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Low Temperature Dissolution Flowsheet for Plutonium Metal

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EXECUTIVE SUMMARY

The H-Canyon flowsheet used to dissolve Pu metal for PuO₂ production utilizes boiling HNO₃. SRNL was requested to develop a complimentary dissolution flowsheet at two reduced temperature ranges. The dissolution and H₂ generation rates of Pu metal were investigated using a dissolving solution at ambient temperature (20-30 °C) and for an intermediate temperature of 50-60 °C. Additionally, the testing included an investigation of the dissolution rates and characterization of the off-gas generated from the ambient temperature dissolution of carbon steel cans and the nylon bags that contain the Pu metal when charged to the dissolver.

The dissolving solution for each experiment consisted of 10 M HNO₃, 0.1 M KF, and 1.5 g/L B. The total dissolution time required in H-Canyon to completely dissolve the Pu metal charge is dependent upon the dissolving solution temperature as well as the surface area of the Pu metal being dissolved. Based on process knowledge using the current flowsheet at boiling (112 - 116 °C), a Pu metal charge was estimated to dissolve in the H-Canyon 6.1D dissolver in 24-36 h. The Pu metal dissolution rate measured at 57 °C is approximately 20 times slower than at boiling, and the dissolution rate at ambient temperature (24 °C) is approximately 80 times slower. Therefore, a Pu metal charge dissolved at 57 °C is estimated to dissolve in the 6.1D dissolver in approximately 20-30 days, and a Pu metal charge dissolved at ambient conditions (24 °C) is estimated to dissolve in about 80-120 days; although, higher ambient temperatures or increased specific surface areas (SSAs) of the Pu metal will result in shorter dissolution times.

Hydrogen concentrations measured by gas chromatography (GC) in the off-gas from the Pu metal dissolutions at the two temperatures were less than detectable (<0.1 vol %). If a conservative H₂ concentration of 0.1 vol % and the total offgas generation rates measured during the laboratory dissolutions performed at 24 and 57 °C are used to estimate the maximum H₂ concentrations for an H-Canyon dissolver charge of 46 kg, comparison of the maximum concentrations with the appropriate lower flammability limit (LFL) for H₂ showed that a dissolver purge is not required for Pu metal dissolutions performed at ambient temperature or 50-60 °C; however, if the dissolver is heated or used to dissolve a full charge at ambient temperature, the continued use of the sparge is recommended to provide agitation in the dissolver.

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LIST OF ABBREVIATIONS

GC	gas chromatography
GPHA	gamma pulse height analysis
H ₂	hydrogen
H ₃ BO ₃	boric acid
HCMA	hot crane maintenance area
HNO ₃	nitric acid
IC	ion chromatography
ICPES	inductively-coupled plasma emission spectroscopy
LFL	lower flammability limit
MA	moving average
MS	mass spectrometer
N ₂	nitrogen
NFPA	National Fire Protection Association
NO	nitric oxide
N ₂ O	nitrous oxide
NO ₂	nitrogen dioxide
PuO ₂	plutonium dioxide
RFETS	Rocky Flats Environmental Technology Site
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
SSA	specific surface area
STP	standard temperature and pressure
UV-vis	ultraviolet-visible

1.0 Introduction

Plutonium metal is dissolved in the H-Canyon 6.1D dissolver to prepare feed solution for PuO₂ production.¹ To prepare a dissolver charge, 3013 Pu storage containers are transferred to HB-Line where the inner containers are cut open and the contents transferred to dissolvable carbon steel cans (containing no polymeric gasket materials). The cans are then bagged out of the glovebox into dissolvable nylon bags and transferred to the Hot Crane Maintenance Area (HCMA). In the HCMA, the carbon steel cans are loaded into stainless steel charging bundles fitted with carbon steel endcaps for dissolution. Once loaded, a bundle is transferred to the 6.1D dissolver and charged to the 10-well insert. Under the current flowsheet, the dissolving solution is heated to boiling after complete charging of the dissolver until the Pu metal is dissolved.¹

During charging of the 6.1D dissolver for the Batch 5 Pu metal dissolution, eight of the ten insert wells had been charged when the Savannah River Site (SRS) entered an operational pause. When deliberate operations in H-Canyon resumed, the ambient-temperature solution in the dissolver was sampled and the analysis indicated that all the Pu metal had dissolved (without heating). Dissolving the Pu metal in the 6.1D dissolver at ambient conditions or at a reduced temperature has advantages. The highly oxidizing and corrosive environment from the use of boiling 10 M HNO₃ containing 0.1 M fluoride as the dissolving solution contributes to the oxidation of Pu(IV) to Pu(VI) and generates metallic impurities as a result of the dissolver corrosion. The use of less aggressive conditions (i.e., lower temperature) would minimize the amount of ferrous sulfamate required to reduce the Pu(VI) prior to anion exchange in HB-Line, reduce the amount of impurities which must be removed during anion exchange, and would likely extend the life of the 6.1D dissolver by reducing the corrosion rate during dissolution. However, increased processing time resulting from slower Pu metal dissolution rates at reduced temperatures would need to be considered.

The dissolution of Pu metal using HNO₃ solutions containing fluoride was investigated at both the Rocky Flats Environmental Technology Site (RFETS) and the SRS. However much of the testing was conducted at HNO₃ concentrations in the 3-5 M range and at elevated temperatures (80-100 °C) to limit the formation of PuO₂ and maximize the rate of dissolution.²⁻⁵ Miner et al. performed lower temperature (23-69 °C) dissolution experiments; however, the Pu metal was dissolved using 1-5 M HNO₃ solutions containing 0.01-0.13 M hydrogen fluoride.⁶ Holcomb performed dissolution experiments using both alpha and delta phase Pu metal. The alpha metal was dissolved using 3-5 M HNO₃ solutions containing fluoride ion as the catalyst; however, Al was also present in the solution at Al to fluoride molar ratios of 0, 0.5, 0.75, and 1.0. The dissolutions were performed at 90-100 °C.⁷ In experiments with delta phase Pu metal, Holcomb demonstrated that a 2-3 M HNO₃ solution containing 0.1 M fluoride at 35-50 °C could be used to dissolve Pu metal in the SRS F-Canyon facility.⁸ The current H-Canyon flowsheet used to dissolve Pu metal for PuO₂ production utilizes the same dissolving solution as the desired low temperature flowsheet (10 M HNO₃, 0.1 M F, and 1.5 g/L B); but, the dissolution is performed at the boiling point of the solution (112 – 116 °C).¹

The dissolution of carbon steel and nylon has been investigated at the Savannah River National Laboratory (SRNL) for previous applications in the SRS F- and H-Canyon facilities. Experimental work was performed to measure the dissolution rate of carbon steel in a 2 M HNO₃ solution containing 0.25 M KF and 2 g/L B for processing Pu-contaminated U metal scrap in H-Canyon. Pierce measured increasing dissolution rates for plate/bar, wire/rivet, and can materials, respectively, at ambient temperature without stirring.⁹ The volume of gas generated at ambient temperature was not measurable due to the solubility of the reaction products. At ambient temperature and 95 °C, the gases generated were predominantly a mixture of NO and NO₂.⁹

The use of dissolvable nylon bags for nuclear material processing applications is based on laboratory experiments used to select and qualify a material for processing Pu-containing residues in F-Canyon.¹⁰⁻¹⁴ A laboratory evaluation was also performed for a nylon bag proposed for use with Pu-containing residues from the RFETS.¹⁵ The most recent evaluation of the dissolution of nylon bags was performed for the processing of Pu-contaminated U scrap in H-Canyon.¹⁶ Nylon bags were dissolved in 2-3 M HNO₃ containing 0.025 M KF and 2 g/L B at varying ratios of nylon mass to solution volume. The experiments were started at ambient conditions and the temperature was slowly increased to the boiling point of the solution. The experiments showed that increasing the HNO₃ concentration lowered the onset temperature for dissolution and increased the dissolution rate. Little acid was consumed during the depolymerization as the nylon broke-up into small pieces which became smaller until they were no longer visible.

1.1 Objectives

Based on the merits of a low temperature flowsheet, SRNL was requested to perform a series of Pu metal dissolution experiments to develop and demonstrate the flowsheet for the 6.1D dissolver at temperatures below boiling.^{17,18} The dissolving chemistry will utilize a 10 M HNO₃ solution, containing 0.1 M fluoride (as KF) and 1.5 g/L B (as boric acid) which is consistent with the dissolving solution used for previous 6.1D dissolutions.¹ The Pu metal dissolution and H₂ generation rates will be measured at 20-30 °C (i.e., ambient conditions) and at an intermediate temperature (50-60 °C). In addition, experiments will be performed to estimate the dissolution and H₂ generation rates of the carbon steel used in the cans and endcaps and the nylon bags used as the primary containment for the Pu-containing cans. The carbon steel and nylon dissolutions will be performed at ambient temperature (20-30 °C), which is representative of the dissolver temperature during charging.

2.0 Experimental Procedure

2.1 Pu Metal

2.1.1 Preparation of Pu Metal

The Pu metal used for the low temperature dissolution flowsheet demonstration experiments is from the same batch of material used to develop the flowsheet currently in use.¹ The samples were prepared from delta-phase metal. A thin oxide coating was present on the surface. The thin sheet-like coupons were cut from larger pieces if necessary using tin snips. For each experiment, the Pu metal mass ranged between 2.5 and 3.3 g. A solution volume between 369 and 489 mL was used to target a final dissolved Pu concentration of 6.75 g/L Pu. The mass and dimensions of the metal pieces used in each of the four experiments are provided in Table 2-1. The dimensions of the metal pieces were measured using a micrometer. Using the measured dimensions, the calculated density of the metal pieces ranged from 13.4 to 15.1 g/cm³.

Table 2-1. Mass and Dimensions of Pu Metal Used in Flowsheet Demonstration Experiments

Experiment	Mass (g)	Length (mm)	Width (mm)	Thickness (mm)	Surface Area (cm²)
Pu-Diss-1	2.4844	29.41	8.28	0.74	5.43
Pu-Diss-2	2.6998	18.26	15.12	0.73	6.01
Pu-Offgas-1	3.1203	20.68	14.95	0.67	6.67
Pu-Offgas-2	3.3031	20.04	17.03	0.71	7.35

2.1.2 Pu Metal Dissolving System

The dissolution vessel and offgas collection system were fabricated from borosilicate glass by the SRNL Glass Shop. A photograph of the equipment is shown in Figure 2-1. The dissolving vessel was made from a 500-mL round-bottom flask. Penetrations were added for a condenser, sample port, thermocouple, and N₂ purge, and the bottom was flattened slightly to facilitate heating and agitation using a hot plate/stirrer (with magnetic stir bar). The apparatus is similar to equipment previously used for Pu metal dissolutions and offgas measurements at SRNL.¹ The solution temperature was controlled using an external thermocouple monitored by the hot plate. A secondary thermometer inserted directly into the dissolving solution through a glass port was used to verify that the target temperature in solution was maintained.



Figure 2-1. Pu Metal Dissolving and Offgas Measurement System

2.1.3 Pu Metal Dissolution Rate Measurements

To perform the Pu metal dissolution rate experiments (Pu-Diss-1 and Pu-Diss-2), the Pu metal was initially placed in a perforated glass basket. The basket was suspended in the dissolver by a glass rod held in place by a compression fitting. The compression fitting allowed adjustment of the basket height. The Pu metal was held above the dissolving solution until the desired solution temperature was reached. Water was not circulated through the condenser during the experiments because the targeted solution temperatures were well below boiling. The dissolving solution consisted of a 10 M HNO₃ solution containing 0.1 M fluoride (from KF) and 1.5 g/L B (from boric acid) which is consistent with the dissolving solution used for the current H-Canyon dissolver flowsheet. The solution was stirred at 250 rpm using the magnetic stir bar. During dissolution rate measurements, the vessel top and basket assembly were detached to periodically remove, weigh and measure the Pu metal coupon. One milliliter solution samples were also taken using a pipette for analysis at that time. The solution samples were analyzed for ²³⁹Pu and ²⁴¹Am by gamma pulse height analysis (GPHA). Samples of the initial and/or final

dissolving solutions were also analyzed for metals by inductively-coupled plasma emission spectroscopy (ICPES), ammonium by ion chromatography (IC), and for free and total acid by titration.

2.1.4 Pu Metal Offgas Measurements

The offgas generated from the dissolution of the Pu metal coupon during Experiments Pu-Offgas-1 and Pu-Offgas-2 exited through the condenser (See Figure 2-1). Viton[®] tubing was used to connect a 40-mL sample bulb and a 2-L Tedlar[®] gas-collection bag to the condenser (Figure 2-2). The sample bulb was connected to the tubing using quick-disconnect fittings which allowed easy replacement of the bulb during dissolution. A final gas sample was also taken by opening an evacuated sample bulb that was connected directly to the dissolver vessel via a sample port using a quick-disconnect fitting. The bulbs were fabricated using glass stopcocks at both ends to prevent leakage following removal from the dissolving system. The Tedlar[®] bag was placed in a water-filled vessel with a graduated cylinder incorporated into the top which allowed measurement of the gas volume by water displacement. Water was not circulated through the condenser.



Figure 2-2. Tedlar[®] Gas-collection Bag

Before the offgas experiments began, air was initially purged from the dissolver and condenser using N₂. A penetration in the dissolver (obscured by the condenser in Figure 2-1) equipped with a quick-disconnect fitting was provided to attach the N₂ supply line. Nitrogen gas (estimated at 200-400 mL/min) was used to purge the dissolver system including gas bulbs with about 2-3 Liters of N₂ prior to lowering the coupon into the HNO₃ solution. Once the N₂ purge was complete, one of the purged gas sample bulbs was put in line with the Tedlar[®] bag to collect the gas sample for analysis by GC. Agitation was then started at 250 rpm. For the moderate temperature (50-60°C) experiment, the hot plate set point was programmed to 55 °C. The volume of water displaced by the offgas was recorded periodically by the rising water column height in the graduated cylinder of the Tedlar[®] bag system during the experiment. When the desired solution temperature was reached, the glass basket containing the Pu metal was lowered into the solution and the water height in the graduated cylinder of the Tedlar[®] bag system was recorded periodically to track the offgas generation. The dissolving solution consisted of a 10 M HNO₃ solution containing 0.1 M fluoride (from KF) and 1.5 g/L B (from boric acid) which is consistent with the dissolving solution used in previous 6.1D dissolutions.

The final solution samples were analyzed for ²³⁹Pu and ²⁴¹Am by gamma pulse height analysis (GPHA) and the gas samples were analyzed by GC. Samples of the final solution following each dissolution were

also analyzed for metals by ICPES and free acid by titration. IC-Cation analysis was also performed to investigate the possibility of ammonium formation.

2.2 Carbon Steel

2.2.1 Carbon Steel Dissolution and Offgas Measurements

The dissolution rate and offgas composition of the carbon steel cans used for Pu metal dissolution were evaluated. The coupons used in the experiments were cut from a carbon steel can supplied by H-Canyon Engineering. The experiments were performed at ambient temperature (20-30 °C), which is representative of the dissolving solution temperature during charging of Pu metal to the dissolver. The dissolving solution consisted of a 10 M HNO₃ solution containing 0.1 M fluoride (from KF) and 1.5 g/L B (from boric acid) which is currently used for the Pu metal dissolving solution in H-Canyon.¹ Online analysis of the offgas by mass spectrometry and Raman Spectroscopy was performed to characterize the offgas and measure the offgas generation rate.

The small scale dissolver system discussed in Section 2.1.2 was modified to permit continuous offgas monitoring. The modified setup, shown in Figure 2-3, allows for an offgas sample port to be connected after the condenser. The mass spectrometer (MS) sample line was connected above the condenser to the offgas sample port. The manometer, also connected to the offgas sample port, acts as a pressure relief device and provides a measurement of the pressure in the system in inches of water. The offgas leaving the condenser subsequently passes through a Raman probe and terminates in a bubbler (beaker containing 700 mL or 3.5 inches of deionized water). The bubbler prevents air in-leakage from the vent side of the system. This configuration allows the offgas analyzers to measure the non-condensable gases such as H₂, N₂, O₂, Ar, NO, N₂O and NO₂ in real time during the experiment.

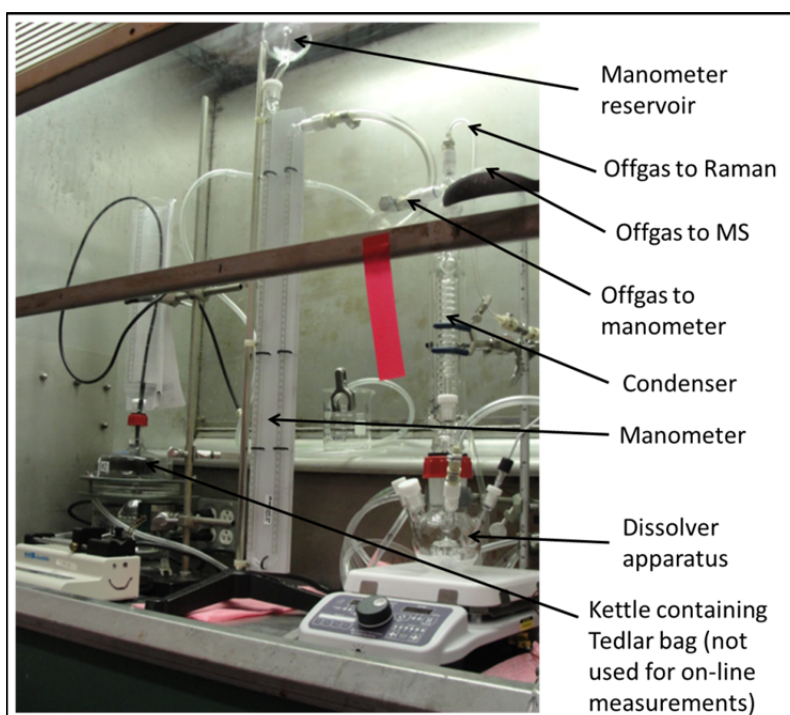


Figure 2-3. Dissolver Setup with Online MS and Raman Offgas Analyzers

The MS used for these experiments was a Monitor Instruments LAB 3000 Cycloidal MS. The Monitor MS samples a portion of the non-condensable offgas stream through a vacuum and outputs the volume percent of the gases in the sample based on its calibration using gas standards. The Monitor MS was calibrated using the gases listed in Table 2-2. The MS is controlled by a computer using Monitor v6.00 software.

To calculate offgas generation rates, an Ar tracer gas was metered into the system through a flow controller at a set rate (10 cm³/min @ STP). The total offgas rate was then calculated by dividing the set input rate by the measured Ar concentration in the offgas.

Table 2-2. Calibration Gases for MS and Raman Analyzers

Supplier	Gas	Ar (%)	N ₂ (%)	N ₂ O (%)	NO ₂ (%)	NO (%)	O ₂ (%)	H ₂ (%)
Air Liquide	20% N ₂ O-80% Ar	80.00	—	20.00	—	—	—	—
Liquid Technology	5% NO ₂ -20% O ₂ -75% Ar	74.89	—	—	4.98	—	20.13	—
Air Liquide	20% NO-80% Ar	80.00	—	—	—	20.00	—	—
Air Liquide	5% N ₂ -10% H ₂ -85% Ar	85.00	5.00	—	—	—	—	10.00
SRNL	Ar*	99.9	—	—	—	—	—	—
SRNL	N ₂ *		99.9	—	—	—	—	—
SRNL	Air*	0.94	78.03	—	—	—	20.99	—

*purity not measured; supplied from SRNL facility gases

The Raman spectrometer non-intrusively analyzes the offgas through a quartz window using the excitation of a laser passing through a fixed portion of the offgas stream. The Raman scattering technique identifies and estimates concentration of gases within the offgas. The Raman was calibrated using standard gases shown in Table 2-2. The Raman was set up to measure concentrations of the offgas species approximately every 12-13 seconds. Since the Raman directly measures the concentrations in the offgas stream, there is zero dead time between the offgas concentration measurement and the reading other than the analysis time of 12-13 seconds. The Raman was controlled by and data was logged using a computer running EZRamanReader v8.3.9 software and an Excel spreadsheet.

2.3 Nylon

2.3.1 *Nylon Dissolution and Offgas Measurements*

A Nylon bag like the ones used for Pu metal dissolution was obtained from H-Canyon Engineering to evaluate nylon dissolution and offgas behavior. A piece of nylon was cut from a bag and dissolved in a solution containing 10 M HNO₃, 0.1 M fluoride (from KF), and 1.5 g/L B (from boric acid) without stirring to measure dissolution rate. A second piece of nylon was dissolved using the dissolver and offgas collection system described in Section 2.1.2 to investigate offgas behavior.

2.4 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

3.0 Results and Discussion

3.1 Pu Metal

3.1.1 General Observations

Prior to starting the dissolution experiments, a Pu metal coupon was placed in the glass dissolver basket as described in Section 2.1.3. Based on the mass of the Pu coupon, the dissolver vessel was filled with the appropriate amount of dissolver solution targeting a final Pu concentration of 6.75 g/L when the coupon was completely dissolved. The stirrer was turned on and the coupon was lowered into the solution representing time zero. At regular intervals (based on the expected dissolution rate for the conditions being tested), the coupon was removed from the vessel to measure the mass and physical dimensions. The solution was sampled at each interval to measure the Pu concentration by GPHA. The offgas measurements were performed in the same fashion without removing the coupon or sampling except the condenser was connected to the Tedlar[®] bag making a sealed system. No visible gas generation was observed from the Pu metal coupons as they dissolved. The plutonium metal dissolved slowly to produce a green opaque solution. After a period of time, light brown gas was observed in the reactor vessel. No visible solids were observed in the vessel following any of the dissolutions.

3.1.2 Sample Analysis

The GPHA for the samples generated during the Pu metal dissolutions are provided in Appendix A. The activities for both ²³⁹Pu and ²⁴¹Am were measured. The ²³⁹Pu activities were converted to total Pu values by assuming the metal coupons were weapons grade Pu containing 94 wt % ²³⁹Pu. Before the concentrations were correlated with the dissolution time, they were corrected for the small change in volume which occurred due to the removal of samples and evaporation losses; although a small concentrating effect would be expected in the 6.1D dissolver due to evaporation. The volumes of solution at the conclusion of Experiments Pu-Diss-1 and Pu-Diss-2 only decreased by 1 and 7 mL respectively as a result of the low temperature of the dissolutions. A small correction was also made for the amount of material removed in samples prior to completing the experiment. The mass of Pu removed in each 1-mL sample ranged from <0.1 mg to approximately 7 mg. The magnitude of the corrections ranged from <1% to approximately 3%. The procedure used to correct the concentrations and the calculated values are provided in Appendix A.

The corrected Pu and Am concentrations as functions of the dissolution time are plotted in Figure 3-1 and Figure 3-2 for Experiments Pu-Diss-1 and Pu-Diss-2, respectively. The error bars in the figures reflect the one sigma uncertainty in the GPHA (Table A-1 and Table A-2). The uncertainties associated with the volume measurements used to calculate the corrected actinide concentrations were found to be insignificant compared to the uncertainties in the radiochemical analyses.

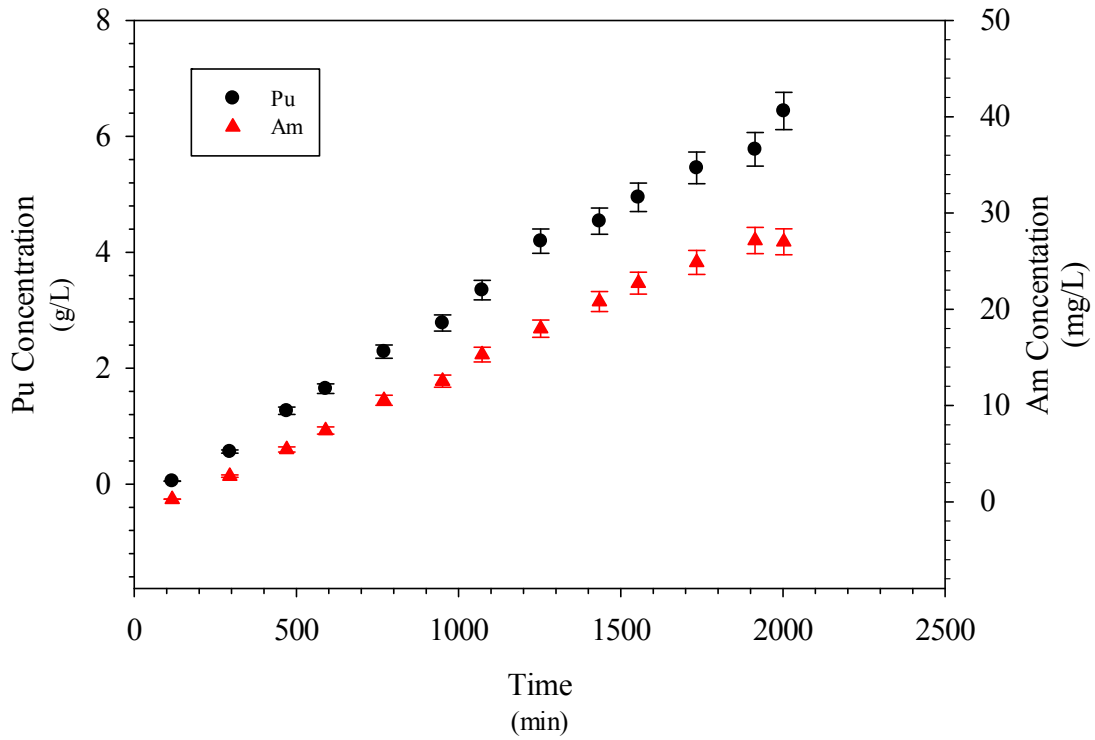


Figure 3-1. Corrected Actinide Concentrations in Experiment Pu-Diss-1

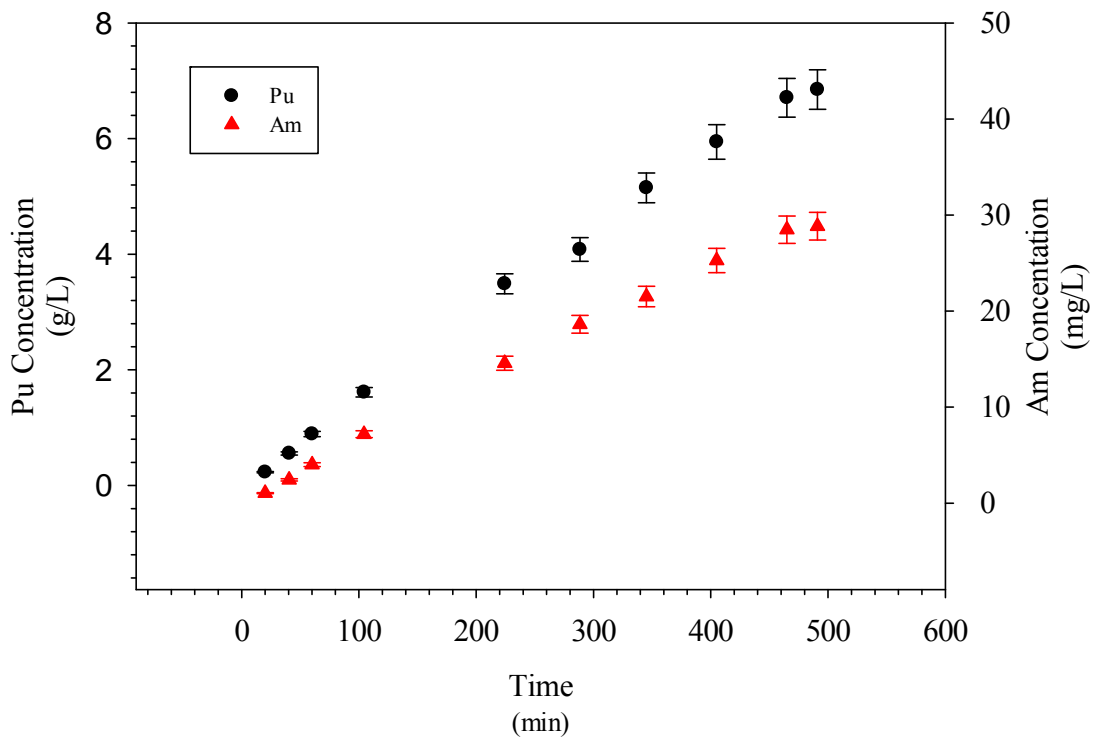


Figure 3-2. Corrected Actinide Concentrations in Experiment Pu-Diss-2

The Pu and Am concentrations (Figure 3-1 and Figure 3-2) are consistent with the observation that complete dissolution was accomplished in each experiment. Each figure shows a steady increase in the actinide concentrations with time which approaches a constant value at the end of the experiment. Table 3-1 provides a comparison of the maximum corrected concentration (at the end of each experiment) to the concentration calculated from the mass of Pu metal and the volume of the dissolving solution used in the experiment. The uncertainties in the Pu concentrations calculated from the metal masses and initial volumes of solution were based on a 1% relative standard deviation in each of the measurements. The calculations are summarized in Appendix A.

