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SANDIA REPORT SAND85-2482 • Unlimited Release • UC-70
Printed December 1985

Liquid Permeability Measurements on Densely Welded Tuff Over the Temperature Range 25° to 90°C

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Prepared by
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for the United States Department of Energy
under Contract DE-AC04-76DP00789



HYDROLOGY DOCUMENT NUMBER 129

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Liquid Permeability Measurements on Densely Welded Tuff over the Temperature Range 25° to 90°C

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ABSTRACT

The Topopah Spring welded unit in the unsaturated zone at Yucca Mountain, on and adjacent to the Nevada Test Site, is currently being studied for consideration as the host unit for a radioactive waste repository. The U. S. Department of Energy is carrying out these studies through the Nevada Nuclear Waste Storage Investigations (NNWSI) project. In support of this effort, liquid permeability experiments, using distilled and deaerated water as the pore fluid, were conducted on a sample of densely welded tuffaceous material from the Nevada Test Site. The primary independent variable was the core temperature, which was systematically increased, then decreased, over the range 25° to 90°C. Confining pressure was maintained constant at 15.2 MPa. Pore water continually flowed through the tuff sample during an extensive three-month test period. The transient pressure decay technique was utilized to measure core permeability. Geochemical analyses of the pore water exiting the core at 90°C showed increased concentrations of silicon, calcium, sodium, potassium and strontium. Fluid mechanic results showed liquid permeability of the tuff core to be $\approx 3 \times 10^{-19} \text{ m}^2$, independent of temperature.

NOMENCLATURE

A	=	core cross-sectional area
C	=	$[K + \Gamma]$
K	=	compressibility of water
k	=	core permeability
L	=	core length
P	=	pressure
P_c	=	confining pressure
\bar{P}_p	=	average pore pressure, $(P_1 + P_2)/2$
ΔP	=	pressure drop along core, $(P_1 - P_2)$
Q	=	volumetric flow rate
T	=	temperature
t	=	time
V	=	volume of reservoir upstream of core

Greek

Γ	=	system compliance
μ	=	viscosity
ρ	=	density

Subscripts

G	=	gas
i	=	initial
L	=	liquid
1	=	upstream of core
2	=	downstream of core

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1 Introduction

The Topopah Spring welded unit in the unsaturated zone at Yucca Mountain, on and adjacent to the Nevada Test Site (NTS), is currently being studied for consideration as the host unit for a radioactive waste repository. The U. S. Department of Energy is carrying out these studies through the Nevada Nuclear Waste Storage Investigations project. Because water flow through unsaturated rock is the principal mechanism for the transport of soluble radionuclides and other contaminants from a repository to the biosphere at an arid site, the determination of the hydrologic properties of Topopah Spring tuff is an essential part of the analyses of radionuclide transport.

In the present investigation, long-time-scale experiments were conducted on a sample of densely welded tuff to quantify the dependence of its liquid permeability on temperature over the range 25° to 90°C. Such dependence could potentially result from thermal expansion and/or geochemical effects in the heated regions near emplaced high-level nuclear waste canisters. Together with previous measurements of liquid permeability and gas permeability of Topopah Spring tuff matrix material [1,2], measurements from the current nonambient temperature experiments provide essential data on the permeability characteristics of welded tuff near a heat-producing waste canister.

2 Experimental Approach

Figure 1 shows a photograph of the experimental apparatus, while Figure 2 shows it in schematic form. This apparatus can be used in the steady-state mode to measure core permeabilities at and above the microdarcy (10^{-18} m^2) level. A transient, pressure decay technique must be used with this apparatus to measure permeabilities below 10^{-18} m^2 . Details of this apparatus have been reported earlier [1]. The one new feature added to this apparatus for the present investigation was the capability to elevate and control the temperature of the entire core-holder assembly. This was accomplished by wrapping the exterior of the core holder with resistance heater tape and insulation, then using a temperature controller/feedback circuit to monitor and control the resultant temperature level. The temperature of the core specimen was measured at each end, as well as at the midlength station, using sheathed thermocouple probes. Temperature uniformity to within 1°C was achieved in each experiment.

A core of unfractured, densely welded, nonlithophysal Topopah Spring tuff from the Busted Butte outcrop at the Nevada Test Site was selected for study (identified as rock #11, A2, #4). The effective porosity of this sample was measured by weighing the core totally dry versus totally saturated, yielding a value of 0.109. The cylindrical core, of diameter 5.54 cm and length 6.36 cm, was placed in the core holder and subjected to a constant confining pressure of 15.2 MPa. Distilled, deaerated water was used as the working (pore) fluid. Inlet pressure to the core sample was cycled once each workday from 0.5 to 13.8 MPa while the exit pressure was maintained at atmospheric condi-

tions. This procedure kept the pore water continually moving through the specimen throughout the entire test sequence.

