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## The Effect of Temperature on Water-Oil Imbibition Relative Permeability

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### ABSTRACT

Silica or limestone porous materials in contact with pure hydrocarbon liquids are strongly hydrophilic. When naturally occurring surface active materials are present in the liquid, they are adsorbed at the solid surface, rendering it less hydrophilic. If the concentration of such material is sufficient, the oil-water-matrix contact angle can be significantly greater than zero. The effect of increasing temperature in such a system will usually be to shift the adsorption equilibrium causing desorption of surface active material and a decrease in contact angle.

A simple geometrical model of a porous medium is used to approximate the effect of variation in wettability, as characterized by the oil-water-matrix contact angle, on imbibition relative permeability. Based on this model and other considerations, we conclude that the following trends will occur in a series of isothermal water floods with temperature increasing;

1. oil-water-matrix contact angle decreases to zero;
2. residual oil saturation decreases;
3. the ratio  $k_{rw}/k_{ro}$  at a fixed water saturation can increase for materials with a low residual oil saturation such as unconsolidated sands;

References and illustrations at end of paper.

4. the ratio  $k_{rw}/k_{ro}$  at a fixed water saturation decreases for materials with a high residual oil saturation such as consolidated sands.

Experimental observations of these conclusions are reported in the literature.

A method is presented for the correction of imbibition relative permeability curves to compensate for changes in temperature, based on the model developed.

### INTRODUCTION

There is considerable experimental evidence that oil-water imbibition relative permeability is strongly temperature dependent. Some doubt exists however as to the nature of this dependency. Poston et al<sup>1\*</sup> observed that at a given water saturation  $k_{rw}/k_{ro}$  increases with increasing temperature in an unconsolidated material. This same observation was made by Davidson<sup>2</sup>. Poston et al also found that as temperature increases, relative permeability to water increases, residual oil saturation decreases, and the oil-water-matrix contact angle, measured in the water phase, decreases.

Habowski<sup>3</sup>, using a consolidated porous medium, also found that residual oil saturation decreased with increasing temperature. He observed that in this type of material,  $k_{rw}/k_{ro}$  decreases with increasing temperature. This would appear to contradict the findings of Poston et al.

The only property of an immiscible oil-water-matrix system that can have a significant effect on relative permeability is wettability, characterized by the oil-water-matrix contact angle. It has previously been demonstrated<sup>4</sup> that relative permeability in a low permeability porous material under normal conditions is independent of other temperature dependent parameters such as viscosity ratio, density difference, and interfacial tension.

In what follows, we demonstrate that the observations of Poston et al and Habowski are in fact not contradictory. By considering a model for the variation of the oil-water-matrix contact angle with temperature, and the resulting effect on discontinuous oil saturation and water relative permeability, all the above observations are fully explicable.

#### A MODEL FOR THE EFFECT OF TEMPERATURE ON WETTABILITY

A force balance at a point where oil, water, and solid phases intersect results in the Dupre equation<sup>5</sup>

$$\sigma_{wo} \cos \theta_c = \sigma_{so} - \sigma_{sw} \quad (1)$$

where  $\sigma_{wo}$  is the water-oil interfacial tension,  $\sigma_{so}$  is the oil-matrix interfacial tension,  $\sigma_{sw}$  is the water-matrix interfacial tension, and  $\theta_c$  is the equilibrium contact angle measured in the water phase. For  $\sigma_{wo}$  less than  $\sigma_{so} - \sigma_{sw}$  spreading occurs and there is no intersection of the liquid-solid interfaces.

When the solid is a polar material such as sandstone or limestone, and the oil is a pure hydrocarbon, spreading of the water will occur. For example, with n-heptane and water on a SiO<sub>2</sub> surface at 25°C<sup>6</sup>,  $\sigma_{so} - \sigma_{sw}$  is 309 dynes/cm and  $\sigma_{wo}$  is 50 dynes/cm.

If the solid is somehow made less polar, it becomes less hydrophilic and  $\sigma_{so} - \sigma_{sw}$  will decrease. Denekas et al<sup>7</sup> have shown that when a crude oil contains certain surface active components, its wetting properties are significantly different from those of a pure hydrocarbon as a result of physical adsorption of these components onto the solid surface. They found that limestone was made oil wet ( $\sigma_{so} - \sigma_{sw} < 0$ ) by organic bases containing nitrogen. Sandstone wettability was affected by surface active materials containing nitrogen, sulfur, and oxygen.

