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Thermal conductivity as a function of saturation for welded and nonwelded tuff

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ABSTRACT: Rock core samples recovered from the proposed repository site at Yucca Mountain, Nevada were used to determine the relationship between thermal conductivity and saturation state for both welded and nonwelded tuffs. Welded tuff was taken from Alcove 5 of the Exploratory Studies Facility at Yucca Mountain, and nonwelded tuffs from four lithostratigraphic units were obtained from the USW SD-7, USW SD-9, and USW SD-12 drillholes. Thermal conductivities were measured for six welded and six nonwelded specimens under dry, saturated, and approximately ten intermediate moisture conditions. All thermal conductivity tests were conducted at 30°C and at atmospheric pressure. A linear and commonly used nonlinear curve were fitted to the data and the goodness of fit determined. The linear relationship provides a better fit to the data, as indicated by the sum of the squared errors.

1 INTRODUCTION

This document contains the results of a suite of laboratory thermal properties tests designed to assess the effect of saturation level on the thermal conductivities of welded and nonwelded tuffs from the proposed repository site at Yucca Mountain, Nevada. Thermal models of the repository require measured values of dry and saturated thermal conductivities and then interpolate between them using either a linear relationship or the following:

$$k_{eq} = k_d + (k_w - k_d)\sqrt{S_l} \qquad (1)$$

where:

k_{eq} = equivalent thermal conductivity at a given saturation level, W / (mK)
k_d = dry thermal conductivity, W / (mK)
k_w = wet thermal conductivity, W / (mK)
S_l = liquid saturation level.

Thermal conductivity increases with moisture content, but other than the results presented here, there are no experimental data to support either the linear relationship or that given in Equation 1. The relationship between conductivity and saturation may vary for different lithologies and may be different for wetting and drying cycles. A testing program was therefore designed to map the thermal conductivity-versus-saturation curve during drying for the repository horizon and also for nonwelded units. Six test specimens were prepared from blocks of welded tuff taken from Alcove 5 within the Main Drift of the Exploratory Studies Facility (ESF), and six additional specimens were prepared from USW SD-7, USW SD-9, and USW SD-12 drillhole cores (abbreviated in the text as SD-7, SD-9, and SD-12) to provide data on four nonwelded lithostratigraphic units. Specimens were tested in the saturated state and then slowly dried. Thermal conductivities were measured at approximately ten intermediate saturation states and then in the oven-dried state. Specimens were then resaturated and retested at the initial conditions to ensure data reproducibility.

2 SAMPLE ACQUISITION AND SPECIMEN PREPARATION

The Alcove 5 specimens tested represent tuff specimens from the TSw2 (Topopah Spring welded Unit 2) thermal/mechanical (T/M) unit and from the Tptpmn (Tertiary, Paintbrush, Topopah Spring tuff, crystal poor, middle nonlithophysal) lithostratigraphic unit. Specimens from the SD-7, SD-9, and SD-12 drillholes represent the CHn1 and CHn2 (Calico Hills nonwelded unit 1 and Calico Hills nonwelded unit 2) T/M units and the Tac4, Tac3, Tac2, and Tacbs (Calico Hills Formation units 4, 3, 2, and basal sandstone) lithostratigraphic units.

All specimens were ground into right circular cylinders. Alcove 5 specimens were 12.7 mm in height and 50.8 mm in diameter, while the nonwelded specimens from the SD drillholes were 50.8 mm in diameter and 6.5 mm in height. The heights of the

nonwelded specimens were reduced to provide data within the calibrated range of the instrumentation.

