale-up of the LEU-modified Cintichem dissolver. For example, the MURR calculations indicate that 7.22E-06 moles of fission gases are produced per gram of irradiated LEU foil. This implies that, if the irradiation was scaled up to 250 grams of LEU-foil, the moles of fission gas produced would be on the order of 2.0E-3 moles.

2. Modeling Results

In an effort to develop a predictive understanding of how the chemistry of the dissolver solution and gas phase will change as increasing masses of uranium are dissolved, the thermodynamic code OLI-ESP version 8.0 was used to simulate the chemistry of the dissolver for different conditions (thermodynamic databases used were "Geochemical," "Corrosion," and "Public"). The models assume that the system is in equilibrium at the end of dissolution. Selected modeling results are shown in Table 1.

The speciation of the generated gas is predicted to change with temperature (Table 1 only shows results for 25°C). When the model is run for a 200°C case, it predicts that the dissolved concentration of NO is 1.8×10^{-3} molar and NO$_2$ is 0.01 molar (it should be noted that the 200°C model run is an upper extreme, most dissolutions will be done at <150°C). For the 25°C case (Table 1), the model predicts that the dissolved concentration of NO is 2.1×10^{-3} molar and NO$_2$ is 7.0×10^{-3} molar. In this case, the dominant NO$_x$ species is predicted to be NO and the amount of gas produced for dissolving 250 grams of uranium metal in 500 mL of 9.4 molar nitric acid (initial acid concentration) is predicted to be 2.1 moles (dominantly NO). This amount of gas will determine the final pressure in the dissolver, as well as the dimension and contents of the cryogenic gas trap. .

For efficient separation of molybdenum using the LEU-Modified Cintichem process, the final nitric-acid concentration and oxidation/reduction potential (ORP) of the dissolver solution should be around 1 molar and approximately 1.0 volts (or greater) respectively. The requirement that the final nitric acid concentration be around 1.0 molar determines the starting acid concentration for a give volume of nitric acid. The volume of nitric acid is minimized for the purpose of waste minimization. The initial nitric acid concentration is thus determined by assuming that 4 moles of HNO$_3$ are consumed for every mole of U metal dissolved, as suggested by reaction (1) above. The calculations indicate that to dissolve 250 grams of uranium metal and end up with a 1 molar acid concentration 500 mL of around 9.4 molar nitric acid would be required. The volume of

nitric acid is also limited by the saturation state of the solution with respect to uranyl nitrate. Our calculations show that the precipitation of U(VI) solids is possible when the concentration of uranium reaches a value of 2.5 molar (approximately 600 g/L) at a concentration of 1 molar nitric acid (calculated using GWB).

Table 1. Thermodynamic modeling results simulating the off-gas composition for the dissolution of uranium metal after the dissolver has cooled to a temperature of 25°C.

U metal (g)	Acid Vol. (mL)	Initial Con. (molar)	ORP (volts)	H_2O (moles)	NO (moles)	NO_2 (moles)	HNO_3 (moles)	HNO_2 (moles)	Vapor (moles)
5	40	3.1	1.030	1.2E-03	0.0404	3.5E-05	3E-08	2.2E-04	0.0418
10	40	5.2	0.975	2.3E-03	0.0816	1.4E-04	2E-07	6.0E-04	0.0847
20	40	9.4	0.996	3.8E-03	0.1632	9.8E-04	2E-06	2.0E-03	0.17
25	50	9.4	0.996	4.7E-03	0.2041	1.2E-03	2E-06	2.5E-03	0.2125
50	100	9.4	0.996	9.4E-03	0.4083	2.4E-03	5E-06	5.0E-03	0.4252
75	150	9.4	0.996	0.0141	0.6124	3.7E-03	7E-06	7.6E-03	0.6377
100	200	9.4	0.996	0.0188	0.8164	4.9E-03	9E-06	0.0101	0.8502
125	250	9.4	0.996	0.0235	1.0205	6.1E-03	1E-05	0.0126	1.0628
150	300	9.4	0.996	0.0283	1.2248	7.3E-03	1E-05	0.0151	1.2755
175	350	9.4	0.996	0.033	1.4288	8.5E-03	2E-05	0.0176	1.488
200	400	9.4	0.996	0.0377	1.6329	9.8E-03	2E-05	0.0201	1.7005
225	450	9.4	0.996	0.0424	1.8372	0.011	2E-05	0.0227	1.9132
250	500	9.4	0.996	0.0471	2.0412	0.0122	2E-05	0.0252	2.1257

