

DIFFUSION OF ENTRAPPED AIR
FROM POROUS MEDIA

by

G. L. Bloomsburg and A. T. Corey

August, 1964



HYDROLOGY PAPERS
COLORADO STATE UNIVERSITY
Fort Collins, Colorado

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ABSTRACT

The purpose of this study was to investigate the entrapment of air in porous media imbibing liquid and to determine both theoretically and experimentally the rate at which air goes into solution and diffuses from the system.

The samples studied were cylindrical cores approximately one inch in diameter and of various lengths up to two and one half inches. Three materials were used, sandstone, a porous ceramic and sintered glass beads. Two liquids, water and a hydrocarbon oil, were used as the liquid imbibed.

Theoretical equations for linear and radial (two dimensional) diffusion were developed. These are based on the assumptions that: (1) the concentration of dissolved air becomes nearly uniform after air is entrapped and (2) the point in the medium at which air dissolves moves from the exterior toward the interior as a front.

The results show that a porous medium will eventually become completely saturated by imbibition even though no liquid is flowing through and the liquid imbibed is saturated with dissolved air at atmospheric pressure. The diffusion process by which the entrapped air moves out is most rapid for fine grained material. This is because the air is entrapped in pockets which maintain a higher concentration of dissolved air in the liquid due to the greater pressure difference across the air-liquid interface associated with smaller radii of curvature.

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NOTATION AND DEFINITIONS

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| <p>A - Area--dimensions: L^2.</p> <p>a - Radius of an air bubble in a liquid--dimensions: L.</p> <p>B - Universal gas constant--dimensions: FL/mole^0k.</p> <p>C - Concentration of dissolved air--dimensions: parts per million by mass.</p> <p>C_o - Assumed uniform concentration of dissolved air in a liquid in a porous medium. Computed directly from the bubbling pressure--dimensions: parts per million by mass.</p> <p>C_B - Concentration of dissolved air at the exterior boundary of the core--dimensions: parts per million by mass.</p> <p>D - Coefficient of diffusion of dissolved air in liquid--dimensions: L^2/T.</p> <p>D_s - Diffusivity coefficient--dimensions: L^2/T.</p> <p>F - Flow rate of air by diffusion--dimensions: L^3/L^2T.</p> <p>f - Functional symbol--dimensions: none.</p> <p>g - Acceleration due to gravity--dimensions: L/T^2.</p> <p>K - Constant of integration--dimensions: none.</p> <p>k - Saturated Permeability - Permeability of a porous medium to a liquid which completely fills all pore spaces. Sometimes called intrinsic permeability--dimensions: L^2.</p> <p>k_e - Effective permeability - the permeability of a porous medium to a particular fluid phase when the medium is occupied by more than one fluid phase--dimensions: L^2.</p> <p>k_r - Relative permeability - the effective permeability divided by the saturated permeability--dimensions: none.</p> <p>L - Length--dimensions: L.</p> <p>M - Molecular weight of a gas--dimensions: M/mole.</p> <p>m - Mass of air--dimensions: FT^2/L.</p> <p>P - Pressure--dimensions: F/L^2.</p> <p>P_b - Bubbling Pressure - the smallest capillary pressure on the drainage cycle at which air permeability is greater than zero--dimensions: F/L^2.</p> | <p>P_c - Capillary pressure - pressure in the nonwetting fluid minus the pressure in the wetting fluid--dimensions: F/L^2.</p> <p>Q - Liquid flow rate--dimensions: L^3/T.</p> <p>q - Liquid volume flux--dimensions: L^3/L^2T.</p> <p>R - Radius--dimensions: L.</p> <p>r - A radial length variable--dimensions: L.</p> <p>S - Wetting fluid saturation, equal to the volume of liquid divided by the volume of voids--dimensions: none.</p> <p>S_o - Initial saturation - the saturation when air becomes trapped--dimensions: none.</p> <p>T - Absolute temperature--dimensions: 0K.</p> <p>t - Time--dimensions: T.</p> <p>V - Volume--dimensions: L^3.</p> <p>V_a - Volume of air in a porous medium at any time--dimensions: L^3.</p> <p>V_{ao} - Initial volume of entrapped air in a porous medium--dimensions: L^3.</p> <p>V_f - Volume of air which has diffused from the porous medium at any time ($V_{ao} - V_a$)--dimensions: L^3.</p> <p>V_l - Volume of liquid in a porous medium--dimensions: L^3.</p> <p>V_v - Volume of voids in a porous medium--dimensions: L^3.</p> <p>x - A length variable--dimensions: L^3.</p> <p>α - Proportionality constant--dimensions: L/T.</p> <p>Δ - Used with any variable to show a small increment--dimensions: none.</p> <p>η - A pore size distribution index--dimensions: none.</p> <p>μ - Fluid viscosity--dimensions: FL^2/T.</p> <p>ξ - Empirical constant--dimensions: none.</p> <p>ρ - Fluid mass density--dimensions: FT^2/L^4.</p> <p>σ - Surface energy of liquid--dimensions: F/L.</p> |
|--|--|

Many of the above symbols are used with various subscripts which are defined at the time of introduction.

DIFFUSION OF ENTRAPPED AIR FROM POROUS MEDIA^{1/}

G. L. Bloomsburg and A. T. Corey^{2/}

INTRODUCTION

Entrapped air can have an effect on several physical properties of porous media containing a liquid. Ordinarily when liquid pressure is greater than atmospheric, media are oftenthought of as being saturated. However, if there is entrapped air, the permeability of media to liquid may be only half that in a completely saturated condition. Entrapped air is also important in consolidation theory for unsaturated soils.

In the field of irrigation, drainage and ground water hydrology, there are many flow problems in which water pressure is equal to or greater than atmospheric but in which there is entrapped air.

There are basically two ways in which air may become trapped in the ground. These are by water moving into an originally dry soil and entrapping air or by air coming out of solution in a saturated system and forming bubbles. Some examples of the first of these are movement outward from an irrigation furrow, seepage from an irrigation canal when water is first turned into a canal, and movement downward from a spreading basin in a ground water recharge operation. An example of the latter is seepage from a canal when temperature or pressure gradients in the soil are such as to allow dissolved

air to come out of solution. Also in cases of decomposition of organic deposits, gases other than air may be formed and cause air pockets in the system.

This study deals with the first situation, that is, with porous media originally containing air imbibing a liquid which entraps air in the system.

At a relatively low liquid saturation, e. g., near the wetting front, there is a continuous gas phase and the flow of air out of media is determined by the rate at which air is displaced by liquid. At some higher liquid saturation the gas phase becomes discontinuous and consists of isolated air bubbles. After this condition is reached, the only way air may leave the system is by going into solution in the liquid and then moving through the liquid by diffusion.

In recent years there have been numerous articles written on the application of a diffusion equation to the problem of water moving into dry soil. This equation is called the "diffusivity equation" and a necessary assumption for its solution is that liquid saturation is a unique function of liquid pressure as long as liquid pressure is constant or continuously increasing. In other words, as long as pressure is constant there is no change of liquid saturation with time, or considering entrapped air, there is little or no diffusion out of the system. However, it has been noted in the course of experimental work that this assumption is not necessarily valid and that air does diffuse out of porous media.

Previous studies concerning the removal of entrapped air have dealt with the removal of air by flowing water only. It is the primary purpose of this study to determine rates at which air diffuses from a system in which the liquid is not flowing and to develop an analytical expression for rate of diffusion as affected by geometry of media pore space, external geometry of the sample, and fluid properties including surface tension of the liquid and solubility and diffusivity of air in the liquid.

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REVIEW OF LITERATURE

Entrapped and Confined Air

There are few articles in the literature dealing with entrapped air in porous media. Zimmerman [30] recognized that air was trapped in a column imbibing from the bottom. The amount was computed by measuring the air which moved out, and subtracting this from the total pore volume. The effect of particle size and chemical constituents on the total capillary rise was also discussed. There was no mention, however, of air diffusing out of the system and no theoretical analysis of the amount of entrapped air.

Christiansen [6] carried out an extensive study on the effect of entrapped air on the permeability of soils. He first investigated whether there was air trapped when a column was wetted from the bottom under a positive pressure and found that it was trapped in this case as well as when water was allowed to flow from the top into the dry soil. The permeability of soil was also investigated as a function of time and was found to decrease slightly initially, then to increase considerably over several days time and after this to decrease gradually. There was no explanation for the slight initial decrease or the gradual decrease at the end, but the increase was attributed to the solution of entrapped air in the flowing liquid which sometimes continued for as long as two months. For soils of low permeability the time for air to dissolve was much longer than for soils of high permeability, which infers that the rate of solution depends on the rate at which water moves through the system. This was also indicated by permeability increasing at the top of the column first and then progressively increasing toward the bottom.

Christiansen made no theoretical analysis of the solution process and conducted all permeability tests at a positive pressure. The porous materials used consisted of agricultural soils, such as clay loam, sandy loam, and loam.

Miller and Miller [20] explained the above mentioned change of permeability with time as being due to first; instability of soil, second; diffusion of air out of entrapped air pockets, and third; micro-organism growth.

Orlob and Radhakrishna [21] used chloride tracer techniques to investigate the effect of entrapped air on various hydraulic properties of media, such as effective porosity, permeability, and hydraulic dispersion. Their experiment was performed using a glass column one meter long with porous materials of various particle sizes. All experiments were with columns under a positive head of about two inches of water. Air was entrapped during experiments by draining the column for various periods of time in order to let air into the various sized pores and then by allowing flow to resume. The authors did not consider diffusion of air out of porous media and were unable (conclusively) to relate the amount of entrapped air to properties of porous material.

Powers [23] discussed the effect of confined air in a porous medium on the rate of water

movement into the medium. By having: 1) the column open at the bottom, 2) the bottom closed and 3) suction applied to the bottom, it was found that confined air did slow the rate of water movement into the soil.

Wilson and Luthin [28] carried out an extensive study of the effect of air flow ahead of a wetting front during infiltration. In this study three degrees of obstruction at the outflow end of a column were examined. These were: 1) no obstruction, 2) partial obstruction and 3) complete obstruction. These three conditions are similar to various conditions under which infiltration occurs in field situations when the surface may be underlain by impermeable layers. A 4.5 cm cylindrical column 31 cm long was placed in a horizontal position and the water flowing in as well as the air flowing out were measured. An interesting result obtained was that the cumulative outflow of air was consistently greater than the inflow of water. This was said to be due to a temperature rise at the wetting front (due to the heat of wetting) which caused air to expand. A more probable explanation is that air adsorbed on dry solid surfaces was displaced by water. Measurements of air pressure during infiltration were obtained and were found initially to be as high as 14 cm of water. When the end of a column was completely obstructed, the pressure increased continuously and approached a maximum final value. In this case infiltration decreased until zero was approached as a limit.

Rawlins and Gardner [24] evacuated air from soil columns before allowing water to enter and found there was slight change in infiltration rates due to presence or absence of air. This fact led the authors to state that no definite conclusions could be drawn as to the effect of air on the process of infiltration. There was no study on the effect of confined air.

Gupta and Swartzendruber [13] used a permeameter to study water flowing through quartz sand in which a pressure change was imposed on the entire system. The volume change resulting from a given pressure change was then used to evaluate the volume of entrapped air in the system. It was found that the volume of entrapped air always decreased as water flowed through the system. The water which was used had been deaerated so a significant amount of air could be dissolved.

Two types of sand were used: one with a particle size range from 0.2 to 0.3 mm and the other from 0.05 to 0.3 mm. There was less air entrapped with a more uniform particle size than with a non-uniform size when the sample was wetted at atmospheric pressure. Hydraulic conductivity decreased over a period of time as was found by Christianson and this was attributed to bacterial activity, since use of a solution of 0.1% phenol solution to stop bacterial activity stopped the decrease. An attempt to determine the amount of entrapped air theoretically from the ideal gas law and the equation for pressure difference across an interface, assuming all air pockets spherical and of the same size, proved to be unsatisfactory.

Peck [22] used the same assumptions as above to determine theoretically changes in capillary pressure with temperature and air pressure. He concluded that due to the effect of temperature on surface tension and the volume of entrapped air, capillary pressure changes. The concentration of dissolved air and the mass of water in soil were assumed to be constant while the volume of water changed due to temperature changes. The same equation as used by Gupta and Swartzendruber was used to determine the volume of entrapped air as a function of capillary pressure. This proved to be accurate to within 20%.

Behavior of Bubbles in Liquids

Dynamics of Bubbles. Entrapped air may move through a liquid in a porous medium in two ways: 1) by movement of air pockets or 2) by pockets dissolving and air in solution moving through the liquid by diffusion. The first method of movement is probably negligible in the experiments carried out in this study but might be important in some field cases as evidenced by bubbles coming out of soil when irrigation water is introduced.

Gardescu [12] discussed the movement of bubbles through capillary spaces by considering the "Jamin effect." That is, resistance to movement for an air bubble is divided into a boundary effect caused by hysteresis of the contact angle of the gas-liquid interface with the solid surface and a resistance due to distortion of gas bubbles when forced through small openings. By a theoretical analysis it was found that resistance due to hysteresis is proportional to the variation of the surface tension and inversely proportional to the radius of the capillary tube. However, resistance due to a change of radius of curvature of the interface when a bubble is forced into a small opening was much greater than resistance due to hysteresis of the contact angle. Actually in the case of air bubbles in water in many porous materials, there should be no hysteresis effect since water wets the surface and bubbles are completely isolated from the solid surface.

Wyckoff and Botset [29] in a comprehensive study of flow of gas-liquid mixtures through unconsolidated sands discussed the movement of bubbles through constrictions. The authors stated that a certain critical force is necessary to force a bubble into a constriction whereas any force causes a bubble to move through a uniform tube. Since the pressure difference across an interface is inversely proportional to the radius of curvature, the critical force to move a bubble depends on the change of radius of curvature of the gas-liquid interface.

Marchessault and Mason [19] considered flow of bubbles through capillary tubes and also mentioned the Jamin effect and hysteresis of the contact angle, but stated that when a liquid wets a solid surface there is no contact angle, as was discussed above. The authors evaluated the force required to move a bubble by considering the viscous resistance of a liquid moving in a film between a bubble and the wall of a capillary tube. The effective film thickness was measured electrically and was correlated with the measured hydrodynamic resistance to flow of the bubble. The experimental data confirmed the resistance equations which were developed.

There have been many papers written concerning the dynamics of growth and shrinking of air bubbles due to pressure and temperature changes.

This is of particular interest in regard to the phenomenon of cavitation. Several of these are Dergarabedian [9], Birkhoff, Margulies and Horning [2], Ma and Wang [17] and Zwick [31].

Dissolution of Bubbles. In recent years there have been several articles written considering, both theoretically and experimentally, the time for a single air bubble to dissolve when immersed in a liquid of large extent.

One of the first of these articles was by Epstein and Plesset [12]. In their development, Fick's first law,

$$\frac{dm}{dt} = AD \frac{\partial C}{\partial r} \quad (2-1)$$

and Fick's second law,

$$\frac{\partial C}{\partial t} = D \left[\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} \right] \quad (2-2)$$

using a spherical coordinate system, were used to develop a theoretical equation for rate of dissolution. In the above equations m is the mass of air going into solution, A is the cross-sectional area through which diffusion occurs, D is the coefficient of diffusion of air in the liquid, C is the concentration of dissolved air in the liquid, and r is the radius measured from the center of the bubble. Neglecting surface tension, the equation is

$$\frac{da}{dt} = \frac{D(C_i - C_s)}{\rho_a} \left[\frac{1}{a} + \frac{1}{(\pi Dt)^{1/2}} \right] \quad (2-3)$$

When surface tension is considered, the equation becomes

$$\frac{da}{dt} = \frac{D(C_i - C_s)}{\rho_a + \frac{4M\sigma}{3BTa}} \left[\frac{1}{a} + \frac{1}{(\pi Dt)^{1/2}} \right] \quad (2-4)$$

where a is the bubble radius, C_i is the uniform dissolved air concentration at the bubble boundary, ρ_a is the air density at atmospheric pressure and the temperature of the system, M is the molecular weight of the gas under standard conditions of temperature and pressure, B is the universal gas constant, T is the absolute temperature and σ is the surface energy of the liquid.

In order to solve these two equations, the last term in the brackets was assumed to be negligible. This is somewhat questionable as the term is certainly not negligible when time is near zero. Epstein and Plesset used the solution of equation (2-4) and values of $D = 2 \times 10^{-5} \text{ cm}^2/\text{sec}$, $\rho_a = 0.0012 \text{ gm/cm}^3$, $C_s = 0.000024$ for a temperature of 22° C , and determined the values in Table 1 for the time for a bubble to dissolve in water.