3 TEST METHODS

Six welded Alcove 5 specimens (three cored vertically and three horizontally) and six nonwelded SD drillhole specimens were saturated in distilled water under vacuum and then tested in the saturated state at 30°C. Intermediate moisture contents were obtained by allowing each specimen to dry slowly in air. Once a target intermediate moisture content was achieved, the specimen was placed in a moisture containment cell and tested. Thermal conductivities were measured at approximately ten intermediate saturation states. Specimens were then oven-dried at 110°C under vacuum. The vacuum was maintained while the specimens cooled to prevent rehydration in moist air. Once measurements were completed on oven-dried specimens, Alcove 5 specimens were resaturated and retested at the initial conditions. Four of the six nonwelded specimens were rehydrated at 100% relative humidity (RH) and tested after reaching stable masses. They were then resaturated and retested. These four tests conducted at the hydrated but not saturated conditions are the only intermediate data points obtained that are representative of the wetting process.

For all specimens, porosity was calculated from the difference in mass between oven-dried and saturated rock. Saturation was then calculated as follows:

$$\text{Saturation} = \frac{M_{\text{Test}} - M_{\text{Dry}}}{M_{\text{Saturated}} - M_{\text{Dry}}} \qquad (2)$$

where:

M_{Test} = Average mass during test (g)
M_{Dry} = Dry mass (g)
$M_{\text{Saturated}}$ = Saturated mass (g).

For nonwelded specimens, this calculation is not necessarily accurate because they contain minerals that absorb water into their structure. The difference between dry and saturated masses therefore does not only include water contained in the pores but also includes water bound in the mineralogy. Porosity therefore should be determined from the difference between the saturated mass and the fully hydrated mass (i.e. the mass measured when minerals are fully hydrated but no water is present in the pore structure). An additional complication is that after the specimens have been dried once, the structures of the zeolitic minerals change and the microstructure is permanently altered; therefore the minerals cannot be rehydrated to their original state. Recognizing that this is an imperfect technique, the nonwelded specimens were treated in the same manner as the welded.

Thermal conductivity measurements were made using the guarded heat flow meter (GHFM). The test specimen was located between two heater plates controlled at different temperatures, producing heat flow through the specimen. The heat flow was measured by a heat flux transducer (HFT) located between the specimen and one heater plate. Radial heat flow losses were minimized in two ways: (1) a cylindrical guard heater surrounded the specimen and was maintained near the mean specimen temperature; (2) specimens with lengths less than 21 mm were used.

The GHFM is calibrated by comparing theoretical values to results obtained using specimens of known thermal conductivity. Calibrations were performed on reference samples of high-purity (99.99%) fused quartz. A range of thermal resistance values was obtained using specimens of different thicknesses (6, 9, 12, 15, 18, and 21 mm) to obtain thermal resistance values that spanned the operating range. Thermal resistance measurements were made at three temperatures (21°, 30°, and 40°C) spanning the test temperature (30°C). Recommended thermal conductivity values for high-purity fused quartz are published by the National Institute of Standards and Technology (NIST).

Calibrations were verified by performing measurements on reference specimens of Pyrex 7740. Recommended thermal conductivity values for Pyrex 7740 are published by the NIST. Verifications were obtained using three specimen sizes (6, 15, and 18 mm) and testing at each of the three temperatures. Verifications were performed periodically throughout the testing program. The verification errors obtained immediately after a calibration but before testing (the pre-test values) show the agreement between results obtained on two types of standards, Pyrex and fused quartz. The pre-test verification errors were below 0.1 W / (mK). This is an indication of the difference in results obtained on the two types of reference materials. The NIST-recommended values for each standard are accurate to only ±5% and so a disparity of 0.125 W / (mK) is within the error range to be expected. The difference between pretest and subsequent verifications is an indication of apparatus reproducibility and drift over time at a specific thermal resistance. Apparatus output was reproducible and drift was not significant [less than 0.02 W / (mK)].

This study represents a change in calibration procedure relative to thermal conductivity measurements made for the Yucca Mountain Project over the past several years. Thermal conductivities reported in Brodsky (1996), Brodsky et al. (1997), Brodsky (1997a), and Brodsky (1997b) were made using the GHFM calibrated with Pyrex 7740 and verified with fused quartz. In the current study, the calibrations were performed using fused quartz, and the verifications were performed with Pyrex 7740. The largest source of error in these measurements is uncertainty in the thermal conductivity values of the standards. This change in calibration procedure was made because the fused quartz data used by the NIST were less scattered over the operating range of the instrument than the Pyrex 7740 data. It was therefore concluded that by calibrating with fused quartz the data might be more accurate. The effect of this change is to increase measured values of thermal conductivity by approximately 0.1 W / (mK), which is just within the error bar reported with previous measurements.

