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Chemical Dissolution of Simulant FCA Cladding and Plates

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EXECUTIVE SUMMARY

The Savannah River Site (SRS) has received some fast critical assembly (FCA) fuel from the Japan Atomic Energy Agency (JAEA) for disposition. Among the JAEA FCA fuel are approximately 7090 rectangular Stainless Steel clad fuel elements. Each element has an internal Pu-10.6Al alloy metal wafer. The thickness of each element is either 1/16 inch or 1/32 inch. The dimensions of each element ranges from 2 inches x 1 inch to 2 inches x 4 inches.

This report discusses the potential chemical dissolution of the FCA clad material or stainless steel. This technology uses nitric acid-potassium fluoride (HNO_3-KF) flowsheets of H-Canyon to dissolve the FCA elements from a rack of materials. Historically, dissolution flowsheets have aimed to maximize Pu dissolution rates while minimizing stainless steel dissolution (corrosion) rates. Because the FCA cladding is made of stainless steel, this work sought to accelerate stainless steel dissolution.

Initial experiments were performed to identify optimal dissolution conditions for stainless steel in terms of HNO_3 molarity (4 M, 7 M, 10 M) and fluoride (F^-) molarity (0.1 M, 0.25 M, 0.4 M). All initial tests had 1.2 g/L B to represent the neutron poison used. Testing was performed at 102 °C, which is just below the boiling point of 4 M HNO_3. The initial experiments showed that higher stainless steel dissolution rates corresponded with low HNO_3 molarity and high F^- molarity. These results were not surprising as the fluoride ion breaks through the oxide layer formed on the stainless steel surface to cause dissolution or corrosion and having lower molar acid reduces the oxidation potential of the solution. For simulant FCA studies performed after the initial studies, the optimal dissolution conditions were 4 M HNO_3 and 0.25 M KF at 102 °C. Using the optimal conditions, long-term dissolutions were performed on simulant FCA fuel elements.

The simulant FCA fuel elements consisted of two stainless steel face plates surrounding an internal Cu plate. The internal Cu plate simulated Pu which would dissolve readily when the cladding was breached during dissolution. These long-term studies included testing to see whether spacing between simulant FCA fuel elements affected the uniform dissolution rate between fuel elements. The long-term studies with the simulant FCA fuel elements included plain stainless steel plates as a control. For one experiment, the combination of the stainless steel plate plus the simulant FCA element provided a stainless steel surface area to liquid volume ratio comparable to those encountered in the H-Canyon dissolver.

The long-term experiments showed that the FCA simulant fuel elements had similar dissolution rates with the plain stainless steel plates. An anomaly occurred when a weld on the FCA simulant element breached. The weld breach allowed the internal Cu plate to dissolve even though the facing had not been breached. As the internal Cu plate rapidly dissolved, there was a sudden mass loss which caused the FCA dissolution rate to increase an order of magnitude. After copper dissolution, the FCA simulant cladding dissolution rate returned to levels comparable to those of the plain stainless steel. It is not known whether all FCA fuel elements could be counted on to fail in a similar manner as they were welded using different weld techniques.

These long-term studies showed that the stainless steel dissolution rate started high (0.006-0.007 mg/(cm^2*min)) and gradually decreased over 30 days to about 0.001 mg/(cm^2*min). Fitting the 30-day incremental dissolution rate data to a power law function and using a surface area of just the face of the FCA element, the estimated time to dissolve the cladding on the FCA plate is about 176 days. This hypothetical case is presented to show how long the dissolution of the stainless steel could take at the tested conditions. Because this is an unacceptably lengthy process cycle time, it is recommended that chemical dissolution of the FCA fuel be not pursued. Alternate dissolution strategies were considered, but the rapid degradation of the dissolution rate and criticality constraints prevent them from being viable approaches.

The dissolution studies with several FCA simulant elements and stainless steel plates spaced 0.5 inch apart showed no significant difference between the dissolution rates of the individual plates due to spacing. Therefore, spacing of 0.5 inches between the FCA fuel elements in a rack should not produce non-uniform dissolution rates.

The simulant FCA cladding or stainless steel plates produced no measurable H_2 offgas during dissolution and so do not have an impact on 60% of the lower flammability limit (LFL) for H_2. The results from these studies confirm that the FCA stainless steel cladding does not contribute to the flammable off gas generation. The dissolution of Pu metal has been examined in prior work.[1,2] The data are relevant although not necessarily identical to the data that would be obtained for Pu-10.6Al. These prior studies showed that for a charge of 46 kg of Pu with a geometry of thin sheets that the generated H_2 was well below the 60% of the lower flammability limit (LFL) with the standard 40 scfm of air for the H-Canyon dissolver.

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LIST OF ABBREVIATIONS

DOE	Department of Energy
FCA	Fast Critical Assembly
FSF	FCA Simulant Fuel
ICPES	inductively-coupled plasma emission spectroscopy
ID	Internal Diameter
JAEA	Japan Atomic Energy Agency
LFL	lower flammability limit
NNSA	National Nuclear Security Administration
OD	Outer Diameter
R&D	research and development
SA	Surface Area
SNF	spent nuclear fuel
SRNL	Savannah River National Laboratory
SRNS	Savannah River Nuclear Solutions
SRS	Savannah River Site
SSP	Stainless Steel Plate

1.0 Introduction

The Savannah River Site (SRS) has received some fast critical assembly (FCA) fuel from the Japan Atomic Energy Agency (JAEA) for disposition. Among the JAEA FCA fuel are approximately 7090 rectangular Stainless Steel clad elements. Each element has an internal Pu-10.6Al alloy metal wafer. The thickness of each element is either 1/16 inch or 1/32 inch. The dimensions of each element ranges from 2 inches x 1 inch to 2 inches x 4 inches.[3]

Savannah River Nuclear Solutions (SRNS) recently completed a Feasibility Study to identify and evaluate options for the disposal of the JAEA FCA fuel using existing technologies.[4] The study was initiated by SRNS at the request of National Nuclear Security Administration (NNSA). Ultimately, the study will provide NNSA with sufficient information to determine the best disposition pathway for the JAEA FCA fuel. To provide a stronger basis for recommending disposition pathways, additional technology and flowsheet development were requested.

The Savannah River National Laboratory (SRNL) was asked to advance the technical maturity of four processing options for the FCA fuel elements.[5,6,7] The four options are 1) oxidation of declad material, 2) chemical dissolution of clad material, 3) electrolytic dissolution of clad material, and 4) actinide alloying of clad material. The SRNL scope is considered non-baseline technical activities. If successful, the tasks proposed by SRNL will demonstrate sufficient technical maturity to support program evaluation and provide input for a future Analysis of Alternatives. Successful completion will also provide sufficient information to estimate production rates for facilities.

This report examines the chemical dissolution of the FCA clad material or stainless steel to estimate dissolution rates and times as well as H_2 generation rates. This technology uses HNO_3-KF flowsheets in H-Canyon to dissolve the FCA cladding and Pu-10.6Al plates from a rack of materials. Historically, dissolution flowsheets have aimed to maximize Pu dissolution rates while minimizing stainless steel dissolution (corrosion) rates.[8] Because the FCA cladding is made of stainless steel, part of the flowsheet would seek to accelerate stainless steel dissolution. The objective is to demonstrate adequate fuel dissolution (stainless steel and Pu-Al) without the expense and hazards associated with a decladding operation. Discussions with the H-area engineering personnel defined “adequate” as a nominal dissolution time of about 30 days.

The generation of H_2 in the offgas during the dissolution of the FCA fuel elements must be shown to be below 60% of the lower flammability limit (LFL) for H_2.[9] It is not expected that the FCA cladding will produce a significant concentration of H_2 in the offgas. The Pu between the two stainless steel plates is also not expected to generate enough H_2 to exceed the 60% of the lower flammability limit (LFL) as demonstrated in prior work.[1,2] As part of this task, SRNL characterized the offgas from the dissolution of FCA cladding (stainless steel) and simulated elements.

2.0 Experimental Procedure

2.1 Surrogate Materials

The fabrication drawings for the FCA fuel do not explicitly identify the stainless steel alloy used. However, an analysis of the FCA stainless steel alloy was provided.[3] As discussed and laid out in the TTQAP[6], the measured composition of the FCA alloy closely resembles that of Alloy 321. Although the FCA cladding resembles 321 stainless steel, the differences between 321 and 304L are minimal such that it was decided that 304L would be used as the simulant FCA alloy for these experiments.[6] Table 2-1 provides a comparison of the FCA material specification, FCA material certification, and the specifications for 321 and 304L stainless steels. Based on the industry standard PREN or Pitting Resistance Equivalent Number [PREN = % Cr + 3.3* % Mo + 16* %N], corrosion resistance depends strongly on the Cr concentration which is similar between the FCA analysis, 304L and 321 alloy.[10]

Table 2-1. FCA Material Certification and Other Stainless Steel Alloys

Element	Specification	Certification	304L Spec	321 Spec
C	Not more than 0.15%	0.050	0.03 max	0.08 max
Si	Not less than 0.20%	0.73	0.75 max	1.0 max
Mn	Not more than 2.00%	1.58	2.00 max	2.00 max
Ni	Not less than 7.0%, not more than 12.0%	10.45	8.0-12.0	9.0-12.0
Cr	Not less than 17.0% not more than 20.0%	17.68	18.0-20.0	17.0-19.0
S	Not more than 0.045%	0.009	0.030 max	0.030 max
P	Not more than 0.045%	0.023	0.045 max	0.045 max
Ti	Not less than four times the carbon content	0.60	n/a	5X C min
Nb	Not less than eight times the carbon content	<detect	n/a	n/a
Fe	Balance	Balance	Balance	Balance

2.2 Dissolution Experiments for Optimal Conditions and Offgas Characterization

Seven 8-hour dissolutions with 304L stainless steel plates (Table 2-2) were performed to evaluate the best conditions for simulant element dissolutions. The impact of HNO_3, F^-, and Al concentrations on dissolution rate were evaluated in these experiments. Of these initial experiments, Experiments 113, 114, 115, and 117 were chosen to perform offgas rate and composition analyses to examine the impact of the stainless steel dissolution on offgas generation. The generation of H_2 during the dissolution of the FCA cladding and plates in an H-Canyon dissolver must be demonstrated to not violate the less than 60% of the LFL operating requirement.[9] A summary of the dissolution conditions and evaluation goals for the experiments are provided in Table 2-2. All experiments were performed at 102°C, which is sub-boiling for all the solutions.

Table 2-2. Initial Dissolution Experiments

Exp[(1)]	Coupon	HNO_3 (M)	F (M)	B (g/L)	Al(M)
113	1	7	0.25	1.2	0
114	2	10	0.1	1.2	0
115	3	4	0.4	1.2	0
116	4	7	0.1	1.2	0
117	5	10	0.4	1.2	0
119	6	4	0.25	1.2	0
120	7	4	0.25	1.2	0.0074

(1) Experimental numbering sequence corresponds to data recording practices

2.2.1 Preparation of Coupons

The 304L Stainless steel coupons were cut by the SRNL Machine Shop from larger stock. The coupons were then weighed and measured. The initial characteristics of the coupons are shown in Table 2-3.

Table 2-3. Stainless Steel Coupon Characteristics

Exp. No.(1)	Mass (g)	Length (mm)	Width (mm)	Thickness (mm)	Surface Area(1) (cm^2)
113	11.087	38.08	25.28	1.47	21.116
114	11.093	38.09	25.31	1.47	21.145
115	11.110	38.07	25.33	1.47	21.150
116	11.105	38.08	25.28	1.47	21.116
117	11.106	38.09	25.28	1.47	21.121
119	11.107	38.08	25.28	1.47	21.116
120	11.103	38.09	25.32	1.47	21.153

(1) Surface areas were calculated for full coupon immersion

The masses, dimensions, and surface areas of the coupons used in the experiments are provided in Table 2-3. The surface area calculations for the stainless steel coupons are illustrated by Equation 1,

$$\text{SA}\left(\text{cm}^2\right) = 2 \cdot w(\text{cm}) \cdot \ell(\text{cm}) + 2 \cdot t(\text{cm}) \cdot \ell(\text{cm}) + 2 \cdot w(\text{cm}) \cdot t(\text{cm}) \tag{1}$$

where *SA* is the surface area of the immersed coupon, *w* is the width, ℓ is the length, and *t* is the thickness, of the coupon. The initial surface areas for the coupons used are shown in Table 2-3.

2.2.2 Dissolving System

The vessels and offgas condenser used to perform the stainless steel plate dissolutions were fabricated from borosilicate glass by the SRNL Glass Shop. Even though the system would be exposed to F^- ion in nitric acid solutions, glass was chosen due to its availability, durability, flexibility for different designs, and impermeability for offgas measurement. For the scoping dissolution experiments, a vessel was made from a 300-ml round-bottom flask with an extended bottom to accommodate 250 ml of solution. The 250-ml vessel had penetrations for a condenser, sample port, thermocouple, and gas purge. For the individual FCA simulant fuel element dissolutions, a vessel was made from a 1000-ml beaker to accommodate 700 ml of solution. For the multiple FCA simulant fuel element dissolutions, a vessel was made from a 2000-ml beaker to accommodate 1500 ml of solution. The 1000-ml and 2000-ml vessels had penetrations for a condenser, sample port, and thermocouple since no offgas was going to be measured.

All dissolution vessels were heated and agitated using a hot plate/stirrer with a magnetic stir bar. The stirring emulates the mixing caused by gas sparging done in the H-Canyon dissolver. During the dissolutions, coupons were charged to the vessel in a glass basket suspended by a glass rod or a stainless steel stand which was held in place by a compression fitting. The compression fitting allows adjustment of the basket or stand height during dissolution. The solution temperature was controlled using an external thermocouple monitored by the hot plate. When offgas was being measured, the system was sealed by connecting a manometer (which acts as a pressure relief device and provides a measurement of the pressure in the system) to the offgas sample port above the condenser which then passes through a cell containing a Raman probe and terminates in a bubbler (i.e., beaker containing 700 mL or 3.5 in of deionized water). The bubbler prevents air in-leakage from the vent side of the system. When characterizing offgas, the Raman spectrometer was used to measure non-condensable gases such as H_2, N_2, O_2, Ar, NO, N_2O and NO_2 in real time during the experiment. A photograph of the dissolution equipment for offgas measurement is shown in Figure 2-1.

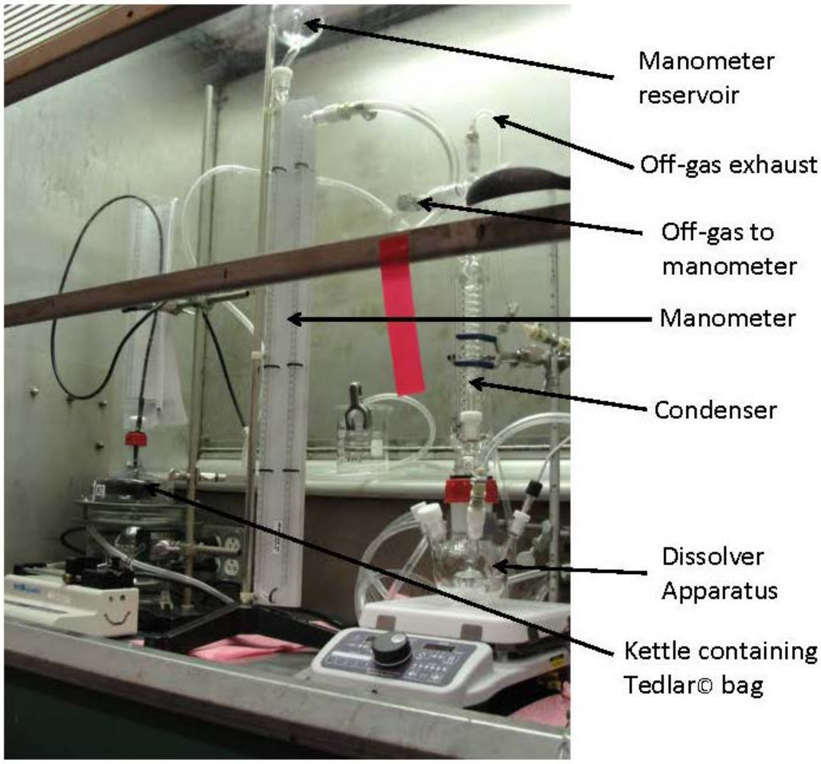


Figure 2-1. Dissolver Setup with Online Raman Offgas Analyzers

2.2.2.1 Raman Spectrometer

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 measures the concentration of gases in the offgas stream. The Raman spectrometer was calibrated using the standard gases shown in Table 2-4. The Raman spectrometer measures the concentrations of the offgas species approximately every 12-13 seconds. Since the Raman spectrometer 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 spectrometer was controlled by and data was logged using a computer running EZRamanReader v8.3.9 software and an Excel spreadsheet.

Table 2-4. Calibration Gases for Raman Analyzer

Supplier	**Gas**	**Ar**	**N_2**	**N_2O**	**NO_2**	**NO**	**O_2**	**H_2**
–	–	(%)	(%)	(%)	(%)	(%)	(%)	(%)
Air Liquide	20% N_2O-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(1)	99.9	–	–	–	–	–	–
SRNL	N_2(1)	–	99.9	–	–	–	–	–
SRNL	Air(1)	0.94	78.03	–	–	–	20.99	–

(1) purity not measured; supplied from SRNL facility gases

To calculate offgas generation rates, a CO_2 tracer gas was metered into the system through a flow controller at a set rate (20 cm^3/min @ 70 °F, 1 atm). The total offgas rate was then calculated by dividing the set input rate by the measured CO_2 concentration in the offgas.

Calibration of the Raman spectrometer and treatment of the data is discussed in Appendix A.

2.2.3 Initial Dissolution Experiments

Prior to performing a dissolution experiment to measure offgas, the dissolving system was checked for leaks by connecting a Tedlar® bag inside a glass kettle filled with water (Figure 2-1) to the dissolver and adding sufficient Ar, N_2, or CO_2 to the bag to generate a column of water 18-28 cm tall. The Tedlar® bag system was then closed and monitored for any observable decrease in the water column height over 2-3 min. After the system integrity was confirmed, the experiment was started.

To perform the dissolutions, a metal coupon was initially placed in a perforated glass basket. For these experiments, the basket containing the coupon was lowered until the coupon was completely immersed at room temperature and the solution was then heated to 102 °C. Chilled water (at 6 °C) was circulated through the condenser during the dissolutions to remove water vapor from the offgas stream before the gas flowed through the Raman cell. For these initial experiments, it took about 30 minutes to heat from room temperature to 102 °C. Once the solution reached 102 °C, the clock was started for sampling at 1, 3, and 8 hours. It was desired that the volume of solution to surface area of the coupon would approximate the volume to surface area ratio in the full-scale dissolver (8.6 mL/cm^2, Appendix B). However, the volume at the lab scale had to be sufficient to fully immerse the coupon. The final volume chosen was 250 mL as it completely immersed the coupon. For Experiment 120, 240 mL was chosen as the amount of solution available was limited and 240 mL was sufficient to cover the coupon. For these chosen volumes, the volume to surface area in the lab scale equipment was 11.3-11.8 mL/cm^2 (Table 2-5).

Table 2-5. Dissolving Solution Volume and Composition

Exp. No.	Initial Volume	Surface Area	Vol/SA
–	(mL)	(cm^2)	(mL/cm^2)
113	250	21.116	11.8
114	250	21.145	11.8
115	250	21.150	11.8
116	250	21.116	11.8
117	250	21.121	11.8
119	250	21.116	11.8
120	240	21.153	11.3

During the experiments, samples at dissolution times of 1, 3, and 8 hours were collected for analysis. Samples were submitted to SRNL Analytical Development (AD) to measure the HNO_3 (total and free acid) and metal concentrations in the dissolving solutions. The free acid and metal concentrations were determined by titration and inductively-coupled plasma emission spectroscopy (ICPES), respectively. In addition, the initial and final volumes of the dissolver solutions and masses of the stainless steel coupons were measured.

2.2.4 Prototypical Dissolution Experiments

Simulant fuel elements of similar dimensions as the FCA fuel elements were fabricated with 304L stainless steel face plates welded onto a frame inside of which was a Cu plate (Figure 2-2). The stainless steel faces were 50.8 mm (2") x 50.8 mm (2") x 0.2286 mm (0.009"). The frame dimensions were 50.8 mm (2") x 50.8 mm (2") x 1.54 mm (0.06"). The Cu plate dimensions were 47.5 mm (1.87") x 47.5 mm (1.87") x 0.8 mm (0.031"). The total dimensions of the FCA simulant fuel elements assembled were 50.8 mm (2") x 50.8 mm (2") x 2.00 mm (0.079") giving a surface area of 55.7 cm^2.

These simulant FCA fuel elements were used in Experiment 121 to examine how long it takes the stainless steel face of the FCA element to breach thus exposing the Cu plate inside. Cu was chosen for the inner plate as it would be detectable by visual observation (blue color), by ICPES (no other Cu sources), and by sudden weight change in the FCA simulant element (easily dissolvable). Experiment 121 used the optimal conditions found in the initial experiments. At various times in the experiment, samples were pulled for ICPES analyses. At the same time, the coupons were weighed and dimensions measured to calculate a dissolution rate. For Experiment 121, it was desired to maintain the volume to surface area at 9 mL/cm^2. However, to get the FCA simulant fuel element completely immersed in the dissolution reactor with some extra head space, 700 mL of solution was needed. This volume was chosen to ensure the FCA simulant fuel element stayed completely immersed for a long duration (about 5 weeks) allowing for sample volume removal and evaporation losses. However, this solution volume to surface area of the FCA simulant element (700 mL/55.7 cm^2 = 12.6 mL/cm^2) was higher than the desired 9 mL/cm^2. Therefore, a stainless steel plate about half the size of the FCA simulant fuel element with dimensions 50.8 mm (2") x 24.6 mm (1") x 1.46 mm (0.06") was added along with the FCA simulant fuel element to the solution. This stainless steel plate had a surface area of 27.2 cm^2 giving a total solution volume to surface area ratio of 700 mL/82.9 cm^2 = 8.4 for Experiment 121, which is closer to the target of 9 mL/cm^2.

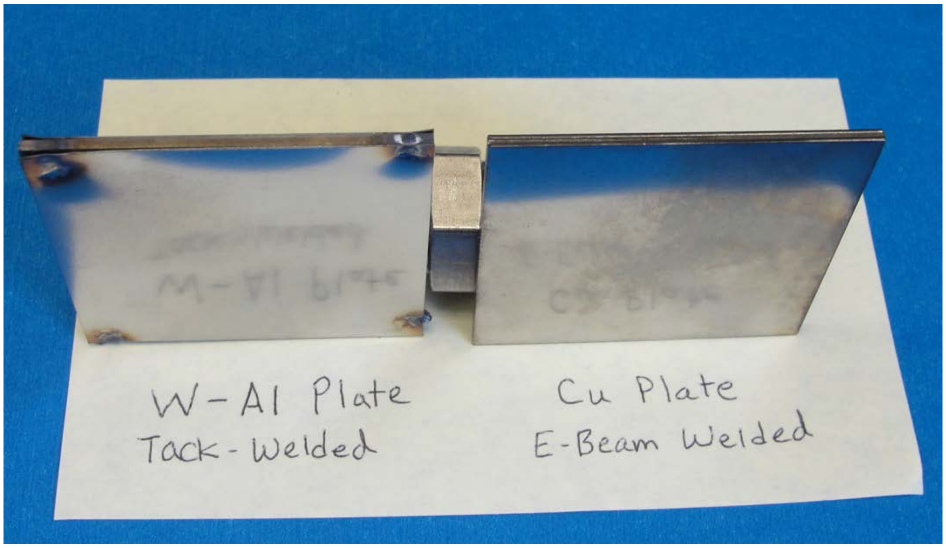


Figure 2-2. FCA Simulant Fuel Elements

Another dissolution experiment (Experiment 122) examined if the spacing of the FCA fuel elements impacted the uniformity of dissolution for multiple elements. Experiment 122 used a stand (Figure 2-3) that had physical characteristics similar to a stand design proposed for the real FCA fuel elements. The stand has support rods that allow adjustment of the perpendicular spacing between the element surfaces to increments of 0.25 inches. Experiment 122 evaluated 0.5-inch spacing. These spacing experiments were performed using the optimal dissolution conditions found in the initial experiments.

The solution volume to cover the elements up to the top of the stand was 1500 ml. At various times in the experiment, samples for ICPES were collected. At the same time, the elements were weighed and their dimensions measured to calculate the dissolution rate. The 1500 ml solution volume allowed some extra head space for sampling and evaporation due to the long duration of the experiment (about 5 weeks).

Due to the additional surface area of the stand (about 284 cm^2), it was decided not to try to match the solution volume to surface area of 9 mL/cm^2. Since the goal of this experiment was to evaluate how spacing affected the dissolution rate uniformity across multiple elements, the volume to surface area did not need to be matched.

Figure 2-3. FCA Simulant Element Stand with FCA Simulant Elements

2.3 <u>Quality Assurance</u>

The 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 <u>Dissolution of Stainless Steel Coupons</u>

Stainless steel coupons were dissolved in seven experiments simulating potential H-Canyon processing scenarios. These seven experiments were used to determine the optimal conditions for dissolving simulant fuel elements. These experiments examined the impact of nitric acid and F^- concentration on the overall dissolution rate. The HNO_3 was varied at 4, 7, and 10M and F^- varied at 0.1, 0.25, and 0.4M as shown in Table 2-2. The Boron concentration was 1.2 g/L for all the experiments. Free and total acid analyses were performed on the initial solutions for the various experiments (Table 3-1) and indicated that the target values were achieved within analytical uncertainty (1σ of 10%).

Table 3-1. Total and Free Acid Analyses for Scoping Experiments

Experiment (HNO_3 Target)	Total Acid (M)	Free Acid (M)	Experiment (HNO_3 Target)	Total Acid (M)	Free Acid (M)
113 (7M)	6.29	6.19	117 (10M)	---	9.4
114 (10M)	10.2	9.76	119 (4M)	---	3.75
115 (4M)	3.96	3.9	120 (4M)	---	3.71
116 (7M)	---	7.05	---	---	---

3.1.1 Rates of Dissolution

Time zero for the dissolution of the stainless steel plates was when the solution reached 102°C after the samples had been lowered into the solution. The stainless steel material in the various experiments had different dissolution rates due to the different HNO_3 and F^- levels. The overall dissolution rates after eight hours are shown in Table 3-2. Based on these initial studies, the conditions that gave the highest dissolution rate of 0.02 mg/min/cm^2 were 4M HNO_3 and 0.4M KF. However, based on H-Canyon processing constraints where KF is limited to 0.23M, the highest practical rate of 0.012 mg/min/cm^2 was achieved with 4M HNO_3 and 0.25M KF. The lower acid may help with dissolution by not tying up the F^- ion as much as the higher HNO_3 concentrations.

Table 3-2. Average Stainless Steel Dissolution Rates for 8 Hours at 102°C

Experiment*	HNO_3 (M)	F^- (M)	Final Mass (g)	Length (mm)	Width (mm)	Depth (mm)	Final SA (cm^2)	Dissolution Rate (mg/(cm^2*min))
113	7	0.25	11.007	38.07	25.28	1.46	21.098	0.008
114	10	0.1	11.052	38.09	25.30	1.46	21.125	0.004
115	4	0.4	10.905	38.06	25.30	1.45	21.096	0.020
116	7	0.1	11.057	38.08	25.27	1.46	21.095	0.005
117	10	0.4	10.960	38.09	25.28	1.47	21.121	0.014
119	4	0.25	10.985	38.07	25.27	1.47	21.103	0.012
120	4	0.25	10.996	38.09	25.31	1.46	21.132	0.011

*All these experiment solutions included 1.2 g/L B

The main constituents of the 304L stainless steel are Fe (66.9 wt%), Cr (19.5 wt%), Ni (10.5 wt%), Mn (2.0 wt%), and Si (1.0 wt%). Potential reactions for the Fe, Cr, and Ni species in nitric acid are:

$$Fe + 6HNO_3 \rightarrow Fe(NO_3)_3 + 3H_2O + 3NO_2 \qquad (2)$$

$$Cr + 6HNO_3 \rightarrow Cr(NO_3)_3 + 3H_2O + 3NO_2 \qquad (3)$$

$$Ni + 4HNO_3 \rightarrow Ni(NO_3)_2 + 2H_2O + 2NO_2 \qquad (4)$$

Based on the ICPES analyses for Fe concentration versus dissolution time for the initial experiments (Figure 3-1), Experiment 115 has the fastest dissolution rate (highest slope) with 4M HNO_3 and 0.4M KF, followed by Experiment 117 with 10M HNO_3 and 0.4M KF, followed by Experiment 119 and 120 with 4M HNO_3 and 0.4M F. This ICPES dissolution data tracks with the mass dissolution data. A complete list of the ICPES results from the initial experiments is in Appendix C.

Based on the dissolution data from the initial experiments, the dissolutions with simulant fuel elements (Experiments 121 and 122) used 4M HNO_3 and 0.25M KF.

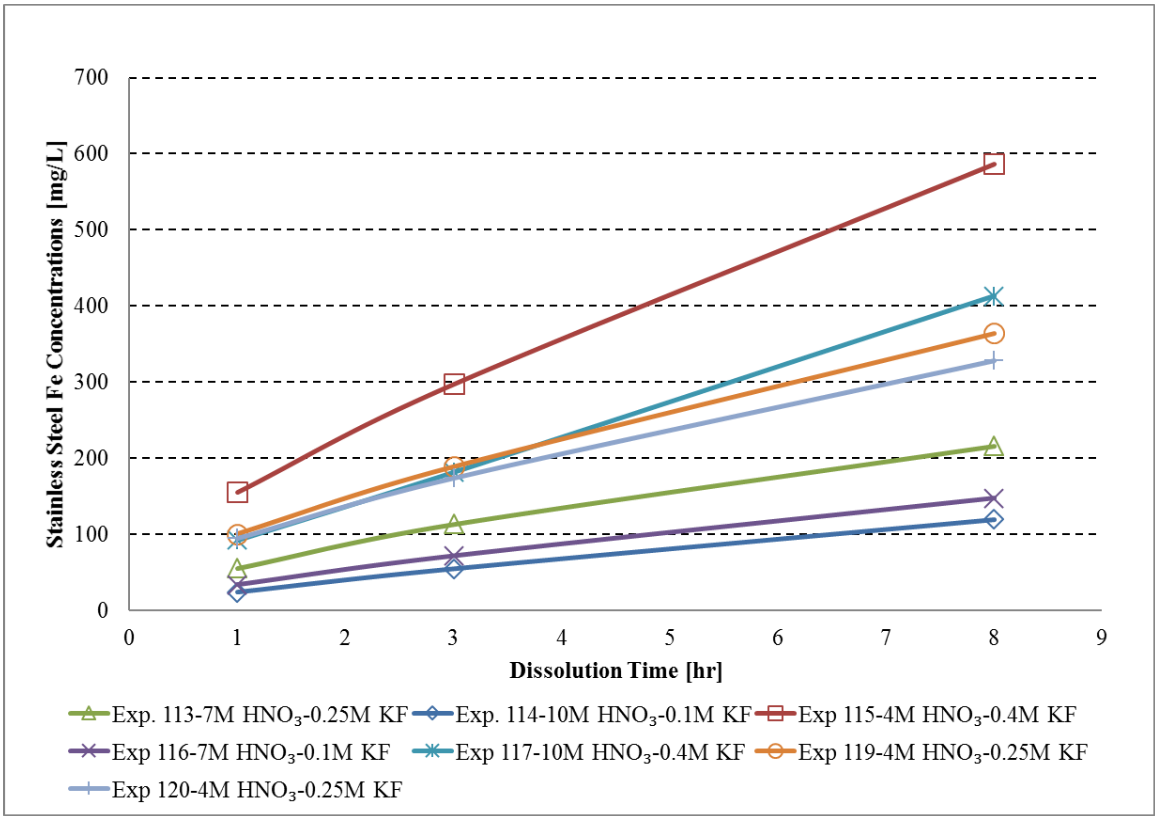


Figure 3-1. Stainless Steel Fe Concentrations for Scoping Experiments

The total offgas generation rate curves (which illustrate the extent of dissolution) for Experiments 113, 114, 115, and 117 are plotted as a function of time in Figure 3-2. The offgas data for Experiment 113, 114, 115, and 117 are listed in Appendix D, E, F, and G, respectively. Peak offgas generation for the stainless steel dissolution Experiments 113, 114, 115, and 117 was approximately 0.04 cc/min/cm^2 with the average offgas rate being between 0.02 and 0.03 cc/min/cm^2. This is a very low total offgas generation when compared to typical Al-1100 dissolution as shown in Figure 3-3 which has a peak total offgas generation ~29 cc/min/cm^2. Based on the peak offgas generation, the stainless steel dissolution is about 725 times slower than the Al-1100 dissolution at peak offgas conditions. The overall dissolution rate of Al 1100 alloy is approximately 19 mg/min/cm^2 (based on Experiment 106) starting with a 7M HNO_3 solution and adding 0.002M Hg at boiling.[11] The highest dissolution rate for the stainless steel was 0.02 mg/min/cm^2. Based on the overall dissolution rate, the stainless steel is approximately 950 times slower than the Al-1100, which tracks with the comparison based on offgas generation.

