FORMATION DAMAGE IN SANDSTONES CAUSED BY CLAY DISPERSION AND MIGRATION

by

D. H. GRAY* and R. W. REX

Chevron Research Company, La Habra, California

ABSTRACT

X-RAY diffraction and electron microscopy were employed in conjunction with core flooding experiments to investigate clay migration phenomena.

Severe water sensitivity or loss of permeability was observed in a suite of sandstones in spite of the almost total absence of montmorillonite or swelling mixed layer clays. Clay migration was found to cause total or partial plugging even in sandstones of 500 millidarcy permeability. Bacterial plugging was ruled out by prefiltering and bactericide treatments of waters.

X-ray diffraction and electron microscopy analyses were performed on the sandstones and produced effluents. The direct cause of damage was displacement of submicroscopic natural clay crystals of needle-shaped mica and hexagonal-shaped kaolinite (Rex, 1965). The mobile clays were identified as authigenic crystals that are present on the pore walls and are dislodged by changes in water chemistry combined with water movement.

Flooding sandstones with alkali metal brines "sensitized" the cores, i.e. triggered clay dispersion upon subsequent flooding with fresh water. Flooding with divalent calcium brine prevented water sensitivity and suppressed the undesirable effect of alkali metal brines. A double layer expansion effect is suggested as the dispersion mechanism.

INTRODUCTION

WATER sensitivity or loss of permeability of sandstones in response to water salinity changes is a well documented, but still incompletely understood phenomena. Johnston and Beeson (1945) were among the first to investigate and report large decreases in permeability of clay-containing sandstones with decreasing salinity of the pore water. Von Engelhardt and Tunn (1954), two research workers in Germany, reported similar observations. The latter extended their work to include the effects of polar and nonpolar solvents under varying pressure gradients; they suggested that anomalous water structure was responsible for permeability deviations in clayey sandstones.

A number of studies have subsequently been carried out to pinpoint the cause and nature of permeability reduction in clay-containing sandstones. Moore (1960) gives a comprehensive summary and introduction to the literature on the subject. The concensus is that water sensitivity or loss of permeability of sandstones can be attributed either to clay swelling in the rock pores, clay particle migration, or a combination of these effects.

* Present address: Dept. of Soil Mechanics, Univ. of California, Berkeley, Calif.

355

Water sensitivity attributable to clay swelling is probably the best documented damage mechanism. Studies by Foster (1955), Dodd *et al.* (1955), Monaghan *et al.* (1959), and White *et al.* (1962) have been made correlating water sensitivity directly to the presence of swelling clays in a sandstone. The degree of damage is believed to be a function of the amount of swelling montmorillonite and mixed layer clay present. Formation damage triggered by swelling of montmorillonite is now a widely recognized phenomenon by the oil industry, and is routinely considered in evaluating "problem" wells by some oil companies.

Water sensitivity attributable to clay particle migration, on the other hand, is not so well understood. There is no obvious correlation between pore water salinity and clay migration as there is between clay swelling and salinity. As a result there has been a tendency to discount the possibility of severe permeability reduction in sandstones which contain negligible amounts of swelling of mixed layer clays. Hewitt (1963) has recognized the possibility of damage from migrating clays and has presented a scheme of analysis for recognizing sandstones which are susceptible to this type of permeability damage.

Clay migration may be, in fact, a more prevalent formation damage mechanism than hitherto suspected. Field evidence and laboratory data are presented in this report to show that clay migration can cause severe permeability reduction even in so-called "clean" sandstones, i.e. those containing only traces of swelling or mixed layer clays. This type of damage can drop a very permeable sandstone to less than 1% of its original permeability in a few hours or less of flow. Significantly, the principal culprits in clay migration damage have been identified by X-ray diffraction and electron microscopy as nonswelling, needle-shaped mica crystals with a partial mixed layer structure and tiny hexagonal kaolinite crystals (Rex, 1965). These needles and hexagons are dislodged from the pore walls in sandstones and are trapped in pore throats where they build up an internal filter cake within the sandstone. Furthermore, our data show that this clay migration can be induced or inhibited by certain combinations of brine and fresh water treatment.