After the instrument was calibrated, the specimens were tested in the same manner as the reference materials. Specimens were placed in moisture containment cells to prevent evaporation of water during the test. Temperature was increased at 1°C / min. to the measurement temperature (30°C). Data were obtained after the instrument had reached steady-state thermal equilibrium as determined by taking readings of the thermocouples and HFT as a function of time until the readings were constant.

4 RESULTS AND DISCUSSION

The objective of this work was to develop a relationship between thermal conductivity and saturation. For the Alcove 5 specimens, the calculation of saturation is straightforward and given in Equation 2. The thermal conductivity data for Alcove 5 are plotted as a function of saturation in Figure 1. Two functional forms were fitted to the data: (1) a line and (2) the functional form given in Equation 1. For both equations, fitting parameters were determined by minimizing the sum of the squared errors (SSE). The fits for Tptpmn are shown in Figure 1 and the fitting parameters (slope, intercept, k_w, and k_d) are given in Table 1. The projected value of k_w for the

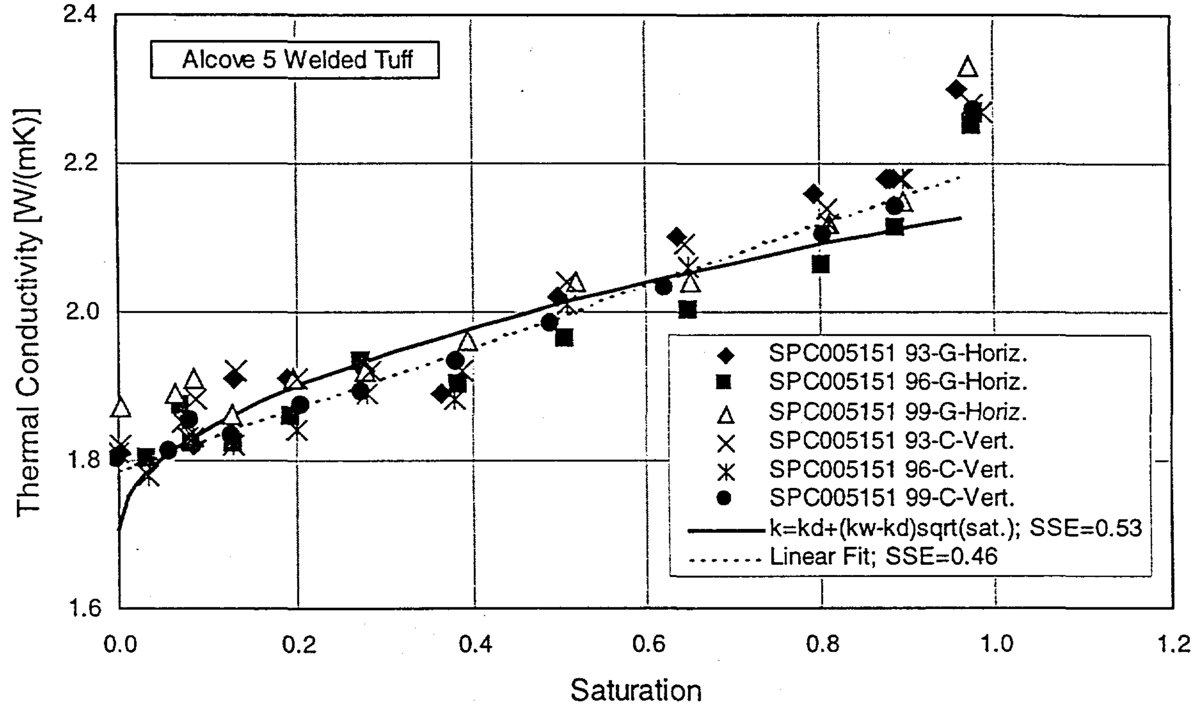

Figure 1. Thermal conductivity as a function of saturation for welded Tptpmn specimens from Alcove 5 of the ESF. Two functional relationships are given as well as the SSE (in conductivity) for each relationship.

