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LETTER REPORT

MEASUREMENT OF UNSATURATED HYDRAULIC CONDUCTIVITY IN THE BANDELIER TUFF AT LOS ALAMOS

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SUMMARY

Hydraulic conductivities, K, were experimentally determined as a function of volumetric water content, θ, in Bandelier Tuff cores from Los Alamos, New Mexico. These data were used to determine the feasibility of applying a new unsaturated flow technology (UFA™) to further hydrologic studies of tuffaceous rocks at Los Alamos. The $K(\theta)$ relationships for eight cores of Bandelier Tuff from boreholes AAA and AAB were measured using the UFA and, together with their in situ water contents, were used to determine transient water flux into these samples at the time of sampling. If the system is at steady-state, then these flux values correspond to the recharge through those points, a situation often encountered in semi-arid to arid regions such as Los Alamos and other sites in the western United States.

Samples AAA 9956, AAB 0011, AAB 0012 and AAB 0040 exhibited fluxes of 6×10^{-8} cm/s, 4.8×10^{-7} cm/s, 2.8×10^{-7} cm/s and 2.4×10^{-8} cm/s, respectively, indicating significant flux. Samples AAB 0063, AAB 0065, AAB 0072 and AAB 0081 had very low water contents suggesting fluxes less than 10^{-10} cm/s, and appear to be close to their residual water contents. Assuming that the samples AAB 0063, AAB 0065, AAB 0072 and AAB 0081 were not accidentally dried out during handling, these results imply that these samples have zero recharge and that redistribution of moisture at these horizons is controlled more by vapor diffusion than by advection. The vapor diffusivities in these cores can be determined using the new UFA gas permeameter. Samples AAA 9956, AAB 0011, AAB 0012 and AAB 0040 appear to be controlled by advection.

Often nonwelded tuff is so friable that cores are collected in a state of partial or total disaggregation without a coherent piece large enough to constitute a whole rock core, a situation which occurs often in the Bandelier Tuff at Los Alamos. Measuring transport properties on samples this friable is accomplished by reassembling, or recompositing, the fragments and powders back into cores to a density as close as possible to the field density. Results from a previous study investigated the validity of this method by measuring hydraulic conductivity using the UFA on both recomposited and whole rock cores of the same core of Bandelier tuff and found the results to be very consistent. These direct measurements on recomposited and whole rock cores were also consistent with van Genuchten/Mualem estimations determined from water retention data for the same material, suggesting that recompositing friable cores is a valid representation of the whole rock in the field.

INTRODUCTION

The scope of this report is to measure unsaturated hydraulic conductivities in eight cores of Bandelier Tuff, to use this information to determine the recharge into the tuff at the time of sampling, and to determine the feasibility of a new unsaturated flow technology as an additional tool for characterizing hydrologic properties.

Modeling the transport of contaminants in vadose zones requires knowledge of the material characteristics under unsaturated conditions. The hydraulic conductivity, K, retardation factor, R_f, and diffusion coefficient, D, are the primary transport parameters that are key input parameters to existing and developing models of contaminant release from repository systems and waste sites (*Pigford and Chambré,* 1988). Knowledge of K, R_f, and D, as a function of the volumetric water content, referred to as the characteristic behaviors of a porous medium, is particularly important in designing defensible remediation strategies for site restoration efforts. An excellent discussion of transport parameters, processes, and mechanisms in partially saturated media can be found in *Wang and Narasimhan* (1993).

However, transport parameters have generally been difficult to obtain as a function of the volumetric water content. Traditional methods of investigating unsaturated systems require very long times to attain homogeneous distributions of water because normal gravity does not provide a large enough driving force relative to the low hydraulic conductivities that characterize unsaturated conditions. The low conductivities of whole rock under normal potential gradients have hampered experimental studies because of the long time scales required for the experiments. Most of the existing transport data on whole rock comes from saturated flow studies using high pressures that reduce experimental times but lose the natural conditions by inducing new fractures, enlarging existing fractures, and affecting pressure-dependent reactions and phase stabilities. *In situ* measurements of unsaturated flow in whole rock in boreholes or at the surface using surface infiltrometers have provided some excellent results (*Kilbury, et al.* 1986), but are restricted to a narrow range of water contents, and the boreholes themselves can severely disturb the system (*Montazer,* 1987). Lin and co-workers at Lawrence Livermore National Laboratory (*Lin and Daily,* 1984) have shown that computed impedance tomography (CIT) has great promise in the study of unsaturated flow, but CIT still has only coarse resolution and is not developed for aqueous chemical studies. Yang and co-workers at the United States Geological Survey Denver have extracted pore water from unsaturated tuff using triaxial compression (*Yang, et al.* 1988) but this method is not designed to investigate unsaturated flow. To address these problems, an unsaturated flow apparatus (UFA™) based on open centrifugation was developed in which hydraulic steady-state is achieved rapidly in most geologic media at low water contents (*Conca and Wright,* 1992; *Wright et al.,* 1994; *Conca,* 1993).

THE UFA METHOD

The application of a centripetal acceleration (or its inertial effect, the centrifugal force) to geologic problems is as old as centrifugation itself (*Russell and Richards*, 1938). However, the use of steady-state centrifugation to fix the water content and to measure unsaturated hydraulic conductivities has only recently been demonstrated (*Conca and Wright,* 1992; *Nimmo et al., 1987*). There are specific advantages to using a centripetal acceleration as a fluid driving force. It is a body force similar to gravity, and so acts simultaneously over the entire system and independently of other driving forces, e.g., gravity or matric suction. The UFA developed for this study consists of a rock core ultracentrifuge with an ultralow constant-rate flow pump which provides any fluid to the sample surface through a rotating seal assembly and microdispersal system (Figure 1). Accelerations up to 20,000 g are attainable at temperatures from -20° to 150°C and flow rates as low as 0.001 ml/hr. The effluent is collected in a transparent, volumetrically-calibrated container at the bottom of the sample assembly which can be observed during centrifugation using a strobe light. The maximum sample volume is 100 cm^3.

