## University of Alberta

# Dynamic Column Investigation of Composite or Consolidated Tailings Release Water Treatment by Mature Fine Tailings 

by
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## To My Wife


#### Abstract

The dynamic column tests were conducted to investigate the physical, chemical, and microbiological processes in mature fine tailings over composite tailings or consolidated tailings (CT) co-disposal systems. For the four testing systems, unamended CT water, CT water amended with $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}, \mathrm{SO}_{4}{ }^{2-}$, or acetate, or West-In Pit water was fed into the columns at two flowrates, approximately $6 \mathrm{~mL} / \mathrm{h}$ and $20 \mathrm{~mL} / \mathrm{h}$. This study showed that different removal efficiencies for $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ were observed from the water in the testing columns. Cation exchange between $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}$ and $\mathrm{Na}^{+}$was identified to be an important mechanism. Most probable number results suggested that sulphate-reducing bacteria outcompeted methanogens in the presence of high $\mathrm{SO}_{4}{ }^{2-}$ concentrations. The flowrates affected the removal efficiencies in the dynamic columns. Low flowrates seemed more favorable for achieving higher removal efficiencies. Therefore, it is recommended that the low flowrates should be further investigated using a flow control pump to achieve more accurate flow rates.


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## TABLE OF CONTENTS

Chapter 1 ..... 1
Introduction ..... 1
1.1 Statement of Problem ..... 1
1.2 Objectives of Research ..... 3
1.3 Thesis Organization ..... 4
Chapter 2 ..... 5
Literature Review ..... 5
2.1 Oil Sands Industry ..... 5
2.1.1 Oil Sands Processing ..... 5
2.1.2 Wastes in Oil Sands Processing ..... 7
2.1.3 Oil Sands Industry in Alberta ..... 8
2.2 Wastes Management in Oil Sands Industry ..... 10
2.2.1 Fine Tailings Management and Challenges ..... 10
2.2.2 Available Alternatives for Fine Tailings Management ..... 12
2.3 MFT ..... 14
2.3.1 Physical Properties of MFT ..... 15
2.3.2 Chemical Properties of MFT ..... 16
2.4 CT ..... 19
2.5 Microbiology in MFT and CT ..... 23
2.6 Summary ..... 27
Chapter 3 ..... 29
Materials and Methods ..... 29
3.1 Dynamic Column Tests ..... 29
3.1.1 Materials ..... 30
3.1.2 Dynamic Column Design ..... 30
3.1.3 Experimental Design. ..... 34
3.1.4 Dynamic Column Sampling. ..... 37
3.1.4.1 Release water sampling ..... 37
3.1.4.2 MFT sampling for MPN analysis ..... 38
3.1.4.3 Freezing and sacrificial sampling of MFT ..... 38
3.1.5 Analysis of MFT Samples from Dynamic Columns ..... 40
3.1.6 Eh Measurement on MFT ..... 40
3.1.7 Sulfide Analysis in Pore Water and Release Water Sample ..... 41
3.1.8 MPN Analysis on MFT. ..... 41
3.1.9 AVS Analysis on MFT Samples ..... 42
3.2 Jar Tests ..... 43
3.3 Static Barrel Mesocosms ..... 44
3.3.1 Experimental Design. ..... 44
3.3.2 Barrel Mesocosms Design and Setup ..... 45
3.3.3 Incubation and Regular Monitoring. ..... 48
3.3.4 Sampling from Barrels ..... 48
Chapter 4 ..... 51
Results and Discussion ..... 51
4.1 Dynamic Column Tests ..... 51
4.1.1 Physical and Chemical Characteristics in System 1 ..... 51
4.1.1.1 Tracer study to characterize the hydraulic properties of System 1 ..... 51
4.1.1.2 Calcium and magnesium concentration analysis ..... 52
4.1.1.3 Sodium concentration analysis ..... 56
4.1.1.4 Sulfate analysis ..... 58
4.1.1.5 Conductivity analysis ..... 59
4.1.1. 6 pH analysis ..... 61
4.1.2 Physical and Chemical Characteristics in System 2 ..... 62
4.1.2.1 Conductivity analysis ..... 63
4.1.2.2 Calcium and magnesium concentration analysis ..... 64
4.1.2.3 Sodium concentration analysis ..... 68
4.1.2.4 Sulfate analysis ..... 70
4.1.2.5 pH analysis ..... 72
4.1.3 Physical and Chemical Characteristics in System 3 (Addition of $\mathrm{Ac}^{-}$) ..... 73
4.1.3.1 Tracer Bromide analysis ..... 73
4.1.3.2 Calcium and magnesium analysis ..... 73
4.1.3.3 Sodium concentration analysis ..... 78
4.1.3.4 Sulfate analysis ..... 79
4.1.3.5 pH analysis ..... 81
4.1.3.6 Conductivity analysis ..... 82
4.1.4 Physical and Chemical Characteristics in System 4 (WIP Water with $\mathrm{Br}^{\circ}$ as Tracer) ..... 84
4.1.4.1 Bromide concentration analysis ..... 84
4.1.4.2 Calcium and magnesium concentration analysis ..... 85
4.1.4.3 Sodium concentration analysis ..... 88
4.1.4.4 Sulfate analysis ..... 90
4.1.4.5 Conductivity analysis ..... 92
4.1.4.6 pH analysis ..... 92
4.1.5 Microbiological Characteristics by MPN and AVS Analyses in all Systems ..... 94
4.1.5.1 MPNs of Methanogens and SRB ..... 94
4.1.5.2 AVS analysis ..... 99
4.2 Jar Test ..... 100
4.3 Barrel Mesocosms ..... 102
4.3.1 Calcium and Magnesium Concentrations and pH ..... 102
4.3.2 Sulfate Analysis ..... 105
4.3.3 Alkalinity Analysis ..... 106
4.3.4 Sodium Ions and Potassium Ions ..... 107
4.3.5 Solids Properties ..... 107
Chapter 5 ..... 110
Conclusions and Recommendations ..... 110
References ..... 113

## LIST OF TABLES

Table 3.1 CT Water and MFT Baseline Sample Notation ..... 30
Table 3.2 Experimental Matrix for Dynamic Column Test ..... 36
Table 3.3 Dynamic Column Sampling Notation ..... 39
Table 3.4 Experimental Matrix for Jar Test. ..... 44
Table 3.5 Jar Sample Notation ..... 44
Table 3.6 Static Barrel Mesocosm System ..... 45
Table 3.7 Notations of Barrel Samples ..... 50
Table 4.1 AVS in Jars and Dynamic Columns ..... 101
Table 4.2 Major Results of Chemical Analysis of MFT Porewater Samples for all Jars
after 128-d Incubation ..... 101
Table 4.3 pH and Concentrations of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ in Barrels ..... 103
Table 4.4 Comparisons of pH and Concentrations of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ between Barrels and Static Columns ..... 104
Table 4.5 $\mathrm{SO}_{4}{ }^{2-}$ Concentrations in the Fractions of Barrels and Static Columns. ..... 105
Table 4.6 Alkalinities in the Fractions of Barrels and Static Columns. ..... 106
Table 4.7 Concentrations of $\mathrm{Na}^{+}$and $\mathrm{K}^{+}$in the Fractions of Barrels and Static Columns107
Table 5.1 Summary of Cations and Anions Removal Efficiency ..... 110

## LIST OF FIGURES

Figure 2.1 Schematic flow diagram of oil sands processes. ..... 6
Figure 3.1 Schematics of dynamic column test system ..... 31
Figure 3.2 Top cover of dynamic column ..... 33
Figure 3.3 Plastic carboy ..... 34
Figure 3.4 Dynamic columns ..... 35
Figure 3.6 Static barrel mesocosms. ..... 46
Figure 3.7 Lids for barrels in System 5 containing CT beneath MFT ..... 47
Figure 3.8 Lids for barrels in System 6 and 7, only MFT or CT, respectively ..... 47
Figure 3.9 Barrels Sampling Using Acrylic Tubing. ..... 49
Figure 4.1 $\mathrm{Br}^{-}$concentration as a function of time in the release water of System 1dynamic columns: (a) at a high flowrate of feed water $(16.5 \mathrm{~mL} / \mathrm{h}$ inColumn DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in ColumnDL-1), where the dashed line represents the $\mathrm{Br}^{-}$concentration in the feedwater.......................................................................................................... 52
Figure 4.2 $\mathrm{Ca}^{2+}$ concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathrm{Ca}^{2+}$ concentration in the feed
$\qquad$
Figure $4.3 \quad \mathrm{Mg}^{2+}$ concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in

Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathrm{Mg}^{2+}$ concentration in the feed water 55

Figure 4.4 $\mathrm{Na}^{+}$concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathrm{Na}^{+}$concentration in the feed water 57

Figure $4.5 \quad \mathrm{SO}_{4}{ }^{2-}$ concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water $(16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feed water 59

Figure 4.6 Conductivity as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the conductivity in the feed water

Figure $4.7 \quad \mathrm{pH}$ as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water $(5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1)............... 62

Figure 4.8 Conductivity as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in

Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the conductivity in the feed water

Figure $4.9 \quad \mathrm{Ca}^{2+}$ concentration as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the $\mathrm{Ca}^{2+}$ concentration in the feed water 66

Figure $4.10 \quad \mathrm{Mg}^{2+}$ concentration as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water $(16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water $(4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the $\mathrm{Mg}^{2+}$ concentration in the feed water 67

Figure $4.11 \mathrm{Na}^{+}$concentration as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the $\mathrm{Na}^{+}$concentration in the feed
$\qquad$
Figure $4.12 \quad \mathrm{SO}_{4}{ }^{2-}$ concentration as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water $(16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feed water 71

Figure 4.13 pH as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2) 72

Figure 4.14 $\mathrm{Br}^{-}$concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathrm{Br}^{-}$concentration in the feed water 74

Figure $4.15 \quad \mathrm{Ca}^{2+}$ concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathrm{Ca}^{2+}$ concentration in the feed water 75

Figure $4.16 \quad \mathrm{Mg}^{2+}$ concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathrm{Mg}^{2+}$ concentration in the feed water 76

Figure 4.17 $\mathrm{Na}^{+}$concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathrm{Na}^{+}$concentration in the feed water

Figure $4.18 \quad \mathrm{SO}_{4}{ }^{2-}$ concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water $(18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feed
$\qquad$

Figure 4.19 pH as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water $(6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3) 82

Figure 4.20 Conductivity as a function in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water $(6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3) 83

Figure 4.21 $\mathrm{Br}^{-}$concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water $(19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathrm{Br}^{-}$concentration in the feed water 85

Figure $4.22 \mathrm{Ca}^{2+}$ concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathrm{Ca}^{2+}$ concentration in the feed water86

Figure $4.23 \quad \mathrm{Mg}^{2+}$ concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in

Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathrm{Mg}^{2+}$ concentration in the feed water 88

Figure 4.24 $\mathrm{Na}^{+}$concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathrm{Na}^{+}$concentration in the feed water 89

Figure $4.25 \quad \mathrm{SO}_{4}{ }^{2-}$ concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feed water 91

Figure 4.26 Conductivity as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the conductivity in the feed water

Figure 4.27
pH as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4);
(b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4) 94
Figure 4.29 MPN of methanogens in high flowrate systems after 60 days of incubation97
Figure 4.30 MPN of methanogens in low flowrate systems after 140 days of incubation ..... 97
Figure 4.31 MPN of SRB in high flowrate systems after 60 days of incubation ..... 98
Figure 4.32 MPN of SRB in low flowrate systems after 140 days of incubation ..... 99
Figure 4.33 Distribution of particle size in different fractions of B1 ..... 108
Figure 4.34 Distribution of particle size in different fractions of B3 ..... 109
Figure 4.35 Distribution of particle size in different fractions of B4 ..... 109

## LIST OF APPENDICES

Appendix A Dynamic System Results. ..... 118
A1 Results of chemical analysis in DH-1 system (sample type: RelW) ..... 119
A2 Results of chemical analysis in DH-2 system (sample type: RelW). ..... 122
A3 Results of chemical analysis in DH-3 system (sample type: RelW). ..... 125
A4 Results of chemical analysis in DH-4 system (sample type: RelW) ..... 128
A5 Results of chemical analysis in DL-1 system (sample type: RelW) ..... 131
A6 Results of chemical analysis in DL-2 system (sample type: RelW) ..... 136
A7 Results of chemical analysis in DL-3 system (sample type: RelW) ..... 140
A8 Results of chemical analysis in DL-4 system (sample type: RelW) ..... 146
A9 Results of baseline chemical analysis for dynamic system. ..... 152
A10 Results of baseline MFT chemical analysis ..... 153
A11 Results of chemical analysis in pore water of D3 \& D4 systems ..... 154
A12 Results of chemical analysis in Jars ..... 155
A13 MPN data of DH \& DL systems ..... 156
A14 MPN data of Jars ..... 157
A15 Results of $\mathrm{S}^{2-}$ analysis in dynamic system (MFT). ..... 157
A16 Results of $\mathrm{S}^{2-}$ analysis in D-3 system ..... 158
A17 Results of $\mathrm{S}^{2-}$ analysis in D-4 system ..... 159
A18 Results of $\mathrm{S}^{2-}$ analysis in Jars. ..... 159
A19 Results of Redox potential analysis in dynamic system ..... 160
A20 Results of Redox potential analysis in Jars ..... 160
A21 Solids content of D3 \& D4 systems ..... 161
A22 Solids content of Jars ..... 161
A23 Volume of CT water flow through D1 column (high flowrate) ..... 162
A24 Volume of CT water flow through D2 column (high flowrate) ..... 164
A25 Volume of CT water flow through D3 column (high flowrate) ..... 166
A26 Volume of CT water flow through D4 column (high flowrate) ..... 168
A27 Volume of CT water flow through D1 column (low flowrate) ..... 170
A28 Volume of CT water flow through D2 column (low flowrate) ..... 174
A29 Volume of CT water flow through D3 column (low flowrate) ..... 178
A30 Volume of CT water flow through D4 column (low flowrate) ..... 182
A31 Change of MFT height in DH system ..... 186
A32 Change of MFT height in DL system ..... 188
A32 Change of MFT height in DL system (cont'd) ..... 189
A32 Change of MFT height in DL system (cont'd) ..... 190
A32 Change of MFT height in DL system (cont'd) ..... 191
Appendix B Jar Test and Static Barrel Mesocosms Results ..... 192
B1 Results of chemical analysis in barrel and C1 system ..... 193
B2 MPN data of barrels \& C1 system ..... 195
B3 Results of $\mathrm{S}^{2-}$ analysis in barrels \& C 1 system ..... 195
B4 Results of Redox potential analysis in barrels ..... 196
B5 Results of Redox potential analysis in C1 system ..... 196
B6 Solids content of barrels \& C1 system ..... 197

## LIST OF ABBREVIATIONS

| $\mathrm{Ac}^{-}$ | acetate |
| :---: | :---: |
| AVS | acid volatile sulfides |
| BDL | below detection limit |
| CT | composite tailings or consolidated tailings |
| EC | electrical conductivity |
| Eh | redox potential |
| g | gram |
| $g$ | gravity |
| $\dot{\mathrm{G}} \mathrm{C}$ | gas chromatography |
| H | height |
| h | hour |
| ID | inside diameter |
| L | liter |
| M | mole per liter |
| MFT | mature fine tailings |
| mg | milligram |
| mL | milliliter |
| MPN | most probable number |
| NST | non-segregating tailings |
| OD | outside diameter |
| ORP | oxidation - reduction potential |

SRB sulfate-reducing bacteria
$t_{\text {incu }} \quad$ incubation time
$t_{R} \quad$ retention time
WIP West-In Pit

## Chapter 1

## Introduction

### 1.1 Statement of Problem

As the demand for oil increases, oil sands production is becoming more and more important for the oil industry. Many oil companies, such as Syncrude Canada Ltd., Suncor Energy Inc. and Shell Canada Ltd., have increased their investments in the oil sands industry to expand oil production. However, the oil industry brings not only billions of dollars of profits, but also environmental issues to the province of Alberta, especially issues related to wastewater and tailings produced from oil sands activities.

Some of the oil sands in northeastern Alberta, Canada, are strip mined and bitumen is separated from sand and clay by an alkaline hot water extraction process (the Clark Hot Water Extraction Process) leaving millions of cubic meters of tailings for disposal (Fedorak et al. 2003). In this hot water extraction process, the froth that is produced by hot water extraction process usually contains about $60 \mathrm{wt} \%$ bitumen, 30 $\mathrm{wt} \%$ water and $10 \mathrm{wt} \%$ solids (Bruce 2006). After bitumen is separated from the froth, the remaining solids and tailings are pumped into tailings ponds. Although the sand portion of the tailings settles rapidly and forms a beach, the fine clay minerals do not settle rapidly (Kasperski 1992). The tailings, a mixture of solids, water, salt, and unrecovered bitumen, are stored in large setting basins called tailings ponds (MacKinnon 1989). By gravitational settling, the coarse sands settle to the bottom of the tailings ponds. The remaining part of the tailings pond consists of tailings liquids, a mixture of clay and silt particles, inorganic and organic compounds, residual bitumen, and water. With time,
these fine tailings dewater and densify in the tailings ponds. After a few years of relatively rapid densification, a dense mature fine tailings (MFT) zone will form at the bottom of the tailings ponds and the release water may be reused in the extraction process (List and Lord 1997). The MFT has a solid content of greater than $30 \mathrm{wt} \%$ however the dewatering and densification of MFT under natural conditions is very slow and it may take hundreds of years for MFT to be consolidated into a trafficable consistency (FTFC 1995).

Any tailings must be held on site because the companies operate under a zerodischarge policy (Fedorak et al. 2003). So the large volume of MFT presents a unique challenge to oil sands companies. To overcome this problem, the oil sand companies have proposed and researched the CT technology (Composite Tailings at Syncrude Canada Ltd. and Consolidated Tailings at Suncor) since 1990 (FTFC 1995; List and Lord 1997), to increase the MFT densification rate and reduce the inventory of existing MFT. In the CT process, MFT is mixed with gypsum $\left(\mathrm{CaSO}_{4}{ }^{2} \mathrm{H}_{2} \mathrm{O}\right)$, which promotes the clay particles to aggregate and the slurry viscosity to increase. Thus, the fine solids and coarse solids stay together at deposition, creating non-segregating tailings (NST) (Matthews et al. 2000). The solids content of the CT can increase from $60 \mathrm{wt} \%$ initially to approximately $75 \mathrm{wt} \%$ within a few weeks or months, because the CT deposit starts dewatering immediately as the slurry is deposited. CT will become trafficable within a few years, leading to a dry landscape at site restoration (Luo 2004).

As a result of the high gypsum dosage ( $1000 \mathrm{mg} / \mathrm{L}$ as an optimum) (Matthews et al. 2000), the CT release water contains high concentrations of calcium $\left(\mathrm{Ca}^{2+}\right)$ and sulfate $\left(\mathrm{SO}_{4}{ }^{2-}\right)$ (MacKinnon et al. 2000). If this water is recycled for use in the extraction process,
these high $\mathrm{Ca}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ can decrease bitumen recovery and cause other process problems. To avoid any detriment to bitumen recovery efficiency and plant integrity, the oil sands companies have been examining various release water treatment options and process modifications to reduce the $\mathrm{Ca}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ concentrations in the recycled release water. One such process modification is the co-disposal of CT and MFT, with the CT being placed beneath MFT. This CT beneath MFT deposition is expected to take advantage of physical, chemical, and anaerobic microbial processes occurring in the MFT to remove, or at least reduce, the high concentrations of $\mathrm{Ca}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ in the CT release water due to ion exchange and SRB changing $\mathrm{SO}_{4}{ }^{2-}$ to $\mathrm{S}^{2-}$. At Syncrude Canada Ltd., a pilot-scale field demonstration test was conducted in 1995 to study CT beneath MFT deposition. The test showed that the CT beneath MFT deposition initiated changes that were beneficial to tailings disposal, with respect to the release water quality and densification rates of both MFT and CT (Shaw et al. 2001). However, prior to adopting the CT beneath MFT deposition scheme as a viable full-scale tailings disposal and management alternative, the physical, chemical, and microbial processes should be investigated and better understood (Shaw et al. 2001).

### 1.2 Objectives of Research

The focus of this research was to investigate, in a laboratory setting, the CT beneath MFT deposition as a possible process modification to improve the quality of release water in terms of $\mathrm{Ca}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ concentrations. Previously, bench-scale column tests were conducted in the Department of Civil \& Environmental Engineering at University of Alberta, to monitor the release water composition and densification of

MFT and CT in static CT beneath MFT systems (Luo 2004). As a continuation of this work, the present study will focus on dynamic column studies to mimic the in-situ conditions in tailings ponds and to investigate the processes occurring in this co-disposal system as the CT release water flows through the MFT layer.

The overall objectives of this project were to conduct dynamic column tests under various laboratory conditions to:
(a) demonstrate whether the disposal of CT beneath MFT is a feasible technology for CT release water treatment, that is for removing or reducing $\mathrm{Ca}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ concentrations in the CT release water by ion exchange and by SRB activity reducing $\mathrm{SO}_{4}{ }^{2-}$ to $\mathrm{S}^{2-}$, and
(b) analyze the physical, chemical, and microbial changes occurring in both the CT and MFT as a result of CT release water flowing through MFT.

The expected study results will lead to a better understanding of these processes and hence the feasibility, in terms of capacity and kinetics, of this CT beneath MFT disposal as a method to improve release water quality.

### 1.3 Thesis Organization

The thesis first presents a literature review in Section 2. Section 3 then describes the materials and methodology used the experiments including the equipment set up, experimental procedures, experimental condition controls, and sample analyses. The experimental results and discussions are presented in Section 4. The conclusions and recommendations for future study are finally summarized in Section 5.

## Chapter 2

## Literature Review

### 2.1 Oil Sands Industry

### 2.1.1 Oil Sands Processing

Oil sands are geological formations that consist of a mixture of bitumen, formation water, coarse sands, and fine particles (clays and silts). Oil sands can vary in grade, fines or clay content, mineralogy, and salt content. Typical oil sands ore contains an average of approximately $10 \mathrm{wt} \%$ of bitumen with the remainder being coarse sands ( $>22 \mu \mathrm{~m}$ in diameter), fine particles ( $<22 \mu \mathrm{~m}$ in diameter), and water (FTFC 1995). The bitumen is molasses-like viscous oil that will not flow unless heated or diluted with lighter hydrocarbons.

The extraction process that is currently being used to separate the bitumen from the oil sands ore is based on the Clark Hot Water Extraction Process, which was developed by Dr. Karl Clark in the 1920s and has been proven to be an efficient and economical process of recovering the bitumen from the oil sands. The schematic flow diagram of the oil sands process, from open surface mining to synthetic crude oil production, is shown in Figure 2.1 (Bruce 2006).

In the Clark Hot Water Extraction Process, prior to bitumen separation, the oil sands are slurried and conditioned in large horizontally rotating tumblers by mixing with hot water, steam, and caustic soda $(\mathrm{NaOH})$ for less than 10 min . The conditioning is optimized at $80^{\circ} \mathrm{C}$ and pH ranging from 9.0 to 11.0 , and this conditioning is achieved by the introduction of steam and the addition of the caustic soda. At this conditioning stage,
bitumen is dislodged from the sand particles, resulting in a loose association of bitumen, sand, and water.


Figure 2.1 Schematic flow diagram of oil sands processes

After the large size particles, such as rocks and lumps of undigested oil sands and clay, are removed from the slurry via vibrating screens, the slurry is further diluted with additional quantities of hot water and pumped into the primary separation vessels, where the bitumen floats to the surface as primary froth and the coarse sands settle to the bottom as primary tailings. The middlings (from the central portion in the primary separation vessels) and the primary tailings are combined and pumped to the tailings oil recovery vessels (the secondary separation vessels) to further recover any remaining
bitumen as secondary froth.
The secondary froth, together with the primary froth, is first de-aerated and heated, then fed to the froth treatment plants. The bitumen froth contains large quantities of water and fine solids (mainly clays), which must be removed, by a froth treatment process, before proceeding to the upgrading process. During the upgrading process, bitumen is converted from the viscous, tar-like material to a low-sulfur synthetic crude oil (a light gold-colored liquid), using coking and hydrocracking technologies. The resulting synthetic crude oil is then transported by pipeline to conventional refineries for further refining.

### 2.1.2 Wastes in Oil Sands Processing

One of the major disadvantages of the Clark Hot Water Extraction Process is that large quantities of tailings are produced during the separation of bitumen from oil sands. Tailings are slurries containing water, coarse sands, fines, and unrecovered bitumen (FTFC 1995).

Due to the "zero discharge" policy, neither the tailings solids nor the released water can be discharged into the environment. Therefore, all of the tailings from the primary and secondary separation vessels and the froth treatment plant are discharged into large settling basins (i.e. tailings ponds), as the "total extraction tailings", with a solids content between 40 and $55 \mathrm{wt} \%$ (Luo, 2004). Upon deposition, most ( $>95 \%$ ) of the coarse solids (particle diameter $>22 \mu \mathrm{~m}$, also called tailings sands) segregate and settle out from the fines fraction (particle diameter $<22 \mu \mathrm{~m}$ ). Tailings sands are usually used to build dykes that surround the tailings ponds. About $50 \%$ of the fines are entrained with
the coarse solids and settle out with them. The remainder of fines (known as fine tailings), consisting mainly of slow-settling fine silt and clay particles, water, and residual bitumen, are carried into the tailings ponds as thin fine tailings, which have a solids content of about 3 to $8 \mathrm{wt} \%$ (MacKinnon and Sethi 1993). The "total extraction tailings" from the oil sands extraction process are thus segregated into tailings sands and fine tailings, which are disposed of separately.

In the tailings ponds, the thin fine tailings settle rapidly to about $20 \mathrm{wt} \%$ solids content over a period of several months, and then consolidate very slowly over 2 to 3 years to approximately $30 \mathrm{wt} \%$ solids content (Mikula et al. 1996). The fine tailings with solids content larger than $30 \mathrm{wt} \%$ are termed "mature fine tailings" (MFT). The produced surface water (i.e. the release water) in the tailings ponds is low in suspended solids and is reused as recycle water in the extraction plant (MacKinnon and Sethi 1993).

The fine tailings, resulting from the bitumen extraction process, are of great concern to the oil sands industry and environmental agencies. This concern is because the fine tailings occupy a large volume, and their densification is very slow. Therefore, since the 1990s, research has been conducted to manage and dispose of fine tailings efficiently, cost effectively and in an environmentally safe manner.

### 2.1.3 Oil Sands Industry in Alberta

Second only to the Saudi Arabia reserves, oil sands deposits in Alberta are described as "Canada's greatest buried energy treasure," and "could satisfy the world's demand for petroleum for the next century" (http://www.energy.gov.ab.ca/89.asp). The oil sands occur in Cretaceous fluvial-estuarine deposits of northeastern Alberta, covering
an area $>140,000 \mathrm{~km}^{2}$. Bitumen is also found in Devonian carbonates (most notably within the Grosmont Formation) but this bitumen has not been commercially produced (http://www.mining-technology.com/project_printable.asp?). It is estimated that the oil sands deposits in Alberta contain over 1.7 trillion barrels of bitumen, of which approximately 300 billion barrels are recoverable with existing technologies. Canada's energy supply can be secured for more than 200 years if the oil sands in Alberta are fully developed (Syncrude 2000a and 2000b).

Annual oil sands production is growing steadily as the industry matures. According to the Canadian Association of Petroleum Producers, in 2005 industry investment in Alberta oil sands totaled approximately $\$ 10$ billion. The marketable oil sands production increased to 966,000 barrels per day in 2005. With anticipated growth, this level of production could reach 3 million barrels per day by 2020 and possibly even 5 million barrels per day by 2030 (http://www.energy.gov.ab.ca/89.asp).

Syncrude Canada Ltd. is now the world's largest producer of crude oil (known as Syncrude Sweet Blend) from oil sands. Syncrude Canada Ltd. - a joint venture of oil and gas companies mining the Athabasca oil sands - holds eight leases covering 258,000 hectares, 40 km north of Fort McMurray. Ranked as the world largest producer of synthetic crude from oil sands - and the biggest single source in Canada - nearly 95 million barrels of the company's "Syncrude Sweet Blend" were shipped in 2006. The consortium runs three separate mines - the original Base Mine and the North Mine, both near Mildred Lake on Lease 17, together with the Aurora Mine some 35 km to the north of Fort McMurray. The upgrader facility, also located on Lease 17, treats oil sands from all three mines (http://www.ags.gov.ab.ca/activities/cbm/alberta_oil_sands.html).

Syncrude Canada Ltd. and Suncor Energy Inc. are currently mining the oil sands using the surface mining operation, and extracting and upgrading the bitumen from the Athabasca oil sands deposit. The two companies produce over 500,000 barrels of crude oil daily from about 1 million tons of ore processed (Luo 2004), providing approximately $20 \%$ of the oil supply for Canada (Fedorak et al. 2003).

### 2.2 Wastes Management in Oil Sands Industry

### 2.2.1 Fine Tailings Management and Challenges

It is of great importance to manage the by-products of oil sands industry, namely, the tailings. The tailings ponds have to be reclaimed once the mining company's leases expire. Fine tailings pose a big challenge to the industry in terms of disposal technologies and management optimization.

Fine tailings management and disposal are creating two challenges for the oil sands industry: (i) the slow self-weight densification rate of the fine tailings and (ii) the large volumes of fine tailings.

The fine tailings have a slow densification rate due to the nature of the fine particles (mainly kaolinite and illite clays) that make up these tailings and due to the chemistry of the porewater found in the tailings. The high $\mathrm{Na}^{+}$content of the fine tailings porewater will cause clay particles to repel each other and remain in suspension in the tailings water, thus prevent the fine tailings from settling. After 2 to 3 years of relatively rapid initial settling and densification, the fine tailings will reach a solid content of approximately $30 \mathrm{wt} \%$ and become MFT. Further densification of the MFT is much slower. It is estimated that the natural densification of MFT to a trafficable surface would
take hundreds of years (FTFC 1995). An empirical equation for describing the solids content increase of fine tailings over time under self-weight conditions, derived from tailings pond data from Syncrude Canada Ltd. is as follows(Sworska et al. 2000):

$$
\begin{equation*}
S_{\mathrm{c}}=18.6+6.35 \ln y \tag{2-1}
\end{equation*}
$$

where $S_{\mathrm{c}}$ is the fine solids content ( $\mathrm{wt} \%$ ) and $y$ is the number of years of settling.
Based on the properties of fine tailings, some studies have investigated the use of coagulation or flocculation to improve the aggregation and settling of tailings. These studies involve taking the tailings and using coagulation or flocculation to thicken them into a paste (Sworska et al. 2000). The tests were carried out using a high molecular weight polyacrylamide flocculant and the effects of pH , polymer dosage and presence of divalent cations on the flocculation of tailings was investigated. The results showed that the addition of the flocculant led to the development of a bimodal size distribution of particles consisting of flocs and dispersed fine particles under alkaline conditions. It was concluded that flocculation was more efficient in the presence of divalent cations over the pH range studied.

The second challenge posed by fine tailings management and disposal is the production and continuous accumulation of large volumes of fine tailings. Because of the high water content in the fine tailings, the fine tailings retain fluid characteristics and must be stored behind geotechnically secure dykes with little possibility of being used as a solid matrix for reclamation. The accumulation and disposal of the fine tailings have been on going since 1967 at Suncor's site and since 1978 at Syncrude Canada Ltd.'s site. Due to "zero-discharge" policy, the fine tailings are stored in large tailings ponds on-site. Consequently, a large inventory (more than 500 million $\mathrm{m}^{3}$ ) of fine tailings has been
accumulated in the tailings ponds at Syncrude Canada Ltd. and Suncor Energy Inc., near Fort McMurray, Alberta (Liu and Fang 1995). The Mildred Lake Settling Basin is Syncrude Canada Ltd.'s largest settling basin and covers about $25 \mathrm{~km}^{2}$, with a water surface area of about $12 \mathrm{~km}^{2}$. Suncor's four tailings ponds cover a total area of about 13 $\mathrm{km}^{2}$, with a water surface area of about $7 \mathrm{~km}^{2}$ (FTFC 1996). Fine tailings are being accumulated at a rate of approximately 20 million $\mathrm{m}^{3} /$ year (Liu and Fang 1995).

Storage and disposal of the large volumes of fine tailings are considered to be the major environmental challenge facing the surface mining oil sands industry (AERCB 1984). Thus, the oil sands companies, government, and research institutions make considerable efforts to identify suitable technologies for reclaiming the existing fine tailings inventories as well as reducing the future volumes of fine tailings that are being continuously produced (FTFC 1995).

### 2.2.2 Available Alternatives for Fine Tailings Management

The fine tailings management and reclamation programmer must meet the following requirements (FTFC 1995):
(i) the direct contact of fine tailings with the environment and the release of contaminants into the environment are restricted;
(ii) the reclaimed area is stable, productive, and biologically self-sustaining.

There are a variety of approaches to dealing with tailings problems (Kasperski 1992) including:
(i) designing retention ponds to store all tailings produced;
(ii) developing physical, mechanical, chemical, biological or geotechnical methods to improve the tailings settling characteristic;
(iii) modifying the oil sands extraction process to minimize tailings production;

Each approach has its advantages and disadvantages. No single reclamation technique can handle the volumes of fine tailings in a technically sound, environmentally friendly, and economically acceptable manner. In fact, the reclamation of the fine tailings requires integrating different techniques. From an overall point of view, the reclamation of the fine tailings will be accomplished through a combination of "dry" and "wet" landscape techniques (FTFC 1995).

The dry approach involves dewatering the fine tailings or incorporating the fine tailings with a solid material so that the deposits are claimed as a land surface suitable for terrestrial vegetation. The dry landscape options include: (i) dewatering of the fine tailings through processes such as evaporation and freeze-thaw; (ii) incorporation of fine tailings with overburden clays; and (iii) creating NST or CT (FTFC 1995). The wet approach maintains the fluid character of MFT in aquatic ecosystems. It is achieved by resorting to a lake system, whereby the fine tailings are capped with a layer of water to be isolated from direct contact with the surrounding environment (FTFC 1995). The objective of this reclamation option is to produce a stable, productive, and self-sustaining cap water zone, which can support viable aquatic ecosystems shortly after water capping. The retention of MFT as a fluid will require stable secure holding areas, whereas terrestrial landscapes may allow for easier reclamation.

In practice, the final strategy will depend on a variety of considerations including availability of economically sound technologies, environmental assessments,
and stakeholder acceptance. Thus, one must have a clear idea whether the goal is to provide maximum amount of recycle water or to minimize the sludge volume, even though these objectives do not have to be exclusive. Also, it is obvious that the proposed methods will be put into use only if they are scientifically, technically, economically and environmentally sound.

To better understand the alternatives in fine tailings management, the physical and chemical properties of MFT and CT will be discussed in the following sections.

### 2.3 MFT

After bitumen extraction, tailings are pumped into tailings ponds or settling basins where the sand fraction settles, and most of the aqueous slurry of fines (silts, clays and residual hydrocarbon) slowly densifies to a suspension called MFT (Siddique et al. 2006), with characteristics of low permeability and slow densification or consolidation rate. Among all the geotechnical and physical MFT properties, solids component plays the most important role. The rate of solids settling and consolidation determines the capacity and lifetime of the containment ponds.

The MFT from the Clark Hot Water Extraction Process have been characterized by a variety of methods (Kasperski 1992; Mikula et al. 1995; Morgenstern and Scott 2000). The common results reveal that the bulk geotechnical and physical properties of the MFT are determined by the MFT component interactions at the microscopic level. Two outstanding issues regarding the structure and properties of MFT are highlighted by the research sponsored by the Fine Tailings Fundamentals Consortium (FTFC): (i) the relative importance of mineral or organic fractions in determining MFT properties, and
(ii) the gel versus floc structure of MFT. The studies have found that the MFT can be described as a suspension whose settled volumes and water holding capacity are determined by the mineral components rather than by the residual bitumen (FTFC 1995).

### 2.3.1 Physical Properties of MFT

The structure of MFT can be explained by two models (FTFC 1995): (i) the bitumen model and (ii) the mineral model. In the bitumen model, the slow densification of MFT slurry is attributed to the presence of residual bitumen and soluble organic surfactants. These components bind the clay particles into a stable aggregate structure that is slow to dewater and consolidate. The mineral model attributes the water holding capacity and stability of MFT to the presence and interaction of fine mineral clays. The clay particles interact with each other and form a three-dimensional gel-like network structure (a "card house-like" structure) in the presence of electrolytes. Other MFT components such as bitumen, water and coarser solids, are trapped in the network. A definite structure of clay mineral flocs or aggregates was observed using cryogenic electron microscopy (Mikula et al. 1996). Consequently, the densification rate of MFT is controlled by the permeability and compressibility of these mineral flocs. Studies since 1991 have favored the mineral model as a model to account for MFT stability (Mikula et al. 1996).

According to the mineral model of MFT structure and stability, the bulk volume and water holding capacity of MFT are determined by the mineral components (FTFC 1995). The amount of ultra-fines in the particle diameter range of 0.2 to $0.3 \mu \mathrm{~m}$ (which originates from the oil sands ores) in MFT contributes to more than $90 \%$ of its water
holding capacity (FTFC 1995). Furthermore, the bulk properties of MFT are a function of the chemistry of the porewater (FTFC 1995). Generally, the MFT from the Clark Hot Water Extraction Process is a weakly flocculated suspension of sodium-rich clays that densifies slowly by self-weight settling.

Tang (1997) reviewed and examined the MFT microstructure using scanning electron microscopy. The MFT has a highly dispersed three-dimensional gel structure with clay particles aligned in edge-to-edge and edge-to-face patterns. The porewater chemistry of the MFT, including pH , alkalinity, salinity, ionic content, and organic matter, may contribute to the formation and stability of the MFT structure. The residual bitumen in MFT could also reduce the MFT hydraulic conductivity, and hence reduce the MFT consolidation rate. The high water holding capacity and slow densification rate of MFT may be explained by this gel structure.

### 2.3.2 Chemical Properties of MFT

Most of the waste materials existing in tailings are derived from the oil sands themselves. The majority of fine mineral solids in the MFT are clays ( $<2 \mu \mathrm{~m}$ in particle diameter), the remainder being silts (from 2 to $22 \mu \mathrm{~m}$ in particle diameter) and fine sands (FTFC 1995). Residual bitumen content is typically in the range of 2 to $10 \mathrm{wt} \%$ (weight of bitumen per weight of dry minerals) (FTFC 1995).

The major anions and cations contained in MFT porewater are chloride $\left(\mathrm{Cl}^{-}\right)$, sulfate $\left(\mathrm{SO}_{4}{ }^{2-}\right)$, bicarbonate $\left(\mathrm{HCO}_{3}{ }^{-}\right)$, sodium $\left(\mathrm{Na}^{+}\right)$, potassium $\left(\mathrm{K}^{+}\right)$, magnesium $\left(\mathrm{Mg}^{2+}\right)$ and calcium $\left(\mathrm{Ca}^{2+}\right)$. Sodium is the predominant cation, accounting for approximately $95 \%$ of the cation equivalents (ion concentration divided over ion equivalent weight) in
the MFT porewater. Most of the $\mathrm{Na}^{+}$comes from the process chemicals. The other cations $\left(\mathrm{K}^{+}, \mathrm{Mg}^{2+}, \mathrm{Ca}^{2+}\right)$ are minor components that represent only about $5 \%$ of the total cation equivalents (MacKinnon 1989).
$\mathrm{HCO}_{3}{ }^{-}$is the most abundant anion and accounts for 50 to $75 \%$ of the anion equivalents. The pH of the MFT porewater ranges from 7.3 to 8.3 (Luo 2004) and thus the dissolved inorganic carbon is present predominantly as $\mathrm{HCO}_{3}{ }^{-} . \mathrm{SO}_{4}{ }^{2-}$ concentrations are affected by microbial processes. In anaerobic environments, $\mathrm{SO}_{4}{ }^{2-}$ is reduced to $\mathrm{S}^{2-}$ by sulfate-reducing bacteria (SRB). Both $\mathrm{Na}^{+}$and $\mathrm{Cl}^{-}$ions are conservative species since their concentration in MFT porewater are not substantially affected by microbiological or chemical interactions (MacKinnon 1989).

The concentrations of nutrients such as nitrite, nitrate, and orthophosphate in the MFT porewater are low. Most of the nitrogen is present as ammonia in the range of 2 to $6 \mathrm{mg} / \mathrm{L}$ (MacKinnon 1989).

The concentrations of dissolved trace metals in fine tailings water are low relative to regulatory guidelines for the protection of freshwater aquatic biota and have been maintained quite constant over time. Usually only aluminum $\left(\mathrm{Al}^{3+}\right)$, barium $\left(\mathrm{Ba}^{2+}\right)$, boron $\left(\mathrm{B}^{3+}\right)$, iron $\left(\mathrm{Fe}^{2+}\right)$, molybdenum $\left(\mathrm{Mo}^{6+}\right)$, strontium $\left(\mathrm{Sr}^{2+}\right)$, and zinc $\left(\mathrm{Zn}^{2+}\right)$ have concentrations above $0.1 \mathrm{mg} / \mathrm{L}$. The trace metal concentrations in the MFT do not appear to be high enough to be detrimental to the environmental acceptability of water quality (MacKinnon 1989).

The major hydrocarbon components of MFT include unrecovered bitumen and naphtha. The concentrations of water-soluble organics released during the hot water extraction process in both the surface water and porewater of the tailings ponds are fairly
low, within the range of 55 to $85 \mathrm{mg} / \mathrm{L}$ (FTFC 1995). Acid-extractable organics account for most of the acute toxicity in tailings pond recycle water. Up to $95 \%$ of the total acid fraction extractable from MFT is composed of naphthenic acids, which have been shown to be toxic to a variety of organisms (FTFC 1995, Clemente and Fedorak 2005).

Chemical, physical, and toxicological properties of the fine tailings and its porewater are a function of the source and composition of the oil sands ores. In addition, both the make-up water used for the extraction process and the chemicals added as process aids during the extraction process can add extra inorganic and organic compounds to the fine tailings stream (FTFC 1995). The composition of the water in the tailings ponds is not constant. Because all the process water and wastes are stored in the tailings ponds and the released water is recycled, the ions in the water of tailings ponds accumulate. Over time, the recycle water from the tailings ponds becomes more brackish (Mikula et al. 1996).

In summary, MFT are a weakly flocculated system of fine clays with a floc or aggregate structure that is not able to support larger particles that could stress the floc structure to initiate consolidation (FTFC 1995). The water holding capacity of the MFT is greatly controlled by the mineral composition and the floc or aggregate mineral structure. Changing the MFT floc structure is the basic mechanism for the creation of NST and other MFT treatment processes. Furthermore, biological activity has been shown in fine tailings and thus it provides an option for reclamation of fine tailings, which will be further discussed in Section 2.5.

### 2.4 CT

The fine tailings and MFT pose the biggest environmental challenge to the oil sands companies in reclaiming the disturbed areas and tailings ponds due to their large volumes and fluid characteristics that require a geotechnically stable enclosure (Luo 2004). As a result, CT process became an attractive tailings management approach. The CT process resorts to chemical amendments to combine the clays and fines in the MFT or thickened tailings with the coarser sand components, and consequently create a NST mixture that will rapidly consolidate (Mikula et al. 2004). NST are a mixture of fines and coarse sand particles which settle simultaneously to form a uniform deposit. The introduction of coarse solids imparts an internal stress that significantly enhances the densification rate of the NST (Luo 2004). Also, the presence of chemical additives (coagulants) is required to produce a non-segregating behavior. The commonly used chemical additive is gypsum. The addition of calcium in the form of gypsum has been known to be an effective coagulant in modifying the MFT properties (FTFC 1995).

Historically, the term of CT has been widely used in place of NST since 1995 (Shaw et al. 1996). Large-scale field tests of the CT approach were started in 1993 at Suncor Energy Inc. and in 1995 at Syncrude Canada Ltd. (List and Lord 1997). In 1996, Suncor Energy Inc. began to use CT technology based on gypsum treatment on a commercial scale to reduce the large volume of fine tailings in its tailings ponds. In 1997 and 1998, Syncrude Canada Ltd. successfully demonstrated a full-scale prototype operation of CT test using gypsum as the coagulant and the results of this work suggested that over $50 \%$ of the extraction tailings could be handled using the CT process (MacKinnon et al. 2000).

Factors affecting the segregation of the CT mixture include total solids content (density), fines content (a fraction of the total solids), particle size gradation, and type and dosage of coagulant aids. These factors can be manipulated individually or in combination to shift the segregation boundaries (Matthews et al. 2000).

Three means of slurry manipulation have been evaluated during the development of the CT process; (i) increasing in solids content by hydrocyclone densification of the extraction tailings, (ii) increasing fines content through enrichment with MFT, and (iii) adjusting the slurry properties through the addition of coagulants (Matthews et al. 2000). Without the addition of chemical coagulants, the tailings would exhibit a gap-graded particle size distribution and lead to the segregation of the fines from the coarse solids during discharge and deposition (Luo 2004). The addition of coagulant aids to the CT mixture (consisting of coarse tailings and MFT) is an essential component of the CT process. Hence, a variety of coagulant aids were assessed, including sulfuric acid, lime $\left(\mathrm{CaO}, \mathrm{Ca}(\mathrm{OH})_{2}\right)$, acid-lime $\left(\mathrm{H}_{2} \mathrm{SO}_{4}-\mathrm{CaO}\right)$, gypsum, sodium aluminate $\left(\mathrm{Na}_{2} \mathrm{Al}_{2} \mathrm{O}_{3}\right)$, alum $\left(48 \% \mathrm{Al}_{2}\left(\mathrm{SO}_{4}\right)_{3} \cdot 14.3 \mathrm{H}_{2} \mathrm{O}\right)$, and organic polymers (polyacrylamides) based on the segregation characteristics, deposit dewatering and densification rates, released water quality, and economics (Luo 2004). The results demonstrated that gypsum is a robust, effective, easy to handle, and readily available coagulant aid to the CT process (Matthews et al. 2000). More recent research investigated the effect of carbon dioxide addition on the strength of the MFT or fluid tailings component and found that carbon dioxide could be a useful CT or NST alternative (Mikula et al. 2004).

Long et al. (2006) measured the forces between a clay fine or silica particle and
a silica wafer in aqueous solutions using an atomic force microscope, in order to provide fundamental insights into the treatment of oil sand tailings. The effect of polymer dosage, solution pH , and addition of calcium and magnesium ions on the interaction and adhesion forces was studied (Luo 2004). The synergy of the polymer and divalent ions significantly enhanced the adhesion between fine solids. The measured adhesion forces correlated well with settling characteristics, that is, the stronger the adhesion, the higher the initial settling rate. This study provided a potential new technology for oil sand tailings treatment using the synergic effect of polymers and divalent cations.

In the CT mixture, most of the fines and porewater come from the MFT. This contribution of MFT is an important factor when predicting the quality of CT release water. The optimum solids content of CT is about $60 \mathrm{wt} \%$ (Luo 2004). When the CT slurry is deposited and starts settling, the CT deposit experiences two stages of densification: the first stage of initial settling and the second stage of long-term selfweight consolidation. The initial settling starts immediately upon deposition of the CT mixture. The volume of the CT deposit decreases substantially with the releasing of its porewater. As the CT deposit densifies to a solids content of $75 \mathrm{wt} \%$ in weeks or months, approximately $50 \%$ of the initial water can be released. The rate of the initial settling depends on the amount and type of fines, chemical addition, mixing procedure, and the hydraulic conductivity of MFT. The long term consolidation of CT begins as the sand grains become in contact with one another and form a sand matrix. Excess water is slowly released from the CT as it consolidates over a long-term. If allowed to dewater and drain, the CT deposit will become trafficable in a few years. (MacKinnon et al. 2000; Tang 1997).

The resulting CT and release waters from the CT process are saline-sodic, with $\mathrm{Na}^{+}, \mathrm{SO}_{4}{ }^{2-}$, and $\mathrm{Cl}^{-}$being the dominant ions. When freshly deposited, the CT deposits are too soft for access by reclamation equipment, and the time required for these deposits to release sufficient water to support traffic is uncertain. A study was conducted by Renault et al. (2003) to determine the suitability of barley (Hordeum vulgare L.) for reclamation of fresh CT deposits and to evaluate benefits of peat amendments. The results showed that amendment of CT with peat improved germination, survival, and growth of barley, but did not prevent leaf injury (probably due to $\mathrm{Na}^{+}$and $\mathrm{Cl}^{-}$and possibly multiple nutrient deficiencies). Therefore, Renault et al. (2003) suggest that field studies should be undertaken to validate their greenhouse results suggesting that barley could be used to improve dewatering of the freshly deposited substrates, reduce soil erosion, and facilitate leaching of ions by root penetration into the substrate.

The CT process produces composite tailings sand, which is a new challenging material for reclamation work. The soil remediation methods show that a reclaimed growing medium should support a healthy plant community that will evolve toward an ecosystem comparable to that of neighboring natural areas. Therefore, Jack pine (Pinus banksiana Lamb.), hybrid poplar (Populus deltoides Bartr. ex Marsh. $\times$ Populus nigra L.) and red clover (Trifolium pratense L.) plants were used in an 8 -week greenhouse bioassay to evaluate the mycorrhizal inoculum potential of CT (Bois et al. 2005). The results showed that CT was devoid of active mycorrhizal propagules. However, on hostile substrates such as saline alkaline CT , controlled inoculation of seedlings in the nursery with selected strains of mycorrhizal fungi could compensate for the low natural
inoculum potential and improve survival and growth of tree seedlings after outplanting (Bois et al. 2005).

In summary, the CT process aims at retaining and consuming the MFT with the coarse solids to eventually create a sustainable and acceptable dry or wet landscape (MacKinnon et al. 2000). It enables the oil sands companies to create broadly diverse landscapes that help fulfill the commitment to the reclamation of the disturbed areas.

### 2.5 Microbiology in MFT and CT

Biodegradation of the residual hydrocarbons (bitumen) in tailings is of great importance for managing oil sands tailings in the long term. MFT and CT are rich in microorganisms (Fedorak et al. 2002). Fedorak et al. (2002) examined three MFT and four CT samples from three oil sands extractions companies and found that each one contains methanogens and SRB.

Active methanogenesis has been found in the Mildred Lake Settling Basin, operated by Syncrude Canada Ltd. (Holowenko 2000). The production of methane may have detrimental effects on the reclamation of the tailings ponds by affecting the tailings settling behavior, producing fugitive emission of low molecular hydrocarbons, and leading to anaerobic conditions in the cap water (Fedorak et al. 2000). Recently, an unexpected increase in the rate of densification of MFT accompanying the microbially mediated production of methane $\left(\mathrm{CH}_{4}\right)$ was reported by Fedorak et al. (2003). The rapid densification observed in methanogenic MFT suggests that microbial activity is able to enhance densification (Fedorak et al. 2003).

Dissimilatory sulfate-reduction is another important biodegradation process
occurring in tailings ponds, in which sulfate is reduced to sulfide by SRB coupled to the oxidation of organic matter (Levett 1991).

$$
\left.\begin{array}{l}
2 \mathrm{CH}_{2} \mathrm{O}_{(\mathrm{aq})}+\mathrm{SO}_{4}^{2-}{ }_{(\mathrm{aq})} \longrightarrow 2 \mathrm{HCO}_{3}^{-}(\mathrm{aq})
\end{array} \mathrm{S}_{(\mathrm{aq})}^{2-}+2 \mathrm{H}^{+}{ }_{(\mathrm{aq})}\right)
$$

where $\mathrm{CH}_{2} \mathrm{O}$ represents the organic substrates. Sulfide is the metabolic product of dissimilatory sulfate reduction and will precipitate in the presence of many metal ions. Hence, SRB activity can be identified by the precipitation of black FeS (Levett 1991).

Microbial activity in a given environment will change the chemical and physical nature of that environment. Due to the release of $\mathrm{CO}_{2}$ and the accumulation of carbonate or bicarbonate from the sulfate reduction process, the aqueous environment will have an elevated alkalinity and metal ions will precipitate as insoluble metal sulfides (e.g. FeS). If no metals are present or if the pH is low enough, the sulfide may be converted to hydrogen sulfide $\left(\mathrm{H}_{2} \mathrm{~S}\right)$ and remain dissolved in the water phase or escape to the atmosphere (Gray 1989).

Sulfate reduction will consume a substantial amount of organic substrates and produce, sometimes via acetate, $\mathrm{CO}_{2}$ (Postgate 1984). Given a high ratio of metabolisable carbon to sulfate, SRB can deplete an environment of sulfate almost completely (Postgate 1984). The disappearance of sulfate in nature over time may be used as an index of the SRB activity (Levett 1991; Postgate 1984).

Sulfite and thiosulfate are intermediates in the normal sulfate reduction process, and can also be used as substitute electron acceptors for the growth and carbon metabolism of some SRB species (Levett 1991; Postgate 1984).

Methanogens and SRB are two important microorganisms affecting the
reclamation of the tailings ponds and MFT slurry (Fedorak et al. 2000).
Methanogens are a morphologically diverse group of anaerobic bacteria unified by their ability to produce methane (Levett 1991). Methanogens grow on a limited number of simple carbon compounds as substrates, including $\mathrm{CO}_{2}, \mathrm{H}_{2}$, formate, methanol, and acetate (Levett 1991). More than half of the biogenic methane in nature is believed to originate from acetate (Postgate 1984). Methanogens are usually considered to be the last players in the decomposition of organic matters in anaerobic ecosystems. That is, they consume the metabolic end products (mainly acetate and $\mathrm{H}_{2}$ ) of other strictly anaerobic bacteria as energy sources and produce $\mathrm{CH}_{4}$ as their waste product. $\mathrm{CO}_{2}$ and $\mathrm{HCO}_{3}{ }^{\circ}$ serve as the terminal electron acceptors during anaerobic respiration by methanogens (Fedorak et al. 2000).

Methanogens are found in a variety of anaerobic habitats including sediments, sludge, and animal waste digesters. They require redox potential (Eh) in the range of -150 to -220 mV to thrive (Fedorak et al. 2000). In general, the presence of oxygen inactivates methanogens, but not every species of methanogen is rapidly killed by oxygen (Levett 1991).

The most common method used to enumerate specific types of methanogens is the most probable number (MPN) technique, where the sample is serially diluted and inoculated into a suitable broth medium with a specific substrate (Levett 1991). The positive and negative tubes (caused by presence or absence of methanogen growth) are scored based on the production of $\mathrm{CH}_{4}$. After incubation, the pattern of positive and negative tubes is checked against the standardized MPN table to determine the MPN of organisms per unit volume of the sample.

SRB also need strict anaerobic conditions for growth. The redox potential (Eh) of the growth media should be around -100 mV (Levett 1991; Postgate 1984). However, some SRB can survive long exposures to oxygen and become active again if the environment becomes anaerobic. SRB can tolerate temperatures from -5 to $+75^{\circ} \mathrm{C}$ and pH values ranging from 5 to 9.5 (Postgate 1984).

The complex interactions between SRB and methanogens have been well studied and involve competition for substrates. Like methanogens, SRB can also use acetate and $\mathrm{H}_{2}$ as energy sources. However, SRB use a wider range of organic compounds as their substrates compared to methanogens (Fedorak et al. 2000). Sulfate is used as the terminal electron acceptor. Therefore, sulfate amendment may stimulate the activity of SRB (Salloum et al. 2002; Fedorak et al. 2002).

SRB and methanogens are strict anaerobes bacteria. Studies have shown that SRB can outcompete methanogens for common substrates, $\mathrm{H}_{2}$ and acetate, except in some particular environments with sulfate deficiency (Postgate 1984). Based on thermodynamic considerations, the utilization of $\mathrm{H}_{2}$ or acetate by SRB yields more energy than the utilization of these substrates by methanogens. Thus in CT, high sulfate concentrations, resulting from the gypsum addition, has the potential to stimulate SRB to out-compete methanogens and consequently cease methanogenesis (Fedorak et al. 2002). It has long been known that methane production in marine sediments occurs only after sulfate has been depleted from the porewater, i.e. methanogenesis begins when sulfate is depleted (Schlesinger 1997). When sulfate is depleted, methanogens carry out the terminal steps in the anaerobic environment (Fedorak et al. 2000). Acetate, products of sulfate reduction, would be expected to favor methanogenesis (Postgate 1984). The
complex relationship between SRB and methanogens is also illustrated by the fact that methane can serve as a substrate for anaerobic oxidation in the presence of SRB (Valentine 2002). In most natural environments there is little or no overlap between the zone of methanogenesis and the zone of sulfate reduction.

In the CT process, a gypsum dosage at about $1000 \mathrm{~g} / \mathrm{m}^{3}$ of CT mixture would result in approximately $1000 \mathrm{mg} \mathrm{SO}_{4}{ }^{2 \pi} / \mathrm{L}$ in the CT porewater (MacKinnon et al. 2000). With the high $\mathrm{SO}_{4}{ }^{2-}$ concentration in the CT porewater, the CT will inhibit methane production by creating an environment that is more suitable for SRB and the SRB would out-compete the methanogens. Fedorak et al. (2002) have shown that the addition of sulfate to fine tailings decreased methane production, and that methanogenesis appeared to have started to a large extent only after the sulfate concentration had dropped to approximately $20 \mathrm{mg} / \mathrm{L}$.

### 2.6 Summary

The oil sands industry is growing rapidly in Alberta. The two major oil sand companies, Syncrude Canada Ltd. and Suncor Energy Inc., are currently mining the oil sands using surface mining operations, and extracting and upgrading the bitumen from the Athabasca oil sands deposit. Oil sands industry operations produce large volumes of fine tailings and the storage and disposal of these tailings has become a major environmental concern currently challenging the industry. The physical and chemical properties of these fine tailings must be understood to identify alternative tailings management processes. The MFT from the Clark Hot Water Extraction Process is characterized by its low permeability and slow consolidation. The most recent studies
have found that the settled volumes and water holding capacity of MFT are determined by the mineral components rather than by the residual bitumen. The MFT has a highly dispersed three-dimensional gel structure with clay particles aligned in edge-to-edge and edge-to-face patterns due to the high water holding capacity. The slow densification rate of MFT may be attributed to this gel structure.

The CT process has been used to treat fine tailings since 1995. The CT process uses chemical amendments to combine the clays and fines in the MFT or thickened tailings with the coarser sand components to create a NST mixture that rapidly consolidates. Ultimately, the CT process can assist to create a sustainable and acceptable dry or wet landscape. The segregation of the CT mixture is affected by the total solids content, the fines content, the particle size gradation, and the type and dosage of coagulant aids.

Biodegradation of the residual hydrocarbons in the tailings is an important factor in managing tailings in the long term. In fact, microbial activity is present in the tailings ponds and the identified microorganisms that significantly affect the reclamation of the tailings are methanogens and SRB. In addition, the complex relationship between these two groups of microorganisms has been addressed by some studies.

Although the oil sand industry is looking for alternative processes to provide the maximum amount of recycle water (of good quality, that is low in $\mathrm{Ca}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ ) and/or minimize the tailings volume, the existing and continually accumulating fine tailings are a big challenge to the oil sands industry. Thus, the oil sands industry, government, and research institutions are making considerable efforts to identify suitable technologies for reclaiming the existing fine tailings inventories and managing release water for recycling.

## Chapter 3

## Materials and Methods

The experimental program consisted of three types of tests:

1. dynamic column tests
2. jar tests
3. barrel mesocosm tests

These three types of tests will be described in the following sections. The materials used in the tests as well as the design of the test apparatus will be presented. The experimental design and the analyses conducted will also be described.

### 3.1 Dynamic Column Tests

Oil sands tailings have been studied extensively in the Department of Biological Sciences and the Department of Civil \& Environmental Engineering at the University of Alberta. These tests involved extensive physical, chemical, and microbiological investigations of the MFT and CT materials using a variety of innovative experimental methods. Fedorak et al. (2000) specifically developed many proven protocols for column setup and analyses to evaluate the CT and MFT under anaerobic conditions. Luo (2004) adapted the method of Fedorak et al. (2000) and designed the static columns to study the physical, chemical, and microbiological changes of MFT and CT materials. In this project, dynamic columns were developed as a further improvement of the static columns, in order to study and monitor the physical, chemical, and microbiological changes which occurred in the release water after having passed through the MFT.

### 3.1.1 Materials

CT water, WIP water and MFT material were delivered in 20 L plastic pails from Syncrude Canada Ltd and kept in a $4^{\circ} \mathrm{C}$ temperature controlled room until use. Several pails of the MFT material and CT water were randomly selected and well mixed in a 100 L plastic container and a 200 L plastic container, respectively, in order to reduce the heterogeneity. The mixed CT water and WIP water will herein be referred to as feed water.

Baseline samples of MFT and feed water (WIP and CT water) were collected and analyzed by Syncrude Canada Ltd for physical, chemical and microbiological characteristics. The results of these analyses are described in Appendix A. The baseline sample notations for Systems 1 to 4 are listed in Table 3.1.

