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Physical and Chemical Stability of Galesville Sandstone at Elevated Temperatures and Pressures

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PHYSICAL AND CHEMICAL STABILITY OF GALESVILLE SANDSTONE AT ELEVATED TEMPERATURES AND PRESSURES

by

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ABSTRACT

Galesville Sandstone is currently being considered as a candidate lithology for a Compressed Air Energy Storage (CAES) demonstration facility. This paper summarizes the results of a preliminary set of laboratory-scale autoclave experiments on a sample of the Galesville from the Media natural gas storage field in Illinois. The dependent variables of interest are percent changes in air permeability, liquid water permeability, and friability (disaggregation potential). The independent variables include temperature (50-300^{o}C), pressure (60-120 bars), time (0.5-2.0 months), oxygen content (0-21%), carbon dioxide content (0.0-0.1%), autoclave type, autoclave stirring frequency, and the humidity conditions. Three humidity conditions were simulated: (1) dry air only, (2) air and water vapor, and (3) liquid water only.

Temperature, humidity and time were the dominant variables. Apparently, solutioning of silica and carbonate cements attributes markedly to the observed increases in friability and permeability for cores treated at elevated temperature in a liquid and/or vapor environment. The lack of confining pressure may also be a significant factor. The average change for air and liquid permeabilities was approximately 30 ± 10 percent for all conditions tested. The average increase in the friability index was 150 percent.

INTRODUCTION

Sandstone aquifers are being suggested as potentially suitable reservoir media for the storage of compressed air at temperatures ranging from 50°C to 300°C. In this "adiabatic" Compressed Air Energy Storage (CAES) concept, the thermal energy of compression is retained, stored, and utilized to minimize the requirements for prime fuel during the production (electrical energy generation) phase of the CAES cycle.

One significant unknown associated with the elevated temperature CAES scheme is the physical and chemical response of a porous media reservoir to the elevated temperatures, pressures, humidities, and oxidation potentials associated with CAES operations. Physical and chemical perturbations may prove to be either advantageous or deleterious to the successful implementation and/or long term operation of any CAES facility. Therefore, a laboratory scale experimental program has been initiated at PNL as a precursor to field investigations. The following experimental categories are included: 1) air-water-rock interaction experiments in autoclaves, 2) thermo-mechanical property testing in specially designed triaxial apparatus, 3) thermal conductivity, specific heat capacities, and thermal expansion coefficient measurements with specially designed equipment, and 4) desaturation, physical, and chemical property testing in a prototype fluid flow facility (in the design phase only).

This short paper covers the results from one set of autoclave tests on the Galesville sandstone. Autoclaves are an expedient means for obtaining preliminary data. However, the following four limitations must be noted: 1) the rock sample cannot be confined under a simulated lithostatic load and therefore is more likely to disaggregate than would be expected under actual field conditions, 2) there is no controlled advective transport of liquids or gases through the sample cores and thus surface reactions tend to dominate, 3) the fluid to rock volume and mass ratios are much higher than would be experienced in an actual CAES reservoir, and 4) the sample size is such that only a small incremental volume of the reservoir can be simulated at any one time. Construction of the flow facility should essentially eliminate the first three of these limitations.

EXPERIMENTAL STRATEGY

The dependent variables of primary interest are: 1) air permeability, 2) liquid water permeability, and 3) rock friability or disaggregation potential. The experimental strategy was to observe percent changes in the dependent variable magnitudes as a function of various combinations of maximum and minimum values of the independent variables. The independent variables are listed in Table 1. The main objectives of this preliminary set of experiments are: 1) identify independent variables that have significant impact on the response of the rock, 2) identify those independent variables that have relatively small influence and therefore can be held constant in future experiments, 3) identify trends in the data pertaining to CAES reservoir design and/or operating criteria.

EXPERIMENTAL RESULTS

Sample Classification

Samples of Galesville sandstone were supplied by Geotechnical Engineering, Inc. and Northern Illinois Gas Company. The four inch diameter well cores are from the Media natural gas storage reservoir and were recovered from a depth of 685 meters. The sandstone is white to grayish-white and has an estimated mean grain size of 0.5 mm.

Petrographic examination reveals that the subangular clasts are equant to subequant in form. The rock as a whole is moderately sorted and exhibits cyclic parallel bedding of thin, coarse grained layers and thicker, fine grained layers. The Galesville sandstone is a quartz arenite consisting of approximately 95% quartz, 3% plagioclase feldspar, 1% kaolinite, and 1% calcite+mica+opaque minerals. The clasts are almost entirely quartz with minor amounts of plagioclase that have been weathered partially to kaolinite. The cementing material is predominantly crystalline quartz and minute quantities of carbonates.

Figure 1 is a micrograph of a typical Galesville sample. Figure 2 presents the pretest air and liquid permeabilities for the Galesville samples used in this preliminary set of autoclave experiments.

TABLE 1. Range of Independent Variables

Variable	Minimum Value	Maximum Value
Pressure (bars)	60	120
Temperature (^{o}C)	50	300
Time (months)	0.5	2.0
Oxygen content (%)	0.0	21.0
Carbon dioxide content (%)	0.0	0.1
Stirring frequency (per day)	0	8
Liquid saturated environment	No	Yes
Air and vapor saturated environment	No	Yes