Table 3-1. Comparison of Pu Concentrations Based on Corrected Value and Metal Mass

Experiment	Temperature	(Corrected) Pu Concentration	(Mass-based) Pu Concentration	Difference
	(°C)	(g/L)	(g/L)	(%)
Pu-Diss-1	24	6.44 ± 0.32	6.73 ± 0.10	4.35
Pu-Diss-2	57	6.85 ± 0.34	6.75 ± 0.10	-1.49

The Pu concentrations for each experiment show good agreement between the concentration measured by GPHA and the concentration based on the mass of Pu metal dissolved and the volume of the dissolving solution. Differences in the values are likely attributed to uncertainties in the radiochemical analyses and to some extent, impurities in the Pu metal.

To calculate the amount of Pu and Am metal dissolved as a function of time, the estimated solution volume and the Pu and Am concentrations at each sample time were used to calculate the mass of Pu and Am in solution. The calculated mass was expressed as a percentage of the total mass dissolved based on the final mass of Pu and Am in solution at the end of the experiment. The calculations are summarized in Appendix A. Figure 3-3 and Figure 3-4 show the mass of Pu and Am dissolved as a function of time for Experiments Pu-Diss-1 and Pu-Diss-2, respectively. The error bars in the figures were calculated using the one sigma uncertainties in the GPHA (Table A-1 and Table A-2) and propagation of error techniques. The uncertainties associated with the volume measurements used to calculate the amounts of the metals dissolved were found to be insignificant compared to the uncertainties in the radiochemical analyses. The error analysis is summarized in Appendix A.

The percentages of Pu and Am dissolved at each sample time during the experiments were very close in magnitude which indicates that the metal dissolved uniformly. The data for each experiment show that complete dissolution essentially coincided with the removal of the final sample. The dissolution times for the two experiments are given in Table 3-2.

Table 3-2. Time Required for Complete Dissolution of Pu Metal Coupons

Experiment	Temperature	Dissolution Time
	(°C)	(h)
Pu-Diss-1	24	33.4
Pu-Diss-2	57	8.2

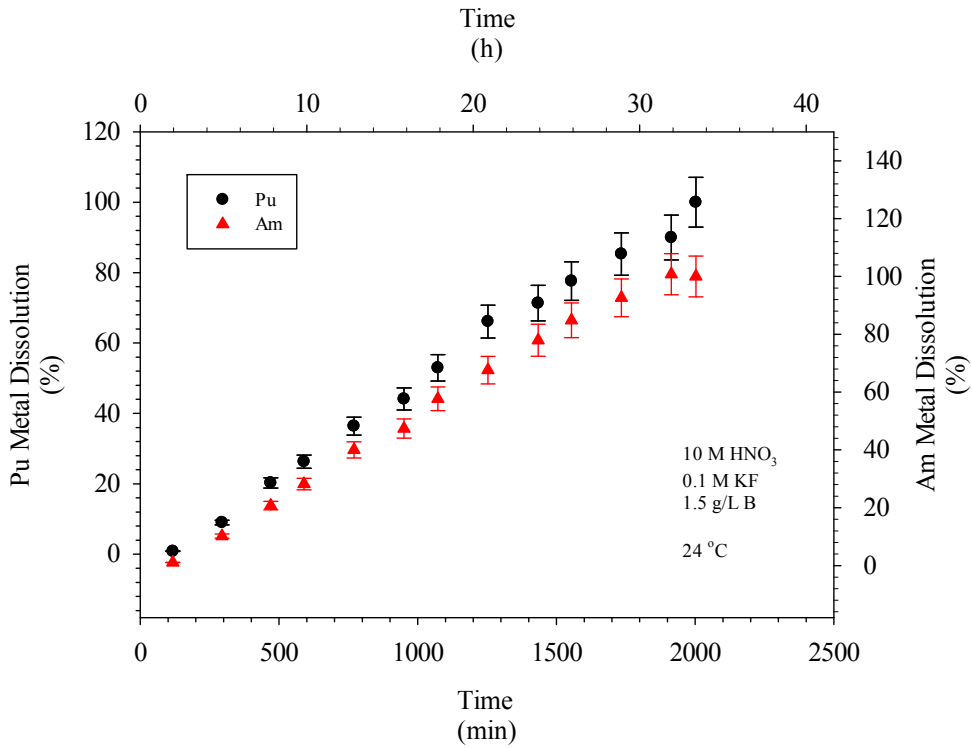


Figure 3-3. Actinide Metal Dissolution in Experiment Pu-Diss-1

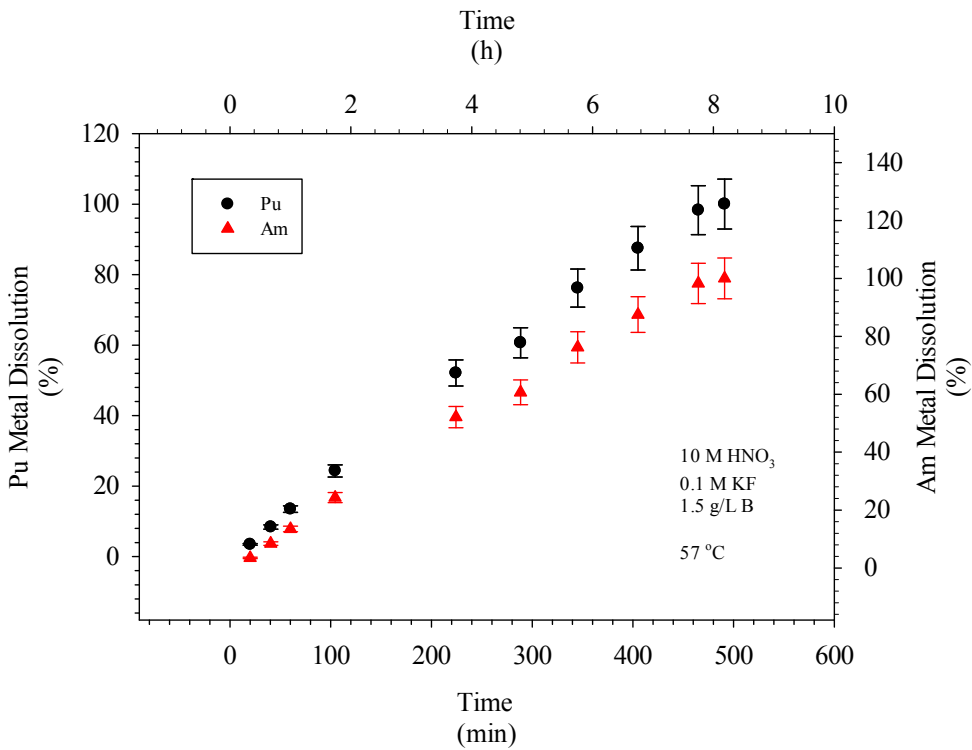


Figure 3-4. Actinide Metal Dissolution in Experiment Pu-Diss-2

3.1.3 Dissolution Rate Measurements

The remaining mass in the Pu metal coupons as a function of time used to calculate the dissolution rates for the ambient (Pu-Diss-1, 24°C) and intermediate temperature (Pu-Diss-2, 57°C) experiments are listed in Table 3-3 and Table 3-4, respectively.

Table 3-3. Pu-Diss-1 Dissolution Time and Weight Measurements (24 °C)

Time (min)	Mass (g)
0	2.4844
117.3	2.4600
294.6	2.2585
470.1	2.0282
590.2	1.8252
770.4	1.5895
950.7	1.3422
1072.9	1.1478
1253.0	0.9236
1434.0	0.6884
1554.0	0.5202
1734.1	0.3155
1914.2	0.0988
2002.8	0.0000

Table 3-4. Pu-Diss-2 Dissolution Time and Weight Measurements (57 °C)

Time (min)	Mass (g)
0	2.6998
20.1	2.5981
40.5	2.4667
60.1	2.3311
104.4	2.0597
164.5	1.7022
224.3	1.3691
288.5	1.0051
345.2	0.7083
405.1	0.4001
465.0	0.1090
491.0	0.0000

The calculated mass to surface area ratio for each sample time is listed for the ambient (Pu-Diss-1, 24°C) and intermediate temperature (Pu-Diss-2, 57°C) dissolution experiments in Table 3-5 and Table 3-6, respectively.

Table 3-5. Pu-Diss-1 Dissolution Time and Pu Mass/Surface Area Ratio (24 °C)

Time (min)	Pu Mass/SA (mg/cm²)
0	457.69
117.3	449.38
294.6	419.65
470.1	383.02
590.2	360.03
770.4	312.27
950.7	275.99
1072.9	228.73
1253.0	208.71
1434.0	152.36
1554.0	118.63
1734.1	78.63
1914.2	24.62
2002.8	0.00

Table 3-6. Pu-Diss-2 Dissolution Time and Pu Mass/Surface Area Ratio (57 °C)

Time (min)	Pu Mass/SA (mg/cm²)
0	449.28
20.1	447.53
40.5	442.22
60.1	418.79
104.4	376.32
164.5	315.00
224.3	261.00
288.5	197.52
345.2	146.51
405.1	97.10
465.0	26.45
491.0	0.00

To calculate the dissolution rates, Figure 3-5 and Figure 3-6 were generated using the data from Table 3-5 and Table 3-6, respectively, where the fitted slope is equal to the dissolution rate in mg/min/cm².

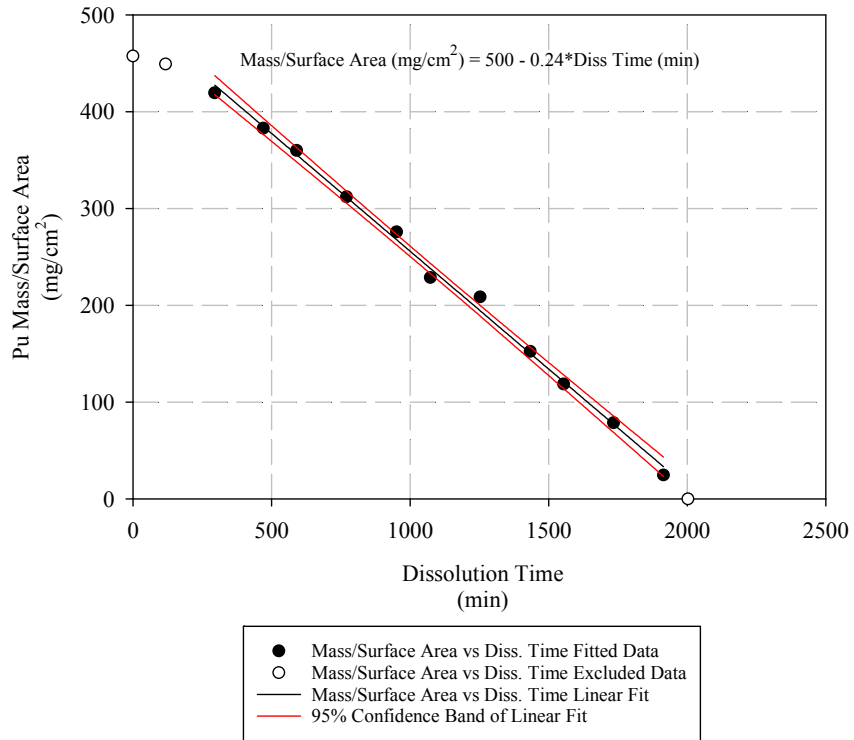


Figure 3-5. Pu Mass to Surface Area versus Dissolution Time at 24 °C (Pu-Diss-1)

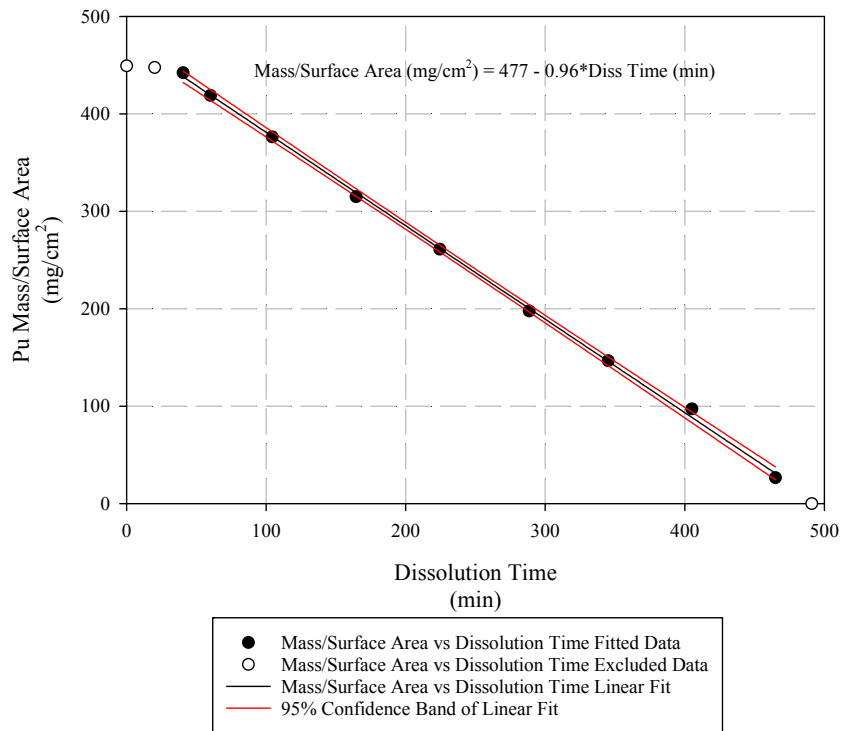


Figure 3-6. Pu Mass to Surface Area versus Dissolution Time at 57 °C (Pu-Diss-2)

Based on a review of the Pu mass to surface area ratio versus dissolution time data at 24°C in Figure 3-5, there was a small induction period of approximately 2 h due to PuO₂ on the surface of the coupon when it was initially placed in the dissolving solution. This conclusion was based on the reduced values for the first two data points. The final data point was not used due to complete dissolution of the Pu metal coupon. A linear fit of the remaining data is shown as equation 1:

$$\frac{M_{\text{Pu}}}{S} = 500 - 0.24D_t \quad (1)$$

where $\frac{M_{\text{Pu}}}{S}$ is the mass to surface area ratio and D_t is the dissolution time. This fit has an adjusted R² of 0.997, indicating the dissolution rate at ambient temperature is fairly linear or constant (not including the induction period). Using the slope from the fit, the dissolution rate at ambient temperature or 24°C is 0.24 ± 0.01 mg/min/cm².

Inspection of the Pu mass to surface area ratio versus dissolution time data at 57°C shows there is a smaller induction period, estimated to be approximately 20 minutes based on the first two data points. The last point was also not used to calculate the dissolution rate due to complete dissolution of the Pu metal coupon. A linear fit of the remaining data is shown as equation 2.

$$\frac{M_{\text{Pu}}}{S} = 477 - 0.96D_t \quad (2)$$

This fit has an adjusted R² of 0.999 indicating the dissolution rate at 57 °C is also fairly linear or constant (not including the induction period). Using the slope from the fit, the dissolution rate at 57°C is 0.96 ± 0.02 mg/min/cm².

3.1.4 Relationship of Pu Metal Dissolution Rate to H-Canyon Processing

The overall dissolution rate of Pu metal is directly proportional to exposed surface area in contact with the dissolving solution. The calculated SSA of the Pu metal coupons used in this work ranged from 2.1-2.2 cm²/g. The SSA of the coupons is greater than two times larger than the DOE 3013 Standard packaging requirement for Pu metal of less than 1 cm²/g.¹⁹ Therefore, the H₂ generation rates that are reported on a Pu mass basis are conservative for all Pu metal that was packaged in 3013 containers.

Holcomb reported the dissolution time of a nominal SRS Pu button as a function of dissolution rate.⁸ The nominal Pu button had a SSA of 0.076 cm²/g. However, Pu metal dissolved in H-Canyon that originates from 3013 containers may have larger SSAs. Plutonium metals and alloys packaged to meet the 3013 Standard have a surface area less than 1 cm²/g. Since the overall dissolution rate is proportional to the Pu metal surface area, the estimated time to completely dissolve a Pu metal charge could range significantly. Assuming a bounding upper SSA of 1 cm²/g for the Pu metal, the overall dissolution rate would be about 13 times faster than the nominal button.

Based on process knowledge using the current flowsheet at boiling (112 - 116 °C),¹ a Pu metal charge was estimated to dissolve in the H-Canyon 6.1D dissolver in 24-36 h. The Pu metal dissolution rate measured at 57 °C is approximately 20 times slower than at boiling, and the dissolution rate at ambient temperature (24 °C) is approximately 80 times slower. Therefore, a Pu metal charge dissolved at 57 °C is estimated to dissolve in the 6.1D dissolver in approximately 20-30 days, and a Pu metal charge dissolved at ambient conditions (24 °C) is estimated to dissolve in about 80-120 days; although, higher ambient temperatures or increased SSAs of the Pu metal will result in shorter dissolution times.

To further evaluate the impact of dissolving temperature, dissolution rates at a broad range of temperatures were compared from available data for similar dissolving solutions in Table 3-7. The estimated H-Canyon processing times are provided for each dissolving temperature.

Table 3-7. Estimated H-Canyon Processing Times from Dissolution Rate Data

Temperature (°C)	HNO ₃ (M)	KF (M)	B (g/L)	Fe (g/L)	Dissolution Rate (mg/min/cm ²)	Est. Processing Time (day)
112 ¹	10	0.1-0.2	1-2	—	20 [*]	1-1.5
100 ¹	10	0.1	1.0	1.9	13.8	1-2
95 ²⁰	6	0.1	1	3.5 ^{**}	5.1	4-6
57	10	0.1	1.5	—	0.96	20-30
24	10	0.1	1.5	—	0.24	80-120

* Estimated dissolution rate¹

** Dissolving solution also contained 0.4 g/L Ni and 0.6 g/L Cr

3.1.5 Pu Metal Offgas Generation

The offgas generation rate during the dissolution of the Pu metal was measured by water displacement in a Tedlar[®] bag system (See Section 2.1.4). A leak check was initially performed by adding sufficient Ar or N₂ to the Tedlar[®] bag system to generate a column of water about 16-cm tall. The Tedlar[®] bag system was then closed and monitored for any observable decrease in the water column height over approximately 3 minutes. After the system integrity was confirmed, the experiment was started. Samples of the offgas were analyzed by GC. The volume correction due to the water pressure on the Tedlar[®] gas bag varied linearly with the amount of gas collected. The volume was also corrected for the tubing volume that was submerged in the water column. The correction factors applied to the volume of offgas collected are provided in Appendix B. The generated offgas volumes measured during Experiments Pu-Offgas-1 (performed at 24 °C) and Pu-Offgas-2 (performed at 57 °C) as a function of the cumulative dissolution time are given in Table 3-8 and Table 3-9, respectively.

Table 3-8. Pu-Offgas-1 Offgas Generation at 24 °C

Cumulative Dissolution Time (min)	Total Offgas Generated (mL)
0.0	0.0
120.6	0.0
240.0	2.7
379.8	2.7
1293.0	5.5
1415.4	5.5
1474.8	11.0
1595.4	11.0
1836.0	11.0
2164.2	11.0
2724.0	11.0
2730.0	11.0

Table 3-9. Pu-Offgas-2 Offgas Generation at 57 °C

Cumulative Reaction Time	Total Offgas Generated
(min)	(mL)
0.0	0.0
61.7	8.2
133.1	22.0
191.4	30.2
252.1	38.5
308.2	46.7
370.4	55.0
428.0	66.0
489.4	74.3
549.4	82.6
578.4	85.4

The overall offgas generation rates based on the data in Table 3-8 and Table 3-9 are 0.004 mL/min for the ambient temperature (24 °C) experiment and 0.15 ml/min for the experiment performed at 57 °C. In earlier work¹ (see Table 3-10, Experiment Pu-5) the offgas generation rate at 100 °C with a similar dissolving solution was 1.6 mL/min. The offgas generation rates at the lower temperatures are orders of magnitude less than at 100 °C. Since the total surface area of the Pu metal pieces dissolved in the offgas experiments was approximately the same (see Section 2.1.1), the offgas generation rates should be proportional to the metal dissolution rates if the dissolution mechanisms are the same. The negligible offgas generation rate observed at 24 °C is consistent with previous observations made by Rudisill et al., where no visible gas generation was observed evolving from Pu metal coupons dissolving in HNO₃ at 20 and 35 °C.¹

The offgas generation rate per mole of dissolved Pu was calculated for Experiment Pu-Offgas-1 and Pu-Offgas-2 since the metal completely dissolved during the offgas collection. The values were calculated using the ideal gas law by assuming the pressure of the gas was 1 atm and the gas was at the temperature of the glovebox. The calculated generation rates are given in Table 3-10. Rudisill et al., using a similar dissolving solution, reported the offgas generation rate per mole of Pu at 100 °C was 0.6 mole offgas/mol Pu.¹ The difference in offgas rate at 100 °C and this work may be indicative of different reaction mechanisms taking place at the lower temperature ranges and perhaps increased solubility of the gas species in the dissolving solution.

Table 3-10. Offgas Generation per Mole of Dissolved Pu

Exp.	Reaction Temp.	HNO₃	KF	B	Fe	Glovebox Temp.	Offgas Gen.
	(°C)	(M)	(M)	(g/L)	(g/L)	(°C)	(mol offgas/mole Pu)
Pu-Offgas-1	24	10	0.1	1.5	—	24	0.035
Pu-Offgas-2	57	10	0.1	1.5	—	25	0.25
Pu-5 ¹	100	10	0.05	1	1.9	18.5	0.6

The mechanisms that impact dissolution and offgas behavior when dissolving Pu metal in HNO₃ between ambient and boiling temperatures are complex and are not well understood. The total offgas rate observed is related to the moles of Pu metal dissolved, the Pu metal dissolution rate, and composition of the gas produced. The primary gases observed during Pu metal dissolution are H₂, NO, NO₂, and N₂O.

Kyser et al. summarized multiple reactions that can take place when dissolving Pu metal in HNO₃.²¹ Additionally, a complex series of solution and gas phase reactions can occur once the NO_x gases are produced.²¹ The subsequent reactions, such as NO₂ reacting with water to reform HNO₃ and HNO₂ (equation 3), can reduce the net volume of offgas measured in the offgas collection system.



Furthermore, increased solubility of gases in the dissolving solution at lower temperatures would lead to a reduced amount of observed offgas. Final solutions from the two offgas experiments were analyzed for ammonium, a possible non-gaseous product from Pu metal dissolution in HNO₃.²¹ Samples from Experiments Pu-Offgas-1 and Pu-Offgas-2 contained only 21 and 5 ppm ammonium, respectively. Therefore, ammonium formation does not significantly account for the decrease in offgas generation at the lower temperatures when compared to boiling conditions.

3.1.6 Pu Metal Offgas Composition

Since the offgas generation was low for the ambient (24°C) and moderate temperature (57°C) dissolution experiments, only one in-line gas bulb was collected at the end of each experiment. A second gas sample was also taken by opening an evacuated sample bulb that was attached directly to the headspace of the dissolver.

The concentrations of H₂, N₂, and O₂ in the offgas samples collected during Experiments Pu-Offgas-1 and Pu-Offgas-2 were obtained by GC. The compositions from the analysis are shown in Table 3-11. Since the H₂ concentrations for both the ambient (24°C) and moderate temperature (57 °C) dissolution experiments were less than detectable (<0.1 vol %) from the in-line and the reactor headspace samples, there was no need to adjust the concentrations to account for dilution from gas in the dissolution vessel, condenser, Viton[®] tubing, and sample bulb.

Oxygen concentrations above the detection limit in the ambient temperature dissolution gas sample bulbs taken at 45.5 h were unexpected as the system was initially purged with N₂ and leak checked. The O₂ is most likely due to a small amount of air in-leakage over the course of the 52.4 h experiment, which was much longer than past experiments performed with the dissolver system. Conversely, the experiment at 57 °C (Pu-Offgas-2) lasted approximately 8 h and the O₂ concentrations were below the GC detection limit.