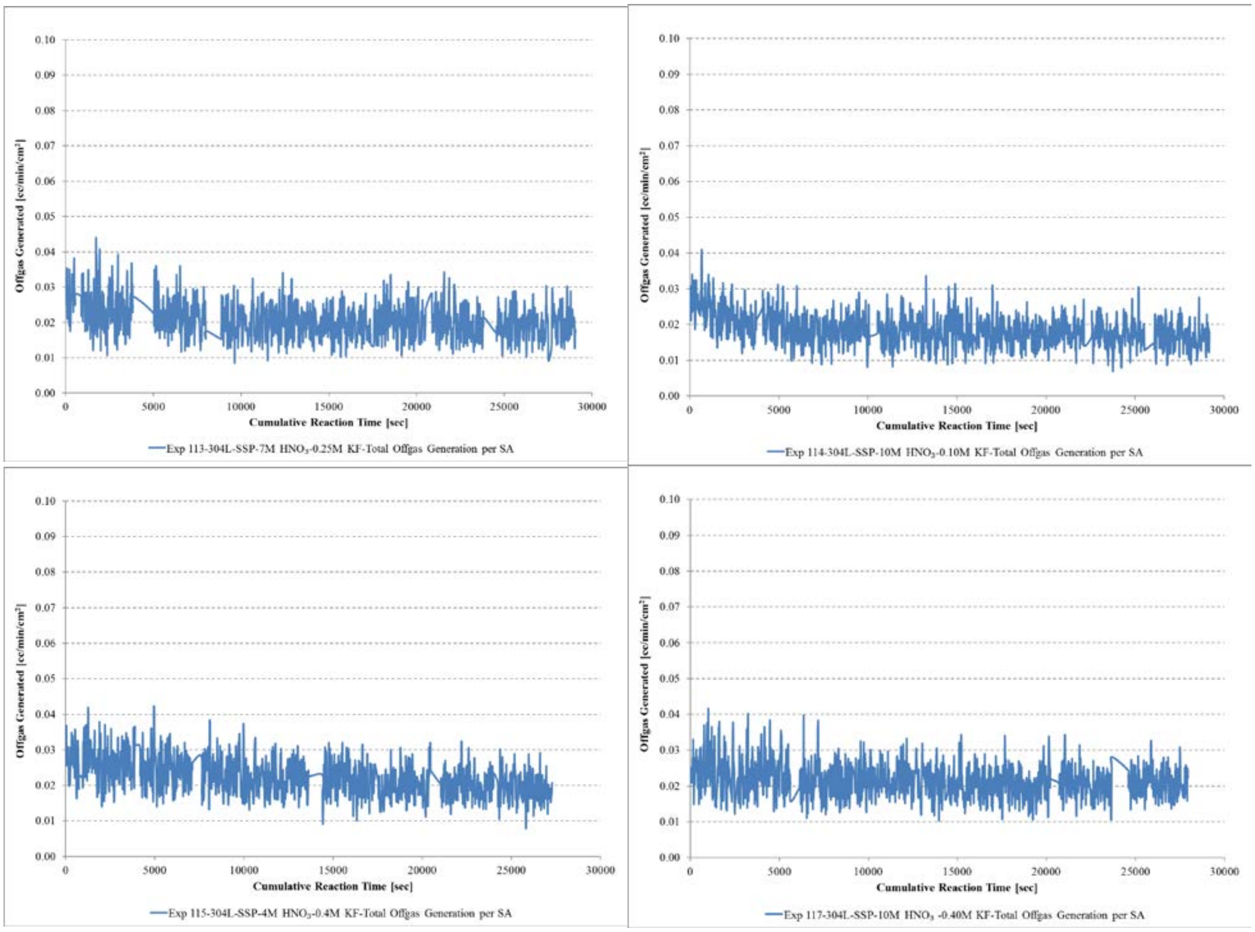


Figure 3-2. Total Offgas Generation Rates for Experiment 113, 114, 115, and 117

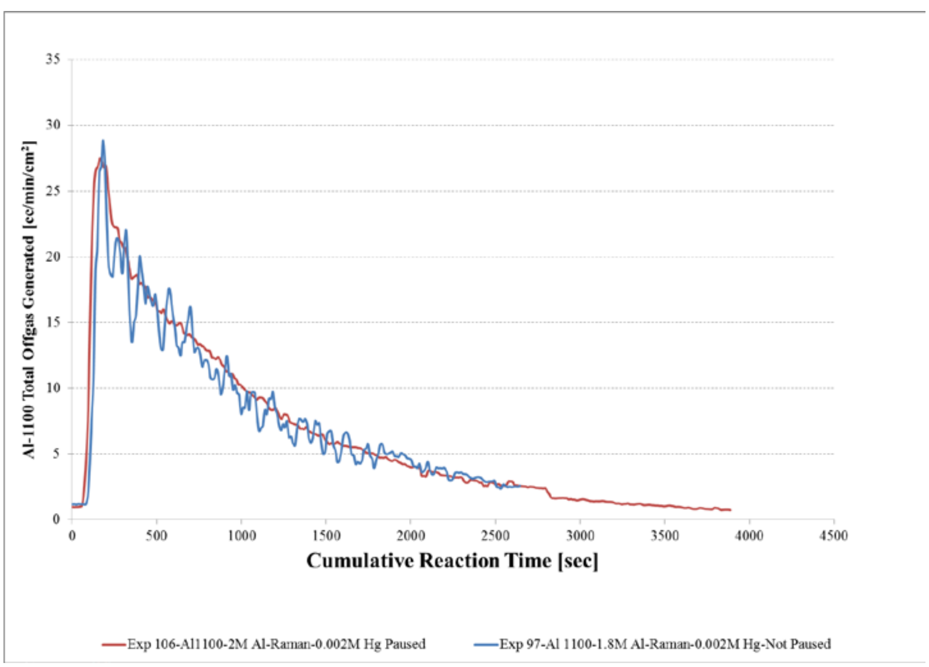


Figure 3-3. Typical Total Offgas Generation Rates for Al-1100 Dissolution[11]

3.1.2 Offgas Characterization

The dissolution of the FCA stainless steel coupons produces low offgas and as a result the H_2 generation rate is inconsequential. Figure 3-4 shows the H_2 generation rates during the dissolution of stainless steel plates (Experiments 113, 114, 115, and 117). In these experiments, stainless steel plates (304 L) were dissolved in 4-10M HNO_3, 0.1-0.4M KF, and 1.2 g /L B. Figure 3-4 shows that very low H_2 is generated by the stainless steel plates (or essentially zero H_2 for the Raman's sensitivity. The conclusion is that dissolution of the FCA element cladding has no flammability concerns.

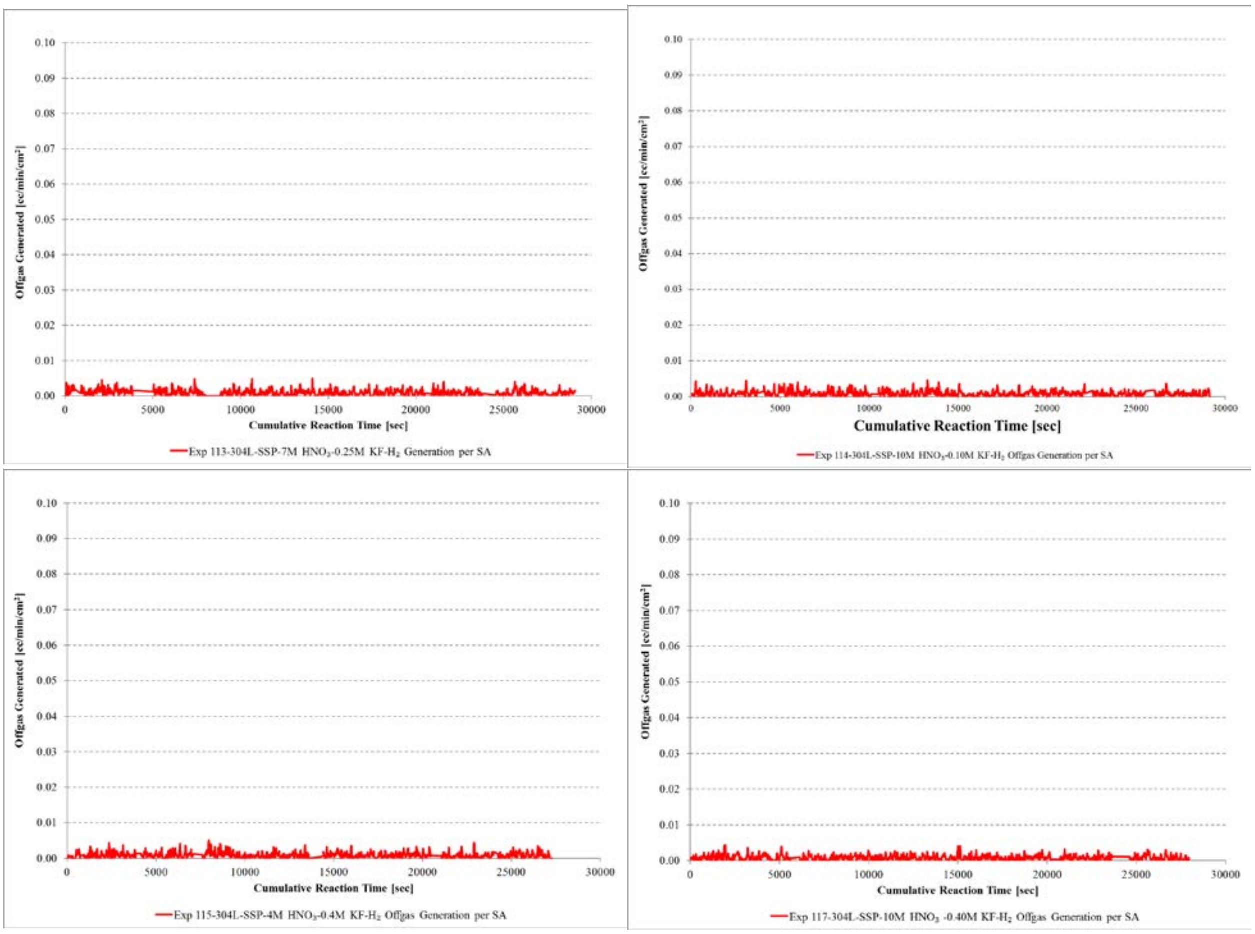


Figure 3-4. H_2 Offgas Generation Rates for Experiment 113, 114, 115, and 117

3.2 <u>Dissolution of FCA Simulant fuel elements</u>

The stainless steel faces on the FCA simulant fuel element have dimensions of 50.8 mm (2") x 50.8 mm (2") x 0.2286 mm (0.009"), giving the element a volume of 0.590 cm^3 and surface area of 26.27 cm^2. Based on the density of 304L Stainless Steel of 8.0 g/cm^3, the stainless steel face has an approximate mass of 4.719 g (4.628 g and 4.640 g have been measured for similar pieces). Based on the mass and surface area of the stainless steel face and the dissolution rate from Experiments 119 and 120 of 0.012 mg/(cm^2*min) for optimal operating conditions of 4M HNO_3 and 0.25M KF, it was estimated that the stainless steel face of the FCA simulant fuel element would breach around 10.4 days. However, that calculation assumes a linear dissolution rate for the duration of the scoping experiments.

To validate this duration, the ICPES analyses from Experiments 119 and 120 with like conditions were examined (Table 3-3). The main constituents of the 304L stainless steel are Fe (66.9 wt%), Cr (19.5 wt%), Ni (10.5 wt%), Mn (2.0 wt%), and Si (1.0 wt%). Using these weight percent values, an assumed dissolution rate of 0.012 mg/(cm^2*min), and other factors in Table 3-4, estimated concentrations of the various species from the stainless steel coupons were computed (Table 3-4). The measured and estimated Fe concentrations (major constituent) versus time are shown in Figure 3-5. Fitting a line to these concentrations versus time and then comparing the slopes provides a comparison of the overall dissolution rates. Based on Experiment 119, the measured dissolution rate was 11% slower than the estimated rate. Based on Experiment 120, the measured dissolution rate was 30% slower than the estimated rate. The ICPES Fe data in Figure 3-5 has a slight curvature so a power law relation was fitted. Power law fits of the Fe concentrations over dissolution time for Experiment 119 and 120 are (Figure 3-6):

$$\text{Exp 119 Fe Conc} = 99.389 * \text{Time}^{0.621} \tag{5}$$

$$\text{Exp 120 Fe Conc} = 93.683 * \text{Time}^{0.600} \tag{6}$$

where the *Fe Conc* is the Fe concentration in mg/L and *Time* is the total dissolution time in hours. Based on the stainless steel face plate mass of 4.719 g which is 66.9% Fe, that gives 3.157 g Fe or 3,157 mg Fe to dissolve in 250 ml for Experiment 119 and 240 ml in Experiment 120. The final target concentrations for Fe are 12,628 mg/L for Experiment 119 and 13,154 mg/L for Experiment 120. Solving the power law Fe concentration relations of Experiment 119 (Equation 5) and Experiment 120 (Equation 6) for these concentrations gives between 108 to 158 days to dissolve the stainless steel face plate. These dissolution times reflect the non-linear nature of the dissolution rate with respect to time. Since Mission Development and Support Engineering indicated a maximum-acceptable duration for dissolution to be about 30 days, it was decided to run the long-term experiments for 35 days to try to capture the breaching of the stainless steel face. If the stainless steel face breached before that time, then the experiment would be stopped.

Table 3-3. ICPES Analyses for Experiments 119 and 120

Experiment	119	119	119	120	120	120
Sample Time	1 hr	3 hr	8 hr	1 hr	3 hr	8 hr
HNO_3(M)	4	4	4	4	4	4
F(M)	0.25	0.25	0.25	0.25	0.25	0.25
Al(M)	0	0	0	0.0074	0.0074	0.0074
Sample ID:	LW6930	LW6931	LW6932	LW6934	LW6935	LW6936
Units:	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
Al	8.95	12.6	14.2	209	212	215
B	1060	1110	1120	967	973	979
Cr	23.2	44.4	85.7	21.7	40.7	77.6
Fe	101	190	368	95.2	175	332
K	9070	9190	9220	8740	8690	8710
Mn	1.88	3.69	7.19	1.77	3.36	6.52
Na	24.5	31.8	35.1	20.7	26.9	30.5
Ni	12.1	23.2	45	11.4	21.2	40.8
Si	263	370	424	214	305	323
Solution Volume (ml)	250	247.81	245.67	240	238.07	235.95

Table 3-4. Estimated Solution Concentrations for Experiments 119 and 120

Experiment	119	119	119	120	120	120
Sample Time (hr)	1 hr	3 hr	8 hr	1 hr	3 hr	8 hr
Sample Time (min)	60	180	480	60	180	480
Plate Mass (g)	11.107	---	10.985	11.103	---	10.996
Surface Area (cm^2)	21.116	21.116	21.116	21.153	21.153	21.153
Overall Dissolution rate (mg/(cm^2*min))	0.012	0.012	0.012	0.012	0.012	0.012
Est. mass dissolved using overall diss. rate (mg)	15.250	45.750	122.000	15.277	45.830	122.214
Est. max dissolution concentration (mg/L)	---	---	---	---	---	---
est Cr	12	36	97	12	38	101
est Fe	41	123	332	43	129	346
est Mn	1	4	10	1	4	10
est Ni	6	19	52	7	20	54
est Si	1	2	5	1	2	5

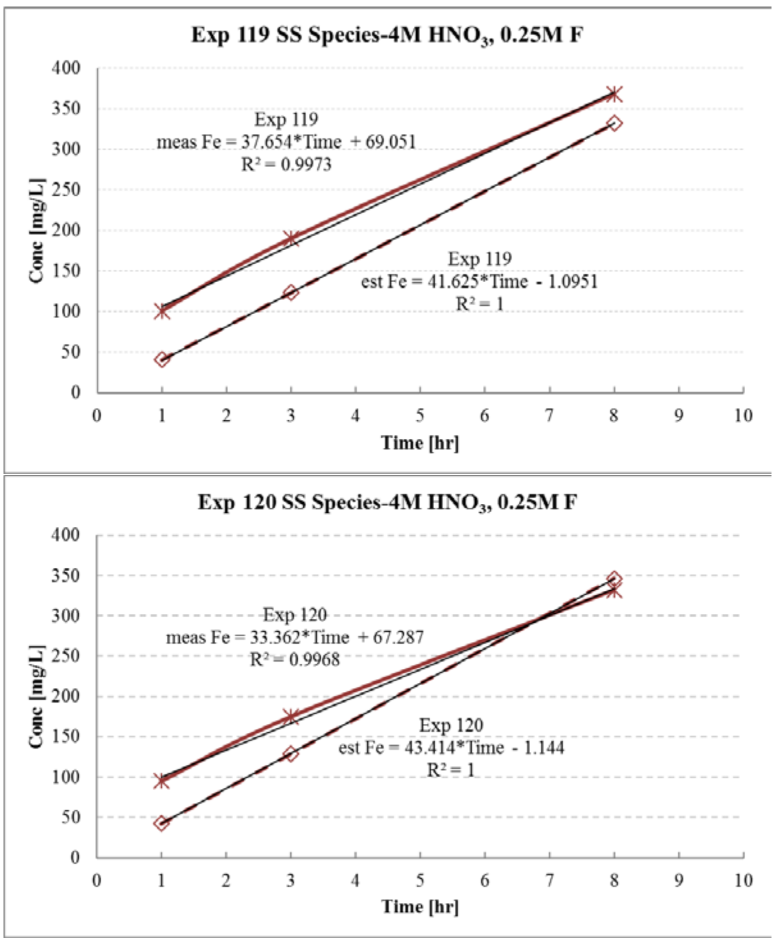


Figure 3-5. Linear Fits of Fe Concentrations for Experiment 119 and 120

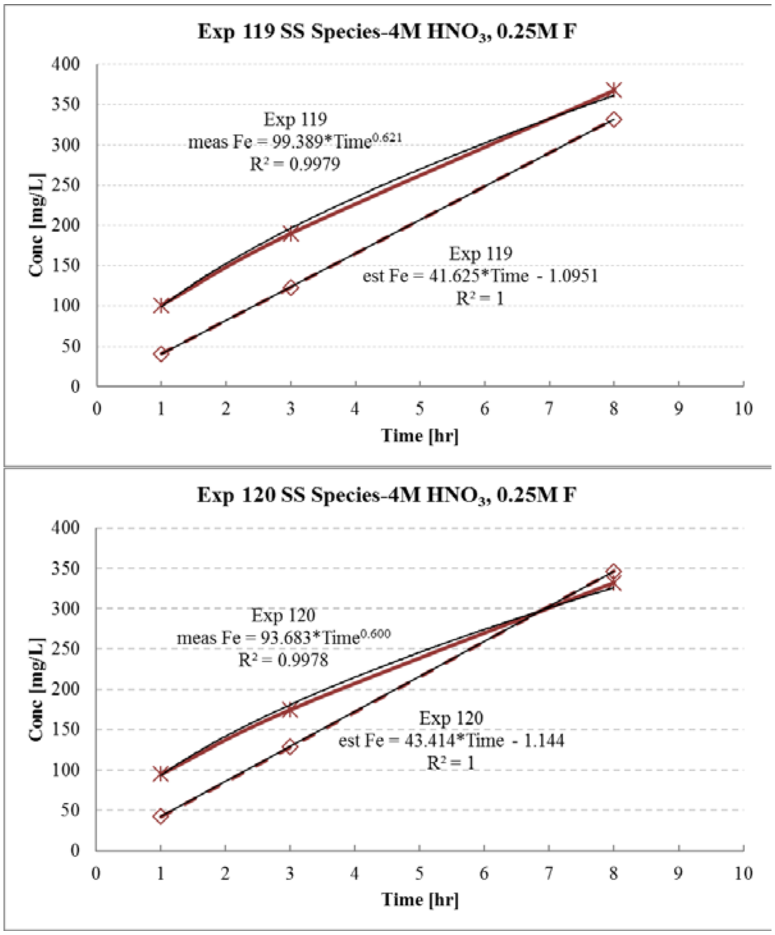


Figure 3-6. Power Fits of Measured Fe Concentrations for Experiment 119 and 120

3.2.1 Single FCA Simulant Fuel Element Dissolution

Experiment 121 was performed to examine how the FCA simulant fuel element dissolved under optimal conditions established in Section 3.1. At various times, samples were pulled for ICPES analyses to track the metal concentrations and for free/total acid analyses in solution to check the acid concentrations. While sampling, the basket with the FCA simulant fuel element and a stainless steel plate was removed from solution to weigh the coupons and measure their dimensions.

The ICPES and free/total acid results for samples obtained during the single FCA simulant fuel element experiments are shown in Table 3-5. The total acid analysis shows that the starting acid concentration is within analytical uncertainty. The free acid analyses show that little acid is consumed by the middle sample (18,679 min), but by the end sample the free acid is down to about 2.4M. This large consumption of acid most likely occurred during the dissolution of the Cu plate:

$$Cu + 4\ HNO_3 \rightarrow Cu(NO_3)_2 + 2\ H_2O + 2\ NO_2 \qquad (7)$$

Looking at the change in the Cu concentration over time (Figure 3-7), there is some Cu in solution at 6 days which steadily grows until 17 days. After 17 days, there is a surge in the Cu concentration which indicates the internal Cu plate has dissolved.

Table 3-5. Total/Free Acid and ICPES Analyses for Experiment 121

Total Time of Dissolution (min)	0	1468	4302	8634	12969	18679	24236	31459	41502	44439
Total Acid (M)	3.81	---	---	---	---	3.74	---	---	---	3.6
Free Acid (M)	3.77	---	---	---	---	3.49	---	---	---	2.35
Free/Total Acid sample ID:	LW7576	---	---	---	---	LW7577	---	---	---	LW7578
ICPES Sample ID:	---	LW7566	LW7567	LW7568	LW7569	LW7570	LW7571	LW7572	LW7573	---
Units:	---	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	---
Al	---	15.6	25.1	40.2	53.3	69.7	82.6	89.5	115	---
B	---	967	1010	1050	1110	1130	1180	1190	1240	---
Cr	---	230	397	580	766	938	1080	1760	1630	---
Cu	---	< 68.4	< 68.4	227	879	1450	2800	31700	23800	
Fe	---	911	1560	2280	2990	3660	4190	6910	6390	---
K	---	8300	8480	8270	8520	8430	8280	11100	8320	---
Mn	---	17.9	31	45.6	58.9	73	85	104	139	---
Na	---	50.6	59	92.2	122	158	187	198	254	---
Ni	---	106	183	269	347	426	507	598	745	---
Si	---	197	175	160	151	134	125	132	122	---
Solution Volume (ml)	700	700	698	695	693	691	684	682	680	678

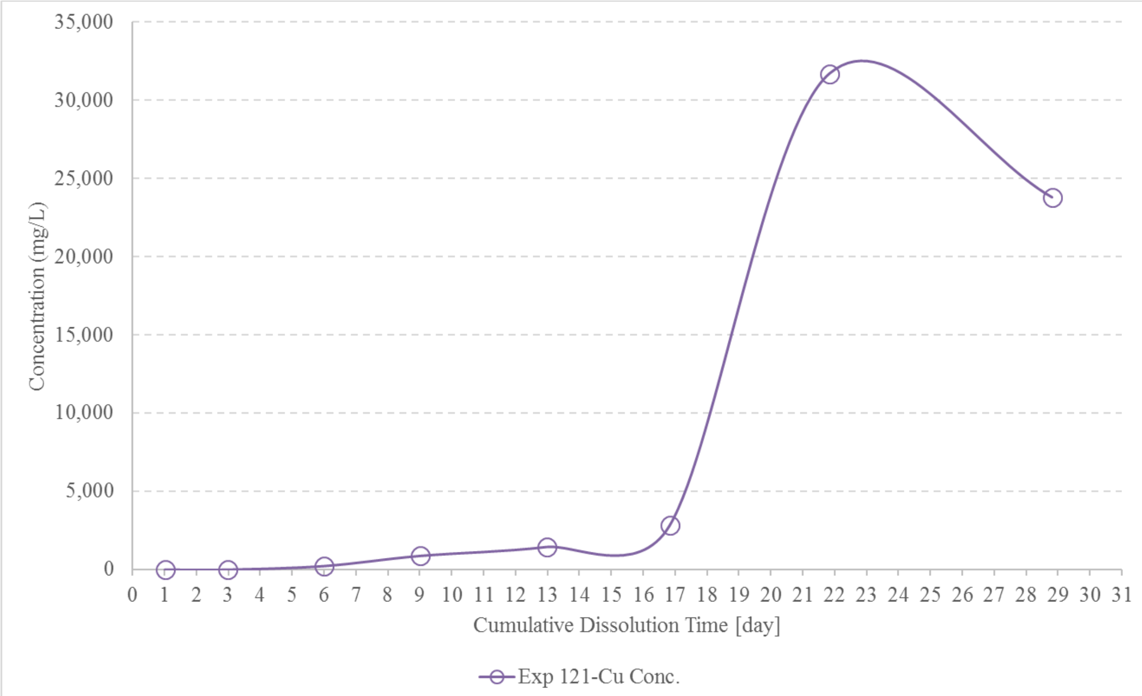


Figure 3-7. Experiment 121 Cu Concentration versus Cumulative Dissolution Time

The coupon mass and dimension data for this experiment are shown in Table 3-6 for the FCA simulant element and in Table 3-7 for the stainless steel plate. The dissolution rates are incremental (from one sample to the next), which gives a more accurate representation of the dissolution rate at that time. The surface area used in the dissolution rate calculation is the average of the initial and final surface areas for that sample. A plot of the incremental dissolutions rates versus cumulative dissolution time are shown in Figure 3-8.

The FCA simulant fuel element first showed signs of a weld failure at 311 hours (18,679 minutes) (Figure 3-9). The dissolution rates show a slight increase for the next sample point at 404 hours (24,236 minutes), which may be due to Cu plate dissolution (Figure 3-10). The weld failure still was not catastrophic, so Cu dissolution was still minor relative to the total weight of the Cu insert. The Cu plate had not dissolved at 404 hours as shown by the small reduction in mass. When the FCA simulant element and stainless steel plate were put back in solution after the 404 hour sampling, a lot of bubbling was observed coming from the FCA simulant fuel element, an indication of substantial weld failure (Figure 3-11). The Cu dissolution was accompanied by the release of significant quantities of orange-brown gas (nitrogen dioxide) in the reaction vessel (Figure 3-12). At this point the Cu plate was dissolving which was confirmed by the sample taken at 524 hours (31,459 minutes) as the dissolution rate jumped up significantly (Figure 3-8). So for the 524 hour sample and those following, the surface area of exposed stainless steel was increased by the inside faces of the front and back face plates of the FCA simulant element. Pictures of the FCA simulant element also showed breaches along the edges (Figure 3-13). At the 692-hour sample point, one stainless steel face had peeled back showing that the internal Cu plate was gone (Figure 3-14). After cleaning the FCA simulant fuel element, the front stainless face had come loose as shown in Figure 3-15.