Table 1. Fitting parameters.

Lithostratigraphic Unit	Number of Specimens	Linear Fit				Fit to Equation 1		
		Intercept or k_d, W/(mK)	Slope	k_w W/(mK)	Sum of Squared Errors	k_d W/(mK)	k_w W/(mK)	Sum of Squared Errors
Tptpmn	6	1.79	0.414	2.20	0.46	1.71	2.14	0.53
Tac4	1	0.52	0.54	1.06	0.007	0.42	0.98	0.011
Tac3	1	0.53	0.55	1.08	0.004	0.43	1.00	0.013
Tac2	2	0.52	0.59	1.11	0.020	0.39	1.03	0.056
Tacbs	2	0.71	0.59	1.31	0.007	0.59	1.21	0.062

linear fit is provided for reference. The sums of the squared errors for both types of functional forms are similar, although the linear equation provides a slightly better fit than Equation 1.

Figure 1 shows that full saturations were generally not achieved during testing. The maximum mass obtained on a saturated specimen was used as the fully saturated mass. Specimens also lost nominal amounts of moisture during the test, and it was the average specimen mass during the test that was used to calculate saturation state. For these reasons, the saturations during testing generally did not reach 1.0.

It is evident from Figure 1 that data obtained at nearly full saturation are above the trend of the remaining data. These data were therefore not used to calculate fitting parameters. The high values obtained at near full saturation are believed to be an artifact of the calibration procedure. This procedure, developed by the equipment manufacturer and used for many years on the Yucca Mountain Project, was used to obtain results on saturated specimens reported in Brodsky et al. (1997) and Brodsky (1997b). The thermal conductivity apparatus is calibrated using dry moisture containment cells. When a specimen with a wet surface is placed in the cell, the conduction across the specimen/cell interface increases. This increase in conduction is attributed to the specimen and results in a slightly higher measured value of thermal conductivity. This problem was noted early in this testing program and several alternative calibration procedures were attempted, including calibrating and testing with wet or flooded moisture containment cells. Unfortunately, these alternative procedures also had inaccuracies associated with them. For specimens with lower saturation states, the specimen surfaces were not wet; thus the dry calibration was appropriate. It is difficult to estimate the error associated with this phenomenon because the database is very limited. Based solely on the data presented here, this error appears to average approximately 0.1–0.2 W / (mK).

Thermal conductivities for nonwelded specimens from lithostratigraphic units Tac4, Tac3, Tac2, and Tacbs are plotted as a function of saturation in Figures 2, 3, 4, and 5, respectively. Again, two functional forms were fitted to the data for each lithostratigraphic unit: (1) a line and (2) the functional form given in Equation 1. As in the case of the welded specimens, the data obtained at full saturation were not used in the fitting procedures. The fits for each lithostratigraphic unit are shown in Figures 2 through 5, and the fitting parameters (slope, intercept, k_w, and k_d) are given in Table 1.

For each lithostratigraphic unit tested here, the SSE for the linear fit is less than that for the fit to Equation 1. The linear fit is clearly better for the nonwelded units, so it is recommended for all the nonwelded units tested in this study. The difference is very small for Tptpmn but the linear fit is nominally better.

One observation made during testing was that when nonsaturated specimens were placed into the GHFM, both the upper and lower surfaces appeared dry. When the specimens were removed, the upper surfaces (cold sides) appeared moist for specimens with higher moisture contents. This implies that moisture redistribution during the test caused some regions to dry and other regions to become more saturated.