Liebermann [16] obtained experimental data for the rate of dissolution and used this to verify the solution of equation (2-3). In this experimental work the concentration of dissolved air in the water was originally about 50% to 60% of the saturation at atmospheric pressure, therefore the effect of surface tension was said to be negligible. Figure 1a shows the experimental data and a theoretical curve which will be discussed below. These data were then used by Liebermann to determine the value of D at 27° C , as $2.9 \times 10^{-5} \text{ cm}^2/\text{sec}$. This seems quite high

TABLE 1

Time for Air Bubbles to Dissolve in Water

Initial Radius (cm)	Time (min)
10^{-3}	0.110
10^{-2}	98.
10^{-1}	96,600.

as a more common value is about 2.0×10^{-5} cm²/sec. The fact that the data do not follow the theoretical curve was attributed to a skin effect due to contaminants on the surface of the bubble.

In part of Liebermann's study, various contaminants such as oleic acid, detergent, mineral oil and sea water were used to determine whether or not the rate of solution was changed. No significant change was observed.

Manley [18] obtained extensive experimental data on the rate of dissolution of bubbles in de-aerated water which are given in Figures 1a and 1b. The theoretical line in each case is the solution of equation (2-4) using the following values for the constants.

$$C_i = 22.5 \times 10^{-6} \text{ (.90) for Manley data}$$

$$= 22.5 \times 10^{-6} \text{ (.40) for Lieberman data}$$

$$C_s = 22.5 \times 10^{-6}$$

$$\rho_a = 0.0012 \text{ gm/cc}$$

$$M = 29 \text{ gm/22.4 liters}$$

$$B = 8.31 \times 10^7 \text{ ergs/}^\circ\text{K mole}$$

$$T = 299^\circ\text{K}$$

$$\sigma = 71.97 \text{ dynes/cm}$$

$$D = 2 \times 10^{-5} \text{ cm}^2/\text{sec}$$

The agreement with the theoretical curves is good except when the bubble becomes quite small where (in nearly all cases) the rate of diffusion becomes slower. Manley gives four possible explanations of this: 1) the effect of solid boundaries and saturated layers of dissolved gas around the bubble, 2) a layer of charged particles on the bubble wall, 3) dipole moments of the molecules on the bubble wall, and 4) the presence of an organic skin material surrounding the bubble.

The first three theories were then discarded because the change of rate of diffusion was found to be independent of the original bubble size, there was a slight change of the diffusion rate in water which has recently been well de-aerated, and the addition of a wetting agent Lissapol N eliminated the reduction. After accepting the presence of an organic skin, Manley theoretically evaluated a diffusion coefficient (having a value of 2.6×10^{-6} cm²/sec or about 1/8 that of air through clean water) for the layer.

Both Liebermann and Manley have taken photographs of a deposit of contamination left when a bubble dissolves and have shown that if the pressure is then lowered so that air will come out of solution, the contamination usually serves as a nucleus for the formation of air bubbles.

Several other authors have considered the organic skin effect. One of these is Brandstaetter [3] who discussed H. Mache's gas bubble method for the quick determination of the diffusion coefficient of gases in liquids. Brandstaetter showed that the apparent dependence of the diffusion coefficient on the bubble radius was in reality due to the presence of contaminants.

Levenspiel [15] also photographed the residue left by the collapse of a bubble and observed that the rate of collapse decreased as the bubble became small.

Fox and Herzfeld [11] wrote about the organic skin of gas bubbles as nuclei for cavitation bubbles. In a theoretical analysis, an equation for the time of dissolution was developed in a slightly different manner than that used by Epstein and Plesset but the computed times were comparable.

In the previously mentioned solutions to the diffusion equations, two assumptions were that the air diffused into an infinite liquid and the liquid at the boundary of the bubble did not move. In the case of many bubbles dissolving in a liquid, neither one of these conditions is strictly satisfied. Therefore, it would be desirable to develop an equation considering a moving boundary at bubble walls and air diffusing into liquid within some finite radius.

Vyrodov [27] considered a solid particle dissolving and diffusing into a finite region. The moving boundary was considered and the solute dissolved into a finite region but a rather complex numerical solution was obtained.

Suquet [26] solved the diffusion equations for an air bubble with several different boundary conditions by the use of a Laplace transformation. One of these conditions was that of air diffusing into a finite volume of liquid; however, air was also diffusing into this volume from the exterior.

THEORY

Surface Energy

One of the important factors controlling the behavior of two or more immiscible fluids in a porous medium is surface energy at the interface between the two fluids. Excellent discussions of surface energy are given by Adams [1], Davies and Rideal [8] and Rouse [25]. Basically, a distinct interface is produced between immiscible fluids by the attractive forces between molecules of each fluid. A molecule in the interior of a fluid is attracted in all directions with equal forces. A molecule on the surface, however, is attracted towards the interior more than towards the surface because there are more molecules in that direction. This causes a maximum number of molecules to be at the interior of the fluid for a given surface area. The result is that fluid drops assume spherical shapes when the surface energy forces are predominant unless the solid boundary of the fluid container prevents this.

A curved interface between two fluids is often treated mathematically as possessing tensile forces parallel to the interface. A theoretical treatment is also possible by work-energy methods.

An equation developed by either method is

$$\Delta P = \sigma \left(\frac{1}{r_1} + \frac{1}{r_2} \right) \quad (3-1)$$

where ΔP is the pressure difference across the interface, σ is the interfacial energy constant, and r_1 and r_2 are the principal radii of curvature of the interface. For a spherical surface such as an isolated air bubble in a liquid, $r_1 = r_2 = r$ or

$$\Delta P = \frac{2\sigma}{r} \quad (3-2)$$

The pressure is greatest on the concave side of the surface which means, in the case of an air bubble in a liquid, that the pressure is greater inside the bubble than in the liquid.

When the interface between two fluids comes in contact with a solid surface, the solid will become wet with one fluid in preference to the other. Thus, when a two fluid system is considered, one fluid is termed the wetting and the other the non-wetting fluid. The concave side of the interface is always on the side of the non-wetting fluid and therefore, considering equation (3-2), the pressure is always less in the wetting fluid than in the non-wetting. The pressure difference (pressure of the non-wetting fluid minus the pressure of the wetting fluid) is called capillary pressure P_c .

Physical Properties of Porous Media

In the development of theoretical equations describing the rate of diffusion of entrapped air from a porous medium, it is desirable to use the fewest number of properties which adequately

describe the particular medium. Brooks and Corey [4] consider that two important parameters of porous media are: 1) the bubbling pressure, an index of the maximum pore size forming a continuous network of flow channels, and 2) the pore-size distribution index, an index of the distribution of sizes of flow channels.

The bubbling pressure is defined as the minimum capillary pressure at which the air permeability is greater than zero. The pore-size distribution index as used by the above authors is the slope of the curve of effective saturation versus capillary pressure on logarithmic paper. The slope of the curve of relative permeability versus capillary pressure on logarithmic paper can also be used for this index since there is a unique relationship between the two slopes. The effective saturation and relative permeability both refer to the liquid phase. The reader is referred to the paper by Brooks and Corey [4] for the definition of "effective saturation."

A curve of these variables for an unconsolidated column of glass beads is shown in Figure 2 on the following page. In Figure 2 the relative permeability k_r is the effective permeability k_e at any particular capillary pressure divided by the saturated permeability k . Thus a value of $k_r = 1$ is for a saturated condition. Both the desired parameters P_b and η may be obtained from this curve.

It should be noted that there is a different relationship for drainage than for imbibition, although the two curves have approximately the same slope. The imbibition curve ordinarily becomes nearly level at a value of k_r approximately equal to 0.50. This part of the imbibition curve is time dependent due to diffusion of entrapped air from the medium. This is indicated by the rise in k_r at $P/\rho g = 40$ cm. over a period of eleven days. The curves are separated in the lower portion because of hysteresis and the "ink bottle" effect (as discussed in the review of literature) and in the upper section by the effect of entrapped air.

It is hypothesized that the bubbling pressure should be an index of the concentration of dissolved air since it is an index of the excess pressure in the air. The value of η might affect the amount of entrapped air since more air might be trapped when the pores have a wider range of sizes. In fact, this has been observed experimentally by Gupta and Swartzendruber [13].

Theoretical Model

The system treated theoretically consists of a homogeneous, isotropic, porous material with easily defined exterior boundaries. A liquid is assumed to fill the sample uniformly throughout with air trapped in many pores. The air pockets are assumed to be completely surrounded by the liquid; that is, the surface of all solid particles of the sample are "wet" by the liquid.

The pressure in the liquid is atmospheric which gives a pressure within the entrapped air that

is greater than atmospheric by an amount given by equation (3-2).

It is known from Henry's law that the equilibrium concentration of dissolved air in a liquid is proportional to the partial pressure of air in contact with the liquid. It is assumed that the original concentration of dissolved air is uniform and equal to the equilibrium concentration for atmospheric pressure. Since the pressure in the air is greater than atmospheric, air tends to go into solution. The air enclosed by interfaces having the smallest radii of curvature is the first to dissolve since the pressure difference is greatest there. As air dissolves, the concentration of dissolved air increases until it reaches some value which is in equilibrium with the air in pockets having the smallest radii of curvature in the medium. It is possible for pockets with larger radii of curvature to increase in size during this time since the liquid may be super saturated with air in comparison to these pockets. The maximum curvature, however, is limited by the maximum pore dimensions.

As the liquid becomes super saturated with air in comparison to atmospheric pressure, air diffuses out of the liquid at the exterior boundary of the sample and a concentration gradient occurs through the sample. As this occurs, the concentration decreases which allows more air to dissolve. Eventually the concentration reaches some value at which all of the air dissolves and the sample is completely saturated even though all the air has not diffused out of the system.

In the case of a flowing liquid phase the liquid leaving the medium has a greater dissolved

air content than that entering and therefore carries the dissolved air out of the sample.

In order to express this mathematically it is assumed that the volume of air in the sample at any time is given by the amount which is originally trapped, less that which is in solution in the liquid, that which has diffused from the exterior boundary and that which has been carried out by a flowing liquid. Expressed in equation form

$$V_a = V_{ao} - \frac{\rho_l}{\rho_a} \int_0^{V_1} C_t dV - A \alpha \frac{\rho_l}{\rho_a} \int_0^t C_B dt - \frac{\rho_l}{\rho_a} \int_0^t C_f Q dt \quad (3-3)$$

where V_a is the volume of air in the sample, V_{ao} is the volume of air originally trapped, ρ_l is the mass density of the liquid, ρ_a is the mass density of the undissolved air, C_t is the dissolved air concentration in the volume element dV at a given time, V_1 is the volume of liquid at any time, A is the exterior area of the sample, C_B is the dissolved air concentration at the boundary of the sample as a function of time, α is a constant of proportionality, C_f is the dissolved air concentration in the liquid flowing out, and Q is volume rate of liquid flow. The unit of concentration used is parts per million by mass; therefore, to change to a volume basis it is necessary to multiply the concentration by the density ratio ρ_l/ρ_a . The proportionality constant α has the dimensions of velocity.

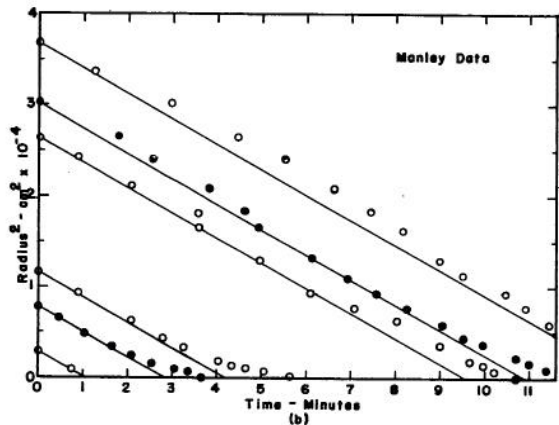
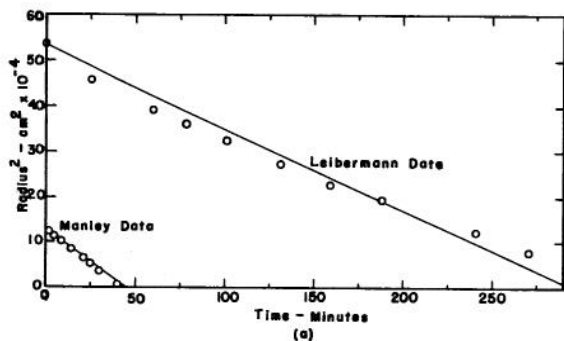


Figure 1. Bubble Radius as a Function of Time

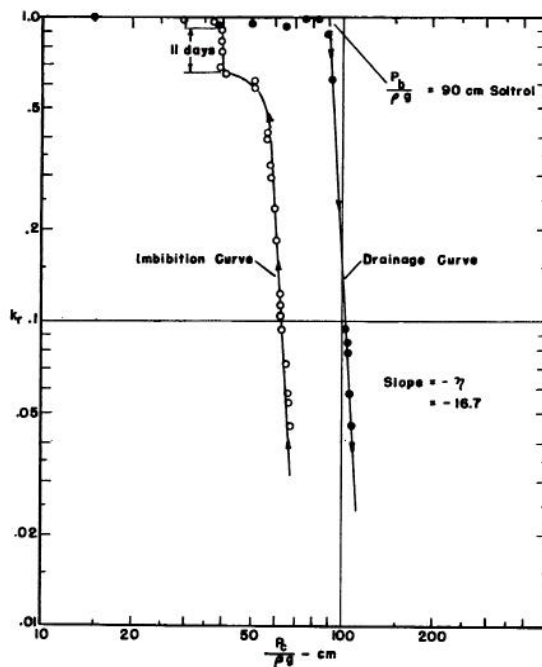


Figure 2. Relative Permeability versus Capillary Pressure Head

Concentration of Dissolved Air

The greatest difficulty in using equation (3-3) is the evaluation of the concentrations C_t , C_B , and C_f . Before evaluating these terms, it is desirable to determine the relative magnitudes of the terms and then to neglect any that are small.

Considering the second term on the right side of equation (3-3) and assuming that C_t is constant throughout the medium, the volume of air, ΔV_a , which can be dissolved in the liquid is

$$\Delta V_a = \frac{\rho_l C_t V_l}{\rho_a} \quad (3-4)$$

It was mentioned previously that the bubbling pressure is used as an index of the excess pressure, and if this is done the value of C_t can be computed directly. The largest value of $P_b/\rho g$ for any of the cores used in this study is 90 cm of oil. The corresponding value of C_t is 18.0 parts per million or $C_t = 18 \times 10^{-6}$. For this core and oil $\rho_l = .753 \text{ gm/cm}^3$ and $\rho_a = 1.01 \times 10^{-3} \text{ gm/cm}^3$. Putting these values into equation (3-4)

$$\begin{aligned} \Delta V_a &= \frac{.753 (18) (10)^{-6} V_l}{(1.01) 10^{-3}} \\ &= 0.0134 V_l \end{aligned}$$

The maximum volume of air which can be dissolved in the liquid is thus of the order of 1% of the liquid volume. The second term in equation (3-3) is, therefore, neglected.

In order to determine the time distribution of concentration at the exterior boundary of the sample, attempts were made to determine the rate at which a single bubble would dissolve into a limited volume of fluid. Various solutions were obtained but none which would satisfy all the boundary conditions. However, if the time for a bubble to dissolve is rapid compared to the time required for the air to diffuse from the sample, the concentration throughout the core will become nearly uniform before appreciable air diffuses from the exterior face.

In order to justify the assumption that the time of dissolution is rapid, consider the core with the largest pore size which was used in this study. This core was made of glass beads and had a "hypothetical pore radius" (this term is explained on page 11) of 0.0061 cm.

The maximum bubble size should be less than 0.01 cm. From Figure 1b the time for a bubble of this size to dissolve in an infinite fluid is less than five minutes. The time could be greater for an air pocket in a porous medium, but even a time of several hours would be small in comparison to the total time of diffusion which in all instances was several hundred hours or greater.

Diffusion of Dissolved Air

The experimental study of diffusion was made with two different boundary conditions: 1) the

side of the core was sealed such that diffusion was from the end of the core (one-dimensional) and 2) the end of the core was sealed such that diffusion was radial (two-dimensional).

In the theoretical development for both these conditions the following ideas are postulated: Soon after air becomes trapped, the liquid in the sample reaches some equilibrium concentration of dissolved air, C_o . The dissolved air at the exterior boundary of the sample begins to diffuse out but only from some incremental thickness. When the air has diffused out of this thickness sufficiently to allow all the air to dissolve, the air in the next increment dissolves, etc. Thus, the point at which air dissolves moves through the sample as a sharply defined front. Towards the interior from the front the same amount of air is entrapped as initially while the concentration of dissolved air is C_o . Towards the exterior from the front there is a concentration gradient with the concentration varying from C_o at the front to C_B at the exterior boundary. In this region all the air is dissolved. This assumes that the distribution of pore sizes in which air is entrapped is uniform.