Table 3.1 CT Water and MFT Baseline Sample Notation

| System | CT Water Baseline Sample Notation | MFT Baseline Sample <br> Notation |
| :---: | :---: | :---: |
| 1 | D1-Base-1 |  |
|  | D1-Base-2 |  |
| 2 | D2-Base-1 |  |
|  | D2-Base-2 | D3-Base-1 |
| 4 | D3-Base-2 |  |
|  | D4-Base-1 |  |

### 3.1.2 Dynamic Column Design

The dynamic column is illustrated in Figure 3.1. An appropriate-sized segment (outside diameter $(\mathrm{OD})=157 \mathrm{~mm}$, inside diameter $(\mathrm{ID})=153 \mathrm{~mm}$; height $(\mathrm{H})=1200$ mm ) of clear acrylic tubing (Johnston Plastics Inc., Edmonton, Alberta) was used as the


Figure 3.1 Schematics of dynamic column test system
column in this project. The bottom of the column was sealed with a $220 \times 220 \times 7 \mathrm{~mm}$ square acrylic plate. A column stopper with a piece of Geo-textile (Nilex Inc., Edmonton, Alberta) was installed onto the bottom at a distance of 100 mm to the bottom acrylic plate. There was a feeding port on the wall between the column stopper and bottom acrylic plate that was used to feed CT water. Under a $\mathrm{N}_{2}$ atmosphere, the column was filled with freshly mixed MFT up to a height of 1000 mm above the column stopper, which gave a headspace height of 100 mm . After thoroughly flushing the headspace of
the MFT-filled column with $\mathrm{N}_{2}$, the column was sealed with a $220 \times 220 \times 7 \mathrm{~mm}$ square acrylic top cover. The top column cover was connected to a 1-L Tedlar ${ }^{8}$ bag (Catalog Number 231-01, Safety Instruments Ltd., Edmonton, Alberta) filled with $\mathrm{N}_{2}$. Finally, a 20-L plastic carboy of feed water was connected to the feeding port via the flow meter by plastic tubing.

In summary, there were four layers (from bottom to top) along the column:

1. Bottom feed water layer of 100 mm in height
2. MFT layer of 1000 mm in height (the total volume of $\mathrm{MFT}=18.4 \mathrm{~L}$ )
3. Release water layer
4. Headspace layer filled with $\mathrm{N}_{2}$ gas

The design of the dynamic column allowed for continuous introduction of feed water in an upflow manner through the feeding port and continuous release of water into the release water layer.

Figure 3.2 shows the top cover of the dynamic column. There were two holes in the top cover. One $3.2-\mathrm{mm}$ NPT threaded hole was used for connecting the 1-L Tedlar ${ }^{(8)}$ bag through a 250 mm long, $3.2-\mathrm{mm}$ ID and $6.4-\mathrm{mm}$ OD Tygon tubing. This Tedlar ${ }^{\circledR}$ bag was used to equilibrate the headspace pressure in case of biogenic gas evolution from the MFT. Another $3.2-\mathrm{mm}$ NPT threaded hole was used to regularly take samples of the release water using 3.2-mm ID stainless steel tubing and syringe.

The column stopper was also made of an acrylic plate with a diameter close to the column inside diameter. Thus, the column stopper would minimize MFT leaking during filling. Many small holes were drilled through the column stopper to allow the feed water to pass through. A layer of Geo-textile was put on the column stopper to
prevent the MFT from blocking these holes and allowing feed to pass through.


Figure 3.2 Top cover of dynamic column

The 20-L plastic carboy served as the reservoir for feed water (Figure 3.3). To ensure anaerobic conditions in the feed water, the reservoir was flushed with $\mathrm{N}_{2}$ and then filled with 20 L of feed water. To equilibrate the headspace, a Tedlar ${ }^{\mathbb{8}}$ bag filled with $\mathrm{N}_{2}$ was connected to the headspace of the reservoir via a hose connector fastened at the cap. The feed water was fed to the column through Tygon tubing from the port near the bottom of the carboy. The flowrate of the feed water was controlled by a combination of valves and a flow meter. Feed water was added every a few days to maintain the water level in the reservoir. The Tedlar ${ }^{\circledR 3}$ bag was also re-filled periodically with $\mathrm{N}_{2}$.


Figure 3.3 Plastic carboy

### 3.1.3 Experimental Design

The dynamic column system (see Figure 3.4) was designed to study and monitor the physical, chemical, and microbiological changes which occurred in the feed water after passing through the MFT. The experimental matrix for the dynamic column tests is presented in Table 3.2. Specifically, feed water without (System 1) or with amendments (Systems 2, 3 and 4) and a conservative tracer (bromide ( $\mathrm{Br}^{-}$), in the form of KBr ) was fed in an upwards fashion into columns filled with MFT. Each type of feed water was fed into columns at two different flowrates: a high flowrate of approximately $20 \mathrm{~mL} / \mathrm{h}$ and a low flowrate of approximately $6 \mathrm{~mL} / \mathrm{h}$. Columns at high flowrate were run for an incubation time of approximately 60 days whereas columns at low flowrate were run for an incubation time of 140 days.


Figure 3.4 Dynamic columns

For System 1, $2500 \mathrm{mg} \mathrm{Br}^{-} / \mathrm{L}$ was added into CT-water of the high flowrate system (System DH-1), and $900 \mathrm{mg} \mathrm{Br} / \mathrm{L}$ was added to CT-water for low flowrate system (System DL-1). For System 2, CT-water was amended with $\mathrm{CaSO}_{4}, \mathrm{MgSO}_{4}$ and $\mathrm{Na}_{2} \mathrm{SO}_{4}$ to achieve approximately $250 \mathrm{mg} \mathrm{Ca}^{2+} / \mathrm{L}, 250 \mathrm{mg} \mathrm{Mg}^{2+} / \mathrm{L}$ and $2500 \mathrm{mg} \mathrm{SO}_{4}{ }^{2-} / \mathrm{L}$. For System 3, CT-water was amended with $300 \mathrm{mg} \mathrm{Br} / \mathrm{L}$ and $100 \mathrm{mg} \mathrm{Ac}^{-} / \mathrm{L}$ to the high flowrate system (System DH-3), and $400 \mathrm{mg} \mathrm{Br}^{-} / \mathrm{L}$ and $100 \mathrm{mg} \mathrm{Ac}^{-} / \mathrm{L}$ to the low flowrate system (System DL-4). The feed water for System 4 was West-In Pit (WIP) water, a different type of water from other systems. This WIP water was amended with 300 mg $\mathrm{Br}^{-} / \mathrm{L}$ for the high flowrate system (System DH-4) and $400 \mathrm{mg} \mathrm{Br}^{-} / \mathrm{L}$ added to the low flowrate system (System DL-4). $\mathrm{Br}^{-}$was used as a tracer to characterize the hydraulic properties of the dynamic columns to verify that the feed water had passed through MFT layer.

Table 3.2 Experimental Matrix for Dynamic Column Test

|  |  | System 1 |  | System 2 |  | System 3 |  | System 4 |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  | High Flowrate | Low <br> Flowrate | High Flowrate | Low <br> Flowrate | High Flowrate | Low Flowrate | High Flowrate | Low Flowrate |
| Name |  | DH-1 | DL-1 | DH-2 | DL-2 | DH-3 | DL-3 | DH-4 | DL-4 |
| Base feed water |  | CT Water | CT Water | CT Water | CT Water | CT Water | CT Water | WIP | WIP |
|  | $\mathrm{Br}^{-}$ | 2500 | 900 | - | - | 300 | 400 | 300 | 400 |
|  | $\mathrm{Ca}^{2+}$ | - | - | 250 | 250 | - | - | - | - |
|  | $\mathrm{Mg}^{2+}$ | - | - | 250 | 250 | - | - | - | - |
|  | $\mathrm{SO}_{4}{ }^{\text {- }}$ | - | - | 2500 | 2500 | - | - | - | - |
|  | $\mathrm{Ac}^{-}$ | - | - | - | - | 100 | 100 | - | - |

*Note: The concentrations provided for the amendments are target concentrations and the actual concentrations were verified by analyses.

[^0]All dynamic columns were kept at room temperature $\left(20^{\circ} \mathrm{C}\right)$ in the dark for about 60 days for high flowrate columns and 140 days for low flowrate columns. The flowrates were measured and adjusted through flow meters (VACT H40407 0035, Fisher Scientific, Edmonton, AB).

### 3.1.4 Dynamic Column Sampling

At the end of the planned incubation periods, that is 60 days and 140 days for the high and low flowrate systems, respectively, the flow through the dynamic columns was stopped and the contents of the columns were analyzed. The analysis involved:

- removing the top cover of the column while maintaining anaerobic condition;
- sampling the release water,
- taking samples of the MFT material (by coring) for MPN analysis; and
- freezing and sacrificially sampling the MFT for chemical analysis.


### 3.1.4.1 Release water sampling

The feed water and release water (the pore water of MFT, and the release water) were sampled and analyzed at predetermined intervals. The maximum ion concentration in the released water was used to calculate the removal efficiency. Therefore, the removal efficiency in this study represents the minimum reduction.

The analyses include: pH , alkalinity, major cations $\left(\mathrm{Mg}^{2+}, \mathrm{Na}^{+}, \mathrm{K}^{+}, \mathrm{NH}_{4}{ }^{+}\right.$, and $\mathrm{Ca}^{2+}$ ), major anions ( $\mathrm{SO}_{4}{ }^{2-}, \mathrm{S}^{2-}, \mathrm{SO}_{3}{ }^{2-}, \mathrm{S}_{2} \mathrm{O}_{3}{ }^{2-}, \mathrm{PO}_{4}{ }^{3-}, \mathrm{NO}_{3}^{-}, \mathrm{NO}_{2}^{-}$, and $\mathrm{Cl}^{-}$), total soluble S (by ICP), and conductivity. The analysis methods are described in Section 3.1.5.

### 3.1.4.2 MFT sampling for MPN analysis

The MPN sampling used a core-sampling method under a $\mathrm{N}_{2}$ atmosphere. The MPN samples were taken from the center of the column using a suitable length piece of rigid plastic tubing (with a $6.4-\mathrm{mm}$ ID). Four MPN samples were taken from each column, that is, each column was separated to four parts in 250 mm of length and the samples were taken from middle of each part. The samples were then transferred to a 60mL syringe or a $90-\mathrm{mL}$ plastic jar filled with $\mathrm{N}_{2}$ and transported to the Department of Biological Sciences at the University of Alberta. All materials used for MPN sampling (plastic tubing, syringe, and jar) were autoclaved at $121^{\circ} \mathrm{C}$ for 20 min before use.

### 3.1.4.3 Freezing and sacrificial sampling of MFT

After the sampling of release water and MFT, the columns were placed in a freezer at a temperature of about $-20^{\circ} \mathrm{C}$ and left for several days to freeze. The column was then broken with hammer and the frozen MFT was cut into four parts of approximately 200 mm in length each. As shown in Figure 3.5, each column was separated into four samples and 20 mm was cut off from either side in each part and discarded. Table 3.3 provides a summary of the sample (with sample notation) taken after sacrificial sampling of the MFT.

The frozen MFT subsamples were then placed in 4-L glass jars (filled with $\mathrm{N}_{2}$ ) at room temperature overnight. Then, subsampling of each MFT sample was performed in a $\mathrm{N}_{2}$ chamber.


Figure 3.5 Sacrificial sampling of MFT after column freezing: (a) Showing sample notation; (b) showing discarded sample (shaded areas)

Table 3.3 Dynamic Column Sampling Notation

| System | 1 | 2 | 3 | 4 |
| :--- | :--- | :--- | :--- | :--- |
| High Flow <br> Rate <br> $(20 \mathrm{~mL} / \mathrm{h})$ | DH-1-U | DH-2-U | DH-3-U | DH-4-U |
|  | DH-1-M-U | DH-2-M-U | DH-3-M-U | DH-4-M-U |
|  | DH-1-M-L | DH-2-M-L | DH-3-M-L | DH-4-M-L |
|  | DH-1-L | DH-2-L | DH-3-L | DH-4-L |
| Low Flow <br> Rate <br> $(6 \mathrm{~mL} / \mathrm{h})$ | DL-1-U | DL-2-U | DL-3-U | DL-4-U |
|  | DL-1-M-U | DL-2-M-U | DL-3-M-U | DL-4-M-U |
|  | DL-1-M-L | DL-2-M-L | DL-3-M-L | DL-4-M-L |
|  | DL-1-L | DL-2-L | DL-3-L | DL-4-L |

Note: where U refers to samples from the upper half zone and L refers to samples from the lower half zone of the MFT layer (refer to Figure 3.5).

The porewater extraction was completed using a combination of centrifugation and filtration procedures. The MFT samples or subsamples were centrifuged at 10000 rpm for 30 min , using a Sorvall RC-5B centrifuge with SS-34 rotor and eight Teflon ${ }^{\mathrm{TM}}$
tubes of 50 mL each. After centrifugation, pre-filtration using a AP 15 Millipore glass fiber filter and vacuum filtration under a $\mathrm{N}_{2}$ atmosphere was performed to remove coarse solids and bitumen. The samples were then filtered through a $0.45 \mu \mathrm{~m}$ Millipore Millex syringe filter (Millipore, Billerica, MA).

### 3.1.5 Analysis of MFT Samples from Dynamic Columns

MFT subsamples were taken under a $\mathrm{N}_{2}$ atmosphere. These samples were analyzed for redox potential, particle size distribution, AVS (acid volatile sulphide) analysis, and also pH , major cations $\left(\mathrm{Mg}^{2+}, \mathrm{Na}^{+}, \mathrm{K}^{+}, \mathrm{NH}_{4}^{+}\right.$, and $\left.\mathrm{Ca}^{2+}\right)$, major anions $\left(\mathrm{SO}_{4}{ }^{2-}, \mathrm{S}^{2-}, \mathrm{SO}_{3}{ }^{2-}, \mathrm{S}_{2} \mathrm{O}_{3}{ }^{2-}, \mathrm{PO}_{4}{ }^{3-}, \mathrm{NO}_{3}{ }^{-}, \mathrm{NO}_{2}{ }^{-}\right.$, and $\mathrm{Cl}^{-}$), total soluble S (by ICP), and conductivity, which were performed by Syncrude Analytical Laboratory at the Edmonton Research Centre, using established company protocols and standard methods (Fedorak et al. 2000).

### 3.1.6 Eh Measurement on MFT

An Accumet Model $50 \mathrm{pH} / \mathrm{Ion} /$ Conductivity Meter with a Cole-Parmer $\mathrm{Ag} / \mathrm{AgCl}$ ORP electrode was used to measure Eh. The ORP electrode was calibrated against a standard ORP solution (from Orion Research Inc., Beverly, MA) before and after the Eh measurements.

The MFT subsamples (slurry) were contained in a $120-\mathrm{mL}$ glass jar and the headspace of the jar was filled with $\mathrm{N}_{2}$ for Eh measurement. The Eh measurements took several hours to stabilize for some samples, therefore Eh measurements were taken every 1 and 2 hours.

### 3.1.7 Sulfide Analysis in Pore Water and Release Water Sample

The sulfide in the release water and porewater samples was analyzed using CHEMetrics sulfide test kit (from CHEMetrics Inc., Calverton, VA), which uses a methylene blue colorimetric method. The measured sulfide was the total acid soluble sulfides including dissolved $\mathrm{H}_{2} \mathrm{~S}, \mathrm{HS}^{-}$, and acid-soluble metallic sulfides in suspension.

A sulfide test kit with two ranges ( 0 to $1 \mathrm{mg} / \mathrm{L}$ and 1 to $10 \mathrm{mg} / \mathrm{L}$, Catalog Number K-9510 Chemetrics Inc.) was used based on the previous sulfide results in MFT and CT porewaters (Fedorak et al. 2000). The minimum detection limit of the test kit was $0.05 \mathrm{mg} / \mathrm{L}$.

The water samples for $\mathrm{S}^{2-}$ analysis were first filtered using a $0.45 \mu \mathrm{~m}$ syringe filter. Water samples were prepared with minimum aeration in a $\mathrm{N}_{2}$ atmosphere and then were analyzed immediately after the preparation to avoid or minimize the oxidation of sulfides.

### 3.1.8 MPN Analysis on MFT

The microbial MPN analyses of SRB and methanogens were conducted in the Department of Biological Science at the University of Alberta. The three-tube MPN method was used to enumerate the SRB and methanogens. Only SRB and methanogens were enumerated because these two groups of microorganisms are considered the most important groups of microorganisms in MFT (Fedorak et al. 2000).

The MPN results were expressed as "MPN/g (dry solids)". Thus unit conversion was necessary for the ratio of dry weight to wet weight of MFT samples because the MPN samples were dispensed as wet materials. The ratio of dry weight to wet weight
was determined in the Environmental Engineering Laboratory by drying the MFT samples at $104^{\circ} \mathrm{C}\left(103\right.$ to $\left.105^{\circ} \mathrm{C}\right)$ oven overnight, following the procedure given in APHA (1995). The weights of the MFT samples were measured before and after drying, and water content was then calculated.

### 3.1.9 AVS Analysis on MFT Samples

AVS are the sum of amorphous iron monosulfide (FeS), mackinawite ( $\mathrm{FeS}_{\mathrm{I}-\mathrm{x}}$ ), greigite ( $\mathrm{Fe}_{3} \mathrm{~S}_{4}$ ) (Duan et al. 1997; Morse et al. 1987), and sulfides of other metals such as ZnS and CdS (Gerard et al. 1998; Lasorsa and Casas 1996). AVS is usually determined by first liberating $\mathrm{H}_{2} \mathrm{~S}$ from the sulfides using cold acid ( 1 to 6 M HCl ), then distilling and trapping the released $\mathrm{H}_{2} \mathrm{~S}$ for quantification by one of several analytical methods. The presence of AVS is an indicator of recent $\mathrm{SO}_{4}{ }^{2-}$ reduction (Kennedy et al. 1999) because AVS species are metastable and reactive, either by oxidation or by subsequent reduction to $\mathrm{FeS}_{2}$ (pyrite). More severe acid treatment, often with heating or with the addition of other chemicals, may release sulfides from other compounds (e.g. pyrite).

The amorphous FeS and mackinawite are quantitatively recovered in cold acid extraction, but the recovery of greigite is incomplete. Under harsher conditions, such as hot HCl , or cold 6 M HCl plus $\mathrm{SnCl}_{2}$, greigite and newly formed fine-grained synthetic pyrite are recovered. $\mathrm{SnCl}_{2}$ in the HCl distillation is present to help diminish the Fe (III) interference that may oxidize sulfide to elemental sulfur ( $\mathrm{S}^{0}$ ) in acid distillation (Duan et al. 1997).

It is critical to obtain the accurate measurement of AVS by using sampling, handling, and analytical techniques that minimize oxidation of sulfides. Sulfide samples should be stored at $4^{\circ} \mathrm{C}$ or frozen at $-20^{\circ} \mathrm{C}$ and analyzed within 2 weeks of sample collection (Lasorsa and Casas 1996) when samples are handled under the $\mathrm{N}_{2}$ atmosphere.

The AVS analysis method for this project used 6 M cold HCl plus $\mathrm{SnCl}_{2}$ to digest the sample, thus, the obtained results were the AVS plus fine-grained (newly formed) synthetic pyrite. The sampling was carried out under a $\mathrm{N}_{2}$ atmosphere. The AVS samples were kept at $4^{\circ} \mathrm{C}$ under a $\mathrm{N}_{2}$ atmosphere during storage and delivery, and were analyzed by ALS Laboratory Group (Vancouver, BC) within 14 days of sample collection.

### 3.2 Jar Tests

To identify the naturally occurring changes within the MFT porewater, jar tests were used as control tests. In the jar test, the same MFT was used as in the dynamic column test. Under anaerobic conditions, 4 L MFT were added to a jar and $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ concentrations were adjusted to different levels comparable to the feed water for all dynamic column systems (See Table 3.4). The jars were stored for the same period of time as the dynamic column tests. The concentrations of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ were monitored and compared with the concentrations in the release water in the dynamic columns. The notation used for the jar test samples are given in Table 3.5.

Under a $\mathrm{N}_{2}$ atmosphere, the MFT in the jar was mixed fully, and samples were taken. All samples were centrifuged and the porewater samples were sent to Syncrude Canada Ltd. laboratory for chemical analysis (the same items with release water).

Table 3.4 Experimental Matrix for Jar Test

| Jar | Concentration | $\mathrm{Br}^{-}(\mathrm{mg} / \mathrm{L})$ | $\mathrm{Ca}^{2+}(\mathrm{mg} / \mathrm{L})$ | $\mathrm{Mg}^{2+}(\mathrm{mg} / \mathrm{L})$ | $\mathrm{SO}_{4}{ }^{2-}(\mathrm{mg} / \mathrm{L})$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| D1 | Initial | 0 | 22 | 14 | 20 |
|  | Amendments | 2500 | 0 | 0 | 1000 |
|  | Final | 2500 | 22 | 14 | 1020 |
| D2 | Initial | 0 | 22 | 14 | 20 |
|  | Amendments | 0 | 140 | 140 | 1790 |
|  | Final | 0 | 162 | 154 | 1800 |
| D3 | Initial | 0 | 22 | 15 | 20 |
|  | Amendments | 320 | 40 | 11 | 975 |
|  | Final | 320 | 62 | 26 | 995 |
| D4 | Initial | 0 | 22 | 15 | 20 |
|  | Amendments | 330 | 0 | 0 | 300 |
|  | Final | 330 | 22 | 15 | 320 |

Note: The concentrations provided for the amendments were target concentrations. The actual concentrations will be verified by analyses.

Table 3.5 Jar Sample Notation

| System | Release Water Sample Notation | MFT Sample Notation |
| :---: | :---: | :---: |
| 1 | Jar D1-1 water | Jar D1-1 |
|  | Jar D1-2 water | Jar D1-2 |
| 2 | Jar D2-1 water | Jar D2-1 |
|  | Jar D2-2 water | Jar D2-2 |
| 3 | Jar D3-1 water | Jar D3-1 |
|  | Jar D3-2 water | Jar D3-2 |
| 4 | Jar D4-1 water | Jar D4-1 |
|  | Jar D4-2 water | Jar D4-2 |

### 3.3 Static Barrel Mesocosms

### 3.3.1 Experimental Design

The static barrel mesocosms consisted of three different systems as shown in
Table 3.6. At barrel filling, System 5 consisted of two barrels, each containing a 25 L

MFT layer on the top of a 50 L CT layer, whereas Systems 6 and 7 contained only one barrel of 50 L of MFT or CT, respectively. No water was added to the barrels. Therefore, all cap water during ensuing incubation was released water.

Table 3.6 Static Barrel Mesocosm System

| System | Barrel Notation | Barrel <br> Configuration | Deposit of the Barrels |
| :---: | :---: | :---: | :---: |
| 5 | B1, B2 | MFT and CT | 25L MFT (U)+ 50L CT(L) |
| 6 | B3 | MFT only | 50 L MFT |
| 7 | B4 | CT only | 50 L CT |

The barrel mesocosms were larger in size than the static column mesocosms. The barrel mesocosm test was designed to provide supplemental information to the static column mesocosm test and to see if the size of the mesocosms would have an effect on the final interpretation of the results. Thus, the analysis of the barrel mesocosms was basically the same as in the static column mesocosms (Luo, 2004). The barrel mesocosms were sacrificially sampled at the end of 2 years of incubation.

Two static column mesocosms were also sacrificially sampled after 2 years of in cubation. These columns served as a comparison to the barrel mesocosms.

### 3.3.2 Barrel Mesocosms Design and Setup

The 110-L Ropak plastic barrels (from Great Western Container Inc., Edmonton, AB ) were used for barrel mesocosms tests (see Figure 3.6). The barrels in System 5 (two barrels) were simply filled first with a 50 L CT layer, and a piece of Geo-grid was placed on the top of the CT, then the barrels were immediately filled with a 25 L MFT layer. No barrel inverting procedure as used in Luo (2004) was used during barrel filling.


Figure 3.6 Static barrel mesocosms

As shown in Figure 3.7, there were three 3.2-mm NPT threaded holes placed in the lids for the barrels in System 5. One was used to connect a 1-L Tedlar ${ }^{(8)}$ bag through about a 250 mm length of $3.2 \mathrm{ID} \times 6.4 \mathrm{OD} \mathrm{mm}$ Tygon tubing to collect the evolved headspace gas from the barrels; one was used to fit a $3.2-\mathrm{mm}$ ID stainless steel tubing which was used to monitor the position change of the CT/MFT interface in an in-situ manner; and the third one was to fit a $3.2-\mathrm{mm}$ ID stainless steel tubing to sample the release water in an in-situ manner when needed.

For the barrels in Systems 6 (one barrel) and 7 (one barrel), there was only one $3.2-\mathrm{mm}$ NPT threaded hole placed in the lid (Figure 3.8). The hole was used to connect a 1-L Tedlar ${ }^{\circledR}$ bag through about a 250 mm length of 3.2 ID $\times 6.4 \mathrm{OD} \mathrm{mm}$ Tygon tubing to collect the evolved headspace gas in the barrels.


Figure 3.7 Lids for barrels in System 5 containing CT beneath MFT


Figure 3.8 Lids for barrels in System 6 and 7, only MFT or CT, respectively

Prior to, and during, the filling process, each barrel was flushed with $\mathrm{N}_{2}$ for several minutes. Then the lid of the barrels was clamped and tightened with a metal ring. The barrel was then flushed with $\mathrm{N}_{2}$ and the Tedlar ${ }^{\circledR}$ bag was attached. The Tedlar ${ }^{\circledR}$ bag in the barrels served as a safety valve allowing for any biogenically evolved gases to escape the barrel and be contained.

### 3.3.3 Incubation and Regular Monitoring

Because of the opaque plastic barrel material, no visual observations of the MFT and CT in the barrels were performed during the incubation. For barrels in System 5, the Geo-grid position changes were regularly recorded by contacting a $3.2-\mathrm{mm}$ ID stainless steel tubing to the Geo-grid piece. For all barrels, the Tedlar ${ }^{\circledR 8}$ bag was regularly checked to see if the Tedlar ${ }^{\circledR}$ bag was fully filled. No analysis of the gas in the Tedlar ${ }^{\circledR}$ bag was performed on the barrel mesocosms.

### 3.3.4 Sampling from Barrels

At the end of the incubation period (2 years), the barrels were sampled and analyzed. Under $\mathrm{N}_{2}$ atmosphere, eight samples were taken from each MFT or CT layer using acrylic tubes $(\mathrm{OD}=50 \mathrm{~mm}, \mathrm{ID}=40 \mathrm{~mm}$; Height $=300 \mathrm{~mm})$. The sampling locations were randomly selected, as shown in Figure 3.9. To take the sample, the acrylic tubes were pushed into the MFT or CT by hand. A rubber stopper was then placed at the bottom to secure the sample. The sample tube was then lifted off the barrel and another rubber stopper was added to the top. All the samples were then frozen at $-20^{\circ} \mathrm{C}$ for further analysis.


Figure 3.9 Barrels Sampling Using Acrylic Tubing

To release the frozen samples, the bottom rubber stopper was removed first and the sampling tube was rinsed with running tap water until the samples slid out. The frozen samples were then cut into two pieces using a knife. The upper portions of all eight samples from one layer were put together into one jar, whereas the lower portions were placed in another jar. After the samples were thawed, the material in each jar was well mixed, sub-sampled and then sent to the Syncrude Canada Ltd. laboratory for chemical analyses. The notations of barrel samples are provided in Table 3.7.

Table 3.7 Notations of Barrel Samples

| Sample | System 5 |  | System 6 | System 7 |
| :---: | :---: | :---: | :---: | :---: |
|  | Barrel 1 | Barrel 2 |  |  |
| Water | B1-release water | B2- release water | B3- release water | B4- release water |
|  | B1-MFT-U pore water | B2-MFT-L <br> pore water | B3-MFT-U pore water | B4-CT-U pore water |
|  | B1-CT-U <br> Water | B2-CT-U <br> Water | B3-MFT-L pore water | B4-CT-L pore water |
|  | B1-CT-L pore water | B2-CT-L pore water |  |  |
| MFT or CT | B1-MFT-U | B2-MFT-U | B3-MFT-U | B4-CT-U |
|  | B1-MFT-L | B2-MFT-L | B3-MFT-L | B4-CT-L |
|  | B1-CT-U | B2-CT-U |  |  |
|  | B1-CT-L | B2-CT-L |  |  |

## Chapter 4

## Results and Discussion

### 4.1 Dynamic Column Tests

Dynamic column tests involved introducing different feed waters in an upflow manner at high and low flows through columns of MFT. Release water and MFT were then analyzed for select physical, chemical and microbiological characteristics. The results of the analyses are presented in the following sections.

### 4.1.1 Physical and Chemical Characteristics in System 1

### 4.1.1.1 Tracer study to characterize the hydraulic properties of System 1

Unamended feed water was fed into System 1. A combination of valves and flow meters was employed to achieve the desired flowrates. The obtained flowrates were calculated to be on average $16.5 \mathrm{~mL} / \mathrm{h}$ and $5.3 \mathrm{~mL} / \mathrm{h}$, respectively. $\mathrm{Br}^{-}$was used as tracer and added to the feed water at $2500 \mathrm{mg} / \mathrm{L}$ for the high flowrate system and at $900 \mathrm{mg} / \mathrm{L}$ for the low flowrate system. As shown in Figure 4.1a, in the high flowrate system, $\mathrm{Br}^{-}$ was detected in the release water on Day 6 and increased steadily to reach the feed water concentration on Day 60. In the low flowrate system, $\mathrm{Br}^{-}$first appeared in the release water on Day 20 and reached the feed concentration of approximately $900 \mathrm{mg} / \mathrm{L}$ on Day 120. Although it took about 40 days and 120 days for the feed water to penetrate through the MFT layer of the high and low flowrate systems, respectively, some feed water may be released as early as Day 6 and Day 20 in these two systems accordingly due to the dispersion via preferential flow pathways.


Figure 4.1 $\quad \mathrm{Br}^{*}$ concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH1); (b) at a low flowrate of feed water $(5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathrm{Br}^{-}$concentration in the feed water

### 4.1.1.2 Calcium and magnesium concentration analysis

The original $\mathrm{Ca}^{2+}$ concentration in the MFT was $20.7 \mathrm{mg} / \mathrm{L}$ and the $\mathrm{Ca}^{2+}$ concentration in the feed water was $31.1 \mathrm{mg} / \mathrm{L}$ (see Appendix A9 and A10). The change in concentration of $\mathrm{Ca}^{2+}$ in the release water for the high and low flowrate systems are illustrated in Figure 4.2.


Figure 4.2 $\mathrm{Ca}^{2+}$ concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathbf{C a}^{\mathbf{2 +}}$ concentration in the feed water

As shown in Figure 4.2a, some data points were missing during the first 5 days because there was not enough water release water to collect a sample. Therefore, the slight drop in $\mathrm{Ca}^{2+}$ concentration from $23.7 \mathrm{mg} / \mathrm{L}$ to $17.5 \mathrm{mg} / \mathrm{L}$ observed on Day 2 was
possibly due to analytical deviation. On Day 6 , the $\mathrm{Ca}^{2+}$ concentration in the release water was consistent with the $\mathrm{Ca}^{2+}$ concentration in MFT, indicating that the release water was mainly MFT pore water. Then, the $\mathrm{Ca}^{2+}$ concentration increased steadily, reflecting the mixing effects of CT water with the MFT pore water. The $\mathrm{Ca}^{2+}$ concentration reached $49.2 \mathrm{mg} / \mathrm{L}$ at Day 40, and increased $64 \%$ within 38 days. This increased concentration indicates that the CT-water was gradually penetrating the MFT layer and moving through the MFT layer to become release water. The reason why the $\mathrm{Ca}^{2+}$ concentration was higher than the feed concentration is unknown. From Day 40 until 60 days of incubation, the $\mathrm{Ca}^{2+}$ concentration fluctuated within the range of 49.2 $\mathrm{mg} / \mathrm{L}$ and $34.5 \mathrm{mg} / \mathrm{L}$, and the average $\mathrm{Ca}^{2+}$ concentration was approximately $43 \mathrm{mg} / \mathrm{L}$.

Compared to the high flowrate system, in the low flowrate system CT water took more time to penetrate the MFT layer (Figure 4.2b). The $\mathrm{Ca}^{2+}$ concentration started to increase from about $20 \mathrm{mg} / \mathrm{L}$ on Day 8 to $30.5 \mathrm{mg} / \mathrm{L}$ on Day 39 , and then fluctuated within a narrow range (from $26 \mathrm{mg} / \mathrm{L}$ to $31 \mathrm{mg} / \mathrm{L}$ ) with an average $\mathrm{Ca}^{2+}$ concentration of $28 \mathrm{mg} / \mathrm{L}$.

For both the high flowrate system and the low flowrate system, the change of $\mathrm{Ca}^{2+}$ concentration with time can be divided into two stages. In the first stage, the $\mathrm{Ca}^{2+}$ concentration increased, which indicated that the CT-water was penetrating through the MFT layer. In the second stage, the average $\mathrm{Ca}^{2+}$ concentration fluctuated within stable ranges, indicating that the MFT pores were saturated with CT water. No apparent $\mathrm{Ca}^{2+}$ removal was observed at the end of the experiment in both high and low flowrate systems.

The original $\mathrm{Mg}^{2+}$ concentration in the MFT was $13.7 \mathrm{mg} / \mathrm{L}$, and $\mathrm{Mg}^{2+}$
concentration in the feed water was $26.4 \mathrm{mg} / \mathrm{L}$ (see Appendix A9). For the high flowrate system (Figure 4.3a), the $\mathrm{Mg}^{2+}$ concentration increased steadily from Day 6 to $28.4 \mathrm{mg} / \mathrm{L}$ on Day 39 , and remained relatively stable at the average $\mathrm{Mg}^{2+}$ concentration of $28 \mathrm{mg} / \mathrm{L}$. As shown in Figure 4.3 b, the $\mathrm{Mg}^{2+}$ in the release water of the low flowrate system


Figure $4.3 \mathbf{M g}^{2+}$ concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathbf{M g}{ }^{\mathbf{2 +}}$ concentration in the feed water

It is interesting to note that both $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ had the same concentration trend and elution timeframe in the same system. These similar profiles are most likely due to the fact that $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ have very similar physical and chemical characteristics: both can be retained by ion exchange, adsorption and chemical precipitation, and both have the same change.

### 4.1.1.3 Sodium concentration analysis

The $\mathrm{Na}^{+}$concentrations in release water for high and low flowrate systems are shown in Figure 4.4a and Figure 4.4 b , respectively. The original $\mathrm{Na}^{+}$concentration in MFT was $1040 \mathrm{mg} / \mathrm{L}$, and $\mathrm{Na}^{+}$concentration in the feed water was $1730 \mathrm{mg} / \mathrm{L}$. For the high flowrate system, the $\mathrm{Na}^{+}$concentrations were approximately $1100 \mathrm{mg} / \mathrm{L}$ during the first 5 days, consistent with the original $\mathrm{Na}^{+}$concentration in MFT. $\mathrm{Na}^{+}$then increased almost linearly until Day 40 to the concentration of $1620 \mathrm{mg} / \mathrm{L}$. From Day 41 to Day 60, the average $\mathrm{Na}^{+}$concentration was stable, close to the feed concentration. The $\mathrm{Na}^{+}$ concentrations also confirmed that the saturation time was 40 days for high flow rate system. For the low flowrate system, the variability in $\mathrm{Na}^{+}$concentrations during the first few testing days was due to difficulty in collecting sufficient sample volume. $\mathrm{Na}^{+}$ concentrations almost linearly increased to $1300 \mathrm{mg} / \mathrm{L}$ on Day 40 . From Day 41 to Day $53, \mathrm{Na}^{+}$fluctuated within the range of $1260 \mathrm{mg} / \mathrm{L}$ and $1310 \mathrm{mg} / \mathrm{L}$, with an average concentration about $1290 \mathrm{mg} / \mathrm{L}$. During Day 54 to Day $60, \mathrm{Na}^{+}$concentration fluctuated but the overall trend was decreasing. From Day 61 to Day 81 , the $\mathrm{Na}^{+}$concentration fluctuated in between $1390 \mathrm{mg} / \mathrm{L}$ and $1350 \mathrm{mg} / \mathrm{L}$, and the average concentration was about $1370 \mathrm{mg} / \mathrm{L}$. From Day 82 to Day $123, \mathrm{Na}^{+}$concentration fluctuated within a wide
range of $1370 \mathrm{mg} / \mathrm{L}$ to $1170 \mathrm{mg} / \mathrm{L}$. The fluctuation and apparent decrease in $\mathrm{Na}^{+}$ concentrations could be attributed to the change of feed water, possible ion exchange reactions in the system, or analytical variability.


Figure $4.4 \quad \mathrm{Na}^{+}$concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathrm{Na}^{+}$concentration in the feed water

Compared with the feed water $\mathrm{Na}^{+}$concentration, $\mathrm{Na}^{+}$concentration was reduced by $1.5 \%$ and $20 \%$ in the high flowrate system and low flowrate system, respectively.

### 4.1.1.4 Sulfate analysis

The original $\mathrm{SO}_{4}{ }^{2-}$ concentration in the MFT was $18 \mathrm{mg} / \mathrm{L}$, and the $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feedwater was $1060 \mathrm{mg} / \mathrm{L}$. The changes in sulfate concentrations followed the same behavior as $\mathrm{Br}^{-}$, indicating that the $\mathrm{SO}_{4}{ }^{2-}$ is not interacting with the MFT. For the high flowrate system (Figure 4.5a), from Day $6, \mathrm{SO}_{4}{ }^{2-}$ concentration started to increase to approximately $680 \mathrm{mg} / \mathrm{L}$ until Day 40 . The average $\mathrm{SO}_{4}{ }^{2-}$ concentration after Day 40 was approximately $625 \mathrm{mg} / \mathrm{L}$.

For the low flowrate system (Figure 4.5b), the initial drop of $\mathrm{SO}_{4}{ }^{2-}$ concentration (from $53 \mathrm{mg} / \mathrm{L}$ on Day 8 to $5 \mathrm{mg} / \mathrm{L}$ on Day 19) was possibly due to sampling and analytical variability. $\mathrm{SO}_{4}{ }^{2-}$ concentration increased almost linearly to 239 $\mathrm{mg} / \mathrm{L}$ from Day 20 to Day 40 and remained relatively constant until Day 60. $\mathrm{SO}_{4}{ }^{2-}$ concentration increased to $330 \mathrm{mg} / \mathrm{L}$ on Day 61 , and then fluctuated in the range of 332 $\mathrm{mg} / \mathrm{L}$ and $245 \mathrm{mg} / \mathrm{L}$ with a slight decreasing trend. From Day 82 until Day $123, \mathrm{SO}_{4}{ }^{2-}$ fluctuated over a wide range of $97 \mathrm{mg} / \mathrm{L}$ to $267 \mathrm{mg} / \mathrm{L}$ with average value of $193 \mathrm{mg} / \mathrm{L}$.

Based on the initial feed water concentration, $\mathrm{SO}_{4}{ }^{2-}$ removal efficiency in the high and low flowrate systems was $35 \%$ and $69 \%$, respectively. The mechanism for sulfate removal was possibly due to the activity of SRB, which will be further discussed in Section 4.5.


Figure 4.5 $\mathrm{SO}_{4}{ }^{\mathbf{2 -}}$ concentration as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the $\mathrm{SO}_{4}{ }^{\mathbf{2 -}}$ concentration in the feed water

### 4.1.1.5 Conductivity analysis

Conductivity is a measurement of the ability of water to pass an electrical current, which is affected mainly by the presence of charged inorganic dissolved solids such as sodium, magnesium, calcium, iron, aluminum, chloride, nitrate, sulfate, and phosphate. Organic compounds do not conduct electrical current very well and have very
low conductivity in water.
The conductivity in the release water of the high flowrate system fluctuated slightly in the range of $4450 \mu \mathrm{~S} / \mathrm{cm}$ to $4140 \mu \mathrm{~S} / \mathrm{cm}$ within the first 14 days (Figure 4.6a).


Figure 4.6 Conductivity as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1), where the dashed line represents the conductivity in the feed water

Then, the conductivity increased to $6740 \mu \mathrm{~S} / \mathrm{cm}$ at Day 42 . From then on, the conductivity varied within a small range of $6570 \mu \mathrm{~S} / \mathrm{cm}$ and $6830 \mu \mathrm{~S} / \mathrm{cm}$. The
conductivity of the low flowrate system fluctuated between $4290 \mu \mathrm{~S} / \mathrm{cm}$ and $3950 \mu \mathrm{~S} / \mathrm{cm}$ from Day 8 to Day 31 (see Figure 4.6b). Then, the conductivity increased from Day 32 to Day 123. Conductivity measurements also confirmed that the hydraulic retention time was approximately 40 and 120 days for the high and low flow rate systems, respectively.

The original conductivity of MFT was $4110 \mu \mathrm{~S} / \mathrm{cm}$, and the conductivity of the feed water was $7360 \mu \mathrm{~S} / \mathrm{cm}$. Therefore, after penetrating the MFT layer, the conductivity of both systems was reduced by $7 \%$ and $20 \%$.

### 4.1.1.6 pH analysis

Figure 4.7 a and 4.7 b present the pH for the high flowrate system and low flowrate system, respectively. As shown in Figure 4.7a, the pH of the high flowrate system increased from 8.04 on Day1 to 8.33 on Day 2; and then decreased linearly to 7.61 on Day 7. From Day 8 until Day 60, the pH varied in a range of 7.47 to 8.03 with the average value of 7.75 . For the low flowrate system, the pH fluctuated in a range of 7.39 to 8.40 with the average value of 7.80 .

It should be noted that the $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ concentrations of the low flowrate system (shown in Figure $4.2 \mathrm{~b}, 4.3 \mathrm{~b}, 4.5 \mathrm{~b}$ and 4.7 b ) also fluctuated during the period of Day 82 to Day 123. One reason for such a fluctuation of these ions concentrations might be because of the pH fluctuation. This fluctuation in pH may have affected the effectiveness of ion exchange between $\mathrm{Na}^{+}$and $\mathrm{Ca}^{2+}$ and between $\mathrm{Na}^{+}$and $\mathrm{Mg}^{2+}$ in the MFT. Moreover, the pH fluctuation might also affect the chemical precipitation of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ with $\mathrm{SO}_{4}{ }^{2-}$. Since the experimental system consisting of MFT and feed water is a very complicated system, it would be very difficult to determine
the causes of the fluctuations of $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}, \mathrm{Na}^{+}, \mathrm{SO}_{4}{ }^{2-}$ and pH without further detailed investigation.


Figure $4.7 \quad \mathbf{p H}$ as a function of time in the release water of System 1 dynamic columns: (a) at a high flowrate of feed water ( $16.5 \mathrm{~mL} / \mathrm{h}$ in Column DH-1); (b) at a low flowrate of feed water ( $5.3 \mathrm{~mL} / \mathrm{h}$ in Column DL-1)

### 4.1.2 Physical and Chemical Characteristics in System 2

For System 2, the feed water was amended so that the concentrations of $\mathrm{Ca}^{2+}$, $\mathrm{Mg}^{2+}$, and $\mathrm{SO}_{4}{ }^{2-}$ were $250 \mathrm{mg} / \mathrm{L}, 250 \mathrm{mg} / \mathrm{L}$, and $2500 \mathrm{mg} / \mathrm{L}$, respectively. The feed water
penetrated through the MFT layer at two different flow rates, and then the release water was sampled and analyzed for $\mathrm{Na}^{+}, \mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}, \mathrm{SO}_{4}{ }^{2-}, \mathrm{pH}$, and conductivity. The flowrates of the feed water were $16.9 \mathrm{~mL} / \mathrm{h}$ and $4.7 \mathrm{~mL} / \mathrm{h}$. Because no $\mathrm{Br}^{-}$was detected in the release water due to some unknown reason, the hydraulic properties of this system were characterized based on the conductivity measurement.

### 4.1.2.1 Conductivity analysis

The conductivity of the high flowrate system increased slowly from $4280 \mu \mathrm{~S} / \mathrm{cm}$ to $4450 \mu \mathrm{~S} / \mathrm{cm}$ within 20 days (Figure 4.8 a ). Then the conductivity increased to 4940 $\mu \mathrm{S} / \mathrm{cm}$ on Day 21. From then on, the conductivity increased slowly to $5190 \mu \mathrm{~S} / \mathrm{cm}$ at Day 60. It was identified from Figure 4.8a that the saturation time was approximately 36 days.

The measured conductivity of the low flowrate system increased steadily from $4020 \mu \mathrm{~S} / \mathrm{cm}$ on Day 5 to $5900 \mu \mathrm{~S} / \mathrm{cm}$ on Day 103 (Figure 4.8 b ). Figure 4.8 b shows that the increase of conductivity was almost linear. Then the conductivity dropped slightly to $5610 \mu \mathrm{~S} / \mathrm{cm}$ on Day 110. Base on Figure 4.8b, it is assumed that the hydraulic retention time was approximately 100 days for this column.

The original conductivity of MFT was $4110 \mu \mathrm{~S} / \mathrm{cm}$. The conductivity of the feed water was $5770 \mu \mathrm{~S} / \mathrm{cm}$. The highest conductivity of the release water of the high flowrate system and low flowrate system was $5200 \mu \mathrm{~S} / \mathrm{cm}$ and $5900 \mu \mathrm{~S} / \mathrm{cm}$, respectively. Therefore, the conductivity was reduced by $10 \%$ after penetrating through the high flowrate system, whereas the conductivity of the low flowrate system increased by $2 \%$, which may be in the range of experimental error.


Figure 4.8 Conductivity as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the conductivity in the feed water

### 4.1.2.2 Calcium and magnesium concentration analysis

The calcium concentration in the MFT layer before introducing feed water was
$23.1 \mathrm{mg} / \mathrm{L}$ for both the high and low flowrate columns. The actual $\mathrm{Ca}^{2+}$ concentration in the feed water was $212 \mathrm{mg} / \mathrm{L}$. As shown in Figure 4.9a, for the high flowrate system,
until Day 6 , the $\mathrm{Ca}^{2+}$ concentration in the release water was comparable to that in the MFT. Then, the $\mathrm{Ca}^{2+}$ concentration increased steadily and reached $65.0 \mathrm{mg} / \mathrm{L}(62 \%$ increases) on Day 35. This result suggests that the feed water was gradually penetrating the MFT layer and accumulating as release water. From Day 35 until 60 days, the $\mathrm{Ca}^{2+}$ concentration fluctuated within the range of $68.3 \mathrm{mg} / \mathrm{L}$ and $42.4 \mathrm{mg} / \mathrm{L}$. A high removal efficiency for $\mathrm{Ca}^{2+}$ (approximately $68 \%$ ) was achieved in this column.

Compared to the high flowrate system, the $\mathrm{Ca}^{2+}$ in the low flowrate system took longer to penetrate the MFT layer (Figure 4.9 b). The $\mathrm{Ca}^{2+}$ concentration fluctuated between $21.5 \mathrm{mg} / \mathrm{L}$ and $25.8 \mathrm{mg} / \mathrm{L}$ from Day 5 to Day 50 . The fluctuation (about 2\%) may be in the experimental error range. Moreover, the average $\mathrm{Ca}^{2+}$ concentration of $34.7 \mathrm{mg} / \mathrm{L}$ suggests that the release water before Day 50 consisted of MFT pore water. After Day 50 , the $\mathrm{Ca}^{2+}$ concentration increased faster and reached $42.4 \mathrm{mg} / \mathrm{L}$ on Day 61 , which seems to imply that the feed water was gradually penetrating through MFT layer and entered the release water. From Day 61 , the $\mathrm{Ca}^{2+}$ concentration increased from 33.0 $\mathrm{mg} / \mathrm{L}$ to $64.0 \mathrm{mg} / \mathrm{L}$. The $\mathrm{Ca}^{2+}$ removal efficiency in the low flow rate column was approximately $70 \%$.

For both the high flowrate system and the low flowrate system, the change of $\mathrm{Ca}^{2+}$ concentration with time can be divided into two stages. In the first stage, the $\mathrm{Ca}^{2+}$ concentration increased, which indicated that the feed water was penetrating through the MFT layer. During the second stage, the $\mathrm{Ca}^{2+}$ concentration fluctuated within a small range, indicating that ion exchange or some other mechanism was occurring between the feed water and the MFT and retained $\mathrm{Ca}^{2+}$ in the MFT layer.


Figure $4.9 \mathbf{C a}^{2+}$ concentration as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the $\mathrm{Ca}^{\mathbf{2 +}}$ concentration in the feed water

The original $\mathrm{Mg}^{2+}$ concentration in MFT was $15.1 \mathrm{mg} / \mathrm{L} . \mathrm{Mg}^{2+}$ in the feed water was $162 \mathrm{mg} / \mathrm{L}$. For the high flowrate system, the $\mathrm{Mg}^{2+}$ concentration increased from 17.6 $\mathrm{mg} / \mathrm{L}$ on Day 2 to $53.4 \mathrm{mg} / \mathrm{L}$ on Day 38 (roughly the hydraulic retention time), as shown in Figure 4.10a. After then, $\mathrm{Mg}^{2+}$ concentration increased from $44 \mathrm{mg} / \mathrm{L}$ to $70 \mathrm{mg} / \mathrm{L}$.


Figure 4.10 $\mathbf{M g}^{\mathbf{2 +}}$ concentration as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the $\mathbf{M g}{ }^{\mathbf{2}}$ concentration in the feed water

In comparison with the high flowrate system, the increase of the $\mathrm{Mg}^{2+}$ concentration in the release water for the low flowrate system was very slow (Figure 4.10 b ). The $\mathrm{Mg}^{2+}$ concentration was relatively constant (ranging from $14.2 \mathrm{mg} / \mathrm{L}$ to 16.7 $\mathrm{mg} / \mathrm{L}$ ) before Day 50 , suggesting the release water before Day 50 consisted mainly of MFT pore water. From Day 51 to Day 84 , the $\mathrm{Mg}^{2+}$ increased slowly from $17.1 \mathrm{mg} / \mathrm{L}$ to
$50.6 \mathrm{mg} / \mathrm{L}$, which implies that the feed water was gradually penetrating the MFT layer and accumulating as release water. After Day 84 , the $\mathrm{Mg}^{2+}$ concentration fluctuated but increased to $65.1 \mathrm{mg} / \mathrm{L}$.

The removal efficiency for $\mathrm{Mg}^{2+}$ was approximately $57 \%$ and $60 \%$ in the high and low flow rate columns, respectively.

Relatively high removal efficiency was observed for $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ in both high and low flow rate columns. The major removal mechanism was possibly ion exchange between $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ with $\mathrm{Na}^{+}$in the MFT layer.

### 4.1.2.3 Sodium concentration analysis

For the high flowrate system (Figure 4.11a), generally, the $\mathrm{Na}^{+}$concentration increased until Day 11, stabled at a plateau from Day 11 to Day 33, decreased from Day 34 to Day 49, and increased during Day 49 to Day 60.

For the low flowrate system (Figure. 4.11b), the $\mathrm{Na}^{+}$concentration can be divided into three stages: stage one was from day 5 to day 54 , where the $\mathrm{Na}^{+}$ concentration slightly increased from $1110 \mathrm{mg} / \mathrm{L}$ to $1190 \mathrm{mg} / \mathrm{L}$; stage two was from Day 55 to Day $89, \mathrm{Na}^{+}$concentration was stable at $1340 \mathrm{mg} / \mathrm{L}$; stage three was from Day 90 to Day 110 , where the $\mathrm{Na}^{+}$concentration increased from $1180 \mathrm{mg} / \mathrm{L}$ to $1320 \mathrm{mg} / \mathrm{L}$ with the trend increasing.

The original $\mathrm{Na}^{+}$concentration in MFT was $1110 \mathrm{mg} / \mathrm{L}$. The sodium concentration in the feed water was $1180 \mathrm{mg} / \mathrm{L}$. The highest $\mathrm{Na}^{+}$concentration in the release water was $1290 \mathrm{mg} / \mathrm{L}$ in the high flowrate system and $1340 \mathrm{mg} / \mathrm{L}$ in the low flowrate system. The high $\mathrm{Na}^{+}$concentration in the release water (higher than the original
concentrations in either the MFT or feed water) may be due to ion exchange between $\mathrm{Ca}^{2+}$ and $\mathrm{Na}^{+}$, or $\mathrm{Mg}^{2+}$ and $\mathrm{Na}^{+}$as expressed in the following equation:

$$
\begin{align*}
& \mathrm{Ca}^{2+}+2 \mathrm{Na}-\mathrm{clay} \longleftrightarrow 2 \mathrm{Na}^{+}+\mathrm{Ca} \text {-clay }  \tag{4-1}\\
& \mathrm{Mg}^{2+}+2 \mathrm{Na}-\mathrm{clay} \longleftrightarrow 2 \mathrm{Na}^{+}+\mathrm{Mg} \text {-clay } \tag{4-2}
\end{align*}
$$



Figure $4.11 \quad \mathrm{Na}^{+}$concentration as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the $\mathbf{N a}^{+}$concentration in the feed water

Thus, a drop in $\mathrm{Ca}^{2+}$ concentration caused by ion exchange between $\mathrm{Ca}^{2+}$ and $\mathrm{Na}^{+}$will result in an increase in the $\mathrm{Na}^{+}$concentration in the porewater and release water from MFT. The same holds true for $\mathrm{Mg}^{2+}$. Therefore a drop in $\mathrm{Ca}^{2+}$ or $\mathrm{Mg}^{2+}$ concentration due to ion exchange will result in a corresponding increase in the $\mathrm{Na}^{+}$concentration as observed here. The increase in $\mathrm{Na}^{+}$concentration suggests that $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ were indeed exchanging with $\mathrm{Na}^{+}$.

### 4.1.2.4 Sulfate analysis

Figure 4.12 shows the sulfate concentration as a function of time in system 2. The $\mathrm{SO}_{4}{ }^{2-}$ concentration in the high flowrate system increased from $117 \mathrm{mg} / \mathrm{L}$ on Day 2 to $1080 \mathrm{mg} / \mathrm{L}$ on Day 36, and then dropped to $647 \mathrm{mg} / \mathrm{L}$ on Day 42 (Figure 4.12a). The decrease in sulfate concentration may be the result of the reduction of sulfate to $\mathrm{S}^{2-}$ by SRB in the MFT layer. After 42 days, the sulfate concentration fluctuated within the range of 700 to $995 \mathrm{mg} / \mathrm{L}$.

The concentration of sulfate on Day 5 in the release water of the low flowrate system was $32.1 \mathrm{mg} / \mathrm{L}$ (Figure 4.12b). From Day 5, the sulfate concentration decreased and reached $4.7 \mathrm{mg} / \mathrm{L}$ on Day 26. From Day 27, sulfate concentration increased rapidly and reached $695 \mathrm{mg} / \mathrm{L}$ on Day 67 . From then on, the sulfate concentration fluctuated within the range of $268 \mathrm{mg} / \mathrm{L}$ and $647 \mathrm{mg} / \mathrm{L}$. Comparison of the high flowrate system and the low flowrate system indicated that the low flowrate system was more effective at decreasing the $\mathrm{SO}_{4}{ }^{2-}$ concentration in release water.

The original $\mathrm{SO}_{4}{ }^{2-}$ concentration in MFT was $30.2 \mathrm{mg} / \mathrm{L}$. The $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feed water was $2160 \mathrm{mg} / \mathrm{L}$. The highest $\mathrm{SO}_{4}{ }^{2-}$ concentration in the
high flowrate system and low flowrate system were $1080 \mathrm{mg} / \mathrm{L}$ and $695 \mathrm{mg} / \mathrm{L}$, respectively. Therefore, the $\mathrm{SO}_{4}{ }^{2-}$ removal efficiency of the high flowrate system and the low flowrate system were $50 \%$ and $68 \%$, respectively.


Figure 4.12 $\mathrm{SO}_{4}{ }^{2-}$ concentration as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2), where the dashed line represents the $\mathrm{SO}_{4}{ }^{\mathbf{2 -}}$ concentration in the feed water

### 4.1.2.5 pH analysis

Figure 4.13 a and 4.13 b show the pH for the high flowrate system and low flowrate system, respectively. As shown in Figure 4.13a, the pH of the high flowrate system fluctuated between 7.76 and 7.97 from Day 2 to Day 11, and then dropped to 7.40

(a)

(b)

Figure 4.13 pH as a function of time in the release water of System 2 dynamic columns: (a) at a high flowrate of feed water ( $16.9 \mathrm{~mL} / \mathrm{h}$ in Column DH-2); (b) at a low flowrate of feed water ( $4.7 \mathrm{~mL} / \mathrm{h}$ in Column DL-2)
until Day 23, and fluctuated within the range of 7.39 and 8.14 during Day 24 and Day 60. The pH of the low flowrate system fluctuated within the range of 7.83 to 8.28 during Day 5 to Day 22, then dropped steadily down to 7.51 at Day 38, and fluctuated in between 7.46 and 8.19 during Day 39 until Day 110 (Figure 4.13b).

### 4.1.3 Physical and Chemical Characteristics in System 3 (Addition of Ac)

In System 3, $\mathrm{Ac}^{-}$was added to feed water at concentration of $100 \mathrm{mg} / \mathrm{L}$ as an organic carbon source to promote the microorganism-mediated reactions. The actual flowrates for this system were $18.7 \mathrm{~mL} / \mathrm{h}$ at the high flowate condition and $6.1 \mathrm{~mL} / \mathrm{h}$ at the low flowate condition.

### 4.1.3.1 Tracer Bromide analysis

In System 3, $\mathrm{Br}^{-}$was added as a tracer at $300 \mathrm{mg} / \mathrm{L}$ for the high flowrate system and $400 \mathrm{mg} / \mathrm{L}$ for the low flowrate system. As presented in Figure 4.14 a and $4.14 \mathrm{~b}, \mathrm{Br}^{-}$ increased steadily, which means that the feed water was gradually penetrating the MFT layer. $\mathrm{Br}^{-}$in the release water was approximately $300 \mathrm{mg} / \mathrm{L}$ after 60 days in the high flowrate system, whereas in low flowrate system, it took 123 days for release water $\mathrm{Br}^{-}$ concentration to reach $400 \mathrm{mg} / \mathrm{L}$. Therefore, it was determined that the approximate saturation times for high and low flowrate systems were respectively 60 and 123 days.

### 4.1.3.2 Calcium and magnesium analysis

Figure 4.15 and 4.16 illustrate the changes in the concentrations of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ in the high and low flowrate systems. The original $\mathrm{Ca}^{2+}$ concentration of the MFT
was $22.8 \mathrm{mg} / \mathrm{L}$ and the $\mathrm{Ca}^{2+}$ in the feed water was $42 \mathrm{mg} / \mathrm{L}$. In the high flowrate system, the $\mathrm{Ca}^{2+}$ concentration varied in a very small range of $23.6 \mathrm{mg} / \mathrm{L}$ to $26.9 \mathrm{mg} / \mathrm{L}$ from Day 3 to Day 37. On Day 38 , the $\mathrm{Ca}^{2+}$ concentration increased from $26.6 \mathrm{mg} / \mathrm{L}$ to $31.8 \mathrm{mg} / \mathrm{L}$, and then varied in the range of $31.8 \mathrm{mg} / \mathrm{L}$ and $23.4 \mathrm{mg} / \mathrm{L}$ from Day 38 to Day 60.


Figure $4.14 \mathrm{Br}^{-1}$ concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathrm{Br}^{-}$concentration in the feed water

In the low flowrate system, the $\mathrm{Ca}^{2+}$ concentration varied in the range of 18.0 $\mathrm{mg} / \mathrm{L}$ to $24.5 \mathrm{mg} / \mathrm{L}$ from Day 3 to Day 63. From Day 64 to Day 114 , the $\mathrm{Ca}^{2+}$ concentration was relatively stable with an average concentration about $22.8 \mathrm{mg} / \mathrm{L}$. The reason for the much lower $\mathrm{Ca}^{2+}$ concentrations between Day 114 and Day 130 could not be identified.


Figure $4.15 \mathrm{Ca}^{2+}$ concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathbf{C a}^{2+}$ concentration in the feed water

The highest $\mathrm{Ca}^{2+}$ concentration of the high flowrate system and the low flowrate system was $31.8 \mathrm{mg} / \mathrm{L}$ and $25.5 \mathrm{mg} / \mathrm{L}$, respectively. In both columns, due to the low concentration of $\mathrm{Ca}^{2+}$, it is difficult to verify that there was significant difference of $\mathrm{Ca}^{2+}$ concentration between feed water and the release water. However, retention of $\mathrm{Ca}^{2+}$ in the MFT layer may have occurred to some extent as a result of sorption, ion exchange and/or precipitation.