Let us assume an oil-water-polar solid system. The oil phase contains a concentration, C, of surface active material capable of being adsorbed at active sites on the solid surface. The Langmuir isotherm may be used to approximate equilibrium during adsorption from solution when adsorption is limited to a monolayer<sup>5</sup>. The fraction of active sites,  $C(s)$ , retaining adsorbed material is

$$C(s) = \frac{bC}{1+bC} \quad (2)$$

where

$$b = b'e^{Q/RT} \quad (3)$$

Here  $b'$  is a constant, Q is the heat of adsorption, R is the gas constant, and T is the absolute temperature.

Healey et al<sup>8</sup> found that the heat of immersion of carbon black in water increased approximately linearly with the fraction of hydrophilic sites. McCaffery and Mungan<sup>13</sup> observed that water wet quartz crystals, when contaminated with certain polar-non-polar materials, gave non-zero oil-water contact angles. We will assume that the adsorption of a surface active molecule converts a strongly hydrophilic site to a less hydrophilic or hydrophobic site. Healey's observation may be generalized to the assumption that  $\sigma_{so} - \sigma_{sw}$  decreases linearly with increasing  $C(s)$ . We may then write

$$\cos \theta_c = \alpha - \beta C(s) \quad (4)$$

where  $\alpha$  and  $\beta$  are positive constants related to the variation of  $\sigma_{so} - \sigma_{sw}$  with  $C(s)$ . Again, a value of  $\alpha - \beta C(s)$  greater than one indicates that spreading of water occurs. A value of  $\alpha - \beta C(s)$  less than minus one indicates that spreading of oil occurs.

Equation (2) and (3) predict a strong dependence of adsorption equilibrium on temperature. Assuming C is constant and sufficiently high to permit the formation of a monolayer,  $C(s)$  will approach unity at low temperatures. The contact angle at this point will be at its maximum value or the surface may be completely oil wet if the surface can be rendered sufficiently hydrophobic. As temperature increases,  $C(s)$  decreases so that  $\theta_c$  also decreases. At some high temperature,  $C(s)$  will be such that  $\theta_c$  is zero. Above this temperature, the surface will be completely water wet.

There will also be some variation in interfacial tension with temperature. The net effect of this on  $\theta_c$ , however, is uncertain. It seems reasonable to assume that this will be small compared with the exponential dependency of the adsorption equilibrium.

Poston et al<sup>1</sup> have observed a variation of contact angle with temperature (Figure 5) similar to that described above.

#### IMBIBITION IN AN IDEALIZED POROUS MEDIUM

Consider a porous medium to be constructed of spheres in a cubic arrangement as shown in Figure 1. The radii of these spheres will be such that the porous medium possesses a characteristic cumulative pore size distribution

function,  $D(R)$  defined such that  $D=0$  at  $R=0$  and  $D=1$  at some maximum pore entry radius,  $R_{\max}$ . While spheres of different sizes are intermixed, we assume for computational purposes that each pore space is bounded by spheres of one size only, forming a unit cell as shown in Figure 2a. The characteristic pore radius,  $R$ , is proportional to the sphere radius,  $a$ .

We will characterize a given porous medium by the residual nonwetting phase saturation following displacement by a completely wetting phase such as the displacement of gas by oil. This value can range from 10-20% for uniform unconsolidated glass beads to about 40% for consolidated sandstones.

The porous medium is assumed to be at an initial water saturation,  $S_{wi}$ , arrived at by the displacement of water by oil. After being heated to a temperature,  $T$ , the oil is subjected to an isothermal displacement by water. The oil-water-matrix contact angle during this displacement measured in the water phase at the temperature  $T$  is  $\theta_c$ . We will first consider the case where the matrix is water wet ( $0^\circ < \theta_c < 90^\circ$ ).

#### THE FORMATION OF DISCONTINUOUS OIL IN THE UNIT CELL - WATER WET

Prior to penetration by water, the unit cell contains oil and irreducible water. The latter is located at the regions of contact of the spherical grains as shown in Figure 2a. As water penetrates, it links with the irreducible water. The resultant configuration (Figure 2b) of the oil-water interface is unstable and is subject to snap-off at the constricted region.