The transient pressure decay technique [1] was used herein to measure core permeability. In this approach, one piston of the positive displacement pump was driven forward at a constant rate, elevating the pressure within the piston chamber (reservoir) upstream of the core holder. Reservoir pressure increases, corresponding to measured volume reductions, were recorded and analyzed to determine the characteristic constant C , equal to the sum of the compressibility of water (K) and the system compliance or expansivity (Γ):

$$C \equiv [K + \Gamma] \quad (1)$$

$$K \equiv \frac{1}{\rho} \left. \frac{\partial \rho}{\partial P} \right|_T \quad (2)$$

$$\Gamma \equiv \frac{1}{V} \left. \frac{\partial V}{\partial P} \right|_T \quad (3)$$

Knowing this system constant, the measured pressure decay rate of the reservoir could then be analyzed to determine core permeability, k , as given in Equation (4):

$$k = \frac{\mu V L C}{A} \ln \left[\frac{\Delta P_i}{\Delta P} \right] / t \quad (4)$$

Figure 3, taken from Reference [1], shows an example of a typical pressure decay response, measured at ambient temperature, on a core of welded tuff taken from the same site (Busted Butte outcrop) as the sample studied here.

Four experiments were conducted during the present investigation. The objective of the first experiment was to measure the liquid permeability of tuff at ambient temperature over a long time period (~ 700 hours) to determine if tuff permeability would vary with time. Results could then be compared with previous tuff results [1] obtained during short-time-scale exposures to liquid flows. The objective of the second experiment was to measure the liquid permeability of welded tuff as core temperature was systematically increased from 25° to 90°C. The objective of the third experiment was to investigate the potential time dependence of liquid permeability while maintaining the tuff sample at an elevated temperature level of 90°C. Finally, the objective of the fourth experiment was to measure the liquid permeability of welded tuff as core temperature was systematically reduced from 90° to 25°C, thereby quantifying potential hysteresis effects. Results are presented in the next section.

3 Results

Before presenting current results, previously measured liquid and gas permeability values for Topopah Spring welded-tuff material, as measured by this author, will be reviewed. During short-time-scale exposures to ambient temperature water flows, the

liquid permeability of this tuff matrix material was measured to be in the range $5-6 \times 10^{-19} \text{ m}^2$ [1]. Gas permeability experiments were also conducted on this same matrix material, using ambient temperature nitrogen as the pore fluid [2]. Results showed gas permeability to be a strong function of average pore pressure (see Figure 4, taken from Reference [2]). Measured gas permeability was found to approach the measured liquid permeability range as the average pore pressure became large, *i.e.*, as noncontinuum gas dynamic (slip-flow) effects became negligible. The combined results of [1] and [2] thus provided a starting point from which to investigate the long-time-scale response of welded tuff to water flows at both ambient and elevated temperature levels.

Initial permeability measurements of the present investigation were made at 25°C over a time period of ~ 700 hours to ensure that the liquid permeability was not changing with time prior to the onset of core heating. Results are shown in Figure 5. Under the stated test conditions, liquid permeability was found to decrease slightly, from $5-6 \times 10^{-19} \text{ m}^2$ to $\sim 3 \times 10^{-19} \text{ m}^2$ over the 700-hour test period. This response could potentially be attributed to matrix material deformation (pore size reductions) under the applied compressive load versus time history.

Upon the attainment of a steady-state k_L value at 25°C , the core temperature was uniformly raised in $\approx 15^\circ\text{C}$ increments to a maximum test temperature of 90°C , each temperature level being held constant for ~ 170 hours. The temperature dependence of water viscosity was taken into account in reducing the data to obtain k_L values. Results are shown in Figure 6. No dependence of k_L on temperature was observed. (Note: The repeatability of these results is demonstrated by the dual measurements of k_L at each above-ambient temperature level set during this increasing-temperature segment of the investigation.) The core was then held at 90°C for a period of ~ 600 hours. Results are shown in Figure 7. Measured k_L values were found to remain constant at $3.0 \times 10^{-19} \text{ m}^2 \pm 10\%$. A systematic reduction in core temperature level was then applied, from 90° to 25°C , with results shown in Figure 6. As before, k_L was found to remain constant, independent of temperature. Measured permeability results were found to be consistent with values reported in References [3-8], obtained on different size cores and by different experimental methods. Based upon the available data within NNWSI, the permeability established in the NNWSI Environmental Assessment for calculations of hydrologic flows in Topopah Spring welded-tuff matrix is $2.3 \times 10^{-18} \text{ m}^2$ (0.72 mm/year at ambient temperature)[8].

During the experiments at 90°C , a sample of the water exiting the core was collected. This sample, along with a sample of the distilled inlet water, were provided to J. Husler, University of New Mexico, for geochemical analyses. Excerpts from his written communication [9] are quoted below.