Four nonwelded specimens were rehydrated after measuring their drying curves by subjecting them to 100% RH conditions until masses were stable. The thermal conductivities measured on rehydrated specimens compared very favorably with those obtained at the same moisture content during drying (Brodsky 1998). These data are consistent with (but not necessarily indicative of) the use of a single curve to characterize thermal conductivity during both wetting and drying cycles.

After specimens were dried and conductivities had been measured as a function of saturation along the drying curve, specimens were resaturated. Because resaturations were incomplete, these final measurements were not always performed at the initial saturations. These measurements were intended as an additional check on the reproducibility of the data. The conductivities and saturations obtained before and after the drying cycle are summarized in Table 2. The data show that the reproducibility of the measurements is very good and well within the accuracy of the data.

5 CONCLUSIONS

Sample material was recovered from blocks in Alcove 5 of the ESF and the SD-7, SD-9, and SD-12 drillholes at Yucca Mountain, Nevada for laboratory thermal conductivity tests. The test objective was to develop a functional relationship between saturation state and thermal conductivity for welded and nonwelded tuffs. The data show that for both the welded (Tptpmn) and nonwelded (Tac4, Tac3, Tac2, and Tacbs) tuffs tested here, a straight line fits the conductivity-versus-saturation data better than a function of the form shown in Equation 1. The SSE was used as the measure of goodness of fit.

The results given in this study differ from those obtained previously using the GHFM because high-purity (99.99%) fused quartz was used as the calibration standard rather than Pyrex 7740. It is expected that this change caused the values of thermal conductivity measured here to increase by approximately 0.1 W / (mK). The change was made because it was thought the new data would be more accurate.

