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EVAPORATIVE WATER LOSS FROM WELDED TUFF*

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ABSTRACT

Welded tuff is one of the many candidate rocks presently being considered as a host medium for the disposal of radioactive waste. In the case where the disposal site lies above the water table, the host rock will in general be only partially saturated. This condition leads to a number of mass transfer processes of interest, including evaporative drying, two-phase water flow due to pressure gradients, capillary movement, plus others. Although these processes have all been known about for decades, it is not clear at this time what the relative importance of each is with regard to geologic media in a waste disposal environment. In particular, there seems to be no data available for tuff that would allow an investigator to sort out mechanisms. This work is intended to be a start in that direction.

This paper reports the measurement of water loss rate for welded tuff at various temperatures due to the action of evaporative drying. The samples are sections of 4.8 cm diameter core cut to lengths of approximately 6 cm. The initial saturation was unknown, but the average initial water content was found to

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be ~7% by weight. The samples were weighed and then inserted in a sealed chamber through which dry nitrogen gas was passed at various flow rates. The chamber could also be heated to any desired temperature. Upon passing through the chamber, the nitrogen gas was forced to flow through four desiccant canisters in series which were periodically weighed to determine the water loss rate.

The resulting data show that the water loss rate declines monotonically with time at a given temperature and increases with increasing temperature as expected. Somewhat surprising, however, is the fact that over 90% of the water from a sample was lost by evaporation at room temperature within 72 hours. All the water loss data, including that taken at temperatures as high as 150°C, are explained to within a factor of two by a simple evaporation front model. The latter assumes the water is lost by the molecular diffusion of water vapor from a receding evaporation front. The motion of the evaporation front seems to depend on mass balance rather than energy balance. Capillary forces and the resulting liquid diffusion are evidently not strong enough to wash out the evaporation front, since the front model seems to fit the data well.

INTRODUCTION

Tuff is one of many candidate rocks currently being considered as a host repository material for the storage of nuclear waste. As is the case with other rocks, tuff usually contains some water and in some cases may contain up to 50% water by volume. This water is mostly contained in the pores (either as a liquid or vapor) and consequently is free to move under the action of density or pressure gradients. Changes in water content influence thermal and mechanical properties, and the moving water may transport radionuclides if such are present. Consequently, it is important to understand the physical processes involved so that the motion of water within a repository may be predicted. The present work involves the drying of welded tuff samples whose boundaries were bathed in a dry nitrogen atmosphere, and consequently, represents a first step towards understanding the overall water transport.

The present group of experiments was partially motivated by previous work¹ done on rock salt, another repository candidate. In that study, the water transport mechanism was believed to be a Darcy flow of water vapor from a receding evaporation front. Here it was intended to measure the water loss rate from heated tuff using a similar apparatus under the assumption that the same mechanism was responsible but operating at a much higher permeability. In retrospect, this mechanism does not seem to provide the best fit to the present experiments, as will be pointed out in the section DISCUSSION OF THEORETICAL MODELS.

A direct application of the results from these preliminary experiments is the drying of mine openings, pillars, and drill holes. It was found that samples of welded tuff 4.8 cm in diameter by 6 cm high lost 90% of their water at ambient temperature in 3 days. Thus, surfaces open to a dry atmosphere may dry out quickly an appreciable distance into the rock.

APPARATUS

The apparatus used in this experiment consisted of a heated stainless steel chamber; a pressurized nitrogen supply together with associated valves and a flow gauge; a thermocouple and datalogger to measure and record sample temperature; and desiccant canisters for collection of water (Figure 1).

The stainless steel chamber was fitted with O-ring seals so as to be airtight. Swagelok fittings were installed on opposite sides of the chamber to allow the inlet and exhaust of nitrogen gas. A Teflon sleeve fitting was installed on the top of the chamber to allow a sheath thermocouple to make thermal contact with the top of the sample. The sample was supported by stainless steel rods so that it would be approximately the same level as the gas inlet and outlet fittings (see Figure 2).

The nitrogen gas was supplied by a pressurized bottle fitted with a standard regulator followed by a 100 psi pressure relief valve to insure safety. A flow control valve and flow gauge were in series between the pressure relief valve and the chamber to control and determine the rate of flow of gas into the chamber.

Four canisters containing removable desiccant cartridges were installed in series as shown in Figure 1 to remove as much moisture as possible from the nitrogen gas stream. Each cartridge was weighed at every weighing interval with the weight gains added to determine the total mass loss from the sample. The low mass gain of the fourth cartridge was a sufficiently small fraction of the total (usually about 1.5%) so as to give assurance that most of the water lost from the sample was collected and measured. The desiccant cartridges were weighed on an analytical balance which has a resolution of 0.1 mg.

The sample chamber was heated by means of heater tape wrapped around its circumference. The amount of power supplied to the tape was adjusted by a

FIGURE 1. SCHEMATIC DIAGRAM OF APPARATUS.