Table 3-6. Experiment 121 FCA Simulant Fuel Element Sample Data

Sample	Initial Mass (g)	Ending Mass (g)	Mass Loss (g)	% Loss	Total Time of Dissolution (min)	Incremental Time of Dissolution (min)	Initial Length (mm)	Initial Width (mm)	Initial Depth (mm)	Initial SA (cm^2)	Ending Length (mm)	Ending Width (mm)	Ending Depth (mm)	Ending SA (cm^2)	Avg SA (cm^2)	Incremental Dissolution Rate (mg/(cm^2*min))
1	28.718	28.255	0.463	1.6	1468	1468	50.80	50.81	1.96	55.61	50.78	50.75	1.93	55.46	55.53	0.0057
2	28.255	28.042	0.213	0.8	4302	2834	50.78	50.75	1.93	55.46	50.75	50.75	1.95	55.48	55.47	0.0014
3	28.042	27.450	0.592	2.1	8634	4332	50.75	50.75	1.95	55.48	50.70	50.72	1.91	55.32	55.40	0.0025
4	27.450	26.952	0.498	1.8	12969	4335	50.70	50.72	1.91	55.32	50.69	50.70	1.91	55.27	55.29	0.0021
5	26.952	25.940	1.012	3.8	18679	5710	50.69	50.70	1.91	55.27	50.68	50.69	1.92	55.27	55.27	0.0032
6	25.940	24.574	1.366	5.3	24236	5557	50.68	50.69	1.92	55.27	50.65	50.67	1.86	55.10	55.19	0.0045
7	24.574	9.650	14.924	60.7	31459	7223	50.65	50.67	1.86	106.43	50.60	50.65	1.82	106.21	106.32	0.0194
8	9.650	8.692	0.958	9.9	41502	10042	50.60	50.65	1.82	106.21	50.55	50.64	1.74	105.92	106.06	0.0009
9	8.692	8.457	0.235	2.7	44439	2937	50.55	50.64	1.74	105.92	50.54	50.62	1.71	105.78	105.85	0.0008

Table 3-7. Experiment 121 Stainless Steel Plate Sample Data

Sample	Initial Mass (g)	Ending Mass (g)	Mass Loss (g)	% Loss	Total Time of Dissolution (min)	Incremental Time of Dissolution (min)	Initial Length (mm)	Initial Width (mm)	Initial Depth (mm)	Initial SA (cm^2)	Ending Length (mm)	Ending Width (mm)	Ending Depth (mm)	Ending SA (cm^2)	Avg SA (cm^2)	Incremental Dissolution Rate (mg/(cm^2*min))
1	13.995	13.705	0.290	2.1	1468	1468	50.81	24.64	1.46	27.24	50.79	24.64	1.46	27.23	27.24	0.0073
2	13.705	13.502	0.203	1.5	4302	2834	50.79	24.64	1.46	27.23	50.76	24.62	1.36	27.04	27.13	0.0026
3	13.502	13.241	0.261	1.9	8634	4332	50.76	24.62	1.36	27.04	50.74	24.58	1.34	26.97	27.00	0.0022
4	13.241	13.032	0.209	1.6	12969	4335	50.74	24.58	1.34	26.97	50.71	24.57	1.38	26.99	26.98	0.0018
5	13.032	12.787	0.245	1.9	18679	5710	50.71	24.57	1.38	26.99	50.71	24.57	1.38	26.99	26.99	0.0016
6	12.787	12.596	0.191	1.5	24236	5557	50.71	24.57	1.38	26.99	50.67	24.52	1.29	26.79	26.89	0.0013
7	12.596	12.408	0.188	1.5	31459	7223	50.67	24.52	1.29	26.79	50.66	24.49	1.33	26.81	26.80	0.0010
8	12.408	12.185	0.223	1.8	41502	10042	50.66	24.49	1.33	26.81	50.62	24.49	1.31	26.76	26.79	0.0008
9	12.185	12.127	0.058	0.5	44439	2937	50.62	24.49	1.31	26.76	50.61	24.47	1.30	26.72	26.74	0.0007

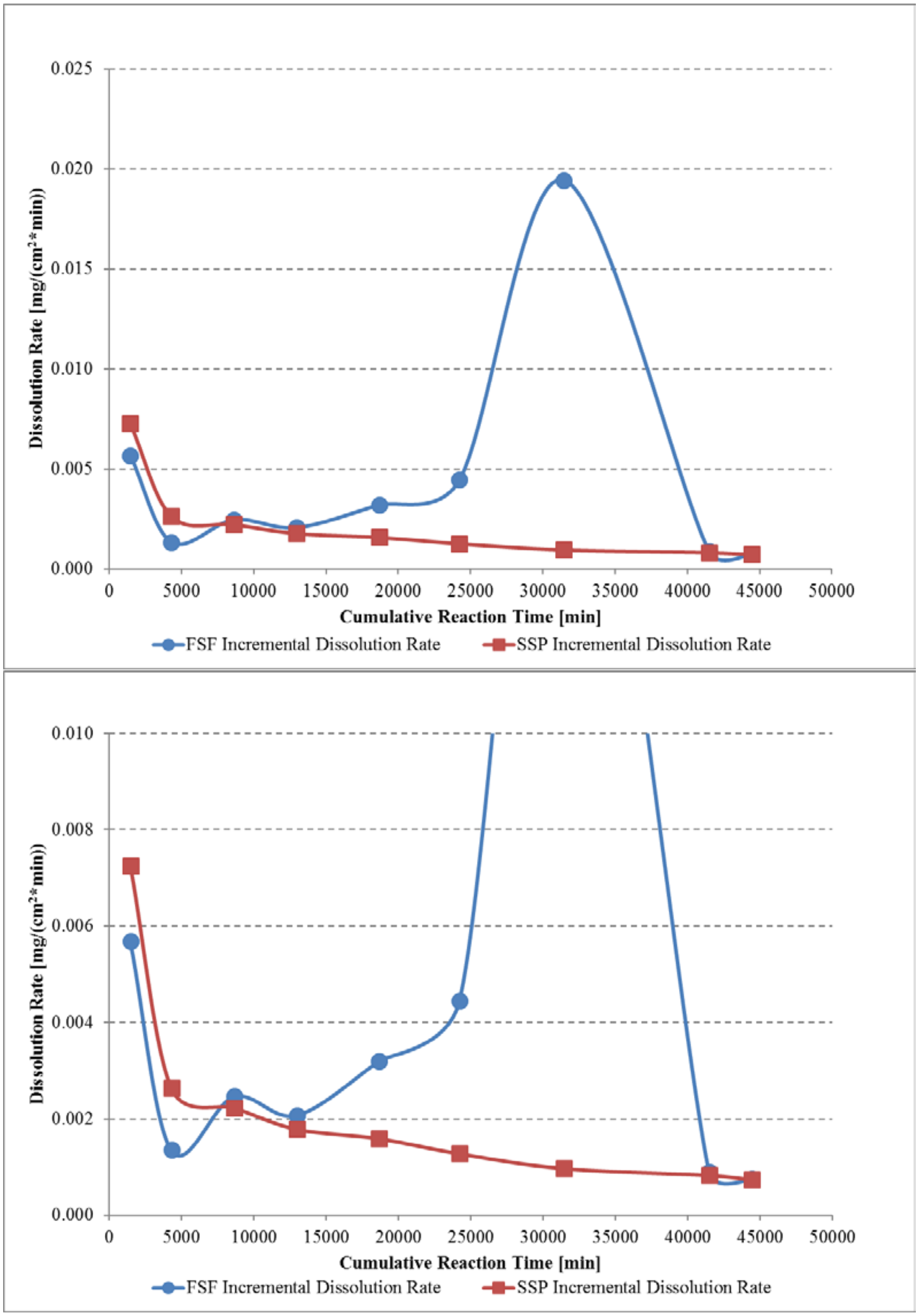


Figure 3-8. Incremental Dissolution Rates Based on Coupon Mass and Dimensions

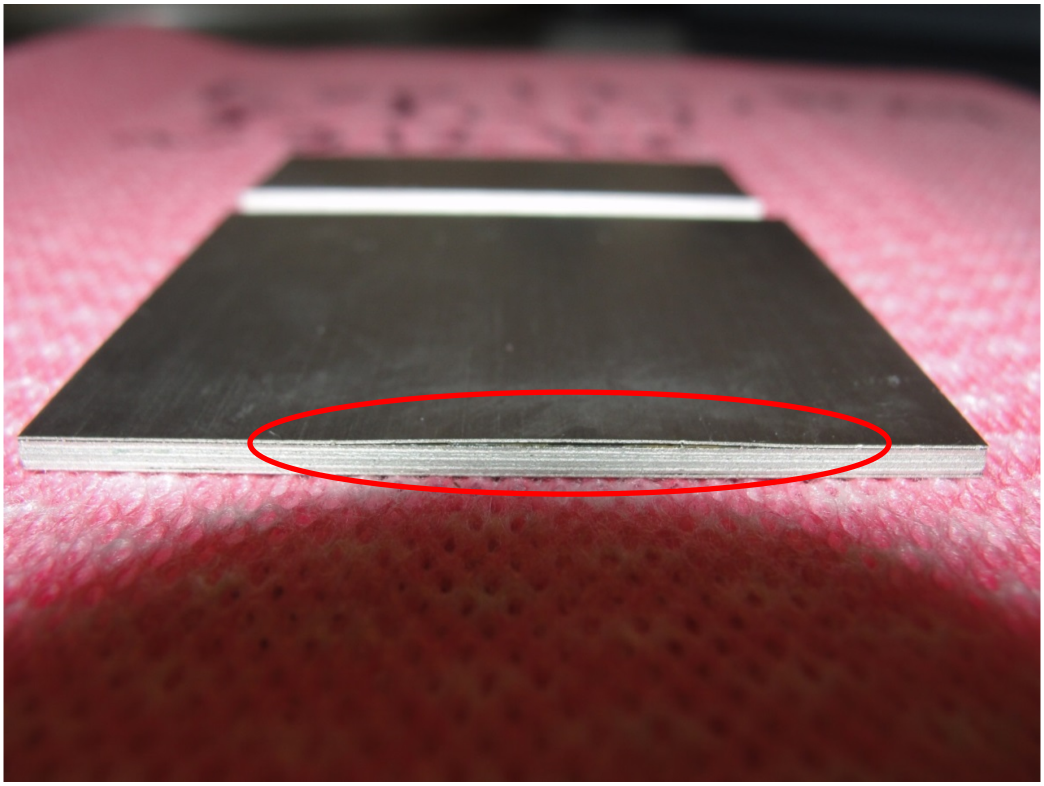


Figure 3-9. Experiment 121 FCA Simulant Element after 311 Hours Showing Weld Breach

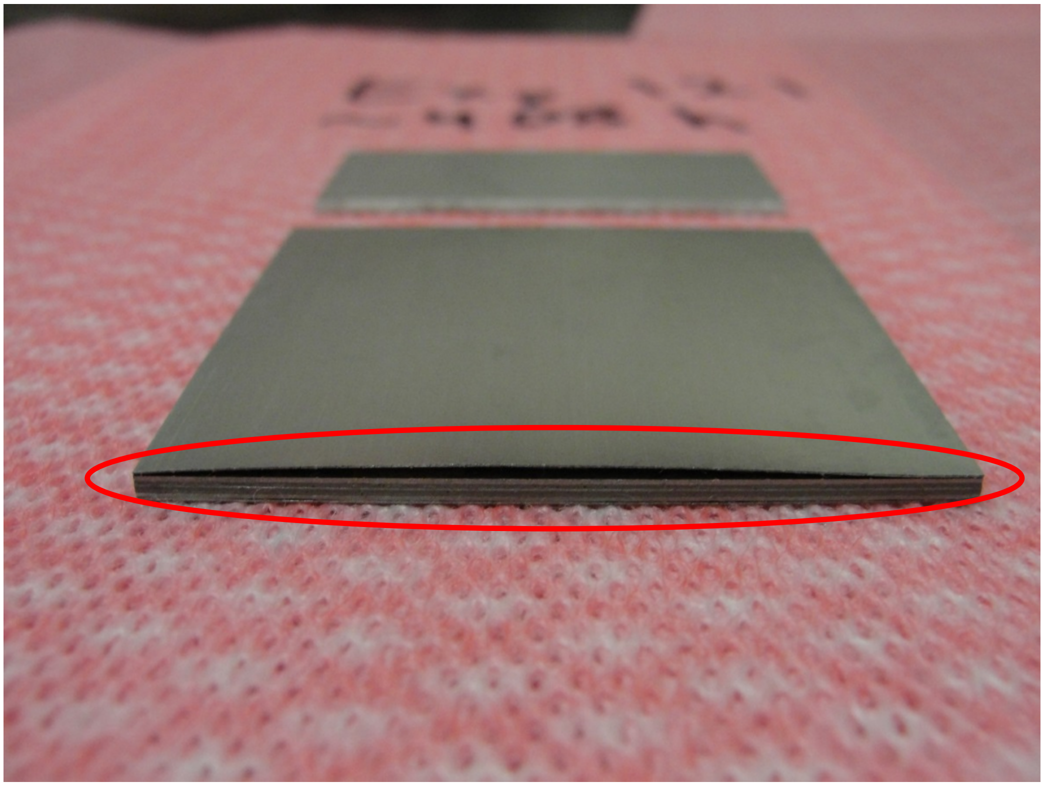


Figure 3-10. Experiment 121 FCA Simulant element after 404 hours Showing Weld Breach

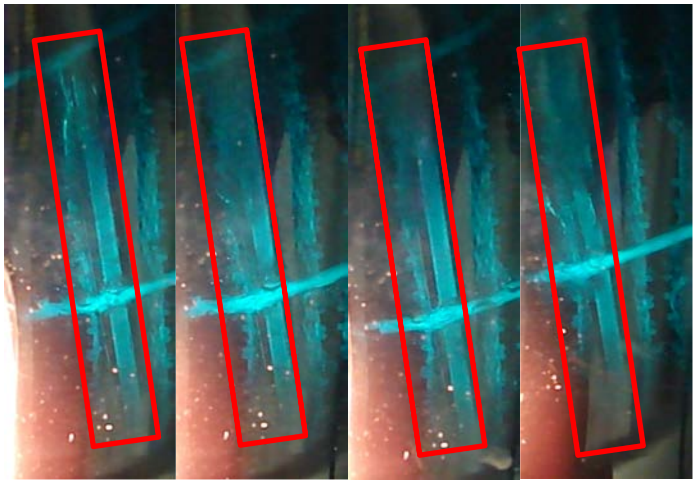


Figure 3-11. Experiment 121 Bubbles from FCA Simulant Element Weld (after 404-hour sample)

Figure 3-12. Experiment 121 Brown Gas in Head Space (after 404-hour sample)

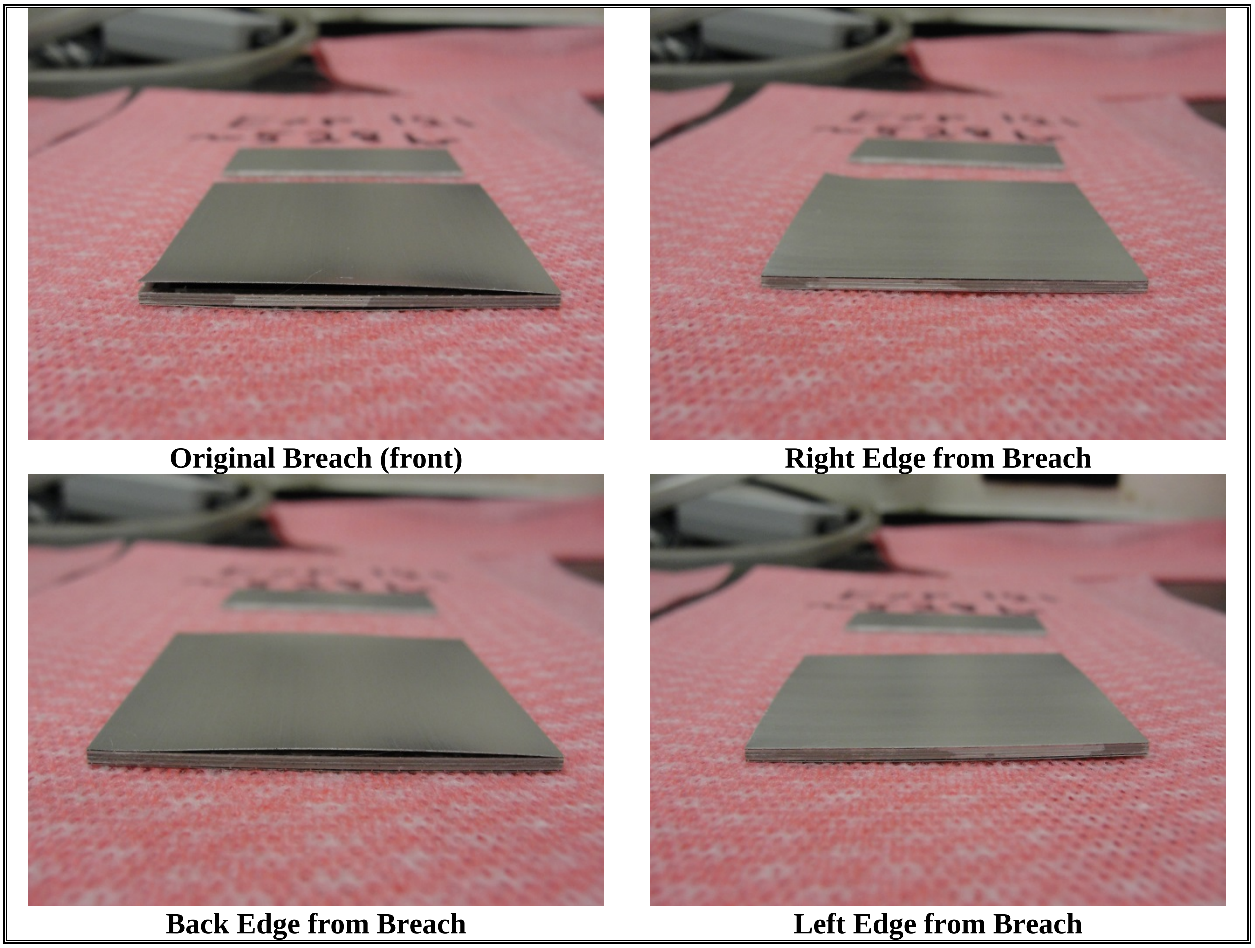


Figure 3-13. Experiment 121 FCA Simulant Element after 524 hours of Dissolution

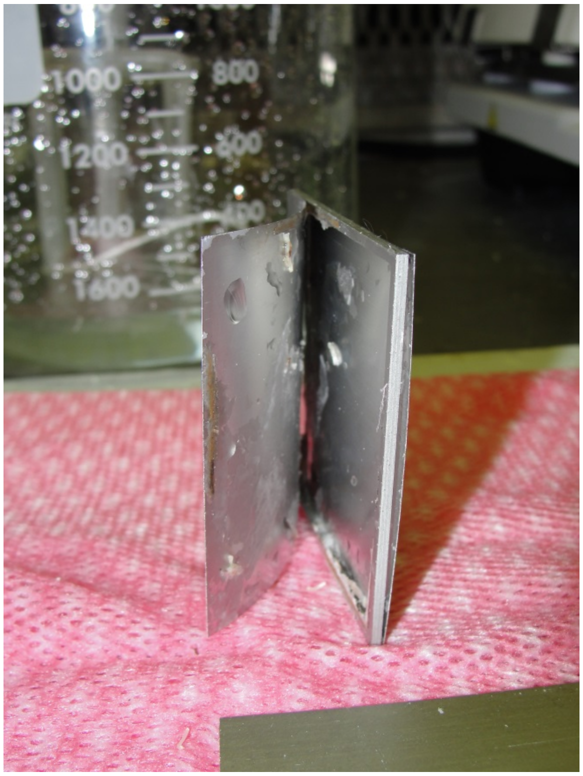


Figure 3-14. Experiment 121 FCA Simulant Element after 692 Hours Dissolution before Cleaning

Figure 3-15. Experiment 121 FCA Simulant Element after 692 Hours Dissolution after Cleaning

Experiment 121 was stopped after the 741-hour (44,439 minutes) sample. As indicated by the incremental dissolution rates for the FCA simulant fuel element at 692 and 741 hours, the FCA simulant fuel element had returned to approximately the same incremental dissolution rate as the stainless steel plate (Figure 3-8). The FCA simulant fuel element dissolution rate for the earlier samples matched the plain stainless steel dissolution rate before the FCA simulant fuel element weld failure and subsequent Cu dissolution (Figure 3-8). Zooming in on the dissolution rate scale, the stainless steel plate dissolution rate dropped sharply at first and then slowed for the duration of the experiment, as expected from the initial experiments (Section 3.2). Since the FCA simulant fuel element dissolution behavior matched the beginning and end of the stainless steel plate, the dissolution rates validate each another. Although the simulant FCA fuel elements experienced breaching at the weld, it is not known whether all FCA fuel elements could be counted on to fail in a similar manner since they were welded using different weld techniques. Consequently, the only certain form of element breach is through the facing.

To estimate the time needed to breach the face of the FCA element, the incremental dissolution rates for the stainless steel plates in Experiments 119, 120, and 121 were examined. The dissolution times and incremental dissolution rates from these experiments are listed in Table 3-8 and plotted in Figure 3-16. A power law fit was made to the dissolution rate data:

$$\text{Diss. Rate} = 0.00628 * \text{time}^{-0.593} \qquad (8)$$

Where *Diss. Rate* is the dissolution rate in mg/(cm^2*min) and *time* is the cumulative dissolution time in days. For an assumed 5.08 cm x 5.08 cm x 0.021 cm (2 in x 2 in x 0.0083 in) stainless steel face of an FCA element, the surface area is 26.23 cm^2 and the metal volume is 0.5419 cm^3. Using 8.0 g/cm^3 as density for stainless steel gives a mass of 4,335 mg to dissolve to breach the face. Using the power law dissolution rate expression (Equation 8) with the surface area of the stainless steel face, the mass dissolved each day was estimated. A table was generated showing the mass dissolved each day and then summed over cumulative days (Appendix H). Using this approach, the mass dissolved reaches 4,335 mg in approximately 176 days indicating that the FCA stainless steel face would breach in a little over 176 days.

Table 3-8. Incremental Dissolution Rates for Experiments 119, 120, 121

Dissolution Time	Incremental Dissolution Rate
days	**mg/(cm^2*min)**
0.33	0.0113
1.02	0.0073
2.99	0.0026
6.00	0.0022
9.01	0.0018
12.97	0.0016
16.83	0.0013
21.85	0.0010
28.82	0.0008
30.86	0.0007

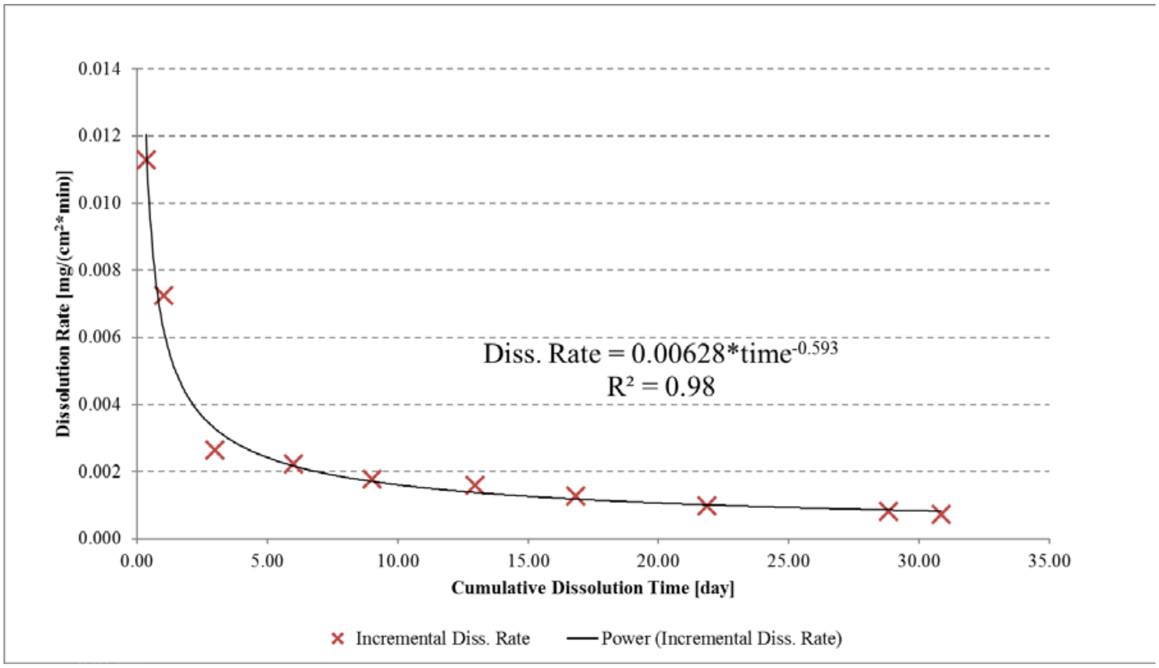


Figure 3-16. Incremental Dissolution Rates for Stainless Steel over time Based on Masses

As a check on the dissolution rate, the ICPES analyses from Experiment 121 were used to estimate the mass of Fe and Cr that had dissolved for each sample point. Knowing the concentrations of these species in stainless steel (Fe 66.9 wt%, Cr 19.5 wt%), the mass of stainless steel dissolved at each sample point can be estimated. Combining these masses with the estimated surface areas for both the stainless steel plate and the FCA simulant element (~ 82.5 cm^2 before the weld breach and ~133.6 cm^2 after the weld breach), the incremental dissolution rate for stainless steel can be estimated at each sample point (Table 3-9). The ICPES results at 21.85 days were not used in the dissolution comparison as the mass of Fe and Cr suddenly jumped up perhaps from contamination after the internal Cu plate dissolved. The ICPES results at 28.82 days were also not used because gave negative mass amounts for Fe and Cr due to the dissolution of the internal Cu plate. Looking at the dissolution rates based on Fe and Cr (Figure 3-17), power law fits gave similar dissolution rate expressions as defined based on the mass measurements.

$$\text{Diss. Rate based on Fe} = 0.007*\text{time}^{-0.642} \quad (6)$$

$$\text{Diss. Rate based on Cr} = 0.006*\text{time}^{-0.626} \quad (7)$$

Where *Diss. Rate* is the dissolution rate in mg/(cm^2*min) and *time* is the cumulative dissolution time in days. Therefore, the ICPES data supports or validates the mass derived dissolution rate.

There was some glass corrosion or etching in the reactor due to the long-term exposure to the F^- ion in solution during Experiment 121 (Figure 3-18, Figure 3-19). Based on the level of F^- ion concentration and recommendations from the SRNL glass shop, it was planned to replace the exposed glassware about half way through the experimental run. The reactor and glass thermowell were changed out after the 404-hour sample due to etching.

Table 3-9. Experiment 121 Incremental Dissolution Rates Based on ICPES

Dissolution Time	Incremental Dissolution Rate based on Cr	Incremental Dissolution Rate based on Fe
days	mg/(cm^2*min)	mg/(cm^2*min)
1.02	0.0068	0.0079
2.99	0.0025	0.0029
6.00	0.0018	0.0021
9.01	0.0018	0.0020
12.97	0.0013	0.0014
16.83	0.0010	0.0011
21.85	0.0024	0.0029

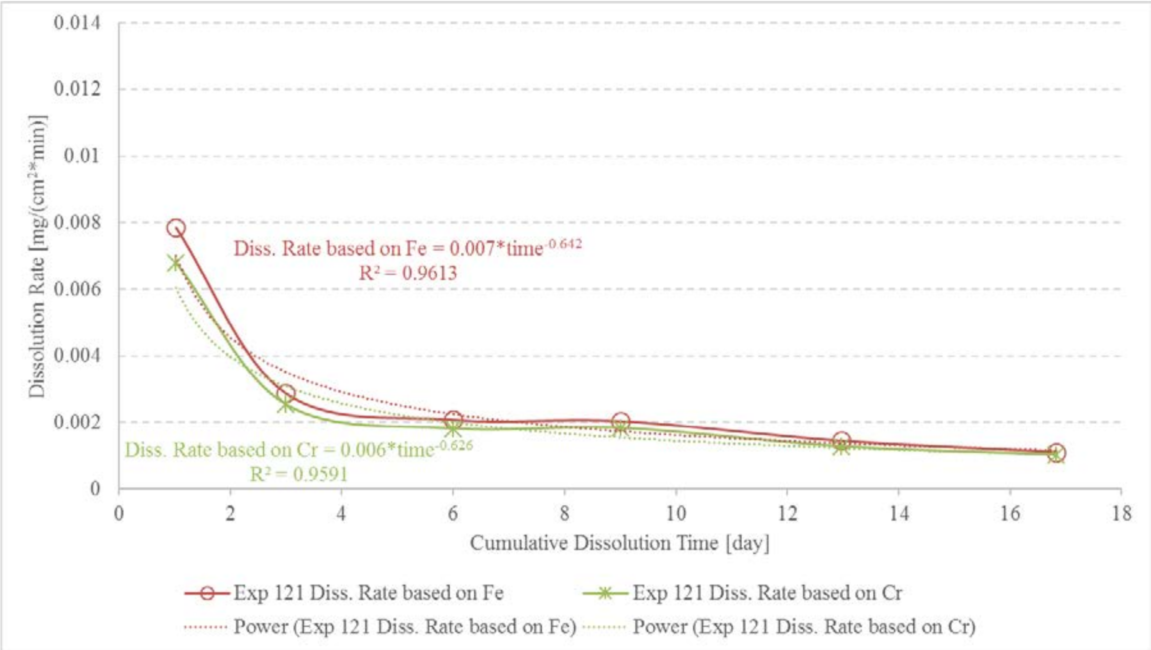


Figure 3-17. Experiment 121 Incremental Dissolution Rates Based on ICPES

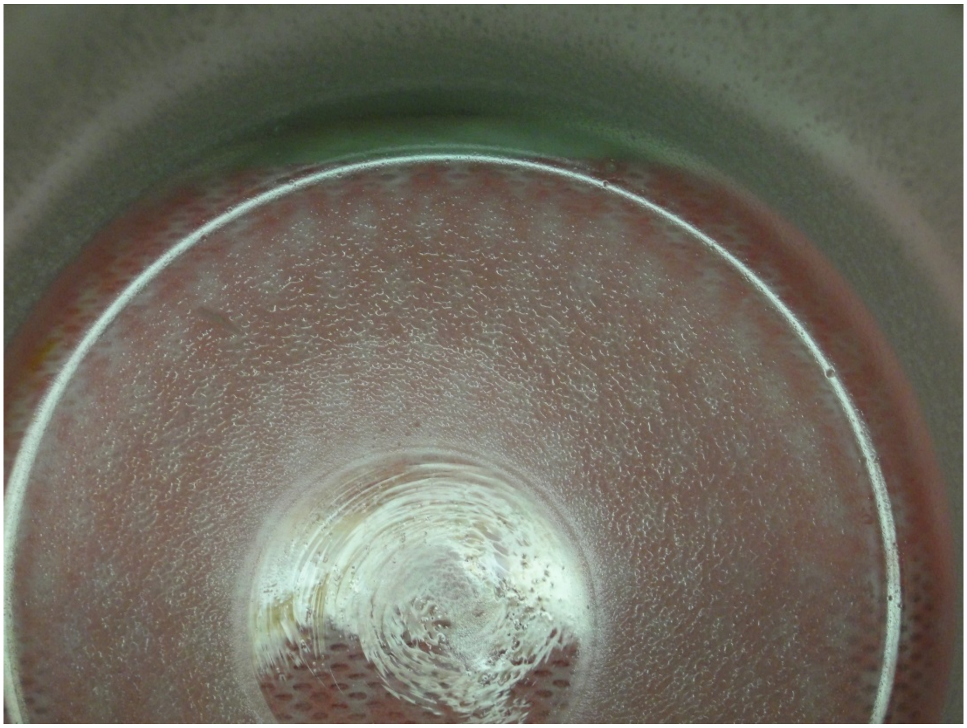


Figure 3-18. Experiment 121 Glass Etching in Reaction Vessel (after 404 hours)

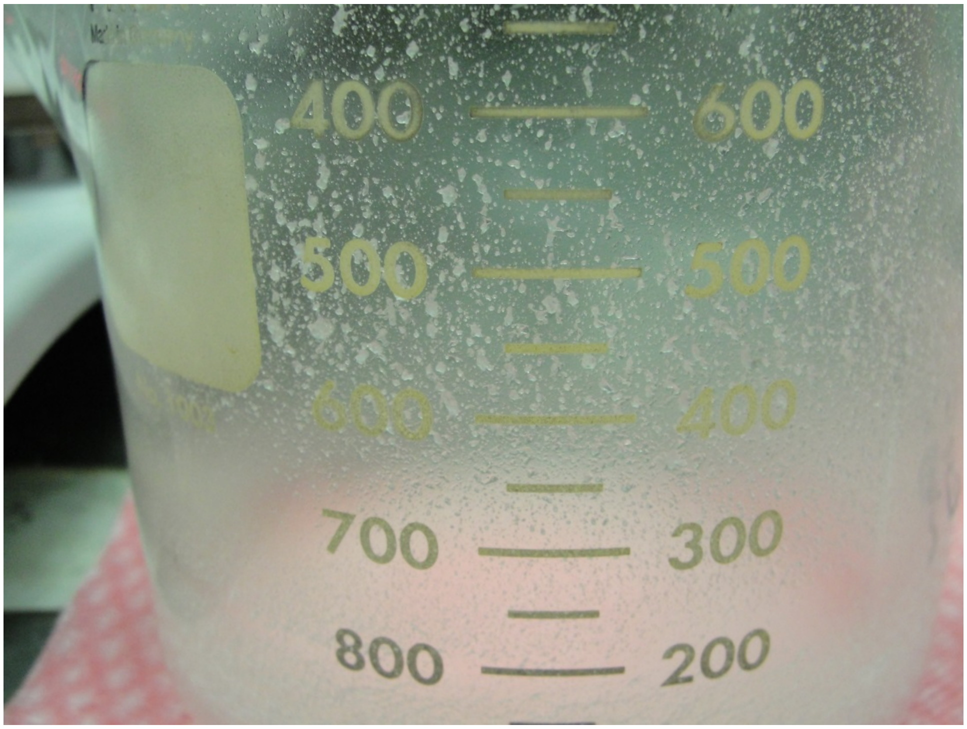


Figure 3-19. Experiment 121 Glass Etching of Reaction Vessel (after 404 Hours)

3.2.2 Single FCA Simulant fuel element Dissolution in Teflon Reactor

There was a concern that the corrosion products of the glassware could form ionic complexes with some of the F^- ion in the form of silicates. This might result in a condition significantly different from H-Canyon dissolution and yield dissolution rates that were biased low. To examine this concern, a dissolution experiment with the same conditions as Experiment 121 was performed except the reactor was Teflon instead of glass (Figure 3-20). Like Experiment 121, there was a FCA Simulant fuel element and a half-sized stainless steel plate for the Teflon Reactor experiment (Figure 3-21). At various times, the coupons were removed from solution to measure their weights. At some of these times, samples were obtained for ICPES analyses to track the metal concentrations in solution. This experiment was performed for a shorter time (8 days) with the objective being to compare the early stages of dissolution with that of Experiment 121. Based on the results of Experiment 121 for the first 8 days of dissolution, the dimensions of the FCA simulant fuel element and stainless steel plate changed at similar rates.