Table 3-11. Composition of Offgas from Pu Metal Dissolutions in 10 M HNO₃

Exp.	Temperature (°C)	H ₂ (vol %)	O ₂ (vol %)	N ₂ (vol %)	Comment
Pu-Offgas-1	24	<0.1	1.7	98	In line with Tedlar [®] Bag and System for whole dissolution
Pu-Offgas-1	24	<0.1	6.5	93	Pulled from reactor head space at end of dissolution
Pu-Offgas-2	57	<0.1	<0.1	99	In line with Tedlar [®] Bag and System for whole dissolution
Pu-Offgas-2	57	<0.1	<0.1	99	Pulled from reactor head space at end of dissolution

3.1.7 Formation of Pu(VI)

Crapse et al. recently performed Pu metal dissolution experiments at temperatures ranging from 95 to 108 °C using 7 to 8.5 M HNO₃ while monitoring the oxidation of Pu(IV) to Pu(VI) using UV-vis spectroscopy.²⁰ The dissolutions were performed with Fe, Cr, and Ni in the solution to simulate the presence of corrosion products during the dissolution of Pu metal in the 6.1D dissolver. Formation of Pu(VI) was presumed to be facilitated by the presence of Cr(VI) formed in the highly oxidizing HNO₃ solutions. It was determined that lowering the temperature to 95 °C from near boiling dramatically decreased the rate of Pu(VI) formation. The rate of formation of Pu(VI) has also been shown to decrease as HNO₃ molarity increases.²² Therefore, Pu metal dissolutions performed using 10 M HNO₃ and at more reduced temperatures (24 and 57 °C), would be expected to generate even less Pu(VI).

Solution samples from the Pu-Offgas-1 and Pu-Offgas-2 experiments will be analyzed by UV-vis spectroscopy to determine if Pu(VI) is present in the solutions. However, it is unlikely that Pu(VI) will be observed. Rudisill et al. sampled and analyzed the solutions from a series of flowsheet demonstration experiments and reported that the estimated Pu(VI) concentration was significantly less than 1 wt %; although, the actual detection limit was not established. The UV-vis spectra were recorded within approximately one week of the dissolution experiments.¹ From these results, it appears that the presence of Cr(VI) (or another strong oxidant) is likely required to oxidize Pu(IV) to Pu(VI) during metal dissolutions. The Pu metal dissolutions in this study were performed using glassware without the presence of potential corrosion products (e.g., Cr) in the solution (similar to the experiments performed by Rudisill et al.); therefore, the absence of Pu(VI) would not be indicative of what to expect during the dissolution of Pu metal in the 6.1D dissolver. Results from the analysis of solutions from experiments performed as part of this study will be provided to H-Canyon engineering when available.

3.2 Carbon Steel

3.2.1 Dissolution Rate

To evaluate the dissolution and offgas behavior of the carbon steel cans used during Pu metal dissolution, a coupon was cut with dimensions of 26.64 mm x 22.15 mm x 0.32 mm and a mass of 1.0493 g from a can supplied by H-Canyon Engineering. A dissolution experiment was performed at ambient temperature (20-30 °C), which is representative of the dissolving solution temperature during charging of Pu metal to the dissolver. The dissolving solution consisted of a 10 M HNO₃ solution containing 0.1 M fluoride (from KF) and 1.5 g/L B (from boric acid) which is currently used for the Pu metal dissolving solution in H-Canyon. The carbon steel coupon dissolved rapidly in 1 minute and 16 seconds without stirring, resulting in a dissolution rate of 68 mg/min/cm². This rate was much faster than rates measured by Pierce for various carbon steel materials dissolved at ambient temperature in 2M HNO₃, 0.025M KF, and 2 g/L B. An uncoated carbon steel can was reported to dissolve at a rate of up to 4 mg/min/cm² without stirring.⁹ The increased rate of dissolution measured in this study is likely due to the different composition of the dissolving solution which contained higher concentrations of HNO₃ and fluoride and a slightly lower concentration of B.

3.2.2 Offgas Composition

3.2.2.1 Mass Spectrometry

Online analysis of the offgas by mass spectrometry was performed during the carbon steel dissolution. The Monitor MS was initially calibrated with a set of calibration gases containing Ar, N₂, N₂O, NO₂, NO, O₂, and H₂. The standard mixtures were also analyzed as a check before the start of the dissolution experiment. A detailed description of the MS calibration is provided in Appendix C. Once the calibration was complete, the system was purged with 100% Ar while the Monitor MS sampled the

system offgas. After purging the system, a set of baseline or zero values for the offgas concentrations were collected.

The offgas concentrations measured by the Monitor MS for the carbon steel dissolution as a function of the dissolution time based on the time when the carbon steel coupon was dropped into the 10 M HNO₃ solution are shown in Figure 3-7. Tabulated MS offgas concentration data are provided in Appendix D. The H₂ and O₂ concentrations were measured to be zero for the entire dissolution. Therefore, there was no H₂ generation rate.

The largest gas concentration measured was from the Ar purge, which was reduced initially from 100 vol % to 93.1 vol % by the end of the dissolution. The Ar concentration remained high since the total offgas generation was low for the carbon steel coupon dissolution. The next largest gas concentration produced was NO which reached a maximum concentration of 5.8 vol % followed by N₂ which reached a maximum of 1.3 vol %. The NO₂ and N₂O concentrations were low, reaching maximums of 0.5 and 0.1 vol %, respectively.

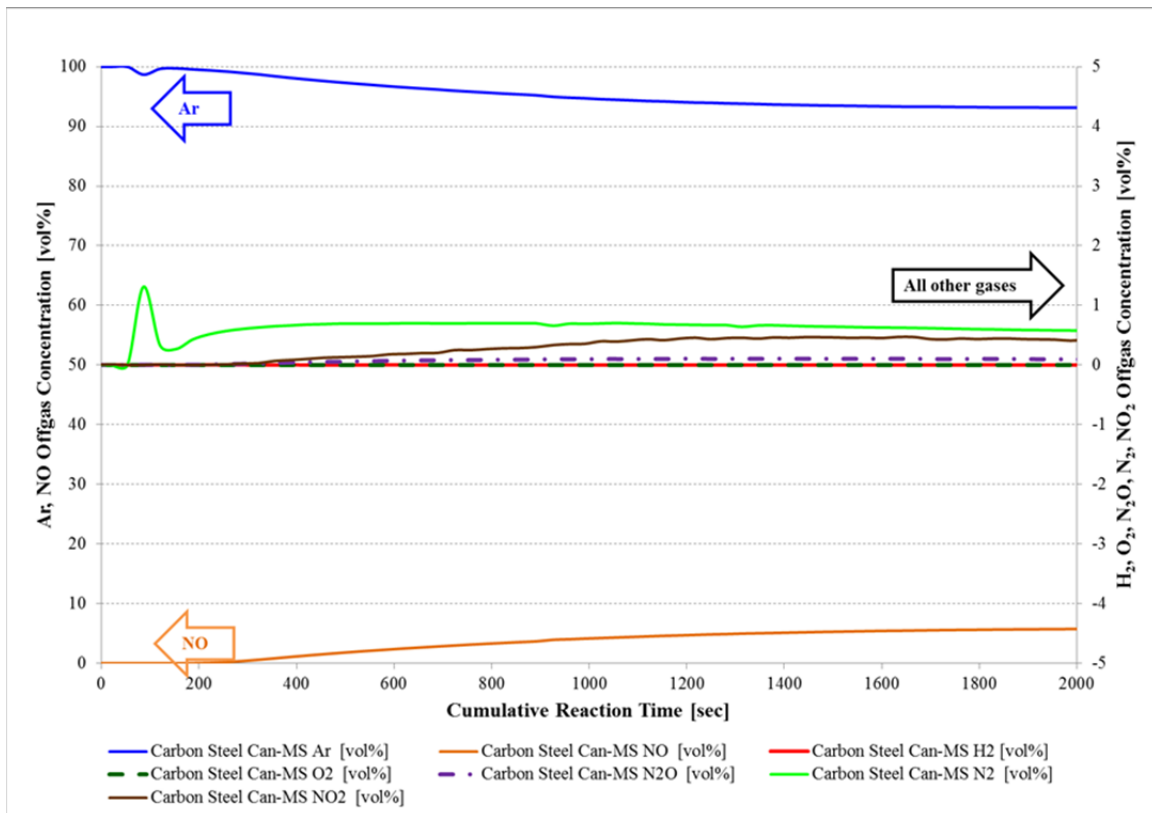


Figure 3-7. MS Analysis for Ambient Temperature Carbon Steel Can Offgas

3.2.2.2 Raman Spectroscopy

The offgas concentrations measured by Raman spectroscopy (which were validated by the Monitor MS concentration measurements) during the carbon steel coupon dissolution are shown in Figure 3-8. The Raman offgas concentration measurements have more noise than the MS values. Therefore, a moving average (MA) of eight data points was used for some of the gases to smooth the offgas profiles. The moving average was calculated using equation 4:

$$y_{MA,t_n} = \frac{\sum_{i=t_n-7}^{t_n} y_i}{8} \quad (4)$$

where t_n is the cumulative dissolution time at step n , y_i is the offgas concentration at time i (vol %), and y_{MA,t_n} is the offgas concentration moving average at time t_n (vol %).

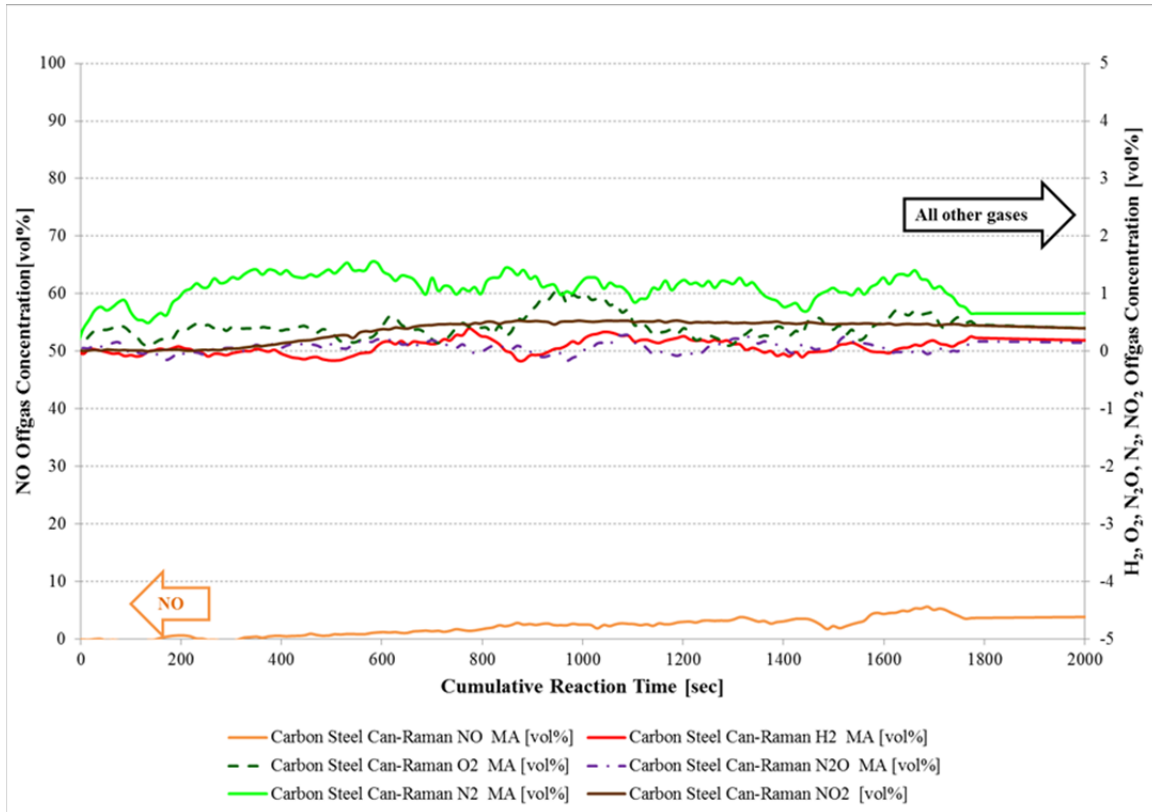


Figure 3-8. Raman Analysis for Ambient Temperature Carbon Steel Can Offgas

The raw and MA offgas concentration data measured by the Raman spectrometer are tabulated in Appendix E. The H₂ and O₂ concentrations, like the concentrations measured by the MS, are essentially zero for the entire dissolution considering the noise in the measurements. Raman spectroscopy cannot detect Ar, but the NO offgas profile is similar to the offgas profile measured by the MS, reaching a maximum NO concentration of 6.2 vol %. The offgas profile for N₂ measured by the Raman spectrometer is similar to the offgas profile measured by the MS, reaching a maximum of 1.6 vol %. The NO₂ concentrations measured by the Raman instrument are also similar to the concentrations measured by the MS, reaching a maximum of 0.5 vol %.

3.3 Nylon

3.3.1 Nylon Dissolution Rate Measurements

Dissolution and offgas measurement experiments were performed to study the dissolution behavior of the nylon bags used as the primary containment for the Pu metal-containing carbon steel cans. To evaluate the dissolution behavior of the nylon bags, a piece cut from a nylon bag supplied by H-Canyon Engineering was dissolved in a solution containing 10 M HNO₃, 0.1 M F, and 1.5 g/L B without stirring.

The nylon weighed 0.0778 g and was completely dissolved in 150 mL of solution over 480 h. The final concentration of dissolved nylon was 0.52 g/L. In another experiment, 0.0764 g of nylon was dissolved in the offgas measurement system described in Section 2.1.2. The water column height did not change over the duration of the experiment, indicating that no measurable offgas was generated. Additionally, there was no evidence of brown gas generated inside the sealed dissolution vessel. Therefore, the nylon bag does not contribute any significant offgas to the Pu metal dissolution process.

3.4 H-Canyon Dissolver Purge

Without controls, the volume of H₂ generated during the dissolution of Pu metal in the H-Canyon dissolver must be maintained below 25% of the LFL. When automatic instrumentation with safety interlocks is provided, the combustion concentration is permitted to be maintained at or below 60% of the LFL.²³ Since the ambient and moderate temperature Pu dissolution experiments generated no detectable H₂ gas, the purge requirements for the 6.1D dissolver were evaluated by assuming the H₂ concentration was conservatively at the detection limit of the gas chromatograph (0.1 vol %). If the H₂ concentration was at the detection limit in the dissolver system, the actual concentrations in the offgas generated during the experiments would be higher due to the dilution from the void volume in the dissolution vessel, condenser, Viton[®] tubing, and sample bulb. The concentration can be estimated by material balance (equation 5),

$$C_{\text{gen}} V_{\text{gen}} = C_{\text{measured}} (V_{\text{void}} + V_{\text{bulb}} + V_{\text{gen}}) \quad (5)$$

where: C_{gen} = concentration of H₂ in the generated gas (vol %)
 C_{measured} = concentration of H₂ measured in the gas sample (vol %)
 V_{gen} = volume of gas collected in the Tedlar[®] bag system (mL)
 V_{void} = void volume of dissolution vessel, condenser, and Viton[®] tubing (mL)
 V_{bulb} = volume of the gas sample bulb (mL).

The calculations are summarized in Table 3-12.

Table 3-12. Estimated H₂ Concentration in Generated Offgas

Experiment	Temp. (°C)	C _{measured} (vol %)	V _{gen} (mL)	V _{void} (mL)	V _{bulb} (mL)	C _{gen} (vol %)
Pu-Offgas-1	24	0.1	11.0	487	40	4.9
Pu-Offgas-2	57	0.1	85.4	460	40	0.7

The moles of H₂ generated in the 6.1D dissolver for a 46 kg charge of Pu metal were estimated from the moles of offgas per mole of Pu dissolved at 24 and 57 °C (Table 3-10) and the generated concentration of H₂ (Table 3-12). The vapor space volume of the 6.1D dissolver is 3655 L,²⁴ which contains 163 moles of air at standard conditions. Therefore, the maximum H₂ concentration in the dissolver can be calculated by assuming that H₂ generated during the Pu metal dissolution displaces air and remains in the dissolver. The presence of water vapor in the dissolver is ignored. The calculations are summarized in Table 3-13.

Table 3-13. Maximum H₂ Concentration in the 6.1D Dissolver

Experiment	Temp. (°C)	H ₂ Gen. per Pu Mass Dissolved (mol/mol)	H ₂ Gen. per Dissolution (mol)	Vapor Space Air (mol)	H ₂ Conc. (vol %)
Pu-Offgas-1	24	0.0017	0.33	163	0.20
Pu-Offgas-2	57	0.0017	0.33	163	0.20

The maximum H₂ concentration calculated for the 6.1D dissolver (Table 3-13) must be compared to the appropriate percentage of the LFL for H₂ at the maximum temperature of the offgas. The LFL for H₂ at 25 °C and 1 atm in air or oxygen is 4 vol %.²⁵ Since the offgas from the 6.1D dissolver flows through the iodine reactor, which operates at 200 °C, the LFL for H₂ must be corrected for the increase in temperature. The LFL is corrected using equation (6),²⁶

$$LFL_T = LFL_{ref} (1 - A(T - T_{ref})) \quad (6)$$

where LFL_T is the LFL at temperature T, LFL_{ref} is the LFL at the reference temperature, A is an empirical coefficient (Zabetakis attenuation factor) equal to 0.0011, and T is the temperature at which the LFL is evaluated. At 200 °C, the LFL of H₂ is 80.8% of the LFL at 25 °C or 3.2 vol %. For comparison to the H₂ concentrations calculated in Table 3-13, the LFL is reduced to 25% (0.80 vol %) and 60% (1.9 vol %) for Pu metal dissolutions performed at ambient temperature and 50-60 °C, respectively. For the ambient temperature dissolution, the H₂ concentration in Table 3-13 is compared to 25% of the H₂ LFL since an interlock would not be operational if steam is not being used to heat the dissolver. The comparison of the maximum H₂ concentrations with the appropriate LFL shows that a dissolver purge is not required for Pu metal dissolutions performed at ambient temperature or 50-60 °C. However, if the dissolver is heated or used to dissolve a full charge at ambient temperature, we recommend the continued use of the sparge to provide agitation in the dissolver.

4.0 Conclusions

The current H-Canyon flowsheet used to dissolve Pu metal for PuO₂ production utilizes boiling HNO₃. The SRNL was requested to develop a complimentary dissolution flowsheet at two reduced temperature ranges. The dissolution and H₂ generation rates of Pu metal were investigated using a dissolving solution at ambient temperature (20-30 °C) and at an intermediate temperature of 50-60 °C. Additionally, the testing included measurement of the ambient temperature dissolution rates and characterization of the off-gas generated from the dissolution of carbon steel cans and the nylon bags that contain the Pu metal when placed in the canyon dissolver.

The dissolving solution for each experiment consisted of 10 M HNO₃, 0.1 M KF, and 1.5 g/L B. The Pu metal dissolving rates were constant throughout the dissolution up to 6.75 g/L Pu, except initially, when the rates were reduced due to the passivation of the metal surface by PuO₂. The Pu metal dissolution rate measured at 57 °C was approximately 20 times slower than at boiling (112 - 116 °C). The dissolution rate at ambient temperature (24 °C) was approximately 80 times slower than the dissolution rate at boiling.

The total dissolution time required in H-Canyon to completely dissolve the Pu metal is dependent upon the dissolving solution temperature as well as the surface area of the Pu metal being dissolved. Based on process knowledge and the current flowsheet at boiling, a Pu metal charge was estimated to dissolve in the H-Canyon 6.1D dissolver in 24-36 h. Therefore, a Pu metal charge dissolved at 57 °C is estimated to dissolve in the 6.1D dissolver in approximately 20-30 days, and a Pu metal charge dissolved at ambient conditions (24 °C) is estimated to dissolve in about 80-120 days; although, higher ambient temperatures or increased SSAs of the Pu metal will result in shorter dissolution times.

Hydrogen concentrations measured by GC in the off-gas from the Pu metal dissolutions at the two temperatures were less than detectable (<0.1 vol %). Dissolution and offgas generation rates were observed to decrease as the temperature decreased. The moles of offgas generated per mole of Pu dissolved also decreased with temperature. Carbon steel can and nylon dissolutions at ambient temperature generated insignificant H₂, and therefore, will not affect offgas flammability.

Using a conservative H₂ concentration of 0.1 vol % and total offgas generation rates measured during Pu metal dissolutions performed at 24 °C and 57 °C, maximum H₂ concentrations were estimated for an H-Canyon dissolver charge of 46 kg of Pu metal. Comparison of the maximum H₂ concentrations with the appropriate LFL showed that a dissolver purge is not required for Pu metal dissolutions performed at ambient temperature or 50-60 °C.

5.0 Flowsheet Recommendations

The use of a 10 M HNO₃ dissolving solution containing 0.1 M KF and 1.5 g/L B at temperatures near 24 and 57 °C effectively dissolve Pu metal at a constant rate up to 6.75 g/L with negligible observed H₂ generation; although, the dissolution rates are much slower than dissolution rates at or near boiling conditions. A dissolver purge is not required for Pu metal dissolutions performed at ambient temperature or 50-60 °C; however, if the dissolver is heated or used to dissolve a full charge at ambient temperature, the continued use of the sparge is recommended to provide agitation in the dissolver.

The low-temperature Pu precipitation issue associated with the use of Gd does not occur for dissolutions with B; however, the B concentration must be maintained below the H₃BO₃ solubility limit and the KF concentration must not exceed the value where B precipitates as KBF₄.

6.0 References

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Appendix A. GPHA Data from Pu Metal Dissolution Rate Measurements

GPHA Data for Pu Dissolutions

Table A-1. GPHA for Pu Dissolution Experiment Pu-Diss-1

Sample ID	Time	²³⁹Pu Activity	1 sigma Uncertainty	²⁴¹Am Activity	1 sigma Uncertainty
	(min)	(dpm/mL)	(%)	(dpm/mL)	(%)
Pu-Diss-1 S1	117	6.69E+06	5.00	2.07E+06	5.00
Pu-Diss-1 S2	295	7.30E+07	5.00	2.04E+07	5.00
Pu-Diss-1 S3	470	1.65E+08	5.00	4.17E+07	5.00
Pu-Diss-1 S4	590	2.15E+08	5.00	5.69E+07	5.00
Pu-Diss-1 S5	770	2.99E+08	5.00	8.10E+07	5.00
Pu-Diss-1 S6	951	3.64E+08	5.00	9.65E+07	5.00
Pu-Diss-1 S7	1073	4.39E+08	5.00	1.18E+08	5.00
Pu-Diss-1 S8	1253	5.51E+08	5.00	1.39E+08	5.00
Pu-Diss-1 S9	1434	5.97E+08	5.00	1.61E+08	5.00
Pu-Diss-1 S10	1554	6.52E+08	5.00	1.76E+08	5.00
Pu-Diss-1 S11	1734	7.20E+08	5.00	1.93E+08	5.00
Pu-Diss-1 S12	1914	7.63E+08	5.00	2.11E+08	5.00
Pu-Diss-1 S13	2003	8.52E+08	5.00	2.10E+08	5.00

Table A-2. GPHA for Pu Dissolution Experiment Pu-Diss-2

Sample ID	Time	²³⁹Pu Activity	1 sigma Uncertainty	²⁴¹Am Activity	1 sigma Uncertainty
	(min)	(dpm/mL)	(%)	(dpm/mL)	(%)
Pu-Diss-2 S1	20	2.96E+07	5.00	7.96E+06	5.00
Pu-Diss-2 S2	41	7.20E+07	5.00	1.85E+07	5.00
Pu-Diss-2 S3	60	1.16E+08	5.00	3.07E+07	5.00
Pu-Diss-2 S4	104	2.11E+08	5.00	5.52E+07	5.00
Pu-Diss-2 S5	165	NA	NA	NA	NA
Pu-Diss-2 S6	224	4.60E+08	5.00	1.13E+08	5.00
Pu-Diss-2 S7	289	5.40E+08	5.00	1.45E+08	5.00
Pu-Diss-2 S8	345	6.84E+08	5.00	1.68E+08	5.00
Pu-Diss-2 S9	405	7.92E+08	5.00	1.98E+08	5.00
Pu-Diss-2 S10	465	8.97E+08	5.00	2.24E+08	5.00
Pu-Diss-2 S11	491	9.17E+08	5.00	2.27E+08	5.00

The Pu and Am activities given in Table A-1 and Table A-2 were converted to a mass basis using the specific activities of ²³⁹Pu and ²⁴¹Am (1.38E+11 and 7.63E+12 dpm/g, respectively) which were calculated from the isotope half-life. The total Pu concentrations were subsequently calculated by assuming the ²³⁹Pu content of weapons grade Pu is 94 wt %. The Pu and Am concentrations for the samples from each dissolution experiment are provided in Table A-3 and Table A-4.

Table A-3. Actinide Concentrations in Samples from Pu Dissolution Experiment Pu-Diss-1

Sample ID	Time	Pu Concentration	1 sigma Uncertainty	Am Concentration	1 sigma Uncertainty
	(min)	(g/L)	(%)	(mg/L)	(%)
Pu-Diss-1 S1	117	0.052	5.00	0.27	5.00
Pu-Diss-1 S2	295	0.56	5.00	2.67	5.00
Pu-Diss-1 S3	470	1.27	5.00	5.47	5.00
Pu-Diss-1 S4	590	1.66	5.00	7.46	5.00
Pu-Diss-1 S5	770	2.30	5.00	10.6	5.00
Pu-Diss-1 S6	951	2.81	5.00	12.6	5.00
Pu-Diss-1 S7	1073	3.38	5.00	15.5	5.00
Pu-Diss-1 S8	1253	4.25	5.00	18.2	5.00
Pu-Diss-1 S9	1434	4.60	5.00	21.1	5.00
Pu-Diss-1 S10	1554	5.03	5.00	23.1	5.00
Pu-Diss-1 S11	1734	5.55	5.00	25.3	5.00
Pu-Diss-1 S12	1914	5.88	5.00	27.7	5.00

Table A-4. Actinide Concentrations in Samples from Pu Dissolution Experiment Pu-Diss-2

Sample ID	Time	Pu Concentration	1 sigma Uncertainty	Am Concentration	1 sigma Uncertainty
	(min)	(g/L)	(%)	(mg/L)	(%)
Pu-Diss-2 S1	20	0.228	5.00	1.04	5.00
Pu-Diss-2 S2	41	0.555	5.00	2.42	5.00
Pu-Diss-2 S3	60	0.894	5.00	4.02	5.00
Pu-Diss-2 S4	104	1.627	5.00	7.23	5.00
Pu-Diss-2 S5	165	NA	NA	NA	NA
Pu-Diss-2 S6	224	3.546	5.00	14.8	5.00
Pu-Diss-2 S7	289	4.163	5.00	19.0	5.00
Pu-Diss-2 S8	345	5.273	5.00	22.0	5.00
Pu-Diss-2 S9	405	6.105	5.00	26.0	5.00
Pu-Diss-2 S10	465	6.915	5.00	29.4	5.00
Pu-Diss-2 S11	491	7.069	5.00	29.8	5.00

Evaporation Rate

The Pu and Am concentrations in each sample were corrected for small changes in volume which occurred due to sample removal and evaporation losses from the dissolver. A small correction was also made for the Pu and Am removed in samples prior to completing the experiment. The volume of each sample removed from the dissolver was 1 mL. The evaporation rate during each experiment was estimated from the initial and final dissolving solution volumes and the total volume removed during sampling. The calculations are summarized in Table A-5.