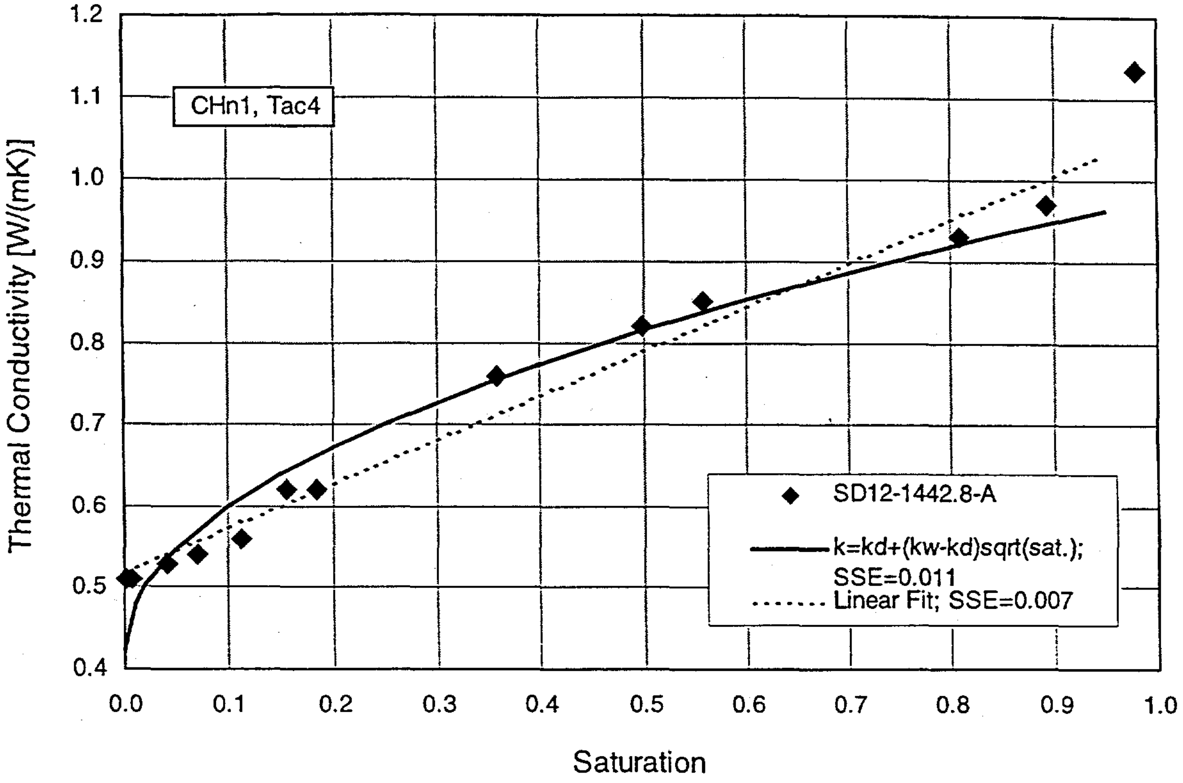

Figure 2. Thermal conductivity as a function of saturation for nonwelded Tac4 specimen from Drillhole SD-12. Two functional relationships are given as well as the SSE (in conductivity) for each relationship.

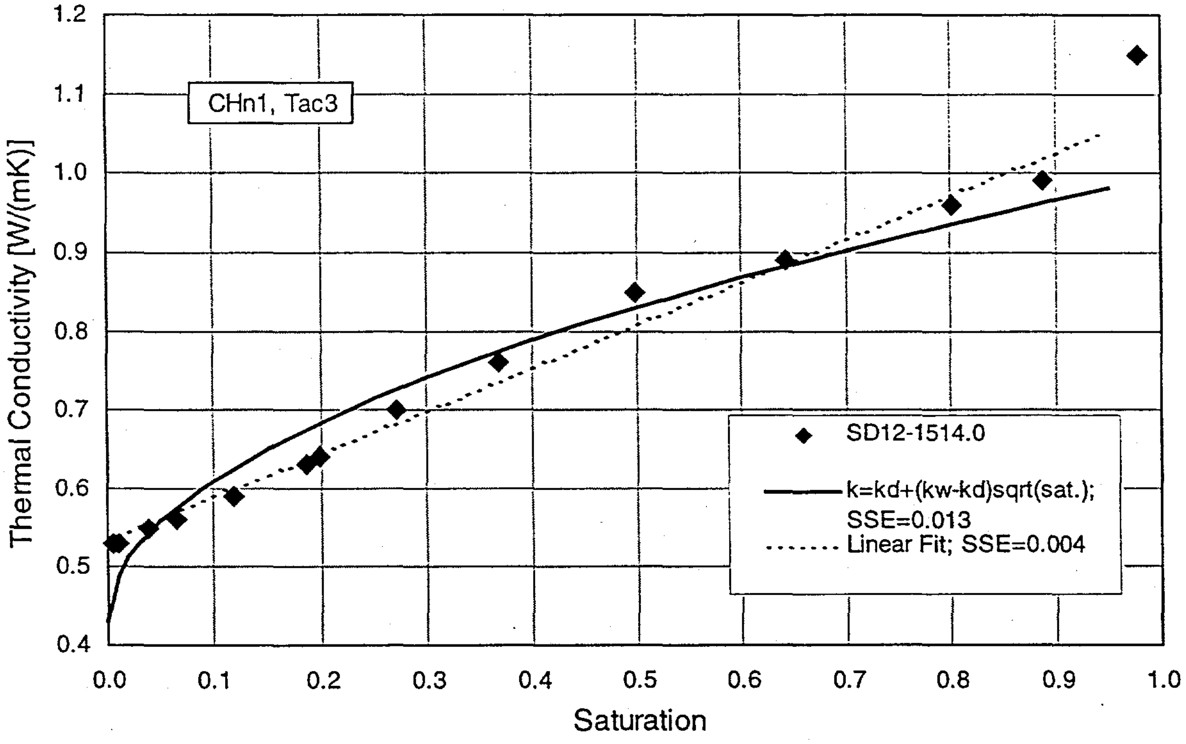

Figure 3. Thermal conductivity as a function of saturation for nonwelded Tac3 specimen from Drillhole SD-12. Two functional relationships are given as well as the SSE (in conductivity) for each relationship.

