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MEASUREMENT OF AMMONIA RELEASE FROM SALTSTONE

J.R. Zamecnik
A.D. Cozzi

February, 2009

Savannah River National Laboratory
Savannah River Nuclear Solutions
Aiken, SC 29808

Prepared for the U.S. Department of Energy under Contract No.
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TABLE OF CONTENTS

LIST OF FIGURES	iii
LIST OF TABLES	iii
ACRONYMS & ABBREVIATIONS	iv
1.0 EXECUTIVE SUMMARY	1
2.0 INTRODUCTION	1
3.0 EXPERIMENTAL.....	2
3.1 Compositions and Sample Preparation.....	2
3.2 Ammonia Sampling.....	6
3.3 Test Matrix.....	8
3.4 Equilibrium Concentrations.....	11
4.0 RESULTS AND DISCUSSION	15
4.1 Analytical Results – Ammonia Recovery	15
4.2 Experimental Results & Discussion.....	16
4.2.1 Mass Losses from Vessels During Tests	16
4.2.2 Ammonia Release Data	17
4.2.3 Data Trends.....	21
4.2.4 Estimation of Chemically Generated Ammonia.....	23
5.0 CONCLUSIONS.....	27
6.0 REFERENCES	28
7.0 APPENDICES.....	29
7.1 Appendix A: Vessel Mass & Volume Data.....	29
7.2 Appendix B: Original Ammonia Generation Data (mg NH₃)	31

LIST OF FIGURES

Figure 3-1. Test Vessels4
 Figure 3-2. Hot Block Apparatus with Sample Vessels5
 Figure 3-3. Hot Blocks in Hood5
 Figure 3-4. Steps Involved in Sampling Equilibrium and Total Vessels8
 Figure 3-5. Henry’s Law Constants in m/atm for PNNL & MCU Compositions.....12
 Figure 3-6. Dimensionless Henry’s Law Constants (g/L/g/L) for PNNL & MCU
 Compositions13
 Figure 3-7. Increase in Dimensionless Henry’s Law Constant for MCU Simulant Due
 to Increase in Ionic Strength14
 Figure 4-1. E-Tests Equilibrium Headspace Ammonia22
 Figure 4-2. T(H) Tests Equilibrium Headspace Ammonia22
 Figure 4-3. Cumulative Ammonia Collected in the Total Tests23
 Figure 4-4. Hypothetical Saltstone Ammonia Concentrations Due to Chemical
 Generation.....24

LIST OF TABLES

Table 3-1. Composition and Properties of MCU Salt Simulant.....2
 Table 3-2. Premix Blend3
 Table 3-3. Representative Data Sheet of the Mixes3
 Table 3-4. Sample Designations7
 Table 3-5. Test Vessel Matrix9
 Table 3-6. Test Vessel Matrix & Samples10
 Table 3-7. Composition and Properties of PNNL & MCU Salt Simulants12
 Table 3-8. Calculated Equilibrium Ammonia in Headspace at 95 °C14
 Table 4-1. Recovery of Silica Gel Sampling Tubes.....15
 Table 4-2. Measured Concentrations of Standard Solutions.....16
 Table 4-3. Mass Losses from Vessels.....17
 Table 4-4. Ratio of Measured to Equilibrium Ammonia Concentration and Fraction
 Initial Ammonia Collected18
 Table 4-5. Total Ammonia Released/Added.....19
 Table 4-6. Comparison of Total Test Equilibrium H Results to Equilibrium Test E
 Results.....20
 Table 4-7. Estimation of Ammonia Chemically Generated.....26
 Table 7-1. Saltstone Sample Masses & Volumes29
 Table 7-2. Liquid Sample Masses & Volumes30

ACRONYMS & ABBREVIATIONS

BQL	Below quantification limit
C	Cumulative ammonia collected on the sample tube installed between measurements (mg)
CBO	Carbon burn out
CLFL	Composite lower flammability limit
DSA	Documented Safety Analysis
E	Equilibrium, or equilibrium amount of ammonia (mg)
E(M/E)	Equilibrium test: (ammonia measured in headspace)/(equilibrium ammonia) (mg/mg)
H	Henry's law constant (units vary) or Headspace, or headspace equilibrium amount of ammonia (mg)
H(M/E)	Total test: (ammonia measured in headspace)/(equilibrium ammonia) (mg/mg)
ISE	Ion selective electrode
L	Liter
M	Molar
m	molal
MCU	Modular Caustic Side Solvent Extraction Unit
PNNL	Pacific Northwest National Laboratory
SDF	Saltstone Disposal Facility
SFT	Salt Feed Tank
SPF	Salt Processing Facility
SRNL	Savannah River National Laboratory
T	Total, or total amount of ammonia collected on the C & H sample tubes
T/E	Total test: (total ammonia released)/(equilibrium ammonia) (mg/mg)
T/I	Total test: (total ammonia released)/(initial ammonia in liquid or saltstone) (mg/mg)
T/I _a	Total test: (total ammonia released)/(initial ammonia in liquid or saltstone adjusted for generation) (mg/mg)

1.0 EXECUTIVE SUMMARY

SRNL was requested by WSRC Waste Solidification Engineering to characterize the release of ammonia from saltstone curing at 95 °C by performing experimental testing. These tests were performed with an MCU-type Tank 50H salt simulant containing 0, 50, and 200 mg/L ammonia. The testing program showed that above saltstone made from the 200 mg/L ammonia simulant, the vapor space ammonia concentration was about 2.7 mg/L vapor at 95 °C. An upper 95% confidence value for this concentration was found to be 3.9 mg/L.

Testing also showed that ammonia was chemically generated from curing saltstone at 95 °C; the amount of ammonia generated was estimated to be equivalent to 121 mg/L additional ammonia in the salt solution feed. Even with chemical generation, the ammonia release from saltstone was found to be lower than its release from salt solution only with 200 mg/L ammonia.

2.0 INTRODUCTION

Tank 50H is the feed tank for low level salt waste to the Saltstone Production Facility (SPF). Salt solution from the SPF Salt Feed Tank (SFT) is mixed with the pre-blended premix materials cement, ground-granulated blast furnace slag, and Class F fly ash in a grout mixer. The resulting saltstone slurry is then pumped to the Saltstone Disposal Facility (SDF), or vault.

The Saltstone Documented Safety Analysis¹ (DSA) is under revision to accommodate changes in the SDF composite lower flammability limit (CLFL) due to the introduction of Isopar[®] L from the Modular CSSX Unit (MCU) facility into Tank 50H.

Washington Savannah River Company (WSRC) Waste Solidification Engineering requested that the Savannah River National Laboratory (SRNL) perform testing to characterize the release of ammonia from saltstone curing at 95 °C.² The test temperature is the maximum temperature that may be achieved in the SDF.³ Testing was conducted per an approved plan.⁴

Ammonia may be introduced into the SDF from several sources. The premix materials slag and fly ash may contain ammonium salts, depending on the source of these materials.⁵ The Tank 50H feed may contain ammonia; specifically, SRNL was requested to test ammonia concentrations of 50 and 200 mg/L in Tank 50H simulant. Ammonia may also be chemically generated within the curing saltstone. Previous qualitative tests have demonstrated ammonia generation from saltstone and from simulant/slag mixtures.⁶ Reduction of nitrite or nitrate by reducing slag components such as Fe⁺² and sulfur are suspected to be the cause of ammonia generation.

A preliminary report giving the recommended ammonia release rate for CLFL calculations has been previously written.⁷

3.0 EXPERIMENTAL

3.1 COMPOSITIONS AND SAMPLE PREPARATION

Saltstone samples were prepared using an MCU-type (Modular CSSX Unit) salt solution spiked with 0, 50 and 200 mg/L ammonia as ammonium chloride. Table 3-1 shows the composition and physical properties of the salt solution used. All chemicals shown in Table 3-1 were reagent grade. The premix materials were cement and ground-granulated blast furnace slag from Holcim, who are the supplier for the SPF. The fly ash was from the SEFA Carbon Burn Out (CBO) process at the SCE&G Wateree power station. This fly ash was used in this work because it, or SEFA CBO fly ash from the Winyah plant, were chosen to be the type used for future SPF operations because of their low ammonia content.

Table 3-2 shows the premix blend used in this study. The ammonia contribution from the premix materials was determined by Eibling to be <2, 10.1, and <2 mg/kg for the CBO fly ash, blast furnace slag, and cement, respectively.⁸ The ammonia content of the slag would introduce about 0.32 mg of ammonia to each saltstone sample tested. The admixtures Daratard 17 (set retarder) and Clear Air 100 (antifoam) were added using the dosages representing recent SPF operations. No ammonia contribution is expected from the admixtures. However, the admixtures were included in the unlikely event that they participate in the reactions that generate ammonia. Table 3-3 is a data sheet representative of the three mixes prepared in this study.

Table 3-1. Composition and Properties of MCU Salt Simulant

Species	Concentration (M, mol/L)
Na ⁺	5.61
OH ⁻	1.37
NO ₃ ⁻	3.31
NO ₂ ⁻	0.37
CO ₃ ⁻²	0.18
SO ₄ ⁻²	0.059
Al ⁺³	0.054
PO ₄ ⁻³	0.012
Weight Percent Solids (Water)	31.5 (68.5)
Density (g/cm ³)	1.26

Table 3-2. Premix Blend

Material	Weight Percent
cement	10
blast furnace slag	45
fly ash (CBO)	45

Table 3-3. Representative Data Sheet of the Mixes

Saltstone Mix Data Sheet			
MIX # 0100	Date: 9/4/2008		
Material	%	WT%	Grams
Waste Solution: <u>MCU simulant</u> Wt% Solids # <u>31.51</u> Grams Water <u>1198.58</u>		46.62	1750.00
Admixture: <u>Daratard 17</u>		0.08	1.60
Admixture: <u>Clear Air 100</u>		0.11	2.20
Admixture: _____			
Premix		53.28	2000.00
Cement (% of Premix)	10	5.33	200.00
Slag (% of Premix)	45	23.98	900.00
Fly Ash (% of Premix)	45	23.98	900.00
Total	100	100.1	3753.80
Water to Premix Ratio	0.60		

Saltstone grouts at the three initial ammonia concentrations were made from the salt solution and premix that were combined and mixed for three minutes with a Rushton blade mixer. Approximately 83 mL of saltstone slurry was then poured into a glass vessel and placed into a Hot Block heating apparatus and heated to 95 °C. A sketch and photo of a test vessel are shown in Figure 3-1. Either a sampling tube or a fibrous filter was placed into the outlet port of the vessel. An air purge could be supplied via the addition port. A Hot Block apparatus (Environmental Express, Mt. Pleasant, SC) filled with vessels is shown in Figure 3-2. Three Hot Blocks were used for the tests. Each was insulated with 1 to 2” of Kaowool M Board (Thermal Ceramics) insulation on the sides and top. The

sample in the vessel was kept at 95 °C down in the Hot Block apparatus, but the headspace above the vessels could not be maintained at this temperature. The headspace temperature was measured in vessels prior to the tests and in one vessel during the tests; the headspace temperature varied from 80-90 °C just below the location of the sampling tube. Heaters for the headspace would have been desirable, but time constraints did not allow for heaters and controllers to be procured.

Vessels containing ~83 mL of salt solution with 200 mg/L ammonia and standard solutions of 200 mg/L ammonia adjusted to pH ~12 were also tested as controls. The 200 mg/L standards were made from ammonium chloride and the pH adjusted with 50 wt% NaOH. For each vessel, the initial empty mass with fittings, with saltstone immediately after filling, and the final mass at the end of the tests were recorded. The headspace in the saltstone vessels was measured by weighing the amount of water required to fill the vessels completely. The headspace in the standard and salt solution vessels was calculated from the total volume and the volume of liquid measured.