The UFA Method is effective because it allows the operator to set the variables in Darcy's Law. Darcy's Law states that the fluid flux equals the hydraulic conductivity times the fluid driving force. Under a centripetal acceleration in which the water is driven by both the matric potential and the centrifugal force per unit volume, $\rho\omega^2 r$, Darcy's Law is given by

$$q = -K(\psi)\ [d\psi/dr - \rho\omega^2 r] \qquad (1)$$

where q is the flux density into the sample; K is the hydraulic conductivity, which is a function of the matric suction (ψ) and therefore of water content (θ); r is the radius from the axis of rotation; ρ is the fluid density; and ω is the rotation speed in radians per second.

Hydraulic conductivity is a function of either the matric potential or the volumetric water content. Above speeds of about 300 rpm, provided that sufficient flux density exists, the matric potential is much less than the acceleration, $d\psi/dr \ll \rho\omega^2 r$. Therefore, Darcy's Law is given by $q = K(\psi)\ [\rho\omega^2 r]$. Rearranging the equation and expressing hydraulic conductivity as a function of volumetric water content, θ, Darcy's Law becomes

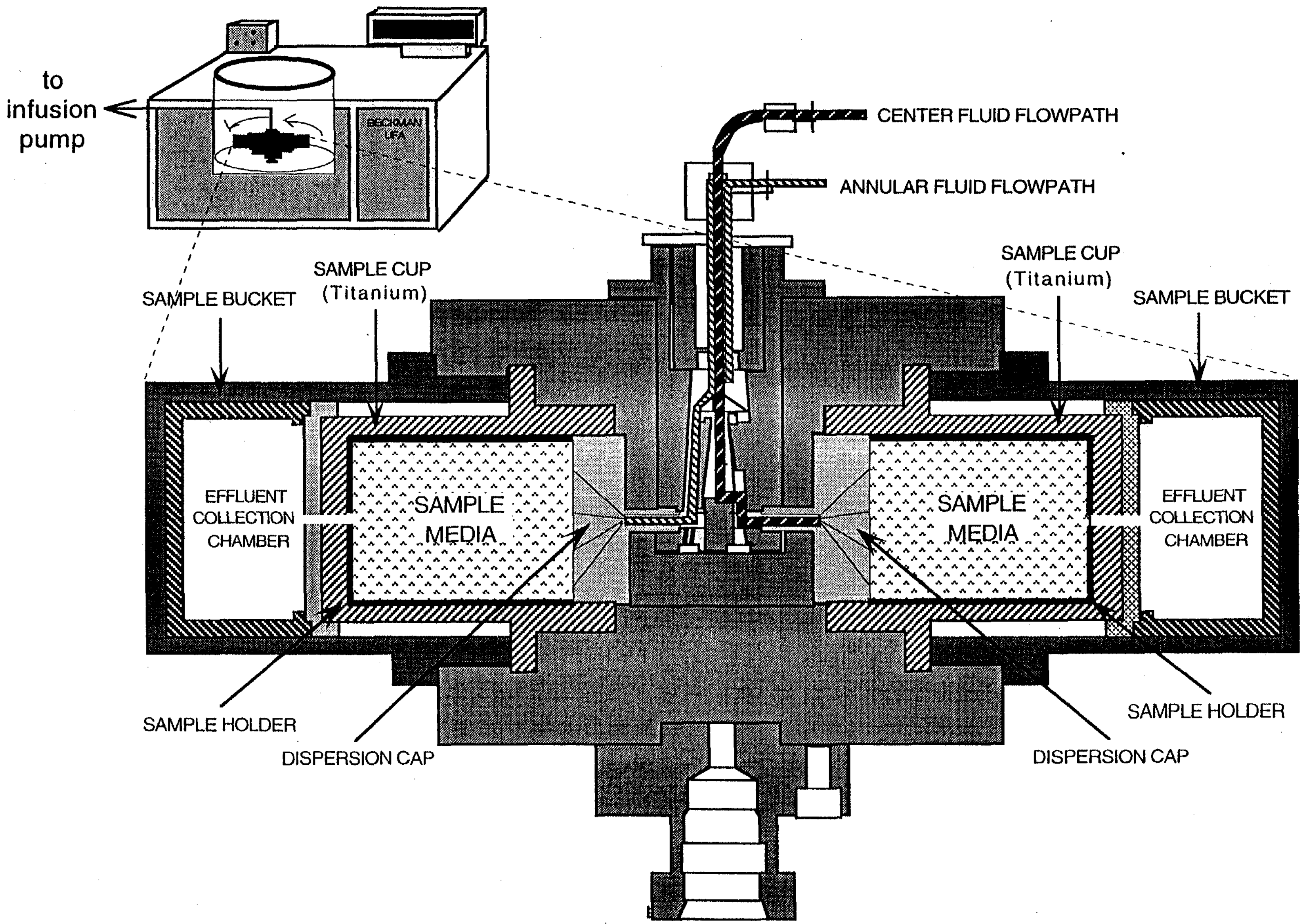


Figure 1. UFA Rotor and Rotating Seal Assembly

$$K(\theta) = q/\rho\omega^2 r \qquad (2)$$

Previous studies have verified equation (2) regarding the linear dependence of K on flux and the second order dependence on rotation speed (*Nimmo and Akstin*, 1988; *Wright et al.*, 1994). The dimensional analysis for equation (2) is

$$cm\ s^{-1} = cm^3\ cm^{-2}\ s^{-1} \div [g\ cm^{-3}\ s^{-2}\ cm \div \{(980.7\ cm\ s^{-2})\ (1\ g\ cm^{-3})\}]$$

One radian per second equals 2π(rpm÷60). A flow rate of 1 ml/hr is 1/3600 $cm^3\ s^{-1}$ divided over the cross-sectional area of the sample. As an example, a whole rock core of Topopah Spring Member tuff accelerated to 7500 rpm with a flow rate into the core of 2 ml/hr achieved hydraulic steady-state in 30 hours with an hydraulic conductivity of 8.28 x 10^{-9} cm/sec at a volumetric water content of 7.0%. Appropriate values of rotation speed and flow rate into the sample are chosen to obtain desired values of flux density, water content, and hydraulic conductivity within the sample. This method provides hydraulic conductivity to within ±8% at a volumetric water content known to within ±2% (*Nimmo and Akstin,* 1988). The high accuracy comes from the tight control on flow rate (±1%) and rotation speed (±5 rpm), and on the ability to precisely measure weight to ±0.001 g.