EXPERIMENTAL PROGRAM

Materials

A preliminary study was made on a series of sandstones from six different localities. Five core suites were oil reservoir rocks from Canada, the U.S.A., and Colombia. The sixth, Berea sandstone, is a building stone quarried in Ohio. The five oilfield suites (Mirador, Paluxy, Granite Wash, and Belly River sandstones) (Table 1) were all originally suspected to be water sensitive based on field performance of wells in these formations. Analytical work showed that these cores were indeed water sensitive, but they did not appear to be damaged by bulk swelling of montmorillonite or mixed layer clays. TABLE 1.-OCCURRENCES OF SANDSTONES STUDIED

				Clay minerals	rais		
Formation	Age	Location	Kaolinite	Illite	Montmorillonite % by weight	Mica needles	Water sensitivity
Navajo	Jurassic	Zion Canyon, Utah	Abundant	Minor	$0.1\pm0.1\%$	Abundant	N.D.*
Wingate	Jurassic	Grand Junction, Colo.	Abundant	Minor	$0.1\pm0.1\%$	Abundant	Severe
Entrada	Jurassic	Grand Junction, Colo.	Abundant	Minor	$0.1\pm0.1\%$	Abundant	Severe
St. Peter	Ordovician	Klondike, Mo.	Abundant	Minor	$0.0\pm0.1\%$	Abundant	N.D.
Roubidoux	Ordovician	Rolla, Mo.	Abundant	Minor	$0.0\pm0.1\%$	Abundant	N.D.
Stevens	Miocene	Wildcat, San Joaquin Valley, Calif.	Abundant	Minor	$0.1\pm0.1\%$	Abundant	Severe
			Abundant	Minor	$0.3\pm0.1\%$	Absent	Severe
Jewett	Miocene	Greeley Field, San Joaquin Valley, Calif.	Abundant	Minor	$0.1\pm0.1\%$	Abundant	N.D.
Wilcox	Eocene	Milam Co., Texas	Abundant	Minor	N.D.*	Abundant	N.D.
Mirador	Tertiary	Rio Zulia Field, Colombia	Abundant	Minor	$0.1\pm0.1\%$	Abundant to sheart	Severe
Paluxy	Eocene	Mize Field	Abundant	Minor	$0.1\pm0.1\%$	Moderate	Severe
Berea	Mississippian	Berea, Ohio	Abundant	Minor	$0.1\pm0.1\%$	Abundant	Severe
"5150" sand		N.E. IAB Field (Coke Co.), Texas	Moderate	Minor	$0.2\pm0.1\%$	N.D.	Severe
Rehm Granite Wash		Hartley Co., Texas	Abundant	Trace	$0.3\pm0.1\%$	N.D.	Severe
Belly River	Cretaceous	Pembina Field, Alberta	Abundant	Minor	$0.1\pm0.1\%$	Abundant	Severe

* N.D. means not determined.

Mica needles and very small kaolinite crystals (Rex, 1965) were found to be the cause of migration damage and the observed water sensitivity in these sandstones. Subsequently, mineralogic analyses were made on additional sandstones, most of which are reservoir sands or aquifers. The occurrence of mica needles in association with small kaolinite crystals is widespread in both geological age and areal extent (Table 1).

Core Flood Tests

Selected cores were first cleaned to remove crude oil. Cleaning procedures can critically affect observed water sensitivity. Solvent extraction with toluene often did not properly clean core plugs, particularly if they were originally air dried, had low initial permeability, or contained asphaltic residues as thin films or "varnish". The latter water-proof residue, if neglected, may cause a false apparent absence of water sensitivity. Flushing in a core cleaning centrifuge with a solvent mixture of chloroform-acetone instead of toluene proved to be a satisfactory method of removing hydrocarbon residues.

Samples for flow tests were cut into one-inch diameter by three-inch long plugs, cleaned, and inserted into standard Hassler sleeve core holders. Separate pieces of the same cores were also used for X-ray, electron microscope, and thin section studies.

Permeant solutions were deaerated and lines purged of trapped gas bubbles to eliminate spurious permeability readings. The permeant solutions were also pre-filtered through a 0.5 micron Millipore filter and treated with 300 ppm formaldehyde to prevent bacterial growth and plugging.

Water sensitivity of sandstone cores was determined by first measuring permeability to air, then to a concentrated artificial brine (or formation brine), and finally to successively more dilute brines, eventually ending with fresh water. In some cases switching directly from a concentrated brine to fresh water was used as a test for water sensitivity. Jones (1964) maintains that this procedure is a more severe test for water sensitivity. Our data support Jones' suggestion.