Longitudinal Diffusion. Consider in Figure 3 the flow of air from the exterior face. This is proportional to the concentration C_B , or

$F = \alpha \frac{C_B \rho_l}{\rho_a}$ where F is the volume flux and α is a constant of proportionality.

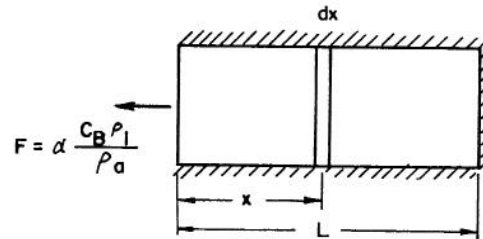


Figure 3. Diagram for Longitudinal Diffusion

It was shown previously that the amount of air which is in solution is small; therefore, the change of amount of dissolved air to the left of the front is negligible with respect to the total amount in the core. The flow from the exterior face is therefore approximately equal to the rate of diffusion through the region x and to the rate at which air goes into solution at the moving front. The rate at which the dissolved air diffuses is

$$F = - \frac{D(C_o - C_B) \rho_l}{x \rho_a} \quad (3-5)$$

where D is the coefficient of diffusion of air in the liquid and x is the distance shown in Figure 3. The rate at which air goes into solution is

$$F = - \frac{V_{ao}}{V} \frac{dx}{dt} \quad (3-6)$$

where V is the total volume of the core. The term V_{ao}/V may be written as

$$\frac{V_{ao}}{V} = \frac{V_{ao}}{V_v} \frac{V_v}{V}$$

but

$$\frac{V_{ao}}{V} = 1 - S_o$$

and

$$\frac{V_v}{V} = \phi$$

where S_o is the original liquid saturation when air becomes trapped, V_v is the volume of voids, and ϕ is the porosity. Substituting into equation (3-6)

$$F = -(1 - S_o) \phi \frac{dx}{dt} \quad (3-7)$$

Combining equations (3-5) and (3-7)

$$\frac{D(C_o - C_B)}{x} \frac{\rho_1}{\rho_a} = (1 - S_o) \phi \frac{dx}{dt}$$

Rearranging terms and integrating

$$\int_0^t (C_o - C_B) dt = \frac{\rho_a (1 - S_o) \phi}{\rho_1 D} \int_0^x x dx,$$

or

$$C_o t - \int_0^t C_B dt = \frac{\rho_a (1 - S_o) \phi}{\rho_1 D} \frac{x^2}{2}$$

Since C_B is an unknown function of time, the integration cannot be completed. The equation may be solved for the integral, or

$$\int_0^t C_B dt = C_o t - \frac{\rho_a (1 - S_o) \phi}{\rho_1 D} \frac{x^2}{2} \quad (3-8)$$

Considering the case of a non-flowing liquid, equation (3-3) may be written as

$$V_a = V_{ao} - \frac{\rho_1}{\rho_a} \int_0^{V_1} C_t dV - A \alpha \frac{\rho_1}{\rho_a} \int_0^t C_B dt$$

But since the second term on the right side was shown to be small,

$$V_a = V_{ao} - A \alpha \frac{\rho_1}{\rho_a} \int_0^t C_B dt,$$

or

$$V_f = A \alpha \frac{\rho_1}{\rho_a} \int_0^t C_B dt \quad (3-9)$$

where V_f is the volume of air which has diffused from the core. Substituting equation (3-8) into (3-9)

$$V_f = A \alpha \frac{\rho_1}{\rho_a} \left[C_o t - \frac{\rho_a (1 - S_o) \phi}{\rho_1 D} \frac{x^2}{2} \right] \quad (3-10)$$

The volume of air which has diffused from the core is proportional to x , or

$$\frac{x}{L} = \frac{V_f}{V_{ao}}$$

Substituting this expression for x into (3-10) and rearranging terms

$$\frac{\rho_a V_f}{\rho_1 A \alpha} = C_o t - \frac{\rho_a \phi L^2 (1 - S_o)}{\rho_1 2D} \left[\frac{V_f}{V_{ao}} \right]^2$$

Dividing by the coefficient of the $\frac{V_f}{V_{ao}}$ term

$$\frac{2DV_{ao}}{\phi(1-S_o)A\alpha L^2} \frac{V_f}{V_{ao}} = \frac{2\rho_1 DC_o t}{\rho_a (1-S_o)\phi L^2} - \left(\frac{V_f}{V_{ao}} \right)^2$$

but

$$\frac{V_{ao}}{\phi(1-S_o)} = V = AL$$

so that

$$\left(\frac{V_f}{V_{ao}} \right)^2 + \frac{2D}{\alpha L} \frac{V_f}{V_{ao}} - \frac{2C_o}{(1-S_o)\phi} \frac{\rho_1 Dt}{\rho_a L^2} = 0$$

Solving this quadratic equation for V_f/V_{ao}

$$\frac{V_f}{V_{ao}} = \frac{1}{2} \left[-\frac{2D}{\alpha L} \pm \sqrt{\left(\frac{2D}{\alpha L} \right)^2 + \frac{8C_o}{(1-S_o)\phi} \frac{\rho_1 Dt}{\rho_a L^2}} \right]$$

or

$$\frac{V_f}{V_{ao}} = \sqrt{\left(\frac{D}{\alpha L} \right)^2 + \frac{2C_o}{(1-S_o)\phi} \frac{\rho_1 Dt}{\rho_a L^2}} - \frac{D}{\alpha L} \quad (3-11)$$

The positive sign is selected for the radical, since from physical considerations V_f/V_{ao} must be positive.

Radial Diffusion

By reasoning analogous to that used for linear diffusion, an equation for radial (two-dimensional) diffusion may be obtained. This equation* is:

$$\frac{Dt}{R^2} = \frac{\frac{V_f}{V_{ao}} + \frac{\alpha R}{2D + \alpha R} \left[1 - \frac{V_f}{V_{ao}} \right] \ln \left[1 - \frac{V_f}{V_{ao}} \right]}{\frac{4C_o \rho_1}{(1-S_o)\phi \rho_a} \frac{\alpha R}{2D + \alpha R}} \quad (3-12)$$

where R is the radius of the core.

*A detailed derivation of this equation is given by George L. Bloomsburg, in a Ph.D. thesis, Colorado State University, 1964.

EXPERIMENTAL PROCEDURE

The porous materials used in the experimental work were consolidated cores of three types, i. e., sandstone, a ceramic material and glass beads. The cores were all approximately one inch in diameter and varied from three fourths to two and one half inches in length. Two different liquids, water and a hydrocarbon oil* were used for the wetting fluid in the ceramic and glass bead cores. Only Soltrol was used in the sandstone cores since most sandstones contain some clay which swells and changes the physical properties of the material being wet with water.

The experimental work on each core consisted of determining the physical properties of the core and measuring the rate of diffusion of entrapped air under several different boundary conditions. The first condition used was that of imbibition from the lower end of the core with the upper end sealed such that all diffusion of air occurred radially from the core. The next condition was that of imbibition through the lower end with the sides sealed such that diffusion occurred longitudinally from the upper end. The last condition was that of liquid flowing from top to bottom through the core with the sides exposed such that there was radial diffusion of air and also air carried out by the flowing fluid.

Determination of Physical Properties of Cores

The physical properties of the cores which were needed were the porosity, bubbling pressure and the pore size distribution index, the significance of which were explained in the section dealing with theory.

The porosity is defined as the ratio of the volume of voids in a material to the total volume of the material. The volume of voids was determined by weighing the oven dried core, then vacuum saturating the core with a liquid of known density and weighing the saturated core to determine the volume of liquid.

The bubbling pressure and pore size distribution index were both determined in the same experiment. A schematic diagram of the apparatus used is shown in Figure 4.

The tensiometer rings consisted of a plastic ring with a groove on the inside connected to the manometer tube. The groove was covered with a porous plastic to act as a capillary barrier. The barrier was in contact with the outside of the core. The use of these capillary barriers and the barriers on the end of the core made it possible to have liquid pressures in the core less than atmospheric without air getting into the manometer, inflow and outflow tubes. The core with tensiometer rings and capillary barriers in place was initially vacuum saturated. It was then placed in a vertical position as shown and the inflow, outflow, and manometer tubes were con-

nected. The supply reservoir was at first higher than the sample to insure that the core did not become desaturated during the period in which the various tubes were being connected.

The first reading was taken with the capillary pressure slightly greater than zero so that the liquid would not flow out of the sample. When the level in the manometer tubes reached an equilibrium with a hydraulic gradient of unity, the outflow was measured per unit time. Darcy's law written in the following form was used to compute the effective permeability

$$k_e = \frac{q \mu_l}{\rho g \Delta H / \Delta L} \quad (4-1)$$

where k_e is the effective permeability, μ_l is the viscosity of the liquid and q is the volume flux per unit area.

The capillary pressure was increased by increments with the sample remaining saturated and the permeability remaining constant until the capillary pressure reached the air entry pressure for the particular medium and liquid. The relative permeability, k_r , was equal to one through this range.

After the sample began to desaturate, the effective permeability was again computed using equation (4-1) but since k_e was less than k , k_r was less than one. A typical curve of k versus P_c is shown in the section dealing with theory. From this curve the bubbling pressure, P_b , and the pore size distribution index η were obtained.

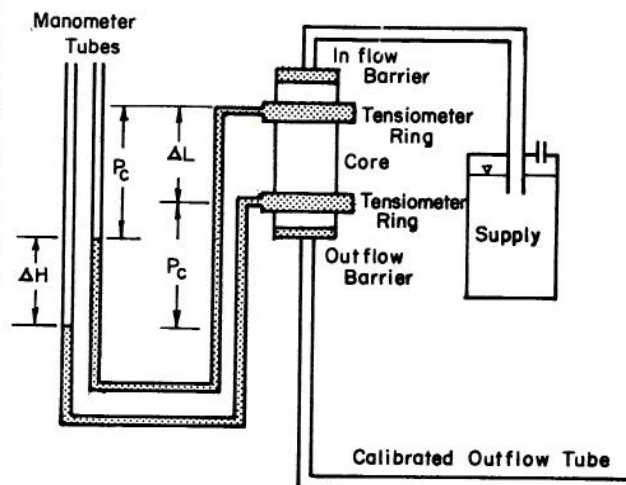


Figure 4. Schematic Diagram of Apparatus for Determining P_b and η

*Soltrol "C" core test fluid. Phillips Petroleum Company, Special Products Div., Bartlesville, Oklahoma.

Diffusion of Air From the Cores

To measure the rate of diffusion of air from the cores, they were placed in a vertical position with the lower end in the liquid (water or Soltrol). They were kept inside a closed plastic container so that there was little evaporation from the exposed surface of the core. Liquid was always on the bottom of the container so that the air in the container was saturated with the liquid vapor.

The cores were oven dried, weighed, and placed in the container. Shortly after the wetting front reached the top of the cores, they were removed and weighed. It was observed that a fairly rapid increase of saturation occurred for a short time after this. Handy [14] also observed this and attributed it to entrapped air being compressed from excess pressure in the bubble. Part of this increase in saturation could also be due to air being dissolved in the liquid. The amount of entrapped air was therefore somewhat less than the volume of empty voids when the wetting front reached the top of the core. When the saturation data were plotted versus time, however, it was relatively easy to obtain a consistent value for

initial saturation by extending a smooth curve back to the ordinate where the abscissa equals zero.

When each core was removed to be weighed, it was placed in an air tight glass bottle to eliminate evaporation during weighing. The lower end of the core, after being removed from the liquid, was "blotted" against a hard smooth surface to remove the excess liquid before being placed in the weighing bottle.

A plastic adhesive tape was used to seal the end or sides of the cores. In some cases there was trouble with liquid collecting under the tape on the outside of the core.

In the flowing liquid case the cores were placed in a plastic container with liquid dripping on the top and flowing out the bottom. This resulted in downward flow at a hydraulic gradient of approximately one and a capillary pressure of zero. The flow rate was measured in some cases so that the total flow through the core could be obtained approximately. The same method was used for weighing the cores as for the non-flowing case.

PRESENTATION OF DATA AND RESULTS

Physical Properties of Cores

Table 2 on the following page gives all physical properties which were determined for the sixteen cores used in the study. All data were obtained at a temperature of $26.0 \pm 2.0^\circ \text{C}$.

In this table, the column labeled pore radius lists a hypothetical radius computed from the bubbling pressure by the equation for pressure difference across a spherical interface. This is

$$\Delta P = \frac{2\sigma}{r}$$

or

$$r = \frac{2\sigma}{\Delta P}$$

However, the bubbling pressure is usually determined as a height of fluid or

$$h = \frac{\Delta P}{\rho g}$$

and

$$r = \frac{2\sigma}{\rho g h}$$

where h is the bubbling pressure head in cm of the liquid. A hypothetical radius has been shown by experience to be an index of an important property of porous media.

The column labeled C_o gives the concentration of dissolved air in parts per million by mass which would be in equilibrium with a spherical bubble of the same radius as the pore radius given in the previous column.

Figure 5 on the following page gives a graphical solution for both C_o and P as a function of bubble radius with water and Soltrol being the liquids considered. The values of solubility of air in water which were used to develop the curve were supplied by Dr. Kemper of the Colorado State University Agronomy Department. The values of solubility of air in Soltrol were determined experimentally as described in Appendix A.

Figures 6 through 9 are the curves of relative permeability versus capillary pressure head for all the cores.

Diffusion of Entrapped Air

Curves of saturation versus time for all cores are shown in Figures 10 through 17. In most cases there are three curves for each core, but in some cases fewer. Cores 360-2, 98-1 and 98-2 were so slow in becoming saturated that it was not possible to use the condition of having the side sealed. The data obtained for this condition with cores 2 and 3 were erratic due to liquid collecting under the tape used for sealing. The weight was corrected when the tape was removed, but it was not always possible to know at what time the correction should be applied.

For most cores complete saturation was obtained fastest with flow and radial diffusion while the slowest was with one-dimensional diffusion only. Exceptions to this occurred with core No. 2 where, in the flowing case the time to reach complete saturation was longer than with radial diffusion only and with core No. 360-2 in which case, for linear diffusion this time was essentially the same as with radial diffusion only. One factor which might have affected the results for No. 2 was a stoppage of flow during one night. The discrepancy in the data from No. 360-2 could have been caused by a slightly greater initial saturation in the linear diffusion case than in the radial case.

With cores S-2 and L-2 (which consist of glass beads) complete saturation with water occurred most rapidly in the linear diffusion case, but this probably was due to a large difference in the initial saturation for the two cases. The data from these four cores which appeared to be erratic were not used in the analysis in the following section.

The saturation when air became trapped was quite consistent for each core regardless of the exterior boundary conditions except when using water. When water was used in the glass bead cores, there was a large difference in the initial saturation for different experimental runs (See Figures 17 and 18).

Only two of the cores (No. 2 and No. 3) were allowed to reach complete saturation during linear diffusion. The reason for this was that liquid which collected under the sealing tape made it appear that complete saturation had been reached. When the tape was removed, however, the cores were found to be only about 95% saturated.