Hydraulic conductivity can be very sensitive to the solution chemistry, especially when samples contain expandable, or swelling, clay minerals. The solution used was a synthetic vadose zone water close to the ionic strength of the porewaters in the field.

Several issues involving flow in an acceleration field have been raised and addressed by Conca and co-workers (1992 and 1994) and Nimmo and co-workers (1987, 1988, and 1991). Compaction from acceleration is negligible for subsurface soils at or near their field densities. Bulk densities in these samples remain constant because a whole-body acceleration does not produce high point pressures. The notable exception is surface soils and recently tilled soils, which can have unusually low bulk densities. Special arrangements must be made to preserve their densities. Whole rock, grout, ceramics or other solids are completely unaffected by these accelerations. Three dimensional deviations of the driving force with position in the sample are less than a factor of two. Theoretically, the situation under which unit gradient conditions are achieved in the UFA, in which the change in the matric potential with radial distance equals zero ($d\psi/dr = 0$), is best at higher fluxes, higher speeds, and/or coarser grain-size (*Nimmo et al.,* 1994) and this is seen in potential gradient measurements in these ranges which all show $d\psi/dr = 0$. The worst case occurs at the lowest fluxes in the finer-grained materials, but even in the worst case, the hydraulic conductivity appears insensitive to small variations in θ or ψ (*Conca and Wright,* 1994).

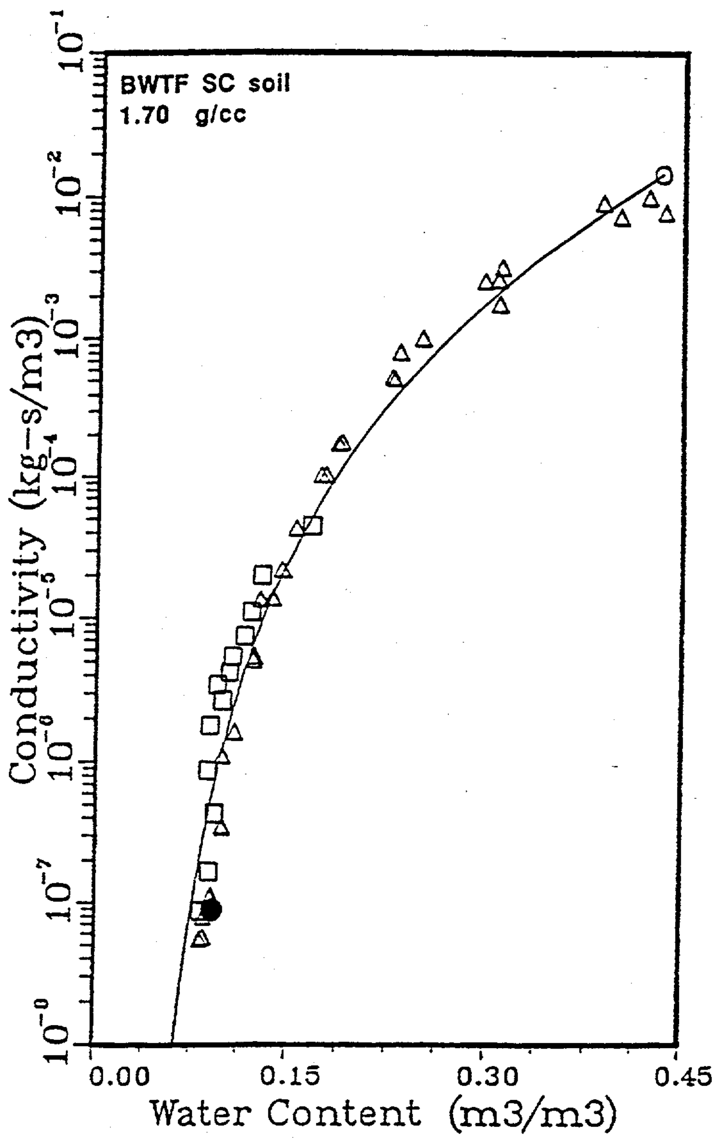
The moisture distribution at steady-state in the UFA is very uniform, to within about 1% in homogeneous systems. In heterogeneous samples or multicomponent systems, such as a whole rock core, each component reaches its own hydraulic conductivity and water content at steady state, as occurs under natural conditions in the field. However, the best verification of the UFA Method must come from investigations of the same materials using several techniques. Several comparisons between the UFA Method and soil columns, van Genuchten/Mualem estimations and lysimeter measurements on the same soils have shown excellent agreement for loams (Figure 2) and even silts (*Nimmo et al.,* 1987; *Wright et al.,* 1994). This reports provides similar validation experiments for whole rock cores (se Figure 7 of this report).

QUALITY ASSURANCE

The results of this project were carried out in accordance with PNL MA-70 Impact Level III, Good Practice Standards procedures, with the proper equipment calibrations by Westinghouse Standards Laboratory, and approved technical procedures in place prior to initiating work. All laboratory tests are described in Laboratory Record Books or cited references to standard tests. However, Impact Level I Technical Procedures are in place for this method. All measurement and test equipment are identified on an M&TE list. All calculations are performed using protocols described in Hand Calculation Procedures or authorized computer-generated algorithms and macros.

RESULTS

The unsaturated hydraulic conductivities of the eight cores of Bandelier Tuff from LANL are given in tabular form as an Appendix, and are also graphed in Figure 3. The shape of the $K(\theta)$ relationship depends upon the pore size distribution of each sample and the connectivity of the pores. There is no obvious relationship between density and $K(\theta)$ in these samples. Sample AAB 0012 was the only sample in this group for which no whole rock core was obtained. Therefore, the fragments and powder were reassembled, or recomposited, back into a core with a density as close as possible to the field density (see discussion below). These eight samples behave similarly within a range of residual water



□ UFA™ measurements made over 3-days

△ Traditional column experiments made over 1-year

● Average of field lysimeter measurements made over 13-years

— Mualem estimation derived from curve-fitted water-retention data made over 6-weeks

Figure 2. Unsaturated Hydraulic Conductivity Curves for the BWTF Soil Obtained Using Four Different Methods.

