Effects of Clays and Clay Minerals on Relative Permeability Curves*

By

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Abstract: It is the purpose of this paper to present the results of experimental studies and discuss about the effects of clays and clay minerals upon relative permeability curves. The relative permeability measurements were carried out on artificial and natural samples. And then, Size Analysis, Differential Thermal Analysis and Spectral Analysis were conducted on an oil sand core in order to examine the clays and clay minerals.

1. Introduction

A number of experimental relative permeability studies were reviewed by this writer in a previous publication, and it was reported that these studies in Oil Reservoir Engineering have made great progress. However, it seems that there remain some questions to be solved in such studies. One of them in two-phase system is ‘Effects of Clays and Clay Minerals on Relative Permeability Curves.’ In particular, most reservoir rocks in Japan are rich in clay minerals. Although the relationship between relative or effective permeability and clay minerals contained in the core is very important, the writer knows of no paper on such subjects, except for a paper reported by Nowak and Krueger. Nowak and Krueger have indicated the effects of aqueous solutions and field mud filtrates upon the effective oil permeabilities of water-sensitive sands from the standpoint of drilling fluid studies. So, it is the purpose of this paper to present the results of experimental studies and discuss about the effects of clays and clay minerals upon relative permeability curves.

2. Experiment

The general technique being used to measure relative permeability in this investigation is very much the same as was reported by this writer in a previous study. This method is similar to the Penn State method and Single Core Dynamic method, except that a mixer is used in order to supply the uniform two liquid mixture to the core. And then, it goes without saying that the influence of the boundary effect has been avoided in this technique.

Throughout all these experiments, a two-phase system, kerosene (oil phase) and aqueous phase, was used. The artificial brine contained on 20,000 and 35,000 p.p.m. by weight of sodium chloride, and kerosene viscosity was approximately 1.7 centipose at 15.0°C (temperature).
These experiments were carried out on artificial and natural samples, that is, (1) cement cores containing bentonite (clay minerals) (2) synthetic cores containing known varying proportions of bentonite to clean sand (3) oil sand cores obtained from an oil field in Japan. Of these samples, oil sand cores were extracted with solvents to insure complete removal of any oily constituents.

Prior to starting any relative permeability tests, the core sample was first fully saturated with brine, and the absolute brine permeability of the core was measured at 100% brine saturation. And then, brine-kerosene mixture was introduced into the core, and was allowed to flow through it. When this flow approached to steady state, two-phase relative permeabilities on a small core sample were accurately determined at a brine saturation. Such several runs were made for the determination of each relative permeability curve.

Moreover, Size Analysis was conducted on an oil sand core in order to determine accurately the quantity of clay in the core and to define grain-size fractions of clastic sediment. To begin with, crushed core (about 30 grams) was analysed by means of Tyler Sieves, and fine-grained clastics (particles less than 0.06 mm in dia.) were graded by Pipette techniques. In this investigation, the Wentworth size classification was adopted for defining grain-size fractions of the oil sand core.

Next, by means of Differential Thermal Analysis, the temperature at which thermal reactions take place in an oil sand core, and also the intensity and character of such reactions were determined in order to know the existence and kind of clay minerals in the core. And then, Spectral Analysis contributed valuable information to research on chemical elements of clays and clay minerals. This analysis was made by the staff of Kawai’s Laboratory (our University) using the spectral analysis apparatus (JACO 3.4 m EBERT type).

3. Results and Discussion

Experiment (1) Effect of brine displacement by oil phase.

Fig. 1 is the plot showing relative permeabilities by using a cement core containing bentonite. The absolute permeability to brine in excess of 1.0 (100%). (Also, this phenomenon was reported by other investigator and co-workers). The curious shape of the oil-brine relative permeability relationships can be explained by using the following concepts. The writer thinks that the swelling effect of clay minerals (bentonite) in the core is different in the presence of oil and brine than when in brine alone, and causes this phenomenon. (In addition, all effective permeabilities in this ex-

Fig. 1. Oil-Brine Relative Permeability on a Cement Core containing Bentonite.
(Absolute Permeability to Brine: 610 md.)
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Experiment were compared to the absolute permeability to brine of the 610 md.). So, the swelling test shown in Fig. 2 was conducted on only bentonite samples in order to examine this effect.

**Fig. 2.** Results of the Swelling Test.

Experiment (2) Effect of variation in aqueous phase.