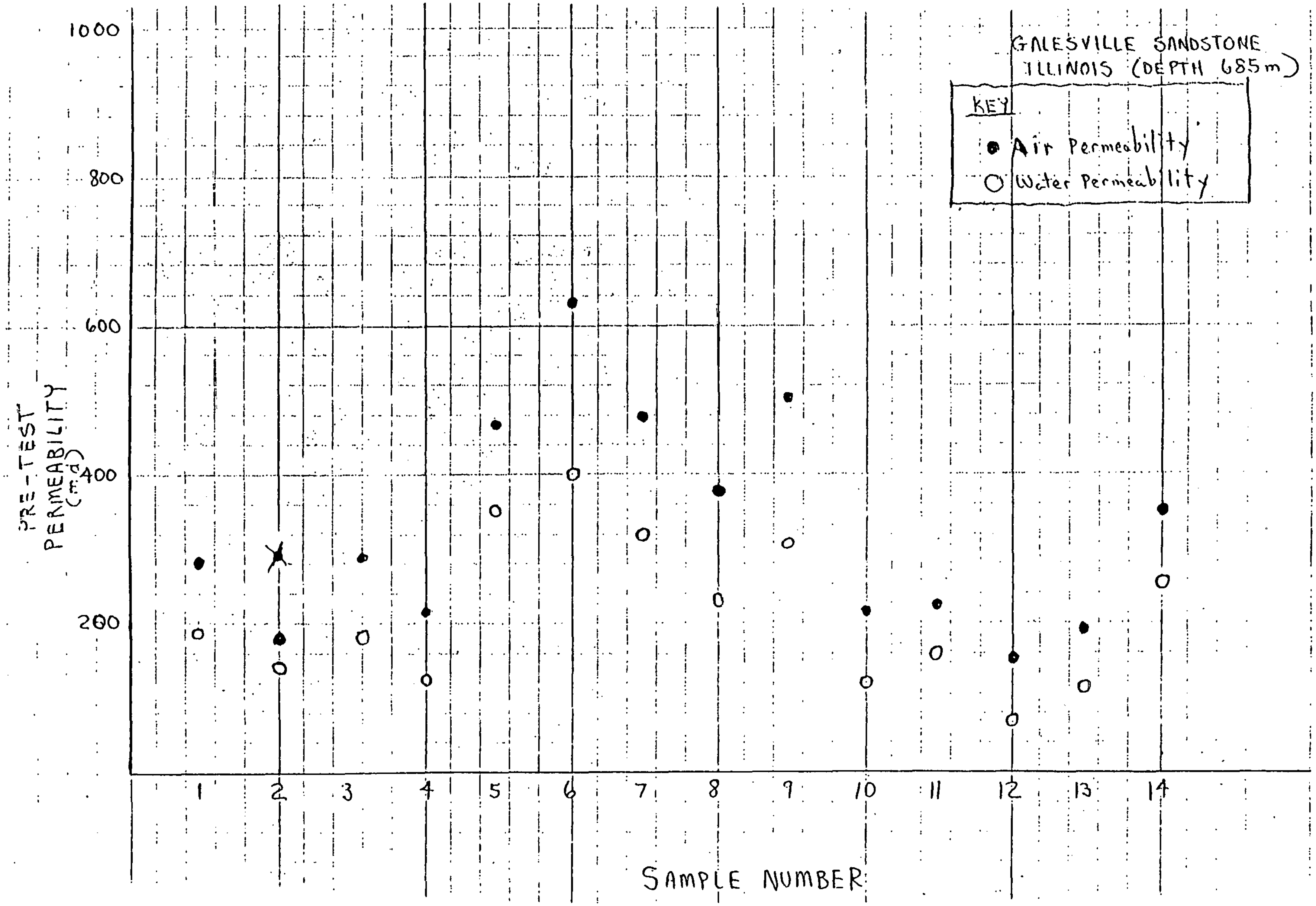
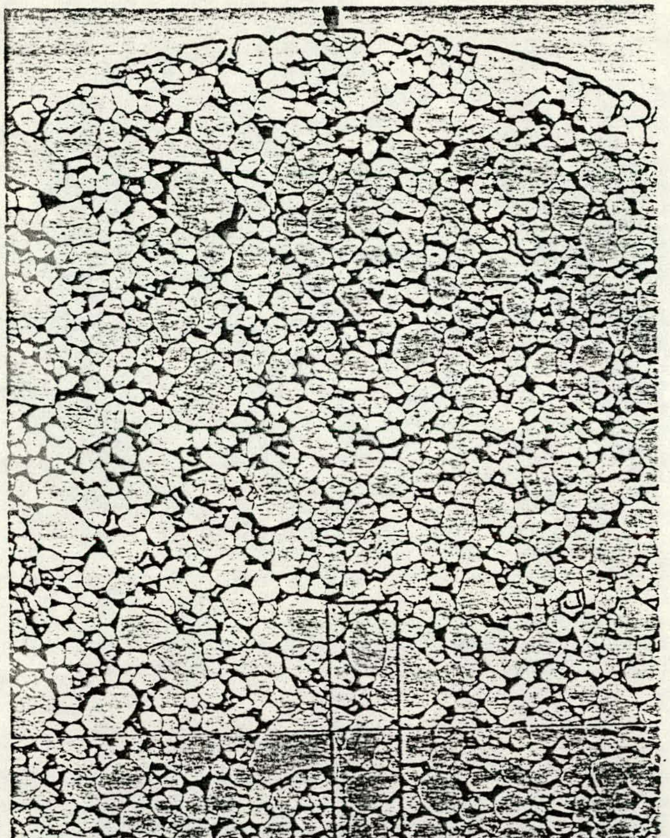
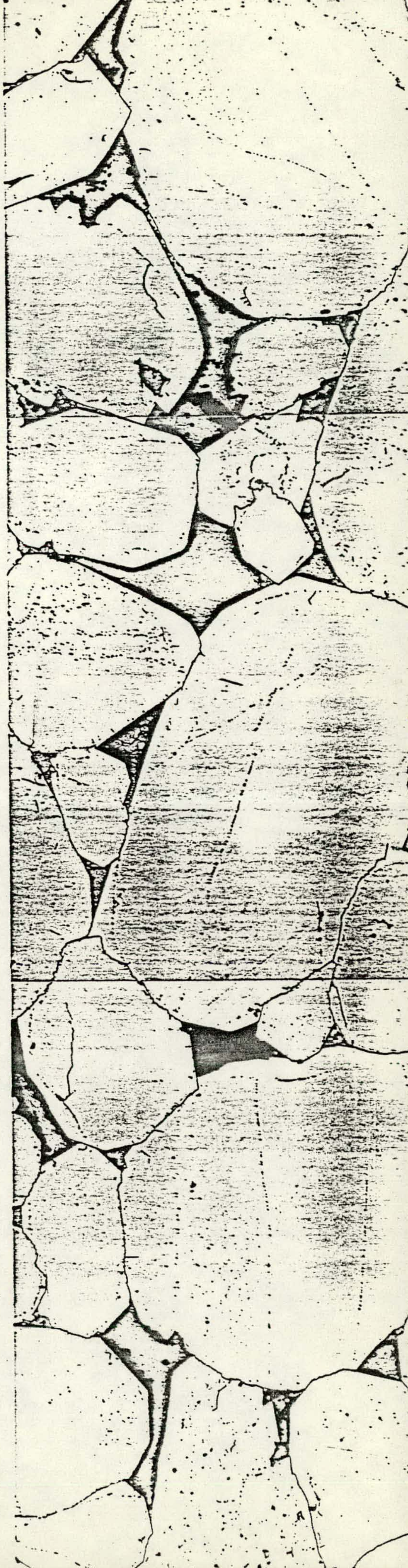


Figure 1. Pre-Test Permeabilities for Galesville Sandstone Samples





SAMPLE - D

Figure 2. Micrograph of Galesville Sandstone

Air Permeability

Table 2 illustrates the percent changes in air permeability for various combinations of the independent variable magnitudes. Figure 3 presents the same data in a series of three dimensional plots. For example in Figure 3a the changes in air permeability are shown in the circles as either increasing (↑) a specified percentage over the pretest level or decreasing (↓) a certain percentage. The independent variables are temperature, time and relative humidity. The sandstone cores were reacted in a totally dry air environment (D), an air and water vapor environment (V), or a totally liquid water environment (L). Figure 4 is a linear response plot of air permeability changes as a function of temperature for dry and wet sandstone cores.

Observations and conclusions are as follows:

- Temperature and humidity are the dominant independent variables
- The length of the experiment has a moderate effect.
- Pressure, oxygen content, carbon dioxide content and autoclave stirring frequency have negligible effects and can be held constant in all future experiments on the Galesville.
- Sample inhomogeneity is the main contributor to the data scatter.
- The mean net air permeability change was approximately 28 ± 10 percent but may be unrealistically high because of the lack of confining stress during the experiments and in some cases severe erosion of the core's cylindrical face.
- The sandstone cores demonstrate good stability with respect to air permeabilities if one or both of the following conditions are met: a) temperatures of 50-100^{0}C, and/or b) negligible humidity.
- The observed increase in porosity and permeability is probably due to the solutioning of silica and carbonate cement since these increases are only observed when the core is saturated with liquid and/or vapor phase water.
- The increase in permeability if it occurs in the field might be beneficial to CAES operations.

TABLE 2. Test Matrix for Air Permeability Screening Experiments

		Pressure (bars)	Temperature (°C)	Time (mnths)	Oxygen (%)	CO_2 (%)	Stirring (Yes-No)	Liquid (Yes-No)	Vapor (Yes-No)	Unassigned			Air Permeability (% change)
Trial	Mean	x_1	x_2	x_3	x_4	x_5	x_6	x_7	x_8	x_9	x_{10}	x_{11}	
1	+	+(120)	+(300)	-(0.5)	+(21)	+(0.1)	+(Y)	-(N)	-(N)	-	+	-	03
2	+	+(120)	-(50)	+(2.0)	+(21)	+(0.1)	-(N)	-(N)	-(N)	+	-	+	02
3	+	-(60)*	+(300)	+(2.0)	+(21)	-(0.0)	- N	- N	+ Y	-	+	+	31
4	+	+(120)	+(300)	+(2.0)	-(0)	-(0.0)	-(N)	+(Y)	-(N)	+	+	-	65
5	+	+(120)	+(300)	-(0.5)	-(0)	-(0.0)	+(Y)	-(N)	+(Y)	+	-	+	76
6	+	+(120)	-(50)	-(0.5)	-(0)	+(0.1)	-(N)	+(Y)	+(Y)	-	+	+	17
7	+	-(60)	-(50)	-(0.5)	+(21)	-(0.0)	+(Y)	+(Y)	-(N)	+	+	+	03
8	+	-(60)	-(50)	+(2.0)	-(0)	+(0.1)	+ Y	- N	+ Y	+	+	-	18
9	+	-(60)*	+(300)	-(0.5)	+(21)	+(0.1)	- N	+ Y	+ Y	+	-	-	96
10	+	+(120)	-(50)	+(2.0)	+(21)	-(0.0)	+ Y	+ Y	+ Y	-	-	-	11
11	+	-(60)*	+(300)	+(2.0)	- 0	+(0.1)	+ Y	+ Y	- N	-	-	+	12
12	+	-(60)	-(50)	-(0.5)	-(0)	-(0.0)	- N	- N	- N	-	-	-	02
SUM +	336	174	283	139	146	148	154	204	249	184	137	141	
SUM -	0	162	53	197	190	188	182	132	87	152	199	195	
OVER-ALL SUM	336	336	336	336	336	336	336	336	336	336	336	336	
DIFF.	336	12	230	- 58	- 44	- 40	- 28	72	162	32	- 62	- 54	
EFFECT	28	2.0	38.3	-9.7	-7.3	-6.7	-4.7	12.0	27.0	5.3	-10.3	-9.0	
RANKING			1	4				3	2				

$$S_{FE} = \sqrt{\frac{UFE_1^2 + UFE_2^2 + \text{---}\ UFE_k^2}{q}} = 8.5 \qquad (MIN) = t \cdot S_{FE} = (2.2)\ (8.5) = 18.7$$