"The two water samples submitted to this laboratory were analyzed by flame atomic absorption spectrophotometry using a Perkin-Elmer Model 303 with Hitachi 165 recorder. All samples and standards were made to contain 1,000 mg/L cesium in 10% HCl. Calibration standards were prepared by dissolving spectrograde pure dry metals or oxides in appropriate acids or fluxes. These were checked by comparing to USGS standard rocks which were dissolved and treated similarly. The Si, Al, Fe, Mg,

Ca, and Ti were analyzed in the N₂O-acetylene flame and Na, K, and Mn were analyzed using air-acetylene. Phosphorus was analyzed by the Molybdenum Blue colorimetric method. The results are given below.”

Table 1: Geochemistry Results

Element	Water In (mg/L)	Water Out (mg/L)
Si	2.4	40
Al	<.2	<.2
Fe	0.5	<.1
Mg	0.5	0.68
Ca	0.25	81
Na	1.5	21
K	<.1	5.9
Ti	<.1	<.1
P	<.1	<.1
Mn	<.02	<.02
Sr	<.01	0.24

These results show increases in the concentrations of silicon, calcium, sodium, potassium and strontium in the pore water after passage through the 90°C tuff core. However, despite these observed geochemical changes in the pore water, liquid permeability was found to be unaffected by temperature increases in the range 25° to 90°C. Similar findings were noted in the recent permeability/geochemistry experiments of Moore, Morrow and Byerlee [5, 6], wherein both distilled and ground water were forced radially outward through hollow-core samples of tuff subjected to radial temperature gradients.

4 Conclusions

Laboratory experiments conducted over a continuous three-month period showed that the liquid permeability of Topopah Spring welded tuff remained essentially constant, independent of temperature, over the range 25° to 90°C. These results indicate that thermal expansion and/or geochemical effects do not influence the liquid permeability of Topopah Spring welded tuffaceous materials. Therefore, it is suggested that these effects can be neglected in both the modeling of hydrological phenomena within Yucca Mountain as well as in the implementation and analyses of future, nonisothermal multiphase transport experiments in Topopah Spring welded tuff [10].