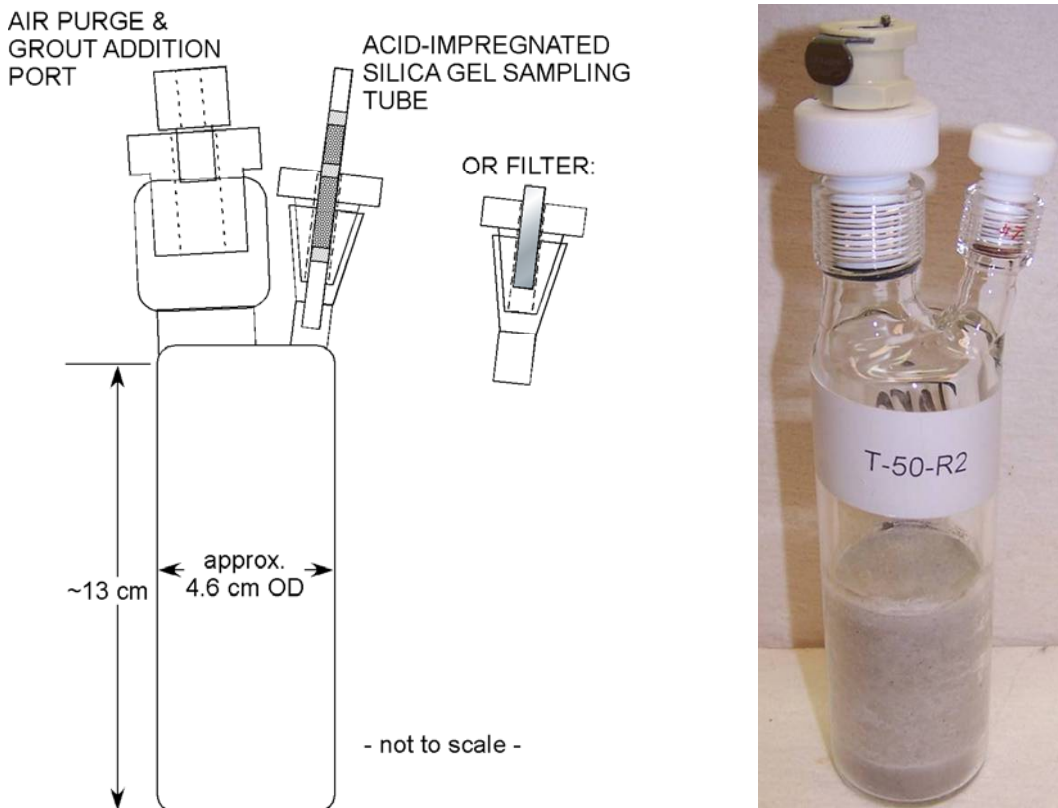


Figure 3-1. Test Vessels



Figure 3-2. Hot Block Apparatus with Sample Vessels



Figure 3-3. Hot Blocks in Hood

3.2 AMMONIA SAMPLING

Two different types of tests were performed. In one test type (called 'Equilibrium', or 'E'), the vessel headspace was open to the atmosphere via a 0.6-cm diameter opening that was plugged with a porous pipette filter. The amount of ammonia in the headspace was measured by placing a sulfuric acid impregnated silica gel tube (SKC Inc. #226-10-06) into the vent in place of the filter and then purging with 2-3 headspace volumes of air. The filter was then re-placed into the vent until the next measurement. These vessels were designated 'E'.

In the second type of test (called 'Total', or 'T') a silica gel tube was immediately placed into the vent and remained there until sampled; this tube was called 'Cumulative' or 'C' or was designated by the letter 'a'. This tube was called 'Cumulative' because it collected the accumulated ammonia evolved between sampling times.

Sampling of the Total vessels consisted of removing the Cumulative tube for analysis, placing a second tube into the vent and purging to sample the headspace as was done for the equilibrium vessels; this second tube was called the total test 'headspace' tube, or 'H', or was designated by the letter 'b'. After purging through the H tube, a new C tube was placed into the vent and remained there until the next T samples were taken. Table 3-4 summarizes the sample designations. (The designations 'a' and 'b' were initially used, but were later renamed to 'C' and 'H' for clarity; however, all lab data reports and data sheets use the former designations, so these are included here.) Figure 3-4 shows a diagram of the sampling process for the E and T vessels.

The silica gel from each sampling tube was transferred to a vial containing ~10 mL of 0.1 M sulfuric acid. The vials were submitted to F/H Lab for analysis of ammonia concentration using an ion selective electrode (ISE).⁹ The weight of dilution acid was recorded for each sample.

Table 3-4. Sample Designations

Sample Tube Description	Designations	Sample Name Example	Comments
Equilibrium Test	Equilibrium E	E-50-R1-F1	Equilibrium 50 mg/L salt solution used Replicate 1 Sampled First on day 1
Total Test (tube that remained in sample port)	Total T a, C	T-200-R3-4a or T-200-R3-4C	Total 200 mg/L salt solution used Replicate 3 Sampled on day 4 'Total' tube (a or C)
Total Test (tube purged to sample headspace)	Total T b, H	T-200-R2-7b or T-200-R2-7H	Total 200 mg/L salt solution used Replicate 2 Sampled on day 7 'Headspace' tube (b or H)

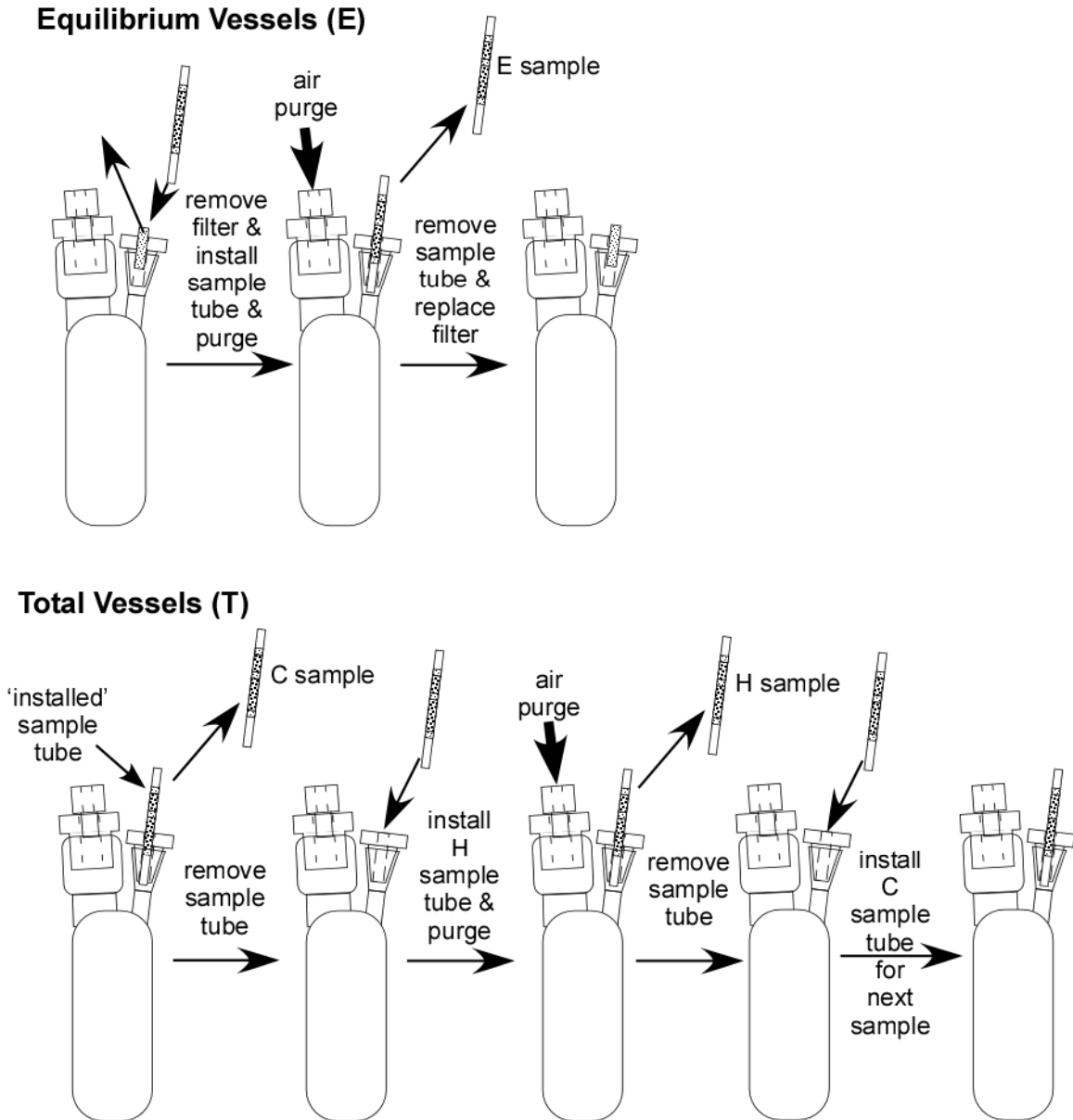


Figure 3-4. Steps Involved in Sampling Equilibrium and Total Vessels

3.3 TEST MATRIX

Replicate Equilibrium and Total saltstone samples were tested with 50 and 200 mg/L NH_3 salt solution. Only Equilibrium type samples were tested with saltstone containing no added ammonia (blanks). Equilibrium and Total tests of salt solution samples and standard solutions at 200 mg/L NH_3 were also tested. The matrix of samples tested is shown in Table 3-5. Samples were taken at various times during the tests. For some vessels, the first sample was taken after one day, whereas for others, the first sample was taken on a subsequent day. Repeated sampling of some vessels was also performed. The

SRNL-STI-2008-00399, Revision 0
SRNS-STI-2008-00120, Revision 0

First sample taken for E vessels (except for standards) was designed by the suffix –F#, where # is the day sampled (1 to 7). If sampled again, the sample had the additional suffix –S# to indicate its Subsequent sampling day. Standards were sampled on days 1, 3, 5, and 7. For T samples, all saltstone and salt solutions were sampled on days 1, 2, 4, and 7, while the standard vessels were sampled on days 1, 3, 5, and 7. The test vessel matrix with sampling days is given in Table 3-6.

Table 3-5. Test Vessel Matrix

Medium	Test Type	Ammonia (mg/L)	Test Designations						
			E-0-R1-F1	E-0-R1-F2	E-0-R1-F3	E-0-R1-F4	E-0-R1-F5	E-0-R1-F6	E-0-R1-F7
Saltstone	Equilibrium	0	E-0-R1-F1	E-0-R1-F2	E-0-R1-F3	E-0-R1-F4	E-0-R1-F5	E-0-R1-F6	E-0-R1-F7
Saltstone	Equilibrium	50	E-50-R1-F1	E-50-R1-F2	E-50-R1-F3	E-50-R1-F4	E-50-R1-F5	E-50-R1-F6	E-50-R1-F7
			E-50-R2-F1	E-50-R2-F2	E-50-R2-F3	E-50-R2-F4	E-50-R2-F5	E-50-R2-F6	E-50-R2-F7
			E-50-R3-F1	E-50-R3-F2	E-50-R3-F3	E-50-R3-F4	E-50-R3-F5	E-50-R3-F6	E-50-R3-F7
Saltstone	Equilibrium	200	E-200-R1-F1	E-200-R1-F2	E-200-R1-F3	E-200-R1-F4	E-200-R1-F5	E-200-R1-F6	E-200-R1-F7
			E-200-R2-F1	E-200-R2-F2	E-200-R2-F3	E-200-R2-F4	E-200-R2-F5	E-200-R2-F6	E-200-R2-F7
			E-200-R3-F1	E-200-R3-F2	E-200-R3-F3	E-200-R3-F4	E-200-R3-F5	E-200-R3-F6	E-200-R3-F7
Saltstone	Total	50	T-50-R1-1	T-50-R2-1	T-50-R3-1				
		200	T-200-R1-1	T-200-R2-1	T-200-R3-1				
Salt	Equilibrium	200	LE-200-F1	LE-200-F2	LE-200-F3				
Salt	Total	200	LT-200-1						
Standard	Equilibrium	200	E-STD-R1-1	E-STD-R2-1					
Standard	Total	200	T-STD-R1-1	T-STD-R2-1					