ORP: oxidation/reduction potential relative to the standard hydrogen electrode.
Initial con. refers to the initial concentration of nitric acid. The final acid concentration in all cases is approximately 1 molar

3. Experimental methods

The Argonne pilot-scale dissolver (Figure 1) is 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 flanged 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 septum, and a pressure relief valve. Hydrostatic pressure tests have established the dissolver vessel pressure rating at > 800 psig at 25°C; however, the individual components of the dissolver can theoretically withstand over 1000 psig. The dissolver is supported within a cylindrical steel sleeve with an open base and a flange at the top. This allows the dissolver to remain vertical during dissolution. An electric heat gun blows hot air into the support sleeve to increase and maintain the temperature. The heat gun (heater turned off) also provides the air for cooling when the dissolution reaction is complete. The temperature of the outer wall of the dissolver is monitored by two thermocouples (one to indicate temperature, and one for temperature over-run).

Dissolution experiments were also performed using a PARR-4754 dissolver (manufactured by the PARR Instrument company) consisting of a 125mL, 316 stainless steel vessel sealed with a PTFE gasket and a steel cap tightened with six compression screws (Figure 1, right). The PARR vessel steel cap contains an opening for a thermocouple and one for a central penetration that connects the vessel body to a gage block. The gage block consists of a 1000 psig rated rupture disk, a pressure gage and a penetration sealed by a needle valve that is used for venting and sampling the gas from the dissolver. This smaller scale dissolver (PARR) was used to collect

data on the temperature of the dissolver solution during the dissolution (the PARR dissolve was equipped with a thermocouple that sits within the dissolver solution, whereas the Argonne dissolve does not have an internal thermocouple). Using the PARR dissolver we monitored both the pressure and temperature profiles during the dissolution experiments. The PARR dissolver was heated using a standard heat tape.
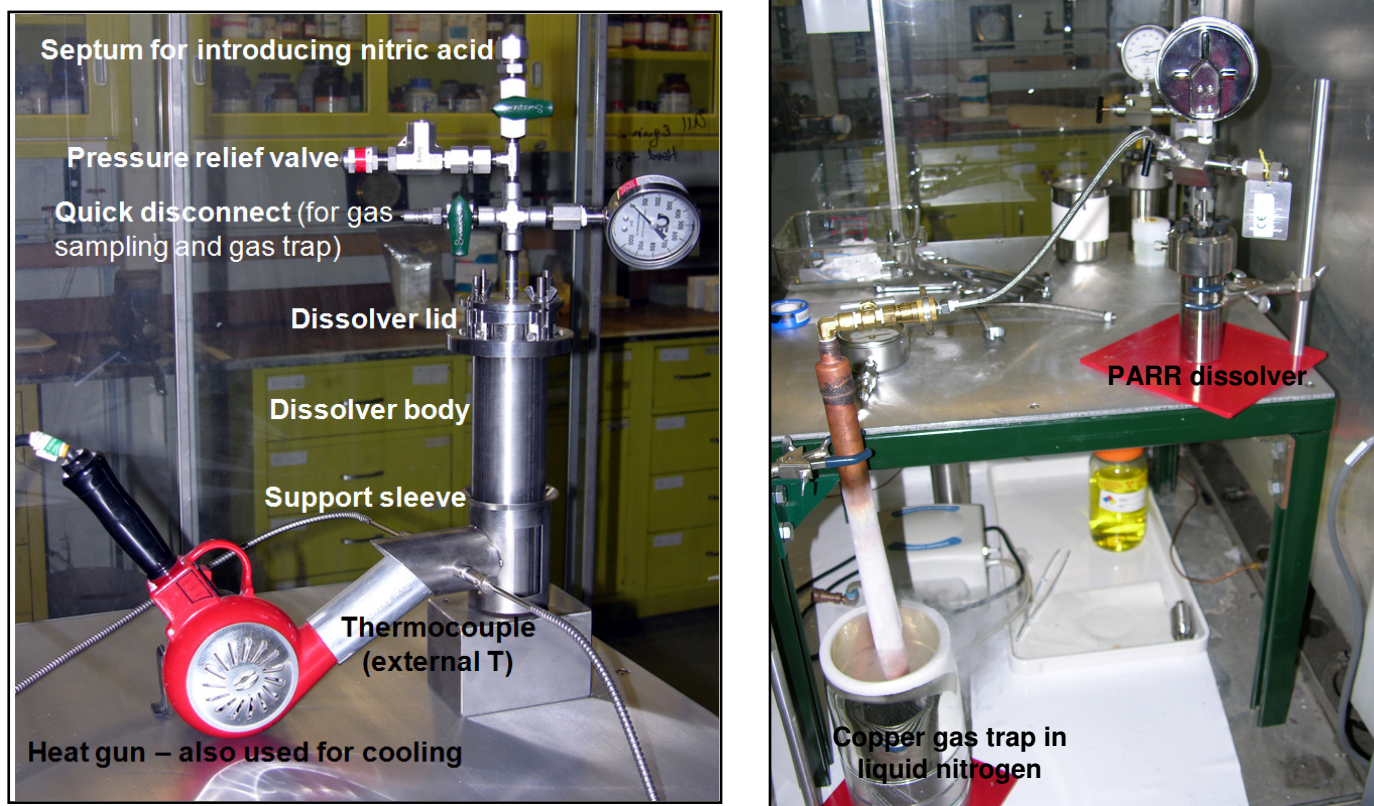