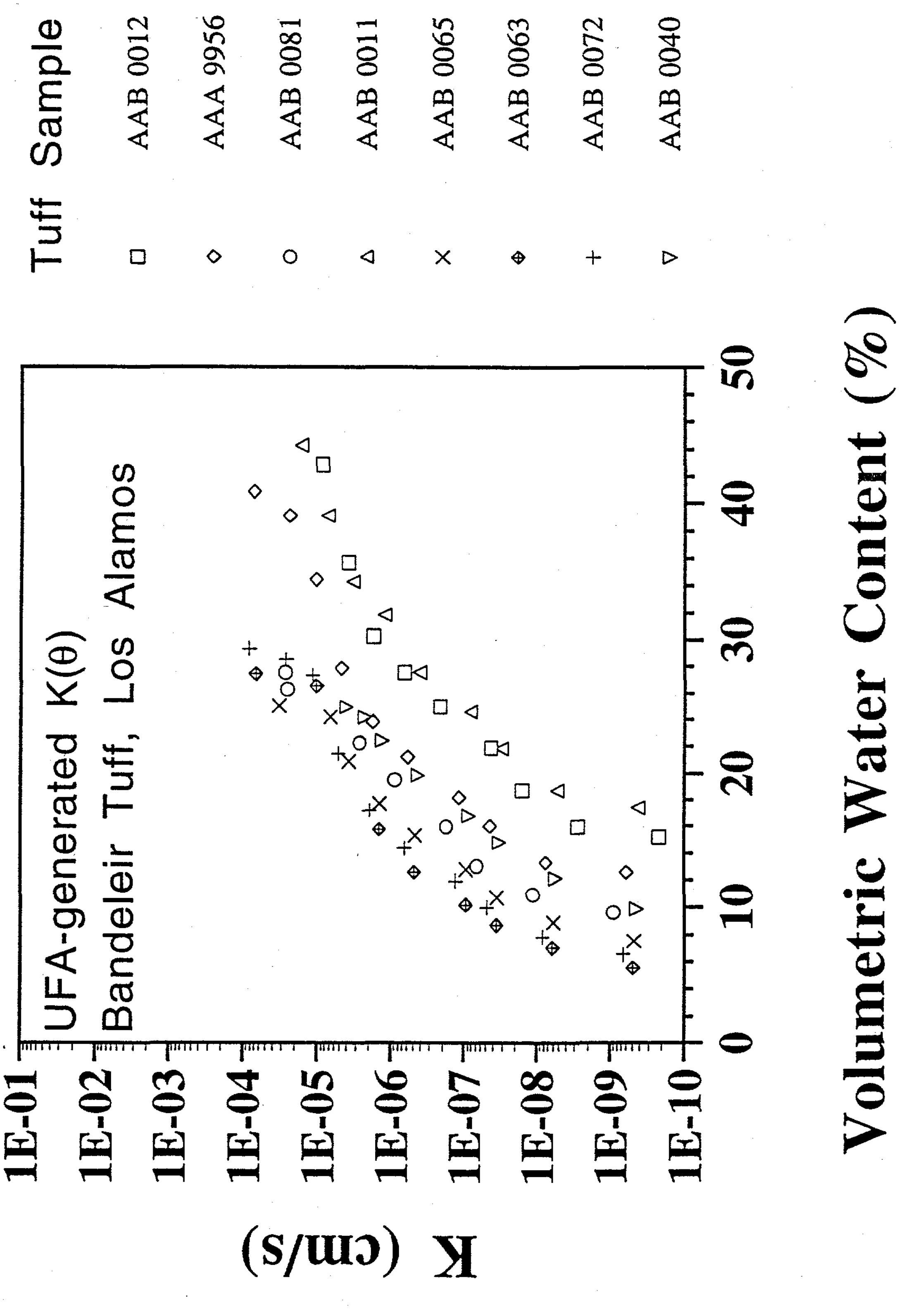


Figure 3. Hydraulic Conductivities of Bandelier Tuff.

contents from about 4 to 17 volume percent, their relative position along the x-axis perhaps reflecting the degree of welding or pore-connectivity within each sample.

Recharge Determinations from $K(\theta)$ and the In Situ Water Content

Recharge is the amount of moisture, usually from precipitation, that passes the upper few meters of soil or rock, past the root zone, and enters the vadose zone. In many situations, the recharge is a critical parameter for modeling contaminant transport and risk assessment, because the recharge is usually the carrier fluid for contaminants of interest and can dominate the rate of transport through the vadose zone. At a minimum, knowledge of the recharge can be used to monitor changes in the system over time and can be used to detect deviations from background.

Often the terms subsurface flux and recharge are used interchangeably, but recharge strictly refers to that flux which eventually enters the saturated zone. When the system has been, or is being, perturbed by anthropogenic events, some transient fluxes may never contribute to the actual recharge, yet these fluxes can play a significant role in overall transport.

The recharge at any point in the subsurface depends upon precipitation rate, type of surface cover, and any lateral fluxes that may occur from perched saturated zones or artificial fluxes within the vadose zone. A recharge less than 10^{-10} cm/s indicates that advection may not be the dominant method of mass transfer within the system, and that vapor transport/molecular diffusion may be as important. Recharge can even have negative values which indicate an annual mean upward migration of moisture, and occurs in very arid regions (*Scanlon,* 1994). Under normal conditions, hydraulic steady state is usually achieved in the vadose zone within meters of the surface, i.e., the recharge is everywhere the same, each material has reached its steady-state volumetric water content for that water flux, and the recharge is equal to the hydraulic conductivity of the materials at that water content. This condition is referred to as unit gradient conditions. Changes to recharge can occur from 1) changing the surface cover through defoliation, emplacement of gravel covers, or surface barriers, 2) disposal of liquid wastes directly above the area, or 3) lateral input of liquid from perched water or migrating plumes, often along boundaries between different units where differences in permeability cause local saturation.