The volume of discontinuous oil formed in this way may or may not be retained in the unit cell. A large volume would not remain discontinuous as a result of interference with oil in adjacent cells. A small volume might itself be unstable or could flow undistorted out of the cell.

The shape of the discontinuous oil-water interface must be constrained so that the mean curvature is constant everywhere on it<sup>4</sup>. Furthermore, the angle of intersection of the interface with the matrix spheres must be  $\theta_c$ . We will assume that when oil is retained in a unit cell, the oil-water interface will be a sphere intersecting the matrix spheres at an angle  $\theta_c$  as shown in Figure 2c. This configuration satisfies the above requirements.

We will further assume that differences in  $S_{or}$  for different porous materials result primarily from the fact that different fractions of the total pore spaces retain oil. The frac-

tion of the pore space volume occupied by oil, when the pore space contains oil will be the same for any porous medium.

It can be shown that if  $a$  is the radius of a matrix sphere in the unit cell, the radius of a retained oil drop is

$$r_o = a(\sqrt{\cos^2 \theta_c + 2} - \cos \theta_c) \quad (5)$$

The fraction of the unit cell pore volume occupied by oil is

$$f_o = \frac{[r_o^3 - 2(2r_o^3 - 3Ar_o^2 + A^3) - 2(2a^3 - 3Ba^2 + B^3)]}{a^3 \left[ \frac{6}{\pi} - 1 \right]} \quad (6)$$

where

$$A = \frac{\sqrt{3}(2a^2 + r_o^2)}{6a} \quad (7)$$

and

$$B = \frac{\sqrt{3}(4a^2 - r_o^2)}{6a} \quad (8)$$

As discussed above, the residual oil saturation is proportional to  $f_o$ .

$$\frac{S_{or}}{S_{or}(\theta_c=0)} = \frac{f_o}{f_o(\theta_c=0)} \quad (9)$$

A plot of  $S_{or}/S_{or}(\theta_c=0)$  is given in Figure 3.

#### ADMISSIBILITY TO FLOW OF WATER IN THE UNIT CELL

We will define a mean hydraulic radius for the unit cell as the ratio of pore volume to surface area<sup>9</sup>. For a single phase occupying the entire pore volume, this is

$$m = \left( \frac{2}{\pi} - \frac{1}{3} \right) a \quad (10)$$

With the retained oil in place, we make the assumption that the oil-water interface retards the flow of water in the same way as does the matrix. We have then for the flow of water,

$$m_w = \frac{(1-f_o) \left( \frac{2}{\pi} - \frac{1}{3} \right) a^3}{r_o^2 + a^2 - 4r_o(r_o - A) - 4a(a - B)} \quad (11)$$

The relative admissibility to flow of water with the trapped oil drop in place is

$$A_{rw} = \left( \frac{m_w}{m} \right)^2 = \left[ \frac{(1-f_o)a^2}{r_o^2 + a^2 - 4r_o(r_o - A) - 4a(a - B)} \right]^2 \quad (12)$$

The relative permeability to water will be proportional to  $A_{rw}$  for small values of  $A_{rw}$  since the flow will be controlled by pores containing trapped oil. We then have

$$\frac{k_{rw}}{k_{rw}(\theta_c=0)} = \frac{A_{rw}}{A_{rw}(\theta_c=0)} \quad (13)$$

A plot of  $k_{rw}/k_{rw}(\theta_c=0)$  vs.  $\theta_c$  is given in Figure 4.

THE DEPENDENCE OF  $k_{rw}/k_{ro}$  ON TEMPERATURE

As demonstrated above, an increase in  $\theta_c$  will increase the trapped oil volume and will decrease the admissibility to flow of water in the unit cell. These effects combine to determine the dependence of  $k_{rw}/k_{ro}$  on  $\theta_c$ .