Figure 1. The Argonne "pilot" scale dissolver (left). PARR dissolver attached to the cryogenic gas trap (cold finger) (right).

The gas trap (copper pipe shown in Figure 1, right) consists of a seamless, type-L copper tube that is sealed at the bottom end by either extruding the copper to a point. The top end consists of a series of copper and brass fittings which are attached to the pipe by a soldered copper pipe reducer. Since the gas trap is operated at liquid nitrogen temperature it is important that the soldering alloys and sealants be capable of handling (not cracking) temperatures down to -196°C. It should be noted that most common soldering alloys (low temperature) are not appropriate for use with the cold trap. The gas traps (cold fingers) manufactured at Argonne are soldered and sealed with a high temperature silver based solder (15% Ag, 5% P, 80% Cu) and an epoxy cement sealant (used to seal brass and copper fittings). Prior to, and during use the gas trap rests in a vertical position, within a dewar of liquid nitrogen (Figure 1). The cryogenic gas trap is attached to the dissolver once the dissolution is complete and the dissolver has cooled to near ambient temperature. The gas trap is immersed in liquid nitrogen throughout the dissolution; therefore, once the valves are opened, the off gas from the dissolver is cryo-pumped into the adsorbent-filled gas trap and thus removed from the headspace of the dissolver. The gas adsorbents that fill the cryogenic gas trap include of 10 cm^3 calcium oxide (at the bottom), 30

cm^3 molecular sieve (13X) and 10 cm^3 calcium sulfate desiccant (at the top). Each layer is separated by a plug of glass wool. The volumes of the gas adsorbents given here are for a gas trap used with Argonne's pilot scale dissolver in which no more 20 grams of uranium foil are usually dissolved. In scaling up the dissolver system the volumes of the gas trap gas adsorbents will be considerably larger.

Although the exact role of each adsorbent is not known water, fission product gases (e.g. xenon, krypton) and nitrogen oxide (NO_x) gases (e.g., HNO_3, HNO_2, NO, N_2O, N_2O_4, NO_2) may be adsorbed by 13X molecular sieve, which is a microporous alkali alumino silicate. It has been found that, due to its charged, "cage-like" structure, the molecular sieve (13X) adsorbs xenon in an irreversible manner; that is it adsorbs xenon at low temperature and does not release it on heating to near 25°C (personal communication with James McGovern, 10/20/2009). Water vapor is also adsorbed by a calcium-sulfate desiccant and acid gases (e.g., HNO_3, HNO_2) are adsorbed by calcium oxide. It is possible that the calcium oxide in the gas trap adsorbs and then reacts with NO_2, HNO_2 and HNO_3 to produce a nitrate and thus irreversibly adsorb these key nitrogen oxide and the acid gases (see reactions 2 to 3). The exact nature of this adsorption process is not understood in detail and is the subject of on-going research at Argonne. The following reactions are suggested by previous work [3].

$Ca(OH)_2 + 2HNO_3 \rightarrow Ca(NO_3)_2 + 2H_2O$ (2)
$Ca(OH)_2 + 2HNO_2 \rightarrow Ca(NO_2)_2 + 2H_2O$ (3)

The question as to whether the gas adsorbents irreversibly adsorb the target gas species, that is if the species remained bound to the sorbent when the cold trap is brought to room temperature, is particularly important because if some or all of the gas species are reversibly adsorbed the cold trap will become pressurized when no longer in use (allowed to reach room temperature). Of particular interest in this regard is the amount of air that is in the dissolver during the dissolution process, as none of the adsorbents will irreversibly adsorb N_2.