Figure 4.16 $\mathbf{M g}^{\mathbf{2 +}}$ concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathbf{M g}{ }^{\mathbf{2 +}}$ concentration in the feed water

As shown in Figure 4.16a, the change in $\mathrm{Mg}^{2+}$ was similar to that of $\mathrm{Ca}^{2+}$ in the high flowrate system. During the first 37 days, $\mathrm{Mg}^{2+}$ fluctuated between $15.8 \mathrm{mg} / \mathrm{L}$ and $17.4 \mathrm{mg} / \mathrm{L}$, while $\mathrm{Mg}^{2+}$ concentration in the feed water was $23.8 \mathrm{mg} / \mathrm{L}$. On Day 38, there was a $16 \%$ increase in $\mathrm{Mg}^{2+}$ concentration to $20.2 \mathrm{mg} / \mathrm{L}$. Then the change of $\mathrm{Mg}^{2+}$ concentration was within the range of $19 \mathrm{mg} / \mathrm{L} \pm 10 \%$. It could be thought that the $\mathrm{Mg}^{2+}$ concentration was stable in this system if an experimental error of approximately $10 \%$ is assumed. The $\mathrm{Mg}^{2+}$ concentration in the feed water was $26.0 \mathrm{mg} / \mathrm{L}$. The highest concentration of $\mathrm{Mg}^{2+}$ in release water was $20.4 \mathrm{mg} / \mathrm{L}$. These results suggest that at least $22 \%$ of the $\mathrm{Mg}^{2+}$ was retained by MFT mostly likely due to sorption, ion exchange and/or precipitation.

The concentration of $\mathrm{Mg}^{2+}$ fed into the low flowrate system was $26.0 \mathrm{mg} / \mathrm{L}$. As the feed water was introduced into System 3 , the $\mathrm{Mg}^{2+}$ in the MFT pore water was slowly released, and therefore the $\mathrm{Mg}^{2+}$ concentration increased in the release water. Subsequently, when the feed water broke through the MFT layer on Day $122, \mathrm{Mg}^{2+}$ in release water was $16.7 \mathrm{mg} / \mathrm{L}$. After that, there was no sharp increase of $\mathrm{Mg}^{2+}$ in release water. More than $30 \% \mathrm{Mg}^{2+}$ was retained in the MFT layer. Ion exchange and precipitation might have contributed to this decrease.

The original $\mathrm{Mg}^{2+}$ concentration of the MFT was $14.8 \mathrm{mg} / \mathrm{L}$. The $\mathrm{Mg}^{2+}$ concentration in the feed water was $23.8 \mathrm{mg} / \mathrm{L}$. The highest $\mathrm{Mg}^{2+}$ concentration of the high flowrate system and the low flowrate system was $20.4 \mathrm{mg} / \mathrm{L}$ and $16.7 \mathrm{mg} / \mathrm{L}$, respectively. Therefore, $\mathrm{Mg}^{2+}$ removal efficiency of the high flowrate system and the low flowrate system was $14 \%$ and $26 \%$, respectively.

### 4.1.3.3 Sodium concentration analysis

Figure 4.17 a and 4.17 b present the $\mathrm{Na}^{+}$concentrations in release water for high and low flowrate columns. The original $\mathrm{Na}^{+}$concentration in the MFT was $1090 \mathrm{mg} / \mathrm{L}$. The $\mathrm{Na}^{+}$concentration in the feed water was $1230 \mathrm{mg} / \mathrm{L}$.


Figure $4.17 \mathrm{Na}^{+}$concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathrm{Na}^{+}$concentration in the feed water

In the high flowrate system, $\mathrm{Na}^{+}$showed a general increasing trend with minor fluctuation before the feed water broke through the MFT layer, from $1036 \mathrm{mg} / \mathrm{L}$ to 1290 $\mathrm{mg} / \mathrm{L}$ (Figure 4.17 a ). As the feed water initially moved through the MFT, the original pore water in the MFT layer was slowly released. With more and more feed water entering the MFT layer, more and more $\mathrm{Na}^{+}$in MFT layer was replaced by $\mathrm{Ca}^{2+}$ or $\mathrm{Mg}^{2+}$ due to ion exchange, and subsequently moved into the release water with the bulk feed water.

As shown in Figure 4.17b, an appreciable increase in the $\mathrm{Na}^{+}$concentration of the release water in the low flow rate system occurred. There was a steady increase from Day 83 to Day 121 when the feed water was approaching the breakthrough point. From Day 106 , the $\mathrm{Na}^{+}$concentration in release water was higher than that in feed water. This result implies that some $\mathrm{Na}^{+}$in the MFT layer was replaced by $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$, and subsequently $\mathrm{Na}^{+}$was released into the release water.

The highest $\mathrm{Na}^{+}$concentrations in the high and the low flowrate columns were $1290 \mathrm{mg} / \mathrm{L}$ and $1330 \mathrm{mg} / \mathrm{L}$, translating to a $5 \%$ and $8 \%$ increase in the $\mathrm{Na}^{+}$concentration respectively.

### 4.1.3.4 Sulfate analysis

The original $\mathrm{SO}_{4}{ }^{2-}$ concentration of the MFT was $17.7 \mathrm{mg} / \mathrm{L}$. The $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feed water was $1035 \mathrm{mg} / \mathrm{L}$. It can be seen from Figure 4.18a, the $\mathrm{SO}_{4}{ }^{2-}$ concentration in the release water for the high flowrate system increased from 93.6 $\mathrm{mg} / \mathrm{L}$ at the beginning of incubation to $600 \mathrm{mg} / \mathrm{L}$ at the end of experiment, due to the gradual penetration of the feed water through the MFT. Because of some limitations in
the chemical analyses, a mass balance could not be conducted. However, the difference between the feed water and release water $\mathrm{SO}_{4}{ }^{2-}$ concentration at breakthrough point indicated that MFT could reduce $\mathrm{SO}_{4}{ }^{2-}$ concentration. The reason for this reduction in $\mathrm{SO}_{4}{ }^{2-}$ concentration might be that SRB in the MFT layer were actively reducing $\mathrm{SO}_{4}{ }^{2-}$ to sulfide ( $\mathrm{S}^{2-}$ ).

(b)

Figure $4.18 \quad \mathrm{SO}_{4}{ }^{2-}$ concentration as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3), where the dashed line represents the $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feed water

The initial sulfate in the release water was $41.1 \mathrm{mg} / \mathrm{L}$ in the low flowrate column (Figure 4.18b). Then, the sulfate concentration in the release water decreased slowly to $6.6 \mathrm{mg} / \mathrm{L}$ on Day 37. A sharp increase occurred on Day 38, from $6.6 \mathrm{mg} / \mathrm{L}$ to $28.3 \mathrm{mg} / \mathrm{L}$. From then, the effluent sulfate increased with minor fluctuation until the breakthrough point. The sulfate concentration at breakthrough was $342 \mathrm{mg} / \mathrm{L}$. Later, the concentration fluctuated within a large range of $183 \mathrm{mg} / \mathrm{L}$ to $496 \mathrm{mg} / \mathrm{L}$.

Generally, before breakthrough, the effluent sulfate at both systems was far lower than that in the feed water before breakthrough point. The sulfate concentration in the feed water was $1015 \mathrm{mg} / \mathrm{L}$, whereas the highest $\mathrm{SO}_{4}{ }^{2-}$ in the release water was 665 $\mathrm{mg} / \mathrm{L}$ and $496 \mathrm{mg} / \mathrm{L}$ in the high and low flowrate columns, respectively. It is believed that the low flowrate system provided more time for sulfate reduction in MFT layer and thus produced lower sulfate concentrations in the release water.

The highest $\mathrm{SO}_{4}{ }^{2-}$ concentration of the high flowrate system and the low flowrate system was $665 \mathrm{mg} / \mathrm{L}$ and $392 \mathrm{mg} / \mathrm{L}$. Therefore, $\mathrm{SO}_{4}{ }^{2-}$ removal efficiency of the high flowrate system and the low flowrate system was $36 \%$ and $62 \%$, respectively.

### 4.1.3.5 pH analysis

As shown in Figure 4.19a, the pH of release water fluctuated within the range of 7.6 to 8.6 , with an overall trend of slight increase. The pH of the release water in the low flowrate system fluctuated within the range of 7.7 to 8.2 during the first 115 days (Figure 4.19b). From Day 116, when approaching the breakthrough point, pH increased to over 8.3. It is possible that the activity of SRB may be contributing to the pH increase in the low flow rate column.


Figure 4.19 pH as a function of time in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3)

### 4.1.3.6 Conductivity analysis

As discussed previously, electronic conductivity provides an indication of the total ions in the solution. At the first 6 days of incubation in high flowrate system (Figure 4.20a), the conductivity increased slightly from $6040 \mu \mathrm{~S} / \mathrm{cm}$ to $6500 \mu \mathrm{~S} / \mathrm{cm}$. This slight increase was likely due to small increases in $\mathrm{Na}^{+}, \mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}$ and $\mathrm{SO}_{4}{ }^{2-}$ at the beginning.

The conductivity decreased to $4860 \mu \mathrm{~S} / \mathrm{cm}$ on Day 23 , and increased to $5210 \mu \mathrm{~S} / \mathrm{cm}$ on Day 29 , and from then on, the conductivity remained relatively constant at $5300 \pm 150$ $\mu \mathrm{S} / \mathrm{cm}$. This result suggests that the total ion concentration substantially did not change much in the last period of incubation. This result was consistent with the results of ion concentration $\left(\mathrm{Na}^{+}, \mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}\right.$ and $\left.\mathrm{SO}_{4}{ }^{2-}\right)$ in this period.


Figure 4.20 Conductivity as a function in the release water of System 3 dynamic columns: (a) at a high flowrate of feed water ( $18.7 \mathrm{~mL} / \mathrm{h}$ in Column DH-3); (b) at a low flowrate of feed water ( $6.1 \mathrm{~mL} / \mathrm{h}$ in Column DL-3)

Unlike the high flowrate system, the low flowrate system showed a decrease in the release water conductivity during the first 28 days (Figure 4.20 b ), from an initial value of $5910 \mu \mathrm{~S} / \mathrm{cm}$ to $4500 \mu \mathrm{~S} / \mathrm{cm}$. In the following days, the conductivity fluctuated within the range of $4540 \mu \mathrm{~S} / \mathrm{cm}$ and $5630 \mu \mathrm{~S} / \mathrm{cm}$ with the average value at $5000 \mu \mathrm{~S} / \mathrm{cm}$. The conductivity of the feed water was $5700 \mu \mathrm{~S} / \mathrm{cm}$, and therefore the lower conductivity of the release water might be attributed to the $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}, \mathrm{CO}_{3}{ }^{2-}$ and $\mathrm{SO}_{4}{ }^{2-}$ reduction resulting from chemical precipitation, ion exchange, sorption and/or $\mathrm{SO}_{4}{ }^{2-}$ reduction by SRB.

The original conductivity of the MFT was $4160 \mu \mathrm{~S} / \mathrm{cm}$. The conductivity of the feed water $5700 \mu \mathrm{~S} / \mathrm{cm}$. Overall, the conductivity of release water at both flowrate conditions was within the range of $4500 \mu \mathrm{~S} / \mathrm{cm}$ and $6500 \mu \mathrm{~S} / \mathrm{cm}$.

### 4.1.4 Physical and Chemical Characteristics in System 4 (WIP Water with $\mathrm{Br}^{-}$as

 Tracer)
### 4.1.4.1 Bromide concentration analysis

For System 4, WIP water with $\mathrm{Br}^{-}$added as the tracer (rather than CT water) was fed into the dynamic columns at two flow rates ( $19.4 \mathrm{~mL} / \mathrm{h}$ and $6.2 \mathrm{~mL} / \mathrm{h}$ ). The tracer concentrations were $300 \mathrm{mg} / \mathrm{L}$ and $400 \mathrm{mg} / \mathrm{L} \mathrm{Br}^{-}$for the high and the low flowrate systems, respectively. Figure 4.21 a and 4.21 b indicate that it took approximately 45 days and 116 days for $\mathrm{Br}^{-}$to reach the respective concentrations in the feed water. In the low flowrate system, it was observed that $\mathrm{Br}^{-7}$ had a sudden increase from $230 \mathrm{mg} / \mathrm{L}$ to 370 $\mathrm{mg} / \mathrm{L}$ during the week of Day 109 to Day 116, which means during this week, the original MFT porewater had been completely replaced with feed water.


Figure 4.21 $\mathrm{Br}^{-}$concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the Br concentration in the feed water

### 4.1.4.2 Calcium and magnesium concentration analysis

The original $\mathrm{Ca}^{2+}$ concentrations in the MFT and feed water were $22.8 \mathrm{mg} / \mathrm{L}$ and $13.7 \mathrm{mg} / \mathrm{L}$, respectively. Both columns showed a similar trend regarding the changes in $\mathrm{Ca}^{2+}$ concentrations (Figure 4.22 a and 4.22 b ). The changes in $\mathrm{Ca}^{2+}$ concentrations
seemed concomitant to follow the general trend of gradual penetration of feed water through the MFT. The initial $\mathrm{Ca}^{2+}$ concentrations were consistent with the $\mathrm{Ca}^{2+}$ in MFT pore water. The subsequent slight decrease in $\mathrm{Ca}^{2+}$ was due to the replacement of MFT porewater with feed water, which had a lower $\mathrm{Ca}^{2+}$ concentration.


Figure 4.22 $\mathrm{Ca}^{2+}$ concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathbf{C a}^{2+}$ concentration in the feed water

At the end of the experiments, $\mathrm{Ca}^{2+}$ concentration in the high flowrate system and the low flowrate system was $17.4 \mathrm{mg} / \mathrm{L}$ and $16.8 \mathrm{mg} / \mathrm{L}$. Therefore, feed of WIP water did not remove $\mathrm{Ca}^{2+}$ from the MFT layer completely. The release water $\mathrm{Ca}^{2+}$ concentration might have eventually approached the $\mathrm{Ca}^{2+}$ in the feed water given a longer incubation time.

The original $\mathrm{Mg}^{2+}$ concentrations in the MFT and feed water were $14.8 \mathrm{mg} / \mathrm{L}$ and $8.2 \mathrm{mg} / \mathrm{L}$ respectively. The $\mathrm{Mg}^{2+}$ concentration decreased gradually in both the high and low flowrate columns (Figure 4.23a and 4.23b), following a similar trend as $\mathrm{Ca}^{2+}$. As discussed previously, it could be attributed to the replacement of MFT pore water by the feed water.

At the end of experiment, $\mathrm{Mg}^{2+}$ in the release water was $11.0 \mathrm{mg} / \mathrm{L}$ for the high flowrate system and $10.9 \mathrm{mg} / \mathrm{L}$ for the low flowrate system. Therefore, it is anticipated that given sufficient time, the feed water would completely replace the MFT porewater and that the release water $\mathrm{Mg}^{2+}$ concentration would approach the $\mathrm{Mg}^{2+}$ concentration in the feed water.

The analysis of feed water also indicated that there was $35.7 \mathrm{mg} / \mathrm{L}$ of $\mathrm{CO}_{3}{ }^{2-}$ and $296 \mathrm{mg} / \mathrm{L}$ of $\mathrm{SO}_{4}{ }^{2-}$ in the WIP water. It is known that the following reactions can occur:

$$
\begin{array}{ll}
\mathrm{Ca}^{2+}(\mathrm{aq})+\mathrm{CO}_{3}{ }^{2-}(\mathrm{aq}) \rightarrow \mathrm{CaCO}_{3}(\mathrm{~s}) \downarrow & \mathrm{Ksp}=3.36 \times 10^{-9} \\
\mathrm{Mg}^{2+}(\mathrm{aq})+\mathrm{CO}_{3}{ }^{2-}(\mathrm{aq}) \rightarrow \mathrm{MgCO}_{3}(\mathrm{~s}) \downarrow & \mathrm{Ksp}=2.38 \times 10^{-6} \tag{4-3}
\end{array}
$$

These reactions could also contribute to the decrease in $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ concentrations over the incubation periods for both high and low flowrate columns.


Figure $4.23 \mathbf{M g}^{\mathbf{2 +}}$ concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathbf{M g}^{\mathbf{2 +}}$ concentration in the feed water

### 4.1.4.3 Sodium concentration analysis

In both high and low flowrate columns, $\mathrm{The}^{\mathrm{Na}}{ }^{+}$concentrations were relatively stable over the incubation period. For the high flowrate system (Figure 4.24a), the $\mathrm{Na}^{+}$ concentrations fluctuated between $960 \mathrm{mg} / \mathrm{L}$ and $1100 \mathrm{mg} / \mathrm{L}$, with an average of 1052 $\mathrm{mg} / \mathrm{L}$; whereas for the low flowrate system (Figure 4.24b), the $\mathrm{Na}^{+}$concentration varied
in the range of $1010 \mathrm{mg} / \mathrm{L}$ to $1150 \mathrm{mg} / \mathrm{L}$, with an average $\mathrm{Na}^{+}$concentration of 1075 $\mathrm{mg} / \mathrm{L}$. Baseline $\mathrm{Na}^{+}$concentrations in MFT and feed water were $1090 \mathrm{mg} / \mathrm{L}$ and 893 $\mathrm{mg} / \mathrm{L}$, respectively. It was anticipated that with the feed water penetrating through the MFT layer, $\mathrm{Na}^{+}$concentrations in the release water should approach $893 \mathrm{mg} / \mathrm{L}$, that is the concentration of $\mathrm{Na}^{+}$in the feed water.


Figure $4.24 \mathrm{Na}^{+}$concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathbf{~ m L} / \mathrm{h}$ in Column DH4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathrm{Na}^{+}$concentration in the feed water

For both columns, a slight increase of $\mathrm{Na}^{+}$concentration was observed towards the end of the incubation period. It is possible that there was ion exchange between $\mathrm{Na}^{+}$ and $\mathrm{Ca}^{2+}$ as well as $\mathrm{Na}^{+}$and $\mathrm{Mg}^{2+}$, in which $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ were retained in the MFT layer and $\mathrm{Na}^{+}$was released. However, since the decreases in $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ were very small, the $\mathrm{Na}^{+}$concentration increase in the release water due to ion exchange would be difficult to detect considering the high baseline concentrations. Therefore, there was no definitive evidence that the ion exchange process occurred in the columns.

### 4.1.4.4 Sulfate analysis

The baseline $\mathrm{SO}_{4}{ }^{2-}$ concentrations in the MFT and the feed water were 17.7 $\mathrm{mg} / \mathrm{L}$ and $296 \mathrm{mg} / \mathrm{L}$ respectively. For the high flowrate column (Figure 4.25 a ), the $\mathrm{SO}_{4}{ }^{2-}$ concentration steadily increased from $19.9 \mathrm{mg} / \mathrm{L}$ on Day 3 to $102 \mathrm{mg} / \mathrm{L}$ on Day 11 , and then remained relatively stable around $100 \mathrm{mg} / \mathrm{L}$ till Day 48. From Day 48 until Day 60, the $\mathrm{SO}_{4}{ }^{2-}$ concentration increased slightly to $145 \mathrm{mg} / \mathrm{L}$.

For the low flowrate system (shown in Figure 4.25b), the $\mathrm{SO}_{4}{ }^{2-}$ concentration decreased gradually from approximately $140 \mathrm{mg} / \mathrm{L}$ to $5 \mathrm{mg} / \mathrm{L}$ on Day 60. From Day 61, the $\mathrm{SO}_{4}{ }^{2-}$ concentration increased almost linearly to $31.9 \mathrm{mg} / \mathrm{L}$ on Day 109. From Day 110 until Day 140 , the $\mathrm{SO}_{4}{ }^{2-}$ concentration increased rapidly to $137 \mathrm{mg} / \mathrm{L}$ and then fluctuated in the range of $123 \mathrm{mg} / \mathrm{L}$ to $197 \mathrm{mg} / \mathrm{L}$. The reasons for initial high $\mathrm{SO}_{4}{ }^{2-}$ concentrations in the release water were unknown. The mechanisms for subsequent decrease in $\mathrm{SO}_{4}{ }^{2-}$ concentrations might include precipitation with $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ as mentioned before and activity of SRB which reduced $\mathrm{SO}_{4}{ }^{2-}$ to $\mathrm{S}^{2-}$. The presence of SRB in System 4 was verified by the MPN and AVS analysis (See section 4.1.5). The increase
in $\mathrm{SO}_{4}{ }^{2-}$ concentration at the final stage suggests that the amount of sulfate fed into the system exceeded the kinetically controlled microbial consumption of $\mathrm{SO}_{4}{ }^{2-}$.

The $\mathrm{SO}_{4}{ }^{2-}$ concentration in the release water at the end of the incubation period was $128 \mathrm{mg} / \mathrm{L}$ for the high flowrate system and $152 \mathrm{mg} / \mathrm{L}$ for the low flowrate system. Therefore, $\mathrm{SO}_{4}{ }^{2-}$ removal efficiencies of $57 \%$ and $49 \%$ were achieved in the high and the low flowrate columns, respectively.

(b)

Figure $4.25 \mathrm{SO}_{4}{ }^{2-}$ concentration as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the $\mathrm{SO}_{4}{ }^{2-}$ concentration in the feed water

### 4.1.4.5 Conductivity analysis

The baseline conductivities of the MFT and feed water were $4160 \mu \mathrm{~S} / \mathrm{cm}$ and $4890 \mu \mathrm{~S} / \mathrm{cm}$ respectively. The conductivities of the release water at the end of the incubation period were $4850 \mu \mathrm{~S} / \mathrm{cm}$ and $4390 \mu \mathrm{~S} / \mathrm{cm}$ in the high and the low flowrate columns, respectively.

Specifically, in the high flowrate column, the conductivity decreased from 5740 $\mu \mathrm{S} / \mathrm{cm}$ on Day 4 to $4330 \mu \mathrm{~S} / \mathrm{cm}$ on Day 21, and then stable at $4630 \mu \mathrm{~S} / \mathrm{cm}$ (Figure 4.26a). The conductivity decrease seem to be the result of a decrease in the $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ concentration since, over this period, $\mathrm{Na}^{+}$concentrations were relatively stable and $\mathrm{Br}^{-}$ and $\mathrm{SO}_{4}{ }^{2-}$ concentration were increasing.

The conductivity of the low flowrate system (Figure 4.26b) decreased from $5480 \mu \mathrm{~S} / \mathrm{cm}$ on Day 3 to $4380 \mu \mathrm{~S} / \mathrm{cm}$ on Day 19. Then, the conductivity seemed to be stabilizing at $4680 \mu \mathrm{~S} / \mathrm{cm}$ until Day 57. The conductivity increased to $4990 \mu \mathrm{~S} / \mathrm{cm}$ on Day 62 and then decreased to $4360 \mu \mathrm{~S} / \mathrm{cm}$ on Day 79. After Day 79, the conductivity was stable at approximately $4450 \mu \mathrm{~S} / \mathrm{cm}$ until Day 140.

The difference between the conductivity of the feed water and release water could be attributed to chemical processes, i.e. precipitation (between $\mathrm{CO}_{3}{ }^{2-}$ and $\mathrm{Ca}^{2+}$ or $\mathrm{Mg}^{2+}$ ) and ion exchange or to microbiological processes.

### 4.1.4.6 $\mathbf{~ p H}$ analysis

Figure 4.27 a and 4.27 b give the pH in the high and low flowrate columns, respectively. The pH of the high flowrate system varied in the range of 7.60 and 8.09
with a slightly increasing trend. For the low flowrate system, the pH ranged from 7.65 to 8.18 with the trend slightly decreasing.


Figure 4.26 Conductivity as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water $(6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4), where the dashed line represents the conductivity in the feed water


Figure 4.27 pH as a function of time in the release water of System 4 dynamic columns: (a) at a high flowrate of feed water ( $19.4 \mathrm{~mL} / \mathrm{h}$ in Column DH-4); (b) at a low flowrate of feed water ( $6.2 \mathrm{~mL} / \mathrm{h}$ in Column DL-4)
4.1.5 Microbiological Characteristics by MPN and AVS Analyses in all Systems

### 4.1.5.1 MPNs of Methanogens and SRB

Enumeration of methanogens and SRB were conducted on all testing systems at the end of incubation period using 3-tube MPN analyses. The results of MPNs are summarized in Appendix A13. As discussed previously, biodegradation of residual
hydrocarbons in the MFT will contribute to the production of $\mathrm{CH}_{4}$ by methanogens and the reduction of $\mathrm{SO}_{4}{ }^{2-}$ by SRB . The microbial activity will affect the physical andchemical properties of MFT and many affect its densification. Recently, Fedorak et al. (2003) reported an unexpected increase in the densification rate of MFT accompanying the microbial $\mathrm{CH}_{4}$ production.

MPN results from the blank jar tests were used as the baseline numbers of methanogens and SRB for the corresponding dynamic columns, as shown in Figure 4.28. This result can be verified since neither hydrocarbon nor terminal electron acceptors (i.e. $\mathrm{CO}_{2}$ or $\mathrm{SO}_{4}{ }^{2-}$ ) were introduced to the MFT, although $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ concentrations in each jar were adjusted. It can be seen from Figure 4.28 that the baseline methanogens and SRB or MPN value in all systems ranged from $10^{5}$ to $10^{6}$ per g dry MFT. This was comparable to previous microbiological charactization of MFT. Fedorak et al. (2000) reported that the MPN for Syncrude Canada Ltd. MFT was mostly in the range of $10^{5}$ to $10^{7} / \mathrm{g}$ (of dry solids) over 1 year of incubation, whereas Holowenko (2000) reported that the MPN of SRB was for the most part in the range of $10^{5}$ to $10^{7} / \mathrm{g}$ (of dry solids) for Syncrude Canada Ltd. MFT. The changes in MPN of methanogens and SRB will be discussed separately in the following sections.

Figures 4.29 and 4.30 present the MPN of methanogens at the end of the incubation period in all high and low flowrate dynamic columns. It seemed that different flow rates did not resulted in changes in MPN. There was also no apparent difference of MPNs along the depth profile of the column. However, a drastic decrease in methanogens was found in Systems 1 and 2, whereas the methanogens showed only slight decrease in Systems 3 and 4. This difference could be attributed to the competition
between methanogens and SRB for substrates and the availability of TEAs. The errors in sampling might also be the reason for the drastic decrease in methanogens in Systems 1 and 2. In Systems 1 and 2, high concentrations of $\mathrm{SO}_{4}{ }^{2-}$ may have stimulated the growth of SRB, which therefore supressed the methanogens.

There are two types of methanogens: (i) acetotrophic methanogens, which can grow on acetate, and (ii) hydrogenotrophic methanogens, which can utilize $\mathrm{H}_{2}$ and bicarbonate. Acetate ( $100 \mathrm{mg} / \mathrm{L}$ ) added to System 3 may have been sufficient to serve as the substrate for both acetotrophic methanogens and SRB. Some other organic compounds might also be utilized by SRB as the carbon source. Therefore, despite the high $\mathrm{SO}_{4}{ }^{2-}$ amendment to System 3, the MPNs of methanogens and SRB were quite similar. Similarly in System 4, it is postulated that no out-competition of SRB over methanogens occurred due to lower $\mathrm{SO}_{4}{ }^{2-}$ concentration and the different composition of organic matter in the feed water (WIP water versus CT water).


Figure 4.28 Background MPNs of methanogens and SRB (as measured in Jar tests)


Figure 4.29 MPN of methanogens in high flowrate systems after 60 days of incubation


Figure 4.30 MPN of methanogens in low flowrate systems after 140 days of incubation

The MPN of SRB in high and low flowrate columns are shown in Figures 4.31 and 4.32, respectively. No apparent difference in the MPN of SRB was observed with the different flow rates. The irregular changes in MPN of SRB along the depth of the columns may be due to the heterogeneity of the MFT materials with regard to the distribution of substrates and microbial population. Slightly lower MPN of SRB in System 3 could be explained by the competition of methanogen enhanced by acetate amendment, as discussed previously. It was assumed that in systems with higher $\mathrm{SO}_{4}{ }^{2-}$ concentrations, the activity of SRB would be enhanced and thus MPN of SRB would increase. In contrast, there was no increase in the estimated SRB numbers in Systems 1 and 2 compared to those from the blank jar tests, indicating that the substrate was the limiting factor in those systems.


Figure 4.31 MPN of SRB in high flowrate systems after $\mathbf{6 0}$ days of incubation


Figure 4.32 MPN of SRB in low flowrate systems after 140 days of incubation

### 4.1.5.2 AVS analysis

Oxidation of the organic matter coupled to sulfate reduction will decrease the concentrations of both substrates and TEAs (i.e. $\mathrm{SO}_{4}{ }^{2-}$ ) and produce biogenic $\mathrm{CO}_{2}$ and reduced sulfur. Thus, the increase in $\mathrm{HCO}_{3}{ }^{-}$and sulfide concentrations indicates the occurrence of SRB activity in the dynamic columns.

The pH in the dynamic columns ranged from 7.2 to 8.8 . In this pH range, the predominant form of sulfide is $\mathrm{HS}^{-}$rather than $\mathrm{H}_{2} \mathrm{~S}$ (Fedorak et al. 2000). In the presence of metals, such as $\mathrm{Fe}^{2+}$, the $\mathrm{HS}^{-}$will further react with the metals to precipitate as metal sulfides, such as FeS. FeS may be further converted to pyrite, as shown in the following equations:

$$
\begin{align*}
& \mathrm{HS}^{-}(\mathrm{aq})+\mathrm{Fe}^{2+}(\mathrm{aq}) \longrightarrow \mathrm{FeS}(\mathrm{~s})+\mathrm{H}^{+}(\mathrm{aq})  \tag{4-4}\\
& \mathrm{FeS}(\mathrm{aq})+\mathrm{S}^{0}(\mathrm{aq}) \longrightarrow \mathrm{FeS}_{2}(\mathrm{~s}) \tag{4-5}
\end{align*}
$$

AVS (mainly the iron sulfides) is an effective sink for reduced sulfur so that the reduced sulfur will not be released to the atmosphere in forms of $\mathrm{H}_{2} \mathrm{~S}$ (Schlesinger, 1997). Due to the meta-stability and reactivity of AVS compounds, its presence is assumed to be an indicator of recent $\mathrm{SO}_{4}{ }^{2-}$ reduction (Kennedy et al. 1999).

Based on the analytical method used by ALS laboratories, results of the analyzed sulfur include both AVS and fine-graded (newly formed) synthetic pyrite. Dissolved sulfide was also measured using the CHEMetrics test kit, which measures dissolved $\mathrm{H}_{2} \mathrm{~S}, \mathrm{HS}^{-}$, and acid soluble metallic sulfides in suspension. There was no appreciable dissolved sulfide in most systems (Appendix A15, A16, and A17).

The detection of AVS for Systems 1 and 2 (see Table 4.1) further verify that sulfate reduction was occurring in all the tested columns. The Systems 3 and 4 were not detected due to lack of funding.

### 4.2 Jar Test

The jars were sampled after 128-day incubation. All samples were centrifuged and the pore water samples were sent to Syncrude Canada Ltd. laboratory for chemical analysis. The concentrations of major cations and anions, pH and EC for all jars were summarized in Table 4.2. Because no samples were analyzed immediately after the setup of jars, the designed concentrations (based on dynamic columns) were assumed to be the initial conditions. The lack of initial concentrations and analytical variability made the interpretation of the results less reliable. However, some trends were still observed.

Table 4.1 AVS in Jars and Dynamic Columns

| System | Sample | $\mathrm{T}_{\text {incu }}(\mathrm{d})$ | AVS $(\mu \mathrm{mol} / \mathrm{g}$ dry $)$ |
| :---: | :---: | :---: | :---: |
|  | DH-1-U | 60 | 0.5 |
|  | DH-1-L | 60 | 2.8 |
| DL-1 | DL-1-U | 140 | 6.34 |
|  | DL-1-M-U | 140 | 7.32 |
|  | DL-1-M-L | 140 | 7.83 |
|  | DL-1-L | 140 | 3.69 |
| DL-2 | DH-2-U | 60 | 5.5 |
|  | DH-2-L | 60 | 4.4 |
|  | DL-2-U | 140 | 10.2 |
|  | DL-2-M-U | 140 | 9.41 |
|  | DL-2-M-L | 140 | 7.76 |
| Jar1 | DL-2-L | 140 | 10.4 |
|  | D1-1 | 128 | 12.2 |
| Jar2 | D1-2 | 128 | 12.6 |
|  | D2-1 | 128 | 13.8 |
| Jar3 | D2-2 | 128 | 13.5 |
|  | D3-1 | 128 | 16.7 |
|  | Jar4 | D3-2 | 128 |

Table 4.2 Major Results of Chemical Analysis of MFT Porewater Samples for all Jars after 128-d Incubation

| Jars | Incub. <br> Time (d) | pH | $\begin{gathered} \mathrm{EC} \\ (\mu \mathrm{~S} / \mathrm{cm}) \end{gathered}$ | $\left(\begin{array}{c} \mathrm{Br}^{-} \\ (\mathrm{mg} / \mathrm{L}) \end{array}\right.$ | $\underset{(\mathrm{mg} / \mathrm{L})}{\mathrm{Na}^{+}}$ | $\begin{gathered} \mathrm{K}^{+} \\ (\mathrm{mg} / \mathrm{L}) \end{gathered}$ | $\begin{gathered} \mathrm{Mg}^{2+} \\ (\mathrm{mg} / \mathrm{L}) \\ \hline \end{gathered}$ | $\begin{gathered} \mathrm{Ca}^{2+} \\ (\mathrm{mg} / \mathrm{L}) \end{gathered}$ | $\begin{gathered} \mathrm{SO}_{4}{ }^{2-} \\ (\mathrm{mg} / \mathrm{L}) \\ \hline \end{gathered}$ | $\begin{gathered} \mathrm{HCO}_{3} \\ \text { (wt } \\ \mathrm{ppm}) \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Jar 1 | $0{ }^{\text {a }}$ | 8.3 | 7360 | 2500 | 1730 | 27.0 | 26.4 | 31.1 | 1060 | 584 |
|  | $128^{\text {b }}$ | 8.3 | 8270 | 3000 | 2285 | 19 | 25 | 32 | 688 | 1565 |
| Jar 2 | $0^{\text {a }}$ | 7.7 | 5250 | 1 | 1070 | 24.9 | 150 | 160 | 1800 | 859 |
|  | $128^{\text {b }}$ | 8.1 | 6205 | 5 | 1560 | 20 | 45 | 44 | 842 | 1695 |
| Jar 3 | $0^{\text {a }}$ | 8.1 | 6430 | 320 | 1240 | 26.5 | 26.0 | 61.7 | 995 | 784 |
|  | $128^{\text {b }}$ | 8.0 | 6765 | 600 | 1730 | 19 | 26 | 35 | 586 | 1920 |
| Jar 4 | $0^{\text {a }}$ | 8.1 | 5270 | 330 | 1090 | 15.0 | 8.1 | 13.7 | 321 | 843 |
|  | $128{ }^{\text {b }}$ | 8.0 | 5490 | 620 | 1375 | 16 | 17 | 25 | 190 | 1340 |

Notes: a: The initial concentrations were estimated from dynamic column results.
b: The 128th day results were average of the duplicate jars.

The concentrations of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ were not amended in Jars 1,3 , and 4. Thus, the changes in $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ were not as much as in Jar 2. In Jar 2, the decrease in $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ concentrations is possibly due to the ion exchange with $\mathrm{Na}^{+}$. Therefore, it is evident that MFT has the capacity to retain a certain amount of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ (when at relatively high concentrations), which verifies the theoretical rationale that MFT could be used to treat CT water. The concentrations of $\mathrm{SO}_{4}{ }^{2-}$ decreased in all jars, which could be explained by the anaerobic microbial-mediated sulfate reduction processes.

### 4.3 Barrel Mesocosms

The investigation of static column mesocosms was undertaken to study and monitor the physical, chemical and microbiological changes occurring in the CT beneath MFT co-disposal system. Four barrels, denoted as B1, B2, B3 and B4 respectively, have different constitutions as presented in Table 3.6. Among them, B1 and B2 were duplicates and consist of both MFT and CT, whereas B3 contained only MFT and B4 contained only CT. Two columns, denoted C1-17 and C1-18, also consisted of MFT and CT, but these were much smaller in size. The sampling and analyses of water samples from different fractions of four barrels and two columns were conducted after 2-year incubation period.

### 4.3.1 Calcium and Magnesium Concentrations and $\mathbf{p H}$

The pH values and the concentrations of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ in the release and pore water samples from the barrels and columns are given in Table 4.3. In Table $4.3 \mathrm{~B} 1 / 2$ represents the average of the results of B 1 and B 2 .

Table 4.3 $\mathbf{~ H H}$ and Concentrations of $\mathrm{Ca}^{\mathbf{2 +}}$ and $\mathrm{Mg}^{\mathbf{2 +}}$ in Barrels

|  | pH |  |  | $\mathrm{Ca}^{2+}(\mathrm{mg} / \mathrm{L})$ |  |  | $\mathrm{Mg}^{2+}(\mathrm{mg} / \mathrm{L})$ |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  | $\mathrm{B} 1 / 2$ | B 3 | B 4 | $\mathrm{~B} 1 / 2$ | B 3 | B 4 | $\mathrm{~B} 1 / 2$ | B 3 | B 4 |
| Release <br> water | 8.05 | 8.23 | 8.24 | 31.2 | 18.20 | 25.5 | 17.6 | 15.3 | 16.9 |
| MFT-U <br> water | 8.18 | 8.09 | NA | 14.1 | 12.10 | NA | 14.5 | 14.8 | NA |
| MFT-L <br> water | 8.20 | 8.11 | NA | 6.65 | 12.50 | NA | 14.1 | 14.1 | NA |
| CT-U water | 8.26 | NA | 8.25 | 18.2 | NA | 23.3 | 14.2 | NA | 14.8 |
| CT-L water | 8.25 | NA | 8.27 | 18.0 | NA | 20.2 | 14.9 | NA | 14.0 |

Note: NA- not applicable

It can be seen from Table 4.3 that the pH of the release water of B 1 and B 2 was around 8.05, which was lower than that of $\mathrm{B} 3(\mathrm{pH} 8.23)$ and $\mathrm{B} 4(\mathrm{pH} 8.24)$. However, the pH values of water samples from the upper layer of the MFT (MFT-U water) and the lower layer of the MFT (MFT-L water) in B1 and B2 were close to those in B3. The water samples from the upper layer of the CT (CT-U water) and lower layer of the CT (CT-L water) had similar pH values to those in B 4 .

The $\mathrm{Ca}^{2+}$ concentrations are important to assess the CT beneath MFT codisposal system. The average $\mathrm{Ca}^{2+}$ concentration in the CT water was above $100 \mathrm{mg} / \mathrm{L}$. In all the barrels, the $\mathrm{Ca}^{2+}$ concentrations in the release water decreased by approximately $70 \%$ compared to the initial $\mathrm{Ca}^{2+}$ concentrations. The release water showed higher $\mathrm{Ca}^{2+}$ concentrations than pore water samples from other fractions of the barrels (Table 4.3). This difference might be explained by the ion exchange occurring in the MFT layer. The concentrations of $\mathrm{Ca}^{2+}$ varied along the depth profile in B 1 and B 2 , indicating that MFT and CT had different physical and chemical characteristics. The $\mathrm{Ca}^{2+}$ concentration in the

B1 and B2 release water was on average $31.2 \mathrm{mg} / \mathrm{L}$, which was higher than those in B3 ( $18.2 \mathrm{mg} / \mathrm{L}$ ) and B4 ( $25.5 \mathrm{mg} / \mathrm{L}$ ). Hence, the high calcium concentrations in the release water of B1 and B2 might be attributed to both MFT and CT layers. Water samples from the lower layer of MFT in B1 and B2 (mean $6.6 \mathrm{mg} / \mathrm{L}$ ) had the lowest calcium concentrations. It is reasonable that B 4 , comprising only CT , showed higher calcium concentration than B3, which just has MFT.

Table 4.3 also shows the $\mathrm{Mg}^{2+}$ concentrations of the water samples from different parts for $\mathrm{B} 1 / 2, \mathrm{~B} 3$ and B 4 . The $\mathrm{Mg}^{2+}$ concentrations are comparable in water samples from each fraction of the barrels.

The effect of the column size on the static column mescocosm was studied by comparing the characteristics of barrels (bigger size) and those of columns (smaller size). The comparisons of pH as well as the concentrations of $\mathrm{Ca}^{2+}$ and $\mathrm{Mg}^{2+}$ between barrels and columns are shown in Table 4.4. The data in the column of C1-17/18 are the averages of measurements of both $\mathrm{C} 1-17$ and $\mathrm{C} 1-18$.

Table 4.4 Comparisons of $\mathbf{p H}$ and Concentrations of $\mathbf{C a}^{\mathbf{2 +}}$ and $\mathbf{M g}^{\mathbf{2 +}}$ between Barrels and Static Columns

|  | pH |  | $\mathrm{Ca}^{2+}(\mathrm{mg} / \mathrm{L})$ |  | $\mathrm{Mg}^{2+}(\mathrm{mg} / \mathrm{L})$ |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  | $\mathrm{B} 1 / 2$ | $\mathrm{C} 1-17 / 18$ | $\mathrm{~B} 1 / 2$ | $\mathrm{C} 1-17 / 18$ | $\mathrm{~B} 1 / 2$ | $\mathrm{C} 1-17 / 18$ |
| Release <br> water | 8.05 | 7.59 | 31.2 | 31.8 | 17.6 | 15.7 |
| MFT-U <br> water | 8.21 | 8.16 | 14.1 | 24.3 | 14.5 | 18.6 |
| MFT-L <br> water | 8.11 | 8.13 | 6.65 | 18.4 | 14.1 | 15.9 |
| CT-U water | 8.17 | 8.36 | 18.2 | 17.8 | 14.2 | 14.3 |
| CT-L water | 8.22 | 8.28 | 18.0 | 21.8 | 14.9 | 14.7 |

The constitutions of B1 and B2 were identical to those of C1-17 and C1-18 except that both B1 and B2 were much bigger than C1-17 and C1-18. Seen in the Table 4.4, the release water in $\mathrm{B} 1 / 2$ has a pH value of 8.05 , which is higher than that in $\mathrm{C} 1-$ 17/18 (7.59). Other than that, the pH values of the water samples from the other four parts are almost same for $\mathrm{B} 1 / 2$ and $\mathrm{C} 1-17 / 18$. As for the $\mathrm{Ca}^{2+}$ concentration, both the $\mathrm{B} 1 / \mathrm{B} 2$ and $\mathrm{C} 1-17 / 18$ had the similar $\mathrm{Ca}^{2+}$ concentrations in the release water. $\mathrm{Ca}^{2+}$ concentrations of both MFT-U water and MFT-L water in C1-17/18 were slightly higher than those in $\mathrm{B} 1 / 2$. The $\mathrm{Mg}^{2+}$ concentrations in water from each part of $\mathrm{B} 1 / 2$ were comparable to their corresponding values in C1-17/18. Therefore, the sample size did not show significant influence in the physical and chemical process in the MFT layers.

### 4.3.2 Sulfate Analysis

Sulfate is one of the major anions accounting for most of the dissolved fraction in the MFT pore water. The sulfate concentrations in the water sample from different parts of barrels and columns are shown in Table 4.5.

Table 4.5 $\mathrm{SO}_{4}{ }^{\mathbf{2 -}}$ Concentrations in the Fractions of Barrels and Static Columns

|  | $\mathrm{SO}_{4}{ }^{2-}(\mathrm{mg} / \mathrm{L})$ |  |  |  |
| :---: | :---: | :---: | :---: | :---: |
|  | $\mathrm{B} 1 / 2$ | B 3 | B 4 | $\mathrm{C} 1-17 / 18$ |
| Release water | 401.5 | 8.01 | 853.0 | 134.5 |
| MFT-U water | 145.2 | 13.1 | - | 86.0 |
| MFT-L water | 148.0 | 6.29 | - | 15.6 |
| CT-U water | 151.0 | - | 665.0 | 98.4 |
| CT-L water | 166.0 | - | 524.0 | 69.1 |

The concentrations of sulfate in B1, B2 and B4 were much higher than those in

B 3 due to the high dose of gypsum in CT . The $\mathrm{SO}_{4}{ }^{2-}$ concentrations in B 1 and B 2 were lower than that in B4, possibly because of the mixing effects of CT water and MFT pore water and the microbial-mediated sulfate reduction process.

It appeared that the size effect had some influence on the $\mathrm{SO}_{4}{ }^{2-}$ concentrations in the columns. In release water, the $\mathrm{SO}_{4}{ }^{2-}$ concentration was $401.5 \mathrm{mg} / \mathrm{L}$ in $\mathrm{B} 1 / 2$ whereas this in $\mathrm{C} 1-17 / 18$ was only $134.5 \mathrm{mg} / \mathrm{L}$. Similar to that in the release water, the $\mathrm{SO}_{4}{ }^{2-}$ concentrations in the other fractions in $\mathrm{B} 1 / 2$ are much lower than those in $\mathrm{C} 1-17 / 18$.

### 4.3.3 Alkalinity Analysis

Both the carbonate and bicarbonate are important constitutions in the water samples from different fractions. According to the pH in all systems, $\mathrm{HCO}_{3}{ }^{-}$should be the predominant species of alkalinity. The alkalinities of water samples (in $\mathrm{mg} \mathrm{HCO}_{3}{ }^{-} / \mathrm{L}$ ) are shown in Table 4.6. For all barrels, the $\mathrm{HCO}_{3}{ }^{-}$concentrations in the release water were higher than those in other fractions. Seen in the Table 4.6, the concentration of $\mathrm{HCO}_{3}{ }^{-}$in the release water in $\mathrm{B} 1 / 2$ was $1141 \mathrm{mg} / \mathrm{L}$, which was close to that in $\mathrm{C} 1-17 / 18$, and the same was in the other fractions. Therefore the size of columns showed no influence on the bicarbonate concentrations in the fractions.

Table 4.6 Alkalinities in the Fractions of Barrels and Static Columns

|  | Alkalinity $(\mathrm{mg} \mathrm{HCO}$ |  |  |  |
| :---: | :---: | :---: | :---: | :---: |
|  | $\left.{ }^{2-} / \mathrm{L}\right)$ |  |  |  |
|  | $\mathrm{B} 1 / 2$ | B 3 | B 4 | $\mathrm{C} 1-17 / 18$ |
| Release water | 1141 | 1272 | 709 | 1340 |
| MFT-U water | 923 | 1000 | - | 1426 |
| MFT-L water | 828 | 1016 | - | 1324 |
| CT-U water | 893 | - | 654 | 992 |
| CT-L water | 852 | - | 706 | 1168 |

### 4.3.4 Sodium Ions and Potassium Ions

The concentrations of $\mathrm{Na}^{+}$and $\mathrm{K}^{+}$in the fractions are shown in Table 4.7. It can be seen that the concentrations of $\mathrm{K}^{+}$in different fractions were much lower than those of $\mathrm{Na}^{+} . \mathrm{Na}^{+}$is one of the major cations in the fractions. The concentrations of $\mathrm{Na}^{+}$in each fraction were similar in all barrels and columns. Therefore, the difference in the make-up of B1/2, B3 and B4 had no influence on the $\mathrm{Na}^{+}$concentrations in different fractions. Whether the system was a barrel or column also had no influence on the $\mathrm{Na}^{+}$ concentrations.

Table 4.7 Concentrations of $\mathrm{Na}^{+}$and $\mathrm{K}^{+}$in the Fractions of Barrels and Static Columns

|  | $\mathrm{Na}^{+}(\mathrm{mg} / \mathrm{L})$ |  |  |  | $\mathrm{K}^{+}(\mathrm{mg} / \mathrm{L})$ |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  | $\mathrm{B} 1 / 2$ | B 3 | B 4 | $\mathrm{C} 1-17 / 18$ | $\mathrm{~B} 1 / 2$ | B 3 | B 4 | $\mathrm{C} 1-17 / 18$ |
| Release <br> water | 1310 | 1080 | 1370 | 1205 | 18 | 15 | 17 | 16 |
| MFT-U <br> water | 1135 | 1030 | NA | 1360 | 15 | 15 | NA | 19 |
| MFT-L <br> water | 1120 | 993 | NA | 1135 | 14 | 14 | NA | 16 |
| CT-U water | 1155 | NA | 1320 | 1165 | 14 | NA | 15 | 14 |
| CT-L water | 1160 | NA | 1300 | 1240 | 15 | NA | 14 | 15 |

Note: NA - not applicable

### 4.3.5 Solids Properties

The particle size of solids in different layers of B1, B3 and B4 are shown in Figure 4.33 to 4.35 . In these figures, the percentage of solid less than certain size increases with the increasing of particle size.

In Figure 4.33, the percentage of solids less than certain particle size in layer of

B1 varied greatly. There were $75.3 \%$ solids with particle size less than $20 \mu \mathrm{~m}$ in the upper layer of MFT, and the corresponding values in lower layer of MFT, upper layer of CT and lower layer of CT were $67.4 \%, 44.8 \%$ and $40.6 \%$ respectively, i.e., the upper layer of MFT had the finest solids and the lower layer of CT had the coarsest solids in B1. However, in B3, the particle size distributions in both the upper layer and lower layer of MFT were almost same (approximately $75 \%$ solids with particle size less than $20 \mu \mathrm{~m}$, Figure 4.34), indicating that MFT is very difficult to settle and densify, whereas the MFT-over-CT disposal system may facilitate the aggregation of fine particles in MFT.

Figure 4.35 gives the particle size distribution of both the upper layer and lower layer in B4. As we can see, the particle size distributions differ greatly. There were higher percentages of particles less than certain size in the upper layer of CT than those in lower layer of CT , due to the densification process.


Figure 4.33 Distribution of particle size in different fractions of B1


Figure 4.34 Distribution of particle size in different fractions of B3


Figure 4.35 Distribution of particle size in different fractions of B4

## Chapter 5

## Conclusions and Recommendations

Based on the results and discussions in the previous sections, the following conclusions can be drawn:

- The obtained flowrates were 5 to $6 \mathrm{~mL} / \mathrm{h}$ for low flowrate systems and 17 to 19 $\mathrm{mL} / \mathrm{h}$ for high flowrate systems. $\mathrm{Br}^{-}$tracer study was used to determine the hydraulic retention time for each system. In system 2, conductivity measurement was used to identify the hydraulic retention time. Due to the variances among all systems, the saturation times varied from 40 to 60 days for high flowrate systems, and from 100 to 123 days for the low flowrate systems.
- Table 5.1 shows the summary of cations and anions removal efficiencies for Systems 1 to 4. For System 1, there was no apparent removal $\mathrm{Ca}^{2+}$. The removal efficiency of $\mathrm{Mg}^{2+}, \mathrm{Na}^{+}, \mathrm{SO}_{4}{ }^{2-}$ and conductivity was $0 \%, 1.5 \%, 35 \%$ and $7 \%$ in the high flowrate system; $28 \%, 20 \%, 69 \%$ and $20 \%$ in the low flowrate system.

Table 5.1 Summary of Cations and Anions Removal Efficiency

| System | Flowrate | $\mathrm{Ca}^{2+}$ <br> $(\%)$ | $\mathrm{Mg}^{2+}$ <br> $(\%)$ | $\mathrm{Na}^{+}$ <br> $(\%)$ | $\mathrm{SO}_{4}{ }^{2-}$ <br> $(\%)$ | Conductivity <br> $(\%)$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| System 1 | high flowrate | 0 | 0 | -1.5 | -35 | -7 |
|  | low flowrate | 0 | -28 | -20 | -69 | -20 |
| System 2 | high flowrate | -68 | -57 | 0 | -50 | -10 |
|  | low flowrate | -70 | -60 | 0 | -68 | -10 |
| System 3 | high flowrate | -24 | -14 | 5 | -36 | 0 |
|  | low flowrate | -39 | -26 | 8 | -62 | 0 |
| System 4 | high flowrate | 0 | 0 | 0 | -57 | 0 |
|  | low flowrate | 0 | 0 | 0 | -49 | -10 |

- For System 2, the high flowrate system achieved $68 \% \mathrm{Ca}^{2+}$ removal efficiency, $57 \% \mathrm{Mg}^{2+}$ removal efficiency, $50 \% \mathrm{SO}_{4}{ }^{2-}$ removal efficiency, and $10 \%$ conductivity removal efficiency; the low flowrate system achieved $70 \% \mathrm{Ca}^{2+}$ removal efficiency, $60 \% \mathrm{Mg}^{2+}$ removal efficiency, $68 \% \mathrm{SO}_{4}{ }^{2-}$ removal efficiency, and $10 \%$ conductivity removal efficiency. In both flowrate systems, feeding the amended CT water to the MFT layer promoted the release of $\mathrm{Na}^{+}$, and more $\mathrm{Na}^{+}$ was released in the low flowrate system. No significant conductivity change in the release water of the low flowrate system was observed.
- For System 3, $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}$, and $\mathrm{SO}_{4}{ }^{2-}$ removal efficiencies were $24 \%, 14 \%$ and $36 \%$, respectively, in the high flowrate system; $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}$, and $\mathrm{SO}_{4}{ }^{2-}$ removal efficiencies were $39 \%, 26 \%$ and $62 \%$, respectively. In the low flowrate system, $\mathrm{Na}^{+}$increased $5 \%$ and $8 \%$ for the high flowrate and low flowrate systems, respectively. The conductivity of the high flowrate system and the low flowrate system was almost equal to the conductivity of the feed water.
- No removal of $\mathrm{Ca}^{2+}, \mathrm{Mg}^{2+}$ and $\mathrm{Na}^{+}$of WIP water (System 4) was observed in the high flowrate system or low flowrate system. $\mathrm{A} \mathrm{SO}_{4}{ }^{2-}$ removal efficiency of $57 \%$ and $49 \%$ was achieved in the high flowrate system and the low flowrate system, respectively. No conductivity reduction of the WIP water was observed in the high flowrate system. The conductivity removal efficiency of the low flowrate system was $10 \%$.
- For all systems fed with CT water, the low flow rates seemed more favorable for achieving higher removal efficiencies.
- The pH of the release water in all systems was in the range of 7.2 to 8.8 .
- MPN results indicated that in Systems 1 and 2, SRB outcompeted methanogens and resulted in a drastic decrease in the MPN of methanogens due to high $\mathrm{SO}_{4}{ }^{2-}$
concentrations. However, in System 3, addition of NaAc in the feed water supported the growth of both SRB and methanogens. MPN analyses also suggested microbial activity of both SRB and methanogens in columns with WIP feed water.
- It was found from the barrel and static column study that the size of the mesocosm had no apparent influence on the cation concentrations, but slight influence on $\mathrm{SO}_{4}{ }^{2-}$ concentration. CT water penetrating through MFT could enhance the densification of fine particles in MFT.

Because of the time limitation of this study, the flow rates used in the experiments were $6 \mathrm{~mL} / \mathrm{h}$ and $20 \mathrm{~mL} / \mathrm{h}$, and the incubation times were 140 days and 60 days, respectively. The lower the flow rate, the higher the removal efficiency. It would be useful to further study lower flow rates in order to investigate if there is a threshold value.

In this study, the flow rate of the feed water was controlled by a combination of valves and flow meter. In order to accurately control the flow rate, it is recommended to use a flow control pump to control the system flow.

Methanogenesis and sulfate reduction were important anaerobic biological processes in MFT deposition. However, MPN analysis can only estimate the number of the relevant microbial populations, but not the microbial activity. Therefore, it is also recommended to analyze the headspace gases in the dynamic columns to detect the methane and/or $\mathrm{H}_{2} \mathrm{~S}$ production, which could verify the occurrence of methanogenesis and/or sulfate reduction processes in the MFT.