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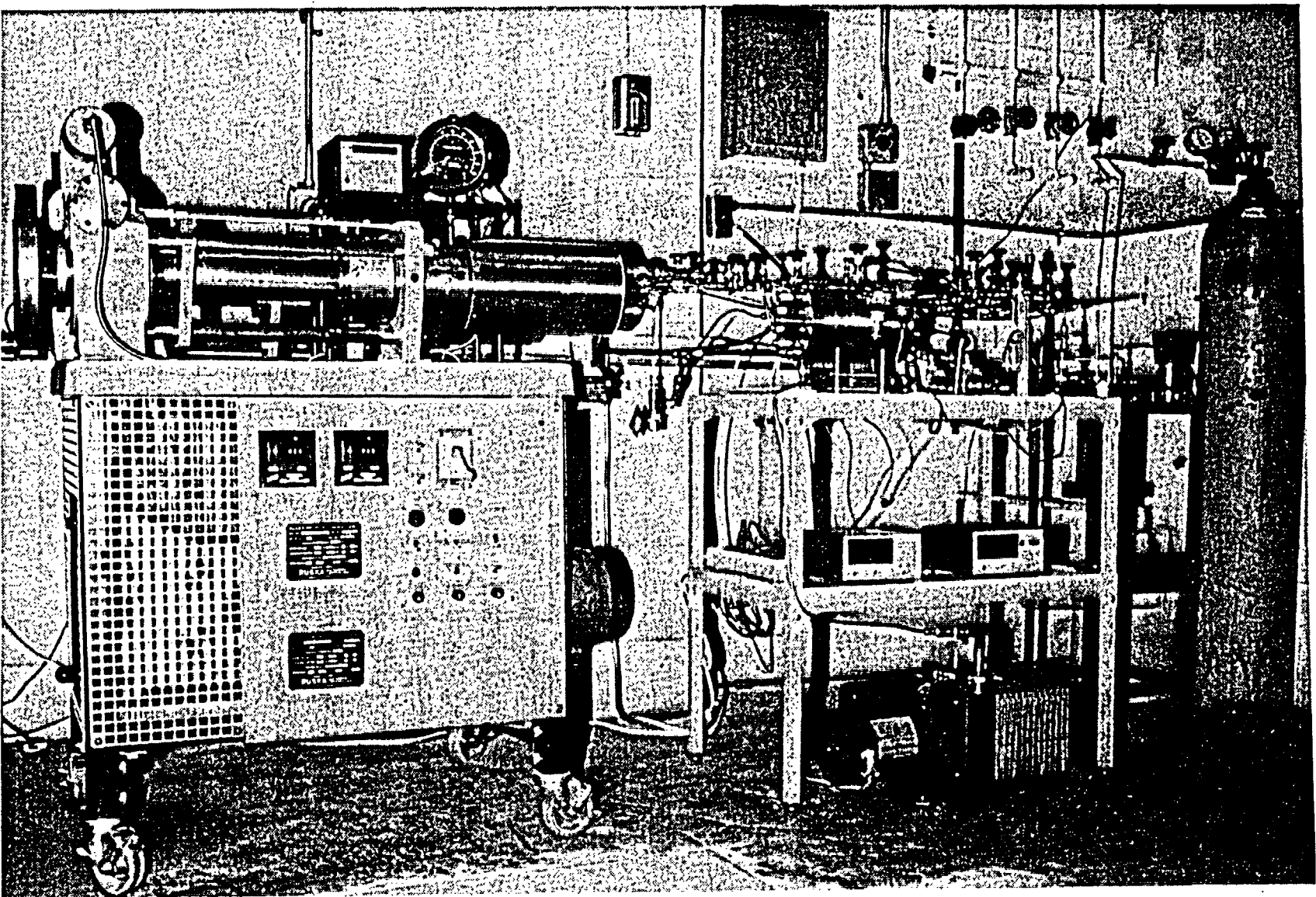
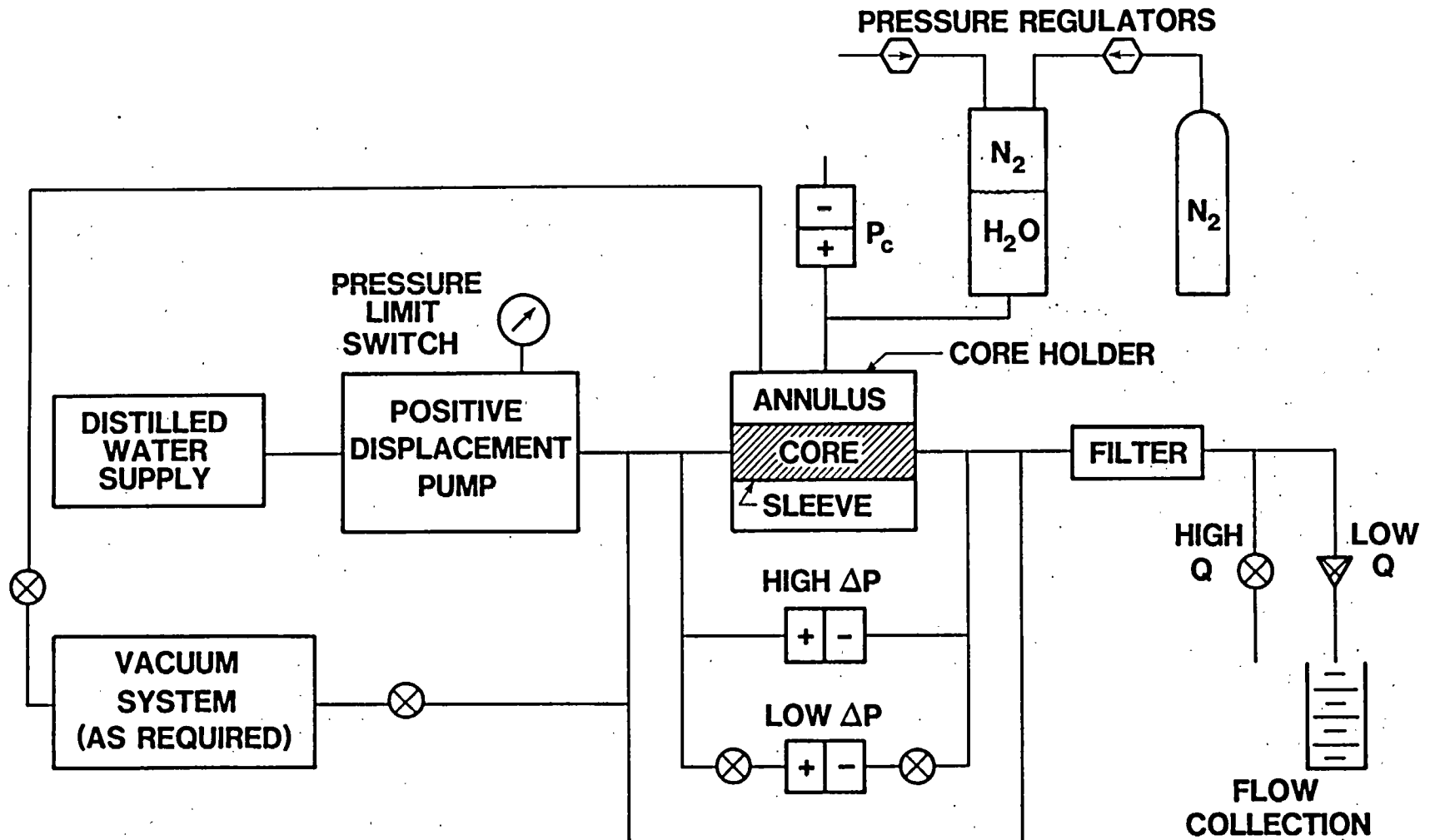