4. Experimental results

Dissolutions were performed using both the Argonne dissolver and the PARR dissolvers (Table 2). All tests involved the use of 40mL nitric acid. The concentration of the acid used was chosen so that the final concentration would be approximately 1 molar [as determined by reaction (1)]. The starting and final nitric acid concentrations are shown in Table 1. The final nitric-acid concentration was determined using a potassium oxalate titration method [4]. All of the uranium foils used for these dissolution experiments were depleted (around 0.2% U-235) and were not irradiated. Foils from the same "batch" (same metallurgical preparation) were used. The foil samples used were approximately 500 microns thick and coated with a thin layer of uranium oxide that was not removed prior to dissolution. Results from the experiments using the Argonne dissolver are summarized in Table 2 and discussed in more detail in [5].

Seven dissolution tests were performed using the PARR dissolver (see Table 2 for conditions). The pressure and temperature profiles for the PARR dissolver tests are shown in Figure 2. The experiments with the PARR dissolver were performed in much the same manner as those done using the Argonne dissolver. The only major difference was that in the PARR experiments the nitric acid was added prior to sealing the vessel, which takes approximately 1 minute. It should

be noted that for tests (5), (6) and (9) the uranium foil was not completely immersed in the acid at the beginning of the experiments (approximately one half of the metal was exposed of the vapor phase during the early stages of the test). The main observations that can be made from the PARR dissolver tests are summarized in Table 2 and shown in Figure 2.   The key observations were that the dissolution of 9.7 grams of uranium results in a maximum pressure of 24.8 atm (350 psig) and an exothermic temperature generation of 45°C above the set point temperature of the dissolver. The maximum pressure and exothermic heat generation were less for lower mass samples.

Table 2. Conditions for dissolution tests discussed in this paper. The solution volume for each test was 40mL and the initial temperature for each test was within the range of 19 to 21°C.

Test	Mass of U_{metal} (g)	Dissolver used	Set point T (^{o}C)	Initial acid con. (M)	Final acid con. (M)**	Max P (atm)	Final P (atm)***	Max T dissolver sol. (^{o}C)
(1)	2.0	Argonne	145	3.0	2.1*	3.0	~1	not measured
(2)	5.58	Argonne	145	3.3	1.1	12.6	3.7	not measured
(3)	3.54	PARR	120	2.6	1.4	7.8	2.0	129
(4)	5.09	PARR	140	3.1	0.9	14.6	3.0	144
(5)	5.17	PARR	125	3.2	1.1	14.5	3.2	148
(6)	7.13	PARR	140	4.0	1.0	19.0	10.4	138
(7)	7.35	PARR	125	4.1	0.9	18.7	10.0	132
(8)	8.23	PARR	125	4.5	1.2	21.4	13.9	153
(9)	9.62	PARR	125	5.0	1.2	24.1	15.3	168
(10)	9.70	PARR	125	5.0	1.1	24.8	16.0	165

*Incomplete dissolution. **The final acidity of the dissolver solution was determined by titration using potassium oxalate method [4]. ***The final pressure of the dissolver is measured when the dissolver solution has reached laboratory ambient temperature (~21°C).

A 7.4 gram sample of depleted uranium (not shown on Table 2) was also dissolved using the PARR dissolver for the purposes of off-gas sampling during the dissolution process.   The pressure and temperature profiles of this dissolution experiment were consistent with those recorded for experiment (7) in Table 2; however, the pressure profile was disrupted by the sampling of off-gas during the dissolution process. Both the dissolver (prior to heating) and the gas sampling cylinders were evacuated to approximately -30 psig prior to the experiment. Off-gas samples were taken at high temperature (125°C, sample J1), at low temperature (10°C, sample J2 – cooled in ice bath), and at moderate temperature (45°C, sample J3). Each of these samples was analyzed by gas chromatography (GC) (Table 3) The higher temperature sample contains more water vapor (as predicted): 4.36 volume %. relative to the lower temperature samples: 0.46 volume % at 10°C, and 0.74 volume.% at 45°C. The higher temperature sample taken during the dissolution process (J1, taken at 125°C) also contains more N_2O than the sample taken once the depleted uranium dissolution experiment was completed (Table 3). It should be noted that data are that the final pressure in the dissolver does not increase in a linear fashion with the amount of uranium dissolve. This deviation from linearity may be due to the variation in the solubility of nitrogen oxide gas species with the total pressure in the dissolver or more likely, by the exothermic reaction of dissolving uranium in nitric acid increasing the temperature of the solution and therefore, the steam pressure. The major gas species present in the PARR experiments was determined by gas chromatography.