Let us assume that imbibition has proceeded to the point where all pore spaces for which  $R$  is less than some  $\hat{R}$  have been penetrated by water. Those pores with  $R < \hat{R}$  can contain either water and trapped oil or water only as discussed in the previous section. Pores with  $R > \hat{R}$  still contain the initial water saturation and continuous oil. The total water saturation at this point is

$$S_w = (1-S_{or})D(\hat{R}) + S_{wi}[1-D(\hat{R})] \quad (14)$$

This may be put in the form

$$S_w^* = [1-S_{or}^*]D(\hat{R}) \quad (15)$$

where

$$S_w^* = \frac{S_w - S_{wi}}{1-S_{wi}} \quad (16)$$

and

$$S_{or}^* = \frac{S_{or}}{1-S_{wi}} \quad (17)$$

We may eliminate  $D(\hat{R})$  by forming the ratio

$$\frac{S_w^*}{S_w^*(\theta_c=0)} = \frac{[1-S_{or}^*]}{[1-S_{or}^*(\theta_c=0)]} \quad (18)$$

The relative permeability to oil at a fixed  $\hat{R}$  will not depend on  $\theta_c$ , so that

$$\frac{k_{rw}/k_{ro}}{k_{rw}/k_{ro}(\theta_c=0)} = \frac{k_{rw}}{k_{rw}(\theta_c=0)} \quad (19)$$

Poston et al measured the variation of  $\theta_c$  with temperature for three different oils. Their results are reproduced in Figure 5. Any crude oil-water-matrix system will have its own characteristic  $\theta_c$ -T relationship which may or may not resemble Figure 5. This relationship, together with equations 14 and 15 form the basis for a method of estimating  $k_{rw}/k_{ro}$  curves for imbibition at any temperature.

A PROCEDURE FOR CORRECTING IMBIBITION  $k_{rw}/k_{ro}$  CURVES FOR USE AT ELEVATED TEMPERATURES

The following procedure is suggested for estimating imbibition relative permeability ratios at an elevated temperatures, T.

1. Determine oil-water imbibition relative permeabilities at ambient temperature together with the oil-water-matrix contact angle\* using the crude oil, water and core in question. Alternatively, determine imbibition relative permeabilities in the core using gas and oil and assume that  $\theta_c=0$  for this system.

2. Obtain the oil-water-matrix contact angle  $\theta_c$  at the temperature T on the core material (quartz for sandstone, calcite for limestone).

3. Determine  $S_{or}$  from Figure 3 and  $k_{rw}/k_{ro}$  ( $\theta_c=0$ ) from Figure 4 for the contact angle,  $\theta_c$ .

4. Choose a point ( $k_{rw}/k_{ro}, S_w$ ) on the curve obtained in step 1. Obtain a corresponding point on the elevated temperature curve by obtaining a new  $S_w$  from equation 18 and a new  $k_{rw}/k_{ro}$  from equation 19, noting that the ratios apply for any two contact angles.

5. Repeat step 4 until the new curve is defined.

This procedure was used to obtain  $k_{rw}/k_{ro}$  curves at different temperatures for two materials. Figure 6 shows the results for an unconsolidated porous medium with  $S_{or} = 15\%$  at  $\theta_c = 0$  and  $S_{wi} = 10\%$ . The curve for  $T=220^\circ\text{F}$  ( $\theta_c=0$ ) is typical of those obtained for uniform unconsolidated glass beads (see for example Naar et al<sup>12</sup>). The  $\theta_c$ -T relationship was obtained from Figure 5.

Figure 7 shows the results of a similar calculation for a consolidated material with  $S_{wi} = 20\%$  and  $S_{or} = 40\%$ . Again, the  $T=220^\circ\text{F}$  curve is typical for consolidated sandstone<sup>12</sup>.

THE OIL WET CASE

As shown in the previous section, it is possible for a normally water wet matrix to become oil wet ( $180^\circ > \theta_c > 90^\circ$ ) as a result of the adsorption of certain surface active materials. Relative permeabilities in this case are similar to those for drainage of a water wet medium (with oil as the wetting phase). While the true residual oil saturation is low, it cannot be achieved in practice since  $k_{rw}/k_{ro}$  becomes large at fairly low water saturations. For

\* A discussion of the various methods available for contact angle measurement is given by Adamson<sup>4</sup>, pp. 268-72. The sessile drop method<sup>10,11</sup> (used by Poston et al to obtain the data shown in Figure 5) is probably most suitable at non-ambient temperatures.

values of  $S_w$  that are of interest, there is little discontinuous trapped oil so that  $k_{rw}/k_{ro}$  is insensitive to contact angle variation and hence to temperature variation while in the oil wet condition.