If both the $K(\theta)$ relationship and the in situ, or field, moisture content, are known with any accuracy, then the recharge, or subsurface flux, can be estimated for the system under unit gradient conditions. Under steady-state conditions, the $K(\theta)$ relationship provides the flux value, or recharge, at any water content, and vice versa. This method of estimating recharge has been recognized as a potentially powerful tool for hydrologic and transport studies (*Gee et al.*, 1992; *Nimmo et al.*, 1994; *Conca and Wright*, 1995). Figures 4 and 5 are plots of hydraulic conductivity versus volumetric water content for different sets of tuff samples. The moisture content of the sample as collected from the field core is marked on the horizontal axis. A dashed line is drawn from the sample's moisture content to the hydraulic conductivity curve. A dotted line is then drawn from this intersection point on the curve to the vertical axis. This point on the vertical axis indicates the subsurface flux through the sample at the time the borehole was drilled. If the water content is so low that the resultant flux is less than 10^{-10} cm/s, then the dashed line points below the horizontal axis. The lines are labeled with the sample number.

It can be seen from Figures 4 and 5 that samples AAA 9956, AAB 0011, AAB 0012 and AAB 0040 have water contents consistent with fluxes of 6×10^{-8} cm/s, 4.8×10^{-7} cm/s, 2.8×10^{-7} cm/s and 2.4×10^{-8} cm/s, respectively, indicating significant flux through these horizons in the neighborhood of 10^{-7} cm/s. Samples AAB 0063, AAB 0065, AAB 0072 and AAB 0081 had very low water contents suggesting fluxes less than 10^{-10} cm/s, and appear to be close to their residual water contents. Assuming that the samples AAB 0063, AAB 0065, AAB 0072 and AAB 0081 were not accidentally dried out during handling, these results imply that these samples have almost zero recharge and that redistribution of moisture at these horizons is controlled more by vapor diffusion than by advection. Samples AAA 9956, AAB 0011, AAB 0012 and AAB 0040 appear to be controlled by advection. The vapor diffusivities in these cores can be determined using the new UFA gas permeameter and may be part of follow-on work.

Because the vadose zones in arid regions can require tens to hundreds of years to achieve unit gradient conditions, recharges determined in this way do not provide any specific information about the timing and/or magnitude of the subsurface flux. As an example, sample AAB 0011 could have experienced a flux of 4.8×10^{-7} cm/s the day before sampling and never have been subjected to any greater flux. Alternatively, 10 years ago the sample could have experienced a momentary flux of 10^{-4} cm/s that caused saturation and now is still draining from that event. However, these fluxes are certainly minimum values. Sample AAB 0011 could not be at this water content without having had a recent flux of at least 4.8×10^{-7} cm/s.

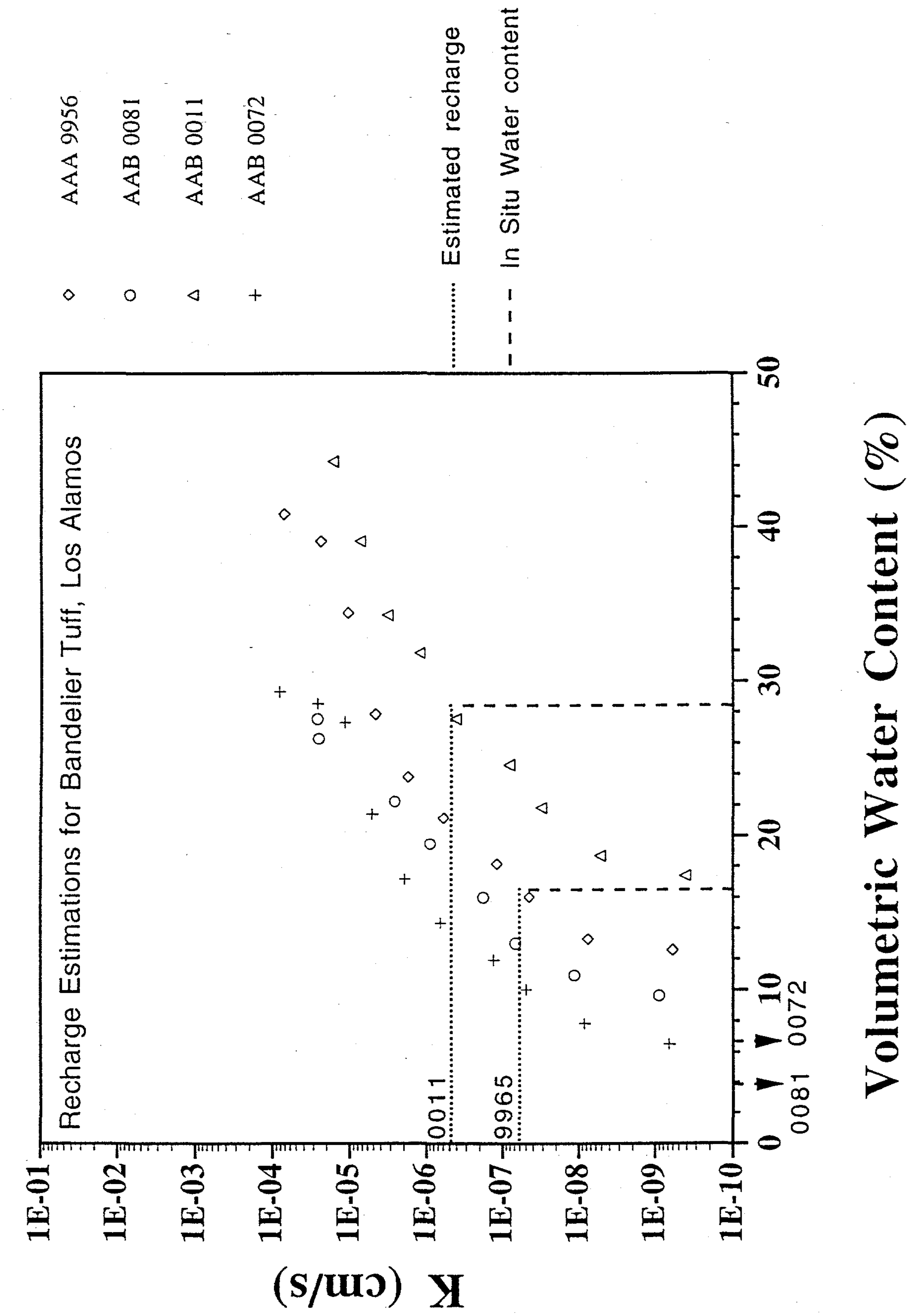


Figure 4. Recharge Estimations for Bandelier Tuff samples.