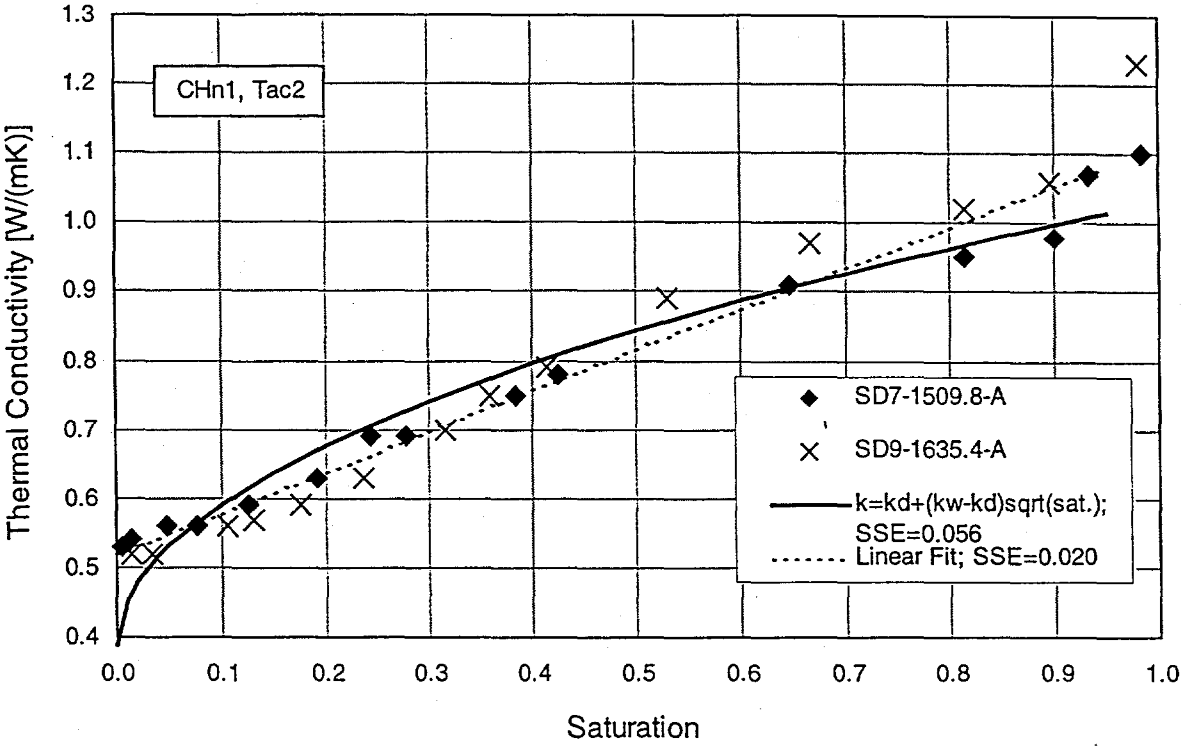

Figure 4. Thermal conductivity as a function of saturation for nonwelded Tac2 specimens from Drillholes SD-7 and SD-9. Two functional relationships are given as well as the SSE (in conductivity) for each relationship.