*Actual pressure increased to prevent flashing

GALESVILLE SANDSTONE

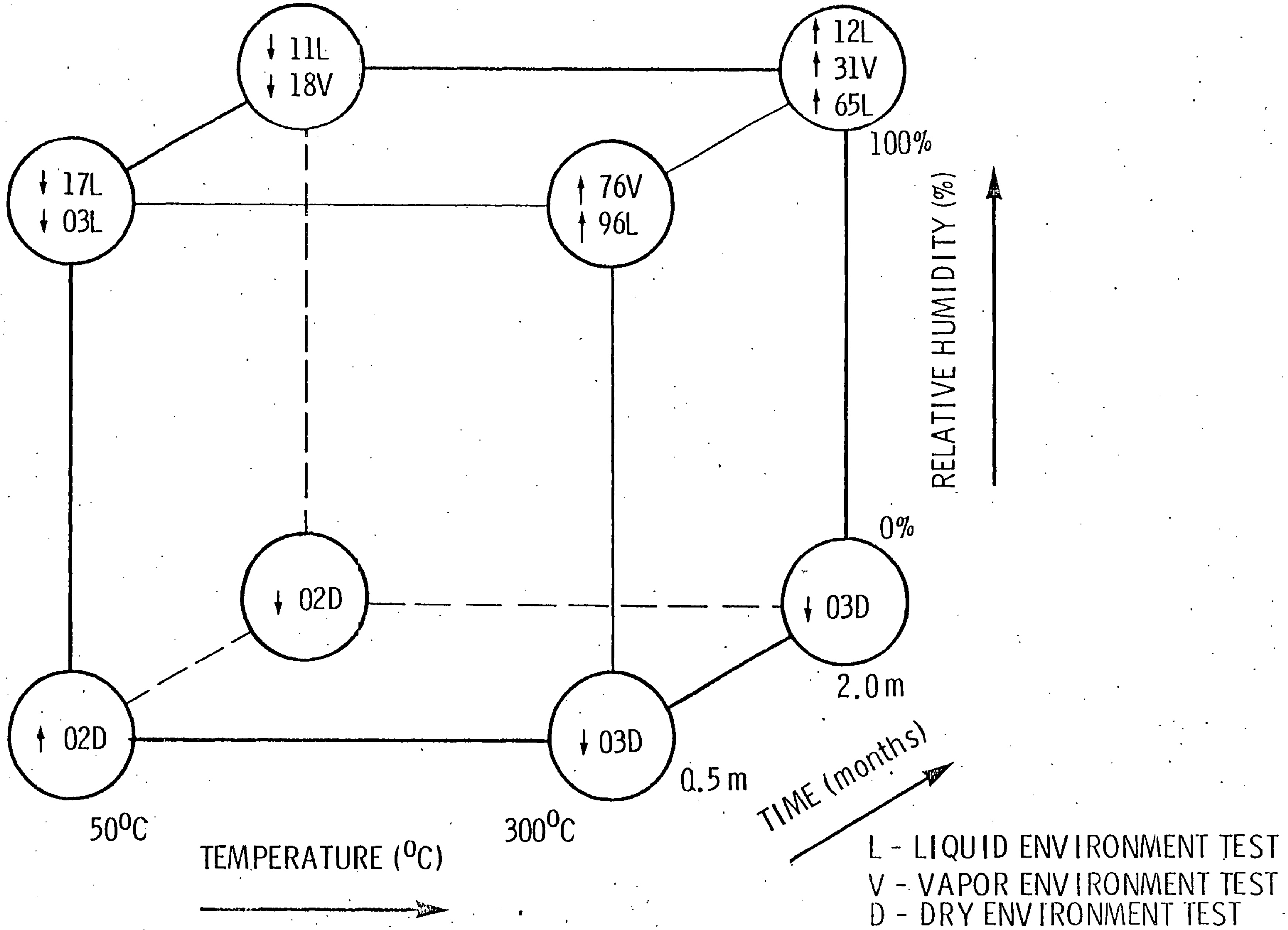


Figure 3a. Percent Changes in Air Permeability for Various Combinations of Temperature, Time and Humidity

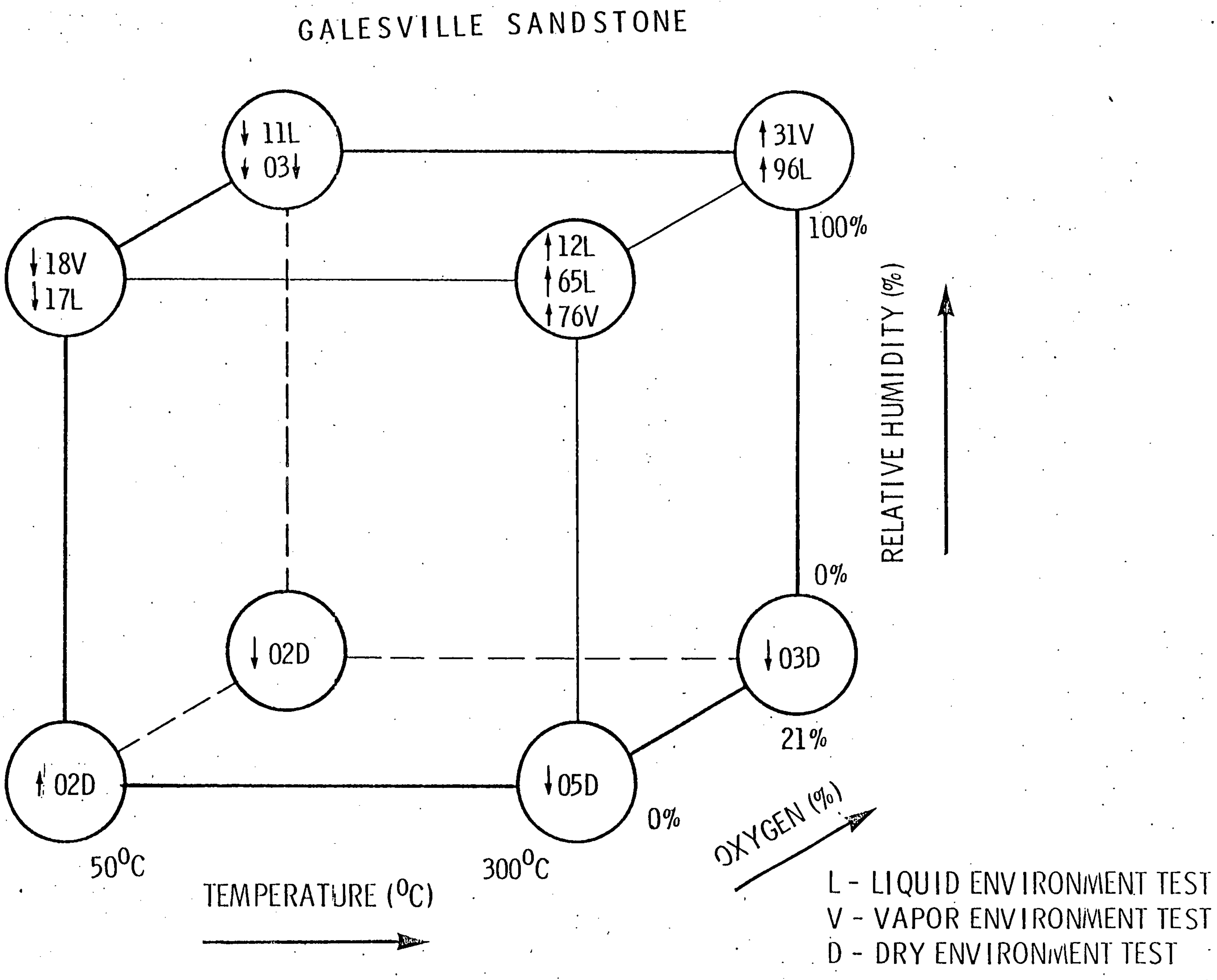


Figure 3b. Percent Changes in Air Permeability for Various Combinations of Temperature, Oxygen and Humicity