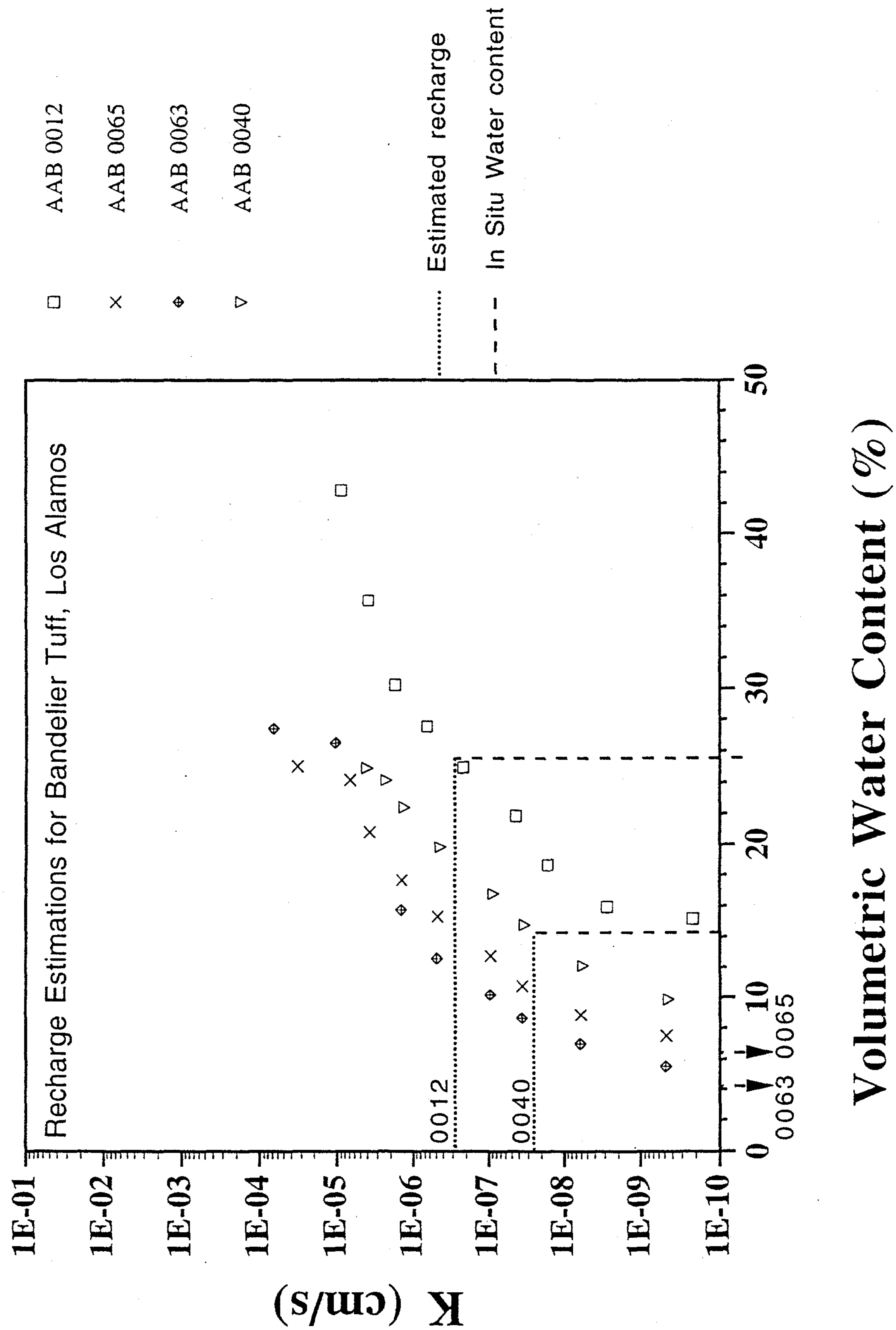


Figure 5. Recharge Estimations for Bandelier Tuff samples.

The greatest source of error for this method of recharge determination is the accuracy of the water content determination and the care taken to preserve the sample and its in situ water content during collection. Because of this need, the more samples obtained, the better the accuracy and ability to detect inconsistencies. Inconsistencies may imply non-steady-state conditions or sampling and handling problems. A single value should be viewed with great skepticism. Regarding this study, eight samples are a small number, but it appears probable that there is a variable recharge distribution at this site. The four samples having fluxes less than 10^{-10} cm/s probably do reflect vanishingly small recharge and are dominated by vapor flow, while the four samples showing larger fluxes probably have significant recharge on the order of 10^{-7} cm/s. If these four samples showing larger fluxes are from shallower depths than the other four samples then they could reflect waste disposal effects. Further investigation of the geological and hydrological aspects of this site should determine whether or not this recharge is natural or anthropogenic. Similar studies on cores from different boreholes at this site would provide greater accuracy and control, and may be used to assemble a subsurface recharge map (*Conca and Wright,* 1995; *Wright et al.,* 1994).

Recompositing of Disaggregated Samples

Because various units within the Bandelier Tuff can be very friable, cores are often collected in a state of partial or total disaggregation without a coherent piece large enough to constitute a whole rock core. Traditionally, measuring transport properties on samples this friable is accomplished by recompositing the fragments and powders back into cores to densities as close as possible to the original field densities. This was done for a sample from another investigation of Bandelier Tuff at Los Alamos, the sample being obtained from borehole G5 at 32 ft. This core was friable enough to have significant amounts of disaggregated material within the core, but also had several coherent pieces large enough to yield a whole rock core for use in the UFA.