TABLE 2
MATERIAL PROPERTIES

Core No.	Material	Fluid	Length (cm)	Diameter (cm)	Dry Weight (Grams)	Saturated Weight (Grams)	Void Volume cm ³	Porosity
1	Alundum	Soltrol	5.07	2.54	55.92	63.29	9.80	.381
2	Sandstone	Soltrol	5.10	2.53	55.95	58.88	3.89	.152
3	Sandstone	Soltrol	5.17	2.52	53.22	57.01	4.90	.190
4	Sandstone	Soltrol	5.52	2.54	62.71	65.67	3.93	.141
5	Sandstone	Soltrol	5.17	2.48	47.88	52.46	6.09	.244
6	Sandstone	Soltrol	5.11	2.55	53.00	57.11	5.46	.209
7	Sandstone	Soltrol	4.54	2.54	52.71	54.81	2.79	.121
8	Sandstone	Soltrol	4.75	2.54	53.36	56.04	3.56	.148
360-1	Alundum RA 360	Soltrol	6.15	2.72	78.76	89.92	14.81	.415
360-2	Alundum RA 360	Water	6.29	2.72	79.02	93.86	14.90	.408
98-1	Alundum RA 98	Soltrol	6.26	2.72	77.88	88.34	13.90	.382
98-2	Alundum RA 98	Water	6.32	2.72	80.29	94.36	14.15	.385
L-1	Glass Beads	Soltrol	4.29	2.65	37.05	42.99	7.89	.336
L-2	Glass Beads	Water	1.79	2.65	15.63	18.88	3.26	.310
S-1	Glass Beads	Soltrol	4.31	2.51	30.37	36.56	8.21	.385
S-2	Glass Beads	Water	4.35	2.50	29.74	37.87	8.15	.382

TABLE 2
MATERIAL PROPERTIES (Continued)

Core No.	$P_b / \rho g$ (cm of fluid)	k_s (cm ² x 10 ⁻⁸)	Pore Radius (cm)	C_o (ppm)	S_o	t_s (Radial) (hr)	t_s (Linear) (hr)	t_s (Flowing) (hr)	
1	49	17.9	1.99	.00130	9.5	.65	1052	--	110
2	80	5.7	.0136	.000727	17.1	.78	185	590	300
3	78	11.4	.0377	.000745	16.7	.75	255	600	167
4	44	3.9	.0027	.00145	8.0	.48	1000	--	--
5	53	8.4	.167	.00120	10.4	.69	780	--	265
6	44	13.9	.429	.00145	8.0	.66	785	--	165
7	48	13.5	.155	.00133	9.4	.48	785	--	287
8	46	7.5	.172	.00139	9.0	.54	1600	--	410
360-1	90	15.2	.631	.00069	18.0	.75	330	--	190
360-2	180	15.2	.631	.00069	4.5	.73	1360	--	--
98-1	46.0	15.1	1.9	.00139	9.0	.65	1260	--	280
98-2	91.	15.1	1.9	.00139	2.25	.69	2780	--	--
L-1	10.5	16.6	47.2	.0061	2.00	.87	792	--	111
L-2	21	16.6	47.2	.0061	0.51	--	1200	--	--
S-1	24	16.1	14.8	.00266	4.7	.89	270	--	89
S-2	48	16.1	14.8	.00266	1.18	--	1075	--	--

