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Evaluation of Changes to the MTR Fuel Dissolution Flowsheet to Increase the Number of L-Bundles Charged to a Dissolver

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EXECUTIVE SUMMARY

Material Test Reactor (MTR) fuels are dissolved in the H-Canyon facility using a Hg-catalyzed, HNO_3 dissolution flowsheet. The flowsheet is based on experimental work in which laboratory-scale experiments were used to provide bounding values for both offgas and H_2 generation rates. The generation of H_2 gas, primarily from the dissolution of Al, is controlled by limiting the surface area exposed to the solution. The number of L-Bundles charged and their submergence level defines the exposed surface area which is directly proportional to the H_2 generation rate. For the MTR fuel dissolution flowsheet, the maximum number of L-Bundles which can be charged to a dissolver was specified as a function of the Al concentration in the solution and the submergence level of the bundle. The MTR fuels are currently dissolved in the 6.1D dissolver. To reach the desired terminal Al concentration, a total of 12 L-Bundles of fuel are dissolved per batch. Depending on the submergence level being used in the dissolver, the 12 bundles are dissolved in three charges of four bundles per charge or in a sequence of three, four, and five bundles. However, if six L-Bundles could be dissolved in the initial charge, a dissolver batch could be completed in two dissolutions instead of three. To assess the feasibility of dissolving six bundles in the first charge, the Savannah River National Laboratory evaluated changes to the MTR flowsheet which increase the number of bundles which can be charged to a dissolver.

Since the development of the existing MTR fuel dissolution flowsheet, improved offgas measurement methods have been developed to more accurately calculate the offgas composition and generation rate during laboratory-scale dissolutions. In the first part of this study, improvements in the calibration model for the Raman spectrometer (used for offgas characterization) and a more effective data smoothing routine were applied to existing data. The uncertainty in the offgas composition data used to define the MTR fuel dissolution flowsheet were subsequently re-evaluated which resulted in an approximate 50% reduction in the uncertainty of the H_2 gas concentration measurements. The reduced uncertainty was used in new calculations to define the maximum number of L-Bundles which can be charged to either the 6.1D or 6.4D dissolver as a function of the Al concentration. With the reduced uncertainty in the H_2 concentration measurements, a flowsheet was defined which supported initially charging 6 bundles to a dissolver. This flowsheet is based on the use of 0.002 M Hg, an L-Bundle immersion depth of 54 in., an air sparge/purge flowrate of 40 scfm, an iodine reactor operating temperature of 200 °C, and not exceeding 60% of the H_2 LFL in the dissolver offgas.

In the second phase of this study, calculations were performed to assess the impact of changing various operating parameters to increase the number of L-Bundles charged to an H-Canyon dissolver. Applying the improved offgas measurement techniques including the reduced uncertainty in the H_2 concentration measurements to the existing data, the number of bundles of MTR fuel which can be charged to either the 6.1D or 6.4D dissolver were calculated based on varying the bundle immersion depth (44-60 in.), air sparge/purge flowrate (40-60 scfm), iodine reactor operating temperature (150-200 °C), and the percentage of the H_2 LFL which must not be exceeded (60-76%). Qualitatively, the percentage of the LFL had the largest influence on the maximum number of bundles which can be charged to a dissolver followed by the air sparge/purge flowrate, L-Bundle immersion depth, and the iodine reactor operating temperature. During the evaluation of each variable, the other variables were held at the baseline flowsheet conditions.

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LIST OF ABBREVIATIONS

DOE	Department of Energy
HFIR	High Flux Isotope Reactor
LFL	lower flammability limit
MTHM	metric tons of heavy metal
MTR	Material Test Reactor
MURR	Missouri University Research Reactor
NIST	National Institute of Standards and Technology
PR	Purge Rate
ROD	Record of Decision
SNF	spent nuclear fuel
SRNL	Savannah River National Laboratory
SRS	Savannah River Site

1.0 Introduction

In 2013, the US Department of Energy (DOE) issued an amended Record of Decision (ROD) for the management of spent nuclear fuel (SNF) at the Savannah River Site (SRS). The amended ROD stated that DOE will manage approximately 3.3 metric tons of heavy metal (MTHM) from the projected inventory of 22 MTHM at SRS by conventional processing at the H-Canyon facility. The 3.3 MTHM includes up to 200 High Flux Isotope Reactor (HFIR) cores, approximately 1000 bundles of Al-clad SNF currently stored at SRS, as well as target residue material containing enriched U.¹ The SNF is from domestic and foreign research reactors and is typically referred to as Material Test Reactor (MTR) fuel. Bundles of MTR fuel containing assemblies fabricated from U-Al alloys (or other U compounds) are currently dissolved in either the 6.1D or 6.4D dissolver in H-Canyon.²

The MTR fuel is dissolved using a Hg-catalyzed, HNO_3 flowsheet. The initial HNO_3 concentration is based on the mass of Al and U charged to the dissolver, but is typically 5 to 7.5 M. A 0.002 M Hg catalyst is gradually added to the dissolver after waiting 45 minutes from when the solution reaches the boiling point. The delay in adding the Hg reduces the initial surge of offgas and the H_2 generation rate by passivating the surface of the Al L-Bundle. The generation of H_2 gas, primarily from the dissolution of Al, is controlled by limiting the number of L-Bundles which are charged to a dissolver. The number of L-Bundles and their submergence level defines the surface area exposed to the solution which is directly proportional to the H_2 generation rate. The maximum number of L-Bundles which can be charged to a dissolver was defined as a function of the Al concentration in the solution during the development of the MTR flowsheet (Table 1-1).² The number of bundles is based on: (1) an immersion depth of 54 in., (2) the addition of two times the uncertainty to the H_2 concentration measurement (for conservatism), (3) adding the Hg after waiting 45 minutes once the solution reaches the boiling point, and (4) not exceeding 60% of the lower flammability limit (LFL) of H_2 at the dissolver conditions.

Table 1-1. Maximum Number of L-Bundles Charged to a Dissolver

Dissolved Al [M]	No. L-Bundles
$0.00 < \text{Al}[\text{M}] \leq 0.15$	4
$0.15 < \text{Al}[\text{M}] \leq 0.23$	3
$0.23 < \text{Al}[\text{M}] \leq 0.35$	4
$0.35 < \text{Al}[\text{M}] \leq 0.38$	5
$0.38 < \text{Al}[\text{M}] \leq 0.65$	6
$0.65 < \text{Al}[\text{M}] \leq 0.74$	8
$0.74 < \text{Al}[\text{M}] \leq 2.00$	10

Material Test Reactor fuels are currently dissolved in the 6.1D dissolver. To reach a terminal Al concentration of nominally 1.7 M, a total of 12 bundles of fuel are selected per batch to provide the required mass of Al. Three dissolutions are performed per batch based on the constraints given in Table 1-1. Currently a charging sequence of 3, 4, and 5 bundles is used to dissolve the 12 bundles; however, H-Canyon Engineering has requested the Savannah River National Laboratory (SRNL) to evaluate changes in the MTR flowsheet which would allow dissolving 6 bundles in each of two charges to reduce the number of dissolutions per batch from three to two.³ The SRNL was requested to evaluate changes to all dissolving parameters including the L-Bundle submergence level, Hg addition rate, Hg concentration, HNO_3 concentration, preliminary heating time prior to Hg addition, operating temperature of the iodine reactor, and the dissolver sparge/purge flowrate. Other options such as increasing the concentration of H_2 in the

offgas to 100% of the LFL and keeping the concentration of H₂ in the offgas to 60% of the LFL without measurement uncertainty should also be considered.

1.1 Study Objectives

Options for increasing the number of L-Bundles which can be charged to the 6.1D dissolver were based on the use of existing data from the development of the MTR fuel dissolution flowsheet.² No new experimental work was performed; therefore, the effects of changes in the Hg addition rate, Hg concentration, HNO₃ concentration, and preliminary heating time prior to Hg addition on the number of bundles which can be charged to the dissolver were not evaluated as part of this study. Although no new dissolution experiments were performed, the existing data from the offgas analyses performed during previous dissolution experiments were re-evaluated. The re-evaluation included the use of an improved Raman calibration model to improve the accuracy of the measured gas concentrations, the utilization of a new data smoothing routine for the Raman data, and a new assessment of the uncertainty of the H₂ gas measurements.

Initially, the effect of the re-evaluation of the existing dissolution data (including the uncertainty in the H₂ concentration measurements) on the number of L-Bundles which can be charged to the 6.1D dissolver was assessed using the results from experiments performed during the development of the original MTR fuel dissolution flowsheet.² In the second phase of this study, the effects of the L-Bundle submergence level, dissolver sparge/purge flowrate, and operating temperature of the iodine reactor on the number of bundles which can be charged was evaluated. The effect of increasing the percentage of the LFL which the H₂ concentration must not exceed was included as part of this evaluation. For each variable, the number of L-Bundles which can be charged to the 6.1D dissolver was calculated over the range of interest. The impact of changing more than one variable at a time was not evaluated at this time.

2.0 Previous Experimental Work

2.1 Technical Basis for MTR Fuel Dissolution Flowsheet

Six scaled dissolution experiments serve as the basis for the MTR fuel dissolution flowsheet (Table 2-1). A description of the methods used to perform the experiments, analysis of the data, and a discussion of the results is provided by Daniel et al.² Five of the experiments were performed using an Al-1100 alloy which provides a bounding estimate for offgas generation including the generation rate of H₂.⁴ One experiment was performed using a 30 wt % U-Al alloy to demonstrate that a terminal Al concentration of 2 M could be achieved in the presence of a significant concentration of U. Although experiments were performed using Hg concentrations greater than 0.002 M, the number of L-Bundles which could be charged to a dissolver was not sufficient (especially at low concentrations of Al) to pursue defining a flowsheet at either 0.003 or 0.004 M Hg.

The technical basis for the MTR fuel dissolution flowsheet is based on the Al-1100 alloy dissolution experiments performed at 0.002 M Hg. In Experiment 97, the Al alloy was held at the boiling point of the solution for 45 minutes prior to the addition of the Hg catalyst. A reduction in the offgas and H₂ generation rates was previously observed following 45 minutes of boiling due to the passivation of the metal surface.⁴ The reduced gas generation rates allowed the dissolution of an additional L-Bundle during the first charge to the dissolver. In subsequent dissolver charges, data from Experiment 106 were used to define the allowable number of L-Bundles which can be charged to a dissolver. In this experiment, the Hg catalyst was metered into the laboratory-scale dissolver containing the Al alloy as soon as the solution reached the boiling point. The combined use of the two data sets eliminates the effects of the passivation of the Al surface following the first charge when the dissolver already contains the Hg catalyst.

Table 2-1. Experimental Basis for the MTR Fuel Dissolution Flowsheet

Exp. No. ⁽¹⁾	Objective (Evaluate ...)	Al Alloy	Hg Conc. (M)	Target Al Conc. (M)
97	Offgas generation rate at 0.002 M Hg with hold on Hg addition	Al-1100	0.002	2.0
98	Offgas generation rate at 0.004 M Hg with hold on Hg addition	Al-1100	0.004	2.0
105	Offgas generation rate at 0.004 M Hg without pausing Hg addition	Al-1100	0.004	2.0
106	Offgas generation rate at 0.002 M Hg without pausing Hg addition	Al-1100	0.002	2.0
107	Offgas generation rate at 0.003 M Hg without pausing Hg addition	Al-1100	0.003	2.0
108	Offgas generation rate at 0.002 M Hg without pausing Hg addition	30 wt. % U-Al	0.002	2.0

(1) Experimental numbering sequence corresponds to data recording practices

2.2 Offgas Characterization for Al-1100 Alloy Dissolutions

During the dissolution of the Al-1100 alloys, which serve as the basis for the MTR fuel dissolution flowsheet, the offgas leaving the dissolving system was characterized by Raman spectroscopy.² The Raman spectrometer was used to measure non-condensable gases such as H₂, N₂, O₂, NO, N₂O, NO₂, CO, and CO₂ in real time during the experiments. The Raman spectrometer was calibrated using the standard gases shown in Table 2-2. To calculate offgas generation rates, a CO₂ tracer gas was metered into the system through a flow controller at a set rate (20, 30, or 50 cm³/min @ 70 °F, 1 atm). The total offgas rate was then calculated by dividing the set input rate by the measured CO₂ concentration in the offgas.

Table 2-2. Calibration Gases for Raman Spectrometer

Supplier	Gas	Ar (%)	N ₂ (%)	N ₂ O (%)	NO ₂ (%)	NO (%)	O ₂ (%)	H ₂ (%)
Air Liquide	20% N ₂ O-80% Ar	80.00	—	20.00	—	—	—	—
Liquid Technology	5% NO ₂ -20% O ₂ -75% Ar	74.89	—	—	4.98	—	20.13	—
Air Liquide	20% NO-80% Ar	80.00	—	—	—	20.00	—	—
Air Liquide	5% N ₂ -10% H ₂ -85% Ar	85.00	5.00	—	—	—	—	10.00
SRNL	N ₂ ⁽¹⁾	—	99.9	—	—	—	—	—
SRNL	Air ⁽¹⁾	0.94	78.03	—	—	—	20.99	—

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

2.2.1 Calibration of the Raman Spectrometer

The Raman spectrometer used during the development of the MTR fuel dissolution flowsheet was calibrated using the set of gases shown in Table 2-2.² 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. As an additional check before and after each experiment, air, 99.9 vol % CO₂, and/or a 2.67 vol % H₂ gas (balance Ar) were analyzed to ensure the instrument was within calibration. If the calibration checks were not acceptable for these gases, the Raman calibration model was adjusted for the gases after the experiment. For the Al dissolutions performed in Experiments 97 and 106, the pre-run calibration checks are shown in Table A-1

and Table A-2 in Appendix A, respectively. The post run checks for Experiment 97 and 106 are shown in Table A-3 and Table A-4 in Appendix A, respectively. The calibration check indicates if the calibration is still valid and provides an indication of the variance of the measurements since the calibration gas is analyzed for several samples. Since the goal of this study was to increase the number of L-Bundles which can be charged to the 6.1D dissolver,³ the uncertainties in the Raman measurements were re-examined to see if they could be reduced.

The concentrations measured by the Raman spectrometer should be positive and sum to 100 vol % except for the 2.67 vol % H₂ gas which is 97.33 vol % Ar (which is not detected by the Raman technique). Since the current MTR fuel dissolution flowsheet was developed,² the Raman calibration model has been improved to give more accurate readings for the various gases. In addition, due to the noise in the Raman signal, the raw readings (i.e., measured intensities) were re-baselined to zero by subtracting out average values representing zero and any raw readings less than zero were fixed to zero. The concentrations measured for H₂, NO₂, N₂, O₂, N₂O, NO, CO₂, CO, H₂O and NH₃ were then normalized to 100 vol % except for the 2.67 vol % H₂ in Ar standard. These re-baselined and normalized Raman readings are the values reported in Table A-1 through Table A-4 except for the 2.67 vol % H₂ gas, where the raw readings are provided. Due to rounding to the nearest hundredth, the numbers in the tables may not sum exactly to 100 vol % but all the decimal places were carried in the calculations performed for this study.

2.2.2 Offgas Generation Rate

In experiments performed to develop the MTR fuel dissolution flowsheet,² the total offgas flow was calculated by dividing the CO₂ tracer flowrate coming into the dissolving vessel (Table 2-3) by the fixed normalized sum of the CO₂ and CO concentrations (measured by the Raman spectrometer). The noise in the concentrations measured by the Raman technique propagates into the total offgas flowrate so moving averages of the total offgas flowrates were performed using equation 1:

$$\text{Offgas flowrate } t_i (\text{cm}^3/\text{min}) = \frac{\sum_{k=t_i-1}^{t_i+1} \text{Offgas flowrate}_k}{3} \quad (1)$$

where Offgas flowrate = offgas generated by the dissolution in cm³/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 flowrates and fixed normalized moving average Raman offgas concentrations from Experiment 97 and 106 are shown in Appendix B and C, respectively.

Table 2-3. CO₂ Tracer Gas Flowrates

Experiment	CO ₂ Flow (cm ³ /min)
97	20
106	20

2.3 Quality Assurance

A Functional Classification of Safety Significant was applied to this work. To match the requested functional classification, this report received technical review by design verification. 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 Re-evaluation of Data

The measured concentrations of the non-condensable gases (e.g., H₂, N₂, O₂, NO, N₂O, NO₂, CO, and CO₂) in the offgas from the Al-1100 alloy dissolutions previously performed² in Experiments 97 and 106 were re-evaluated in light of the improvements in the Raman spectrometer calibration model and the data smoothing routine. The re-evaluation of the existing data also included a new assessment of the uncertainties in the measured concentrations of H₂ and other gases.

3.1.1 Data Smoothing

As previously discussed, the gas concentrations measured by the Raman spectrometer should be positive and sum to 100 vol % except in cases where there is significant Ar present. Due to the noise in the Raman signal, the raw readings from Experiments 97 and 106 were re-baselined to zero by subtracting out average values representing zero and any raw readings less than zero were fixed to zero. The concentrations for H₂, NO₂, N₂, O₂, N₂O, NO, CO₂, CO, H₂O and NH₃ were then normalized to 100 vol %. Even with these corrections, significant noise is associated with the concentrations measured by the Raman spectrometer. To reduce the noise, moving averages of the fixed and normalized readings were performed using equation 2:

$$\text{Gas}_{t_i}(\text{vol}\%) = \frac{\sum_{k=t_i-3}^{t_i+3} \text{Gas}_k}{7} \quad (2)$$

where Gas = NO₂, N₂, O₂, N₂O, NO, CO₂, CO, H₂O and NH₃ concentrations (vol %)

t_i = time at integer time step i

k = integer time step t_{i-3}, t_{i-2}, t_{i-1}, t_i, t_{i+1}, t_{i+2}, and t_{i+3}.

The moving averages do not eliminate all the noise but smooth the concentrations so comparisons and calculations can be more easily performed. The fixed, normalized, and moving average gas concentrations are reported in Appendix B and Appendix C for Experiments 97 and 106, respectively. Due to rounding to the nearest hundredth, the numbers in the appendices may not sum to exactly 100 vol % but extended decimal places were carried in the calculations subsequently performed to define the number of L-Bundles which can be charged to an H-Canyon dissolver.

3.1.2 Uncertainty in Offgas Concentration Measurements

The measured concentrations of the gases used to calibrate the Raman spectrometer (Table A-1 through Table A-4) should be constant since the spectrometer is analyzing constant sources (calibration gases) without being connected to the dissolution equipment. To estimate the measurement uncertainties of the gas analyses performed by the Raman spectrometer, the pre-run and post-run check values in Table A-1 through Table A-4 were compared to the standard values for only Experiments 97 and 106. In the current MTR fuel dissolution flowsheet,² a weighted average of the offgas measurement uncertainties from all Al-1100 alloy dissolutions performed (Table 2-1) was used. Since each experiment has its own uncertainty defined by the pre-run and post-run checks, a less conservative approach was selected to evaluate the measurement uncertainties for each experiment rather than use the weighted average.