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## Appendix A Dynamic System Results

A1 Results of chemical analysis in DH-1 system (sample type: ReIW)

| $\begin{gathered} \text { Sample } \\ \text { ID } \\ \hline \end{gathered}$ | Incub. Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}^{2+}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{2-}$ | $\begin{gathered} \mathbf{C O}_{3}{ }^{2-} \\ (\mathbf{w t ~ p p m}) \\ \hline \end{gathered}$ | $\begin{array}{\|c} \mathrm{HCO}_{3}^{-} \\ \text {(wt ppm) } \end{array}$ | $\begin{array}{\|c\|} \hline \text { Total } \\ \text { Cations } \\ \hline \end{array}$ | $\begin{gathered} \text { Total } \\ \text { Anions } \\ \hline \end{gathered}$ | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \mathrm{pH} \\ \hline \end{gathered}$ | $\mathrm{S}^{\mathbf{2 -}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DH-1-1 | 1 | 8.04 | 4350 | 49 | 1180 | 29.2 | 15.3 | 23.7 | 840 | 94 | 0.0 | 1510 | 54.51 | 50.98 | 1.07 | 8.21 | 28.5 |
| DH-1-3 | 2 | 8.33 | 4140 | 0 | 1070 | 16.5 | 12.2 | 17.5 | 800 | 61 | 0.0 | 1450 | 48.84 | 47.58 | 1.03 | 8.30 | 19.4 |
| DH-1-5 | 5 | 7.98 | 4270 | 0 | 1110 | 15.4 | 13.4 | 19.5 | 810 | 23 | 0.0 | 1530 | 50.75 | 48.37 | 1.05 | 7.87 | 8.7 |
| DH-1-6 | 6 | 7.66 | 4190 | 39 | 1120 | 16.4 | 15.2 | 23.1 | 800 | 40 | 0.0 | 1540 | 51.54 | 49.09 | 1.05 | 7.61 | 14.5 |
| DH-1-7 | 7 | 7.61 | 4430 | 160 | 1120 | 16.3 | 16.0 | 23.9 | 800 | 127 | 0.0 | 1520 | 51.64 | 52.07 | 0.99 | 7.67 | 37.1 |
| DH-1-8 | 8 | 7.64 | 4450 | 160 | 1090 | 15.7 | 15.3 | 23.1 | 800 | 119 | 0.0 | 1520 | 50.22 | 51.91 | 0.97 | 7.67 | 32.1 |
| DH-1-9 | 9 | 7.58 | 4290 | 140 | 1080 | 15.6 | 15.0 | 22.7 | 800 | 88 | 0.0 | 1550 | 49.74 | 51.51 | 0.97 | 7.65 | 25.9 |
| DH-1-10 | 10 | 7.57 | 4310 | 170 | 1090 | 15.5 | 15.2 | 22.7 | 800 | 127 | 0.0 | 1570 | 50.19 | 53.02 | 0.95 | 7.81 | 34.8 |
| DH-1-11 | 11 | 7.57 | 4340 | 170 | 1090 | 15.7 | 15.2 | 22.8 | 800 | 113 | 0.0 | 1510 | 50.20 | 51.74 | 0.97 | 7.62 | 29.5 |
| DH-1-12 | 12 | 7.59 | 4270 | 170 | 1170 | 16.9 | 16.4 | 24.6 | 780 | 98 | 0.0 | 1530 | 53.90 | 51.19 | 1.05 | 7.64 | 32.8 |
| DH-1-13 | 13 | 7.73 | 4180 | 180 | 1160 | 16.9 | 16.6 | 25.4 | 800 | 98 | 0.0 | 1640 | 53.52 | 53.69 | 1.00 | 7.63 | 30.2 |
| DH-1-14 | 14 | 7.66 | 4220 | 190 | 1150 | 16.8 | 16.7 | 25.6 | 780 | 94 | 0.0 | 1500 | 53.10 | 50.87 | 1.04 | 7.78 | 29.3 |
| DH-1-15 | 15 | 7.68 | 4290 | 230 | 1190 | 17.9 | 17.5 | 26.7 | 790 | 117 | 0.0 | 1510 | 54.99 | 52.28 | 1.05 | 7.65 | 36.1 |
| DH-1-16 | 16 | 7.64 | 4310 | 250 | 1170 | 17.1 | 17.1 | 26.3 | 800 | 142 | 0.0 | 1490 | 54.05 | 53.01 | 1.02 | 7.63 | 38.7 |
| DH-1-17 | 17 | 7.71 | 4330 | 290 | 1180 | 17.1 | 17.3 | 26.5 | 790 | 151 | 0.0 | 1470 | 54.51 | 53.08 | 1.03 | 7.79 | 43.4 |
| DH-1-18 | 18 | 7.66 | 4530 | 360 | 1190 | 17.1 | 17.7 | 27.3 | 790 | 173 | 0.0 | 1460 | 55.02 | 54.24 | 1.01 | 7.74 | 51.4 |
| DH-1-19 | 19 | 7.69 | 4500 | 400 | 1200 | 17.4 | 18.0 | 27.8 | 800 | 193 | 0.0 | 1450 | 55.51 | 55.26 | 1.00 | 7.68 | 55.5 |
| DH-1-20 | 20 | 7.78 | 4580 | 420 | 1230 | 17.7 | 18.2 | 27.8 | 790 | 200 | 0.0 | 1430 | 56.84 | 55.05 | 1.03 | 7.80 | 59.9 |
| DH-1-21 | 21 | 7.73 | 4650 | 410 | 1220 | 17.7 | 18.1 | 27.8 | 730 | 175 | 0.0 | 1440 | 56.40 | 52.88 | 1.07 | 7.67 | 55.4 |
| DH-1-22 | 22 | 7.59 | 4580 | 470 | 1220 | 18.2 | 18.6 | 28.6 | 790 | 197 | 0.0 | 1530 | 56.49 | 57.24 | 0.99 | 7.54 | 57.1 |
| DH-1-23 | 23 | 7.63 | 4750 | 510 | 1220 | 17.8 | 18.8 | 29.0 | 770 | 220 | 0.0 | 1420 | 56.52 | 55.85 | 1.01 | 7.66 | 66.0 |
| DH-1-24 | 24 | 7.54 | 4850 | 590 | 1220 | 17.8 | 19.2 | 29.7 | 780 | 241 | 0.0 | 1490 | 56.58 | 58.70 | 0.96 | 7.52 | 72.4 |
| DH-1-25 | 25 | 7.51 | 4720 | 670 | 1300 | 19.0 | 20.6 | 32.0 | 770 | 294 | 0.0 | 1430 | 60.33 | 59.53 | 1.01 | 7.48 | 80.5 |
| DH-1-26 | 26 | 7.66 | 4990 | 710 | 1310 | 19.0 | 20.5 | 32.2 | 770 | 273 | 0.0 | 1370 | 60.76 | 58.60 | 1.04 | 7.85 | 82.0 |
| DH-1-27 | 27 | 7.52 | 5350 | 820 | 1330 | 19.5 | 21.7 | 33.9 | 830 | 288 | 0.0 | 1360 | 61.83 | 61.80 | 1.00 | 8.12 | 98.1 |

$0 Z 1$

| 0.081 | $\varepsilon{ }^{\text {¢ }} \cdot \stackrel{L}{ }$ | 660 | S $\angle 9.9$ | 61＇9L | 0SOI | 00 | 819 | $08 L$ | $9.9 t$ | I6z | I＇zz | 0¢91 | 0002 | 0659 | $08^{\circ} \mathrm{L}$ | zs | zs－l－Ha |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 006 tl | £ั゙L | 860 | てE＇LL | $65^{\prime} S L$ | 0901 | $0 \cdot 0$ | IS9 | $0<L$ | 8 8t | $\dagger^{\dagger} 82$ |  | 0791 | 0002 | 0¢59 | $68^{\circ} \mathrm{L}$ | Is | IS－l－Ha |
| 0.6 tI | でし | 660 | £¢9L | $8 S^{\prime}$ S $L$ | $0<01$ | 00 | S09 | 0LL | 6 6t | £ $8 \%$ | $\varepsilon \downarrow$ | 0291 | 0002 | 0099 | $\varepsilon L^{\circ} L$ | OS | OS－I－HG |
| ${ }^{0} 161$ | LİL | 860 | 889 9 | Lて＇SL | 0＜01 | 00 | 279 | 0LL | $0 \cdot 9 t$ | 687 | 8 Iz | 0191 | $000 z$ | 0LS9 | $28^{\circ} \mathrm{L}$ | $6{ }^{6}$ | $6 \mathrm{t}-\mathrm{HC}$ |
| 0＇LLI | S2＇L | ＋6．0 | L6：08 | L8＇s | 0zil | 00 | $6 \angle 9$ | 608 |  |  | L．0z | 0 0¢91 | 0802 | $0 ¢ 89$ | ts＇L | $8{ }^{8}$ | 8t－l－HO |
| 0＇291 | I＇L | 960 | 6と＇6L | z¢＇9L | 020I | $00^{\circ}$ | S99 | ¢08 | $\downarrow$ ¢ | 8.2 | 802 | 0ヶ91 | ISOz | $09 \angle 9$ | ＋8． L | Lt | Lt－I－Ha |
| 0＇991 | $0{ }^{\circ} \mathrm{L}$ | 560 | L6\％ 6 | $86.5 L$ | 0011 | $0 \cdot 0$ | $8 \angle 9$ | 808 | tot | \％ 87 | $0^{\circ} \mathrm{I}$ | 0 0¢9 | 6202 | $07 \angle 9$ | $6 L^{\circ} \mathrm{L}$ | $9+$ | 9t－l－Ha |
| 0＇191 | £1＇L | S6． 0 | £964 | $85^{\circ} \mathrm{SL}$ | 0901 | $00^{\circ}$ | 789 | 618 | $\varepsilon$ ¢ | 188 | 「Iz | 0291 | £z0z | $00 \angle 9$ | E0＇8 | St | st－1－Ha |
| 0． 591 | \＄6． | $\angle 60$ | 66 LL | 06\％$¢$ | 0001 | 00 | 899 | 608 | 0 ¢ $\downarrow$ | 0.8 z | $z^{\prime \prime} \mathrm{I}$ | 0 0¢91 | £96I | $00 \angle 9$ | E0＇8 | to | tt－l－Ha |
| 0＇ZLI | $\varepsilon \underbrace{1} L$ | 86.0 | £6\％LL | 50．9L | 0＜01 | 00 | Ls9 | 218 | $0 \cdot \mathrm{st}$ | $9 \cdot 8 z$ | がIz | 0¢91 | 0861 | $00 \angle 9$ | $\pm 8 . \mathrm{L}$ | £ | Et－I－HG |
| 0 0＇SLI | $\varepsilon \Sigma^{\circ} \mathrm{L}$ | 96.0 | 198 L | $\varepsilon 9$ SL | sL0I | 00 | 299 | L18 | $0 \cdot \mathrm{St}$ | 887 | 「•Iz | 0791 | 6561 | 0t $\angle 9$ | $Z L L$ | てt | zt－I－Ha |
| 0981 | てIL | 660 | 0 OSL | £0＇s | $0{ }_{0} \mathrm{I}$ | 00 | SI9 | $08 L$ | L L L | －0¢ | z＇zz | 0091 | 0081 | 0959 | ILL | It | lt－I－HO |
| 0．981 | zでL | 660 | E9＇9L | L0．9L | 0911 | 00 | LI9 | 008 | 76t | 0．1E | 6 zz | 0791 | 0081 | 0¢t9 | 18.4 | $0{ }^{0}$ | 0t－I－HG |
| 094I | HL | 960 | 89 SL | 86． 2 | 0 LII | 00 | 119 | 008 | 8 加 | †「8z | $¢^{\prime} \mathrm{Iz}$ | 09¢1 | 0081 | 01 ¢9 | ＋5゙L | $6 \varepsilon$ | 6ع－І－Ha |
| 0685 | $9 \varepsilon^{\circ} \mathrm{L}$ | 860 | ¢ ¢ $て<$ | 5600 | 08 II | 00 | 6SS | $0<L$ | 1で | 9＇9z | 902 | 0zs | 0091 | 0819 | $t L L$ | $8 \varepsilon$ | 8ع－І－Ha |
| 0＇z91 | tic | $\angle 60$ | 0LTL | $9{ }^{\circ} \mathrm{OL}$ | 0911 | 00 | LtS | 008 | くで | $\tau \cdot L \tau$ | $0 \cdot 1 z$ | 00s | 0091 | 0129 | $L S \cdot L$ | $\llcorner\varepsilon$ | L£－І－HG |
| 0．651 | 61 L | $00 \cdot 1$ | 95：0L | £ $5^{\circ} 0 \mathrm{~L}$ | $0<11$ | 00 | 60S | 062 | でて | 8.97 | $80 \tau$ | 0ISI | 00 S I | 0809 | 9SL | $9 \varepsilon$ | 9ع－І－HG |
| 0．LSI | zz＇L | $00^{\circ} \mathrm{I}$ | $97^{\circ} \mathrm{L}$ | $8 \varepsilon^{\circ} \mathrm{LL}$ | 002I | 00 | 26t | 018 | 07t | 9.97 | ${ }^{1} 17$ | 0¢SI | 00SI | 096s | ZS＇L | ¢£ | ¢ع－І－Ha |
| 0.681 | $\angle t L$ | 960 | 0ヶ゙69 | 18.99 | 0t2I | $0 \cdot 0$ | tot | 008 | L•8 | $1 \cdot 9 z$ | Loz | 0¢tl | 00t 1 | 086s | $9 S^{\circ} \mathrm{L}$ | $\pm \varepsilon$ | เモ－І－Hの |
| 0＇ti | 98. | 660 | E699 | s199 | OtCI | $0 \cdot 0$ | でけ | 082 | でゅ $\varepsilon$ | ${ }^{0.97}$ | 802 | 02tl | 00EI | 0865 | $95^{\circ} \mathrm{L}$ | £์ | £ $\varepsilon$－І－Нの |
| 0＜EI | $9 \varepsilon^{\circ} \mathrm{L}$ | $\angle 60$ | £ $\chi^{\prime \prime} 89$ | S6S9 | 0szi | 00 | 97t | 018 | †68 | 8 Sz | ${ }^{\text {to }} 0$ | 0ıti | 00\＆I | 058S | くがL | zย | zع－І－нの |
| 062I | $09 . L$ | 860 | て1＇L9 | 08：59 | 06ZI | 00 | L8E | 088 | 18¢ | 6 ² |  | 0 tbl | 0021 | 0عLS | ZS．L | İ | เ $\varepsilon$－І－Hव |
| 0971 | IL•8 | 860 | 95： 9 | 9t゙t9 | 00.1 | ¢ 88 | 6LE | 088 | LLE | L＇って | $0.0 \tau$ | 08\＆1 | 0011 | 0t9s | LS＇L | ${ }_{0}$ | 0¢－I－HO |
| 0 LII | 19．2 | 10.1 | L0＇ャ9 | 0＜t＇9 | 00¢I | $0 \cdot 0$ | ISE | 078 | $\varepsilon \varsigma \varepsilon$ | $6 \cdot \mathcal{L}$ | 861 | $06 \varepsilon 1$ | 0001 | 0Lss | $6 t^{\circ} \mathrm{L}$ | 62 | 6z－І－Ha |
| 0． 21 | tS L $L$ | E0＇ | $L \tau^{\circ} ¢ 9$ | 61＇s9 | 0ャ\＆ | 00 | SIE | 0 088 | 898 |  | S 02 | 00tI | 076 | 09ts | $6 t^{\circ} \mathrm{L}$ | 82 | 82－1－HO |
| ${ }_{2} \mathrm{~S}$ | $\begin{aligned} & \mathrm{H}^{\mathrm{d}} \\ & \text { Y/V } \end{aligned}$ |  | $\begin{gathered} \text { suoliv } \\ \text { [Elou } \end{gathered}$ | $\begin{gathered} \text { suople] } \\ \text { [Elol } \end{gathered}$ |  |  | ${ }_{7}{ }^{\text {b }}$ | ． 10 | ${ }_{+z^{\text {B }} \text { P }}$ | $+z^{8} \mathrm{~N}$ | ${ }_{+}+$ | ${ }^{\text {E }} \mathbf{N}$ | $+^{19}$ | puos | $\mathrm{H}^{\text {d }}$ |  | $\underset{\text { ofdurs }}{\text { III }}$ |



## A1 Results of chemical analysis in DH-1 system (sample type: RelW) (cont'd)


A2 Results of chemical analysis in DH-2 system (sample type: RelW)

| $\begin{gathered} \text { Sample } \\ \text { ID } \\ \hline \end{gathered}$ | Incub. Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | Mg ${ }^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | Cr | $\mathrm{SO}_{4}{ }^{2}$ | $\begin{gathered} \mathbf{C O}_{3}{ }^{2-} \\ (\mathrm{wtpm}) \end{gathered}$ | $\begin{gathered} \mathbf{H C O}_{3}^{-} \\ \text {(wt ppm) } \\ \hline \end{gathered}$ | $\begin{gathered} \text { Total } \\ \text { Cations } \\ \hline \end{gathered}$ | $\begin{gathered} \text { Total } \\ \text { Anions } \\ \hline \end{gathered}$ | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \\ \hline \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \\ \hline \end{gathered}$ | $\mathrm{S}^{\mathbf{2}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DH-2-2 | d | 7.91 | 4280 | 0 | 1060 | 16.2 | 17.6 | 24.7 | 800 | 117 | 0 | 1540 | 49.20 | 50.23 | 0.98 | 7.87 | 34 |
| DH-2-3 | 3 | 7.88 | 4110 | 0 | 1150 | 17.0 | 16.6 | 24.7 | 800 | 67 | 0 | 1570 | 53.05 | 49.69 | 1.07 | 7.85 | 22 |
| DH-2-4 | 4 | 7.83 | 4120 | 0 | 1110 | 16.3 | 15.6 | 23.1 | 800 | 75 | 0 | 1550 | 51.13 | 49.51 | 1.03 | 7.84 | 24 |
| DH-2-5 | 5 | 7.76 | 4260 | 0 | 1070 | 15.8 | 15.8 | 23.4 | 790 | 139 | 0 | 1520 | 49.41 | 50.08 | 0.99 | 7.82 | 44 |
| DH-2-6 | 6 | 7.76 | 4320 | 0 | 1120 | 16.5 | 16.6 | 24.7 | 750 | 183 | 0 | 1500 | 51.74 | 49.57 | 1.04 | 7.80 | 61 |
| DH-2-7 | 7 | 7.90 | 4190 | 0 | 1180 | 17.7 | 18.0 | 27.3 | 790 | 220 | 0 | 1500 | 54.62 | 51.43 | 1.06 | 7.79 | 68 |
| DH-2-8 | 8 | 7.80 | 4260 | 0 | 1180 | 17.6 | 18.6 | 28.3 | 780 | 253 | 0 | 1480 | 54.72 | 51.51 | 1.06 | 7.73 | 82 |
| DH-2-9 | 9 | 7.76 | 4160 | 0 | 1230 | 18.1 | 19.3 | 29.5 | 780 | 269 | 0 | 1480 | 57.03 | 51.84 | 1.10 | 7.75 | 77 |
| DH-2-10 | 10 | 7.78 | 4220 | 0 | 1220 | 18.1 | 19.3 | 29.5 | 780 | 294 | 0 | 1470 | 56.59 | 52.20 | 1.08 | 7.74 | 91 |
| DH-2-11 | 11 | 7.97 | 4380 | 0 | 1240 | 18.5 | 19.9 | 30.4 | 790 | 317 | 0 | 1470 | 57.57 | 52.96 | 1.09 | 7.82 | 105 |
| DH-2-12 | 12 | 7.83 | 4360 | 0 | 1230 | 18.4 | 19.8 | 30.3 | 780 | 340 | 0 | 1440 | 57.12 | 52.66 | 1.08 | 7.81 | 115 |
| DH-2-13 | 13 | 7.83 | 4340 | 0 | 1240 | 18.5 | 20.0 | 30.6 | 780 | 363 | 0 | 1430 | 57.58 | 52.98 | 1.09 | 7.78 | 120 |
| DH-2-14 | 14 | 7.80 | 4380 | 0 | 1220 | 18.2 | 19.8 | 30.3 | 780 | 387 | 0 | 1410 | 56.68 | 53.15 | 1.07 | 7.87 | 129 |
| DH-2-15 | 15 | 7.75 | 4390 | 0 | 1220 | 18.2 | 20.3 | 31.0 | 780 | 415 | 0 | 1410 | 56.75 | 53.73 | 1.06 | 8.06 | 138 |
| DH-2-16 | 16 | 7.69 | 4460 | 0 | 1240 | 18.6 | 21.6 | 32.9 | 770 | 495 | 0 | 1370 | 57.83 | 54.46 | 1.06 | 7.71 | 164 |
| DH-2-17 | 17 | 7.69 | 4470 | 0 | 1240 | 18.6 | 22.2 | 33.5 | 770 | 529 | 0 | 1320 | 57.91 | 54.35 | 1.07 | 7.69 | 177 |
| DH-2-18 | 18 | 7.68 | 4400 | 0 | 1240 | 18.8 | 22.2 | 33.6 | 770 | 504 | 0 | 1350 | 57.93 | 54.32 | 1.07 | 7.70 | 168 |
| DH-2-19 | 19 | 7.63 | 4470 | 0 | 1250 | 19.0 | 22.7 | 34.3 | 770 | 518 | 0 | 1350 | 58.44 | 54.61 | 1.07 | 7.68 | 176 |
| DH-2-20 | 20 | 7.49 | 4450 | 0 | 1260 | 19.2 | 23.6 | 35.7 | 760 | 548 | 0 | 1330 | 59.03 | 54.63 | 1.08 | 7.82 | 190 |
| DH-2-21 | 21 | 7.44 | 4940 | 0 | 1230 | 19.7 | 26.7 | 39.1 | 830 | 657 | 0 | 1290 | 58.16 | 58.34 | 1.00 | 7.48 | 216 |
| DH-2-22 | 22 | 7.46 | 4850 | 0 | 1230 | 20.0 | 28.7 | 41.5 | 810 | 708 | 0 | 1250 | 58.46 | 58.11 | 1.01 | 7.57 | 235 |
| DH-2-23 | 23 | 7.40 | 4970 | 0 | 1240 | 20.5 | 31.4 | 45.2 | 810 | 778 | 0 | 1200 | 59.32 | 58.70 | 1.01 | 7.68 | 258 |
| DH-2-24 | 24 | 7.66 | 4990 | 0 | 1250 | 21.0 | 34.6 | 48.5 | 810 | 836 | 0 | 1180 | 60.19 | 59.58 | 1.01 | 7.80 | 276 |
| DH-2-25 | 25 | 7.52 | 4970 | 0 | 1260 | 21.3 | 37.4 | 39.7 | 780 | 850 | 0 | 1140 | 60.43 | 58.37 | 1.04 | 7.66 | 292 |
| DH-2-26 | 26 | 7.42 | 4990 | 0 | 1240 | 21.0 | 37.4 | 51.5 | 790 | 824 | 0 | 1170 | 60.14 | 58.60 | 1.03 | 7.54 | 280 |


| IIE | IE＇L | $0^{\circ} \mathrm{I}$ | $8{ }^{\circ} 09$ | てt＇09 | $0 ¢ 11$ | 0 | 2\＆6 | 008 | t＇ss | 0.85 | $\tau^{\prime} ¢ \tau$ | 00ZI | 0 | 096t | £1＇8 | IS | Is－z－HO |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| เ\＆E | $19 . L$ | $86^{\circ}$ | S6＇19 | 10＇19 | 0tI I | 0 | S66 | 008 | $\varsigma$ ¢ร | £ 9¢ | 6 tz | $61 z 1$ | 0 | 066t | $\pm 8 \cdot \mathrm{~L}$ | 0s | $0 \mathrm{c}-\mathrm{z}-\mathrm{HO}$ |
| $66 \varepsilon$ | でし | 10.1 | 66：8s | 14\％ 6 | 0til | 0 | 088 | 08 L | ع 89 | 8 zs | で¢ | 0811 | 0 | 0tos | $\varepsilon \varsigma^{\circ} \mathrm{L}$ | $6{ }^{6}$ | $6 t-\tau-\mathrm{Ha}$ |
| ISE | L6L | 260 | 50.09 | El＇ss | 092I | 0 | 0¢8 | 0LL | 7\％ 6 | L＇St | $0 \cdot$ \％ | 0011 | 0 | 056t | IS＇L | $8{ }^{\circ}$ | $8 \mathrm{t}-\mathrm{z}$－ HG |
| $87 \tau$ | $8 E^{\circ} \mathrm{L}$ | $\varepsilon 60$ | 67\％ 6 | Et＇ss | 0611 | 0 | Ss8 | 082 | 6.45 | 8 加 | げて | 01It | 0 | $006 t$ | $19 . L$ | $\angle t$ | Lt－z－HG |
| slz | StiL | $00^{\circ} \mathrm{I}$ | ti\％ 6 | 26．85 | 002I | 0 | 078 | 08 L | 9．95 | E0s | 0 －$\varepsilon$ | 08II | 0 | 088t | $68^{\circ} \mathrm{L}$ | $9{ }^{\text {d }}$ | 9t－z－HO |
| zsz | でL | 960 | $98 . L S$ | 09＇ss | 09II | 0 | 018 | 082 | で¢ | Stt | $0 \cdot 1 z$ | 0zil | 0 | $006 t$ | 78.2 | st | st－z－HG |
| 192 | $\angle E \cdot L$ | 660 | 8L＇8S | t08s | 09II | 0 | t58 | 082 | 5 ¢t |  | $0 \tau z$ | 08II | 0 | $016{ }^{\text {t }}$ | tis | tt | toz－HO |
| 192 |  | $00^{\circ} \mathrm{I}$ | 97：8s | t0 08 | 0121 | 0 | E08 | $0<1$ | 8 ¢ร | ILt | 8 \％ | 0LII | 0 | 028t | $58 . L$ | Et | $\varepsilon t-z-\mathrm{HO}$ |
| ££Z | 19. | ¢0＇I | 1ど85 | 80.09 | 09¢1 | 0 | Lt9 | 008 | L．9S | 8 8t | L＇ル | 0zzı | 0 | ozos | zs＇L | 2t | てt－z－H0 |
| 6 c | $8 \varepsilon^{\circ} \mathrm{L}$ | $00^{\circ} \mathrm{I}$ | 2で09 | －t＇09 | 0szI | 0 | 862 | 078 | 9 zs | z＇st | $6.1 z$ | 0¢zI | 0 | 0cos | $98^{\circ} \mathrm{L}$ | 10 | $\mathrm{t}-\mathrm{z}-\mathrm{HO}$ |
| 9¢Z | 29.1 | 20． | 95\％6S | 0＜09 | 092I | 0 | 662 | 062 | bts | $\varepsilon \angle L$ | $8^{\circ} \mathrm{I} Z$ | 0¢ZI | 0 | 0LOS | 68. | ${ }^{0}$ | $0 \mathrm{t}-\mathrm{z}-\mathrm{HO}$ |
| ${ }^{\text {t62 }}$ | $66^{\circ} \mathrm{L}$ | 20.1 | ＋66s | 8609 | 04 II | 0 | 206 | 08 L | ع＇6s | Lis | 9 zz | 0zzı | 0 | OtIS | 96 L | $6 \varepsilon$ | $6 \varepsilon-z-\mathrm{HO}$ |
| L0E | $1 \mathrm{H}^{\text {L }} \mathrm{L}$ | $00^{\circ} 1$ | SL＇19 | $8 \mathrm{t}^{\prime} 19$ | 0 LII | 0 | 666 | 062 | $0 \cdot 89$ | $\downarrow$ ¢¢ | 8 zz | 0zzı | 0 | 0tIs | 88 L | $8 \varepsilon$ | $8 \varepsilon-z-H \sigma$ |
| 662 | $00^{\circ} \mathrm{L}$ | $00^{\circ} \mathrm{I}$ | $85^{\circ}$ | 0＜${ }^{\circ} 19$ | 0601 | 0 | 0801 | 062 | L09 | S＇0s | て＇ı | 0tzı | 0 | 08IS | $86 . L$ | L£ | Lع－z－нの |
| 882 | $87^{\circ} \mathrm{L}$ | 960 | z9\％ 29 | 86.65 | 0601 | 0 | 0801 | 062 | £6S | L＇9t | 002 | 0ıZI | 0 | 002s | $98 . \mathrm{L}$ | $9 \varepsilon$ | $9 \varepsilon$－z－HO |
| $97 \varepsilon$ | 669 | 10.1 | てく＇19 | ャモ゙て9 | 0601 | 0 | 0S01 | $08 \angle$ | $\varsigma ¢ 9$ | L＇6t | $\varepsilon ̇ z \tau$ | oszı | 0 | 09IS | $t L L$ | $\stackrel{\varsigma}{ }$ | ¢ع－z－нの |
| $91 \varepsilon$ | L0＇L | 10.1 | LS＇19 | Lモ̇9 | 0 0¢II | 0 | 866 | 062 | 979 | L＇9t | でzz | 097I | 0 | ObIS |  | $\downarrow \varepsilon$ | ャะ－z－HG |
| $61 \varepsilon$ | 11 L | 20.1 | ャレ＇19 | LL＇z9 | 00II | 0 | 0801 | 062 | t＇z9 | £．9t | £zz | 0LZI | 0 | 0EOS | $99^{\circ} \mathrm{L}$ | £є | £ $\varepsilon$－z－Ha |
| $00 \varepsilon$ | to 2 | 66.0 | 28＇19 | 8609 | $0 \varepsilon \mathrm{E}_{\mathrm{I}}$ | 0 | 0101 | 062 | t8s | で¢ | がız | 0tzI | 0 | 010s | $L 9 . L$ | z | zع－z－Ha |
| 882 | 56．L | 96.0 | $60^{\circ} 19$ | 8L＇85 | $0 \varepsilon$ II | 0 | SL6 | 062 | 9 ts | て．0t | ¢0z | 00zI | 0 | 00IS | L9\％L | I¢ | І¢－z－HO |
| L0E | $\pm て ゙ L$ | 20.1 | LL＇09 | ${ }_{01}$ I 79 | 02.1 | 0 | ts6 | 008 | でLS | 9 It | L＇Iz | 0LZI | 0 | 000s |  | ${ }^{0}$ | 0¢－z－Ha |
| $\angle 82$ | $\varepsilon \varepsilon \cdot L$ | L60 | 88.09 | 8L＇85 | OSII | 0 | zz6 | 018 | tis | $0 \cdot L \varepsilon$ | £0z | 0ızI | 0 | 090s | ${ }^{\text {tS }} \mathrm{L} \mathrm{L}$ | 62 | 6z－z－HO |
| 682 | $5 \square^{\circ} \mathrm{L}$ | 20.1 | 5865 | 6019 | 0 LII | 0 | $0 \angle 8$ | 008 | 90s | L．8E | $\varsigma^{\prime} \mathrm{L}$ | 09zI | 0 | 010s | $6 \varepsilon^{\circ} \mathrm{L}$ | 82 | $88^{\text {8－z－Ha }}$ |
| $t \angle \tau$ | $\varepsilon \varsigma^{\circ} \mathrm{L}$ | $00^{-1}$ | 8L6\％ | 00.09 | 002I | 0 | $0 \varepsilon 8$ | 018 | tos | ¢．9E | $0^{\circ} \mathrm{I}$ I | 0 － 21 | 0 | 0s6t | 2 t L | $\angle Z$ | Lz－z－Hの |
| ${ }_{7} \mathrm{~S}$ | $\begin{gathered} \mathrm{H}^{\mathrm{d}} \\ \text { MTV } \end{gathered}$ |  | suo！uv ${ }^{[810, L}$ | suolib ${ }^{[810} \mathbf{C l}$ |  | $\begin{array}{\|c\|} \hline(\mathrm{udd} 3 \mathrm{M}) \\ -{ }_{-}^{2} \mathrm{O} \\ \hline \end{array}$ | $z^{\text {b }} \mathrm{OS}$ | －15 | $+2^{80}$ | $+z^{8} \mathbf{W}$ | ＋+ | ${ }^{\text {E }} \mathrm{N}$ | ${ }_{+}{ }^{\text {d }}$ | puod | $\mathrm{H}^{\text {d }}$ |  | $\underset{\text { Pdures }}{\text { aI }}$ |


A2 Results of chemical analysis in DH-2 system (sample type: RelW) (cont'd)

| $\underset{\text { ID }}{\text { Sample }}$ | Incub. Time (d) | pH | Cond | $\mathbf{B r}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{2}$ | $\begin{gathered} \mathrm{CO}_{3}{ }^{2-} \\ \text { (wt ppm) } \end{gathered}$ | $\begin{gathered} \mathbf{H C O}_{3}^{-} \\ \text {(wt ppm) } \end{gathered}$ | Total Cations | Total Anions | $\begin{aligned} & \text { Ratio } \\ & \text { Cat/Ani } \end{aligned}$ | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \\ \hline \end{gathered}$ | $\mathrm{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DH-2-52 | 52 | 8.06 | 4970 | 0 | 1180 | 23.9 | 57.0 | 56.4 | 790 | 795 | 0 | 1220 | 59.49 | 58.82 | 1.01 | 7.68 | 291 |
| DH-2-53 | 53 | 7.87 | 5020 | 0 | 1230 | 21.3 | 52.6 | 42.9 | 770 | 784 | 0 | 1210 | 60.55 | 57.86 | 1.05 | 7.70 | 260 |
| DH-2-54 | 54 | 7.68 | 5030 | 0 | 1225 | 21.8 | 53.7 | 50.6 | 800 | 854 | 0 | 1110 | 60.82 | 58.52 | 1.04 | 7.17 | 318 |
| DH-2-55 | 55 | 7.72 | 5060 | 0 | 1210 | 22.6 | 55.0 | 42.4 | 800 | 763 | 0 | 1100 | 59.89 | 56.46 | 1.06 | 7.21 | 270 |
| DH-2-56 | 56 | 7.65 | 5080 | 0 | 1180 | 21.7 | 54.0 | 43.1 | 770 | 700 | 0 | 1260 | 58.52 | 56.93 | 1.03 | 7.87 | 274 |
| DH-2-57 | 57 | 8.02 | 5130 | 0 | 1280 | 24.7 | 65.4 | 62.2 | 810 | 854 | 0 | 1190 | 64.85 | 60.17 | 1.08 | 7.28 | 328 |
| DH-2-58 | 58 | 8.00 | 5100 | 0 | 1260 | 24.8 | 65.7 | 63.9 | 800 | 897 | 0 | 1110 | 64.09 | 59.42 | 1.08 | 7.00 | 338 |
| DH-2-59 | 59 | 7.81 | 5130 | 0 | 1280 | 25.4 | 67.2 | 56.4 | 760 | 990 | 0 | 1130 | 64.72 | 60.56 | 1.07 | 7.20 | 360 |
| DH-2-60 | 60 | 7.83 | 5190 | 0 | 1290 | 25.3 | 69.5 | 51.2 | 770 | 1240 | 0 | 1280 | 65.09 | 68.51 | 0.95 | 8.30 | 408 |

A3 Results of chemical analysis in DH-3 system (sample type: RelW)

| Sample ID | Incub. Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{\text {2 }}$ | $\begin{gathered} \mathbf{C O}_{3}{ }^{2-} \\ (\mathbf{w t ~ p m}) \end{gathered}$ | $\begin{array}{r} \mathrm{HCO}_{3}^{-} \\ \text {(wt ppm } \end{array}$ | Total Cations | Total Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \\ \hline \end{gathered}$ | $\mathrm{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DH-3-3 | 3 | 7.92 | 6040 | 23 | 1036 | 16.5 | 15.8 | 23.6 | 810 | 93.6 | 0.0 | 1330 | 48.0 | 46.9 | 1.02 | 7.64 | 35.6 |
| DH-3-4 | 4 | 7.78 | 6180 | 45 | 1070 | 16.5 | 16.5 | 24.8 | 790 | 181 | 0.0 | 1360 | 49.6 | 48.9 | 1.01 | 7.35 | 67.1 |
| DH-3-5 | 5 | 7.84 | 6100 | 59 | 1057 | 16.4 | 16.7 | 25.2 | 790 | 233 | 0.0 | 1390 | 49.0 | 50.6 | 0.97 | 7.45 | 84.8 |
| DH-3-6 | 6 | 7.77 | 6500 | 60 | 1070 | 16.5 | 17.0 | 25.5 | 800 | 235 | 0.0 | 1400 | 49.6 | 51.1 | 0.97 | 7.78 | 85.7 |
| DH-3-7 | 7 | 7.78 | 6410 | 67 | 1058 | 16.3 | 16.9 | 25.5 | 790 | 247 | 0.0 | 1380 | 49.1 | 50.9 | 0.97 | 7.47 | 90.4 |
| DH-3-8 | 8 | 7.85 | 6290 | 68 | 1081 | 16.7 | 17.0 | 25.7 | 800 | 256 | 0.0 | 1380 | 50.1 | 51.3 | 0.98 | 7.48 | 93.2 |
| DH-3-9 | 9 | 7.74 | 6350 | 64 | 1070 | 16.5 | 17.3 | 26.5 | 770 | 239 | 0.0 | 1380 | 49.7 | 50.1 | 0.99 | 7.44 | 88.5 |
| DH-3-10 | 10 | 7.75 | 6070 | 66 | 1080 | 16.3 | 16.9 | 25.6 | 800 | 235 | 0.0 | 1380 | 50.1 | 50.9 | 0.98 | 7.41 | 85.6 |
| DH-3-11 | 11 | 7.78 | 5900 | 62 | 1070 | 16.4 | 16.7 | 25.3 | 800 | 222 | 0.0 | 1380 | 49.6 | 50.6 | 0.98 | 7.42 | 81.0 |
| DH-3-12 | 12 | 7.78 | 5980 | 61 | 1060 | 16.3 | 16.7 | 25.2 | 790 | 221 | 0.0 | 1390 | 49.2 | 50.4 | 0.98 | 7.45 | 80.3 |
| DH-3-13 | 13 | 7.75 | 5800 | 53 | 1050 | 16.0 | 15.7 | 24.1 | 800 | 156 | 0.0 | 1430 | 48.6 | 49.9 | 0.97 | 7.45 | 57.7 |
| DH-3-14 | 14 | 7.77 | 5910 | 54 | 1051 | 16.1 | 15.8 | 24.1 | 810 | 162 | 0.0 | 1450 | 48.6 | 50.6 | 0.96 | 7.50 | 57.1 |
| DH-3-15 | 15 | 7.54 | 5350 | 83 | 1150 | 17.2 | 17.2 | 26.9 | 790 | 246 | 0.0 | 1400 | 53.2 | 51.4 | 1.04 | 7.39 | 87.1 |
| DH-3-16 | 16 | 7.64 | 5430 | 75 | 1150 | 17.1 | 16.6 | 25.8 | 810 | 244 | 0.0 | 1420 | 53.1 | 52.1 | 1.02 | 7.47 | 76.5 |
| DH-3-17 | 17 | 7.64 | 5400 | 76 | 1130 | 16.9 | 16.6 | 25.7 | 810 | 254 | 0.0 | 1420 | 52.2 | 52.3 | 1.00 | 7.47 | 76.7 |
| DH-3-19 | 19 | 7.56 | 5540 | 110 | 1160 | 17.3 | 16.8 | 26.0 | 790 | 306 | 0.0 | 1360 | 53.6 | 52.3 | 1.02 | 7.40 | 108.0 |
| DH-3-20 | 20 | 7.55 | 5480 | 110 | 1150 | 17.4 | 16.9 | 26.1 | 790 | 305 | 0.0 | 1370 | 53.2 | 52.4 | 1.01 | 7.40 | 109.0 |
| DH-3-21 | 21 | 7.60 | 5560 | 99 | 1150 | 17.0 | 16.6 | 25.6 | 790 | 270 | 0.0 | 1330 | 53.1 | 50.9 | 1.04 | 7.45 | 96.9 |
| DH-3-22 | 22 | 7.61 | 5480 | 100 | 1150 | 17.2 | 16.6 | 25.5 | 800 | 271 | 0.0 | 1320 | 53.1 | 51.1 | 1.04 | 7.44 | 97.7 |
| DH-3-23 | 23 | 7.96 | 4860 | 92 | 1150 | 17.6 | 17.0 | 26.7 | 750 | 298 | 0.0 | 1380 | 53.2 | 51.1 | 1.04 | 7.61 | 101.0 |
| DH-3-24 | 24 | 7.94 | 4850 | 94 | 1165 | 17.9 | 17.0 | 26.6 | 760 | 293 | 0.0 | 1400 | 53.9 | 51.6 | 1.04 | 7.67 | 102.0 |
| DH-3-25 | 25 | 7.89 | 4890 | 120 | 1180 | 18.0 | 17.5 | 27.2 | 750 | 300 | 0.0 | 1370 | 54.6 | 51.3 | 1.06 | 7.65 | 121.0 |
| DH-3-26 | 26 | 7.86 | 4840 | 120 | 1170 | 17.8 | 17.4 | 27.0 | 760 | 303 | 0.0 | 1370 | 54.1 | 51.7 | 1.05 | 7.79 | 119.0 |
| DH-3-27 | 27 | 7.72 | 4890 | 120 | 1160 | 17.3 | 17.2 | 26.7 | 760 | 305 | 0.0 | 1380 | 53.6 | 51.9 | 1.03 | 7.62 | 117.0 |
| DH-3-28 | 28 | 7.89 | 4890 | 120 | 1180 | 17.3 | 17.1 | 26.9 | 760 | 312 | 37.8 | 1370 | 54.5 | 53.1 | 1.03 | 8.46 | 121.0 |
| DH-3-29 | 29 | 7.70 | 5210 | 160 | 1150 | 17.0 | 17.2 | 26.5 | 700 | 400 | 0.0 | 1310 | 53.2 | 51.5 | 1.03 | 7.34 | 160.0 |

A3 Results of chemical analysis in DH-3 system (sample type: RelW) (cont'd)

| Sample ID | Incub. <br> Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{\mathbf{2 -}}$ | $\underset{(\mathbf{w t ~ p p m})}{\mathrm{CO}_{3}{ }^{\mathbf{2 -}}}$ | $\begin{array}{\|c\|} \mathbf{H C O}_{3}^{-} \\ (\mathbf{w t ~ p p m}) \\ \hline \end{array}$ | Total <br> Cations | Total <br> Anions | $\begin{array}{\|c\|} \text { Ratio } \\ \text { Cat/Ani } \\ \hline \end{array}$ | $\begin{gathered} \text { ALK } \\ \ldots \mathbf{p H} \end{gathered}$ | $\mathrm{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DH-3-30 | 30 | 7.70 | 5220 | 160 | 1150 | 16.6 | 17.1 | 26.4 | 700 | 400 | 0.0 | 1260 | 53.2 | 50.7 | 1.05 | 7.34 | 154.0 |
| DH-3-31 | 31 | 7.72 | 5180 | 160 | 1150 | 16.7 | 17.1 | 26.5 | 720 | 382 | 0.0 | 1360 | 53.2 | 52.5 | 1.01 | 7.67 | 145.0 |
| DH-3-32 | 32 | 7.70 | 5230 | 170 | 1130 | 16.6 | 17.0 | 26.3 | 720 | 390 | 0.0 | 1310 | 52.3 | 52.0 | 1.01 | 7.62 | 145.0 |
| DH-3-33 | 33 | 7.88 | 5240 | 180 | 1130 | 16.5 | 17.0 | 26.3 | 720 | 413 | 0.0 | 1300 | 52.3 | 52.4 | 1.00 | 7.55 | 151.0 |
| DH-3-34 | 34 | 7.89 | 5270 | 200 | 1170 | 16.8 | 17.8 | 27.1 | 720 | 431 | 0.0 | 1290 | 54.1 | 52.9 | 1.02 | 7.63 | 164.0 |
| DH-3-35 | 35 | 7.88 | 5270 | 190 | 1130 | 16.5 | 17.3 | 26.4 | 710 | 432 | 0.0 | 1300 | 52.3 | 52.7 | 0.99 | 7.66 | 158.0 |
| DH-3-36 | 36 | 7.94 | 5290 | 200 | 1110 | 16.1 | 16.9 | 25.9 | 720 | 444 | 0.0 | 1290 | 51.4 | 53.2 | 0.97 | 7.60 | 155.0 |
| DH-3-37 | 37 | 7.92 | 5280 | 200 | 1130 | 16.6 | 17.4 | 26.6 | 730 | 444 | 0.0 | 1280 | 52.3 | 53.3 | 0.98 | 7.60 | 160.0 |
| DH-3-38 | 38 | 8.27 | 5330 | 240 | 1210 | 17.6 | 20.2 | 31.8 | 740 | 566 | 0.0 | 1190 | 56.3 | 55.1 | 1.02 | 7.30 | 198.0 |
| DH-3-39 | 39 | 8.32 | 5320 | 240 | 1190 | 17.2 | 19.9 | 31.1 | 740 | 564 | 0.0 | 1175 | 55.4 | 54.9 | 1.01 | 7.11 | 198.0 |
| DH-3-40 | 40 | 8.17 | 5340 | 240 | 1230 | 17.5 | 20.0 | 31.2 | 750 | 528 | 0.0 | 1320 | 57.2 | 56.8 | 1.01 | 7.76 | 187.0 |
| DH-3-41 | 41 | 8.18 | 5340 | 240 | 1240 | 17.5 | 19.9 | 31.0 | 740 | 534 | 0.0 | 1230 | 57.6 | 55.1 | 1.04 | 7.63 | 186.0 |
| DH-3-42 | 42 | 8.15 | 5290 | 240 | 1230 | 17.5 | 19.5 | 30.4 | 750 | 527 | 0.0 | 1250 | 57.1 | 55.6 | 1.03 | 7.65 | 184.0 |
| DH-3-43 | 43 | 8.13 | 5320 | 240 | 1220 | 17.3 | 19.1 | 29.7 | 740 | 524 | 0.0 | 1250 | 56.6 | 55.3 | 1.02 | 7.68 | 180.0 |
| DH-3-44 | 44 | 7.88 | 5260 | 260 | 1220 | 18.9 | 19.2 | 27.1 | 790 | 494 | 0.0 | 1510 | 56.5 | 60.5 | 0.93 | 8.11 | 162.0 |
| DH-3-45 | 45 | 7.92 | 5250 | 250 | 1190 | 17.1 | 18.6 | 28.1 | 770 | 500 | 0.0 | 1310 | 55.1 | 56.7 | 0.97 | 7.22 | 160.0 |
| DH-3-46 | 46 | 7.95 | 5310 | 250 | 1140 | 16.4 | 17.9 | 27.4 | 770 | 545 | 0.0 | 1180 | 52.8 | 55.5 | 0.95 | 7.56 | 169.0 |
| DH-3-47 | 47 | 7.94 | 5320 | 250 | 1150 | 16.2 | 17.7 | 27.0 | 760 | 538 | 88.8 | 1090 | 53.2 | 56.6 | 0.94 | 8.80 | 168.0 |
| DH-3-48 | 48 | 7.70 | 5310 | 240 | 1170 | 16.8 | 18.1 | 28.0 | 760 | 653 | 0.0 | 1120 | 54.2 | 56.4 | 0.96 | 7.62 | 210.0 |
| DH-3-49 | 49 | 7.70 | 5320 | 240 | 1170 | 17.0 | 18.4 | 28.5 | 770 | 665 | 0.0 | 1140 | 54.3 | 57.2 | 0.95 | 7.60 | 213.0 |
| DH-3-50 | 50 | 8.63 | 5290 | 270 | 1200 | 19.0 | 20.3 | 31.3 | 760 | 520 | 0.0 | 1360 | 55.9 | 57.9 | 0.97 | 7.70 | 305.0 |
| DH-3-51 | 51 | 8.47 | 5410 | 270 | 1200 | 19.0 | 20.4 | 31.3 | 770 | 399 | 0.0 | 1350 | 55.9 | 55.5 | 1.01 | 7.54 | 196.0 |
| DH-3-52 | 52 | 8.50 | 5410 | 270 | 1240 | 18.8 | 20.2 | 30.7 | 770 | 384 | 0.0 | 1440 | 57.6 | 56.7 | 1.02 | 7.82 | 198.0 |
| DH-3-53 | 53 | 8.49 | 5400 | 270 | 1260 | 18.6 | 19.7 | 29.7 | 780 | 441 | 0.0 | 1360 | 58.4 | 56.8 | 1.03 | 7.70 | 219.0 |
| DH-3-54 | 54 | 8.48 | 5380 | 270 | 1270 | 19.0 | 20.4 | 30.9 | 770 | 444 | 0.0 | 1360 | 58.9 | 56.6 | 1.04 | 7.81 | 217.0 |
| DH-3-55 | 55 | 8.01 | 5400 | 250 | 1290 | 18.0 | 19.1 | 29.4 | 760 | 636 | 0.0 | 1170 | 59.6 | 57.0 | 1.05 | 7.75 | 214.0 |

A3 Results of chemical analysis in DH-3 system (sample type: RelW) (cont'd)

| Sample ID | Incub. <br> Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{\text {2- }}$ | $\begin{gathered} \mathrm{CO}_{3}{ }^{2-} \\ (\text { wt ppm }) \end{gathered}$ | $\begin{gathered} \mathrm{HCO}_{3}^{-} \\ (\text {wt ppm) } \end{gathered}$ | Total Cations | Total Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \\ \hline \end{gathered}$ | $\mathbf{S}^{\mathbf{2}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DH-3-56 | 56 | 8.01 | 5420 | 260 | 1270 | 17.9 | 19.1 | 29.3 | 760 | 641 | 0.0 | 1140 | 58.7 | 56.7 | 1.04 | 7.69 | 215.0 |
| DH-3-57 | 57 | 8.42 | 5280 | 260 | 1265 | 17.5 | 17.8 | 26.5 | 760 | 301 | 0.0 | 1540 | 58.3 | 56.2 | 1.04 | 7.75 | 167.0 |
| DH-3-58 | 58 | 8.40 | 5350 | 280 | 1220 | 15.8 | 16.3 | 23.4 | 770 | 341 | 0.0 | 1430 | 56.0 | 55.7 | 1.00 | 7.68 | 119.0 |
| DH-3-59 | 59 | 8.47 | 5360 | 300 | 1240 | 18.8 | 19.3 | 26.0 | 830 | 495 | 0.0 | 1410 | 57.3 | 60.6 | 0.95 | 7.80 | 166.0 |
| DH-3-60 | 60 | 8.37 | 5470 | 300 | 1270 | 18.7 | 19.6 | 25.7 | 820 | 600 | 0.0 | 1370 | 58.6 | 61.8 | 0.95 | 8.03 | 205.0 |

A4 Results of chemical analysis in DH-4 system (sample type: RelW)

| $\begin{gathered} \text { Sample } \\ \hline \end{gathered}$ | Incub. <br> Time <br> (d) | pH | Cond | Br ${ }^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | Mg ${ }^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | Cr | $\mathrm{SO}_{4}{ }^{2}$ | $\begin{gathered} \mathbf{C O}_{3}{ }^{2-} \\ (\mathbf{w t ~ p p m}) \\ \hline \end{gathered}$ | $\mathrm{HCO}_{3}$ (wt ppm) | $\begin{array}{\|c\|} \hline \text { Total } \\ \text { Cations } \\ \hline \end{array}$ | Total Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \end{gathered}$ | $\mathbf{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DH-4-3 | 3 | 8.04 | 5540 | 11 | 1100 | 16.7 | 15.2 | 23.0 | 830 | 19.9 | 0.0 | 1550 | 50.7 | 49.3 | 1.03 | 7.62 | 8.7 |
| DH-4-4 | 4 | 7.74 | 5740 | 24 | 1090 | 16.6 | 15.6 | 24.0 | 830 | 35.3 | 0.0 | 1500 | 50.3 | 49.0 | 1.03 | 7.47 | 14.7 |
| DH-4-5 | 5 | 7.76 | 5540 | 24 | 1110 | 16.8 | 15.5 | 23.9 | 830 | 40.1 | 0.0 | 1500 | 51.2 | 49.1 | 1.04 | 7.48 | 14.8 |
| DH-4-6 | 6 | 7.72 | 5560 | 42 | 1110 | 16.7 | 15.6 | 24.1 | 820 | 57.6 | 0.0 | 1450 | 51.2 | 48.6 | 1.05 | 7.48 | 22.9 |
| DH-4-7 | 7 | 7.73 | 5450 | 43 | 1100 | 16.7 | 15.5 | 24.0 | 830 | 58.0 | 0.0 | 1490 | 50.7 | 49.6 | 1.02 | 7.52 | 22.9 |
| DH-4-8 | 8 | 7.60 | 5090 | 53 | 1110 | 16.7 | 15.2 | 23.3 | 830 | 63.4 | 0.0 | 1460 | 51.1 | 49.3 | 1.04 | 7.51 | 24.5 |
| DH-4-9 | 9 | 7.60 | 5050 | 52 | 1110 | 16.5 | 15.6 | 24.0 | 830 | 64.2 | 0.0 | 1470 | 51.2 | 49.5 | 1.03 | 7.52 | 24.6 |
| DH-4-10 | 10 | 7.76 | 5150 | 52 | 1100 | 16.3 | 15.1 | 23.2 | 830 | 62.0 | 0.0 | 1470 | 50.7 | 49.4 | 1.03 | 7.59 | 24.1 |
| DH-4-11 | 11 | 7.62 | 5130 | 100 | 1080 | 16.4 | 14.5 | 22.5 | 820 | 102 | 0.0 | 1360 | 49.7 | 48.8 | 1.02 | 7.55 | 38.3 |
| DH-4-12 | 12 | 7.61 | 5140 | 100 | 1090 | 16.3 | 14.5 | 22.4 | 820 | 104 | 0.0 | 1350 | 50.1 | 48.6 | 1.03 | 7.48 | 38.6 |
| DH-4-13 | 13 | 7.61 | 5230 | 100 | 1090 | 16.6 | 14.5 | 22.4 | 820 | 102 | 0.0 | 1380 | 50.1 | 49.1 | 1.02 | 7.53 | 39.1 |
| DH-4-14 | 14 | 7.74 | 5170 | 100 | 1090 | 15.9 | 14.3 | 22.1 | 810 | 102 | 0.0 | 1350 | 50.1 | 48.3 | 1.04 | 7.56 | 38.1 |
| DH-4-15 | 15 | 7.73 | 5110 | 100 | 1090 | 15.9 | 14.4 | 22.1 | 820 | 104 | 0.0 | 1350 | 50.1 | 48.6 | 1.03 | 7.54 | 38.3 |
| DH-4-16 | 16 | 7.93 | 4520 | 110 | 1070 | 16.3 | 14.5 | 22.5 | 790 | 91.3 | 0.0 | 1360 | 49.3 | 47.8 | 1.03 | 7.78 | 37.3 |
| DH-4-17 | 17 | 7.99 | 4540 | 110 | 1130 | 16.8 | 14.7 | 22.7 | 800 | 91.7 | 0.0 | 1390 | 51.9 | 48.6 | 1.07 | 7.74 | 38.2 |
| DH-4-18 | 18 | 8.08 | 4460 | 120 | 1090 | 16.6 | 14.4 | 22.3 | 790 | 94.1 | 0.0 | 1380 | 50.1 | 48.3 | 1.04 | 7.81 | 39.2 |
| DH-4-19 | 19 | 8.07 | 4460 | 120 | 1090 | 16.5 | 14.4 | 22.2 | 790 | 95.1 | 0.0 | 1350 | 50.1 | 47.9 | 1.05 | 7.82 | 38.8 |
| DH-4-20 | 20 | 7.95 | 4510 | 120 | 1130 | 17.3 | 14.7 | 22.5 | 800 | 95.0 | 0.0 | 1340 | 51.9 | 48.0 | 1.08 | 7.84 | 40.3 |
| DH-4-21 | 21 | 8.09 | 4330 | 140 | 1110 | 16.3 | 14.1 | 21.7 | 800 | 92.6 | 0.0 | 1330 | 50.9 | 48.0 | 1.06 | 7.72 | 38.1 |
| DH-4-22 | 22 | 7.82 | 4700 | 130 | 1060 | 15.5 | 13.5 | 20.7 | 760 | 88.0 | 0.0 | 1310 | 48.6 | 46.3 | 1.05 | 7.68 | 36.1 |
| DH-4-23 | 23 | 7.78 | 4700 | 150 | 1060 | 15.3 | 13.2 | 20.1 | 780 | 96.4 | 0.0 | 1290 | 48.6 | 47.0 | 1.03 | 7.64 | 38.7 |
| DH-4-24 | 24 | 7.76 | 4700 | 150 | 1050 | 14.9 | 13.3 | 20.6 | 780 | 97.6 | 0.0 | 1290 | 48.2 | 47.0 | 1.02 | 7.91 | 38.3 |
| DH-4-25 | 25 | 7.75 | 4670 | 170 | 1060 | 14.8 | 13.0 | 20.3 | 770 | 97.5 | 0.0 | 1310 | 48.6 | 47.3 | 1.03 | 7.70 | 37.9 |
| DH-4-26 | 26 | 7.76 | 4670 | 160 | 1060 | 15.0 | 13.0 | 20.1 | 780 | 96.2 | 0.0 | 1320 | 48.6 | 47.6 | 1.02 | 7.68 | 38.2 |
| DH-4-27 | 27 | 7.68 | 4650 | 160 | 1060 | 15.0 | 12.8 | 19.9 | 770 | 97.2 | 0.0 | 1290 | 48.5 | 46.9 | 1.04 | 7.56 | 38.1 |


| £ $\downarrow$ ¢ | ILL | $0^{\circ} \mathrm{I}$ | ［＇L | $0 \cdot \angle$ | 0 OSI | 00 | 801 | 062 | t＇81 | S．II | 9 q ¢ | 0¢01 | $00 \varepsilon$ | 0t9t | $96 . L$ | 25 | 7s－t－HG |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| ¢ 88 | tScL | 460 | $6 \cdot 9$ | sst | 02 II | 00 | tol | $08 L$ | †91 | LOL | て¢ı | 0001 | 062 | 099t | $88 . \mathrm{L}$ | IS | Is－t－HC |
| $\tau \cdot 8 \varepsilon$ | $9 L^{\circ} \mathrm{L}$ | 860 | $\varsigma 9 t$ | 9 St | 08II | 00 | 666 | 092 | $\tau<1$ | ［＇II | $\varsigma ¢ \varepsilon$ | 0001 | 062 | 0¢9t | $88^{\circ} \mathrm{L}$ | 05 | $0 \mathrm{~s}-\mathrm{t}-\mathrm{HC}$ |
|  | 90゙L | 860 | 89 | $0 \cdot 9$ | 06 II | 00 | $\llcorner\cdot 96$ | OLL | でLI | III | $9 \varepsilon 1$ | 0101 | 062 | 0S9t | E8 ${ }^{\circ} \mathrm{L}$ | 60 | $6 \mathrm{t}-\mathrm{t}$－ HG |
| 8 \％ 1 ¢ | 29\％ | 660 | L＇9t | ［9t | 0811 | 00 | 2.66 | $0 \angle L$ | L LI | がい | 6 El | 0101 | 062 | 059t | 58L | $8{ }^{+}$ | $8 \mathrm{t}-\mathrm{t}-\mathrm{HC}$ |
| 9 9 $\downarrow 2$ | L9\％ | L60 | 8＇97 | s＇st | 0 IZI | 0.0 | $0 \mathcal{L}$ | 08L | $\tau \cdot L$ | て＇II | L¢ı | 866 | 082 | 00St | 96.4 | $\angle t$ | $\angle \mathrm{t}-\mathrm{t}$－HG |
| 0 0．sz | セLL | $66^{\circ}$ | 0＇Lt | 9.97 | 0zzı | $0 \cdot 0$ | szL | 08 L | 6LL | $\varsigma^{\prime \prime}$ | 0 ¢ 1 | 0201 | $08 z$ | 0z9t | 96.4 | $9 t$ | $9 \mathrm{t}-\mathrm{t}$－ HO |
| 888 | $65^{\circ} \mathrm{L}$ | 20.1 | 6 什 | 9．st | 0 OLI | $0 \cdot 0$ | 0 0．8 | 09 L | 9 LI | $\varepsilon \pm$ | L¢ $¢$ | 666 | $0<z$ | 009t | 68 L | st | St－t－Hd |
| ＋6\％ | 19.4 | 86.0 | $\varsigma 9 t$ | LSt | 0121 | 00 | I＇t8 | $0 L L$ | 8 LI | $9 \cdot 11$ | 6 EL | 0001 | $09 z$ | 0tSt | $8 L^{\circ} \mathrm{L}$ | to | tot－HC |
| $\varepsilon \% \%$ | 09.2 | 86.0 | 0.97 | でSt | 081I | 0.0 | 9＇48 | $0 L L$ | $\dagger$ ¢ | $\varepsilon{ }^{\prime \prime}$ | $9 \varepsilon \mathrm{~L}$ | 066 | 092 | 019t | $8 L^{\circ} \mathrm{L}$ | $\varepsilon{ }^{\text {t }}$ | ¢t－t－Ha |
| $\tau て \varepsilon$ | 09.2 | 16.0 | て＇St | 8 ¢ $\downarrow$ | 0121 | 0.0 | 6.88 | 0tL | ZLL | 0 II | $0 \varepsilon 1$ | 096 | 0zz | 0LSt | ILLL | 2t | てr－b－Ha |
| $\varsigma ¢ \mathcal{L}$ | 9 $9^{\circ} \mathrm{L}$ | 00.1 | ISt | $\varepsilon$ ¢ $\dagger$ | 0611 | $00^{\circ}$ | £＇\％8 | OSL | 181 | 9 II | 9 EI | 066 | $0 z z$ | 06St | ELL | It | It －-Ha |
| $8 \angle E$ | 89. | 960 | £ 9 ${ }^{\text {¢ }}$ | $\varepsilon \downarrow \square$ | 0szi | 00 | 866 | OSL | 8 Ll | ナ！ | 1＇ย1 | 896 | 012 | 009t | L8． | ${ }^{0}$ | 0t－t－ra |
| 9 CE | ＋9 2 | $\angle 60$ | $0 \cdot 9$ | t＇tt | $0<Z 1$ | 00 | 6 ¢6 | 0\＆L | 181 | S＇II | で๕ | $0<6$ | $01 z$ | 08tr | $98 . \mathrm{L}$ | $6 \varepsilon$ | $6 \varepsilon$－-HO |
| 9＇ท | $89^{\circ} \mathrm{L}$ | 860 | I＇St | tot | 0121 | 00 | ¢ 88 | 0tL | $6 \angle 1$ | がい | İ£ | $0<6$ | 012 | 08St | 68.2 | $8 \varepsilon$ | $8 \varepsilon \cdot \downarrow$－HG |
| で9E | $65^{\circ} \mathrm{L}$ | 20.1 | 0．st | 8.5 | 002I | 00 | ع：88 | 0tL | $\varepsilon 61$ | でて | 9 9ı | 0001 | 0iz | 08St | LLL | $\angle \varepsilon$ | L\＆－t－HG |
| $90 \varepsilon$ | Sc L | 860 | $88 t$ | $0.8 t$ | 0szl | 00 | ${ }^{\text {¢ }}$ L6 | $0 ¢ 8$ | 881 | 0 0ı | 0 0t | 0s01 | 0tz | 06St | $S L \cdot L$ | $9 \varepsilon$ | 9£－$\downarrow$－ HO |
| $00 \varepsilon$ | $15 \cdot L$ | 660 | $98 t$ | 6 Lt | Otz 1 | 00 | $\varsigma^{\text {c }} 16$ | 088 | 981 | 611 | 6 ¢ | OSOL | 0ヶて | 019t | $8 L^{\circ} \mathrm{L}$ | ¢ $\varepsilon$ | ¢ $\varepsilon$－$\downarrow$ Ha |
| 6 ¢ 5 | 0LL | 660 | $8.8 \dagger$ | t－8t | 0tzI | 00 | 801 | 088 | 681 | 0 ZI | $8 \cdot \varepsilon 1$ | 0901 | 0\＆z | 009t | SLL | $\downarrow \varepsilon$ | เ¢－$\downarrow$－ HG |
| ¢＇SE | SLLL | 960 | 688 |  | oczi | 00 | L01 | 078 | £ 81 | $8{ }^{\text {8 }}$ I | $8 \cdot \mathcal{1}$ | 0¢0I | 0\＆z | 019t | ¢8\％$L$ | $\varepsilon \varepsilon$ | £ ¢－b－HG |
|  | 192 | 860 | $\varepsilon 6 t$ | 5 5 | 0szI | 00 | 811 | 088 | L81 | 0 ZI | IttI | 0901 | $0 \downarrow \tau$ | 029t | $6 L^{\circ} \mathrm{L}$ | て¢ | z£－t－Hd |
| $6 \cdot \varepsilon \varepsilon$ | 69.2 | 201 | ［8t | 685 | 002I | 00 | 811 | 078 | $\varepsilon 61$ | $\varepsilon \not \subset 1$ | $\varepsilon \downarrow \square$ | 0LOI | 0\＆z | 029t | $78 . \mathrm{L}$ | IE | I $\varepsilon$－-HG |
| S＇LE | $\varepsilon L L$ | 20.1 | L＇9t | 96 | 0szI | 00 | 0.26 | $08 L$ | ＋61 | s＇z | $s+1$ | 0＋01 | 081 | 019t | $68{ }^{\circ}$ | ${ }^{0 \varepsilon}$ | $0 \varepsilon-b-\mathrm{Ha}$ |
| 99\％ | ILL | 860 |  | L＇9t | 06ZI | $0 \cdot 0$ | †86 | $08 L$ | 681 | £＇ı | でゅI | 0z01 | 081 | 099t | $\angle 8.2$ | $6 乙$ | $6 \tau-\mathrm{t}-\mathrm{HO}$ |
| I 8 ¢ | 19． L | ＋0． 1 | 8.97 | $9.8 t$ | 092I | 00 | 8.26 | $08 L$ | zoz |  | 0 ¢ s | 0901 | $0<1$ | 099b | L9\％ | 82 | $82-t-\mathrm{HC}$ |
| ${ }_{-2} \mathrm{~S}$ | $\begin{gathered} \mathrm{H}^{\mathrm{d}} \\ \text { y/V } \end{gathered}$ | $\begin{aligned} & \text { !पY ABJ } \\ & \text { opey } \end{aligned}$ | $\begin{aligned} & \text { suo!uv } \\ & \text { [BłoIU } \end{aligned}$ | $\begin{gathered} \text { suope] } \\ \text { [घto } \end{gathered}$ |  |  | ${ }_{-}^{5} \mathrm{OS}$ | 13 | ${ }_{+2^{\text {E }} \text { D }}$ | $+2^{8} \mathrm{~N}$ | ＋${ }^{\text {Y }}$ | ${ }_{+}^{\text {E }}$ N | $+{ }^{18}$ | puo？ | $\mathrm{H}^{\text {d }}$ | $\begin{gathered} (\mathbf{p}) \\ \substack{\text { כwilit } \\ \text { qnoul }} \end{gathered}$ | $\underset{{ }_{\text {Pdures }}}{\mathbf{d I I}}$ |



| 9 \％$\downarrow$ | 0¢：8 | 20.1 | L＇9t | 8 $2 t$ | $0 ¢$ II | 00 | 821 | 08 L | † $\llcorner$ | 0．II | L＇zI | OSOL | 082 | 058t | 06. | 09 | 09－t－HO |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| I＇st | Sc8 | 20.1 | S＇Lt | $L \cdot 8 t$ | 09 II | $0 \cdot 0$ | 8 ¢ | 06 L | ［81 | £ル | 0 ¢ $\varepsilon$ | 0201 | $0 \angle Z$ | 028t | $88^{\circ} \mathrm{L}$ | 65 | $6 \mathrm{~S}-\mathrm{t} \mathrm{HO}$ |
| t－st | LE8 | to 1 | $1 \cdot \mathrm{~L}$ | 16t | 0601 | I＇$\varepsilon$ | StI | 08 L | 081 | ャII | でદ | 0801 | 082 | 066t | $98 . \mathrm{L}$ | ZL | $8 s^{-t-H 0}$ |
| $68 \varepsilon$ | L9．L | $66^{\circ}$ | 1．8t | 8.2 | OtII | 00 | Sti | 008 | 081 |  | でદı | OSOI | 018 | 0ZIt | \＆6． 2 | Ls | Ls－t－HO |
| $0 \cdot 6 \varepsilon$ | 16.4 | 00.1 | $て ゙ く t$ |  | 0zII | 00 | 621 | 06 L | 081 | でI | 6 ZI | 0t0I | 01E | 029t | $98^{\circ} \mathrm{L}$ | 95 | 9s－t－HO |
| $0 \cdot 6 \varepsilon$ | LLLL | 660 | $\varepsilon \angle t$ | 6.97 | 0¢ı | 00 | szı | $06 L$ | L LI | ITI | 6 ZI | 0ع0I | $01 \varepsilon$ | 089t | $\angle 8.2$ | ss | ss－t－HO |
| 8 \％ | SLL | 10.1 | $0 \cdot 2$ | † $\stackrel{\text { ¢ }}{ }$ | $0<\mathrm{II}$ | 00 | £6 | 08 L | 8 21 | $\tau$ | †＇¢ | 0t0 1 | $01 \varepsilon$ | 029t | 96. | ts | ts－t－HO |
| I＇tE | $8 L^{\circ}$ | $\mathrm{EO}^{\circ} \mathrm{I}$ | ［ $2 \cdot$ | ع 8 | OSII | $0 \cdot$ | 801 | 062 | 981 | L＇II | 6 ¢ 1 | 0901 | $00 \varepsilon$ | 0t9t | $96 \%$ | ¢s | ¢ $¢$－+ Ho |
| ${ }_{7} \mathrm{~S}$ | $\begin{aligned} & \mathrm{Hd}^{\mathrm{y}} \\ & \mathrm{y} T \mathrm{~V} \end{aligned}$ |  | $\begin{gathered} \text { Suoluy } \\ \text { [BIOIO } \end{gathered}$ | $\begin{gathered} \text { sLoug } \\ \text { [घ1oL } \end{gathered}$ |  | $\begin{gathered} (\text { (mddin }) \\ z^{\mathbf{E} O D} \end{gathered}$ | $\tau^{\text {b }} \mathrm{OS}$ | ．10 | $+\varepsilon^{\text {e }}$ O | $+2^{8} \mathrm{~N}$ | ＋+ | ${ }_{+}{ }^{\text {E }} \mathrm{N}$ | ＋ 19 | puod | $\mathrm{H}^{\text {d }}$ | $\stackrel{\text {（p）}}{\text { aw！．}}$ <br> －qnouI | $\underset{\text { Id IUurs }}{\text { III }}$ |