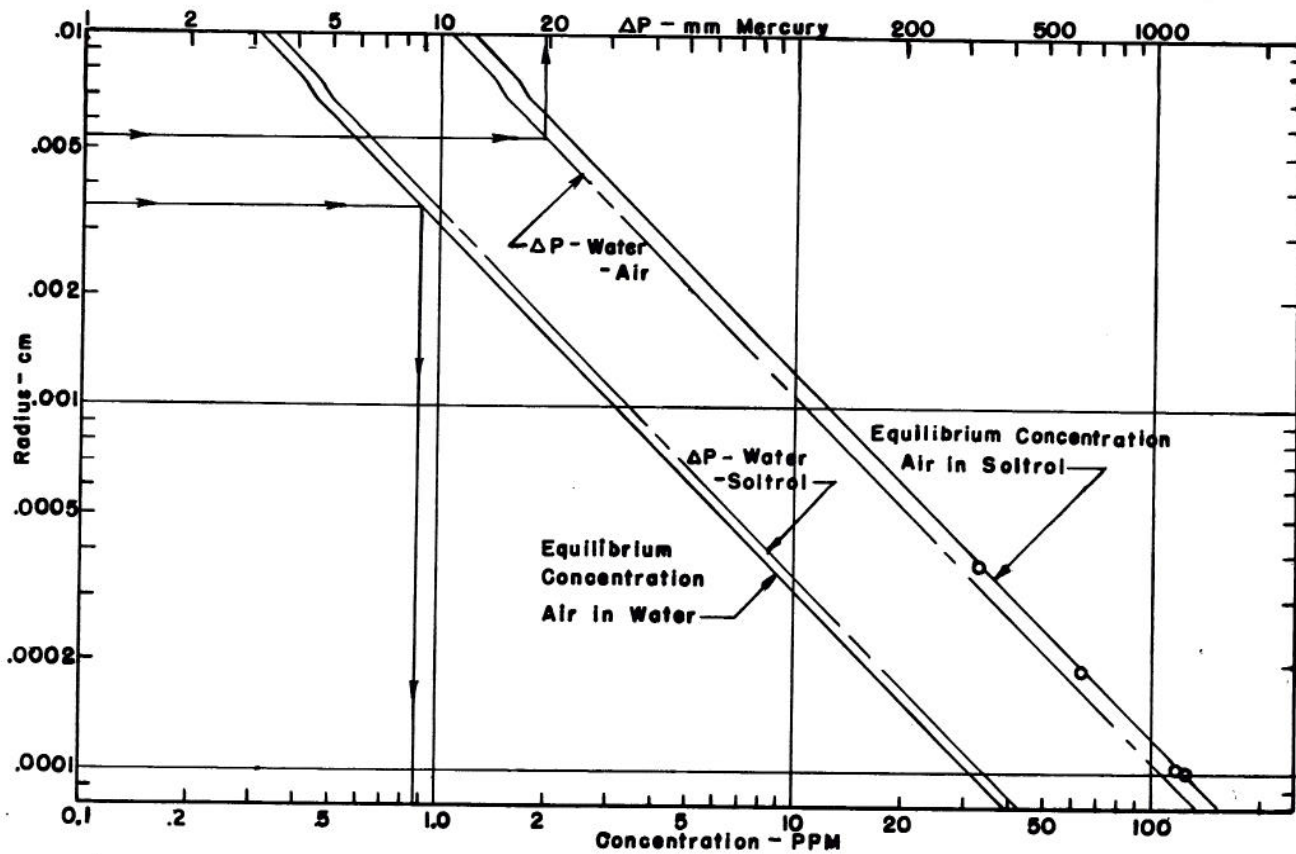


Figure 5. Concentration and ΔP as a Function of Bubble Radius

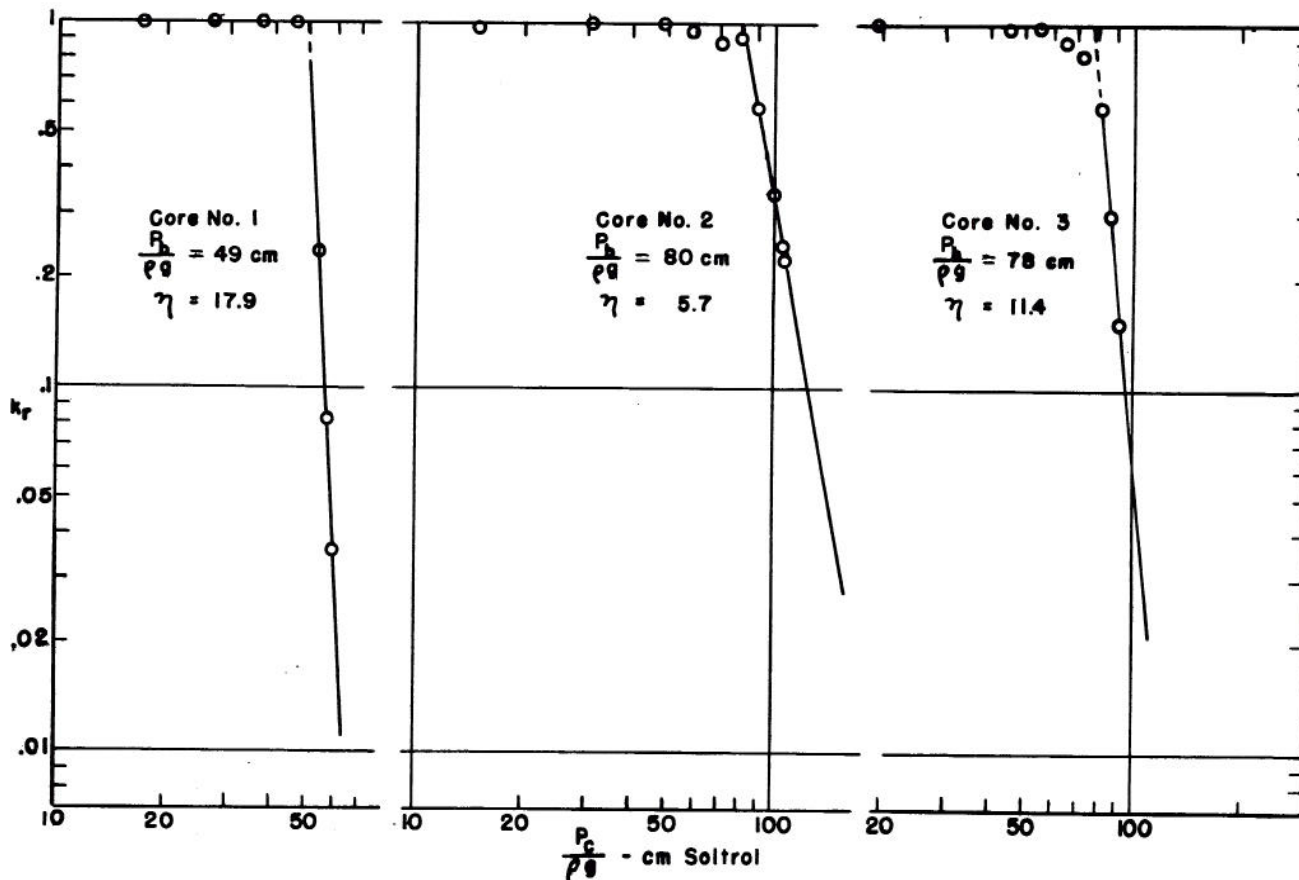


Figure 6. Relative Permeability versus Capillary Pressure Head

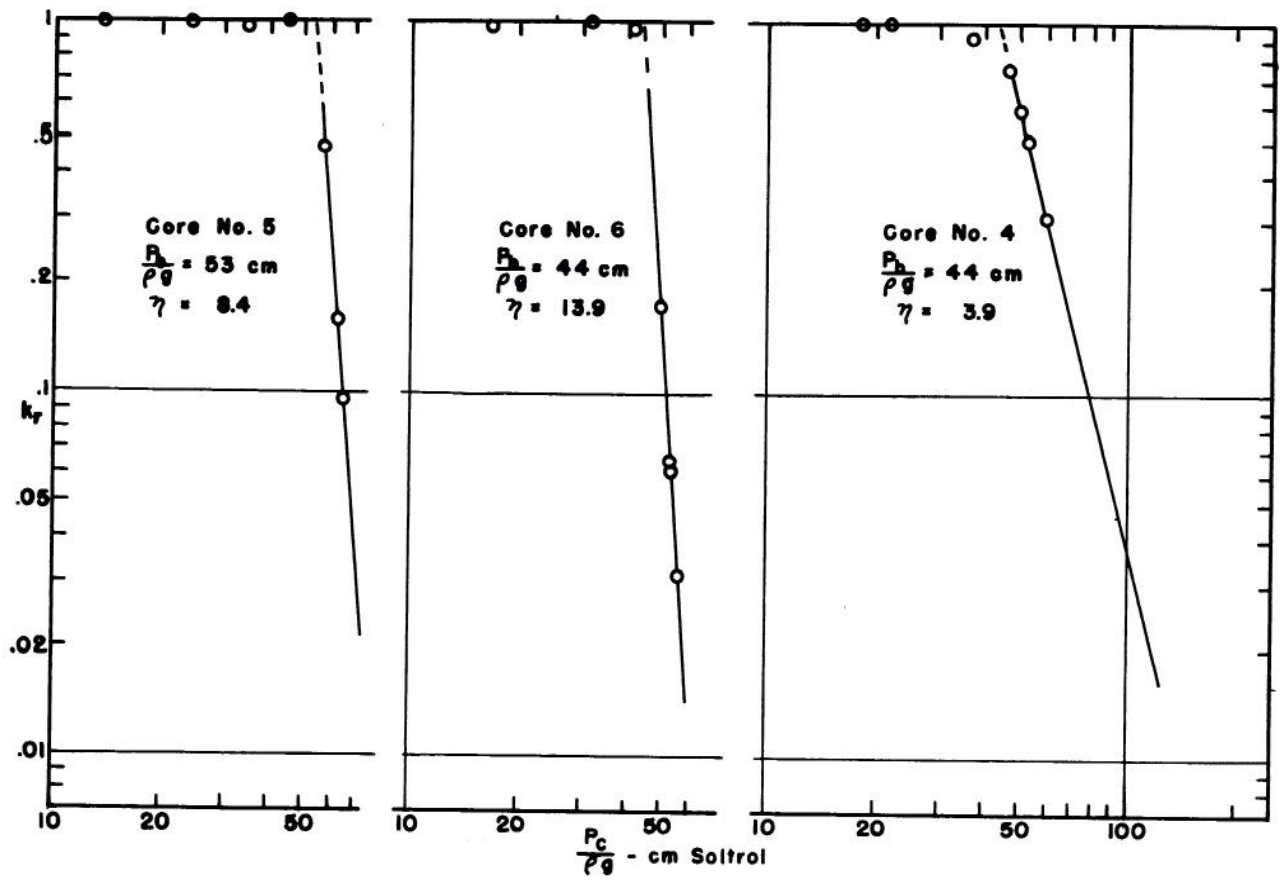


Figure 7. Relative Permeability versus Capillary Pressure Head

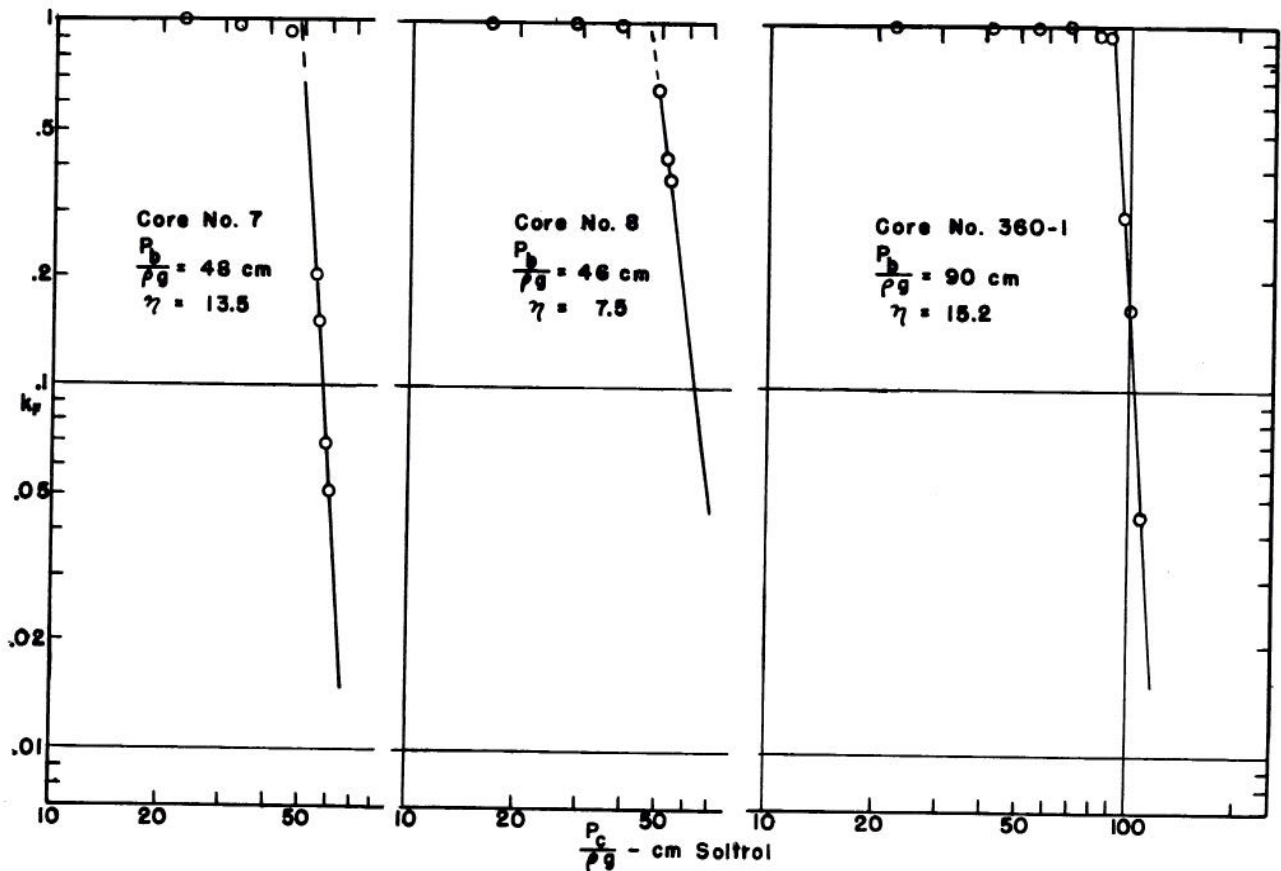


Figure 8. Relative Permeability versus Capillary Pressure Head

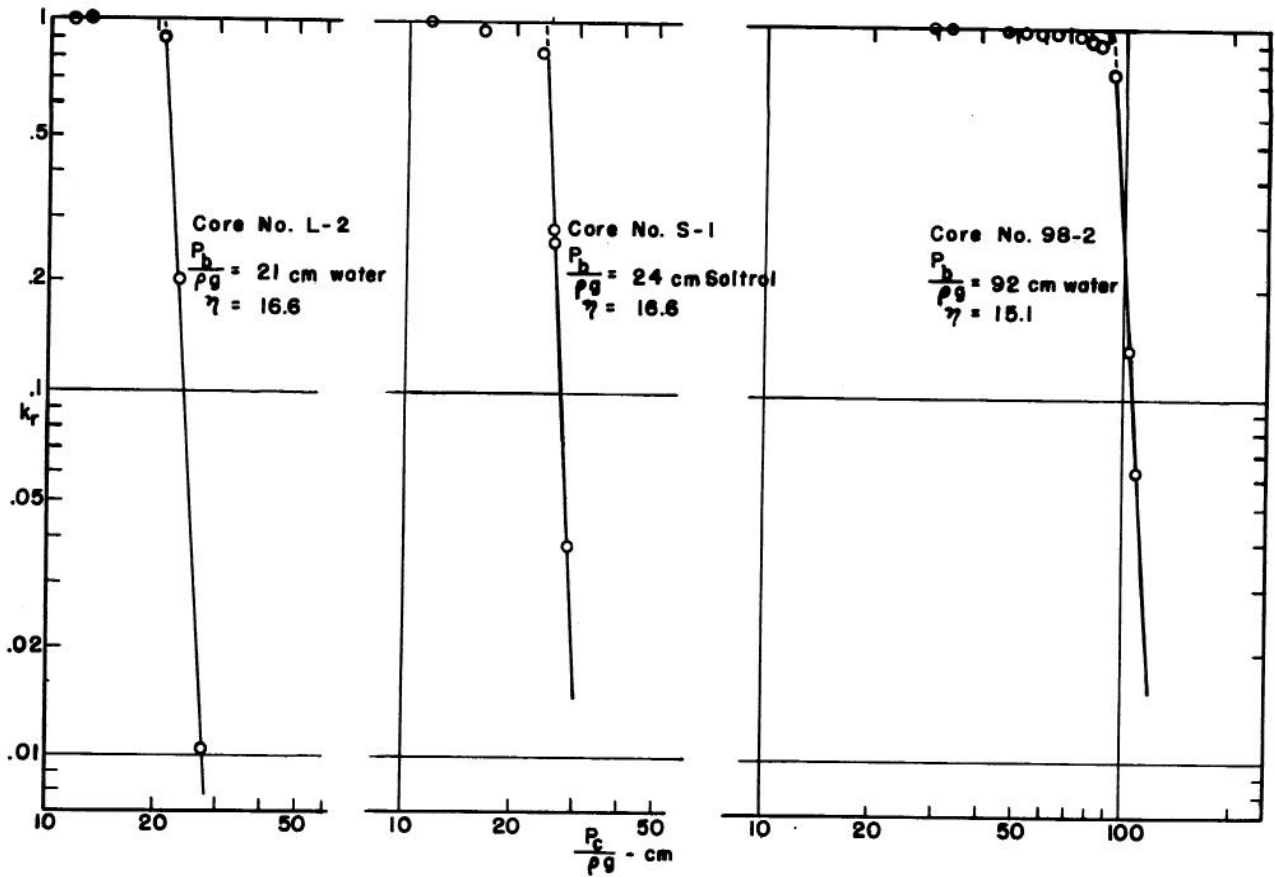


Figure 9. Relative Permeability versus Capillary Pressure Head

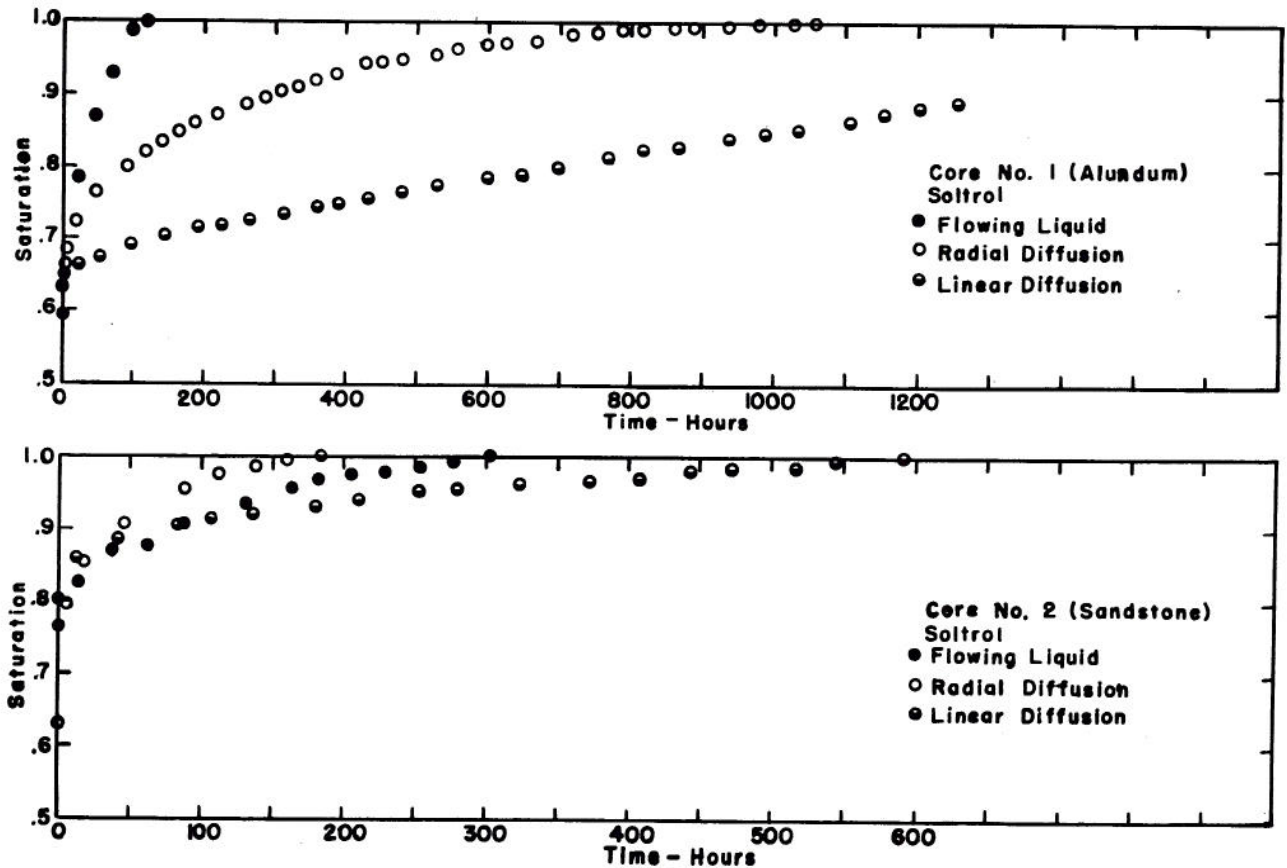


Figure 10. Saturation versus Time

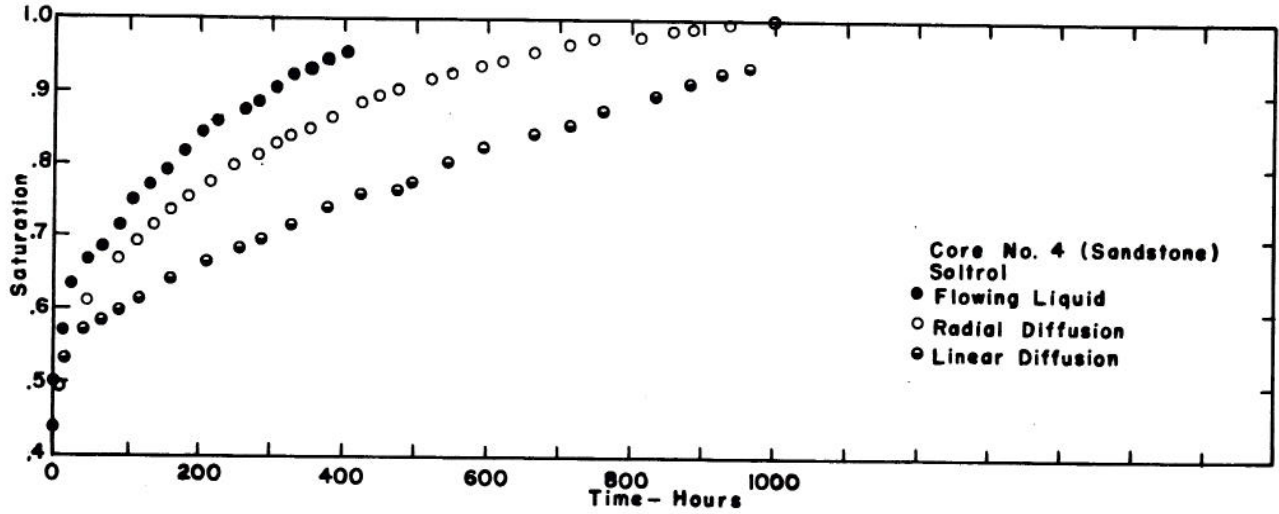
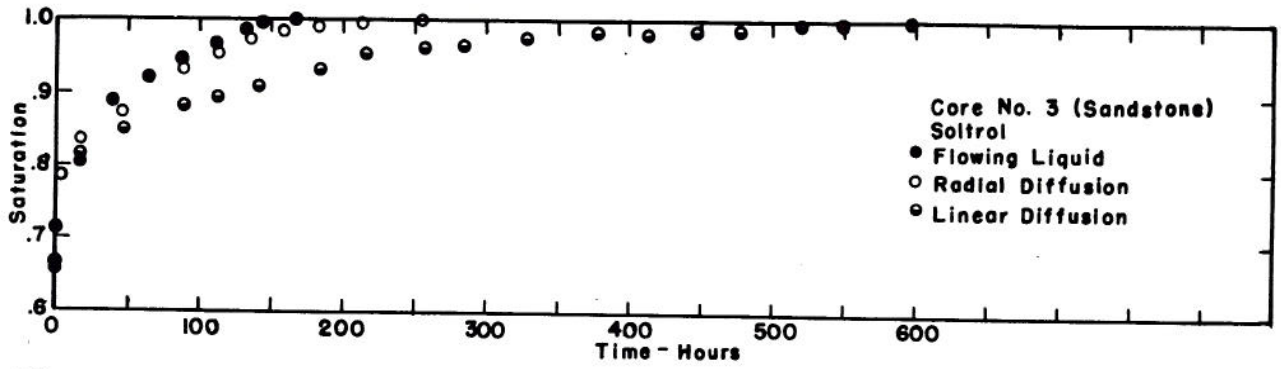


Figure 11. Saturation versus Time

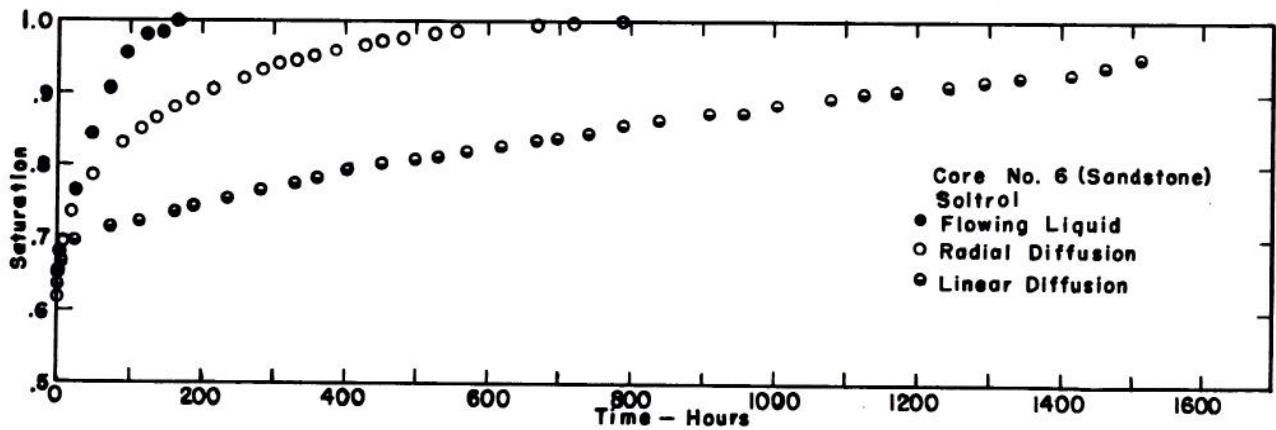
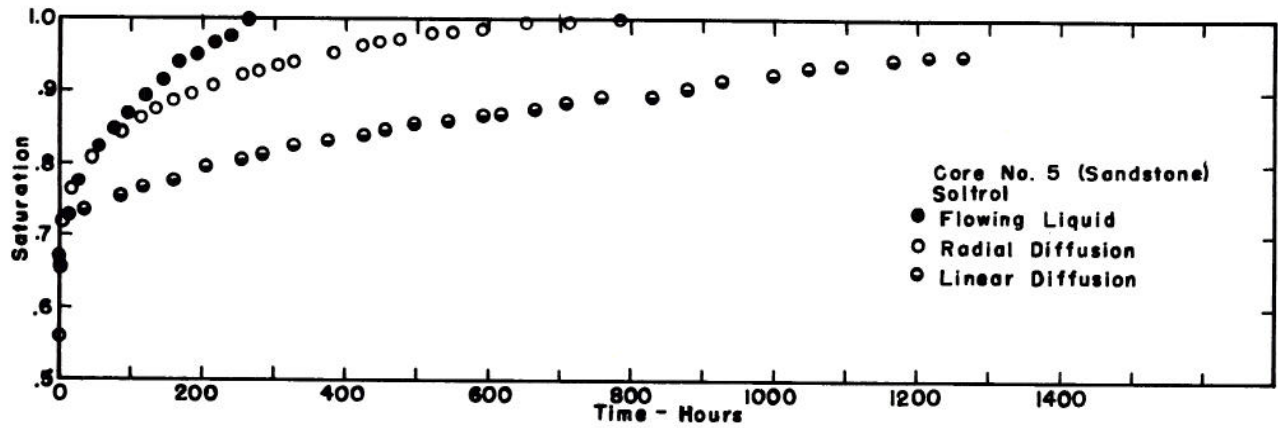


Figure 12. Saturation versus Time

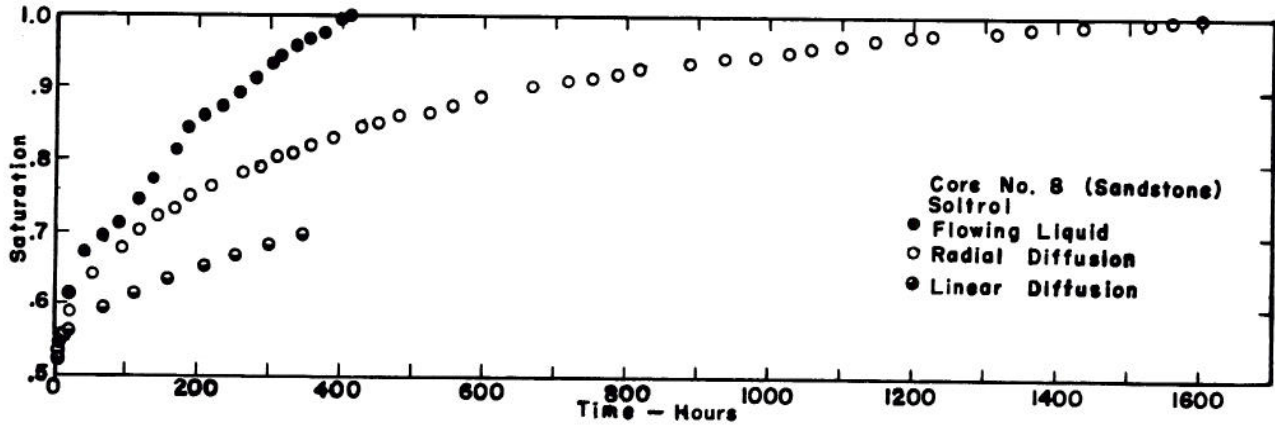
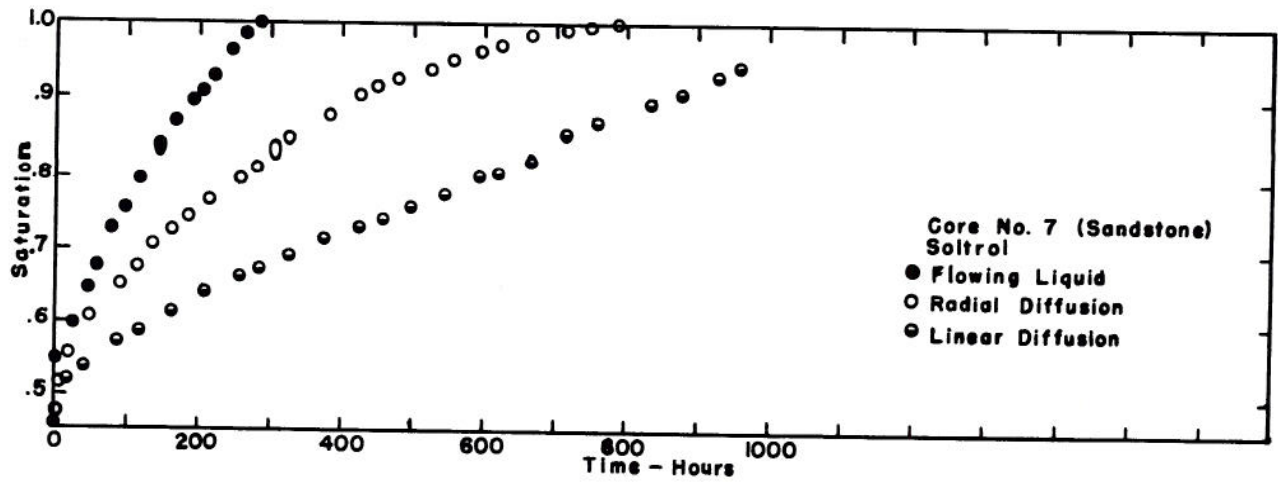