Each of the sandstones investigated exhibited severe water sensitivity, that is, permeability to fresh water less than a third of permeability to air. This water sensitivity index was adopted by White, *et al.* (1962) of the Bureau of Mines and later used by Hewitt (1963) in their respective studies of water sensitivity of sandstones. The sandstones reported on here typically showed a specific permeability to concentrated brine (50,000 ppm NaCl) of about half that to air. Equilibrium permeability to brine was quickly established whereas with the fresh water, permeability dropped in exponential fashion and leveled off slowly with continued fluid flow.

Susceptibility to induced water sensitivity was determined by comparing permeability of cores to fresh water *before* and *after* saturation with a monovalent cation brine. Usually a brine of 50,000 ppm NaCl was used for initial saturation and flooding tests. Other salts studied include KCl, LiCl, CsCl,

FORMATION DAMAGE IN SANDSTONES CAUSED BY CLAY DISPERSION 359

 $BbCl, NH_4Cl, AgNO_3, Mg(NO_3)_2, NaNO_3, and CaCl_2.$ Our detailed study of the influence of various ions on the degree of formation damage will be described in a separate paper.

Criteria for Damage

Distinguishing clay *swelling* damage from clay *migration* damage is always an interpretive problem. A steady, usually rapid decrease in permeability with decreasing salinity of the flowing liquids is generally a consequence of clay swelling; however, water sensitivity caused by particle migration will also be manifest in this fashion, but sometimes in a more irregular manner. Damage caused by particle plugging was detected by noting a temporary change (usually an increase) in permeability when fluid flow direction was reversed. The Berea and the Entrada sandstones actually produced dispersed clays before permeability decline occurred. This is considered to be the best available evidence for clay migration damage as opposed to clay swelling damage.

In most, but not all, cases permanent loss of air permeability accompanied clay migration damage, especially in the less permeable cores. This was indicated by the difference between initial and final air permeabilities. The permanent loss of permeability suggested the occurrence of a structural change in the pore network, such as closing off pore channels or expansion and realignment of clay particles within the pores. This differs from conventional swelling clay damage where a considerable portion of the original air permeability can be restored in a swelling damaged core by redrying.

Our X-ray analyses show that swelling clays were either absent or present at very low concentrations in the rocks studied thereby providing further assurance that we are dealing with a clay migration phenomena.

RESULTS

Core Floods

Characteristic water sensitivity behavior is shown in Figs. 1 through 5. Many more flow tests were run than are presented in this report; however, the results shown are typical of the water sensitive behavior of each of the sandstone studied.

Berea sandstone was included in this study because it was known to be water sensitive, contained barely a trace of swelling clay, resembled quite closely the other sandstones studied in its clay mineralogy, and was available in quantity.

We found that Berea sandstone could be made highly water sensitive, even to the point of complete cessation of flow, as a result of flooding with a monovalent cation brine followed by flooding with fresh water (Fig. 3). One per cent calcium chloride solution had the opposite effect preventing subsequent decrease in permeability to fresh water (Fig. 4). The flow behavior

of Berea sandstone was not only strongly dependent on the ion in solution, but also on the sequence or order of flooding from one liquid to another. For example, a sodium-saturated core was sensitive to fresh water but if sodium chloride brine was first displaced with a calcium chloride brine the core was no longer sensitive to fresh water (Fig. 4). This demonstrates that the *potential*

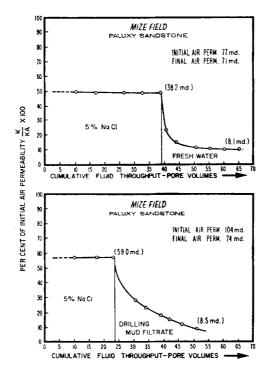


FIG. 1. Water sensitivity displayed by Paluxy sandstone showing reduced permeability to fresh water and mud filtrate.

for damage is reversible and controlled by the type of adsorbed cation. Unfortunately, the damage itself is not reversible by simply changing the pore water composition.

