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QA: QA

Civilian Radioactive Waste Management System
Management & Operating Contractor

The Determination of Diffusion Coefficient of Invert Materials

TDR-EBS-MD-000002 REV 00

January 18, 2000

Prepared for:

U.S. Department of Energy
Yucca Mountain Site Characterization Office
P.O. Box 30307
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Prepared by:

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Under Contract Number
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Civilian Radioactive Waste Management System
Management & Operating Contractor

The Determination of Diffusion Coefficient of Invert Materials

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January 18, 2000

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1. OBJECTIVE AND SCOPE

The Engineered Barrier System (EBS) Testing Department is performing tests in the Department of Energy's Atlas Facility to evaluate the performance of various means for increasing the time for breakthrough of radionuclides from the waste package to the base of the invert. This includes testing various barriers in the invert as a means of increasing breakthrough time through the process of diffusion. A diffusion barrier may serve as an invert material for the emplacement drifts. The invert material may consist of crushed tuff from the repository excavation at Yucca Mountain or silica sand.

The objective of this report is to determine the diffusion coefficient of the crushed tuff and silica sand invert materials specified by the EBS Testing Department. The laboratory derived information from the testing was used in the Nernst-Einstein equation (Jurinak et al. 1987, p. 626) to determine the diffusion coefficient of the invert material. This report transmits the results and describes the methodology and interpretation.

The scope of this report is to determine the diffusion coefficients of the invert materials mentioned above using the centrifuge at UFA Ventures. Standard laboratory procedures, described in Section 2 of this report, were used. The diffusion coefficients are to be determined over a range of moisture contents. The report contains the diffusion coefficients calculated by the Nernst-Einstein equation (Jurinak et al. 1987, p. 626) that become a part of the Technical Database. Raw data is also included in the report, however this data does not become part of the Technical Database as per Section 3.23 of AP-SIII.3Q *Submittal and Incorporation of Data to the Technical Data Management System*. A sieve analysis of the samples was not conducted as part of this report, but sieve analysis may be accomplished as part of other reports. Two samples of crushed tuff and two samples of silica sand were tested.

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2. QUALITY ASSURANCE

This report was prepared under Administrative Procedure AP-3.11Q *Technical Reports* in accordance with an approved development plan (CRWMS M&O 1999a). A Quality Administrative Procedure QAP-2-0 *Conduct of Activities* Activity Evaluation (CRWMS M&O 1999b) was completed for this report and the report was found to be quality affecting. The *Quality Assurance Requirements and Description* (DOE 1998) is applicable to this report.

The specific Technical Procedures used to produce the data for the report are the YMP-approved *Determining the Aqueous Molecular Diffusion Coefficient in Porous Media Using the Nernst-Einstein Method* (LANL 1999) and *Procedure for Using the UFA to Measure the Matric Potential of Core Sample* (UFA Ventures 1997).

The data for this report comes from the UFA Ventures, Inc. scientific notebook. This notebook will eventually be submitted to the Yucca Mountain Project records center in accordance with the applicable procedures. A subset of the scientific notebook has been submitted to the records center for data tracking purposes entitled *Portion of UFA Ventures, Inc. Scientific Notebook* (Conca 1999).

A commercial spreadsheet program assisted in routine calculations. The program is used to perform the data reduction calculations presented in Attachment A. The software program is appropriate for this application, and is not a controlled source of information.

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3. DESCRIPTION OF THE TEST CONFIGURATION (PLANNED)

The test plan is to use the centrifuge at UFA Ventures and to use the established laboratory procedures discussed in Section 2 of this report. The test will be conducted by placing a saturated sample contained in an electrical conductivity cell in a centrifuge. The centrifuge is used to drive off water from the sample to a set volumetric moisture content as required for different stages of the experiment. The water is removed from a test sample by subjecting the sample to high centrifugal forces produced by the centrifuge and is thus equivalent to high gravitational forces. The electrical conductivity of the sample is measured across the electrical conductivity cell. This information is used in the Nernst-Einstein equation to determine the diffusion coefficient of the invert material at various volumetric moisture contents. All information is to be collected in a Scientific Notebook (to be submitted under the appropriate procedure at a future date). This report contains a subset of the information contained in the Scientific Notebook. The use of the Nernst-Einstein equation is described below.