Table 3-6. Test Vessel Matrix & Samples

HOT BLOCK 1					
Date Sampled					
9/5 Fri	T-200-R1-1	T-200-R2-1	T-200-R3-1		
9/6 Sat	T-200-R1-2	T-200-R2-2	T-200-R3-2		
9/7 Sun					
9/8 Mon	T-200-R1-4	T-200-R2-4	T-200-R3-4		
9/9 Tue					
9/10 Wed					
9/11 Thu	T-200-R1-7	T-200-R2-7	T-200-R3-7		
9/5 Fri	E-0-R1-F1	E-0-R1-F2	E-0-R1-F3	E-0-R1-F4	E-0-R1-F5
9/6 Sat	E-0-R1-F1-S3	E-0-R1-F2-S4	E-0-R1-F3	E-0-R1-F4	E-0-R1-F5
9/7 Sun					
9/8 Mon					
9/9 Tue					
9/10 Wed					
9/11 Thu	E-0-R1-F1-S7	E-0-R1-F2-S7	E-0-R1-F3-S7	E-0-R1-F4-S7	
9/5 Fri	E-200-R3-F1	E-200-R3-F2	E-200-R3-F3	E-200-R3-F4	E-200-R3-F5
9/6 Sat	E-200-R3-F1-S3	E-200-R3-F2-S4	E-200-R3-F3	E-200-R3-F4	E-200-R3-F5
9/7 Sun					
9/8 Mon					
9/9 Tue					
9/10 Wed					
9/11 Thu	E-200-R3-F1-S7	E-200-R3-F2-S7	E-200-R3-F3-S7	E-200-R3-F4-S7	
9/5 Fri	E-200-R2-F1	E-200-R2-F2	E-200-R2-F3	E-200-R2-F4	E-200-R2-F5
9/6 Sat	E-200-R2-F1-S3	E-200-R2-F2-S4	E-200-R2-F3	E-200-R2-F4	E-200-R2-F5
9/7 Sun					
9/8 Mon					
9/9 Tue					
9/10 Wed					
9/11 Thu	E-200-R2-F1-S7	E-200-R2-F2-S7	E-200-R2-F3-S7	E-200-R2-F4-S7	
9/5 Fri	E-200-R1-F1	E-200-R1-F2	E-200-R1-F3	E-200-R1-F4	E-200-R1-F5
9/6 Sat	E-200-R1-F1-S3	E-200-R1-F2-S4	E-200-R1-F3	E-200-R1-F4	E-200-R1-F5
9/7 Sun					
9/8 Mon					
9/9 Tue					
9/10 Wed					
9/11 Thu	E-200-R1-F1-S7	E-200-R1-F2-S7	E-200-R1-F3-S7	E-200-R1-F4-S7	
HOT BLOCK 2					
Date Sampled					
9/5 Fri	T-50-R1-1	T-50-R2-1	T-50-R3-1	LT-200-1	
9/6 Sat	T-50-R1-2	T-50-R2-2	T-50-R3-2	LT-200-2	
9/7 Sun					
9/8 Mon	T-50-R1-4	T-50-R2-4	T-50-R3-4	LT-200-4	
9/9 Tue					
9/10 Wed					
9/11 Thu	T-50-R1-7	T-50-R2-7	T-50-R3-7	LT-200-7	
9/5 Fri					LE-200-F1
9/6 Sat					LE-200-F1-S3
9/7 Sun					
9/8 Mon					
9/9 Tue	E-0-R1-F6	E-0-R1-F7			LE-200-F1-S6
9/10 Wed					
9/11 Thu					
9/5 Fri			E-50-R3-F1	E-50-R3-F2	E-50-R3-F3
9/6 Sat			E-50-R3-F1-S3	E-50-R3-F2-S4	E-50-R3-F3
9/7 Sun					
9/8 Mon					
9/9 Tue					
9/10 Wed	E-200-R3-F6	E-200-R3-F7	E-50-R3-F1-S7	E-50-R3-F2-S7	E-50-R3-F3-S7
9/11 Thu					
9/5 Fri			E-50-R2-F1	E-50-R2-F2	E-50-R2-F3
9/6 Sat			E-50-R2-F1-S3	E-50-R2-F2-S4	E-50-R2-F3
9/7 Sun					
9/8 Mon					
9/9 Tue					
9/10 Wed	E-200-R2-F6	E-200-R2-F7	E-50-R2-F1-S7	E-50-R2-F2-S7	E-50-R2-F3-S7
9/11 Thu					
9/5 Fri			E-50-R1-F1	E-50-R1-F2	E-50-R1-F3
9/6 Sat			E-50-R1-F1-S3	E-50-R1-F2-S4	E-50-R1-F3
9/7 Sun					
9/8 Mon					
9/9 Tue					
9/10 Wed	E-200-R1-F6	E-200-R1-F7	E-50-R1-F1-S7	E-50-R1-F2-S7	E-50-R1-F3-S7
9/11 Thu					

Table 3-6. Test Vessel Matrix & Samples (continued)

HOT BLOCK 3					
Date Sampled					
9/5 Fri	T-STD-R1-1	T-STD-R2-1			
9/6 Sat					
9/7 Sun	T-STD-R1-3	T-STD-R2-3			
9/8 Mon					
9/9 Tue	T-STD-R1-5	T-STD-R2-5			
9/10 Wed					
9/11 Thu	T-STD-R1-7	T-STD-R2-7			
9/5 Fri					
9/6 Sat	LE-200-F2	LE-200-F3			
9/7 Sun					
9/8 Mon	LE-200-F2-S4				
9/9 Tue					
9/10 Wed					
9/11 Thu	LE-200-F2-S7	LE-200-F3-S7			
9/5 Fri					
9/6 Sat					
9/7 Sun					
9/8 Mon	E-50-R3-F4	E-50-R3-F5	E-50-R3-F6	E-50-R3-F7	
9/9 Tue					
9/10 Wed	E-50-R3-F4-S7				
9/11 Thu					
9/5 Fri					E-STD-R1-1
9/6 Sat					E-STD-R1-3
9/7 Sun	E-50-R2-F4	E-50-R2-F5	E-50-R2-F6	E-50-R2-F7	E-STD-R1-5
9/8 Mon					E-STD-R1-7
9/9 Tue					
9/10 Wed	E-50-R2-F4-S7				
9/11 Thu					
9/5 Fri					E-STD-R2-1
9/6 Sat					E-STD-R2-3
9/7 Sun					E-STD-R2-5
9/8 Mon	E-50-R1-F4	E-50-R1-F5	E-50-R1-F6	E-50-R1-F7	E-STD-R2-7
9/9 Tue					
9/10 Wed	E-50-R1-F4-S7				
9/11 Thu					

3.4 EQUILIBRIUM CONCENTRATIONS

The concentrations of ammonia over salt solutions and pH 12 water at 95 °C were estimated using the OLI Systems, Inc. StreamAnalyzer aqueous electrolyte simulation software (StreamAnalyzer, version 2.0.58, OLI Systems, Inc., Morris Plains, NJ). The calculations were done using the Aqueous (H+) thermodynamic framework and the Public database; the zeolite and GIBBSITE databases were also used in some calculations, but were found to have no effect on the calculated vapor-liquid equilibria.

The Henry's law constant for ammonia over the MCU simulant salt solution was calculated for the solution composition given in Table 3-1. To validate the OLI prediction of the Henry's law constant for ammonia, OLI predictions for a tank waste simulant composition tested by the Pacific Northwest National Laboratory (PNNL) were compared with values given by PNNL.¹⁰ The PNNL simulant composition (SY1-SIM-93B) is shown in Table 3-7 along with the MCU simulant composition.

Table 3-7. Composition and Properties of PNNL & MCU Salt Simulants

Species	Concentration (M)	
	MCU Simulant	PNNL Simulant
Na ⁺	5.61	6.63
OH ⁻	1.37	3.40
NO ₃ ⁻	3.31	1.69
NO ₂ ⁻	0.37	2.00
CO ₃ ⁻²	0.18	0.20
SO ₄ ⁻²	0.059	0
Al ⁺³	0.054	0.43
PO ₄ ⁻³	0.012	0

The PNNL simulant Henry's law constant was reported to be described by the following equation:

$$\ln H = -7.357 + 3330.1/T - 0.002139 T$$

where T is temperature (K) and H is the Henry's law constant in m/atm (mol NH₃/kg water)/(atm NH₃).

A plot of the Henry's law constant in these units from the PNNL model, predicted by OLI from the PNNL composition, and predicted for the MCU simulant composition are given in Figure 3-5.

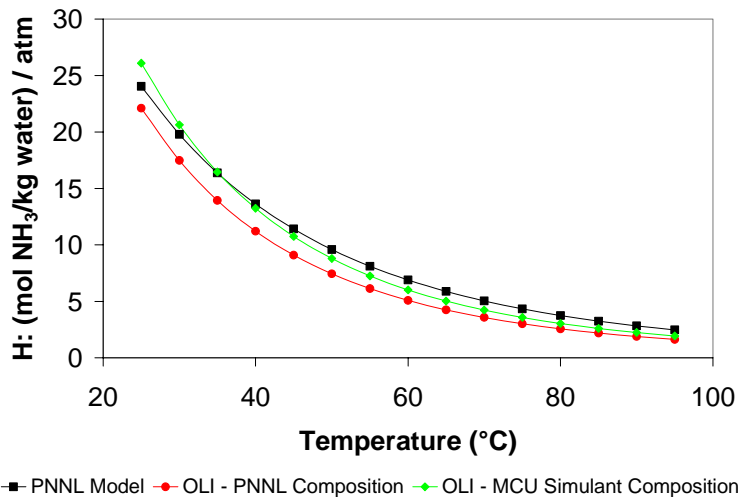


Figure 3-5. Henry's Law Constants in m/atm for PNNL & MCU Compositions

The Henry's law constants in the more convenient dimensionless units of:

$$(g \text{ NH}_3/L)_{\text{vapor}} / (g \text{ NH}_3/L)_{\text{solution}}$$

are shown in Figure 3-6. In these units, 1.94 mol/kg/atm is equivalent to 0.0209 (g/L/g/L). Note that the denominator (g NH₃/L)_{solution} is the value at the actual temperature; i.e., because the density of the solution decreases with increasing temperature, the volume increases and the liquid phase NH₃ concentration in g/L decreases.

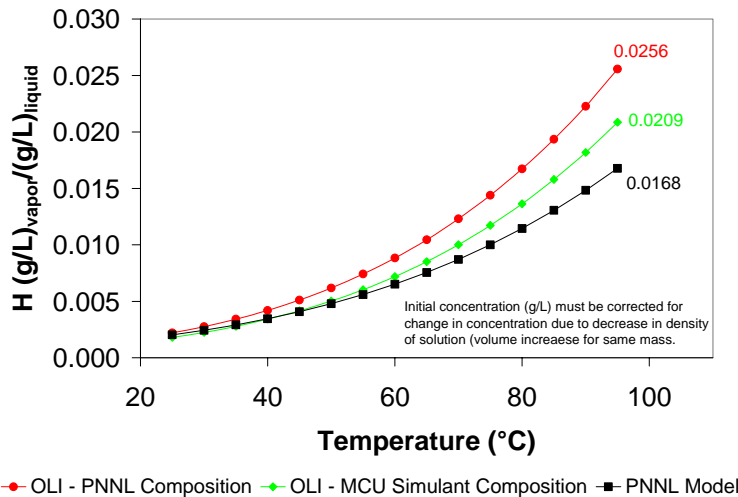


Figure 3-6. Dimensionless Henry's Law Constants (g/L/g/L) for PNNL & MCU Compositions

The OLI prediction for the PNNL composition is about 9% higher at 25 °C and 52% higher at 95 °C. Therefore, the OLI model predicts higher ammonia concentrations in the vapor than the correlation, which is conservative. At 95 °C, the predicted dimensionless Henry's law constant for the MCU simulant is 0.0209.

The expected concentrations of ammonia in the vapor space over salt solution at 95 °C, using this Henry's law constant, are shown in Table 3-8. The ammonia concentration above dilute ammonia in water at pH 12 is also shown (as predicted by OLI). Note that the NH₃ concentration in solution, C_{NH₃}, at 95 °C must be used in:

$$y_{\text{NH}_3} \left(\frac{g}{L} \right) = H \left(\frac{g/L}{g/L} \right) C_{\text{NH}_3} \left(\frac{g}{L} \right)$$

because the Henry's law constants in the table are for 95 °C. At 95 °C, the concentration of NH₃ in salt solution is the concentration at 25 °C / 1.0452, where 1.0452 is the ratio of the density at 25 °C to that at 95 °C. For water, this ratio is 1.0364.