As discussed above, as temperature is increased beyond a certain point, a transition from an oil wet to a water wet condition will occur. From this point on, there will be significant dependence of  $k_{rw}/k_{ro}$  on temperature.

#### DISCUSSION OF RESULTS

The structure of real porous materials is far too complex for a simple geometrical analysis such as this to provide anything but an approximation of the effect of temperature on imbibition relative permeability. Depending on water saturation, the  $k_{rw}/k_{ro}$  ratio can span several orders of magnitude with temperature changes that are of practical interest. In this case, such an approximation can be of value.

In summary, if the oil-water-matrix contact angle decreases to zero as temperature increases as predicted by the adsorption model, the following are results of the above analysis.

At a fixed penetration radius,  $\hat{R}$ , trapped oil saturation decreases and relative permeability to water increases with increasing temperature. Relative permeability to oil remains constant. If we refer now to a fixed water saturation, relative permeability to oil increases and relative permeability to water may either increase or decrease with increasing temperature, depending on the magnitude of  $S_{Or}(\theta_c = 0)$  for the material in question.

Figure 6 shows a situation in which the increase in  $S_w$  at a fixed  $\hat{R}$  is relatively small since the residual oil saturation is small. The shift in the  $k_{rw}/k_{ro}$  curves is due, for the most part, to the increase in  $k_{rw}$  with decreasing  $\theta_c$  given by Figure 4. Figure 7 shows the case where  $S_{Or}(\theta_c = 0)$  is high. An increase in temperature here causes a large increase in  $S_w$  at a fixed  $\hat{R}$ . The net result of this is that at a fixed  $S_w$ , the number of pores permeable to water is decreased significantly. The resulting decrease in  $k_{rw}$  cancels the increase shown in Figure 4, causing the behavior shown in Figure 7.

Thus  $k_{rw}/k_{ro}$  at a given water saturation increases with increasing temperature for low residual oil porous media and decreases with increasing temperature for high residual oil materials. This follows the experimental observations of Poston et al<sup>1</sup> and Davidson<sup>2</sup> in unconsolidated sands and of Habowski<sup>3</sup> in consolidated sands.

The simple adsorption model does not allow effects such as pyrolysis in the oil that may create new surface active species or crystallographic rearrangement in the solid that may affect the adsorption equilibrium. Either of these may cause deviation from the trends predicted.

In this work, we did not consider the effect of wettability changes on the irreducible wetting phase saturation following drainage. The more interesting case, in terms of applications, is that in which the initial wetting phase saturation is established before the temperature is increased. The results given here should be applicable to displacement during hot water and steamflooding, and in a water bank during forward combustion.

#### CONCLUSIONS

1. Variation in oil-water imbibition relative permeabilities with temperature in low permeability porous media under normal conditions can result from changes in wettability with temperature.
2. A method is developed for approximating the effect of temperature variation on imbibition  $k_{rw}/k_{ro}$  vs saturation curves for any porous medium.
3. In a series of isothermal water floods with temperature increasing, the following trends will be observed for a fixed water saturation
  - (a) oil-water-matrix contact angle decreases to zero.
  - (b) trapped oil saturation decreases.
  - (c) the ratio  $k_{rw}/k_{ro}$  can increase for materials with a low residual oil saturation, such as unconsolidated sands.
  - (d) the ratio  $k_{rw}/k_{ro}$  decreases for materials having a high residual oil saturation, such as consolidated sands.
4. As temperature increases beyond a certain temperature corresponding to complete water wetting, there will be no further effect on the displacement.