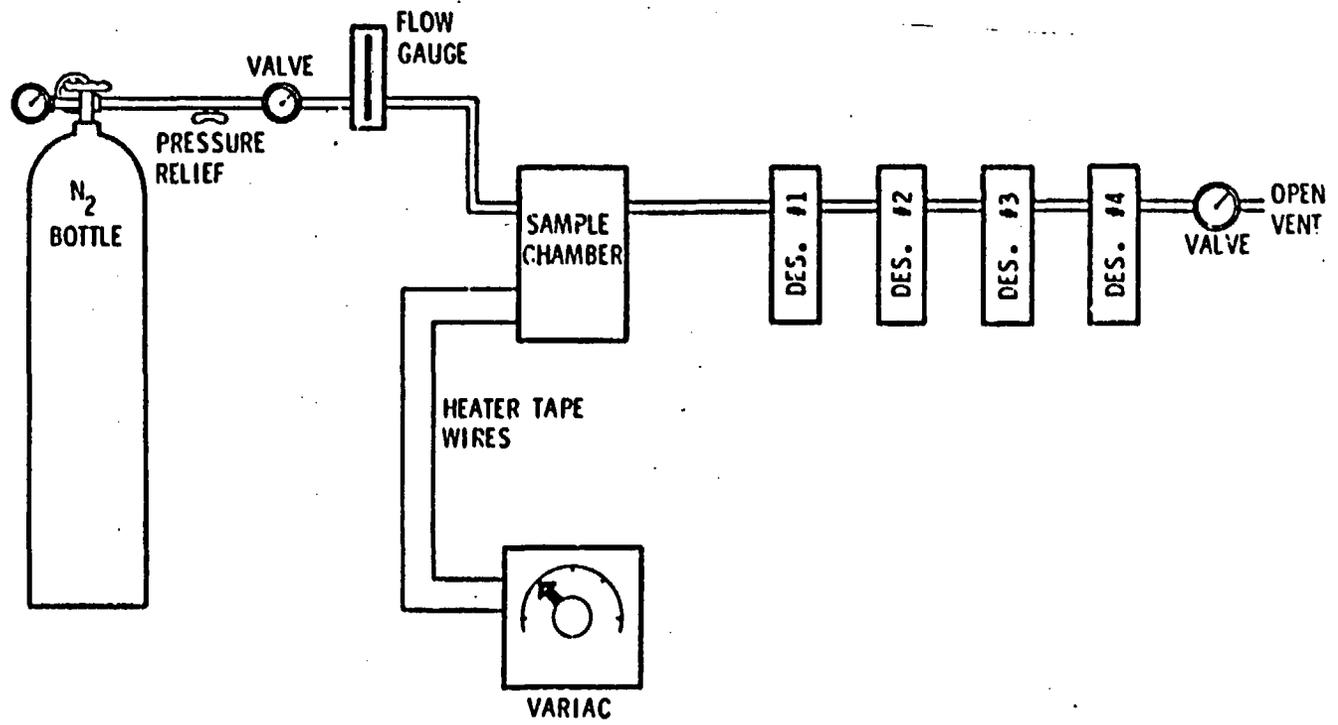
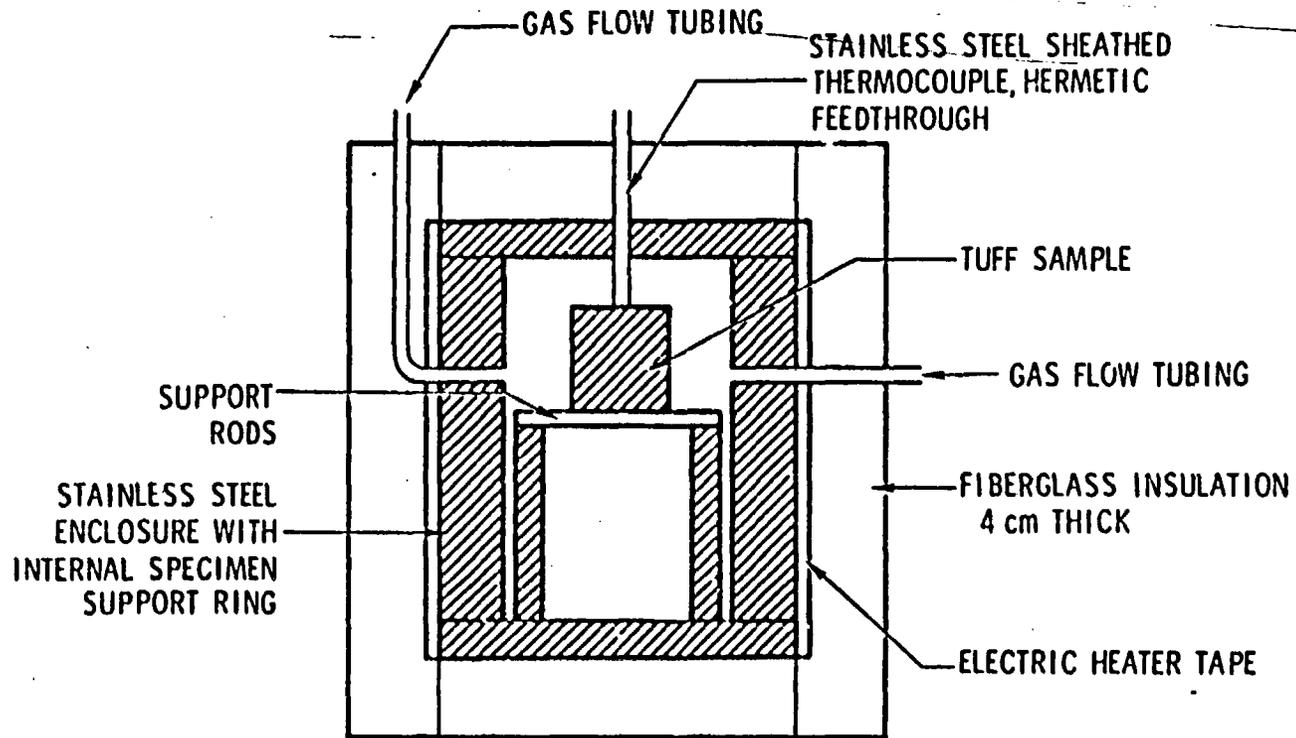