Figure 13. Saturation versus Time

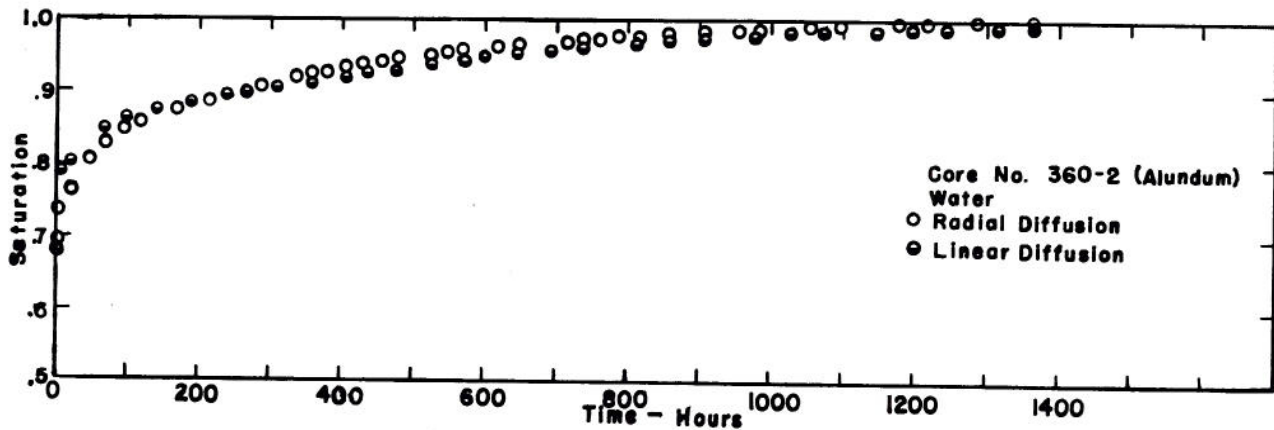
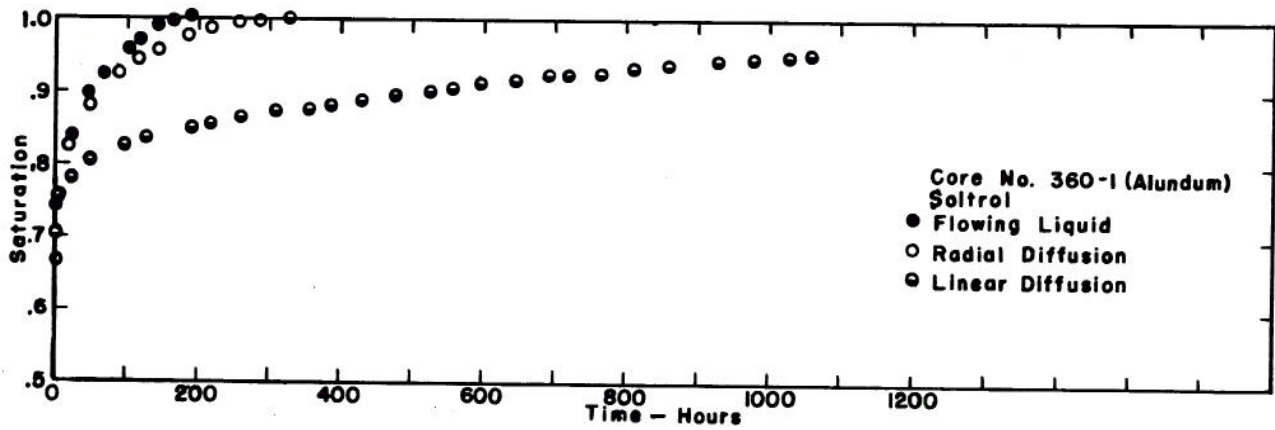


Figure 14. Saturation versus Time

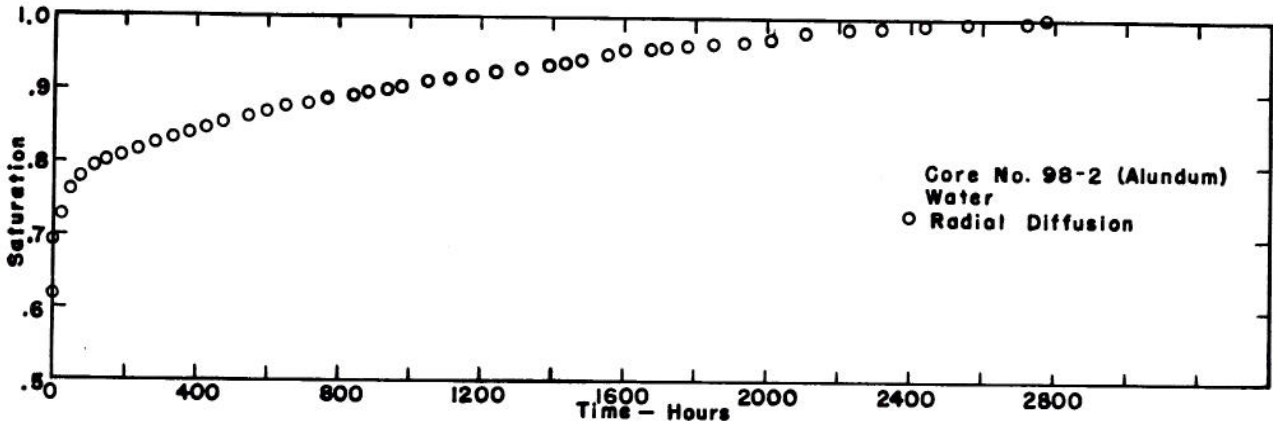
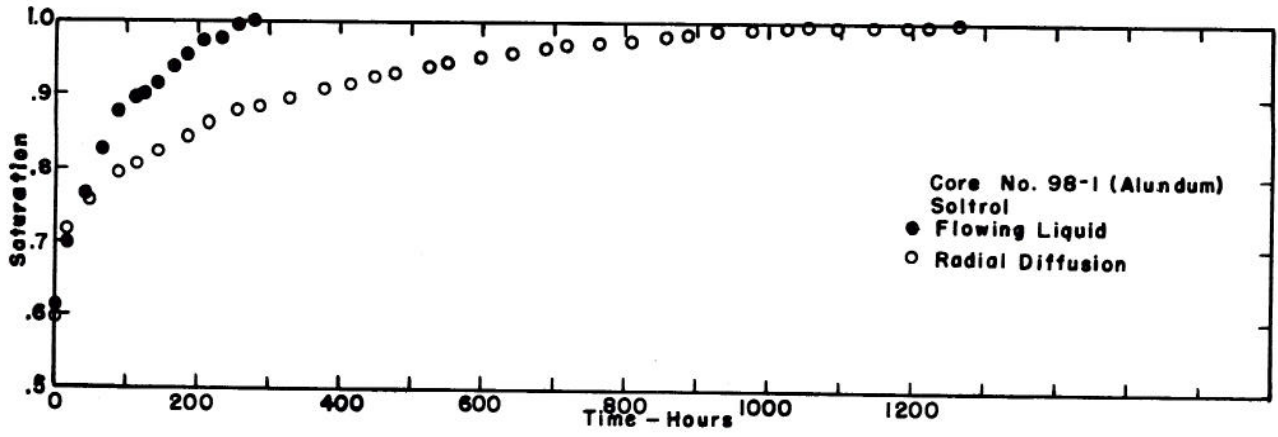


Figure 15. Saturation versus Time

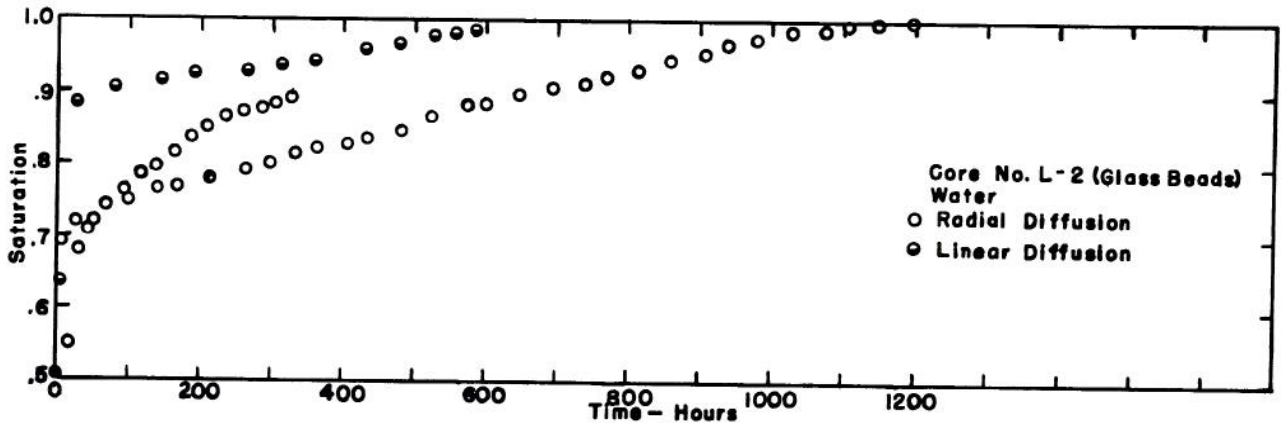
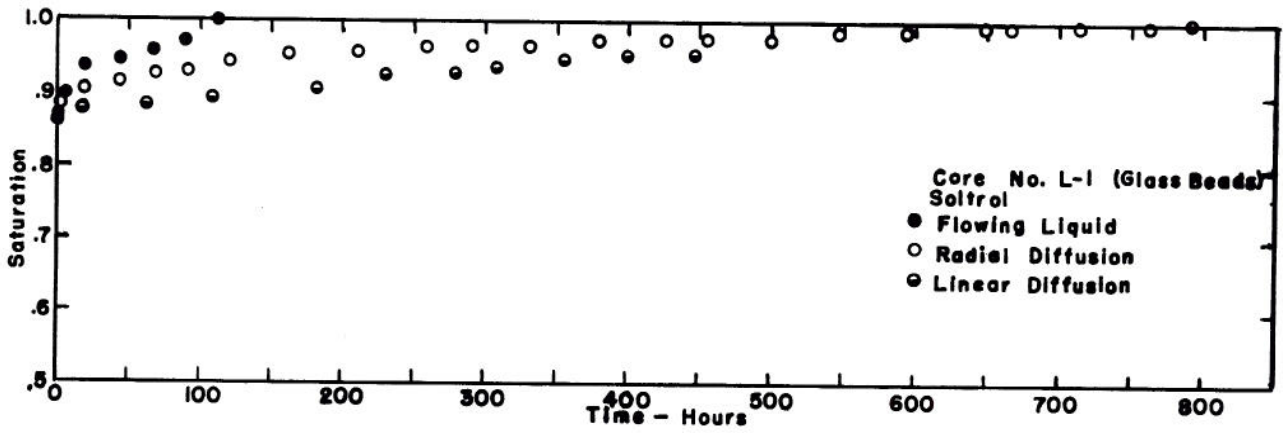


Figure 16. Saturation versus Time

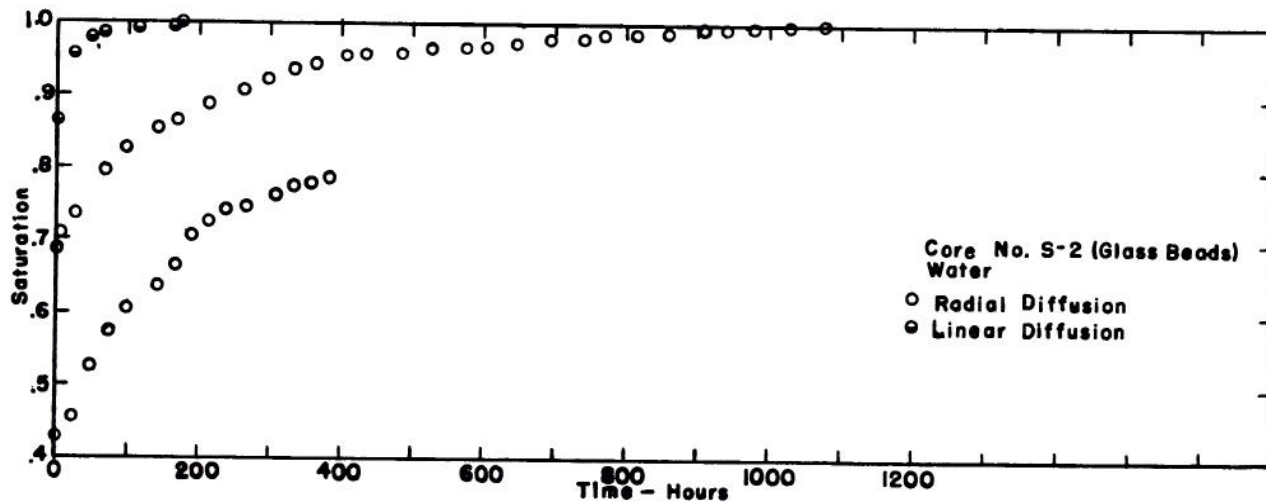
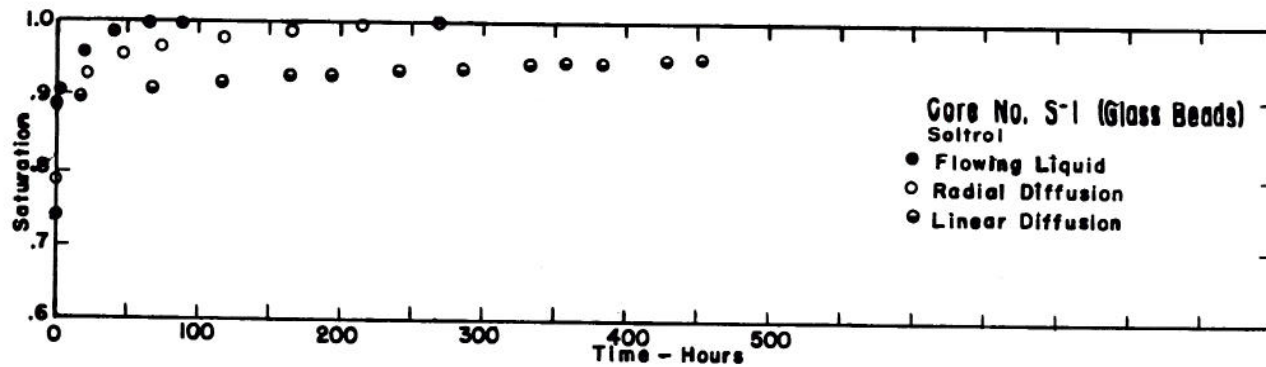


Figure 17. Saturation versus Time

ANALYSIS OF RESULTS

Amount of Entrapped Air

It was stated in the previous section that the amount of entrapped air was inconsistent when water was the imbibing liquid in the glass bead cores. It is felt that this was due to water not completely wetting the surface of the glass beads so that not all of the air in the core (when the wetting front reached the top) was necessarily trapped. It is known that very slight contamination of the surface of glass will greatly alter its wettability to water. The cores had been washed in acetone to remove contaminants but this apparently had little effect.