Table A-5. Evaporation Rate during Pu Dissolution Experiments

Experiment No.	Initial Volume (mL)	Final Volume (mL)	Volume Removed (mL)	Evaporated Volume (mL)	Dissolution Time (min)	Evaporation Rate (mL/min)
Pu-Diss-1	369	353	15	1	2003	0.00050
Pu-Diss-2	400	380	13	7	491	0.014

The calculations in Table A-5 assume the evaporation rate was constant during the dissolving experiments. The estimated volumes of solution in the dissolver for each dissolution experiment prior to the removal of the sample are given in Table A-6 and Table A-7.

Table A-6. Estimated Dissolver Volumes for Pu Dissolution Experiment Pu-Diss-1

Sample ID	Dissolution Time (min)	Dissolver Volume (mL)
Pu-Diss-1 S1	117	369
Pu-Diss-1 S2	295	368
Pu-Diss-1 S3	470	367
Pu-Diss-1 S4	590	366
Pu-Diss-1 S5	770	365
Pu-Diss-1 S6	951	364
Pu-Diss-1 S7	1073	362
Pu-Diss-1 S8	1253	361
Pu-Diss-1 S9	1434	360
Pu-Diss-1 S10	1554	359
Pu-Diss-1 S11	1734	358
Pu-Diss-1 S12	1914	357
Pu-Diss-1 S13	2003	356

Table A-7. Estimated Dissolver Volumes for Pu Dissolution Experiment Pu-Diss-2

Sample ID	Dissolution Time (min)	Dissolver Volume (mL)
Pu-Diss-2 S1	20	400
Pu-Diss-2 S2	41	398
Pu-Diss-2 S3	60	397
Pu-Diss-2 S4	104	396
Pu-Diss-2 S5	165	394
Pu-Diss-2 S6	224	392
Pu-Diss-2 S7	289	390
Pu-Diss-2 S8	345	388
Pu-Diss-2 S9	405	386
Pu-Diss-2 S10	465	384
Pu-Diss-2 S11	491	383

Corrected Actinide Concentrations

The corrected Pu and Am concentrations were calculated by adjusting for the change in volume and accounting for the small amount of material removed from the dissolving solution in each sample. The generalized expression used to calculate the corrected concentrations ($C_{An_{corrected_t}}$) at sample time t is given as equation A.1,

$$C_{An_{corrected_t}} = \frac{C_{An_t} V_t + \sum_{i=1}^{t-1} C_{An_i} V_{s_i}}{V_0} \quad (A.1)$$

where C_{An_t} and C_{An_i} are measured concentrations (Table A-3 and Table A-4), V_t is the estimated volume (Table A-6 and Table A-7), V_{s_i} is the volume of the sample removed (i.e., 1 mL), and V_0 is the initial volume (Table A-5). The corrected concentrations for the actinides are given in Table A-8 and Table A-9. The uncertainties given in the tables reflect the one sigma uncertainties in the GPHA (Table A-1 and Table A-2). The uncertainties associated with the volume measurements used to calculate the corrected actinide concentrations were found to be insignificant compared to the uncertainties in the radiochemical analyses.

Table A-8. Corrected Actinide Concentrations for Pu Dissolution Experiment Pu-Diss-1

Sample ID	Dissolution Time	Pu_{corrected} Concentration	1 sigma Uncertainty	Am_{corrected} Concentration	1 sigma Uncertainty
	(min)	(g/L)	(g/L)	(mg/L)	(mg/L)
Pu-Diss-1 S1	117	0.052	0.003	0.271	0.01
Pu-Diss-1 S2	295	0.561	0.028	2.67	0.13
Pu-Diss-1 S3	470	1.27	0.06	5.44	0.27
Pu-Diss-1 S4	590	1.65	0.08	7.41	0.37
Pu-Diss-1 S5	770	2.29	0.11	10.5	0.5
Pu-Diss-1 S6	951	2.78	0.14	12.5	0.6
Pu-Diss-1 S7	1073	3.35	0.17	15.3	0.8
Pu-Diss-1 S8	1253	4.19	0.21	18.0	0.9
Pu-Diss-1 S9	1434	4.54	0.23	20.8	1.0
Pu-Diss-1 S10	1554	4.95	0.25	22.7	1.1
Pu-Diss-1 S11	1734	5.46	0.27	24.9	1.2
Pu-Diss-1 S12	1914	5.78	0.29	27.1	1.4
Pu-Diss-1 S13	2003	6.44	0.32	27.0	1.4

Table A-9. Corrected Actinide Concentrations for Pu Dissolution Experiment Pu-Diss-2

Sample ID	Dissolution Time	Pu _{corrected} Concentration	1 sigma Uncertainty	Am _{corrected} Concentration	1 sigma Uncertainty
	(min)	(g/L)	(g/L)	(mg/L)	(mg/L)
Pu-Diss-2 S1	20	0.228	0.011	1.04	0.05
Pu-Diss-2 S2	41	0.553	0.028	2.42	0.12
Pu-Diss-2 S3	60	0.890	0.044	4.00	0.20
Pu-Diss-2 S4	104	1.61	0.08	7.17	0.36
Pu-Diss-2 S5	165	NA	NA	NA	NA
Pu-Diss-2 S6	224	3.49	0.17	14.6	0.7
Pu-Diss-2 S7	289	4.08	0.20	18.6	0.9
Pu-Diss-2 S8	345	5.15	0.26	21.5	1.1
Pu-Diss-2 S9	405	5.94	0.30	25.3	1.3
Pu-Diss-2 S10	465	6.71	0.34	28.5	1.4
Pu-Diss-2 S11	491	6.85	0.34	28.8	1.4

Uncertainty in Actinide Concentrations Based on Metal Mass

The expected concentration of Pu at the end of the dissolution ($C_{Pu_{final}}$) was calculated from the mass of metal (M_{Pu}) (Table 2-1) and the initial volume (V_0) of solution used in the experiment (Table A-5). The calculation is illustrated by equation A.2.

$$C_{Pu_{final}} = \frac{M_{Pu}}{V_0} \quad (A.2)$$

To calculate the uncertainty in the expected concentration, the variance in the expected Pu final concentration ($V_{C_{Pu_{final}}}$) is initially calculated using equation A.3,

$$V_{C_{Pu_{final}}} = \left(\frac{\partial C_{Pu_{final}}}{\partial M_{Pu}} \right)^2 V_{M_{Pu}} + \left(\frac{\partial C_{Pu_{final}}}{\partial V_0} \right)^2 V_{V_0} \quad (A.3)$$

where $V_{M_{Pu}}$ and V_{V_0} are the variances in the mass of metal and the initial volume of solution used in the experiment. The one sigma uncertainty in the final concentration ($s_{C_{Pu_{final}}}$) is subsequently calculated from equation A.4.

$$s_{C_{Pu_{final}}} = \sqrt{\left(\frac{1}{V_0} \right)^2 V_{M_{Pu}} + \left(-\frac{M_{Pu}}{V_0^2} \right)^2 V_{V_0}} \quad (A.4)$$

The uncertainties in the Pu concentrations calculated from the metal masses and initial volume of solution were based on a 1% relative standard deviation in each of the measurements. The uncertainties are given in Table 3-1.

Mass of Pu/Am Metal Dissolved

To calculate the amount of Pu and Am metal dissolved as a function of time, the estimated solution volume (Table A-6 and Table A-7) and the corrected Pu and Am concentrations (Table A-8 and Table A-9) at each sample time were used to calculate the mass of Pu and Am in solution (M_{An_t}) (equation A.5).

$$M_{An_t} = C_{An_{corrected_t}} V_t \left(\frac{1}{1000} \right) \quad (A.5)$$

The calculated mass was expressed as a percentage of the total mass dissolved based on the final mass of Pu and Am in solution at the end of the experiment. The calculation is illustrated by equation A.6,

$$D_t = \frac{M_{An_t}}{M_{An_{final}}} (100) \quad (A.6)$$

where D_t is the percentage of the actinide metal dissolved and $M_{An_{final}}$ is the final actinide mass in solution. The calculations are summarized in Table A-10 and Table A-11.

Table A-10. Actinide Metal Dissolved during Experiment Pu-Diss-1

Sample ID	Dissolution Time (min)	Pu Mass Dissolved (g)	1 sigma Uncertainty (g)	Am Mass Dissolved (mg)	1 sigma Uncertainty (mg)	Pu Dissolved (%)	Am Dissolved (%)
Pu-Diss-1 S1	117	0.019	0.001	0.10	0.005	0.83	1.04
Pu-Diss-1 S2	295	0.206	0.010	0.98	0.049	9.01	10.2
Pu-Diss-1 S3	470	0.464	0.023	2.00	0.100	20.3	20.7
Pu-Diss-1 S4	590	0.603	0.030	2.71	0.136	26.3	28.2
Pu-Diss-1 S5	770	0.834	0.042	3.85	0.192	36.4	40.0
Pu-Diss-1 S6	951	1.01	0.05	4.56	0.23	44.1	47.4
Pu-Diss-1 S7	1073	1.21	0.06	5.54	0.28	52.9	57.7
Pu-Diss-1 S8	1253	1.52	0.08	6.50	0.33	66.1	67.6
Pu-Diss-1 S9	1434	1.63	0.08	7.49	0.37	71.3	77.9
Pu-Diss-1 S10	1554	1.78	0.09	8.16	0.41	77.6	84.8
Pu-Diss-1 S11	1734	1.95	0.10	8.91	0.45	85.3	92.6
Pu-Diss-1 S12	1914	2.06	0.10	9.69	0.48	90.0	100
Pu-Diss-1 S13	2003	2.29	0.11	9.6	0.5	100	100

Table A-11. Actinide Metal Dissolved during Experiment Pu-Diss-2

Sample ID	Dissolution Time	Pu Mass Dissolved	1 sigma Uncertainty	Am Mass Dissolved	1 sigma Uncertainty	Pu Dissolved	Am Dissolved
	(min)	(g)	(g)	(mg)	(mg)	(%)	(%)
Pu-Diss-2 S1	20	0.091	0.005	0.417	0.021	3.48	3.77
Pu-Diss-2 S2	41	0.220	0.011	0.963	0.048	8.41	8.72
Pu-Diss-2 S3	60	0.353	0.02	1.59	0.08	13.5	14.4
Pu-Diss-2 S4	104	0.638	0.03	2.84	0.14	24.3	25.7
Pu-Diss-2 S5	165	NA	NA	NA	NA	NA	NA
Pu-Diss-2 S6	224	1.37	0.07	5.71	0.29	52.1	51.7
Pu-Diss-2 S7	289	1.59	0.08	7.26	0.36	60.7	65.8
Pu-Diss-2 S8	345	2.00	0.10	8.35	0.42	76.2	75.6
Pu-Diss-2 S9	405	2.29	0.11	9.76	0.49	87.5	88.4
Pu-Diss-2 S10	465	2.58	0.13	10.95	0.55	98.3	99.1
Pu-Diss-2 S11	491	2.62	0.1	11.0	0.6	100	100

Uncertainties in the Masses of Pu and Am Metal Dissolved

The amounts of Pu and Am metal dissolved as functions of time were calculated as percentages of the total mass dissolved based on the final mass of Pu and Am in solution (Table A-10 and Table A-11). To calculate the uncertainty in the percentage of the actinide metals dissolved, the variance in D_t (V_{D_t}) is initially calculated using equation A.7,

$$V_{D_t} = \left(\frac{\partial D_t}{\partial M_{An_t}} \right)^2 V_{M_{An_t}} + \left(\frac{\partial D_t}{\partial M_{An_{final}}} \right)^2 V_{M_{An_{final}}} \quad (A.7)$$

where $V_{M_{An_t}}$ and $V_{M_{An_{final}}}$ are the variances in the mass of the actinide in solution at time (t) and the final actinide mass in solution, respectively. The one sigma uncertainty (s_{D_t}) is subsequently calculated from equation A.8.

$$s_{D_t} = \sqrt{\left(\frac{100}{M_{An_{final}}} \right)^2 V_{M_{An_t}} + \left(-\frac{100 M_{An_t}}{M_{An_{final}}^2} \right)^2 V_{M_{An_{final}}}} \quad (A.8)$$

The mass of Pu and Am metal dissolved as a function of time was calculated using equation A.5. The variance in the mass of the actinides ($V_{M_{An_t}}$) is initially calculated using equation A.9.

$$V_{M_{An_t}} = \left(\frac{dM_{An_t}}{dC_{An_{corrected_t}}} \right)^2 V_{C_{An_{corrected_t}}} \quad (A.9)$$

The one sigma uncertainties in the dissolved masses are subsequently calculated from equation A.10 and are provided in Table A-11 and Table A-12 with the calculated Pu and Am masses.

$$S_{M_{\text{Act}}_t} = \sqrt{\left(\frac{V_t}{1000}\right)^2 V_{C_{\text{Act}}^{\text{corrected}_t}}} \quad (\text{A.10})$$

The uncertainties in the amount of Pu and Am metal dissolved as a function of time were calculated using equation A.8 and the data in Table A-10 and Table A-11. The results from the calculations are given in Table A-12 and Table A-13 for experiments Pu-Diss-1 and Pu-Diss-2, respectively.

Table A-12. Uncertainties in the Actinide Metal Dissolved during Experiment Pu-Diss-1

Sample ID	Dissolution Time (min)	Pu Dissolved (%)	1 sigma Uncertainty (%)	Am Dissolved (%)	1 sigma Uncertainty (%)
Pu-Diss-1 S1	117	0.83	0.06	1.04	0.07
Pu-Diss-1 S2	295	9.01	0.64	10.2	0.7
Pu-Diss-1 S3	470	20.3	1.4	20.7	1.5
Pu-Diss-1 S4	590	26.3	1.9	28.2	2.0
Pu-Diss-1 S5	770	36.4	2.6	40.0	2.8
Pu-Diss-1 S6	951	44.1	3.1	47.4	3.3
Pu-Diss-1 S7	1073	52.9	3.7	57.7	4.1
Pu-Diss-1 S8	1253	66.1	4.7	67.6	4.8
Pu-Diss-1 S9	1434	71.3	5.0	77.9	5.5
Pu-Diss-1 S10	1554	77.6	5.5	84.8	6.0
Pu-Diss-1 S11	1734	85.3	6.0	92.6	6.5
Pu-Diss-1 S12	1914	90.0	6.4	101	7.1
Pu-Diss-1 S13	2003	100	7	100	7.1

Table A-13. Uncertainties in the Actinide Metal Dissolved during Experiment Pu-Diss-2

Sample ID	Dissolution Time (min)	Pu Dissolved (%)	1 sigma Uncertainty (%)	Am Dissolved (%)	1 sigma Uncertainty (%)
Pu-Diss-2 S1	20	3.48	0.25	3.77	0.27
Pu-Diss-2 S2	41	8.41	0.6	8.72	0.62
Pu-Diss-2 S3	60	13.5	1.0	14.4	1.02
Pu-Diss-2 S4	104	24.3	1.7	25.7	1.8
Pu-Diss-2 S5	165	NA	NA	NA	NA
Pu-Diss-2 S6	224	52.1	3.7	51.7	3.7
Pu-Diss-2 S7	289	60.7	4.3	65.8	4.7
Pu-Diss-2 S8	345	76.2	5.4	75.6	5.3
Pu-Diss-2 S9	405	87.5	6.2	88.4	6.2
Pu-Diss-2 S10	465	98.3	7.0	99.1	7.0
Pu-Diss-2 S11	491	100	7	100	7.1

Appendix B. Tedlar[®] Bag Measurement Method

The off-gas volume generated during a Pu metal dissolution experiment was tracked by recording height changes in the water column of the Tedlar[®] Bag system. The water column heights were then used to calculate the volumes of generated gas. The volumes of gas were then corrected for the slight pressure created by the rising water column above the Tedlar[®] bag collection system as shown in Figure B-1. The volume was also corrected for the tubing volume that is submerged in the water column. The inner diameter of the water column on top of the Tedlar[®] Bag system was 5.88 cm giving a cross-sectional area of 27.19 cm². The Viton[®] tubing that went down through the water column had an outer diameter of 0.6 cm with a cross-sectional area of 0.283 cm². The total cross-sectional area of water in the column was then 27.19-0.283=26.91 cm². Therefore, the offgas volume generated before a pressure correction based on the water column height is given by equation B.1:

$$V_{\text{tube}_{\text{corr}}} = 26.91H \quad (\text{B.1})$$

where $V_{\text{tube}_{\text{corr}}}$ is the volume of offgas generated before the pressure correction and H is the height of the water column.

The pressure/volume correction to the offgas generation ($V_{\text{correction}}$) is based on the height of water above the Tedlar[®] Bag which follows the correlation given by equation B.2.

$$V_{\text{correction}} = 9.672 \times 10^{-5}H + 2.027 \times 10^{-2} \quad (\text{B.2})$$

Therefore, combining Equation B.1 and B.2 gives the offgas generated for the height of water column (V_{offgas}) corrected for the displacement of the Viton[®] tubing and the extra weight of the water column (equation B.3):

$$V_{\text{offgas}} = 26.91 H \left(1 + \left(9.672 \times 10^{-5}H + 2.027 \times 10^{-2} \right) \right) \quad (\text{B.3})$$

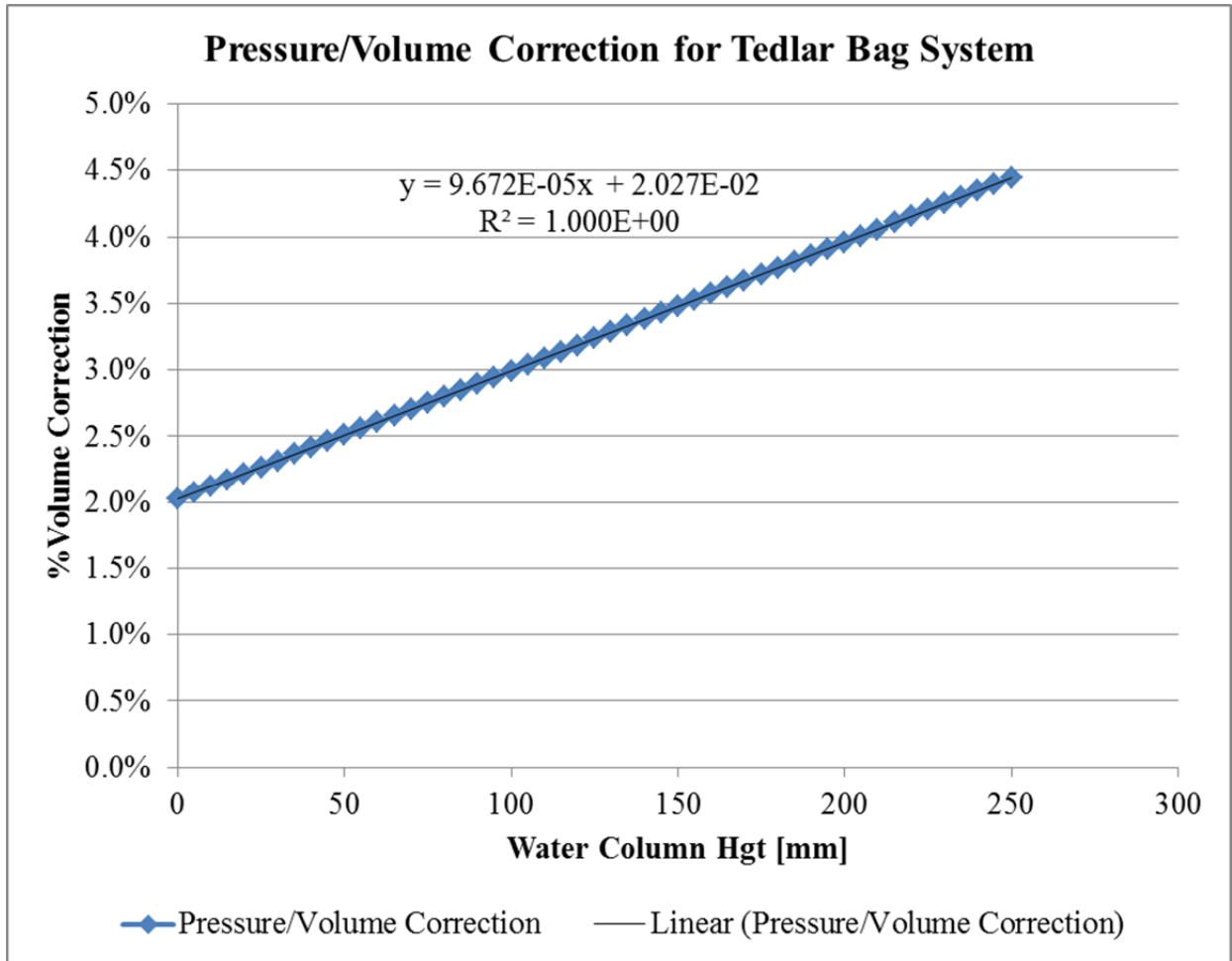


Figure B-1. Pressure/Volume Correction for Tedlar® Bag System

Appendix C. Monitor MS Calibration and Sampling Method

The Monitor MS was calibrated using a set of calibration gases as shown in Table 2-2 before the start of each dissolution experiment. The calibration gases were then sampled back through the MS to check the calibration. For the carbon steel can dissolution, the calibration checks are shown in Table C-1. The calibration check tells us if the calibration was successful and provides an indication of the variance of the measurements since the calibration gas is read for several samples.

Table C-1. Pre-run Check of Calibration Gases for Carbon Steel Can Dissolution

Gas	Ar	N ₂	N ₂ O	NO ₂	NO	O ₂	H ₂
	%	%	%	%	%	%	%
20% N ₂ O-80% Ar	79.32	0.00	20.34	0.31	0.01	0.00	0.02
	79.35	0.00	20.34	0.27	0.03	0.00	0.02
	79.39	0.00	20.28	0.28	0.03	0.00	0.02
	79.38	0.00	20.31	0.26	0.03	0.00	0.02
	79.28	0.00	20.41	0.26	0.03	0.00	0.02
4.98% NO ₂ -20.13% O ₂ -74.89% Ar	76.28	0.27	0.03	4.93	0.00	18.47	0.02
	75.67	0.27	0.03	5.58	0.02	18.41	0.02
	75.56	0.27	0.02	5.70	0.02	18.40	0.03
	75.56	0.27	0.02	5.70	0.02	18.41	0.02
	75.54	0.27	0.03	5.74	0.01	18.39	0.03
20% NO-80% Ar	80.01	0.12	0.05	0.12	19.67	0.00	0.03
	80.04	0.11	0.05	0.08	19.69	0.00	0.03
	80.06	0.11	0.05	0.06	19.70	0.00	0.02
	80.06	0.11	0.05	0.05	19.70	0.00	0.03
	80.05	0.11	0.05	0.07	19.70	0.00	0.03
5% N ₂ -10% H ₂ -85% Ar	84.86	4.77	0.01	0.00	0.01	0.00	10.36
	84.76	4.76	0.01	0.03	0.00	0.00	10.44
	84.77	4.75	0.01	0.02	0.00	0.00	10.44
	84.80	4.73	0.01	0.01	0.00	0.00	10.44
	84.83	4.71	0.01	0.01	0.00	0.00	10.43
100% Ar	99.94	0.03	0.00	0.01	0.00	0.00	0.02
	99.93	0.05	0.00	0.00	0.00	0.00	0.02
	99.94	0.03	0.00	0.01	0.00	0.00	0.02
	99.95	0.02	0.01	0.00	0.00	0.00	0.02
	99.94	0.02	0.00	0.02	0.00	0.00	0.02
100% N ₂	0.00	97.83	0.05	0.00	0.00	2.09	0.03
	0.00	97.88	0.05	0.02	0.00	2.02	0.03
	0.00	97.98	0.05	0.00	0.00	1.95	0.03
	0.00	97.98	0.05	0.03	0.00	1.90	0.03
	0.00	98.08	0.05	0.00	0.00	1.85	0.03
Air (78.0% N ₂ -21.0% O ₂ -0.9% Ar)	0.67	75.62	0.09	0.02	0.00	23.56	0.04
	0.67	75.57	0.09	0.02	0.00	23.61	0.04
	0.67	75.56	0.10	0.02	0.00	23.62	0.04
	0.67	75.57	0.09	0.01	0.00	23.62	0.04
	0.67	75.51	0.10	0.02	0.00	23.67	0.04

Once the calibration is complete, the system is purged with 100% Ar while the Monitor MS is sampling the system offgas. By purging the system with 100% Ar, a set of baseline or zero values for the offgas concentrations can be collected and analyzed. The baseline values for the carbon steel can dissolution experiment are shown in Table C-2. These baseline values represent zero or 100% concentrations within the accuracy of the Monitor MS. In other words, the raw N₂, N₂O, NO₂, NO, O₂, and H₂ MS readings are re-baselined by subtracting off the average baseline values shown in Table C-2. The raw Ar MS reading is re-baselined to 100% by dividing by the average baseline value shown in Table C-2 while limiting the maximum to 100%. These re-baseline steps are not necessary in dissolutions which generate a lot of offgas, but become necessary when there is little offgas generation like during the dissolution of the carbon steel coupon.

Table C-2. Monitor MS Baseline Offgas Concentrations

Sample	Ar	N ₂	N ₂ O	NO ₂	NO	O ₂	H ₂
	%	%	%	%	%	%	%
1	99.68	0.21	0.02	0.01	0.00	0.00	0.07
2	99.71	0.20	0.02	0.01	0.00	0.00	0.06
3	99.74	0.18	0.02	0.00	0.00	0.00	0.06
4	99.74	0.16	0.02	0.01	0.00	0.00	0.06
5	99.76	0.15	0.02	0.01	0.00	0.00	0.07
6	99.77	0.14	0.02	0.00	0.00	0.00	0.07
Average	99.73	0.17	0.02	0.01	0.00	0.00	0.07
Re-baseline	100	0	0	0	0	0	0

At the end of the dissolution experiment, the H₂ calibration gas is re-sampled by the MS to see if the instrument is still reading the offgas concentrations accurately. Table C-3 shows multiple samples of the H₂ calibration gas read by the Monitor MS. Comparing the pre and post run calibration checks of the H₂ calibration gas shows that the Monitor MS is reading offgas concentrations accurately and the offgas concentrations measured during the experiment are valid.