Figures 6 and 7 show the direct hydraulic conductivity measurements using the UFA on the recomposited and whole rock cores. Also shown is the estimated hydraulic conductivity from the van Genuchten/Mualem relationships curve-fitted to water retention data for the same material (courtesy of Daniel B. Stephens and Associates). The difference between Figure 6 and Figure 7 is the value for the residual water content chosen for the curve fitting routine, 1.5% and 4.5% by volume. The choice of 4.5% results in a better fit, but the

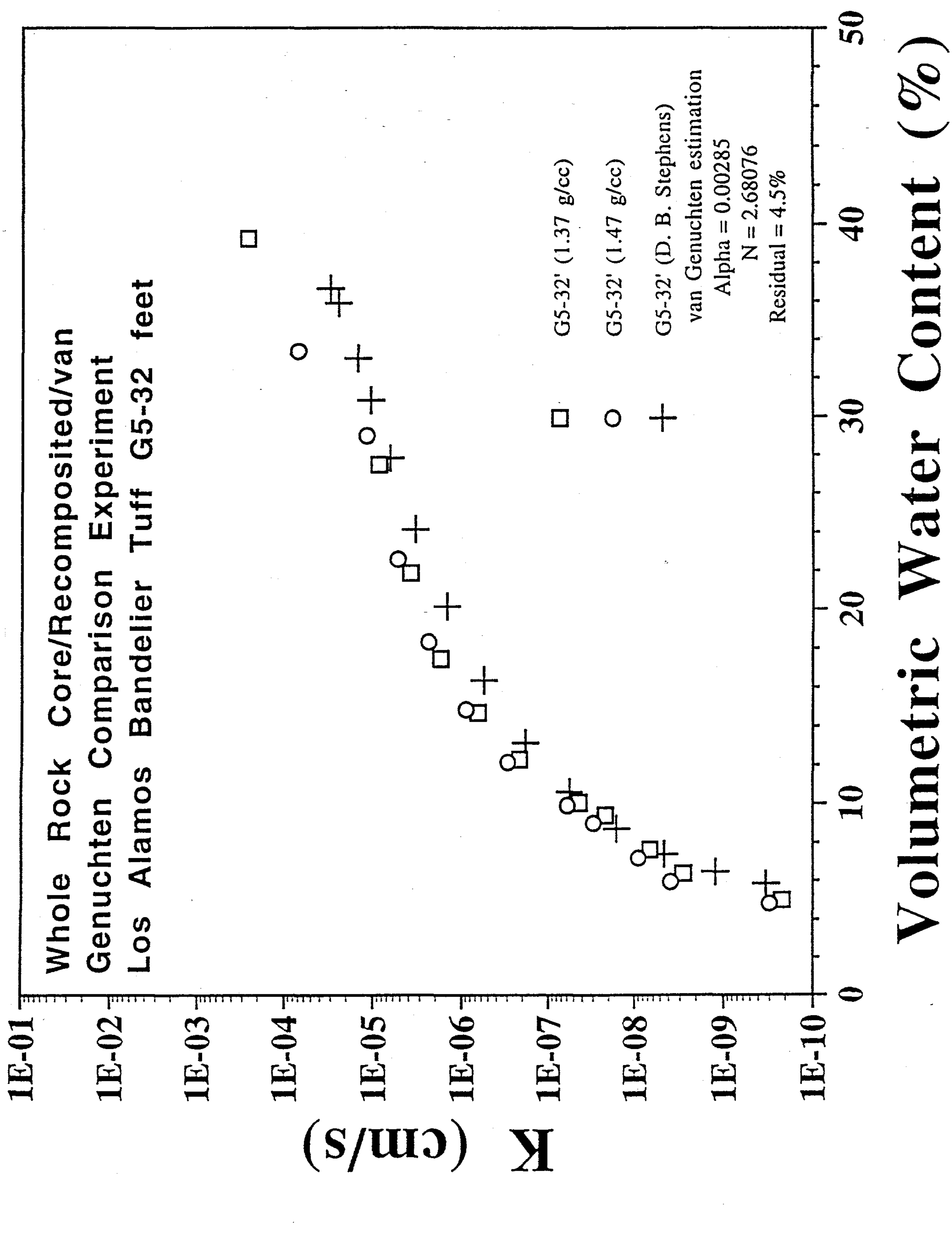


Figure 6. Comparisons of K(θ) for Whole Rock and Recomposites.