The ICPES results for samples pulled during the single FCA simulant fuel element experiments in a Teflon reactor are shown in Table 3-10. The Cu concentration (Figure 3-22) versus time for the Teflon Reactor experiment is similar to the glass reactor initially but its last data point at 8 days is lower because the FCA simulant fuel weld had not been breached as in Experiment 121.

The coupon mass data for this experiment are shown in Table 3-11 for the FCA simulant fuel element and in Table 3-12 for the stainless steel plate. The change in dimensions were assumed to be negligible between samples. The dissolution rates are incremental to represent dissolution from one sample to the next. The surface area used in the dissolution rate calculation is the initial value except for the last sample which used the average of the initial and final surface areas. Plots of the incremental dissolutions rates versus cumulative dissolution time for the Teflon and glass reactor dissolutions are shown in Figure 3-23. The dissolution rates for the FCA simulant fuel element and the stainless steel plates in the Teflon reactor are similar to the dissolution rates in Experiment 121 during the same time frame. Based on these results, the etching of the glass reactor from the F^- is not interfering with the stainless steel dissolution rates in a significant manner. The dissolution rates measured in these experiments also validate the values obtained in the longer-term Experiment 121.

As a check on the dissolution rate, the ICPES analyses from the Teflon Reactor Experiment were used to estimate the mass of Fe and Cr that had dissolved for each sample point. Knowing the concentrations of these species in stainless steel (Fe 66.9 wt%, Cr 19.5 wt%), the mass of stainless steel dissolved at each sample point can be estimated. Combining these masses with the estimated surface areas for both the stainless steel plate and the FCA simulant element (~ 82.8 cm^2), the incremental dissolution rate for stainless steel can be estimated at each sample point (Table 3-13). Looking at the dissolution rates based on Fe and Cr (Figure 3-24) from the Teflon experiment compared to Experiment 121, the dissolution rates are basically the same indicating that the dissolved Si from the glassware due to the F^- did not bias the dissolution results.

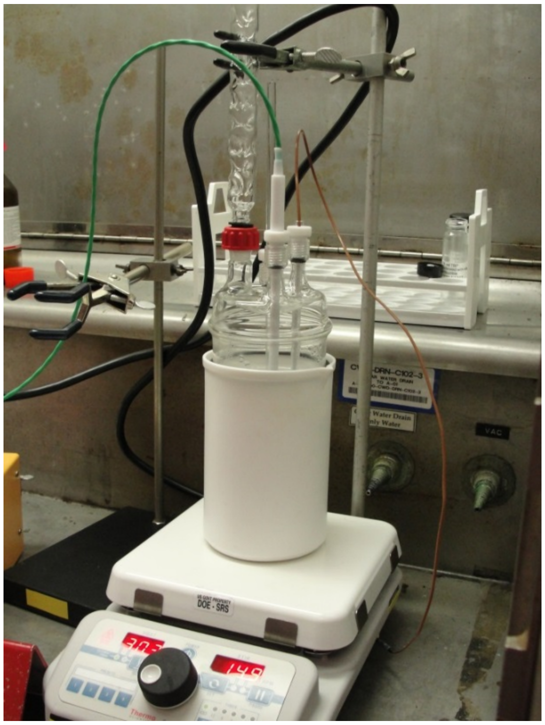


Figure 3-20. Teflon Reactor Dissolution Experimental Setup

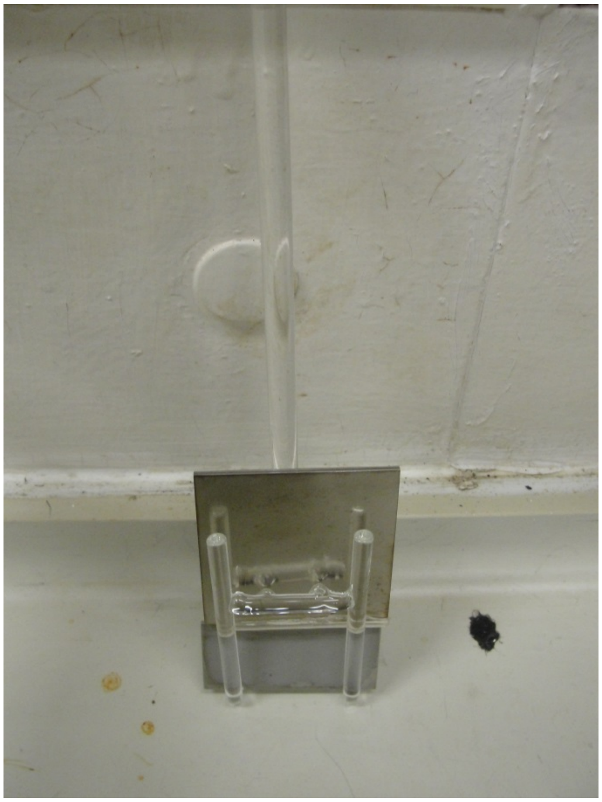


Figure 3-21. Teflon Reactor Dissolution FCA Simulant Element (top) and Stainless Steel Plate (bottom) in Holder

Table 3-10. ICPES Analyses for Teflon Reactor Experiment

Total Time of Dissolution (min)	2801	5849	11384
ICPES Sample ID:	LW7929	LW7930	LW7931
Units:	mg/L	mg/L	mg/L
Al	15.2	16.3	24.3
B	1110	1110	1180
Cr	304	435	656
Cu	10.4	57.8	281
Fe	1210	1730	2640
K	8520	8430	8970
Mn	23.9	34.3	52.1
Na	48.1	44.9	65.1
Ni	153	220	332
Si	324	306	267
Solution Volume (ml)	750	750	685

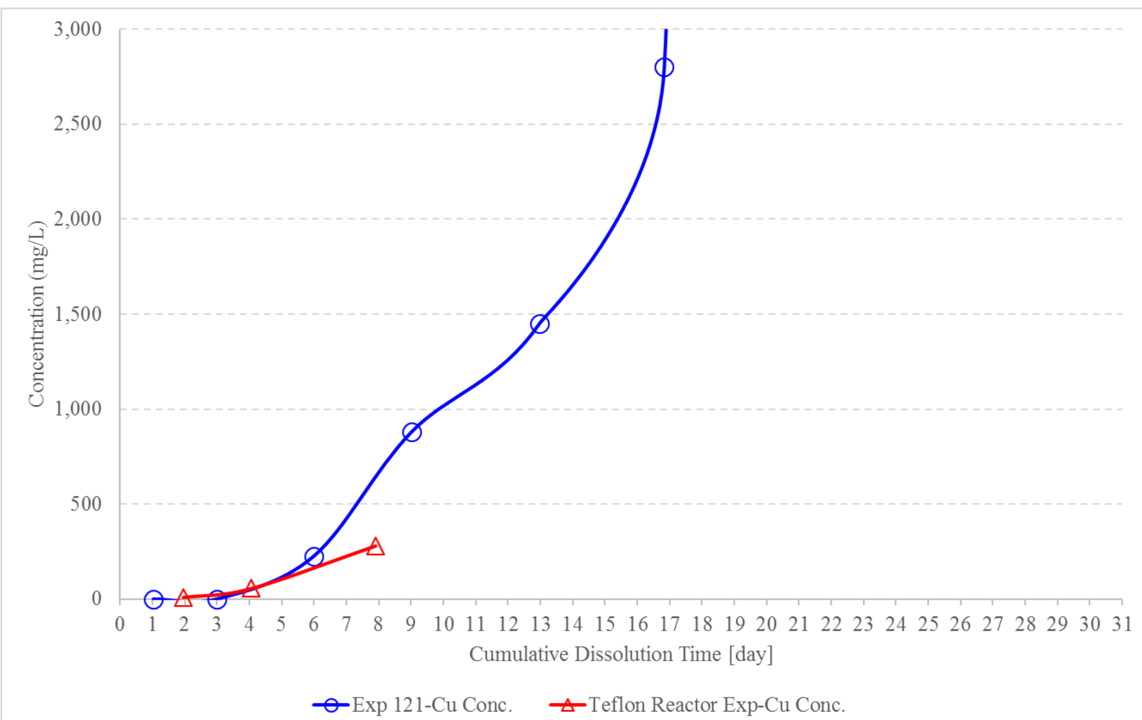


Figure 3-22. Comparison of Cu Concentration for Teflon reactor and Experiment 121 Glass Reactor

Table 3-11. Teflon Reactor Dissolution Experiment FCA Simulant Fuel Element Sample Data

Sample	Initial Mass (g)	Ending Mass (g)	Mass Loss (g)	% Loss	Total Dissolution Time (min)	Incremental Dissolution Time (min)	Initial Length (mm)	Initial Width (mm)	Initial Depth (mm)	Initial SA (cm^2)	Ending Length (mm)	Ending Width (mm)	Ending Depth (mm)	Ending SA (cm^2)	Avg SA (cm^2)	Incremental Dissolution Rate (mg/(cm^2*min))
1	28.6080	28.0784	0.5294	1.9	1390	1390	50.80	50.80	2.00	55.68	---	---	---	---	---	0.0068
2	28.0784	27.9202	0.1582	0.6	2801	1411	50.80	50.80	2.00	55.68	---	---	---	---	---	0.0020
3	27.9202	27.7640	0.1562	0.6	5849	3048	50.80	50.80	2.00	55.68	---	---	---	---	---	0.0009
4	27.7640	27.4830	0.2810	1.0	8772	2923	50.80	50.80	2.00	55.68	---	---	---	---	---	0.0017
5	27.4830	27.1295	0.3535	1.3	11384	2612	50.80	50.80	2.00	55.68	50.68	50.72	1.95	55.37	55.52	0.0024

Table 3-12. Teflon Reactor Dissolution Experiment Stainless Steel Plate Sample Data

Sample	Initial Mass (g)	Ending Mass (g)	Mass Loss (g)	% Loss	Total Dissolution Time (min)	Incremental Dissolution Time (min)	Initial Length (mm)	Initial Width (mm)	Initial Depth (mm)	Initial SA (cm^2)	Ending Length (mm)	Ending Width (mm)	Ending Depth (mm)	Ending SA (cm^2)	Avg SA (cm^2)	Incremental Dissolution Rate (mg/(cm^2*min))
1	13.9338	13.6200	0.3138	2.3	1390	1390	50.79	24.83	1.34	27.25	---	---	---	---	---	0.0083
2	13.6200	13.5023	0.1177	0.9	2801	1411	50.79	24.83	1.34	27.25	---	---	---	---	---	0.0031
3	13.5023	13.3308	0.1715	1.3	5849	3048	50.79	24.83	1.34	27.25	---	---	---	---	---	0.0021
4	13.3308	13.1885	0.1423	1.1	8772	2923	50.79	24.83	1.34	27.25	---	---	---	---	---	0.0018
5	13.1885	13.0818	0.1067	0.8	11384	2612	50.79	24.83	1.34	27.25	50.71	24.76	1.39	27.21	27.23	0.0015

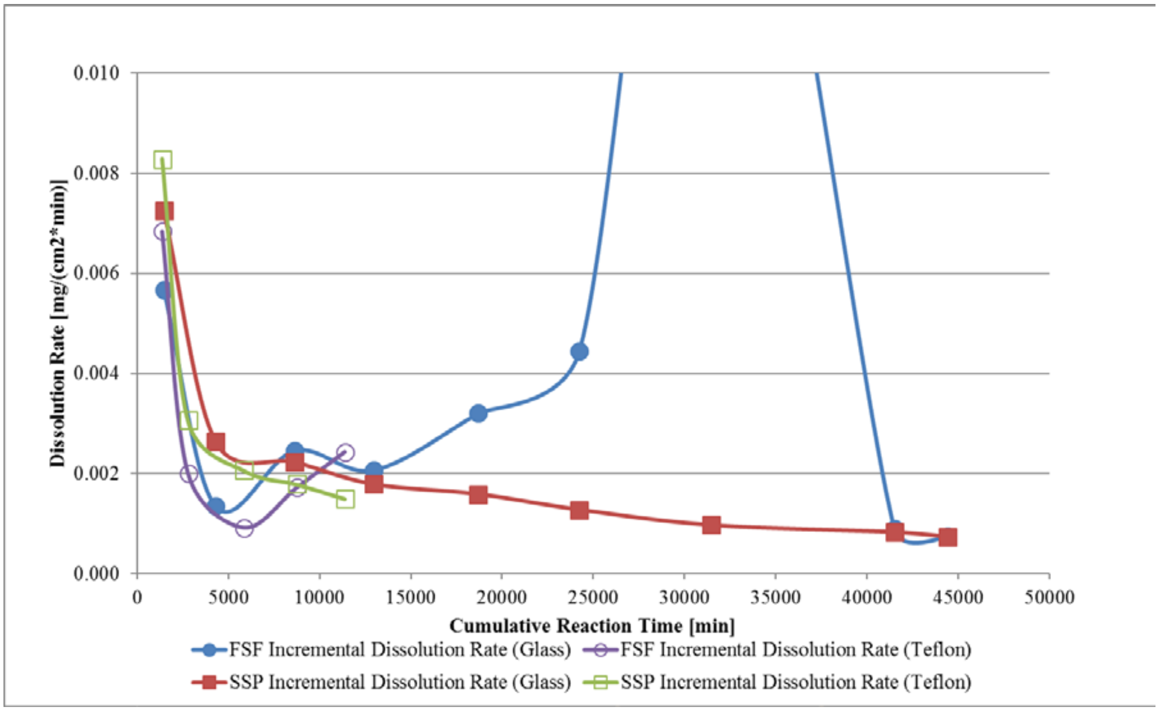


Figure 3-23. Incremental Dissolution Rates based on Coupon Mass and Dimensions

Table 3-13. Teflon Reactor Experiment Incremental Dissolution Rates based on ICPES

Dissolution Time	Incremental Dissolution Rate based on Cr	Incremental Dissolution Rate based on Fe
days	$mg/(cm^2*min)$	$mg/(cm^2*min)$
1.95	0.0050	0.0058
4.06	0.0020	0.0023
7.91	0.0014	0.0017

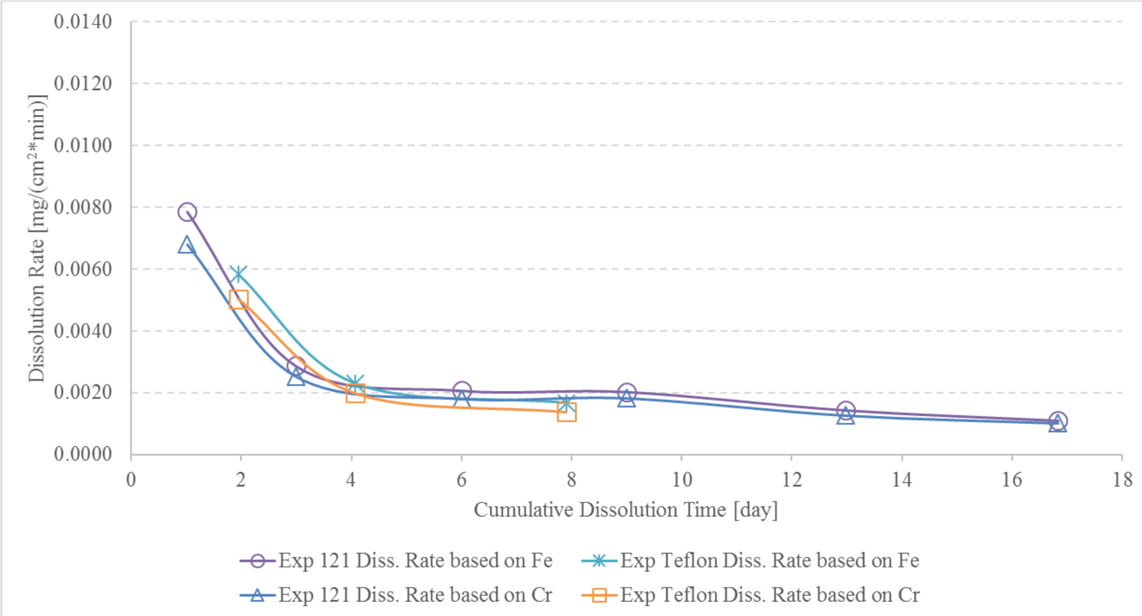


Figure 3-24. Teflon Reactor Incremental Dissolution Rates over time based on ICPES

3.2.3 Dissolution of Multiple FCA Simulant Fuel Elements

After observing in Experiment 121 where the weld breached on the FCA simulant fuel element, it was decided to include stainless steel plates in the multiple FCA simulant fuel element dissolution test, Experiment 122. The reason was that if a FCA simulant fuel element weld breached before the facing breached, then the mass data would be biased high or low but the rates of the stainless steel plates would be unaffected and could therefore be used as a reference point for tracking the dissolution rates. The ordering of the FCA Simulant fuel elements (FSF) and the stainless steel plates (SSP) in the stand is shown in Figure 3-25.

At various times in Experiment 122, samples were pulled for ICPES analyses to track the metal concentrations in solution for comparison with the weight-loss data (Table 3-14). Also at these sample points, the stand with the simulant fuel elements was removed from solution to weigh the coupons and measure their dimensions. The coupon mass and dimension data for this experiment at the various sample points is shown in Table 3-15. The dissolution rates are incremental, representing from one sample to the next, which gives a more accurate presentation of the dissolution rate at that time compared to a cumulative rate.

The ICPES and free/total acid results for samples obtained during the multiple FCA simulant fuel element experiments are shown in Table 3-14. The total acid analysis shows that the starting acid concentration is within analytical uncertainty. The free acid analyses show that not much acid is consumed by the later sample time (19,899 min) nor at the end of the experiment (24,151 min). These results make sense as none of the FCA simulant fuel elements appeared to be significantly breached allowing the dissolution of the internal Cu plate which consumes HNO_3 to yield NOx. These observations track with the Cu concentration versus total dissolution time (Figure 3-26). Comparing Experiment 121 Cu concentration where the internal Cu Plate dissolved, one can see that in Experiment 122 the internal Cu plates were still primarily intact.

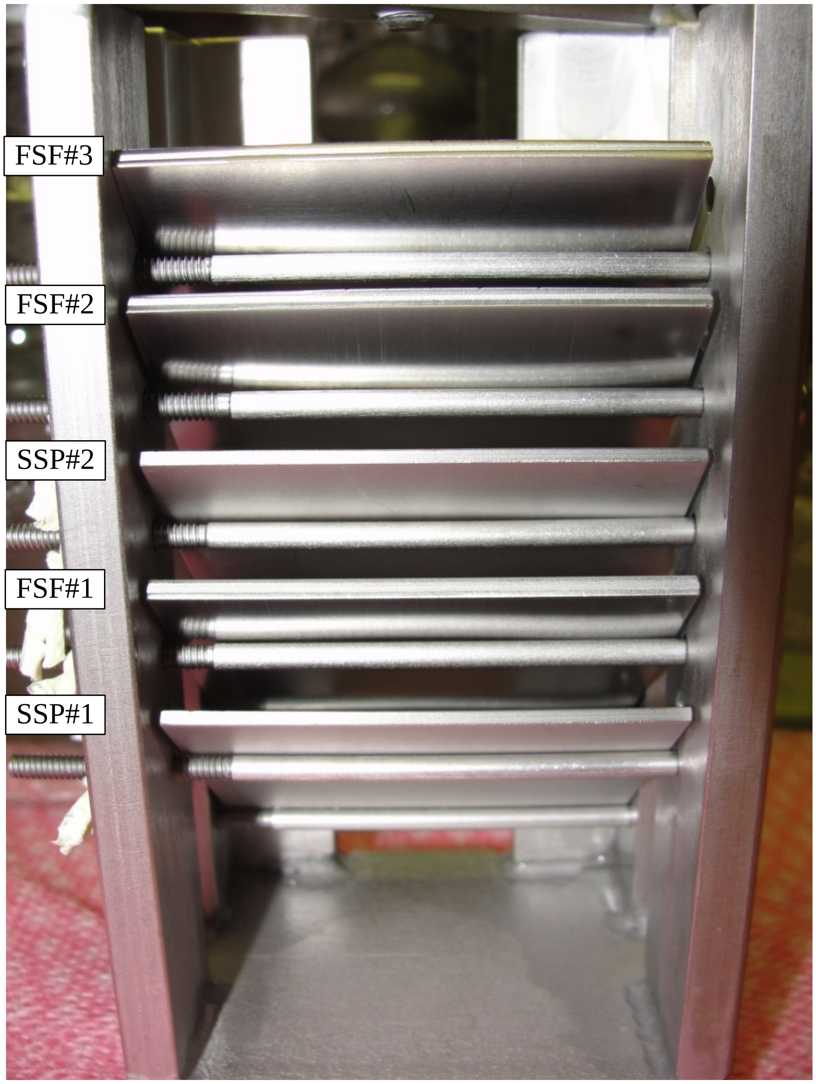


Figure 3-25. Experiment 122 Stand with Multiple Simulant Elements at 0.5-inch spacing

Table 3-14. Total/Free Acid and ICPES Analyses for Experiment 122

Total Time of Dissolution (min)	0	1446	4310	8558	12792	19899	24151
Total Acid (M)	3.95	---	---	---	---	3.84	3.86
Free Acid (M)	3.80	---	---	---	---	3.79	3.77
Free/Total Acid sample ID:	LW7933	---	---	---	---	LW7934	LW7935
ICPES Sample ID:	---	LW7923	LW7924	LW7925	LW7926	LW7927	LW7928
Units:	---	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
Al	---	9.55	11.5	14.4	16.9	21.1	23.1
B	---	2010	2010	2050	2060	2080	2100
Cr	---	257	656	1170	1600	2230	2570
Cu	---	13.7	31.8	53.9	72.7	98.9	114
Fe	---	1040	2660	4670	6370	8820	10100
K	---	5600	5650	5640	5690	5790	5700
Mn	---	19.6	50.8	89.4	122	169	192
Na	---	20.1	24.3	30.5	36.6	46.1	51.1
Ni	---	131	337	598	809	1120	1290
Si	---	98.9	92.3	145	81.1	62.7	71.2
Solution Volume (ml)	1500	1500	1496	1493	1491	1489	1483

Table 3-15. Experiment 122 Dissolution Rate Sample Data

Coupon	Initial Mass (g)	Ending Mass (g)	Mass Loss (g)	% Mass Loss	Total Time of Dissolution (min)	Incremental Time of Dissolution (min)	Initial Length (mm)	Initial Width (mm)	Initial Depth (mm)	Initial SA (cm^2)	Ending Length (mm)	Ending Width (mm)	Ending Depth (mm)	Ending SA (cm^2)	Avg SA (cm^2)	Incremental Dissolution Rate (mg/(cm^2*min))
FSF#1	28.618	28.490	0.128	0.4%	1446	1446	50.88	50.81	2.08	55.94	50.86	50.80	2.08	55.90	55.92	0.0016
FSF#1	28.490	28.299	0.191	0.7%	4310	2864	50.86	50.80	2.08	55.90	50.85	50.80	2.04	55.81	55.85	0.0012
FSF#1	28.299	28.071	0.228	0.8%	8558	4248	50.85	50.80	2.04	55.81	50.80	50.78	2.06	55.79	55.80	0.0010
FSF#1	28.071	27.886	0.185	0.7%	12792	4234	50.80	50.78	2.06	55.79	50.79	50.78	2.00	55.65	55.72	0.0008
FSF#1	27.886	27.629	0.257	0.9%	19899	7107	50.79	50.78	2.00	55.65	50.77	50.75	2.00	55.59	55.62	0.00065
FSF#1	27.629	27.506	0.123	0.4%	24151	4252	50.77	50.75	2.00	55.59	50.76	50.74	1.98	55.53	55.56	0.00052
FSF#2	28.656	28.532	0.124	0.4%	1446	1446	50.80	50.81	2.33	56.36	50.79	50.80	2.32	56.31	56.34	0.0015
FSF#2	28.532	28.343	0.189	0.7%	4310	2864	50.79	50.80	2.32	56.31	50.79	50.80	2.27	56.21	56.26	0.0012
FSF#2	28.343	28.118	0.225	0.8%	8558	4248	50.79	50.80	2.27	56.21	50.77	50.79	2.24	56.12	56.17	0.0009
FSF#2	28.118	27.936	0.182	0.6%	12792	4234	50.77	50.79	2.24	56.12	50.75	50.77	2.15	55.90	56.01	0.0008
FSF#2	27.936	27.679	0.257	0.9%	19899	7107	50.75	50.77	2.15	55.90	50.72	50.75	2.08	55.71	55.80	0.00065
FSF#2	27.679	27.554	0.125	0.5%	24151	4252	50.72	50.75	2.08	55.71	50.71	50.74	2.13	55.79	55.75	0.00053
FSF#3	28.724	28.603	0.121	0.4%	1446	1446	50.88	50.81	2.22	56.22	50.84	50.83	2.18	56.12	56.17	0.0015
FSF#3	28.603	28.416	0.187	0.7%	4310	2864	50.84	50.83	2.18	56.12	50.83	50.83	2.09	55.93	56.03	0.0012
FSF#3	28.416	28.194	0.222	0.8%	8558	4248	50.83	50.83	2.09	55.93	50.83	50.80	2.16	56.03	55.98	0.0009
FSF#3	28.194	28.018	0.176	0.6%	12792	4234	50.83	50.80	2.16	56.03	50.82	50.80	2.06	55.82	55.93	0.0007
FSF#3	28.018	27.787	0.231	0.8%	19899	7107	50.82	50.80	2.06	55.82	50.80	50.77	2.04	55.73	55.78	0.00058
FSF#3	27.787	27.834*	-0.047	-0.2%	24151	4252	50.80	50.77	2.04	55.73	50.80	50.77	2.01	55.66	55.70	-0.00020
SSP#1	29.732	29.582	0.150	0.5%	1446	1446	50.78	50.79	1.47	54.56	50.78	50.78	1.46	54.54	54.55	0.0019
SSP#1	29.582	29.383	0.199	0.7%	4310	2864	50.78	50.78	1.46	54.54	50.76	50.79	1.46	54.53	54.53	0.0013
SSP#1	29.383	29.151	0.232	0.8%	8558	4248	50.76	50.79	1.46	54.53	50.76	50.77	1.45	54.48	54.51	0.0010
SSP#1	29.151	28.962	0.189	0.6%	12792	4234	50.76	50.77	1.45	54.48	50.75	50.77	1.44	54.45	54.47	0.0008
SSP#1	28.962	28.700	0.262	0.9%	19899	7107	50.75	50.77	1.44	54.45	50.73	50.75	1.43	54.40	54.42	0.00068
SSP#1	28.700	28.562	0.138	0.5%	24151	4252	50.73	50.75	1.43	54.40	50.72	50.75	1.42	54.36	54.38	0.00060
SSP#2	29.661	29.513	0.148	0.5%	1446	1446	50.73	50.78	1.48	54.52	50.78	50.72	1.46	54.48	54.50	0.0019
SSP#2	29.513	29.310	0.203	0.7%	4310	2864	50.78	50.72	1.46	54.48	50.78	50.72	1.45	54.46	54.47	0.0013
SSP#2	29.310	29.082	0.228	0.8%	8558	4248	50.78	50.72	1.45	54.46	50.71	50.77	1.44	54.42	54.44	0.0010
SSP#2	29.082	28.898	0.184	0.6%	12792	4234	50.71	50.77	1.44	54.42	50.70	50.76	1.45	54.41	54.42	0.0008
SSP#2	28.898	28.638	0.260	0.9%	19899	7107	50.70	50.76	1.45	54.41	50.69	50.74	1.43	54.35	54.38	0.00067
SSP#2	28.638	28.502	0.136	0.5%	24151	4252	50.69	50.74	1.43	54.35	50.68	50.73	1.42	54.31	54.33	0.00059

*FSF weld may have been breached and solution entered weld area, thus artificially driving mass up.

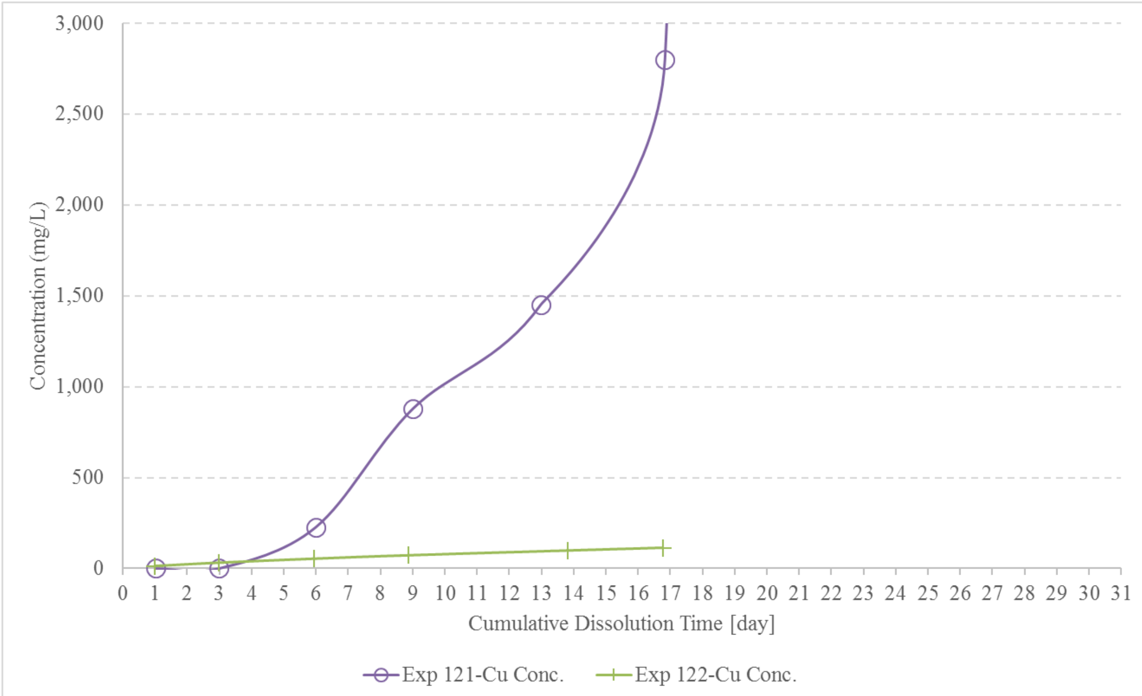


Figure 3-26. Experiment 121 and 122 Cu Concentration versus Dissolution Time

As seen in Figure 3-27, the incremental dissolution rates for the stainless steel plates (SSP#1, SSP#2) are similar at each sample time. The incremental dissolution rates for the FCA simulant fuels (FSF#1, FSF#2, FSF#3) are also similar at each of the sample times. It appears that the FSF#3 dissolution rate is drifting away from FSF#1 and FSF#2 at 19,899 minutes (332 hours). The last sample point at 24,151 minutes (403 hours) for FSF#3 indicates that the weld may have been breached as its mass increased from the prior sample point (liquid inside of the element).

Pictures of the edge of FSF#3 look intact until 403 hours where there is some discoloration, which may indicate a breach of the weld (Figure 3-28). The sample mass at 332 hours may have been biased high due to the breach (allowing liquid inside of the simulant element), thus giving a lower dissolution rate as observed from weight-loss data. The uniformity of dissolution rates between the five plates indicates that the 0.5-inch spacing between the FCA fuel elements does not appear to negatively affect the relative dissolution rate among the plates. Also, the rack design appears appropriate for the application. The rack design allows for testing at 0.25-inch spacing, but that does not appear to be warranted due to lengthy dissolution time observed for Experiment 121.