Table C-3. Post-run Check of Calibration Gas for Carbon Steel Dissolution

Gas	Ar	N ₂	N ₂ O	NO ₂	NO	O ₂	H ₂
	%	%	%	%	%	%	%
5% N ₂ -10% H ₂ -85% Ar	84.30	4.74	0.03	0.01	0.01	0.00	10.36
	84.38	4.74	0.02	0.01	0.00	0.00	10.44
	84.20	4.75	0.02	0.01	0.00	0.00	10.44
	84.15	4.76	0.03	0.01	0.00	0.00	10.44
	84.17	4.75	0.02	0.00	0.00	0.00	10.43

Appendix D. MS Offgas Data for Carbon Steel

Table D-1. Raw Monitor MS Offgas Concentration Data for Room Temp Carbon Steel Dissolution

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	Ar	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:08:32	0	n/a	n/a	n/a	n/a	n/a	n/a	n/a
12/16/15 9:08:51	19	0.07	0.12	0.00	0.00	99.77	0.03	0.02
12/16/15 9:09:25	53	0.07	0.17	0.00	0.00	99.74	0.02	0.01
12/16/15 9:09:58	86	0.07	1.48	0.00	0.00	98.42	0.03	0.00
12/16/15 9:10:32	120	0.07	0.50	0.00	0.00	99.39	0.02	0.02
12/16/15 9:11:05	153	0.07	0.44	0.00	0.00	99.47	0.02	0.00
12/16/15 9:11:39	187	0.06	0.60	0.01	0.00	99.30	0.03	0.01
12/16/15 9:12:13	221	0.07	0.69	0.07	0.00	99.14	0.03	0.01
12/16/15 9:12:47	255	0.07	0.74	0.19	0.00	98.95	0.04	0.02
12/16/15 9:13:20	288	0.07	0.78	0.37	0.00	98.71	0.04	0.03
12/16/15 9:13:54	322	0.07	0.80	0.59	0.00	98.46	0.05	0.04
12/16/15 9:14:27	355	0.07	0.82	0.83	0.00	98.15	0.05	0.08
12/16/15 9:15:00	388	0.07	0.84	1.07	0.00	97.87	0.06	0.09
12/16/15 9:15:34	422	0.07	0.85	1.30	0.00	97.61	0.07	0.11
12/16/15 9:16:08	456	0.07	0.86	1.52	0.00	97.35	0.07	0.13
12/16/15 9:16:41	489	0.07	0.86	1.73	0.00	97.12	0.08	0.14
12/16/15 9:17:15	523	0.07	0.87	1.94	0.00	96.90	0.08	0.15
12/16/15 9:17:48	556	0.07	0.87	2.13	0.00	96.69	0.09	0.16
12/16/15 9:18:22	590	0.07	0.87	2.32	0.00	96.47	0.09	0.19
12/16/15 9:18:55	623	0.07	0.87	2.49	0.00	96.28	0.10	0.19
12/16/15 9:19:29	657	0.07	0.87	2.66	0.00	96.10	0.10	0.21
12/16/15 9:20:02	690	0.07	0.87	2.82	0.00	95.93	0.10	0.21
12/16/15 9:20:36	724	0.07	0.87	2.98	0.00	95.73	0.11	0.26
12/16/15 9:21:09	757	0.07	0.87	3.13	0.00	95.57	0.11	0.26
12/16/15 9:21:43	791	0.06	0.87	3.28	0.00	95.41	0.11	0.28
12/16/15 9:22:16	824	0.07	0.87	3.41	0.00	95.25	0.11	0.29
12/16/15 9:22:50	858	0.06	0.87	3.55	0.00	95.12	0.11	0.29
12/16/15 9:23:23	891	0.06	0.87	3.67	0.00	94.97	0.12	0.31
12/16/15 9:23:59	927	0.06	0.83	3.94	0.00	94.71	0.12	0.34
12/16/15 9:24:31	959	0.07	0.86	4.02	0.00	94.57	0.12	0.36
12/16/15 9:25:03	991	0.06	0.86	4.13	0.00	94.46	0.12	0.36
12/16/15 9:25:36	1024	0.06	0.87	4.23	0.00	94.32	0.12	0.40
12/16/15 9:26:08	1056	0.06	0.87	4.32	0.00	94.22	0.12	0.40

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	Ar	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:26:40	1088	0.06	0.87	4.41	0.00	94.11	0.12	0.42
12/16/15 9:27:12	1120	0.07	0.86	4.50	0.00	94.01	0.13	0.44
12/16/15 9:27:44	1152	0.06	0.85	4.60	0.00	93.94	0.12	0.42
12/16/15 9:28:17	1185	0.07	0.85	4.67	0.00	93.84	0.13	0.45
12/16/15 9:28:49	1217	0.07	0.84	4.75	0.00	93.74	0.13	0.46
12/16/15 9:29:21	1249	0.06	0.84	4.83	0.00	93.70	0.12	0.44
12/16/15 9:29:53	1281	0.06	0.84	4.89	0.00	93.62	0.13	0.46
12/16/15 9:30:25	1313	0.06	0.81	4.98	0.00	93.56	0.13	0.46
12/16/15 9:30:58	1346	0.06	0.84	5.03	0.00	93.50	0.13	0.45
12/16/15 9:31:30	1378	0.07	0.84	5.08	0.00	93.42	0.13	0.47
12/16/15 9:32:02	1410	0.07	0.83	5.14	0.00	93.37	0.13	0.46
12/16/15 9:32:34	1442	0.07	0.82	5.19	0.00	93.32	0.13	0.47
12/16/15 9:33:06	1474	0.07	0.82	5.25	0.00	93.27	0.13	0.47
12/16/15 9:33:38	1506	0.06	0.81	5.29	0.00	93.23	0.13	0.47
12/16/15 9:34:10	1538	0.07	0.81	5.34	0.00	93.20	0.13	0.46
12/16/15 9:34:42	1570	0.07	0.80	5.38	0.00	93.16	0.13	0.47
12/16/15 9:35:14	1602	0.07	0.80	5.43	0.00	93.12	0.13	0.46
12/16/15 9:35:46	1634	0.07	0.80	5.46	0.00	93.07	0.13	0.48
12/16/15 9:36:19	1667	0.07	0.79	5.50	0.00	93.04	0.13	0.48
12/16/15 9:36:51	1699	0.06	0.79	5.53	0.00	93.05	0.12	0.44
12/16/15 9:37:23	1731	0.06	0.78	5.57	0.00	93.03	0.12	0.44
12/16/15 9:37:55	1763	0.07	0.78	5.59	0.00	92.99	0.12	0.45
12/16/15 9:38:27	1795	0.07	0.77	5.62	0.00	92.98	0.13	0.44
12/16/15 9:38:59	1827	0.07	0.77	5.65	0.00	92.95	0.12	0.45
12/16/15 9:39:31	1859	0.07	0.76	5.66	0.00	92.93	0.12	0.45
12/16/15 9:40:03	1891	0.07	0.76	5.69	0.00	92.93	0.12	0.44
12/16/15 9:40:35	1923	0.06	0.76	5.71	0.00	92.92	0.12	0.44
12/16/15 9:41:07	1955	0.07	0.75	5.72	0.00	92.91	0.12	0.43
12/16/15 9:41:39	1987	0.07	0.75	5.74	0.00	92.91	0.12	0.42
12/16/15 9:42:11	2019	0.07	0.75	5.75	0.00	92.89	0.12	0.43
12/16/15 9:42:44	2052	0.07	0.74	5.77	0.00	92.89	0.12	0.41
12/16/15 9:43:16	2084	0.06	0.74	5.78	0.00	92.90	0.12	0.41
12/16/15 9:43:48	2116	0.06	0.73	5.79	0.00	92.88	0.12	0.42
12/16/15 9:44:20	2148	0.07	0.73	5.79	0.00	92.88	0.12	0.42
12/16/15 9:44:52	2180	0.06	0.73	5.81	0.00	92.89	0.11	0.40
12/16/15 9:45:24	2212	0.06	0.72	5.81	0.00	92.89	0.11	0.40
12/16/15 9:45:56	2244	0.06	0.72	5.81	0.00	92.89	0.11	0.40
12/16/15 9:46:29	2277	0.07	0.71	5.81	0.00	92.87	0.11	0.42

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	Ar	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:47:01	2309	0.07	0.71	5.82	0.00	92.89	0.11	0.41
12/16/15 9:47:33	2341	0.07	0.70	5.82	0.00	92.89	0.11	0.41
12/16/15 9:48:06	2374	0.06	0.70	5.83	0.00	92.90	0.11	0.40
12/16/15 9:48:38	2406	0.07	0.70	5.82	0.00	92.90	0.11	0.40
12/16/15 9:49:10	2438	0.07	0.69	5.82	0.00	92.91	0.11	0.40
12/16/15 9:49:42	2470	0.07	0.69	5.82	0.00	92.92	0.11	0.40
12/16/15 9:50:14	2502	0.07	0.68	5.82	0.00	92.93	0.11	0.39
12/16/15 9:50:46	2534	0.07	0.68	5.82	0.00	92.95	0.11	0.39
12/16/15 9:51:18	2566	0.07	0.67	5.82	0.00	92.96	0.10	0.39
12/16/15 9:51:50	2598	0.07	0.67	5.80	0.00	92.96	0.10	0.40
12/16/15 9:52:23	2631	0.07	0.67	5.80	0.00	92.99	0.10	0.38
12/16/15 9:52:55	2663	0.07	0.66	5.80	0.00	93.00	0.10	0.37
12/16/15 9:53:27	2695	0.07	0.66	5.79	0.00	93.00	0.10	0.38
12/16/15 9:53:59	2727	0.06	0.66	5.79	0.00	93.03	0.10	0.36
12/16/15 9:54:31	2759	0.06	0.65	5.78	0.00	93.05	0.10	0.35
12/16/15 9:55:03	2791	0.07	0.65	5.77	0.00	93.06	0.10	0.36
12/16/15 9:55:35	2823	0.07	0.65	5.76	0.00	93.08	0.10	0.35
12/16/15 9:56:08	2856	0.07	0.65	5.75	0.00	93.06	0.10	0.38
12/16/15 9:56:40	2888	0.07	0.64	5.74	0.00	93.10	0.09	0.36
12/16/15 9:57:12	2920	0.07	0.64	5.73	0.00	93.12	0.09	0.35
12/16/15 9:57:44	2952	0.07	0.64	5.72	0.00	93.14	0.09	0.35
12/16/15 9:58:16	2984	0.07	0.64	5.70	0.00	93.14	0.10	0.36
12/16/15 9:58:48	3016	0.07	0.63	5.69	0.00	93.17	0.09	0.35
12/16/15 9:59:20	3048	0.07	0.63	5.68	0.00	93.18	0.09	0.36
12/16/15 9:59:52	3080	0.07	0.63	5.66	0.00	93.20	0.09	0.35
12/16/15 10:00:25	3113	0.06	0.62	5.66	0.00	93.23	0.09	0.34
12/16/15 10:00:57	3145	0.06	0.62	5.64	0.00	93.25	0.09	0.33
12/16/15 10:01:29	3177	0.07	0.62	5.63	0.00	93.27	0.09	0.32
12/16/15 10:02:01	3209	0.06	0.62	5.61	0.00	93.30	0.09	0.33
12/16/15 10:02:33	3241	0.07	0.62	5.59	0.00	93.29	0.09	0.35
12/16/15 10:03:05	3273	0.06	0.61	5.58	0.00	93.33	0.08	0.33
12/16/15 10:03:37	3305	0.07	0.61	5.57	0.00	93.35	0.08	0.33
12/16/15 10:04:10	3338	0.07	0.61	5.55	0.00	93.36	0.08	0.34
12/16/15 10:04:42	3370	0.07	0.60	5.54	0.00	93.40	0.08	0.32
12/16/15 10:05:14	3402	0.07	0.60	5.51	0.00	93.39	0.09	0.34
12/16/15 10:05:46	3434	0.07	0.60	5.50	0.00	93.41	0.08	0.34
12/16/15 10:06:18	3466	0.07	0.60	5.48	0.00	93.44	0.08	0.34
12/16/15 10:06:51	3499	0.07	0.59	5.46	0.00	93.46	0.08	0.34

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	Ar	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:07:23	3531	0.07	0.59	5.45	0.00	93.48	0.08	0.34
12/16/15 10:07:55	3563	0.07	0.59	5.44	0.00	93.52	0.08	0.31
12/16/15 10:08:27	3595	0.06	0.58	5.42	0.00	93.53	0.08	0.33
12/16/15 10:08:59	3627	0.07	0.58	5.41	0.00	93.57	0.08	0.30
12/16/15 10:09:31	3659	0.07	0.58	5.38	0.00	93.58	0.07	0.32
12/16/15 10:10:03	3691	0.06	0.58	5.37	0.00	93.61	0.07	0.30
12/16/15 10:10:35	3723	0.06	0.57	5.36	0	93.63	0.07	0.31
12/16/15 10:11:09	3757	0.06	0.57	5.34	0	93.64	0.07	0.32
12/16/15 10:11:41	3789	0.06	0.57	5.32	0	93.66	0.08	0.31
12/16/15 10:12:14	3822	0.06	0.57	5.30	0	93.69	0.07	0.30
12/16/15 10:12:46	3854	0.07	0.55	5.29	0	93.71	0.07	0.31
12/16/15 10:13:18	3886	0.07	0.57	5.27	0	93.73	0.07	0.30
12/16/15 10:13:50	3918	0.07	0.57	5.24	0	93.73	0.07	0.32
12/16/15 10:14:22	3950	0.07	0.57	5.23	0	93.78	0.07	0.30
12/16/15 10:14:54	3982	0.06	0.56	5.22	0	93.81	0.07	0.29
12/16/15 10:15:26	4014	0.07	0.56	5.19	0	93.81	0.07	0.30
12/16/15 10:15:59	4047	0.07	0.56	5.18	0	93.83	0.07	0.30
12/16/15 10:16:31	4079	0.07	0.55	5.16	0	93.86	0.07	0.29
12/16/15 10:17:04	4112	0.07	0.55	5.14	0	93.86	0.07	0.31
12/16/15 10:17:36	4144	0.07	0.55	5.13	0	93.88	0.07	0.31
12/16/15 10:18:08	4176	0.06	0.25	5.14	0	94.26	0.06	0.23
12/16/15 10:18:40	4208	0.07	0.14	3.88	0	95.65	0.05	0.22
12/16/15 10:19:12	4240	0.06	0.09	2.62	0	97.01	0.04	0.18
12/16/15 10:19:44	4272	0.07	0.06	1.75	0	97.93	0.03	0.17
12/16/15 10:20:20	4308	0.07	0.05	1.09	0	98.62	0.03	0.15
12/16/15 10:20:54	4342	0.07	0.04	0.80	0	98.92	0.03	0.14
12/16/15 10:21:29	4377	0.07	0.04	0.65	0	99.11	0.03	0.11
12/16/15 10:22:03	4411	0.07	0.04	0.56	0	99.18	0.03	0.13
12/16/15 10:22:36	4444	0.07	0.04	0.51	0	99.27	0.02	0.10
12/16/15 10:23:10	4478	0.07	0.04	0.47	0	99.30	0.03	0.10
12/16/15 10:23:44	4512	0.07	0.04	0.45	0	99.34	0.02	0.09
12/16/15 10:24:18	4546	0.06	0.04	0.43	0	99.35	0.02	0.09
12/16/15 10:24:51	4579	0.07	0.04	0.42	0	99.37	0.02	0.09
12/16/15 10:25:25	4613	0.06	0.04	0.40	0	99.38	0.02	0.10
12/16/15 10:25:58	4646	0.07	0.04	0.39	0	99.37	0.03	0.11
12/16/15 10:26:32	4680	0.06	0.03	0.39	0	99.41	0.02	0.08

Table D-2. Re-Baselined Monitor MS Offgas Concentration Data for Room Temp Carbon Steel Dissolution

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	Ar	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:08:32	0	n/a	n/a	n/a	n/a	n/a	n/a	n/a
12/16/15 9:08:51	19	0.00	0.00	0.00	0.00	100.00	0.00	0.01
12/16/15 9:09:25	53	0.00	0.00	0.00	0.00	100.00	0.00	0.00
12/16/15 9:09:58	86	0.00	1.31	0.00	0.00	98.68	0.00	0.00
12/16/15 9:10:32	120	0.00	0.33	0.00	0.00	99.66	0.00	0.01
12/16/15 9:11:05	153	0.00	0.27	0.00	0.00	99.73	0.00	0.00
12/16/15 9:11:39	187	0.00	0.43	0.01	0.00	99.56	0.00	0.00
12/16/15 9:12:13	221	0.00	0.52	0.07	0.00	99.40	0.01	0.00
12/16/15 9:12:47	255	0.00	0.57	0.19	0.00	99.21	0.01	0.01
12/16/15 9:13:20	288	0.00	0.60	0.37	0.00	98.98	0.02	0.02
12/16/15 9:13:54	322	0.00	0.63	0.59	0.00	98.72	0.02	0.03
12/16/15 9:14:27	355	0.00	0.65	0.83	0.00	98.41	0.03	0.07
12/16/15 9:15:00	388	0.00	0.66	1.07	0.00	98.13	0.04	0.08
12/16/15 9:15:34	422	0.00	0.68	1.30	0.00	97.87	0.05	0.10
12/16/15 9:16:08	456	0.00	0.69	1.52	0.00	97.61	0.05	0.12
12/16/15 9:16:41	489	0.00	0.69	1.73	0.00	97.38	0.05	0.13
12/16/15 9:17:15	523	0.00	0.70	1.94	0.00	97.16	0.06	0.14
12/16/15 9:17:48	556	0.00	0.69	2.13	0.00	96.95	0.07	0.15
12/16/15 9:18:22	590	0.00	0.70	2.32	0.00	96.72	0.07	0.18
12/16/15 9:18:55	623	0.00	0.70	2.49	0.00	96.54	0.07	0.19
12/16/15 9:19:29	657	0.00	0.70	2.66	0.00	96.35	0.08	0.20
12/16/15 9:20:02	690	0.00	0.70	2.82	0.00	96.19	0.08	0.20
12/16/15 9:20:36	724	0.00	0.70	2.98	0.00	95.98	0.08	0.25
12/16/15 9:21:09	757	0.00	0.70	3.13	0.00	95.83	0.08	0.25
12/16/15 9:21:43	791	0.00	0.70	3.28	0.00	95.66	0.09	0.27
12/16/15 9:22:16	824	0.00	0.70	3.41	0.00	95.51	0.09	0.28
12/16/15 9:22:50	858	0.00	0.70	3.55	0.00	95.37	0.09	0.28
12/16/15 9:23:23	891	0.00	0.70	3.67	0.00	95.23	0.09	0.30
12/16/15 9:23:59	927	0.00	0.66	3.94	0.00	94.96	0.10	0.33
12/16/15 9:24:31	959	0.00	0.69	4.02	0.00	94.82	0.10	0.35
12/16/15 9:25:03	991	0.00	0.69	4.13	0.00	94.71	0.10	0.36
12/16/15 9:25:36	1024	0.00	0.70	4.23	0.00	94.57	0.10	0.40
12/16/15 9:26:08	1056	0.00	0.70	4.32	0.00	94.47	0.10	0.39
12/16/15 9:26:40	1088	0.00	0.70	4.41	0.00	94.37	0.10	0.41
12/16/15 9:27:12	1120	0.00	0.69	4.50	0.00	94.26	0.10	0.43

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	Ar	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:27:44	1152	0.00	0.68	4.60	0.00	94.19	0.10	0.42
12/16/15 9:28:17	1185	0.00	0.68	4.67	0.00	94.09	0.11	0.44
12/16/15 9:28:49	1217	0.00	0.67	4.75	0.00	93.99	0.11	0.46
12/16/15 9:29:21	1249	0.00	0.67	4.83	0.00	93.95	0.10	0.43
12/16/15 9:29:53	1281	0.00	0.67	4.89	0.00	93.87	0.10	0.45
12/16/15 9:30:25	1313	0.00	0.64	4.98	0.00	93.81	0.11	0.46
12/16/15 9:30:58	1346	0.00	0.66	5.03	0.00	93.75	0.10	0.44
12/16/15 9:31:30	1378	0.00	0.67	5.08	0.00	93.67	0.11	0.46
12/16/15 9:32:02	1410	0.00	0.66	5.14	0.00	93.62	0.10	0.46
12/16/15 9:32:34	1442	0.00	0.65	5.19	0.00	93.57	0.10	0.47
12/16/15 9:33:06	1474	0.00	0.65	5.25	0.00	93.52	0.10	0.47
12/16/15 9:33:38	1506	0.00	0.64	5.29	0.00	93.48	0.10	0.46
12/16/15 9:34:10	1538	0.00	0.64	5.34	0.00	93.45	0.10	0.45
12/16/15 9:34:42	1570	0.00	0.63	5.38	0.00	93.40	0.10	0.46
12/16/15 9:35:14	1602	0.00	0.63	5.43	0.00	93.37	0.10	0.45
12/16/15 9:35:46	1634	0.00	0.63	5.46	0.00	93.32	0.10	0.47
12/16/15 9:36:19	1667	0.00	0.62	5.50	0.00	93.29	0.10	0.47
12/16/15 9:36:51	1699	0.00	0.62	5.53	0.00	93.30	0.10	0.43
12/16/15 9:37:23	1731	0.00	0.61	5.57	0.00	93.28	0.10	0.43
12/16/15 9:37:55	1763	0.00	0.61	5.59	0.00	93.24	0.10	0.45
12/16/15 9:38:27	1795	0.00	0.60	5.62	0.00	93.22	0.10	0.43
12/16/15 9:38:59	1827	0.00	0.60	5.65	0.00	93.19	0.10	0.44
12/16/15 9:39:31	1859	0.00	0.59	5.66	0.00	93.18	0.10	0.44
12/16/15 9:40:03	1891	0.00	0.59	5.69	0.00	93.17	0.10	0.43
12/16/15 9:40:35	1923	0.00	0.59	5.71	0.00	93.16	0.10	0.43
12/16/15 9:41:07	1955	0.00	0.58	5.72	0.00	93.16	0.10	0.43
12/16/15 9:41:39	1987	0.00	0.58	5.74	0.00	93.16	0.10	0.41
12/16/15 9:42:11	2019	0.00	0.58	5.75	0.00	93.13	0.10	0.42
12/16/15 9:42:44	2052	0.00	0.57	5.77	0.00	93.14	0.09	0.41
12/16/15 9:43:16	2084	0.00	0.57	5.78	0.00	93.15	0.09	0.40
12/16/15 9:43:48	2116	0.00	0.56	5.79	0.00	93.13	0.10	0.41
12/16/15 9:44:20	2148	0.00	0.56	5.79	0.00	93.13	0.09	0.41
12/16/15 9:44:52	2180	0.00	0.55	5.81	0.00	93.14	0.09	0.39
12/16/15 9:45:24	2212	0.00	0.55	5.81	0.00	93.14	0.09	0.40
12/16/15 9:45:56	2244	0.00	0.55	5.81	0.00	93.14	0.09	0.40
12/16/15 9:46:29	2277	0.00	0.54	5.81	0.00	93.12	0.09	0.41
12/16/15 9:47:01	2309	0.00	0.54	5.82	0.00	93.14	0.09	0.40
12/16/15 9:47:33	2341	0.00	0.53	5.82	0.00	93.13	0.09	0.41

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	Ar	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:48:06	2374	0.00	0.53	5.83	0.00	93.15	0.09	0.40
12/16/15 9:48:38	2406	0.00	0.53	5.82	0.00	93.15	0.09	0.39
12/16/15 9:49:10	2438	0.00	0.52	5.82	0.00	93.16	0.08	0.39
12/16/15 9:49:42	2470	0.00	0.52	5.82	0.00	93.17	0.08	0.39
12/16/15 9:50:14	2502	0.00	0.51	5.82	0.00	93.18	0.08	0.39
12/16/15 9:50:46	2534	0.00	0.51	5.82	0.00	93.19	0.08	0.38
12/16/15 9:51:18	2566	0.00	0.50	5.82	0.00	93.20	0.08	0.38
12/16/15 9:51:50	2598	0.00	0.50	5.80	0.00	93.20	0.08	0.39
12/16/15 9:52:23	2631	0.00	0.50	5.80	0.00	93.23	0.08	0.37
12/16/15 9:52:55	2663	0.00	0.49	5.80	0.00	93.24	0.08	0.36
12/16/15 9:53:27	2695	0.00	0.49	5.79	0.00	93.24	0.08	0.38
12/16/15 9:53:59	2727	0.00	0.49	5.79	0.00	93.28	0.08	0.35
12/16/15 9:54:31	2759	0.00	0.48	5.78	0.00	93.30	0.07	0.35
12/16/15 9:55:03	2791	0.00	0.48	5.77	0.00	93.31	0.07	0.35
12/16/15 9:55:35	2823	0.00	0.48	5.76	0.00	93.32	0.07	0.34
12/16/15 9:56:08	2856	0.00	0.48	5.75	0.00	93.31	0.07	0.37
12/16/15 9:56:40	2888	0.00	0.47	5.74	0.00	93.34	0.07	0.36
12/16/15 9:57:12	2920	0.00	0.47	5.73	0.00	93.37	0.07	0.34
12/16/15 9:57:44	2952	0.00	0.47	5.72	0.00	93.39	0.07	0.34
12/16/15 9:58:16	2984	0.00	0.46	5.70	0.00	93.39	0.07	0.35
12/16/15 9:58:48	3016	0.00	0.46	5.69	0.00	93.42	0.07	0.34
12/16/15 9:59:20	3048	0.00	0.46	5.68	0.00	93.43	0.07	0.35
12/16/15 9:59:52	3080	0.00	0.45	5.66	0.00	93.45	0.07	0.34
12/16/15 10:00:25	3113	0.00	0.45	5.66	0.00	93.48	0.06	0.33
12/16/15 10:00:57	3145	0.00	0.45	5.64	0.00	93.50	0.06	0.33
12/16/15 10:01:29	3177	0.00	0.45	5.63	0.00	93.52	0.07	0.32
12/16/15 10:02:01	3209	0.00	0.45	5.61	0.00	93.55	0.06	0.32
12/16/15 10:02:33	3241	0.00	0.45	5.59	0.00	93.53	0.07	0.35
12/16/15 10:03:05	3273	0.00	0.44	5.58	0.00	93.58	0.06	0.32
12/16/15 10:03:37	3305	0.00	0.44	5.57	0.00	93.60	0.06	0.32
12/16/15 10:04:10	3338	0.00	0.43	5.55	0.00	93.61	0.06	0.33
12/16/15 10:04:42	3370	0.00	0.43	5.54	0.00	93.64	0.06	0.32
12/16/15 10:05:14	3402	0.00	0.43	5.51	0.00	93.64	0.06	0.34
12/16/15 10:05:46	3434	0.00	0.43	5.50	0.00	93.66	0.06	0.33
12/16/15 10:06:18	3466	0.00	0.42	5.48	0.00	93.69	0.06	0.33
12/16/15 10:06:51	3499	0.00	0.42	5.46	0.00	93.71	0.06	0.33
12/16/15 10:07:23	3531	0.00	0.42	5.45	0.00	93.73	0.06	0.33
12/16/15 10:07:55	3563	0.00	0.42	5.44	0.00	93.77	0.06	0.30