FIGURE 2. SECTION VIEW OF SAMPLE CHAMBER.



variac in order to control the sample temperature. The latter was measured by a chromel-alumel thermocouple encased in a 1/16" metal sheath. The thermocouple was lowered through the Teflon fitting until it made contact with the sample, after which the fitting was tightened. The thermocouple leads were connected directly to a Fluke 2240B datalogger which gave periodic printouts of the temperature accurate to better than 0.1°C.

The samples were sections of welded tuff core 4.8 cm in diameter and 6 cm in height. The core was from the welded Grouse Canyon Member of the Belted Range tuffs located in Southwestern Nevada. The samples were kept in foil sealed with bee's wax until just prior to their placement in the chamber. Upon removing the foil, excess water on the surface of the sample was removed with a paper towel, after which the sample was weighed on an analytical balance and placed in the chamber.

EXPERIMENTAL PROCEDURE AND RESULTS

Basically, the experiment consisted of inserting a wet tuff sample into the chamber, turning on the flow of N₂ gas, applying power to the heater tape if desired, and periodically weighing the desiccant canisters. The sample temperature history was monitored throughout the duration of each run via an automatic periodic printout from the Fluke datalogger. The canisters were weighed at intervals ranging from as little as 20 minutes during working hours to 12 hours when the experiment was left overnight. The N₂ flow rate was varied during the experiment, depending on the water loss rate, in such a way as to not use up the bottle too quickly while still providing sufficient flow to keep the atmosphere surrounding the sample as dry as possible.

The capacity of the N₂ gas to transport water vapor to the canisters may be determined as a function of flow rate by a simple calculation in which the water vapor density at 100% relative humidity is multiplied by the flow rate. The relative humidity existing in the flow stream at a given time during a run may

be estimated by dividing the above "capacity" by the measured water loss rate. Relative humidities obtained in this way were usually below 20%. Exceptions are the first few hours of each run, where the mass loss rates are always high (ideally approaching infinity) and the relevant humidities may have approached 100%. Flow rates used varied from 0.4 l/min to 20 l/min.