Figure 1: Photograph of Experimental Apparatus

Figure 2: Schematic of Experiment



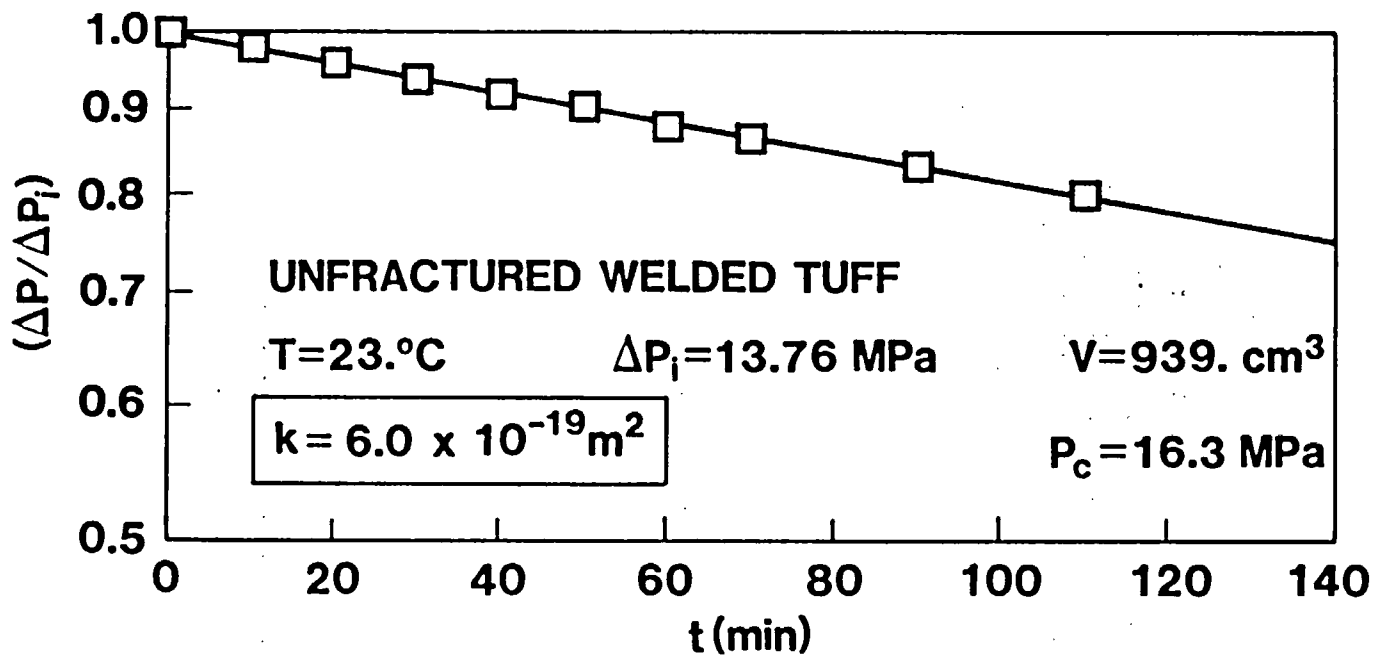


Figure 3: Example of Pressure Decay Curve

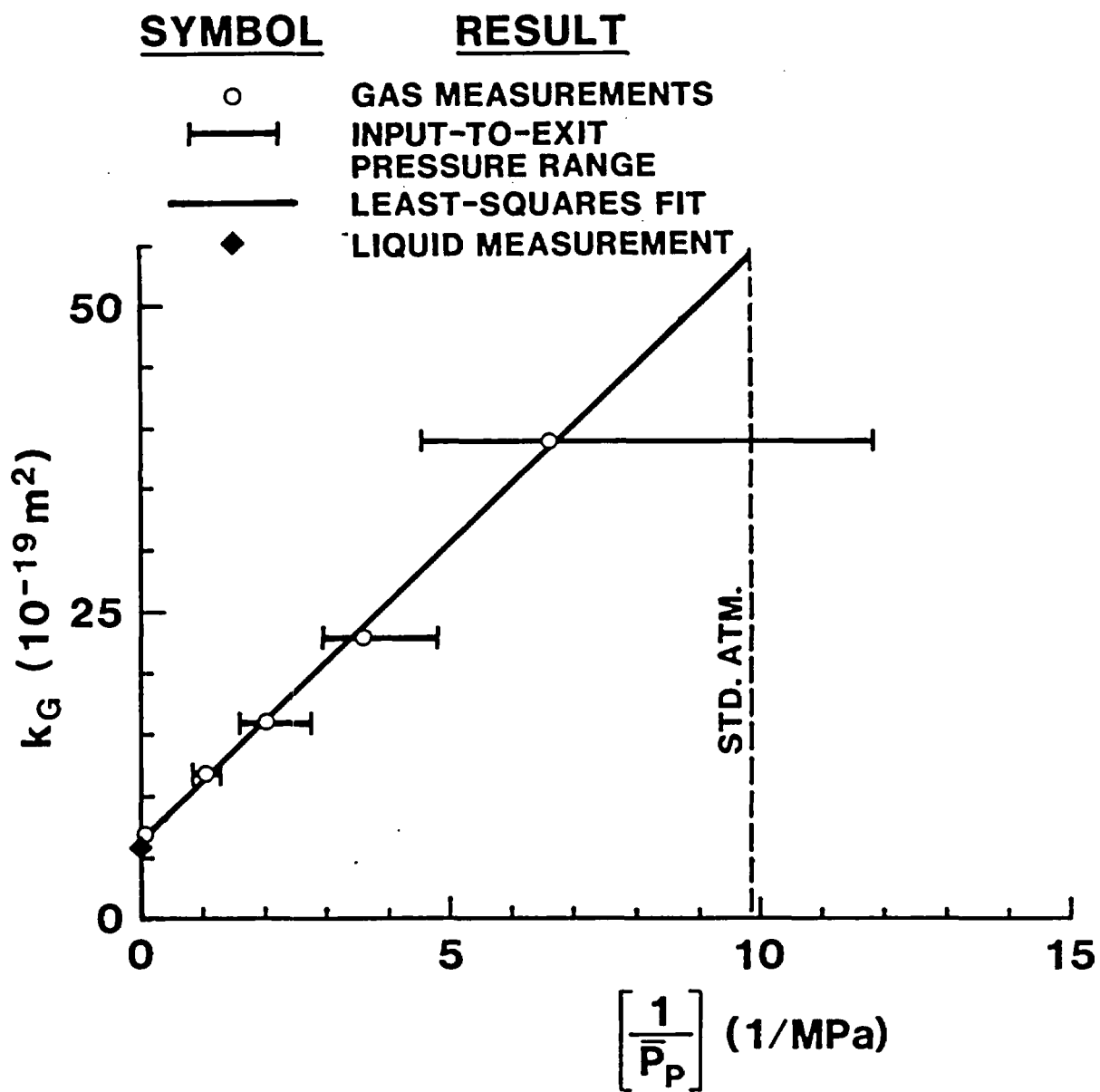


Figure 4: Summary of Gas Permeability Measurements Extrapolated to Infinite Pore Pressure