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	Ar	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:08:27	3595	0.00	0.41	5.42	0.00	93.78	0.06	0.32
12/16/15 10:08:59	3627	0.00	0.41	5.41	0.00	93.82	0.05	0.30
12/16/15 10:09:31	3659	0.00	0.41	5.38	0.00	93.83	0.05	0.31
12/16/15 10:10:03	3691	0.00	0.41	5.37	0.00	93.86	0.05	0.29
12/16/15 10:10:35	3723	0.00	0.40	5.36	0.00	93.88	0.05	0.30
12/16/15 10:11:09	3757	0.00	0.40	5.34	0.00	93.89	0.05	0.31
12/16/15 10:11:41	3789	0.00	0.40	5.32	0.00	93.91	0.05	0.30
12/16/15 10:12:14	3822	0.00	0.40	5.30	0.00	93.94	0.05	0.30
12/16/15 10:12:46	3854	0.00	0.38	5.29	0.00	93.96	0.05	0.31
12/16/15 10:13:18	3886	0.00	0.40	5.27	0.00	93.98	0.05	0.29
12/16/15 10:13:50	3918	0.00	0.40	5.24	0.00	93.98	0.05	0.31
12/16/15 10:14:22	3950	0.00	0.39	5.23	0.00	94.03	0.05	0.29
12/16/15 10:14:54	3982	0.00	0.39	5.22	0.00	94.06	0.05	0.28
12/16/15 10:15:26	4014	0.00	0.39	5.19	0.00	94.06	0.05	0.29
12/16/15 10:15:59	4047	0.00	0.38	5.18	0.00	94.08	0.05	0.29
12/16/15 10:16:31	4079	0.00	0.38	5.16	0.00	94.11	0.05	0.28
12/16/15 10:17:04	4112	0.00	0.38	5.14	0.00	94.11	0.05	0.30
12/16/15 10:17:36	4144	0.00	0.38	5.13	0.00	94.13	0.05	0.30
12/16/15 10:18:08	4176	0.00	0.08	5.14	0.00	94.51	0.04	0.22
12/16/15 10:18:40	4208	0.00	0.00	3.88	0.00	95.91	0.03	0.21
12/16/15 10:19:12	4240	0.00	0.00	2.62	0.00	97.27	0.01	0.17
12/16/15 10:19:44	4272	0.00	0.00	1.75	0.00	98.19	0.01	0.16
12/16/15 10:20:20	4308	0.00	0.00	1.09	0.00	98.88	0.01	0.14
12/16/15 10:20:54	4342	0.00	0.00	0.80	0.00	99.18	0.00	0.13
12/16/15 10:21:29	4377	0.00	0.00	0.65	0.00	99.38	0.00	0.10
12/16/15 10:22:03	4411	0.00	0.00	0.56	0.00	99.44	0.01	0.12
12/16/15 10:22:36	4444	0.00	0.00	0.51	0.00	99.54	0.00	0.09
12/16/15 10:23:10	4478	0.00	0.00	0.47	0.00	99.57	0.00	0.10
12/16/15 10:23:44	4512	0.00	0.00	0.45	0.00	99.60	0.00	0.09
12/16/15 10:24:18	4546	0.00	0.00	0.43	0.00	99.62	0.00	0.09
12/16/15 10:24:51	4579	0.00	0.00	0.42	0.00	99.63	0.00	0.08
12/16/15 10:25:25	4613	0.00	0.00	0.40	0.00	99.64	0.00	0.09
12/16/15 10:25:58	4646	0.00	0.00	0.39	0.00	99.63	0.00	0.10
12/16/15 10:26:32	4680	0.00	0.00	0.39	0.00	99.68	0.00	0.08

Appendix E. Raman Offgas Data for Carbon Steel

Table E-1. Raw Raman Offgas Concentration Data for Room Temp Carbon Steel Dissolution

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:08:32	0	n/a	n/a	n/a	n/a	n/a	n/a
12/16/15 9:08:37	5	-0.24	2.05	-0.19	0.37	0.22	0.01
12/16/15 9:08:49	17	0.45	1.11	0.49	0.89	0.12	0.01
12/16/15 9:09:00	28	-0.05	1.06	0.28	0.67	0.23	0.02
12/16/15 9:09:12	40	-0.30	0.86	-0.50	0.65	-0.02	0.01
12/16/15 9:09:24	52	-0.10	0.63	-1.29	0.15	0.00	0.03
12/16/15 9:09:36	64	-0.13	0.66	-0.86	-0.04	0.22	0.02
12/16/15 9:09:48	76	-0.08	1.00	-1.28	0.48	0.04	0.02
12/16/15 9:10:00	88	-0.21	-0.31	0.34	0.17	-0.28	0.02
12/16/15 9:10:12	100	-0.11	0.42	0.02	-0.38	-0.69	0.02
12/16/15 9:10:24	112	0.25	0.12	0.30	0.30	0.34	0.01
12/16/15 9:10:35	123	0.13	0.95	-0.04	-0.26	0.05	0.01
12/16/15 9:10:47	135	0.24	0.48	1.38	0.29	0.12	0.00
12/16/15 9:10:59	147	0.05	1.42	-0.66	0.61	0.01	0.01
12/16/15 9:11:11	159	0.09	1.24	1.75	0.41	-0.40	0.02
12/16/15 9:11:23	171	-0.24	0.69	0.50	0.59	-0.35	0.03
12/16/15 9:11:35	183	0.02	1.44	1.51	0.21	0.13	0.03
12/16/15 9:11:47	195	0.13	1.06	0.57	0.77	0.00	0.03
12/16/15 9:11:59	207	-0.01	1.09	0.12	0.20	0.06	0.01
12/16/15 9:12:11	219	0.01	1.27	-1.26	0.29	0.18	0.01
12/16/15 9:12:23	231	-0.19	1.22	-1.72	0.71	0.24	0.02
12/16/15 9:12:35	243	-0.04	0.85	-0.95	0.28	-0.26	0.03
12/16/15 9:12:46	254	-0.40	1.34	0.13	0.61	-0.18	0.02
12/16/15 9:12:58	266	0.13	1.84	0.79	0.15	0.19	0.03
12/16/15 9:13:10	278	-0.01	0.80	0.22	0.43	0.14	0.02
12/16/15 9:13:22	290	-0.04	1.22	-0.52	0.19	0.10	0.04
12/16/15 9:13:34	302	0.27	1.74	0.84	0.74	0.06	0.05
12/16/15 9:13:46	314	0.17	1.05	0.71	0.11	0.13	0.05
12/16/15 9:13:58	326	-0.05	1.86	1.15	0.67	-0.16	0.07
12/16/15 9:14:10	338	-0.05	1.36	-0.44	0.30	0.18	0.08
12/16/15 9:14:22	350	-0.09	1.49	0.83	0.42	0.26	0.09
12/16/15 9:14:34	362	-0.04	1.14	-0.69	0.51	-0.11	0.09
12/16/15 9:14:46	374	-0.20	1.47	1.94	0.32	0.32	0.11

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:14:58	386	0.20	1.11	0.39	0.18	0.33	0.12
12/16/15 9:15:10	398	-0.21	1.23	0.78	0.37	-0.48	0.14
12/16/15 9:15:22	410	-0.15	1.59	0.12	0.27	0.44	0.15
12/16/15 9:15:34	422	-0.32	1.14	1.66	0.66	0.10	0.16
12/16/15 9:15:46	434	-0.14	1.11	-0.31	0.65	0.19	0.18
12/16/15 9:15:58	446	-0.23	1.44	1.53	0.55	0.14	0.19
12/16/15 9:16:10	458	0.18	1.58	1.46	0.09	-0.06	0.19
12/16/15 9:16:22	470	-0.12	1.20	0.30	-0.17	0.17	0.22
12/16/15 9:16:34	482	-0.11	1.59	-0.77	0.64	0.04	0.24
12/16/15 9:16:46	494	-0.37	1.71	1.03	0.17	0.03	0.25
12/16/15 9:16:58	506	-0.18	1.06	2.02	-0.36	0.09	0.27
12/16/15 9:17:10	518	-0.26	1.86	1.26	-0.16	-0.17	0.28
12/16/15 9:17:23	531	0.17	1.86	0.47	0.70	0.18	0.28
12/16/15 9:17:35	543	0.04	0.41	1.53	0.32	0.73	0.23
12/16/15 9:17:47	555	0.52	1.58	0.97	0.35	0.27	0.32
12/16/15 9:17:59	567	-0.01	1.18	0.56	0.37	0.36	0.35
12/16/15 9:18:11	579	0.02	2.75	0.84	0.53	-0.19	0.34
12/16/15 9:18:23	591	0.47	1.59	1.81	0.90	0.39	0.37
12/16/15 9:18:35	603	0.36	-0.17	2.50	0.72	-0.07	0.39
12/16/15 9:18:47	615	-0.18	1.43	0.63	0.96	-0.10	0.38
12/16/15 9:18:59	627	-0.21	1.06	1.11	0.54	0.27	0.41
12/16/15 9:19:11	639	0.47	1.12	0.28	-0.69	0.05	0.40
12/16/15 9:19:23	651	-0.01	1.45	1.06	-0.01	0.25	0.39
12/16/15 9:19:35	663	0.45	0.80	2.70	0.08	-0.08	0.42
12/16/15 9:19:48	676	-0.11	1.61	1.29	0.51	0.47	0.44
12/16/15 9:20:00	688	0.40	0.64	2.42	0.34	0.47	0.45
12/16/15 9:20:12	700	0.19	2.11	1.56	-0.06	0.21	0.45
12/16/15 9:20:24	712	-0.04	-0.39	1.32	0.45	-0.37	0.47
12/16/15 9:20:36	724	0.34	1.74	-0.32	0.81	0.02	0.47
12/16/15 9:20:48	736	0.29	0.99	1.70	0.12	-0.27	0.48
12/16/15 9:21:00	748	0.73	0.42	3.27	1.03	0.02	0.47
12/16/15 9:21:13	761	0.44	1.61	1.56	0.73	0.42	0.47
12/16/15 9:21:25	773	0.85	1.24	0.19	0.02	-0.28	0.46
12/16/15 9:21:37	785	-0.05	1.19	2.97	0.14	0.01	0.49
12/16/15 9:21:49	797	-0.42	1.16	2.95	-0.02	0.33	0.48
12/16/15 9:22:01	809	-0.16	1.42	2.87	0.45	0.12	0.51
12/16/15 9:22:13	821	-0.10	2.20	0.81	-0.04	0.41	0.51
12/16/15 9:22:26	834	-0.30	1.08	4.74	1.22	-0.45	0.48

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:22:38	846	-0.17	1.69	2.02	-0.22	0.11	0.51
12/16/15 9:22:50	858	0.13	1.43	3.25	1.20	0.03	0.51
12/16/15 9:23:02	870	-0.14	0.44	3.13	1.71	0.21	0.53
12/16/15 9:23:14	882	-0.13	1.88	0.58	0.12	-0.35	0.53
12/16/15 9:23:27	895	0.30	-0.03	3.71	1.08	-0.13	0.52
12/16/15 9:23:39	907	-0.11	1.70	1.64	1.20	-0.21	0.53
12/16/15 9:23:51	919	-0.02	0.77	2.22	0.98	-0.01	0.52
12/16/15 9:24:03	931	0.04	1.27	5.31	1.05	-0.31	0.51
12/16/15 9:24:16	944	0.26	1.76	-0.31	0.84	0.42	0.47
12/16/15 9:24:28	956	0.24	0.06	3.30	1.56	0.08	0.51
12/16/15 9:24:40	968	0.23	0.90	2.96	-0.10	-0.78	0.52
12/16/15 9:24:52	980	0.43	1.65	2.60	1.20	0.04	0.52
12/16/15 9:25:04	992	0.02	1.05	2.64	0.77	0.23	0.53
12/16/15 9:25:17	1005	0.67	2.67	1.51	1.73	0.60	0.52
12/16/15 9:25:29	1017	0.39	0.91	1.67	0.13	0.07	0.51
12/16/15 9:25:41	1029	0.19	1.09	0.81	1.18	0.42	0.52
12/16/15 9:25:54	1042	0.53	0.39	4.24	0.84	0.51	0.53
12/16/15 9:26:06	1054	0.18	0.86	1.90	0.66	-0.20	0.53
12/16/15 9:26:18	1066	0.02	0.42	5.50	0.12	0.46	0.53
12/16/15 9:26:30	1078	0.14	1.55	3.85	-0.01	0.11	0.53
12/16/15 9:26:43	1091	0.06	0.19	1.91	1.02	0.27	0.53
12/16/15 9:26:55	1103	-0.15	1.37	1.15	-0.31	-0.45	0.51
12/16/15 9:27:07	1115	0.59	1.42	0.15	0.51	0.11	0.52
12/16/15 9:27:20	1128	0.23	1.19	2.03	0.54	-0.43	0.52
12/16/15 9:27:32	1140	0.26	1.80	2.15	-0.10	-0.21	0.51
12/16/15 9:27:44	1152	0.02	0.67	5.26	0.81	0.11	0.53
12/16/15 9:27:57	1165	0.34	1.60	4.20	0.27	0.17	0.50
12/16/15 9:28:09	1177	0.33	0.24	4.41	0.27	-0.01	0.53
12/16/15 9:28:21	1189	0.25	1.04	4.07	0.50	0.09	0.53
12/16/15 9:28:34	1202	0.08	1.95	1.99	0.43	-0.09	0.51
12/16/15 9:28:46	1214	0.12	0.85	0.36	-0.33	0.10	0.49
12/16/15 9:28:58	1226	0.07	1.39	0.89	-0.30	-0.68	0.51
12/16/15 9:29:11	1239	0.33	1.31	4.65	0.26	0.20	0.50
12/16/15 9:29:23	1251	-0.21	1.37	5.53	1.08	0.87	0.51
12/16/15 9:29:35	1263	0.61	0.67	3.64	0.02	0.30	0.49
12/16/15 9:29:48	1276	0.09	1.20	4.82	-0.16	0.29	0.50
12/16/15 9:30:00	1288	0.41	0.93	3.79	-0.20	0.35	0.50
12/16/15 9:30:12	1300	-0.43	1.65	3.66	0.65	0.31	0.49

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:30:25	1313	0.11	1.68	3.67	0.58	0.14	0.49
12/16/15 9:30:37	1325	-0.70	0.36	0.27	0.15	-0.24	0.50
12/16/15 9:30:50	1338	0.60	1.72	1.79	0.86	-0.51	0.49
12/16/15 9:31:02	1350	-0.48	0.42	2.91	0.15	0.45	0.49
12/16/15 9:31:14	1362	0.23	-0.21	4.38	0.16	0.61	0.50
12/16/15 9:31:27	1375	0.29	0.74	1.29	-0.15	-0.05	0.51
12/16/15 9:31:39	1387	-0.23	0.65	5.91	0.99	0.22	0.52
12/16/15 9:31:52	1400	-0.15	0.82	4.60	0.42	0.07	0.49
12/16/15 9:32:04	1412	-0.24	1.86	5.37	-0.38	-0.71	0.49
12/16/15 9:32:17	1425	-0.12	0.69	2.14	0.77	-0.16	0.48
12/16/15 9:32:29	1437	-0.09	0.73	1.78	0.43	0.52	0.48
12/16/15 9:32:41	1449	0.14	0.42	2.68	1.95	0.26	0.50
12/16/15 9:32:54	1462	0.32	1.38	1.14	0.46	0.11	0.50
12/16/15 9:33:06	1474	0.33	1.44	-3.51	-0.11	0.03	0.48
12/16/15 9:33:19	1487	-0.14	1.04	-0.06	-0.32	0.04	0.48
12/16/15 9:33:31	1499	0.14	1.26	8.75	0.22	0.06	0.47
12/16/15 9:33:44	1512	0.32	1.26	2.49	0.16	0.66	0.48
12/16/15 9:33:56	1524	-0.02	0.65	4.75	-0.42	0.38	0.47
12/16/15 9:34:09	1537	0.10	0.37	4.71	1.06	-0.15	0.48
12/16/15 9:34:21	1549	-0.18	1.31	4.98	0.59	-0.05	0.48
12/16/15 9:34:33	1561	-0.13	0.60	3.64	0.68	0.14	0.49
12/16/15 9:34:46	1574	-0.08	2.44	4.88	1.20	-0.14	0.48
12/16/15 9:34:58	1586	-0.23	1.47	2.49	0.27	-0.23	0.47
12/16/15 9:35:11	1599	0.11	2.14	7.19	0.36	-0.13	0.47
12/16/15 9:35:23	1611	0.20	0.36	3.79	0.96	-0.05	0.48
12/16/15 9:35:36	1624	0.46	1.81	5.21	0.57	0.51	0.46
12/16/15 9:35:49	1637	0.34	0.60	7.37	0.76	-0.16	0.47
12/16/15 9:36:01	1649	-0.11	1.11	4.42	0.22	0.16	0.48
12/16/15 9:36:14	1662	0.23	1.31	7.48	1.30	-0.16	0.47
12/16/15 9:36:26	1674	-0.20	1.27	4.59	0.71	0.17	0.47
12/16/15 9:36:39	1687	0.28	1.27	5.13	0.42	-0.72	0.48
12/16/15 9:36:51	1699	0.30	1.02	2.83	0.37	0.29	0.45
12/16/15 9:37:04	1712	-0.26	0.64	5.63	-0.68	0.29	0.46
12/16/15 9:37:16	1724	0.34	0.91	2.84	0.14	-0.08	0.47
12/16/15 9:37:29	1737	0.08	0.04	2.88	1.74	0.12	0.47
12/16/15 9:37:41	1749	0.38	0.01	1.00	0.74	0.13	0.47
12/16/15 9:37:54	1762	0.56	0.95	3.69	0.54	0.61	0.45
12/16/15 9:38:06	1774	0.40	0.40	5.38	0.91	0.36	0.46

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:38:19	1787	0.07	1.30	5.37	-0.05	-0.34	0.45
12/16/15 9:44:10	2138	-0.19	1.14	4.49	-0.21	-0.08	0.37
12/16/15 9:44:21	2149	0.02	1.47	1.96	0.77	-0.13	0.41
12/16/15 9:44:33	2161	-0.03	2.10	5.56	0.16	-0.28	0.40
12/16/15 9:44:44	2172	0.23	0.89	4.71	1.21	-0.29	0.41
12/16/15 9:44:55	2183	-0.02	1.85	5.67	-0.39	-0.21	0.38
12/16/15 9:45:07	2195	0.12	0.75	5.67	0.91	0.38	0.40
12/16/15 9:45:18	2206	-0.27	1.52	3.01	-0.31	0.02	0.39
12/16/15 9:45:29	2217	0.00	-0.09	7.90	-0.26	0.02	0.40
12/16/15 9:45:41	2229	0.01	0.65	5.56	-0.50	0.26	0.41
12/16/15 9:45:52	2240	0.14	0.45	3.61	0.77	-0.09	0.40
12/16/15 9:46:03	2251	0.12	1.93	6.08	0.45	-0.05	0.40
12/16/15 9:46:15	2263	0.56	1.31	7.50	0.31	-0.27	0.39
12/16/15 9:46:26	2274	-0.32	1.37	4.85	-0.10	-0.23	0.39
12/16/15 9:46:38	2286	0.05	0.89	5.27	0.92	0.39	0.40
12/16/15 9:46:49	2297	-0.08	1.08	4.85	0.36	-0.14	0.40
12/16/15 9:47:00	2308	-0.08	1.76	3.94	0.14	0.18	0.41
12/16/15 9:47:12	2320	0.61	0.57	4.62	-0.28	0.60	0.40
12/16/15 9:47:23	2331	0.54	0.88	4.32	1.26	0.06	0.40
12/16/15 9:47:35	2343	-0.35	1.61	4.77	0.39	0.87	0.41
12/16/15 9:47:46	2354	-0.20	0.95	5.25	0.25	0.29	0.39
12/16/15 9:47:57	2365	0.42	0.76	4.88	0.85	0.47	0.38
12/16/15 9:48:09	2377	-0.01	-0.19	4.56	-0.21	0.19	0.39
12/16/15 9:48:20	2388	0.11	0.36	5.99	-0.30	0.16	0.40
12/16/15 9:48:32	2400	-0.12	0.84	6.16	-0.54	0.14	0.37
12/16/15 9:48:43	2411	0.03	0.73	3.69	0.32	0.02	0.39
12/16/15 9:48:55	2423	-0.01	0.63	4.13	-0.12	0.31	0.38
12/16/15 9:49:06	2434	-0.06	0.84	6.74	0.45	0.42	0.37
12/16/15 9:49:18	2446	0.02	0.54	5.55	-0.30	0.57	0.38
12/16/15 9:49:29	2457	0.27	0.57	6.70	-0.28	0.31	0.37
12/16/15 9:49:41	2469	-0.10	0.11	4.45	-0.18	0.23	0.38
12/16/15 9:49:52	2480	0.24	1.00	6.03	-0.60	0.33	0.37
12/16/15 9:50:04	2492	0.24	1.49	7.24	0.16	-0.23	0.35
12/16/15 9:50:15	2503	-0.05	1.05	5.01	0.79	0.15	0.37
12/16/15 9:50:27	2515	-0.15	1.53	5.85	0.43	0.17	0.38
12/16/15 9:50:38	2526	-0.30	0.91	5.15	0.65	0.15	0.37
12/16/15 9:50:50	2538	-0.28	1.50	4.82	0.81	-0.16	0.38
12/16/15 9:51:01	2549	-0.12	0.78	2.55	0.52	0.44	0.38

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:51:13	2561	0.22	0.96	4.75	0.51	0.52	0.37
12/16/15 9:51:24	2572	0.44	0.86	3.24	0.47	0.10	0.36
12/16/15 9:51:36	2584	-0.08	0.47	6.12	-0.31	0.12	0.34
12/16/15 9:51:48	2596	-0.15	0.88	3.17	0.64	0.25	0.36
12/16/15 9:51:59	2607	-0.49	0.11	3.80	0.72	-0.15	0.38
12/16/15 9:52:11	2619	0.50	-0.10	5.38	0.25	-0.15	0.37
12/16/15 9:52:22	2630	0.24	0.09	4.12	0.09	0.00	0.36
12/16/15 9:52:34	2642	0.16	0.79	4.46	0.62	-0.71	0.37
12/16/15 9:52:45	2653	0.00	1.20	4.14	-0.08	0.13	0.37
12/16/15 9:52:57	2665	-0.17	2.01	7.65	0.49	-0.21	0.36
12/16/15 9:53:09	2677	0.12	-0.13	5.94	-0.39	-0.26	0.37
12/16/15 9:53:20	2688	0.26	-0.23	5.83	1.15	0.03	0.36
12/16/15 9:53:32	2700	0.52	0.74	4.28	0.07	0.37	0.37
12/16/15 9:53:43	2711	0.15	0.39	6.89	0.50	-0.39	0.36
12/16/15 9:53:55	2723	0.25	0.49	7.49	0.73	-0.20	0.36
12/16/15 9:54:06	2734	0.06	-0.60	5.42	0.73	0.25	0.36
12/16/15 9:54:18	2746	0.11	0.54	5.42	0.31	0.00	0.35
12/16/15 9:54:30	2758	-0.07	1.01	5.84	0.15	0.15	0.36
12/16/15 9:54:41	2769	0.21	0.70	5.59	0.45	-0.19	0.35
12/16/15 9:54:53	2781	0.34	1.70	6.94	0.62	0.08	0.36
12/16/15 9:55:04	2792	0.04	0.26	3.09	-0.92	-0.15	0.34
12/16/15 9:55:16	2804	0.05	1.39	4.84	0.05	0.53	0.35
12/16/15 9:55:28	2816	0.02	1.28	5.47	0.65	0.13	0.35
12/16/15 9:55:39	2827	0.40	0.84	5.73	0.21	0.10	0.34
12/16/15 9:55:51	2839	0.44	1.15	2.93	0.81	0.23	0.34
12/16/15 9:56:03	2851	-0.17	0.08	4.33	0.00	0.47	0.35
12/16/15 9:56:14	2862	0.05	0.74	4.66	0.71	0.07	0.35
12/16/15 9:56:26	2874	-0.40	1.12	5.87	-0.21	0.47	0.36
12/16/15 9:56:38	2886	0.15	0.96	6.00	0.04	0.32	0.36
12/16/15 9:56:49	2897	0.60	1.83	5.74	0.82	0.20	0.34
12/16/15 9:57:01	2909	-0.05	1.32	5.66	-0.27	0.17	0.33
12/16/15 9:57:13	2921	-0.01	1.27	5.53	0.08	-0.44	0.36
12/16/15 9:57:24	2932	0.24	1.21	5.38	-0.22	0.26	0.32
12/16/15 9:57:36	2944	-0.03	0.85	6.84	-0.07	0.19	0.34
12/16/15 9:57:48	2956	-0.16	1.85	2.36	0.16	-0.15	0.37
12/16/15 9:57:59	2967	-0.20	0.71	4.03	0.84	-0.07	0.33
12/16/15 9:58:11	2979	0.22	0.93	5.86	0.18	0.06	0.33
12/16/15 9:58:23	2991	-0.24	0.94	3.19	0.36	0.09	0.34