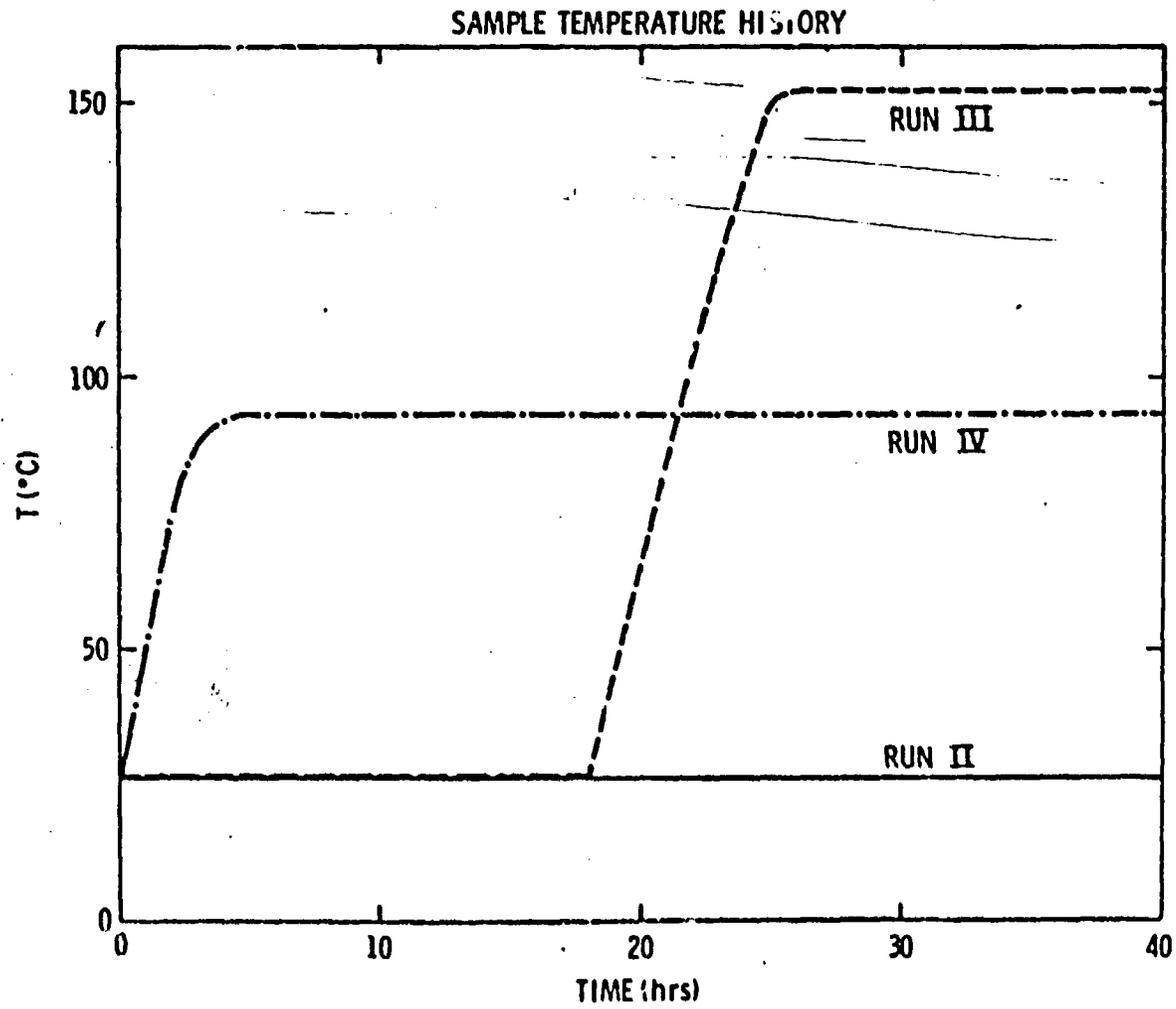
The desiccant canisters were weighed in the same order each time in a procedure that took 7 to 8 minutes. In this manner, the time interval between weighings for each canister was identical. A new canister was inserted whenever the mass gain for a given canister began to approach its maximum of 6 g. While on the analytical balance, each canister absorbed water from the atmosphere, thus registering mass gain which was somewhat high. The magnitude of this error was measured by successively weighing a canister which was sealed between weighings. The mass gain per weighing averaged 1.5×10^{-3} g. Other background included water trapped on chamber and fitting surfaces and was measured to be 2×10^{-6} g/min. The absorption error mentioned above would add to about 1% total error in the mass flow rate. The background measurement indicates that flow rates below 2×10^{-4} g/min may be in error, although rates as low as 7×10^{-5} g/min were recorded at times. These figures are well below the usual range of interest, 10^{-3} to 10^{-1} g/min.

The total mass loss at the end of each run, as determined by the total canister weight gain, was checked by comparing with before and after sample weights. The two results always agreed to better than 2%.

The temperature versus time profiles for the three experiments to be discussed (denoted runs II through IV) are shown in Figure 3. Run II was held at room temperature for 72 hours and then heated to 180°C to drive off most of the remaining water. Run III was heated to 150°C after 18 hours at room temperature, and Run IV was heated immediately to 92°C.

The water loss rate for Run II is shown in Figure 4. The data indicates a steady decrease in loss rate dropping more than an order of magnitude after 50 hours. At 72 hours the total mass loss was 91% of the total for the entire ex-

FIGURE 3. SAMPLE TEMPERATURE HISTORY FOR EACH RUN.