Figure 5: Liquid Permeability as a Function of Time for T = 25°C

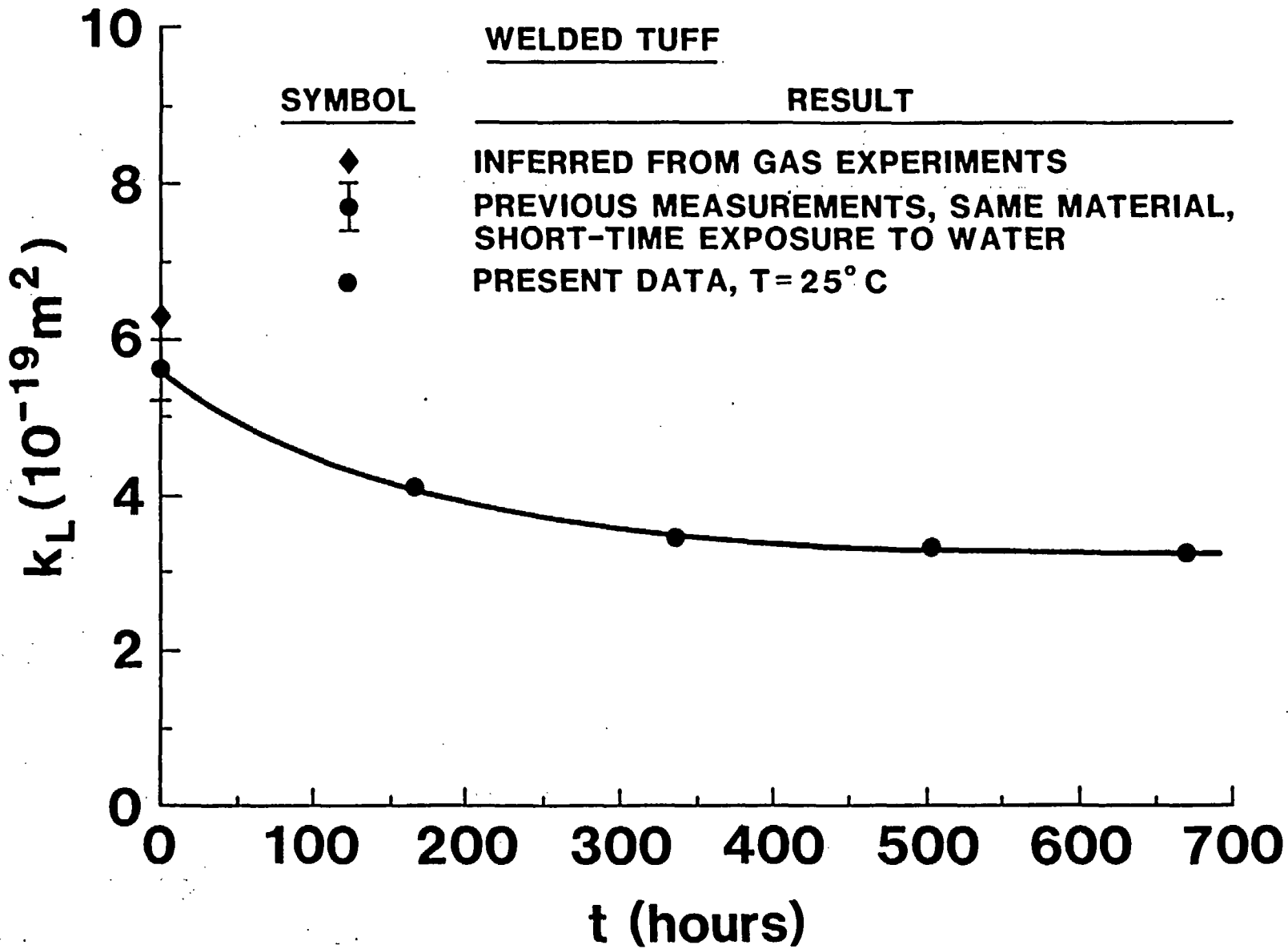
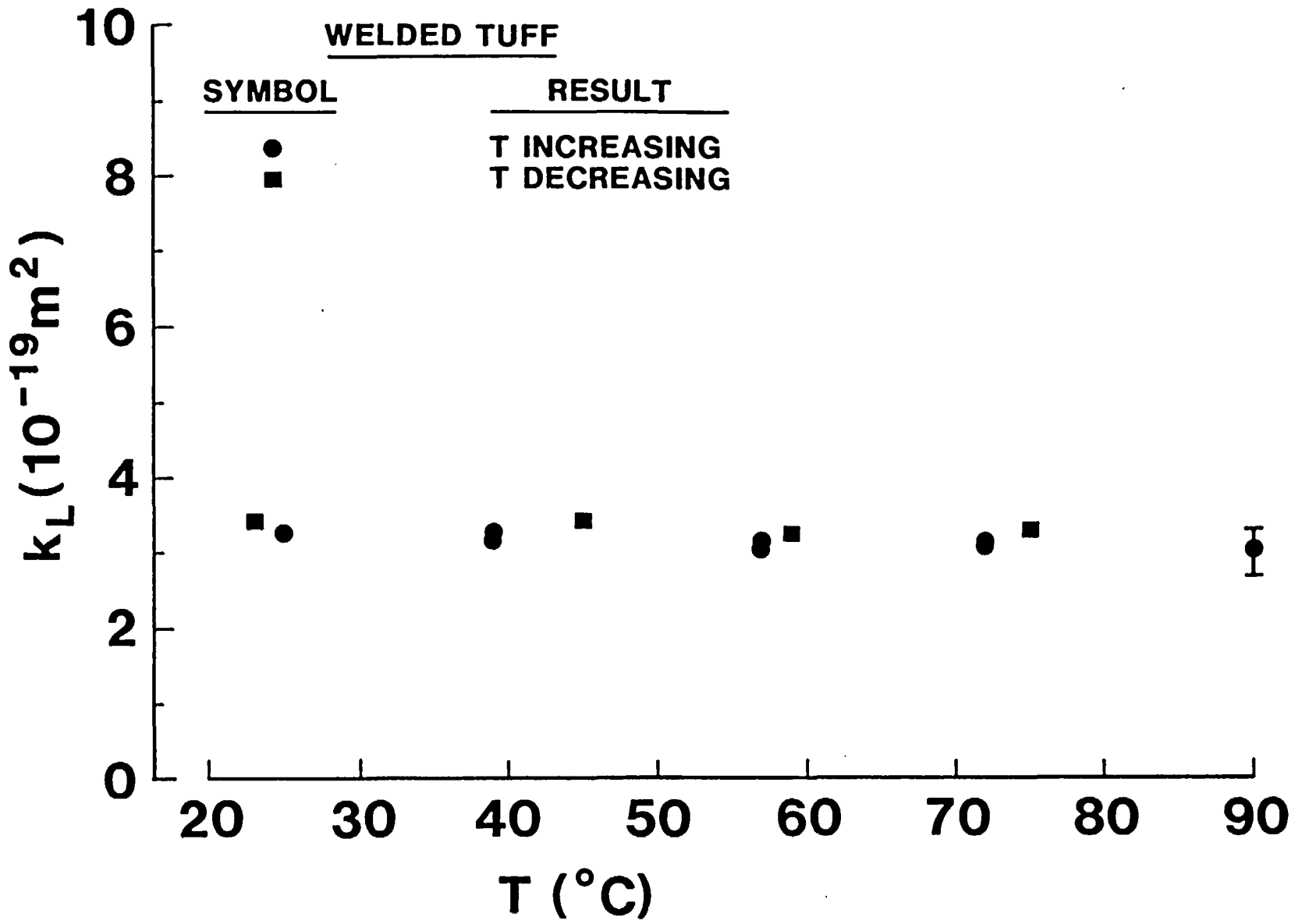