Table 3-8. Calculated Equilibrium Ammonia in Headspace at 95 °C

Solution (NH ₃ Conc. at 25 ; 95 °C)	Henry's Law Constant* (m/atm)	Henry's Law Constant* (g/L/g/L)	Ammonia* (atm)	Ammonia* (mg/L)
Salt Solution (50 ; 47.84mg/L)	1.94	0.02086	0.00177	0.998
Salt Solution (200 ; 191.4 mg/L)	1.94	0.02086	0.00708	3.99
Water, pH 12 (200 ; 193.0 mg/L)	4.52	0.00761	0.00261	1.47

* at 95 °C

The equilibrium concentration of ammonia above saltstone was assumed to be the same as the concentration above the salt solution. No adjustment for the effect of the saltstone matrix on the equilibrium was done because the nature of this adjustment was not known. It has been speculated that the effective Henry's law constant would increase because the ionic species in the pore liquor would be more concentrated due to water evaporation and incorporation into the saltstone matrix; such an increase in the Henry's law constant due to increased ionic strength is expected, as shown in Figure 3-7. For this figure, the dimensionless Henry's law constant was predicted using OLI as a function of water removed from 100% initial mass to 85%.

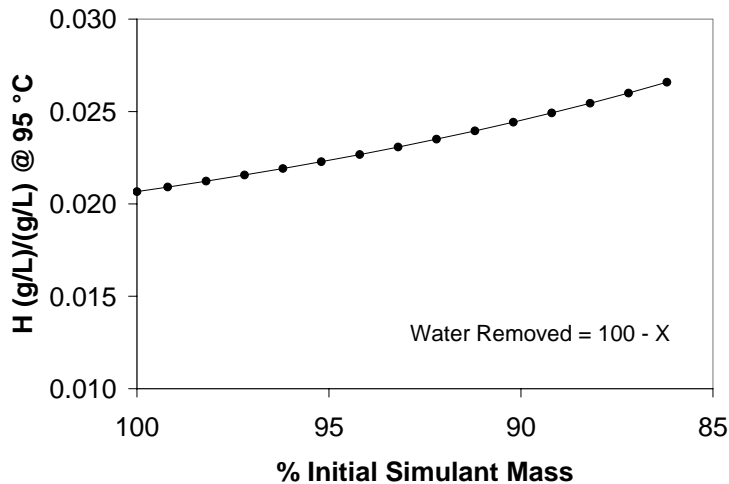


Figure 3-7. Increase in Dimensionless Henry's Law Constant for MCU Simulant Due to Increase in Ionic Strength

The equilibrium concentration of ammonia in the vapor could also be expected to be higher because the ammonia concentration in the pore liquor would increase as water is incorporated into the saltstone matrix. Counteracting these two effects that would increase the amount of ammonia in the vapor would be the possible decrease in diffusivity of the ammonia in the saltstone due to the development of the pore structure.

4.0 RESULTS AND DISCUSSION

4.1 ANALYTICAL RESULTS – AMMONIA RECOVERY

Standard samples were submitted to the F/H Laboratory for ammonia analysis. These included samples of premix raw materials extracted into dilute sulfuric acid and samples recovered from the acid-impregnated silica gel sample tubes. The sample tubes were spiked with several amounts of ammonia using NH₄Cl solutions adjusted to pH 12. The premix samples were analyzed as received and also with 100 mg/L spikes of ammonia. The lab also analyzed 40 mg/L standard solutions.

The recovery of 21 spiked raw material extracts and the 40 mg/L standards had a mean of 90.7% and a standard deviation of 4.3. Silica gel sample tubes were spiked with approximately 7, 80, and 970 µg of ammonia to give extract concentrations of about 0.7, 8, and 97 mg/L. The recovery of these samples is shown in Table 4-1. For concentrations near 100 mg/L, the recovery was 91.5%. These samples were also spiked by the laboratory with 100 mg/L ammonia; these spike recoveries were 90.7±0.8%. For samples at greater than 40 mg/L in the extract analyzed (as extracted, spiked with additional ammonia, or as a lab standard), the recovery was approximately 91% (90.7, 91.5, 90.7). Therefore, silica gel sample tube results with extract concentrations of about 100 mg/L and greater should be divided by about 0.91 to adjust for sample recovery. The samples with concentrations near 8 mg/L had recoveries of only about 80%, suggesting that at lower measured values, the concentrations measured should perhaps be divided by about 0.80. However, these data were also divided by 0.91 because the higher standard deviation of 19.2% on this mean of 79.3% incorporated 91%.

Table 4-1. Recovery of Silica Gel Sampling Tubes

Ammonia in Sample Extract (mg/L)	Number of Samples	% Recovery Mean	% Recovery Standard Deviation
<1	6	BQL	NA
7.1 to 8.6	7	79.3	19.2
94.5 to 100	3	91.5	3.0

BQL: below quantification limit

The standard solutions used for the saltstone tests were also analyzed. These standard solutions included 50 and 200 mg/L ammonia in simulant and a standard 200 mg/L ammonia in water adjusted to pH 12. The measured concentrations and recovery adjusted concentrations are shown in Table 4-2. All of the adjusted concentrations are within 10% of the target concentrations and so were assumed to be at the target values. These results also demonstrate that the 91% recovery value used was reasonable.

Table 4-2. Measured Concentrations of Standard Solutions

Solution	Measured Ammonia (mg/L)	Measured Ammonia adjusted for Recovery (mg/L)
50 mg/L Simulant	41	45
200 mg/L Simulant	173	190
200 mg/L Standard	178	195

4.2 EXPERIMENTAL RESULTS & DISCUSSION

4.2.1 Mass Losses from Vessels During Tests

During the seven-day duration of the tests, all vessels lost mass due to evaporation of water. The amount of water evaporated depended on the type of sample. The mean mass losses are tabulated in Table 4-3. All of the Total T samples lost less water than the comparable Equilibrium E samples. The E samples were open to the atmosphere through the filter, whereas the T samples quickly became more isolated from the atmosphere by condensed water in the sample tube. It appears that the condensed water in the sample tube decreased the rate of evaporation of the water in all samples. For this reason, the T samples may have actually been closer to liquid-vapor equilibrium than the Equilibrium samples. Note that the actual Saltstone vault is open to the atmosphere through (relatively) small vents; therefore, the vault behavior might be somewhere between the E and T tests.

The other trend that is obvious from the data is that the relative volatility of water was highest in the liquid standard for both the E and T tests. The saltstone samples had the lowest volatility. The liquid standard is expected to have the highest volatility because it has the lowest ionic strength. The salts in the simulant raise the boiling point and decrease the vapor pressure at a given temperature. The lower apparent volatility of water from the saltstone relative to the salt solution is consistent with the possible increase in ionic strength due to incorporation of water into the saltstone matrix and the expected lower diffusivity of water in saltstone pore structure relative to salt solution. The saltstone, liquid, and headspace volumes are summarized in Appendix 7.1. Because the volume of the liquid samples decreased during the experiments, an interpolated volume based on the initial and final volumes was used for most calculations.

Table 4-3. Mass Losses from Vessels

Sample Type	Mean % Mass Lost
Saltstone T (T-50, T-200)	1.9
Salt Solution T (LT-200)	3.3
Liquid Standard T (T-STD)	7.0
Saltstone E (E-0, E-50, E-200)	6.3
Salt Solution E (LE-200)	12.1
Liquid Standard E (E-STD)	18.2

4.2.2 Ammonia Release Data

The experimental data collected was the quantity of ammonia collected on the silica gel sampling tube. For the equilibrium E tests, these data and the measured volumes of the vessel headspaces were combined to give a concentration of ammonia in the headspace in mg/L. For the total T tests, the headspace H purge samples are also expressed in mg/L, whereas for the cumulative C sample tubes that remained on the vessels, the results are reported in mg evolved. For comparison of results, the ratio of (Measured Equilibrium)/Equilibrium (M/E) is useful; Measured Equilibrium (M) means the amount of ammonia actually measured in the headspace; Equilibrium (E) is the calculated amount of ammonia in the headspace based on the Henry's law constants from Section 3.4.

Note that neither Measured Equilibrium nor Equilibrium are actually equilibrium values. The measured 'equilibrium' was lower than the actual equilibrium concentration for several reasons. Because the headspace was not necessarily at the same temperature as the liquid, convection occurred in the headspace such that equilibrium would not necessarily exist between the bulk vapor and the liquid surface. The bulk evaporation of water from the surface would also lower the apparent equilibrium concentration. For saltstone, the equilibrium concentration of ammonia in the vapor could be higher because the ammonia concentration in the pore liquor would increase as water is incorporated into the saltstone matrix, whereas the equilibrium concentration could be lower due to a decrease in diffusivity of the ammonia in the saltstone due to the development of the pore structure.

The M/E value can be calculated for the E tests 'E(M/E)' and the T test headspace samples 'H(M/E). These ratio results are shown in Table 4-4. These values are the average values found from several measurements for each sample. The original experimental results (mg ammonia) are tabulated in Appendix 7.2. The ratios of the headspace samples from the Total tests to the expected equilibrium amounts H(M/E) are

also shown in Table 4-4 column (a). The total ammonia collected ratioed to both the amount initially present (T/I) in the saltstone or liquid and the adjusted (for chemical generation of ammonia) amount (T/I_a) are shown in columns (c) and (d), respectively. The adjustments for chemical generation are covered in Section 4.2.4.

Ranges of ratios are shown for the LE, T-STD, and LT-200 samples. For these samples the expected equilibrium was calculated based on several assumptions for the liquid composition and quantity. During the tests, both the volume and concentration of ammonia in the liquids decreased, so the headspace volume increased and the equilibrium concentration of ammonia in the headspace decreased. The drop in ammonia concentration in the liquid was estimated two ways; for the Total samples, the remaining ammonia was estimated from the amount collected on the sample tubes; for all liquids, the amount remaining was also estimated by linear interpolation of the initial and final concentrations. The equilibrium amounts of ammonia in the headspace were then calculated from these calculated liquid compositions. The higher Measured/Equilibrium values result from using the lower calculated liquid ammonia concentrations because the calculated equilibrium amount is smaller. Use of these lower values is probably most accurate. For the saltstone samples, no such corrections were made because the concentration of ammonia in the saltstone was not known as a function of time.

Table 4-4. Ratio of Measured to Equilibrium Ammonia Concentration and Fraction Initial Ammonia Collected

Test Type	Material	Sample Designation	Mean NH ₃ Measured in Headspace (mg/L)	a	b	c	d
				<u>Original Measured</u> Equilibrium	<u>Adjusted Measured</u> Equilibrium	<u>(Total Collected)</u> (Total Initially Added) (T/I)	<u>(Total Collected)</u> (Adjusted Total Initially Added) (T/I _a)
				<u>E(M/E)</u>	<u>E(M/E_a)</u>		
E	Saltstone	E-0	0.84	NA	0.35		NA
E	Saltstone	E-50	0.83	0.83	0.24		
E	Saltstone	E-200	1.30	0.33	0.20		
E	Standard	E-STD ^α	0.29	0.19	NA		
E	Simulant Liquid	LE-200 ^α	1.07	0.27-0.32	NA		
				<u>H(M/E)</u>	<u>H(M/E_a)</u>		
T	Saltstone	T-50	2.03	2.03	0.60	2.17	0.64
T	Saltstone	T-200	2.68	0.67	0.43	0.74	0.46
T	Simulant Liquid	LT-200	4.20	1.05-1.84	NA	0.36-0.76	NA
T	Standard	T-STD	1.66	1.17-1.35	NA	0.09-0.55	NA

^α one data point

The M/E values for the E tests in column (a) of Table 4-4 are consistent (0.19 to 0.33, except for the E-50 tests). These data suggest that the behavior of the E experimental system was consistent between the liquid and saltstone tests. Although these data only indicate headspace concentrations of about 30% of equilibrium, the results are consistent. It is clear that the E experiments did not measure the actual vapor pressure of ammonia

above the samples, so the data could not be used to determine a Henry’s law constant. (For the saltstone vault, the assumption has been made that the ammonia in the headspace is the equilibrium amount above the salt solution feed.)

The E-0 and E-50 samples had the same amount of ammonia in the headspace even though the only initial ammonia source for the E-0 samples was the small amount present in the slag premix component. Therefore, it is evident that a significant amount of ammonia was chemically generated from the saltstone. Table 4-5 summarizes for the Total tests the ratio of the total ammonia released to that initially added (T/I). For the liquid samples, the V values are based on the initial amount and the cumulative total of the sample tubes; the L values are based on the initial amount and the final measured liquid. From these data, it is apparent that there was more ammonia released from the T-50 tests than was initially present in the sample. The T-200 data also tend to indicate more ammonia was evolved than initially present (compare T-200 (0.74) to LT-200 (0.36) or T-STD (0.09) on same basis (V)). Although 0.74 by itself does not indicate more ammonia released than initially present (a value > 1.0 would), the fact that the T-200 value is more than twice the LT-200 value indicates something very different occurs, which could be attributed to chemical generation.