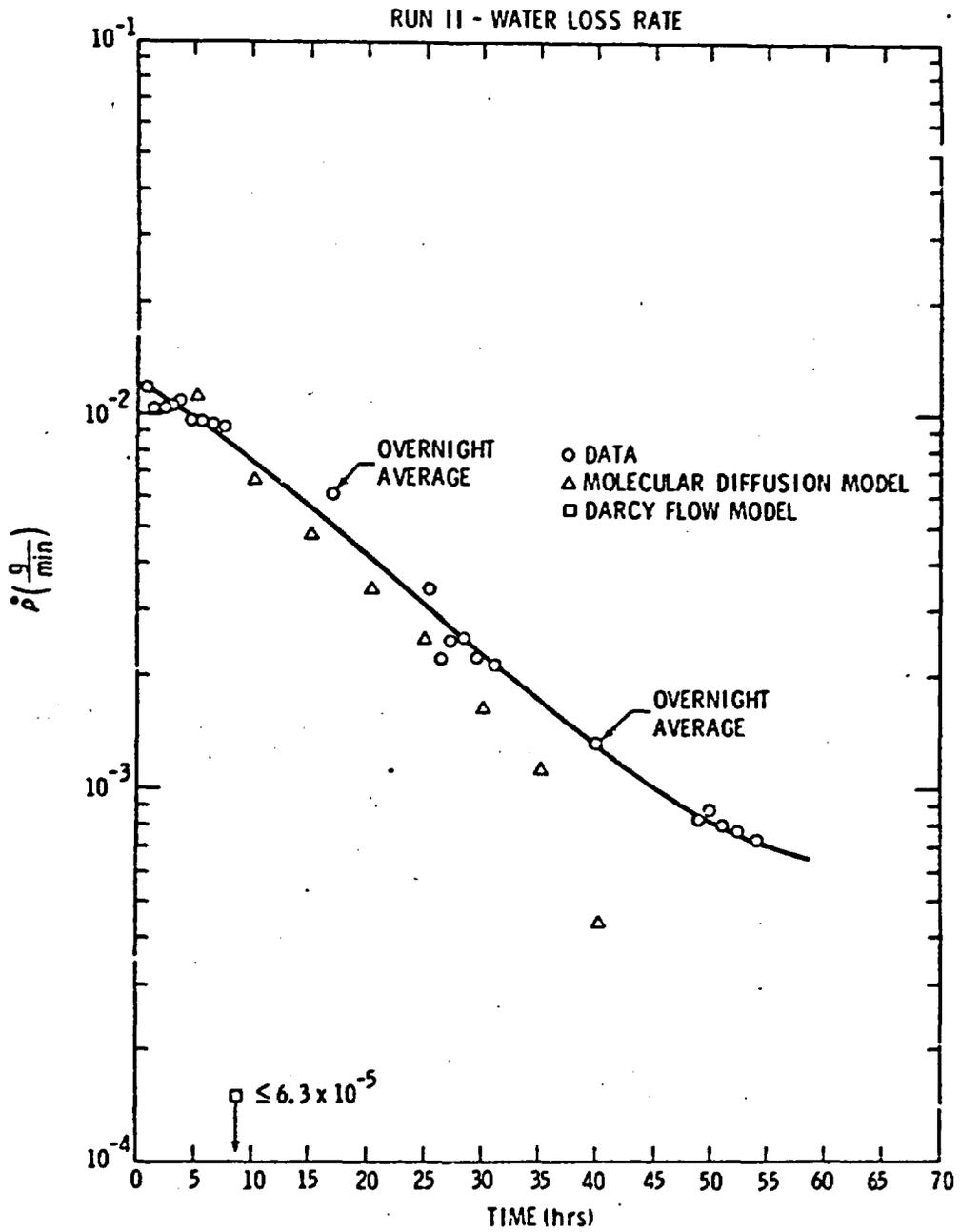


FIGURE 4. WATER LOSS RATE FOR RUN II -- THE CURVE REPRESENTS AN APPROXIMATE FIT TO THE DATA.

periment. The remaining 9% was then driven off by heating the sample chamber to 180°C. The water loss rate for this period of time (not shown) peaked at 8×10^{-3} g/min and then quickly fell to background levels. Apparently the remaining 9% was either loosely bound water or free water trapped in tightly sealed pores from which it was released very slowly at ambient temperature.

The data for Run III (shown in Figure 5) initially behaved similarly to Run I but then responded with a well defined peak as the heat was turned on at 18 hours. Due to the effect of heating, more water per initial sample weight was lost in 40 hours than in 75 hours with Run II. This effect is enhanced further in Run IV where the same percent water loss occurred in 24 hours. Run IV (Figure 6) produced the highest measured loss rates (>0.09 g/min) of the three tests due to the fact that the sample temperature was increased immediately while the regions near the sample surface were still wet. The average total water loss of the three samples expressed as a percent of the initial weight was 7.3%.

DISCUSSION OF THEORETICAL MODELS

The Darcy flow model was the first attempt to fit the data described in the previous section. Since this model gave good agreement with experiments in the rock salt case,¹ it was expected to apply here also with a larger permeability and porosity. Although this model is described in detail in Reference 1, a quick review will be given here.