#### NOMENCLATURE

- |          |  |
|----------|--|
| a        | radius of a matrix sphere in the unit cell |
| A        | defined by equation 7                      |
| $A_{rw}$ | relative admissibility to flow of water    |

b, b'	constants in Langmuir isotherm equation
B	defined by equation 8
C	bulk concentration of surface active material
C <sub>(s)</sub>	surface concentration of surface active material
D(R)	cumulative pore size distribution
f <sub>o</sub>	fraction of cell pore volume occupied by trapped oil
k <sub>ro</sub>	relative permeability to oil
k <sub>rw</sub>	relative permeability to water
m	mean hydraulic radius for flow of water with no oil trapped
m <sub>w</sub>	mean hydraulic radius for flow of water with oil trapped
Q	heat of adsorption
r <sub>o</sub>	radius of trapped oil droplet
R	pore entry radius or gas constant
S <sub>or</sub>	residual oil saturation
S <sub>or</sub> *	reduced residual oil saturation defined by equation 17
S <sub>w</sub>	water saturation
S <sub>w</sub> *	reduced water saturation defined by equation 16
S <sub>wi</sub>	initial water saturation
T	temperature
α, β	positive constants in equation 4
θ <sub>c</sub>	oil-water-matrix contact angle

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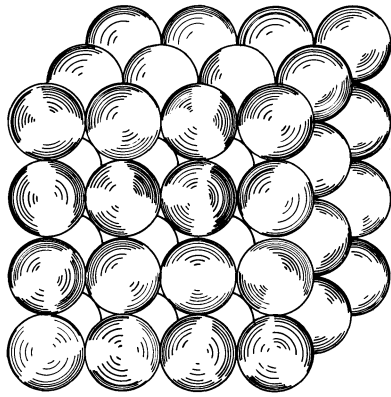