Sample standard deviations of the measured concentrations for H₂, CO₂, O₂, and N₂ with respect to the calibrated concentrations for the pre-run and post-run data were calculated for both Experiments 97 and 106. The sample standard deviations were then doubled (i.e., 2σ) to provide an approximate 95% confidence limit for the uncertainty in the Raman spectroscopy concentration measurements. Table 3-1 shows the sample standard deviations of the measured concentrations with respect to their calibrated values for Experiments 97 and 106. For the CO₂, H₂ and O₂ gases, the 2σ values or twice the standard deviation was

< 2 vol %. The 2σ value for N_2 was < 3 vol %. For the key offgas measurement performed for H_2 , a 2σ of 0.60 vol % bounds both Experiments 97 and 106. For the subsequent calculations performed in this report to determine the number of L-Bundles which can be charged to an H-Canyon dissolver, a 2σ for the H_2 measurement of 0.60 vol % was used. In the current MTR flowsheet,² the 2σ uncertainty used for the H_2 measurement was 1.14 vol %; therefore, implementing the new approaches for analyzing the data results in a reduction of the uncertainty in the measurement by approximately 50%. The reduction in uncertainty will allow more L-Bundles to be charged to a dissolver and enhance the benefits from changing the operating parameters for the dissolvers (e.g., L-Bundle submergence level, dissolver sparge/purge flowrate, operating temperature of the iodine reactor, and percentage of the LFL which must not be exceeded).

Table 3-1. Uncertainty of Measured Concentrations with Respect to Calibrated Values

Exp.	Gas	Pre-Run		Post-Run	
		Standard Deviation (σ) (vol %)	2*Standard Deviation (2σ) (vol %)	Standard Deviation (σ) (vol %)	2*Standard Deviation (2σ) (vol %)
97	CO ₂	0.55	1.10	0.39	0.78
97	N ₂	1.39	2.78	NA	NA
97	O ₂	0.74	1.48	NA	NA
97	H ₂	0.13	0.26	0.15	0.30
106	CO ₂	0.36	0.72	0.36	0.72
106	N ₂	0.97	1.94	1.34	2.68
106	O ₂	0.75	1.50	0.93	1.86
106	H ₂	0.23	0.46	0.29	0.58

3.2 Effect of Hg Addition on Offgas Generation

During the development of the flowsheet for the dissolution of HFIR fuel, an observation was made that holding an Al-1100 alloy coupon in boiling HNO₃ solution for 45 min prior to starting the Hg addition significantly reduced the initial surge of offgas and the H_2 generation rate.⁴ Although it was not necessary to use a hold time as part of the HFIR flowsheet, a 45 minute hold time was incorporated into the MTR fuel dissolution flowsheet allowing an additional L-Bundle to be added to the dissolver during the first charge.² In Experiment 97, Al-1100 was dissolved in 7 M HNO₃ and the addition of Hg (to a final concentration of 0.002 M) was not started until 45 min after the solution reached the boiling point. In Experiment 106, Al-1100 was dissolved in 7 M HNO₃ and the addition of Hg (to the same concentration) was started immediately after reaching the solution boiling point. The H_2 generation rates for the two experiments (using the new Raman calibration model and data smoothing routine) are shown in Figure 3-1. The data demonstrate that holding the solution at the boiling point for 45 min and then starting the Hg addition reduces the H_2 generation rate especially during the peak H_2 generation at the beginning of the dissolution.

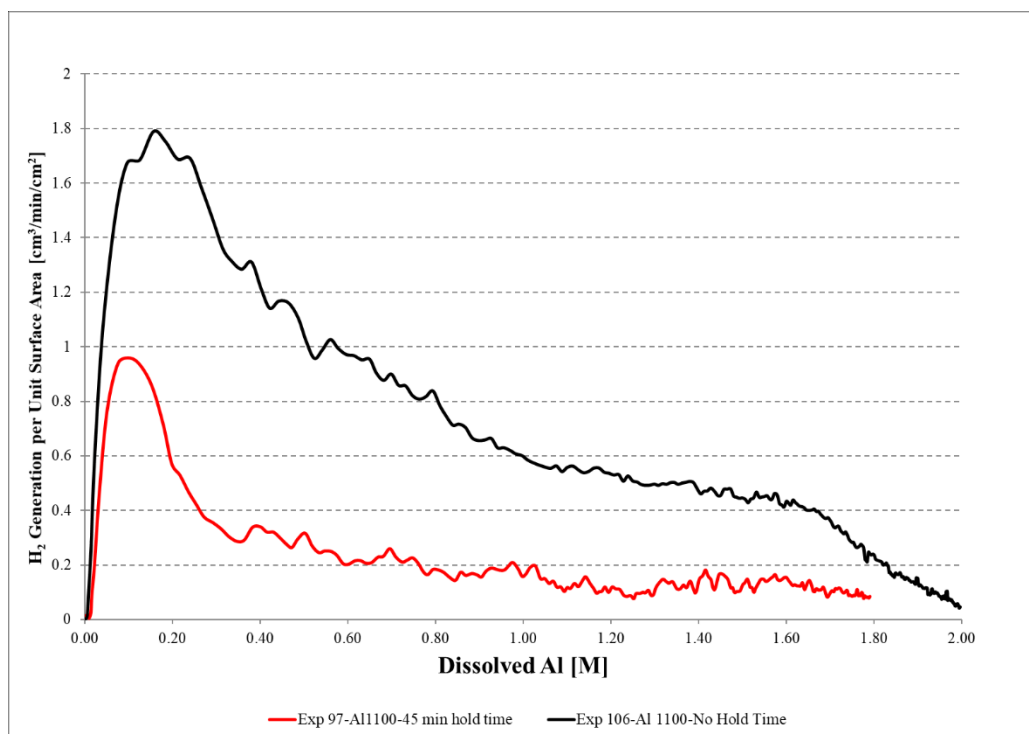


Figure 3-1. Effect of Hold Time on H₂ Generation Rate for Experiments 97 and 106

3.3 Re-evaluation of Flammable Gas Generation at Existing Flowsheet Conditions

The rate of flammable gas generation from MTR fuel is proportional to the surface area exposed to the dissolving solution. Although, most research reactor fuel assemblies are fabricated from multiple plates, the spacing between plates is sufficiently close that fresh dissolving solution cannot continuously fill the small void between adjacent surfaces and dissolve Al. The offgas generation from the dissolution of Al also hinders the flow of fresh solution to the close surfaces. During the development of a dissolution flowsheet for SRS U-Al alloy tubular fuel, Caracciolo⁵ demonstrated that the dissolution rate for two concentric fuel tubes containing a 16 wt % U-Al alloy was essentially the same as for a single tube of the same alloy. This result indicated that the outside surface area of the outer tube controlled the dissolution rate. The MTR fuel assemblies are packaged in L-Bundles which dissolve prior to the fuel. Since the outer surface area of an L-Bundle is greater than the outer surface area of an MTR fuel assembly (similar to Missouri University Research Reactor (MURR) fuel assemblies), the surface area of the L-Bundle provides a bounding value for the H₂ generation rate.⁶ For previous dissolution flowsheets, the peak offgas generation rate during the dissolution of fuel assemblies was based on the outer surface area of the L-Bundle.^{2,4,6-10}

The H₂ generation rates calculated from the Experiments 97 and 106 data were re-evaluated based on the improvements in the Raman spectrometer calibration model and the new data smoothing routine. The recalculated generation rates are plotted as a function of the dissolved Al concentration in Figure 3-1. The H₂ generation rates were calculated from the measured offgas generation rates, measured H₂ concentrations, and the measured surface area of the Al-1100 coupons which were dissolved. The concentration of Al in the dissolving solution as a function of time was estimated using the method described by Almond et al.⁸ The figure shows that the H₂ generation rates surge after the start of the Hg addition. The figure also shows that holding for 45 minutes at boiling before starting the Hg addition reduces the initial surge in the H₂ generation rate.

The criterion of not exceeding 60% of the H₂ LFL in the dissolver offgas (at 200 °C) was used for the flammable gas calculations in this report which is consistent with the criterion used for other fuel dissolution flowsheets.^{2,4,6-10} A maximum concentration of 60% of the LFL is permitted by NFPA 69 when automatic instrumentation with safety interlocks is provided.¹¹ If a low air sparge/purge flowrate is measured in an H-Canyon dissolver, an interlock closes the steam supply to the dissolver coils.¹² The H₂ LFL is corrected to a temperature of 200 °C due to the iodine reactor in the dissolver offgas stream which operates at this temperature. Lower flammability limit data reported by Scott et al.¹³ for air, H₂, NO, and N₂O mixtures were used to calculate the LFL for comparison to the H₂ concentrations calculated for an H-Canyon dissolver. The H₂ LFL data were correlated using a second order polynomial to allow interpolation at varying concentrations of air (Table 3-2).²

Table 3-2. Correlations for H₂ LFL Data

NO:N ₂ O Ratio (vol %/vol %)	Correlation
$\frac{NO}{N_2O} \geq 2.57$	$H_2 \text{ LFL (vol\%)} = 6.038 + 3.174E-02 \cdot (\text{Air vol\%}) - 5.401E-04 \cdot (\text{Air vol\%})^2$
$1 \leq \frac{NO}{N_2O} < 2.57$	$H_2 \text{ LFL (vol\%)} = 3.425 + 1.207E-01 \cdot (\text{Air vol\%}) - 1.186E-03 \cdot (\text{Air vol\%})^2$
$\frac{NO}{N_2O} < 1$	$H_2 \text{ LFL (vol\%)} = 5.479 - 1.067E-03 \cdot (\text{Air vol\%}) - 1.400E-04 \cdot (\text{Air vol\%})^2$

The NO:N₂O volume ratios measured during laboratory dissolution experiments are used to estimate the H₂ LFL in the offgas from an H-Canyon dissolver. The NO and N₂O concentrations measured during Experiments 97 and 106 were re-evaluated based on the improvements in the Raman spectrometer calibration model and the new data smoothing routine. The recalculated NO:N₂O ratios are shown in Figure 3-2. The LFL data for H₂ show that normally higher NO:N₂O ratios result in higher values for the H₂ LFL; likewise, lower NO:N₂O ratios normally will result in lower values of the H₂ LFL.

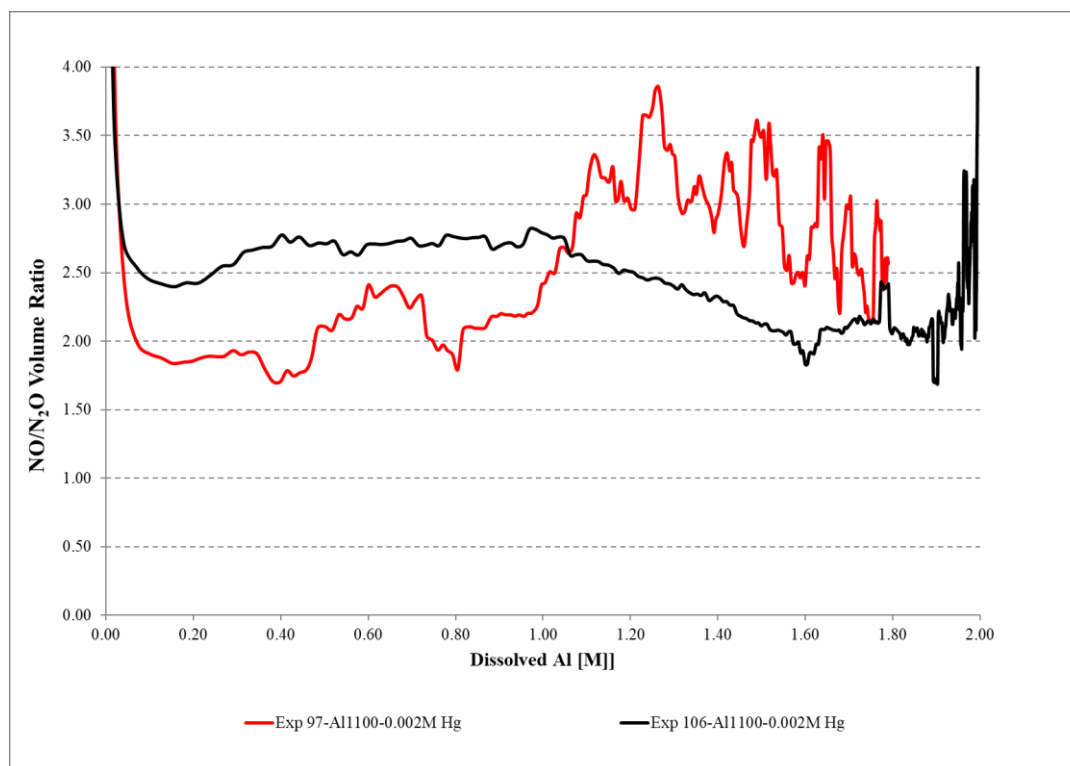


Figure 3-2. NO:N₂O Volume Ratio from the Dissolution of Al-1100 Alloy

The maximum concentration of H₂ calculated during the dissolution of MTR fuel must be compared to the appropriate percentage of the LFL for H₂ at the maximum temperature of the offgas. Since the offgas from the H-Canyon dissolvers flows through an iodine reactor, which operates at 200 °C, the LFL for H₂ must be corrected for the increase in temperature. The LFL is corrected using equation 3:¹⁴

$$LFL_T = LFL_{ref}(1 - A \cdot (T - T_{ref})) \quad (3)$$

where LFL_T is the LFL at temperature T (°C), LFL_{ref} is the LFL at the reference temperature (T_{ref}), A is an empirical coefficient (Zabetakis attenuation factor) equal to 0.0011, and T (°C) is the temperature at which the LFL is evaluated. Since the H₂ LFL decreases with increasing temperature, a reduction in the reactor operating temperature will increase the L-Bundle surface area which can be charged to a dissolver. For this study, the SRNL was asked to evaluate the impact of operating the iodine reactor between 150 and 200 °C on the number of L-Bundles which can be charged to a dissolver. The H₂ LFL data reported by Scott et al.¹³ were measured at 28 °C; therefore, equation 3 can be rewritten as equation 4 for use in this evaluation.

$$LFL_T = LFL_{28^\circ C}(1 - 0.0011 \cdot (T - 28)) \quad (4)$$

The H₂ generation rates calculated for Experiments 97 and 106 (Figure 3-1) were used to predict the H₂ concentration in the offgas stream from an H-Canyon dissolver. The experiments were performed using 0.002 M Hg and targeted a final Al concentration of 1.8 to 2 M. The offgas generation rate for an L-Bundle was based on the outer surface area of the bundle since the surface area is bounding for MURR-like fuel.⁷ For the normal 54 in. immersion of the L-Bundle in an H-Canyon dissolver, the exposed outer surface area per L-Bundle is 6.4546 ft².⁶ For L-Bundle immersion depths of 44 to 60 in. (at 2 in. increments), the associated outer surface area per bundle is shown in Table 3-3.

Table 3-3. L-Bundle Surface Areas for Immersion Depths

Immersion Depth (in)	L-Bundle Surface Area (ft²)
44	5.3638
46	5.5819
48	5.8001
50	6.0183
52	6.2364
54	6.4546
56	6.6728
58	6.8909
60	7.1091

The uncertainties associated with the calculations performed to determine the number of L-Bundles that can be charged to an H-Canyon dissolver were not evaluated; however, the effect of the new uncertainties in the H₂ concentration measurements from Experiments 97 and 106 on the number of L-Bundles which can be charged to a dissolver was examined. To estimate the H₂ concentration in the dissolver offgas stream, the total offgas generation rate was initially predicted for a single L-Bundle (equation 5) based on its surface area (SA) using the values given in Table 3-3 for the desired immersion depth.

$$\text{Predicted Offgas Rate (scfm)} = \text{Measured Offgas Rate} \left(\frac{\text{scfm}}{\text{ft}^2} \right) \cdot \text{SA}(\text{ft}^2) \quad (5)$$

The predicted H₂ generation rate for the L-Bundle was calculated by multiplying the predicted offgas rate by the measured H₂ concentration plus the 2σ_{H2} uncertainty (Table 3-1) in the H₂ measurement (equation 6).

$$\text{Predicted H}_2 \text{ Offgas Rate (scfm)} = (\text{Measured H}_2 \text{ (vol\%)} + 2\sigma_{H_2}) \cdot \text{Predicted Offgas Rate (scfm)} \quad (6)$$

The H₂ measurement uncertainty (2σ_{H2}) is assumed constant over the range of measurements based on prior testing and comparison with mass spectrometer data. The predicted H₂ concentration in the dissolver offgas stream was subsequently calculated from the predicted H₂ generation rate, the predicted (total) offgas rate, and the volumetric flowrate of air used to sparge (i.e., mix) the solution and purge the dissolver (PR) (equation 7). A dissolver sparge/purge flowrate of 40 scfm is used during SNF dissolution in the H-Canyon dissolvers; however, the SRNL was asked to evaluate the impact of purge rates between 40 and 60 scfm on the number of L-Bundles which can be charged to a dissolver.

$$\text{Predicted H}_2 \text{ Conc (vol \%)} = \frac{\text{Predicted H}_2 \text{ Generation Rate (scfm)}}{\text{Predicted Offgas Rate (scfm)} + \text{PR (scfm)}} \cdot \left(\frac{100 \text{ vol\%}}{1} \right) \quad (7)$$

To demonstrate how the recalculated uncertainties in the H₂ concentration measurements affect the number of L-Bundles which can be charged to a dissolver, the measured H₂ concentrations in Experiments 97 and 106 were increased by 0.60 vol % (2σ uncertainty). The predicted H₂ concentration (with air dilution) in the dissolver offgas stream is compared with 60% of the calculated H₂ LFL at 200 °C in Figure 3-3 and Figure 3-4, respectively. The figures show the number of L-Bundles (at a 54 in. immersion depth) which can be charged to a dissolver without exceeding the calculated LFL over an Al concentration range from 0 to 2 M. The sharp decrease and increase in 60% of the temperature-corrected LFL of H₂ at nominally 0.3 and 1.1 M Al, respectively, in Figure 3-4 is an artifact of a change in the correlation used to calculate the LFL (Table 3-2). The move from one data set to another results in an irregular shift in the calculated LFL. The number of L-Bundles that can be charged depends on the concentration of Hg used and whether the Hg is added when the boiling point of the solution is reached or delayed for 45 minutes after boiling is achieved. Figure 3-4 shows that when the Hg addition is started at boiling for a final concentration of 0.002 M, 6 L-Bundles can be added to the dissolver in the first charge since the predicted H₂ concentration is below 60% of the LFL. The margin between the predicted H₂ concentration and the calculated LFL is greater for 0.002 M Hg when the dissolving solution was allowed to boil for 45 minutes prior to starting the Hg addition (Figure 3-3). The number of L-Bundles which can be charged to a dissolver on Figure 3-3 does not increase with the Al concentration since these data are only valid for the first dissolver charge when the Hg addition is delayed.

The results from the re-evaluation of the data from the Al-1100 dissolution experiments performed to develop the MTR fuel dissolution flowsheet are shown in Table 3-4. The table provides the maximum number of L-Bundles which can be charged to a dissolver as a function of the Al concentration at a 54 in. immersion level. The initial charge of L-Bundles is based on the addition of 0.002 M Hg when the boiling point of the solution is reached (Experiment 106). The allowable number of L-Bundles for higher concentrations of Al is also based on Experiment 106 in which 0.002 M Hg was added as soon as the solution began to boil.