At the conclusion of experiment (10) a gas trap, immersed in liquid nitrogen and containing 10 cm^3 calcium oxide, 30 cm^3 molecular sieve (13X) and 10 cm^3 calcium sulfate desiccant was attached to the vessel (Figure 2, right). The gas trap had been evacuated to approximately 30 psig before use. When the valves were opened between the dissolver and the cryogenic gas trap, the pressure in the dissolver reached zero (gauge does not record negative values) within seconds. The pressure in the gas trap was less than -30 psig while it was immersed in liquid nitrogen. After 15 minutes the gas trap was removed from the liquid nitrogen dewar and allowed to warm to laboratory ambient temperature (21°C). A pressure gauge was attached to the gas trap during warm up to determine the volume of gas that desorbed from the gas adsorbents once they reached ambient temperature. As the gas trap warmed its internal pressure increased to 100 psig. It is not clear if the remainder of the gas was irreversibly adsorbed onto the gas trap chemicals or whether the differences in volume between the dissolver volume and gas trap produced this pressure decrease. It should be noted however that when the used gas trap was vented at laboratory ambient temperatures the gas was dark brown suggesting that NO_2 or N_2O_4 was not irreversibly adsorbed by the gas trap adsorbents.
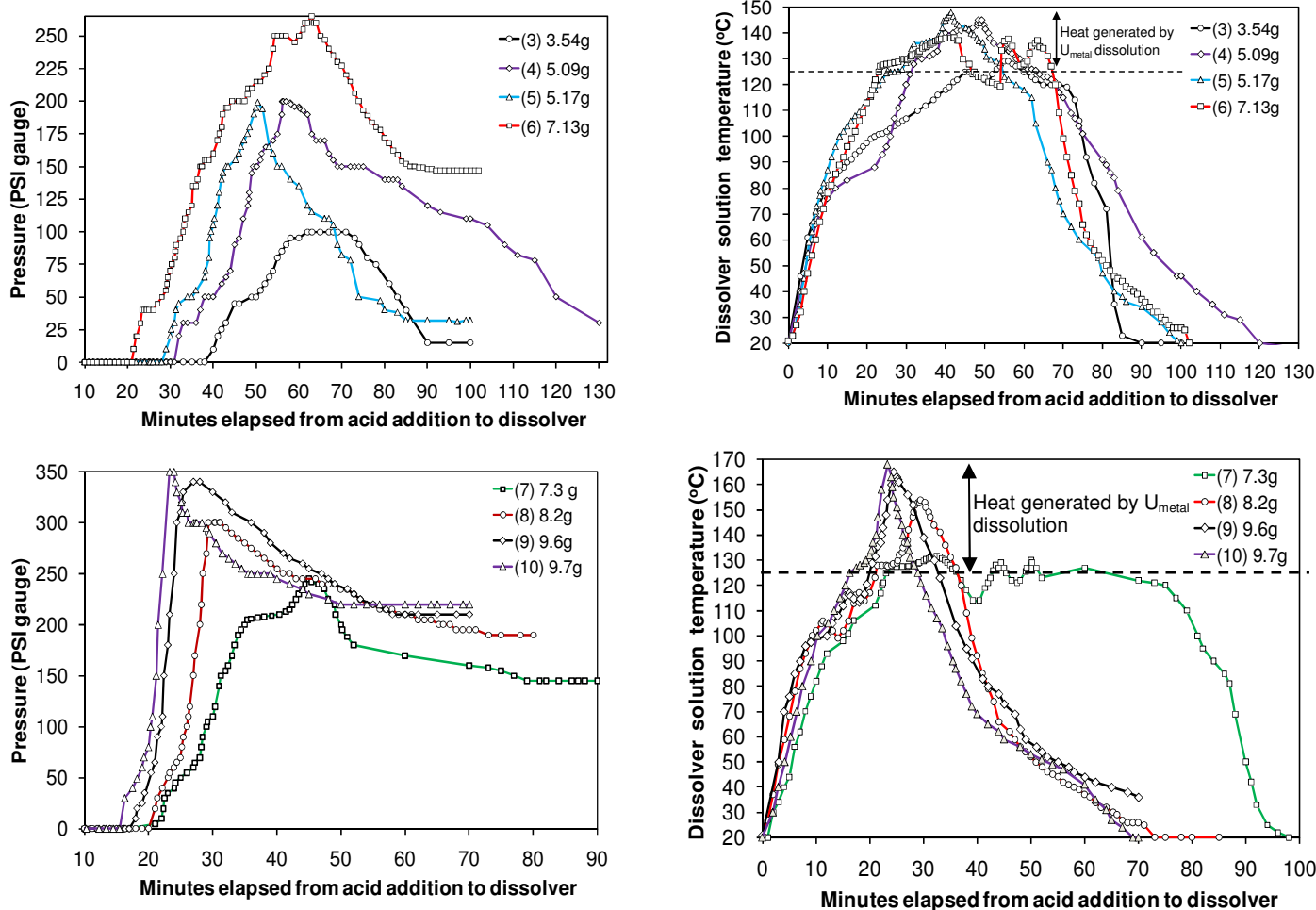