GALESVILLE SANDSTONE

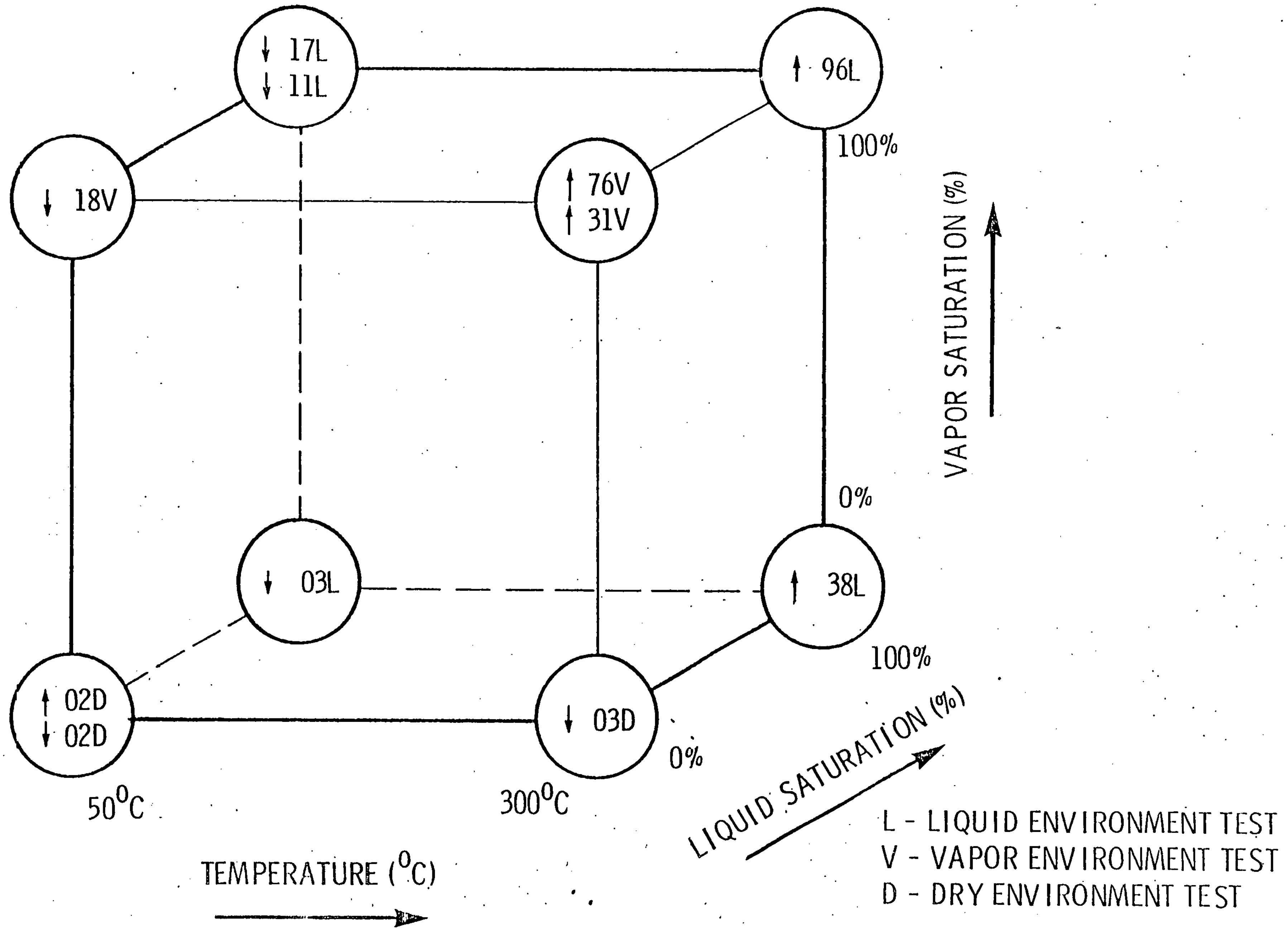


Figure 3c. Percent Change in Air Permeability for Various Combinations of Temperature, Liquid Saturation, and Vapor Saturation

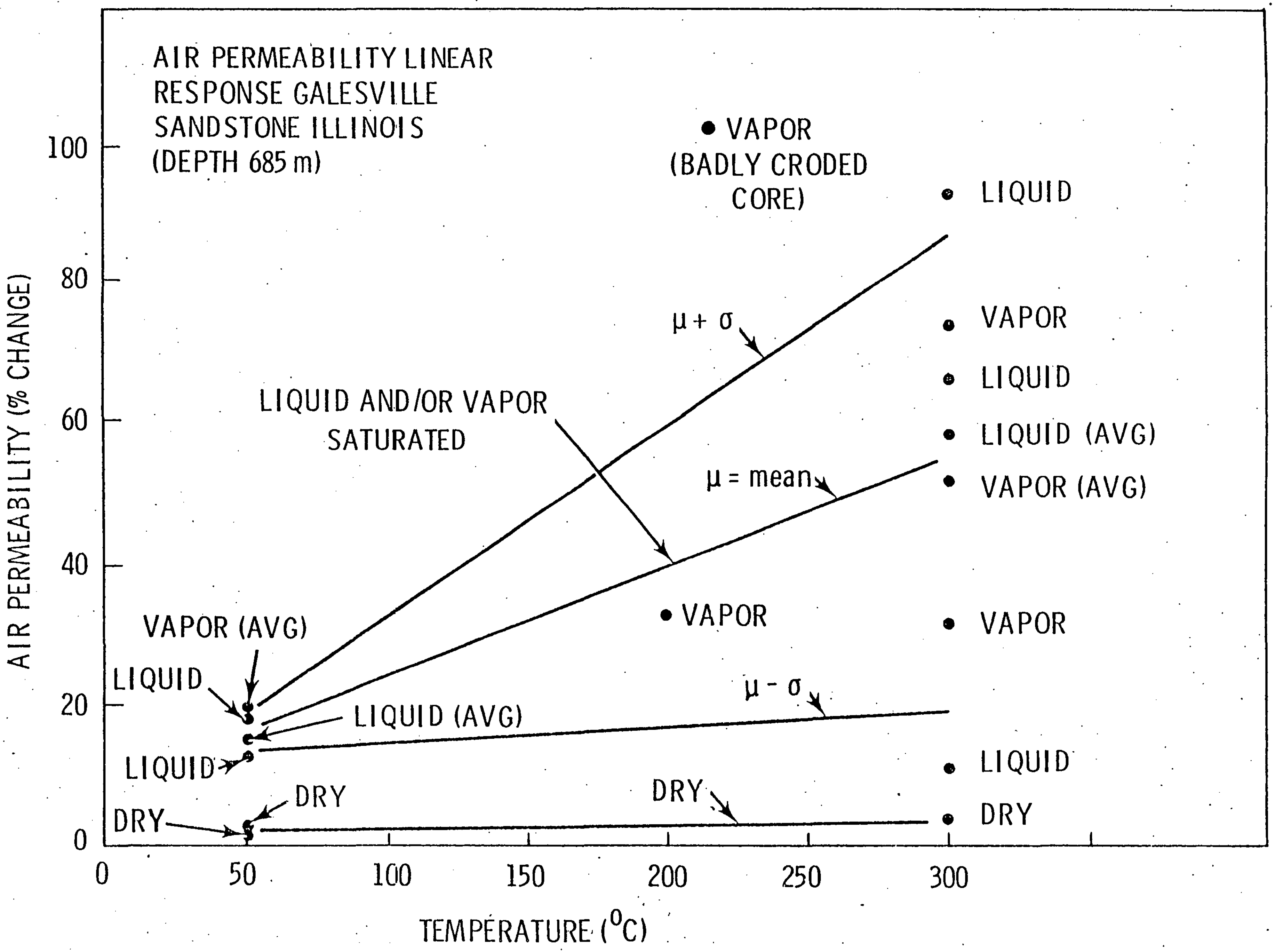


Figure 4. Air Permeability Variations Versus Temperature

Liquid (Water) Permeability

Figure 5 presents the changes in liquid water permeability for various combinations of temperature, time, humidity, and oxygen content. The primary observations are as follows:

- Humidity, time, and temperature are the dominant independent variables.
- A more rapid response is observed for a vapor dominated test than for a liquid dominated one.
- Oxygen content, carbon dioxide content, pressure, and stirring frequency have essentially negligible contributions to the observed changes.
- The mean increase in liquid permeability was approximately 32 ± 10 percent.