A5 Results of chemical analysis in DL-1 system (sample type: RelW)

| Sample ID | Incub. Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}^{\text {2 }}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathbf{S O}_{4}{ }^{2}$ | $\begin{array}{\|c\|} \mathbf{C O}_{3}{ }^{2-} \\ (\mathrm{wt} \text { ppm }) \\ \hline \end{array}$ | $\begin{array}{\|c} \mathbf{H C O}_{3}{ }^{-} \\ \text {(wt ppm) } \end{array}$ | $\begin{gathered} \text { Total } \\ \text { Cations } \end{gathered}$ | $\begin{gathered} \text { Total } \\ \text { Anions } \end{gathered}$ | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \ldots \\ \hline \end{gathered}$ | $\mathrm{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-1-8 | 8 | 7.83 | 4230 | 10 | 1140 | 15.8 | 14.5 | 22.3 | 800 | 53 | 0 | 1550 | 52.29 | 49.16 | 1.06 | 7.92 | 14.3 |
| DL-1-9 | 9 | 7.93 | 4190 | 0 | 1090 | 15.1 | 13.9 | 20.9 | 810 | 28 | 0 | 1560 | 49.98 | 48.97 | 1.02 | 8.01 | 9.7 |
| DL-1-10 | 10 | 7.79 | 4260 | 0 | 1090 | 15.1 | 13.7 | 20.8 | 800 | 15 | 0 | 1570 | 49.96 | 48.59 | 1.03 | 7.76 | 6.5 |
| DL-1-11 | 11 | 7.89 | 4130 | 4 | 1120 | 15.6 | 13.9 | 21.1 | 810 | 12 | 0 | 1590 | 51.31 | 49.19 | 1.04 | 7.95 | 5.8 |
| DL-1-12 | 12 | 7.74 | 4040 | 4 | 1050 | 14.5 | 13.1 | 19.8 | 810 | 10 | 0 | 1580 | 48.11 | 48.98 | 0.98 | 7.77 | 5.3 |
| DL-1-13 | 13 | 7.91 | 4180 | 0 | 1060 | 14.5 | 13.0 | 19.5 | 800 | 11 | 0 | 1450 | 48.52 | 46.54 | 1.04 | 7.44 | 5.6 |
| DL-1-14 | 14 | 7.91 | 4140 | 4 | 1050 | 14.1 | 12.8 | 19.3 | 800 | 8 | 0 | 1570 | 48.05 | 48.49 | 0.99 | 8.14 | 4.9 |
| DL-1-15 | 15 | 7.72 | 4130 | 4 | 1110 | 15.7 | 14.1 | 21.5 | 800 | 6 | 0 | 1590 | 50.91 | 48.78 | 1.04 | 7.76 | 4.4 |
| DL-1-16 | 16 | 7.74 | 4130 | 5 | 1120 | 16.1 | 14.5 | 22.2 | 800 | 5 | 0 | 1590 | 51.43 | 48.77 | 1.05 | 7.86 | 4.3 |
| DL-1-17 | 17 | 7.91 | 4140 | 5 | 1080 | 16.3 | 13.2 | 20.2 | 800 | 5 | 0 | 1570 | 49.48 | 48.44 | 1.02 | 8.05 | 4.2 |
| DL-1-18 | 18 | 8.07 | 3970 | 0 | 1120 | 16.3 | 14.6 | 22.2 | 790 | 6 | 0 | 1570 | 51.44 | 48.11 | 1.07 | 8.10 | 4.1 |
| DL-1-19 | 19 | 7.88 | 3990 | 10 | 1120 | 18.3 | 16.0 | 24.4 | 800 | 5 | 0 | 1590 | 51.72 | 48.83 | 1.06 | 7.98 | 3.7 |
| DL-1-20 | 20 | 7.71 | 4040 | 15 | 1125 | 17.1 | 15.6 | 23.5 | 810 | 10 | 0 | 1610 | 51.83 | 49.61 | 1.04 | 7.74 | 4.4 |
| DL-1-21 | 21 | 7.73 | 3990 | 29 | 1130 | 17.5 | 15.8 | 24.0 | 800 | 15 | 0 | 1600 | 52.10 | 49.43 | 1.05 | 8.06 | 6.4 |
| DL-1-22 | 22 | 7.75 | 4060 | 38 | 1140 | 17.5 | 15.6 | 23.7 | 800 | 24 | 0 | 1650 | 52.50 | 50.55 | 1.04 | 7.85 | 8.6 |
| DL-1-23 | 23 | 7.78 | 4040 | 53 | 1160 | 16.9 | 15.1 | 23.1 | 800 | 33 | 0 | 1680 | 53.28 | 51.41 | 1.04 | 7.96 | 10.7 |
| DL-1-24 | 24 | 7.85 | 4100 | 61 | 1160 | 17.6 | 15.8 | 24.1 | 790 | 36 | 0 | 1710 | 53.41 | 51.79 | 1.03 | 7.91 | 11.3 |
| DL-1-25 | 25 | 7.90 | 4100 | 68 | 1140 | 16.8 | 15.0 | 23.1 | 790 | 41 | 0 | 1610 | 52.40 | 50.34 | 1.04 | 7.94 | 12.2 |
| DL-1-26 | 26 | 7.80 | 4120 | 72 | 1160 | 17.3 | 15.6 | 23.7 | 790 | 38 | 0 | 1580 | 53.36 | 49.83 | 1.07 | 7.84 | 11.5 |
| DL-1-27 | 27 | 7.63 | 3950 | 92 | 1170 | 17.4 | 15.6 | 23.9 | 790 | 47 | 0 | 1560 | 53.81 | 49.94 | 1.08 | 7.85 | 13.9 |
| DL-1-28 | 28 | 7.59 | 4070 | 130 | 1190 | 17.4 | 16.1 | 24.5 | 790 | 70 | 0 | 1550 | 54.75 | 50.72 | 1.08 | 7.71 | 20.9 |
| DL-1-29 | 29 | 7.63 | 4290 | 160 | 1180 | 17.2 | 16.3 | 24.7 | 770 | 91 | 0 | 1770 | 54.34 | 54.58 | 1.00 | 7.76 | 28.4 |
| DL-1-30 | 30 | 7.66 | 4260 | 170 | 1200 | 17.7 | 16.5 | 25.3 | 780 | 95 | 0 | 1630 | 55.27 | 52.78 | 1.05 | 7.93 | 29.6 |
| DL-1-31 | 31 | 7.73 | 4250 | 180 | 1190 | 17.6 | 16.4 | 24.9 | 790 | 97 | 0 | 1520 | 54.80 | 51.41 | 1.07 | 7.86 | 29.1 |
| DL-1-32 | 32 | 7.79 | 4630 | 250 | 1220 | 17.9 | 17.3 | 26.7 | 850 | 152 | 0 | 1500 | 56.28 | 54.79 | 1.03 | 7.71 | 53.3 |

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| 0＇tt | $0 \varepsilon^{\circ} \angle$ | ${ }^{7} 60$ | ＋185 | t6 ${ }^{\text {ts }}$ | 0 ¢ 1 | 0 | £ Z | $0 \varepsilon 8$ | L＇9z | $\tau<1$ | L＇91 | 06II | 0ts | 026t | 76 L | LS | Ls－I－Ta |
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| $\varepsilon^{\prime}$＇IS | $L L^{\circ} \mathrm{L}$ | $00^{\circ} 1$ | 80 LS | $\pm$ ¢ $\angle ¢$ | 0itl | 0 | siz | 018 | 8.82 | ＋81 | $9 \cdot L I$ | 0ヶてI | 0ts | 016t | ع18 | 95 | 9s－I－70 |
| †＇s9 | $\mathrm{HIL}_{2}$ | 660 | 6785 | $6 L \cdot L S$ | 0itl | 0 | $0 \downarrow$ ¢ | $0 ¢ 8$ | 062 | 581 | L＇LI | 0szI | oss | 06Lt | 08L | ss | ¢s－i－Ta |
| 0 IL | $00^{\circ} \mathrm{L}$ | L60 | L6LS | 9E．9S | 00tr | 0 | szz | 0 ¢8 | 6 LZ | 8.2 | C＇LI | 0zzI | 0ts | 008t | LLL | ts | ts－I－Ta |
| 565 | $9 \mathrm{~F}^{\circ} \mathrm{L}$ | 660 | £ $\underbrace{09}$ | $8{ }^{\text {¢ }} 65$ | $0 ¢ t 1$ | 0 | £ $\downarrow$ | 828 | ¢ 8 \％ | で81 | L L L | 06zı | 195 | OSIS | $68^{\circ} \mathrm{L}$ | $\varepsilon \varsigma$ | £s－I－Ta |
| $\varepsilon \angle S$ | It L | 660 | 60.09 | 25．6s | $0 ¢ \square 1$ | 0 | IEz | 088 | $8.8 z$ | ¢ 81 | $9 \cdot 2$ | 062I | ILS | 00IS | ＋8L | zs | 2s－I－Ta |
| 9.95 | $\mathfrak{E t}$ ¢ | 860 | 0009 | 0685 | 0¢tI | 0 | ¢tz | 698 | $\tau \cdot L Z$ | ¢ $¢ 1$ | 8.91 | 08ZI | s9s | 0zIS | 588 | is | IS－I－Ta |
| ¢89 | $15{ }^{\circ} \mathrm{L}$ | 860 | 6809 | $8{ }^{\text {t }} 65$ | $0 ¢ \square 1$ | 0 | $9 \downarrow て$ | 188 | $58 \%$ | z＇8I | S＇LI | 06zI | 19¢ | 060s | 66 L | os | OS－I－Ta |
| 699 | $09^{\circ} \mathrm{L}$ | 10.1 | tt 65 | z¢09 | 02tI | 0 | LĖ | 028 | ¢ 8 \％ | 081 | $\varepsilon \angle L$ | 0IEI | tos | 060s | 88L | $6{ }^{6}$ | $6 \mathrm{t}-\mathrm{I}-\mathrm{Ta}$ |
| $0 \cdot \mathrm{ZL}$ | LS＇L | $00^{\prime} 1$ | £ Z09 | LE09 | $0 ¢ \downarrow 1$ | 0 | $6 \downarrow 2$ | 828 | $\llcorner\cdot 8 \tau$ | $\downarrow \cdot 81$ | $\tau \angle L$ | 01¢1 | 95s | 01IS | $8 L \cdot L$ | $8{ }^{\text {t }}$ | $8{ }^{6}-\mathrm{I}-7 \mathrm{Ca}$ |
| L＇z8 | 20．8 | 660 | เ9＇09 | 8669 | 0¢tl | 0 | ¢92 | 968 | $\varepsilon \cdot 6 \tau$ | 981 | I＇LI | 00\＆ 1 | L9S | 0615 | t9 L | $\angle t$ | Lt－I－7d |
| 0.98 | $79^{\circ} \mathrm{L}$ | 20.1 | LL：8S | 8965 | 0ztI | 0 | $8 \downarrow \tau$ | 078 | ¢0E | ع61 | 6.21 | 06ZI | 0ts | 010s | 69 L | $9{ }^{\text {t }}$ | $9 \mathrm{er-r}-7 \mathrm{C}$ |
| \％＇08 | t90 $L$ | 20 ${ }^{\circ}$ | $9{ }^{\circ} \mathrm{LS}$ | S1．8s | 02tI | 0 | ¢£ぇ | 018 | $\downarrow \bullet 8 \tau$ | 181 | $0 \angle 1$ | 097ı | 00¢ | 0s6t | $69^{\circ} \mathrm{L}$ | st | St－I－7a |
| 892 | $65^{\circ} \mathrm{L}$ | 660 | 29\％8 | L6LS | 0zt1 | 0 | Stz | OS8 | L＇9z | z LI | z＇91 | 0921 | 0IS | 086t | 6SL | tt | 比 I －7 |
| 9 ¢8 | L9 ${ }^{\circ}$ | $\varepsilon^{0} \mathrm{I}$ | セが8 | 8E09 | $01+1$ | 0 | osz | OS8 | 88. | t 81 | SLL | 0ıEı | 00s | 0¢6t | 29.2 | Et | \＆t－I－7a |
| 908 | OS $L$ | 10 I | 07：8s | £0＇6s | 02tl | 0 | Itz | 098 | ¢ 88 | 181 | 9.2 | 08 z | 09t | 068t | 2SL | zt | てt－I－Ta |
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| 978 | $z L L$ | t0 I | ZL＇LS | L66S | $01 t 1$ | 0 | $6 \varepsilon \tau$ | OS8 | 1＇62 | ¢ 81 | 8 LI | 00¢ı | 09t | 008t | 68 L | ${ }^{0}+$ | $0 \mathrm{t}-\mathrm{I}-7 \mathrm{C}$ |
| $0 \cdot L L$ | $18 . L$ | $\varepsilon^{\circ} \mathrm{I}$ |  | ${ }^{6 Z^{\prime} 65}$ | 02th | 0 | t£ | OS8 | ¢ 08 | 561 | 681 | 08zI | $0{ }^{\circ} \mathrm{t}$ | 068t | ＋LL | $6 \varepsilon$ | 6ع－1－7a |
| 989 | $\varepsilon L L$ | $\varepsilon_{0} \mathrm{I}$ | $6 \varepsilon^{\circ} 95$ | z7\％8 | 0tol | 0 | 661 | 058 | $9.8 z$ | －81 | 581 | 09zı | 08E | 028t | $9 L L$ | $8 \varepsilon$ | 8¢－I－Ta |
| 80 L | $s L L^{\circ}$ | 10.1 | 0L＇ss | 8E＇95 | $00 t 1$ | 0 | 661 | 0 088 | L－Lz | 081 | 9.41 | 0zzı | 0LE | 06Lt | $8 L^{\circ} \mathrm{L}$ | $\llcorner\varepsilon$ | LE－I－Ta |
| $\angle \cdot \mathscr{L}$ | ${ }^{16} \mathrm{~L}$ | 20． 1 | 09．9s | 8L＇LS | 0¢tI | 0 | LIz | 058 | † 8 C | －81 | 881 | 0szI | $08 \varepsilon$ | 008t | 96 L | $9 \varepsilon$ | 98－I－7a |
| $\varepsilon \cdot \varepsilon L$ | $08 . \mathrm{L}$ | E0＇ 1 | S0＇9s | 08． LS | 0¢bl | 0 | 912 | $0 \pm 8$ | 8.82 | 981 | ［＇81 | 0szl | $09 \varepsilon$ | 0LLt | 18 L | ¢£ | ¢ $\varepsilon$－-70 |
| I＇tL | $\pm L \cdot L$ | £0＇ I |  | 1695 | 0 0tol | 0 | 902 | $0 ¢ 8$ | $5 \cdot 8 \mathrm{z}$ | 581 | て＇81 | 0¢zI | $0 \downarrow \varepsilon$ | 08Lt | ZLL | $\pm \varepsilon$ | $\pm \varepsilon$－ |
| 6 LS | $t L^{\circ} \mathrm{L}$ | ＋0， 1 | 91＇SS | tでLs | 0＜ti | 0 | 891 | 098 | $\downarrow$－ | L＇LI | 681 | 0ャzı | $0 \angle z$ | 0L9t | E8L | £ | £ $\varepsilon$－ $\mathrm{I}-\mathrm{Ta}$ |
| ${ }_{-2} \mathrm{~S}$ | $\begin{aligned} & \text { Hd } \\ & \text { YTV } \end{aligned}$ |  |  | $\begin{aligned} & \text { suope] } \\ & \text { [E70. } \end{aligned}$ | （ <br> ．${ }^{\text {© OOH }}$ |  | ${ }_{-2}{ }^{\text {b }}$ OS | －10 | ${ }_{+1}{ }^{\text {B }}$ O | $+z^{8} \mathrm{~W}$ | ${ }_{+}+\mathrm{H}$ | ${ }^{\text {b }}$ N | $\ddagger^{\text {d／}}$ | puos | $\mathrm{H}^{\text {d }}$ |  | II Pdures |

A5 Results of chemical analysis in DL－1 system（sample type：RelW）（cont＇d）

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A5 Results of chemical analysis in DL－1 system（sample type：RelW）（cont＇d）

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| $0 \cdot 1 z z$ | $16^{\circ} \mathrm{L}$ | 860 | 01＇29 | $00 \cdot 19$ | 0tSI | 0 | $6 \varepsilon \tau$ | OSL | 8.82 | soz | 6.41 | 0z¢I | 098 | 0L9S | 918 | £ $\downarrow$ | \＆モI－I－Tの |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 0 0¢II | $79^{\circ} \mathrm{L}$ | 10.1 | しく19 | 09 \％9 | 01 sl | 0 | ゅャて | OSL | £92 | £0z | 081 | 09¢ | 098 | 0L9s | 128 | z | てzI－I－TG |
| 6.6 | $\varepsilon L L$ | $66^{\circ}$ | 9209 | L009 | 00s 1 | 0 | ¢ヵて | OEL | †＇6て | 561 | SLI | 00\＆1 | 078 | 0L9S | 16.4 | IZ1 | IZI－I－Ta |
| †＇99 | $\varepsilon 9 \%$ | S60 | S0＇29 | 0685 | 0 lSI | 0 | tst | OSL | く๕ $\downarrow$ | S61 | ［＇LI | 0871 | 028 | 0695 | 278 | 0zI | 02I－I－Ta |
| $0 \cdot \downarrow L$ | $\varepsilon ¢^{\circ} \mathrm{L}$ | 860 | LS 09 | 81.65 | 0ist | 0 | $\varepsilon ⿺ 𠃊$ | OSL | で0¢ | I＇61 | $\llcorner\cdot 91$ | 08Z1 | 078 | 0t9s | 99 L | 611 | 61－I－7a |
| でし | 9S＇L | ＋60 |  | t0．9s | 0zs | 0 | 6t1 | OSL | $6 . z z$ | † 21 | L＇sı | 0zzI | 078 | 06ss | $68 . \mathrm{L}$ | 8 I | 8L－I－7a |
| $0 \cdot \varepsilon<1$ | tS 8 | S60 | £8＇19 | 88.85 | 0btI | III | ISI | OSL | 8 8 $\downarrow$ | 161 | $\varepsilon ้ L I$ | 08ZI | 078 | 009S | 908 | LII | LIL－ITG |
| 0＇z0E | LT＇L | 960 | 9＋＇09 | 86 LS | ${ }_{0 \downarrow \text { ¢ }}$ | 0 | ＋81 | OSL | $80 \varepsilon$ | 961 | $8{ }^{\circ} \mathrm{LI}$ | oszi | 078 | 0985 | $6 \mathrm{t}^{2} \mathrm{~L}$ | 9 II | 91－I－7a |
| 0．8LZ | £̇L | 960 | 69.65 | $00^{\prime} \angle S$ | 09tI | 0 | 912 | OSL | ¢ 88 | £81 | $0 \cdot \mathrm{LI}$ | 0ヶてI | 018 | 0985 | tSL | SII | SH－ITCd |
| 0901 | STLL | 960 | で09 | E6LS | $00 t 1$ | 0 | L97 | OSL | ¢0¢ | ع61 | L＇LI | 0szi | 018 | 0985 | $89^{\circ} \mathrm{L}$ | †II | tul－I－Ta |
| 000E | 19 L | \＄6\％ | 95：85 | 68 ss | 0LSI | 0 | 0 t | $0 Z L$ | $9 . L z$ | 8LL | ¢91 | 0IZI | 0LL | 088S | 59．L | ElI | £ |
| 685 | ＋8\％ | 160 | 976s | si＇ts | 0ss | 0 | $\llcorner 6$ | 092 | S＜L | 6 61 | z＇91 | 0＜II | 058 | 098s | $98^{\circ} \mathrm{L}$ | ZII | 2H－I－7a |
| 0791 | $7 \dot{t} L$ | $\pm 60$ | ts＇6s | £6． SS | 00SI | 0 | 9 l | 092 | く－92 | 981 | でLI | 01ZI | 078 | OSLS | $86 . \mathrm{L}$ | III | 1II－I－7a |
| 0．9SI | $00^{\circ} \mathrm{L}$ | 560 | ¢9．09 | でしく | 08t | 0 | 122 | 092 |  | 161 | t＇LI | 0ャてI | 0¢8 | ofzs | 20＇8 | 0 I | 0ヶI－I－7a |
| 0．1II | $0{ }^{\text {b }} \mathrm{L} L$ | L60 | 95：09 | 5 St 8 S | 0btI | 0 |  | 092 | で1\＆ | 661 | L＇LI | 09ZI | $0 ¢ 8$ | $0 t \angle S$ | $69^{\circ} \mathrm{L}$ | 601 | 601－1－7a |
| －¢ 9 | $87^{\circ} \mathrm{L}$ | S6．0 | $0 \downarrow \cdot 19$ | 68.85 | 0ISI | ， | $0 z z$ | 0LL | LOE | 961 | $\varepsilon \angle L$ | 09ZI | $0 ¢ 8$ | 0LLS | $8 L^{\circ} \mathrm{L}$ | 801 | 801－1－7a |
| ${ }^{2} \mathrm{~S}$ | $\begin{gathered} \mathrm{H}^{\mathrm{d}} \\ \mathrm{y} 7 \mathrm{~V} \end{gathered}$ |  | $\begin{gathered} \text { suopur } \\ \text { fibiol } \end{gathered}$ | suolies ［ ${ }^{\text {Bl7OL }}$ |  |  | ${ }_{2}{ }^{\text {b }}$ | 10 | $+2^{\text {E }} \mathrm{O}$ | $+i^{8} \mathbf{W}$ | ＋${ }^{\text {Y }}$ | ${ }^{\text {B }}$ N | ＋ 18 | puos | $\mathrm{H}^{\text {d }}$ |  | CII Pdurs |


A6 Results of chemical analysis in DL-2 system (sample type: RelW)

| Sample ID | Incub. <br> Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathrm{Mg}^{2+}$ | $\mathrm{Ca}^{2+}$ | Cr | $\mathrm{SO}_{4}{ }^{2}$ | $\begin{gathered} \mathbf{C O}_{3}{ }^{2-} \\ (\mathbf{w t p p m}) \end{gathered}$ | $\begin{gathered} \mathbf{H C O}_{3}^{-} \\ (\text {(wt ppm) }) \end{gathered}$ | $\begin{gathered} \text { Total } \\ \text { Cations } \end{gathered}$ | $\begin{gathered} \text { Total } \\ \text { Anions } \\ \hline \end{gathered}$ | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \end{gathered}$ | $\mathrm{S}^{\mathbf{2}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-2-5 | 5 | 8.10 | 4020 | 0.01 | 1110 | 16.3 | 14.2 | 21.5 | 800 | 32.1 | 0 | 1540 | 50.94 | 48.45 | 1.05 | 8.05 | 10.10 |
| DL-2-7 | 7 | 7.92 | 3960 | 0.01 | 1140 | 16.3 | 14.6 | 22.1 | 790 | 22.6 | 0 | 1550 | 52.30 | 48.13 | 1.09 | 7.82 | 8.96 |
| DL-2-8 | 8 | 7.93 | 3930 | 0.01 | 1130 | 16.7 | 14.9 | 22.7 | 780 | 9.7 | 0 | 1580 | 51.94 | 48.08 | 1.08 | 7.85 | 5.40 |
| DL-2-9 | 9 | 8.10 | 4030 | 0.01 | 1120 | 16.4 | 14.8 | 22.7 | 800 | 7.0 | 0 | 1580 | 51.48 | 48.58 | 1.06 | 8.15 | 4.83 |
| DL-2-10 | 10 | 7.92 | 4070 | 0.01 | 1160 | 16.9 | 15.2 | 23.3 | 800 | 5.2 | 0 | 1580 | 53.30 | 48.55 | 1.10 | 7.91 | 4.24 |
| DL-2-11 | 11 | 7.90 | 3990 | 0.01 | 1130 | 16.3 | 14.8 | 22.6 | 800 | 4.6 | 0 | 1590 | 51.91 | 48.70 | 1.07 | 7.78 | 4.04 |
| DL-2-12 | 12 | 7.90 | 4070 | 0.01 | 1140 | 16.5 | 14.9 | 22.7 | 800 | 3.9 | 0 | 1580 | 52.36 | 48.52 | 1.08 | 7.95 | 3.95 |
| DL-2-13 | 13 | 7.93 | 3990 | 0.01 | 1140 | 16.4 | 14.9 | 22.7 | 800 | 3.5 | 0 | 1580 | 52.36 | 48.51 | 1.08 | 7.87 | 3.85 |
| DL-2-14 | 14 | 7.83 | 4040 | 0.01 | 1130 | 16.2 | 14.7 | 22.5 | 800 | 3.3 | 0 | 1620 | 51.90 | 49.16 | 1.06 | 7.84 | 3.76 |
| DL-2-15 | 15 | 7.92 | 4040 | 0.01 | 1140 | 16.3 | 14.8 | 22.6 | 800 | 3.2 | 0 | 1590 | 52.35 | 48.67 | 1.08 | 7.93 | 3.69 |
| DL-2-16 | 16 | 7.90 | 4080 | 0.01 | 1130 | 16.1 | 14.5 | 22.2 | 800 | 2.1 | 0 | 1560 | 51.86 | 48.15 | 1.08 | 8.06 | 3.58 |
| DL-2-17 | 17 | 7.85 | 3990 | 0.01 | 1145 | 17.0 | 15.3 | 23.4 | 800 | 1.9 | 0 | 1580 | 52.66 | 48.48 | 1.09 | 8.13 | 3.53 |
| DL-2-18 | 18 | 7.88 | 4080 | 0.01 | 1140 | 16.5 | 14.8 | 22.6 | 800 | 2.1 | 0 | 1580 | 52.35 | 48.48 | 1.08 | 8.11 | 3.58 |
| DL-2-19 | 19 | 8.03 | 4210 | 0.01 | 1090 | 15.7 | 14.6 | 13.3 | 850 | 2.7 | 0 | 1580 | 49.68 | 49.90 | 1.00 | 8.01 | 5.67 |
| DL-2-20 | 20 | 8.10 | 4300 | 0.01 | 1100 | 15.7 | 14.6 | 21.5 | 870 | 2.1 | 0 | 1580 | 50.52 | 50.45 | 1.00 | 7.91 | 5.09 |
| DL-2-21 | 21 | 8.06 | 4300 | 0.01 | 1100 | 20.0 | 14.3 | 22.1 | 870 | 2.4 | 0 | 1520 | 50.64 | 49.48 | 1.02 | 8.12 | 4.13 |
| DL-2-22 | 22 | 8.28 | 4140 | 0.01 | 1140 | 17.0 | 15.3 | 23.7 | 880 | 2.8 | 0 | 1600 | 52.46 | 51.08 | 1.03 | 7.83 | 3.86 |
| DL-2-23 | 23 | 7.96 | 4320 | 0.01 | 1100 | 16.0 | 15.0 | 23.1 | 870 | 4.1 | 0 | 1620 | 50.64 | 51.15 | 0.99 | 7.75 | 4.50 |
| DL-2-24 | 24 | 7.78 | 4320 | 0.01 | 1180 | 17.6 | 16.1 | 24.9 | 870 | 1.9 | 0 | 1600 | 54.34 | 50.78 | 1.07 | 7.63 | 3.60 |
| DL-2-25 | 25 | 7.88 | 4290 | 0.01 | 1120 | 16.5 | 15.2 | 23.7 | 850 | 2.9 | 0 | 1600 | 51.57 | 50.23 | 1.03 | 7.77 | 3.83 |
| DL-2-26 | 26 | 7.84 | 4300 | 0.01 | 1120 | 16.3 | 15.1 | 23.4 | 870 | 4.7 | 0 | 1590 | 51.54 | 50.67 | 1.02 | 7.71 | 4.41 |
| DL-2-27 | 27 | 7.83 | 4310 | 1.70 | 1180 | 16.8 | 15.2 | 23.6 | 850 | 14.9 | 0 | 1590 | 54.18 | 50.34 | 1.08 | 7.76 | 6.01 |
| DL-2-28 | 28 | 7.69 | 4350 | 0.01 | 1100 | 15.6 | 14.3 | 21.9 | 850 | 14.1 | 0 | 1590 | 50.51 | 50.30 | 1.00 | 7.69 | 6.81 |
| DL-2-29 | 29 | 7.66 | 4410 | 0.01 | 1110 | 15.8 | 14.6 | 22.3 | 850 | 20.3 | 0 | 1600 | 51.00 | 50.60 | 1.01 | 7.63 | 8.92 |
| DL-2-30 | 30 | 7.74 | 4370 | 0.01 | 1120 | 15.9 | 14.7 | 22.5 | 850 | 29.8 | 0 | 1590 | 51.45 | 50.63 | 1.02 | 7.75 | 11.60 |
| DL-2-31 | 31 | 7.73 | 4420 | 0.01 | 1130 | 15.8 | 14.8 | 22.6 | 850 | 41.0 | 0 | 1600 | 51.90 | 51.03 | 1.02 | 7.62 | 15.50 |


| 0079I | $9 \varepsilon^{\circ} \mathrm{L}$ | S0． | ¢S6S | L5\％9 | 08SI | 0 | 076t | 0 088 | ¢＇9E | $0 \times \tau$ | 8＇81 | 0ヶ¢ | 000 | 068t | $08 . \mathrm{L}$ | LS | Ls－z－Td |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 00．9SI | $8 L^{\circ} \mathrm{L}$ | $40^{\circ} \mathrm{I}$ | 2985 | 1s\％9 | 0ssi | 0 | 0てくt | 0¢8 | 6.61 | 6 zz | L－81 | 09¢I | $00^{\circ} 0$ | 0＜8t | 08 L | 95 | 9s－z－Ta |
| 00．6tI | ¢ $¢$ | ＋0．1 | 6Z\％65 | $60^{*} 19$ | 0091 | 0 | 0．ist | 078 | $6 \downarrow$ ¢ | szz | 981 | 0z\＆I | $00^{\circ} 0$ | 0＜8t | 99. | ss | ss－z－7a |
| 00＇L0I | $L \varepsilon^{\circ} \mathrm{L}$ | 560 | 6185 | zz＇ss | 0191 | 0 | 0．0se | 028 | $\varepsilon{ }^{\prime} 6 z$ | て＇61 | 191 | 0611 | 000 | 088t | $85^{\circ} \mathrm{L}$ | $\dagger$ ¢ | ts－z－Td |
| 00．00I | $9 \varepsilon \%$ | ＋60 | ＋0．85 | 69 ＇ts | 0291 | 0 | 0 0¢ $¢$ | $0<8$ | ちゃ8て | 981 | 1＇91 | 08II | 000 | 008t | E9L | $\varepsilon \varsigma$ | £s－z－Ta |
| 0ャレ6 | ULL | 560 | $0{ }^{\circ} \angle \mathrm{LS}$ | LSts | 0091 | 0 | 0．02E | 048 |  | 6 LI | L－SI | 08II | 000 | 09Lt | $\angle L L$ | zs | zs－z－Ta |
| 08＇16 | $6 S^{\circ} \mathrm{L}$ | ＋60 | $8 \chi^{\circ} \mathrm{LS}$ | Ls¢s | 0191 | 0 | 0．90ع | 028 | ¢9\％ | 1 21 | 9 s ı | 091I | 000 | 0ZLt | $E L L$ | Is | ［s－z－7a |
| 0¢98 | 0LL | 460 | Lz＇LS | 69．¢s | 0291 | 0 | 0862 | $0<8$ |  | L＇91 | 9 s I | 01zI | 000 | 0ZLt | $56 . L$ | 0 ¢ | 0s－z－7a |
| 0L＇6L | 678 | L60 | L0＇LS | 8Z¢¢ | 0 0¢9 | 0 | 0． 297 | 088 | $0 \cdot 9 z$ | 9.91 | †＇91 | 002I | 000 | 089t | 608 | $6{ }^{6}$ | 6 t －て－7a |
| 0S08 |  | 160 | ts＇9s | ＋8＇ts | 0191 | 0 | 0．Lsz | 088 | $0 \cdot 97$ | 9.91 | t¢91 | 0611 | 000 | 099t | \＄6L | $8{ }^{\text {b }}$ | 8 b －-70 |
| $00^{\prime} \mathrm{I}$ L | L6L | 860 | t6＇ss | 99.75 | 0291 | 0 | $0 \downarrow$－${ }^{\text {a }}$ | $0<8$ | でって | L＇si | 8 s | 06II | 000 | 08St | 28.1 | $\angle t$ | $\angle \mathrm{t}-\mathrm{z}-\mathrm{Ta}$ |
| 00＇IL | $9 L^{\prime} L$ | S60 | £8＇s¢ | ¢0¢¢ | 0191 | 0 | 0．$๕ z \tau$ | 088 |  | † 91 | ¢91 | $0 \mathrm{O}_{1} \mathrm{I}$ | 000 | 0LSt | ＋8．L | $9+$ | 9 9 －でフa |
| 0LOL | $89^{\circ} \mathrm{L}$ | 560 | LLSS | ¢0¢ร | 0191 | 0 | 0．0zz | 088 | I＇sz | z91 | L＇91 | OSII | 000 | 0tSt | 08： | st | stoz－Ta |
| 0でIL | L8L | 660 | sscs | 26．ts | 0091 | 0 | 0．1iz | 088 | £9z | 1＜L | $0<1$ | 06It | 100 | 09St | ＋6． | t | toz－Ta |
| 06¢9 | 68 L | $\angle 60$ | 2s＇ts | t9\％s | 0091 | 0 | 0.561 | 098 |  | ¢91 | L＇91 | 0tit | 100 | osst | 58.2 | £t | $\varepsilon \leftarrow て-\mathrm{Ta}$ |
| 01＇t9 | SL＇L | 20.1 | 86 zs | 00．ts | 0zsi | 0 | 0.181 | 098 | 8 cz | 891 | $0 \angle 1$ | 0LII | 100 | Ottt | ELL | zt | ででTの |
| 0¢¢¢ | ULL | 560 | E1＇9s | L0¢s | 06SI | 0 | 0．9zz | 006 | $\dagger$ ¢ $¢$ | ¢91 | 991 | 0sil | 100 | 08tt | $S L \cdot L$ | It | け－でTU |
| 0ヵ¢¢ | 2LL | 860 | て£＇ts | 6て＇\＆s | 08 SI | 0 | 0691 | 888 | 9＇દz | てSI | 8 ו | 091t | 100 | 09st | $19 . L$ | ${ }_{0}$ | $0 \downarrow て ゙ T \mathrm{a}$ |
| 08＇ss | $\varepsilon L \cdot L$ | 860 | 20＇ss | S6¢¢ | 0091 | 0 | 0．591 | 006 | †¢く | 991 | 891 | 0LII | 100 | 09St | $2 S^{\circ} \mathrm{L}$ | $6 \varepsilon$ | $6 \varepsilon-z-70$ |
| 0695 |  | $00^{\circ} \mathrm{I}$ | Iz＇ts | \＆t＇ts | 08S 1 | 0 | $0 \cdot 991$ | 288 | L＇sz | 891 | $0<1$ | 08II | 100 | OSSt | ISLL | $8 \varepsilon$ | $8 \varepsilon-て-10$ |
| 069t | LLL | 860 | LIts | zoes | 06S 1 | 0 | 00t1 | 168 | $0 ¢ \Sigma$ | z＇91 | t＇91 | OSII | 100 | 0ist | 79. | L¢ | L¢－z－7a |
| 09\％6を | $6 L^{\circ} \mathrm{L}$ | 860 | $8 \varepsilon \cdot \varepsilon$ ¢ | 80\％s | 06SI | 0 | 0911 | 188 | どャて | 8 SI | E＇91 | 0¢II | 100 | 0Ltt | 19.4 | $9 \varepsilon$ | 9 S －でフa |
| 0S＂9£ | $08^{\circ} \mathrm{L}$ | $00 \cdot 1$ | z¢๕ร | $8{ }^{\text {¢ }}$ ¢ $¢$ | 06S 1 | 0 | 0．${ }^{0} 1$ | L68 | I＇sz | $\varepsilon 91$ | L＇91 | 0911 | 100 | 08tt | $99^{\circ} \mathrm{L}$ | $\varsigma \varepsilon$ | s¢－でTa |
| 08＊1¢ | L9． | 960 | £¢ ¢ | zi＇is | 06s 1 | 0 | 0.611 | $\angle 88$ | 9 9 $\varepsilon$ | †＇SI | 9 s | 0III | 100 | 00st | $69^{\circ} \mathrm{L}$ | $\downarrow \varepsilon$ | เ¢－て－Ta |
| 0¢0\％ | ＋9．L | 660 | $80^{\circ} \mathrm{IS}$ | ILOS | 06SI | 0 | 6 ＋9 | 078 | 0.02 | z＇єı | L $\varepsilon 1$ | 01t1 | 100 | 08tt | $69^{\circ} \mathrm{L}$ | $\varepsilon \varepsilon$ | £¢－z－Ta |
| $09 \cdot \angle 1$ | 29.4 | $\angle 6.0$ | $9 \varepsilon^{\prime \prime}$ IS | to 0s | 06s 1 | 0 | $\tau$ Is | 098 | $9 \cdot 1 z$ | でャ1 | I＇si | 0601 | 100 | 0t\＆t | ILL | て¢ | 2¢－z－7a |
| ${ }_{-2} \mathrm{~S}$ | $\begin{aligned} & \mathrm{H}^{\mathrm{d}} \\ & \text { yTV } \end{aligned}$ |  | $\begin{gathered} \text { suogur } \\ \text { [घto } \end{gathered}$ | $\begin{gathered} \text { suoped } \\ \text { Lexol } \end{gathered}$ | $\left\|\begin{array}{c} (\mathrm{udd} \boldsymbol{M}) \\ { }_{-}^{\boldsymbol{\varepsilon}} \mathbf{O O H} \end{array}\right\|$ |  | ${ }_{2} \mathrm{OS}$ | 13 | $+z^{\text {E }} \mathrm{S}$ | ${ }^{+8}{ }^{8} \mathrm{~N}$ | ＋ | ${ }_{+}{ }^{\text {EN }}$ | ${ }^{18}$ | puo？ | $\mathrm{H}^{\text {d }}$ |  | al ${ }^{\text {dumb }}$ |

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A6 Results of chemical analysis in DL-2 system (sample type: RelW) (cont'd)

| Sample ID | Incub. <br> Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | Cl | $\mathbf{S O}_{4}{ }^{\text {2 }}$ | $\underset{(\mathbf{w t ~ p p m})}{\mathbf{C O}_{3}^{\mathbf{2}^{-}}}$ | $\begin{gathered} \mathrm{HCO}_{3} \\ (\mathrm{wt} \mathrm{ppm}) \end{gathered}$ | Total Cations | Total <br> Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | $\underset{\mathbf{p H}}{\substack{\text { ALK }}}$ | $\mathbf{S}^{\mathbf{2}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-2-58 | 58 | 7.64 | 4900 | 0.00 | 1320 | 18.9 | 25.2 | 37.8 | 830 | 520.0 | 0 | 1540 | 61.87 | 59.46 | 1.04 | 7.37 | 169.00 |
| DL-2-59 | 59 | 7.64 | 4960 | 0.00 | 1330 | 19.4 | 26.4 | 39.3 | 830 | 545.0 | 0 | 1540 | 62.49 | 59.98 | 1.04 | 7.59 | 182.00 |
| DL-2-60 | 60 | 7.56 | 4980 | 0.00 | 1360 | 19.5 | 27.5 | 40.7 | 830 | 590.0 | 0 | 1510 | 63.96 | 60.43 | 1.06 | 7.39 | 194.00 |
| DL-2-61 | 61 | 7.58 | 5040 | 0.00 | 1320 | 19.7 | 28.9 | 42.4 | 800 | 605.0 | 0 | 1520 | 62.42 | 60.06 | 1.04 | 7.35 | 201.00 |
| DL-2-62 | 62 | 7.83 | 4940 | 0.00 | 1330 | 19.7 | 29.3 | 38.1 | 850 | 600.0 | 0 | 1470 | 62.68 | 60.54 | 1.04 | 7.23 | 200.00 |
| DL-2-63 | 63 | 8.03 | 4930 | 0.00 | 1340 | 19.9 | 30.1 | 36.5 | 850 | 595.0 | 0 | 1480 | 63.10 | 60.60 | 1.04 | 7.16 | 199.00 |
| DL-2-64 | 64 | 7.75 | 4890 | 0.00 | 1330 | 20.2 | 31.0 | 39.4 | 850 | 625.0 | 0 | 1530 | 62.90 | 62.05 | 1.01 | 7.17 | 205.00 |
| DL-2-65 | 65 | 7.92 | 4900 | 0.00 | 1350 | 20.5 | 31.8 | 34.7 | 860 | 635.0 | 0 | 1480 | 63.61 | 61.72 | 1.03 | 7.36 | 208.00 |
| DL-2-66 | 66 | 7.81 | 4940 | 0.00 | 1320 | 20.2 | 32.7 | 35.6 | 850 | 640.0 | 0 | 1490 | 62.41 | 61.70 | 1.01 | 7.26 | 214.00 |
| DL-2-67 | 67 | 7.56 | 5070 | 0.00 | 1370 | 21.1 | 36.3 | 46.2 | 850 | 695.0 | 0 | 1460 | 65.44 | 62.36 | 1.05 | 7.44 | 230.00 |
| DL-2-68 | 68 | 7.90 | 5070 | 0.00 | 1360 | 21.3 | 37.5 | 41.8 | 850 | 680.0 | 0 | 1510 | 64.89 | 62.86 | 1.03 | 7.22 | 225.00 |
| DL-2-69 | 69 | 8.19 | 5140 | 0.00 | 1320 | 19.3 | 33.4 | 34.5 | 850 | 442.0 | 0 | 1590 | 62.39 | 59.22 | 1.05 | 7.93 | 187.00 |
| DL-2-70 | 70 | 7.85 | 5270 | 0.00 | 1310 | 19.8 | 34.9 | 35.6 | 850 | 457.0 | 0 | 1640 | 62.15 | 60.35 | 1.03 | 7.32 | 146.00 |
| DL-2-71 | 71 | 8.07 | 5190 | 0.00 | 1350 | 20.1 | 35.4 | 36.9 | 850 | 435.0 | 0 | 1640 | 64.01 | 59.89 | 1.07 | 7.43 | 200.00 |
| DL-2-72 | 72 | 7.98 | 5200 | 0.00 | 1340 | 19.6 | 35.0 | 37.8 | 840 | 556.0 | 0 | 1540 | 63.57 | 60.49 | 1.05 | 7.64 | 165.00 |
| DL-2-73 | 73 | 8.10 | 5230 | 0.00 | 1360 | 20.4 | 36.0 | 41.5 | 840 | 432.0 | 0 | 1620 | 64.73 | 59.22 | 1.09 | 7.37 | 272.00 |
| DL-2-74 | 74 | 7.61 | 5310 | 0.00 | 1330 | 19.4 | 36.0 | 45.0 | 830 | 548.0 | 0 | 1720 | 63.57 | 62.99 | 1.01 | 7.47 | 141.00 |
| DL-2-75 | 75 | 7.68 | 5300 | 0.00 | 1330 | 18.1 | 34.6 | 39.6 | 820 | 647.0 | 0 | 1650 | 63.15 | 63.63 | 0.99 | 7.34 | 141.00 |
| DL-2-76 | 76 | 8.03 | 5250 | 0.00 | 1320 | 18.4 | 36.8 | 40.0 | 820 | 549.0 | 0 | 1690 | 62.93 | 62.24 | 1.01 | 7.47 | 225.00 |
| DL-2-77 | 77 | 7.69 | 5300 | 0.00 | 1320 | 18.5 | 38.0 | 47.6 | 820 | 585.0 | 0 | 1790 | 63.41 | 64.63 | 0.98 | 7.94 | 695.00 |
| DL-2-78 | 78 | 7.75 | 5350 | 0.00 | 1330 | 18.7 | 38.7 | 47.8 | 830 | 563.0 | 0 | 1800 | 63.92 | 64.62 | 0.99 | 7.95 | 766.00 |
| DL-2-79 | 79 | 7.71 | 5340 | 0.00 | 1330 | 19.2 | 39.7 | 48.7 | 830 | 540.0 | 0 | 1780 | 64.06 | 63.81 | 1.00 | 8.02 | 874.00 |
| DL-2-80 | 80 | 7.81 | 5320 | 0.00 | 1315 | 20.0 | 42.5 | 52.2 | 810 | 528.0 | 0 | 1810 | 63.84 | 63.49 | 1.01 | 7.99 | 706.00 |
| DL-2-81 | 81 | 8.07 | 5240 | 0.00 | 1335 | 19.7 | 41.0 | 47.8 | 830 | 612.0 | 0 | 1610 | 64.36 | 62.52 | 1.03 | 7.36 | 245.00 |
| DL-2-82 | 82 | 7.46 | 5420 | 0.00 | 1320 | 18.3 | 40.8 | 48.3 | 800 | 567.0 | 0 | 1800 | 63.68 | 63.86 | 1.00 | 8.11 | 1020.00 |
| DL-2-83 | 83 | 7.71 | 5540 | 0.00 | 1330 | 22.2 | 50.1 | 51.0 | 780 | 560.0 | 0 | 1760 | 65.12 | 62.49 | 1.04 | 7.55 | 207.00 |


| 008 Ca | E8 2 | $90^{\circ} \mathrm{I}$ | t0 29 | Ss＇s9 | 0281 | 0 | 0 Sts | 0tL | £＇8t | 8＇19 | $0 \cdot \varepsilon z$ | 0z¢ı | 000 | 019S | E8L | 011 | 011－z－7a |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 00．0101 | ZLL | ＋0＇ 1 | 98 ＇19 | Et＇t9 | 0981 | 0 | 0 s 0 s | 0tL | 6.95 | E＇6S | く＇IZ | 06ZI | $00^{\circ}$ | 089S | $8 L L$ | 601 | $601-z-70$ |
| 00.929 | $68 . \mathrm{L}$ | $80^{\circ} \mathrm{I}$ | L0＇19 | ャで99 | 0181 | 0 | 002s | 0EL | 9．9s | I＇s9 | 0 －$\varepsilon z$ | 0z\＆1 | 000 | 069s | $18^{\circ} \mathrm{L}$ | 80 I | $80 \mathrm{I}-7-\mathrm{Ta}$ |
| 00＇798 | $58 . \mathrm{L}$ | $\angle 0^{\circ} \mathrm{I}$ | £0＇19 | E0＇s9 | $0<L \mathrm{I}$ | 0 | 098s | 0＋L | 0 ¢ $¢$ | ¢¢¢ | z＇zz | 00¢I | 000 | 0s9s | 58．${ }^{\circ}$ | LOI | L01－z－7a |
| 00＇t9 | E8L | $90^{\circ} \mathrm{I}$ | $0{ }_{\text {¢ }}$ 19 | £์＇s9 | 0881 | 0 | 088s | 0EL | ［8t | 9 ＇t9 | してz | 01є1 | $00^{\circ}$ | 0¢9s | $68^{\circ} \mathrm{L}$ | 90 I | $901-z-7 \mathrm{~T}$ |
| 00＾ท ${ }^{\text {¢ }}$ | 08． L | 80.1 | ¢ $\varepsilon^{-19}$ | $90 \cdot 99$ | 0181 | 0 | 008s | 0tL | 8 ts | I＇t9 | Ľz | 0Z£I | $00^{\circ}$ | 0s9s | $6 L^{\prime} \mathrm{L}$ | S0I | S01－z－7a |
| 00．0801 | L8． | ${ }^{50} 1$ | \＆L＇19 | てL＇ャ9 | 0581 | 0 | 0＇E6t | OSL | ¢＇9s | 6 \％9 | \＆¢ | 06ZI | $00 \%$ | 009s | $6 L^{\circ} \mathrm{L}$ | tol | ＋01－でTa |
| 00．0sII | 028 | $\mathrm{EO}^{\circ} \mathrm{I}$ | LS6S | IS＇19 | 0641 | 0 | 00st | 0t $\angle$ | 6＇$¢ 9$ | 99¢ | $9 \cdot 1 z$ | 0zzI | $00 \cdot 0$ | 006s | zS＇L | E01 | を0⿺－て－7a |
| 00 It8 | 518 | ＋0． 1 | 6t＇8S | 0609 | 0LLI | 0 | 0ıtı | 0tL | 695 | ¢¢¢ | ¢゙ı | 0zzI | $00^{\circ} 0$ | 0585 | 6S＇L | z0I | 201－z－7a |
| 00＇L69 | S1＇8 | E0＇I | z7＇8S | 21009 | 0881 | 0 | 0z9E | 0t $\angle$ | ［9t | 805 | 1•Iz | 0zてI | $00^{\circ} 0$ | 0¢8S | ELL | 101 | 101－z－7a |
| 00．060I | t18 | ${ }^{\text {E }}$－ 1 | $97^{\prime} 85$ | 26．6s | $0+81$ | 0 | 0 0tte | OSL | ［8t | $\varepsilon \angle t$ | Loz | 0zて1 | $00^{\circ} 0$ | 0SLs | LLL | 001 | 00I－z－7a |
| 00． 29 L | 97＇8 | ${ }^{0} 0.1$ | L6LS | LE09 | 0981 | 0 | $0 \mathrm{~S} 0 \varepsilon$ | OSL | 8 St | L＇8t | 「1て | 0¢ZI | $00^{\circ} 0$ | 0tLs | $9 L \cdot L$ | 66 | $66-2-7 \mathrm{I}$ |
| 00．0LS | $\mathrm{Sl}^{8} 8$ | $0^{-1}$ | $85^{\circ} \mathrm{LS}$ | $6 \mathrm{t}^{\circ} \mathrm{LS}$ | 0981 | 0 | 0.982 | OSL | $58 \varepsilon$ | $0 \cdot \mathrm{st}$ | 6.61 | 081 | $00^{\circ} 0$ | 0ILS | $96 . L$ | 86 | $86-z-7 \mathrm{~d}$ |
| 00＇ut | £18 | ${ }^{\text {E O }}$ I | Itics | 88：8S | 0681 | 0 | 0897 | 0tL | £0t | 6 6t | 0.02 | 01ZI | $00^{\circ}$ | 069s | 86 L | L6 | L6－z－7d |
| $00 \cdot \mathrm{zs}$ | $z L \cdot L$ | ${ }^{+0} 1$ | IE 8 S | 88.09 | 0181 | 0 |  | 094 | でで | 0 \％ | z＇6I | 097I | 000 | 029s | $69^{\circ} \mathrm{L}$ | 96 | 96－z－7a |
| $00 \mathrm{L8z}$ | 9¢ ${ }^{\circ}$ | $00^{-1}$ | 5165 | セ¢゙6s | 0ZLI | 0 | 08st | 092 | して¢ | ガt | $9.0 z$ | 0¢ZI | 000 | 09ss | 96. | S6 | ¢6－2－7d |
| 00 ¢ $<2$ | $85^{\prime} \mathrm{L}$ | $00 \cdot 1$ | 9685 | t＜$<85$ | 0 0¢9 | 0 | 007s | 092 | 0．9を | S＂St | でı | 01ZI | 000 | 0StS | $\mathrm{Sl}^{18}$ | ${ }^{\text {t6 }}$ | ャ6－z－7a |
| 00 $28 \varepsilon$ | $97^{\circ} \mathrm{L}$ | $90^{\circ} \mathrm{I}$ | L988 | 0079 | 08sı | 0 | $0 \cdot \mathrm{StS}$ | 094 | 60t | 0．0s | żz | 0＜ZI | 000 | 0¢Ss | ¢18 | E6 | \＆6－z－7a |
| 00＇661 | $88 . \mathrm{L}$ | 20.1 | 9L＇6s | L909 | 0781 | 0 | $0 \cdot \varepsilon 6 \varepsilon$ | 094 | L＇It | 6 6t | 7\％61 | OSZI | 000 | 0z9s | $89^{\circ} \mathrm{L}$ | 26 | 26－z－7a |
| 00＇t9¢ | $6 L^{\circ} \mathrm{L}$ | 860 | $0 \chi^{\circ} 09$ | 27\％ 6 | 0881 | 0 | 0．tzt | 0LL | 2＇8t | t＇6t | toz | 00ZI | 000 | 0¢9s | 19.4 | 16 | 16－z－7a |
| 00 TLZ | E6：L | 10.1 | £ ${ }^{\circ} 65$ | E965 | 0¢81 | 0 | 0688 | OSL | 0．9t | tos | toz | 0ızı | $00 \cdot 0$ | 009s | $99 . L$ | 06 | 06－z－7a |
| 00－86乏 | Et－8 | L0． 1 | S8＇19 | 96．s9 | $0<L I$ | 0 | 0 0¢¢s | 0LL | L＇LS | 6.05 | z＇zz | 0t£ | 000 | 009s | IS＇L | 68 | 68－z－7a |
| 00．98I | $56 . \mathrm{L}$ | ＋0． 1 | \＆1＇z9 | 99 ＇¢9 | 0＜81 | 0 | 001s | 0¢ $\llcorner$ | s ＇ts | 8.20 | 6．1z | 0¢£ | 000 | 08S | $2 S^{\circ} L$ | 88 | $88-z-7 \mathrm{a}$ |
| 00 LEt | $99 \cdot L$ | L0．${ }^{\text {I }}$ | $69 \% 5$ | ${ }^{\square} 6$ ¢ $¢ 9$ | 0¢91 | 0 | 088s | $08 L$ | 6.4 | でをt | $\varsigma$ 江 | 0て£ | $00^{\circ}$ | 06zs | \＆18 | $\angle 8$ | L8－z－7 |
| $00 \angle L \square$ | $9 t^{\circ} \mathrm{L}$ | $60^{\circ} \mathrm{I}$ | 58\％6 | 80＇s9 | 06SI | 0 | 0－29s | 08L | －8 ${ }^{\text {t }}$ | L＇St | $\tau$ て̇ | 0¢EI | $00^{\circ} 0$ | 08£ | E08 | 98 | 98－でフa |
| 00 zlz | $86 . L$ | 10.1 | \＆0＇t9 | ¢どャ9 | 0¢81 | 0 | 0 － 995 | 062 | 69t | $9.8 t$ | 0 \％z | 0てEI | 000 | 019s | $99^{\circ} \mathrm{L}$ | 58 | ¢8－z－7a |
| 00 ¢61 | ULL | S0＇I | 6179 | zI＇s9 | 0181 | 0 | 0.905 | 08 L | los | 9.05 | \＆z | 0¢£ | $00^{\circ}$ | 019s | $9 L L$ | ャ8 | ＋8－z－7 |
| $\overbrace{2} \mathrm{~S}$ | $\begin{aligned} & \mathrm{H}^{\mathrm{d}} \\ & \text { YTV } \end{aligned}$ |  | $\begin{gathered} \text { suopuv } \\ \text { I\#10 } \end{gathered}$ |  |  | $\begin{array}{\|c} (\operatorname{modd} \mp M) \\ -{ }^{6} \mathbf{O} \mathrm{O} \end{array}$ | $z^{\text {b }}$ OS | IJ | $+z^{8}$ | ${ }_{+2^{8} \mathrm{C}}$ | ＋+ | ${ }_{+}^{\text {E }}$ N | ${ }^{19}$ | puo？ | $\mathrm{H}^{\text {d }}$ | $\stackrel{\text {（p）awill }}{ }$ －qnouI | II Pdues |