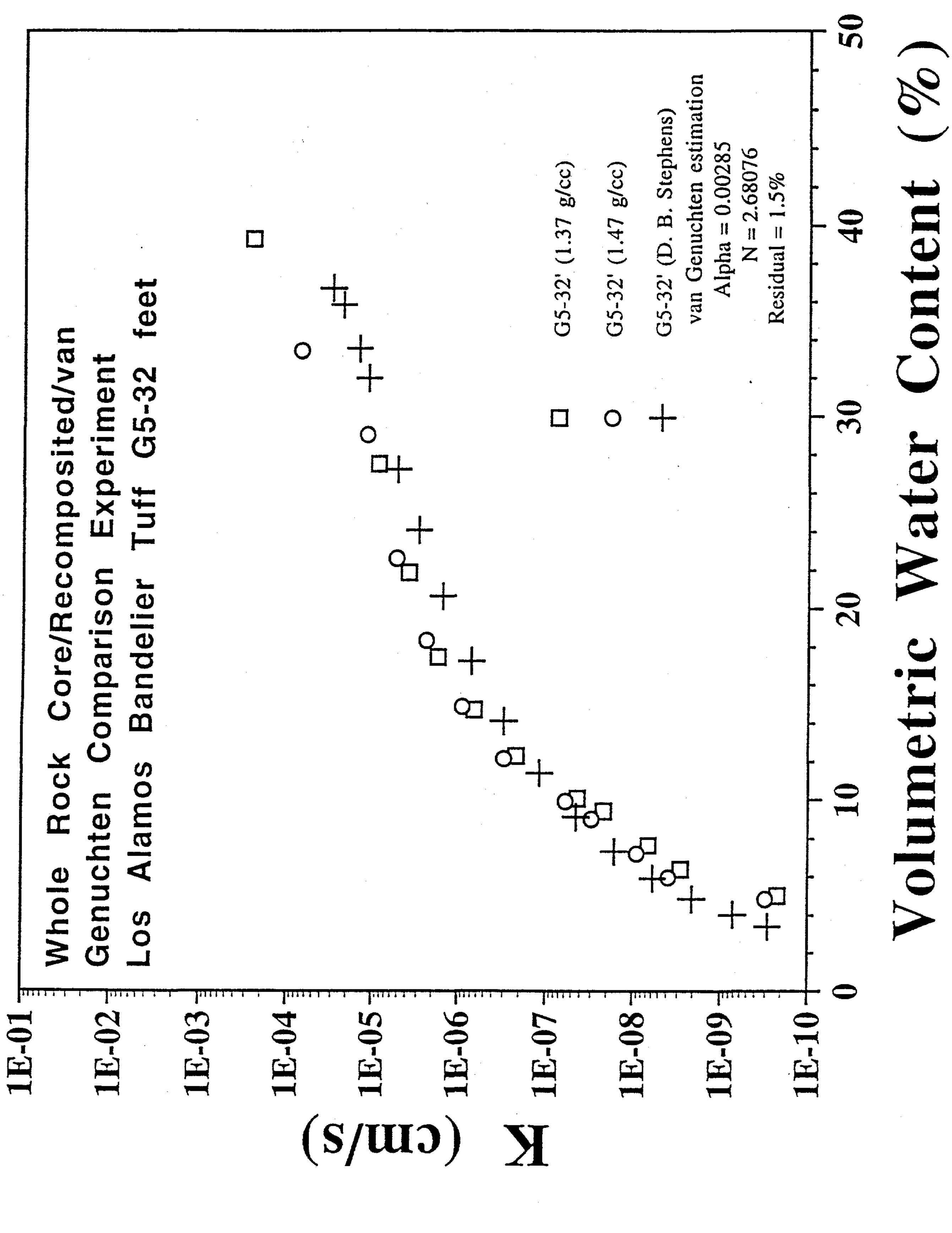


Figure 7. Comparisons of K(θ) for Whole Rock and Recomposites.

1.5% choice still is a reasonable fit for this type of comparison, especially considering that applying van Genuchten/Mualem relationships to rocks is still unpredictable. In fact, these results suggest that van Genuchten/Mualem relationships can be used for whole rock cores. Figure 6 indicates that recompositing friable cores is a valid representation of the whole rock in the field, i.e., the pore connectivities are not significantly different in the whole rock core as in the recomposited core. Often, the grains and fragments of friable nonwelded tuffs are not appreciably cemented or bonded together and, thus, the system is closer to being a soil than a rock. Of course, recompositing will not be valid for rocks that are coherent and for which whole rock cores can be obtained, such as welded tuffs and other hard rocks.

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APPENDIX

Hydraulic Conductivites of Eight Bandelier Tuff Cores

Determined Using the UFA Method

	1	2	3	4	5
	Samples	Volumetric Water Content (%)	K (cm/s)	Column 4	Column 5
1	Tuff	54.11	2.2000E-04		
2	AAB 0012	42.83	8.7000E-06		
3	13.5'-14.0'	35.68	3.8700E-06		
4	1.17 g/cc	30.22	1.7400E-06		
5	Recomposited	27.53	6.5300E-07		
6	initial vol. water	24.94	2.1800E-07		
7	content = 25.3%	21.86	4.3500E-08		
8		18.68	1.6400E-08		
9		15.93	2.7800E-09		
10		15.14	2.1900E-10		
11		14.83	3.9000E-11		
12					
13	Tuff	40.85	7.1900E-05		
14	AAA 9956	39.06	2.3800E-05		
15		34.42	1.0600E-05		
16	1.28 g/cc	27.82	4.7500E-06		
17	Whole Rock	23.79	1.7800E-06		
18	initial vol. water	21.16	5.9400E-07		
19	content = 16.4%	18.15	1.1900E-07		
20		15.98	4.4900E-08		
21		13.27	7.6000E-09		
22		12.55	6.0000E-10		
23					
24	Tuff	27.51	2.6800E-05		
25	AAB 0081	26.25	2.5800E-05		
26		22.23	2.6900E-06		
27	1.42 g/cc	19.47	8.9800E-07		
28	Whole Rock	15.95	1.8000E-07		
29	initial vol. water	13.00	6.7900E-08		
30	content = 6.4%	10.93	1.1500E-08		
31		9.61	9.0700E-10		
32					
33	Tuff	51.00	1.1300E-04		
34	AAB 0011	50.79	4.1200E-05		
35		44.24	1.6600E-05		
36	1.15 g/cc	39.07	7.3600E-06		
37	Whole Rock	34.26	3.3100E-06		
38	initial vol. water	31.81	1.2400E-06		
39	content = 28.3%	27.52	4.1400E-07		
40		24.57	8.2800E-08		
41		21.83	3.1300E-08		
42		18.69	5.3000E-09		

	1	2	3	4	5
	Samples	Volumetric Water Content (%)	K (cm/s)	Column 4	Column 5
43		17.38	4.1800E-10		
44					
45	Tuff	25.00	3.2900E-05		
46	AAB 0065	24.16	6.7400E-06		
47		20.84	3.7900E-06		
48	1.85 g/cc	17.66	1.4200E-06		
49	Whole Rock	15.29	4.7400E-07		
50	initial vol. water	12.74	9.4800E-08		
51	content = 6.4%	10.73	3.5800E-08		
52		8.89	6.0600E-09		
53		7.50	4.7900E-10		
54					
55	Tuff	27.40	6.7000E-05		
56	AAB 0063	26.51	1.0500E-05		
57		15.72	1.4500E-06		
58	1.74 g/cc	12.55	4.8400E-07		
59	Whole Rock	10.17	9.6800E-08		
60	initial vol. water	8.68	3.6600E-08		
61	content = 4.2%	7.01	6.1900E-09		
62		5.52	4.8900E-10		
63					
64	Tuff	29.30	8.2700E-05		
65	AAB 0072	28.51	2.6300E-05		
66		27.29	1.1700E-05		
67	1.44 g/cc	21.42	5.2600E-06		
68	Whole Rock	17.17	1.9700E-06		
69	initial vol. water	14.32	6.5800E-07		
70	content = 3.8%	11.91	1.3200E-07		
71		9.99	4.9700E-08		
72		7.83	8.4200E-09		
73		6.52	6.6500E-10		
74					
75	Tuff	24.90	4.1000E-06		
76	AAB 0040	24.14	2.2900E-06		
77		22.41	1.3200E-06		
78	1.62 g/cc	19.83	4.4000E-07		
79	Whole Rock	16.77	8.8000E-08		
80	initial vol. water	14.77	3.3300E-08		
81	content = 14.2%	12.10	5.6300E-09		
82		9.86	4.4500E-10		