Matrix Friability Index

Friability refers to the potential for a sandstone or other rock to disaggregate. It is somewhat analagous to the grain to grain contact strength. The measurements were made with a modified modulus of rupture apparatus common to soil physics applications. Table 3 presents the results of the test matrix. Figure 6 is a set of three dimensional representations of the percent changes in the friability index after treatment of the samples in the autoclave. The linear response plot of friability as a function of temperature is shown in Figure 7. Figure 8 presents some photographs of the cores before and after testing under various conditions. The main observations are as follows:

- Temperature, time, and liquid saturation are the dominant independent variables.
- Oxygen content and vapor saturation also have significant impacts.
- Pressure, carbon dioxide content, and stirring frequency are apparently insignificant.
- All combinations of independent variables tested resulted in some increase in friability.
- The lack of confining stress on the core contributes to the disaggregation.

TABLE 3. Test Matrix for Friability Screening Experiments

Trial	Mean	Pressure (bars) x_1	Temperature (°C) x_2	Time (mnths) x_3	Oxygen (%) x_4	CO_2 (%) x_5	Stirring (Yes-No) x_6	Liquid (Yes-No) x_7	Vapor (Yes-No) x_8	Unassigned x_9	x_{10}	x_{11}	Friability Index (% change)
1	+	+(120)	+(300)	-(0.5)	+(21)	+(0.1)	+(Y)	-(N)	-(N)	-	+	-	27
2	+	+(120)	-(50)	+(2.0)	+(21)	+(0.1)	-(N)	-(N)	-(N)	+	-	+	41
3	+	-(60)*	+(300)	+(2.0)	+(21)	-(0.0)	- N	- N	+ Y	-	+	+	222
4	+	+(120)	+(300)	+(2.0)	-(0)	-(0.0)	-(N)	+(Y)	-(N)	+	+	-	474
5	+	+(120)	+(300)	-(0.5)	-(0)	-(0.0)	+(Y)	-(N)	+(Y)	+	-	+	185
6	+	+(120)	-(50)	-(0.5)	-(0)	+(0.1)	-(N)	+(Y)	+(Y)	-	+	+	29
7	+	-(60)	-(50)	-(0.5)	+(21)	-(0.0)	+(Y)	+(Y)	-(N)	+	+	+	43
8	+	-(60)	-(50)	+(2.0)	-(0)	+(0.1)	+ Y	- N	+ Y	+	+	-	26
9	+	-(60)*	+(300)	-(0.5)	+(21)	+(0.1)	- N	+ Y	+ Y	+	-	-	171
10	+	+(120)	-(50)	+(2.0)	+(21)	-(0.0)	+ Y	+ Y	+ Y	-	-	-	61
11	+	-(60)*	+(300)	+(2.0)	- 0	+(0.1)	+ Y	+ Y	- N	-	-	+	480
12	+	-(60)	-(50)	-(0.5)	-(0)	-(0.0)	- N	- N	- N	-	-	-	31
SUM +	1790	817	1559	1304	565	774	822	1258	694	940	821	1000	
SUM -	0	973	231	486	1225	1016	968	532	1096	850	969	790	
OVER-ALL SUM	1790	1790	1790	1790	1790	1790	1790	1790	1790	1790	1790	1790	
DIFF.	1790	-156	1328	818	-660	-242	-146	726.0	-402	90	-148	210	
EFFECT	149.2	-26.0	221.3	136.3	-110.0	-40.3	-24.3	121.0	-67.0	15.0	-24.7	35.0	
RANKING			1	2	4			3					

$$S_{FE} = \sqrt{\frac{UFE_1^2 + UFE_2^2 + \text{---}\ UFE_k^2}{q}} = 26.2 \qquad (MIN) = t \cdot S_{FE} = (2.2)\ (26.2) = 57.6$$

*Actual pressure increased to prevent flashing

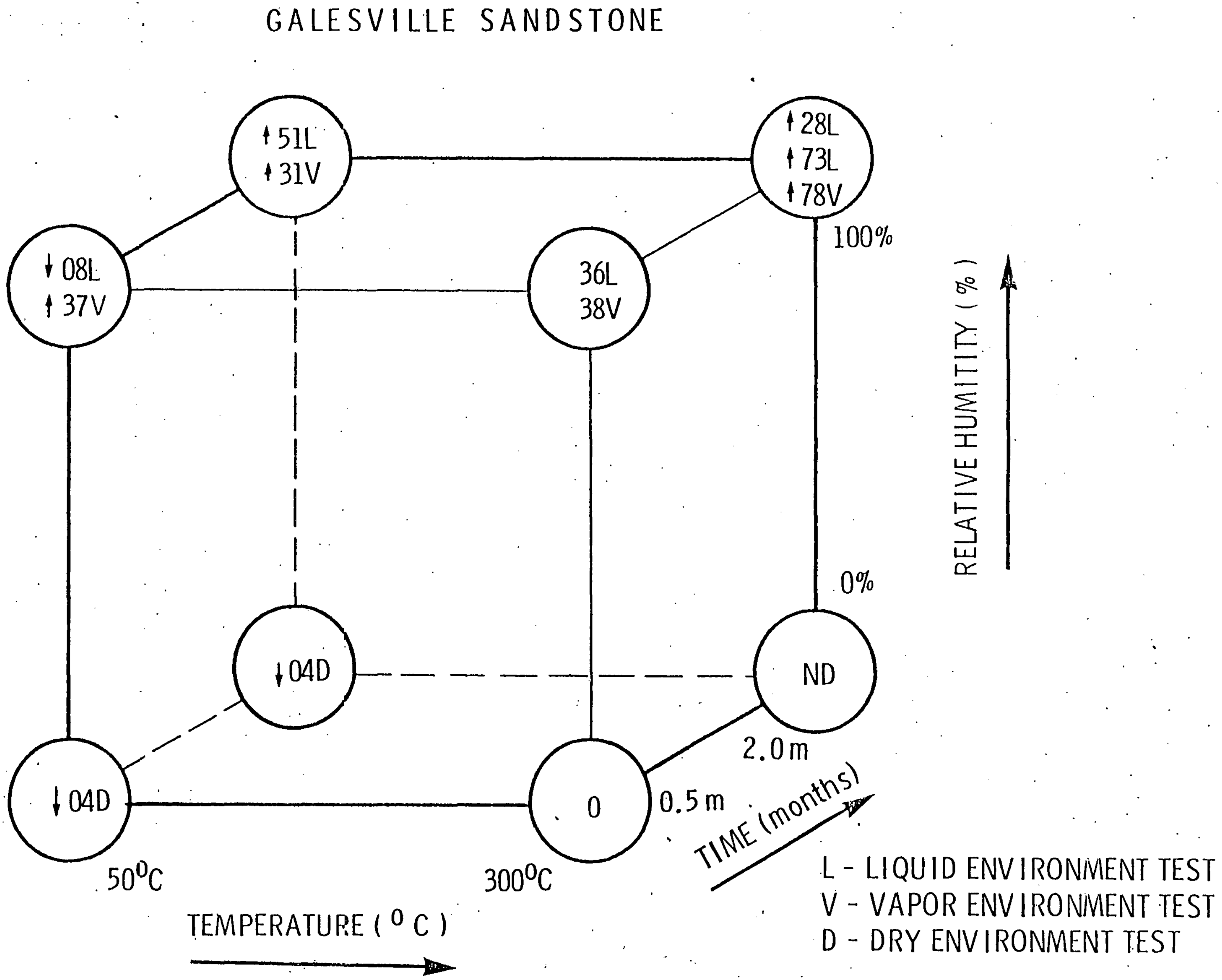


Figure 5. Percent Change in Liquid Permeability for Various Combinations of Temperature, Time and Relative Humidity