The aqueous diffusion coefficient is a measure of how ions migrate through subsurface soils and rocks. Diffusion often controls the ultimate source of a contaminant, i.e., how it comes out of the waste form or how it gets through a containment barrier or liner (invert). Once a contaminant gets out into the far-field environment, advective flow usually becomes the dominant transport process. Diffusion becomes important in unsaturated environments where the flow of water is very slow.

Direct and indirect methods have been used to determine diffusion coefficients in porous media. Both techniques employ steady-state and transient-state techniques. Fick's first law forms the basis of steady-state experiments, while Fick's second law describes the transient-state (Helfferich 1962, pp. 257 & 259). Unfortunately, it has been extremely difficult to measure diffusion coefficients below 10^{-8} cm^2/s, using direct steady-state techniques (tracer diffusion tests) because it is difficult to maintain proper boundary conditions in the measuring apparatus, and the tests take an extremely long time. This necessitates an indirect method to measure diffusion coefficients in unsaturated or relatively impermeable soils and rocks. The most widely used indirect method is to measure electrical conductivity in under steady state conditions. The method chosen for this work is the Unsaturated Flow Apparatus (UFA) because of its speed in obtaining results and its accuracy (Conca and Wright 1992, p. 7).

The UFA is a large ultracentrifuge in which core samples are centrifuged at a constant rate. Fluid can be delivered to the samples while the sample is being centrifuged, or the sample can be initially saturated and centrifuged at various speeds to drain to a specific volumetric moisture content. The sample holder can be an electrical conductivity cell to allow for measuring an electrical conductivity value for use in the Nernst-Einstein method. When the target volumetric moisture content is reached, the sample holder is removed and the electrical conductivity measured using a standard conductivity bridge.

Electrical conductivity cells used in the UFA have two stainless steel electrodes in contact with the sample. Once electrical conductivity is measured, it is related to the diffusion coefficient through the Nernst-Einstein equation (Jurinak et al. 1987, p. 626) which is given by:

$$D_i = \frac{RT\Lambda_i}{F^2 Z_i} = \frac{(8.31)xTx\Theta Gt_i}{(9.65x10^4)^2 Z_i C_i} = 8.92 \times 10^{-10} \times T \times \frac{\Theta Gt_i}{Z_i C_i} \quad \frac{\text{Kelvin joule mole}^2 \text{ 1000 cm}^3 \text{ mhos}}{\text{Kelvin coul}^2 \text{ mole mole cm}}$$

$$D_i = 8.92 \times 10^{-7} \times T \times \frac{\Theta Gt_i}{Z_i C_i} \text{ cm}^2/\text{s} \qquad \text{(Eq.1)}$$

Where:

D_i is the diffusion coefficient of the i^{th} ion (cm^2/s)

R is the gas constant (8.31 J/Kelvin mole) (Lide 1997, p. 1-1)

T is absolute temperature (Kelvin)

F is Faraday's constant (9.65×10^4 coul/mole) (Lide 1997, p. 1-1)

Λ_i is the equivalent conductance of the entire system [(mhos L)/(mole cm)] (note:1L = 1000 cm^3)

Z_i is the charge number on the i^{th} ion (dimensionless)

t_i is the transport or transference number of the i^{th} ion (dimensionless)

C_i is the molar concentration of the i^{th} ion (C_i is multiplied by the appropriate activity coefficient at high ionic strengths) (mole/ L)

Θ is the cell constant for the conductivity cell sample holder (cm^{-1})

G is the measured conductance on a conductivity bridge (mhos)

Also note that a joule = volt amp sec; mho = amp $volt^{-1}$; and coul = amp sec

All of these parameters are known or easily measured in the laboratory.

4. DESCRPTION OF TEST CONFIGURATION (AS-BUILT)

The as-built test configuration was the same as the planned test configuration. See the Test Results Section of this report for a discussion of events that occurred during the test that affected test results. The test was conducted as per the plan using the UFA Ventures centrifuge and the laboratory procedures mentioned in Section 2 of this report. Measurements were taken until a low conductance was achieved, indicating a very low level of moisture in the sample. The information was collected in the UFA Ventures scientific notebook. A subset of the scientific notebook (Conca 1999) is presented in Appendix A.

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5. SUMMARY OF DATA FROM THE TEST

The data are shown in the tables in Appendix A. Calibration of instruments was accomplished according to the standard laboratory procedures mentioned in Section 2 of this report.

From similar kinds of measurements to those described in this report, we infer that the error on data included in this report is within ±7% (Nimmo et al.1987, p.131). Conca and Wright (1992, p. 10) indicate that the electrical conductivity and centrifuge method can be used for volumetric water contents from 0 to 50% and for diffusion coefficients from 10^{-4} to 10^{-10}.