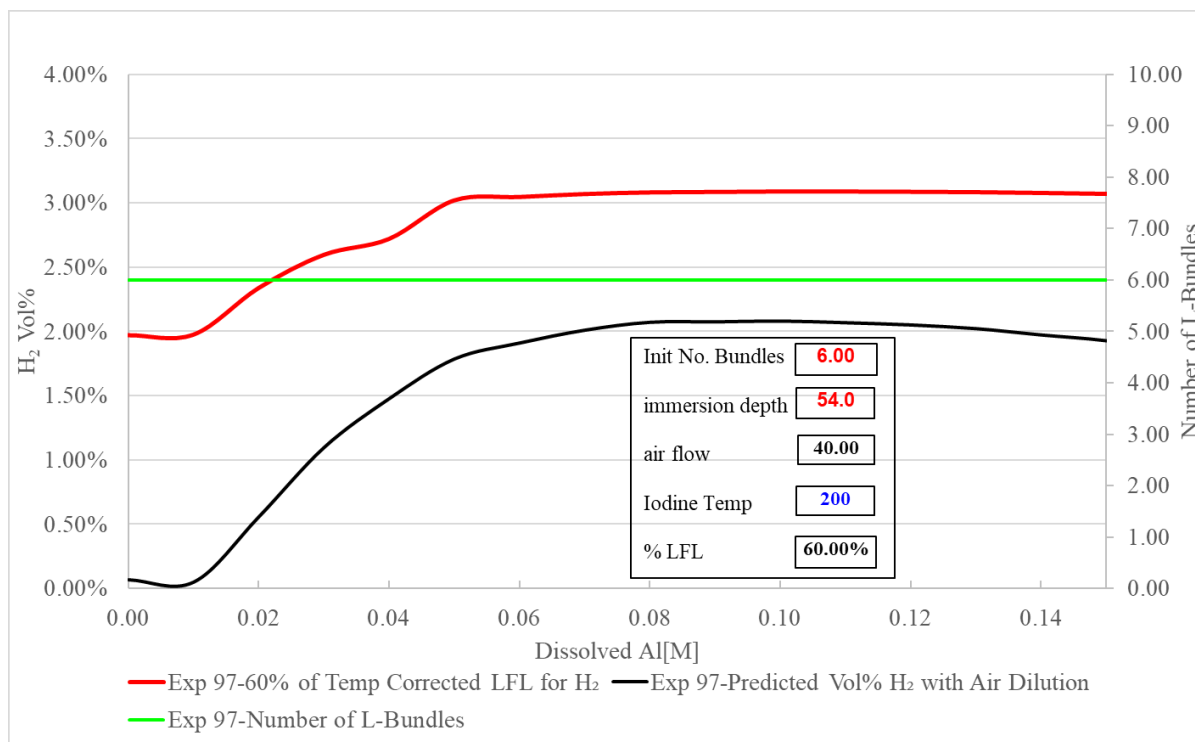


Figure 3-3. H₂ LFL Comparison for Exp. 97 – 7 M HNO₃, 0.002 M Hg, 45 min Hold Time, and 54 in. Immersion

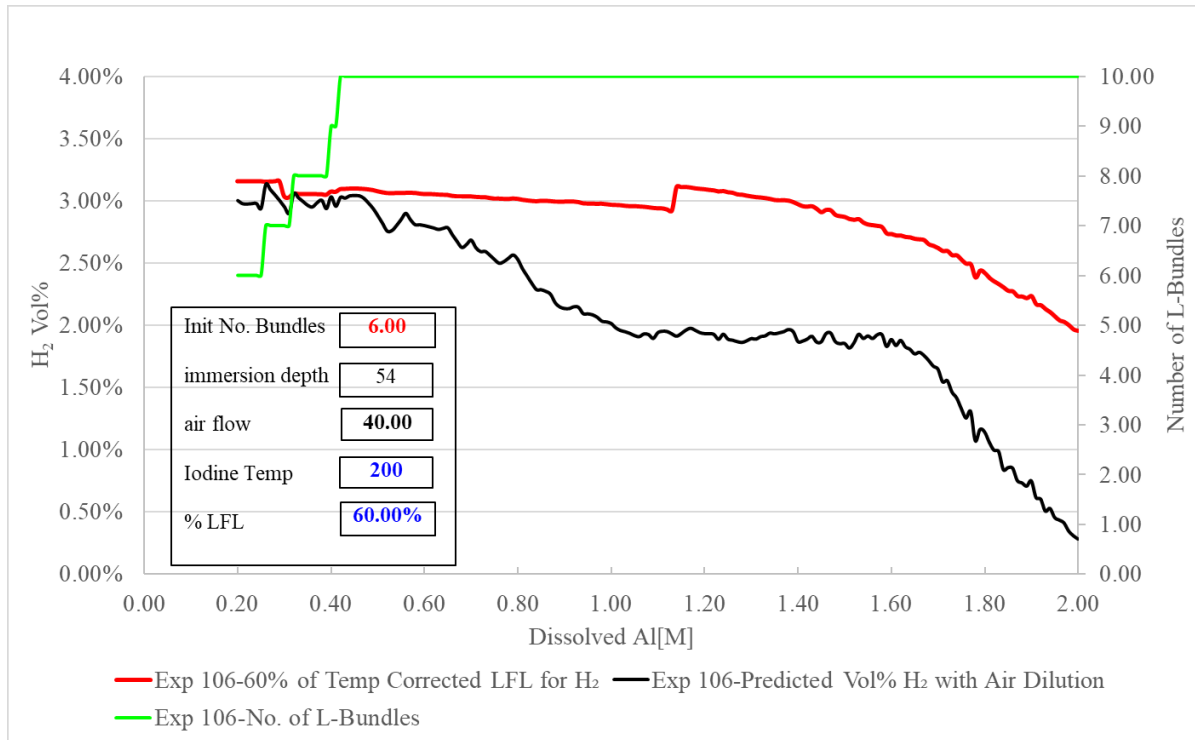


Figure 3-4. H₂ LFL Comparison for Exp. 106 – 7 M HNO₃, 0.002 M Hg, No Hold Time, and 54 in. Immersion

Table 3-4. Maximum Number of L-Bundles Allowed based on Al Concentration and H₂ vol % + 0.60 vol % Uncertainty with 0.002 M Hg at a 54 in. Immersion Depth

Dissolved Al [M]	No. L-Bundles
$0.00 < \text{Al[M]} \leq 0.25$	6
$0.25 < \text{Al[M]} \leq 0.31$	7
$0.31 < \text{Al[M]} \leq 0.39$	8
$0.39 < \text{Al[M]} \leq 0.41$	9
$0.41 < \text{Al[M]} \leq 2.00$	10

Other aspects of the flammable gas calculations have built-in conservatisms which further address the uncertainty in the calculations. The calculated H₂ concentration in the offgas stream is compared to 60% of the LFL which provides a layer of conservatism. The saturated water vapor in the offgas stream is ignored and would further dilute the H₂ concentration. Given the conservative nature of the experimental design and the calculations performed, the maximum number of L-Bundles that can be charged to an H-Canyon dissolver using nominally 0.002 M Hg to catalyze the dissolution adequately incorporates the many uncertainties associated with the experimental and modeling work.

3.4 Effect of Changes in Conditions on the Allowable Number of L-Bundles Charged to a Dissolver

In the second phase of this study, the L-Bundle submergence level, dissolver sparge/purge flowrate, operating temperature of the iodine reactor, and the percentage of the LFL which the H₂ concentration must not exceed were systematically varied to calculate the effect on the number of bundles which can be charged to a dissolver. The range of interest for each variable is shown in Table 3-5. Increases in the LFL beyond 60% of the calculated value were evaluated in this study; although, the concentration of H₂ in the offgas from the dissolvers is constrained to 60% of the LFL based on the guidance in NFPA 69 that was used to develop the dissolution flowsheets.^{2,4} The use of NFPA 69 no longer applies to the dissolver offgas, but H-Canyon will continue to use the standard as a guide to establish H₂ concentration limits in the dissolvers and dissolver offgas systems.¹⁵ During this evaluation, the maximum number of L-Bundles which can be charged to a dissolver was calculated using the methodology discussed in Section 3.3 over the range of the variable while holding the other variables at their normal value (i.e., 54 in. immersion depth, 40 scfm sparge/purge flowrate, iodine reactor operating temperature of 200 °C, and 60% of the calculated H₂ LFL). The impact of changing more than one variable at a time was not evaluated at this time.

Table 3-5. Range of Variables for Calculating Maximum Allowable Number of L-Bundles

Variable	Range
Immersion Depth	44-60 in.
Air Sparge/Purge	40-60 scfm
Iodine Reactor Temperature	150-200 °C
% LFL	60-76%

3.4.1 Effect of Immersion Depth on Flammable Gas Calculations

The immersion depth of the L-Bundles in an H-Canyon dissolver is normally controlled at 54 in. However, it may be advantageous to increase the solution level to increase the amount of Al which can be dissolved in a batch and the amount of solution which must be evaporated before the minimum liquid level is reached in the smaller 6.1D dissolver. If the solution level in a dissolver increases, the increased L-Bundle immersion depth increases the exposed outer surface area which will lower the maximum number of L-Bundles which can be charged to a dissolver as a function of the Al concentration. Alternatively, the immersion depth of the L-Bundles could be decreased below the normal 54 in. level reducing the exposed surface area and increasing the maximum number of bundles which can be charged to a dissolver.

The maximum number of L-Bundles based on maintaining the H₂ concentration below 60% of the LFL at 200 °C with a 40 scfm sparge/purge flowrate was calculated for immersion depths of 44 to 60 in. at 2 in. increments following the same method as discussed in Section 3.3. The results of these calculations are shown in Table 3-6. As the immersion depth increases, the maximum number of L-Bundles goes down slightly for each Al concentration range; however, by increasing the immersion depth, the amount of Al which can be dissolved per batch slightly increases. A reduction in the number of L-Bundles which can be charged at low Al concentrations in the dissolver can be recovered when a larger number of bundles is allowed at higher concentrations, thus maintaining the efficiency of the dissolver. The effect of changing the L-Bundle immersion depth is shown graphically in Figure 3-5 as a function of the Al concentration. The immersion depth (44 to 60 in.) was normalized prior to plotting the data.

The number of L-Bundles which can be charged to a dissolver above a 56-in. immersion level is restricted to five bundles due to the increasing surface area (Table 3-6). However, if a minimum of six bundles is desired, the offgas data from Experiment 97 can be used to calculate the maximum number of bundles in the initial charge. If the dissolution flowsheet for the initial charge of MTR fuel is defined using this data, the H-Canyon dissolving solution must be held at the boiling point for 45 minutes before starting the Hg addition. The transition from the Experiment 97 to the Experiment 106 data occurs at the Al concentration where 6 L-Bundles are initially allowed at immersion depths of 58 and 60 in. (0.17 and 0.19 M, respectively) (Table 3-6). Using the H₂ generation rates calculated from the Experiment 97 and 106 data, the impact of the immersion depth on the number of L-Bundles which can be charged to a dissolver is shown in Table 3-7. The effect of changing the L-Bundle immersion depth is shown graphically in Figure 3-6 as a function of the Al concentration. As in Figure 3-5, the immersion depth (44 to 60 in.) was normalized prior to plotting the data. The primary difference in the number of bundles which can be charged to a dissolver given in Table 3-6 and Table 3-7 is that holding the solution at the boiling point for 45 minutes prior to the Hg addition allows 6 bundles to be initially charged at immersion depths of 58 and 60 in. This approach is consistent with the approach used for the current MTR fuel dissolution flowsheet in which the data from both Experiments 97 and 106 were used to define the number of bundles which can be charged to a dissolver.²

**Table 3-6. Maximum Number of L-Bundles Based on Al Concentration and H₂ vol % + 0.60 vol %
Uncertainty with 0.002 M Hg at Varying L-Bundle Immersion Depths using Exp 106 Data**

Immersion Depth (in.)	Dissolved Al [M]	No. L-Bundles
44	$0.00 < \text{Al}[\text{M}] \leq 0.20$	7
	$0.20 < \text{Al}[\text{M}] \leq 0.27$	8
	$0.27 < \text{Al}[\text{M}] \leq 0.33$	9
	$0.33 < \text{Al}[\text{M}] \leq 2.00$	10
46	$0.00 < \text{Al}[\text{M}] \leq 0.25$	7
	$0.25 < \text{Al}[\text{M}] \leq 0.31$	8
	$0.31 < \text{Al}[\text{M}] \leq 0.38$	9
	$0.38 < \text{Al}[\text{M}] \leq 2.00$	10
48	$0.00 < \text{Al}[\text{M}] \leq 0.26$	7
	$0.26 < \text{Al}[\text{M}] \leq 0.32$	8
	$0.32 < \text{Al}[\text{M}] \leq 0.39$	9
	$0.39 < \text{Al}[\text{M}] \leq 2.00$	10
50	$0.00 < \text{Al}[\text{M}] \leq 0.19$	6
	$0.19 < \text{Al}[\text{M}] \leq 0.27$	7
	$0.27 < \text{Al}[\text{M}] \leq 0.38$	8
	$0.39 < \text{Al}[\text{M}] \leq 0.40$	9
	$0.40 < \text{Al}[\text{M}] \leq 2.00$	10
52	$0.00 < \text{Al}[\text{M}] \leq 0.24$	6
	$0.24 < \text{Al}[\text{M}] \leq 0.30$	7
	$0.30 < \text{Al}[\text{M}] \leq 0.38$	8
	$0.38 < \text{Al}[\text{M}] \leq 0.40$	9
	$0.40 < \text{Al}[\text{M}] \leq 2.00$	10
54	$0.00 < \text{Al}[\text{M}] \leq 0.25$	6
	$0.25 < \text{Al}[\text{M}] \leq 0.31$	7
	$0.31 < \text{Al}[\text{M}] \leq 0.39$	8
	$0.39 < \text{Al}[\text{M}] \leq 0.41$	9
	$0.41 < \text{Al}[\text{M}] \leq 2.00$	10
56	$0.00 < \text{Al}[\text{M}] \leq 0.26$	6
	$0.26 < \text{Al}[\text{M}] \leq 0.33$	7
	$0.33 < \text{Al}[\text{M}] \leq 0.39$	8
	$0.39 < \text{Al}[\text{M}] \leq 0.41$	9
	$0.41 < \text{Al}[\text{M}] \leq 2.00$	10
58	$0.00 < \text{Al}[\text{M}] \leq 0.17$	5
	$0.17 < \text{Al}[\text{M}] \leq 0.27$	6
	$0.27 < \text{Al}[\text{M}] \leq 0.38$	7
	$0.38 < \text{Al}[\text{M}] \leq 0.40$	8
	$0.40 < \text{Al}[\text{M}] \leq 0.47$	9
	$0.47 < \text{Al}[\text{M}] \leq 2.00$	10
60	$0.00 < \text{Al}[\text{M}] \leq 0.19$	5
	$0.19 < \text{Al}[\text{M}] \leq 0.30$	6
	$0.30 < \text{Al}[\text{M}] \leq 0.39$	7
	$0.39 < \text{Al}[\text{M}] \leq 0.40$	8
	$0.40 < \text{Al}[\text{M}] \leq 0.48$	9
	$0.48 < \text{Al}[\text{M}] \leq 2.00$	10

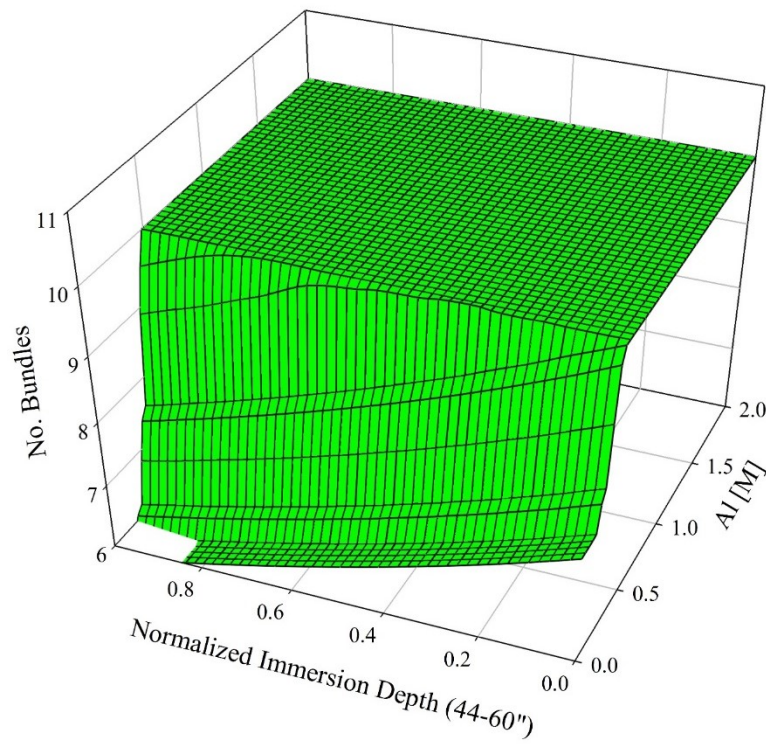


Figure 3-5. Maximum Number of L-Bundles as Functions of Normalized Immersion Depth (44 to 60 in.) and Al Concentration using Exp 106 Data

Table 3-7. Maximum Number of L-Bundles Based on Al Concentration and H₂ vol % + 0.60 vol % Uncertainty with 0.002 M Hg at Varying L-Bundle Immersion Depths using Exp 97 and 106 Data
(Initial Charge held at the solution boiling point for 45 minutes prior to starting Hg addition.)