A7 Results of chemical analysis in DL-3 system (sample type: RelW)

| Sample ID | Incub. <br> Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | Mg ${ }^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | Cl | $\mathbf{S O}_{4}{ }^{\text {2- }}$ | $\begin{gathered} \mathrm{CO}_{3}{ }^{2-} \\ (\mathrm{wt} \mathrm{ppm}) \end{gathered}$ | $\begin{gathered} \mathbf{H C O}_{3}^{-} \\ (\mathrm{wt} \mathrm{ppm}) \end{gathered}$ | $\underset{\text { Tatal }}{\text { Tans }}$ | Total Anions | $\begin{aligned} & \text { Ratio } \\ & \text { Cat/Ani } \end{aligned}$ | $\underset{\underset{\mathrm{pH}}{\mathrm{ALK}}}{\substack{\text { and }}}$ | $\mathbf{S}^{2+}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-3-3 | 3 | 8.07 | 5720 | 5 | 1021 | 15.4 | 14.2 | 21.0 | 830 | 41.1 | 0.0 | 1510 | 47.0 | 49.1 | 0.96 | 7.64 | 15.80 |
| DL-3-4 | 4 | 8.06 | 5900 | 9 | 1021 | 15.7 | 13.9 | 20.6 | 820 | 36.9 | 0.0 | 1510 | 47.0 | 48.7 | 0.96 | 7.70 | 15.30 |
| DL-3-5 | 5 | 7.78 | 5910 | 1 | 1027 | 15.1 | 14.1 | 21.3 | 830 | 14.3 | 0.0 | 1550 | 47.3 | 49.1 | 0.96 | 7.48 | 7.37 |
| DL-3-6 | 6 | 7.79 | 5630 | 1 | 1030 | 15.4 | 14.2 | 21.5 | 830 | 14.3 | 0.0 | 1590 | 47.4 | 49.8 | 0.95 | 7.57 | 7.42 |
| DL-3-7 | 7 | 7.80 | 5630 | 1 | 1030 | 15.1 | 14.1 | 21.3 | 820 | 14.2 | 0.0 | 1520 | 47.4 | 48.3 | 0.98 | 7.53 | 7.55 |
| DL-3-8 | 8 | 7.79 | 5830 | 1 | 1035 | 15.1 | 14.1 | 21.3 | 830 | 15.4 | 0.0 | 1550 | 47.6 | 49.1 | 0.97 | 7.57 | 7.60 |
| DL-3-9 | 9 | 7.78 | 5780 | 1 | 1045 | 14.9 | 14.1 | 21.2 | 830 | 14.4 | 0.0 | 1550 | 48.1 | 49.1 | 0.98 | 7.58 | 7.67 |
| DL-3-10 | 10 | 7.78 | 5600 | 1 | 1045 | 15.0 | 14.1 | 21.3 | 840 | 17.3 | 0.0 | 1520 | 48.1 | 49.0 | 0.98 | 7.47 | 7.68 |
| DL-3-11 | 11 | 7.90 | 5510 | 1 | 1060 | 14.9 | 14.1 | 21.2 | 830 | 13.8 | 0.0 | 1550 | 48.7 | 49.1 | 0.99 | 7.60 | 7.47 |
| DL-3-12 | 12 | 7.90 | 5610 | 1 | 1090 | 16.1 | 15.4 | 23.9 | 830 | 14.2 | 0.0 | 1530 | 50.3 | 48.8 | 1.03 | 7.57 | 7.59 |
| DL-3-13 | 13 | 7.79 | 5490 | 6 | 1100 | 16.1 | 15.3 | 23.7 | 830 | 5.0 | 0.0 | 1560 | 50.7 | 49.1 | 1.03 | 7.57 | 6.71 |
| DL-3-14 | 14 | 7.77 | 5410 | 5 | 1120 | 16.3 | 15.2 | 23.4 | 830 | 11.9 | 0.0 | 1560 | 51.6 | 49.3 | 1.05 | 7.50 | 6.83 |
| DL-3-15 | 15 | 7.65 | 5090 | 5 | 1110 | 16.1 | 14.8 | 22.6 | 830 | 13.7 | 0.0 | 1580 | 51.0 | 49.6 | 1.03 | 7.48 | 6.91 |
| DL-3-16 | 16 | 7.77 | 5060 | 5 | 1100 | 16.4 | 14.7 | 22.5 | 830 | 11.3 | 0.0 | 1550 | 50.6 | 49.1 | 1.03 | 7.59 | 6.62 |
| DL-3-17 | 17 | 7.77 | 5111 | 6 | 1110 | 16.2 | 14.7 | 22.4 | 830 | 9.8 | 0.0 | 1560 | 51.0 | 49.2 | 1.04 | 7.60 | 6.43 |
| DL-3-19 | 19 | 7.82 | 5300 | 6 | 1130 | 16.5 | 14.7 | 22.5 | 830 | 8.2 | 0.0 | 1560 | 51.9 | 49.2 | 1.05 | 7.69 | 6.25 |
| DL-3-20 | 20 | 7.82 | 5280 | 6 | 1110 | 16.2 | 14.6 | 22.2 | 840 | 10.5 | 0.0 | 1550 | 51.0 | 49.4 | 1.03 | 8.02 | 6.14 |
| DL-3-21 | 21 | 7.83 | 5140 | 6 | 1120 | 16.3 | 14.7 | 22.4 | 840 | 9.8 | 0.0 | 1620 | 51.5 | 50.5 | 1.02 | 7.70 | 6.11 |
| DL-3-22 | 22 | 7.84 | 5160 | 6 | 1110 | 16.1 | 14.7 | 22.3 | 850 | 6.9 | 0.0 | 1570 | 51.0 | 49.9 | 1.02 | 7.70 | 6.12 |
| DL-3-23 | 23 | 8.04 | 4630 | 5 | 1100 | 15.8 | 15.6 | 24.5 | 780 | 8.7 | 0.0 | 1630 | 50.8 | 48.9 | 1.04 | 7.75 | 5.50 |
| DL-3-24 | 24 | 8.06 | 4740 | 6 | 1090 | 15.9 | 15.3 | 23.9 | 780 | 8.6 | 0.0 | 1600 | 50.3 | 48.5 | 1.04 | 7.78 | 5.54 |
| DL-3-25 | 25 | 8.05 | 4640 | 9 | 1100 | 16.3 | 15.6 | 24.3 | 780 | 7.6 | 0.0 | 1590 | 50.8 | 48.3 | 1.05 | 7.68 | 5.10 |
| DL-3-26 | 26 | 8.03 | 4490 | 9 | 1120 | 16.5 | 15.4 | 23.8 | 790 | 5.0 | 0.0 | 1630 | 51.6 | 49.2 | 1.05 | 7.75 | 5.16 |
| DL-3-27 | 27 | 8.01 | 4580 | 9 | 1115 | 16.8 | 15.4 | 23.7 | 790 | 5.2 | 0.0 | 1590 | 51.4 | 48.5 | 1.06 | 7.80 | 5.19 |
| DL-3-28 | 28 | 7.99 | 4500 | 13 | 1120 | 16.5 | 15.3 | 23.6 | 800 | 5.3 | 0.0 | 1590 | 51.6 | 48.9 | 1.06 | 7.74 | 5.27 |
| DL-3-29 | 29 | 7.86 | 4780 | 14 | 1040 | 14.8 | 13.2 | 19.7 | 800 | 6.4 | 0.0 | 1650 | 47.7 | 49.9 | 0.96 | 7.69 | 5.56 |

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| $0 \varepsilon^{\circ} \angle 1$ | $\dagger_{L} L^{\circ}$ | 460 | tils | 96t | 08SI | 00 | ${ }^{6}$ It | 0ヶ8 | z $7 \%$ | でゅ | I＇si | 0801 | $\angle L$ | 0¢6t | £ 8.2 | Ss | ¢¢－₹－7a |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| LL\％ | $L L \cdot L$ | 960 | s＇Is | 96 t | 0091 | 00 | 2．0z | 058 | 1•Iて | 6 ¢ 1 | ¢＇sı | 0801 | $\dagger L$ | 068t | $78 . L$ | ts | ャร－¢－7d |
| SL＇6 | ZLLL | 960 | s＇Is | 96 | 0091 | 00 | $1 \cdot 1 z$ | 058 |  | $6 \mathrm{\varepsilon}$ I | 9＇sı | 0801 | $\varepsilon L$ | 088t | $98 . \mathrm{L}$ | £ร | £ร－£－7］ |
| 018 8 | $8 L^{\circ} \mathrm{L}$ | 860 | £＇Is | 0.05 | 06SI | 00 | 9 Cl | 098 |  | 0 － 1 | L＇si | 060I | Is | 0＜8t | $88^{\circ} \mathrm{L}$ | zs | $2 s-\varepsilon-7 \mathrm{a}$ |
| $\angle 8.6$ | $6 L^{\circ} \mathrm{L}$ | $\angle 60$ | s＇is | 0.05 | 0091 | 00 | E．61 | 098 | がレて | I＇ti | 6s | 060I | 6 t | 098t | ${ }^{5} 6 . L$ | 15 | ıs－£－70 |
| $00^{\circ} 01$ | LL＇L | $\angle 60$ | z＇zs | LOS | 0z91 | 00 | 9 zz | $0 \angle 8$ | E\％1 | 6 zl | 9＊t | OLII | 0 ¢ | 068t | $\varepsilon 6^{\circ} \mathrm{L}$ | 0 S | $05^{-\varepsilon-10}$ |
| 0ャ01 | 18. | 560 | $0.7 s$ | 560 | 0t91 | 00 | 097 | 058 | $01 \%$ | $\llcorner\mathcal{L} \mathfrak{L}$ | 0 ¢ | 0801 | 6 t | 0LLt | 96 L | $6{ }^{6}$ | $6 t-\varepsilon-7 \mathrm{C}$ |
| 0801 | 8L＇L | ＋60 | sıls | $9.8 t$ | 06SI | 00 | †Lz | 098 | 9.02 | $\varsigma \stackrel{\varepsilon}{1}$ | 6 tI | 0901 | $6 t$ | 06Lt | 96 L | $8{ }^{\text {t }}$ | $8 t-\varepsilon-7 \mathrm{~T}$ |
| 08\％1 | $\downarrow L^{\circ} \mathrm{L}$ | 560 | $\pm$ IS | $9.8 t$ | 06S 1 | 00 | 197 | 098 | £ $0 \sim$ | $\varepsilon \varepsilon \boxed{1}$ | ガち | 0901 | 6 t | 008t | $88^{\circ} \mathrm{L}$ | Lt | $\angle t-\varepsilon-7 \mathrm{~T}$ |
| $00 \cdot \mathrm{II}$ | $69^{\circ} \mathrm{L}$ | 960 | 9 is | ＋6t | 08SI | 00 | $\varepsilon \% z$ | $0 \angle 8$ | ¢61 | 8＇zı | $6 \varepsilon 1$ | 0801 | 6 t | $018 t$ | $\pm 6 . L$ | $9{ }^{+}$ | $9 t-\varepsilon-7 \mathrm{C}$ |
| 0s＇II | $L 9^{\circ} \mathrm{L}$ | 860 | £＇Is | s．0s | 085 1 | 00 | $\varepsilon L L$ | 098 | 8.17 | でか1 | I＇sI | 00II | Is | 008t | 68 L | St | St－$\varepsilon-7 \mathrm{~T}$ |
| 06 II | $59 \cdot L$ | 660 | $\downarrow$ IS | $0 \cdot \mathrm{IS}$ | 06SI | 0.0 | L＇レE | 058 | $0 \cdot \varepsilon \tau$ | 6tI | $\mathfrak{C}$ | 0III | os | 008t | $58 . L$ | to |  |
| 0t゙II | SL＇L | $10^{\circ} \mathrm{I}$ | E0S | 9.05 | 06SI | 00 | $s s z$ | 078 | $\downarrow \downarrow z$ | 9 91 | $\downarrow$ ¢ | 0011 | $\angle \mathrm{t}$ | OLLt | 98 L | Et | $\mathfrak{E t - \varepsilon}$－Ta |
| 0s\％ol | $\varepsilon L^{\prime} L$ | 660 | ros | L＇6t | 08SI | 00 | ¢9\％ | 078 | 881 | ¢＇zı | $\downarrow$ †¢ | 0601 | $\angle t$ | 06Lt | 58. | てt | てt－$\varepsilon$－ $7 a$ |
| $00^{\prime \prime}$ | $0 L^{\circ} \mathrm{L}$ | $\varepsilon^{\circ} \mathrm{I}$ | ros | ${ }^{\circ} \mathrm{IS}$ | 08SI | 00 | LSz | 078 | szz | 9＊1 | $\dagger$ ¢ | 0211 | ${ }^{8+}$ | 06Lt | $8 L^{\circ} \mathrm{L}$ | It | It－を－Ta |
| $00 \cdot 11$ | $99^{\circ} \mathrm{L}$ | $00^{\circ}$ | s．os | s．os | 0091 | 00 | L＇9z | 078 | 0 0 $z$ | ガちI | z＇si | 00II | $\angle t$ | 08Lt | E8 ${ }^{\circ} \mathrm{L}$ | $0{ }^{0}$ | $00^{-\varepsilon-1 / 7}$ |
| 2L6 | LE： 8 | $00^{\prime}$ | $\angle 0 S$ | 6.05 | 0z91 | 00 | ゅで | 028 | $6 \cdot 1 z$ |  | ISI | 01II | $9 \downarrow$ | $018{ }^{\circ}$ | $8 L^{\circ} \mathrm{L}$ | $6 \varepsilon$ | $6 \varepsilon-\varepsilon-7 a$ |
| 0001 | LL＇L | 20.1 | 805 | 8.15 | 0z91 | 00 | と：8z | 078 | $z z z$ | ぢカI | E＇si | 0 0ıI | $8{ }^{\text {b }}$ | 0785 | $8 L^{\circ} \mathrm{L}$ | $8 \varepsilon$ | $8 \varepsilon-\varepsilon \cdot 7 \mathrm{~T}$ |
| zI＇s | $t \angle L$ | 860 | S＇6t | 9.85 | 06SI | 00 | $9 \cdot 9$ | 028 | z＇0z | $\bigcirc \mathcal{E} 1$ | 6.71 | 0901 | $\angle 1$ | 08Lt | 96 L | LE | LE－$\varepsilon-7 \mathrm{~T}$ |
| 29 ¢ | IL＇L | 660 | ［6t | L＇st | 08SI | 00 | 02 | 018 | ${ }^{\circ} \mathrm{OR}$ | 6 6I | ［＇si | 0901 | 4 | 008t | 56.4 | $9 \varepsilon$ | $9 \varepsilon-\varepsilon-7 \mathrm{~T}$ |
| $s z \mathrm{~s}$ | $L L L$ | $00^{\circ} \mathrm{I}$ | 1＇6t | $0 \cdot 6$ | 08SI | 00 | L＇9 | 018 | 008 | $\vdash \mathcal{L}$ | 8 tl | 0201 | 91 | 06Lt | 16.4 | ¢ร | ¢ $\varepsilon$－$\varepsilon-7 \mathrm{~T}$ |
| $0 \varepsilon \cdot \varsigma$ | ZLL | 660 | L＇6t | 16t | 0z91 | 00 | $\llcorner 9$ | 018 |  | 0 ¢ ${ }^{\text {d }}$ | ¢ SI | 0LOI | 91 | 008t | $\varepsilon 6^{\circ} \mathrm{L}$ | $\downarrow \varepsilon$ | $\downarrow$ เร－$\varepsilon$－ 7 d |
| Its | $89^{\circ} \mathrm{L}$ | $20 \cdot 1$ | 2\％6t | ros | 06SI | 00 | ¢9 | 018 | ゅてz | L＇t | L－si | 0601 | sı | 08Lt | 68 L | £์ | $\varepsilon \varepsilon-\varepsilon-T \mathrm{G}$ |
| $8 \varepsilon^{\prime} \mathrm{S}$ | $9 L^{\circ} \mathrm{L}$ | 20.1 | 16t | 00 s | 08SI | $0 \cdot 0$ | $\downarrow 9$ | 018 | I＇Iz | Iti | $\varsigma ¢$ | 0601 | 91 | 088t | $58 . \mathrm{L}$ | て¢ | て¢－£－7 |
| 60 S | $\mathcal{E} L \cdot L$ | 960 | 685 | $て ゙ く \downarrow$ | 06SI | 00 | $\varepsilon 9$ | 008 | 161 | 6 zl | 9 tI | 0EOI | SI | 06Lt | $98 . \mathrm{L}$ | เع | เ $\varepsilon$－$\varepsilon-7 \mathrm{C}$ |
| 07＇s | $9 L \cdot L$ | 660 | 685 | S 85 | 06SI | $0 \cdot 0$ | $\llcorner\cdot 9$ | 008 | t61 | İદ1 | $8 \mathfrak{t 1}$ | 0901 | SI | 088t | L8＇L | $0 \varepsilon$ | $0 \varepsilon-\varepsilon-7 \mathrm{~J}$ |
| $\overbrace{}^{-} \mathrm{S}$ | $\begin{aligned} & \mathbf{H d}^{\mathrm{d}} \\ & \text { yTV } \end{aligned}$ | $\begin{aligned} & \hline!\overline{\text { UVABD }} \\ & \text { ongy } \end{aligned}$ | $\begin{array}{\|c} \hline \text { suoluv } \\ \text { Iघiol } \end{array}$ | $\begin{gathered} \text { suolige } \\ \text { IEqo. } \end{gathered}$ |  | $\left\|\begin{array}{c} (\text { (udd } 3 M) \\ -200 \end{array}\right\|$ | $\overbrace{}^{\text {ºs }}$ | －10 | $+i^{\text {B }}$ O | $+z^{8} \mathbf{N}$ | ＋+ | ${ }^{\text {B }} \mathrm{N}$ | ${ }^{18}$ | puob | $\mathrm{H}^{\text {d }}$ |  | II Pdurs |



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| 00 LtI | 06.2 | 660 | て＇LS | t＇9s | 01 tI | $0 \cdot 0$ | Ltt | OLL | 9 9 $\varepsilon$ | ¢91 | z＇91 | 0¢zi | 0sz | 090s | 678 | 901 | $90 \mathrm{I}-\varepsilon-\mathrm{Ta}$ |
| 0001 It | 96.4 | 660 | $5 \cdot 95$ | L＇ss | 0itl | $0 \cdot 0$ | 97t | 092 | $\varepsilon \leq ร$ | －＇91 | 1＇91 | 0ızı | osz | 00IS | LI＇8 | s0I | S0I－-7 Ta |
| 00.9 LI | L6L | $00^{\circ} \mathrm{I}$ | 9.5 | L＇ts | 00t 1 | $0 \cdot 0$ | $65 \varepsilon$ | $0 \angle L$ | ¢＇£ | z＇91 | \＆．91 | 06II | $00 z$ | 060S | 918 | tor | toi－E－Ta |
| $00^{\circ} \mathrm{E} L$ | $9{ }^{1} 8$ | 660 | $L$＇ts | I＇ts | 08SI | 00 | $0 z z$ | OLL | 0 0 $\varepsilon$ | 6 bl | z＇sı | 08II | $00 z$ | ${ }^{066} \mathrm{t}$ | $8 L^{\circ} \mathrm{L}$ | E0I | £01－-7 Ta |
| $00^{\circ} \mathrm{t}$ L | t18 | 460 | 8 ＇s | z＇¢ | 0LSI | 00 | 0¢z | OSL |  | 0 －si | $\angle \cdot \mathrm{SI}$ | 09II | 0¢z | 068t |  | z01 |  |
| $06 \cdot 98$ | L0＇8 | 860 | 0 ¢s | $8 \times 5$ | OISI | 00 |  | 008 | $\varepsilon \downarrow \tau$ | L＇SI | 8 SI | 0LII | $01 z$ | 026t | $\pm L L$ | 101 | 101－-7 Ta |
| 0¢＇98 | E0\％8 | 560 | $9 . \mathrm{s}$ | 0 0 \％ | 00SI | 00 | ¢9z | 018 | $8.7 \tau$ | 0.91 | 8 SI | 0 SLI | 0ız | 086t | $\varepsilon L \cdot L$ | 00I | $001-\varepsilon-7 \mathrm{~T}$ |
| $08: 86$ | t18 | 960 | $s t s$ | ゅis | 0 ¢ ${ }^{\text {I }}$ | $0 \cdot 0$ | sLz | 018 | $\downarrow$ ¢ $\downarrow$ | z＇si | z＇91 | 0tIt | $00 z$ | 066t | 89.4 | 66 | 66－\＆－7a |
| 0¢886 | 808 | S60 | 0 ＇ts | sis | 0 ¢ $\dagger 1$ | $0 \cdot 0$ | £92 | 008 | $0 \cdot \varepsilon \tau$ | 6 tl | 0.91 | 0zil | $00 z$ | 086t | L9．L | 86 | $86-\varepsilon-7 \mathrm{~T}$ |
| 0686 | t18 | 660 | $\varsigma ¢ \varsigma$ | 875 | 0 ¢ $\dagger 1$ | $0 \cdot 0$ | tsz | 062 | $\tau$ ¢ | I＇si | で91 | 0SII | $00 z$ | 026t | ILL | L6 | L6－$\varepsilon$－ 7 a |
| $0 \downarrow$＋ 9 | £0．8 | 960 | 8 ＇ts | ¢ $¢ \mathrm{~s}$ | ossi | 00 | S6I | 018 | \＆$\because 乙$ | 8.91 | 0 －sı | 0blit | $00 z$ | 086t | $68^{\circ} \mathrm{L}$ | 96 | 96－$\varepsilon$－ 70 |
| 0602 | $88^{\circ} \mathrm{L}$ | S60 | L＇ts | 6.15 | ${ }_{0+S} 1$ | $0 \cdot 0$ | 012 | 008 | $6 . z z$ | 67t | 8 SI | 0عII | $00 z$ | 0ع6t | $78 . \mathrm{L}$ | S6 | ¢6－-7 Ta |
| 08＇SL | $58 . \mathrm{L}$ | ＋60 | 8．ss | $\downarrow$ ¢ | 0091 | 00 | $0 z z$ | 008 | $0 \cdot \varepsilon z$ | 6 tl | $\downarrow$＇si | 0ヵII | $00 z$ | 0＜8t | S6． L | ${ }^{\text {t6 }}$ | เ6－$\varepsilon$－ 7 a |
| 0く＇E9 | $98^{\circ} \mathrm{L}$ | ＋6．0 | †＇Ss | ${ }_{6}$ IS | 0191 | 00 | 061 | 008 | †¢ | 0 SI | t－sI | 0¢II | $00 z$ | 0¢8t | $66^{\circ} \mathrm{L}$ | £6 | £6－－-T |
| 0＜＇E9 | L0．8 | £60 | L＇ss | 6.15 | 08SI | 00 | 061 | 088 | I＇£z | I＇si | 9＇st | 0عII | $00 z$ | 028t | S6． L | 76 | 26－\＆－7a |
| 0¢゙K | S0．8 | E60 | 9．ss | sis | Stst | 00 | 012 | 088 | L̇z | $8{ }^{\text {b }}$ I |  | OZII | $00 z$ | $0+8 t$ | $\varepsilon 6 L$ | 16 | $16-\varepsilon-7 \mathrm{~T}$ |
| 00\％$\angle 9$ | 86.2 | S6．0 | 0 ＇ts | z＇is | 08tI | 00 | L6I | 088 | 8 zz | 6＋1 | $\tau$＇si | sili | 08I | 08Lt | L9LL | 06 | 06－$\varepsilon$－ 7 T |
| 00\％ 29 | It 8 | ${ }^{6} 60$ | $\varepsilon$ vts | z＇IS | 06tI | 0.0 | soz | 088 | 8 zz | 6tt | $\tau$＇si | Sili | 08I | 008t | ZLLL | 68 | 68－£－7a |
| 0t＇6t | 20.8 | ＋6．0 | I＇ts | 0 ots | ossi | 00 | 9ti | 0¢8 | く̇z | L＇ti | r＇si | 01II | 081 | 08Lt | 0LL | 88 | 88－$\varepsilon$－ 7 T |
| 0¢0s | $20 \cdot 8$ | £60 | sts | 9.05 | 09SI | 0.0 | LtI | $0+8$ | I $\varepsilon \tau$ | 6 ta | t＇sı | 0011 | 081 | 08Lt | $\varepsilon L \cdot L$ | $\angle 8$ | L8－$\varepsilon$－-T |
| $096 \varepsilon$ | $90 \cdot 8$ | 260 | L＇Ss | 0 is | 0991 | 0.0 | SII | 058 | 8＇zz | L＇ti | 6SI | 01II | $0<1$ | 0¢Lt | 6LL | 98 | 98－を－7］ |
| 0¢68 | tis 8 | 260 | 0 ＇ts | L－6t | 00s 1 | rse | zzI | $0+8$ | 8＇zz | 8 tl | I＇si | 0801 | 091 | 068＞ | SLLL | 58 | ¢8－$\varepsilon$－-T |
| 09 tot | 9 c 8 | 060 | $\dagger$ ¢s | L＇6t | 0¢SI |  | İI | 088 | $0 \cdot \varepsilon z$ | 8 tI | I＇si | 0801 | $0<1$ | 010s | $\varepsilon L L$ | ¢8 | เ8－$\varepsilon$－ 7 T |
| $0 \varepsilon$ ¢ $\dagger$ | 9£＇8 | 060 | £＇s¢ | 866 | $0 ¢ \mathcal{S}$ | ¢ 91 | $0 \varepsilon I$ | 088 | $\tau$ \％ | 0 SI | $\tau$ ¢ ${ }^{\text {s }}$ | 0801 | $0<1$ | 0ع6t | SLLL | £8 | £8－₹－Ta |
| 0Sttot | £¢．8 | 160 | 8 ts | Ios | ossi | $0 \cdot 0$ | £ยı | $0 \angle 8$ | し＇zz | L＇tı | 6 bl | 0601 | $0<1$ | 09St | $\varepsilon L \cdot L$ | 28 | 28－£－7 |
| ${ }_{-2} \mathrm{~S}$ | $\begin{aligned} & \text { Hed }^{\text {YTV }} \end{aligned}$ |  | $\begin{gathered} \text { suopuy } \\ \text { [B]OL } \end{gathered}$ |  |  |  | ${ }_{7}^{7} \mathrm{OS}$ | － 1 | $+2^{\text {b }}$ S | $+z^{8} \mathrm{~L}$ | ${ }_{+}+$ | ${ }^{\text {B }} \mathrm{N}$ | $\pm 19$ | puos | $\mathrm{H}^{\text {d }}$ |  | （II Pdues |

A7 Results of chemical analysis in DL-3 system (sample type: RelW) (cont'd)

| Sample ID | Incub. Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | Cl | $\mathrm{SO}_{4}{ }^{2}$ | $\begin{array}{\|c\|} \mathbf{C O}_{3}{ }^{2-} \\ (\mathbf{w t} \text { ppm }) \\ \hline \end{array}$ | $\begin{array}{\|c\|} \mathbf{H C O}_{3}{ }^{-} \\ \text {(wt ppm) } \end{array}$ | $\begin{gathered} \text { Total } \\ \text { Cations } \\ \hline \end{gathered}$ | Total <br> Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \\ \hline \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \end{gathered}$ | $\mathbf{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-3-108 | 108 | 8.08 | 5070 | 260 | 1250 | 16.5 | 16.3 | 24.2 | 770 | 387 | 0.0 | 1410 | 57.3 | 56.1 | 1.02 | 7.97 | 189.00 |
| DL-3-109 | 109 | 8.44 | 5080 | 260 | 1250 | 16.4 | 16.1 | 18.7 | 770 | 432 | 0.0 | 1390 | 57.0 | 56.7 | 1.01 | 7.89 | 386.00 |
| DL-3-110 | 110 | 8.16 | 5090 | 260 | 1230 | 16.5 | 16.1 | 24.2 | 780 | 485 | 0.0 | 1400 | 56.5 | 58.3 | 0.97 | 7.95 | 193.00 |
| DL-3-111 | 111 | 8.22 | 5090 | 260 | 1240 | 16.4 | 16.1 | 23.3 | 780 | 466 | 0.0 | 1370 | 56.8 | 57.4 | 0.99 | 7.88 | 149.00 |
| DL-3-112 | 112 | 8.17 | 5070 | 260 | 1260 | 16.2 | 16.0 | 23.6 | 790 | 410 | 0.0 | 1440 | 57.7 | 57.7 | 1.00 | 7.99 | 211.00 |
| DL-3-113 | 113 | 8.13 | 5140 | 260 | 1290 | 16.0 | 15.9 | 23.5 | 780 | 373 | 0.0 | 1460 | 59.0 | 56.9 | 1.04 | 8.09 | 172.00 |
| DL-3-114 | 114 | 8.21 | 5010 | 210 | 1310 | 15.8 | 15.6 | 22.7 | 830 | 351 | 0.0 | 1470 | 59.8 | 57.4 | 1.04 | 8.13 | 141.00 |
| DL-3-116 | 116 | 8.36 | 5090 | 270 | 1290 | 16.2 | 16.1 | 12.2 | 820 | 154 | 43.2 | 1570 | 58.5 | 56.9 | 1.03 | 8.48 | 161.00 |
| DL-3-117 | 117 | 8.67 | 5240 | 290 | 1310 | 16.3 | 16.4 | 9.8 | 820 | 269 | 143.0 | 1350 | 59.2 | 59.2 | 1.00 | 8.90 | 309.00 |
| DL-3-118 | 118 | 8.79 | 5170 | 280 | 1310 | 16.7 | 16.3 | 9.3 | 790 | 346 | 85.5 | 1420 | 59.2 | 59.1 | 1.00 | 8.75 | 235.00 |
| DL-3-119 | 119 | 8.43 | 5300 | 290 | 1300 | 16.6 | 16.5 | 13.8 | 810 | 340 | 88.2 | 1430 | 59.0 | 59.9 | 0.99 | 8.70 | 115.00 |
| DL-3-120 | 120 | 8.60 | 5220 | 290 | 1300 | 16.4 | 16.2 | 9.8 | 810 | 220 | 142.0 | 1360 | 58.8 | 58.1 | 1.01 | 8.90 | 224.00 |
| DL-3-121 | 121 | 8.59 | 5170 | 280 | 1320 | 16.3 | 17.0 | 14.3 | 800 | 161 | 83.1 | 1450 | 59.9 | 55.9 | 1.07 | 8.71 | 194.00 |
| DL-3-122 | 122 | 8.59 | 5210 | 290 | 1280 | 16.3 | 16.7 | 12.9 | 800 | 161 | 77.4 | 1460 | 58.1 | 56.0 | 1.04 | 8.69 | 262.00 |
| DL-3-123 | 123 | 8.59 | 5010 | 380 | 1280 | 16.0 | 16.5 | 16.1 | 860 | 342 | 0.0 | 1490 | 58.2 | 60.5 | 0.96 | 7.89 | 131.00 |
| DL-3-124 | 124 | 8.56 | 5130 | 370 | 1260 | 16.2 | 16.4 | 15.9 | 860 | 343 | 0.0 | 1560 | 57.4 | 61.6 | 0.93 | 8.19 | 136.00 |
| DL-3-125 | 125 | 8.64 | 5070 | 370 | 1250 | 16.3 | 16.3 | 15.5 | 870 | 279 | 0.0 | 1490 | 56.9 | 59.4 | 0.96 | 8.06 | 146.00 |
| DL-3-126 | 126 | 8.54 | 5140 | 370 | 1240 | 15.9 | 15.8 | 15.2 | 860 | 231 | 0.0 | 1450 | 56.4 | 57.4 | 0.98 | 7.96 | 84.30 |
| DL-3-127 | 127 | 8.40 | 5100 | 370 | 1230 | 15.4 | 15.0 | 13.9 | 860 | 318 | 0.0 | 1480 | 55.8 | 59.7 | 0.93 | 7.86 | 92.90 |
| DL-3-128 | 128 | 8.36 | 5220 | 370 | 1260 | 16.0 | 15.6 | 14.4 | 860 | 392 | 0.0 | 1480 | 57.2 | 61.3 | 0.93 | 7.93 | 108.00 |
| DL-3-129 | 129 | 8.25 | 5090 | 370 | 1260 | 16.1 | 15.9 | 16.9 | 860 | 464 | 0.0 | 1460 | 57.4 | 62.5 | 0.92 | 8.08 | 142.00 |
| DL-3-130 | 130 | 8.23 | 5170 | 360 | 1300 | 16.4 | 16.1 | 17.1 | 850 | 448 | 0.0 | 1450 | 59.1 | 61.5 | 0.96 | 7.84 | 174.00 |
| DL-3-131 | 131 | 8.06 | 5190 | 370 | 1290 | 16.8 | 17.0 | 22.4 | 850 | 410 | 0.0 | 1450 | 59.1 | 60.9 | 0.97 | 7.89 | 148.00 |
| DL-3-132 | 132 | 8.04 | 5180 | 370 | 1320 | 16.8 | 16.9 | 22.1 | 850 | 425 | 0.0 | 1450 | 60.3 | 61.2 | 0.99 | 7.84 | 149.00 |
| DL-3-133 | 133 | 8.01 | 5200 | 380 | 1310 | 16.6 | 16.6 | 21.8 | 880 | 446 | 0.0 | 1450 | 59.9 | 62.6 | 0.96 | 7.86 | 149.00 |
| DL-3-134 | 134 | 8.37 | 5620 | 370 | 1320 | 20.1 | 17.0 | 25.5 | 860 | 299 | 0.0 | 1530 | 60.6 | 60.2 | 1.01 | 7.97 | 178.00 |

A7 Results of chemical analysis in DL-3 system (sample type: RelW) (cont'd)

| Sample ID | Incub. <br> Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{2-}$ | $\begin{gathered} \mathbf{C O}_{3}{ }^{2-} \\ \text { (wt ppm) } \end{gathered}$ | $\begin{gathered} \mathrm{HCO}_{3}^{-} \\ \text {(wt ppm) } \end{gathered}$ | Total Cations | Total <br> Anions | Ratio Cat/Ani | $\begin{gathered} \mathbf{A L K} \\ \ldots \mathbf{p H} \\ \hline \end{gathered}$ | $\mathbf{S}^{\mathbf{2 -}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-3-135 | 135 | 8.41 | 5630 | 380 | 1330 | 17.4 | 17.2 | 25.6 | 860 | 325 | 0.0 | 1560 | 61.0 | 61.3 | 0.99 | 8.17 | 186.00 |
| DL-3-136 | 136 | 8.57 | 5440 | 380 | 1320 | 17.2 | 16.7 | 21.5 | 850 | 183 | 0.0 | 1560 | 60.3 | 58.1 | 1.04 | 7.82 | 269.00 |
| DL-3-137 | 137 | 8.50 | 5410 | 380 | 1320 | 17.5 | 16.8 | 21.5 | 840 | 194 | 0.0 | 1550 | 60.3 | 57.9 | 1.04 | 8.08 | 268.00 |
| DL-3-138 | 138 | 8.59 | 5400 | 380 | 1290 | 16.0 | 17.7 | 23.8 | 850 | 182 | 0.0 | 1550 | 59.2 | 57.9 | 1.02 | 7.76 | 210.00 |
| DL-3-139 | 139 | 8.12 | 5380 | 380 | 1320 | 16.1 | 17.5 | 23.3 | 850 | 496 | 0.0 | 1430 | 60.4 | 62.5 | 0.97 | 7.93 | 168.00 |
| DL-3-140 | 140 | 8.12 | 5410 | 390 | 1310 | 16.0 | 17.3 | 23.0 | 850 | 462 | 0.0 | 1430 | 60.0 | 61.9 | 0.97 | 7.98 | 170.00 |

A8 Results of chemical analysis in DL-4 system (sample type: RelW)

| Sample ID | Incub. <br> Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{\text {2- }}$ | $\begin{gathered} \mathrm{CO}_{3}{ }^{2} \\ \text { (wt ppm) } \\ \hline \end{gathered}$ | $\mathrm{HCO}_{3}{ }^{-}$ <br> (wt ppm) | Total Cations | Total Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \\ \hline \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \text { pH } \end{gathered}$ | $\mathrm{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-4-3 | 3 | 7.95 | 5480 | 9 | 1100 | 16.4 | 14.5 | 21.7 | 840 | 32.5 | 0.0 | 1470 | 50.5 | 48.5 | 1.04 | 7.65 | 13.2 |
| DL-4-4 | 4 | 7.83 | 5340 | 93 | 1070 | 15.9 | 14.0 | 21.4 | 810 | 139.0 | 0.0 | 1300 | 49.2 | 48.2 | 1.02 | 7.52 | 51.7 |
| DL-4-5 | 5 | 7.81 | 5320 | 94 | 1070 | 16.0 | 13.9 | 21.3 | 810 | 140.0 | 0.0 | 1280 | 49.2 | 47.9 | 1.03 | 7.50 | 52.1 |
| DL-4-6 | 6 | 7.93 | 5400 | 90 | 1090 | 16.3 | 14.1 | 21.6 | 820 | 138.0 | 0.0 | 1300 | 50.1 | 48.4 | 1.03 | 7.60 | 49.5 |
| DL-4-7 | 7 | 7.93 | 5320 | 87 | 1080 | 16.0 | 14.1 | 21.5 | 820 | 135.0 | 0.0 | 1280 | 49.6 | 48.0 | 1.03 | 7.53 | 49.6 |
| DL-4-8 | 8 | 7.91 | 4980 | 86 | 1080 | 15.9 | 14.0 | 21.2 | 810 | 129.0 | 0.0 | 1300 | 49.6 | 47.9 | 1.04 | 7.65 | 48.0 |
| DL-4-9 | 9 | 7.89 | 4980 | 86 | 1080 | 16.0 | 14.0 | 21.2 | 820 | 130.0 | 0.0 | 1300 | 49.6 | 48.2 | 1.03 | 7.64 | 48.4 |
| DL-4-10 | 10 | 7.94 | 4970 | 82 | 1090 | 16.1 | 14.0 | 21.1 | 830 | 125.0 | 0.0 | 1310 | 50.0 | 48.5 | 1.03 | 7.65 | 45.0 |
| DL-4-11 | 11 | 7.94 | 5000 | 76 | 1080 | 16.1 | 13.9 | 21.0 | 830 | 106.0 | 0.0 | 1340 | 49.6 | 48.5 | 1.02 | 7.68 | 39.0 |
| DL-4-12 | 12 | 7.94 | 4990 | 77 | 1070 | 16.0 | 13.9 | 21.0 | 830 | 105.0 | 0.0 | 1340 | 49.1 | 48.5 | 1.01 | 7.70 | 39.1 |
| DL-4-13 | 13 | 7.95 | 5020 | 76 | 1100 | 16.1 | 13.9 | 21.0 | 830 | 105.0 | 0.0 | 1350 | 50.4 | 48.6 | 1.04 | 7.71 | 39.3 |
| DL-4-14 | 14 | 7.84 | 4990 | 67 | 1110 | 16.0 | 13.9 | 21.0 | 830 | 69.4 | 0.0 | 1460 | 50.9 | 49.6 | 1.03 | 7.66 | 26.9 |
| DL-4-15 | 15 | 7.84 | 5000 | 68 | 1100 | 15.9 | 13.8 | 20.9 | 830 | 69.7 | 0.0 | 1410 | 50.4 | 48.8 | 1.03 | 7.69 | 26.4 |
| DL-4-16 | 16 | 8.12 | 4510 | 62 | 1110 | 16.6 | 14.2 | 21.5 | 800 | 58.0 | 0.0 | 1480 | 50.9 | 48.8 | 1.04 | 7.73 | 24.5 |
| DL-4-17 | 17 | 8.12 | 4510 | 62 | 1110 | 16.3 | 14.1 | 21.5 | 800 | 57.1 | 0.0 | 1510 | 50.9 | 49.3 | 1.03 | 7.72 | 24.4 |
| DL-4-18 | 18 | 8.12 | 4540 | 61 | 1120 | 16.7 | 14.1 | 21.4 | 800 | 47.8 | 0.0 | 1530 | 51.4 | 49.4 | 1.04 | 7.67 | 20.9 |
| DL-4-19 | 19 | 8.15 | 4380 | 61 | 1110 | 16.5 | 14.8 | 22.9 | 800 | 47.5 | 0.0 | 1540 | 51.1 | 49.5 | 1.03 | 7.84 | 21.0 |
| DL-4-20 | 20 | 8.16 | 4520 | 61 | 1100 | 16.0 | 14.3 | 22.0 | 800 | 47.6 | 0.0 | 1550 | 50.5 | 49.7 | 1.02 | 7.71 | 20.4 |
| DL-4-21 | 21 | 8.09 | 4470 | 60 | 1110 | 16.2 | 14.3 | 21.9 | 810 | 43.1 | 0.0 | 1510 | 51.0 | 49.2 | 1.04 | 7.94 | 18.4 |
| DL-4-22 | 22 | 8.08 | 4720 | 61 | 1070 | 15.7 | 13.5 | 20.3 | 800 | 41.8 | 0.0 | 1450 | 49.1 | 47.9 | 1.02 | 7.90 | 17.8 |
| DL-4-23 | 23 | 8.14 | 4720 | 60 | 1040 | 14.7 | 13.2 | 20.1 | 810 | 38.0 | 0.0 | 1460 | 47.7 | 48.3 | 0.99 | 8.02 | 15.7 |
| DL-4-24 | 24 | 8.14 | 4700 | 60 | 1060 | 15.5 | 13.6 | 20.7 | 800 | 38.3 | 0.0 | 1450 | 48.7 | 47.9 | 1.02 | 7.82 | 16.6 |
| DL-4-25 | 25 | 7.99 | 4690 | 68 | 1040 | 14.9 | 13.4 | 20.4 | 860 | 22.2 | 0.0 | 1460 | 47.7 | 49.5 | 0.96 | 7.88 | 11.1 |
| DL-4-26 | 26 | 7.98 | 4680 | 69 | 1020 | 14.4 | 12.6 | 19.1 | 800 | 22.5 | 0.0 | 1460 | 46.7 | 47.8 | 0.98 | 7.80 | 10.6 |
| DL-4-27 | 27 | 8.00 | 4690 | 70 | 1030 | 14.6 | 12.9 | 19.5 | 820 | 22.5 | 0.0 | 1510 | 47.2 | 49.2 | 0.96 | 7.81 | 10.7 |
| DL-4-28 | 28 | 8.00 | 4730 | 70 | 1040 | 14.8 | 12.9 | 19.4 | 830 | 23.0 | 0.0 | 1460 | 47.6 | 48.7 | 0.98 | 7.88 | 10.8 |

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| $7^{\prime} 6$ | 50＇8 | 960 | ع＇6t | $\varepsilon \cdot L \square$ | ${ }^{0} 5 ¢ 1$ | 00 | 861 | $0<8$ | $\varepsilon 81$ | 6．11 | 8 8 | Scor | 081 | 08¢t | $00 \cdot 8$ | 08 | 08－t－7a |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 96 | 818 | 960 | S6t | S゙Lt | 06E1 | 0.0 | I＇6I | 098 | 981 | I＇zı | 6 ¢ 1 | 0t0I | $0<1$ | 09¢t | $L 6 . L$ | 6 L | 6L－b－7a |
| 06 | 818 | 10.1 | $\varepsilon \% t$ | L－8t | 0¢¢ı | 00 | $8 \cdot 91$ | 078 | 561 | s＇zı | İti | s901 | $0<1$ | 0¢tt | $98^{\circ} \mathrm{L}$ | $8 L$ | 8L－b－Ta |
| 16 | Iz\％ | $00^{\prime}$ | $18 t$ | 0.85 | 09¢ | 0.0 | ع91 | 0¢8 | 961 | 9 zl | $00^{\circ}$ | Osot | 0＜1 | 0ISt | $58 . \mathrm{L}$ | LL | LL－b－7a |
| 06 | $\mathrm{Lt}^{\text {¢ }} 8$ | 10.1 | $0.8 t$ | ¢ 8 t | $0<\mathrm{II}$ | I＇88 | 061 | $0{ }^{0} 8$ | L＇61 | 9 zl | $0{ }^{\text {b }} \mathrm{l}$ | ssor | 0LI | 0¢9t | E8\％ | 92 | 9L－- －7d |
| t8 | ¢t．8 | 660 | 1＇6t | 58 t | $00 ¢ 1$ | $0 \cdot 0 \varepsilon$ | $\varepsilon z z$ | 098 | 0.02 | 8＇z1 | でもl | 0901 | 0LI | tet | $9 L^{\circ} \mathrm{L}$ | SL | $\varsigma L-\downarrow \mathrm{Td}$ |
| $\mathrm{t}^{\text {8 }}$ | 2t－8 | $00^{\circ} \mathrm{I}$ | \％$\%$ | \％$\%$ | $00 \varepsilon 1$ | $6 \cdot \varepsilon \varepsilon$ | ¢91 | 0¢8 | ¢61 | szı | 0ヶt | ssot | 091 | 019 ${ }^{\text {d }}$ | $18 . \mathrm{L}$ | tL | カL－t－Ta |
| ¢8 | 0s•8 | 20.1 | tit | ${ }^{\circ} \mathrm{s} 8$ | 0¢ZI | 0 ＇st | L＇ti | 078 | 2＇61 | ャ＇zı | 0 0t | 0901 | 091 | 079 | $88 . \mathrm{L}$ | $\varepsilon\llcorner$ | $\varepsilon L-\downarrow-7 \mathrm{~T}$ |
| $8{ }^{8} 5$ | $9{ }^{\text {2 }} 8$ | 960 | LOS | 685 | 0¢ะI |  | 98 | 016 | 961 | 9 zl | ガか | 0LOI | OSı | OSL ${ }^{\text {d }}$ | 28.2 | UL | zL－b－Ta |
| 6 S | Is＇8 | 860 | E0s | 16t | 01 ¢ı | 0 ＇st | $0{ }^{\prime \prime}$ | 006 | t＇6I | ¢ $\mathrm{c}^{\text {I }}$ | でャI | ¢LOI | 0sı | 09Lt | $98 . \mathrm{L}$ | IL | LL－V－7a |
| L＇9 | Lt．8 | $\angle 60$ | LOS | E＇6t | 0 0¢ะ | $8^{\prime}$ LE | $s^{\prime \prime} \downarrow$ | 016 | 161 | $\varepsilon \chi 1$ | 0 －t | 0801 | OSI | 06Lt | 508 | $0 L$ | 0L－t－7a |
| 89 | $6 c^{8}$ | 860 | 9.05 | S＇6t | $0 ¢ \varepsilon$ I | て＇ıE | $0 \cdot 01$ | 016 | ع＇61 | †てı |  | 5801 | 0si | 018t | ＋0．8 | 69 | 69－ヶ7d |
| 69 | $00^{8}$ | $\angle 60$ | 0.15 | \＆＇6t | 0¢¢I | 1＇9\％ | 0 ZI | 076 | 561 | 9 za | 「けI | 0801 | OSI | 08St | 56 L | 89 | 89－t－7a |
| 69 | ¢ $¢ 8$ | $\varepsilon^{6} 0$ | $0^{\circ} \mathrm{IS}$ |  | 08 E I | til | $\angle 01$ | 076 | 981 | 0 zl | $9 \cdot \varepsilon I$ | 0t01 | OSI | OSL ${ }_{\text {d }}$ | 00＇8 | $\llcorner 9$ | L9－ヶ7 |
| 69 | เ¢ 8 | \＄60 | ${ }^{\circ} \mathrm{IS}$ | S＇8t | 01 l | 00 | 601 | 076 | ¢61 | $\downarrow$ ¢ | sti | 0901 | 0tI | 008t | 88 L | 99 | 99－b－7a |
| 69 | $66^{8} 8$ | 560 | LOS | $08 t$ | оєє | 908 | $8 \cdot 8$ | 026 | 681 |  | L＇$\varepsilon 1$ | OSOI | 0tI | 06Lt | \＄6． | ¢9 | ¢9－b－7a |
| L＇9 | ¢s．8 | $10^{\circ} \mathrm{I}$ | 160 | 665 | 06ZI | どで | ¢8 | 088 | 102 | $0 \varepsilon 1$ | ¢＇ti | 0601 | 0¢I | 076t | 66 L | เ9 | カ9－b－7a |
| 8.9 | tif | $0^{\circ} \mathrm{I}$ | $\varepsilon \% 6$ | ti6t | 0z¢ | 698 | S9 | S 28 | 661 | $0 \varepsilon 1$ | $s{ }^{\text {cti }}$ | 0801 | 0¢I | 006t | 06 L | £9 | ¢9－t－Ta |
| 8.9 | LE8 | 860 | L＇6t | $68 t$ | $06 \varepsilon 1$ | 081 | ¢01 | 028 | L．61 | 8 zl | $\varepsilon \underbrace{\circ} \downarrow$ | 0LOI | 0¢I | 066t | 16 L | 29 | 29－t－7a |
| ¢99 | ガ8 | 10.1 | $56 t$ | 667 | 09 EL | ¢＇t¢ | $\tau$ | 598 | $9.0 z$ | ャ¢ı | 8 tI | 0601 | 0zI | 0¢6t | 56.2 | 19 | 19－b－7a |
| ¢＇9 | $66^{8}$ | 860 | $\varepsilon \cdot 0 \varsigma$ | S 66 | 06 E | て＇š | 0 ¢ | 068 | $\dagger 0 z$ | $\tau \cdot \varepsilon 1$ | $9{ }^{\text {9 }} \mathrm{t}$ | 0801 | 0zI | 026t | S6L | 09 | 09－t－7a |
| 19 | 688 | $00^{\circ}$ | los | 660 | 08 El | 0 ロヶて | $0 \cdot \mathrm{~s}$ | 068 | z＇0z | 1¢ | ¢＇tI | 0601 | 0zI | 0¢6t | 68 L | 65 | 6s－t－7a |
| $\varepsilon \cdot 9$ |  | $10 \cdot 1$ | 66 | £ $0 ¢$ | 09 E I | ＋＇62 | 8.01 | 588 | soz | $\varepsilon \varepsilon 1$ | 9tı | 001 I | 0z1 | 088t | 06 L | 92 | 8s－b－7a |
| 8．5 | LS8 | $00^{\circ}$ | $0 \cdot 0 ¢$ | $66 t$ | 06ZI | L＇$¢$ | $8 \cdot 8$ | 006 | s．0z | て＇\＆ı | 6 tl | 0601 | 021 | 069t | 108 | Ls | Ls－t－7a |
| $\llcorner\cdot 9$ | 198 | 10.1 | 8.20 | ع \％$\quad$ | 06II | してદ | ${ }^{+} \mathrm{I}$ | 016 | 9 Cl | がい | 0 －${ }^{\text {L }}$ | 0901 | 011 | 01Lt | 21.8 | 95 | 9s－b－7a |
| 1.4 | LS8 | $\angle 60$ | $0 \cdot 15$ | t＇6t | 0¢£ | $0 \cdot \mathrm{IS}$ | がII | 076 | zoz | I＇$\varepsilon 1$ | 6tı | 0801 | 011 | 002t | ${ }^{\text {t1 }} 8$ | ss | ss－t－7a |
| $-_{2} \mathrm{~S}$ | $\begin{aligned} & \mathrm{H}^{\mathrm{d}} \\ & \text { HTV }^{2} \end{aligned}$ | $\begin{aligned} & \text { !UVABT } \\ & \text { opey } \end{aligned}$ | suoluy <br> ${ }^{[170} \mathbf{L}$ | $\begin{array}{\|c} \hline \text { suopb } \\ \text { [E10 } \end{array}$ | $\begin{array}{\|c\|c\|} \hline(\text { udd } \mathfrak{M}) \\ -{ }^{-} \mathbf{O O H} \\ \hline \end{array}$ | $\left\lvert\, \begin{gathered} \left(\left.\begin{array}{c} \text { (udd } \mathfrak{z M}) \\ -\mathbf{O} \end{array} \right\rvert\,\right. \end{gathered}\right.$ | ${ }_{-}^{5} \mathrm{~F}$ OS | ． 10 | ${ }_{+8} \mathrm{~B}^{\mathrm{B}} \mathrm{O}$ | $+2^{8} \mathrm{~W}$ | ${ }_{+}+$ | ${ }^{\mathbf{8}} \mathbf{N}$ | ＋${ }^{\text {dg }}$ | puo， | $\mathrm{H}^{\text {d }}$ |  | OII Pdurs |


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| 0＇si | 0ع＊8 | 20.1 | $0.8 t$ | 88 t | 06ZI | 00 | 80¢ | $0 ¢ 8$ | 181 | 8＇II | $\tau ¢ \stackrel{ }{ }$ | 0L0I | 0\＆z | 0Stt | ${ }^{2} 8^{\circ} \mathrm{L}$ | 901 | $90 \mathrm{I}-\mathrm{F}-7 \mathrm{C}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 0＇tI | 188 | 20 I | I＇8t | $16 t$ | 00¢ | 00 | く＇İ | $00^{08}$ | 0LI | でı | 0 ¢t | 0801 | $0 z z$ | 0Stt | $28 . \mathrm{L}$ | s01 | ¢01－b－7a |
| 0ヤ1 | $67^{8}$ | 10.1 | L＇Lt | で8t | 06ZI | 0.0 | くıを | 088 | £ L | £ı | 8 ¢ | 0901 | $0 z z$ | 0Stt | 18．${ }^{\circ}$ | tol | ャ0I－ャ－7a |
| 6¢1 | Lて8 | 00.1 | L＇L | 8 L | 08zI | 0.0 | $6 \angle L$ | 0¢8 | 0LI | でı | $\varsigma ¢ 1$ | OSOI | 0zz | 00tt | $58 . \mathrm{L}$ | E0I | ع0I－ャ－7a |
| $\downarrow$ ¢¢ | sz\％ | 860 | $1.8 t$ | $0 \cdot 2$ | 0IEI | 00 | I＇sz | 0¢8 | 6 LI | L＇II | 9＊t | 0EOI | $0 z z$ | 0ztt | $68^{\circ} \mathrm{L}$ | $z 01$ | 201－b－7a |
| $\varsigma \mathfrak{c l}$ | $0{ }^{0} 8$ | $00 \cdot 1$ | $\dagger 8 t$ | $\dagger 8 t$ | $00 \varepsilon 1$ | 00 | $6 \angle Z$ | $0{ }^{0} 8$ | 981 | İI | 6tı | 0901 | 0\＆z | OStt | 76.4 | 101 | 101－＊－7a |
| 9 9＇II | 978 | 660 | z＇8t | S＇Lt | $00 \varepsilon 1$ | 00 | ¢¢z | $00^{0} 8$ | L＇81 | İI | ぢゅ | 0t01 | $0 z z$ | OStt | 16.4 | 001 | 001－t－7a |
| silit | 188 | 660 | ¢ $8 \checkmark$ | 6.2 | $01 \varepsilon \frac{1}{}$ | 00 | L＇zz | $0 \vdash 8$ | 9 LI | SII | $て ゙ ゅ 1$ | 0s0I | $0 z \tau$ | 08tt | 50.8 | 66 | $66-7 \mathrm{Ta}$ |
| 6 bl | 18.8 | 660 | z＇8t | 6.2 | 00¢ 1 | 00 | 862 | 078 | 081 | 8II | $\mathfrak{s t}$ | OSOI | 0Iz | 06tr | 208 | 86 | $86-7$－7 |
| 0 SI | $1 \varepsilon 8$ | $00 \cdot 1$ | L＇8t | $\stackrel{\circ}{\circ} \mathrm{t}$ | 0 0ıย | 00 | 662 | 058 | L＇LI | 9 ll | †＇tI | 0LOI | OIz | 00tt | 208 | 16 | L6－t－7a |
| L－sI | £ \％ 8 | $\angle 60$ | L＇8t | $\dagger$ ¢ | $01 \varepsilon 1$ | 00 | 6 6を | 058 | 8．LI | L＇ı | $\varepsilon \downarrow \square$ | 0t0I | OIz | OStt | $20 \cdot 8$ | 96 | 96－t－Ta |
| 891 | £ \％ 8 | $00^{\circ} \mathrm{I}$ | $\varepsilon: 8 t$ | ¢ \％$\quad$ t | 00¢ 1 | 00 | 8 8¢ | 0ヶ8 | ELI | かい | ガロI | 0901 | 01z | OStt | 86.4 | S6 | ¢6－t－TU |
| ［＇sI | で8 | 20.1 | ¢ $\angle t$ | ¢ $¢$ | $00 \varepsilon 1$ | 0.0 | 682 | 028 | 981 | İて | 6bl | 0901 | 002 | 0ttt | E0＇8 | ${ }^{+6}$ |  |
| ぐゅI | $00^{8}$ | ＋0． 1 | $\varepsilon \angle L$ | で6t | $00 \varepsilon 1$ | 00 | $00 \varepsilon$ | 018 | $\varepsilon \cdot 81$ | 6It | 9＇tl | 0801 | 002 | 06Et | 90.8 | £6 | £6－t－7a |
| ぐゅ | ャで8 | to 1 |  | L＇6t | 0 0ıE | 00 | 908 | 028 | ¢ 81 | 0 OI | ISI | 0601 | 012 | 0ztt | 10.8 | 26 | 26－b－7a |
| 9＊tI | ャで8 | $\mathrm{EO}^{\circ} \mathrm{I}$ | 085 | \＆\％ 6 | $00 \varepsilon 1$ | 0.0 | I－1¢ | $0 ¢ 8$ | $5 \cdot 81$ | 0 OL | 8 tl | 0801 | 012 | $00^{0+t b}$ | 10.8 | 16 | 16－b－7a |
| ¢゙っI | 91.8 | ＋0． 1 | SLt | E\％ 6 | 06ZI | 0.0 | ¢6\％ | 078 | 981 | 0て1 | $8{ }^{8} \mathrm{~b}$ | 0801 | 012 | 08Et | S0．8 | 06 | 06－t－7a |
| 9 9ı | 818 | $\mathrm{E}_{0} \mathrm{I}$ | LLL | \＆\％ 6 | $01 \varepsilon 1$ | 0.0 | $\varepsilon 87$ | 078 | L＇81 | 1 İ | 9＇t | 0801 | 002 | 09tt | $28 . \mathrm{L}$ | 68 | 68－t－7a |
| 9＇£ | 618 | E0． 1 | $0.8 t$ | $\varepsilon 6 t$ | $01 \varepsilon!$ | 0.0 | 562 | 088 | 9：81 | İı | Lbl | 0801 | 002 | OStt | 18 L | 88 | $88-\downarrow-7 \mathrm{a}$ |
| 1\％ | 02＇8 | E0＇ | 08 t | $\varepsilon 6 t$ | ${ }_{0} \downarrow \varepsilon$ I | $0 \cdot 0$ | $9 \downarrow 1$ | 088 | 061 | どて1 | tol | 0801 | 061 | 08tt | $88^{\circ} \mathrm{L}$ | 48 | L8－b－7a |
| $1 \%$ | £̇8 | 660 | 2＇8t | $0.8 t$ | 0¢EI | 0.0 | $\varepsilon \angle L$ | 0¢8 | 061 | でてI | ちゅ！ | 0s0I | 061 | 09tt | 86.4 | 98 | 98－b－7a |
| S．L | £18 | $\angle 60$ | 685 | $\downarrow$ ¢ | OSEI | 00 | †てI | 098 | 8．LI | 9 ll | $9 \mathrm{\varepsilon}$ L | 0t0I | 081 | 0LEt | $\angle 6.4$ | 58 | 58－b－7a |
| $9{ }^{-2}$ | 008 | 960 | $66 t$ | $6 \angle t$ | 0itl | 00 | ¢csı | 098 | ［＇81 | 8.1 | L $\mathcal{L}$ | 0s0I | 081 | 0LEt | 56.4 | เ8 | ＋8－t－7a |
| 0.6 | $66 \%$ | $\angle 60$ | $1{ }^{1} 6$ | SLt | 09EI | 00 | 1く1 | 098 | $\dagger 81$ | 6 LI | 6 6I | 0t01 | 081 | 06Et | 66 L | ¢8 | ¢8－b－7a |
| $0 \%$ | 918 | $\angle 60$ | E．6t | $0 \cdot 8$ | 0s¢ı | 00 | £81 | $0<8$ | 0.61 | £ てI | 0ヶt | 0soi | 081 | 06Et | 90.8 | 28 | 28－t－7a |
| £6 | 408 | 860 | ¢6t | 585 | 09¢I | $0 \cdot 0$ | $\varepsilon 61$ | $0 \angle 8$ | 5\％61 |  | $て ゙ \downarrow$ | 0901 | 081 | 06Et | 508 | 18 | 18－t－7a |
| ${ }_{2} \mathrm{~S}$ | $\begin{aligned} & \mathrm{H}^{\mathrm{d}} \\ & \mathrm{yTV} \end{aligned}$ |  | $\begin{gathered} \text { suolug } \\ \text { IBłol } \end{gathered}$ |  | $\begin{array}{\|c\|} \hline(\text { mad } 7 \mathrm{M}) \\ -_{-} \mathbf{O O H} \\ \hline \end{array}$ |  | $\tau^{5} \mathrm{OS}$ | IV | $+2^{8} 3$ | ${ }_{+2^{8} \mathrm{~N}}$ | ＋${ }^{\text {I }}$ | ${ }^{\text {B }}$ N | ${ }^{18}$ | puo？ | $\mathrm{H}^{\text {d }}$ |  | CI गldues |