As a check on the dissolution rates, the ICPES analyses from Experiment 122 were used to estimate the mass of Fe and Cr that had dissolved for each sample point. Knowing the concentrations of these species in stainless steel (Fe 66.9 wt%, Cr 19.5 wt%), the mass of stainless steel dissolved at each sample point can be estimated. Combining these masses with the estimated surface areas for all the stainless steel plates, all the FCA simulant elements, and the stand (about 561 cm^2), the incremental dissolution rate for stainless steel can be estimated at each sample point (Table 3-16). Looking at the dissolution rates based on Fe and Cr (Figure 3-29) for both Experiment 121 and 122, the estimated dissolution rates from about Day 3 forward are very close. Due to the extra amount of stainless steel surface available in Experiment 122 (about seven times that of Experiment 121), the initial dissolution rate for Day 1 was lowered due to the 0.25M F- ions having more surface area to attack relative to the volume of the solution. It is expected that if the F- to surface area ratio in Experiment 122 was like that in Experiment 121, the initial dissolution values would have been closer. These results do support the prior result that the spacing of the FCA simulant fuel elements does not impact the dissolution rates of the elements relative to each other.

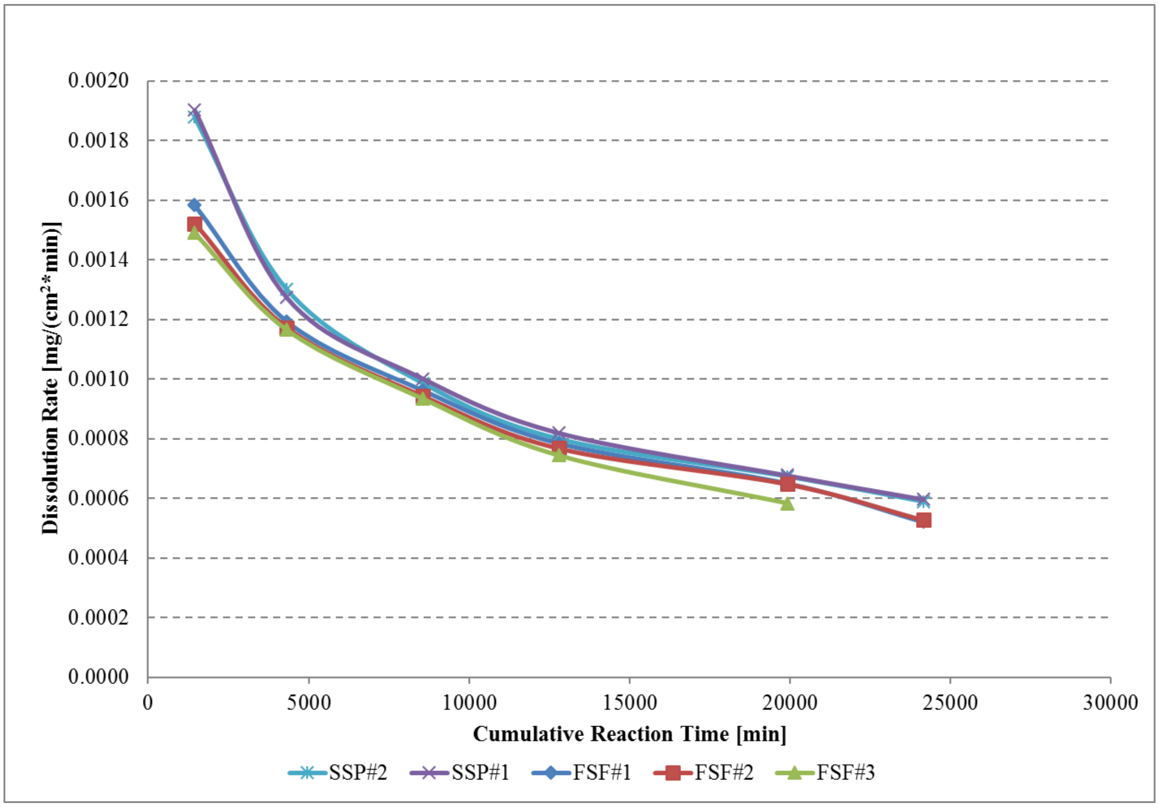
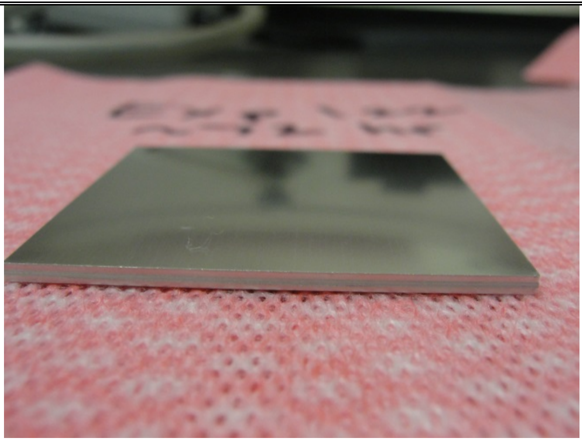
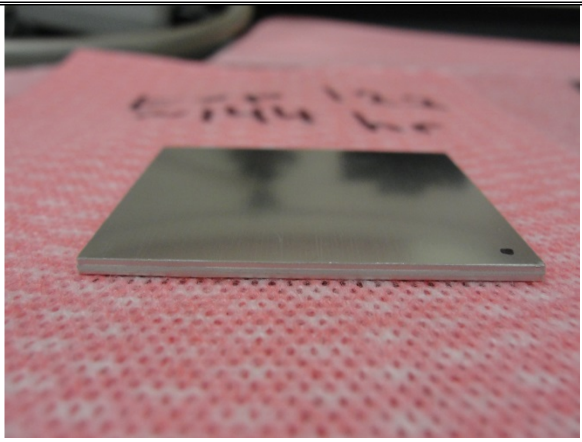


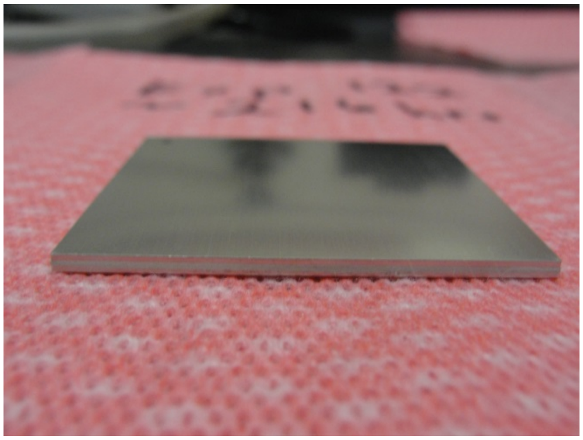
Figure 3-27. Incremental Dissolution Rates based on Coupon Mass and Dimensions



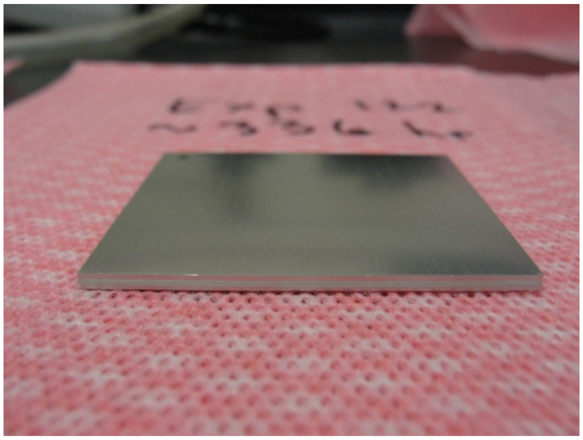
73 hours dissolution



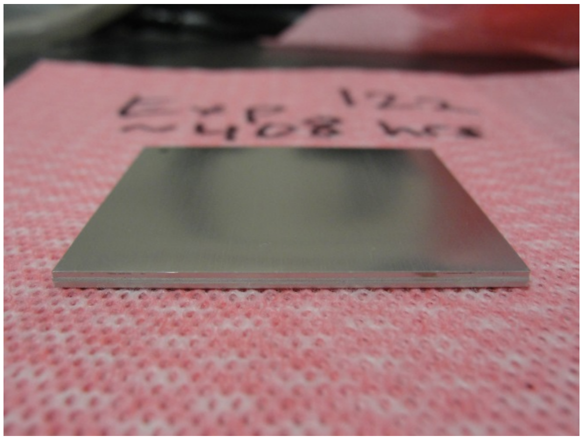
143 hrs dissolution



213 hours dissolution



332 hours dissoluiton



403 hours dissolution

Figure 3-28. Experiment 122 FSF#3 Edge Pictures at Sample Points

Table 3-16. Experiment 122 Incremental Dissolution Rates based on ICPES

Dissolution Time	Incremental Dissolution Rate based on Cr	Incremental Dissolution Rate based on Fe
days	mg/(cm^2*min)	mg/(cm^2*min)
1.00	0.0024	0.0029
2.99	0.0019	0.0022
5.94	0.0016	0.0019
8.88	0.0014	0.0016
13.82	0.0012	0.0014
16.77	0.0011	0.0012

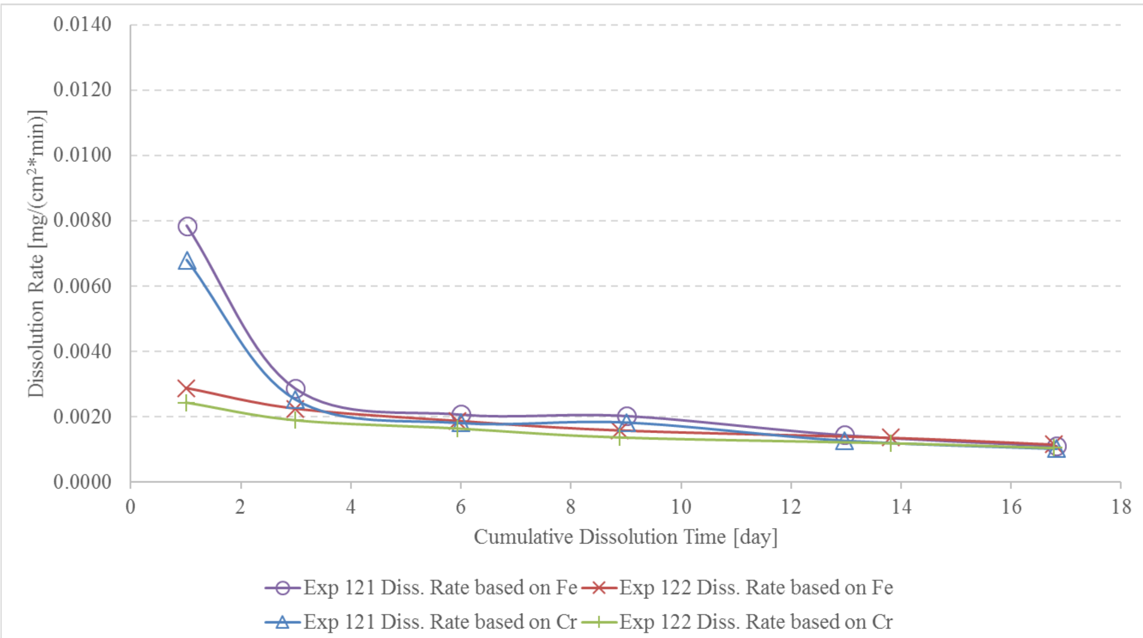


Figure 3-29. Experiment 121 & 122 Incremental Dissolution Rates over time based on ICPES

4.0 Conclusions

Initial experiments were performed to identify optimal dissolution conditions for stainless steel in terms of HNO_3 molarity (4M, 7M, 10M) and F^- molarity (0.1M, 0.25M, 0.4M). All scoping tests used 1.2 g/L B to represent the neutron poison. The initial experiments showed that higher stainless steel dissolution rates corresponded with low HNO_3 and high F^- concentrations. These results were not surprising as the F^- ion breaks through the oxide layer formed on the stainless steel surface to cause dissolution or corrosion and having lower molar acid reduces the oxidation potential of the solution. From these initial experiments, the optimal dissolution conditions within H-Canyon constraints were determined to be 4M HNO_3 and 0.25M KF at 102 °C. Using these optimal conditions, long-term dissolutions were performed on FCA simulant fuel elements.

The simulant FCA fuel elements consisted of two stainless steel face plates surrounding an internal Cu plate. The internal Cu plate simulated Pu which would readily dissolve if the stainless steel cladding was breached.

These long-term studies included testing to see whether spacing between simulant FCA fuel elements affected the uniform dissolution rate between fuel elements. The long-term studies with the simulant FCA fuel elements included plain stainless steel plates as a control. The experiments showed that the FCA simulant fuel element had similar dissolution rates with the plain stainless steel plates. One anomaly occurred when the weld on the FCA simulant breached, allowing the internal Cu plate to dissolve even though the facing had not been breached. It is not known whether all FCA fuel elements could be counted on to fail in a similar manner as they were welded using different weld techniques. When the internal Cu plate dissolved, there was a sudden mass loss which caused the calculated FCA dissolution rate to increase an order of magnitude. The FCA simulant fuel element dissolution rate then returned to similar levels of the plain stainless steel after the Cu dissolution was complete.

These studies showed that the stainless steel dissolution rate started high (0.006-0.007 mg/(cm^2*min)) and gradually decreased over 30 days to about 0.001 mg/(cm^2*min). Assuming an FCA plate has a stainless steel face plate of 2 inches (50.8 mm) by 2 inches (50.8 mm) by 0.0083 inch (0.21 mm) with a density of 8 g/cm^3, that gives a theoretical mass of 4.34 g. Fitting the 30-day incremental dissolution rate data to a power law function and using a surface area of just the face of the FCA element of 26.2 cm^2 the estimated time to dissolve the cladding on the FCA plate is about 176 days. This hypothetical case is presented to show how long the dissolution of the stainless steel could take based on test results for 4M HNO_3, 0.25M KF, and 1.2 g/L B at 102 °C.

An alternate strategy was considered that involved dissolving the fuel elements for 20 days, removing the dissolver solution before the dissolution rate degrades so much, and adding a new batch of dissolver solution. Calculations show that each 20-day dissolution cycle would remove about 60 mg/cm^2 of stainless steel; 160-168 mg/cm^2 of penetration are required to breach the cladding. This would result in two 20-day dissolution cycles and a 10 to 15-day dissolution cycle to breach the elements. Furthermore, approximately five days would be required between each dissolution cycle to cool, transfer, reload, and heat the dissolver solution. The net result is a 60 to 65-day dissolution cycle with a three-fold increase in waste volume. This duration is still considered to be outside of the practical length of time (30 days) for dissolution of a batch of FCA fuel elements. It should be noted that the breaching of some elements does not guarantee that all elements breach at the same time or that all of the Pu dissolves readily from partially-breached elements. Hence, significant additional uncertainty exists regarding the duration of a full dissolution cycle beyond breaching of the fuel elements.

Another approach could involve dissolving with elevated fluoride levels using an alternate fluoride source, such as calcium fluoride (CaF_2), because of the 0.23M limit for using KF. Aside from the fact that using CaF_2 is more difficult for H-Canyon than KF, the use of high fluoride concentrations introduces a different process risk – that of fluoride salt precipitation. Of particular concern is plutonium fluoride (PuF_3 or PuF_4) precipitation, which can result in plutonium solids at the bottom of the H-Canyon dissolver.[12,13,14] This presents a considerable criticality hazard. Attempts to use complexing agents, such as aluminum, to prevent the formation of PuF_3 or PuF_4, would complex the fluoride ions and greatly diminish the stainless steel dissolution rate, thereby eliminating the benefit of using elevated fluoride concentrations.

The dissolution studies with several FCA simulant and stainless steel plates spaced 0.5 inches apart showed no significant difference between the dissolution rates of the individual plates due to spacing. Therefore, spacing of 0.5 inches between the FCA fuel elements in a rack of similar design should produce uniform dissolution rates for multiple samples.

The FCA cladding or stainless steel plates produced no significant H_2 offgas during dissolution. The gas generation does not challenge 60% of the lower flammability limit (LFL) for H_2. The results from these studies confirm that the FCA cladding does not contribute to the flammable off gas generation.

The dissolution of Pu has been examined in prior work.[1,2] The data are relevant although not necessarily identical to the data that would be obtained for Pu-10.6Al. These prior studies showed that for a charge of 46 kg of Pu with a geometry of thin sheets, the generated H_2 will be well below the 60% of the LFL with the standard 40 scfm of air for the H-Canyon dissolver.

5.0 Flowsheet Recommendations

The stainless steel cladding on FCA fuel elements can be dissolved in H-Canyon using standard HNO_3-KF flowsheets. Dissolution of 0.008-in of cladding is required to breach the cladding of 72% of the FCA fuel and expose the Pu-Al to chemical dissolution; the remaining 28% of the fuel elements were fabricated with 0.005-in cladding. Using 4M HNO_3, 0.25M KF, and 1.2 g/L B – which is the optimal conditions for HNO_3-KF in H-Canyon – it is estimated to take about 176 days to breach the fuel cladding at the facing of 2 inch by 2 inch by 0.0083 inch. It should be noted, though, that after the cladding is breached and the Pu-Al dissolved, pieces of the FCA fuel frames and the FCA fuel holders would need to be manually removed from the dissolver.

Because this is an unacceptably lengthy process cycle time, it is recommended that chemical dissolution of the FCA fuel be not pursued.

6.0 References

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Appendix A. Raman Calibration and Sampling Method

The Raman spectrometer was calibrated using a set of calibration gases as shown in Table 2-4. Due to the nature of the Raman technique, the instrument only needs to be calibrated once for the intensities (or quantities) of the calibration gases. The wavelengths for the various calibration gases are known and also remain fixed. The Raman calibration is also linear over its range. As an additional check before and after each experiment, air, 99.9 vol % CO_2, and/or a 2.67 vol % H_2 gas (balance Ar) were analyzed using the Raman cell to ensure the calibration was still good. If the calibration checks were off for these gases, the Raman calibration model was adjusted for those gases after the run. For the stainless steel dissolutions performed in Experiments 113, 114, 115, and 117, the calibration checks are shown in Table A-1. The calibration check indicates if the calibration was successful and provides an indication of the variance of the measurements since the calibration gas is analyzed for several samples.

The Raman readings should be positive and sum to 100% except for the 2.67 vol % H_2 gas which is 97.33 vol % Ar (which is not detected by the Raman spectrometer). Due to the noise in the Raman signal, any raw readings that are less than zero are fixed to zero and then all the gas readings for H_2, NO_2, N_2, O_2, N_2O, NO, CO_2, CO, H_2O and NH_3 are normalized to 100% except for the 2.67 vol % H_2 gas. These fixed and normalized Raman readings are the values reported in Table A-1 except for the 2.67 vol % H_2 gas where the raw readings are provided. Due to rounding to the nearest hundredth, the numbers in the table may not sum exactly to 100 vol % but all the decimal places were carried in the calculations performed for this report.

The total offgas flow is calculated from the fixed normalized sum of the CO_2 and CO concentrations divided into the CO_2 tracer flow rate coming into the system as shown in Table A-2. The noise in the concentrations measured by the Raman spectrometer propagates into the total offgas flow rate so moving averages of the total offgas flow rates were performed using equation A-1:

$$\text{Offgas flow rate}_{\,t_i}(\text{cm}^3/\text{min}) = \frac{\sum_{k=t_{i-1}}^{t_{i+1}} \text{Offgas flow rate}_k}{3} \qquad \text{(A-1)}$$

where Offgas flow rate = offgas generated by the dissolution in cm^3/min
t_i = time at integer time step i
k= integer time step t_{i-1}, t_i, and t_{i+1}.

The moving average offgas flow rates and fixed normalized moving average Raman offgas concentrations for Experiment 113, 114, 115, and 117 are shown in Appendix D, E, F and G, respectively.

Table A-1. Pre-run Check of Calibration Gases for Dissolution Experiments 113-117

Gas Description	Exp.	H_2 (%)	NO_2 (%)	N_2 (%)	O_2 (%)	N_2O (%)	NO (%)	CO_2 (%)	CO (%)	H_2O (%)	NH_3 (%)
99.9% CO_2	113	0.07	0.38	0.00	0.00	0.00	0.00	**99.12**	0.21	0.00	0.22
	113	0.00	0.19	0.47	1.03	0.00	0.17	**97.70**	0.00	0.00	0.42
	113	0.02	0.27	0.44	0.11	0.00	0.00	**98.18**	0.00	0.38	0.60
	113	0.12	0.25	0.44	0.91	0.00	0.00	**98.28**	0.00	0.00	0.00
	113	0.00	0.24	0.00	0.00	0.00	0.00	**98.85**	0.00	0.79	0.12
	113	0.02	0.19	0.00	0.20	0.00	0.00	**99.58**	0.00	0.00	0.00
	114	0.00	0.19	0.00	0.23	0.00	0.00	**99.17**	0.12	0.11	0.18
	114	0.35	0.22	0.00	0.35	0.00	0.00	**97.80**	0.84	0.00	0.45
	114	0.00	0.23	0.25	0.95	0.00	0.00	**97.37**	0.86	0.33	0.00
	114	0.00	0.33	0.28	0.28	0.00	0.22	**98.59**	0.00	0.30	0.00
	114	0.00	0.19	0.00	0.00	0.00	0.00	**98.09**	1.72	0.00	0.00
	114	0.00	0.24	0.55	0.67	0.00	0.00	**97.20**	0.58	0.34	0.42
	115	0.00	0.16	0.00	0.00	0.00	0.85	**97.64**	1.16	0.00	0.20
	115	0.00	0.31	0.20	0.16	0.00	0.00	**99.19**	0.00	0.13	0.00
	115	0.00	0.24	0.18	0.82	0.00	0.87	**97.67**	0.22	0.00	0.00
	115	0.00	0.30	0.55	0.00	0.00	0.00	**97.65**	0.00	0.77	0.73
	115	0.00	0.17	0.00	0.39	0.00	0.00	**99.25**	0.00	0.20	0.00
	115	0.00	0.18	0.44	0.26	0.00	0.44	**98.68**	0.00	0.00	0.00
	117	0.23	0.18	0.26	0.64	0.00	0.00	**98.38**	0.14	0.17	0.00
	117	0.46	0.27	0.19	0.00	0.00	0.00	**98.53**	0.00	0.00	0.55
	117	0.00	0.24	0.00	1.37	0.00	0.00	**97.60**	0.48	0.31	0.00
	117	0.00	0.25	0.35	0.00	0.00	0.00	**99.16**	0.00	0.25	0.00
	117	0.21	0.21	0.00	0.86	0.00	0.00	**98.61**	0.02	0.09	0.00
	117	0.20	0.22	0.12	0.85	0.00	0.00	**97.91**	0.00	0.14	0.56
Air (78.0% N_2-21.0% O_2-0.9% Ar)	113	0.00	0.05	**75.86**	**20.76**	0.00	0.00	3.15	0.00	0.17	0.00
	113	0.00	0.02	**74.25**	**19.38**	0.00	1.40	4.20	0.41	0.34	0.00
	113	0.48	0.00	**75.45**	**20.70**	0.00	0.00	2.44	0.85	0.07	0.00
	113	0.34	0.00	**75.27**	**20.39**	0.00	0.00	3.12	0.00	0.72	0.16
	113	0.34	0.00	**77.17**	**20.19**	0.00	0.62	1.41	0.27	0.01	0.00
	113	0.00	0.00	**76.59**	**19.94**	0.00	1.27	2.20	0.00	0.00	0.00
	114	0.16	0.00	**76.28**	**20.89**	0.00	0.00	1.38	0.83	0.08	0.37
	114	0.05	0.00	**74.57**	**23.70**	0.00	0.00	1.25	0.15	0.00	0.27
	114	0.19	0.00	**76.57**	**20.63**	0.00	0.00	2.17	0.23	0.21	0.00
	114	0.00	0.01	**74.17**	**23.62**	0.00	0.00	2.02	0.00	0.00	0.18
	114	0.11	0.01	**75.02**	**20.17**	0.00	0.36	3.18	0.45	0.71	0.00
	114	0.00	0.00	**77.20**	**20.83**	0.00	0.00	1.74	0.04	0.00	0.19
	115	0.00	0.02	**77.83**	**20.92**	0.00	0.00	1.24	0.00	0.00	0.00
	115	0.00	0.00	**74.22**	**21.65**	0.00	0.00	2.67	0.94	0.19	0.34
	115	0.00	0.00	**76.38**	**20.72**	0.00	1.69	0.00	0.88	0.33	0.00
	115	0.00	0.00	**74.55**	**21.10**	0.00	0.57	3.57	0.00	0.05	0.16
	115	0.22	0.00	**74.93**	**21.25**	0.00	0.00	2.60	0.65	0.00	0.36
	115	0.00	0.00	**74.70**	**23.29**	0.00	0.00	1.47	0.35	0.20	0.00
	117	0.00	0.02	**76.51**	**21.05**	0.00	0.00	2.22	0.19	0.00	0.00
	117	0.10	0.00	**77.76**	**20.17**	0.00	0.00	1.93	0.03	0.00	0.00
	117	0.00	0.02	**75.02**	**20.78**	0.00	0.00	2.93	0.91	0.34	0.00
	117	0.00	0.00	**76.20**	**20.58**	0.00	0.00	2.91	0.00	0.31	0.00
	117	0.25	0.00	**74.70**	**22.63**	0.00	0.00	2.35	0.00	0.00	0.06
	117	0.00	0.01	**76.92**	**20.62**	0.00	0.92	1.51	0.00	0.03	0.00

Gas Description	Exp.	H_2 (%)	NO_2 (%)	N_2 (%)	O_2 (%)	N_2O (%)	NO (%)	CO_2 (%)	CO (%)	H_2O (%)	NH_3 (%)
2.67% H_2-97.33% Ar	113	**3.15**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	113	**3.42**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	113	**2.67**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	113	**3.15**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	113	**3.07**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	113	**3.01**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	114	**3.33**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	114	**2.41**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	114	**2.94**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	114	**2.86**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	114	**3.35**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	114	**3.09**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	115	**3.32**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	115	**2.91**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	115	**2.56**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	115	**2.98**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	115	**2.72**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	115	**2.96**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	117	**2.92**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	117	**3.16**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	117	**2.28**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	117	**3.46**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	117	**2.35**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	117	**2.98**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

The values in Table A-1 should be constant since the Raman is reading constant sources (calibration gases) without being connected to the dissolution equipment. The variance seen in these values represent the instrument noise and should not be counted as part of the process noise or variance. To estimate the measurement uncertainty of the Raman spectroscopy, the pre-run check values of Table A-1 were compared to the standard values across Experiments 113 to 117. Sample standard deviations of the measured concentrations with respect to the calibrated concentrations for the data were calculated. These sample standard deviations were then doubled to get an idea of the variability in the Raman spectroscopy concentration measurements. Table A-3 shows the sample standard deviation of the measured concentrations with respect to their calibrated values. For the H_2 gas, the 2σ values or twice the standard deviation is 0.62 vol%. The 2σ value for O_2 and N_2 is < 5 vol % and the 2σ value for CO_2 is < 4 vol %.

Table A-2. Raman Tracer Gas Flow Rates

Experiment	CO_2 Flow
---	(cm^3/min)
113	20
114	20
115	20
117	20

Table A-3. Standard Deviation of Raman Concentrations with Respect to Calibrated Values

Gas	Standard Deviation (σ)	2*Standard Deviation (2σ)
---	(vol %)	(vol %)
CO_2	1.61	3.21
N_2	2.31	4.62
O_2	0.57	1.14
H_2	0.31	0.62

Appendix B. Volume to Surface Area Estimate for FCA Elements in H-Canyon Dissolver

To scale the lab experiments to the full-scale dissolver, the volume of solution to surface area of immersed stainless steel was chosen as the scale factor. This scale factor was chosen since the solution has a uniform F^- concentration and the F^- is what corrodes the stainless steel based on the surface area available.

To determine the volume of solution to stainless steel surface area for the H-Canyon dissolver, the following assumptions were made:

1. 8-ft tall Vessel of 8-ft ID filled to 7 ft or 7,500 Liter.
2. Steam Coils in Vessel from DP-500, Figure 7-11, has a surface area of 460 ft^2 or 427,354 cm^2.
3. 10 Well Inserts immersed 5 ft with 6.625 in OD and 5.501 in ID.
4. Endpoint concentration of 3.5 g/L Pu
5. Each FCA Element has 35.3 g Pu in a 2 in x 2 in x 1/16 in plate.
6. Each FCA Element Holder is 30 in (76.2 cm) tall with ¼ in spacing between plates (Figure B-1) and has
   a. 12 faces (inside and outside faces for 4 columns) of 2 cm x 76.2 cm
   b. 8 faces (edges of 4 columns) of 1/8 cm x 76.2 cm
   c. 1 base plate with 2 faces of 7.5 cm x 7.5 cm and with 4 edges of 7.5 cm x ¼ cm
   d. 190 pins that are 5.25 cm long and 0.318 cm OD
7. Sparge Ring of 2-in Sch 80 pipe (2.375 in OD) and 5-ft diameter
8. 8 Coil Supports of 1-in Sch 80 pipe (1.315 in OD) and 4-ft long

Figure B-1. Theoretical FCA Fuel Element Holder Front and Back View

Based on the assumptions above, the surface areas of the various pieces in the dissolver can be estimated as follows.

For the Dissolver Vessel, the ID is 8 ft (243.84 cm) and the solution height is 7 ft (213.36 cm) so the exposed surface area is:

$$Vessel\ SA = \frac{3.1416*243.84^2}{4} + 3.1416*243.84*213.36 = 46{,}698 + 163{,}444 = 210{,}142\ \text{cm}^2 \qquad \text{(B-1)}$$

For the well insert, the OD is 6.625 in (16.83 cm) and the ID is 5.501 in (13.97 cm) and when it is immersed 5 ft (152.4 cm) the exposed surface area is:

$$Well\ Insert\ SA = 3.1416*16.83*152.4 + 3.1416*13.97*152.4 = 8{,}058 + 6{,}689 = 14{,}746.4\ \text{cm}^2 \qquad \text{(B-2)}$$

So for 10 Well Inserts the total exposed surface area is 147,464 cm^2.

To reach an endpoint concentration of 3.5 g/L Pu in 7,500 L translates to 26,250 g Pu. Since each FCA element contains 35.3 g Pu then that translates into 744 FCA elements. The surface area of one FCA element is:

$$FCA\ Element\ SA = 2*2*2 + 2*\frac{1}{16}*4 = 8.5\ \text{in}^2 = 54.84\ \text{cm}^2 \qquad \text{(B-3)}$$

So for 744 FCA elements the total exposed surface area is 40,801 cm^2.

The FCA element holder that is 30 in (76.2 cm) tall has the following surface area terms:

$$FCA\ Holder\ Column\ Face\ SA = 2*76.2 = 152.4\ \text{cm}^2 \qquad \text{(B-4)}$$

$$FCA\ Holder\ Column\ Edges\ SA = \frac{1}{8}*76.2 = 9.525\ \text{cm}^2 \qquad \text{(B-5)}$$

$$FCA\ Holder\ Base\ Plate\ SA = 2*7.5*7.5 + 4*\frac{1}{4}*7.5 = 120\ \text{cm}^2 \qquad \text{(B-6)}$$

$$FCA\ Holder\ Pin\ SA = 3.1416*0.318*5.25 = 5.245\ \text{cm}^2 \qquad \text{(B-7)}$$

For each FCA element holder, the 12 column faces have a surface area of 1,828.8 cm^2, the 8 column edges have a surface area of 76.2 cm^2, the base plate has a surface area is 120 cm^2, and the 190 pins have a surface area of 996.5 cm^2, giving a total surface area of 3,021.5 cm^2. For 8 FCA element holders the total surface area is 24,172 cm^2.