Cores of Berea and Entrada sandstones produced milky dispersions of native clay during core flood tests while permeability decline was observed. Other sandstone cores with initial air permeabilities below about 200–300 millidarcys did not seem to produce any large amount of effluent when showing a permeability decline. The mobile material was probably trapped in the pore throats of the low permeability sandstones and, although moving, most of the clay particles had a mean free path short with respect to the three inches length of the cores. FORMATION DAMAGE IN SANDSTONES CAUSED BY CLAY DISPERSION 361

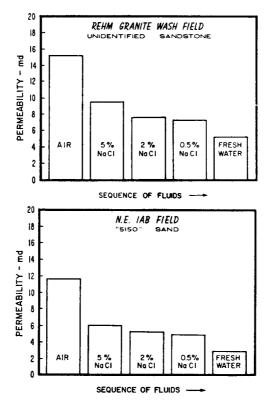


FIG. 2. Water sensitivity displayed by sandstones from Rehm Granite Wash and N.E. IAB Fields showing reduction in permeability with decreasing salinity.

Migratory Clays

Samples of the milky clay effluents were collected in a number of experiments and electron microscope and X-ray studies were undertaken to identify the constituent minerals. They consisted almost entirely of clay minerals with only rare grains of quartz silt evident. Grain and crystal morphology was observed with the electron microscope. X-ray diffraction and electron diffraction phase identifications were made on both effluent and separated fractions of original core material to relate crystal morphology to mineralogy (Fig. 6).

The Berea sandstone effluent consisted of a mass of small hexagonal crystals of kaolinite, usually of 0.1 to 0.3 microns maximum diameter, trapped in a felt-like aggregate of needle- or blade-shaped crystals of mica. These mica crystals usually show a degree of chloritic mixed layer structure. The mica needles usually ranged from 1 to 5 microns in length, 0.1 to 0.3 microns in width, and about 0.005 to 0.5 microns in thickness. This geometry

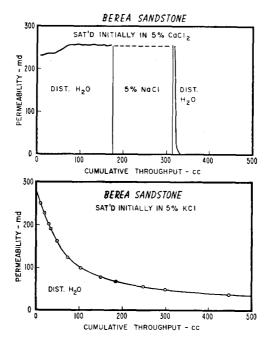


FIG. 3. Effect of saturating Berea sandstone with various cations on the subsequent permeability to distilled water.

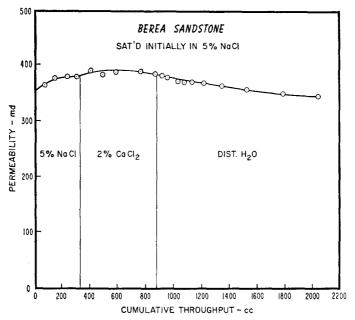


FIG. 4. Inhibiting effect of calcium chloride in preventing serious reduction of permeability to distilled water in Berea sandstone.

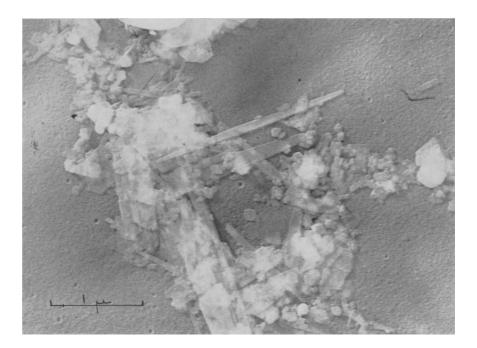


PLATE 1. Electron micrograph of the clay effluent produced from core of Berea sandstone.

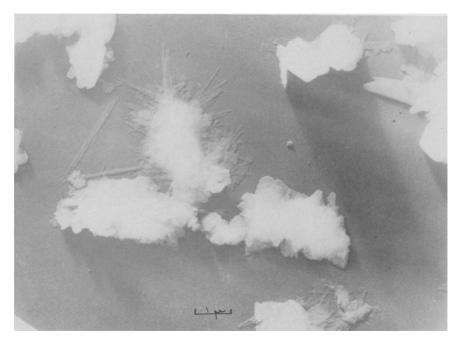


PLATE 2. Electron micrograph of authigenic mica needles and hexagonal kaolinite crystals from the water sensitive Mirador sandstone.

FORMATION DAMAGE IN SANDSTONES CAUSED BY CLAY DISPERSION 363

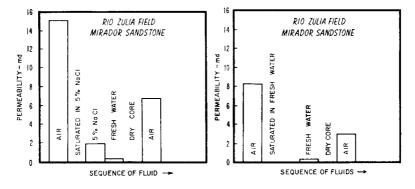


FIG. 5. Water sensitivity exhibited by Mirador sandstone, Rio Zulia field.