GALESVILLE SANDSTONE

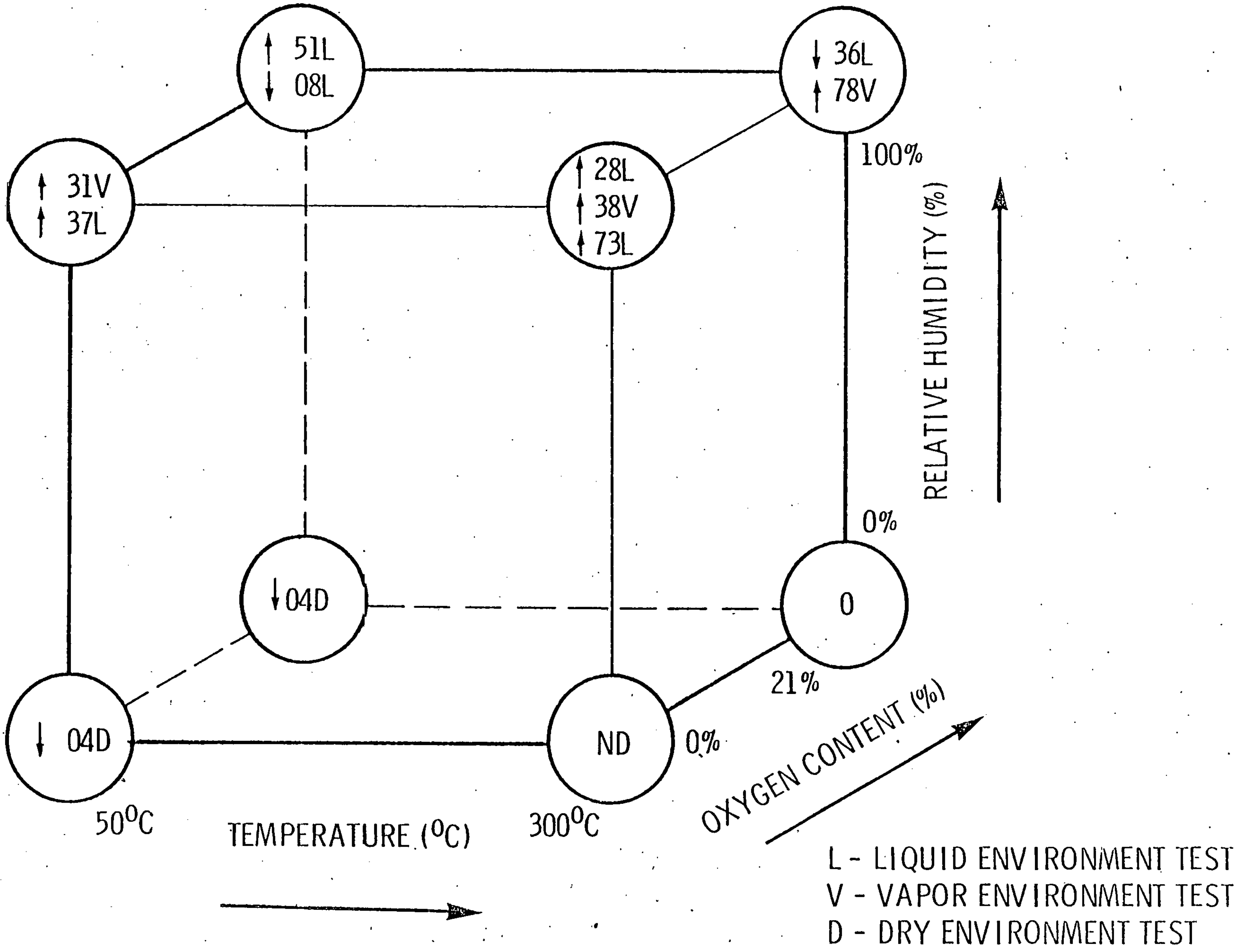
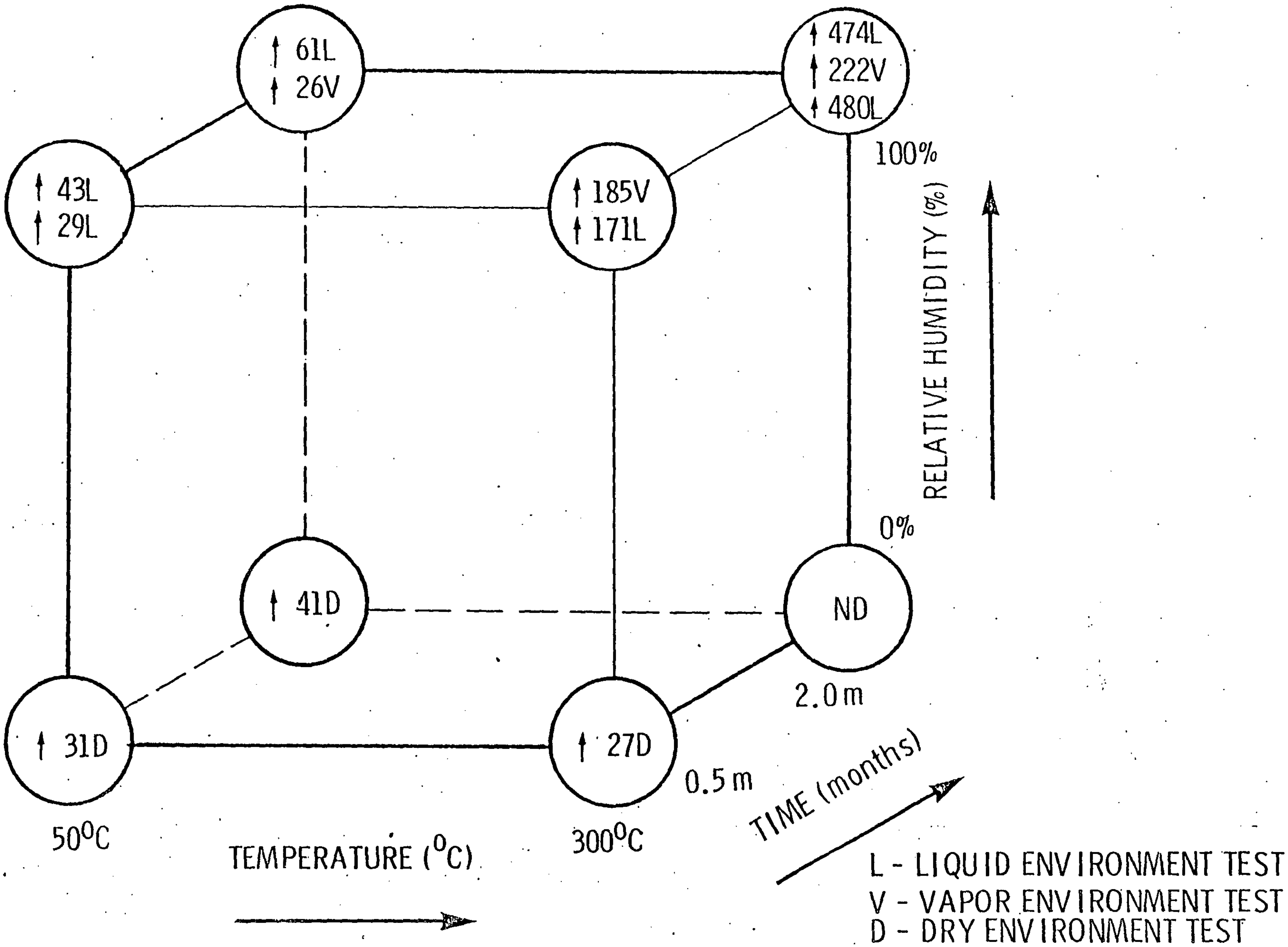