The sparge ring is 2.375 in (6.0325 cm) OD and 5-ft (152.4 cm) in diameter. To calculate its surface area one needs to convert the 152.4 cm diameter into a length which is the circumference or π*diameter. The surface area for the sparge ring then is calculated as follows:

$$Sparge\ Ring\ SA = 3.1416*6.0325*(3.1416*152.4) = 9{,}074\ \text{cm}^2 \qquad \text{(B-8)}$$

The coil supports are 1.315 in (3.34 cm) OD and 4 ft (121.92 cm) long. Each coil support has a surface area as follows:

$$Coil\ Support\ SA = 3.1416*3.34*121.92 = 1{,}279.3\ \text{cm}^2 \qquad \text{(B-9)}$$

For the 8 coil supports that gives a total surface area of 10,234 cm^2.

Combing all the surface area terms defined above gives a total surface area of 869,241 cm^2 (Table B-1). This surface area is based on 7,500 L or 7,500,000 mL in the dissolver, so the volume to surface area for the FCA elements is approximately 8.6 mL/cm^2.

Table B-1. Surface Area Estimate for FCA Fuel Elements in H-Canyon Dissolver

Item	Count	Surface Area (cm^2)
Vessel	1	210,142
Well Insert	10	147,464
FCA Element	744	40,801
FCA Element Holder	8	24,172
Steam Coils	---	427,354
Sparge Ring	1	9,074
Coil Support	8	10,234
Total	---	869,241

Appendix C. ICPES Data for Dissolutions

Table C-1. ICPES Analyses for Experiments 113 and 114

Experiment	113	113	113	114	114	114
Sample Time	1 hr	3 hr	8 hr	1 hr	3 hr	8 hr
Sample ID:	LW6910	LW6911	LW6912	LW6914	LW6915	LW6916
Units:	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
Al	6.05	8.24	9.91	< 4.82	< 4.82	< 4.82
B	996	989	955	1190	1180	1160
Cr	13.5	27.7	53.8	5.65	13.1	29.4
Fe	55.9	114	218	23.9	55	121
K	8770	8610	8290	3840	3780	3680
Mn	0.933	1.97	3.98	0.366	0.88	2.08
Na	19.4	24.5	27.8	7.45	9.49	10.9
Ni	7.16	14.6	28.2	< 10.5	< 10.5	15.6
Si	174	189	184	25.2	30.4	28.1
Solution Volume (ml)	250	248.07	246.28	250	248.1	246.29

Table C-2. ICPES Analyses for Experiments 115 and 116

Experiment	115	115	115	116	116	116
Sample Time	1 hr	3 hr	8 hr	1 hr	3 hr	8 hr
Sample ID:	LW6918	LW6919	LW6919	LW6922	LW6923	LW6924
Units:	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
Al	17.7	24.5	27.7	3.55	4.9	6.07
B	1260	1270	1300	1220	1220	1230
Cr	39.5	75.6	149	7.94	17.4	36.9
Fe	156	299	593	34.1	72.2	149
K	< 131	< 131	< 131	3880	3870	3870
Mn	2.43	4.98	10.9	0.515	1.19	2.64
Na	9250	9210	9340	13.9	15	17
Ni	20.5	39.7	76.7	< 10.5	9.31	19.6
Si	513	706	798	60.6	45	45.7
Solution Volume (ml)	250	247.98	245.74	250	247.94	246.12

Table C-3. ICPES Analyses for Experiment 117

Experiment	117	117	117
Sample Time	1 hr	3 hr	8 hr
Sample ID:	LW6926	LW6927	LW6928
Units:	mg/L	mg/L	mg/L
Al	12.9	15.7	19.9
B	1440	1270	1280
Cr	23.4	46.2	107
Fe	93	183	418
K	< 131	< 131	< 131
Mn	1.14	2.89	7
Na	10500	9290	9930
Ni	10.4	24.1	54.8
Si	249	227	333
Solution Volume (ml)	250	247.94	246.12

Table C-4. ICPES Analyses for Experiments 119 and 120

Experiment	119	119	119	120	120	120
Sample Time	1 hr	3 hr	8 hr	1 hr	3 hr	8 hr
HNO_3(M)	4	4	4	4	4	4
F(M)	0.25	0.25	0.25	0.25	0.25	0.25
Al(M)	0	0	0	0.0074	0.0074	0.0074
Sample ID:	LW6930	LW6931	LW6932	LW6934	LW6935	LW6936
Units:	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
Al	8.95	12.6	14.2	209	212	215
B	1060	1110	1120	967	973	979
Cr	23.2	44.4	85.7	21.7	40.7	77.6
Fe	101	190	368	95.2	175	332
K	9070	9190	9220	8740	8690	8710
Mn	1.88	3.69	7.19	1.77	3.36	6.52
Na	24.5	31.8	35.1	20.7	26.9	30.5
Ni	12.1	23.2	45	11.4	21.2	40.8
Si	263	370	424	214	305	323
Solution Volume (ml)	250	247.81	245.67	240	238.07	235.95

Appendix D. Raman Offgas Data for Experiment 113

As discussed in Appendix A, the Raman readings should be positive and sum to 100% except in cases where there is significant Ar present. Due to the noise in the Raman signal, any raw readings that are less than zero are fixed to zero and then all the gas readings for H_2, NO_2, N_2, O_2, N_2O, and NO are normalized to 100 vol %. Even with these corrections, the Raman readings have noise in them. To reduce this noise, moving averages of the fixed and normalized readings were performed using equation D-1:

$$\text{Gas}_{t_i}(\text{vol\%}) = \frac{\sum_{k=t_{i-2}}^{t_{i+2}} \text{Gas}_k}{5} \quad \text{(D-1)}$$

where Gas = H_2, NO_2, N_2, O_2, N_2O, NO concentrations (vol %)
t_i = time at integer time step i
k= integer time step t_{i-2}, t_{i-1}, t_i, t_{i+1}, and t_{i+2}.

These moving averages do not eliminate all the noise but smooth the values so comparisons and calculations can be performed. Due to the length of the experiment, 8 hours, and the sampling of the Raman about every 11-12 seconds, approximately every 10 points or 2 minutes of the fixed, normalized, and moving average Raman gas concentrations are reported in Table D-1. A complete set of the data is maintained in SRNL ELN. Due to rounding to the nearest hundredth, the numbers in the table may not sum to exactly 100 vol % but all the decimal places were carried in the calculations performed for this report.

Table D-1. Fixed Normalized Moving Average Raman Data for Experiment 113

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
0	96.64	0.57	3.47	10.17	58.74	15.55	2.72	9.34
125	96.9	0.64	10.09	13.41	36.46	30.83	0	9.2
237	97.23	0.57	1.62	10.17	50.22	27.18	7.53	3.28
363	96.58	0.71	8.86	9.59	39.82	28.29	9.98	3.45
477	96.15	0.8	7.03	7.75	41.57	24.43	7.87	11.36
553	97.13	0.59	6.69	12.03	46.52	29.39	0.24	5.12
865	97.19	0.58	3.07	11.97	55.33	24.26	0.24	5.12
989	96.81	0.66	4.82	9.13	25.19	45.09	9	6.77
1103	97.78	0.45	0	15.58	40.39	34.42	8.3	1.3
1229	97.58	0.5	3.92	15.19	34.18	44.19	2.51	0
1343	97.13	0.59	6.76	13.32	42.39	18.97	5.08	13.47
1470	97.35	0.55	5.44	16.04	41.26	21.66	3.39	12.2
1586	96.79	0.67	0	20.47	21.97	33.22	3.64	20.7
1703	97.25	0.57	2.9	13.56	36.52	27.4	4.77	14.85
1832	96.58	0.72	3.48	12.99	32.46	34.31	12.38	4.38
1950	97	0.62	4.88	10.88	16.42	33.4	1.86	32.56

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
2068	97.34	0.55	8.38	13.3	41.8	20.06	1.29	15.18
2187	97.91	0.43	5.24	18.12	42.41	30.41	1.5	2.31
2306	98.4	0.33	4.49	22.21	24.73	20.43	10.6	17.55
2427	97.83	0.44	5.34	12.98	40.14	8.44	9.55	23.56
2547	96.8	0.66	6.47	11.5	44.13	27.16	2.29	8.45
2668	96.8	0.66	4.99	10.34	10.29	32.94	6.32	35.12
2791	98.55	0.29	5.18	25.67	37.96	13.13	10.05	8.01
2913	98.53	0.3	15.86	19.88	25.99	31.05	1.93	5.3
3023	98.21	0.37	4.38	19.05	11.39	8.87	21.06	35.25
3147	97.92	0.43	6.05	12.83	12.78	29.43	7.85	31.06
3271	97.87	0.44	7.5	19.31	37.47	31.89	3.47	0.36
3383	98.1	0.39	1.67	22.05	26.22	27.06	5.36	17.64
3508	97.03	0.61	6.62	12.66	31	27.38	13.41	8.93
3633	98.38	0.33	4.4	15.03	25.51	28.54	5.33	21.19
3747	96.26	0.78	7.01	13.97	36.89	22.76	9.13	10.24
3848	97.19	0.58	5.35	20.25	16.37	21.8	23.45	12.79
5010	97.68	0.48	5.59	16.86	22.54	24.73	18.49	11.79
5135	96.61	0.7	3.22	11.27	30.97	16.94	13.91	23.69
5248	96.97	0.63	4.1	12.97	22.24	37.23	4	19.45
5373	97.94	0.42	2.48	15.94	11.65	39.19	8.81	21.94
5488	97.98	0.41	0	16.53	32.28	30.64	0.19	20.36
5614	97.64	0.48	6.42	17.93	28.8	29.46	5.07	12.32
5730	98.06	0.4	2.1	17.08	22.26	36.13	16.6	5.82
5847	98.55	0.3	0.5	22.04	24.65	23.64	19.3	9.87
5976	97.61	0.49	2.87	13.47	14.33	42.47	15.26	11.6
6093	97.68	0.48	10.28	11.66	25.65	26.26	6.39	19.77
6212	97.66	0.48	6.21	11.65	12.14	15.16	13.81	41.04
6330	97.31	0.56	4.39	14.56	28.88	25.8	13.86	12.51
6450	97.11	0.6	9.9	14.03	25.14	18.2	12.5	20.23
6570	98.37	0.33	3.76	24.44	26.77	31.09	1.07	12.87
6690	97.8	0.45	3.33	14.29	27.06	27.99	9.57	17.76
6812	97.56	0.5	5.2	20.81	23.49	26.5	15.34	8.67
6933	97.83	0.44	0.3	20.49	32.34	10.19	13.12	23.55
7056	98.07	0.39	4.89	12.13	25.93	25.82	8.85	22.38
7166	97.83	0.45	12.52	13.38	25.87	32.3	7.59	8.35
7178	98.25	0.36	6.46	13.69	29.12	27.71	5.5	17.53
7289	97.22	0.57	2.22	12.65	29.76	17.53	18.09	19.75

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
7413	97.49	0.51	13.5	11.84	7.64	27.6	11.69	27.72
7538	97.45	0.52	0.9	12.41	29.83	30.2	7.03	19.62
7650	98.52	0.3	0	21.96	30.59	41.14	6.31	0
7776	98	0.41	1.69	11.78	27.96	28.94	7.81	21.83
7889	97.99	0.41	0	12.38	19.85	30.81	13.79	23.18
7990	98.18	0.37	0	22.65	39.34	22.37	6.96	8.68
8841	98.41	0.32	0	24.3	31.11	26.59	6.96	11.04
8966	97.85	0.44	2.77	24.47	22.93	21.52	8.7	19.62
9079	98.2	0.37	9.77	13.9	21.59	21.64	15.12	17.98
9204	98.31	0.35	0	25.75	27.85	16.6	21.31	8.5
9318	98.39	0.33	0.55	31.94	16.51	19.69	16.73	14.57
9444	97.63	0.49	0.25	15.21	30.25	21.3	4.99	27.99
9559	97.66	0.48	6.04	11.9	25.07	29.97	11.25	15.77
9687	98.23	0.36	9.65	17.86	11.86	39.74	7.17	13.72
9803	97.91	0.43	3.51	24.63	29.83	17.87	15.31	8.85
9920	97.66	0.48	11.56	12.74	17.7	30.49	20.88	6.63
10038	98.14	0.38	2.12	24.62	12.47	11.63	11.94	37.22
10168	97.97	0.41	5.55	14.45	15.35	22.77	16.61	25.27
10287	98.11	0.39	7.71	17.76	25.5	35.86	4.32	8.85
10406	98.45	0.32	11.87	20.11	9.86	14.63	24.42	19.12
10526	97.95	0.42	5.12	18.05	27.99	21.58	15.32	11.94
10647	96.77	0.67	15.53	11.13	31.18	20.86	12.44	8.86
10768	98.23	0.36	9.96	18.29	32.02	17.44	12.28	10
10890	98.03	0.4	2.36	19.16	18.85	41.64	6.16	11.82
10902	98.46	0.31	2.31	21.08	16.98	31.93	13.87	13.83
11012	98.32	0.34	2.86	23.94	24.82	33.81	14.56	0
11135	97.5	0.51	7.68	15.83	14.31	25.51	18.71	17.96
11246	97.08	0.6	4.92	13.89	21.46	22.67	15.67	21.39
11258	97.45	0.53	3.46	10.25	27.35	31.28	14.41	13.26
11377	98.51	0.3	1.28	22.86	29.72	24.34	17.07	4.73
11490	98.12	0.38	6.76	20.21	20.55	18.63	20.4	13.46
11614	97.34	0.55	9.99	15.93	14.92	21.56	17.32	20.28
11728	98.29	0.35	11.47	26.07	17.94	19.27	24.13	1.13
11854	98.27	0.35	0.57	27.79	15.98	19.24	11.61	24.81
11969	98.24	0.36	2.12	16.08	25.3	23.81	17.52	15.17
12096	98.47	0.31	2.25	29.49	30.37	14.43	21.86	1.6
12225	97.77	0.46	0	14.87	34.5	27.83	18.12	4.68

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12343	97.2	0.58	2.58	10.2	24.27	24.22	10.66	28.08
12461	97.71	0.47	0.6	12.55	26.73	28.27	20.53	11.32
12579	98.33	0.34	2.87	19.9	23.53	25.23	22.12	6.35
12698	98.3	0.35	2.76	14.76	8.31	31.33	16.72	26.12
12817	98.16	0.37	2.26	15.81	33.36	28.72	16.28	3.57
12937	98.56	0.29	0	18.98	24.79	26.72	28.41	1.11
12949	98.65	0.27	0.26	17.7	25.95	30.45	24.53	1.11
13069	97.41	0.53	2.43	11.85	11.95	31.04	27.62	15.12
13081	97.74	0.46	2.43	11.47	10.85	34.15	26.87	14.23
13202	97.75	0.46	2.82	17.41	27.92	28.62	13.85	9.36
13324	98.2	0.37	4.9	19.35	23.28	23.26	20.65	8.55
13446	97.49	0.51	7.64	12.63	15.74	22.78	13.87	27.34
13569	98.07	0.39	8.6	18.51	23.68	19.09	25.29	4.83
13692	98.16	0.38	3.71	15.58	28.07	19.68	16.82	16.14
13816	97.84	0.44	7.72	15.1	28.75	21.09	17.21	10.13
13940	97.65	0.48	2.88	17.74	20.3	19.44	32.74	6.91
14065	98.63	0.28	8.02	24.78	25.4	14.93	14.92	11.95
14191	97.8	0.45	8.57	13.14	30.63	16.62	15.17	15.87
14440	98.38	0.33	5.13	17.61	30.61	13.16	11.76	21.74
14553	97.92	0.42	2.37	17.2	29.64	20.8	21.72	8.27
14667	97.4	0.53	3.62	11.97	28.68	18.89	22.98	13.87
14781	97.79	0.45	1.77	15.15	21.38	28.78	22.01	10.91
14896	97.35	0.55	3.71	21.73	12.75	27.59	10.45	23.77
15011	97.67	0.48	6.84	15.52	40.57	18.49	10.18	8.39
15127	97.57	0.5	10.19	17.45	29.11	27.56	8.36	7.33
15243	97.67	0.48	0.39	11.7	39.47	14.11	6.28	28.05
15361	97.91	0.43	10.49	17.17	23.18	27.39	6.79	14.98
15478	98.3	0.35	6.34	17.16	26.13	26.92	13.09	10.36
15596	98.38	0.33	3.83	15.89	27	15.08	20.11	18.09
15715	97.73	0.47	1.35	30.89	2.35	20.82	10.84	33.74
15834	97.89	0.43	2.11	13.92	24.48	23.8	20.98	14.71
15954	98.88	0.23	2.91	23.64	36.17	17.36	11.32	8.6
16073	97.58	0.5	1.59	18.34	16.74	33.02	16.62	13.69
16194	98.02	0.4	7.87	16.63	19.69	25.36	16.58	13.87
16315	98.11	0.39	4.89	16.29	21.05	26.12	8.6	23.06
16436	97.99	0.41	8.86	15.09	31.12	22.61	16.56	5.76
16558	98.25	0.36	0.2	22.58	21.55	13.31	27.26	15.1

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
16681	98.17	0.37	1.68	15.58	25.09	24.21	20.43	13.02
16804	98.26	0.35	5.24	20.34	14.12	43.9	11.03	5.37
16928	98.31	0.34	3.61	15.68	12.93	35.43	16.14	16.22
17052	97.47	0.52	4.1	14.77	22.54	30.09	20.35	8.15
17177	98.4	0.32	5.36	21.33	7.13	19.52	32.08	14.57
17303	98.32	0.34	13.25	18.79	13.29	22.77	14.05	17.85
17572	97.75	0.46	2.59	16.32	26.55	18.64	21.7	14.2
17684	98.48	0.31	3.86	17.09	27.34	19.35	22.32	10.03
17797	98.16	0.38	4.78	16.19	12.31	27.8	23.21	15.71
17911	98.29	0.35	4.77	14.9	32.69	20.27	23.59	3.79
18025	98.19	0.37	6.53	15.41	24.36	21.57	19.4	12.73
18140	97.74	0.46	1.65	12.24	47.36	21.21	15.82	1.71
18255	97.25	0.57	7.22	11.97	30.42	14.94	16.99	18.46
18371	97.52	0.51	7.35	15.57	27.37	16.42	15.42	17.88
18487	97.67	0.48	1.98	10.58	36.87	17	20.91	12.67
18604	97.99	0.41	3.71	16.28	21.39	24.87	24.54	9.21
18722	97.44	0.53	0.77	14.83	21.8	24.98	24.41	13.21
18840	98.23	0.36	3.9	18.96	10.17	15.83	32.35	18.8
18958	97.25	0.57	8.2	12.37	19.58	27.74	18.45	13.67
19078	98.13	0.38	5.71	14.6	32.08	15.72	8.62	23.26
19198	98.11	0.39	11.22	15.28	6.46	32.08	22.36	12.61
19318	97.91	0.43	1.23	22.07	19.81	26.74	7.14	23.01
19439	98.08	0.39	5.45	10.31	17.07	17.84	20.4	28.93
19561	98.48	0.31	1.28	24.8	9.19	49.76	6.69	8.28
19684	98.16	0.37	11.72	15.09	25.27	15	20.18	12.74
19808	97.91	0.43	0	16.92	30.06	19.74	18.99	14.29
19933	98.08	0.39	6.01	15.38	19.83	29.98	21.55	7.25
20058	97.7	0.47	3.57	16.41	31.06	8.86	23.45	16.65
20183	98.27	0.35	3.31	13.88	40.05	21.8	16.96	4.01
20308	98	0.41	3.32	14.85	29.6	19.7	12.8	19.74
20434	98.04	0.4	1.2	18.02	17.71	27.46	12.48	23.13
20867	97.38	0.54	1.42	13.43	28.17	21.89	8.58	26.51
20980	98.21	0.37	16.57	15.18	16.21	21.43	17.58	13.03
21093	97.07	0.6	6.61	12.32	24.82	16.27	19.95	20.02
21206	97.9	0.43	1.8	12.41	24.77	18.62	15.63	26.77
21320	97.81	0.45	5.58	17.25	32.25	11.15	23.12	10.64
21435	98.42	0.32	6.31	13.24	26.95	18.77	24.63	10.1

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
21550	96.76	0.67	11.68	9.57	27.5	17.79	11.15	22.31
21666	97.89	0.43	6.14	17.78	15.53	32.19	10.69	17.67
21783	98.28	0.35	6.16	15.84	27.75	26.23	9.88	14.14
21900	97.91	0.43	0.84	14.38	22.93	22.88	20.74	18.24
22017	98.1	0.39	7.25	12.94	28.46	35.85	11.13	4.37
22135	97.26	0.56	4.22	8.73	18.24	18.38	21.94	28.5
22254	98.7	0.26	3.88	19.66	15.92	37.17	15.99	7.38
22373	98.41	0.32	5.4	16.43	14.67	23.55	20.55	19.41
22493	98.04	0.4	5.54	15.27	22.93	17.53	22.1	16.63
22613	97.63	0.49	0	12.81	15.92	23.72	19.57	27.99
22734	97.28	0.56	2.32	12.54	28.59	17.36	20.23	18.96
22856	97.56	0.5	10.66	18.49	12.26	18.02	22.48	18.09
22978	97.8	0.45	4.91	14.14	23.83	17.97	22.97	16.18
23100	98.5	0.31	6.81	29.68	23.38	9.36	22.34	8.43
23223	98.48	0.31	8.05	25.33	2.13	23.61	27.84	13.04
23347	98.23	0.36	5.51	16.02	25.97	38.68	8.11	5.71
23471	97.57	0.5	4.43	10.18	23.33	25.04	21.8	15.21
23596	97.47	0.52	2.37	14.5	19.4	18.72	24.01	21
23722	98.35	0.34	0.71	27.6	10.64	15.31	20.1	25.64
24557	98.46	0.31	4.72	16	11.7	32.82	24.62	10.14
24669	97.36	0.54	4.12	13.74	31.69	18.41	10.77	21.27
24782	97.83	0.44	3.27	12.95	11.8	38.76	17.51	15.72
24896	98.62	0.28	2.41	19.47	11.27	24.61	35.73	6.51
25010	97.68	0.48	3.99	14.53	4.66	31.03	23.7	22.09
25125	97.96	0.42	0.73	12.59	23.98	13.77	24.48	24.45
25240	98.21	0.36	4.1	17.45	3.9	51.94	14.64	7.98
25356	98.03	0.4	0.56	21.16	9.19	29.54	20.11	19.45
25472	97.3	0.56	3.08	11.12	22.53	23.08	19.06	21.12
25589	98.32	0.34	11.2	25.46	5.58	31.38	13.23	13.14
25707	97.79	0.45	13.35	13.98	31.76	12.6	23.93	4.37
25825	98.56	0.29	0.56	21.94	21.38	19.63	25.8	10.7
25944	98.1	0.39	6	15.03	17.81	32.02	22.59	6.56
26063	98.4	0.32	17.19	17.78	19.39	16.26	20.77	8.61
26183	97.85	0.44	14.16	13.24	18.68	19.24	18.04	16.63
26303	98.1	0.39	5.93	13.47	10.91	30.97	22.43	16.29
26424	98.16	0.38	2.79	17.87	15.92	13.28	32.94	17.2
26546	97.7	0.47	1.5	11.14	12.04	34.28	20.65	20.38

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
26669	98.11	0.39	6.94	13.2	28.24	8.32	19.66	23.63
26792	98.01	0.41	12.08	14.84	25.87	16.21	20.78	10.22
26916	98.46	0.31	9.18	17.28	7.85	20.15	25.54	20
27040	98.36	0.33	1.74	17.38	13.51	23.71	28.98	14.68
27165	98.73	0.26	3.23	21.31	12.73	19.36	30.4	12.98
27290	98.27	0.35	7.15	15.27	20.12	19.05	17.26	21.15
27416	97.85	0.44	1.88	12.03	9.65	35.79	24.84	15.81
27706	97.59	0.5	0.66	15.27	22.15	18.81	31.38	11.72
27818	97.49	0.51	2.28	12.84	7.5	29.05	33.59	14.75
27931	97.82	0.45	3.74	14.81	18.25	21.98	18.25	22.97
28045	98.53	0.3	3.96	19.92	35.34	18.02	14.89	7.87
28159	97.84	0.44	9.42	11.39	22.73	23.49	22.96	10.01
28274	98.06	0.4	5.48	14.09	22.52	16.27	23.55	18.09
28389	98.53	0.3	5.96	25.55	8.23	21.24	27.61	11.4
28505	98.43	0.32	8.06	13.77	25.4	32.61	12.78	7.39
28621	98.47	0.31	4.67	15.34	19.06	27.37	22.44	11.12
28738	98.28	0.35	6.7	13.04	19.53	11.69	19.04	29.99
28856	97.75	0.46	4.4	12.62	22.1	28.85	21.36	10.66
28974	97.81	0.45	5.55	15.1	35.61	23.01	14.81	5.92
29093	97.67	0.48	7.7	11.36	16.75	29.62	27.13	7.43
29212	97.22	0.57	5.72	15.06	17.64	21.72	23.36	16.49

Appendix E. Raman Offgas Data for Experiment 114

As discussed in Appendix A, the Raman readings should be positive and sum to 100% except in cases where there is significant Ar present. Due to the noise in the Raman signal, any raw readings that are less than zero are fixed to zero and then all the gas readings for H_2, NO_2, N_2, O_2, N_2O, and NO are normalized to 100 vol %. Even with these corrections, the Raman readings have noise in them. To reduce this noise, moving averages of the fixed and normalized readings were performed using equation D-1.

These moving averages do not eliminate all the noise but smooth the values so comparisons and calculations can be performed. Due to the length of the experiment, 8 hours, and the sampling of the Raman about every 11-12 seconds, approximately every 10 points or 2 minutes of the fixed, normalized, and moving average Raman gas concentrations are reported in Table E-1. A complete set of the data is maintained in SRNL ELN. Due to rounding to the nearest hundredth, the numbers in the table may not sum to exactly 100 vol % but all the decimal places were carried in the calculations performed for this report.

Table E-1. Fixed Normalized Moving Average Raman Data for Experiment 114

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
0	97.31	0.56	2.07	9.99	56.23	24.6	0	7.11
123	96.74	0.68	1.23	8.97	54.25	27.83	0	7.73
246	96.88	0.64	10.56	7.93	54.18	16.85	2.11	8.37
370	97.03	0.61	4.28	8.67	52.35	29.36	0	5.35
495	97.56	0.5	8.87	10.81	53.59	22.4	0	4.34
620	97.45	0.53	4.28	9.49	45.48	34.2	0	6.56
778	97.71	0.47	2.24	10.16	38.06	43.1	0	6.44
892	97.37	0.54	5.06	12.06	48.18	34.69	0	0
1005	97.29	0.56	2.52	10.57	38.71	36.21	1.92	10.07
1119	97.39	0.54	9.62	9.57	37.22	34.71	0.73	8.15
1233	98.17	0.37	2.65	12.3	50.5	20.66	0	13.9
1348	97.53	0.51	2.9	11.34	53.55	19.63	0	12.57
1463	97.65	0.48	5.58	12.11	49.99	31.76	0	0.56
1578	98.06	0.4	1.33	13.64	53.05	23.9	0	8.07
1695	97.84	0.44	6.78	13.82	25.64	35.71	1.41	16.63
1812	97.04	0.61	1.51	11.99	38.66	38	0	9.84
1929	97.65	0.48	1.85	11.17	35.58	26.03	0.89	24.48
2047	97.77	0.46	5.63	12.89	29.29	29.13	0.03	23.04
2166	98.11	0.39	2.04	14.86	63.38	16.28	0	3.43
2285	97.84	0.44	1.92	13.09	46.57	22.2	0	16.23
2404	97.43	0.53	2.34	12.45	34.5	28.95	11.27	10.49
2525	98.25	0.36	3.7	15.28	44.32	20.67	3.72	12.3

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
2646	97.43	0.53	3.28	13.84	55.7	19.67	0.08	7.43
2767	97.84	0.44	6.09	15.23	36.39	33.2	1.05	8.03
2889	97.56	0.5	2.26	14.44	30.96	22.61	3.21	26.51
3012	97.55	0.5	0	15.23	41.06	33.33	1.31	9.06
3135	98	0.41	6.66	11.88	36.33	24.67	0	20.46
3258	97.7	0.47	2.56	16.87	44.23	33.73	2.61	0
3383	97.84	0.44	7.44	15.78	29.44	37.94	5.07	4.34
3508	97.72	0.47	5.29	15.28	33.98	33.14	6.42	5.89
3633	98.42	0.32	8.35	16.01	22.93	20.51	11.52	20.69
4074	97.44	0.53	6.33	9.13	31.77	34.72	6.29	11.75
4187	97.79	0.45	3.82	13.1	38.04	30.8	4.14	10.1
4300	98.4	0.32	8.07	13.19	40.49	14.17	1.93	22.16
4414	97.52	0.51	1.31	12.55	41.51	31.28	4.26	9.09
4528	97.66	0.48	0.67	16.6	43.67	24.13	6.04	8.89
4643	97.73	0.47	8.39	17.87	39.68	22.59	4.33	7.14
4758	98.5	0.3	0	16.03	57.12	24.92	0	1.93
4873	97.7	0.47	0.5	17.37	48.2	9.04	2.78	22.12
4990	97.57	0.5	5.21	14.65	46.85	14.52	2.75	16.01
5107	97.79	0.45	4.93	12.5	30.94	21.75	7.55	22.34
5224	96.94	0.63	12.07	17.87	19.33	33.99	2.73	14.02
5342	98.76	0.25	0	25.99	28.55	25.28	8.66	11.51
5461	97.35	0.54	3.52	11.21	50.58	14.73	5.4	14.57
5580	97.88	0.43	12.31	15.41	29.18	36	5.75	1.35
5700	98.96	0.21	16.04	20.57	23.07	7.95	1.73	30.65
5821	98.47	0.31	1.3	24.13	36.15	10.53	10.72	17.17
5942	97.53	0.51	4.84	14.97	30.87	23.77	4.85	20.7
6064	98.26	0.35	6.21	26.28	36	28.63	0.61	2.27
6186	98.29	0.35	2.48	16.33	34.57	26.48	20.13	0
6310	97.63	0.49	0	14.72	35.64	7.65	2.13	39.86
6433	97.43	0.53	11.63	11.34	30.68	24.13	6.6	15.62
6558	97.79	0.46	5.45	18.08	29.74	28.13	6.17	12.43
6682	98.34	0.34	6.05	14	41.25	19.32	0	19.38
6808	98.11	0.39	2.38	30.1	40.9	10.85	10.17	5.6
6934	98.74	0.25	1.77	19.04	40.18	22.31	3.55	13.14
7123	97.54	0.5	3.72	12.06	43.79	17.9	0.06	22.46
7237	98.72	0.26	7.07	16.64	44.51	18.96	5.56	7.25
7351	98.76	0.25	0	35.95	26.44	25.92	8.7	2.99