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6. DISCUSSION OF THE TEST RESULTS

All four samples drained very rapidly with a resulting drop in the diffusion coefficient to very low values (Tables A-1 through A-4). The lowest value for the tuff was 1.25 x 10^{-10} cm^2/s, and for the sand was 2.18 x 10^{-10} cm^2/s. The tuff began to disintegrate at higher revolutions per minute (RPM) and the electrical conductivity could no longer be measured with any accuracy. The disintegration occurred when the sharp angular points of the tuff broke down under the high pressures and the material at the bottom of the sample holder became noticeably smaller than the material at the top of the cup. This did not occur in the silica sand because the particles were all well rounded.

On Tables A-2 and A-4 the 5000 RPM setting was mistakenly set at 4500 RPM. This error had no effect on the results.

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7.1 DOCUMENTS CITED

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7.2 CODES, STANDARDS, REGULATIONS, AND PROCEDURES

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8. APPENDIX

There are two appendices to this document.

Appendix A (5 Pages):	Tables of Data and Graph of Data
Appendix B (4 pages):	Document Input Reference Sheets

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APPENDIX A

TABLES OF DATA AND GRAPH OF DATA

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Table A-1. Crushed Tuff

Speed (RPM)	Total Weight (gm)	Volumetric Moisture Content (%)	Conductance (μmho) (G)	Temperature (°K)	Diffusion Coefficient (cm^2/s)
300	248.59	32.13	30,000	296	2.02E-06
600	243.70	18.15	8,000	297	5.40E-07
800	240.59	9.26	600	297	4.05E-08
1000	239.81	7.03	100	297	6.75E-09
1200	239.79	6.97	110	298	7.45E-09
1500	239.76	6.89	100	296	6.73E-09
1800	239.71	6.75	80	298	5.42E-09
2100	239.67	6.63	65	297	4.39E-09
2500	239.67	6.63	55	301	3.76E-09
3000	239.53	6.23	50	299	3.40E-09
3500	239.45	6.00	50	302	3.43E-09
4000	239.29	5.55	30	299	2.04E-09
5000	239.26	5.46	30	299	2.04E-09
6000	239.20	5.29	Not Available		
7000	239.14	5.12	Not Available		
8000	239.10	5.00	Not Available		
9000	239.05	4.86	Not Available		
10000	238.99	4.69	Not Available		

Notes: Client ID = EBS-TCO-007, Lab ID = 99-0059

Weights: Assembly= 150.66gm; Holder/Epoxy= 46.11gm; Tare= 112.78gm; Soil/Cup/Tare= 199.47gm (at end of test)

Θ = Cell Constant = 0.301 (measured), Cell Dimensions = 3.00cm dia, 4.94cm long

t_i = Transference Number = 0.4883 for 1.0M KCl (Robinson 1965, p. 159)

C_i = Molar Concentration = 0.58 for 1.0M KCl (Langmir 1997, p. 128)

Z_i = Charge Number = 1 (Number of positive ions K^+ =1)

Sample moisture calculation:

Total Weight-Assembly-Holder/Epoxy = 248.59-150.66-46.11 = 51.82 gm

Soil/Cup/Tare-Tare-Holder/Epoxy = 199.47-112.78-46.11 =40.58 gm

Dry Bulk Density = $40.58/\,(3.00^2/4 \times \pi \times 4.94) = 1.16$ gm/cm^3

Volumetric Moisture Content = $[(51.82/40.58) - 1] \times 1.16 \times 100\% = 32.13\%$

Sample calculation using Equation 1:

$$D_i = 8.92 \times 10^{-7} \times T \times \frac{\Theta G t_i}{Z_i C_i} \ \ cm^2/s$$

$$D_i = 8.92 \times 10^{-7} \times 296 \times \frac{(0.301)(30{,}000)(0.4883)}{(1)\,(0.58)\,(10^6)} \ \ cm^2/s$$

$$D_i = 2.02 \times 10^{-6} \ \ cm^2/s$$

Note that Conductance is in μmhos, hence 10^6 factor in equation.