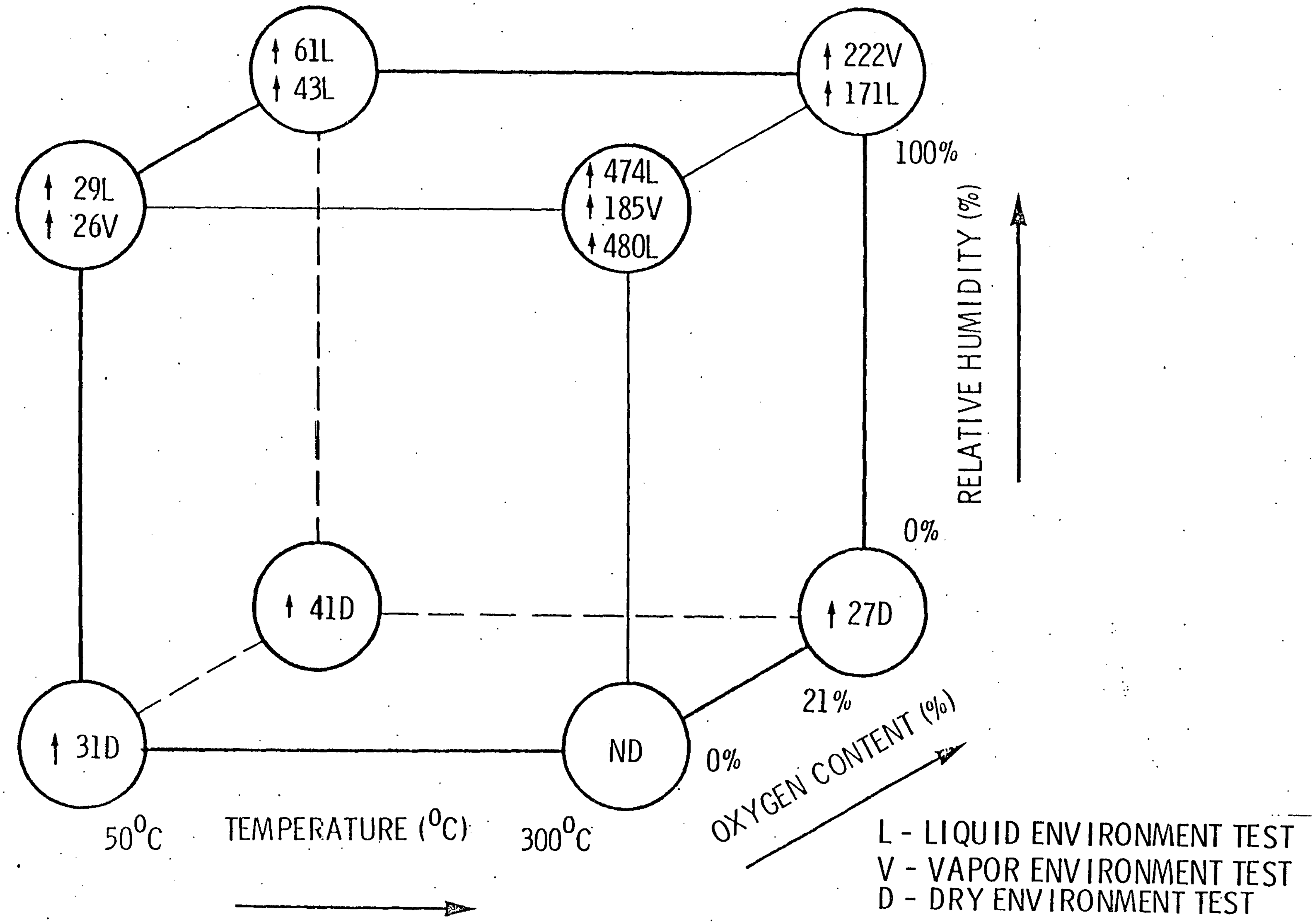
Figure 5b. Percent Change in Liquid Permeability for Various Combinations of Temperature, Oxygen Content, and Relative Humidity



GALESVILLE SANDSTONE

6a. Percent Changes in Friability Index for Various Combinations of Temperature, Time, and Humidity

GALESVILLE SANDSTONE



6b. Percent Changes in Friability Index for Various Combinations of Temperature, Oxygen Content, and Relative Humidity

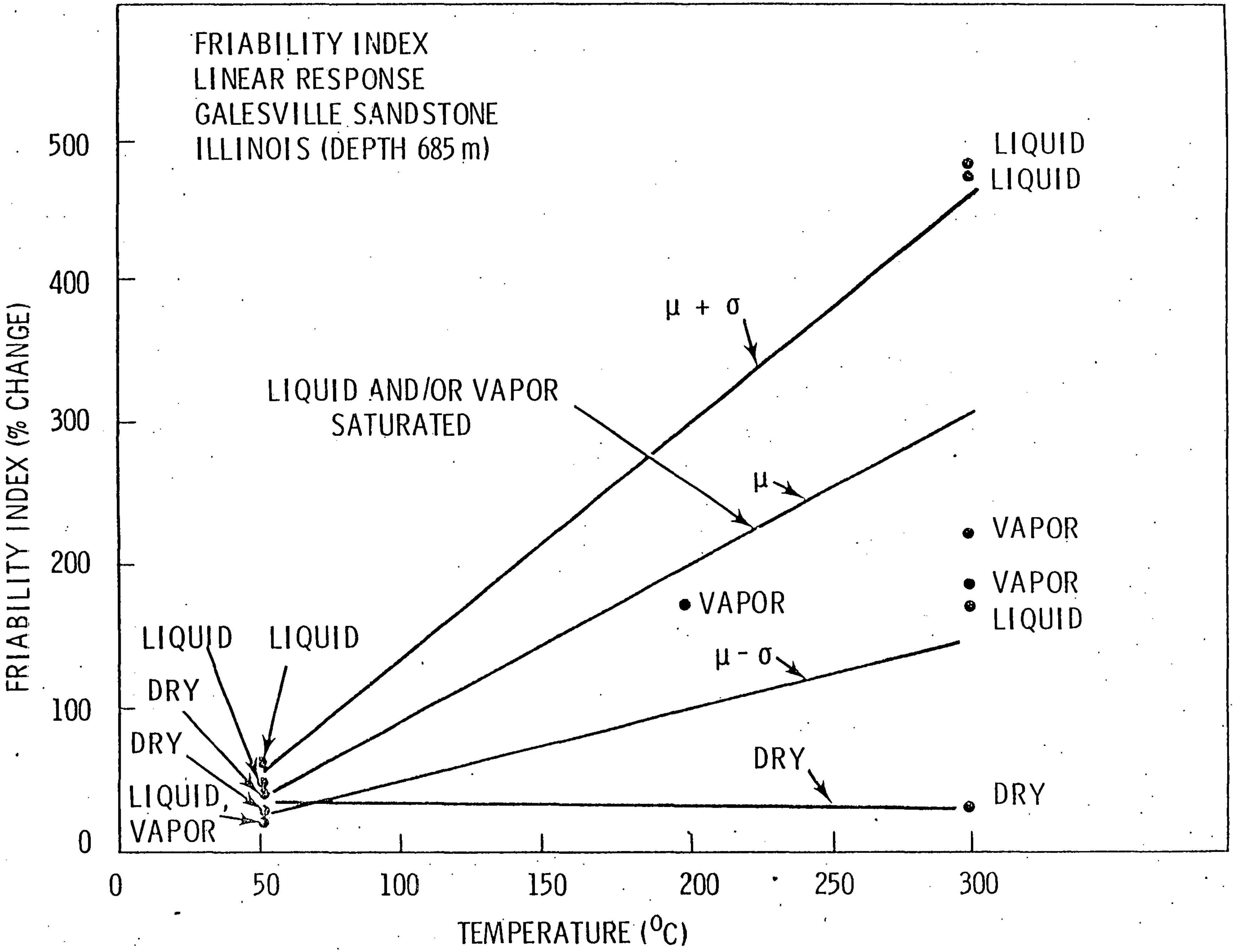
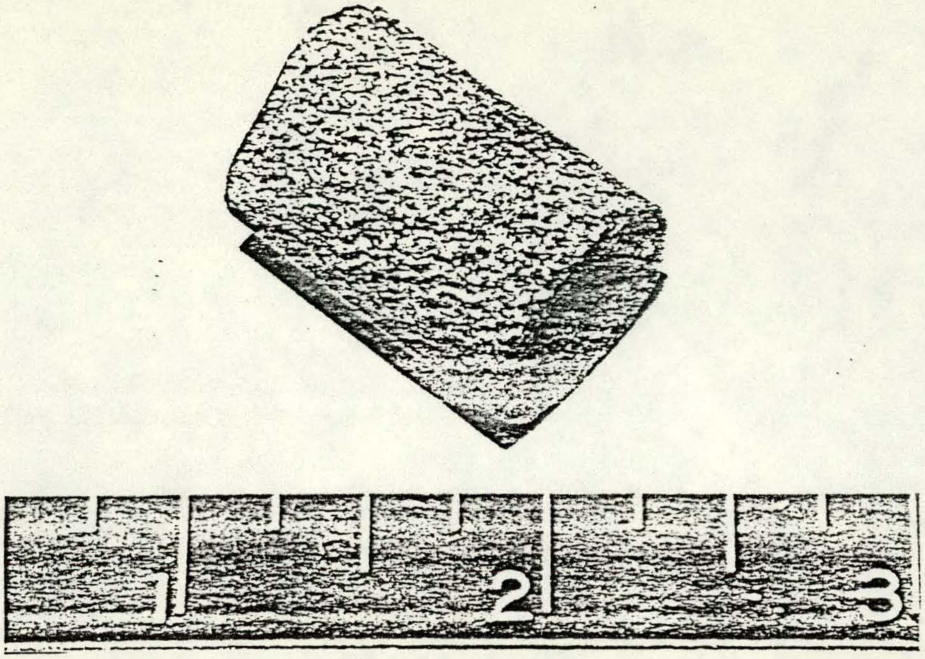
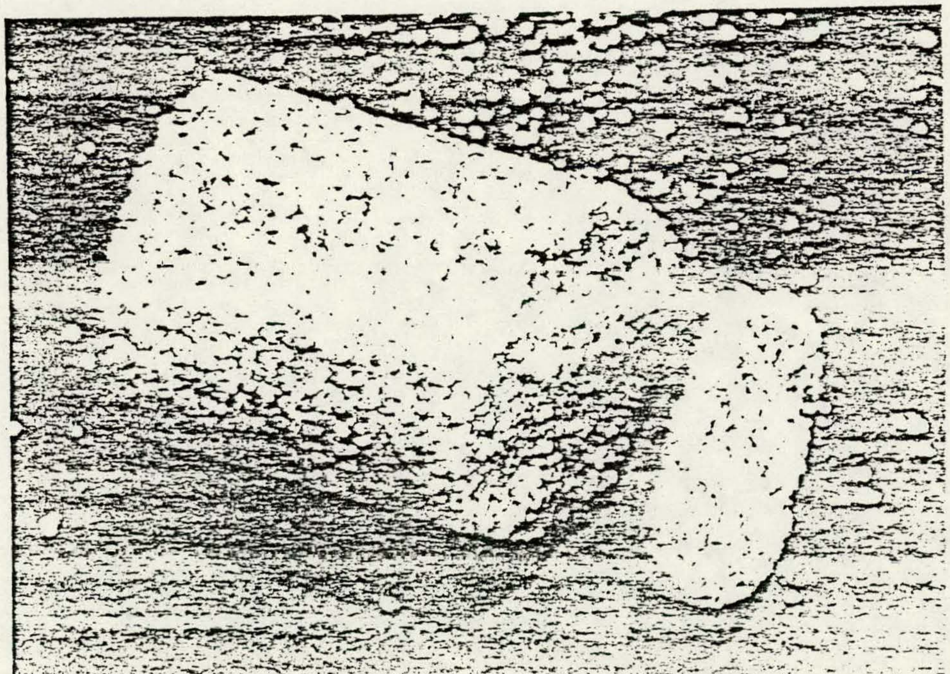