A8 Results of chemical analysis in DL-4 system (sample type: RelW) (cont'd)

| Sample 1D | Incub. <br> Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathbf{N a}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{2}$ | $\begin{gathered} \mathrm{CO}_{3}{ }^{2-} \\ \text { (wt ppm) } \end{gathered}$ | $\begin{array}{\|c} \mathbf{H C O}_{3}^{-} \\ \text {(wt ppm) } \end{array}$ | Total <br> Cations | Total <br> Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \\ \hline \end{gathered}$ | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \\ \hline \end{gathered}$ | $\mathrm{S}^{\mathbf{2}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-4-107 | 107 | 7.89 | 4480 | 230 | 1090 | 14.9 | 12.2 | 18.6 | 830 | 29.5 | 0.0 | 1290 | 49.7 | 48.0 | 1.04 | 8.31 | 15.1 |
| DL-4-108 | 108 | 8.12 | 4570 | 230 | 1060 | 15.2 | 11.8 | 18.2 | 850 | 34.9 | 0.0 | 1290 | 48.4 | 48.7 | 0.99 | 8.27 | 17.6 |
| DL-4-109 | 109 | 8.13 | 4560 | 230 | 1070 | 15.2 | 11.8 | 18.3 | 840 | 31.9 | 0.0 | 1300 | 48.8 | 48.5 | 1.01 | 8.28 | 15.2 |
| DL-4-110 | 110 | 7.67 | 4550 | 290 | 1070 | 13.7 | 11.7 | 18.2 | 840 | 137.0 | 0.0 | 1120 | 48.8 | 48.5 | 1.01 | 8.08 | 50.4 |
| DL-4-111 | 111 | 7.71 | 4530 | 290 | 1060 | 13.4 | 11.6 | 18.1 | 830 | 134.0 | 0.0 | 1120 | 48.3 | 48.2 | 1.00 | 8.12 | 49.7 |
| DL-4-112 | 112 | 7.67 | 4540 | 280 | 1080 | 13.7 | 11.6 | 18.0 | 830 | 134.0 | 0.0 | 1090 | 49.2 | 47.5 | 1.03 | 8.14 | 50.4 |
| DL-4-113 | 113 | 7.65 | 4600 | 290 | 1090 | 13.8 | 11.9 | 18.6 | 830 | 138.0 | 0.0 | 1140 | 49.7 | 48.6 | 1.02 | 8.12 | 50.8 |
| DL-4-114 | 114 | 7.83 | 4530 | 290 | 1070 | 13.7 | 11.8 | 18.4 | 830 | 123.0 | 0.0 | 1170 | 48.8 | 48.7 | 1.00 | 8.14 | 46.4 |
| DL-4-115 | 115 | 7.82 | 4440 | 290 | 1100 | 13.8 | 11.8 | 18.5 | 830 | 126.0 | 0.0 | 1140 | 50.1 | 48.3 | 1.04 | 8.16 | 46.8 |
| DL-4-116 | 116 | 7.83 | 4510 | 370 | 1100 | 14.1 | 11.7 | 18.2 | 890 | 177.0 | 0.0 | 1130 | 50.1 | 51.9 | 0.96 | 7.99 | 49.5 |
| DL-4-117 | 117 | 7.80 | 4470 | 370 | 1100 | 13.7 | 11.5 | 17.8 | 890 | 175.0 | 0.0 | 1130 | 50.0 | 51.9 | 0.96 | 8.05 | 48.0 |
| DL-4-118 | 118 | 7.84 | 4440 | 370 | 1110 | 13.7 | 11.9 | 18.8 | 890 | 196.0 | 0.0 | 1100 | 50.5 | 51.8 | 0.98 | 8.01 | 53.8 |
| DL-4-119 | 119 | 7.81 | 4360 | 370 | 1100 | 14.0 | 11.9 | 18.8 | 890 | 195.0 | 0.0 | 1100 | 50.1 | 51.8 | 0.97 | 8.12 | 54.5 |
| DL-4-120 | 120 | 7.78 | 4450 | 370 | 1110 | 13.6 | 11.7 | 18.4 | 900 | 194.0 | 0.0 | 1100 | 50.5 | 52.1 | 0.97 | 8.03 | 53.9 |
| DL-4-121 | 121 | 7.80 | 4480 | 370 | 1120 | 14.0 | 11.8 | 18.4 | 890 | 192.0 | 0.0 | 1100 | 51.0 | 51.7 | 0.99 | 7.99 | 54.9 |
| DL-4-122 | 122 | 7.91 | 4470 | 370 | 1110 | 14.4 | 12.0 | 18.7 | 890 | 194.0 | 0.0 | 1120 | 50.6 | 52.1 | 0.97 | 7.99 | 56.0 |
| DL-4-123 | 123 | 7.88 | 4510 | 370 | 1100 | 14.1 | 11.8 | 18.4 | 900 | 193.0 | 0.0 | 1110 | 50.1 | 52.2 | 0.96 | 8.07 | 55.6 |
| DL-4-124 | 124 | 7.87 | 4490 | 380 | 1110 | 14.2 | 11.9 | 18.5 | 900 | 197.0 | 0.0 | 1110 | 50.5 | 52.4 | 0.96 | 8.00 | 55.5 |
| DL-4-125 | 125 | 7.95 | 4430 | 380 | 1100 | 14.1 | 11.4 | 17.7 | 890 | 132.0 | 0.0 | 1140 | 50.0 | 51.3 | 0.98 | 7.85 | 36.5 |
| DL-4-126 | 126 | 7.91 | 4400 | 380 | 1110 | 14.3 | 11.3 | 17.4 | 890 | 133.0 | 0.0 | 1140 | 50.4 | 51.3 | 0.98 | 7.92 | 37.2 |
| DL-4-127 | 127 | 7.85 | 4680 | 380 | 1130 | 13.0 | 11.4 | 18.1 | 890 | 160.0 | 0.0 | 1120 | 51.3 | 51.5 | 1.00 | 7.95 | 44.1 |
| DL-4-128 | 128 | 7.83 | 4690 | 390 | 1120 | 12.9 | 11.4 | 18.1 | 890 | 162.0 | 0.0 | 1120 | 50.9 | 51.7 | 0.98 | 7.94 | 43.1 |
| DL-4-129 | 129 | 7.83 | 4700 | 380 | 1135 | 13.3 | 11.5 | 18.2 | 890 | 182.0 | 0.0 | 1100 | 51.6 | 51.6 | 1.00 | 7.95 | 49.2 |
| DL-4-130 | 130 | 7.81 | 4750 | 390 | 1129 | 13.4 | 11.4 | 18.1 | 900 | 179.0 | 0.0 | 1100 | 51.3 | 52.0 | 0.99 | 7.89 | 49.2 |
| DL-4-131 | 131 | 7.77 | 4630 | 390 | 1150 | 13.5 | 11.5 | 18.3 | 900 | 181.0 | 0.0 | 1100 | 52.2 | 52.0 | 1.00 | 7.90 | 49.4 |
| DL-4-132 | 132 | 7.91 | 4640 | 390 | 1135 | 13.4 | 11.5 | 18.1 | 900 | 184.0 | 0.0 | 1090 | 51.6 | 51.9 | 0.99 | 8.20 | 50.5 |

A8 Results of chemical analysis in DL-4 system (sample type: RelW) (cont'd)

| Sample ID | Incub. <br> Time (d) | pH | Cond | $\mathbf{B r}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{\mathbf{2 -}}$ | $\begin{gathered} \mathbf{C O}_{3}^{2-} \\ \text { (wt ppm) } \end{gathered}$ | $\begin{gathered} \mathbf{H C O}_{3}^{-} \\ \text {(wt ppm) } \end{gathered}$ | Total Cations | Total <br> Anions | Ratio Cat/Ani | $\begin{gathered} \text { ALK } \\ \mathbf{p H} \\ \hline \end{gathered}$ | $\mathrm{S}^{\mathbf{2 -}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-4-133 | 133 | 7.91 | 4670 | 390 | 1120 | 12.9 | 11.3 | 17.9 | 900 | 186.0 | 0.0 | 1090 | 50.9 | 52.0 | 0.98 | 7.91 | 49.2 |
| DL-4-134 | 134 | 7.85 | 4620 | 390 | 1110 | 12.5 | 11.1 | 17.7 | 900 | 183.0 | 0.0 | 1100 | 50.4 | 52.1 | 0.97 | 8.12 | 47.6 |
| DL-4-135 | 135 | 7.76 | 4660 | 390 | 1130 | 12.8 | 11.3 | 17.9 | 890 | 183.0 | 0.0 | 1090 | 51.3 | 51.6 | 0.99 | 7.99 | 48.2 |
| DL-4-136 | 136 | 7.87 | 4410 | 360 | 1077 | 13.5 | 11.5 | 18.3 | 870 | 168.0 | 0.0 | 1100 | 49.0 | 50.5 | 0.97 | 7.97 | 48.6 |
| DL-4-137 | 137 | 7.89 | 4400 | 360 | 1081 | 13.1 | 11.3 | 17.8 | 890 | 173.0 | 0.0 | 1100 | 49.2 | 51.2 | 0.96 | 7.97 | 48.8 |
| DL-4-138 | 138 | 7.85 | 4410 | 360 | 1075 | 12.9 | 11.2 | 17.5 | 890 | 165.0 | 0.0 | 1110 | 48.9 | 51.2 | 0.95 | 7.97 | 48.5 |
| DL-4-139 | 139 | 7.93 | 4410 | 360 | 1090 | 12.8 | 10.9 | 16.9 | 880 | 149.0 | 0.0 | 1120 | 49.5 | 50.8 | 0.97 | 8.08 | 42.7 |
| DL-4-140 | 140 | 7.95 | 4390 | 360 | 1081 | 12.7 | 10.9 | 16.8 | 840 | 152.0 | 0.0 | 1120 | 49.1 | 49.7 | 0.99 | 7.98 | 42.4 |

## A9 Results of baseline chemical analysis for dynamic system

| Sampling Date | Sample ID | Sample Type | Incub. Time <br> (d) | pH | Cond | $\mathbf{B r}^{+}$ | $\mathbf{N a}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{2-}$ | $\left.\begin{array}{c} \mathrm{CO}_{3}{ }^{2-} \\ (\text { wt } \mathrm{ppm}) \end{array}\right)$ | $\left(\begin{array}{c} \mathrm{HCO}_{3}^{-} \\ (\text {wt ppm) } \end{array}\right.$ | Total Cations | Total Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | ALK pH | $\mathrm{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 17-Jun-04 | CT RW from Site | PW | 0 | 7.63 | 4500 | 0.0 | 1140 | 26.1 | 28.1 | 113 | 690 | 1060 | 0.0 | 727 | 58.23 | 53.44 | 1.09 | 7.45 | 351.0 |
| 26-Jul-04 | D2-BASE-1 | water | 0 | 7.69 | 5250 | 3.8 | 1070 | 24.9 | 106 | 157 | 700 | 1620 | 0.0 | 859 | 63.84 | 67.60 | 0.94 | 7.72 | 482.0 |
| 26-Jul-04 | D2-BASE-2 | water | 0 | 7.76 | 5770 | 1.4 | 1080 | 25.0 | 162 | 212 | 680 | 2160 | 0.0 | 785 | 71.70 | 77.04 | 0.93 | 7.58 | 709.0 |
| 28-Jul-04 | $\begin{gathered} \text { JPang-WIP- } \\ 0704 \end{gathered}$ | water | 0 | 7.92 | 2970 | 0.0 | 893 | 12.3 | 7.3 | 14.3 | 720 | 307 | 0.0 | 863 | 40.47 | 40.83 | 0.99 | 7.92 | 97.4 |
| 19-Aug-04 | CT <br> WATER <br> BASE-1 | water | 0 | 8.25 | 7360 | 2500 | 1730 | 27.0 | 26.4 | 31.1 | 750 | 1060 | 0.0 | 584 | 79.66 | 83.65 | 0.95 | 7.78 | 355.0 |
| 19-Aug-04 | CT WATER BASE-2-1 | water | 0 | 8.40 | 5260 | 18.0 | 1180 | 27.8 | 115 | 58 | 760 | 1660 | 0.0 | 436 | 64.50 | 63.36 | 1.02 | 7.84 | 546.0 |
| 19-Aug-04 | CT <br> WATER BASE-2-2 | water | 0 | 7.93 | 5930 | 0.0 | 1160 | 26.9 | 172 | 155 | 749 | 2229 | 0.0 | 540 | 73.21 | 76.39 | 0.96 | 7.48 | 753.0 |
| 03-Feb-05 | D3-BASE | water | 0 | 8.07 | 6430 | 320 | 1240 | 26.5 | 26.0 | 61.7 | 720 | 995 | 0.0 | 784 | 59.84 | 57.86 | 1.03 | 7.43 | 346.0 |
| 03-Feb-05 | D4-BASE | water | 0 | 8.07 | 5270 | 330 | 1090 | 15.0 | 8.1 | 13.7 | 760 | 321 | 0.0 | 843 | 49.14 | 46.04 | 1.07 | 7.47 | 114.0 |
| 23-Mar-05 | D3-Base-60 | water | 59 | 8.07 | 5700 | 340 | 1230 | 25.0 | 23.8 | 42.0 | 820 | 1035 | 0.0 | 842 | 58.20 | 62.71 | 0.93 | 7.74 | 332.0 |
| 31-Mar-05 | D4-Base-60 | water | 59 | 8.38 | 4890 | 300 | 1100 | 14.7 | 8.2 | 13.7 | 770 | 296 | 35.7 | 869 | 49.57 | 47.04 | 1.05 | 8.53 | 111.0 |

## A10 Results of baseline MFT chemical analysis

| Sampling Date | Sample ID | Sample <br> Type | Incub. Time <br> (d) | pH | Cond | $\mathbf{B r}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{2-}$ | $\underset{(\text { wt ppm })}{\mathrm{CO}_{3}{ }^{\mathbf{2 -}}}$ | $\begin{gathered} \mathrm{HCO}_{3}^{-} \\ (\text {wt ppm) } \end{gathered}$ | Total Cations | Total Anions | Ratio Cat/Ani | $\begin{gathered} \mathbf{A L K} \\ \mathbf{p H} \end{gathered}$ | $S^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 12-Jul-04 | MFT <br> Mixed | Slurry | 0 | 7.48 | 4110 | 0.0 | 1040 | 16.3 | 13.7 | 20.7 | 800 | 18 | 0 | 1610 | 47.81 | 49.29 | 0.97 | 7.95 | 6.27 |
| 19-Aug-04 | MFT BASE 2 | Slurry | 0 | 7.59 | 4110 | 0.0 | 1110 | 17.5 | 15.1 | 23.1 | 868 | 30.7 | 0 | 1560 | 51.12 | 50.66 | 1.01 | 7.90 | 9.71 |
| 19-Aug-04 | $\begin{gathered} \text { MFT } \\ \text { BASE } 4 \\ \hline \end{gathered}$ | Slurry | 0 | 7.67 | 4160 | 0.0 | 1090 | 17.5 | 14.8 | 22.8 | 868 | 17.7 | 0 | 1570 | 50.21 | 50.56 | 0.99 | 8.05 | 6.15 |

A11 Results of chemical analysis in pore water of D3 \& D4 systems

| Sample ID | Sample Type | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | Mg ${ }^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathbf{S O}_{4}{ }^{2-}$ | $\begin{array}{\|c\|} \mathbf{C O}_{(\mathrm{wt} \mathrm{pmm}}{ }^{2}{ }^{2} \\ \hline \end{array}$ | $\begin{array}{\|l\|l} \mathbf{H C O}_{3} \\ \text { (wt ppm) } \end{array}$ | $\begin{array}{\|c} \text { Total } \\ \text { Cations } \end{array}$ | $\begin{gathered} \text { Total } \\ \text { Anions } \\ \hline \end{gathered}$ | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \\ \hline \end{gathered}$ | Alkalinity expressed as $\mathrm{HCO}_{3}$ | $\begin{gathered} \mathbf{A L K} \\ \mathbf{p H} \end{gathered}$ | $\mathbf{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-3-R | RawW | 8.05 | 5600 | 420 | 1120 | 24.5 | 24.6 | 39.6 | 930 | 1220 | 0.0 | 752 | 53.4 | 69.2 | 0.77 | 616 | 8.0 | 329.0 |
| DL-4-R | RawW | 8.57 | 4440 | 390 | 1000 | 14.3 | 7.8 | 12.4 | 920 | 386 | 0.0 | 853 | 45.1 | 52.8 | 0.85 | 699 | 8.2 | 108.0 |
| DH-3-Top | Rel W | 7.91 | 4510 | 210 | 1200 | 17.9 | 22.0 | 41.4 | 740 | 590 | 0.0 | 1230 | 56.5 | 55.9 | 1.01 | 1008 | 7.6 | 205.0 |
| DH-3-U | Porew | 8.36 | 4300 | 220 | 1130 | 14.5 | 13.7 | 16.3 | 820 | 51.0 | 0.0 | 1480 | 51.5 | 51.2 | 1.01 | 1213 | 7.4 | 50.5 |
| DH-3-M-U | Porew | 8.62 | 4070 | 260 | 1050 | 14.7 | 10.9 | 11.4 | 760 | 58.1 | 0.0 | 1330 | 47.5 | 47.7 | 1.00 | 1090 | 7.6 | 68.3 |
| DH-3-M-L | PoreW | 8.63 | 3920 | 290 | 1050 | 15.3 | 10.9 | 11.7 | 720 | 80.3 | 0.0 | 1200 | 47.5 | 45.3 | 1.05 | 984 | 7.6 | 124.0 |
| DH-3-L | Porew | 8.73 | 4280 | 300 | 1050 | 16.8 | 13.5 | 16.9 | 750 | 205 | 0.0 | 1360 | 48.1 | 51.4 | 0.93 | 1115 | 7.5 | 230.0 |
| DL-3-Top | Rel W | 7.68 | 5680 | 380 | 1150 | 15.7 | 20.5 | 36.4 | 720 | 920 | 0.0 | 950 | 53.9 | 59.8 | 0.90 | 779 | 8.2 | 299.0 |
| DL-3-U | PoreW | 8.43 | 5570 | 500 | 1140 | 11.3 | 14.4 | 13.9 | 870 | 239 | 0.0 | 141 | 51.7 | 58.9 | 0.88 | 1156 | 8.1 | 201.0 |
| DL-3- M-U | Porew | 8.22 | 5530 | 500 | 1120 | 11.3 | 15.6 | 14.9 | 830 | 295 | 0.0 | 1330 | 51.0 | 57.6 | 0.89 | 1090 | 7.7 | 121.0 |
| DL-3- M-L | PoreW | 8.16 | 5260 | 480 | 1060 | 12.2 | 14.6 | 14.6 | 800 | 226 | 0.0 | 1250 | 48.3 | 53.7 | 0.90 | 1025 | 7.6 | 201.0 |
| DL-3-L | Porew | 8.19 | 5430 | 490 | 1110 | 14.2 | 16.6 | 18.2 | 810 | 443 | 0.0 | 1170 | 50.9 | 57.4 | 0.89 | 959 | 7.8 | 140.0 |
| DH-4-Top | Rel W | 8.04 | 3970 | 260 | 955 | 12.9 | 11.3 | 17.9 | 760 | 204 | 0.0 | 1040 | 43.7 | 46.0 | 0.95 | 852 | 7.6 | 69.7 |
| DH-4 | PoreW | 8.36 | 3880 | 290 | 950 | 13.3 | 8.3 | 9.7 | 810 | 13.5 | 0.0 | 1060 | 42.8 | 44.1 | 0.9 | 869 | 7.7 | 8.7 |
| DH-4-M-U | PoreW | 8.47 | 3790 | 300 | 963 | 12.2 | 7.4 | 8.9 | 800 | 13.7 | 0.0 | 998 | 43.2 | 42.9 | 1.01 | 818 | 7.6 | 9.7 |
| DH-4-M-L | Porew | 8.42 | 3850 | 300 | 990 | 13.2 | 7.4 | 9.2 | 780 | 11.5 | 0.0 | 1070 | 44.5 | 43.5 | 1.02 | 877 | 7.7 | 8.1 |
| DH-4-L | Porew | 8.51 | 3760 | 300 | 975 | 13.0 | 6.8 | 8.4 | 790 | 12.1 | 0.0 | 1000 | 43.7 | 42.6 | 1.02 | 820 | 7.7 | 8.1 |
| DL-4-Top | Rel W | 7.84 | 4560 | 390 | 950 | 11.8 | 10.2 | 15.5 | 830 | 71.4 | 0.0 | 170 | 43.2 | 48.9 | 0.88 | 959 | 8.3 | 31.1 |
| DL-4-U | Porew | 8.13 | 4620 | 430 | 922 | 11.7 | 10.2 | 12.9 | 810 | 9.0 | 0.0 | 1130 | 41.9 | 46.9 | 0.89 | 926 | 8.2 | 9.4 |
| DL-4-M-U | PoreW | 8.28 | 4610 | 460 | 906 | 10.5 | 8.8 | 11.3 | 810 | 13.9 | 0.0 | 1120 | 41.0 | 47.2 | 0.87 | 918 | 8.3 | 11.2 |
| DL-4-M-L | PoreW | 8.23 | 4480 | 430 | 900 | 11.7 | 9.0 | 11.3 | 800 | BDL | 8.4 | 1060 | 40.7 | \#VALUE! | \#Value! | 883 | 8.3 | 9.1 |
| DL-4-L | PoreW | 8.26 | 4550 | 460 | 920 | 11.6 | 8.4 | 10.8 | 810 | 14.2 | 0.0 | 1070 | 41.5 | 46.4 | 0.90 | 877 | 8.3 | 9.7 |

A12 Results of chemical analysis in Jars

| System | $\begin{array}{\|c} \text { Sample } \\ \text { ID } \end{array}$ | Sample Type | Incub. Time (d) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}^{\text {2+ }}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{\mathbf{2}}$ | $\begin{array}{\|c} \mathrm{CO}_{3}{ }^{2-} \\ \text { (wt ppm) } \end{array}$ | $\begin{array}{\|c} \mathbf{H C O}_{3}^{-} \\ \text {(wt ppm) } \end{array}$ | Total Cations | Total Anions | $\begin{array}{\|c\|} \hline \text { Ratio } \\ \text { Cat/Ani } \\ \hline \end{array}$ | Alkalinity expressed as $\mathrm{HCO}_{3}^{-}$ | $\begin{gathered} \text { ALK } \\ \mathrm{pH} \end{gathered}$ | $\mathrm{S}^{\mathbf{2 -}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Jar 1 | Jar D1-1 | PoreW | 128 | 8.37 | 8340 | 3000 | 2300 | 19.7 | 25.4 | 31.0 | 1100 | 703 | 0.0 | 1580 | 104.2 | 109.0 | 0.96 | 1295 | 7.5 | 285 |
| Jar 1 | Jar D1-2 | PoreW | 128 | 8.21 | 8200 | 3000 | 2270 | 18.9 | 24.5 | 32.1 | 1100 | 673 | 0.0 | 1550 | 102.8 | 107.9 | 0.95 | 1270 | 7.8 | 241 |
| Jar 2 | Jar D2-1 | PoreW | 128 | 8.19 | 6150 | 8.9 | 1530 | 20.0 | 42.5 | 41.4 | 1000 | 797 | 0.0 | 1750 | 72.6 | 73.6 | 0.99 | 1434 | 8.0 | 277 |
| Jar 2 | Jar D2-2 | PoreW | 128 | 7.92 | 6260 | 0.1 | 1590 | 20.3 | 47.2 | 45.8 | 1000 | 887 | 0.0 | 1640 | 75.9 | 73.5 | 1.03 | 1344 | 7.6 | 494 |
| Jar 3 | Jar D3-1 | PoreW | 128 | 8.03 | 7390 | 700 | 1900 | 20.1 | 29.8 | 37.2 | 1000 | 590 | 0.0 | 2090 | 87.5 | 83.5 | 1.05 | 1713 | 7.5 | 533 |
| Jar 3 | Jar D3-2 | PoreW | 128 | 7.90 | 6140 | 500 | 1560 | 17.9 | 22.5 | 32.9 | 1000 | 582 | 0.0 | 1750 | 71.8 | 75.2 | 0.95 | 1434 | 7.8 | 198 |
| Jar 4 | Jar D4-1 | PoreW | 128 | 8.14 | 5880 | 810 | 1500 | 17.0 | 19.0 | 28.0 | 1100 | 370 | 0.0 | 1590 | 68.6 | 74.9 | 0.92 | 1303 | 8.3 | 131 |
| Jar 4 | Jar D4-2 | PoreW | 128 | 7.83 | 5100 | 430 | 1250 | 15.4 | 14.6 | 21.5 | 1100 | 9.9 | 0.0 | 1090 | 57.0 | 54.4 | 1.05 | 893 | 7.8 | 7.1 |

## A13 MPN data of DH \& DL systems

| System | Sample ID | t (d) | Methanogen MPN/g DRY WT | SRB MPN/g DRY WT |
| :---: | :---: | :---: | :---: | :---: |
| DH-1 | DH-1-U | 60 | $4.52 \mathrm{E}+01$ | $8.94 \mathrm{E}+04$ |
|  | DH-1-M-U | 60 |  | $5.04 \mathrm{E}+05$ |
|  | DH-1-M-L | 60 |  | $9.41 \mathrm{E}+04$ |
|  | DH-1-L | 60 | $1.81 \mathrm{E}+01$ | $9.05 \mathrm{E}+04$ |
| DH-2 | DH-2-U | 60 | $4.59 \mathrm{E}+01$ | $4.26 \mathrm{E}+04$ |
|  | DH-2-M-U | 60 |  | $1.99 \mathrm{E}+05$ |
|  | DH-2-M-L | 60 |  | $9.41 \mathrm{E}+04$ |
|  | DH-2-L | 60 | $9.36 \mathrm{E}+01$ | $5.06 \mathrm{E}+05$ |
| DH-3 | DH-3-U | 60 | $5.11 \mathrm{E}+04$ | $9.30 \mathrm{E}+04$ |
|  | DH-3-M-U | 60 | $5.49 \mathrm{E}+04$ | $5.10 \mathrm{E}+04$ |
|  | DH-3-M-L | 60 | $4.74 \mathrm{E}+04$ | $2.00 \mathrm{E}+04$ |
|  | DH-3-L | 60 | $5.04 \mathrm{E}+04$ | $4.90 \mathrm{E}+04$ |
| DH-4 | DH-4-U | 60 | $5.46 \mathrm{E}+04$ | $2.09 \mathrm{E}+05$ |
|  | DH-4-M-U | 60 | $2.10 \mathrm{E}+05$ | $9.55 \mathrm{E}+04$ |
|  | DH-4-M-L | 60 | $4.34 \mathrm{E}+04$ | $5.10 \mathrm{E}+05$ |
|  | DH-4-L | 60 | $5.30 \mathrm{E}+04$ | $9.82 \mathrm{E}+04$ |
| DL-1 | DL-1-U | 140 | BDL | $2.04 \mathrm{E}+04$ |
|  | DL-1-M-U | 140 |  | $9.55 \mathrm{E}+03$ |
|  | DL-1-M-L | 140 |  | $2.13 \mathrm{E}+04$ |
|  | DL-1-L | 140 | $1.89 \mathrm{E}+01$ | $1.80 \mathrm{E}+04$ |
| DL-2 | DL-2-U | 140 | BDL | $5.15 \mathrm{E}+04$ |
|  | DL-2-M-U | 140 |  | $9.57 \mathrm{E}+03$ |
|  | DL-2-M-L | 140 |  | $8.56 \mathrm{E}+03$ |
|  | DL-2-L | 140 | $7.55 \mathrm{E}+00$ | $2.01 \mathrm{E}+04$ |
| DL-3 | DL-3-U | 140 | $5.11 \mathrm{E}+04$ | $4.91 \mathrm{E}+04$ |
|  | DL-3-M-U | 140 | $1.97 \mathrm{E}+04$ | $4.47 \mathrm{E}+05$ |
|  | DL-3-M-L | 140 | $4.61 \mathrm{E}+05$ | $4.61 \mathrm{E}+03$ |
|  | DL-3-L | 140 | $5.32 \mathrm{E}+04$ | $8.81 \mathrm{E}+03$ |
| DL-4 | DL-4-U | 140 | $8.05 \mathrm{E}+04$ | $4.13 \mathrm{E}+04$ |
|  | DL-4-M-U | 140 | $4.17 \mathrm{E}+04$ | $4.39 \mathrm{E}+04$ |
|  | DH-L-M-L | 140 | $4.78 \mathrm{E}+04$ | $7.21 \mathrm{E}+04$ |
|  | DL-4-L | 140 | $1.80 \mathrm{E}+05$ | $4.64 \mathrm{E}+04$ |

## A14 MPN data of Jars

| System | Sample ID | $\mathbf{t}(\mathbf{d})$ | Methanogen MPN/g DRY WT | SRB MPN/g DRY WT |
| :---: | :---: | :---: | :---: | :---: |
|  | JAR D1-1 | 140 | $1.83 \mathrm{E}+05$ | $4.96 \mathrm{E}+05$ |
|  | JAR D1-2 | 140 | $9.57 \mathrm{E}+04$ | $9.23 \mathrm{E}+04$ |
| 2 | JAR D2-1 | 140 | $3.35 \mathrm{E}+05$ | $2.24 \mathrm{E}+05$ |
|  | JAR D2-2 | 140 | $9.75 \mathrm{E}+04$ | $5.08 \mathrm{E}+05$ |
| 3 | JAR D3-1 | 140 | $5.02 \mathrm{E}+05$ | $5.35 \mathrm{E}+05$ |
|  | JAR D3-2 | 140 | $8.89 \mathrm{E}+04$ | $1.97 \mathrm{E}+05$ |
| 4 | JAR D4-1 | 140 | $2.07 \mathrm{E}+05$ | $2.10 \mathrm{E}+05$ |
|  | JAR D4-2 | 140 | $2.07 \mathrm{E}+05$ | $9.42 \mathrm{E}+04$ |

A15 Results of $\mathbf{S}^{\mathbf{2 -}}$ analysis in dynamic system (MFT)

| $\mathbf{1 D}$ | $\mathbf{s}^{\mathbf{2}}$ | $\mathbf{I D}$ | $\mathbf{s}^{\mathbf{2}}$ | $\mathbf{1 D}$ | $\mathbf{s}^{\mathbf{2}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| DL-1-U | BDL | DH-3-U | BDL | DH-4-U | BDL |
| DL-1-M-U | BDL | DH-3-M-U | BDL | DH-4-M-U | BDL |
| DL-1-M-L | BDL | DH-3-M-L | BDL | DH-4-M-L | BDL |
| DL-1-L | BDL | DH-3-L | BDL | DH-4-L | BDL |
| DL-2-U | BDL | DL-3-U | BDL | DL-4-U | BDL |
| DL-2-M-U | BDL | DL-3-M-U | BDL | DL-4-M-U | BDL |
| DL-2-M-L | BDL | DL-3-M-L | BDL | DL-4-M-L | BDL |
| DL-2-L | BDL | DL-3-L | BDL | DL-4-L | BDL |

A16 Results of $\mathbf{S}^{2-}$ analysis in D-3 system

| ID | Day | $\mathbf{s}^{\mathbf{2 -}}$ | ID | Day | $\mathbf{S}^{\mathbf{2}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| DH-3-3 | 3 | BDL | DL-3-3 | 3 | BDL |
| DH-3-7 | 7 | BDL | DL-3-7 | 7 | BDL |
| DH-3-14 | 14 | BDL | DL-3-14 | 14 | BDL |
| DH-3-21 | 21 | BDL | DL-3-21 | 21 | BDL |
| DH-3-28 | 28 | BDL | DL-3-28 | 28 | BDL |
| DH-3-35 | 35 | BDL | DL-3-35 | 35 | BDL |
| DH-3-42 | 42 | 0.8 | DL-3-42 | 42 | BDL |
| DH-3-49 | 49 | 1.0 | DL-3-49 | 49 | BDL |
| DH-3-56 | 56 | 2.0 | DL-3-56 | 56 | BDL |
| DH-3-60 | 60 | BDL | DL-3-63 | 63 | BDL |
|  |  |  | DL-3-70 | 70 | BDL |
|  |  |  | DL-3-77 | 77 | BDL |
|  |  |  | DL-3-84 | 84 | BDL |
|  |  |  | DL-3-91 | 91 | BDL |
|  |  |  | DL-3-98 | 98 | BDL |
|  |  |  | DL-3-105 | 105 | BDL |
|  |  |  | DL-3-112 | 112 | 1.0 |
|  |  |  | DL-3-119 | 119 | 0.5 |
|  |  |  | DL-3-126 | 126 | BDL |
|  |  |  | DL-3-133 | 133 | BDL |
|  |  |  |  | DL-3-140 | 140 |


| ع\％ | て－tairs |
| :---: | :---: |
| s s | ［－tarer |
| TOE | $\tau$－$\varepsilon$ are ${ }^{\text {d }}$ |
| Tag | ［－¢GIET |
| TG日 | $\tau-z a^{\text {rep }}$ |
| Tロя | ［－zarer |
| TGQ | z－IGres |
| TGG | I－IGrer |
| ${ }_{-\tau} \mathrm{S}$ | （II |


| ID | Day | $\mathbf{S}^{\mathbf{2 -}}$ | ID | Day | $\mathbf{S}^{\mathbf{2 -}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| DH－4－3 <br> DH－4－7 <br> DH－4－14 <br> DH－4－21 <br> DH－4－28 <br> DH－4－35 <br> DH－4－42 <br> DH－4－49 <br> DH－4－56 <br> DH－4－60 | 3 | BDL | DL－4－3 | 3 | BDL |
|  | 7 | BDL | DL－4－7 | 7 | BDL |
|  | 14 | BDL | DL－4－14 | 14 | BDL |
|  | 21 | BDL | DL－4－21 | 21 | BDL |
|  | 28 | BDL | DL－4－28 | 28 | BDL |
|  | 35 | 1.0 | DL－4－35 | 35 | BDL |
|  | 42 | 2.0 | DL－4－42 | 42 | BDL |
|  | 49 | 2.5 | DL－4－49 | 49 | BDL |
|  | 56 | 3.0 | DL－4－56 | 56 | BDL |
|  | 60 | 3.0 | DL－4－63 | 63 | BDL |
|  |  |  | DL－4－70 | 70 | BDL |
|  |  |  | DL－4－77 | 77 | BDL |
|  |  |  | DL－4－84 | 84 | BDL |
|  |  |  | DL－4－91 | 91 | BDL |
|  |  |  | DL－4－98 | 98 | BDL |
|  |  |  | DL－4－105 | 105 | BDL |
|  |  |  | DL－4－112 | 112 | 4.5 |
|  |  |  | DL－4－119 | 119 | BDL |
|  |  |  | DL－4－126 | 126 | 2.5 |
|  |  |  | DL－4－133 | 133 | BDL |
|  |  |  | DL－4－140 | 140 | BDL |



## A19 Results of Redox potential analysis in dynamic system

| ID | Eh (mV) |  | ID | Eh (mV) |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  | 1 hr | 2 hr |  | 1 hr | 2 hr |
| DH-1-U | -141.1 | -153.3 | DH-3-U | -140.0 | -155.0 |
| DH-1-M-U | -174.4 | -178.8 | DH-3-M-U | 55.0 | 9.0 |
| DH-1-M-L | -150.4 | -161.0 | DH-3-M-L | -130.0 | -165.0 |
| DH-1-L | -147.7 | -166.9 | DH-3-L | -200.0 | -204.0 |
| DL-1-U | -153.0 | -165.0 | DL-3-U | -165.0 | -178.0 |
| DL-1-M-U | -142.0 | -153.0 | DL-3-M-U | -190.0 | -200.0 |
| DL-1-M-L | -172.0 | -178.0 | DL-3-M-L | -194.0 | -201.0 |
| DL-1-L | -154.0 | -164.0 | DL-3-L | -196.0 | -199.0 |
| DH-2-U | -181.0 | -182.8 | DH-4-U | 17.2 | -9.0 |
| DH-2-M-U | -178.8 | -182.6 | DH-4-M-U | 46.0 | 13.0 |
| DH-2-M-L | -194.1 | -198.0 | DH-4-M-L | 24.0 | -13.0 |
| DH-2-L | -180.7 | -183.8 | DH-4-L | 21.1 | 0.0 |
| DL-2-U | -193.0 | -195.0 | DL-4-U | 65.0 | 47.0 |
| DL-2-M-U | -183.0 | -189.0 | DL-4-M-U | -53.0 | -78.0 |
| DL-2-M-L | -187.0 | -189.0 | DL-4-M-L | 39.0 | 15.0 |
| DL-2-L | -198.0 | -199.0 | DL-4-L | -125.0 | -150.0 |

A20 Results of Redox potential analysis in Jars

| ID | Eh (mV) |  |
| :---: | :---: | :---: |
|  | $\mathbf{1 ~ h r}$ | $\mathbf{2 ~ h r}$ |
| D1-1 | -178 | -190 |
| D1-2 | -175 | -184 |
| D2-1 | -198 | -206 |
| D2-2 | -208 | -211 |
| D3-1 | -201 | -208 |
| D3-2 | -180 | -186 |
| D4-1 | -188 | -194 |
| D4-2 | -91 | -102 |

A21 Solids content of D3 \& D4 systems

| Sample ID | $\begin{gathered} \text { Solids } \\ (\mathrm{g} / 100 \mathrm{~g}) \end{gathered}$ | $\begin{aligned} & \text { Bitumen } \\ & (\mathrm{g} / \mathbf{1 0 0} \mathrm{g}) \\ & \hline \end{aligned}$ | $22 \mu \mathrm{~m}$ | $11 \mu \mathrm{~m}$ | $5.5 \mu \mathrm{~m}$ | $2.8 \mu \mathrm{~m}$ | $1.4 \mu \mathrm{~m}$ | $0.5 \mu \mathrm{~m}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Base | 38.8 | 3.35 | 73.8 | 59.0 | 42.9 | 28.3 | 14.2 | 2.5 |
| DH-3-U |  |  |  |  |  |  |  |  |
| DH-3-M-U |  |  |  |  |  |  |  |  |
| DH-3-M-L |  |  |  |  |  |  |  |  |
| DH-3-L | 42.3 | 3.63 | 76.7 | 62.3 | 46.2 | 30.4 | 14.6 | 2.4 |
| DL-3--U | 42.0 | 3.65 | 74.0 | 60.2 | 44.3 | 28.5 | 13.4 | 2.2 |
| DL-3--M-U | 42.8 | 3.20 | 75.7 | 62.9 | 45.8 | 27.1 | 11.6 | 1.9 |
| DL-3--M-L | 43.1 | 3.44 | 73.4 | 59.6 | 43.8 | 27.6 | 12.6 | 2.0 |
| DL-3--L | 43.3 | 3.67 | 74.7 | 60.7 | 45.1 | 29.3 | 13.9 | 2.3 |
| DH-4-U | 42.0 | 3.65 | 74.0 | 60.2 | 44.3 | 28.5 | 13.4 | 2.2 |
| DH-4-M-U | 42.8 | 3.20 | 75.7 | 62.9 | 45.8 | 27.1 | 11.6 | 1.9 |
| DH-4-M-L | 43.1 | 3.44 | 73.4 | 59.6 | 43.8 | 27.6 | 12.6 | 2.0 |
| DH-4-L | 43.3 | 3.67 | 74.7 | 60.7 | 45.1 | 29.3 | 13.9 | 2.3 |
| DL-4--U | 44.8 | 3.58 | 74.6 | 61.2 | 44.8 | 27.2 | 11.7 | 1.7 |
| DL-4--M-U | 43.4 | 3.58 | 74.4 | 60.9 | 44.9 | 28.3 | 12.7 | 1.8 |
| DL-4--M-L | 45.1 | 3.67 | 77.9 | 64.4 | 47.8 | 29.5 | 12.7 | 1.8 |
| DL-4--L | 45.0 | 3.55 | 77.1 | 64.0 | 47.2 | 28.1 | 11.6 | 1.7 |

## A22 Solids content of Jars

| Sample ID | Solids <br> $(\mathbf{g} / \mathbf{1 0 0} \mathbf{g})$ | Bitumen <br> $(\mathbf{g} / \mathbf{1 0 0} \mathbf{g})$ | $\mathbf{2 2 \mu \mathrm { m }}$ | $\mathbf{1 1} \mu \mathrm{m}$ | $\mathbf{5 . 5 \mu \mathrm { m }}$ | $\mathbf{2 . 8} \boldsymbol{\mu \mathrm { m }}$ | $\mathbf{1 . 4} \boldsymbol{\mu \mathrm { m }}$ | $\mathbf{0 . 5} \boldsymbol{\mu \mathrm { m }}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| D1-Jar Base | 38.0 | 3.32 | 78.7 | 63.1 | 45.9 | 30.0 | 14.6 | 2.4 |
| D2-Jar Base | 38.0 | 3.27 | 74.0 | 59.8 | 43.6 | 27.7 | 12.9 | 1.9 |
| D3\&D4 Jar | 38.8 | 3.35 | 73.8 | 59.0 | 42.9 | 28.3 | 14.2 | 2.5 |
| Base | 39.4 | 3.1 | 74.8 | 61.4 | 45.4 | 28.2 | 12.2 | 1.8 |
| Jar D1-1 | 38.5 | 3.3 | 76.4 | 62.2 | 45.9 | 29.1 | 13.0 | 1.9 |
| Jar D1-2 | 38.1 | 3.3 | 77.8 | 63.4 | 46.7 | 29.1 | 12.7 | 1.8 |
| Jar D2-1 | 38.2 | 3.3 | 75.7 | 61.9 | 45.5 | 27.8 | 12.0 | 1.7 |
| Jar D2-2 | 37.5 | 3.2 | 75.4 | 61.0 | 44.8 | 28.3 | 13.0 | 2.1 |
| Jar D3-1 | 37.5 |  |  |  |  |  |  |  |
| Jar D3-2 | 37.8 | 3.1 | 78.1 | 65.1 | 47.5 | 27.2 | 11.4 | 2.3 |
| Jar D4-1 | 39.0 | 3.4 | 77.7 | 63.3 | 46.7 | 29.2 | 13.0 | 2.1 |
| Jar D4-2 | 38.9 | 3.3 | 77.3 | 63.6 | 47.1 | 29.0 | 12.4 | 1.7 |

## A23 Volume of CT water flow through D1 column (high flowrate)

| No. | Day | $\mathbf{V}$ (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DH-1-1 | 1 | 0 | 0 |
| DH-1-2 | 2 | 0 | 0 |
| DH-1-3 | 3 | 60 | 60 |
| DH-1-4 | 4 | 0 | 60 |
| DH-1-5 | 5 | 240 | 300 |
| DH-1-6 | 6 | 600 | 900 |
| DH-1-7 | 7 | 440 | 1340 |
| DH-1-8 | 8 | 360 | 1700 |
| DH-1-9 | 9 | 455 | 2155 |
| DH-1-10 | 10 | 380 | 2535 |
| DH-1-11 | 11 | 420 | 2955 |
| DH-1-12 | 12 | 330 | 3285 |
| DH-1-13 | 13 | 460 | 3745 |
| DH-1-14 | 14 | 465 | 4210 |
| DH-1-15 | 15 | 270 | 4480 |
| DH-1-16 | 16 | 395 | 4875 |
| DH-1-17 | 17 | 410 | 5285 |
| DH-1-18 | 18 | 480 | 5765 |
| DH-1-19 | 19 | 480 | 6245 |
| DH-1-20 | 20 | 170 | 6415 |
| DH-1-21 | 21 | 350 | 6765 |
| DH-1-22 | 22 | 410 | 7175 |
| DH-1-23 | 23 | 330 | 7505 |
| DH-1-24 | 24 | 520 | 8025 |
| DH-1-25 | 25 | 510 | 8535 |
| DH-1-26 | 26 | 200 | 8735 |
| DH-1-27 | 27 | 520 | 9255 |
| DH-1-28 | 28 | 520 | 9775 |
| DH-1-29 | 29 | 440 | 10215 |
| DH-1-30 | 30 | 300 | 10515 |
| DH-1-31 | 31 | 350 | 10865 |
| DH-1-32 | 32 | 490 | 11355 |
| DH-1-33 | 33 | 200 | 11555 |
| DH-1-34 | 34 | 430 | 11985 |
| DH-1-35 | 35 | 500 | 12485 |
| DH-1-36 | 36 | 480 | 12965 |

A23 Volume of CT water flow through D1 column (high flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DH-1-37 | 37 | 410 | 13375 |
| DH-1-38 | 38 | 500 | 13875 |
| DH-1-39 | 39 | 500 | 14375 |
| DH-1-40 | 40 | 300 | 14675 |
| DH-1-41 | 41 | 420 | 15095 |
| DH-1-42 | 42 | 460 | 15555 |
| DH-1-43 | 43 | 450 | 16005 |
| DH-1-44 | 44 | 460 | 16465 |
| DH-1-45 | 45 | 450 | 16915 |
| DH-1-46 | 46 | 500 | 17415 |
| DH-1-47 | 47 | 450 | 17865 |
| DH-1-48 | 48 | 340 | 18205 |
| DH-1-49 | 49 | 500 | 18705 |
| DH-1-50 | 50 | 460 | 19165 |
| DH-1-51 | 51 | 500 | 19665 |
| DH-1-52 | 52 | 450 | 20115 |
| DH-1-53 | 53 | 440 | 20555 |
| DH-1-54 | 54 | 510 | 21065 |
| DH-1-55 | 55 | 300 | 21365 |
| DH-1-56 | 56 | 480 | 21845 |
| DH-1-57 | 57 | 420 | 22265 |
| DH-1-58 | 58 | 400 | 22665 |
| DH-1-59 | 59 | 450 | 23115 |
| DH-1-60 | 60 | 700 | 23815 |
| Flowrate | 1440 hrs | 23815 ml | $16.54 \mathrm{ml} / \mathrm{h}$ |

## A24 Volume of CT water flow through D2 column (high flowrate)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DH-2-1 | 1 | 0 | 0 |
| DH-2-2 | 2 | 355 | 355 |
| DH-2-3 | 3 | 355 | 710 |
| DH-2-4 | 4 | 400 | 1110 |
| DH-2-5 | 5 | 570 | 1680 |
| DH-2-6 | 6 | 300 | 1980 |
| DH-2-7 | 7 | 420 | 2400 |
| DH-2-8 | 8 | 460 | 2860 |
| DH-2-9 | 9 | 430 | 3290 |
| DH-2-10 | 10 | 400 | 3690 |
| DH-2-11 | 11 | 475 | 4165 |
| DH-2-12 | 12 | 470 | 4635 |
| DH-2-13 | 13 | 285 | 4920 |
| DH-2-14 | 14 | 320 | 5240 |
| DH-2-15 | 15 | 375 | 5615 |
| DH-2-16 | 16 | 310 | 5925 |
| DH-2-17 | 17 | 175 | 6100 |
| DH-2-18 | 18 | 300 | 6400 |
| DH-2-19 | 19 | 510 | 6910 |
| DH-2-20 | 20 | 300 | 7210 |
| DH-2-21 | 21 | 500 | 7710 |
| DH-2-22 | 22 | 575 | 8285 |
| DH-2-23 | 23 | 520 | 8805 |
| DH-2-24 | 24 | 480 | 9285 |
| DH-2-25 | 25 | 505 | 9790 |
| DH-2-26 | 26 | 510 | 10300 |
| DH-2-27 | 27 | 380 | 10680 |
| DH-2-28 | 28 | 430 | 11110 |
| DH-2-29 | 29 | 500 | 11610 |
| DH-2-30 | 30 | 500 | 12110 |
| DH-2-31 | 31 | 480 | 12590 |
| DH-2-32 | 32 | 420 | 13010 |
| DH-2-33 | 33 | 480 | 13490 |
| DH-2-34 | 34 | 460 | 13950 |
| DH-2-35 | 35 | 450 | 14400 |
| DH-2-36 | 36 | 370 | 14770 |

A24 Volume of CT water flow through D2 column (high flowrate) (cont'd)

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| $\stackrel{\text { E }}{\text { E }}$ |  | $\frac{2}{3}$ |
|  |  |  |
| \% |  | 20 |

## A25 Volume of CT water flow through D3 column (high flowrate)

| No. | Day | $V(\mathrm{ml})$ | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DH-3-1 | 1 | 0 | 0 |
| DH-3-2 | 2 | 0 | 0 |
| DH-3-3 | 3 | 480 | 480 |
| DH-3-4 | 4 | 480 | 960 |
| DH-3-5 | 5 | 480 | 1440 |
| DH-3-6 | 6 | 480 | 1920 |
| DH-3-7 | 7 | 480 | 2400 |
| DH-3-8 | 8 | 480 | 2880 |
| DH-3-9 | 9 | 480 | 3360 |
| DH-3-10 | 10 | 480 | 3840 |
| DH-3-11 | 11 | 480 | 4320 |
| DH-3-12 | 12 | 480 | 4800 |
| DH-3-13 | 13 | 450 | 5250 |
| DH-3-14 | 14 | 420 | 5670 |
| DH-3-15 | 15 | 480 | 6150 |
| DH-3-16 | 16 | 480 | 6630 |
| DH-3-17 | 17 | 480 | 7110 |
| DH-3-18 | 18 | 480 | 7590 |
| DH-3-19 | 19 | 480 | 8070 |
| DH-3-20 | 20 | 480 | 8550 |
| DH-3-21 | 21 | 480 | 9030 |
| DH-3-22 | 22 | 480 | 9510 |
| DH-3-23 | 23 | 480 | 9990 |
| DH-3-24 | 24 | 480 | 10470 |
| DH-3-25 | 25 | 480 | 10950 |
| DH-3-26 | 26 | 480 | 11430 |
| DH-3-27 | 27 | 480 | 11910 |
| DH-3-28 | 28 | 480 | 12390 |
| DH-3-29 | 29 | 480 | 12870 |
| DH-3-30 | 30 | 480 | 13350 |
| DH-3-31 | 31 | 480 | 13830 |
| DH-3-32 | 32 | 480 | 14310 |
| DH-3-33 | 33 | 480 | 14790 |
| DH-3-34 | 34 | 480 | 15270 |
| DH-3-35 | 35 | 480 | 15750 |
| DH-3-36 | 36 | 450 | 16200 |

A25 Volume of CT water flow through D3 column (high flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DH-3-37 | 37 | 480 | 16680 |
| DH-3-38 | 38 | 480 | 17160 |
| DH-3-39 | 39 | 480 | 17640 |
| DH-3-40 | 40 | 480 | 18120 |
| DH-3-41 | 41 | 480 | 18600 |
| DH-3-42 | 42 | 400 | 19000 |
| DH-3-43 | 43 | 350 | 19350 |
| DH-3-44 | 44 | 480 | 19830 |
| DH-3-45 | 45 | 480 | 20310 |
| DH-3-46 | 46 | 480 | 20790 |
| DH-3-47 | 47 | 450 | 21240 |
| DH-3-48 | 48 | 400 | 21640 |
| DH-3-49 | 49 | 350 | 21990 |
| DH-3-50 | 50 | 480 | 22470 |
| DH-3-51 | 51 | 480 | 22950 |
| DH-3-52 | 52 | 480 | 23430 |
| DH-3-53 | 53 | 480 | 23910 |
| DH-3-54 | 54 | 480 | 24390 |
| DH-3-55 | 55 | 400 | 24790 |
| DH-3-56 | 56 | 350 | 25140 |
| DH-3-57 | 57 | 500 | 25640 |
| DH-3-58 | 58 | 450 | 26090 |
| DH-3-59 | 59 | 400 | 26490 |
| DH-3-60 | 60 | 450 | 26940 |
| Flowrate | 1440 hrs | 26940 | $18.71 \mathrm{ml} / \mathrm{h}$ |

## A26 Volume of CT water flow through D4 column (high flowrate)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DH-4-1 | 1 | 0 | 0 |
| DH-4-2 | 2 | 0 | 0 |
| DH-4-3 | 3 | 300 | 300 |
| DH-4-4 | 4 | 480 | 780 |
| DH-4-5 | 5 | 480 | 1260 |
| DH-4-6 | 6 | 480 | 1740 |
| DH-4-7 | 7 | 480 | 2220 |
| DH-4-8 | 8 | 480 | 2700 |
| DH-4-9 | 9 | 480 | 3180 |
| DH-4-10 | 10 | 480 | 3660 |
| DH-4-11 | 11 | 480 | 4140 |
| DH-4-12 | 12 | 480 | 4620 |
| DH-4-13 | 13 | 480 | 5100 |
| DH-4-14 | 14 | 480 | 5580 |
| DH-4-15 | 15 | 480 | 6060 |
| DH-4-16 | 16 | 480 | 6540 |
| DH-4-17 | 17 | 480 | 7020 |
| DH-4-18 | 18 | 480 | 7500 |
| DH-4-19 | 19 | 480 | 7980 |
| DH-4-20 | 20 | 480 | 8460 |
| DH-4-21 | 21 | 480 | 8940 |
| DH-4-22 | 22 | 480 | 9420 |
| DH-4-23 | 23 | 480 | 9900 |
| DH-4-24 | 24 | 480 | 10380 |
| DH-4-25 | 25 | 480 | 10860 |
| DH-4-26 | 26 | 480 | 11340 |
| DH-4-27 | 27 | 480 | 11820 |
| DH-4-28 | 28 | 480 | 12300 |
| DH-4-29 | 29 | 480 | 12780 |
| DH-4-30 | 30 | 480 | 13260 |
| DH-4-31 | 31 | 480 | 13740 |
| DH-4-32 | 32 | 480 | 14220 |
| DH-4-33 | 33 | 480 | 14700 |
| DH-4-34 | 34 | 480 | 15180 |
| DH-4-35 | 35 | 480 | 15660 |
| DH-4-36 | 36 | 480 | 16140 |

A26 Volume of CT water flow through D4 column (high flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DH-4-37 | 37 | 480 | 16620 |
| DH-4-38 | 38 | 480 | 17100 |
| DH-4-39 | 39 | 480 | 17580 |
| DH-4-40 | 40 | 480 | 18060 |
| DH-4-41 | 41 | 480 | 18540 |
| DH-4-42 | 42 | 480 | 19020 |
| DH-4-43 | 43 | 480 | 19500 |
| DH-4-44 | 44 | 480 | 19980 |
| DH-4-45 | 45 | 480 | 20460 |
| DH-4-46 | 46 | 480 | 20940 |
| DH-4-47 | 47 | 480 | 21420 |
| DH-4-48 | 48 | 480 | 21900 |
| DH-4-49 | 49 | 480 | 22380 |
| DH-4-50 | 50 | 480 | 22860 |
| DH-4-51 | 51 | 480 | 23340 |
| DH-4-52 | 52 | 490 | 23830 |
| DH-4-53 | 53 | 500 | 24330 |
| DH-4-54 | 54 | 500 | 24830 |
| DH-4-55 | 55 | 550 | 25380 |
| DH-4-56 | 56 | 450 | 25830 |
| DH-4-57 | 57 | 500 | 26330 |
| DH-4-58 | - 58 | 550 | 26880 |
| DH-4-59 | 59 | 500 | 27380 |
| DH-4-60 | 60 | 550 | 27930 |
| Flowrate | 1440 hrs | 27930 | $19.40 \mathrm{ml} / \mathrm{h}$ |

A27 Volume of CT water flow through D1 column (low flowrate)

| No. | Day | $\mathbf{V}$ (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-1-1 | 1 | 0 | 0 |
| DL-1-2 | 2 | 0 | 0 |
| DL-1-3 | 3 | 0 | 0 |
| DL-1-4 | 4 | 0 | 0 |
| DL-1-5 | 5 | 0 | 0 |
| DL-1-6 | 6 | 0 | 0 |
| DL-1-7 | 7 | 0 | 0 |
| DL-1-8 | 8 | 150 | 150 |
| DL-1-9 | 9 | 100 | 250 |
| DL-1-10 | 10 | 165 | 415 |
| DL-1-11 | 11 | 180 | 595 |
| DL-1-12 | 12 | 125 | 720 |
| DL-1-13 | 13 | 110 | 830 |
| DL-1-14 | 14 | 100 | 930 |
| DL-1-15 | 15 | 150 | 1080 |
| DL-1-16 | 16 | 135 | 1215 |
| DL-1-17 | 17 | 65 | 1280 |
| DL-1-18 | 18 | 110 | 1390 |
| DL-1-19 | 19 | 130 | 1520 |
| DL-1-20 | 20 | 175 | 1695 |
| DL-1-21 | 21 | 190 | 1885 |
| DL-1-22 | 22 | 150 | 2035 |
| DL-1-23 | 23 | 150 | 2185 |
| DL-1-24 | 24 | 130 | 2315 |
| DL-1-25 | 25 | 100 | 2415 |
| DL-1-26 | 26 | 145 | 2560 |
| DL-1-27 | 27 | 150 | 2710 |
| DL-1-28 | 28 | 145 | 2855 |
| DL-1-29 | 29 | 140 | 2995 |
| DL-1-30 | 30 | 140 | 3135 |
| DL-1-31 | 31 | 120 | 3255 |
| DL-1-32 | 32 | 165 | 3420 |
| DL-1-33 | 33 | 80 | 3500 |
| DL-1-34 | 34 | 150 | 3650 |
| DL-1-35 | 35 | 125 | 3775 |
| DL-1-36 | 36 | 135 | 3910 |
|  |  |  |  |

## A27 Volume of CT water flow through D1 column (low flowrate) (cont'd)

| No. | Day | $V(\mathrm{ml})$ | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-1-37 | 37 | 160 | 4070 |
| DL-1-38 | 38 | 90 | 4160 |
| DL-1-39 | 39 | 110 | 4270 |
| DL-1-40 | 40 | 140 | 4410 |
| DL-1-41 | 41 | 150 | 4560 |
| DL-1-42 | 42 | 100 | 4660 |
| DL-1-43 | 43 | 150 | 4810 |
| DL-1-44 | 44 | 140 | 4950 |
| DL-1-45 | 45 | 90 | 5040 |
| DL-1-46 | 46 | 130 | 5170 |
| DL-1-47 | 47 | 145 | 5315 |
| DL-1-48 | 48 | 80 | 5395 |
| DL-1-49 | 49 | 80 | 5475 |
| DL-1-50 | 50 | 70 | 5545 |
| DL-1-51 | 51 | 175 | 5720 |
| DL-1-52 | 52 | 150 | 5870 |
| DL-1-53 | 53 | 100 | 5970 |
| DL-1-54 | 54 | 100 | 6070 |
| DL-1-55 | 55 | 170 | 6240 |
| DL-1-56 | 56 | 150 | 6390 |
| DL-1-57 | 57 | 100 | 6490 |
| DL-1-58 | 58 | 140 | 6630 |
| DL-1-59 | 59 | 200 | 6830 |
| DL-1-60 | 60 | 150 | 6980 |
| DL-1-61 | 61 | 150 | 7130 |
| DL-1-62 | 62 | 150 | 7280 |
| DL-1-63 | 63 | 140 | 7420 |
| DL-1-64 | 64 | 140 | 7560 |
| DL-1-65 | 65 | 200 | 7760 |
| DL-1-66 | 66 | 100 | 7860 |
| DL-1-67 | 67 | 130 | 7990 |
| DL-1-68 | 68 | 150 | 8140 |
| DL-1-69 | 69 | 130 | 8270 |
| DL-1-70 | 70 | 160 | 8430 |
| DL-1-71 | 71 | 140 | 8570 |
| DL-1-72 | 72 | 180 | 8750 |

A27 Volume of CT water flow through D1 column (low flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-1-73 | 73 | 110 | 8860 |
| DL-1-74 | 74 | 130 | 8990 |
| DL-1-75 | 75 | 130 | 9120 |
| DL-1-76 | 76 | 160 | 9280 |
| DL-1-77 | 77 | 160 | 9440 |
| DL-1-78 | 78 | 150 | 9590 |
| DL-1-79 | 79 | 170 | 9760 |
| DL-1-80 | 80 | 150 | 9910 |
| DL-1-81 | 81 | 100 | 10010 |
| DL-1-82 | 82 | 140 | 10150 |
| DL-1-83 | 83 | 130 | 10280 |
| DL-1-84 | 84 | 60 | 10340 |
| DL-1-85 | 85 | 90 | 10430 |
| DL-1-86 | 86 | 170 | 10600 |
| DL-1-87 | 87 | 140 | 10740 |
| DL-1-88 | 88 | 135 | 10875 |
| DL-1-89 | 89 | 140 | 11015 |
| DL-1-90 | 90 | 140 | 11155 |
| DL-1-91 | 91 | 150 | 11305 |
| DL-1-92 | 92 | 100 | 11405 |
| DL-1-93 | 93 | 140 | 11545 |
| DL-1-94 | 94 | 145 | 11690 |
| DL-1-95 | 95 | 150 | 11840 |
| DL-1-96 | 96 | 120 | 11960 |
| DL-1-97 | 97 | 80 | 12040 |
| DL-1-98 | 98 | 60 | 12100 |
| DL-1-99 | 99 | 40 | 12140 |
| DL-1-100 | 100 | 75 | 12215 |
| DL-1-101 | 101 | 75 | 12290 |
| DL-1-102 | 102 | 140 | 12430 |
| DL-1-103 | 103 | 140 | 12570 |
| DL-1-104 | 104 | 150 | 12720 |
| DL-1-105 | 105 | 140 | 12860 |
| DL-1-106 | 106 | 130 | 12990 |
| DL-1-107 | 107 | 135 | 13125 |
| DL-1-108 | 108 | 145 | 13270 |

A27 Volume of CT water flow through D1 column（low flowrate）（cont＇d）

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A28 Volume of CT water flow through D2 column (low flowrate)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-2-1 | 1 | 0 | 0 |
| DL-2-2 | 2 | 0 | 0 |
| DL-2-3 | 3 | 0 | 0 |
| DL-2-4 | 4 | 0 | 0 |
| DL-2-5 | 5 | 130 | 130 |
| DL-2-6 | 6 | 0 | 130 |
| DL-2-7 | 7 | 190 | 320 |
| DL-2-8 | 8 | 190 | 510 |
| DL-2-9 | 9 | 150 | 660 |
| DL-2-10 | 10 | 160 | 820 |
| DL-2-11 | 11 | 160 | 980 |
| DL-2-12 | 12 | 150 | 1130 |
| DL-2-13 | 13 | 155 | 1285 |
| DL-2-14 | 14 | 160 | 1445 |
| DL-2-15 | 15 | 150 | 1595 |
| DL-2-16 | 16 | 150 | 1745 |
| DL-2-17 | 17 | 160 | 1905 |
| DL-2-18 | 18 | 115 | 2020 |
| DL-2-19 | 19 | 100 | 2120 |
| DL-2-20 | 20 | 90 | 2210 |
| DL-2-21 | 21 | 70 | 2280 |
| DL-2-22 | 22 | 160 | 2440 |
| DL-2-23 | 23 | 190 | 2630 |
| DL-2-24 | 24 | 180 | 2810 |
| DL-2-25 | 25 | 120 | 2930 |
| DL-2-26 | 26 | 150 | 3080 |
| DL-2-27 | 27 | 150 | 3230 |
| DL-2-28 | 28 | 150 | 3380 |
| DL-2-29 | 29 | 150 | 3530 |
| DL-2-30 | 30 | 120 | 3650 |
| DL-2-31 | 31 | 150 | 3800 |
| DL-2-32 | 32 | 90 | 3890 |
| DL-2-33 | 33 | 140 | 4030 |
| DL-2-34 | 34 | 150 | 4180 |
| DL-2-35 | 35 | 130 | 4310 |
| DL-2-36 | 36 | 115 | 4425 |