Immersion Depth (in.)	Dissolved Al [M]	No. L-Bundles
44	$0.00 < \text{Al}[\text{M}] \leq 0.20$	7
	$0.20 < \text{Al}[\text{M}] \leq 0.27$	8
	$0.27 < \text{Al}[\text{M}] \leq 0.33$	9
	$0.33 < \text{Al}[\text{M}] \leq 2.00$	10
46	$0.00 < \text{Al}[\text{M}] \leq 0.25$	7
	$0.25 < \text{Al}[\text{M}] \leq 0.31$	8
	$0.31 < \text{Al}[\text{M}] \leq 0.38$	9
	$0.38 < \text{Al}[\text{M}] \leq 2.00$	10
48	$0.00 < \text{Al}[\text{M}] \leq 0.26$	7
	$0.26 < \text{Al}[\text{M}] \leq 0.32$	8
	$0.32 < \text{Al}[\text{M}] \leq 0.39$	9
	$0.39 < \text{Al}[\text{M}] \leq 2.00$	10
50	$0.00 < \text{Al}[\text{M}] \leq 0.27$	7
	$0.27 < \text{Al}[\text{M}] \leq 0.38$	8
	$0.39 < \text{Al}[\text{M}] \leq 0.40$	9
	$0.40 < \text{Al}[\text{M}] \leq 2.00$	10
52	$0.00 < \text{Al}[\text{M}] \leq 0.24$	6
	$0.24 < \text{Al}[\text{M}] \leq 0.30$	7
	$0.30 < \text{Al}[\text{M}] \leq 0.38$	8
	$0.38 < \text{Al}[\text{M}] \leq 0.40$	9
	$0.40 < \text{Al}[\text{M}] \leq 2.00$	10
54	$0.00 < \text{Al}[\text{M}] \leq 0.25$	6
	$0.25 < \text{Al}[\text{M}] \leq 0.31$	7
	$0.31 < \text{Al}[\text{M}] \leq 0.39$	8
	$0.39 < \text{Al}[\text{M}] \leq 0.41$	9
	$0.41 < \text{Al}[\text{M}] \leq 2.00$	10
56	$0.00 < \text{Al}[\text{M}] \leq 0.26$	6
	$0.26 < \text{Al}[\text{M}] \leq 0.33$	7
	$0.33 < \text{Al}[\text{M}] \leq 0.39$	8
	$0.39 < \text{Al}[\text{M}] \leq 0.41$	9
	$0.41 < \text{Al}[\text{M}] \leq 2.00$	10
58	$0.00 < \text{Al}[\text{M}] \leq 0.27$	6
	$0.27 < \text{Al}[\text{M}] \leq 0.38$	7
	$0.38 < \text{Al}[\text{M}] \leq 0.40$	8
	$0.40 < \text{Al}[\text{M}] \leq 0.47$	9
	$0.47 < \text{Al}[\text{M}] \leq 2.00$	10
60	$0.00 < \text{Al}[\text{M}] \leq 0.30$	6
	$0.30 < \text{Al}[\text{M}] \leq 0.39$	7
	$0.39 < \text{Al}[\text{M}] \leq 0.40$	8
	$0.40 < \text{Al}[\text{M}] \leq 0.48$	9
	$0.48 < \text{Al}[\text{M}] \leq 2.00$	10

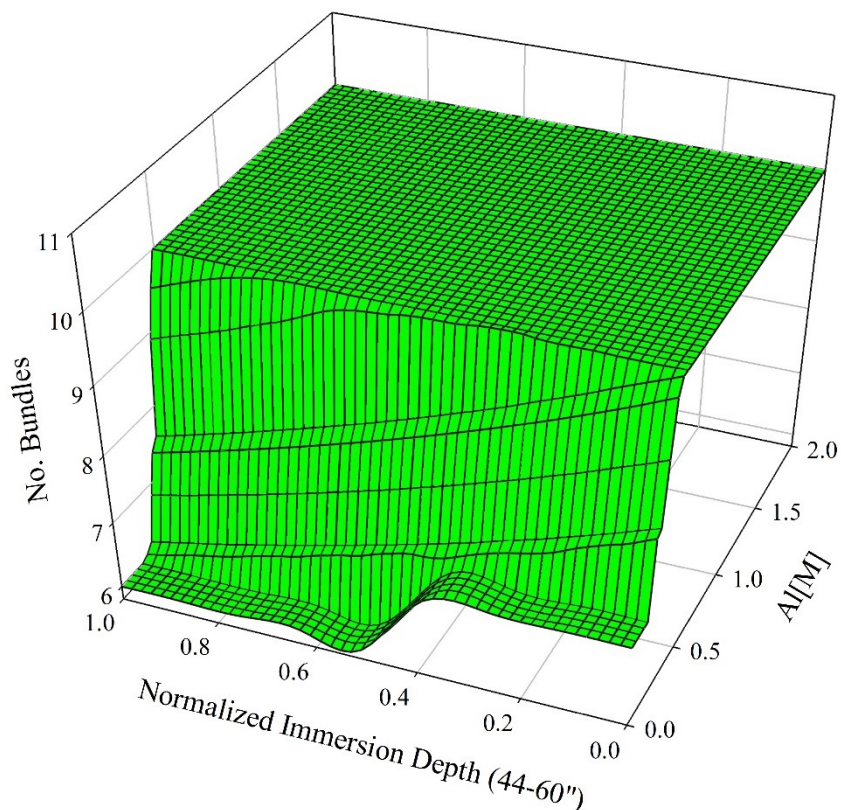


Figure 3-6. Maximum Number of L-Bundles as Functions of Normalized Immersion Depth (44 to 60 in.) and Al Concentration using Exp 97 and 106 Data

(Initial Charge held at the solution boiling point for 45 minutes prior to starting Hg addition.)

3.4.2 Effect of Sparge/Purge Flowrate on Flammable Gas Calculations

The air sparge/purge rate for an H-Canyon dissolver is normally controlled at 40 scfm. However, it may be advantageous to increase the flowrate to increase the number of L-Bundles that can be added to the dissolver during each charge. Increasing the air sparge/purge flowrate dilutes the H_2 in the offgas thus allowing more L-Bundles to be charged while staying below 60% of the calculated H_2 LFL. The maximum number of L-Bundles based on maintaining the H_2 concentration below 60% of the LFL at 200 °C and 54 in. immersion was calculated for air sparge/purge flowrates of 40 to 60 scfm at 2 scfm increments following the same method as discussed in Section 3.3. The results of these calculations are shown in Table 3-8. As the air sparge/purge rate increases, the maximum number of L-Bundles increases for each Al concentration range. The effect of increasing the air sparge/purge rate is shown graphically in Figure 3-7 as a function of the Al concentration. The sparge/purge flowrate (40 to 60 scfm) was normalized prior to plotting the data.

Table 3-8. Maximum Number of L-Bundles Allowed based on Al Concentration and H₂ vol % + 0.60 vol % Uncertainty at Varying Air Sparge/Purge Rates using Exp 106 Data

Air Sparge/Purge Rate (scfm)	Dissolved Al [M]	No. L-Bundles
40	$0.00 < \text{Al}[\text{M}] \leq 0.25$	6
	$0.25 < \text{Al}[\text{M}] \leq 0.31$	7
	$0.31 < \text{Al}[\text{M}] \leq 0.39$	8
	$0.39 < \text{Al}[\text{M}] \leq 0.41$	9
	$0.41 < \text{Al}[\text{M}] \leq 2.00$	10
42	$0.00 < \text{Al}[\text{M}] \leq 0.20$	6
	$0.20 < \text{Al}[\text{M}] \leq 0.30$	7
	$0.30 < \text{Al}[\text{M}] \leq 0.38$	8
	$0.38 < \text{Al}[\text{M}] \leq 0.40$	9
	$0.40 < \text{Al}[\text{M}] \leq 2.00$	10
44	$0.00 < \text{Al}[\text{M}] \leq 0.16$	6
	$0.16 < \text{Al}[\text{M}] \leq 0.26$	7
	$0.26 < \text{Al}[\text{M}] \leq 0.32$	8
	$0.32 < \text{Al}[\text{M}] \leq 0.39$	9
	$0.39 < \text{Al}[\text{M}] \leq 2.00$	10
46	$0.00 < \text{Al}[\text{M}] \leq 0.25$	7
	$0.25 < \text{Al}[\text{M}] \leq 0.31$	8
	$0.31 < \text{Al}[\text{M}] \leq 0.38$	9
	$0.38 < \text{Al}[\text{M}] \leq 2.00$	10
48	$0.00 < \text{Al}[\text{M}] \leq 0.20$	7
	$0.20 < \text{Al}[\text{M}] \leq 0.27$	8
	$0.27 < \text{Al}[\text{M}] \leq 0.38$	9
	$0.38 < \text{Al}[\text{M}] \leq 2.00$	10
50	$0.00 < \text{Al}[\text{M}] \leq 0.17$	7
	$0.17 < \text{Al}[\text{M}] \leq 0.26$	8
	$0.26 < \text{Al}[\text{M}] \leq 0.31$	9
	$0.31 < \text{Al}[\text{M}] \leq 2.00$	10
52	$0.00 < \text{Al}[\text{M}] \leq 0.25$	8
	$0.25 < \text{Al}[\text{M}] \leq 0.30$	9
	$0.31 < \text{Al}[\text{M}] \leq 2.00$	10
54	$0.00 < \text{Al}[\text{M}] \leq 0.20$	8
	$0.20 < \text{Al}[\text{M}] \leq 0.27$	9
	$0.27 < \text{Al}[\text{M}] \leq 2.00$	10
56	$0.00 < \text{Al}[\text{M}] \leq 0.17$	8
	$0.17 < \text{Al}[\text{M}] \leq 0.26$	9
	$0.26 < \text{Al}[\text{M}] \leq 2.00$	10
58	$0.00 < \text{Al}[\text{M}] \leq 0.24$	9
	$0.24 < \text{Al}[\text{M}] \leq 2.00$	10
60	$0.00 < \text{Al}[\text{M}] \leq 0.20$	9
	$0.20 < \text{Al}[\text{M}] \leq 2.00$	10

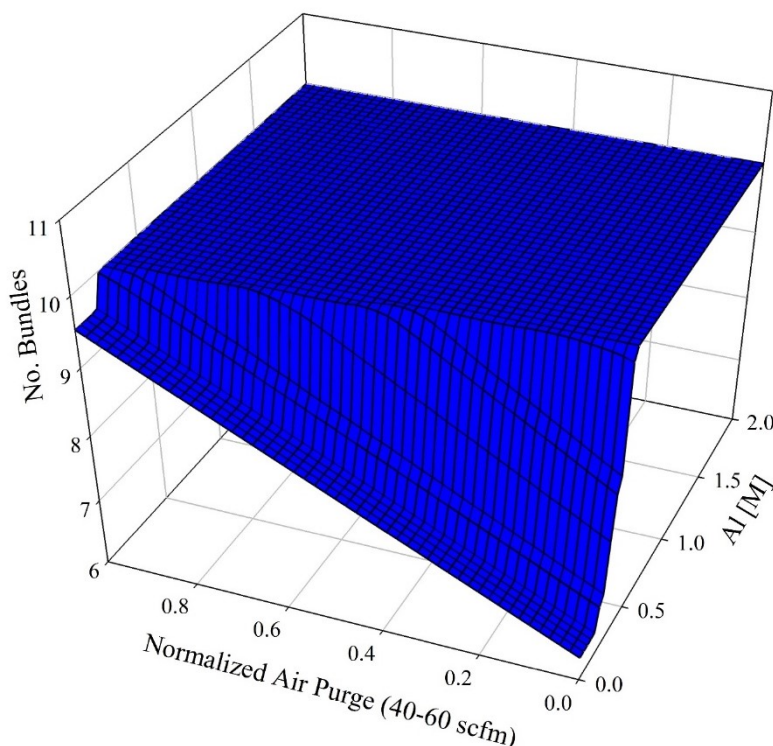


Figure 3-7. Maximum Number of L-Bundles as Functions of Normalized Air Sparge/Purge (40 to 60 scfm) and Al Concentration using Exp 106 Data

3.4.3 Effect of the Iodine Reactor Operating Temperature on Flammable Gas Calculations

The steam-heated iodine reactors are operated to maintain the temperature of the offgas and remove volatile I_2 primarily from short-cooled irradiated fuel. The operating temperature is nominally 200 °C. The maximum number of L-Bundles which can be charged to a dissolver is based on maintaining the H_2 concentration below 60% of the LFL at 200 °C and 54 in. immersion with an air sparge/purge rate of 40 scfm. Decreasing the operating temperature of the reactor below 200 °C will increase the calculated LFL of H_2 which increases with decreasing temperature (equation 4). If the iodine reactor temperature is varied from 150 to 200 °C at 10 °C increments, the maximum number of L-Bundles increases for each Al concentration range as the temperature decreases. The results of these calculations are shown in Table 3-9. The effect of decreasing the iodine reactor operating temperature is shown graphically in Figure 3-8 as a function of the Al concentration. The operating temperature (150 to 200 °C) was normalized prior to plotting the data.

Table 3-9. Maximum Number of L-Bundles Based on Al Concentration and H₂ vol % + 0.60 vol % Uncertainty at Varying Iodine Reactor Operating Temperatures using Exp 106 Data

Iodine Reactor Operating Temperature (°C)	Dissolved Al [M]	No. L-Bundles
150	$0.00 < \text{Al}[\text{M}] \leq 0.25$	7
	$0.25 < \text{Al}[\text{M}] \leq 0.30$	8
	$0.30 < \text{Al}[\text{M}] \leq 0.38$	9
	$0.38 < \text{Al}[\text{M}] \leq 2.00$	10
160	$0.00 < \text{Al}[\text{M}] \leq 0.26$	7
	$0.26 < \text{Al}[\text{M}] \leq 0.31$	8
	$0.31 < \text{Al}[\text{M}] \leq 0.38$	9
	$0.38 < \text{Al}[\text{M}] \leq 2.00$	10
170	$0.00 < \text{Al}[\text{M}] \leq 0.18$	6
	$0.18 < \text{Al}[\text{M}] \leq 0.27$	7
	$0.27 < \text{Al}[\text{M}] \leq 0.32$	8
	$0.32 < \text{Al}[\text{M}] \leq 0.39$	9
	$0.39 < \text{Al}[\text{M}] \leq 2.00$	10
180	$0.00 < \text{Al}[\text{M}] \leq 0.20$	6
	$0.20 < \text{Al}[\text{M}] \leq 0.28$	7
	$0.28 < \text{Al}[\text{M}] \leq 0.38$	8
	$0.38 < \text{Al}[\text{M}] \leq 0.40$	9
	$0.40 < \text{Al}[\text{M}] \leq 2.00$	10
190	$0.00 < \text{Al}[\text{M}] \leq 0.24$	6
	$0.24 < \text{Al}[\text{M}] \leq 0.30$	7
	$0.30 < \text{Al}[\text{M}] \leq 0.38$	8
	$0.38 < \text{Al}[\text{M}] \leq 0.40$	9
	$0.40 < \text{Al}[\text{M}] \leq 2.00$	10
200	$0.00 < \text{Al}[\text{M}] \leq 0.25$	6
	$0.25 < \text{Al}[\text{M}] \leq 0.31$	7
	$0.31 < \text{Al}[\text{M}] \leq 0.39$	8
	$0.39 < \text{Al}[\text{M}] \leq 0.41$	9
	$0.41 < \text{Al}[\text{M}] \leq 2.00$	10

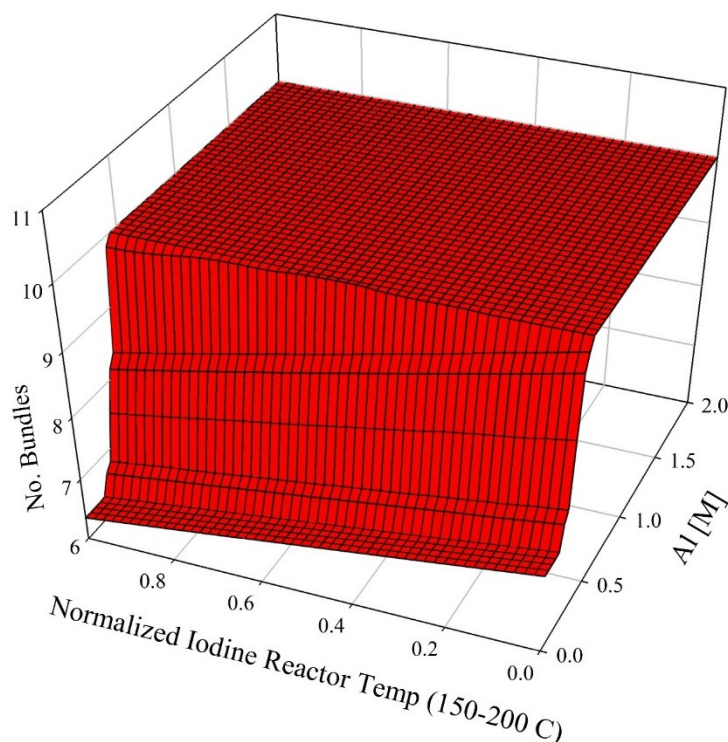


Figure 3-8. Maximum Number of L-Bundles as Functions of Normalized Iodine Reactor Temperature (150 to 200 °C) and Al Concentration using Exp 106 Data

3.4.4 Effect of LFL Percentage on Flammable Gas Calculations

The criterion of not exceeding 60% of the H_2 LFL in the dissolver offgas (at 200 °C) was used for the flammable gas calculations in this study which is consistent with the criterion used for other fuel dissolution flowsheets.^{2,4,6-10} A maximum concentration of 60% of the LFL is permitted by NFPA 69 when automatic instrumentation with safety interlocks is provided.¹¹ If a low air sparge/purge flowrate is measured in an H-Canyon dissolver, an interlock closes the steam supply to the dissolver coils.¹² Therefore, the maximum number of L-Bundles which can be charged to a dissolver for the existing MTR fuel dissolution flowsheet is based on maintaining the H_2 concentration below 60% of the LFL at 200 °C and 54 in. immersion with an air sparge/purge rate of 40 scfm. If the percentage of the LFL is varied from 60 to 76% at 1% increments, the maximum number of L-Bundles increases for each Al concentration range. The results of these calculations are shown in Table 3-10. The effect of increasing the percentage of the LFL is shown graphically in Figure 3-9 as a function of the Al concentration. The percentage of the LFL (60 to 76%) was normalized prior to plotting the data.

Table 3-10. Maximum Number of L-Bundles based on Al Concentration and H₂ vol % + 0.60 vol % Uncertainty at Varying Percentages of the LFL using Exp 106 Data

% LFL	Dissolved Al [M]	No. L-Bundles	% LFL	Dissolved Al [M]	No. L-Bundles
60	$0.00 < \text{Al}[\text{M}] \leq 0.25$	6	66	$0.00 < \text{Al}[\text{M}] \leq 0.19$	7
	$0.25 < \text{Al}[\text{M}] \leq 0.31$	7		$0.19 < \text{Al}[\text{M}] \leq 0.27$	8
	$0.31 < \text{Al}[\text{M}] \leq 0.39$	8		$0.27 < \text{Al}[\text{M}] \leq 0.31$	9
	$0.39 < \text{Al}[\text{M}] \leq 0.41$	9		$0.31 < \text{Al}[\text{M}] \leq 2.00$	10
	$0.41 < \text{Al}[\text{M}] \leq 2.00$	10	67	$0.00 < \text{Al}[\text{M}] \leq 0.17$	7
61	$0.00 < \text{Al}[\text{M}] \leq 0.24$	6		$0.17 < \text{Al}[\text{M}] \leq 0.26$	8
	$0.24 < \text{Al}[\text{M}] \leq 0.30$	7		$0.26 < \text{Al}[\text{M}] \leq 0.30$	9
	$0.30 < \text{Al}[\text{M}] \leq 0.38$	8		$0.30 < \text{Al}[\text{M}] \leq 2.00$	10
	$0.38 < \text{Al}[\text{M}] \leq 0.40$	9	68	$0.00 < \text{Al}[\text{M}] \leq 0.25$	8
	$0.40 < \text{Al}[\text{M}] \leq 2.00$	10		$0.25 < \text{Al}[\text{M}] \leq 0.29$	9
62	$0.00 < \text{Al}[\text{M}] \leq 0.19$	6		$0.29 < \text{Al}[\text{M}] \leq 2.00$	10
	$0.19 < \text{Al}[\text{M}] \leq 0.27$	7	69	$0.00 < \text{Al}[\text{M}] \leq 0.24$	8
	$0.27 < \text{Al}[\text{M}] \leq 0.33$	8		$0.24 < \text{Al}[\text{M}] \leq 0.28$	9
	$0.33 < \text{Al}[\text{M}] \leq 0.39$	9		$0.28 < \text{Al}[\text{M}] \leq 2.00$	10
	$0.39 < \text{Al}[\text{M}] \leq 2.00$	10	70	$0.00 < \text{Al}[\text{M}] \leq 0.19$	8
63	$0.00 < \text{Al}[\text{M}] \leq 0.26$	7		$0.19 < \text{Al}[\text{M}] \leq 0.27$	9
	$0.26 < \text{Al}[\text{M}] \leq 0.31$	8		$0.27 < \text{Al}[\text{M}] \leq 2.00$	10
	$0.31 < \text{Al}[\text{M}] \leq 0.39$	9	71	$0.00 < \text{Al}[\text{M}] \leq 0.18$	8
	$0.39 < \text{Al}[\text{M}] \leq 2.00$	10		$0.18 < \text{Al}[\text{M}] \leq 0.26$	9
64	$0.00 < \text{Al}[\text{M}] \leq 0.25$	7		$0.26 < \text{Al}[\text{M}] \leq 2.00$	10
	$0.25 < \text{Al}[\text{M}] \leq 0.30$	8	72	$0.00 < \text{Al}[\text{M}] \leq 0.25$	9
	$0.30 < \text{Al}[\text{M}] \leq 0.38$	9		$0.25 < \text{Al}[\text{M}] \leq 2.00$	10
	$0.38 < \text{Al}[\text{M}] \leq 2.00$	10	73	$0.00 < \text{Al}[\text{M}] \leq 0.24$	9
	$0.00 < \text{Al}[\text{M}] \leq 0.24$	7		$0.24 < \text{Al}[\text{M}] \leq 2.00$	10
65	$0.24 < \text{Al}[\text{M}] \leq 0.28$	8	74	$0.00 < \text{Al}[\text{M}] \leq 0.19$	9
	$0.28 < \text{Al}[\text{M}] \leq 0.32$	9		$0.19 < \text{Al}[\text{M}] \leq 2.00$	10
	$0.32 < \text{Al}[\text{M}] \leq 2.00$	10	75	$0.00 < \text{Al}[\text{M}] \leq 0.17$	9
				$0.17 < \text{Al}[\text{M}] \leq 2.00$	10
			76	$0.00 < \text{Al}[\text{M}] \leq 2.00$	10