Figure 2. Pressure and temperature profiles from the depleted uranium dissolution experiments using the PARR-4754 dissolver experiments using the PARR-4754 dissolver

Table 3. Concentrations of gases in dissolver off-gas samples as determined by gas chromatography

Peaks	J1 (125°C) Vol.%	J2 (10°C) Vol.%	J3 (45°C) Vol.%
NO_2	93.23	98.47	97.84
N_2	2.23	1.02	1.37
H_2O	4.36	0.46	0.74
N_2O	0.09	0.03	0.02
O_2	0.09	0.02	0.03

No corrosion of the stainless steel (304 for the Argonne dissolver and 316 for the PARR dissolver) was observed during these experiments discussed in this report.

5. Conclusions from modeling and experimental studies

The main conclusion that pertains to scaling up the dissolver to a system capable of digesting 250 grams of irradiated uranium foil is that the temperature of the dissolver must be monitored and controlled externally. Even our "small scale" tests in which 9.7 grams of unirradiated depleted uranium was dissolved in 40mL of nitric acid in a 125mL dissolver reached temperatures 45°C over the experimental set point [resulting in total pressures of approximately 360 psig (24.8 atm)]. Heat management is particularly important since the irradiated targets will be thermally hot due to the decay short half life fission products produced. Due to the exothermic nature of the uranium metal dissolution in nitric acid and the decay heat it is concluded that a larger scale dissolver must have an active heating/cooling system (monitored and controlled externally).

Other important observation made from these experiments include: Of the nitrogen oxide gases, only NO_2 and minor amounts of N_2O were measured in the off-gas samples. The cryogenic gas trap used to evacuate the nitrogen oxide gases from the dissolver head space became pressurized once it was removed from the liquid nitrogen. More data are needed on the role of the gas adsorbents within the gas trap and the maximum pressure it will reach following use and warm up. This is a key topic for designing a scaled-up version of the dissolver system.

6. Implication of experimental and modeling results for dissolver scale-up

The modeling results (Table 2) for runs in which the dissolver was assumed to have cooled down to near ambient temperature were used to estimate the pressure inside dissolvers of different volumes (Table 4). These calculations thus estimate the final pressure in the dissolver after the dissolution is complete and the dissolver cooled. The ideal gas law was used for these estimates which make the results only guideline or "ball-park" values, as this calculation assumes that the dissolver solution is incompressible and they ignore the solubility of the nitrogen oxide and acid gases present. This yields a conservative estimate as indicated by the comparison between the calculation and our experimental results. After a number of iterations, it was determined that a dissolver volume of 1.3 liters would be sufficient as long as its pressure rating is approximately 100 atm (1500 psig). This volume minimizes the size of the dissolver, while accommodating the required solution volume and head space.

Our experimental results show that once the nitric-acid solution in the dissolver has been heated to 125°C the dissolution reaction becomes self-sustaining and if the dissolver is not actively

cooled, the temperature (and thus the pressure) in the dissolver with spike within a matter of minutes. Experimental results also showed that uranium metal dissolution reaction proceeds at an adequate rate when the temperature of the dissolver solution is 150°C to 170°C. An adequate rate is defined as one that will allow complete dissolution in approximately 30 minutes. The vapor pressure of pure water at 150°C is around 5 atm, 70 psig. At 170°C the water vapor pressure is around 8 atm, 115 psig.