Table 4-5. Total Ammonia Released/Added

		Ammonia Released / Initially Added (T/I)						
Test		Replicates				Mean		
Saltstone	T-50	2.37	2.20	1.95		2.17		
Saltstone	T-200	0.72	0.80	0.69		0.74		
Liquid	LT-200	V	L	-	-	V	L	
		0.36	0.76			0.36	0.76	
Liquid	T-STD	V	L	V	L	-	V	L
		0.19	0.37	0.09	0.55		0.14	0.46

- saltstone values based on ammonia measured by vapor sample tubes
- liquid values based on ammonia measured by vapor sample tubes (V), and on difference between initial and final ammonia concentrations in liquid (L)

Table 4-6 shows that the amount of ammonia found in the headspace {H(M/E)} of both the liquid and saltstone T samples was significantly greater than for the corresponding E samples {E(M/E)}. As mentioned previously, it appears that the H(M/E) ratios from the Total samples are somewhat more representative of the equilibrium amount of ammonia than are the E samples. Specifically, the H(M/E) results for the LT-200 and T-STD samples were 1.05-1.84 and 1.17-1.35, respectively, which are reasonably close to the equilibrium value of 1.0. The saltstone T-50 and T-200 samples have higher M/E values than the saltstone E samples. This ratio is even higher for the liquid samples. The liquid samples had H/E ratios of ~5-7; if saltstone behaved the same as the simulant and standard liquids, the H/E ratios for the T-50 and T-200 samples would be expected to be similar. However, these values are much lower, indicating that the amount of ammonia in the vapor of the saltstone samples was less than the amount present in corresponding liquid samples.

Even though the T samples do not give an actual measurement of the vapor pressure or Henry's law constant for the liquid samples, the data definitely show that the amount of ammonia above the saltstone was less than for liquid with the same composition, even with increased ammonia content due to chemical generation of ammonia from the saltstone.

Table 4-6. Comparison of Total Test Equilibrium H Results to Equilibrium Test E Results

Sample Type	Total Test Equilibrium H(M/E)		Equilibrium Test E(M/E)	H/E Ratio
STD	1.17-1.35	>	0.19	~7
L	1.05-1.84	>	0.27-0.32	~5
T-50	2.03	>	0.83	2.4
T-200	0.67	>	0.33	2.0

H/E is (headspace equilibrium from T test)/(measured headspace equilibrium from E test)

The saltstone T-200 data forms the basis for the calculation of the expected amount of ammonia in the headspace above curing saltstone made from salt solution with 200 mg/L ammonia. This value is 2.68 mg/L vapor in Table 4-4. An upper 95% confidence interval on this mean value is:

$$\bar{y} + t(\alpha, n-1) \times \frac{\sigma}{\sqrt{n}}$$

where \bar{y} is the mean and $t(\alpha, n-1) = 1.796$ is the upper α -tail of the Student's t distribution with n-1 degrees of freedom. For the present data (n=12), this equation is:

$$2.68 + 1.796 \times \frac{2.36}{\sqrt{12}} = 3.90$$

Therefore, the upper 95% confidence estimate on the concentration of ammonia in the headspace is 3.90 mg/L vapor.

For the saltstone equilibrium E data at 200 mg/L initial NH₃, the average headspace NH₃ concentration was 1.30 mg/L vapor. The upper 95% confidence interval on this value is 1.39 mg/L vapor (n=39, $\sigma=0.336$, $t(0.05, 38) = 1.684$). Recall, however, that the E test concentrations were found to be significantly lower than expected due to the higher evaporation rate of the water, and are thus not a conservative estimate.

The equilibrium value calculated from Henry's law for salt solution with 200 mg/L ammonia is 3.99 mg/L vapor.

4.2.3 Data Trends

Figure 4-1 shows that the E equilibrium test headspace ammonia amounts for both 50 and 200 mg/L saltstone increase in the first 4 days, then drop between days 4 and 7; the blank saltstone has approximately constant ammonia amounts. Figure 4-2 shows this same data for the T(H) total equilibrium samples and the T(H) liquid samples. The liquid sample results all drop slowly in time, whereas the 200 mg/L saltstone shows a peak at 4 days (solid red line); the approximate average value is shown by the dotted red line. The 50 mg/L saltstone data do not show any peaking trend. Due to the significant spread of the replicate data points, the average equilibrium amounts of ammonia were used for the calculations in Section 4.2.2. These results are for vessels open to the atmosphere, so the headspace amounts would be expected to be proportional to the vapor pressure. This behavior suggests two possible explanations: 1) ammonia by chemical generation increases in the first several days, resulting in a higher concentration in the saltstone and thus higher concentration in the vapor, and then declines; 2) ammonia generation increases, but at about day 4, the permeability of the saltstone decreases such that the amount of ammonia released decreases. The second explanation is consistent with the known setting behavior of saltstone, but this consistency does not rule out the first explanation. The liquid samples, not shown, started with higher release rates and then declined. As expected, there was no intermediate peak in ammonia evolution. The cumulative ammonia collected versus time for the total T tests is shown in Figure 4-3. The amount of ammonia collected is very linear in time for all four types of samples. The linearity of these total evolved results suggest that the high peak values for 200 mg/L saltstone (Figure 4-2) were probably biased high.