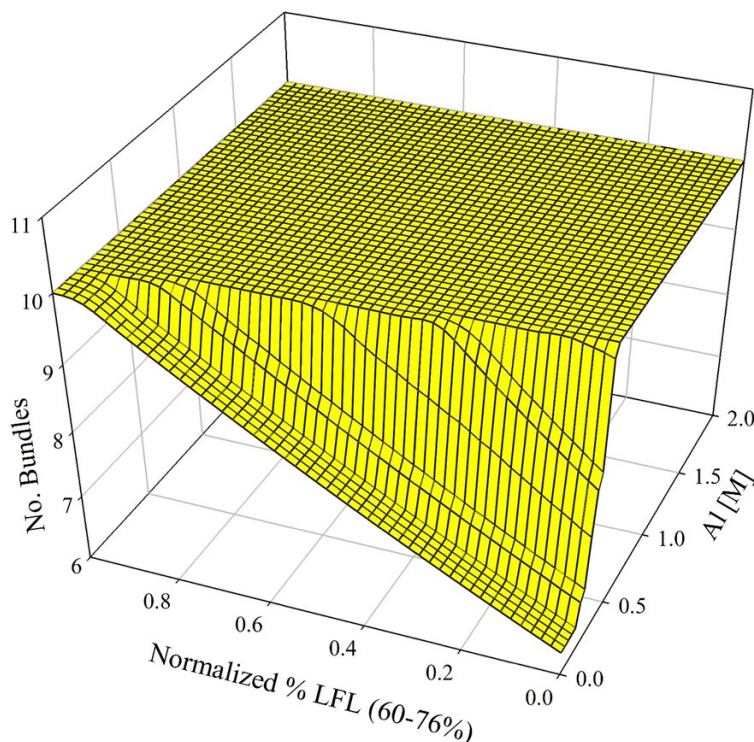


Figure 3-9. Maximum Number of L-Bundles as Functions of Normalized Percentage of the LFL (60 to 76%) and Al Concentration using Exp 106 Data

3.5 Relative Effect of Flowsheet Variables on the Number of L-Bundles Charged to a Dissolver

To compare the impact of the four dissolver parameters on the maximum number of L-Bundles which can be charged to a dissolver while staying below the calculated H_2 LFL, data from the three dimensional plots of the normalized parameters (Figure 3-5 to Figure 3-9) were used to construct a two dimensional plot of their relative influence. To construct the plot, the changes in the number of bundles over the normalized range of each variable at zero Al were plotted on Figure 3-10. Zero Al in the dissolving solutions corresponds to the front edge of the three-dimensional plots when the number of L-Bundles which can be charged to a dissolver is at the minimum value. The absolute value of the slope provides a measure of the relative influence of each parameter on the maximum number of L-Bundles which can be charged to a dissolver. The larger the magnitude of the slope, the larger the change in the number of bundles over the range of the parameter. For each parameter plot on Figure 3-10, the other three parameters were held at their existing flowsheet values (i.e., 54 in. immersion depth, 40 scfm sparge/purge flowrate, iodine reactor operating temperature of 200 °C, and 60% of the calculated H_2 LFL).² The percentage of the LFL had the largest influence on the maximum number of bundles followed by the air sparge/purge flowrate, L-Bundle immersion depth, and the iodine reactor operating temperature.

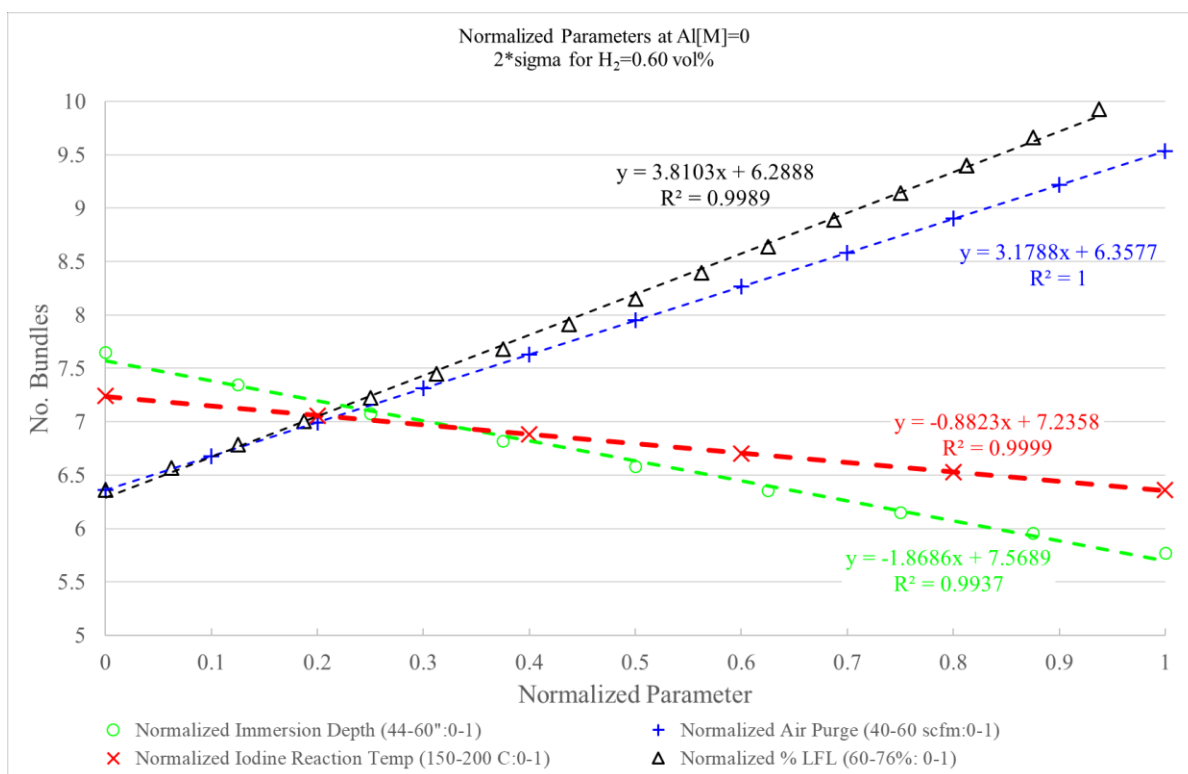


Figure 3-10. Maximum Number of L-Bundles as a Function of Normalized Dissolver Parameters at 0 M Al

4.0 Conclusions

Since the development of the existing MTR fuel dissolution flowsheet, improved offgas measurement methods have been developed to more accurately calculate the offgas composition and generation rate during laboratory dissolutions. In the first part of this study, improvements in the Raman spectrometer calibration model and a more effective data smoothing routine were applied to existing data. The uncertainty in the offgas composition data used to define the MTR fuel dissolution flowsheet were subsequently re-evaluated which resulted in an approximate 50% reduction in the uncertainty of the H₂ gas concentration measurements. The reduced uncertainty was used in new calculations to define the maximum number of L-Bundles which can be charged to either the 6.1D or 6.4D dissolver as a function of the Al concentration. With the reduced uncertainty in the H₂ concentration measurements, a flowsheet was developed which supported initially charging 6 bundles to a dissolver and the addition of the Hg catalyst as soon as the solution reached the boiling point (Table 3-4). This flowsheet is based on an L-Bundle immersion depth of 54 in., an air sparge/purge flowrate of 40 scfm, an iodine reactor operating temperature of 200 °C, and not exceeding 60% of the H₂ LFL.

In the second phase of this study, calculations were performed to assess the impact of changing various dissolver operating parameters to increase the number of L-Bundles charged to an H-Canyon dissolver. Applying the improved offgas measurement techniques including the reduced uncertainty in the H₂ concentration measurements, the number of bundles of MTR fuel which can be charged to either the 6.1D or 6.4D dissolver were calculated based on varying the bundle immersion depth (44-60 in.), air sparge/purge flowrate (40-60 scfm), iodine reactor operating temperature (150-200 °C), and the percent of the H₂ LFL which must not be exceeded (60-76%). Qualitatively, the percentage of the LFL had the largest influence on the maximum number of bundles which can be charged to a dissolver followed by the air sparge/purge flowrate, L-Bundle immersion depth, and the iodine reactor operating temperature. During the evaluation

of each variable, the other variables were held at the baseline flowsheet conditions. The impact of changing more than one variable at a time was not evaluated.

The effect of varying the immersion level on the maximum number of L-Bundles which can be charged to a dissolver was evaluated for two flowsheet options. Table 3-6 provides the charging limitations based on starting the Hg addition as soon as the solution reaches the boiling point. Since only 5 bundles can be charged at immersion levels of 58 and 60 in., a second flowsheet option was developed using offgas generation data based on starting the Hg addition 45 minutes after the solution reaches its boiling point. Table 3-7 provides the maximum number of bundles which can be charged to the dissolver based on this restriction. The effects of the air sparge/purge flowrate, the iodine reactor operating temperature, and the percent of the H₂ LFL which must not be exceeded on the number of L-Bundles which can be charged to a dissolver are provided in Table 3-8, Table 3-9, and Table 3-10, respectively.

5.0 Flowsheet Recommendations

Flowsheet recommendations were developed for the dissolution of MTR fuels using either the 6.4D or 6.1D dissolver. The process conditions are summarized below which are required to ensure that the predicted H₂ concentration in the offgas from dissolution is less than a specified percentage of the H₂ LFL at an L-Bundle immersion depth, air sparge/purge flowrate, and iodine reactor operating temperature evaluated by this study.

- The baseline immersion depth of the L-Bundles in the dissolver solution is 54 in., but flammable gas calculations were performed for immersion depths of 44-60 in.
- A dissolver air sparge/purge flowrate of 40 scfm is normally used in the H-Canyon dissolvers, but flammable gas calculations were performed for air sparge/purge flowrates of 40-60 scfm.
- The iodine reactor operating temperature is normally 200 °C, but flammable gas calculations were performed for temperatures of 150-200 °C.
- The percentage of the H₂ LFL which must not be exceeded in the dissolver offgas is normally 60%, but flammable gas calculations were performed for H₂ LFL percentages of 60-76%.
- The initial HNO₃ concentration shall be based on the mass of Al and U charged to the dissolver. The concentration is typically 5 to 7.5 M.
- A Hg concentration of nominally 0.002 M shall be used to catalyze the dissolution of the MTR fuel. Variations in the Hg concentration added to the dissolver shall not exceed $\pm 25\%$ of the required concentration (0.0020 ± 0.0005). The Hg shall be added to the 6.4D dissolver as described in Procedure 221-H-4101.¹⁶ A similar Hg addition method shall be used for the 6.1D dissolver.
 - If the Hg is added when the boiling point of the solution is reached, the number of L-Bundles which can be charged to a dissolver as a function of the Al concentration is given by Table 3-4. These limitations are based on an L-Bundle immersion depth of 54 in., an air sparge/purge flowrate of 40 scfm, an iodine reactor operating temperature of 200 °C, and not exceeding 60% of the H₂ LFL.
 - If the Hg is added 45 minutes after the solution boils, the number of L-Bundles which can be charged to a dissolver as a function of the Al concentration is given by Table 3-7. These limitations are based on L-Bundle immersion depths of 44-60 in., an air sparge/purge flowrate of 40 scfm, an iodine reactor operating temperature of 200 °C, and not exceeding 60% of the H₂ LFL.
- The HNO₃ concentration in the dissolver solution at the completion of the dissolution of a batch of MTR fuel shall be greater than 0.5 M.

Based on past MTR fuel dissolution history, the recommended flowsheet should result in an estimated dissolution time of 24 to 60 h for a dissolver charge. The dissolution time would be expected to increase as the Al concentration in the dissolver also increases.

The maximum number of L-Bundles which can be charged to an H-Canyon dissolver as a function of the Al concentration over the evaluated ranges of the bundle immersion depth (44-60 in.), air sparge/purge flowrate (40-60 scfm), iodine reactor operating temperature (150-200 °C), and the percent of the H₂ LFL which must not be exceeded (60-76%) are provided in Table 3-6, Table 3-8, Table 3-9, Table 3-10, respectively. Increases in the LFL beyond 60% of the calculated value were evaluated in this study; although, the concentration of H₂ in the offgas from the dissolvers is constrained to 60% of the LFL based on the guidance in NFPA 69 that was used to develop the MTR and HFIR dissolution flowsheets. The use of NFPA 69 no longer applies to the dissolver offgas, but H-Canyon will continue to use the standard as a guide to establish H₂ concentration limits in the dissolvers and dissolver offgas systems.¹⁵ When the number of L-Bundles which can be charged to a dissolver is based on the limitations provided in the table for a specific variable, the other three variables must be at the baseline operating conditions (i.e., 54 in. immersion depth, 40 scfm sparge/purge flowrate, iodine reactor operating temperature of 200 °C, and 60% of the calculated H₂ LFL). For these dissolver operating scenarios, the Hg catalyst may be added as soon as the solution reaches its boiling point.

6.0 References

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Appendix A. Calibration Data for Raman Spectrometer

For the Al dissolutions performed in Experiment 97 and 106, the pre-run calibration checks are shown in Table A-1 and Table A-2, respectively. The post run checks for Experiment 97 and 106 are shown in Table A-3 and Table A-4, respectively.

Table A-1. Pre-run Check of Calibration Gases for Al Dissolution Experiment 97

Gas Description	Exp.	H ₂ (%)	NO ₂ (%)	N ₂ (%)	O ₂ (%)	N ₂ O (%)	NO (%)	CO ₂ (%)	CO (%)	H ₂ O (%)	NH ₃ (%)
99.9% CO ₂	97	0.00	0.00	0.00	0.34	0.00	0.05	99.38	0.00	0.23	0.00
	97	0.00	0.00	0.05	0.03	0.00	0.00	99.69	0.20	0.03	0.00
	97	0.00	0.00	0.00	0.62	0.00	0.00	99.36	0.00	0.00	0.01
	97	0.02	0.00	0.00	0.00	0.00	0.00	99.94	0.00	0.00	0.03
	97	0.19	0.00	0.14	0.09	0.00	0.00	99.50	0.00	0.07	0.00
	97	0.00	0.00	0.00	0.34	0.00	0.00	99.66	0.00	0.00	0.00
	97	0.00	0.00	0.00	0.51	0.00	0.00	99.49	0.00	0.00	0.00
	97	0.15	0.00	0.10	0.00	0.00	0.00	99.34	0.37	0.03	0.00
	97	0.00	0.01	0.00	0.51	0.00	0.00	99.47	0.00	0.02	0.00
	97	0.00	0.00	0.00	0.72	0.00	0.00	98.80	0.44	0.04	0.00
	Avg.	0.04	0.00	0.03	0.32	0.00	0.00	99.46	0.10	0.04	0.00
Air (78.0% N ₂ -21.0% O ₂ -0.9% Ar)	97	0.16	0.00	77.76	21.00	0.21	0.35	0.19	0.00	0.33	0.00
	97	0.00	0.01	78.14	21.34	0.11	0.00	0.31	0.00	0.08	0.00
	97	0.04	0.00	79.81	19.96	0.02	0.00	0.00	0.00	0.17	0.00
	97	0.00	0.01	79.72	20.21	0.00	0.00	0.07	0.00	0.00	0.00
	97	0.00	0.00	79.33	20.18	0.08	0.00	0.00	0.36	0.01	0.05
	97	0.00	0.01	79.48	20.51	0.00	0.00	0.00	0.00	0.00	0.00
	97	0.00	0.01	79.35	20.47	0.11	0.00	0.00	0.00	0.06	0.00
	97	0.00	0.00	79.50	20.07	0.00	0.00	0.43	0.00	0.00	0.00
	97	0.00	0.01	79.20	20.26	0.00	0.00	0.00	0.53	0.00	0.00
	97	0.00	0.01	79.23	20.32	0.00	0.00	0.31	0.00	0.14	0.00
	Avg.	0.02	0.01	79.15	20.43	0.05	0.04	0.13	0.09	0.08	0.00
2.67% H ₂ -97.33% Ar	97	2.57	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.70	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.84	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.63	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.34	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.64	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.70	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.69	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Avg.	2.65	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Table A-2. Pre-run Check of Calibration Gases for Al Dissolution Experiment 106

Gas Description	Exp.	H ₂ (%)	NO ₂ (%)	N ₂ (%)	O ₂ (%)	N ₂ O (%)	NO (%)	CO ₂ (%)	CO (%)	H ₂ O (%)	NH ₃ (%)
99.9% CO ₂	106	0.00	0.00	0.00	0.00	0.00	0.00	99.95	0.00	0.00	0.05
	106	0.00	0.00	0.02	0.18	0.00	0.00	99.47	0.11	0.22	0.00
	106	0.00	0.00	0.16	0.00	0.00	0.00	99.73	0.11	0.00	0.00
	106	0.00	0.00	0.16	0.00	0.00	0.00	99.84	0.00	0.00	0.00
	106	0.01	0.00	0.00	0.00	0.00	0.00	99.57	0.35	0.00	0.07
	106	0.00	0.00	0.73	0.00	0.00	0.00	99.27	0.00	0.00	0.00
	106	0.00	0.00	0.02	0.00	0.00	0.00	99.94	0.03	0.00	0.00
	106	0.00	0.00	0.11	0.00	0.00	0.00	99.40	0.49	0.00	0.00
	106	0.04	0.00	0.06	0.00	0.00	0.00	99.90	0.00	0.00	0.00
	106	0.04	0.01	0.00	0.00	0.00	0.00	99.48	0.46	0.00	0.00
	Avg.	0.01	0.00	0.13	0.02	0.00	0.00	99.65	0.15	0.02	0.01
Air (78.0% N ₂ -21.0% O ₂ -0.9% Ar)	106	0.07	0.00	78.52	20.81	0.00	0.00	0.60	0.00	0.00	0.00
	106	0.00	0.00	77.49	20.39	1.38	0.74	0.00	0.00	0.00	0.00
	106	0.00	0.00	76.46	20.71	0.00	0.00	2.13	0.70	0.00	0.00
	106	0.00	0.00	77.66	21.60	0.02	0.00	0.73	0.00	0.00	0.00
	106	0.19	0.00	78.89	20.62	0.00	0.00	0.29	0.01	0.00	0.00
	106	0.00	0.00	78.14	19.71	0.00	0.00	2.15	0.00	0.00	0.00
	106	0.18	0.00	79.58	19.65	0.00	0.00	0.58	0.00	0.00	0.00
	106	0.01	0.00	77.01	20.58	0.00	0.00	2.36	0.00	0.00	0.04
	106	0.00	0.00	77.95	21.26	0.00	0.00	0.00	0.76	0.03	0.00
	106	0.00	0.00	79.11	20.43	0.00	0.00	0.15	0.32	0.00	0.00
	Avg.	0.05	0.00	78.08	20.58	0.14	0.07	0.90	0.18	0.00	0.00
2.67% H ₂ -97.33% Ar	106	2.46	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.56	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.52	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.53	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.23	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.64	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.74	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.43	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.71	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Avg.	2.52	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Table A-3. Post-run Check of Calibration Gases for Al Dissolution Experiment 97