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
7465	97.74	0.46	5.82	15.67	43.76	23.19	5.06	6.49
7580	97.72	0.47	1.1	17.44	35.76	32.14	1.43	12.14
7696	97.97	0.42	15.72	15.88	23.56	12.58	8.87	23.39
7811	98.32	0.34	5.66	23.52	11.42	46.83	8.49	4.07
7928	98.51	0.3	1.59	28.97	17.57	41.62	8.82	1.44
8045	97.92	0.42	2.52	11.14	31.65	22.9	9.6	22.19
8162	98.38	0.33	1.01	23.99	15.75	30.7	11.61	16.93
8280	98.32	0.34	1.59	18.42	17.59	43.66	17.12	1.62
8399	97.85	0.44	1.62	16.76	19.08	42.17	3.17	17.21
8518	97.9	0.43	4.29	14.84	28.67	28.89	14.68	8.63
8638	98.4	0.32	0	17.44	35.46	31.96	6.49	8.66
8758	97.82	0.45	3.5	14.45	12.91	38.4	10.82	19.92
8879	98.69	0.27	11.15	20.03	31.23	33.08	4.5	0
9001	98.5	0.31	9.85	16.36	26.23	39.71	4.98	2.88
9123	98.45	0.32	4.57	15.79	31.58	18.89	2.14	27.02
9245	98.81	0.24	7.81	20.35	25.33	21.36	1.26	23.89
9368	97.81	0.45	5.12	10.37	22.92	32.89	5.2	23.5
9492	97.22	0.57	3.91	11.77	38.59	16.83	9.98	18.92
9617	98.19	0.37	2.4	16.23	28.55	28.08	8.96	15.78
9742	98.12	0.38	4.22	14.32	30.6	27.68	7.85	15.34
9867	97.48	0.52	3.74	16.06	22.57	26.41	16.68	14.54
9993	98.55	0.29	0.8	30.76	44.18	14.55	2.38	7.34
10512	98.18	0.37	3.95	13.12	18.29	40.78	10.29	13.56
10625	97.58	0.5	1.96	17.78	33.53	17.04	10.31	19.37
10739	98.46	0.31	16.68	17.77	23.09	22.15	7.67	12.63
10853	97.92	0.43	0.6	20.29	31.59	16.79	23.93	6.79
10967	98.43	0.32	7.46	18.76	24.21	24.06	12.75	12.76
11083	97.08	0.6	7.87	11.31	35.33	17.31	12.51	15.66
11198	98.71	0.26	10.9	21.52	26.47	20.91	7.11	13.08
11314	97.75	0.46	1.45	12.52	44.89	23.33	7.98	9.83
11431	98.83	0.24	5.24	20.73	23.42	31.34	12.41	6.85
11548	98.27	0.35	7.61	19.03	29.05	24.99	6.17	13.15
11665	98.68	0.27	7.19	20.36	11.54	27.72	3.29	29.89
11783	98.28	0.35	1.28	16.21	20.91	32.78	12.99	15.81
11902	97.74	0.46	0.36	12.27	29.27	29.37	4.22	24.51
12022	98.32	0.34	1.33	22.76	18.31	44.09	2.49	11.02
12142	98.01	0.41	8.1	13.29	19.03	44.1	12.83	2.65

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12263	98.1	0.39	1.68	13.95	22.22	25.83	16.39	19.93
12385	97.49	0.52	4.03	14.77	25.14	39.03	9.4	7.64
12507	97.54	0.5	8.83	14.26	34.3	21.8	5.06	15.75
12629	98.48	0.31	1.57	16.85	20.54	32.3	2.03	26.7
12752	98.36	0.33	4.79	15.82	22.91	30.88	4.39	21.22
12876	97.79	0.45	3.31	13.69	49.92	22.64	4.78	5.65
13054	98.64	0.28	3.25	30.53	22.89	24.38	9.47	9.48
13167	98.24	0.36	5.31	16.4	27.05	13.59	11.61	26.05
13280	97.04	0.62	15.56	12.34	19.23	31.3	4.1	17.48
13394	98.5	0.31	10.6	24.12	37.69	15.13	5.05	7.4
13509	98.93	0.22	17.7	29.61	8.91	16.86	9.79	17.13
13624	98.11	0.39	7.49	15.95	13.03	44.29	15.92	3.33
13740	98.54	0.3	4.06	21.51	37	23.43	7.49	6.52
13856	98.72	0.26	1.94	21.04	29.46	21.74	10.76	15.07
13973	98.37	0.33	5.36	15.37	35.94	24.45	12.32	6.56
14091	97.67	0.48	0.87	14.94	35.28	17.05	22.94	8.91
14209	98.53	0.3	3.13	21.92	32.99	19.79	10.88	11.29
14328	98.02	0.4	8.74	13.34	30.26	21.6	9.19	16.87
14447	98.26	0.35	6.31	26.36	19.37	32.85	11.26	3.85
14567	97.16	0.59	2.21	14.32	18.38	35.88	6.74	22.47
14687	97.72	0.47	4.99	11.81	34.47	23.06	9	16.68
14808	97.26	0.56	4.31	12.96	49.59	16.06	14.06	3.03
14929	98.84	0.24	2.09	23.08	29.57	19.88	12.79	12.59
15051	97.62	0.49	12.6	13.1	12.82	47.48	9.34	4.67
15174	97.97	0.42	6.69	12.25	22.92	26.1	10.88	21.16
15297	98.09	0.39	3.89	18.7	30.5	30.79	6.75	9.38
15420	98.23	0.36	0	18.3	16.04	37.57	14.95	13.14
15545	97.19	0.58	1.8	10.29	19.42	27.44	16.35	24.7
15669	98.02	0.4	2.75	18.08	34.92	16.3	13.6	14.35
15795	97.52	0.51	0.09	10.72	26.24	23.86	11.97	27.12
15920	98.3	0.35	0.62	17.87	27.2	27.45	22.46	4.39
16145	97.85	0.44	0.39	16.59	25.99	30.61	12.76	13.65
16260	97.89	0.43	6.45	14.48	21.71	44.41	11.86	1.1
16373	98.14	0.38	5.1	15.47	34.27	15.86	11.87	17.42
16487	97.87	0.44	1.31	18.81	25.13	42.73	8.77	3.26
16602	99.04	0.19	0	30.01	33.87	8.6	14.16	13.36
16717	98.55	0.29	2.61	15.47	8.97	19.96	8.14	44.86

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
16833	97.86	0.44	0	24.26	18.95	30.33	17.59	8.87
16949	98.66	0.27	10.73	21.32	4.34	30.26	9.25	24.1
17067	98.26	0.35	2.49	20.16	28.25	16.79	9.54	22.77
17186	98.6	0.28	11.9	16.96	37.19	13.75	9.51	10.68
17305	98.35	0.34	4.29	21.59	10.44	32.42	17.29	13.96
17425	97.97	0.42	3.64	12.75	39.33	30.35	5.67	8.27
17545	97.88	0.43	2.67	18.2	31.91	24.08	10.01	13.13
17665	98	0.41	0	14.6	24.78	23.62	7.93	29.07
17786	98.47	0.31	3.24	17.81	42.57	14.44	21.69	0.25
17907	98.4	0.33	2.62	14.84	11.23	34.42	23.61	13.26
18029	98.11	0.39	6.8	14.81	28.33	32.73	17.32	0
18151	98.6	0.29	8.48	26.02	29.6	19.46	6.92	9.52
18274	98.27	0.35	0.79	14.77	39	24.3	14.2	6.94
18398	97.82	0.45	6.95	15.52	19.64	27.44	17.07	13.39
18522	98.01	0.41	2.97	14.4	30.44	29.36	9.73	13.1
18647	98.61	0.28	0	20.02	27.69	17.43	25.54	9.32
18772	98.19	0.37	6.1	19.71	30.45	27.55	13.29	2.9
18898	97.57	0.5	4.55	13.63	28.23	24.58	17.06	11.95
19025	98.32	0.34	9.57	16.61	21.7	29.2	22.92	0
19241	97.91	0.43	5.26	12.58	30.46	15.03	28.88	7.78
19354	98.44	0.32	0.34	18.78	34.18	30.08	9.64	6.98
19468	98.34	0.34	1.26	20.15	19.93	26.78	30.75	1.12
19581	98.68	0.27	6.73	20.75	12.3	39.47	7.1	13.66
19696	97.98	0.41	5.21	17.46	18.74	35.54	11.91	11.13
19810	97.99	0.41	7.66	16.43	30.97	29.48	6.82	8.64
19926	98.66	0.27	7.65	17.88	40.7	16.72	8.42	8.63
20041	98.47	0.31	0	16.84	19.39	33.6	19.34	10.84
20161	98.29	0.35	0.96	23.9	30.24	32.9	9.28	2.72
20284	98.35	0.34	11.07	16.17	39.14	12.63	20.99	0
20407	98.33	0.34	8.51	14.95	17.31	25.04	18.39	15.8
20530	98.63	0.28	10.66	14.96	34.36	26.11	12.59	1.33
20655	98.33	0.34	1.15	19.84	33.8	24.04	18.06	3.11
20779	98.03	0.4	6.71	14.4	22.28	26.01	20	10.6
20903	98.22	0.36	13.07	15.59	14.92	19.76	34.63	2.03
21023	98.73	0.26	3.18	18.9	12.72	32.11	16.84	16.26
21145	97.69	0.47	2.23	15.68	17.22	41.23	19.8	3.84
21267	98.01	0.41	4.03	16.22	37.82	24.28	7.41	10.24

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
21389	98.58	0.29	2.94	17.69	22.47	23.19	23.55	10.16
21512	98.33	0.34	6.72	14.63	47.79	12.12	11.41	7.33
21635	98.37	0.33	3.46	14.44	37.36	15.3	11.41	18.03
21759	98.03	0.4	3.91	14.48	32.62	29.35	10.32	9.32
21884	98.11	0.39	1.02	29.03	24.44	24.04	21.47	0
22009	97.95	0.42	3.72	15.21	16.03	21.89	25.79	17.36
22135	98.26	0.36	6.04	19.01	29.02	19.16	11.44	15.33
22642	99.04	0.19	7.26	32.57	34.16	7.84	13.83	4.34
22755	98.08	0.39	0.7	15.52	19.6	26.09	25.66	12.42
22868	98.26	0.35	5.93	11.72	18.79	19.49	19.65	24.42
22982	97.96	0.42	2.01	17.63	8.92	27.48	21.74	22.22
23098	98.31	0.35	6.18	22.47	16.12	15.16	20.13	19.94
23213	97.97	0.41	2.65	15.64	15.13	31.09	20.39	15.1
23329	98.31	0.34	4.28	17.94	24.59	31.42	9.93	11.85
23446	98.72	0.26	4.46	22.19	20.03	26.35	21.4	5.57
23564	98.32	0.34	6.99	21.12	27.09	19.48	13.98	11.34
23682	98.52	0.3	9	19.62	29.91	17.89	16.12	7.46
23800	98.49	0.31	0.43	21.69	22.29	21.29	13.65	20.64
23919	97.81	0.45	0	15.68	36.42	29.96	15.8	2.14
24039	98.43	0.32	6.44	15.42	21.32	26.44	4.64	25.74
24159	98.26	0.35	7.53	15.84	21.32	22.49	15.56	17.26
24280	98.31	0.34	5.35	16.52	35.72	30.86	7.43	4.12
24401	97.96	0.42	7.63	16.13	27.93	25.8	19.21	3.29
24523	97.91	0.43	0.79	14.68	31.44	26.68	23.97	2.44
24646	98.4	0.33	4.3	16.24	25.19	31.02	20.18	3.06
24769	97.91	0.43	4.41	13.98	15.79	36.34	16.48	12.99
24892	97.79	0.45	4.05	19.36	26.1	34.76	10.29	5.43
25016	98	0.41	7.35	14.85	19.97	19.43	23.47	14.93
25141	98.43	0.32	2.09	21.2	24.78	34.26	13.85	3.83
25267	98.24	0.36	8.93	14.48	15.61	42.85	14.72	3.42
25393	98.14	0.38	2.36	19.15	22.16	33.88	11.48	10.96
25519	98.59	0.29	6.92	17.88	11.31	33.19	28.02	2.69
26139	98.24	0.36	4.17	18.1	19	35.75	20.51	2.47
26252	98.24	0.36	0	13.58	25.38	26.85	23.33	10.87
26365	98.55	0.29	3.31	21.06	7.48	38.05	18.61	11.5
26479	98.63	0.28	4.11	22.31	30.8	18.4	13.08	11.28
26593	98.02	0.4	2.06	17.09	38.35	6.04	6.54	29.92

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
26708	97.5	0.51	13.06	12.47	18.82	32.64	6.73	16.28
26824	98.4	0.33	9.43	23.54	20.2	18.81	16.07	11.94
26940	98.35	0.34	4.27	21.26	10.11	41.5	19.97	2.89
27057	97.61	0.49	0	12.23	16.42	33.1	23.86	14.38
27174	98.08	0.39	4.86	15.44	28.61	28.29	16.34	6.46
27292	98.32	0.34	4.64	16.82	31.5	21.47	11.16	14.4
27410	98.11	0.39	5.65	15.16	27.69	30.83	20.34	0.33
27529	97.62	0.49	3.72	17.21	27.61	23.77	7.62	20.08
27649	98.11	0.39	1.58	14.37	18.51	41.49	14.96	9.08
27769	98.48	0.31	4.17	20.46	13.62	33.04	17.42	11.29
27890	98.78	0.25	3.24	23.57	28.65	24.32	10.65	9.57
28011	98.93	0.22	6.09	20.95	12.78	21.51	27.94	10.73
28133	99.06	0.19	0.62	29.25	28.94	19.02	22.16	0
28256	98.39	0.33	3.75	14.65	30.7	10.13	6.66	34.1
28380	98.19	0.37	10	16.16	20.23	22.63	20.71	10.25
28504	98.77	0.25	5.64	23.19	12.15	39.49	19.54	0
28628	98.39	0.33	0	22.25	16.45	34.38	23.5	3.42
28787	97.99	0.41	6.76	12.12	31.31	27.63	14.57	7.61
28900	98.3	0.35	2.14	16.66	11.63	27.15	33.14	9.28
29014	98.01	0.41	5.45	13.64	26.64	27.83	14.19	12.25
29127	98.04	0.4	6.15	20.47	37.21	19.11	14.34	2.72

Appendix F. Raman Offgas Data for Experiment 115

As discussed in Appendix A, the Raman readings should be positive and sum to 100% except in cases where there is significant Ar present. Due to the noise in the Raman signal, any raw readings that are less than zero are fixed to zero and then all the gas readings for H_2, NO_2, N_2, O_2, N_2O, and NO are normalized to 100 vol %. Even with these corrections, the Raman readings have noise in them. To reduce this noise, moving averages of the fixed and normalized readings were performed using equation D-1.

These moving averages do not eliminate all the noise but smooth the values so comparisons and calculations can be performed. Due to the length of the experiment, 8 hours, and the sampling of the Raman about every 11-12 seconds, approximately every 10 points or 2 minutes of the fixed, normalized, and moving average Raman gas concentrations are reported in Table F-1. A complete set of the data is maintained in SRNL ELN. Due to rounding to the nearest hundredth, the numbers in the table may not sum to exactly 100 vol % but all the decimal places were carried in the calculations performed for this report.

Table F-1. Fixed Normalized Moving Average Raman Data for Experiment 115

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
0	97.19	0.54	0.24	11.66	30.42	18.55	15.55	23.57
123	97.2	0.58	2.44	12.33	41.86	22.05	0	21.33
247	97.38	0.54	0.06	27.11	39.45	23.54	0	9.85
371	96.53	0.72	0	13.71	31.2	31.3	0	23.79
496	97.71	0.47	4.17	15.42	42.33	19.13	0	18.95
621	98.1	0.39	9.24	21.04	35.42	18.01	0	16.29
747	97.66	0.48	3.46	26.86	24.03	39.09	0	6.56
1014	96.33	0.76	1.77	15.11	39.56	30.01	0	13.54
1127	96.6	0.71	0.77	14.29	29.44	31.9	0	23.6
1240	96.63	0.7	0	15.94	31.17	27.34	0	25.55
1354	96.83	0.66	1.09	17.77	34.75	30.47	0	15.9
1469	97.57	0.5	2.74	18.89	30.78	24.43	0	23.16
1584	97.32	0.55	4.51	17.72	40.73	32.8	0	4.24
1699	97.16	0.59	9.29	22.48	22.17	22.49	0	23.57
1816	97.22	0.57	2.49	18.83	35.36	28.71	0	14.61
1932	96.48	0.73	5.92	21.28	32.6	26.89	0	13.31
2050	97.8	0.45	4.25	27.59	32.82	3.58	0	31.75
2168	98.2	0.37	4.4	31.46	28.65	18.29	0	17.2
2287	96.98	0.62	3.87	16.72	22.56	29.46	0	27.39
2407	97.85	0.44	3	31.08	18.15	18.14	0	29.63
2527	97.24	0.57	8.66	21.22	27.74	22.9	0	19.48
2648	96.95	0.63	8.89	29.2	18.52	14.62	0	28.78

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
2769	97.49	0.52	5.16	27.35	18.31	26.7	0	22.48
2891	97.46	0.52	2.71	19.79	30.97	17.03	0	29.5
3013	97.25	0.57	6.68	33	26.58	11.02	0	22.73
3136	97	0.62	8.33	17.38	27.39	9.47	0	37.43
3260	96.66	0.69	3.29	25.57	23.82	16.86	0	30.46
3384	96.46	0.73	5.83	16.33	24.46	15.72	0	37.67
3509	97.45	0.52	1.98	22.02	23.73	24.8	0	27.46
3634	97.48	0.52	3.16	20.53	32.9	16.68	0	26.72
3760	97.45	0.52	1.43	31.52	22.53	11.16	0	33.36
3887	96.79	0.66	4.29	17.64	38.95	17.36	0	21.77
4250	97.03	0.61	2.61	28.19	30.97	13.76	0	24.47
4363	97.56	0.5	4.54	24.91	24.72	24.16	0	21.67
4476	97.26	0.56	6.58	22.49	36.33	17.93	0	16.66
4590	97.23	0.57	3.23	19.17	22.38	25.56	0	29.66
4705	97.77	0.46	2.9	20.72	27.83	27.63	0	20.93
4820	96.65	0.7	2.22	17.51	21.15	26.52	0	32.6
4935	96.85	0.65	3.24	23.03	17.78	16.97	0.46	38.52
5052	97.31	0.55	3.3	24.32	22.25	25.8	0	24.33
5170	97.39	0.54	3.77	24.07	20.75	19.54	0.5	31.37
5288	98.07	0.39	8.22	31.94	24.69	19.38	0	15.76
5406	97.5	0.51	0	23.99	35.67	15.08	0	25.26
5524	97.9	0.43	0	21.98	40.53	14.71	0	22.77
5643	98.17	0.37	1.56	30.37	24.67	20.07	0	23.32
5763	97	0.62	4.77	19.45	31.56	16.65	0	27.57
5883	97.09	0.6	1.78	20.35	31.05	22.01	0	24.8
6004	97.81	0.45	4.43	27.53	23.14	31.83	0	13.07
6125	98.2	0.37	9.36	30.35	5.25	32.85	0	22.18
6247	97.51	0.51	0.54	23.72	19.58	8.96	0	47.19
6369	97.34	0.55	9.54	18.41	14.19	34.87	0	22.98
6492	97.01	0.62	0.51	20.8	40.66	18.3	0	19.72
6616	97.53	0.51	6.79	28.99	22.59	16.49	0	25.14
6740	97.21	0.58	0.79	21.76	18.01	13.48	0	45.96
6865	97.72	0.47	6.56	32.97	27.65	18.22	0	14.6
6990	97.86	0.44	3.7	22.82	23.52	16.72	0	33.24
7116	97.28	0.56	4.47	20.94	31.47	20.88	4.13	18.11
7709	97.8	0.45	0.02	27.52	20.04	19.17	0	33.25
7821	97.86	0.44	5.83	19.46	33.89	23.21	0	17.61

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
7935	97.41	0.53	11.79	17.83	10.94	16.61	0	42.83
8049	97.89	0.43	3.33	28.11	14.42	8.96	0.36	44.82
8163	98.03	0.4	7.15	33.62	15.38	30.81	0	13.04
8278	97.21	0.57	8.1	20.43	21.02	15.65	0	34.81
8394	97.49	0.52	0.17	24.45	43.97	17.89	0	13.53
8510	97.18	0.58	8.8	17.09	30.11	20.81	0	23.19
8627	97.93	0.42	16.55	23.11	19.39	25.18	0	15.77
8744	97.66	0.48	7.72	20.83	30.49	12.44	0	28.52
8862	98	0.41	3.89	30.83	14.46	25.79	0	25.04
8981	97.58	0.5	9.66	20.89	14.38	24.43	0	30.64
9100	98.22	0.36	5.35	29.3	28.48	12.66	0	24.21
9219	98.14	0.38	2.36	27.4	28.51	12.5	0	29.24
9340	97.56	0.5	10.21	20.75	22.65	19.43	0	26.95
9460	97.69	0.47	2.04	28.1	36.08	14.83	0	18.94
9582	98.25	0.36	2.56	37.26	39.25	12.34	1.39	7.2
9704	97.54	0.51	4.37	20.43	29.02	21.19	0	24.99
9826	98.26	0.36	0	26.46	13.5	38.68	0	21.35
9949	97.3	0.56	0.88	32.11	21.13	30.21	0	15.68
10072	97.26	0.57	0	31.93	22.17	12.02	4.99	28.89
10197	97.84	0.44	4.65	23.27	15.46	35	0	21.62
10321	98	0.41	4.52	25.26	29.51	36.44	0	4.28
10446	98.08	0.39	2.2	25.97	24.86	16.58	0	30.38
10572	97.16	0.59	0.71	17.98	14.75	41.09	0	25.47
10727	97.99	0.41	8.24	25.11	16.54	22.21	1.7	26.2
10840	98.29	0.35	1.89	23.15	20.93	23.77	0	30.25
10954	97.38	0.54	8.03	20.58	23.79	27.24	0	20.35
11068	97.54	0.5	1.01	22.33	34	19.52	0	23.15
11182	98.11	0.38	0.56	42.93	10.77	37.26	0	8.48
11298	98.13	0.38	1.59	24.06	27.22	31.21	0.79	15.13
11414	98.46	0.31	1.34	28.91	15.49	14.63	0.62	39.01
11531	98.01	0.41	2.45	27.86	20.22	17.51	1.45	30.5
11649	97.69	0.47	10.88	28.33	20.7	15.82	0	24.27
11767	96.79	0.66	9.04	17.52	31.37	19.92	1.09	21.06
11886	97.96	0.42	4.81	31.33	22.62	21.76	0	19.49
12005	97.94	0.42	0	24.16	21.41	46.84	0.16	7.43
12125	98.42	0.32	0	30.38	20.02	43.8	0	5.8
12246	97.96	0.42	4.05	34.17	30.15	24.34	1.04	6.24

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
12368	97.97	0.42	4.78	25.95	29.47	32.7	0	7.1
12490	97.3	0.55	4.35	16.56	28.79	14.7	1.72	33.88
12613	97.8	0.45	4.98	24.65	23.27	31.56	0	15.55
12737	97.65	0.48	8.27	21.26	19.81	9.21	0	41.46
12861	97.58	0.5	8.11	28.72	16.15	25.54	0.79	20.7
12986	97.92	0.43	3.98	26.17	21.08	13.82	0	34.95
13111	98.08	0.39	7.68	27.85	7.63	38.29	0	18.54
13237	97.65	0.48	1.26	23	20.05	31.27	0	24.42
13364	97.44	0.53	11.6	18.81	32.75	16.18	0	20.66
13491	97.86	0.44	0	21.62	22.44	23.25	0	32.69
13619	97.67	0.48	0.1	20.26	28.21	23.03	0.64	27.76
14488	97.59	0.49	2.9	29.22	15.01	20.49	0	32.37
14601	96.97	0.63	8.42	25.96	9.34	28.14	0	28.15
14714	97.89	0.43	6.23	23.04	20.78	33.69	0	16.26
14828	97.24	0.57	7.41	20.62	22.93	30.97	0	18.07
14942	97.93	0.42	4.87	28.25	13.71	18.68	0	34.49
15058	97.06	0.61	2.02	23.42	15.96	35.1	0	23.5
15173	97.34	0.55	4.94	21.89	26.22	17.02	0	29.94
15289	97.13	0.59	4.19	20.59	32.38	30.79	0	12.04
15406	97.53	0.51	8.67	19.79	23.08	17.43	0	31.03
15524	97.28	0.56	1.33	26.14	11.35	33.75	0	27.43
15642	97.86	0.44	5.12	25.05	20.58	24	0	25.26
15760	97.43	0.53	2.12	17.83	19.12	18.59	0	42.35
15879	97.19	0.58	3.88	21.33	28.34	17.16	0	29.29
15999	97.65	0.48	15.6	23.82	17.17	19.64	0.24	23.53
16119	98.82	0.24	1.08	33.88	20.09	27.46	0.12	17.37
16240	97.55	0.5	1.26	18.03	10.54	27.48	0	42.69
16361	98.3	0.35	1.59	42.1	31.99	10.32	0	14
16483	97.96	0.42	2.01	21.87	21.63	33.54	0	20.95
16605	98.31	0.34	3.21	29.26	14.62	36.73	0	16.18
16728	97.77	0.46	2.27	22.11	29.25	31.41	0	14.97
16852	97.83	0.44	3.46	20.74	31.23	36.19	0	8.38
16976	98.12	0.39	5.8	33.86	4.32	25.13	0	30.89
17101	97.57	0.5	8.2	22.63	13.86	25.01	0	30.3
17226	97.54	0.5	7.98	32.26	16.51	9.63	0	33.62
17352	97.36	0.54	0	21.4	12.93	34.23	0.19	31.25
17606	98.36	0.33	6.98	27.29	43.76	17.87	0.27	3.83

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
17718	98.08	0.39	1.57	28.9	16.23	20.81	0	32.49
17832	98.68	0.27	5.49	40.87	36	6.57	0	11.08
17946	97.49	0.52	2.26	33.58	15.01	22.14	0	27.01
18060	98.1	0.39	4.67	27.94	7.34	33.68	0	26.38
18175	97.81	0.45	2.78	21.96	21.2	44.37	0.85	8.84
18291	98	0.41	1.63	32.68	16.72	18.34	0	30.63
18407	97.76	0.46	0.12	21.53	34.69	25.27	0	18.39
18524	98.15	0.38	8.7	29.81	18.27	24.14	0	19.08
18641	98.35	0.34	7.88	30.14	37.46	13.48	3.81	7.21
18759	97.49	0.52	5.2	16.44	16.26	28.17	0	33.93
18878	97.52	0.51	1.14	21.39	13.27	30.45	0	33.75
18997	97.68	0.48	4.64	18.99	34.97	26.14	3.47	11.79
19116	98.58	0.29	10.18	33.35	6.85	17.55	0.91	31.16
19236	97.71	0.47	4.09	21.45	26.21	30.54	0	17.71
19357	97.8	0.45	5.92	25.23	22.96	29.95	0.65	15.29
19479	98.12	0.38	8.19	22.95	10.95	42.69	0	15.23
19600	98.5	0.31	3.84	31.57	7.29	33.93	0	23.37
19723	98	0.41	3.55	23.42	11.49	29.93	0	31.62
19846	97.98	0.41	3.92	25.26	29.92	19.6	0.44	20.86
19970	98.09	0.39	0.62	21.69	19.55	30.08	3.43	24.62
20094	97.78	0.45	5.71	26.07	4.11	46.71	0	17.4
20218	98.22	0.36	1.48	33.13	17.97	26.21	0	21.21
20344	97.91	0.43	6.69	29.9	18.56	29.34	0	15.51
20469	97.51	0.51	3.36	19.8	22.62	30.69	0	23.54
21129	98.15	0.38	0.78	36.55	24.2	12.41	0	26.06
21242	97.52	0.51	6.25	18.71	21.84	20.21	0.65	32.34
21356	97.72	0.47	4.47	31.4	36.42	18.36	0	9.35
21470	97.76	0.46	9.03	22.6	26.55	29.43	0.11	12.29
21584	98.09	0.39	2.9	38.13	11.52	24.25	0	23.2
21699	97.73	0.47	0.53	23.25	34.4	18.49	0	23.33
21815	97.92	0.43	0.21	29.77	9.11	27.04	0	33.87
21931	97.7	0.47	5.41	19.73	25.26	22.7	0	26.9
22048	97.76	0.46	6.16	20.88	16.2	27.11	2.81	26.83
22165	98.11	0.39	0.57	25.32	19.39	32.93	0	21.8
22283	97.95	0.42	3.59	21.79	27.72	6.73	0	40.17
22402	98.57	0.29	2.27	41.07	7.26	1.88	3.06	44.45
22521	97.4	0.54	1.59	24.15	38.12	9.79	1.94	24.41

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
22640	97.58	0.5	2.32	19.74	18.77	31.5	0	27.67
22761	97.87	0.44	2.61	26.72	15.46	25.68	2.81	26.72
22881	97.74	0.46	18.02	18.43	26.82	22.07	0	14.65
23003	98.28	0.35	7.26	28.81	20.68	29.59	2.56	11.11
23124	97.62	0.49	1.73	20.03	31.56	33.62	0	13.07
23247	97.86	0.44	2.19	23.32	29.03	23.66	0	21.8
23370	98.1	0.39	0.5	28.21	19.33	35.82	0	16.13
23494	97.59	0.5	3.92	25.98	27.12	19.5	3.46	20.02
23619	97.94	0.42	3.75	22.78	14.4	40.16	1.27	17.64
23745	98.26	0.36	2.74	25.57	19.04	29.24	0	23.42
23871	98.38	0.33	3.43	20.13	19.87	18.54	1.03	37.02
23997	97.58	0.5	7.23	22.8	31.23	12.32	0.38	26.04
24332	98.31	0.34	5.49	34.88	28.77	13.61	0	17.25
24445	97.68	0.48	1.47	22.62	16.35	34.55	0.22	24.78
24558	98.45	0.32	1.77	28.05	29.14	18.99	9.9	12.14
24672	98.01	0.41	0.64	21.49	26.13	19.47	0	32.27
24787	98.22	0.36	6.18	31.15	29.65	29.05	0	3.97
24902	98.32	0.34	3.14	24.6	20.41	31.28	3.25	17.32
25017	98.18	0.37	5.05	23.31	26.11	29.64	3.09	12.8
25134	97.7	0.47	4.53	19.25	18.49	30.68	0.2	26.86
25250	98.02	0.4	9.33	24.76	22.49	17.12	2.77	23.53
25368	98.33	0.34	9.15	30.07	16.49	35.35	0.23	8.71
25485	97.96	0.42	6.66	19.04	22.55	28.19	0	23.57
25604	98.38	0.33	0	28.69	25.6	35.78	0	9.93
25723	97.64	0.48	0.44	18.76	25.74	16.85	0	38.21
25842	97.82	0.45	5.71	26.17	13.31	24.36	0	30.44
25963	97.04	0.61	1.06	19.85	13.44	27.98	0	37.67
26083	97.85	0.44	5.27	22.83	40.73	21.83	2.81	6.53
26205	97.64	0.48	8.31	26.91	15.47	21.97	2.98	24.36
26326	98.31	0.34	0	30.54	32.53	14.33	3.76	18.84
26449	98.37	0.33	9.77	29.64	11.84	32.7	0.91	15.15
26572	98.34	0.34	5.51	26.94	15.92	25.97	2.01	23.65
26695	98	0.41	12.09	25.11	36.33	18.32	8.15	0
26819	98.59	0.29	0	32.04	16.29	33.99	0	17.68
26944	97.74	0.46	0	22.67	20.49	13.16	0	43.68
27069	97.91	0.43	10.5	27.25	23.9	23.8	0	14.55
27195	98.42	0.32	0	24.33	15.31	31.53	0	28.83

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
27354	97.62	0.49	0.53	23	39.08	8.96	1.43	27
27467	98.34	0.34	9.94	26.85	18.43	24.59	0	20.18
27582	97.38	0.54	12.46	22.58	14.67	22.11	2.5	25.69
27696	97.26	0.56	1.41	16.97	12.79	25.79	0	43.04
27811	98.7	0.26	11.23	29.3	31.92	16.66	0	10.89
27927	98.09	0.39	1.38	31.08	9.25	30.27	0.72	27.31
28044	97.61	0.49	5.63	17.69	16.05	34.68	0.98	24.97
28160	98.24	0.36	5.99	25.43	23.76	24.36	0	20.46
28278	97.7	0.47	8.74	17.33	16.51	20.92	0.58	35.91
28396	97.98	0.41	1.21	20.25	15.95	45.24	0	17.35
28515	98.27	0.35	0.76	24.18	33.66	38.11	0	3.29
28634	98.01	0.41	4.43	20.47	27.1	17.18	0.51	30.31
28754	98.44	0.32	6.25	33.87	14.59	17.24	0	28.05
28874	98.76	0.25	3.2	30.42	25.29	30.34	0	10.74
28995	96.48	0.74	11.04	25.41	10.03	17.51	16.08	19.93
29116	98.38	0.33	0	37.39	17.82	13.76	0	31.03

Appendix G. Raman Offgas Data for Experiment 117

As discussed in Appendix D, the Raman readings should be positive and sum to 100% except in cases where there is significant Ar present. Due to the noise in the Raman signal, any raw readings that are less than zero are fixed to zero and then all the gas readings for H_2, NO_2, N_2, O_2, N_2O, and NO are normalized to 100 vol %. Even with these corrections, the Raman readings have noise in them. To reduce this noise, moving averages of the fixed and normalized readings were performed using equation D-1.