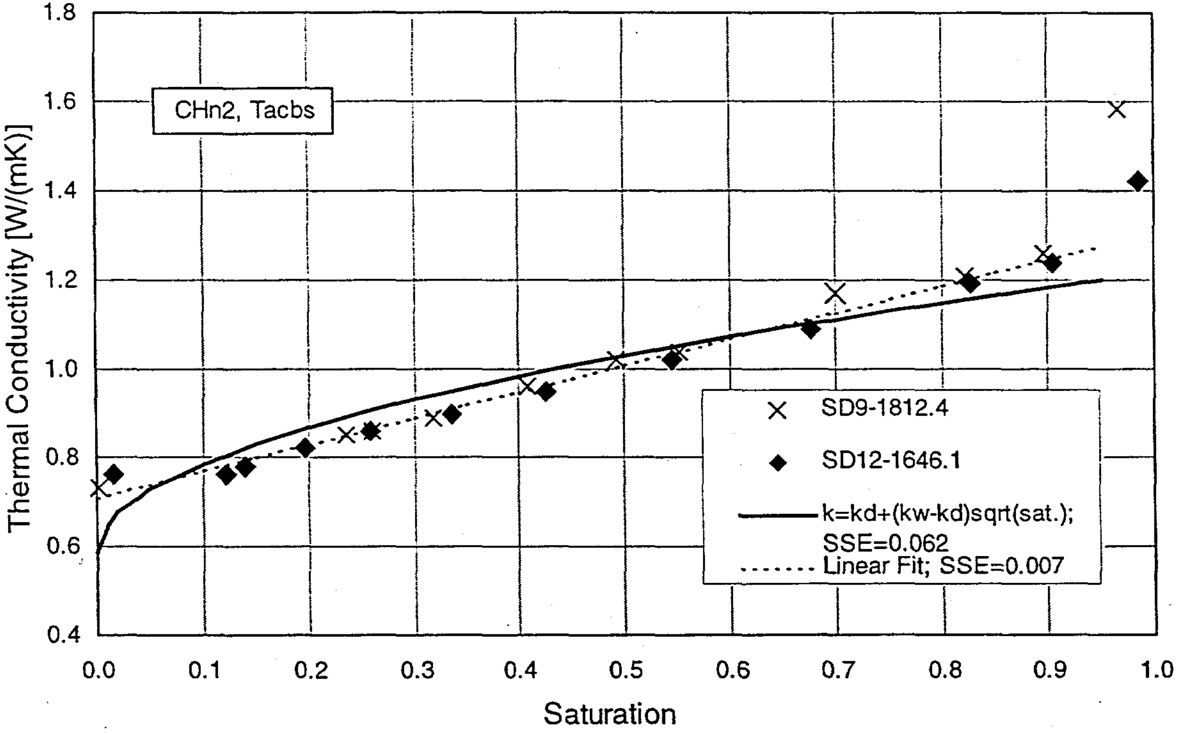

Figure 5. Thermal conductivity as a function of saturation for nonwelded Tacbs specimens from Drillholes SD-9 and SD-12. Two functional relationships are given as well as the SSE (in conductivity) for each relationship.

Table 2. Thermal conductivities under saturated conditions.

Specimen ID	Initial Measurements		Final Measurements	
	Saturation %	Thermal Conductivity W / (mK)	Saturation %	Thermal Conductivity W / (mK)
SPC005151 93-G-Horiz.	96	2.3	91	2.31
SPC005151 96-G-Horiz.	98	2.25	94	2.29
SPC005151 99-G-Horiz.	97	2.33	85	2.34
SPC005151 93-C-Vert.	98	2.28	91	2.31
SPC005151 96-C-Vert.	99	2.27	93	2.35
SPC005151 99-C-Vert.	98	2.27	94	2.33
SD12-1442.8-A	98	1.13	97	1.07
SD12-1514.0	98	1.15	94	1.07
SD7-1509.8-A	98	1.10	93	1.07
SD9-1635.4-A	98	1.23	97	1.21
SD12-1646.1	99	1.42	89	1.29
SD9-1812.4	97	1.58	No Data	No Data

It was also noted that fully saturated specimens very likely provide overestimates of thermal conductivity because of the effect of water on interface thermal resistance in the apparatus. It is difficult to estimate the error associated with this phenomenon because the database is very limited. Based only on the data presented here (the difference between conductivities measured on saturated specimens and k_w determined from the linear fit to the data), this error appears to average approximately 0.1–0.2 W/(mK).

Data obtained after rehydrating four nonwelded specimens at 100% RH compare favorably with data obtained at the same moisture content during the drying cycle. Data obtained after fully resaturating specimens also compare favorably with data obtained before drying. These comparisons are indicative of good reproducibility of results and are also consistent with (but not necessarily indicative of) the same functional relationship between thermal conductivity and saturation during wetting and drying cycles.

7. REFERENCES

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