Gas Description	Exp.	H ₂ (%)	NO ₂ (%)	N ₂ (%)	O ₂ (%)	N ₂ O (%)	NO (%)	CO ₂ (%)	CO (%)	H ₂ O (%)	NH ₃ (%)
99.9% CO ₂	97	0.00	0.00	0.00	0.31	0.00	0.00	99.46	0.00	0.24	0.00
	97	0.00	0.01	0.00	0.00	0.00	0.00	99.99	0.00	0.00	0.00
	97	0.00	0.01	0.00	0.75	0.00	0.00	99.17	0.00	0.03	0.05
	97	0.00	0.00	0.00	0.18	0.00	0.00	99.57	0.24	0.00	0.00
	97	0.12	0.00	0.00	0.59	0.00	0.00	99.28	0.00	0.00	0.02
	97	0.00	0.00	0.00	0.14	0.00	0.12	99.73	0.00	0.00	0.00
	97	0.10	0.00	0.00	0.29	0.00	0.00	99.58	0.00	0.00	0.02
	97	0.00	0.00	0.00	0.18	0.00	0.00	99.82	0.00	0.00	0.00
	97	0.00	0.00	0.00	0.19	0.00	0.00	99.81	0.00	0.00	0.00
	97	0.00	0.01	0.00	0.07	0.00	0.00	99.92	0.00	0.00	0.00
	Avg.	0.02	0.00	0.00	0.27	0.00	0.01	99.63	0.02	0.03	0.01
Air (78.0% N ₂ -21.0% O ₂ -0.9% Ar)	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	97	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Avg.	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
2.67% H ₂ -97.33% Ar	97	2.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.85	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.56	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.87	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.79	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.62	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.47	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	97	2.63	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Avg.	2.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

NA=Not Available

Table A-4. Post-run Check of Calibration Gases for Al Dissolution Experiment 106

Gas Description	Exp.	H ₂ (%)	NO ₂ (%)	N ₂ (%)	O ₂ (%)	N ₂ O (%)	NO (%)	CO ₂ (%)	CO (%)	H ₂ O (%)	NH ₃ (%)
99.9% CO ₂	106	0.33	0.00	0.00	0.00	0.00	0.00	99.60	0.07	0.00	0.00
	106	0.05	0.00	0.00	0.00	0.00	0.00	99.95	0.00	0.00	0.00
	106	0.00	0.00	0.00	0.00	0.00	0.00	99.75	0.25	0.00	0.00
	106	0.00	0.00	0.00	0.00	0.00	0.00	99.97	0.03	0.00	0.00
	106	0.00	0.01	0.37	0.00	0.00	0.13	99.34	0.16	0.00	0.00
	106	0.00	0.00	0.00	0.00	0.00	0.00	99.93	0.00	0.00	0.07
	106	0.00	0.00	0.00	0.00	0.00	0.00	99.17	0.83	0.00	0.00
	106	0.00	0.00	0.00	0.00	0.00	0.00	99.91	0.00	0.00	0.09
	106	0.00	0.00	0.00	0.00	0.00	0.00	99.44	0.56	0.00	0.00
	106	0.00	0.00	0.00	0.00	0.00	0.00	99.86	0.14	0.00	0.00
	Avg.	0.00	0.00	0.04	0.00	0.00	0.00	99.69	0.00	0.00	0.00
Air (78.0% N ₂ -21.0% O ₂ -0.9% Ar)	106	0.00	0.00	78.65	20.15	0.00	0.00	0.47	0.73	0.00	0.00
	106	0.00	0.00	78.80	20.13	0.00	1.08	0.00	0.00	0.00	0.00
	106	0.00	0.00	79.26	20.47	0.00	0.00	0.27	0.00	0.00	0.00
	106	0.00	0.00	79.72	19.60	0.00	0.00	0.00	0.68	0.00	0.00
	106	0.00	0.00	79.29	20.05	0.00	0.00	0.00	0.66	0.00	0.00
	106	0.00	0.00	79.33	20.55	0.00	0.00	0.12	0.00	0.00	0.00
	106	0.00	0.00	79.85	19.90	0.00	0.00	0.00	0.26	0.00	0.00
	106	0.00	0.00	78.94	20.03	0.00	0.00	0.49	0.54	0.00	0.00
	106	0.00	0.00	78.15	20.59	0.00	0.89	0.21	0.15	0.00	0.00
	106	0.00	0.00	79.65	20.22	0.00	0.13	0.00	0.00	0.00	0.00
	Avg.	0.00	0.00	79.16	20.17	0.00	0.00	0.00	0.00	0.00	0.00
2.67% H ₂ -97.33% Ar	106	2.93	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.82	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.44	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.14	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.58	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.25	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	106	2.62	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Avg.	2.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Appendix B. Raman Offgas Data for Experiment 97

The fixed, normalized, and moving average Raman gas concentration measurements for Experiment 97 are reported in Table B-1. 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 B-1. Fixed Normalized Moving Average Raman Data for Experiment 97

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
2	0.001	79.24	5.27	1.43	2.61	1.39	5.24	0.00	89.33
13	0.002	78.80	5.41	1.43	2.64	1.36	4.87	0.00	89.69
24	0.003	79.45	5.19	1.43	2.66	1.38	5.66	0.00	88.87
35	0.004	78.01	5.64	1.65	2.64	2.12	5.26	0.00	88.33
47	0.005	78.23	5.57	1.58	2.65	1.72	5.21	0.00	88.83
58	0.006	78.42	5.50	0.49	2.48	1.72	5.05	0.00	90.25
69	0.008	78.49	5.48	0.90	2.58	1.80	3.75	2.06	88.92
80	0.009	78.47	5.49	1.29	2.53	2.27	3.45	5.66	84.79
92	0.010	74.19	7.15	2.02	2.46	3.05	3.22	9.50	79.74
103	0.011	60.44	16.48	2.73	2.39	3.26	2.72	13.73	75.18
114	0.016	44.63	29.58	3.30	2.38	2.83	3.22	18.07	70.19
126	0.026	28.88	55.78	4.08	2.42	3.51	2.52	22.41	65.05
137	0.037	21.71	79.75	4.72	2.49	4.11	2.06	26.71	59.90
148	0.057	17.59	94.23	4.93	2.46	4.25	2.78	28.81	56.76
160	0.080	17.09	97.47	4.87	2.63	4.12	2.71	29.50	56.17
171	0.101	17.30	95.90	4.79	2.83	4.21	2.34	29.81	56.01
182	0.129	18.26	91.01	4.63	3.15	4.68	2.33	30.02	55.19
194	0.156	19.73	82.80	4.27	3.37	5.17	2.51	29.74	54.94
205	0.177	21.73	72.09	3.92	3.62	5.35	3.06	29.45	54.60
216	0.196	22.37	69.52	3.77	3.83	5.69	3.61	28.89	54.21
228	0.212	22.90	67.34	3.45	4.03	6.06	3.32	28.76	54.38
239	0.228	22.68	68.24	3.07	4.20	6.14	3.87	28.64	54.07
250	0.246	22.09	70.59	2.64	4.32	6.48	4.32	28.46	53.79
262	0.265	21.94	71.18	2.47	4.38	6.54	4.52	28.00	54.08
273	0.285	22.25	69.95	2.35	4.48	6.90	4.97	28.02	53.28
284	0.304	23.34	65.86	2.29	4.49	7.05	5.34	27.67	53.16
296	0.321	23.73	64.39	2.21	4.57	7.03	5.36	27.80	53.02
307	0.337	23.62	64.79	2.23	4.75	7.27	6.15	28.39	51.22
319	0.360	23.20	66.23	2.53	4.93	7.46	6.51	29.02	49.56
330	0.379	24.70	61.69	2.73	5.00	7.30	6.62	28.97	49.37

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
		(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
341	0.393	26.23	56.69	2.80	5.07	7.38	6.52	28.09	50.14
353	0.406	27.07	53.94	2.94	5.21	7.65	6.29	28.37	49.54
364	0.418	27.37	53.20	2.80	5.27	7.50	5.79	28.39	50.26
376	0.433	28.72	50.02	2.74	5.29	7.46	5.92	28.15	50.44
387	0.447	28.76	49.89	2.63	5.18	7.25	6.20	27.30	51.44
398	0.463	27.11	54.64	2.67	5.16	6.99	6.23	25.51	53.44
410	0.484	24.88	60.49	2.59	5.14	6.48	7.89	25.07	52.83
421	0.501	26.42	56.68	2.35	5.09	6.14	8.83	25.19	52.40
433	0.515	26.82	55.36	2.20	5.06	5.87	8.98	24.40	53.51
444	0.531	28.12	51.60	2.42	5.16	5.79	8.85	24.58	53.19
456	0.548	28.47	50.89	2.42	5.21	5.47	9.24	24.50	53.16
467	0.562	29.53	47.93	2.42	5.21	5.18	9.27	23.93	53.99
479	0.578	29.83	47.20	2.16	5.15	5.15	9.54	24.08	53.93
490	0.593	28.59	49.97	2.02	5.26	5.42	8.77	23.02	55.51
502	0.609	29.40	48.12	2.22	5.32	5.44	8.81	23.53	54.68
513	0.623	29.86	47.11	2.28	5.32	4.91	8.78	23.53	55.18
525	0.635	29.49	48.08	2.13	5.27	4.69	10.10	23.02	54.79
536	0.646	29.67	47.69	2.18	5.25	4.71	10.17	22.82	54.87
548	0.659	28.81	49.70	2.29	5.28	4.67	9.69	22.99	55.10
559	0.673	29.10	48.94	2.35	5.28	4.59	9.76	23.41	54.60
571	0.690	27.77	52.04	2.48	5.27	4.48	9.21	24.23	54.33
582	0.706	28.37	50.59	2.25	5.23	4.47	9.32	23.86	54.86
594	0.722	30.00	47.03	2.22	5.18	4.50	9.27	23.66	55.16
605	0.735	30.88	44.88	2.41	5.48	5.77	9.28	25.35	51.71
617	0.748	30.59	45.55	2.46	5.54	6.09	9.19	25.50	51.21
628	0.760	31.64	43.77	2.34	5.61	6.64	9.16	25.97	50.28
640	0.772	32.59	41.85	2.08	5.58	6.49	9.51	25.68	50.66
651	0.783	33.55	39.76	2.06	5.53	6.47	9.57	26.04	50.33
663	0.796	32.52	41.51	2.20	5.53	6.50	8.66	26.57	50.53
675	0.809	31.11	44.52	2.03	5.60	6.84	9.44	27.23	48.87
686	0.822	29.78	47.50	1.82	5.30	5.68	9.18	25.30	52.72
698	0.838	28.87	49.34	1.55	5.22	5.84	9.26	25.16	52.97
709	0.852	30.90	45.25	1.58	5.19	5.87	9.71	25.08	52.56
721	0.865	31.08	44.81	1.93	5.18	6.10	9.15	25.09	52.55
732	0.875	31.01	44.95	1.79	5.27	6.46	9.32	24.90	52.27
744	0.889	30.97	45.02	1.87	5.41	6.02	10.36	24.02	52.32
756	0.902	34.48	38.69	2.10	5.45	6.18	10.12	23.95	52.21

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
		(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
768	0.912	35.66	36.33	2.13	5.51	6.16	9.62	23.92	52.65
780	0.923	35.33	36.90	2.39	5.53	6.02	9.94	23.83	52.28
792	0.936	35.30	36.93	2.53	5.60	5.77	9.69	23.93	52.49
804	0.947	35.09	37.32	2.47	5.66	6.16	9.72	23.87	52.12
816	0.958	37.02	34.40	2.61	5.60	6.24	9.83	23.72	52.00
828	0.969	37.12	34.27	2.63	5.46	6.98	9.18	23.80	51.94
840	0.978	37.12	34.27	2.83	5.64	6.78	8.45	23.82	52.47
852	0.989	35.29	37.02	2.81	5.60	7.20	8.22	23.74	52.43
865	1.002	37.76	34.31	2.68	5.58	7.21	7.27	23.69	53.56
877	1.011	39.84	31.07	2.54	5.46	7.38	6.78	22.83	55.02
890	1.020	39.04	32.56	2.54	5.38	7.25	6.65	22.82	55.36
902	1.031	34.89	37.78	2.52	5.36	7.37	6.37	22.35	56.02
914	1.043	33.08	40.53	2.40	5.37	7.17	5.90	22.62	56.53
926	1.054	35.84	36.12	2.07	5.17	7.02	6.43	21.58	57.73
938	1.065	35.94	35.95	2.08	5.18	7.12	6.11	21.58	57.94
950	1.075	38.78	31.95	2.06	5.27	7.71	6.01	21.67	57.28
962	1.085	38.11	32.84	2.14	5.31	7.82	6.18	21.33	57.22
974	1.094	39.53	30.71	1.92	5.60	8.50	6.35	19.74	57.89
985	1.103	39.94	30.26	1.97	5.63	7.95	5.67	20.20	58.57
997	1.112	44.29	25.72	2.00	5.64	7.99	5.55	19.42	59.39
1009	1.118	42.96	27.70	2.11	5.61	7.93	4.67	19.59	60.09
1020	1.126	42.50	28.20	2.01	5.57	7.64	4.76	18.84	61.18
1032	1.135	38.09	32.83	2.01	5.49	7.09	4.12	18.65	62.65
1044	1.144	41.18	28.78	2.09	5.42	7.00	3.85	18.90	62.74
1056	1.152	38.84	31.97	2.17	5.23	6.34	2.82	19.87	63.57
1067	1.160	38.57	32.36	2.41	5.32	7.11	3.69	19.43	62.04
1079	1.171	39.33	31.62	2.08	5.43	7.48	3.33	19.62	62.06
1091	1.179	43.72	26.40	2.15	5.32	7.90	4.79	18.69	61.15
1103	1.186	47.85	21.83	2.21	5.45	7.96	4.95	19.75	59.68
1115	1.192	46.53	23.17	2.19	5.45	7.86	5.32	19.62	59.56
1126	1.198	46.12	23.53	2.20	5.50	7.48	5.60	19.01	60.21
1138	1.206	42.07	27.99	2.12	5.38	7.48	6.11	19.61	59.29
1150	1.214	43.36	26.68	1.87	5.26	6.49	5.17	20.08	61.13
1162	1.222	39.69	31.03	1.91	5.19	6.22	5.69	20.43	60.55
1174	1.230	39.83	30.82	1.82	5.23	6.24	4.53	20.73	61.46
1185	1.240	38.38	32.33	1.70	4.92	6.01	4.62	19.30	63.45
1197	1.248	41.23	28.76	1.55	4.99	6.59	5.24	17.57	64.06

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
		(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
1209	1.256	41.56	28.30	1.49	5.00	6.82	5.42	17.48	63.79
1221	1.264	44.16	25.79	1.71	4.97	6.81	5.03	17.57	63.92
1233	1.271	45.77	24.33	1.57	5.10	7.08	5.06	17.22	63.96
1244	1.276	45.22	25.10	1.90	5.00	6.99	5.05	16.78	64.28
1256	1.283	46.63	24.05	1.98	4.99	7.11	4.80	16.71	64.41
1268	1.291	44.89	25.65	1.94	5.16	7.51	4.67	17.22	63.50
1280	1.297	48.44	21.57	2.28	5.11	7.21	4.29	18.35	62.77
1292	1.303	47.23	22.47	2.38	5.06	7.27	3.56	18.60	63.13
1304	1.309	49.23	20.66	2.14	5.02	7.05	4.60	18.30	62.88
1316	1.314	49.95	20.04	2.28	4.85	7.02	4.56	18.62	62.65
1327	1.319	45.94	24.22	2.40	4.93	6.93	4.98	18.57	62.19
1339	1.326	43.24	26.79	2.50	4.96	7.09	5.00	19.87	60.58
1351	1.335	43.84	26.32	2.79	4.89	7.08	4.65	20.48	60.11
1363	1.341	44.88	25.00	2.80	4.95	7.20	4.42	20.41	60.21
1375	1.348	44.17	25.86	2.57	4.92	7.67	5.32	19.73	59.79
1387	1.356	45.91	24.66	2.81	4.93	8.15	3.95	19.95	60.22
1399	1.363	49.74	21.14	2.87	4.91	8.68	4.23	19.21	60.10
1411	1.367	50.09	20.71	2.68	4.75	8.58	3.59	19.74	60.65
1423	1.374	47.62	22.49	2.49	4.71	7.75	3.88	19.30	61.87
1435	1.380	43.37	26.51	2.37	4.76	8.02	4.16	19.58	61.11
1447	1.387	44.37	25.39	2.30	4.82	7.82	4.09	20.05	60.92
1459	1.395	43.63	26.03	2.70	4.92	7.17	2.85	20.66	61.71
1470	1.401	50.08	20.61	2.84	4.95	7.06	3.86	20.83	60.47
1482	1.406	54.85	16.88	2.89	4.87	6.97	3.55	21.55	60.18
1494	1.410	54.26	17.43	3.13	4.93	6.40	4.84	20.77	59.93
1506	1.415	47.97	22.66	3.24	4.85	6.64	4.78	20.46	60.02
1518	1.423	46.58	23.55	3.39	4.83	6.49	4.19	19.88	61.23
1530	1.429	46.05	24.08	3.74	4.71	6.30	4.37	18.79	62.08
1542	1.435	50.22	20.20	3.90	4.64	6.36	5.10	18.29	61.71
1554	1.441	52.50	18.81	3.87	4.79	6.70	3.92	19.02	61.69
1566	1.445	57.26	15.04	3.88	4.81	6.45	4.27	18.73	61.86
1578	1.449	57.50	14.88	3.67	4.74	7.12	2.68	19.91	61.89
1590	1.453	53.18	17.84	3.69	4.78	7.13	2.30	20.08	62.02
1602	1.459	48.07	21.98	3.74	4.71	6.95	2.79	20.20	61.61
1614	1.466	45.80	23.78	3.50	4.56	7.05	3.12	21.44	60.32
1626	1.472	45.66	23.90	3.30	4.85	7.57	2.89	22.05	59.35
1638	1.478	47.55	22.06	3.30	4.69	7.18	3.36	21.08	60.39