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:58:35	3003	-0.50	1.48	4.00	-0.11	-0.48	0.34
12/16/15 9:58:46	3014	-0.28	0.81	5.14	0.49	0.15	0.34
12/16/15 9:58:58	3026	-0.16	0.44	5.03	0.19	0.06	0.34
12/16/15 9:59:10	3038	0.41	0.70	4.78	0.69	0.05	0.32
12/16/15 9:59:21	3049	-0.19	0.52	4.39	-0.09	0.17	0.35
12/16/15 9:59:33	3061	0.25	0.57	3.45	-0.05	0.64	0.33
12/16/15 9:59:45	3073	-0.19	0.90	5.50	-0.37	-0.13	0.34
12/16/15 9:59:57	3085	0.23	-0.17	7.04	0.19	0.02	0.33
12/16/15 10:00:08	3096	-0.01	0.70	5.72	-0.73	0.30	0.34
12/16/15 10:00:20	3108	0.37	0.92	4.53	0.29	0.30	0.33
12/16/15 10:00:32	3120	0.11	0.50	4.94	0.85	0.07	0.32
12/16/15 10:00:44	3132	-0.14	0.56	6.04	-0.11	0.30	0.33
12/16/15 10:00:56	3144	0.23	0.61	5.27	0.81	0.42	0.34
12/16/15 10:01:07	3155	0.33	1.86	6.16	-0.13	-0.57	0.32
12/16/15 10:01:19	3167	0.22	0.00	5.18	0.64	-0.07	0.33
12/16/15 10:01:31	3179	-0.34	0.86	4.72	-0.23	-0.05	0.35
12/16/15 10:01:43	3191	-0.05	1.18	6.02	-0.24	0.30	0.33
12/16/15 10:01:54	3202	0.23	0.88	5.27	-0.51	0.06	0.33
12/16/15 10:02:06	3214	-0.32	2.19	5.35	-0.39	0.04	0.33
12/16/15 10:02:18	3226	0.24	0.67	5.81	-0.55	0.33	0.33
12/16/15 10:02:30	3238	0.04	-0.39	5.05	0.56	-0.16	0.34
12/16/15 10:02:42	3250	-0.05	0.97	6.93	-0.18	0.10	0.32
12/16/15 10:02:54	3262	0.13	1.05	6.42	0.02	-0.10	0.32
12/16/15 10:03:05	3273	-0.25	0.90	7.00	0.44	0.02	0.34
12/16/15 10:03:17	3285	-0.16	0.91	6.64	-0.32	-0.05	0.33
12/16/15 10:03:29	3297	-0.26	-0.36	6.41	0.35	-0.34	0.34
12/16/15 10:03:41	3309	-0.15	0.26	4.97	0.13	0.01	0.31
12/16/15 10:03:53	3321	-0.01	0.45	4.44	0.63	0.15	0.33
12/16/15 10:04:05	3333	0.47	-0.54	5.61	0.18	0.18	0.29
12/16/15 10:04:17	3345	-0.28	-0.17	5.48	0.94	-0.31	0.33
12/16/15 10:04:28	3356	-0.30	1.02	3.93	0.10	-0.10	0.34
12/16/15 10:04:40	3368	0.20	1.56	4.11	0.32	0.12	0.29
12/16/15 10:04:52	3380	0.19	0.46	3.60	0.77	0.21	0.33
12/16/15 10:05:04	3392	0.29	0.24	2.72	1.30	-0.06	0.31
12/16/15 10:05:16	3404	-0.02	0.92	4.80	0.22	-0.37	0.32
12/16/15 10:05:28	3416	0.52	0.77	5.82	1.50	-0.45	0.32
12/16/15 10:05:40	3428	0.29	1.78	5.46	0.82	0.14	0.33
12/16/15 10:05:52	3440	-0.24	0.30	5.91	0.84	-0.05	0.32

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:06:03	3451	0.23	0.27	3.49	0.53	0.12	0.32
12/16/15 10:06:15	3463	-0.24	1.83	4.84	0.86	0.29	0.31
12/16/15 10:06:27	3475	0.07	1.43	4.41	0.10	0.32	0.31
12/16/15 10:06:39	3487	-0.01	1.76	4.52	0.40	0.14	0.32
12/16/15 10:06:51	3499	0.12	1.47	2.57	0.56	-0.46	0.31
12/16/15 10:07:03	3511	-0.03	0.50	3.82	0.18	0.20	0.32
12/16/15 10:07:15	3523	-0.08	0.34	5.50	-0.57	0.73	0.32
12/16/15 10:07:27	3535	0.23	0.85	4.34	0.62	0.13	0.32
12/16/15 10:07:39	3547	0.00	0.35	6.03	1.00	0.34	0.32
12/16/15 10:07:51	3559	0.00	0.65	3.44	0.68	0.46	0.31
12/16/15 10:08:03	3571	-0.11	0.56	2.22	-0.01	0.10	0.31
12/16/15 10:08:14	3582	0.29	1.22	2.88	-0.33	0.36	0.30
12/16/15 10:08:26	3594	0.14	0.92	5.03	-0.16	0.00	0.30
12/16/15 10:08:38	3606	0.31	0.95	6.04	0.02	0.18	0.31
12/16/15 10:08:50	3618	0.04	1.13	3.91	-0.64	0.39	0.32
12/16/15 10:09:02	3630	0.49	1.39	4.28	0.87	0.03	0.31
12/16/15 10:09:14	3642	0.05	1.93	3.61	0.31	0.23	0.30
12/16/15 10:09:26	3654	0.16	0.98	3.84	0.07	-0.04	0.30
12/16/15 10:09:38	3666	0.28	1.36	5.51	-0.20	-0.12	0.31
12/16/15 10:09:50	3678	-0.34	1.47	4.60	0.56	-0.10	0.30
12/16/15 10:10:02	3690	0.46	0.75	5.65	1.08	0.50	0.33
12/16/15 10:10:14	3702	-0.20	1.16	4.96	-0.07	0.06	0.29
12/16/15 10:10:26	3714	0.16	0.57	4.78	-0.14	-0.60	0.31
12/16/15 10:10:38	3726	0.39	1.32	4.38	0.22	-0.32	0.31
12/16/15 10:10:50	3738	0.38	0.78	4.11	0.13	0.04	0.30
12/16/15 10:11:02	3750	0.09	1.23	4.37	0.15	0.08	0.31
12/16/15 10:11:14	3762	0.01	0.17	3.27	0.11	0.09	0.31
12/16/15 10:11:26	3774	0.57	1.51	4.95	-0.26	0.35	0.31
12/16/15 10:11:38	3786	0.07	0.94	4.21	-0.24	-0.27	0.30
12/16/15 10:11:50	3798	0.24	0.46	3.74	0.93	0.03	0.31
12/16/15 10:12:02	3810	-0.08	0.33	6.56	1.34	0.23	0.30
12/16/15 10:12:15	3823	-0.13	0.20	4.81	-0.62	0.67	0.30
12/16/15 10:12:27	3835	-0.25	0.54	5.03	-0.42	0.20	0.31
12/16/15 10:12:39	3847	-0.07	0.82	6.37	0.22	-0.09	0.29
12/16/15 10:12:51	3859	0.35	0.58	3.66	0.71	0.03	0.30
12/16/15 10:13:03	3871	-0.04	0.37	4.70	-0.15	0.13	0.29
12/16/15 10:13:15	3883	0.55	0.09	4.50	0.83	-0.11	0.31
12/16/15 10:13:27	3895	0.16	1.37	5.08	0.82	0.02	0.29

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:13:39	3907	0.06	0.31	3.64	0.19	0.09	0.31
12/16/15 10:13:51	3919	-0.18	0.43	4.55	0.52	0.05	0.29
12/16/15 10:14:03	3931	0.11	0.77	6.02	0.94	0.47	0.31
12/16/15 10:14:15	3943	0.20	1.42	4.86	0.33	0.49	0.31
12/16/15 10:14:27	3955	0.17	1.21	4.48	-0.09	-0.58	0.32
12/16/15 10:14:39	3967	0.36	0.07	4.35	0.10	0.05	0.30
12/16/15 10:14:52	3980	-0.23	0.41	4.22	0.88	0.07	0.31
12/16/15 10:15:04	3992	0.05	0.45	4.80	0.04	0.35	0.28
12/16/15 10:15:16	4004	-0.03	1.14	5.53	0.14	0.61	0.28
12/16/15 10:15:28	4016	-0.21	0.87	4.63	0.90	0.16	0.33
12/16/15 10:15:40	4028	0.31	0.14	4.25	0.61	-0.29	0.28
12/16/15 10:15:52	4040	0.11	1.04	5.69	-0.16	0.63	0.29
12/16/15 10:16:04	4052	0.02	0.33	4.90	0.63	-0.02	0.31
12/16/15 10:16:16	4064	0.16	-0.08	3.92	0.87	-0.01	0.27
12/16/15 10:16:29	4077	-0.54	0.44	5.07	0.26	0.17	0.07
12/16/15 10:16:41	4089	0.36	0.34	4.55	0.03	-0.44	0.06
12/16/15 10:16:53	4101	-0.09	1.25	4.32	0.19	0.07	0.09
12/16/15 10:17:05	4113	0.15	0.20	4.89	0.22	-0.01	0.10
12/16/15 10:17:17	4125	0.47	0.94	6.18	0.35	-0.24	0.12
12/16/15 10:17:30	4138	0.16	0.52	3.01	-0.17	-0.24	0.14
12/16/15 10:17:42	4150	-0.11	0.35	2.75	-0.23	0.22	0.16
12/16/15 10:17:54	4162	0.34	0.58	1.80	-0.06	-0.16	0.16
12/16/15 10:18:06	4174	-0.15	0.32	1.27	-0.09	0.01	0.17
12/16/15 10:18:18	4186	0.57	-0.08	0.46	1.06	-0.12	0.17
12/16/15 10:18:30	4198	-0.20	0.50	0.10	0.92	0.15	0.18
12/16/15 10:18:43	4211	0.41	0.09	0.70	-0.14	-0.09	0.17
12/16/15 10:18:55	4223	0.19	-0.09	1.53	-0.10	-0.23	0.14
12/16/15 10:19:07	4235	0.26	-0.29	0.68	-0.60	0.05	0.15
12/16/15 10:19:19	4247	0.13	0.46	-6.74	0.16	0.16	0.17
12/16/15 10:19:32	4260	0.39	-0.26	0.83	-0.16	0.22	0.16
12/16/15 10:19:44	4272	-0.37	0.29	-1.63	0.17	0.32	0.16
12/16/15 10:19:56	4284	0.25	0.12	0.14	-0.06	0.04	0.06
12/16/15 10:20:08	4296	-0.12	0.16	-0.73	0.76	-0.06	0.18
12/16/15 10:20:20	4308	0.04	0.06	-0.57	0.32	-0.10	0.14
12/16/15 10:20:33	4321	0.11	0.19	0.63	0.59	0.39	0.15
12/16/15 10:20:45	4333	-0.16	0.69	0.07	-0.14	0.23	0.15
12/16/15 10:20:57	4345	0.02	-0.37	-0.35	0.04	-0.40	0.15
12/16/15 10:21:10	4358	0.35	0.97	0.60	0.44	-0.05	0.15

Date/Time	Cumulative Reaction Time	H ₂	N ₂	NO	O ₂	N ₂ O	NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:21:22	4370	-0.22	0.26	0.11	0.42	0.22	0.14
12/16/15 10:21:34	4382	0.19	-0.45	0.11	-0.39	-0.45	0.13
12/16/15 10:21:46	4394	0.15	0.33	0.34	-0.14	0.07	0.13
12/16/15 10:21:59	4407	0.31	0.39	-0.37	0.42	-0.09	0.14
12/16/15 10:22:11	4419	0.17	0.07	0.01	-0.17	-0.40	0.15
12/16/15 10:22:23	4431	-0.24	-0.40	-0.30	0.07	-0.14	0.14
12/16/15 10:22:36	4444	-0.27	0.04	-0.23	0.07	-0.07	0.14
12/16/15 10:22:48	4456	0.35	0.18	-0.63	1.13	0.20	0.14
12/16/15 10:23:00	4468	-0.05	0.09	0.15	0.36	0.29	0.14
12/16/15 10:23:13	4481	0.13	0.27	0.17	-0.03	-0.46	0.14
12/16/15 10:23:25	4493	-0.01	0.11	0.36	-0.11	-0.21	0.15
12/16/15 10:23:37	4505	-0.23	0.25	0.06	-0.14	0.35	0.15
12/16/15 10:23:49	4517	-0.01	-0.42	1.06	0.14	-0.05	0.14
12/16/15 10:24:02	4530	0.23	-0.31	-0.48	0.52	-0.08	0.13
12/16/15 10:24:14	4542	-0.42	0.12	0.69	-0.56	-0.19	0.12
12/16/15 10:24:27	4555	-0.05	0.62	-0.49	0.13	0.09	0.13
12/16/15 10:24:39	4567	-0.11	-0.15	-0.15	0.45	0.58	0.14
12/16/15 10:24:51	4579	0.05	-0.13	-0.67	0.28	-0.08	0.13
12/16/15 10:25:04	4592	-0.20	-0.63	-1.67	0.41	0.20	0.13
12/16/15 10:25:16	4604	0.06	-0.08	0.75	0.21	0.13	0.14
12/16/15 10:25:28	4616	0.61	-0.51	0.51	-0.18	0.12	0.13
12/16/15 10:25:41	4629	0.35	0.11	-0.08	0.63	-0.15	0.14
12/16/15 10:25:53	4641	-0.01	0.06	-1.48	0.61	-0.10	0.14
12/16/15 10:26:06	4654	0.10	-0.34	-2.40	-0.05	0.07	0.14
12/16/15 10:26:18	4666	-0.15	0.04	-1.54	0.02	0.09	0.13
12/16/15 10:26:30	4678	0.16	0.00	0.71	-0.28	0.07	0.14
12/16/15 10:26:43	4691	-0.19	-0.59	0.50	0.07	-0.51	0.15
12/16/15 10:26:55	4703	-0.01	0.17	-0.83	1.01	0.23	0.13
12/16/15 10:27:08	4716	-0.10	-0.53	0.24	0.16	0.07	0.13
12/16/15 10:27:20	4728	0.01	0.04	-0.55	0.05	0.14	0.14
12/16/15 10:27:32	4740	0.07	-0.28	-1.08	0.38	-0.51	0.12
12/16/15 10:27:45	4753	0.10	0.07	-2.78	0.13	-0.30	0.11
12/16/15 10:27:57	4765	0.13	-0.12	-0.01	-0.07	0.06	0.13
12/16/15 10:28:10	4778	-0.82	0.27	1.67	0.17	0.11	0.12
12/16/15 10:28:22	4790	1.25	0.06	-0.43	-0.68	0.11	0.13
12/16/15 10:28:35	4803	0.37	-0.97	-0.46	0.45	0.01	0.13
12/16/15 10:28:47	4815	-0.45	0.09	1.26	0.68	0.25	0.13
12/16/15 10:29:00	4828	0.26	-0.35	-0.14	0.02	-0.16	0.13

Date/Time	Cumulative Reaction Time	H₂	N₂	NO	O₂	N₂O	NO₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:29:12	4840	0.06	0.04	0.07	0.11	0.09	0.12
12/16/15 10:29:25	4853	-0.12	0.97	1.32	-0.01	0.09	0.13
12/16/15 10:29:37	4865	-0.28	0.38	-1.63	-0.59	0.42	0.12
12/16/15 10:29:50	4878	-0.31	0.02	-0.88	0.02	-0.11	0.11
12/16/15 10:30:02	4890	0.17	-0.14	0.04	0.38	-0.21	0.13

Table E-2. Moving Average Raman Offgas Concentration Data for Room Temp Carbon Steel Dissolution

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:08:32	0	n/a	n/a	n/a	n/a	n/a	n/a
12/16/15 9:08:37	5	-0.04	0.39	-0.09	0.14	0.06	0.02
12/16/15 9:08:49	17	0.04	0.55	-0.11	0.27	0.05	0.02
12/16/15 9:09:00	28	0.03	0.71	0.00	0.36	0.11	0.02
12/16/15 9:09:12	40	0.01	0.77	0.02	0.37	0.08	0.01
12/16/15 9:09:24	52	-0.01	0.71	-0.23	0.37	0.09	0.02
12/16/15 9:09:36	64	-0.04	0.77	-0.11	0.41	0.15	0.02
12/16/15 9:09:48	76	-0.04	0.86	-0.23	0.45	0.16	0.02
12/16/15 9:10:00	88	-0.08	0.88	-0.38	0.42	0.07	0.02
12/16/15 9:10:12	100	-0.07	0.68	-0.35	0.32	-0.05	0.02
12/16/15 9:10:24	112	-0.09	0.56	-0.37	0.25	-0.02	0.02
12/16/15 9:10:35	123	-0.07	0.54	-0.41	0.13	-0.04	0.02
12/16/15 9:10:47	135	0.00	0.49	-0.18	0.09	-0.02	0.02
12/16/15 9:10:59	147	0.02	0.59	-0.10	0.15	-0.02	0.02
12/16/15 9:11:11	159	0.04	0.66	0.23	0.20	-0.10	0.01
12/16/15 9:11:23	171	0.02	0.63	0.45	0.22	-0.15	0.02
12/16/15 9:11:35	183	0.05	0.85	0.59	0.22	-0.10	0.02
12/16/15 9:11:47	195	0.08	0.92	0.66	0.37	-0.01	0.02
12/16/15 9:11:59	207	0.05	1.05	0.64	0.35	-0.05	0.02
12/16/15 9:12:11	219	0.04	1.09	0.49	0.42	-0.03	0.02
12/16/15 9:12:23	231	-0.02	1.18	0.10	0.47	-0.02	0.02
12/16/15 9:12:35	243	-0.03	1.11	0.06	0.43	-0.05	0.02
12/16/15 9:12:46	254	-0.09	1.12	-0.14	0.46	-0.02	0.02
12/16/15 9:12:58	266	-0.04	1.27	-0.10	0.40	0.04	0.02
12/16/15 9:13:10	278	-0.05	1.19	-0.26	0.43	0.05	0.02
12/16/15 9:13:22	290	-0.07	1.21	-0.40	0.36	0.06	0.02
12/16/15 9:13:34	302	-0.03	1.29	-0.31	0.42	0.06	0.03
12/16/15 9:13:46	314	-0.01	1.26	-0.06	0.40	0.05	0.03
12/16/15 9:13:58	326	0.01	1.34	0.29	0.40	0.00	0.04
12/16/15 9:14:10	338	0.00	1.40	0.36	0.40	0.06	0.04
12/16/15 9:14:22	350	0.04	1.42	0.45	0.37	0.11	0.05
12/16/15 9:14:34	362	0.02	1.33	0.26	0.42	0.08	0.06
12/16/15 9:14:46	374	0.00	1.42	0.47	0.41	0.10	0.07
12/16/15 9:14:58	386	0.03	1.40	0.59	0.41	0.13	0.08
12/16/15 9:15:10	398	-0.03	1.34	0.58	0.36	0.06	0.09
12/16/15 9:15:22	410	-0.07	1.41	0.51	0.38	0.10	0.10

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:15:34	422	-0.11	1.32	0.57	0.38	0.13	0.12
12/16/15 9:15:46	434	-0.12	1.29	0.59	0.42	0.13	0.13
12/16/15 9:15:58	446	-0.14	1.28	0.68	0.44	0.12	0.14
12/16/15 9:16:10	458	-0.11	1.33	0.94	0.39	0.12	0.15
12/16/15 9:16:22	470	-0.10	1.30	0.74	0.33	0.10	0.17
12/16/15 9:16:34	482	-0.14	1.36	0.60	0.38	0.07	0.18
12/16/15 9:16:46	494	-0.16	1.42	0.63	0.36	0.13	0.20
12/16/15 9:16:58	506	-0.16	1.35	0.87	0.28	0.09	0.21
12/16/15 9:17:10	518	-0.15	1.44	0.82	0.18	0.05	0.23
12/16/15 9:17:23	531	-0.11	1.54	0.91	0.18	0.05	0.24
12/16/15 9:17:35	543	-0.08	1.41	0.91	0.15	0.13	0.24
12/16/15 9:17:47	555	-0.04	1.41	0.85	0.19	0.17	0.26
12/16/15 9:17:59	567	-0.02	1.41	0.88	0.25	0.19	0.28
12/16/15 9:18:11	579	-0.01	1.55	1.09	0.24	0.16	0.29
12/16/15 9:18:23	591	0.10	1.54	1.18	0.33	0.21	0.31
12/16/15 9:18:35	603	0.16	1.38	1.24	0.47	0.19	0.32
12/16/15 9:18:47	615	0.17	1.33	1.16	0.61	0.20	0.33
12/16/15 9:18:59	627	0.12	1.23	1.24	0.59	0.21	0.35
12/16/15 9:19:11	639	0.18	1.32	1.09	0.46	0.12	0.37
12/16/15 9:19:23	651	0.11	1.30	1.10	0.41	0.12	0.38
12/16/15 9:19:35	663	0.17	1.25	1.37	0.38	0.07	0.39
12/16/15 9:19:48	676	0.15	1.11	1.42	0.38	0.15	0.40
12/16/15 9:20:00	688	0.15	0.99	1.50	0.31	0.16	0.41
12/16/15 9:20:12	700	0.12	1.28	1.38	0.21	0.19	0.42
12/16/15 9:20:24	712	0.14	1.05	1.47	0.15	0.16	0.43
12/16/15 9:20:36	724	0.21	1.13	1.29	0.18	0.13	0.44
12/16/15 9:20:48	736	0.19	1.12	1.47	0.28	0.09	0.45
12/16/15 9:21:00	748	0.28	0.99	1.74	0.41	0.06	0.46
12/16/15 9:21:13	761	0.28	1.09	1.60	0.49	0.12	0.46
12/16/15 9:21:25	773	0.40	1.04	1.46	0.43	0.03	0.47
12/16/15 9:21:37	785	0.34	1.11	1.53	0.40	-0.03	0.47
12/16/15 9:21:49	797	0.27	0.99	1.70	0.41	-0.02	0.47
12/16/15 9:22:01	809	0.25	1.22	1.90	0.41	0.04	0.48
12/16/15 9:22:13	821	0.20	1.28	2.04	0.30	0.09	0.48
12/16/15 9:22:26	834	0.12	1.29	2.42	0.44	0.07	0.49
12/16/15 9:22:38	846	0.01	1.45	2.26	0.28	0.08	0.49
12/16/15 9:22:50	858	-0.03	1.43	2.47	0.34	0.03	0.49
12/16/15 9:23:02	870	-0.15	1.33	2.84	0.55	0.10	0.50

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:23:14	882	-0.16	1.41	2.54	0.55	0.05	0.51
12/16/15 9:23:27	895	-0.07	1.26	2.64	0.69	0.00	0.51
12/16/15 9:23:39	907	-0.06	1.30	2.49	0.78	-0.05	0.51
12/16/15 9:23:51	919	-0.05	1.12	2.66	0.91	-0.10	0.52
12/16/15 9:24:03	931	-0.01	1.14	2.73	0.89	-0.08	0.52
12/16/15 9:24:16	944	0.04	1.15	2.44	1.02	-0.04	0.51
12/16/15 9:24:28	956	0.05	0.98	2.45	1.07	-0.04	0.51
12/16/15 9:24:40	968	0.10	1.04	2.42	0.84	-0.16	0.51
12/16/15 9:24:52	980	0.17	1.01	2.68	0.98	-0.11	0.51
12/16/15 9:25:04	992	0.14	1.15	2.54	0.94	-0.07	0.51
12/16/15 9:25:17	1005	0.23	1.27	2.53	1.00	0.04	0.51
12/16/15 9:25:29	1017	0.29	1.28	2.46	0.90	0.05	0.51
12/16/15 9:25:41	1029	0.30	1.26	1.90	0.91	0.14	0.51
12/16/15 9:25:54	1042	0.34	1.09	2.47	0.91	0.15	0.52
12/16/15 9:26:06	1054	0.33	1.19	2.29	0.80	0.11	0.52
12/16/15 9:26:18	1066	0.30	1.13	2.61	0.83	0.27	0.52
12/16/15 9:26:30	1078	0.27	1.12	2.77	0.68	0.28	0.53
12/16/15 9:26:43	1091	0.27	1.01	2.67	0.71	0.28	0.53
12/16/15 9:26:55	1103	0.17	0.85	2.63	0.45	0.15	0.52
12/16/15 9:27:07	1115	0.20	0.91	2.44	0.50	0.15	0.52
12/16/15 9:27:20	1128	0.20	0.92	2.59	0.42	0.05	0.52
12/16/15 9:27:32	1140	0.17	1.10	2.33	0.30	-0.04	0.52
12/16/15 9:27:44	1152	0.15	1.08	2.75	0.32	0.00	0.52
12/16/15 9:27:57	1165	0.19	1.23	2.59	0.34	-0.04	0.52
12/16/15 9:28:09	1177	0.21	1.06	2.66	0.38	-0.05	0.52
12/16/15 9:28:21	1189	0.23	1.17	2.93	0.31	-0.08	0.52
12/16/15 9:28:34	1202	0.26	1.24	3.03	0.40	-0.03	0.52
12/16/15 9:28:46	1214	0.20	1.17	3.06	0.30	-0.03	0.52
12/16/15 9:28:58	1226	0.18	1.19	2.92	0.19	-0.06	0.52
12/16/15 9:29:11	1239	0.19	1.13	3.23	0.24	-0.01	0.51
12/16/15 9:29:23	1251	0.16	1.22	3.26	0.27	0.08	0.51
12/16/15 9:29:35	1263	0.20	1.10	3.19	0.24	0.10	0.51
12/16/15 9:29:48	1276	0.17	1.22	3.24	0.19	0.14	0.51
12/16/15 9:30:00	1288	0.19	1.21	3.21	0.10	0.17	0.50
12/16/15 9:30:12	1300	0.12	1.17	3.42	0.13	0.22	0.50
12/16/15 9:30:25	1313	0.12	1.27	3.83	0.24	0.22	0.50
12/16/15 9:30:37	1325	0.03	1.15	3.75	0.30	0.28	0.50
12/16/15 9:30:50	1338	0.06	1.20	3.40	0.37	0.19	0.50