A28 Volume of CT water flow through D2 column (low flowrate) (cont'd)

| No. | Day | $V(\mathrm{ml})$ | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-2-37 | 37 | 150 | 4575 |
| DL-2-38 | 38 | 200 | 4775 |
| DL-2-39 | 39 | 150 | 4925 |
| DL-2-40 | 40 | 100 | 5025 |
| DL-2-41 | 41 | 155 | 5180 |
| DL-2-42 | 42 | 140 | 5320 |
| DL-2-43 | 43 | 130 | 5450 |
| DL-2-44 | 44 | 130 | 5580 |
| DL-2-45 | 45 | 90 | 5670 |
| DL-2-46 | 46 | 145 | 5815 |
| DL-2-47 | 47 | 110 | 5925 |
| DL-2-48 | 48 | 145 | 6070 |
| DL-2-49 | 49 | 140 | 6210 |
| DL-2-50 | 50 | 150 | 6360 |
| DL-2-51 | 51 | 140 | 6500 |
| DL-2-52 | 52 | 200 | 6700 |
| DL-2-53 | 53 | 100 | 6800 |
| DL-2-54 | 54 | 130 | 6930 |
| DL-2-55 | 55 | 140 | 7070 |
| DL-2-56 | 56 | 135 | 7205 |
| DL-2-57 | 57 | 170 | 7375 |
| DL-2-58 | 58 | 150 | 7525 |
| DL-2-59 | 59 | 180 | 7705 |
| DL-2-60 | 60 | 110 | 7815 |
| DL-2-61 | 61 | 130 | 7945 |
| DL-2-62 | 62 | 130 | 8075 |
| DL-2-63 | 63 | 160 | 8235 |
| DL-2-64 | 64 | 150 | 8385 |
| DL-2-65 | 65 | 140 | 8525 |
| DL-2-66 | 66 | 170 | 8695 |
| DL-2-67 | 67 | 150 | 8845 |
| DL-2-68 | 68 | 100 | 8945 |
| DL-2-69 | 69 | 120 | 9065 |
| DL-2-70 | 70 | 100 | 9165 |
| DL-2-71 | 71 | 80 | 9245 |
| DL-2-72 | 72 | 60 | 9305 |


| No. | Day | $\mathbf{V}$ (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-2-73 | 73 | 90 | 9395 |
| DL-2-74 | 74 | 90 | 9485 |
| DL-2-75 | 75 | 80 | 9565 |
| DL-2-76 | 76 | 60 | 9625 |
| DL-2-77 | 77 | 95 | 9720 |
| DL-2-78 | 78 | 90 | 9810 |
| DL-2-79 | 79 | 60 | 9870 |
| DL-2-80 | 80 | 70 | 9940 |
| DL-2-81 | 81 | 60 | 10000 |
| DL-2-82 | 82 | 145 | 10145 |
| DL-2-83 | 83 | 140 | 10285 |
| DL-2-84 | 84 | 140 | 10425 |
| DL-2-85 | 85 | 120 | 10545 |
| DL-2-86 | 86 | 40 | 10585 |
| DL-2-87 | 87 | 40 | 10625 |
| DL-2-88 | 88 | 120 | 10745 |
| DL-2-89 | 89 | 140 | 10885 |
| DL-2-90 | 90 | 140 | 11025 |
| DL-2-91 | 91 | 150 | 11175 |
| DL-2-92 | 92 | 140 | 11315 |
| DL-2-93 | 93 | 90 | 11405 |
| DL-2-94 | 94 | 60 | 11465 |
| DL-2-95 | 95 | 150 | 11615 |
| DL-2-96 | 96 | 140 | 11755 |
| DL-2-97 | 97 | 140 | 11895 |
| DL-2-98 | 98 | 140 | 12035 |
| DL-2-99 | 99 | 140 | 12175 |
| DL-2-100 | 100 | 150 | 12325 |
| DL-2-101 | 101 | 140 | 12465 |
| DL-2-102 | 102 | 135 | 12600 |
| DL-2-103 | 103 | 150 | 12750 |
| DL-2-104 | 104 | 130 | 12880 |
| DL-2-105 | 105 | 140 | 13020 |
| DL-2-106 | 106 | 140 | 13160 |
| DL-2-107 | 107 | 140 | 13300 |
| DL-2-108 | 108 | 140 | 13440 |
|  |  |  |  |

A28 Volume of CT water flow through D2 column (low flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-2-109 | 109 | 110 | 13550 |
| DL-2-110 | 110 | 100 | 13650 |
| DL-2-111 | 111 | 70 | 13720 |
| DL-2-112 | 112 | 70 | 13790 |
| DL-2-113 | 113 | 60 | 13850 |
| DL-2-114 | 114 | 60 | 13910 |
| DL-2-115 | 115 | 60 | 13970 |
| DL-2-116 | 116 | 60 | 14030 |
| DL-2-117 | 117 | 60 | 14090 |
| DL-2-118 | 118 | 60 | 14150 |
| DL-2-119 | 119 | 60 | 14210 |
| DL-2-120 | 120 | 60 | 14270 |
| DL-2-121 | 121 | 60 | 14330 |
| DL-2-122 | 122 | 50 | 14380 |
| DL-2-123 | 123 | 50 | 14430 |
| DL-2-124 | 124 | 60 | 14490 |
| DL-2-125 | 125 | 50 | 14540 |
| DL-2-126 | 126 | 50 | 14590 |
| DL-2-127 | 127 | 50 | 14640 |
| DL-2-128 | 128 | 100 | 14740 |
| DL-2-129 | 129 | 60 | 14800 |
| DL-2-130 | 130 | 60 | 14860 |
| DL-2-131 | 131 | 60 | 14920 |
| DL-2-132 | 132 | 80 | 15000 |
| DL-2-133 | 133 | 90 | 15090 |
| DL-2-134 | 134 | 90 | 15180 |
| DL-2-135 | 135 | 90 | 15270 |
| DL-2-136 | 136 | 80 | 15350 |
| DL-2-137 | 137 | 90 | 15440 |
| DL-2-138 | 138 | 140 | 15580 |
| DL-2-139 | 139 | 140 | 15720 |
| DL-2-140 | 140 | 140 | 15860 |
| Flowrate | 3360 hrs | 15860 | $4.72 \mathrm{ml} / \mathrm{h}$ |


| No. | Day | $\mathbf{V}(\mathbf{m l})$ | Sum-V $(\mathbf{m l})$ |
| :---: | :---: | :---: | :---: |
| DL-3-1 | 1 | 0 | 0 |
| DL-3-2 | 2 | 0 | 0 |
| DL-3-3 | 3 | 140 | 140 |
| DL-3-4 | 4 | 100 | 240 |
| DL-3-5 | 5 | 140 | 380 |
| DL-3-6 | 6 | 140 | 520 |
| DL-3-7 | 7 | 140 | 660 |
| DL-3-8 | 8 | 140 | 800 |
| DL-3-9 | 9 | 140 | 940 |
| DL-3-10 | 10 | 140 | 1080 |
| DL-3-11 | 11 | 140 | 1220 |
| DL-3-12 | 12 | 140 | 1360 |
| DL-3-13 | 13 | 140 | 1500 |
| DL-3-14 | 14 | 140 | 1640 |
| DL-3-15 | 15 | 140 | 1780 |
| DL-3-16 | 16 | 140 | 1920 |
| DL-3-17 | 17 | 140 | 2060 |
| DL-3-18 | 18 | 140 | 2200 |
| DL-3-19 | 19 | 140 | 2340 |
| DL-3-20 | 20 | 140 | 2480 |
| DL-3-21 | 21 | 140 | 2620 |
| DL-3-22 | 22 | 140 | 2760 |
| DL-3-23 | 23 | 140 | 2900 |
| DL-3-24 | 24 | 140 | 3040 |
| DL-3-25 | 25 | 140 | 3180 |
| DL-3-26 | 26 | 140 | 3320 |
| DL-3-27 | 27 | 140 | 3460 |
| DL-3-28 | 28 | 140 | 3600 |
| DL-3-29 | 29 | 140 | 3740 |
| DL-3-30 | 30 | 140 | 3880 |
| DL-3-31 | 31 | 140 | 4020 |
| DL-3-32 | 32 | 140 | 4160 |
| DL-3-33 | 33 | 140 | 4300 |
| DL-3-34 | 34 | 140 | 4440 |
| DL-3-35 | 35 | 140 | 4580 |
| DL-3-36 | 36 | 150 | 4730 |
|  |  |  |  |

A29 Volume of CT water flow through D3 column (low flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-3-37 | 37 | 150 | 4880 |
| DL-3-38 | 38 | 150 | 5030 |
| DL-3-39 | 39 | 140 | 5170 |
| DL-3-40 | 40 | 150 | 5320 |
| DL-3-41 | 41 | 150 | 5470 |
| DL-3-42 | 42 | 150 | 5620 |
| DL-3-43 | 43 | 145 | 5765 |
| DL-3-44 | 44 | 165 | 5930 |
| DL-3-45 | 45 | 150 | 6080 |
| DL-3-46 | 46 | 140 | 6220 |
| DL-3-47 | 47 | 145 | 6365 |
| DL-3-48 | 48 | 145 | 6510 |
| DL-3-49 | 49 | 140 | 6650 |
| DL-3-50 | 50 | 140 | 6790 |
| DL-3-51 | 51 | 135 | 6925 |
| DL-3-52 | 52 | 145 | 7070 |
| DL-3-53 | 53 | 150 | 7220 |
| DL-3-54 | 54 | 160 | 7380 |
| DL-3-55 | 55 | 150 | 7530 |
| DL-3-56 | 56 | 145 | 7675 |
| DL-3-57 | 57 | 150 | 7825 |
| DL-3-58 | 58 | 150 | 7975 |
| DL-3-59 | 59 | 165 | 8140 |
| DL-3-60 | 60 | 145 | 8285 |
| DL-3-61 | 61 | 150 | 8435 |
| DL-3-62 | 62 | 160 | 8595 |
| DL-3-63 | 63 | 150 | 8745 |
| DL-3-64 | 64 | 145 | 8890 |
| DL-3-65 | 65 | 155 | 9045 |
| DL-3-66 | 66 | 140 | 9185 |
| DL-3-67 | 67 | 135 | 9320 |
| DL-3-68 | 68 | 140 | 9460 |
| DL-3-69 | 69 | 150 | 9610 |
| DL-3-70 | 70 | 160 | 9770 |
| DL-3-71 | 71 | 145 | 9915 |
| DL-3-72 | 72 | 150 | 10065 |

## A29 Volume of CT water flow through D3 column (low flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-3-73 | 73 | 145 | 10210 |
| DL-3-74 | 74 | 140 | 10350 |
| DL-3-75 | 75 | 100 | 10450 |
| DL-3-76 | 76 | 145 | 10595 |
| DL-3-77 | 77 | 160 | 10755 |
| DL-3-78 | 78 | 150 | 10905 |
| DL-3-79 | 79 | 150 | 11055 |
| DL-3-80 | 80 | 155 | 11210 |
| DL-3-81 | 81 | 160 | 11370 |
| DL-3-82 | 82 | 145 | 11515 |
| DL-3-83 | 83 | 150 | 11665 |
| DL-3-84 | 84 | 150 | 11815 |
| DL-3-85 | 85 | 145 | 11960 |
| DL-3-86 | 86 | 150 | 12110 |
| DL-3-87 | 87 | 150 | 12260 |
| DL-3-88 | 88 | 150 | 12410 |
| DL-3-89 | 89 | 155 | 12565 |
| DL-3-90 | 90 | 150 | 12715 |
| DL-3-91 | 91 | 160 | 12875 |
| DL-3-92 | 92 | 155 | 13030 |
| DL-3-93 | 93 | 160 | 13190 |
| DL-3-94 | 94 | 160 | 13350 |
| DL-3-95 | 95 | 150 | 13500 |
| DL-3-96 | 96 | 160 | 13660 |
| DL-3-97 | 97 | 150 | 13810 |
| DL-3-98 | 98 | 165 | 13975 |
| DL-3-99 | 99 | 155 | 14130 |
| DL-3-100 | 100 | 150 | 14280 |
| DL-3-101 | 101 | 160 | 14440 |
| DL-3-102 | 102 | 145 | 14585 |
| DL-3-103 | 103 | 150 | 14735 |
| DL-3-104 | 104 | 185 | 14920 |
| DL-3-105 | 105 | 160 | 15080 |
| DL-3-106 | 106 | 160 | 15240 |
| DL-3-107 | 107 | 165 | 15405 |
| DL-3-108 | 108 | 165 | 15570 |

A29 Volume of CT water flow through D3 column (low flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-3-109 | 109 | 165 | 15735 |
| DL-3-110 | 110 | 150 | 15885 |
| DL-3-111 | 111 | 145 | 16030 |
| DL-3-112 | 112 | 150 | 16180 |
| DL-3-113 | 113 | 150 | 16330 |
| DL-3-114 | 114 | 165 | 16495 |
| DL-3-115 | 115 | 160 | 16655 |
| DL-3-116 | 116 | 150 | 16805 |
| DL-3-117 | 117 | 170 | 16975 |
| DL-3-118 | 118 | 175 | 17150 |
| DL-3-119 | 119 | 165 | 17315 |
| DL-3-120 | 120 | 160 | 17475 |
| DL-3-121 | 121 | 165 | 17640 |
| DL-3-122 | 122 | 155 | 17795 |
| DL-3-123 | 123 | 150 | 17945 |
| DL-3-124 | 124 | 150 | 18095 |
| DL-3-125 | 125 | 160 | 18255 |
| DL-3-126 | 126 | 165 | 18420 |
| DL-3-127 | 127 | 155 | 18575 |
| DL-3-128 | 128 | 150 | 18725 |
| DL-3-129 | 129 | 145 | 18870 |
| DL-3-130 | 130 | 150 | 19020 |
| DL-3-131 | 131 | 160 | 19180 |
| DL-3-132 | 132 | 150 | 19330 |
| DL-3-133 | 133 | 155 | 19485 |
| DL-3-134 | 134 | 150 | 19635 |
| DL-3-135 | 135 | 150 | 19785 |
| DL-3-136 | 136 | 150 | 19935 |
| DL-3-137 | 137 | 155 | 20090 |
| DL-3-138 | 138 | 140 | 20230 |
| DL-3-139 | 139 | 140 | 20370 |
| DL-3-140 | 140 | 145 | 20515 |
| Flowrate | 3360 hrs | 20515 | $6.11 \mathrm{ml} / \mathrm{h}$ |



A30 Volume of CT water flow through D4 column (low flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-4-37 | 37 | 150 | 4980 |
| DL-4-38 | 38 | 155 | 5135 |
| DL-4-39 | 39 | 140 | 5275 |
| DL-4-40 | 40 | 160 | 5435 |
| DL-4-41 | 41 | 150 | 5585 |
| DL-4-42 | 42 | 145 | 5730 |
| DL-4-43 | 43 | 150 | 5880 |
| DL-4-44 | 44 | 150 | 6030 |
| DL-4-45 | 45 | 150 | 6180 |
| DL-4-46 | 46 | 140 | 6320 |
| DL-4-47 | 47 | 165 | 6485 |
| DL-4-48 | 48 | 160 | 6645 |
| DL-4-49 | 49 | 150 | 6795 |
| DL-4-50 | 50 | 165 | 6960 |
| DL-4-51 | 51 | 150 | 7110 |
| DL-4-52 | 52 | 155 | 7265 |
| DL-4-53 | 53 | 150 | 7415 |
| DL-4-54 | 54 | 145 | 7560 |
| DL-4-55 | 55 | 155 | 7715 |
| DL-4-56 | 56 | 150 | 7865 |
| DL-4-57 | 57 | 170 | 8035 |
| DL-4-58 | 58 | 165 | 8200 |
| DL-4-59 | 59 | 145 | 8345 |
| DL-4-60 | 60 | 165 | 8510 |
| DL-4-61 | 61 | 160 | 8670 |
| DL-4-62 | 62 | 150 | 8820 |
| DL-4-63 | 63 | 165 | 8985 |
| DL-4-64 | 64 | 140 | 9125 |
| DL-4-65 | 65 | 150 | 9275 |
| DL-4-66 | 66 | 145 | 9420 |
| DL-4-67 | 67 | 140 | 9560 |
| DL-4-68 | 68 | 150 | 9710 |
| DL-4-69 | 69 | 140 | 9850 |
| DL-4-70 | 70 | 145 | 9995 |
| DL-4-71 | 71 | 150 | 10145 |
| DL-4-72 | 72 | 160 | 10305 |

A30 Volume of CT water flow through D4 column (low flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-4-73 | 73 | 145 | 10450 |
| DL-4-74 | 74 | 160 | 10610 |
| DL-4-75 | 75 | 165 | 10775 |
| DL-4-76 | 76 | 160 | 10935 |
| DL-4-77 | 77 | 160 | 11095 |
| DL-4-78 | 78 | 160 | 11255 |
| DL-4-79 | 79 | 150 | 11405 |
| DL-4-80 | 80 | 145 | 11550 |
| DL-4-81 | 81 | 150 | 11700 |
| DL-4-82 | 82 | 160 | 11860 |
| DL-4-83 | 83 | 150 | 12010 |
| DL-4-84 | 84 | 150 | 12160 |
| DL-4-85 | 85 | 160 | 12320 |
| DL-4-86 | 86 | 160 | 12480 |
| DL-4-87 | 87 | 165 | 12645 |
| DL-4-88 | 88 | 160 | 12805 |
| DL-4-89 | 89 | 165 | 12970 |
| DL-4-90 | 90 | 150 | 13120 |
| DL-4-91 | 91 | 165 | 13285 |
| DL-4-92 | 92 | 145 | 13430 |
| DL-4-93 | 93 | 160 | 13590 |
| DL-4-94 | 94 | 165 | 13755 |
| DL-4-95 | 95 | 150 | 13905 |
| DL-4-96 | 96 | 145 | 14050 |
| DL-4-97 | 97 | 150 | 14200 |
| DL-4-98 | 98 | 155 | 14355 |
| DL-4-99 | 99 | 160 | 14515 |
| DL-4-100 | 100 | 155 | 14670 |
| DL-4-101 | 101 | 165 | 14835 |
| DL-4-102 | 102 | 160 | 14995 |
| DL-4-103 | 103 | 150 | 15145 |
| DL-4-104 | 104 | 160 | 15305 |
| DL-4-105 | 105 | 160 | 15465 |
| DL-4-106 | 106 | 155 | 15620 |
| DL-4-107 | 107 | 150 | 15770 |
| DL-4-108 | 108 | 155 | 15925 |

A30 Volume of CT water flow through D4 column (low flowrate) (cont'd)

| No. | Day | V (ml) | Sum-V (ml) |
| :---: | :---: | :---: | :---: |
| DL-4-109 | 109 | 150 | 16075 |
| DL-4-110 | 110 | 145 | 16220 |
| DL-4-111 | 111 | 140 | 16360 |
| DL-4-112 | 112 | 150 | 16510 |
| DL-4-113 | 113 | 160 | 16670 |
| DL-4-114 | 114 | 155 | 16825 |
| DL-4-115 | 115 | 160 | 16985 |
| DL-4-116 | 116 | 145 | 17130 |
| DL-4-117 | 117 | 165 | 17295 |
| DL-4-118 | 118 | 150 | 17445 |
| DL-4-119 | 119 | 155 | 17600 |
| DL-4-120 | 120 | 150 | 17750 |
| DL-4-121 | 121 | 165 | 17915 |
| DL-4-122 | 122 | 150 | 18065 |
| DL-4-123 | 123 | 145 | 18210 |
| DL-4-124 | 124 | 145 | 18355 |
| DL-4-125 | 125 | 155 | 18510 |
| DL-4-126 | 126 | 150 | 18660 |
| DL-4-127 | 127 | 160 | 18820 |
| DL-4-128 | 128 | 145 | 18965 |
| DL-4-129 | 129 | 155 | 19120 |
| DL-4-130 | 130 | 150 | 19270 |
| DL-4-131 | 131 | 160 | 19430 |
| DL-4-132 | 132 | 145 | 19575 |
| DL-4-133 | 133 | 150 | 19725 |
| DL-4-134 | 134 | 140 | 19865 |
| DL-4-135 | 135 | 150 | 20015 |
| DL-4-136 | 136 | 140 | 20155 |
| DL-4-137 | 137 | 155 | 20310 |
| DL-4-138 | 138 | 150 | 20460 |
| DL-4-139 | 139 | 140 | 20600 |
| DL-4-140 | 140 | 140 | 20740 |
| Flowrate | 3360 hrs | 20740 | $6.17 \mathrm{ml} / \mathrm{h}$ |


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## A31 Change of MFT height in DH system (cont'd)

| No | $\mathbf{t}_{\text {incu }}$ <br> $(\mathbf{d})$ | $\mathbf{H}$ <br> $(\mathbf{c m})$ | $\mathbf{N o}$ | $\mathbf{t}_{\text {incu }}$ <br> $(\mathbf{d})$ | $\mathbf{H}$ <br> $(\mathbf{c m})$ | $\mathbf{N o}$ | $\mathbf{t}_{\text {incu }}$ <br> $(\mathbf{d})$ | $\mathbf{H}$ <br> $(\mathbf{c m})$ | $\mathbf{N o}$ | $\mathbf{t}_{\text {incu }}$ <br> $(\mathbf{d})$ | $\mathbf{H}$ <br> $(\mathbf{c m})$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DH-1-45 | 45 | -0.8 | DH-2-45 | 45 | -0.1 | DH-3-45 | 45 | -2.5 | DH-4-45 | 45 | -1.8 |
| DH-1-46 | 46 | -0.7 | DH-2-46 | 46 | -0.1 | DH-3-46 | 46 | -2.4 | DH-4-46 | 46 | -1.5 |
| DH-1-47 | 47 | -0.8 | DH-2-47 | 47 | -0.2 | DH-3-47 | 47 | -2.3 | DH-4-47 | 47 | -1.4 |
| DH-1-48 | 48 | -0.7 | DH-2-48 | 48 | -0.1 | DH-3-48 | 48 | -2.1 | DH-4-48 | 48 | -1.3 |
| DH-1-49 | 49 | -1.1 | DH-2-49 | 49 | 0.1 | DH-3-49 | 49 | -2.0 | DH-4-49 | 49 | -1.3 |
| DH-1-50 | 50 | -1.0 | DH-2-50 | 50 | -0.5 | DH-3-50 | 50 | -2.0 | DH-4-50 | 50 | 1.1 |
| DH-1-51 | 51 | -1.0 | DH-2-51 | 51 | -0.3 | DH-3-51 | 51 | -2.0 | DH-4-51 | 51 | -1.0 |
| DH-1-52 | 52 | -0.7 | DH-2-52 | 52 | -0.1 | DH-3-52 | 52 | -2.7 | DH-4-52 | 52 | -1.0 |
| DH-1-53 | 53 | -0.7 | DH-2-53 | 53 | 0.0 | DH-3-53 | 53 | -2.7 | DH-4-53 | 53 | -1.0 |
| DH-1-54 | 54 | -0.9 | DH-2-54 | 54 | -0.2 | DH-3-54 | 54 | -2.7 | DH-4-54 | 54 | -1.0 |
| DH-1-55 | 55 | -0.3 | DH-2-55 | 55 | -0.1 | DH-3-55 | 55 | -2.6 | DH-4-55 | 55 | -1.0 |
| DH-1-56 | 56 | -0.9 | DH-2-56 | 56 | -0.1 | DH-3-56 | 56 | -2.5 | DH-4-56 | 56 | -1.0 |
| DH-1-57 | 57 | -0.6 | DH-2-57 | 57 | -0.2 | DH-3-57 | 57 | 2.1 | DH-4-57 | 57 | -0.9 |
| DH-1-58 | 58 | -0.5 | DH-2-58 | 58 | -0.2 | DH-3-58 | 58 | -2.0 | DH-4-58 | 58 | -1.0 |
| DH-1-59 | 59 | -1.2 | DH-2-59 | 59 | -0.2 | DH-3-59 | 59 | -2.0 | DH-4-59 | 59 | -1.0 |
| DH-1-60 | 60 | -1.5 | DH-2-60 | 60 | 0.3 | DH-3-60 | 60 | -2.0 | DH-4-60 | 60 | -1.0 |

Note: when $\mathrm{t}=0$, the height of MFT $=0$

| No | $\mathbf{t}_{\text {incu }}$ <br> (d) | $\begin{gathered} \mathrm{H} \\ (\mathrm{~cm}) \\ \hline \end{gathered}$ | No | $\mathbf{t}_{\text {incu }}$ <br> (d) | $\begin{gathered} \mathbf{H} \\ (\mathrm{cm}) \end{gathered}$ | No | $\mathbf{t}_{\text {incu }}$ <br> (d) | $\begin{gathered} \mathrm{H} \\ (\mathrm{~cm}) \end{gathered}$ | No | $\mathrm{t}_{\text {incu }}$ <br> (d) | $\begin{gathered} \mathrm{H} \\ (\mathrm{~cm}) \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-1-1 | 1 | 0.5 | DL-2-1 | 1 | 0.0 | DL-3-1 | 1 | -0.1 | DL-4-1 | 1 | 0.0 |
| DL-1-2 | 2 | 1.5 | DL-2-2 | 2 | -0.4 | DL-3-2 | 2 | -0.2 | DL-4-2 | 2 | 0.1 |
| DL-1-3 | 3 | 1.6 | DL-2-3 | 3 | -0.9 | DL-3-3 | 3 | 0.0 | DL-4-3 | 3 | 0.4 |
| DL-1-4 | 4 | 1.8 | DL-2-4 | 4 | -1.9 | DL-3-4 | 4 | 0.1 | DL-4-4 | 4 | -0.6 |
| DL-1-5 | 5 | 1.4 | DL-2-5 | 5 | -2.0 | DL-3-5 | 5 | 0.2 | DL-4-5 | 5 | -0.7 |
| DL-1-6 | 6 | 1.4 | DL-2-6 | 6 | -2.1 | DL-3-6 | 6 | 1.1 | DL-4-6 | 6 | -2.0 |
| DL-1-7 | 7 | 1.4 | DL-2-7 | 7 | -0.9 | DL-3-7 | 7 | 0.2 | DL-4-7 | 7 | -2.4 |
| DL-1-8 | 8 | 1.5 | DL-2-8 | 8 | -0.8 | DL-3-8 | 8 | 0.0 | DL-4-8 | 8 | -2.5 |
| DL-1-9 | 9 | 1.7 | DL-2-9 | 9 | -0.6 | DL-3-9 | 9 | -0.3 | DL-4-9 | 9 | -2.2 |
| DL-1-10 | 10 | 1.9 | DL-2-10 | 10 | -0.5 | DL-3-10 | 10 | -0.3 | DL-4-10 | 10 | -2.3 |
| DL-1-11 | 11 | 1.8 | DL-2-11 | 11 | -0.8 | DL-3-11 | 11 | -0.4 | DL-4-11 | 11 | -2.0 |
| DL-1-12 | 12 | 1.8 | DL-2-12 | 12 | -0.4 | DL-3-12 | 12 | -0.5 | DL-4-12 | 12 | -1.6 |
| DL-1-13 | 13 | 1.6 | DL-2-13 | 13 | 0.0 | DL-3-13 | 13 | 0.0 | DL-4-13 | 13 | -1.7 |
| DL-1-14 | 14 | 1.9 | DL-2-14 | 14 | -0.4 | DL-3-14 | 14 | 0.0 | DL-4-14 | 14 | -1.7 |
| DL-1-15 | 15 | 1.8 | DL-2-15 | 15 | -0.4 | DL-3-15 | 15 | -0.1 | DL-4-15 | 15 | -1.6 |
| DL-1-16 | 16 | 1.8 | DL-2-16 | 16 | 0.3 | DL-3-16 | 16 | -0.2 | DL-4-16 | 16 | -1.6 |
| DL-1-17 | 17 | 1.7 | DL-2-17 | 17 | 0.7 | DL-3-17 | 17 | 0.4 | DL-4-17 | 17 | -1.6 |
| DL-1-18 | 18 | 1.6 | DL-2-18 | 18 | 1.0 | DL-3-18 | 18 | -0.6 | DL-4-18 | 18 | -1.6 |
| DL-1-19 | 19 | 1.9 | DL-2-19 | 19 | -4.0 | DL-3-19 | 19 | -0.5 | DL-4-19 | 19 | -1.6 |
| DL-1-20 | 20 | 1.7 | DL-2-20 | 20 | -4.5 | DL-3-20 | 20 | 0.7 | DL-4-20 | 20 | -1.5 |
| DL-1-21 | 21 | 1.6 | DL-2-21 | 21 | -3.0 | DL-3-21 | 21 | -1.0 | DL-4-21 | 21 | -1.5 |
| DL-1-22 | 22 | 1.5 | DL-2-22 | 22 | -2.0 | DL-3-22 | 22 | -0.8 | DL-4-22 | 22 | -2.0 |
| DL-1-23 | 23 | 1.4 | DL-2-23 | 23 | -2.4 | DL-3-23 | 23 | -0.5 | DL-4-23 | 23 | -2.6 |
| DL-1-24 | 24 | 1.4 | DL-2-24 | 24 | 2.0 | DL-3-24 | 24 | -0.3 | DL-4-24 | 24 | -2.0 |
| DL-1-25 | 25 | 1.3 | DL-2-25 | 25 | 1.8 | DL-3-25 | 25 | -0.2 | DL-4-25 | 25 | 1.8 |
| DL-1-26 | 26 | 1.9 | DL-2-26 | 26 | -2.0 | DL-3-26 | 26 | 0.2 | DL-4-26 | 26 | -1.8 |
| DL-1-27 | 27 | 1.2 | DL-2-27 | 27 | -2.1 | DL-3-27 | 27 | -0.4 | DL-4-27 | 27 | -2.2 |
| DL-1-28 | 28 | 1.1 | DL-2-28 | 28 | -2.0 | DL-3-28 | 28 | 0.6 | DL-4-28 | 28 | -2.8 |
| DL-1-29 | 29 | 0.9 | DL-2-29 | 29 | -2.0 | DL-3-29 | 29 | -0.9 | DL-4-29 | 29 | -2.9 |
| DL-1-30 | 30 | 0.6 | DL-2-30 | 30 | -1.8 | DL-3-30 | 30 | -1.0 | DL-4-30 | 30 | -3.0 |
| DL-1-31 | 31 | 0.5 | DL-2-31 | 31 | -2.0 | DL-3-31 | 31 | -1.0 | DL-4-31 | 31 | -3.0 |
| DL-1-32 | 32 | 0.3 | DL-2-32 | 32 | 2.0 | DL-3-32 | 32 | -1.2 | DL-4-32 | 32 | -3.0 |
| DL-1-33 | 33 | 0.1 | DL-2-33 | 33 | -1.7 | DL-3-33 | 33 | 1.2 | DL-4-33 | 33 | -3.0 |
| DL-1-34 | 34 | 0.5 | DL-2-34 | 34 | -2.0 | DL-3-34 | 34 | -1.3 | DL-4-34 | 34 | -3.3 |
| DL-1-35 | 35 | 0.4 | DL-2-35 | 35 | -2.0 | DL-3-35 | 35 | -1.5 | DL-4-35 | 35 | -2.8 |
| DL-1-36 | 36 | 0.5 | DL-2-36 | 36 | -2.0 | DL-3-36 | 36 | -1.0 | DL-4-36 | 36 | -2.5 |
| DL-1-37 | 37 | 0.4 | DL-2-37 | 37 | -1.5 | DL-3-37 | 37 | -0.5 | DL-4-37 | 37 | -2.9 |
| DL-1-38 | 38 | 0.3 | DL-2-38 | 38 | -2.0 | DL-3-38 | 38 | -0.9 | DL-4-38 | 38 | -3.0 |
| DL-1-39 | 39 | 0.3 | DL-2-39 | 39 | -1.8 | DL-3-39 | 39 | -1.1 | DL-4-39 | 39 | -3.0 |
| DL-1-40 | 40 | 0.3 | DL-2-40 | 40 | -1.6 | DL-3-40 | 40 | -1.3 | DL-4-40 | 40 | -3.0 |
| DL-1-41 | 41 | 0.0 | DL-2-41 | 41 | -1.8 | DL-3-41 | 41 | -1.6 | DL-4-41 | 41 | -3.2 |
| DL-1-42 | 42 | 0.0 | DL-2-42 | 42 | -1.8 | DL-3-42 | 42 | -1.7 | DL-4-42 | 42 | -3.5 |
| DL-1-43 | 43 | 0.0 | DL-2-43 | 43 | -1.0 | DL-3-43 | 43 | -1.8 | DL-4-43 | 43 | -3.4 |
| DL-1-44 | 44 | 0.0 | DL-2-44 | 44 | -0.6 | DL-3-44 | 44 | -2.0 | DL-4-44 | 44 | -3.3 |
| DL-1-45 | 45 | -0.1 | DL-2-45 | 45 | 0.4 | DL-3-45 | 45 | -2.1 | DL-4-45 | 45 | -2.3 |



A32 Change of MFT height in DL system (cont'd)

| No | $t_{\text {incu }}$ <br> (d) | $\begin{gathered} \mathbf{H} \\ (\mathrm{cm}) \end{gathered}$ | No | $t_{\text {incu }}$ <br> (d) | $\begin{gathered} \mathbf{H} \\ (\mathrm{cm}) \end{gathered}$ | No | $\mathbf{t}_{\text {incu }}$ <br> (d) | $\begin{gathered} \mathbf{H} \\ (\mathrm{cm}) \end{gathered}$ | No | $t_{\text {incu }}$ <br> (d) | $\begin{gathered} \mathrm{H} \\ (\mathrm{~cm}) \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-1-91 | 91 | 1.5 | DL-2-91 | 91 | -2.5 | DL-3-91 | 91 | -1.1 | DL-4-91 | 91 | -3.0 |
| DL-1-92 | 92 | 1.7 | DL-2-92 | 92 | -2.5 | DL-3-92 | 92 | -1.0 | DL-4-92 | 92 | -3.2 |
| DL-1-93 | 93 | 2.0 | DL-2-93 | 93 | 2.1 | DL-3-93 | 93 | -1.1 | DL-4-93 | 93 | -3.3 |
| DL-1-94 | 94 | 2.0 | DL-2-94 | 94 | 4.1 | DL-3-94 | 94 | -1.2 | DL-4-94 | 94 | -3.3 |
| DL-1-95 | 95 | 2.0 | DL-2-95 | 95 | 4.5 | DL-3-95 | 95 | -1.8 | DL-4-95 | 95 | -3.4 |
| DL-1-96 | 96 | 2.1 | DL-2-96 | 96 | 6.5 | DL-3-96 | 96 | -2.1 | DL-4-96 | 96 | -3.6 |
| DL-1-97 | 97 | 2.5 | DL-2-97 | 97 | 6.4 | DL-3-97 | 97 | -2.4 | DL-4-97 | 97 | -3.7 |
| DL-1-98 | 98 | 1.5 | DL-2-98 | 98 | 6.4 | DL-3-98 | 98 | -2.5 | DL-4-98 | 98 | -3.8 |
| DL-1-99 | 99 | 2.0 | DL-2-99 | 99 | 6.5 | DL-3-99 | 99 | -2.6 | DL-4-99 | 99 | -2.2 |
| DL-1-100 | 100 | -0.4 | DL-2-100 | 100 | 5.8 | DL-3-100 | 100 | -1.7 | DL-4-100 | 100 | -2.3 |
| DL-1-101 | 101 | 0.6 | DL-2-101 | 101 | 5.6 | DL-3-101 | 101 | -1.6 | DL-4-101 | 101 | -2.2 |
| DL-1-102 | 102 | 1.2 | DL-2-102 | 102 | 5.5 | DL-3-102 | 102 | -1.7 | DL-4-102 | 102 | -2.1 |
| DL-1-103 | 103 | 1.0 | DL-2-103 | 103 | 5.4 | DL-3-103 | 103 | -1.8 | DL-4-103 | 103 | -2.5 |
| DL-1-104 | 104 | 0.9 | DL-2-104 | 104 | 5.3 | DL-3-104 | 104 | -1.9 | DL-4-104 | 104 | -3.0 |
| DL-1-105 | 105 | 0.8 | DL-2-105 | 105 | 5.3 | DL-3-105 | 105 | -2.0 | DL-4-105 | 105 | -3.4 |
| DL-1-106 | 106 | 0.7 | DL-2-106 | 106 | 5.3 | DL-3-106 | 106 | -2.1 | DL-4-106 | 106 | -3.4 |
| DL-1-107 | 107 | 0.7 | DL-2-107 | 107 | 5.5 | DL-3-107 | 107 | -2.2 | DL-4-107 | 107 | -3.5 |
| DL-1-108 | 108 | 0.5 | DL-2-108 | 108 | 5.5 | DL-3-108 | 108 | -2.5 | DL-4-108 | 108 | -3.4 |
| DL-1-109 | 109 | 0.5 | DL-2-109 | 109 | 5.4 | DL-3-109 | 109 | -2.7 | DL-4-109 | 109 | -3.2 |
| DL-1-110 | 110 | 0.4 | DL-2-110 | 110 | 5.5 | DL-3-110 | 110 | -2.5 | DL-4-110 | 110 | -2.5 |
| DL-1-111 | 111 | 0.3 | DL-2-111 | 111 | 5.5 | DL-3-111 | 111 | -2.4 | DL-4-111 | 111 | 2.0 |
| DL-1-112 | 112 | 0.3 | DL-2-112 | 112 | 6.2 | DL-3-112 | 112 | -2.4 | DL-4-112 | 112 | -1.8 |
| DL-1-113 | 113 | 0.3 | DL-2-113 | 113 | 6.3 | DL-3-113 | 113 | -2.5 | DL-4-113 | 113 | -1.6 |
| DL-1-114 | 114 | 0.5 | DL-2-114 | 114 | 6.2 | DL-3-114 | 114 | -2.6 | DL-4-114 | 114 | -1.6 |
| DL-1-115 | 115 | 0.3 | DL-2-115 | 115 | 6.3 | DL-3-115 | 115 | -2.5 | DL-4-115 | 115 | -1.5 |
| DL-1-116 | 116 | 0.0 | DL-2-116 | 116 | 6.3 | DL-3-116 | 116 | -2.6 | DL-4-116 | 116 | 1.5 |
| DL-1-117 | 117 | 0.0 | DL-2-117 | 117 | 6.2 | DL-3-117 | 117 | -2.2 | DL-4-117 | 117 | -1.4 |
| DL-1-118 | 118 | 0.0 | DL-2-118 | 118 | 6.1 | DL-3-118 | 118 | -2.0 | DL-4-118 | 118 | -1.4 |
| DL-1-119 | 119 | 0.0 | DL-2-119 | 119 | 6.1 | DL-3-119 | 119 | -2.0 | DL-4-119 | 119 | -1.4 |
| DL-1-120 | 120 | 1.2 | DL-2-120 | 120 | 6.2 | DL-3-120 | 120 | -2.0 | DL-4-120 | 120 | -1.3 |
| DL-1-121 | 121 | 0.0 | DL-2-121 | 121 | 6.2 | DL-3-121 | 121 | -2.0 | DL-4-121 | 121 | -1.4 |
| DL-1-122 | 122 | -0.1 | DL-2-122 | 122 | 6.2 | DL-3-122 | 122 | -2.0 | DL-4-122 | 122 | -1.8 |
| DL-1-123 | 123 | -0.2 | DL-2-123 | 123 | 6.2 | DL-3-123 | 123 | -2.0 | DL-4-123 | 123 | -2.0 |
| DL-1-124 | 124 | -0.2 | DL-2-124 | 124 | 6.2 | DL-3-124 | 124 | -2.0 | DL-4-124 | 124 | -3.5 |
| DL-1-125 | 125 | 0.3 | DL-2-125 | 125 | 6.2 | DL-3-125 | 125 | -1.9 | DL-4-125 | 125 | -3.6 |
| DL-1-126 | 126 | 0.5 | DL-2-126 | 126 | 6.2 | DL-3-126 | 126 | -1.8 | DL-4-126 | 126 | -4.0 |
| DL-1-127 | 127 | 0.0 | DL-2-127 | 127 | 6.2 | DL-3-127 | 127 | -1.8 | DL-4-127 | 127 | -4.5 |
| DL-1-128 | 128 | 0.0 | DL-2-128 | 128 | 6.2 | DL-3-128 | 128 | -1.8 | DL-4-128 | 128 | -4.7 |
| DL-1-129 | 129 | 0.0 | DL-2-129 | 129 | 6.2 | DL-3-129 | 129 | -2.0 | DL-4-129 | 129 | -4.5 |
| DL-1-130 | 130 | 0.0 | DL-2-130 | 130 | 6.2 | DL-3-130 | 130 | -2.5 | DL-4-130 | 130 | -4.5 |
| DL-1-131 | 131 | 0.0 | DL-2-131 | 131 | 6.2 | DL-3-131 | 131 | -2.8 | DL-4-131 | 131 | -4.8 |
| DL-1-132 | 132 | 0.0 | DL-2-132 | 132 | -2.1 | DL-3-132 | 132 | -2.9 | DL-4-132 | 132 | -5.0 |
| DL-1-133 | 133 | 0.0 | DL-2-133 | 133 | -2.1 | DL-3-133 | 133 | -3.2 | DL-4-133 | 133 | -5.0 |
| DL-1-134 | 134 | 0.0 | DL-2-134 | 134 | -2.1 | DL-3-134 | 134 | -2.5 | DL-4-134 | 134 | -5.0 |
| DL-1-135 | 135 | 0.0 | DL-2-135 | 135 | -2.1 | DL-3-135 | 135 | -2.1 | DL-4-135 | 135 | -5.0 |

## A32 Change of MFT height in DL system (cont'd)

| No | $\mathbf{t}_{\text {incu }}$ <br> $(\mathbf{d})$ | $\mathbf{H}$ <br> $(\mathbf{c m})$ | $\mathbf{N o}$ | $\mathbf{t}_{\text {incu }}$ <br> $(\mathbf{d})$ | $\mathbf{H}$ <br> $(\mathbf{c m})$ | No | $\mathbf{t}_{\text {incu }}$ <br> $(\mathbf{d})$ | $\mathbf{H}$ <br> $(\mathbf{c m})$ | No | $\mathbf{t}_{\text {incu }}$ <br> $(\mathbf{d})$ | $\mathbf{H}$ <br> $(\mathbf{c m})$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| DL-1-136 | 136 | 0.0 | DL-2-136 | 136 | -2.1 | DL-3-136 | 136 | -2.4 | DL-4-136 | 136 | -5.0 |
| DL-1-137 | 137 | 0.0 | DL-2-137 | 137 | -2.1 | DL-3-137 | 137 | -2.6 | DL-4-137 | 137 | -5.0 |
| DL-1-138 | 138 | 0.0 | DL-2-138 | 138 | -2.1 | DL-3-138 | 138 | -2.7 | DL-4-138 | 138 | -5.0 |
| DL-1-139 | 139 | 0.0 | DL-2-139 | 139 | -2.1 | DL-3-139 | 139 | -2.5 | DL-4-139 | 139 | -5.0 |
| DL-1-140 | 140 | 0.0 | DL-2-140 | 140 | -2.8 | DL-3-140 | 140 | -2.5 | DL-4-140 | 140 | -5.0 |

Note: when $\mathrm{t}=0$, the height of MFT $=0$

# Appendix B Jar Test and Static Barrel Mesocosms Results 

## B1 Results of chemical analysis in barrel and C1 system

| System | Sample ID | Sample Type | Incub. Time (y) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{2-}$ | $\begin{gathered} \mathrm{CO}_{3}^{2-} \\ (\mathrm{wt} \mathrm{ppm}) \end{gathered}$ | $\begin{array}{\|} \mathbf{H C O}_{3}^{-} \\ \text {(wt ppm) } \end{array}$ | Total Cations | Total Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \end{gathered}$ | Alkalinity expressed as $\mathrm{HCO}_{3}^{-}$ | ALK pH | $\mathbf{S}^{\mathbf{2 -}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Barrel 1 | B1-TOP | RelW | 2 | 7.99 | 5700 | 170 | 1340 | 18.1 | 21.1 | 35.8 | 1100 | 445 | 0.0 | 1400 | 62.3 | 65.3 | 0.95 | 1148 | 8.3 | 160 |
| Barrel 1 | B1-MFT-U | PoreW | 2 | 8.20 | 5040 | 230 | 1130 | 13.6 | 11.0 | 14.9 | 1070 | 96.3 | 0.0 | 1260 | 51.1 | 55.7 | 0.92 | 1033 | 8.0 | 27.3 |
| Barrel 1 | B1-CT-U | PoreW | 2 | 8.15 | 4860 | 210 | 1170 | 14.5 | 10.1 | 17.9 | 1100 | 135 | 0.0 | 1130 | 53.0 | 54.9 | 0.96 | 926 | 7.9 | 132 |
| Barrel 1 | B1-CT-L | PoreW | 2 | 8.28 | 4870 | 270 | 1190 | 15.1 | 9.9 | 17.4 | 1200 | 161 | 0.0 | 1080 | 53.8 | 58.2 | 0.92 | 885 | 7.8 | 106 |
| Barrel 2 | B2-TOP | RelW | 2 | 8.10 | 5480 | 180 | 1280 | 17.0 | 19.0 | 26.5 | 1000 | 358 | 21.6 | 1340 | 59.0 | 60.6 | 0.97 | 1134 | 8.4 | 131 |
| Barrel 2 | B2-MFT-U | PoreW | 2 | 8.21 | 7830 | 210 | 1140 | 15.4 | 10.6 | 13.3 | 1100 | 194 | 0.0 | 992 | 51.5 | 53.9 | 0.96 | 813 | 8.0 | 65.1 |
| Barrel 2 | B2-MFT-L | Porew | 2 | 8.20 | 7800 | 210 | 1120 | 14.1 | 10.1 | 13.3 | 1100 | 148 | 0.0 | 1010 | 50.6 | 53.3 | 0.95 | 828 | 8.1 | 48.5 |
| Barrel 2 | B2-CT-U | Porew | 2 | 8.19 | 4890 | 210 | 1140 | 13.8 | 10.2 | 18.5 | 1100 | 167 | 0.0 | 1050 | 51.7 | 54.3 | 0.95 | 861 | 7.9 | 80.3 |
| Barrel 2 | B2-CT-L | PoreW | 2 | 8.15 | 4810 | 210 | 1130 | 14.7 | 10.0 | 18.5 | 1100 | 171 | 0.0 | 1000 | 51.3 | 53.6 | 0.96 | 820 | 7.8 | 71.7 |
| Barrel 3 | B3-TOP | RelW | 2 | 8.23 | 4580 | 0 | 1080 | 15.3 | 13.7 | 18.2 | 900 | 8.0 | 40.2 | 1470 | 49.4 | 51.0 | 0.97 | 1272 | 8.5 | 7.2 |
| Barrel 3 | B3-MFT-U | PoreW | 2 | 8.09 | 4220 | 6.3 | 1030 | 14.8 | 9.8 | 12.1 | 1000 | 13.1 | 0.0 | 1220 | 46.6 | 48.5 | 0.96 | 1000 | 8.1 | 6.6 |
| Barrel 3 | B3-MFT-L | PoreW | 2 | 8.11 | 4220 | 0.0 | 993 | 14.1 | 9.7 | 12.5 | 1000 | 6.3 | 0.0 | 1240 | 45.0 | 48.6 | 0.92 | 1016 | 8.0 | 5.9 |
| Barrel 4 | B4-TOP | RelW | 2 | 8.24 | 6100 | 270 | 1370 | 16.9 | 18.3 | 25.5 | 1100 | 853 | 13.5 | 838 | 62.8 | 66.3 | 0.95 | 709 | 8.4 | 305 |
| Barrel 4 | B4-CT-U | PoreW | 2 | 8.25 | 5580 | 330 | 1320 | 14.8 | 13.2 | 23.3 | 1200 | 665 | 0.0 | 798 | 60.0 | 64.9 | 0.93 | 654 | 7.9 | 221 |
| Barrel 4 | B4-CT-L | PoreW | 2 | 8.27 | 5460 | 340 | 1300 | 14.0 | 11.3 | 20.2 | 1200 | 524 | 0.0 | 861 | 58.8 | 63.1 | 0.93 | 706 | 7.9 | 194 |
| C1 | C1-17-Top | PoreW | 2 | 7.54 | 5470 | 130 | 1160 | 15.1 | 20.3 | 31.0 | 950 | 137 | 0.0 | 1630 | 54.1 | 58.0 | 0.93 | 1336 | 7.6 | 49.7 |
| C1 | C1-17-U | Relw | 2 | 8.17 | 6730 | 180 | 1340 | 18.6 | 21.3 | 23.5 | 1200 | 104 | 0.0 | 1790 | 61.7 | 67.6 | 0.91 | 1467 | 7.7 | 37.8 |

B1 Results of chemical analysis in barrel and C1 system (cont'd)

| System | Sample ID | Sample Type | Incub. Time (y) | pH | Cond | $\mathrm{Br}^{+}$ | $\mathrm{Na}^{+}$ | $\mathbf{K}^{+}$ | $\mathbf{M g}{ }^{\mathbf{2 +}}$ | $\mathrm{Ca}^{2+}$ | $\mathrm{Cl}^{-}$ | $\mathrm{SO}_{4}{ }^{\text {2- }}$ | $\begin{gathered} \mathrm{CO}_{3}{ }^{2-} \\ (\mathrm{wt} \mathrm{ppm}) \end{gathered}$ | $\begin{gathered} \mathrm{HCO}_{3}{ }^{-} \\ \text {(wt ppm) } \end{gathered}$ | Total Cations | Total Anions | $\begin{gathered} \text { Ratio } \\ \text { Cat/Ani } \\ \hline \end{gathered}$ | Alkalinity expressed as $\mathrm{HCO}_{3}^{-}$ | $\begin{gathered} \text { ALK } \\ \text { pH } \\ \hline \end{gathered}$ | $\mathrm{S}^{2-}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| C1 | C1-17-M-U | PoreW | 2 | 8.14 | 5270 | 150 | 1090 | 15.7 | 12.6 | 17.0 | 1000 | 15.9 | 0.0 | 1790 | 49.7 | 59.7 | 0.83 | 1467 | 7.6 | 8.6 |
| C1 | C1-17-M-L | PoreW | 2 | 8.36 | 5450 | 190 | 1130 | 13.9 | 12.4 | 18.5 | 1100 | 72.8 | 0.0 | 1290 | 51.4 | 56.0 | 0.92 | 1057 | 7.7 | 26.4 |
| C1 | C1-17-L | PoreW | 2 | 8.23 | 5500 | 210 | 1240 | 15.0 | 13.2 | 22.3 | 1100 | 52.4 | 0.0 | 1540 | 56.5 | 59.9 | 0.94 | 1262 | 7.5 | 22.3 |
| C1 | C1-18-Top | PoreW | 2 | 7.64 | 5510 | 150 | 1250 | 16.2 | 21.2 | 32.6 | 960 | 132 | 0.0 | 1640 | 58.2 | 58.6 | 0.99 | 1344 | 7.6 | 50.6 |
| Cl | C1-18-U | PoreW | 2 | 8.14 | 6140 | 180 | 1380 | 18.5 | 20.5 | 25.1 | 1200 | 67.9 | 0.0 | 1690 | 63.4 | 65.2 | 0.97 | 1385 | 7.6 | 27.6 |
| Cl | C1-18-M-U | PoreW | 2 | 8.12 | 5330 | 180 | 1180 | 16.1 | 14.0 | 19.8 | 1000 | 15.2 | 0.0 | 1440 | 53.9 | 54.3 | 0.99 | 1180 | 8.2 | 8.3 |
| Cl | C1-18-M-L | PoreW | 2 | 8.35 | 5370 | 210 | 1200 | 14.6 | 11.5 | 17.0 | 1100 | 124 | 0.0 | 1130 | 54.4 | 54.7 | 0.99 | 926 | 7.6 | 52.9 |
| CI | C1-18-L | PoreW | 2 | 8.32 | 5790 | 250 | 1240 | 14.4 | 13.7 | 21.2 | 1100 | 85.8 | 0.0 | 1310 | 56.5 | 57.4 | 0.98 | 1074 | 7.5 | 35.8 |

B2 MPN data of barrels \& C1 system

| System | Sample | $\mathbf{t}(\mathrm{d})$ | Methanogen MPN/g DRY WT | SRB MPN/g DRY WT |
| :---: | :---: | :---: | :---: | :---: |
| 5 | B1-MFT | 750 | $1.94 \mathrm{E}+04$ | $3.82 \mathrm{E}+04$ |
|  | B2-MFT | 750 | $4.17 \mathrm{E}+04$ | $1.55 \mathrm{E}+05$ |
|  | B1-CT | 750 | $7.18 \mathrm{E}+02$ | $2.09 \mathrm{E}+03$ |
|  | B2-CT | 750 | $7.26 \mathrm{E}+03$ | $4.14 \mathrm{E}+02$ |
| 6 | B3-MFT | 750 | $1.83 \mathrm{E}+04$ | $3.96 \mathrm{E}+04$ |
|  | B4-CT | 750 | $4.79 \mathrm{E}+03$ | $1.05 \mathrm{E}+02$ |
| C1 | C1-17-U | 910 | $2.41 \mathrm{E}+04$ | $6.28 \mathrm{E}+04$ |
|  | C1-17-M-U | 910 | $8.81 \mathrm{E}+03$ | $4.44 \mathrm{E}+02$ |
|  | C1-17-M-L | 910 | $5.76 \mathrm{E}+02$ | $3.53 \mathrm{E}+01$ |
|  | C1-17-L | 910 | $3.38 \mathrm{E}+02$ | $5.03 \mathrm{E}+02$ |
|  | C1-18-U | 910 | $4.13 \mathrm{E}+04$ | $2.07 \mathrm{E}+04$ |
|  | C1-18-M-U | 910 | $5.44 \mathrm{E}+03$ | $4.43 \mathrm{E}+02$ |
|  | C1-18-M-L | 910 | $1.83 \mathrm{E}+03$ | $1.94 \mathrm{E}+01$ |
|  | C1-18-L | 910 | $2.00 \mathrm{E}+03$ | $1.81 \mathrm{E}+02$ |

B3 Results of $\mathbf{S}^{\mathbf{2 -}}$ analysis in barrels $\boldsymbol{\&} \mathbf{C} 1$ system

| ID | $\mathbf{S}^{2-}$ | ID | $\mathbf{s}^{\mathbf{2}^{-}}$ |
| :---: | :---: | :---: | :---: |
| B1-MFT-U | BDL | C1-17-U | BDL |
| B1-MFT-L | BDL | C1-17-M-U | BDL |
| B1-CT-U | BDL | C1-17-M-L | BDL |
| B1-CT-L | BDL | C1-17-L | BDL |
| B2-MFT-U | BDL | C1-18-U | BDL |
| B2-MFT-L | BDL | C1-18-M-U | BDL |
| B2-CT-U | BDL | C1-18-M-L | BDL |
| B2-CT-L | BDL | C1-18-L | BDL |
| B3-MFT-U | BDL |  |  |
| B3-MFT-L | BDL |  |  |
| B4-CT-U | BDL |  |  |
| B4-CT-L | BDL |  |  |

B4 Results of Redox potential analysis in barrels

| ID | Eh (mV) |  | ID | Eh (mV) |  | 1 D | Eh (mV) |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  | 1 hr | 2 hr |  | 1 hr | 2 hr |  | 1 hr | 2 hr |
| B1-MFT-U | 174 | 96 | B2-MFT-U | 297 | 199 | B3-MFT-U | 84 | 57 |
| B1-MFT-L | 189 | 116 | B2-MFT-L | 185 | 116 | B3-MFT-L | 54 | 34 |
| B1-CT-U | 337 | 253 | B2-CT-U | 324 | 245 | B4-CT-U | 449 | 238 |
| B1-CT-L | 222 | 153 | B2-CT-L | 226 | 136 | B4-CT-L | 334 | 257 |

B5 Results of Redox potential analysis in C1 system

| ID | Eh (mV) |  | ID | Eh (mV) |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  | $\mathbf{1 ~ h r}$ | $\mathbf{2 ~ h r}$ |  | $\mathbf{1} \mathbf{~ h r}$ | $\mathbf{2 ~ h r}$ |
| C1-17-U | 89 | 50 | C1-18-U | 105 | 53 |
| C1-17-M-U | 87 | 52 | C1-18-M-U | -1 | -39 |
| C1-17-M-L | 290 | 215 | C1-18-M-L | 280 | 230 |
| C1-17-L | 165 | 110 | C1-18-L | 61 | 48 |

B6 Solids content of barrels $\boldsymbol{\&} \mathbf{C} 1$ system

| System | Sample ID | Solids <br> $(\mathrm{g} / \mathbf{1 0 0 g})$ | Bitumen <br> $(\mathrm{g} / \mathbf{1 0 0 g})$ | $\mathbf{2 2} \boldsymbol{\mu \mathrm { m }}$ | $\mathbf{1 1} \boldsymbol{\mu \mathrm { m }}$ | $\mathbf{5 . 5 \mu \mathrm { m }}$ | $\mathbf{2 . 8} \boldsymbol{\mu \mathrm { m }}$ | $\mathbf{1 . 4} \boldsymbol{\mu \mathrm { m }}$ | $\mathbf{0 . 5} \boldsymbol{\mu \mathrm { m }}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Barrel 1 | B1-MFT-U | 43.5 | 3.6 | 75.3 | 61.0 | 44.9 | 28.7 | 12.9 | 1.7 |
| Barrel 1 | B1-MFT-L | 51.5 | 3.0 | 67.4 | 55.1 | 40.5 | 25.0 | 10.7 | 1.3 |
| Barrel 1 | B1-CT-U | 76.5 | 0.60 | 44.8 | 37.7 | 26.8 | 15.1 | 6.7 | 1.9 |
| Barrel 1 | B1-CT-L | 76.8 | 1.36 | 40.6 | 33.2 | 24.6 | 15.6 | 7.1 | 1.2 |
| Barrel 2 | B2-MFT-U | 53.4 | 3.70 | 79.0 | 66.2 | 47.8 | 27.3 | 11.2 | 2.0 |
| Barrel 2 | B2-MFT-L | 53.3 | 3.18 | 74.6 | 62.6 | 45.9 | 26.8 | 11.4 | 2.1 |
| Barrel 2 | B2-CT-U | 76.2 | 0.42 | 40.3 | 34.0 | 23.4 | 12.7 | 5.9 | 2.0 |
| Barrel 2 | B2-CT-L | 75.8 | 1.44 | 26.8 | 21.7 | 16.1 | 10.6 | 5.3 | 0.9 |
| Barrel 3 | B3-MFT-U | 47.0 | 3.33 | 79.9 | 66.7 | 49.3 | 29.4 | 12.3 | 1.9 |
| Barrel 3 | B3-MFT-L | 47.9 | 3.85 | 78.8 | 65.1 | 48.5 | 30.4 | 13.4 | 1.9 |
| Barrel 4 | B4-CT-U | 74.3 | 0.78 | 34.8 | 29.4 | 20.5 | 11.1 | 5.1 | 1.8 |
| Barrel 4 | B4-CT-L | 76.4 | 0.59 | 46.9 | 39.6 | 27.1 | 14.4 | 6.5 | 2.1 |
| C1 | C1-17-U | 42.9 | 3.42 | 74.1 | 60.5 | 44.5 | 28.4 | 13.0 | 1.9 |
| C1 | C1-17-M-U | 49.8 | 4.01 | 72.0 | 59.0 | 43.1 | 26.4 | 11.7 | 1.8 |
| C1 | C1-17-M-L | 78.3 | 0.17 | 37.3 | 31.2 | 21.6 | 12.1 | 5.5 | 1.4 |
| C1 | C1-17-L | 78.7 | 0.16 | 38.3 | 32.0 | 22.2 | 12.3 | 5.4 | 1.4 |
| C1 | C1-18-U | 41.9 | 3.61 | 75.3 | 60.9 | 44.6 | 28.7 | 13.3 | 1.9 |
| C1 | C1-18-M-U | 47.3 | 3.85 | 74.9 | 60.4 | 44.3 | 28.9 | 13.8 | 2.1 |
| C1 | C1-18-M-L | 76.3 | 1.11 | 28.6 | 23.5 | 17.5 | 10.7 | 4.8 | 0.8 |
| C1 | C1-18-L | 78.5 | 0.51 | 34.5 | 29.1 | 20.5 | 11.4 | 5.2 | 1.5 |


[^0]:    - not added