For simplicity, the cylindrical sample is modeled as an isothermal sphere with two distinct regions (one liquid water, one air and water vapor) separated by an evaporation front as shown in Figure 7. For the Darcy flow model, the water vapor is considered to be a fluid flow which moves according to Darcy's law in a quasi-steady state manner. The boundary conditions are taken to be $p =$ equilibrium vapor pressure at the evaporation front and $p = 0$ at the outer surface. Thus, the effect of foreign gases (such as air) is ignored. The evap-

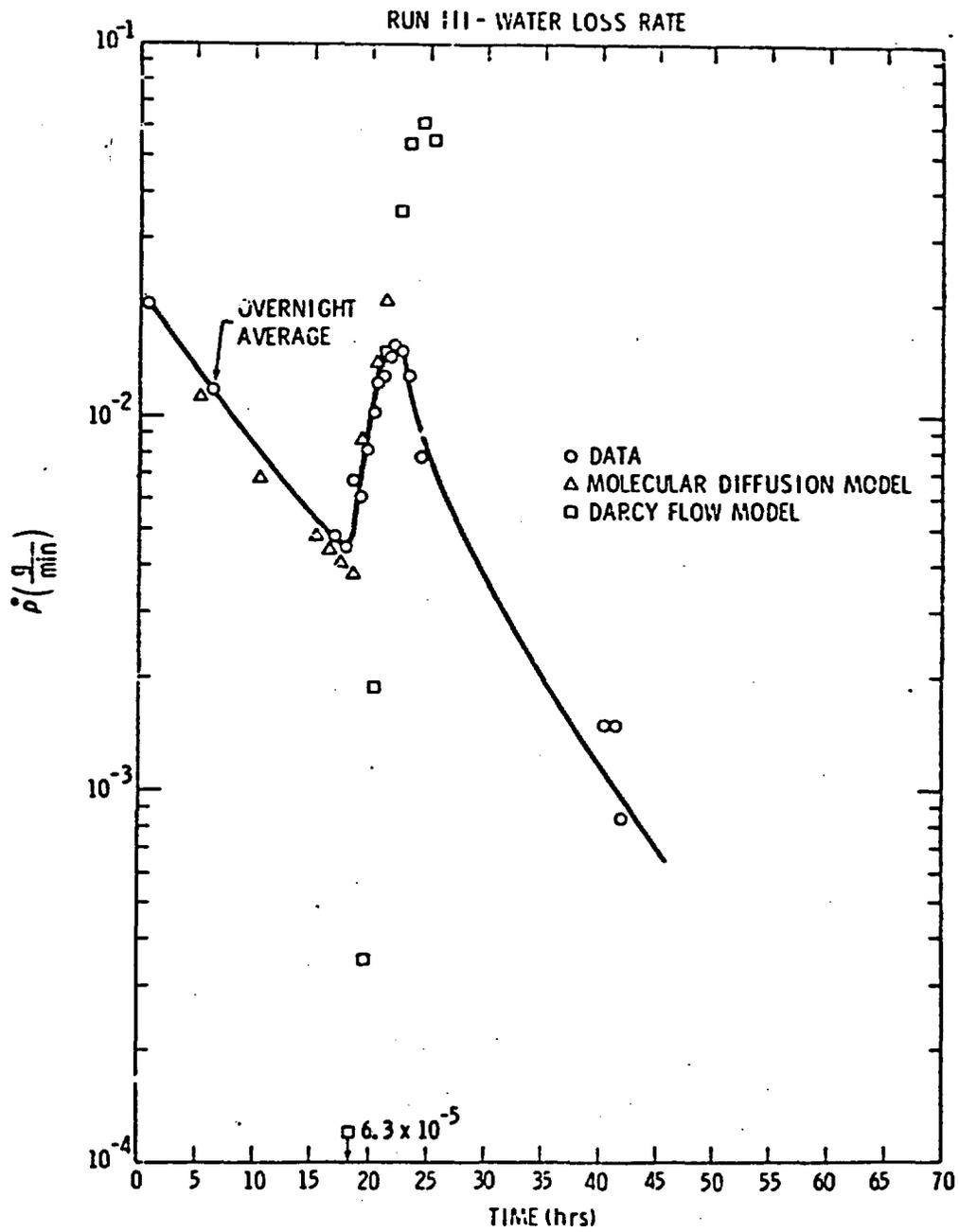


FIGURE 5. WATER LOSS RATE FOR RUN III -- THE CURVE REPRESENTS AN APPROXIMATE FIT TO THE DATA.