Table A-2. Crushed Tuff (duplicate)

Speed (RPM)	Total Weight (gm)	Volumetric Moisture Content (%)	Conductance (μmho) (G)	Temperature (°K)	Diffusion Coefficient (cm^2/s)
300	235.37	8.29	360	297	2.24E-08
600	235.11	7.54	110	296	6.81E-09
800	235.05	7.36	100	297	6.21E-09
1000	235.00	7.22	70	299	4.38E-09
1200	234.87	6.84	35	299	2.19E-09
1500	234.62	6.11	25	297	1.55E-09
1800	234.38	5.41	16	298	9.97E-10
2100	234.05	4.45	10	296	6.19E-10
2500	233.77	3.64	8	299	5.00E-10
3000	232.62	0.29	2	296	1.24E-10
3500	232.59	0.20	2	299	1.25E-10
4000	232.58	0.17	Not Taken		
4500*	232.56	0.12	Not Taken		
6000	232.56	0.12	Not Taken		
7000	232.54	0.06	Not Taken		
8000	232.54	0.06	Not Taken		
9000	232.54	0.06	Not Taken		
10000	232.54	0.06	Not Taken		

Notes: Dry Bulk Density = 1.04 g/cm^3, Client ID = EBS-TCO-007, Lab ID = 99-0059 duplicate
Weights: Assembly= 149.68gm; Holder/Epoxy= 46.42gm; Tare= 114.69gm;
Soil/Cup/Tare= 197.53gm (at end of test)
Cell Constant= 0.277, Cell Dimensions = 2.99cm dia, 4.89cm long
* Experimental error, should have been 5000 RPM.

Table A-3. Silica Sand

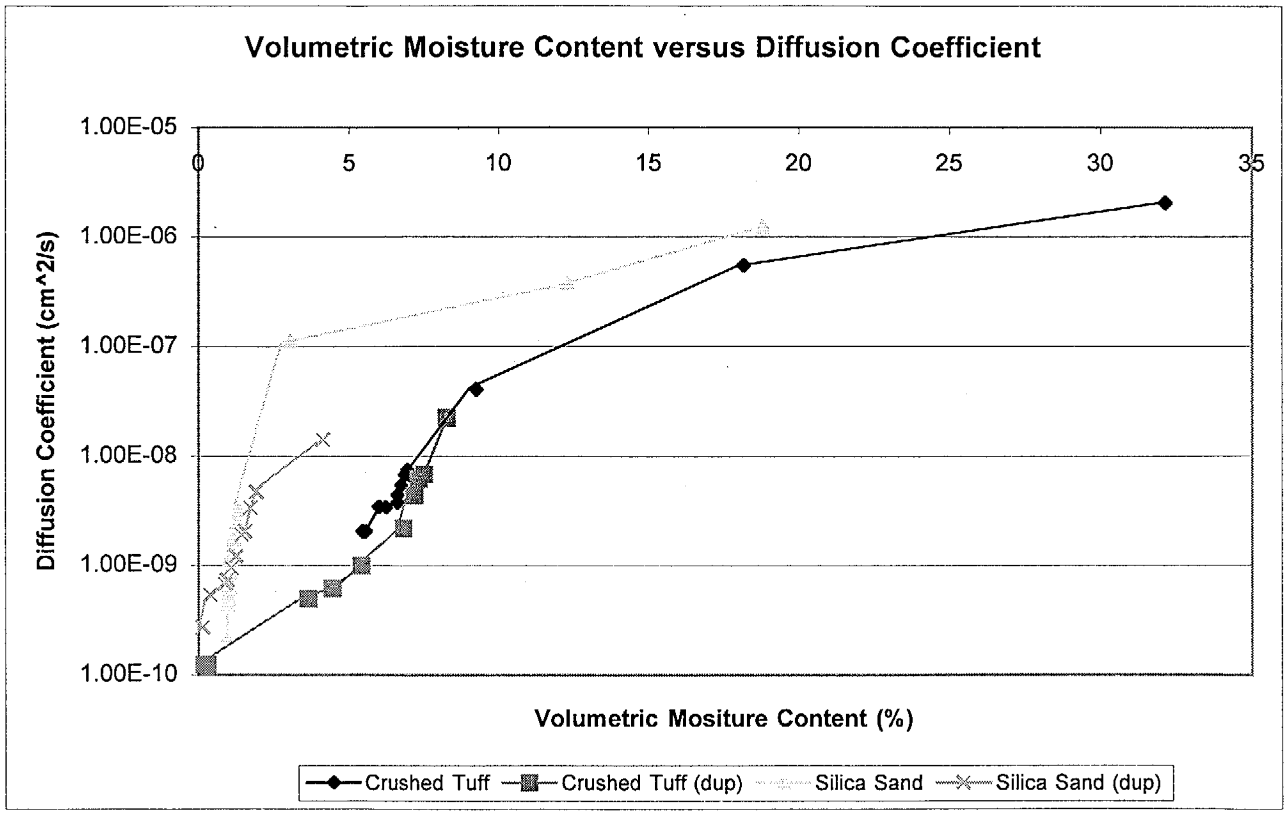
Speed (RPM)	Total Weight (gm)	Volumetric Moisture Content (%)	Conductance (μmho) (G)	Temperature (°K)	Diffusion Coefficient (cm^2/s)
300	254.55	18.78	20,000	296	1.24E-06
600	252.33	12.30	6,000	297	3.73E-07
800	249.16	3.04	1,800	297	1.12E-07
1000	248.58	1.34	50	297	3.11E-09
1200	248.56	1.29	40	298	2.49E-09
1500	248.52	1.17	28	296	1.73E-09
1800	248.51	1.14	28	298	1.75E-09
2100	248.51	1.14	20	297	1.24E-09
2500	248.50	1.11	20	301	1.26E-09
3000	248.48	1.05	20	299	1.25E-09
3500	248.48	1.05	18	302	1.14E-09
4000	248.48	1.05	14	299	8.76E-10
5000	248.47	1.02	11	299	6.88E-10
6000	248.47	1.02	10	299	6.25E-10
7000	248.46	0.99	9	298	5.61E-10
8000	248.46	0.99	8	298	4.99E-10
9000	248.45	0.96	7	298	4.36E-10
10000	248.45	0.96	3.5	298	2.18E-10