We aim to design the dissolver so that its cooling system will be able to maintain a steady state temperature of the dissolver solution with a maximum of 110°C; however, 100°C is desired since it will lead to a lower maximum working pressure during dissolver operation. Therefore, the maximum pressure that the production dissolver will need to be rated for is 64 atm from reaction off-gas + 8 atm dissolver solution vapor pressure: approximately 72 atm, 1045 psig. This is one of the key design criteria that will be used in the fabrication design of the production scale dissolver. Based on experimental and modeling results plus the simple calculations discussed above the following design criteria must be used for the production scale dissolver: (1) Low corrosion rates at 170°C in 10 molar nitric acid, (2) Maximum working pressure of 72 atm (1045 psig) or using a safety factor of 1.5x a maximum pressure rating of 108 atm (1570 psig), (3) Active cooling to maintain dissolver solution at 170°C. It should be noted that we took a very conservative approach to this conceptual design. We are currently working on designing cooling system for the dissolver that will keep the temperature closer to 105°C during the reaction.

Table 4. Experimentally measured and calculated pressures of off-gas produced during the dissolution of different amounts of uranium foil.

Dissolver Vol. (L)	Acid Vol. (L)	U (g)	U (moles)	Gas (moles)	Measured P (atm)	Calc. P (atm)	P (psig)
0.36	0.04	5.58	0.023	0.047	3.7	4	38
0.15	0.04	3.54	0.015	0.030	2.0	7	83
0.15	0.04	5.09	0.021	0.043	3.0	10	125
0.15	0.04	5.17	0.022	0.043	3.2	10	127
0.15	0.04	7.13	0.030	0.060	10.4	13	181
0.15	0.04	7.35	0.031	0.062	10.0	14	187
0.15	0.04	8.23	0.035	0.069	13.9	15	211
0.15	0.04	9.62	0.040	0.081	15.3	18	250
0.15	0.04	9.70	0.041	0.082	16.0	18	252
1.3	0.04	20	0.084	0.168	----	3	33
1.3	0.05	25	0.105	0.210	----	4	46
1.3	0.1	50	0.210	0.420	----	9	111
1.3	0.15	75	0.315	0.630	----	13	182
1.3	0.2	100	0.420	0.840	----	19	260
1.3	0.25	125	0.525	1.050	----	24	345
1.3	0.3	150	0.630	1.260	----	31	439
1.3	0.35	175	0.735	1.470	----	38	542
1.3	0.4	200	0.840	1.680	----	46	657
1.3	0.45	225	0.945	1.891	----	54	785
1.3	0.5	250	1.050	2.101	----	64	930

"Gas" includes a mixture of vapor phase H_2O, NO, NO_2 HNO_3 and HNO_2.

7. Vessel design and operation

Figure 3 shows conceptual renderings of the production scale dissolver which was designed based on the experimetal and modeling work discussed above. The dissolver vessel consists of an approximately 32 cm hight, 304L cylinder consisting of two or more welded segments. The outer diameter of the vessel is approximately 14 cm. The dissolver lid, which is attached to a hinged support rod (Figure 3, right), is sealed using two semi-circle clamps which are used to seal the vessel by tighening 10 quarter inch hexagonal bolts. Between the lid and the vessel body there is an O-ring gasket. The gasket material is a subject of ongoing research, but will probaly consist of an annealed metal. The lid assembly is penetrated by a valved port used to evacuate and remove the off-gas from the vessel. Solenoid valves will be used for all ports on the dissolver (lid port, acid inlet, product outlet). The lid will also have a penetration into which a thermocouple can be inserted. Within the dissolver, there are acid channels into which the acid is pumped from a separate reservoir. Within these channels, the acid is heated by hot air or a water jacket prior to being pumped into the vessel containing the irradiated LEU foil. The hot acid is pushed from the channels into the vessel using an inert gas or more acid if the process will be repeated continually. The acid reservoir consists of a simple 304L stainless steel reservoir and a simple piston/plunger mechanism. This design should make the addition of acid into the vessel relatively easy to accomplish by remote operation. Once heated, the acid is pumped into the vessel in which the uranium foil is contained within a mesh basket (304L). The basket is used so that the foils can be readily lowered into the vessel by remote operation at the beginning of the process. Once the dissolution is complete, the dissolver solution (product) is removed from the vessel by a valved port at the bottom of the dissolver. Again, all valves will be of the solenoid type. The entire dissolver system (lid, vessel, acid reservoir) is supported by a balanced steel assembly.