This problem was not encountered to as great an extent with water in the ceramic cores since all contaminants were burned out by heating the cores to approximately 1700° F. When these cores were allowed to stand in water for some time there was (apparently) microbiological growth because after being dried, water would not enter them immediately unless they were reheated. The wettability of oil is apparently not affected to as great an extent by commonly occurring contaminants.

Considering the cores in which the amount of entrapped air was consistent, (see Table 2), the glass bead cores with Soltrol had the least entrapped air while the sandstone cores had the most. Among the cores of similar materials, there appears to be less air entrapped in the cores with higher bubbling pressure and higher η values; however, there are exceptions to this.

It appears that the most significant variable affecting the amount of entrapped air may be particle shape or smoothness because the glass beads (smooth round particles) had the least entrapped air. When water is the imbibing fluid, wettability may be equally important.

Theoretical Analysis

Linear Diffusion. Equation (3-11) was given as the theoretical equation for the linear diffusion case, i. e.,

$$\frac{V_f}{V_{ao}} = \sqrt{\left(\frac{D}{\alpha L}\right)^2 + \frac{2C_o \rho_1}{(1-S_o)\phi \rho_a} \frac{Dt}{L^2}} - \frac{D}{\alpha L} \quad (3-11)$$

If V_f/V_{ao} is plotted versus $\left(\frac{Dt}{L^2}\right)^{1/2}$ the curve approaches a straight line of slope

$$\sqrt{\frac{2C_o \rho_1}{(1-S_o)\phi \rho_a}}$$

when the time variable becomes large relative to $(D/\alpha L)^2$. This straight line intersects the time axis at $V_f/V_{ao} = -D/\alpha L$. Actually because of two factors

$$\frac{2C_o \rho_1}{(1-S_o)\phi \rho_a}$$

is only an index of the slope. The two factors are: 1) the concentration C_o itself is only an index obtained from the bubbling pressure of the medium, and 2) the value of D used here is affected by the tortuosity of the liquid paths and the porosity of the porous medium.

Figures 18 and 19 show the relationship of V_f/V_{ao} versus $\left(\frac{Dt}{L^2}\right)^{1/2}$ for all the cores used in the study. Only a small number of data are shown for each core. In most cases the data approach a straight line which intersects the time axis at a negative value of V_f/V_{ao} .

The exceptions to this are cores No. 2, 3, and 360-1 which have the highest bubbling pressure. The data from these three all show a tendency to form curves which are concave downward. This is not predicted by the theoretical equation. For this reason, lines were not drawn through the points and the slope was not determined. The data for cores No. 2 and 3 appear to be somewhat erratic in that there are straight line portions and abrupt curves. It is thought this is partially due to experimental error. As previously mentioned, liquid collected under the tape which was used for sealing and the weight was corrected after the tape was removed. The correction was applied proportionately along the curve but perhaps, in some cases, this was incorrect.

The number which appears with each core identification number is the value of

$$\frac{2C_o \rho_1}{(1-S_o)\rho_a}$$

which is a parameter (appearing in equation (3-11) multiplied by the porosity. There is a tendency for the curves with larger values of this parameter to be steeper. An exception to this is core No. 4 which has a smaller value than any of the sandstone cores but a steeper slope than No. 5, 6, 7, or 8. This core has a low value of η (see Table 2) which would indicate a wide range of pore sizes. In this case the assumption of a uniform pore size for entrapped air may not be valid.

Radial Diffusion. The equation developed for the radial case is

$$\frac{Dt}{R^2} = \frac{\frac{V_f}{V_{ao}} + \left(\frac{\alpha R}{2D + \alpha R}\right) \left(1 - \frac{V_f}{V_{ao}}\right) \ln \left(1 - \frac{V_f}{V_{ao}}\right)}{\frac{4\rho_1 C_o}{\rho_a (1-S_o)\phi} \left(\frac{\alpha R}{2D + \alpha R}\right)} \quad (3-12)$$

This is difficult to work with unless an approximation is made for the numerator. Figure 20 shows a plot of the numerator versus values of V_f/V_{ao} for two values of the parameter $\frac{\alpha R}{2D + \alpha R}$. It should be noted that the value of this parameter is between 0 and 1 depending on the relative magnitude of α and D .

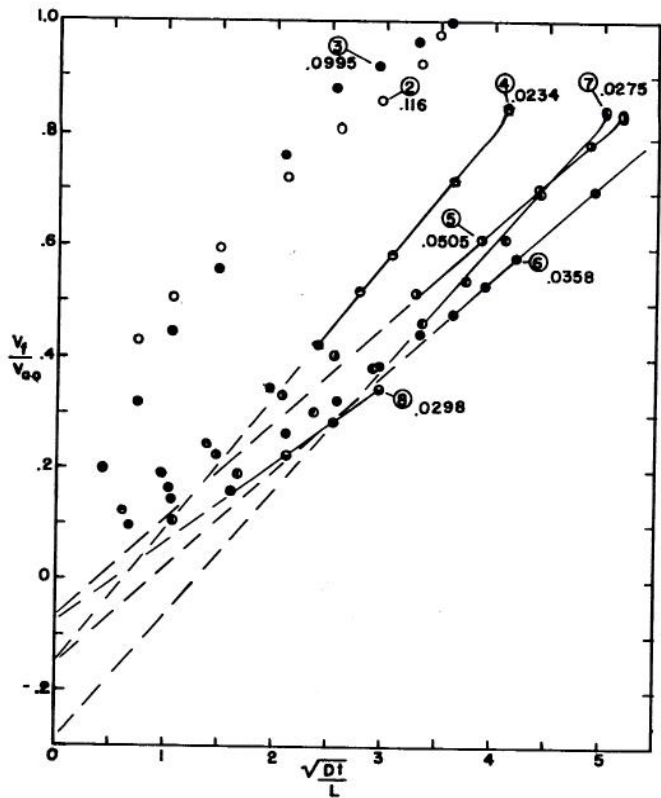


Figure 18. Plot of Dimensionless Variables Linear Diffusion - Sandstone Cores

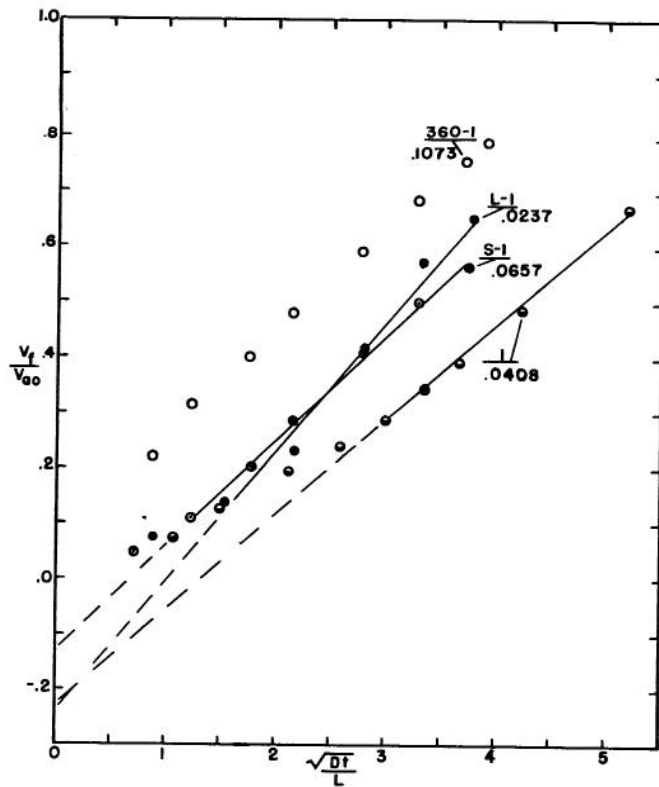


Figure 19. Plot of Dimensionless Variables Linear Diffusion - Ceramic and Glass Bead Cores

On the same figure are shown curves with

$$f\left(\frac{V_f}{V_{ao}}\right) = \left(\frac{V_f}{V_{ao}}\right)^{1/\xi}$$

for two values of ξ . These two functions are not identical but are somewhat similar. If the numerator of equation (3-12) is replaced by $\left(\frac{V_f}{V_{ao}}\right)^{1/\xi}$ where ξ varies from 1 to approximately $1/3$, the equation is

$$\frac{Dt}{R^2} = \frac{\left(\frac{V_f}{V_{ao}}\right)^{1/\xi}}{\frac{4\rho_1 C_o}{\rho_a(1-S_o)\phi} \frac{\alpha R}{2D+\alpha R}}$$

or

$$\frac{V_f}{V_{ao}} = \left[\frac{4\rho_1 C_o}{\rho_a(1-S_o)\phi} \frac{\alpha R}{2D+\alpha R} \right]^\xi \left(\frac{Dt}{R^2}\right)^\xi \quad (6-1)$$

A plot of V_f/V_{ao} versus Dt/R^2 on logarithmic paper is a straight line of slope ξ with the value of the term in brackets given by the value of V_f/V_{ao} at $Dt/R^2 = 1$.

Figure 21 shows a plot of V_f/V_{ao} versus Dt/R^2 for the sandstone cores used in the study. In most cases the data are close to a straight line but

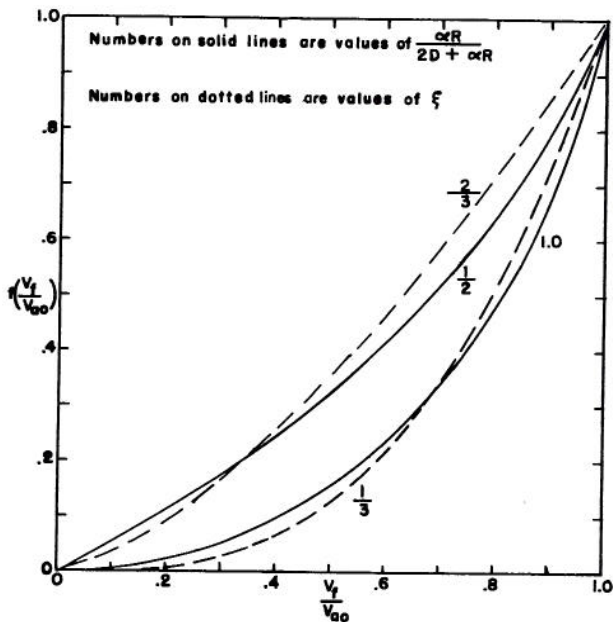


Figure 20. Plot of $f\left(\frac{V_f}{V_{ao}}\right)$ versus $\frac{V_f}{V_{ao}}$

curve away as the value of $V_f/V_{ao} = 1$ is approached. The slope of the line in most cases is approximately $1/2$, which is between the theoretical limits of $1/3$ and 1 for ξ .

Figure 22 shows the same data for the ceramic and glass bead cores. For several cores the data follow a straight line better than in the previous figure but still curve as $V_f/V_{ao} = 1$ is approached. This is probably due to the substitution

of $\left(\frac{V_f}{V_{ao}}\right)^{1/\xi}$ for $\frac{V_f}{V_{ao}} + \frac{\alpha R}{2D + \alpha R} \left[1 - \frac{V_f}{V_{ao}}\right] \ln \left[1 - \frac{V_f}{V_{ao}}\right]$ in equation (3-12) to obtain equation (6-1).

Analysis of Flowing Liquid Case

The computed average concentration of dissolved air in the oil which flowed through the various cores is shown in Figure 23. The amount of oil was either measured or computed by assuming a hydraulic gradient of one and estimating the value of relative permeability k_r . The amount of air which had been removed by the oil was estimated from the difference in saturation at a particular time between the radial case and the flowing case. The values of concentration are not considered to be very accurate; however, the concentration is consistently less than was listed in Table 2. In most cases the concentration appears to decrease with time which would be expected. The concentrations in the ceramic and glass bead cores appear to be less than in the sandstone cores regardless of bubbling pressure.

An improvement in this part of the study possibly could be made by having the sides of the cores sealed. By this method nearly all the air which left the system would be carried out by the moving fluid.

Considering the slight differences in concentration for the different cores, it appears that in the flowing liquid case, the most important variable affecting the time for complete saturation to be reached is the permeability of the sample. The permeability controls the flow rate of liquid and varies by a factor of 2×10^4 in the cores used in this study. The concentration given in Table 2 varies by a factor of less than 10 in the cores in which Soltrol was used.

Summary

In general, the experimental results substantiate the theory which was developed. It was not possible to obtain a completely analytical solution because the exact values of several parameters were not determined. These were: 1) the actual concentration of dissolved air, C_o , 2) the value of the proportionality constant α , and 3) the value of tortuosity and its effect on the coefficient of diffusion, D .

The bubbling pressure was used as an index of C_o but perhaps some value of capillary pressure from the imbibition curve would be a better index. This would give a lower value of C_o for each core which is also indicated by the results from the flowing liquid experiments.

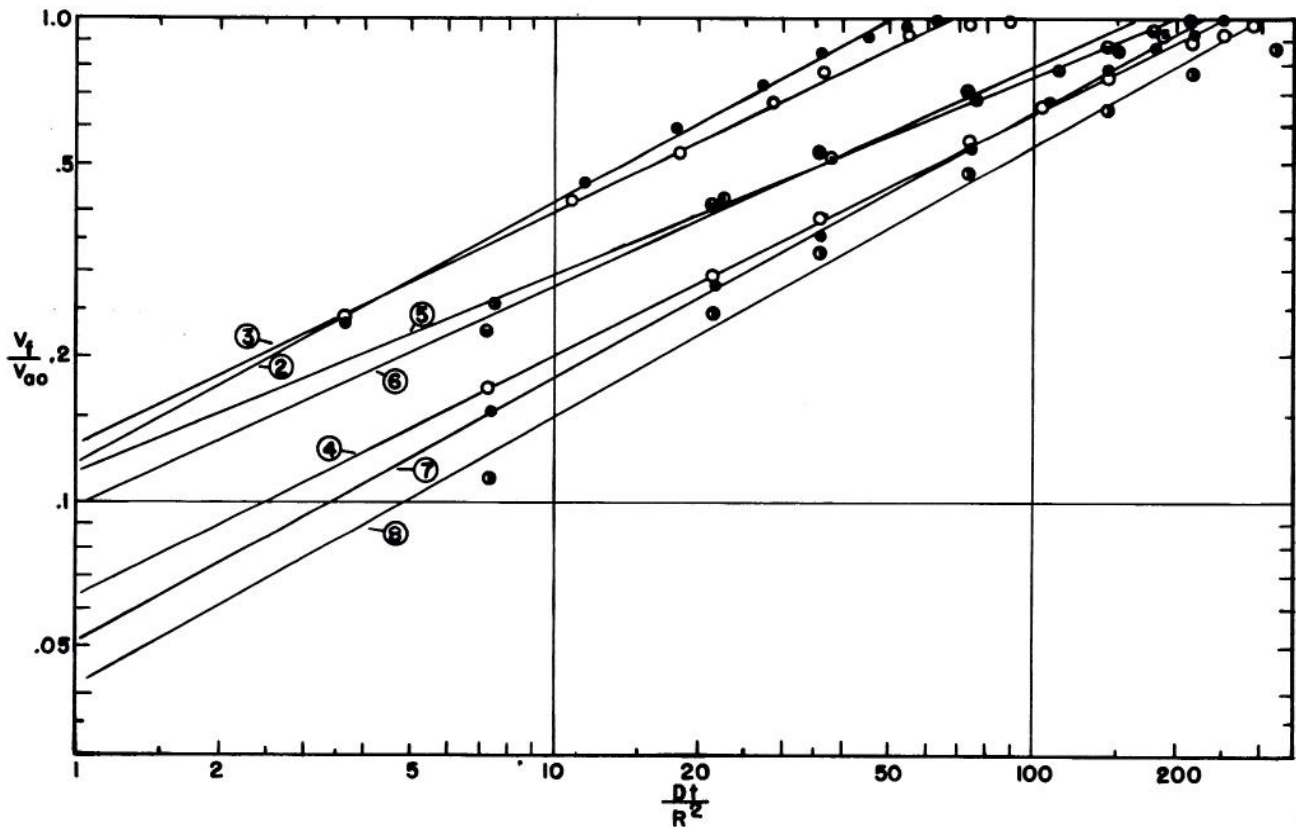


Figure 21. Logarithmic Plot of Dimensionless Variables Radial Diffusion - Sandstone Cores