Notes: Dry Bulk Density = 1.61 g/cm^3, Client ID = EBS-TCO-008, Lab ID = 99-0060
Weights: Assembly= 148.57gm; Holder/Epoxy= 44.43gm; Tare= 132.30gm
Soil/Cup/Tare= 231.85gm (at end of test)
Cell Constant = 0.277, Cell Dimensions = 2.99cm dia, 4.89cm long

Table A-4. Silica Sand (duplicate)

Speed (RPM)	Total Weight (gm)	Volumetric Moisture Content (%)	Conductance (μmho) (G)	Temperature (°K)	Diffusion Coefficient (cm^2/s)
300	251.96	4.13	210	297	1.42E-08
600	251.19	1.92	70	296	4.71E-09
800	251.12	1.72	50	297	3.38E-09
1000	251.06	1.55	30	299	2.04E-09
1200	251.02	1.44	28	299	1.90E-09
1500	250.96	1.26	18	297	1.22E-09
1800	250.90	1.09	14	298	9.48E-10
2100	250.85	0.95	11	296	7.40E-10
2500	250.83	0.89	10	299	6.80E-10
3000	250.66	0.40	8	296	5.38E-10
3500	250.57	0.14	4	299	2.72E-10
4000	250.56	0.11	Not Taken		
4500*	250.55	0.09	Not Taken		
6000	250.55	0.09	Not Taken		
7000	250.54	0.06	Not Taken		
8000	250.54	0.06	Not Taken		
9000	250.54	0.06	Not Taken		
10000	250.54	0.06	Not Taken		

Notes: Dry Bulk Density = 1.61 g/cm^3, Client ID = EBS-TCO-008, Lab ID = 99-0060 duplicate
Weights: Assembly= 148.91gm; Holder/Epoxy= 44.48gm; Tare= 119.72gm
Soil/Cup/Tare= 221.33gm (at end of test)
Cell Constant = 0.301, Cell Dimensions = 3.00cm dia, 4.94cm long
* Experimental error, should have been 5000 RPM.


Volumetric Moisture Content versus Diffusion Coefficient
Diffusion Coefficient (cm^2/s)
1.00E-05
1.00E-06
1.00E-07
1.00E-08
1.00E-09
1.00E-10
0
5
10
15
20
25
30
35
Volumetric Mositure Content (%)
Crushed Tuff
Crushed Tuff (dup)
Silica Sand
Silica Sand (dup)

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APPENDIX B

DOCUMENT INPUT REFERENCE SHEETS

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OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT
DOCUMENT INPUT REFERENCE SYSTEM

1. Document Identifier No./Rev.:	Change:	Title:
TDR-EBS-MD-000002 Rev. 00		THE DETERMINATION OF DIFFUSION COEFFICIENT OF INVERT MATERIALS