Comparisons of the results on the different systems (that is, the fresh water-kerosene system and brine-kerosene system) may be seen in Figs. 3 and 4. The term "1st curve" designates the data obtained on the brine-kerosene system. And, the tests on the fresh water-kerosene system gave the behavior graphed as the "2nd curve."

First explain the curves of Fig. 3 on the synthetic cores containing known proportion of bentonite to sand. After the brine displacement by fresh water, a lot of bentonite in the core were washed away during the test on the fresh water-kerosene flow. (Accordingly, this run was a rapid and inaccurate measurement of determining the data). This phenomenon also may be observed in loose oil sand reservoirs where the surface water is used as injection water in the water flooding operations. If so, on account of this phenomenon occurs the "Sand Trouble" in both producing wells and injection ones.

**Fig. 3.** Relative Permeability Curves on the Synthetic Cores

- **1st Curve (Brine-Oil System):**
  - Absolute Brine Permeability: 15. darcy
  - Absolute Water Permeability: 7.4 darcy

- **2nd Curve (Fresh Water-Oil System):**
  - Absolute Brine Permeability: 14. darcy
  - Absolute Water Permeability: 7.4 darcy

Next, Fig. 4, in which the point number represents the order of measurements, shows the curves determined on a cement core (containing bentonite) of 35 md. permeability (absolute perm. to brine). The data graphed were taken on one core during two flow test. And, the "2nd curve" data obtained 15 days after brine displacement by the fresh water. It seems that the difference between point 2 and point 8 shows the existence of hysteresis in relative permeability measurement on the core containing clay minerals, although
the writer has reported\(^8\) that there is no significant variation of the curves with hysteresis test on unconsolidated clean sand samples. So, it is thought that this hysteresis is certainly due to the presence of clay minerals.

Fig. 4. Effective Permeability on a Cement Core containing Bentonite.
(Absolute Permeability to Brine of the 35 md).

![Graph of Effective Permeability](image)

1st Curve: Brine-Oil System
2nd Curve: Fresh Water-Oil System

Fig. 3 and 4 demonstrate the swelling effect of clay minerals in the cores. The main feature to be noted from the "2nd curve" is the increase in the irreducible aqueous phase saturation and the decreased effective permeability. That is, measurement on the fresh water-kerosene system gave effective permeabilities to kerosene and fresh water which were lower than the results obtained on the brine-kerosene system.

Experiment (3) Effect of variations in the contents of clays and clay minerals.

Fig. 5 shows how the relative permeability curves change for variations in clay mineral contents. Natural cores contain a lot of constituents such as quartz, feldspar, mica, clay minerals and cementing materials, and unknown varying proportions of each other in natural cores make matter worse. Therefore, in order to keep above variable under control, synthetic cores containing only unconsolidated sand and bentonite (known weight percent) were used in this tests.

Fig. 5. Relative Permeabilities on Synthetic Cores.

![Graph of Relative Permeabilities](image)

<table>
<thead>
<tr>
<th></th>
<th>Sand (mesh)</th>
<th>Bentonite</th>
<th>Absolute Brine Perm. (darce)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Curves 1</td>
<td>-60~+100</td>
<td>0</td>
<td>34</td>
</tr>
<tr>
<td>Curves 2</td>
<td>-60~+100</td>
<td>1% (wt.)</td>
<td>26</td>
</tr>
<tr>
<td>Curves 3</td>
<td>-60~+100</td>
<td>3% (wt.)</td>
<td>15</td>
</tr>
<tr>
<td>Curves 4</td>
<td>-60~+100</td>
<td>5% (wt.)</td>
<td>8.1</td>
</tr>
</tbody>
</table>

Two points shown in Fig. 5 immediately attract attention. (a) Although the curves 1 obtained on unconsolidated clean sands agree very well with similar results reported by Leverett\(^9\) in experiments with long unconsolidated sand columns, the curves 2 on the core containing a bit of bentonite (1% wt.) deviate markedly from the smooth curves showed for clean sands. (b) As the weight percentage of bentonite in the core becomes higher, relative permeability curves are shifted to higher brine saturation.