FIGURE 1 ARRANGEMENT OF SPHERES IN AN IDEALIZED POROUS MEDIUM

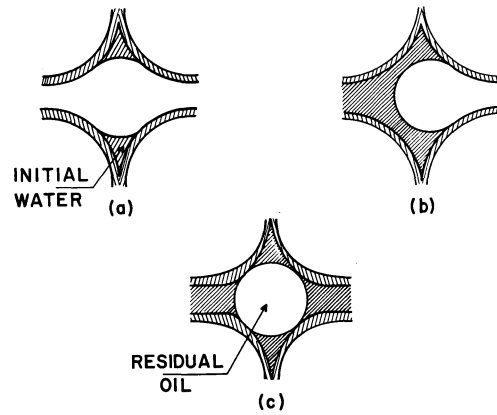


FIGURE 2 PENETRATION OF THE UNIT CELL BY WATER

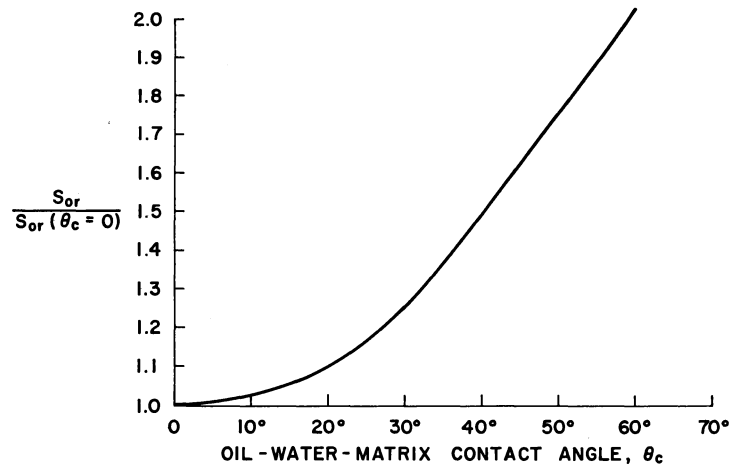


FIGURE 3 DEPENDENCE OF RESIDUAL OIL SATURATION ON CONTACT ANGLE

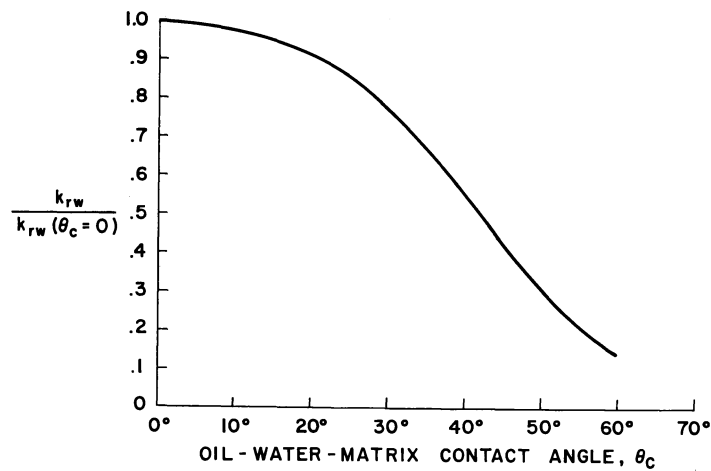


FIGURE 4 DEPENDENCE OF WATER RELATIVE PERMEABILITY ON CONTACT ANGLE

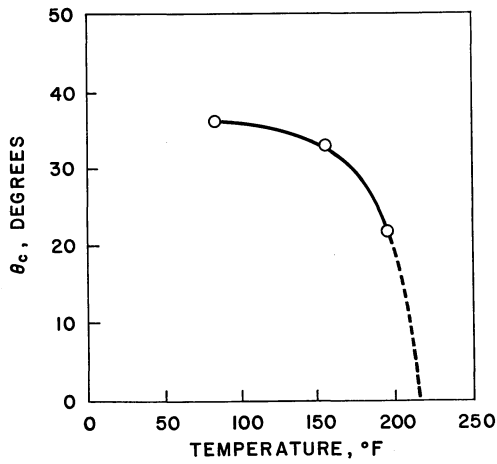


FIGURE 5 CONTACT ANGLE VS. TEMPERATURE FOR AN OIL-WATER-GLASS SYSTEM, AVERAGE FOR THREE OILS AFTER POSTON ET AL<sup>1</sup>

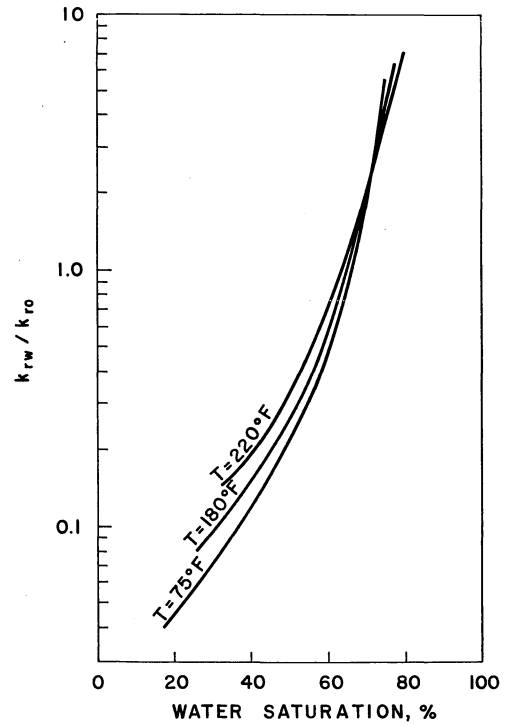


FIGURE 6 RELATIVE PERMEABILITY RATIO VS WATER SATURATION - UNCONSOLIDATED MATERIAL WITH  $S_{or}(\theta_c = 0) = 15\%$ ,  $S_{wi} = 10\%$

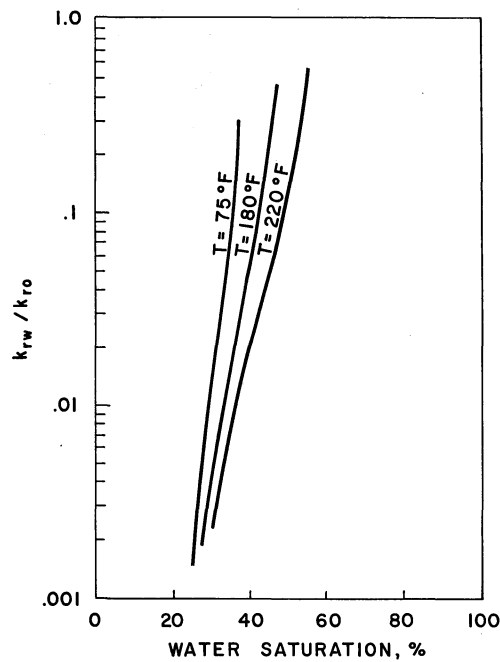


FIGURE 7 RELATIVE PERMEABILITY RATIO VS. WATER SATURATION - CONSOLIDATED MATERIAL WITH  $S_{or}(\theta_c = 0) = 40\%$ ,  $S_{wi} = 20\%$