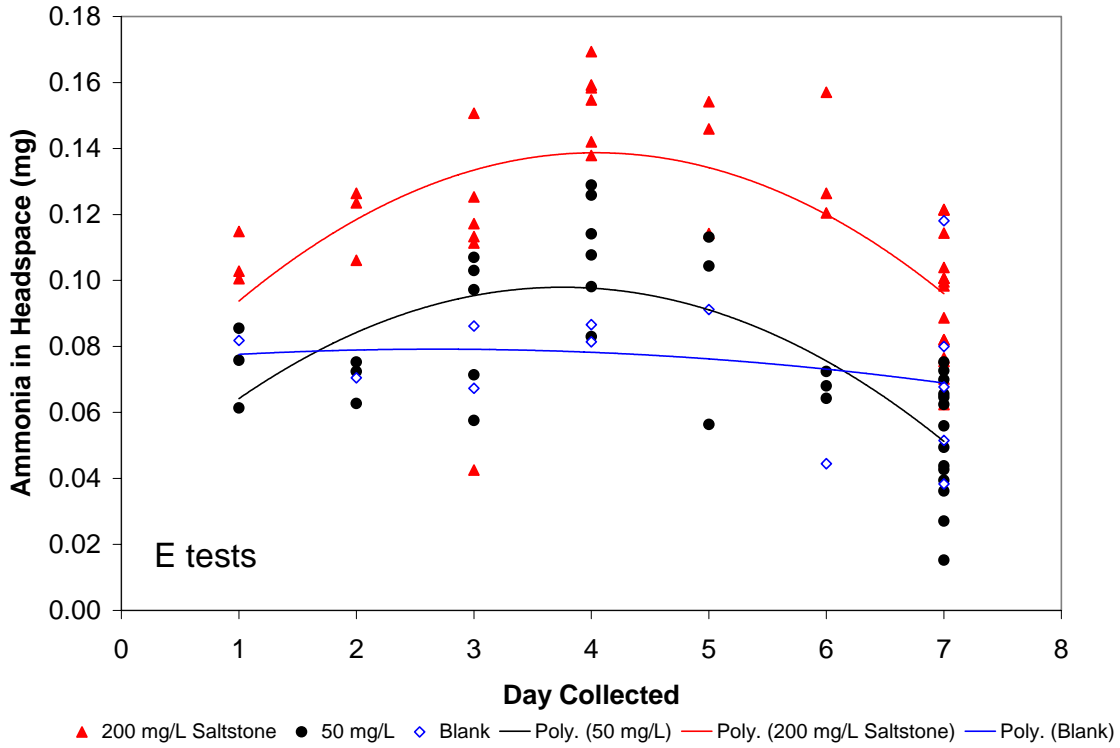


Figure 4-1. E-Tests Equilibrium Headspace Ammonia

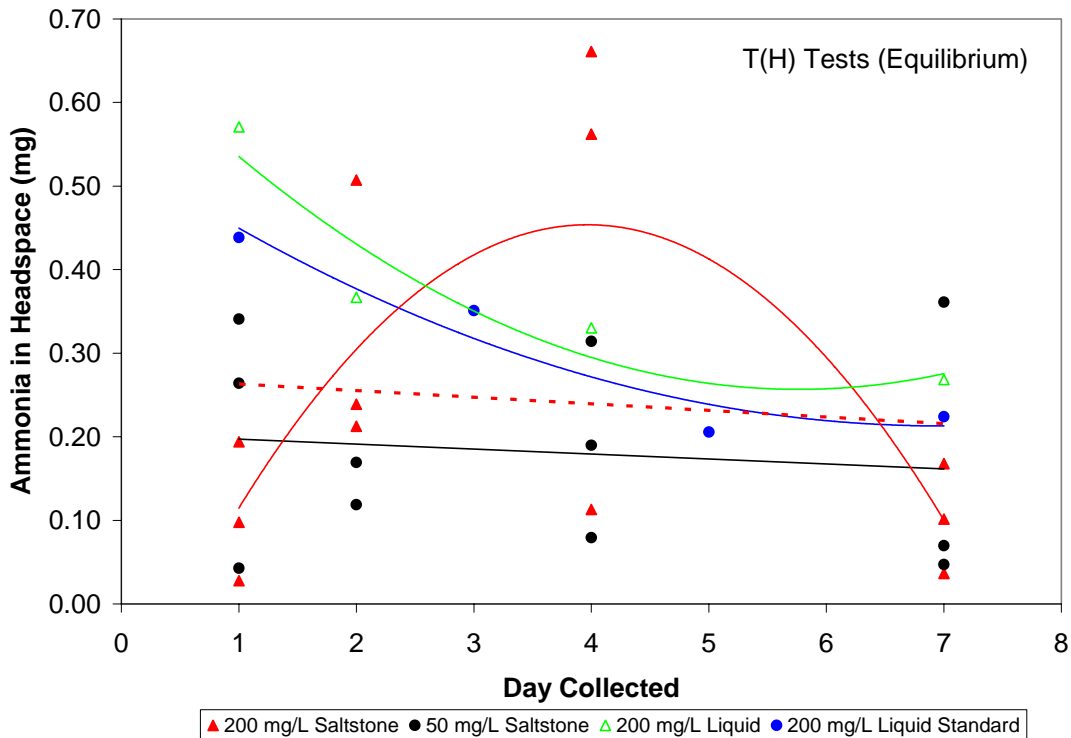


Figure 4-2 T(H) Tests Equilibrium Headspace Ammonia

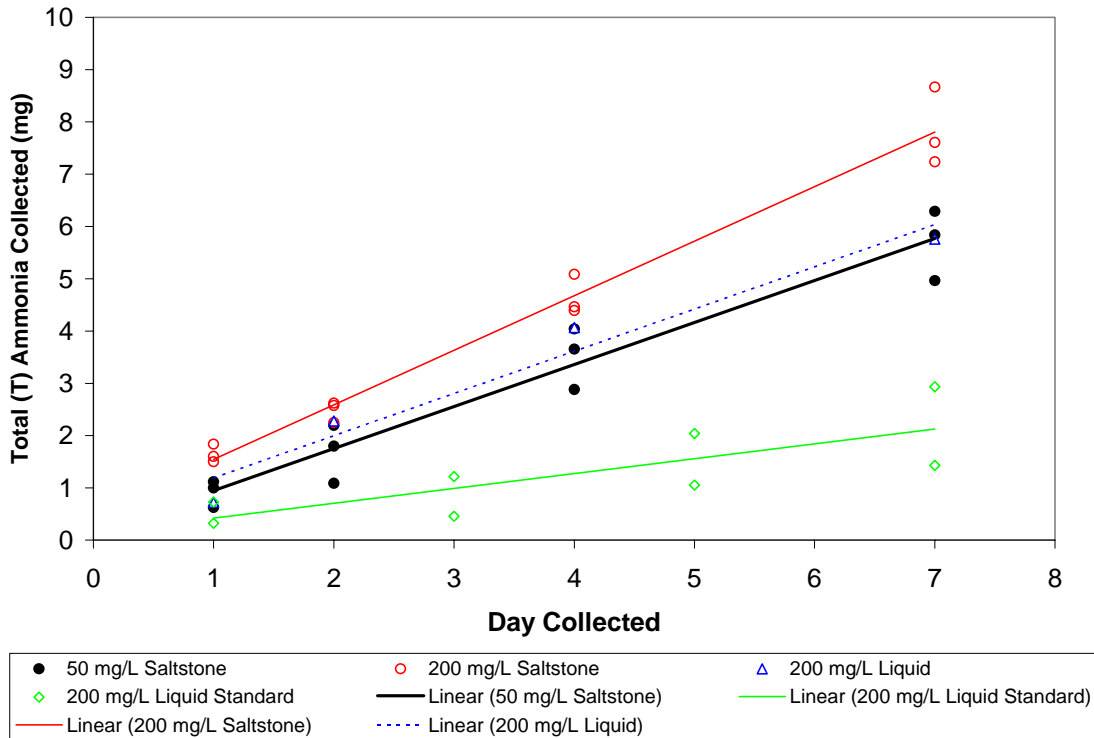


Figure 4-3. Cumulative Ammonia Collected in the Total Tests

4.2.4 Estimation of Chemically Generated Ammonia

The data for the blanks and the 50 mg/L salt saltstone show that ammonia was generated chemically within the saltstone. If it were assumed that all of the ammonia generated were present initially, the initial ammonia concentrations would be higher than the actual initial concentrations. In reality, the chemically generated ammonia was probably generated over the course of the experiment and not all at once. Therefore, the chemically generated ammonia would replace some or all of the ammonia lost to the headspace, and the concentration of ammonia in the saltstone would decrease slower than if there were no chemical reaction.

Figure 4-4 shows hypothetical ammonia concentration curves for saltstone with initially 0, 50, and 200 mg/L ammonia in the salt solution. The solid symbols show a 120 mg/L increase in ammonia for each initial concentration; for these curves, assume that there was no loss of ammonia to the headspace. The open triangles show what the actual concentration could have been with ammonia loss to the headspace; the larger symbols show an overall increase, the medium symbols show a decrease, and the small symbols show a larger increase such as might occur with no chemical generation. The actual concentration of ammonia within the saltstone cannot be determined, so which type of curve actually applies is not known. It could also be possible to have an initial increase in ammonia in the saltstone solution, followed by a decrease due to slowing generation of ammonia.

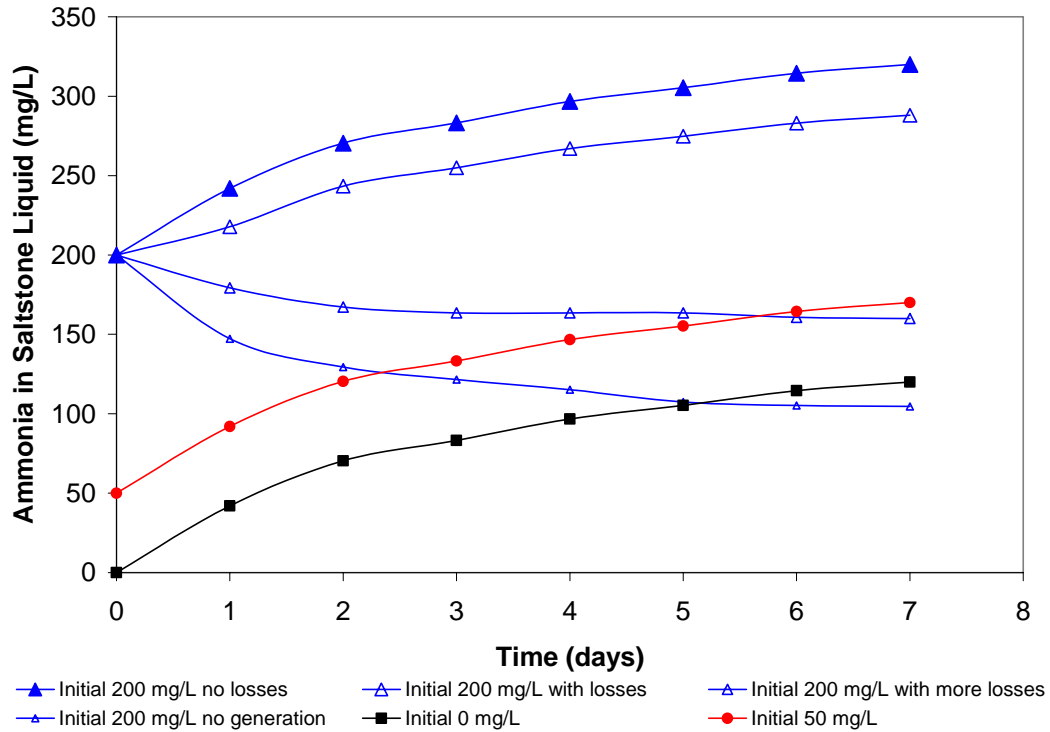


Figure 4-4. Hypothetical Saltstone Ammonia Concentrations Due to Chemical Generation

Because the actual concentration versus time curve for the salt solution in saltstone is unknown and cannot be determined, actual graphs of concentration versus time cannot be plotted. However, as an approximation to the behavior of the ammonia release from the saltstone, it could be assumed that the initial concentration at each salt ammonia concentration was the actual concentration plus some chemically generated amount:

$$x_0 = 0 + \delta_0$$

$$x_{50} = 50 + \delta_{50}$$

$$x_{200} = 200 + \delta_{200}$$

where x_i = initial concentration of ammonia with chemical generation

δ_i = ammonia generated

For simplicity, assume the amount of ammonia generated is independent of the initial concentration (this is not necessarily true), so the δ_i are equal. In Table 4-4, the H(M/E) values were tabulated for each test type. The H(M/E) for 50 mg/L saltstone was 2.03 and this value for 200 mg/L was 0.69. The equilibrium E concentration assumed in the denominator appears to be too low due to the ammonia chemically generated, so we need to assume a higher adjusted equilibrium concentration E_a of ammonia due to the higher adjusted concentration in the liquid (initial + δ).

The vapor pressures of ammonia above each sample, calculated from the Henry's law constant for the solution, are shown in column 2 in Table 4-7. Column 3 gives the corresponding mass of equilibrium NH_3 in the headspace (H or E) for an average vessel; column 4 shows the total mass of NH_3 in the original saltstone or liquid sample (I). In column 5, the total ammonia collected T is given and the mean for each sample type is in column 6. Note that 'Total NH_3 Collected' here is the sum of the cumulative and headspace amounts ($T = C + H$).

The Mean Collected (measured) / Expected Equilibrium T/E in column 7 is the ratio of columns 6/3. The T/E values for the Liquid Standard and 200 mg/L Salt solutions are expected to be the same if the amount of ammonia collected is proportional to the vapor pressure, so these values were averaged to give 16.4 mg/mg. This same ratio for the Saltstone 200 and 50 mg/L samples is 22.0 and 64.1, respectively. If the assumption above applies, and we assume that the vapor pressure above the saltstone and the salt solution have the same dependence on liquid concentration, then these values should both equal 16.4. The Adjusted P_{vap} (NH_3) values in column 9, 0.954 for T-200 and 0.694 for T-50) give ratios of 16.4.

If we now assume the saltstone samples all had the same amount of *additional* initial NH_3 (δ), then the adjusted T/E_a ratios for T-200 and T-50 samples cannot be exactly 16.4 and the P_{vap}^{adj} values in column 11 cannot equal those in column 9. The closest P_{vap}^{adj} values for a single value of δ are 1.13 and 0.600, respectively, for T-200 and T-50. These values were chosen because they make the mean of (Mean Collected / New Equilibrium), in column 13, equal to 16.4. The increase in initial concentration of NH_3 is 121 mg/L, so the estimated initial concentrations of NH_3 increase from 50 to 171, 200 to 321, and 0 to 121 mg/L.

Columns 14 and 15 show the original and adjusted E(M/E_a) ratios for the Equilibrium tests; the values for both 50 and 200 mg/L are closer to each other, and are closer to the values for the standard and simulant liquids (0.25 and 0.27-0.33, respectively). The H(M/E_a) ratios for the Total samples are also closer; the values for T-50 and T-200 are 0.60 (vs. 2.03) and 0.44 (vs. 0.69), respectively. These values are summarized in column (b) of Table 4-4. The E(M/E_a) value for the blank saltstone is 0.35, which is reasonably consistent with the other initial concentrations. The Total Collected/Total Initially Added (T/I and T/I_a) values also are closer after the addition of more initial ammonia, as shown in columns c & d of Table 4-4. The new values are all similar, except for the standard liquid, which is expected to be lower due to the lower vapor pressure of ammonia for the standard.

Therefore, an increase of about 121 mg/L of initial ammonia approximately accounts for the observed ammonia release data. This means that about 121 mg/L of ammonia may have been generated from each saltstone sample over the course of seven days.

Table 4-7. Estimation of Ammonia Chemically Generated

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
			E or H	I	T	T	T/E	T/E			P_{vap}^{adj}	E_a	T/ E_a	E(M/E)	E(M/ E_a)
Sample	Initial Conc (mg/L)	P_{vap} (NH ₃) (vol%)	Expected Equilibrium NH ₃ in Headspace (mg)	Total NH ₃ Initially Added (mg)	Total NH ₃ Collected (mg)	Mean Total NH ₃ Collected (mg)	Mean Collected / Expected Equil (mg/mg)	Mean Collected / Expected Equil (mg/mg)	Adjusted P_{vap} (NH ₃) (vol%)	New Initial Conc (mg/L)	P_{vap} Calculated Based on New Initial Conc (vol%)	Adjusted Expected Equilibrium NH ₃ Amount (mg)	Mean Collected / New Equil (mg/mg)	Equilibrium Samples (Meas/ Equil) original	Equilibrium Samples (Meas/ Equil) adjusted
Liquid Standard (T-STD)	200	0.195	0.110	15.2	2.67 1.21	1.94	17.6	16.4							
Salt 200 mg/L (LT)	200	0.708	0.382	16.0	5.76	5.76	15.1								
Saltstone 200 mg/L (T-200)	200	0.708	0.356	10.6	7.61 8.67 7.24	7.84	22.0	16.4	0.954	321	1.13	0.566	13.8	0.326	0.202
Saltstone 50 mg/L (T-50)	50	0.177	0.0889	2.62	6.29 5.84 4.96	5.70	64.1	16.4	0.694	171	0.600	0.302	18.9	0.827	0.244
Saltstone 0 mg/L	0									121	0.425	0.214			0.348

Expect these values to be equal if amount collected is proportional to vapor pressure

Ammonia to added to all initial concentrations (δ): 121

These values should = 16.4

Values that make mean = 16.4

Mean: 16.4

Closer to same value as expected

Adjust Ammonia to add to all initial concentrations so Mean Value = 16.4

New P_{vap} close to Adjusted P_{vap} , given same Ammonia to add.

Mean 0.265

5.0 CONCLUSIONS

The ammonia concentration above saltstone made from the 200 mg/L ammonia simulant was found to be about 2.7 mg/L at 95 °C. An upper 95% confidence value for this concentration was found to be 3.9 mg/L. This concentration, 3.9 mg/L, is approximately equal to the equilibrium concentration of ammonia above the simulant solution.

OLI simulations of the simulant solution composition were used to predict the Henry's law constant and the equilibrium vapor concentration of ammonia as a function of temperature. The OLI simulation was checked versus a correlation developed by PNNL for a particular waste composition. The OLI predictions were found to be about 50% higher than this correlation.

Recovery of ammonia from vapor samples collected on acid-impregnated silica gel sampling tubes is about 91% when the concentration of ammonia in the acid extract is above 40 mg/L. For concentrations of less than 10 mg/L, the recovery was found to be about 80%.

The tests did not measure the actual vapor equilibrium concentrations, but measurements on standard solutions of simulant and ammonia in water gave higher than equilibrium values for the vapor concentration (Total tests), indicating that the measurements were biased high. Results for saltstone samples were found to be lower than the corresponding equilibrium, so the actual amount would be even less when the bias is accounted for. Therefore, the measured vapor concentrations of ammonia above saltstone were conservatively high.