Therefore in the study of dirty sands, additional variables are involved, but for clean sand the new variable to be introduced is the contents of clay minerals. (As "Dirty Sand" is the same meaning as "Muddy
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Sand” which is geological term, and might be interpreted as the sand containing clay minerals, the term “Dirtiness” used in this paper may be construed to mean the degree of containing clay minerals. This variable, namely “Dirtiness” is very unfavorable to ultimate oil recovery, that is, the irreducible brine saturation in the cores of low clay mineral content will in general be less than that in the cores of high content. Of course, this point would vary greatly with the properties of clay minerals in the core and the chemical elements of the core.

Also, it is imagined from the results shown in Fig. 5 and the previous report3) that the core containing clays would give relative permeability curves having the same tendency as the curves showed for the core containing clay minerals. (*The writer reported that the brine permeability dropped more rapidly with decreasing brine saturation in the sand samples having wide distribution of grain size than having narrow one).

Experiment (4) Relative permeability curves on a natural core and the results of clays and clay minerals tests.

The consolidated oil sand cores obtained from an oil field in Japan are used in this investigation, and the data for one of these cores is shown in Fig. 6. However, the curves in Fig 6 are not necessarily representative in the field behavior of relative permeability, for an oil reservoir is mostly a heterogeneous formation. The relative permeability curves in Fig. 6 deviate from those for unconsolidated clean sand core. Probably, this deviation is chiefly dependent upon the wide variation of grain size and the “Dirtiness” of reservoir rock, as this reason is known from the following experiments.

Fig. 7 is illustrations of Size Analysis data — the histogram and the cumulative frequency curves — on the same core as used in the relative permeability measurement (Fig. 6). The data means that the median diameter of this core is 0.215 mm and its geometric coefficient of sorting is 1.49. And, it is determined that this sediment (core) is well sorted, and containing clay percent of the core is about 1.4% (by weight). Moreover, according to Shepard,10) the sediment type of this core becomes “Sand”. It is obvious that this core has a wide range of grain size and contains a bit of clay, although it does not necessarily follow that clay minerals are contained in this core. However, it is sure that clay mineral exists in this core, as known from the results of the following clay mineral tests and absolute permeability measurements (in which air, water and brine absolute permeabilities of the core are different).

Fig. 8 shows a Differential Thermal Curve for clay and silt contained in the core. The behavior of this curve will predict in the existence of montmorillonite (clay mineral) and limonite in this core. And then, it seems likely that the exothermic reaction between about 200°C and 800°C relates to oil contained. Next, a photograph of the arc spectrum is also shown in Fig. 9. From the result of this spectral analysis, the following chemical elements were obtained.
Fig. 7. Cumulative-Frequency Curve and Histogram of a Natural Core.

Fig. 8. Differential Thermal Curve.

Fig. 9. A Photograph of the Arc Spectrum.

Major Component: Si, Al, Fe, Ca, Mg
Minor Component: Na, Mn, Ba, Cu, Ti
Trace Component: K, Zn, Cr, Ca, Sr, Zr
Uncertain Component: Sn, Co, P, B

As the general formula\(^{11}\) of montmorillonite group is

\[(X,Y)_{2-3}Z_2O_{10}(OH)_2\cdot mH_2O\cdot (W_w)\]

where \(X\): Al, Fe\(^{'''}\), Mn\(^{'''}\), Cr\(^{'''}\)
\(Y\): Mg, Fe\(^{'''}\), Mn\(^{'''}\), Ni, Zn, Li
\(Z\): Si, Al
\(W\): K, Na, Ca

the writer imagines the formula \((Al,Mg)_{2-3}\ Si_2O_{10}(OH)_2\cdot mH_2O\cdot Ca_w\) for the montmorillonite in this core.

4. Conclusion

The relative permeability curves on the core containing clays and clay minerals deviate markedly from the smooth curves showed for the unconsolidated clean sand samples having uniform grain size. Although this deviation would vary greatly with the properties of clay minerals in the core and the chemical elements of the core, the deviation is also dependent upon the amount of clays and clay minerals contained in the core. Moreover, relative permeability characteri-
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stics of the core containing clay minerals are influenced by the swelling effect of clay minerals, and the swelling phenomenon occurs in the displacement of oil by brine and that of brine by fresh water.

In the majority of oil reservoir cores in Japan there exist clays and clay minerals. Accordingly, investigations on the relative permeability must not be indifferent to clays and clay minerals. So, in studying the relative permeability curves on natural cores, it is not only favorable but also necessary that the results of clays and clay minerals tests be provided.

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