These moving averages do not eliminate all the noise but smooth the values so comparisons and calculations can be performed. Due to the length of the experiment, 8 hours, and the sampling of the Raman about every 11-12 seconds, approximately every 10 points or 2 minutes of the fixed, normalized, and moving average Raman gas concentrations are reported in Table G-1. A complete set of the data is maintained in SRNL ELN. Due to rounding to the nearest hundredth, the numbers in the table may not sum to exactly 100 vol % but all the decimal places were carried in the calculations performed for this report.

Table G-1. Fixed Normalized Moving Average Raman Data for Experiment 117

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
8	98.15	0.47	4	10.2	51.52	20.08	6.44	7.77
122	97.15	0.59	3.02	11.58	59.46	20.1	4.09	1.75
238	97.41	0.53	0	15.67	38.39	28.5	4.2	13.24
353	97.43	0.53	4.03	11.17	48.49	21.7	10.68	3.93
470	97.66	0.48	0.3	18.1	31.85	35.37	1.32	13.06
587	97.27	0.57	0.99	12.11	52.11	17.3	8.26	9.23
704	97.52	0.51	4.88	9.96	48.32	21.07	2.16	13.61
823	97.82	0.45	3.11	17.37	38.48	30.45	5.41	5.17
941	96.2	0.79	1.25	10.39	33.54	26.81	3.45	24.57
1060	97.56	0.5	3.21	14.24	28.6	40.74	3.59	9.63
1180	97.02	0.62	4.97	11.62	47.65	16.35	10.77	8.64
1301	96.94	0.63	8.67	13.94	36.76	28.76	6.51	5.37
1423	98.46	0.31	1.95	27.38	30.73	11.52	27.21	1.22
1544	97.86	0.44	5.11	15.2	49.78	17.3	7.2	5.41
1666	97.37	0.54	2.94	17.09	8.83	35.38	1.55	34.21
1790	97.95	0.42	0	14.51	42.55	31.18	6.59	5.17
1914	97.38	0.54	12.55	15.17	33.29	14.85	12.41	11.73
2038	98.63	0.28	0	28.38	0.12	21.95	23.05	26.51
2163	97.87	0.44	3.98	14.66	29.07	13.93	18.47	19.89
2289	97.73	0.47	3.06	22.88	30.31	19.42	13.42	10.91
2415	97.22	0.57	6.58	10.44	28.52	18.26	12.77	23.43
2684	97.79	0.45	7.97	19.86	29.56	27.23	3.93	11.45

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
2797	98.56	0.29	0.07	18.63	24.87	16.21	19.44	20.77
2910	97.03	0.61	1.69	15.13	12.64	30.14	5.55	34.84
3024	97.75	0.46	1.9	17.29	27.32	11.08	5.51	36.9
3138	97.08	0.6	2.99	15.58	13.07	20.29	21.55	26.52
3253	98.03	0.4	5.74	17.18	29.05	19.89	17.36	10.78
3368	97.63	0.49	2.86	16.29	21.9	19.91	18.96	20.08
3484	97.17	0.58	7.49	14.62	18.09	26.41	13.77	19.62
3600	98.43	0.32	1.94	19.12	26.78	36.92	2.16	13.07
3717	97.9	0.43	4	19.36	25.13	24.86	11.07	15.58
3835	97.39	0.54	7.61	18.16	26.45	22.45	19.47	5.86
3953	97.29	0.56	0	15.81	19.01	21.3	19.86	24.02
4072	98.19	0.37	7.47	22.18	32.9	27.73	7.85	1.87
4191	97.87	0.44	5.78	17.61	28.27	28.03	7.83	12.49
4311	97.79	0.45	2.5	17.15	27.82	22.12	13.14	17.28
4431	97.9	0.43	2.79	19.22	11.74	33.75	11.28	21.22
4552	97.48	0.52	1.7	16.83	28.59	18.11	14.17	20.59
4674	97.91	0.43	0	20.64	10.91	35.21	20.3	12.94
4796	97.78	0.45	0.22	28.76	19.41	29.59	19.5	2.52
4918	97.29	0.56	2.53	14.86	17.94	32.01	8.16	24.5
5042	97.29	0.56	5.16	19.88	22.18	24.37	10.53	17.89
5165	98.12	0.39	7.78	19.41	28.1	17.22	7.71	19.78
5290	98.11	0.39	1.96	22.32	14.82	28.27	24.76	7.86
5415	97.69	0.47	8.79	19.02	17.99	32.72	4.07	17.4
5540	97.52	0.51	1.81	19.22	38.57	23.36	13.81	3.22
6152	97.6	0.49	1.64	22.51	13.1	31.51	7.6	23.63
6264	98.66	0.27	0	20.63	21.38	21.61	12.06	24.32
6377	97.54	0.51	3.82	14.89	21.42	25.61	8.35	25.92
6491	97.99	0.41	1.37	20.9	36.92	10.93	13.98	15.9
6605	97.61	0.49	7.89	25.79	17.02	12.59	14.43	22.28
6720	98.05	0.4	7.47	19.79	22.73	26.16	10.18	13.68
6835	97.87	0.44	3.12	18.76	7.98	32.67	4.33	33.14
6951	98.01	0.41	2.59	21.71	30.7	10.67	5.36	28.97
7067	97.24	0.57	2.87	14.89	29.04	20.89	7.67	24.65
7184	97.43	0.53	4.94	13.7	14.59	26.93	22.89	16.95
7302	98.29	0.35	2.27	22.72	27.23	11.16	7.1	29.52
7420	98.56	0.29	1.31	22.54	24.1	18.09	21.6	12.35
7539	97.5	0.51	6.68	15.86	23.17	22.53	12.97	18.8

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
7658	98.25	0.36	1.08	23.77	31.18	7.69	12.99	23.28
7778	97.5	0.51	0	17.18	32.07	21.37	14.02	15.36
7898	98.3	0.35	2.44	21.32	12.08	14.09	13.95	36.12
8019	97.77	0.46	9.95	15.97	14.24	13.68	14.34	31.83
8141	97.83	0.44	2.57	19.22	20.75	27.62	23.99	5.86
8263	98.1	0.39	3.3	19.71	44.18	18.42	9.76	4.64
8385	98.69	0.27	10.53	23.21	15.8	22.38	17.09	10.98
8509	97.44	0.53	6.17	17.18	7.48	20.09	12.1	36.98
8632	98.54	0.3	4.14	20.2	13.65	37.77	14.32	9.92
8757	98.49	0.31	6.44	18.4	15.67	25.19	22.67	11.63
8882	97.24	0.57	0.64	15.41	16.13	30.26	12.9	24.66
9007	97.27	0.56	5.71	14.83	13.75	31.32	7.71	26.67
9247	98.12	0.38	7.12	19.36	30.28	12.65	19.25	11.34
9359	98.18	0.37	5.69	19.5	31.16	19.38	19.84	4.43
9472	98.08	0.39	2.54	16.84	24.18	14.66	18.67	23.11
9586	97.94	0.42	3.9	19.86	14	15.81	23.52	22.9
9700	97.63	0.49	6.23	17.76	14.02	15.89	24.77	21.33
9814	97.02	0.61	3.66	13.72	28.24	8.87	16.14	29.37
9930	97.76	0.46	4.43	15.61	23.9	29.24	5.51	21.31
10045	98.32	0.34	10.48	19.69	17	25.25	19.47	8.11
10162	97.85	0.44	1.05	20.93	18.62	17.6	26.13	15.68
10279	97.61	0.49	2.19	15.97	15.08	21.36	16.42	28.97
10396	98.46	0.31	7.88	25.39	30.07	15.68	11.2	9.78
10514	97.51	0.51	5.64	17.2	12.46	31.75	8.57	24.39
10633	97.98	0.41	5.94	15.41	14.89	20.82	17.86	25.08
10752	98.11	0.39	1.1	15.36	31.4	12.87	16.84	22.43
10872	97.94	0.42	1.77	14.93	13.41	15.6	27.81	26.47
10993	98.15	0.38	4.64	18.35	9.97	24.09	17.82	25.13
11114	97.62	0.49	3.26	21.37	10.12	18.79	14.03	32.44
11235	98.06	0.4	9.74	17.59	18.4	19.74	20.12	14.41
11381	98.17	0.37	8.05	18.78	21.99	23.15	27.95	0.08
11493	98.17	0.37	8.88	20.55	21.54	15.52	19.9	13.6
11606	97.1	0.6	8.26	14.52	23.01	24.39	16.4	13.43
11720	98.08	0.39	0.58	18.05	23.84	31.86	10.17	15.49
11834	97.09	0.6	4.65	14.71	30.08	20.28	8.91	21.37
11949	96.77	0.67	6.95	14.31	23.15	23.87	9.37	22.35
12064	98.18	0.37	4.15	21.43	29.84	19.5	9.41	15.67

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	**CO_2 and CO**	**Total Offgas Flow**	**H_2**	**NO_2**	**N_2**	**O_2**	**N_2O**	**NO**
(sec)	**(vol %)**	**(cm^3/min)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**	**(vol %)**
12180	98.02	0.41	4.11	15.09	23.38	19.46	17.71	20.25
12297	97.61	0.49	3.93	18.45	24.68	26.24	18.73	7.97
12414	98.38	0.33	3.81	23.33	12.32	27.5	23.79	9.25
12531	98.31	0.35	4.37	27.77	24.26	24.39	14.55	4.67
12649	97.57	0.5	3.49	14.96	11.05	21.47	17.7	31.33
12768	97.83	0.44	1.46	16.61	33.46	29.47	15.49	3.52
12887	98.16	0.38	0.2	19.39	9.22	21.04	24.29	25.86
13007	97.03	0.61	7.29	12.84	27.41	14.78	12.84	24.84
13128	97.62	0.49	1.86	14.75	11.69	19.88	18.36	33.46
13249	97.51	0.51	5.83	17.1	20.52	19.24	16.86	20.44
13370	97.63	0.49	2.62	18.46	29.65	19.46	14.34	15.48
13492	97.48	0.52	3.78	15.23	25.52	20.66	14.71	20.11
13615	98.27	0.35	9.03	20.88	19.43	13.55	19.49	17.61
13738	97.75	0.46	0.35	17.3	15.46	24.74	12.38	29.77
13862	97.86	0.44	5.88	18.12	19.47	16.27	24.78	15.47
13986	98.11	0.39	2.85	22.96	28.53	26.19	14.11	5.36
14111	97.71	0.47	3.67	21.79	21.49	22.51	14.47	16.08
14237	97.32	0.55	5.91	15.15	18.03	15.5	14.11	31.31
14470	97.64	0.48	1.56	16.86	14.55	38.48	20.69	7.86
14583	98.12	0.38	8.19	21.52	33.62	13.72	9.51	13.44
14696	97.8	0.45	3.81	16.19	18.81	25.25	17.51	18.43
14809	97.09	0.6	3.82	13.97	26.09	20.74	17.64	17.74
14923	98.19	0.37	3.31	24.68	13.54	19.69	15.13	23.64
15038	98.24	0.36	8.65	24.24	12	26.15	26.28	2.68
15153	97.87	0.44	11.59	17.65	25.38	22.75	18.31	4.32
15269	97.88	0.44	0	23.24	21.12	19.39	23.68	12.58
15386	98.11	0.38	7.64	14.51	27.92	22.03	19.75	8.16
15503	97.69	0.48	4.27	17.84	18.99	33.02	15.81	10.07
15620	97.29	0.56	7.41	19.62	12.72	16.33	25.42	18.5
15738	97.81	0.45	11.53	19.87	11.18	7.66	39.62	10.14
15857	97.77	0.46	4.04	15.89	20.5	20.23	11.24	28.11
15976	97.05	0.61	5.02	15.62	36.32	13.63	12.6	16.81
16096	97.21	0.57	0.68	13.56	10.81	25.06	19.16	30.72
16217	97.64	0.48	2.24	17.66	23.89	29.09	16.25	10.88
16337	97.99	0.41	6.84	15.56	21.08	26.31	17.16	13.05
16459	98.46	0.31	4.83	21.72	32.21	12.15	14.8	14.29
16581	97.92	0.42	5	18.8	32.69	22.89	18.96	1.66

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
16704	97.76	0.46	2.77	21.5	10.79	28.3	19.91	16.73
16827	98.17	0.37	5.64	18.95	15.19	33.74	20.18	6.29
16950	98.39	0.33	6.96	31.75	8.43	20.49	23.02	9.35
17075	98.07	0.39	3.06	17.13	28.16	14.97	19.16	17.52
17200	97.9	0.43	7.5	19.18	17.84	20.81	20.37	14.29
17343	98.33	0.34	2.01	22.19	26.71	19.89	25.13	4.07
17455	97.64	0.48	1.56	20.91	11.74	24.33	18.77	22.69
17569	97.54	0.5	1.44	15.29	14.27	20.08	23.8	25.12
17683	97.64	0.49	1.59	14.86	10.41	25.28	18.14	29.73
17797	97.71	0.47	0.99	14.27	30.75	7.13	19.22	27.64
17913	98.65	0.28	0.49	27.19	23.88	11.92	14.95	21.58
18028	98.06	0.4	9.9	17.58	26.67	13.07	20.6	12.18
18145	97.64	0.48	2.4	15.29	8.98	31.23	26.15	15.94
18261	97.98	0.41	5.94	19.36	15.08	39.94	13.05	6.64
18379	98.31	0.34	1.81	17.88	5.03	17.18	33.43	24.68
18497	97.87	0.44	4.49	23.41	25.3	22.96	17.8	6.04
18615	97.9	0.43	3.36	18.27	19.92	29.89	20.52	8.03
18734	98.12	0.38	10.82	16.78	23.3	7.66	16.8	24.65
18854	97.88	0.43	4.82	17.84	5.88	32.53	23.04	15.89
18974	98.48	0.31	7.8	24.2	13.51	22.09	17.72	14.68
19095	97.79	0.45	2.84	18.54	28	26.1	15.66	8.86
19216	98.46	0.32	2.44	27.69	10.03	12.18	23.74	23.91
19338	97.86	0.44	5.32	15.87	12.84	26.23	17.91	21.82
19462	97.69	0.47	4.26	23.67	21.29	13.5	17.32	19.97
19586	97.63	0.49	2.95	17.54	8.19	31.2	28.67	11.45
19711	97.86	0.44	12.26	19.51	4.32	29.43	27.3	7.18
19836	96.81	0.66	1.21	11.43	26.32	22.98	16.55	21.51
19961	97.82	0.45	2.63	14.9	22.78	29.15	18.23	12.31
20087	98.82	0.24	10.29	23.55	12.5	17.35	19.92	16.39
20213	97.46	0.52	0	17.94	32.19	20.65	19.04	10.19
20778	97.69	0.47	4.27	16.16	16.55	16.26	21.19	25.56
20891	97.24	0.57	2.5	12.98	20.75	19.04	19.03	25.71
21006	97.08	0.6	3.23	11.99	16.9	30.83	24.87	12.18
21121	98.41	0.33	3.48	19.72	10.62	15.54	25.97	24.66
21237	97.23	0.57	1.67	19.25	18.92	17.95	28.71	13.49
21352	98.22	0.36	1.74	16.97	11.52	19.66	30.98	19.13
21469	97.95	0.42	6.58	16.09	11.21	21.46	20.08	24.58

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
21585	98.29	0.35	6.64	18.45	8.57	24.69	27.96	13.7
21703	98.04	0.4	2.5	20.6	15.99	29.96	28.03	2.92
21821	97.87	0.44	5.27	14.5	20.18	15.46	20.69	23.9
21939	97.97	0.41	5.15	26.07	11.96	26.09	15.52	15.21
22059	98.02	0.4	5.08	19.12	10.33	24.69	25.68	15.09
22178	97.97	0.42	1	17.16	18.9	27.74	24.22	10.98
22298	97.46	0.52	5.39	14.26	18.62	22.01	24.48	15.24
22419	97.86	0.44	1.75	15.3	18.58	29.75	9.41	25.22
22541	98.15	0.38	5.01	21.11	9.78	13.42	38.65	12.04
22663	98.35	0.34	5.54	16.89	13.41	31.71	21.66	10.79
22785	97.97	0.41	0.99	17.43	13.12	17.14	28.74	22.57
22908	98.1	0.39	7.76	20.47	19.49	16.39	31.42	4.47
23032	98.13	0.38	7.3	20.21	12.84	14.72	36.33	8.6
23156	97.62	0.49	3.01	18.63	21.56	14.35	36.7	5.76
23281	98.44	0.32	4.17	21.92	28.07	6.53	30.88	8.44
23406	97.44	0.52	3.11	22.39	21.27	9.41	24.83	18.99
23532	98.44	0.32	3.19	19.41	17.12	21.79	25.99	12.5
23658	97.41	0.53	3.96	25.72	8.5	26.22	27.49	8.12
24669	98.09	0.39	2.8	16.13	23.02	10.84	22.94	24.26
24782	97.84	0.44	1.68	13.65	16.25	7.66	26.62	34.13
24895	97.44	0.52	3.82	13.81	18.39	6.08	28.21	29.7
25009	97.82	0.45	3.36	15.66	13.3	32.31	17.08	18.28
25123	97.85	0.44	1.28	16.27	6.18	32.58	23.04	20.64
25238	97.78	0.45	4.64	16.31	13.04	23.62	10.81	31.58
25354	97.83	0.44	5.35	16.06	10.31	12.33	25.35	30.6
25470	97.61	0.49	7.09	12.24	28.69	16.54	18.68	16.75
25587	98.33	0.34	11.14	19.38	18.56	13.21	22.9	14.82
25704	97.88	0.43	9.73	14.05	17.5	9.59	20.79	28.34
25822	97.83	0.44	1.34	14.96	21.48	17.54	22.45	22.23
25940	97.87	0.44	3.57	14.54	11.44	15.95	21.76	32.74
26059	97.9	0.43	2.28	18.14	20.71	28.12	17.84	12.9
26179	97.68	0.48	1.64	15.21	17.54	24.16	27.22	14.23
26299	98.49	0.31	1.25	18.36	11.06	33.14	21.58	14.61
26420	98.01	0.41	1.39	16.99	6.07	30.44	28.29	16.82
26541	97.75	0.46	2.46	14.89	25.12	20.56	20.65	16.32
26663	98.05	0.4	8.26	16.75	17.61	19.13	20.42	17.83
26785	97.75	0.46	2.34	16.76	29.03	15.39	30.77	5.71

---	Tracer	Offgas Flow and Concentrations without tracers and water						
Cumulative Reaction Time	CO_2 and CO	Total Offgas Flow	H_2	NO_2	N_2	O_2	N_2O	NO
(sec)	(vol %)	(cm^3/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
26908	97.45	0.52	3.83	16.68	18.67	21.78	20.18	18.86
27031	97.99	0.41	3.08	20.6	14.47	16.6	32.27	12.96
27156	97.73	0.47	1.17	20.22	10.2	26.28	36.08	6.04
27280	98.16	0.37	5.8	16.21	6.57	28.02	17.5	25.9
27405	98.21	0.36	2.7	17.13	3.32	15.15	18.75	42.95
27531	97.96	0.42	2.8	18.65	5.81	12.61	34.84	25.3
27756	98.21	0.36	8.65	16.4	14.7	14.78	30.38	15.08
27869	97.42	0.53	2.98	23.63	13.07	25.67	22.62	12.03
27986	97.77	0.46	5.51	13.24	25.44	7.22	33.11	15.48
28105	97.82	0.45	6.63	18.52	17.98	22.23	28.41	6.23
28224	97.21	0.57	1.91	13.83	6.93	24.23	22.07	31.03
28340	97.95	0.42	4.62	16.31	11.3	33.66	17.75	16.37
28457	97.68	0.48	14.36	13.14	21.98	8.54	23.78	18.21
28574	96.99	0.62	6.76	11.97	10.79	14.45	31.61	24.42
28691	97.21	0.57	1.06	20.94	12.61	16.88	23.21	25.29
28809	98.2	0.37	4.86	19.92	15.28	29.56	24.4	5.97
28928	97.84	0.44	2.4	18.91	6.14	28.99	39.67	3.89
29047	97.99	0.41	4.38	15.08	16.42	20.62	32.84	10.66

Appendix H. Estimated Mass Dissolved for Stainless Steel Face of FCA Element

Based on the incremental dissolution rate power law function discussed in Section 3.2.1 and the assumed 2" x 2" x 0.0083" (5.08 cm x 5.08 cm x 0.021 cm) stainless steel face on the FCA element with 26.23 cm^2 surface area, the dissolved mass was estimated for each day and for cumulative days as shown in Table H-1.

Table H-1. Estimated Mass Dissolved for Stainless Steel Face of FCA Element

Time (day)	Incremental Dissolution Rate ($mg/(cm^2*min)$)	Time (min)	Mass Dissolved (mg)	Cumulative Mass Dissolved (mg)
1	0.0063	1440	237.23	237
2	0.0042	1440	157.28	395
3	0.0033	1440	123.66	518
4	0.0028	1440	104.27	622
5	0.0024	1440	91.34	714
6	0.0022	1440	81.98	796
7	0.0020	1440	74.82	871
8	0.0018	1440	69.13	940
9	0.0017	1440	64.46	1004
10	0.0016	1440	60.56	1065
11	0.0015	1440	57.23	1122
12	0.0014	1440	54.35	1176
13	0.0014	1440	51.83	1228
14	0.0013	1440	49.60	1278
15	0.0013	1440	47.62	1325
16	0.0012	1440	45.83	1371
17	0.0012	1440	44.21	1415
18	0.0011	1440	42.74	1458
19	0.0011	1440	41.39	1500
20	0.0011	1440	40.15	1540
21	0.0010	1440	39.00	1579
22	0.0010	1440	37.94	1617
23	0.0010	1440	36.95	1654
24	0.0010	1440	36.03	1690
25	0.0009	1440	35.17	1725
26	0.0009	1440	34.36	1759
27	0.0009	1440	33.60	1793
28	0.0009	1440	32.89	1826
29	0.0009	1440	32.21	1858
30	0.0008	1440	31.57	1889
31	0.0008	1440	30.96	1920
32	0.0008	1440	30.38	1951

Time (day)	Incremental Dissolution Rate (mg/(cm^2*min))	Time (min)	Mass Dissolved (mg)	Cumulative Mass Dissolved (mg)
33	0.0008	1440	29.83	1981
34	0.0008	1440	29.31	2010
35	0.0008	1440	28.81	2039
36	0.0008	1440	28.33	2067
37	0.0007	1440	27.88	2095
38	0.0007	1440	27.44	2122
39	0.0007	1440	27.02	2149
40	0.0007	1440	26.62	2176
41	0.0007	1440	26.23	2202
42	0.0007	1440	25.86	2228
43	0.0007	1440	25.50	2254
44	0.0007	1440	25.15	2279
45	0.0007	1440	24.82	2304
46	0.0006	1440	24.50	2328
47	0.0006	1440	24.19	2352
48	0.0006	1440	23.89	2376
49	0.0006	1440	23.60	2400
50	0.0006	1440	23.32	2423
51	0.0006	1440	23.05	2446
52	0.0006	1440	22.78	2469
53	0.0006	1440	22.53	2491
54	0.0006	1440	22.28	2514
55	0.0006	1440	22.04	2536
56	0.0006	1440	21.80	2558
57	0.0006	1440	21.57	2579
58	0.0006	1440	21.35	2600
59	0.0006	1440	21.14	2622
60	0.0006	1440	20.93	2643
61	0.0005	1440	20.72	2663
62	0.0005	1440	20.53	2684
63	0.0005	1440	20.33	2704
64	0.0005	1440	20.14	2724
65	0.0005	1440	19.96	2744
66	0.0005	1440	19.78	2764
67	0.0005	1440	19.60	2784
68	0.0005	1440	19.43	2803
69	0.0005	1440	19.26	2822
70	0.0005	1440	19.10	2841
71	0.0005	1440	18.94	2860

Time (day)	Incremental Dissolution Rate (mg/(cm^2*min))	Time (min)	Mass Dissolved (mg)	Cumulative Mass Dissolved (mg)
72	0.0005	1440	18.78	2879
73	0.0005	1440	18.63	2898
74	0.0005	1440	18.48	2916
75	0.0005	1440	18.33	2935
76	0.0005	1440	18.19	2953
77	0.0005	1440	18.05	2971
78	0.0005	1440	17.91	2989
79	0.0005	1440	17.78	3006
80	0.0005	1440	17.65	3024
81	0.0005	1440	17.52	3042
82	0.0005	1440	17.39	3059
83	0.0005	1440	17.26	3076
84	0.0005	1440	17.14	3093
85	0.0005	1440	17.02	3110
86	0.0004	1440	16.90	3127
87	0.0004	1440	16.79	3144
88	0.0004	1440	16.68	3161
89	0.0004	1440	16.56	3177
90	0.0004	1440	16.46	3194
91	0.0004	1440	16.35	3210
92	0.0004	1440	16.24	3226
93	0.0004	1440	16.14	3243
94	0.0004	1440	16.04	3259
95	0.0004	1440	15.94	3275
96	0.0004	1440	15.84	3290
97	0.0004	1440	15.74	3306
98	0.0004	1440	15.64	3322
99	0.0004	1440	15.55	3337
100	0.0004	1440	15.46	3353
101	0.0004	1440	15.37	3368
102	0.0004	1440	15.28	3383
103	0.0004	1440	15.19	3399
104	0.0004	1440	15.10	3414
105	0.0004	1440	15.02	3429
106	0.0004	1440	14.93	3444
107	0.0004	1440	14.85	3459
108	0.0004	1440	14.77	3473
109	0.0004	1440	14.69	3488
110	0.0004	1440	14.61	3503

Time (day)	Incremental Dissolution Rate (mg/(cm^2*min))	Time (min)	Mass Dissolved (mg)	Cumulative Mass Dissolved (mg)
111	0.0004	1440	14.53	3517
112	0.0004	1440	14.45	3532
113	0.0004	1440	14.38	3546
114	0.0004	1440	14.30	3560
115	0.0004	1440	14.23	3574
116	0.0004	1440	14.16	3589
117	0.0004	1440	14.08	3603
118	0.0004	1440	14.01	3617
119	0.0004	1440	13.94	3631
120	0.0004	1440	13.87	3645
121	0.0004	1440	13.81	3658
122	0.0004	1440	13.74	3672
123	0.0004	1440	13.67	3686
124	0.0004	1440	13.61	3699
125	0.0004	1440	13.54	3713
126	0.0004	1440	13.48	3726
127	0.0004	1440	13.42	3740
128	0.0004	1440	13.35	3753
129	0.0004	1440	13.29	3766
130	0.0004	1440	13.23	3780
131	0.0003	1440	13.17	3793
132	0.0003	1440	13.11	3806
133	0.0003	1440	13.05	3819
134	0.0003	1440	13.00	3832
135	0.0003	1440	12.94	3845
136	0.0003	1440	12.88	3858
137	0.0003	1440	12.83	3871
138	0.0003	1440	12.77	3883
139	0.0003	1440	12.72	3896
140	0.0003	1440	12.66	3909
141	0.0003	1440	12.61	3921
142	0.0003	1440	12.56	3934
143	0.0003	1440	12.50	3946
144	0.0003	1440	12.45	3959
145	0.0003	1440	12.40	3971
146	0.0003	1440	12.35	3984
147	0.0003	1440	12.30	3996
148	0.0003	1440	12.25	4008
149	0.0003	1440	12.20	4020

Time (day)	Incremental Dissolution Rate (mg/(cm^2*min))	Time (min)	Mass Dissolved (mg)	Cumulative Mass Dissolved (mg)
150	0.0003	1440	12.15	4033
151	0.0003	1440	12.11	4045
152	0.0003	1440	12.06	4057
153	0.0003	1440	12.01	4069
154	0.0003	1440	11.97	4081
155	0.0003	1440	11.92	4093
156	0.0003	1440	11.88	4105
157	0.0003	1440	11.83	4116
158	0.0003	1440	11.79	4128
159	0.0003	1440	11.74	4140
160	0.0003	1440	11.70	4152
161	0.0003	1440	11.66	4163
162	0.0003	1440	11.61	4175
163	0.0003	1440	11.57	4186
164	0.0003	1440	11.53	4198
165	0.0003	1440	11.49	4209
166	0.0003	1440	11.45	4221
167	0.0003	1440	11.41	4232
168	0.0003	1440	11.36	4244
169	0.0003	1440	11.32	4255
170	0.0003	1440	11.29	4266
171	0.0003	1440	11.25	4278
172	0.0003	1440	11.21	4289
173	0.0003	1440	11.17	4300
174	0.0003	1440	11.13	4311
175	0.0003	1440	11.09	4322
176	0.0003	1440	11.06	4333
177	0.0003	1440	11.02	4344
178	0.0003	1440	10.98	4355
179	0.0003	1440	10.95	4366
180	0.0003	1440	10.91	4377
181	0.0003	1440	10.87	4388
182	0.0003	1440	10.84	4399
183	0.0003	1440	10.80	4410
184	0.0003	1440	10.77	4420
185	0.0003	1440	10.73	4431
186	0.0003	1440	10.70	4442
187	0.0003	1440	10.67	4452
188	0.0003	1440	10.63	4463

Time (day)	Incremental Dissolution Rate (mg/(cm^2*min))	Time (min)	Mass Dissolved (mg)	Cumulative Mass Dissolved (mg)
189	0.0003	1440	10.60	4474
190	0.0003	1440	10.57	4484
191	0.0003	1440	10.53	4495
192	0.0003	1440	10.50	4505
193	0.0003	1440	10.47	4516
194	0.0003	1440	10.44	4526
195	0.0003	1440	10.40	4537
196	0.0003	1440	10.37	4547
197	0.0003	1440	10.34	4557
198	0.0003	1440	10.31	4568
199	0.0003	1440	10.28	4578
200	0.0003	1440	10.25	4588

Distribution:

T. B. Brown, 773-A
M. E. Cercy, 773-42A
D. A. Crowley, 773-43A
D. E. Dooley, 773-A
W. G. Dyer, 703-2H
A. P. Fellinger. 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. A. McGuire, 773-42A
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
G. T. Chandler, 773-A
M. K. Hackney, 730-2B
R. R. Livingston, 730-2B
S. J. Robertson II, 704-2H
K. P. Burrows, 704-24
P. M. Palmer, 704-2H
C. M. Hadden, 704-2H
W. H. Clifton Jr, 704-2H
A. C. Carraway, 211-18H
T. L. Tice, 221-H
J. B. Schaade, 703-H
R. T. Burns, 221-H
S. A. Yano, 704-2H
J. R. Lint, 704-2H
A. D. Meredith, 704-2H
K. J. Usher, 704-2H
J. L. Varble, 703-H

Records Administration (EDWS)