Ammonia release from saltstone appears to be lower than predicted by simple liquid-vapor equilibrium. Likely causes could be decreased volatility due to increased ionic strength in the pore liquid and decreased diffusivity or permeation due to the development of the pore structure of the saltstone.

The amount of ammonia released from the 50 mg/L ammonia saltstone was greater than the amount added and the blank saltstone also generated ammonia. Estimates showed that approximately 121 mg/L of ammonia on an initial simulant basis was generated by chemical generation in saltstone. When the amount of ammonia in the saltstone was adjusted by this amount, the Measured/Equilibrium (M/E) and Total/Initial (T/I) values for saltstone, salt simulant, and the ammonia standards were found to be similar.

The amount of ammonia found in the headspace in some of the saltstone equilibrium headspace samples at 50 and 200 mg/L initial ammonia peaked at 4 days, suggesting that the chemical generation rate of ammonia may have peaked, or that the release rate decreased due to decreased diffusivity from the development of pore structure. Although peaks were seen in some of the data, the statistical average headspace values were used for the calculations due to the large scatter in the data.

6.0 REFERENCES

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7.0 APPENDICES

7.1 APPENDIX A: VESSEL MASS & VOLUME DATA

Table 7-1. Saltstone Sample Masses & Volumes

Vessel	Tare Wt (g)	Initial Full Wt (g)	Final Full Wt (g)	Initial Saltstone Wt (g)	Final Saltstone or Liquid Wt (g)	Mass Loss (g)	Mass Loss % Initial	Mean	Final Headspace Volume (mL)
E-0-R1-F1	201.9	333.9	325.2	132.0	123.3	8.7	6.59%		88.7
E-0-R1-F2	204.5	342.4	332.8	137.9	128.3	9.6	6.96%		87.1
E-0-R1-F3	203.3	336.5	327	133.2	123.7	9.5	7.13%		88.5
E-0-R1-F4	208.5	343.6	335.6	135.1	127.1	8.0	5.92%		90.0
E-0-R1-F5	203.5	338.9	332.2	135.4	128.7	6.7	4.95%		87.9
E-0-R1-F6	203.4	338.6	329.6	135.2	126.2	9.0	6.66%		87.9
E-0-R1-F7	206.9	345.7	336	138.8	129.1	9.7	6.99%	6.46%	91.4
E-200-R1-F1	192.8	325.7	319.3	132.9	126.5	6.4	4.82%		88.3
E-200-R1-F2	190.5	325.2	317.9	134.7	127.4	7.3	5.42%		87.7
E-200-R1-F3	189.5	321.1	314	131.6	124.5	7.1	5.40%		85.9
E-200-R1-F4	185.7	317.1	310	131.4	124.3	7.1	5.40%		86.0
E-200-R1-F5	190.6	321.3	315.4	130.7	124.8	5.9	4.51%		91.2
E-200-R1-F6	191.5	323.1	317.2	131.6	125.7	5.9	4.48%		91.8
E-200-R1-F7	191.1	323.8	316.8	132.7	125.7	7.0	5.28%		93.3
E-200-R2-F1	187.6	319.2	310.8	131.6	123.2	8.4	6.38%		87.8
E-200-R2-F2	187.9	321.9	313.1	134.0	125.2	8.8	6.57%		89.2
E-200-R2-F3	187.0	320.9	311.7	133.9	124.7	9.2	6.87%		83.9
E-200-R2-F4	191.1	328.3	319.9	137.2	128.8	8.4	6.12%		88.7
E-200-R2-F5	190.3	322.9	316	132.6	125.7	6.9	5.20%		86.3
E-200-R2-F6	189.8	325.8	318.2	136.0	128.4	7.6	5.59%		88.3
E-200-R2-F7	190.1	325.2	316.5	135.1	126.4	8.7	6.44%		89.2
E-200-R3-F1	189.5	325.9	317.6	136.4	128.1	8.3	6.09%		85.2
E-200-R3-F2	184.3	312.9	302.8	128.6	118.5	10.1	7.85%		91.5
E-200-R3-F3	190.7	326.6	317.2	135.9	126.5	9.4	6.92%		91.2
E-200-R3-F4	188.6	325.7	317	137.1	128.4	8.7	6.35%		84.5
E-200-R3-F5	189.6	318.3	310.9	128.7	121.3	7.4	5.75%		91.1
E-200-R3-F6	188.1	320.3	311.8	132.2	123.7	8.5	6.43%		89.4
E-200-R3-F7	188.6	323.6	314.2	135.0	125.6	9.4	6.96%	5.94%	87.7
E-50-R1-F1	190.0	328.7	321.1	138.7	131.1	7.6	5.48%		86.1
E-50-R1-F2	190.0	318.7	311.2	128.7	121.2	7.5	5.83%		90.6
E-50-R1-F3	189.9	324.3	318.4	134.4	128.5	5.9	4.39%		88.3
E-50-R1-F4	186.8	319.2	311.6	132.4	124.8	7.6	5.74%		88.0
E-50-R1-F5	203.6	338.7	330.5	135.1	126.9	8.2	6.07%		91.9
E-50-R1-F6	201.6	328.7	319.4	127.1	117.8	9.3	7.32%		93.2
E-50-R1-F7	203.6	334.5	325.4	130.9	121.8	9.1	6.95%		95.1
E-50-R2-F1	204.2	337.8	328.5	133.6	124.3	9.3	6.96%		89.9
E-50-R2-F2	199.4	331	322.1	131.6	122.7	8.9	6.76%		89.0
E-50-R2-F3	202.6	338.1	330.4	135.5	127.8	7.7	5.68%		86.4
E-50-R2-F4	203.1	335.5	326.3	132.4	123.2	9.2	6.95%		87.6
E-50-R2-F5	204.3	336.1	325.4	131.8	121.1	10.7	8.12%		97.4
E-50-R2-F6	201.3	331.1	320.5	129.8	119.2	10.6	8.17%		90.1
E-50-R2-F7	206.9	333.8	333.3	126.9	126.4	0.5	0.39%		94.4

Vessel	Tare Wt (g)	Initial Full Wt (g)	Final Full Wt (g)	Initial Saltstone Wt (g)	Final Saltstone or Liquid Wt (g)	Mass Loss (g)	Mass Loss % Initial	Mean	Final Headspace Volume (mL)
E-50-R3-F1	198.4	332.7	322.8	134.3	124.4	9.9	7.37%		87.6
E-50-R3-F2	202.3	340.7	330.9	138.4	128.6	9.8	7.08%		89.9
E-50-R3-F3	204.6	340.6	332.1	136.0	127.5	8.5	6.25%		88.7
E-50-R3-F4	202.0	335.9	325.9	133.9	123.9	10.0	7.47%		89.0
E-50-R3-F5	206.2	341.8	331	135.6	124.8	10.8	7.96%		89.6
E-50-R3-F6	207.3	345	333.6	137.7	126.3	11.4	8.28%		87.8
E-50-R3-F7	201.1	338.2	328.5	137.1	127.4	9.7	7.08%	6.49%	86.8
Saltstone Equilibrium Samples			Mean	133.7	125.3	8.4	6.25%		89.1
			Stdev	3.0	2.9	1.8	1.30%		2.7
T-200-R1	207.5	345.2	342.6	137.7	135.1	2.6	1.89%		91.9
T-200-R2	207.4	348	345.8	140.6	138.4	2.2	1.56%		90.7
T-200-R3	209.7	347.5	344.7	137.8	135.0	2.8	2.03%		90.4
T-50-R1	206.9	344.8	342.7	137.9	135.8	2.1	1.52%		90.3
T-50-R2	202.4	335.7	333	133.3	130.6	2.7	2.03%		88.3
T-50-R3	198.3	331.8	328.4	133.5	130.1	3.4	2.55%		87.1
Saltstone Total Samples			Mean	136.8	134.2	2.63	1.93%		89.8
			Stdev	2.85	3.21	0.47	0.37%		1.8

Table 7-2. Liquid Sample Masses & Volumes

Vessel	Tare Wt (g)	Initial Full Wt (g)	Final Full Wt (g)	Initial Saltstone Wt (g)	Final Saltstone or Liquid Wt (g)	Mass Loss (g)	Mass Loss % Initial	Final Headspace Volume (mL)	Initial Headspace Volume (mL)	Total Vessel Volume (mL)	Initial Liquid Density (25 °C) (kg/L)	Final Liquid Density (25 °C) (kg/L)	Initial Liquid Volume (mL)	Final Liquid Volume (mL)
E-STD-R1	207.5	283.3	270.1	75.8	62.6	13.2	17.41%	104.8	91.6	167.4	1.00	1.00	75.8	62.6
E-STD-R2	205.2	282.5	267.8	77.3	62.6	14.7	19.02%	106.5	91.8	169.1	1.00	1.00	77.3	62.6
Equilibrium Tests			Mean	76.6	62.6	14.0	18.22%					①		
Standards			Stdev	1.1	0.0	1.1								
LE-200-F1	205.2	303.3	293.6	98.1	88.4	9.7	9.89%	102.2	92.8	171.3	1.25	1.28	78.5	69.1
LE-200-F2	200.9	292.8	280.5	91.9	79.6	12.3	13.38%	101.1	89.3	162.8	1.25	1.29	73.5	61.7
LE-200-F3	203.4	300.3	287.7	96.9	84.3	12.6	13.00%	100.8	88.6	166.1	1.25	1.29	77.5	65.3
Equilibrium Tests			Mean	95.6	84.1	11.5	12.06%	101.4	90.2					
Simulant			Stdev	3.3	4.4	1.6								
Equilibrium Tests						Mean	14.54%							
All Liquids						Stdev	3.66%							
LT-200	206.6	306.6	303.3	100.0	96.7	3.3	3.30%	93.2	89.9	169.9	1.25	1.26	80.0	76.7
T-STD-R1	204.8	280.3	275.9	75.5	71.1	4.4	5.83%	102.2	97.8	173.3	1.00	1.00	75.5	71.1
T-STD-R2	205.0	281.7	275.5	76.7	70.5	6.2	8.08%	98.0	91.8	168.5	1.00	1.00	76.7	70.5
Total Tests			Mean	76.1	70.8	5.30	6.96%	100.1	94.8					
Standards			Stdev	0.85	0.42	1.27	1.59%							
Total Tests						Mean	5.74%							
All Liquids						Stdev	2.39%							

① final density for simulant estimated from final mass by OLI simulation

7.2 APPENDIX B: ORIGINAL AMMONIA GENERATION DATA (MG NH₃)

☆ e.g., E-200-R2-F2-S4

<RQ values not included in Mean, StDev

Sample	☆ First Day (F)	Second Day (S)	Measured Ammonia in Sample (mg)	Measured Ammonia in Sample (mg/L)	Ammonia at Equilibrium (mg)	Measured / Equilibrium
E-50-R1	1	1	0.0855	0.993	0.0860	0.994
E-50-R1	2	2	0.0724	0.799	0.0905	0.800
E-50-R1	1	3	0.1030	1.20	0.0905	1.14
E-50-R1	3	3	0.0576	0.652	0.0882	0.652
E-50-R1	2	4	0.0981	1.08	0.0882	1.11
E-50-R1	4	4	0.129	1.46	0.0879	1.47
E-50-R1	5	5	0.113	1.23	0.0918	1.23
E-50-R1	6	6	0.0680	0.730	0.0931	0.731
E-50-R1	1	7	0.0726	0.843	0.0860	0.844
E-50-R1	2	7	0.0656	0.762	0.0905	0.725
E-50-R1	3	7	0.0753	0.875	0.0882	0.853
E-50-R1	4	7	0.0699	0.812	0.0879	0.795
E-50-R1	7	7	0.0153	0.161	0.0950	0.161
E-50-R2	1	1	0.0758	0.843	0.0898	0.844
E-50-R2	2	2	0.0627	0.705	0.0889	0.705
E-50-R2	1	3	0.0972	1.08	0.0898	1.082
E-50-R2	3	3	0.0714	0.826	0.0863	0.827
E-50-R2	2	4	0.0830	0.932	0.0889	0.933
E-50-R2	4	4	0.126	1.44	0.0875	1.44
E-50-R2	5	5	0.1043	1.07	0.0973	1.072
E-50-R2	6	6	0.0642	0.713	0.0900	0.713
E-50-R2	1	7	0.0494	0.550	0.0898	0.550
E-50-R2	2	7	0.0362	0.403	0.0889	0.407
E-50-R2	3	7	0.0427	0.475	0.0863	0.495
E-50-R2	4	7	0.0395	0.439	0.0875	0.451
E-50-R2	7	7	0.0270	0.286	0.0943	0.287
E-50-R3	1	1	0.0614	0.700	0.0875	0.701
E-50-R3	2	2	0.0753	0.838	0.0898	0.838
E-50-R3	1	3	0.1070	1.22	0.0875	1.22
E-50-R3	3	3	<RQ	<RQ	0.0886	<RQ
E-50-R3	2	4	0.1077	1.20	0.0898	1.20
E-50-R3	4	4	0.114	1.28	0.0889	1.28
E-50-R3	5	5	0.0564	0.629	0.0895	0.629
E-50-R3	6	6	0.0724	0.825	0.0877	0.825
E-50-R3	1	7	0.0624	0.713	0.0875	0.713
E-50-R3	2	7	0.0646	0.738	0.0898	0.720
E-50-R3	3	7	<RQ	<RQ	0.0886	<RQ
E-50-R3	4	7	0.0438	0.500	0.0889	0.492
E-50-R3	7	7	0.0559	0.644	0.0867	0.644
Mean			0.0737	0.828	0.0893	0.826
Stdev			0.0272	0.306		0.304
E-200-R1	1	1	0.115	1.30	0.353	0.325
E-200-R1	2	2	0.124	1.41	0.351	0.352
E-200-R1	1	3	0.117	1.33	0.353	0.332
E-200-R1	3	3	0.111	1.30	0.343	0.324
E-200-R1	2	4	0.159	1.82	0.351	0.454