Figure 6: Liquid Permeability as a Function of Temperature



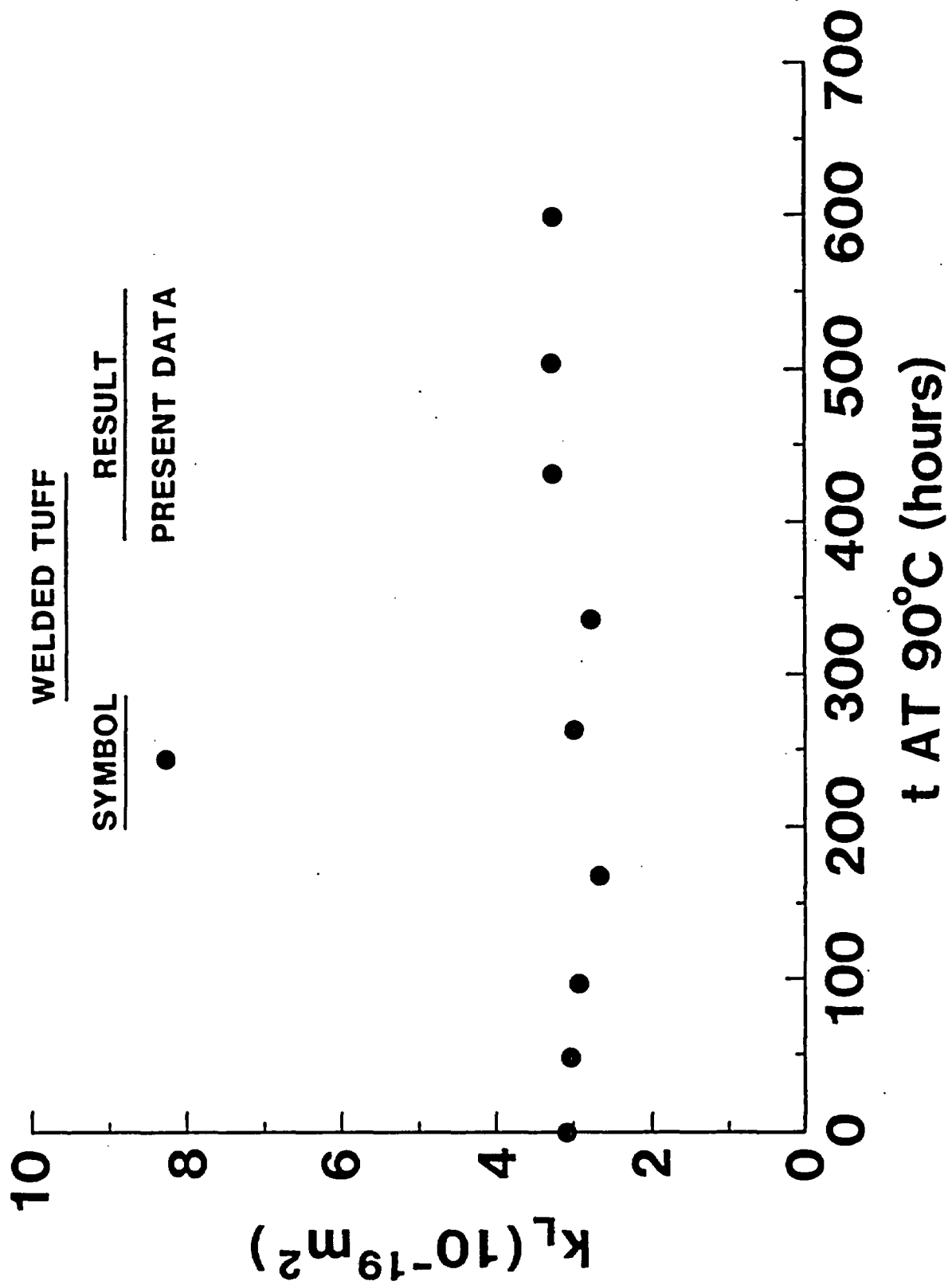


Figure 7: Liquid Permeability as a Function of Time for $T = 90^\circ C$

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6310	Central File
6311	L. W. Scully
6311	L. Perrine
6312	F. W. Bingham
6312	N. K. Hayden
6312	B. S. Langkopf
6312	R. R. Peters
6312	R. W. Prindle
6313	T. E. Blejwas
6313	E. A. Klavetter
6314	J. R. Tillerson
6315	S. Sinnock
6332	WMT Library (20)
6430	N. R. Ortiz
3141	S. A. Landenberger (5)
3151	W. L. Garner (3)
8024	P. W. Dean
3154-3	C. H. Dalin (28) for DOE/OSTI