Input Document			4. Input Status	5. Section Used in	6. Input Description	7. TBV/TBD Priority	8. TBVDue To		
2a.	2. Technical Product Input Source Title and Identifier(s) with Version	3. Section					Unqual.	From Uncontrolled Source	Un-Confirmed
1	CRWMS M&O 1999a. Development Plan (DP) Checklist and Cover Sheet: Development Plan for Data Reporting in Support of the Engineered Barrier System Testing Program. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19990802.0315.	N/A	N/A - Reference Only	2	Development Plan	N/A	N/A	N/A	N/A
2	CRWMS M&O 1999b. *Engineered Barrier Systems Performance Testing for SR and LA (12012383MT).* Activity Evaluation. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19990630.0473.	N/A	N/A - Reference Only	2	Activity Evaluation	N/A	N/A	N/A	N/A
3	DOE (U.S. Department of Energy) 1998. *Quality Assurance Requirements and Description.* DOE/RW-0333P, Rev. 8. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.19980601.0022.	N/A	N/A - Reference Only	2	Procedure	N/A	N/A	N/A	N/A

4	AP-3.11Q, Rev. 0, ICN 0. *Technical Reports.* Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.19990701.0620.	N/A	N/A - Reference Only	2	Procedure	N/A	N/A	N/A	N/A
5	LANL-CST-DP-124, R0. *Determining the Aqueous Molecular Diffusion Coefficient in Porous Media Using the Nernst-Einstein Method.* Los Alamos, New Mexico: Los Alamos National Laboratory. TIC: 245581; 245583.	N/A	N/A - Reference Only	2	Procedure	N/A	N/A	N/A	N/A
6	QAP-2-0, Rev. 5. *Conduct of Activities.* Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19980826.0209.	N/A	N/A - Reference Only	2	Procedure	N/A	N/A	N/A	N/A
7	Conca, J.L. and Wright, J. 1992. "Diffusion and Flow in Gravel, Soil, and Whole Rock." *Applied Hydrogeology, 1*, 5-24. Hanover, Germany: Verlag Heinz Heise GmbH. TIC: 224081.	p. 7 &10	N/A – Reference Only	3&5	Description for UFA Machine	N/A	N/A	N/A	N/A
8	Helfferich, F. 1962. *Ion Exchange.* New York, New York: McGraw-Hill. TIC: 217683.	pp. 257 & 259	N/A - Reference Only	3	Fick's first and second laws	N/A	N/A	N/A	N/A
9	Jurinak, J.J.; Sandhu, S.S.; and Dudley, L.M. 1987. "Ionic Diffusion Coefficients as Predicted by Conductometric Techniques." *Soil Science Society of America Journal, 51*, 625-630. Madison, Wisconsin: Soil Science Society of America. TIC: 236974.	p. 626	N/A - Reference Only	1 & 3	Nernst-Einstein Equation	N/A	N/A	N/A	N/A

10	Langmuir, D. 1997. *Aqueous Environmental Geochemistry.* Upper Saddle River, New Jersey: Prentice Hall. TIC: 237107.		N/A - Reference Only	App. A	Molar Concentration of 1.0 M KCl		N/A	N/A	N/A
		P. 128	N/A - Reference Only	App. A	Molar Concentration of 1.0M KCl	N/A	N/A	N/A	N/A
11	Lide, D.R. and Frederikse, H.P.R., eds. 1997. *CRC Handbook of Chemistry and Physics.* 78th Edition. Boca Raton, Florida: CRC Press. TIC: 243741.	p. 1-1	N/A - Reference Only	3	Gas & Faraday's constants	N/A	N/A	N/A	N/A
12	Nimmo, J.R.; Rubin, J.; and Hammermeister, D.P. 1987. "Unsaturated Flow in a Centrifugal Field: Measurement of Hydraulic Conductivity and Testing of Darcy's Law." *Water Resources Research, 23* (1), 124-134. Washington, D.C.: American Geophysical Union. TIC: 216671.	p. 131	N/A - Reference Only	5	Error on Data	N/A	N/A	N/A	N/A
13	Robinson, R.A. and Stokes, R.H. 1965. *Electrolyte Solutions, the Measurement and Interpretation of Conductance, Chemical Potential and Diffusion in Solutions of Simple Electrolytes.* 2nd Edition. London, England: Butterworths & Company. TIC: 242575.	p. 159	N/A - Reference Only	App. A	Transference Number for 1.0M KCl	N/A	N/A	N/A	N/A
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