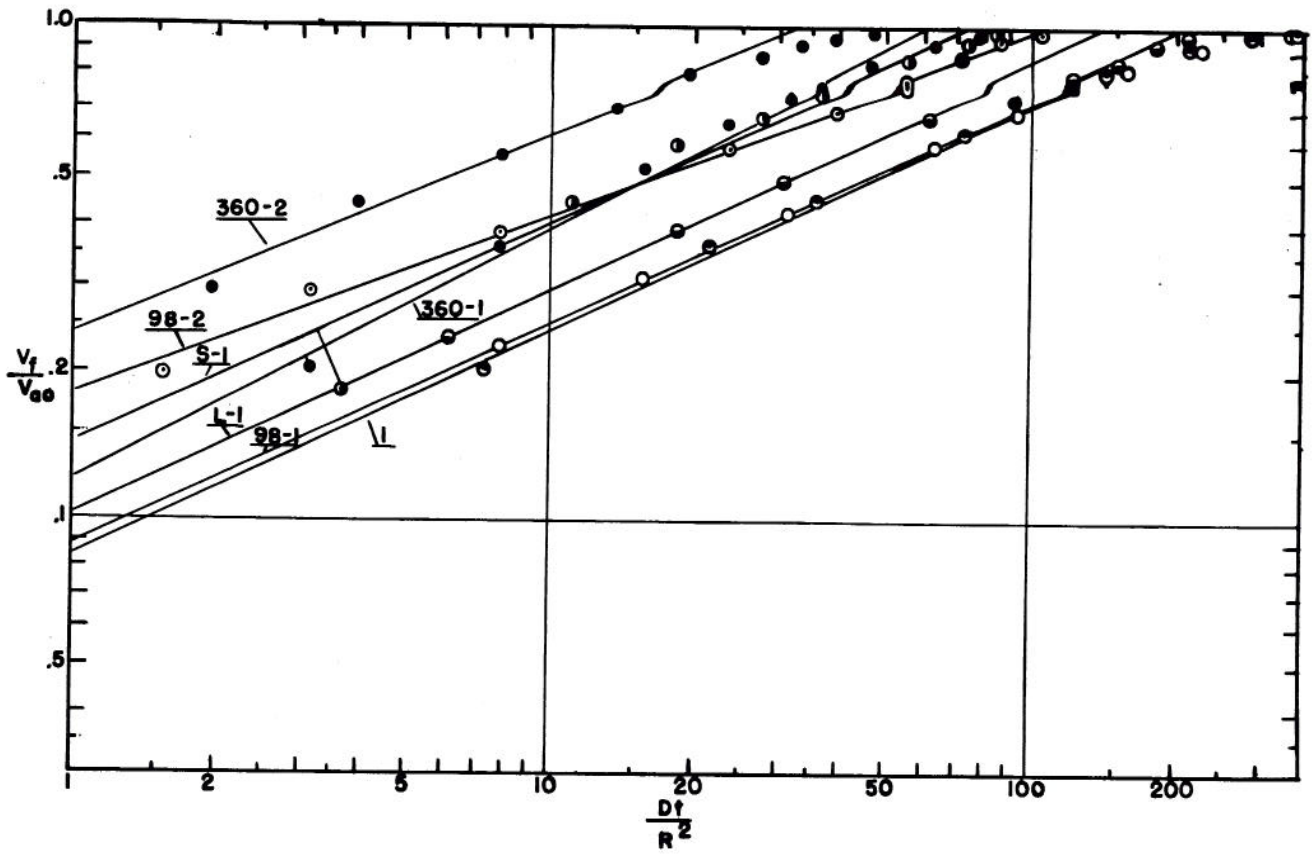


Figure 22. Logarithmic Plot of Dimensionless Variables Radial Diffusion - Ceramic and Glass Bead Cores

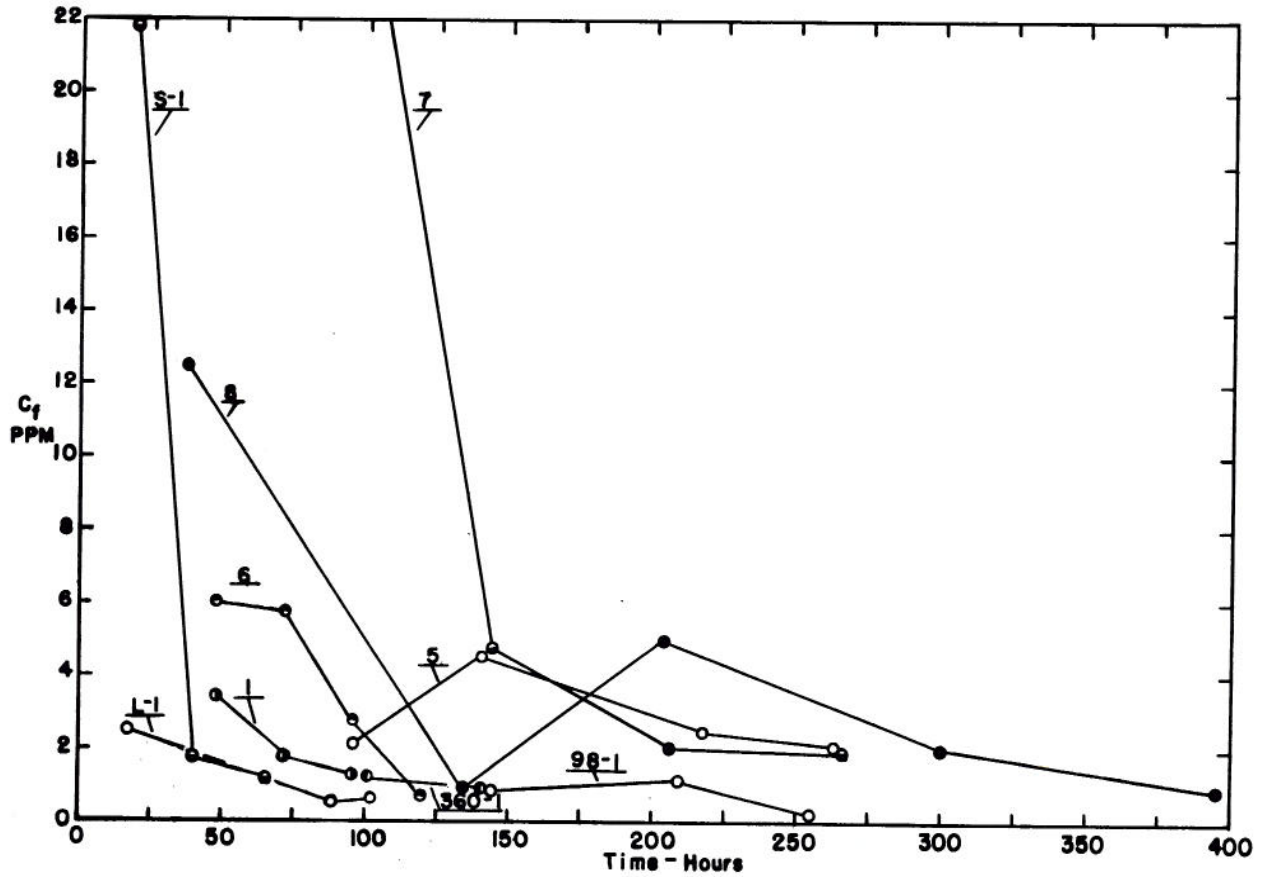


Figure 23. Concentration of Dissolved Air in Effluent From Cores

CONCLUSIONS AND RECOMMENDATIONS

The study presented in this paper should be regarded as a pilot study on the subject of diffusion of entrapped air from porous media. There are many aspects of the problem that need to be studied in more detail and under more rigorously controlled conditions.

Among the aspects that need to be studied more intensively are the effect of grain shape and smoothness on the amount of entrapped air and also the effect of wettability. Such studies will require experiments in which these variables are controlled and characterized more fully than was the case for this study.

An effort should be made to determine by direct measurement the distribution of undissolved air in porous media as a function of time. This is necessary to establish definitely the validity of the assumption made here that the locus of points at which air dissolves moves through media as a distinct front.

Direct measurements of diffusivity D , concentration, C_0 , and the constant of proportionality α would make possible completely analytical solutions for both linear and radial diffusion. It would then be possible to compare solutions directly with experimental results. Perhaps use of a pure

gas rather than a mixture of gases (air) would expedite the necessary measurements.

Despite the shortcomings of the study presented, the authors feel that the results justify the following interesting conclusions:

1. Porous media imbibing liquid at atmospheric pressure eventually become completely saturated even though there is no liquid flowing through the porous material.
2. Entrapped air moves out more rapidly from fine grained material than from coarse grained material, provided that dissolved air is not removed by flowing liquid.
3. When water is the liquid being imbibed in a porous medium, an important factor determining the amount of air initially entrapped is the surface condition of the solid particles. This causes the wettability to be variable and also the amount of entrapped air to be inconsistent.
4. Media which have more uniform pores, higher bubbling pressures, and consist of smoother particles tend to have less air entrapped.

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APPENDIX A

Determination of Diffusivity and Solubility of Air in Soltrol

No data were available on the diffusivity and solubility of air in soltrol; thus, it was necessary to obtain these data experimentally. The schematic diagram below shows the equipment used for this experiment.

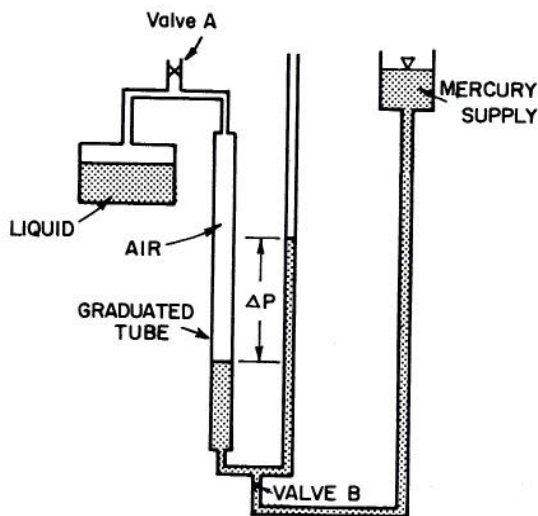


Figure 24. Schematic Diagram of Apparatus to Determine Solubility and Diffusivity of Air in Soltrol

The method of operation was as follows. The container was filled with liquid (water or Soltrol) which had been in contact with air at atmospheric pressure for some time. It was assumed that the concentration of dissolved air was uniform. The mercury in the vertical tubes was initially near the bottom of the tubes. Valve A was then closed and valve B opened slightly. As mercury flowed into the tubes, the air in the container was compressed and a pressure differential ΔP was developed. By noting the pressure differential, the immediate volume change and the atmospheric pressure, the initial quantity of air could be computed.

At any time later the total amount of undissolved air was computed from the volume change and total pressure. The amount which had gone into solution was obtained by subtraction.

In order to obtain the solubility of air in the oil, it was necessary to leave the apparatus at a given pressure differential until equilibrium was reached and no more air went into solution. Several values of solubility were obtained at different pressures and these were plotted in Figure 5. These measurements could be repeated to within 2%.

The coefficient of diffusion was more difficult to obtain. The theoretical relationship for one-dimensional diffusion of a solute into a solvent is given in Figure 25. For this relationship the concentration in the solvent was initially uniform and less than C_c . This was without flow of any type in the solvent.

With the equipment used it was impossible to eliminate all convective mixing even when the container of the liquid was protected from direct radiation.

The effect of convective mixing is shown for water in Figure 25 with the air going into solution much faster than the theory would indicate. This was not nearly so pronounced with the Soltrol, perhaps because the container was better protected from direct radiation. The value of $\frac{Dt}{L^2}$ for the points shown was calculated with an assumed D of 15×10^{-5} cm^2/sec . The points are from several different experiments using different sized containers for the oil.

The value for D of 15×10^{-5} cm^2/sec is probably a little small, but the correct value is certainly not larger than about 18×10^{-5} . If this value is used, some of the experimental points are to the right of the theoretical line. It is concluded, therefore, that the correct value of D is between 15×10^{-5} and 18×10^{-5} cm^2/sec , and a value of 16×10^{-5} cm^2/sec was used in all calculations in this study.

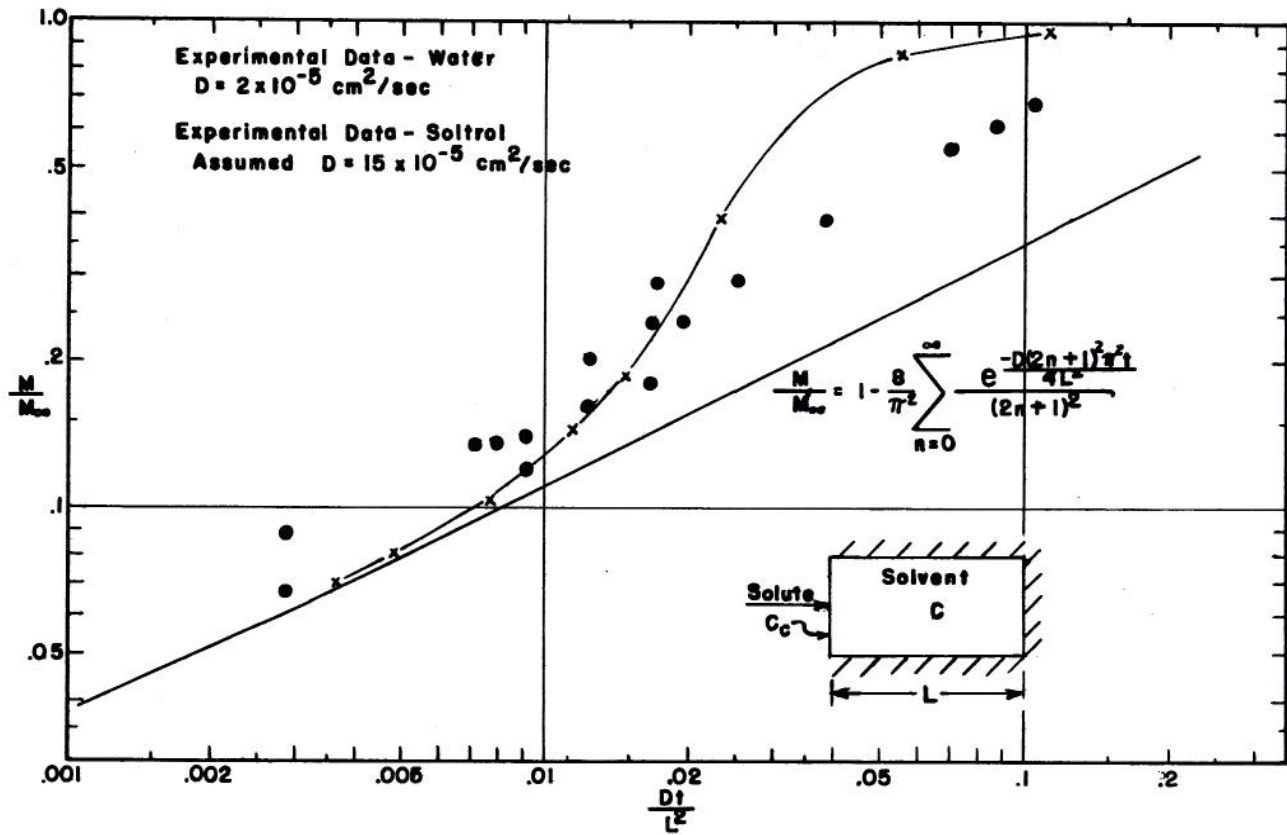


Figure 25. Theoretical Relationship For One-Dimensional Diffusion

Key Words: Theory, Diffusion, Entrapped Air, Saturation, Porous Media.

Abstract: The purpose of this study was to investigate the entrapment of air in porous media imbibing liquid and to determine both theoretically and experimentally the rate at which air goes into solution and diffuses from the system. The samples studied were cylindrical cores approximately one inch in diameter and of various lengths up to two and one half inches. Three materials were used, sandstone, a porous ceramic and sintered glass beads. Two liquids, water and a hydrocarbon oil, were used as

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the liquid imbibed. Theoretical equations for linear and radial (two dimensional) diffusion were developed. These are based on the assumptions that: (1) the concentration of dissolved air becomes nearly uniform after air is entrapped and (2) the point in the medium at which air dissolves moves from the exterior toward the interior as a front. The results show that a porous medium will eventually become completely saturated by imbibition even though no liquid is flowing through and the liquid imbibed is saturated with dissolved air at atmospheric pressure. The diffusion process by which the entrapped air moves out is most rapid for fine grained material. This is because the air is entrapped in pockets which maintain a higher concentration of dissolved air in the liquid due to the greater pressure difference across the air-liquid interface associated with smaller radii of curvature.

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