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
		(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
1650	1.484	54.47	17.71	3.28	4.72	7.47	3.67	19.83	61.05
1662	1.488	55.41	16.92	3.44	4.84	7.76	3.67	17.97	62.33
1674	1.492	59.26	14.25	3.48	4.79	7.75	4.54	17.82	61.61
1687	1.497	57.73	14.96	3.42	4.86	8.02	3.95	17.47	62.28
1699	1.501	57.24	15.37	3.33	4.85	8.27	4.39	17.16	62.00
1711	1.505	56.48	15.81	3.35	4.45	8.14	4.40	17.61	62.05
1723	1.510	51.42	19.15	3.29	4.41	8.34	4.89	17.62	61.46
1735	1.515	51.41	19.17	3.51	4.38	8.38	4.84	17.40	61.49
1747	1.520	49.19	20.69	3.56	4.25	8.30	6.05	18.62	59.22
1759	1.526	54.27	17.33	3.43	4.34	8.40	5.96	16.99	60.89
1771	1.530	54.94	16.79	3.48	4.26	8.11	6.64	17.51	60.00
1783	1.534	59.22	14.14	3.53	4.26	7.50	6.09	18.62	60.00
1795	1.538	55.28	16.57	3.49	4.26	7.29	6.16	18.75	60.06
1808	1.543	53.31	18.30	3.52	4.26	7.02	6.00	18.64	60.56
1820	1.548	48.96	21.00	3.47	4.28	7.07	5.80	20.59	58.79
1832	1.554	49.24	20.79	3.56	4.29	6.80	5.09	20.94	59.33
1844	1.559	51.86	18.58	3.75	4.20	6.79	5.00	22.64	57.63
1857	1.564	52.63	18.01	3.95	4.18	6.61	5.42	22.70	57.14
1869	1.569	52.23	18.30	4.10	4.17	6.85	5.90	21.78	57.19
1881	1.574	52.05	18.43	4.22	4.33	7.24	6.64	22.65	54.91
1893	1.579	51.78	18.63	4.40	4.29	7.29	6.61	22.59	54.82
1905	1.584	53.96	17.14	4.29	4.20	7.13	7.65	21.99	54.74
1917	1.588	54.10	17.04	4.17	4.13	7.35	8.83	21.56	53.96
1929	1.593	53.96	17.14	4.24	4.16	7.16	8.47	21.94	54.03
1942	1.598	51.81	18.61	4.10	4.18	6.87	8.20	21.89	54.76
1954	1.603	52.60	18.06	4.24	4.24	6.95	8.53	22.32	53.71
1966	1.608	53.56	17.38	4.10	4.11	6.79	8.38	21.15	55.47
1978	1.612	55.26	16.20	4.09	4.12	6.87	8.48	21.24	55.19
1990	1.617	55.31	16.17	3.85	4.05	6.75	11.92	19.16	54.27
2002	1.621	55.79	15.85	4.00	4.07	6.77	11.32	19.28	54.56
2015	1.626	57.35	14.94	4.14	4.04	6.94	11.89	18.90	54.09
2027	1.630	57.17	15.05	4.09	4.08	7.67	11.42	18.95	53.78
2039	1.633	57.63	14.77	4.15	4.06	8.15	12.14	16.20	55.29
2051	1.637	56.98	15.13	4.44	4.02	8.28	11.37	16.60	55.29
2064	1.642	61.56	12.68	4.33	3.88	8.36	10.51	16.20	56.72
2076	1.645	61.26	12.87	4.63	4.02	8.52	6.76	18.84	57.23
2088	1.648	58.86	14.45	4.63	4.11	8.87	6.86	16.95	58.58

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
		(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
2100	1.653	55.39	16.20	4.40	4.12	8.80	6.57	17.06	59.06
2112	1.657	59.84	13.94	4.46	4.05	9.03	6.55	17.27	58.64
2125	1.660	62.41	12.27	4.25	4.09	9.44	4.53	20.62	57.07
2137	1.663	62.20	12.39	4.09	4.20	9.39	4.54	21.22	56.56
2149	1.667	59.41	13.69	4.13	4.35	9.49	5.41	22.15	54.47
2162	1.671	60.17	13.27	4.07	4.31	9.65	4.93	21.83	55.20
2174	1.675	60.61	13.02	4.16	4.21	9.63	4.66	23.63	53.72
2186	1.679	60.05	13.32	4.11	4.25	10.61	4.54	23.82	52.66
2198	1.683	59.23	13.77	4.31	4.45	10.87	5.21	20.60	54.57
2211	1.687	60.97	12.84	4.03	4.35	10.41	5.84	19.95	55.42
2223	1.690	66.52	10.41	4.09	4.25	10.85	5.64	19.34	55.83
2235	1.693	66.87	10.22	4.03	4.26	10.89	4.96	19.05	56.79
2248	1.696	65.94	10.70	4.17	4.24	11.10	4.70	18.96	56.83
2260	1.699	61.79	12.39	4.03	4.23	10.85	5.02	19.12	56.75
2272	1.702	61.56	12.52	4.22	4.16	10.17	4.99	18.87	57.59
2285	1.706	61.47	12.57	4.04	3.96	10.05	5.54	21.52	54.89
2297	1.710	61.18	12.71	4.48	3.92	9.93	5.90	20.83	54.94
2309	1.713	61.91	12.31	4.50	3.97	9.77	6.73	20.76	54.28
2322	1.717	62.30	12.10	4.77	4.01	10.27	7.84	20.75	52.36
2334	1.720	62.72	11.90	4.79	4.06	10.20	8.18	20.89	51.88
2346	1.723	64.57	11.01	4.75	4.10	10.30	7.77	20.84	52.24
2359	1.726	65.69	10.46	4.70	4.11	10.63	7.49	20.73	52.34
2371	1.729	65.75	10.43	4.71	4.13	10.64	7.10	21.39	52.04
2383	1.732	64.91	10.81	4.75	4.19	10.23	7.58	21.85	51.40
2396	1.736	64.58	10.97	4.86	4.28	11.03	7.83	22.43	49.58
2408	1.739	64.87	10.83	4.07	4.28	11.12	8.13	22.24	50.16
2421	1.742	66.41	10.15	4.44	4.33	11.34	8.91	22.40	48.58
2433	1.745	67.97	9.45	4.62	4.36	11.76	8.43	22.44	48.40
2445	1.748	68.80	9.07	4.69	4.38	11.39	9.15	22.43	47.96
2458	1.751	68.38	9.25	4.81	4.40	11.22	8.90	22.38	48.28
2470	1.754	67.93	9.44	4.62	4.40	11.76	8.45	22.52	48.25
2483	1.757	67.79	9.50	5.02	4.47	11.04	8.71	18.89	51.86
2495	1.759	67.34	9.70	5.66	4.36	10.28	8.88	18.43	52.38
2507	1.762	72.05	8.00	5.47	4.31	9.96	8.83	17.74	53.69
2520	1.765	71.70	8.16	5.74	4.47	10.05	8.84	18.33	52.59
2532	1.767	72.62	7.76	5.63	4.42	10.23	9.04	18.15	52.53
2545	1.770	70.01	8.62	5.75	4.55	10.50	10.31	18.10	50.79

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO₂ and CO	Total Offgas Flow	H₂	NO₂	N₂	O₂	N₂O	NO
(sec)	(M)	(vol %)	(cm³/min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
2557	1.772	70.73	8.31	5.73	4.54	10.42	10.76	17.71	50.84
2570	1.775	72.15	7.75	4.98	4.36	9.95	10.64	20.41	49.66
2582	1.777	70.86	8.25	5.29	4.46	10.05	10.90	20.54	48.77
2595	1.780	71.56	7.96	5.19	4.44	10.64	10.91	20.10	48.72
2607	1.782	71.30	8.06	5.06	4.32	10.52	11.05	19.15	49.89
2620	1.785	71.46	7.99	5.15	4.41	10.45	10.54	20.19	49.26
2632	1.787	72.12	7.74	5.12	4.33	9.83	9.85	19.65	51.21
2645	1.790	71.40	8.02	5.27	4.45	10.61	8.51	19.95	51.21

Appendix C. Raman Offgas Data for Experiment 106

The fixed, normalized, and moving average Raman gas concentration measurements for Experiment 106 are reported in Table C-1. 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 C-1. Fixed Normalized Moving Average Raman Data for Experiment 106

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
0	0.001	79.13	5.017	0.23	0.20	1.14	0.26	0.00	98.17
11	0.002	80.79	5.012	0.14	0.18	1.02	0.12	0.00	98.54
23	0.003	79.98	5.150	0.34	0.17	0.79	0.00	0.00	98.70
35	0.004	77.86	5.239	0.54	0.21	1.16	0.00	0.44	97.66
46	0.005	79.92	5.355	0.81	0.18	0.45	0.00	3.27	95.28
58	0.006	78.88	5.601	1.29	0.14	0.45	0.00	6.71	91.41
69	0.007	75.69	9.939	1.92	0.14	0.73	0.00	10.36	86.86
81	0.010	52.58	20.882	2.47	0.11	1.13	0.00	14.06	82.23
93	0.015	34.37	39.028	3.20	0.11	1.23	0.00	18.10	77.36
104	0.024	24.73	79.696	3.68	0.10	1.78	0.00	21.77	72.66
116	0.041	12.50	122.403	4.30	0.04	1.81	0.00	25.31	68.54
128	0.065	10.74	149.021	4.88	0.04	1.91	0.00	26.30	66.87
139	0.089	12.44	159.199	5.16	0.06	2.41	0.00	26.72	65.65
151	0.116	10.49	158.272	5.22	0.07	2.57	0.00	26.92	65.21
163	0.145	10.90	160.051	5.49	0.09	2.55	0.00	27.03	64.85
174	0.170	12.04	155.636	5.53	0.10	2.92	0.00	26.68	64.77
186	0.199	11.28	151.342	5.47	0.11	2.78	0.00	26.75	64.89
198	0.228	11.72	151.320	5.48	0.11	2.80	0.00	26.28	65.32
209	0.252	12.05	142.544	5.46	0.12	2.94	0.05	25.77	65.66
221	0.277	13.24	131.959	5.46	0.12	2.71	0.05	25.74	65.91
233	0.296	14.40	122.623	5.44	0.12	2.62	0.21	25.10	66.51
244	0.309	14.49	120.147	5.37	0.12	2.65	0.21	25.00	66.65
256	0.327	13.93	119.932	5.26	0.11	2.54	0.21	24.93	66.95
268	0.346	14.47	119.658	5.38	0.11	2.47	0.27	24.84	66.94
280	0.364	14.58	115.175	5.17	0.11	2.40	0.27	24.37	67.69
291	0.384	15.37	113.604	4.93	0.10	2.47	0.22	24.78	67.51
303	0.405	14.98	116.582	4.92	0.10	2.49	0.22	24.53	67.75
315	0.420	13.69	115.626	4.93	0.09	2.55	0.06	24.97	67.41
327	0.434	15.72	111.366	4.87	0.08	2.55	0.12	24.84	67.53
338	0.453	16.56	102.958	4.87	0.09	2.61	0.12	24.86	67.45

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
350	0.472	16.55	98.384	4.78	0.08	2.59	0.07	24.79	67.70
362	0.490	17.62	99.320	4.88	0.08	2.69	0.07	25.39	66.89
374	0.508	16.19	99.779	5.05	0.07	2.76	0.07	25.19	66.86
385	0.526	16.36	100.075	4.88	0.07	2.88	0.07	25.37	66.74
397	0.544	17.48	96.229	4.96	0.07	2.98	0.07	24.82	67.11
409	0.561	17.85	96.369	4.93	0.07	3.07	0.05	24.76	67.13
421	0.579	16.30	94.637	4.94	0.06	2.97	0.05	24.81	67.17
433	0.593	18.31	93.728	5.00	0.07	3.16	0.05	24.68	67.04
444	0.606	18.30	90.082	4.92	0.06	3.19	0.05	24.60	67.17
456	0.623	17.90	90.218	4.77	0.06	3.16	0.05	24.62	67.34
468	0.639	18.24	89.817	4.92	0.06	3.28	0.05	24.43	67.26
480	0.656	18.50	88.137	4.78	0.06	3.28	0.05	24.82	67.00
492	0.672	18.75	87.844	4.79	0.06	3.22	0.00	24.83	67.09
503	0.687	18.39	84.553	4.77	0.06	3.25	0.00	24.74	67.17
515	0.703	20.36	84.657	4.69	0.06	3.23	0.00	24.88	67.15
527	0.718	18.69	83.582	4.80	0.06	3.07	0.00	24.40	67.67
539	0.734	18.95	85.377	4.82	0.06	3.10	0.00	24.43	67.58
551	0.750	19.31	82.173	4.69	0.06	3.00	0.00	24.57	67.68
563	0.763	20.53	80.211	4.56	0.07	3.00	0.00	24.63	67.75
575	0.775	20.07	79.021	4.43	0.06	3.02	0.04	24.61	67.84
586	0.789	19.99	80.413	4.37	0.07	3.10	0.04	24.58	67.84
598	0.804	19.69	79.488	4.35	0.07	3.01	0.04	24.58	67.95
610	0.818	20.65	78.350	4.18	0.06	3.15	0.04	25.18	67.39
622	0.833	20.70	78.306	4.12	0.07	3.15	0.04	25.10	67.53
634	0.847	19.72	78.853	4.10	0.06	3.27	0.04	24.91	67.61
646	0.862	20.30	78.250	4.17	0.06	3.26	0.04	24.86	67.61
658	0.876	21.10	75.054	4.12	0.06	3.19	0.00	25.10	67.53
670	0.890	21.77	74.748	4.14	0.06	3.18	0.00	24.95	67.66
682	0.904	20.50	74.488	4.09	0.06	3.33	0.00	24.23	68.29
694	0.919	21.27	74.611	4.00	0.06	3.35	0.00	24.25	68.34
706	0.933	21.69	72.896	4.05	0.06	3.48	0.00	24.36	68.05
717	0.943	21.64	72.522	3.96	0.06	3.40	0.03	24.49	68.06
729	0.954	21.53	71.931	3.92	0.06	3.52	0.11	24.61	67.77
741	0.968	22.11	70.413	3.95	0.06	3.74	0.11	24.49	67.65
753	0.981	22.76	70.643	3.89	0.06	3.86	0.11	24.55	67.52
765	0.994	21.37	69.997	3.89	0.06	3.94	0.29	25.30	66.52
777	1.007	22.60	69.257	3.99	0.07	4.03	0.29	25.21	66.41

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
789	1.020	23.35	67.960	3.92	0.07	4.01	0.39	25.22	66.39
801	1.032	22.29	67.227	4.08	0.07	4.07	0.36	25.47	65.95
813	1.045	23.17	67.077	4.12	0.06	3.97	0.27	25.54	66.03
825	1.057	23.47	64.116	4.20	0.07	3.91	0.27	25.54	66.00
837	1.069	24.75	63.754	4.14	0.07	3.87	0.27	25.73	65.92
849	1.080	23.47	63.146	4.22	0.07	3.95	0.09	25.77	65.90
861	1.093	23.98	63.533	4.29	0.06	3.92	0.09	25.91	65.73
873	1.104	24.39	62.024	4.40	0.07	4.10	0.00	26.15	65.29
885	1.116	24.78	60.098	4.41	0.07	4.25	0.00	25.92	65.34
897	1.128	25.77	59.928	4.39	0.07	4.32	0.00	25.97	65.25
909	1.139	24.55	58.411	4.45	0.07	4.47	0.00	25.97	65.05
921	1.150	26.27	57.989	4.50	0.07	4.75	0.00	26.09	64.59
933	1.160	26.20	55.827	4.46	0.07	4.84	0.03	26.15	64.44
945	1.171	26.67	56.799	4.55	0.07	5.00	0.03	26.20	64.15
957	1.181	25.30	54.980	4.54	0.07	4.95	0.03	26.14	64.27
969	1.192	28.21	54.322	4.56	0.08	4.96	0.03	26.10	64.28
981	1.202	27.39	52.237	4.65	0.08	5.13	0.03	26.10	64.02
993	1.212	27.47	51.964	4.65	0.08	5.23	0.03	26.27	63.74
1006	1.222	28.54	50.865	4.76	0.07	5.17	0.03	26.31	63.66
1018	1.232	28.69	50.060	4.87	0.07	5.10	0.00	26.43	63.53
1030	1.241	28.41	49.277	4.90	0.07	5.25	0.00	26.54	63.23
1042	1.250	29.53	49.049	4.99	0.07	5.41	0.00	26.23	63.30
1054	1.260	28.97	48.412	5.03	0.07	5.67	0.00	26.40	62.82
1066	1.269	29.21	47.995	5.13	0.08	5.73	0.00	26.54	62.52
1078	1.278	30.09	46.968	5.25	0.08	5.85	0.03	26.57	62.22
1090	1.286	30.32	46.176	5.27	0.08	5.95	0.03	26.50	62.17
1102	1.295	30.26	46.146	5.32	0.08	6.27	0.03	26.48	61.82
1114	1.304	30.13	46.302	5.33	0.08	6.34	0.03	26.30	61.92
1127	1.314	30.11	45.942	5.41	0.08	6.32	0.03	26.72	61.43
1139	1.323	30.76	44.849	5.50	0.07	6.43	0.03	26.54	61.43
1151	1.331	31.70	43.768	5.38	0.08	6.45	0.03	26.43	61.62
1163	1.339	31.66	41.875	5.40	0.08	6.51	0.00	26.52	61.49
1175	1.348	33.70	41.259	5.60	0.08	6.77	0.14	26.57	60.85
1187	1.355	32.65	41.106	5.63	0.08	6.85	0.14	26.52	60.78
1199	1.363	31.89	41.659	5.68	0.08	6.93	0.14	26.70	60.48
1212	1.372	32.78	40.697	5.67	0.08	7.18	0.14	26.66	60.28
1224	1.380	34.27	38.607	5.78	0.09	7.16	0.14	27.10	59.73

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
1236	1.387	35.43	37.306	6.00	0.08	7.62	0.14	27.04	59.12
1248	1.394	35.02	38.835	6.03	0.09	7.63	0.18	27.12	58.95
1260	1.402	31.77	38.878	6.04	0.09	7.65	0.13	27.18	58.92
1273	1.411	35.35	38.442	6.12	0.09	7.61	0.13	27.31	58.75
1285	1.418	35.85	35.850	6.20	0.09	7.82	0.13	27.23	58.53
1297	1.425	36.25	35.387	6.23	0.10	7.92	0.13	27.31	58.32
1309	1.431	36.24	34.831	6.27	0.10	8.09	0.13	27.25	58.18
1321	1.438	36.95	34.708	6.31	0.10	8.11	0.13	27.41	57.94
1334	1.445	36.49	34.220	6.32	0.10	8.38	0.80	26.99	57.41
1346	1.452	37.22	33.043	6.37	0.10	8.58	0.74	26.95	57.26
1358	1.458	39.53	33.152	6.54	0.10	8.83	0.74	27.13	56.66
1370	1.465	36.28	33.002	6.63	0.09	8.90	0.74	27.18	56.47
1383	1.472	37.54	33.611	6.82	0.10	9.01	0.74	27.07	56.26
1395	1.478	38.15	31.902	6.90	0.10	9.16	0.79	26.94	56.12
1407	1.484	40.00	31.339	7.04	0.10	9.24	0.79	26.93	55.91
1419	1.490	38.77	30.789	7.19	0.10	9.44	0.06	27.13	56.08
1432	1.497	39.39	30.877	7.23	0.10	9.66	0.05	27.23	55.73
1444	1.503	39.79	30.318	7.16	0.10	9.83	0.05	26.99	55.87
1456	1.509	40.07	29.924	7.24	0.10	9.95	0.05	26.92	55.74
1468	1.515	40.33	30.358	7.47	0.11	10.40	0.05	27.48	54.50
1481	1.521	38.78	30.011	7.48	0.11	10.51	0.00	27.46	54.44
1493	1.527	40.92	27.863	7.48	0.10	10.45	0.00	27.37	54.59
1505	1.533	46.35	26.974	7.66	0.10	10.74	0.00	27.96	53.54
1518	1.539	40.89	26.104	7.76	0.10	10.70	0.00	27.79	53.66
1530	1.544	43.24	26.694	7.97	0.11	10.75	0.00	28.66	52.51
1542	1.549	44.52	26.193	8.03	0.11	10.75	0.00	28.62	52.50
1555	1.554	42.19	26.076	7.90	0.10	10.34	0.00	28.01	53.65
1567	1.560	43.57	26.665	8.07	0.10	10.36	0.05	27.90	53.52
1579	1.566	42.84	26.092	8.09	0.10	10.57	0.05	27.90	53.29
1592	1.572	43.78	25.830	8.03	0.10	10.33	0.05	27.34	54.15
1604	1.577	44.32	25.523	8.00	0.10	10.40	0.05	27.34	54.11
1616	1.582	43.70	25.459	7.99	0.10	10.55	0.05	26.36	54.94
1629	1.588	43.97	25.378	7.95	0.10	10.62	0.05	26.32	54.95
1641	1.593	44.56	24.961	7.92	0.11	10.96	0.05	26.22	54.74
1653	1.598	44.93	24.855	7.91	0.11	11.08	0.00	26.07	54.83
1666	1.603	44.28	24.709	7.96	0.11	11.28	0.00	26.06	54.60
1678	1.608	45.00	24.651	8.01	0.11	11.48	0.00	26.01	54.39