SRNL-STI-2008-00399, Revision 0
 SRNS-STI-2008-00120, Revision 0

Sample	★ First Day (F)	Second Day (S)	Measured Ammonia in Sample (mg)	Measured Ammonia in Sample (mg/L)	Ammonia at Equilibrium (mg)	Measured / Equilibrium
E-200-R1	4	4	0.138	1.60	0.344	0.401
E-200-R1	5	5	0.154	1.69	0.365	0.423
E-200-R1	6	6	0.120	1.31	0.367	0.328
E-200-R1	1	7	0.100	1.13	0.353	0.282
E-200-R1	2	7	0.101	1.14	0.351	0.287
E-200-R1	3	7	0.104	1.18	0.343	0.303
E-200-R1	4	7	0.077	0.867	0.344	0.223
E-200-R1	7	7	0.121	1.30	0.373	0.325
E-200-R2	1	1	0.101	1.15	0.351	0.287
E-200-R2	2	2	0.126	1.42	0.357	0.355
E-200-R2	1	3	0.113	1.29	0.351	0.323
E-200-R2	3	3	0.125	1.49	0.335	0.374
E-200-R2	2	4	0.158	1.78	0.357	0.444
E-200-R2	4	4	0.169	1.91	0.355	0.478
E-200-R2	5	5	0.146	1.69	0.345	0.423
E-200-R2	6	6	0.157	1.78	0.353	0.445
E-200-R2	1	7	0.082	0.935	0.351	0.234
E-200-R2	2	7	0.075	0.860	0.357	0.212
E-200-R2	3	7	0.062	0.711	0.335	0.186
E-200-R2	4	7	0.101	1.15	0.355	0.284
E-200-R2	7	7	0.114	1.28	0.357	0.321
E-200-R3	1	1	0.103	1.21	0.341	0.302
E-200-R3	2	2	0.106	1.16	0.366	0.290
E-200-R3	1	3	0.151	1.77	0.341	0.442
E-200-R3	3	3	0.043	0.467	0.365	0.117
E-200-R3	2	4	0.142	1.55	0.366	0.388
E-200-R3	4	4	0.155	1.83	0.338	0.458
E-200-R3	5	5	0.114	1.25	0.364	0.314
E-200-R3	6	6	0.126	1.41	0.357	0.354
E-200-R3	1	7	0.122	1.43	0.341	0.357
E-200-R3	2	7	0.070	0.823	0.366	0.192
E-200-R3	3	7	0.089	1.04	0.365	0.243
E-200-R3	4	7	0.098	1.15	0.338	0.291
E-200-R3	7	7	0.082	0.935	0.351	0.234
Mean			0.115	1.30	0.352	0.326
Stdev			0.030	0.34		0.085
E-0-R1	1	1	0.0818	0.923	NA	NA
E-0-R1	2	2	0.0705	0.809	NA	NA
E-0-R1	1	3	0.0862	0.971	NA	NA
E-0-R1	3	3	0.0673	0.760	NA	NA
E-0-R1	2	4	0.0866	0.994	NA	NA
E-0-R1	4	4	0.0814	0.905	NA	NA
E-0-R1	5	5	0.0912	1.04	NA	NA
E-0-R1	6	6	0.0445	0.506	NA	NA
E-0-R1	1	7	0.0515	0.581	NA	NA
E-0-R1	2	7	0.0384	0.432	NA	NA
E-0-R1	3	7	0.1181	1.33	NA	NA
E-0-R1	4	7	0.0800	0.902	NA	NA
E-0-R1	7	7	0.0677	0.741	NA	NA
Mean			0.0742	0.838	NA	NA
Stdev			0.0205	0.232	NA	NA

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 SRNS-STI-2008-00120, Revision 0

Sample	★ First Day (F)	Second Day (S)	Measured Ammonia in Sample (mg)	Measured Ammonia in Sample (mg/L)	Ammonia at Equilibrium (mg)	Measured / Equilibrium
T-50-R1T		1	0.888		0.0902	9.85
T-50-R1T		2	1.016		0.0902	11.26
T-50-R1T		4	1.711		0.0902	18.96
T-50-R1T		7	2.110		0.0902	23.4
T-50-R2T		1	0.833		0.0902	9.23
T-50-R2T		2	0.759		0.0902	8.42
T-50-R2T		4	1.851		0.0902	20.52
T-50-R2T		7	2.333		0.0902	25.86
T-50-R3T		1	0.644		0.0882	7.30
T-50-R3T		2	0.508		0.0882	5.76
T-50-R3T		4	1.891		0.0882	21.43
T-50-R3T		7	2.241		0.0882	25.40
Mean			1.40			
Stdev			0.68			
T-50-R1H		1	0.341	3.77	0.0902	3.78
T-50-R1H		2	0.169	1.87	0.0902	1.88
T-50-R1H		4	0.314	3.48	0.0902	3.48
T-50-R1H		7	0.361	4.00	0.0902	4.00
T-50-R2H		1	0.264	2.99	0.0882	2.99
T-50-R2H		2	0.119	1.34	0.0882	1.35
T-50-R2H		4	0.190	2.15	0.0882	2.15
T-50-R2H		7	0.070	0.789	0.0882	0.79
T-50-R3H		1	0.043	0.491	0.0870	0.49
T-50-R3H		2	<RQ	<RQ	0.0870	<RQ
T-50-R3H		4	0.079	0.910	0.0870	0.91
T-50-R3H		7	0.047	0.541	0.0870	0.54
Mean			0.182	2.03		2.03
Stdev			0.121	1.34		1.34
T-200-R1T		1	1.80		0.367	4.91
T-200-R1T		2	0.83		0.367	2.26
T-200-R1T		4	1.93		0.367	5.25
T-200-R1T		7	3.24		0.367	8.81
T-200-R2T		1	1.25		0.363	3.45
T-200-R2T		2	0.83		0.363	2.30
T-200-R2T		4	2.20		0.363	6.06
T-200-R2T		7	3.21		0.363	8.84
T-200-R3T		1	1.54		0.361	4.25
T-200-R3T		2	0.78		0.361	2.17
T-200-R3T		4	2.26		0.361	6.25
T-200-R3T		7	2.95		0.361	8.17
Mean			1.90			
Stdev			0.90			

SRNL-STI-2008-00399, Revision 0
SRNS-STI-2008-00120, Revision 0

Sample	★ First Day (F)	Second Day (S)	Measured Ammonia in Sample (mg)	Measured Ammonia in Sample (mg/L)	Ammonia at Equilibrium (mg)	Measured / Equilibrium
T-200-R1H		1	0.194	2.11	0.367	0.528
T-200-R2H		1	0.028	0.302	0.367	0.075
T-200-R3H		1	0.098	1.06	0.367	0.266
T-200-R1H		2	0.213	2.31	0.367	0.579
T-200-R2H		2	0.507	5.59	0.363	1.399
T-200-R3H		2	0.239	2.63	0.363	0.659
T-200-R1H		4	0.562	6.20	0.363	1.551
T-200-R2H		4	0.661	7.29	0.363	1.824
T-200-R3H		4	0.113	1.25	0.361	0.313
T-200-R1H		7	0.036	0.403	0.361	0.101
T-200-R2H		7	0.102	1.12	0.361	0.281
T-200-R3H		7	0.168	1.86	0.361	0.464
Mean			0.243	2.68		0.670
Stdev			0.214	2.36		0.590
LE-200	1	1	0.324	3.32	0.390	0.830
LE-200	1	3	0.0490	0.503	0.390	0.126
LE-200	1	6	<RQ	<RQ	0.390	<RQ
LE-200	2	2	0.0618	0.650	0.380	0.163
LE-200	2	4	0.0268	0.282	0.380	0.070
LE-200	2	7	<RQ	<RQ	0.380	<RQ
LE-200	3	3	0.0565	0.597	0.378	0.149
LE-200	3	7	<RQ	<RQ	0.378	<RQ
Mean			0.104	1.070		0.268
Stdev			0.124	1.265		0.316
LT-200T		1	0.152		0.366	0.415
LT-200T		2	1.191		0.366	3.26
LT-200T		4	1.457		0.366	3.98
LT-200T		7	1.422		0.366	3.89
Mean			1.055			
Stdev			0.614			
LT-200H		1	0.571	6.24	0.366	1.56
LT-200H		2	0.367	4.01	0.366	1.003
LT-200H		4	0.330	3.61	0.366	0.903
LT-200H		7	0.268	2.93	0.366	0.734
Mean			0.384	4.20		1.050
Stdev			0.131	1.43		0.358
E-STD-R1		1	0.175	1.79	0.1540	1.14
E-STD-R1		3	0.029	0.290	0.1540	0.185
E-STD-R1		5	<RQ	<RQ	0.1540	<RQ
E-STD-R1		7	<RQ	<RQ	0.1540	<RQ
E-STD-R2		1	<RQ	<RQ	0.1565	<RQ
E-STD-R2		3	<RQ	<RQ	0.1565	<RQ
E-STD-R2		5	<RQ	<RQ	0.1565	<RQ
E-STD-R2		7	<RQ	<RQ	0.1565	<RQ
Mean			0.102	1.038		0.662
Stdev			0.104	1.06		0.674

SRNL-STI-2008-00399, Revision 0
 SRNS-STI-2008-00120, Revision 0

Sample	★ First Day (F)	Second Day (S)	Measured Ammonia in Sample (mg)	Measured Ammonia in Sample (mg/L)	Ammonia at Equilibrium (mg)	Measured / Equilibrium
T-STD-R1T		1	0.287		0.1469	1.95
T-STD-R1T		3	0.138		0.1469	0.94
T-STD-R1T		5	0.621		0.1469	4.23
T-STD-R1T		7	0.669		0.1469	4.55
T-STD-R2T		1	0.295		0.1394	2.12
T-STD-R2T		3	0.116		0.1394	0.83
T-STD-R2T		5	0.570		0.1394	4.09
T-STD-R2T		7	0.351		0.1394	2.52
Mean			0.381			
Stdev			0.201			
T-STD-R1H		1	0.439	4.39	0.1469	2.98
T-STD-R1H		3	0.351	3.51	0.1469	2.39
T-STD-R1H		5	0.206	2.06	0.1469	1.40
T-STD-R1H		7	0.224	2.24	0.1469	1.52
T-STD-R2H		1	0.0259	0.273	0.1394	0.186
T-STD-R2H		3	0.0226	0.238	0.1394	0.162
T-STD-R2H		5	0.0228	0.240	0.1394	0.163
T-STD-R2H		7	0.0299	0.315	0.1394	0.214
Mean			0.165	1.66		0.181
Stdev			0.166	1.65		1.12

Distribution (electronic)

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R.E. Eibling
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