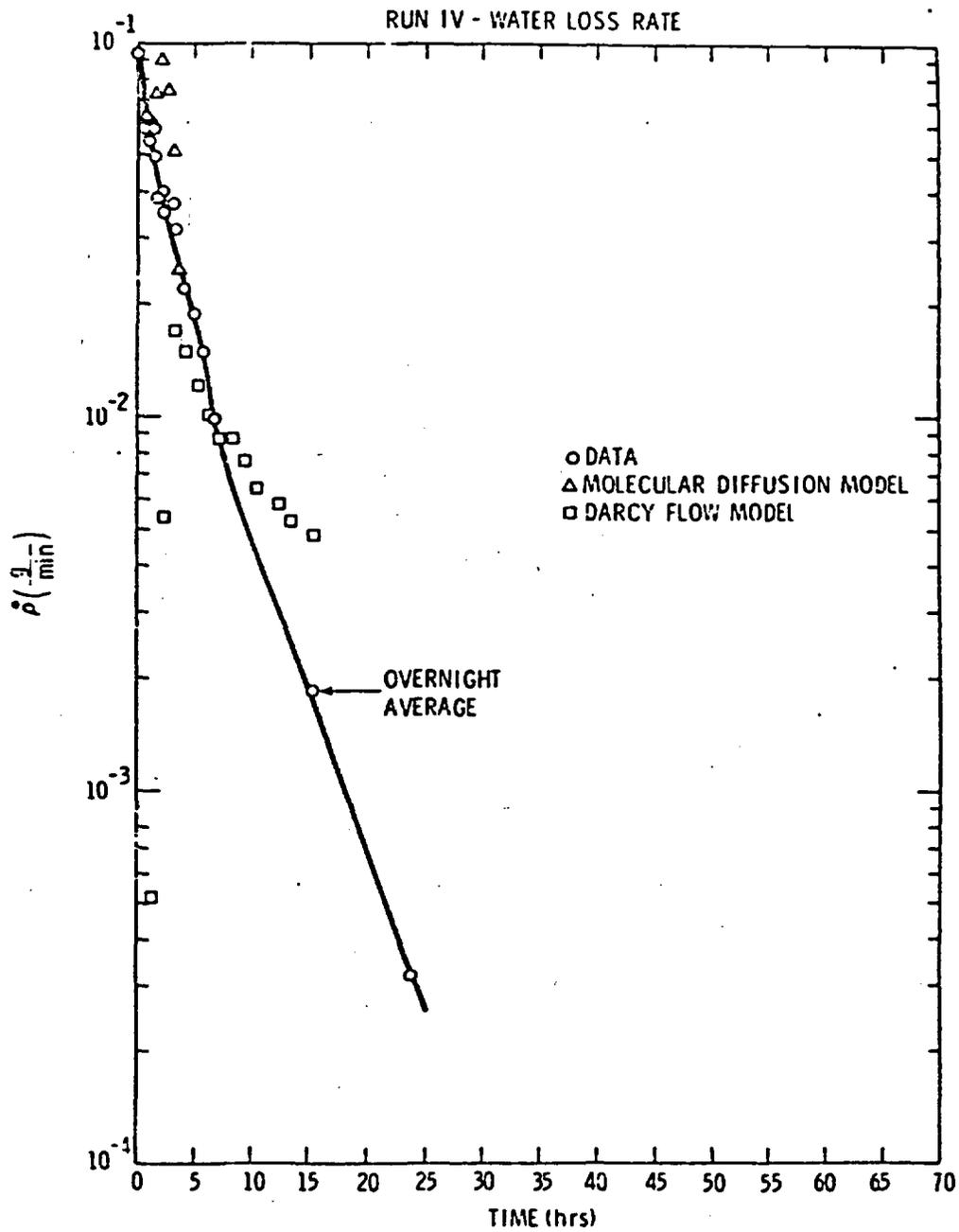


FIGURE 6. WATER LOSS RATE FOR RUN IV -- THE CURVE
 REPRESENTS AN APPROXIMATE FIT TO THE DATA.

poration front is then allowed to recede at a rate determined by mass balance:

$$u = - \frac{\rho_v v}{\rho_w \phi} \quad (1)$$

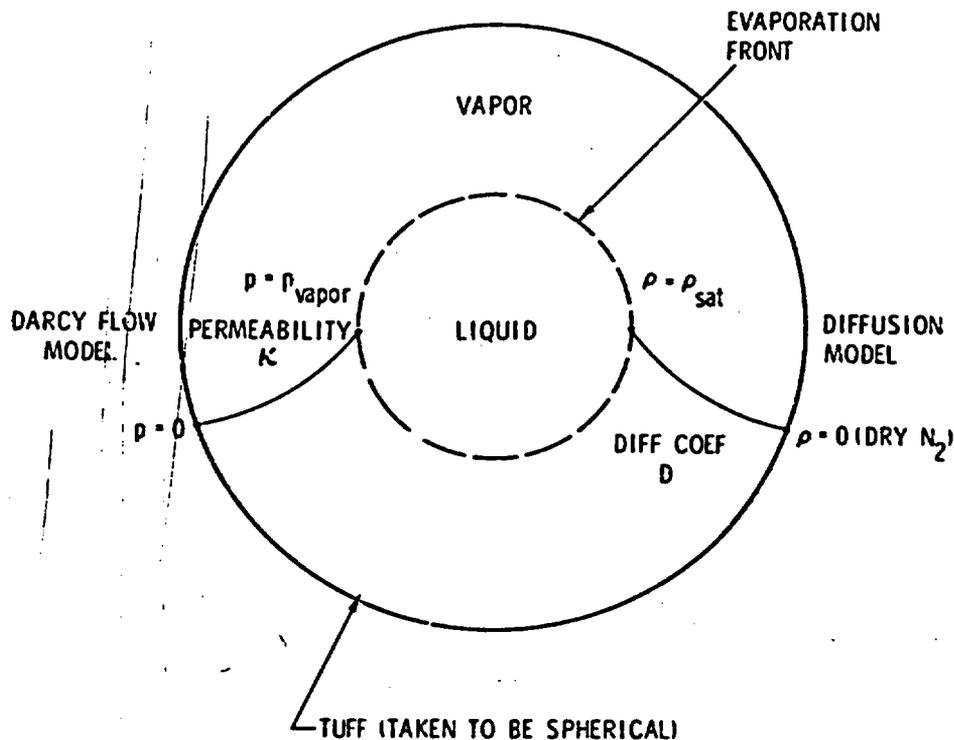


Figure 7. EVAPORATION FRONT MODELS -- THE TWO MODELS DIFFER ONLY IN THE HANDLING OF THE VAPOR REGION.

where $\rho_{v,w}$ = density of water (vapor, liquid), v = vapor velocity at the front, ϕ = porosity.

Another model used, the molecular diffusion model, is similar to the latter except that in the vapor region, transport is assumed to proceed via diffusion rather than bulk motion. The diffusion equation is thus solved in this region with boundary conditions ρ_v = equilibrium saturation density at the front and $\rho_v = 0$ at the outer surface (dry boundary). The front recession is calculated in a similar manner to the Darcy flow model.

In both cases, the sphere was assumed isothermal with latent heat effects ignored. This simplification is not ordinarily made in drying theory calculations

and, in fact, it is often assumed that evaporation takes place at a constant temperature known as the evaporation temperature.² For small samples, however, sample temperature is externally controlled with the latent heat being a small perturbation. This may be seen by making an estimate of the temperature gradient necessary to supply the heat of vaporization. The result for the present case is $\sim 0.4^\circ\text{C}/\text{cm}$ which would induce nonuniformities of only $\sim 1^\circ\text{C}$. Consequently, this effect is small and may be neglected.

Comparisons with the data for both models may be seen in Figures 4 through 6. The permeability chosen for the Darcy flow model was $5 \times 10^{-13} \text{ cm}^2$ which gave a reasonable fit for high temperature data as well as measured values for tuff.³ However, it may be seen immediately that a much higher value is necessary to fit the room temperature data. This strong temperature dependence is the downfall of the Darcy flow model, since no permeability may be found which will fit all the data. The reason for the sensitivity to temperature of this model stems from the strong temperature dependence of the saturated vapor pressure; the flux varies approximately as the square of this pressure.

The molecular diffusion model seems to fare much better, since the diffusion coefficient is much less temperature sensitive. The predicted loss rate is within a factor of two of the measured value except for nearly dry conditions. The latter problem is due to the evaporation front reaching the origin after which the model predicts zero mass loss rate. This is indicated on the graphs as the place where the triangles cease to be plotted. Of course, the data shows no such dramatic drop-off. The good agreement with experiment elsewhere for the diffusion model is the more encouraging when it is realized that, unlike the Darcy flow model which uses permeability as a fitting parameter, there are no undetermined parameters. The diffusion constant for water vapor in air is well known as a function of temperature. Consequently, the diffusion model output is a clear prediction.

While the loss mechanism(s) are still poorly understood, it would appear that the evaporation is diffusion dominated at low temperatures and perhaps

Darcy flow dominated at sufficiently high temperatures. At this point, the crossover temperature is unknown but would appear to be above 150°C. The single evaporation front hypothesis is almost certainly an over-simplification since there appears to be some water which may remain bound or trapped after the front has moved past.

CONCLUSION

Herein, we report a preliminary experiment in which the water loss rate of heated samples of welded tuff was measured as a function of time. The results clearly show a declining loss rate as the sample dries from the outside inwards. Although the rate is higher at high temperature, a significant loss of water at room temperature may occur in a few days.

Theoretical investigations indicate that molecular diffusion is the primary loss mechanism, at least over the temperature range surveyed here. Predictions based on a diffusion model and a receding evaporation front give fairly good agreement with the data for temperatures ranging from room temperature to 150°C.

Future experiments should be conducted on samples cut so as to offer true one-dimensional geometry together with greater depth. This would allow a more careful comparison with theory. From the theoretical side, some attempt must be made to combine the diffusion and Darcy flow phenomena with each included in a more exact manner. Also, some allowance should be made for "tight" pores.

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