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
		(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
1691	1.613	45.11	24.590	8.08	0.11	11.57	0.00	26.03	54.20
1703	1.618	44.46	24.492	7.94	0.11	11.58	0.19	26.02	54.16
1715	1.623	45.30	24.152	8.07	0.11	11.76	0.19	25.94	53.93
1728	1.628	46.17	23.199	8.19	0.11	11.88	0.23	25.82	53.78
1740	1.633	47.47	22.611	8.24	0.11	11.86	0.23	26.00	53.57
1753	1.638	47.19	22.344	8.18	0.11	11.97	0.25	25.93	53.56
1765	1.642	47.04	22.320	8.15	0.11	12.03	0.25	25.60	53.85
1777	1.646	47.55	22.248	8.25	0.12	12.28	0.29	25.54	53.53
1790	1.651	47.43	21.654	8.28	0.12	12.29	0.10	25.43	53.78
1802	1.656	49.10	21.314	8.17	0.13	12.50	0.10	25.27	53.83
1815	1.660	48.74	20.743	8.05	0.13	12.56	0.06	25.14	54.06
1827	1.664	49.43	20.618	8.01	0.13	13.02	0.06	24.91	53.86
1840	1.669	49.56	20.701	8.10	0.13	13.22	0.04	24.82	53.68
1852	1.673	48.44	20.772	8.08	0.14	13.63	0.04	24.92	53.19
1865	1.678	49.17	20.159	7.94	0.14	13.89	0.00	24.52	53.51
1877	1.682	51.92	19.531	8.04	0.14	14.21	0.00	24.52	53.10
1890	1.686	50.76	19.302	8.00	0.13	14.54	0.17	24.48	52.67
1902	1.690	50.02	19.708	8.01	0.14	14.92	0.22	24.56	52.16
1914	1.694	50.33	19.638	7.79	0.14	15.52	0.22	24.39	51.95
1927	1.698	51.03	18.857	7.68	0.14	15.85	0.22	24.18	51.94
1940	1.702	53.13	18.607	7.58	0.14	16.13	0.22	24.13	51.79
1952	1.706	51.30	18.069	7.69	0.14	16.59	0.22	24.09	51.27
1965	1.710	53.22	18.092	7.67	0.13	17.08	0.22	23.82	51.08
1977	1.714	53.04	17.278	7.66	0.14	17.53	0.05	23.60	51.01
1990	1.717	54.73	16.961	7.65	0.14	18.10	0.01	23.62	50.49
2002	1.721	54.60	16.675	7.82	0.14	18.08	0.01	23.52	50.43
2015	1.725	54.28	16.843	7.85	0.15	18.46	0.01	23.48	50.06
2027	1.728	53.98	16.831	8.05	0.15	18.82	0.00	23.17	49.80
2040	1.732	54.65	16.389	8.13	0.15	19.84	0.00	20.94	50.94
2052	1.735	56.30	16.206	7.99	0.15	20.05	0.00	20.99	50.81
2065	1.739	54.79	13.506	7.99	0.16	20.08	0.00	21.18	50.59
2077	1.742	70.19	13.470	7.94	0.15	20.30	0.00	21.15	50.45
2090	1.745	56.47	13.173	7.90	0.15	20.50	0.03	21.03	50.39
2103	1.748	56.16	15.281	7.93	0.15	20.72	0.03	20.90	50.27
2115	1.751	57.44	15.207	7.79	0.15	20.79	0.03	20.83	50.40
2128	1.755	56.83	15.008	7.76	0.15	20.15	0.03	23.21	48.71
2140	1.758	57.12	14.940	7.87	0.15	20.36	0.03	23.33	48.26

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
		(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
2153	1.761	57.78	14.711	7.98	0.15	20.92	0.03	23.20	47.72
2165	1.763	57.96	14.291	7.92	0.15	21.00	0.03	22.96	47.94
2178	1.767	59.25	13.619	7.92	0.16	21.21	0.00	22.82	47.90
2236	1.780	61.35	13.088	7.96	0.16	21.66	0.00	22.97	47.23
2248	1.783	60.77	12.664	7.91	0.16	22.23	0.00	23.08	46.62
2260	1.786	61.57	12.530	7.82	0.16	22.69	0.00	22.85	46.48
2272	1.788	62.12	12.585	7.95	0.16	23.09	0.00	22.52	46.29
2283	1.791	60.47	12.439	7.95	0.16	23.50	0.00	22.50	45.89
2294	1.793	62.41	12.512	8.15	0.16	23.99	0.00	22.38	45.32
2306	1.796	61.70	11.955	8.16	0.16	24.61	0.00	22.36	44.71
2317	1.798	63.68	11.900	8.15	0.16	24.80	0.00	22.16	44.73
2328	1.800	62.73	11.596	7.29	0.17	25.10	0.00	22.65	44.79
2339	1.803	63.49	11.715	7.34	0.17	24.78	0.20	22.53	44.98
2351	1.805	62.97	11.201	7.27	0.17	25.07	0.20	22.60	44.69
2362	1.807	65.92	11.302	7.26	0.17	25.00	0.20	22.42	44.95
2373	1.810	62.88	11.157	7.06	0.18	24.92	0.20	22.50	45.15
2384	1.812	63.84	11.281	7.09	0.19	25.03	0.20	22.28	45.22
2396	1.814	65.13	10.955	6.94	0.19	24.77	0.20	22.31	45.60
2407	1.816	64.87	10.562	7.89	0.18	24.67	0.20	21.61	45.45
2418	1.819	66.34	10.580	7.92	0.19	25.38	0.00	21.66	44.85
2430	1.821	65.01	10.461	7.79	0.20	25.62	0.01	21.55	44.83
2441	1.823	65.63	10.401	7.80	0.19	25.88	0.20	21.39	44.53
2453	1.825	66.74	10.210	7.92	0.19	25.95	0.48	21.12	44.32
2464	1.827	66.25	10.346	7.96	0.18	26.20	0.48	21.16	44.01
2475	1.829	64.77	10.567	8.00	0.18	26.76	0.48	21.24	43.34
2487	1.831	65.29	10.298	7.91	0.18	26.53	0.48	21.32	43.57
2498	1.833	68.07	9.934	7.77	0.18	26.62	0.48	21.26	43.69
2509	1.835	67.14	9.754	7.83	0.17	26.63	0.48	21.19	43.70
2521	1.837	66.47	9.955	7.77	0.17	26.34	0.28	21.18	44.25
2532	1.839	66.70	9.515	7.85	0.17	26.90	0.00	21.42	43.65
2543	1.841	70.25	9.503	7.64	0.17	26.66	0.00	21.33	44.20
2555	1.843	66.55	9.419	7.70	0.18	26.72	0.00	21.35	44.05
2566	1.845	67.26	9.780	7.73	0.18	27.17	0.00	21.27	43.66
2577	1.847	67.67	9.805	7.69	0.18	27.14	0.11	21.38	43.51
2589	1.850	66.38	9.805	7.60	0.18	27.17	0.11	21.35	43.59
2600	1.852	67.27	9.710	7.60	0.18	27.61	0.14	21.51	42.96
2612	1.854	68.33	9.395	7.51	0.19	27.29	0.14	21.31	43.57

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
2623	1.856	68.53	9.347	7.55	0.19	27.62	0.14	21.29	43.21
2634	1.858	67.60	9.243	7.44	0.19	28.00	0.14	20.71	43.53
2646	1.859	69.07	9.205	7.31	0.19	28.32	0.14	20.61	43.43
2657	1.861	68.80	9.178	7.41	0.19	28.38	0.47	20.35	43.20
2669	1.863	67.78	9.080	7.52	0.18	28.56	0.47	20.07	43.18
2680	1.865	69.77	9.141	7.45	0.18	28.66	0.44	19.98	43.28
2691	1.866	68.37	8.844	7.54	0.18	29.15	0.44	19.90	42.79
2703	1.868	69.89	8.965	7.17	0.18	27.78	0.44	23.71	40.71
2714	1.870	68.91	8.676	7.35	0.17	27.61	0.48	23.84	40.55
2726	1.872	70.45	9.940	7.55	0.17	27.75	0.48	23.65	40.40
2737	1.874	61.71	9.770	7.65	0.18	27.97	0.04	23.53	40.63
2749	1.876	70.14	9.729	7.63	0.18	28.03	0.04	23.81	40.32
2760	1.877	70.76	8.270	7.60	0.19	28.45	0.04	23.62	40.10
2772	1.879	71.35	8.261	7.34	0.19	28.48	0.34	23.68	39.97
2783	1.881	70.20	8.143	7.69	0.20	29.88	0.34	19.42	42.48
2795	1.883	71.66	8.274	7.52	0.21	29.98	0.30	19.25	42.74
2806	1.884	70.36	8.200	7.50	0.21	29.99	0.30	19.59	42.41
2818	1.886	70.75	8.097	7.39	0.21	30.03	0.30	19.52	42.56
2829	1.888	72.47	7.901	7.38	0.21	30.47	0.30	19.65	41.98
2841	1.889	71.85	7.647	7.40	0.21	30.15	0.67	19.65	41.93
2852	1.891	72.71	7.535	7.56	0.21	30.50	0.37	19.56	41.81
2864	1.893	73.36	7.657	7.32	0.22	30.59	0.37	20.17	41.34
2875	1.894	70.91	7.718	7.26	0.21	31.91	0.37	20.14	40.12
2887	1.896	72.23	7.801	7.27	0.20	32.07	0.37	19.84	40.25
2898	1.897	72.70	7.807	7.37	0.20	32.21	0.37	19.79	40.05
2910	1.899	70.87	7.740	7.36	0.22	32.07	0.43	19.41	40.51
2921	1.901	72.75	7.640	7.46	0.22	32.07	0.28	19.28	40.70
2933	1.902	73.51	7.116	6.36	0.23	32.51	0.45	19.03	41.41
2944	1.904	75.05	7.188	6.52	0.23	32.53	0.45	18.72	41.55
2956	1.905	72.18	6.789	6.84	0.23	31.32	0.45	18.85	42.32
2968	1.907	76.89	6.833	6.97	0.24	31.55	0.57	18.46	42.22
2979	1.908	74.68	6.529	6.80	0.24	32.39	0.57	17.94	42.06
2991	1.910	74.64	6.832	6.85	0.22	32.03	0.95	18.09	41.86
3002	1.911	74.30	6.971	6.59	0.23	32.53	0.73	18.24	41.69
3014	1.913	73.53	7.199	7.59	0.23	31.84	0.56	18.55	41.22
3026	1.914	72.78	7.178	7.74	0.23	31.98	1.24	18.27	40.54
3037	1.916	74.48	7.007	7.43	0.23	32.23	1.24	18.19	40.68

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
3049	1.918	74.95	6.768	7.25	0.23	31.99	1.12	18.47	40.94
3060	1.919	74.72	6.444	7.37	0.23	31.46	1.12	19.13	40.69
3072	1.920	77.28	6.409	7.36	0.23	32.04	0.68	19.11	40.58
3084	1.922	75.24	6.337	7.76	0.23	31.95	0.68	18.69	40.69
3095	1.923	75.33	6.486	7.71	0.23	32.45	0.68	18.24	40.69
3107	1.924	75.97	6.393	7.58	0.24	32.22	0.00	18.86	41.09
3118	1.926	76.05	6.368	7.49	0.24	32.38	0.00	18.65	41.24
3130	1.927	75.54	6.446	7.54	0.25	32.77	0.00	18.76	40.68
3142	1.929	75.30	6.356	7.57	0.25	32.71	0.00	18.40	41.06
3153	1.930	76.83	6.218	7.50	0.26	33.01	0.00	18.16	41.07
3165	1.932	76.73	6.125	7.34	0.25	33.30	0.00	17.83	41.28
3177	1.933	76.10	6.106	6.92	0.24	33.54	0.00	17.94	41.36
3188	1.934	77.01	6.059	7.01	0.24	34.17	0.00	17.09	41.49
3200	1.936	77.15	5.769	7.35	0.23	34.24	0.00	17.07	41.11
3212	1.937	78.71	5.590	7.26	0.22	33.95	0.00	16.40	42.16
3223	1.938	78.63	5.549	7.38	0.19	35.75	0.00	17.49	39.18
3235	1.940	77.52	5.584	7.21	0.19	36.26	0.00	16.96	39.39
3247	1.941	78.39	5.216	7.02	0.20	36.19	0.00	17.21	39.38
3259	1.942	82.19	5.226	7.58	0.21	35.86	0.00	17.80	38.55
3270	1.943	77.43	5.187	7.83	0.22	36.26	0.00	18.68	37.00
3282	1.945	78.75	5.423	7.75	0.23	36.32	0.00	18.77	36.93
3294	1.946	79.86	5.064	7.66	0.24	36.86	0.00	18.76	36.49
3305	1.947	80.80	5.021	7.47	0.27	35.27	0.32	17.58	39.08
3317	1.948	79.14	4.958	7.74	0.27	34.60	0.32	17.50	39.57
3329	1.949	80.48	5.147	8.07	0.32	34.81	0.32	17.52	38.95
3341	1.951	78.99	5.252	8.31	0.33	36.20	0.32	16.15	38.69
3353	1.952	78.17	5.203	7.38	0.30	33.17	0.41	14.16	44.58
3364	1.953	80.96	4.961	7.47	0.29	32.93	0.62	13.82	44.86
3376	1.954	81.33	6.728	7.49	0.32	32.94	0.94	14.09	44.22
3388	1.955	64.75	6.849	7.46	0.33	32.82	0.62	13.88	44.89
3400	1.957	79.79	6.982	7.31	0.33	33.10	0.62	14.17	44.47
3411	1.958	80.03	4.958	6.96	0.28	32.93	0.62	13.97	45.24
3423	1.959	80.59	4.857	7.24	0.27	31.92	0.62	14.56	45.40
3435	1.960	80.77	4.840	7.95	0.30	35.08	0.53	16.27	39.88
3447	1.961	80.19	4.727	7.64	0.30	35.70	0.32	16.34	39.71
3459	1.962	81.70	4.763	7.82	0.28	35.80	0.00	16.56	39.55
3470	1.963	80.43	4.643	7.92	0.27	35.73	0.00	16.41	39.67

Cumulative Reaction Time	Est. Al Conc.	Tracer	Offgas Flow and Concentrations without tracers and water						
		CO ₂ and CO	Total Offgas Flow	H ₂	NO ₂	N ₂	O ₂	N ₂ O	NO
(sec)	(M)	(vol %)	(cm ³ /min)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)	(vol %)
3482	1.964	81.37	4.560	8.02	0.26	35.84	0.00	16.54	39.33
3494	1.965	82.54	4.376	8.32	0.27	36.12	0.00	16.88	38.41
3506	1.966	82.25	4.446	7.67	0.27	35.50	0.00	16.73	39.83
3518	1.968	80.68	4.507	7.75	0.27	35.15	0.00	16.14	40.69
3530	1.969	81.92	4.595	7.81	0.28	35.56	0.00	15.72	40.64
3541	1.970	81.36	4.424	7.84	0.27	35.31	0.00	15.34	41.24
3553	1.971	82.39	4.337	7.80	0.26	35.43	0.24	15.26	41.00
3565	1.972	82.80	4.105	7.62	0.29	35.56	0.24	14.55	41.74
3577	1.973	83.73	4.079	7.86	0.29	36.81	0.24	14.37	40.43
3589	1.974	82.66	4.051	7.64	0.29	37.78	0.24	13.71	40.35
3601	1.975	83.09	3.774	7.66	0.29	38.18	0.24	13.87	39.75
3613	1.976	86.74	3.675	7.83	0.30	37.73	0.24	13.96	39.94
3625	1.977	83.68	3.589	7.73	0.32	38.52	0.24	12.89	40.29
3637	1.978	84.00	3.795	7.40	0.34	39.80	0.00	12.68	39.77
3649	1.979	84.47	3.598	7.69	0.34	40.76	0.53	13.57	37.11
3660	1.980	85.81	3.378	7.28	0.35	40.17	0.53	13.06	38.62
3672	1.980	86.40	3.219	7.66	0.36	39.98	0.58	12.85	38.57
3684	1.981	86.21	3.279	7.40	0.37	40.20	0.58	12.31	39.15
3696	1.982	85.15	3.515	7.58	0.37	41.24	0.58	12.37	37.85
3708	1.983	83.83	3.709	7.58	0.37	41.34	0.58	13.51	36.63
3720	1.984	84.10	3.616	7.61	0.35	38.83	0.58	17.32	35.31
3732	1.985	86.17	3.411	7.37	0.35	38.47	0.05	16.60	37.17
3744	1.986	86.06	3.629	7.31	0.35	38.44	0.05	16.69	37.16
3756	1.987	81.85	3.617	7.66	0.34	38.28	0.00	17.03	36.69
3768	1.987	86.30	3.605	7.94	0.36	38.94	0.00	17.10	35.66
3780	1.988	86.19	3.311	7.58	0.36	38.80	0.00	16.08	37.17
3792	1.989	84.91	3.606	7.45	0.38	38.95	0.00	15.28	37.94
3804	1.990	83.13	3.501	7.26	0.41	41.31	0.00	11.98	39.04
3816	1.991	87.37	3.213	7.44	0.43	42.54	0.00	11.02	38.57
3828	1.992	88.14	2.846	7.76	0.44	42.19	0.00	10.81	38.80
3840	1.993	87.12	2.767	7.41	0.44	43.10	0.00	10.04	39.01
3852	1.994	88.29	2.907	7.49	0.44	42.28	0.00	9.22	40.57
3864	1.995	86.54	2.869	7.42	0.38	41.54	0.28	9.31	41.06
3876	1.995	87.55	2.886	7.64	0.38	41.02	0.28	8.67	42.01
3888	1.996	88.09	2.782	8.27	0.37	41.58	0.28	7.77	41.72

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