Operation of the production-scale dissolver is envisioned to involve a number of steps, all of which can be performed by remote operations: The first step is to lower the basket (containing irradiated LEU) into the vessel. The second step is to seal the vessel lid. The third step is to push the acid from the acid reservoir into acid channels in which it will be heated to approximately $100^{o}C$ by hot air or a water jacket. The fourth step is to push the hot acid from the channels into the vessel; this begins the dissolution process. The hot acid is vacuum-drawn into the channels. The dissolver solution (product) is then drained or pumped out using the port at the base of the dissolver.
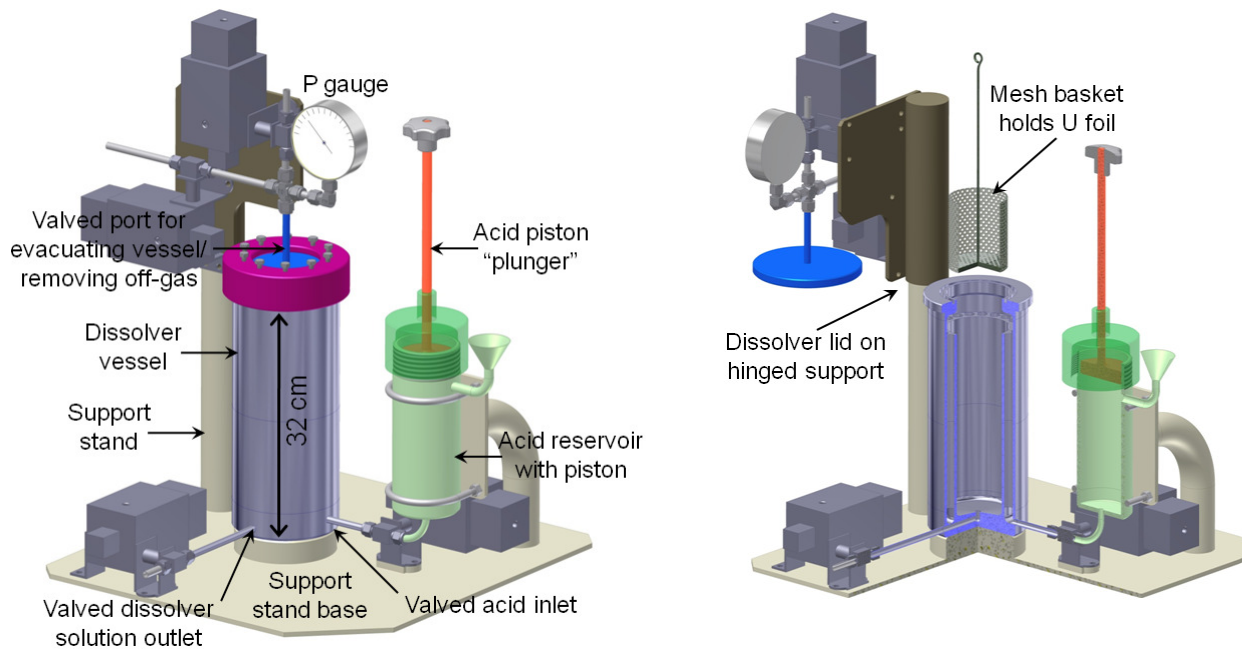
Figure 3. Conceptual rendering of the production dissolver design. Diagram on right is a cut-away view showing the dissolver lid on its hinged support rod and the 304L mesh basket that will hold the irradiated LEU foil.

8. On-going and future work

On-going work includes calculations and experiments that will allow us to design and/or optimize a method for removing and capturing the off-gas from the dissolver vessel. Near term future work includes optimizing the conceptual design presented in this report and developing fabrication drawings for the production scale dissolver. We are looking into to ways of optimizing the dissolver system design so that the dissolutions could be performed at approximately ambient pressures and temperatures near 105°C. The scaled-up dissolver system will be fabricated at Argonne and tested cold using a mock-up hot cell facility. We will also irradiate uranium foils using the Argonne linac to produce fission products to measure their behavior during dissolution and trapping.

9. Acknowledgements

10. References

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