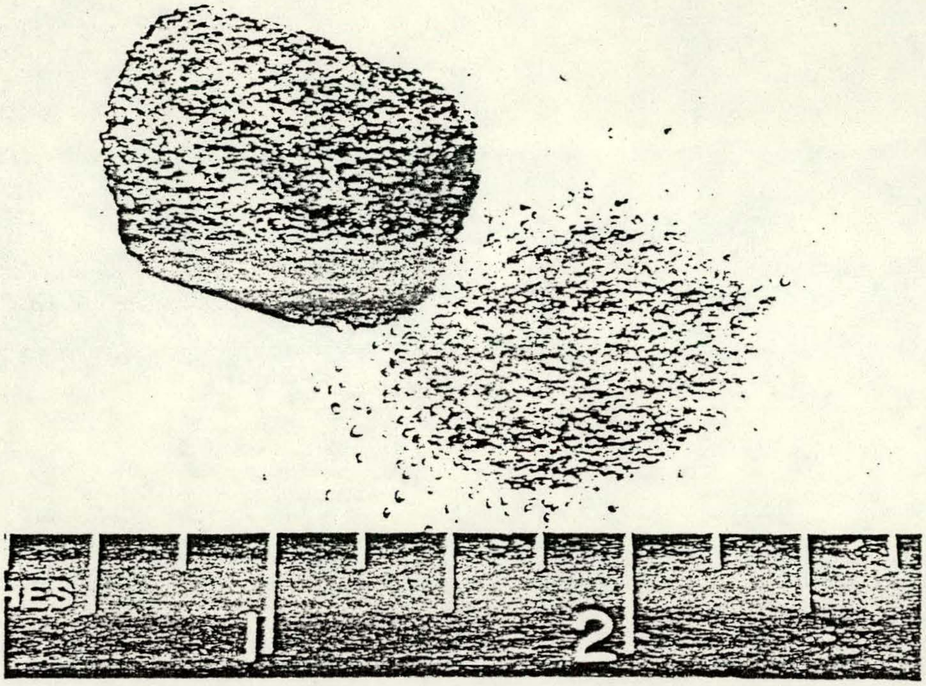
Figure 7. Friability Index Variations Versus Temperature



(50^{0}C, 336 hours, Liquid)



(300^{0}C, 1488 hours, Liquid)



($300^{o}C$, 336 hours, Vapor)

Figure 8a, 8b, 8c. Increasing Friability of Galesville Samples Reacted in a High Humidity Environment at Elevated Temperature.

- The disaggregation is probably due to solutioning of the interstitial cements (silica and carbonate).
- The disaggregation probably accounts for the increased permeabilities.
- Disaggregation may not be a problem for a CAES reservoir, however, the potential for consolidation and associated subsidence and for particle plugging must be assessed.

SUMMARY

The objective of this experimental task is to investigate how anticipated CAES operating conditions might perturb the mineralogical and physical characteristics of host rock formations. The first phase of the program involves reacting sandstone cores in an autoclave facility supporting one of three environments: 1) totally dry air, 2) partially saturated (air and water vapor), and 3) totally liquid saturated.

The dry air tests and partially saturated tests are designed to simulate CAES field conditions. The liquid saturated tests provide a vehicle for real time monitoring of groundwater chemical changes that may be attributable to solution, hydrolytic, and redox reactions. Similar reactions may take place in the partially saturated tests but are more difficult to detect during the experiment. The liquid tests may also simulate isolated, low permeability regions within the reservoir.

The physical properties of primary interest include porosity, air permeability, liquid permeability, and friability. Significant changes in Galesville sandstone properties were observed for certain combinations of temperature, humidity, and experiment length. The main observations and conclusions are as follows:

- The Galesville sandstone (Media natural gas storage field in Illinois) appears to be a reasonable CAES candidate rock. It is a clean sandstone (almost 95% silica) and supports an air permeability ranging from 100 to 1500 md.
- The physical property changes observed during the experiments may be due, totally or in part, to the lack of confining stress on the sample. The autoclaves cannot reproduce the effective

stress regime that will actually exist in a CAES reservoir. The flow facility which is currently in the design phase should correct this short coming. Similar comments can be made concerning the fluid to solid volume ratio.

- The observed increases in porosity, permeability, and friability are well correlated with increased temperature, increased humidity, and increased testing time. The property changes are apparently insensitive to the fluid pressure, the presence of oxygen or carbon dioxide, and the autoclave type or stirring frequency.
- The changes are apparently not a result of mechanical degradation such as microcracking or differential thermal expansion and contraction. This conclusion is based on the fact that negligible changes were observed for a dry air environment regardless of the testing temperature. However, this was not true for the case in which a dry core was thermally cycled from 250-300°C several times. The air and liquid water permeabilities were not significantly altered, but the friability increased by a factor of two.
- The changes in physical properties are not due to hydrolytic or redox reactions.
- The changes are apparently due to solutioning of the silica cement that bonds the individual grains. Such solutioning is at least as active in an air and water vapor environment as it is in a liquid water environment.
- The cement is almost exclusively silica, but some carbonate material exists and is rapidly removed from the sandstone as well. Silica solubility increases from 14 mg/l at 50°C to 800 mg/l at 300°C. Carbonate has a retrograde solubility, i.e., decreases with temperature.
- Removal of the cement increases the porosity and permeability of the sandstone, but unfortunately also increases its friability or disaggregation potential. This could conceivably result in particle plugging of the formation and/or consolidation and subsidence.

- Air has no effect on the Galesville sandstone, but is very detrimental to well casing alloys, if any moisture is present.
- The sandstone exhibits good stability at all temperatures between 50 and 300°C if tested with dry air. However, if the rock is exposed to vapor and/or liquid water at temperatures above 100°C, significant physical degradation may occur. Such degradation may be beneficial to CAES operations, however.
- Preliminary tests on Berea sandstone and Mt. Simon sandstone reveal trends in physical property changes similar to those observed for Galesville sandstone.
- One caprock type for the Media natural gas storage field in Illinois is the Eau Claire which is predominantly shale and siltstone. Samples of this rock showed reasonable chemical and physical stability when heated to 300°C in an air and/or water vapor environment.