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:31:02	1350	0.03	1.08	3.07	0.26	0.14	0.49
12/16/15 9:31:14	1362	-0.02	0.97	3.16	0.28	0.17	0.50
12/16/15 9:31:27	1375	0.00	0.91	2.72	0.28	0.13	0.50
12/16/15 9:31:39	1387	-0.08	0.88	2.99	0.43	0.12	0.50
12/16/15 9:31:52	1400	-0.04	0.77	3.10	0.40	0.09	0.50
12/16/15 9:32:04	1412	-0.09	0.80	3.31	0.28	-0.02	0.50
12/16/15 9:32:17	1425	-0.01	0.84	3.55	0.35	-0.01	0.49
12/16/15 9:32:29	1437	-0.10	0.71	3.55	0.30	0.12	0.49
12/16/15 9:32:41	1449	-0.02	0.71	3.52	0.53	0.10	0.50
12/16/15 9:32:54	1462	-0.01	0.91	3.11	0.56	0.03	0.50
12/16/15 9:33:06	1474	-0.01	1.00	2.51	0.57	0.04	0.49
12/16/15 9:33:19	1487	0.01	1.05	1.77	0.40	0.02	0.49
12/16/15 9:33:31	1499	0.04	1.10	2.29	0.38	0.02	0.48
12/16/15 9:33:44	1512	0.11	1.03	1.93	0.44	0.19	0.48
12/16/15 9:33:56	1524	0.12	1.02	2.25	0.30	0.26	0.48
12/16/15 9:34:09	1537	0.15	0.98	2.62	0.37	0.17	0.48
12/16/15 9:34:21	1549	0.11	1.09	2.91	0.20	0.13	0.48
12/16/15 9:34:33	1561	0.05	0.99	3.22	0.23	0.14	0.48
12/16/15 9:34:46	1574	0.00	1.12	4.27	0.40	0.12	0.48
12/16/15 9:34:58	1586	-0.01	1.17	4.59	0.47	0.08	0.48
12/16/15 9:35:11	1599	-0.02	1.28	4.39	0.49	0.06	0.48
12/16/15 9:35:23	1611	-0.03	1.17	4.55	0.59	-0.03	0.48
12/16/15 9:35:36	1624	0.03	1.31	4.61	0.71	-0.01	0.48
12/16/15 9:35:49	1637	0.06	1.34	4.94	0.67	-0.01	0.48
12/16/15 9:36:01	1649	0.07	1.32	4.87	0.63	0.01	0.48
12/16/15 9:36:14	1662	0.11	1.41	5.35	0.70	-0.02	0.47
12/16/15 9:36:26	1674	0.10	1.26	5.32	0.64	0.01	0.47
12/16/15 9:36:39	1687	0.16	1.23	5.65	0.66	-0.05	0.47
12/16/15 9:36:51	1699	0.19	1.09	5.10	0.66	0.01	0.47
12/16/15 9:37:04	1712	0.13	1.13	5.33	0.46	0.05	0.47
12/16/15 9:37:16	1724	0.11	1.02	5.04	0.40	-0.03	0.47
12/16/15 9:37:29	1737	0.08	0.95	4.48	0.53	0.01	0.47
12/16/15 9:37:41	1749	0.14	0.81	4.05	0.59	0.01	0.47
12/16/15 9:37:54	1762	0.18	0.76	3.57	0.50	0.10	0.46
12/16/15 9:38:06	1774	0.26	0.65	3.67	0.52	0.13	0.46
12/16/15 9:38:19	1787	0.23	0.66	3.70	0.46	0.17	0.46
12/16/15 9:44:10	2138	0.17	0.67	3.91	0.39	0.13	0.45
12/16/15 9:44:21	2149	0.21	0.78	3.45	0.57	0.07	0.44

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:44:33	2161	0.16	0.93	3.79	0.57	0.05	0.44
12/16/15 9:44:44	2172	0.18	1.03	4.02	0.51	0.00	0.43
12/16/15 9:44:55	2183	0.13	1.26	4.60	0.37	-0.05	0.42
12/16/15 9:45:07	2195	0.07	1.24	4.85	0.41	-0.08	0.41
12/16/15 9:45:18	2206	-0.01	1.38	4.55	0.26	-0.12	0.40
12/16/15 9:45:29	2217	-0.02	1.20	4.87	0.23	-0.07	0.40
12/16/15 9:45:41	2229	0.01	1.14	5.01	0.20	-0.03	0.40
12/16/15 9:45:52	2240	0.02	1.02	5.21	0.20	-0.02	0.40
12/16/15 9:46:03	2251	0.04	0.99	5.28	0.24	0.01	0.40
12/16/15 9:46:15	2263	0.08	1.05	5.63	0.12	0.01	0.40
12/16/15 9:46:26	2274	0.05	0.99	5.52	0.16	0.01	0.40
12/16/15 9:46:38	2286	0.04	1.00	5.47	0.16	0.01	0.40
12/16/15 9:46:49	2297	0.06	0.95	5.71	0.25	-0.01	0.40
12/16/15 9:47:00	2308	0.05	1.18	5.21	0.29	0.01	0.40
12/16/15 9:47:12	2320	0.13	1.17	5.09	0.32	0.05	0.40
12/16/15 9:47:23	2331	0.18	1.22	5.18	0.38	0.07	0.40
12/16/15 9:47:35	2343	0.12	1.18	5.02	0.38	0.18	0.40
12/16/15 9:47:46	2354	0.02	1.14	4.74	0.37	0.25	0.40
12/16/15 9:47:57	2365	0.11	1.06	4.74	0.49	0.34	0.40
12/16/15 9:48:09	2377	0.11	0.93	4.65	0.35	0.32	0.40
12/16/15 9:48:20	2388	0.13	0.84	4.79	0.26	0.35	0.40
12/16/15 9:48:32	2400	0.12	0.72	5.07	0.18	0.35	0.39
12/16/15 9:48:43	2411	0.05	0.74	4.95	0.25	0.28	0.39
12/16/15 9:48:55	2423	-0.02	0.71	4.93	0.08	0.31	0.39
12/16/15 9:49:06	2434	0.02	0.62	5.18	0.09	0.25	0.38
12/16/15 9:49:18	2446	0.05	0.56	5.21	0.02	0.28	0.38
12/16/15 9:49:29	2457	0.03	0.54	5.44	-0.12	0.26	0.38
12/16/15 9:49:41	2469	0.02	0.58	5.43	-0.12	0.27	0.38
12/16/15 9:49:52	2480	0.03	0.66	5.43	-0.16	0.29	0.38
12/16/15 9:50:04	2492	0.08	0.74	5.57	-0.07	0.24	0.37
12/16/15 9:50:15	2503	0.07	0.78	5.73	-0.01	0.26	0.37
12/16/15 9:50:27	2515	0.05	0.89	5.95	0.06	0.24	0.37
12/16/15 9:50:38	2526	0.02	0.90	5.75	0.08	0.21	0.37
12/16/15 9:50:50	2538	-0.02	1.02	5.66	0.22	0.12	0.37
12/16/15 9:51:01	2549	-0.06	1.05	5.14	0.32	0.13	0.37
12/16/15 9:51:13	2561	-0.02	1.15	5.18	0.41	0.17	0.37
12/16/15 9:51:24	2572	0.00	1.13	4.83	0.54	0.14	0.37
12/16/15 9:51:36	2584	-0.04	1.01	4.69	0.48	0.19	0.37

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:51:48	2596	-0.05	0.99	4.46	0.46	0.20	0.37
12/16/15 9:51:59	2607	-0.09	0.81	4.20	0.50	0.16	0.37
12/16/15 9:52:11	2619	0.01	0.68	4.23	0.45	0.12	0.37
12/16/15 9:52:22	2630	0.07	0.51	4.14	0.36	0.14	0.37
12/16/15 9:52:34	2642	0.11	0.51	4.38	0.37	0.00	0.36
12/16/15 9:52:45	2653	0.08	0.54	4.31	0.30	-0.05	0.36
12/16/15 9:52:57	2665	0.00	0.68	4.86	0.30	-0.09	0.36
12/16/15 9:53:09	2677	0.03	0.61	4.83	0.29	-0.14	0.37
12/16/15 9:53:20	2688	0.08	0.47	5.17	0.35	-0.17	0.37
12/16/15 9:53:32	2700	0.20	0.55	5.23	0.27	-0.10	0.37
12/16/15 9:53:43	2711	0.16	0.61	5.42	0.30	-0.13	0.36
12/16/15 9:53:55	2723	0.16	0.66	5.84	0.38	-0.15	0.36
12/16/15 9:54:06	2734	0.15	0.48	5.96	0.40	-0.04	0.36
12/16/15 9:54:18	2746	0.16	0.40	6.11	0.45	-0.05	0.36
12/16/15 9:54:30	2758	0.18	0.28	5.89	0.41	-0.01	0.36
12/16/15 9:54:41	2769	0.19	0.38	5.85	0.51	0.00	0.36
12/16/15 9:54:53	2781	0.20	0.62	5.98	0.44	0.01	0.36
12/16/15 9:55:04	2792	0.14	0.56	5.83	0.32	-0.06	0.35
12/16/15 9:55:16	2804	0.12	0.69	5.58	0.26	0.06	0.35
12/16/15 9:55:28	2816	0.10	0.78	5.33	0.25	0.10	0.35
12/16/15 9:55:39	2827	0.14	0.97	5.36	0.19	0.08	0.35
12/16/15 9:55:51	2839	0.18	1.04	5.05	0.25	0.11	0.35
12/16/15 9:56:03	2851	0.17	0.93	4.87	0.23	0.15	0.35
12/16/15 9:56:14	2862	0.15	0.93	4.75	0.27	0.18	0.35
12/16/15 9:56:26	2874	0.05	0.86	4.62	0.16	0.23	0.35
12/16/15 9:56:38	2886	0.07	0.94	4.98	0.28	0.29	0.35
12/16/15 9:56:49	2897	0.14	1.00	5.09	0.38	0.25	0.35
12/16/15 9:57:01	2909	0.13	1.01	5.12	0.26	0.25	0.35
12/16/15 9:57:13	2921	0.08	1.06	5.09	0.25	0.19	0.35
12/16/15 9:57:24	2932	0.05	1.07	5.40	0.12	0.19	0.34
12/16/15 9:57:36	2944	0.07	1.16	5.71	0.11	0.16	0.34
12/16/15 9:57:48	2956	0.04	1.30	5.42	0.04	0.13	0.35
12/16/15 9:57:59	2967	0.07	1.25	5.19	0.17	0.06	0.34
12/16/15 9:58:11	2979	0.08	1.25	5.18	0.19	0.03	0.34
12/16/15 9:58:23	2991	-0.03	1.14	4.86	0.13	0.01	0.34
12/16/15 9:58:35	3003	-0.09	1.16	4.65	0.15	-0.07	0.34
12/16/15 9:58:46	3014	-0.12	1.10	4.60	0.20	0.01	0.34
12/16/15 9:58:58	3026	-0.17	1.00	4.56	0.25	-0.02	0.34

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 9:59:10	3038	-0.11	0.98	4.30	0.35	-0.04	0.34
12/16/15 9:59:21	3049	-0.12	0.82	4.55	0.32	0.00	0.34
12/16/15 9:59:33	3061	-0.06	0.80	4.48	0.21	0.09	0.34
12/16/15 9:59:45	3073	-0.11	0.79	4.43	0.14	0.07	0.34
12/16/15 9:59:57	3085	-0.05	0.66	4.92	0.12	0.06	0.34
12/16/15 10:00:08	3096	0.01	0.56	5.13	0.04	0.16	0.34
12/16/15 10:00:20	3108	0.09	0.57	5.05	0.02	0.18	0.33
12/16/15 10:00:32	3120	0.12	0.58	5.04	0.10	0.18	0.33
12/16/15 10:00:44	3132	0.05	0.56	5.20	0.00	0.21	0.33
12/16/15 10:00:56	3144	0.11	0.58	5.31	0.11	0.24	0.33
12/16/15 10:01:07	3155	0.11	0.74	5.65	0.10	0.09	0.33
12/16/15 10:01:19	3167	0.17	0.63	5.61	0.23	0.10	0.33
12/16/15 10:01:31	3179	0.10	0.75	5.32	0.17	0.09	0.33
12/16/15 10:01:43	3191	0.09	0.81	5.36	0.24	0.09	0.33
12/16/15 10:01:54	3202	0.07	0.81	5.45	0.14	0.06	0.33
12/16/15 10:02:06	3214	0.02	1.02	5.50	-0.02	0.05	0.33
12/16/15 10:02:18	3226	0.07	1.03	5.47	-0.07	0.06	0.33
12/16/15 10:02:30	3238	0.04	0.91	5.45	-0.11	-0.01	0.33
12/16/15 10:02:42	3250	0.00	0.79	5.54	-0.11	0.07	0.33
12/16/15 10:02:54	3262	-0.02	0.93	5.70	-0.19	0.07	0.33
12/16/15 10:03:05	3273	0.00	0.93	5.98	-0.10	0.07	0.33
12/16/15 10:03:17	3285	-0.02	0.90	6.06	-0.12	0.03	0.33
12/16/15 10:03:29	3297	-0.08	0.74	6.20	-0.01	-0.02	0.33
12/16/15 10:03:41	3309	-0.06	0.50	6.15	0.06	-0.02	0.33
12/16/15 10:03:53	3321	-0.09	0.47	5.98	0.20	-0.05	0.33
12/16/15 10:04:05	3333	-0.03	0.46	6.05	0.16	0.00	0.32
12/16/15 10:04:17	3345	-0.06	0.31	5.87	0.30	-0.06	0.32
12/16/15 10:04:28	3356	-0.12	0.31	5.56	0.31	-0.06	0.32
12/16/15 10:04:40	3368	-0.06	0.39	5.20	0.29	-0.04	0.32
12/16/15 10:04:52	3380	-0.02	0.34	4.82	0.43	-0.01	0.32
12/16/15 10:05:04	3392	0.05	0.41	4.36	0.55	0.02	0.32
12/16/15 10:05:16	3404	0.07	0.49	4.34	0.56	-0.02	0.32
12/16/15 10:05:28	3416	0.13	0.53	4.51	0.67	-0.10	0.32
12/16/15 10:05:40	3428	0.11	0.82	4.49	0.75	-0.10	0.32
12/16/15 10:05:52	3440	0.12	0.88	4.54	0.73	-0.07	0.32
12/16/15 10:06:03	3451	0.18	0.79	4.49	0.79	-0.04	0.32
12/16/15 10:06:15	3463	0.13	0.82	4.58	0.85	-0.02	0.32
12/16/15 10:06:27	3475	0.11	0.94	4.68	0.77	-0.01	0.32

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:06:39	3487	0.08	1.13	4.90	0.66	0.02	0.32
12/16/15 10:06:51	3499	0.09	1.20	4.62	0.70	0.01	0.32
12/16/15 10:07:03	3511	0.02	1.17	4.37	0.54	0.09	0.32
12/16/15 10:07:15	3523	-0.02	0.99	4.38	0.36	0.16	0.32
12/16/15 10:07:27	3535	0.04	1.05	4.18	0.34	0.18	0.32
12/16/15 10:07:39	3547	0.01	1.07	4.50	0.39	0.21	0.32
12/16/15 10:07:51	3559	0.04	0.92	4.33	0.37	0.23	0.32
12/16/15 10:08:03	3571	0.01	0.81	4.05	0.36	0.20	0.32
12/16/15 10:08:14	3582	0.05	0.74	3.85	0.26	0.23	0.31
12/16/15 10:08:26	3594	0.05	0.67	4.16	0.17	0.29	0.31
12/16/15 10:08:38	3606	0.10	0.73	4.44	0.15	0.29	0.31
12/16/15 10:08:50	3618	0.11	0.83	4.24	0.15	0.25	0.31
12/16/15 10:09:02	3630	0.15	0.90	4.23	0.18	0.23	0.31
12/16/15 10:09:14	3642	0.15	1.09	3.93	0.09	0.22	0.31
12/16/15 10:09:26	3654	0.17	1.14	3.98	0.01	0.16	0.31
12/16/15 10:09:38	3666	0.22	1.23	4.39	-0.01	0.13	0.31
12/16/15 10:09:50	3678	0.14	1.27	4.60	0.10	0.07	0.31
12/16/15 10:10:02	3690	0.18	1.24	4.68	0.26	0.13	0.31
12/16/15 10:10:14	3702	0.12	1.27	4.54	0.25	0.12	0.31
12/16/15 10:10:26	3714	0.13	1.20	4.65	0.31	-0.01	0.31
12/16/15 10:10:38	3726	0.12	1.19	4.66	0.23	-0.05	0.31
12/16/15 10:10:50	3738	0.16	1.05	4.73	0.21	-0.07	0.31
12/16/15 10:11:02	3750	0.15	1.08	4.79	0.22	-0.06	0.31
12/16/15 10:11:14	3762	0.12	0.93	4.51	0.25	-0.03	0.31
12/16/15 10:11:26	3774	0.23	0.94	4.56	0.15	0.02	0.31
12/16/15 10:11:38	3786	0.18	0.96	4.38	-0.01	-0.07	0.31
12/16/15 10:11:50	3798	0.24	0.87	4.23	0.11	-0.08	0.31
12/16/15 10:12:02	3810	0.21	0.84	4.45	0.30	0.03	0.31
12/16/15 10:12:15	3823	0.14	0.70	4.50	0.19	0.15	0.31
12/16/15 10:12:27	3835	0.06	0.67	4.62	0.12	0.17	0.31
12/16/15 10:12:39	3847	0.04	0.62	4.87	0.13	0.15	0.30
12/16/15 10:12:51	3859	0.09	0.67	4.92	0.21	0.14	0.30
12/16/15 10:13:03	3871	0.01	0.53	4.89	0.22	0.12	0.30
12/16/15 10:13:15	3883	0.07	0.42	4.92	0.35	0.14	0.30
12/16/15 10:13:27	3895	0.06	0.54	5.09	0.34	0.14	0.30
12/16/15 10:13:39	3907	0.08	0.54	4.73	0.20	0.12	0.30
12/16/15 10:13:51	3919	0.07	0.56	4.69	0.34	0.04	0.30
12/16/15 10:14:03	3931	0.12	0.59	4.82	0.51	0.07	0.30

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:14:15	3943	0.15	0.67	4.63	0.52	0.15	0.30
12/16/15 10:14:27	3955	0.13	0.75	4.73	0.42	0.07	0.30
12/16/15 10:14:39	3967	0.18	0.71	4.69	0.46	0.06	0.30
12/16/15 10:14:52	3980	0.08	0.75	4.65	0.46	0.08	0.31
12/16/15 10:15:04	3992	0.07	0.63	4.62	0.37	0.12	0.30
12/16/15 10:15:16	4004	0.06	0.74	4.85	0.36	0.19	0.30
12/16/15 10:15:28	4016	0.05	0.79	4.86	0.41	0.20	0.31
12/16/15 10:15:40	4028	0.08	0.71	4.64	0.37	0.11	0.30
12/16/15 10:15:52	4040	0.07	0.67	4.74	0.30	0.13	0.30
12/16/15 10:16:04	4052	0.05	0.55	4.79	0.39	0.19	0.30
12/16/15 10:16:16	4064	0.02	0.54	4.74	0.49	0.19	0.30
12/16/15 10:16:29	4077	-0.02	0.54	4.85	0.41	0.20	0.27
12/16/15 10:16:41	4089	0.02	0.53	4.82	0.41	0.10	0.24
12/16/15 10:16:53	4101	0.01	0.54	4.67	0.42	0.03	0.21
12/16/15 10:17:05	4113	0.06	0.46	4.70	0.33	0.01	0.19
12/16/15 10:17:17	4125	0.08	0.56	4.94	0.30	0.02	0.17
12/16/15 10:17:30	4138	0.09	0.49	4.60	0.30	-0.09	0.15
12/16/15 10:17:42	4150	0.07	0.50	4.34	0.19	-0.06	0.13
12/16/15 10:17:54	4162	0.09	0.58	4.07	0.07	-0.08	0.11
12/16/15 10:18:06	4174	0.14	0.56	3.60	0.03	-0.10	0.12
12/16/15 10:18:18	4186	0.17	0.51	3.08	0.16	-0.06	0.14
12/16/15 10:18:30	4198	0.15	0.42	2.56	0.25	-0.05	0.15
12/16/15 10:18:43	4211	0.19	0.40	2.03	0.20	-0.06	0.16
12/16/15 10:18:55	4223	0.15	0.28	1.45	0.15	-0.06	0.16
12/16/15 10:19:07	4235	0.16	0.17	1.16	0.10	-0.02	0.16
12/16/15 10:19:19	4247	0.19	0.19	-0.02	0.14	-0.03	0.16
12/16/15 10:19:32	4260	0.20	0.08	-0.14	0.13	0.02	0.16
12/16/15 10:19:44	4272	0.17	0.08	-0.51	0.16	0.06	0.16
12/16/15 10:19:56	4284	0.13	0.10	-0.55	0.02	0.08	0.15
12/16/15 10:20:08	4296	0.14	0.06	-0.65	0.00	0.05	0.15
12/16/15 10:20:20	4308	0.10	0.06	-0.81	0.06	0.05	0.15
12/16/15 10:20:33	4321	0.08	0.09	-0.92	0.15	0.12	0.15
12/16/15 10:20:45	4333	0.03	0.21	-1.00	0.20	0.15	0.15
12/16/15 10:20:57	4345	0.02	0.11	-0.20	0.19	0.08	0.14
12/16/15 10:21:10	4358	0.01	0.26	-0.23	0.26	0.04	0.14
12/16/15 10:21:22	4370	0.03	0.26	-0.01	0.30	0.03	0.14
12/16/15 10:21:34	4382	0.02	0.19	-0.02	0.26	-0.03	0.15
12/16/15 10:21:46	4394	0.06	0.21	0.12	0.14	-0.01	0.14

Date/Time	Cumulative Reaction Time	MA H ₂	MA N ₂	MA NO	MA O ₂	MA N ₂ O	MA NO ₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:21:59	4407	0.09	0.25	0.14	0.15	-0.01	0.14
12/16/15 10:22:11	4419	0.10	0.24	0.07	0.06	-0.11	0.14
12/16/15 10:22:23	4431	0.09	0.10	0.02	0.09	-0.16	0.14
12/16/15 10:22:36	4444	0.05	0.15	0.03	0.09	-0.12	0.14
12/16/15 10:22:48	4456	0.05	0.05	-0.12	0.18	-0.08	0.14
12/16/15 10:23:00	4468	0.08	0.03	-0.12	0.17	-0.08	0.14
12/16/15 10:23:13	4481	0.07	0.12	-0.11	0.21	-0.08	0.14
12/16/15 10:23:25	4493	0.05	0.09	-0.11	0.22	-0.11	0.14
12/16/15 10:23:37	4505	-0.02	0.08	-0.05	0.15	-0.06	0.14
12/16/15 10:23:49	4517	-0.04	0.01	0.08	0.19	-0.01	0.14
12/16/15 10:24:02	4530	0.02	0.03	0.06	0.24	-0.01	0.14
12/16/15 10:24:14	4542	0.00	0.04	0.17	0.16	-0.02	0.14
12/16/15 10:24:27	4555	-0.05	0.09	0.19	0.04	-0.03	0.14
12/16/15 10:24:39	4567	-0.06	0.06	0.15	0.05	0.00	0.14
12/16/15 10:24:51	4579	-0.07	0.01	0.05	0.09	0.05	0.14
12/16/15 10:25:04	4592	-0.09	-0.08	-0.21	0.15	0.10	0.13
12/16/15 10:25:16	4604	-0.05	-0.12	-0.12	0.20	0.08	0.13
12/16/15 10:25:28	4616	0.02	-0.13	-0.19	0.16	0.10	0.13
12/16/15 10:25:41	4629	0.04	-0.08	-0.14	0.17	0.09	0.13
12/16/15 10:25:53	4641	0.09	-0.09	-0.41	0.32	0.10	0.13
12/16/15 10:26:06	4654	0.11	-0.21	-0.65	0.29	0.10	0.13
12/16/15 10:26:18	4666	0.10	-0.19	-0.82	0.24	0.04	0.13
12/16/15 10:26:30	4678	0.12	-0.17	-0.65	0.17	0.05	0.13
12/16/15 10:26:43	4691	0.12	-0.17	-0.38	0.13	-0.04	0.14
12/16/15 10:26:55	4703	0.11	-0.13	-0.58	0.23	-0.02	0.14
12/16/15 10:27:08	4716	0.02	-0.14	-0.61	0.27	-0.03	0.13
12/16/15 10:27:20	4728	-0.02	-0.14	-0.67	0.20	0.01	0.14
12/16/15 10:27:32	4740	-0.01	-0.19	-0.62	0.17	-0.05	0.13
12/16/15 10:27:45	4753	-0.01	-0.14	-0.67	0.19	-0.09	0.13
12/16/15 10:27:57	4765	0.02	-0.16	-0.47	0.18	-0.10	0.13
12/16/15 10:28:10	4778	-0.10	-0.12	-0.35	0.24	-0.09	0.13
12/16/15 10:28:22	4790	0.08	-0.04	-0.47	0.14	-0.01	0.13
12/16/15 10:28:35	4803	0.12	-0.18	-0.42	0.07	-0.04	0.13
12/16/15 10:28:47	4815	0.08	-0.11	-0.30	0.14	-0.02	0.13
12/16/15 10:29:00	4828	0.11	-0.15	-0.24	0.13	-0.05	0.12
12/16/15 10:29:12	4840	0.11	-0.11	-0.10	0.10	0.02	0.13
12/16/15 10:29:25	4853	0.08	0.00	0.41	0.08	0.07	0.13
12/16/15 10:29:37	4865	0.03	0.06	0.21	0.02	0.12	0.13

Date/Time	Cumulative Reaction Time	MA H₂	MA N₂	MA NO	MA O₂	MA N₂O	MA NO₂
	(sec)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12/16/15 10:29:50	4878	0.10	0.03	-0.11	0.00	0.09	0.12
12/16/15 10:30:02	4890	-0.04	0.00	-0.05	0.13	0.05	0.12

Distribution:

R. T. Burns, 221-H
J. L. Bodkin, 221-H
W. H. Clifton, Jr. 704-2H
W. G. Dyer, 704-2H
S. L. Garrison, 704-2H
J. R. Lint, 704-2H
R. M. Mobley, 703-H
P. M. Palmer, 221-H
S. J. Robertson, 704-2H
J. B. Schaade, 704-2H
C. M. Hadden, 704-2H
D. E. Welliver, 221-H
K. P. Burrows, 704-2H
P. M. Almond, 773-A
K. P. Crapse, 773-A
W. E. Daniel, 999-W
E. A. Kyser, 773-A
R. A. Pierce, 773-A
T. S. Rudisill, 773-A
M. C. Thompson, 773-A
N. M. Askew, 703-41A
T. B. Brown, 773-A
M. E. Cercy, 773-42A
D. A. Crowley, 773-43A
D. E. Dooley, 773-A
A. P. Fellingner, 773-42A
S. D. Fink, 773-A
C. C. Herman, 773-A
D. T. Hobbs, 773-A
E. N. Hoffman, 999-W
J. E. Hyatt, 773-A
K. M. Kostelnik, 773-42A
B. B. Looney, 773-42A
D. H. McGuire, 999-W
T. O. Oliver, 773-42A
F. M. Pennebaker, 773-42A
G. N. Smoland, 773-42A
B. J. Wiedenman, 773-42A
W. R. Wilmarth, 773-A
P. R. Jackson, 703-46A

Records Administration (EDWS)