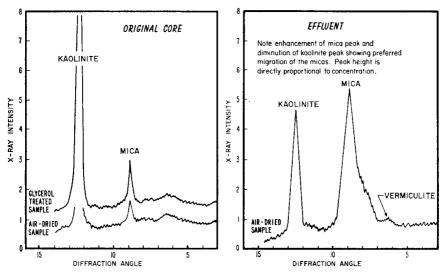


FIG. 6. X-ray patterns of clay minerals in the Berea sandstone and of effluent produced from a sensitized core.

makes them highly susceptible to the interlocking which was commonly observed under the electron microscope (Plates 1 and 2). Furthermore, they are readily maintained in suspension by Brownian motion. Some laboratory preparations in fresh water were observed to remain in suspension for many months without settling. This ease of dispersion and suspension stability enhances the probability of migration by the mica needles and the smallest kaolinite crystals. The similarity of behavior of these two different clay minerals is not surprising because the colloidal properties of kaolinite approach those of micas as the kaolinite grain size becomes smaller and crystal edge effects become more important. However, not all micas nor all kaolinites

participate in this migration phenomenon. Our data suggest that only the authigenic blade- or needle-shaped mica crystals with mixed layer properties and only the finest size crystals of authigenic kaolinite are mobile. Detailed X-ray studies indicate that small quantities of vermiculite and very chloritic mixed layer micas accompany the predominant kaolinite and slightly mixed layer micas in the mobile fraction. The abundant detrital micas and kaolinites common in shales do not appear to contribute to clay migration damage. The effect of various alkali cations is mineral selective. Kaolinite is readily dispersed by lithium while not dispersed by cesium treatment whereas the mixed layer clays are dispersed by both lithium and cesium.

The abundance of mixed layer mica needles in core effluent of high permeability sandstones led to a direct search for mica needles and very fine kaolinite crystals in less permeable sandstones which showed water sensitivity, no montmorillonitic swelling clays, but without obvious clay effluent during core flood tests. The results showed a direct correlation. Numerous montmorillonite-free water sensitive sands showed mica needles and fine kaolinite crystals in varying degrees of abundance (Table 1).

INTERPRETATION

Mechanism of Dispersion of Clay Particles

The mechanism most reasonable to us for clay dispersion is that a certain proportion of the mixed layer mica needles and small kaolinite crystals grow on the surfaces of other minerals such as quartz. These clay crystals would be held in place only by weak van der Waal forces and the coulombic attraction of bonds shared by doubly charged ions such as calcium. The replacement of divalent ions with monovalent ions does not cause clay dispersion until pore water salinity is reduced. Then the monovalent cations at the exchange sites become more strongly hydrolyzed and the silicate surfaces become mutually repulsive because of their strong negative charge (Foster, 1955). This mutual repulsion by the expansion of the double layer is suggested to provide the pressures necessary to disperse the particles and hold them in suspension in a sol state. The occasional observed formation of stable sols of mixed layer mica needles is evidence in support of this mechanism (van Olphen, 1963). We suspect that mixed layer and vermiculite crystal surfaces contribute significantly to this process because of their lower charge density compared to muscovite.

A recent paper by F. O. Jones (1964) describes work on formation damage by clay migration. He included data on the Berea, Paluxy, Yegua, Nellie Bly, and Second Frontier sandstones. His data match the data of this study to such a close degree that it is almost certain that we are both describing the same phenomenon. Jones (1964) presents data to show that damage can be avoided in water sensitive cores if solution concentration is changed very slowly by small steps. This suggests that on a geological time scale where water flow rates and solution composition changes are gradual, we would not expect FORMATION DAMAGE IN SANDSTONES CAUSED BY CLAY DISPERSION 365

natural clay migration. Jones suggests that the rate sensitivity of the dispersion phenomenon indicates that osmotic pressures play a significant role in dispersing mobile clays. Our data indicate that going from a high ionic strength calcium brine to distilled water produces no significant clay dispersion even though osmotic gradients should be large. For this reason we prefer to consider the dispersion phenomenon to be a combination effect of local shear caused by moving pore water and changes in double layer thickness. We prefer to consider the rate effect of slow versus fast salinity change as influencing the rate of expansion of the double layer. Fast expansion is viewed as generating sufficient local shear to permit dislodgment of weakly bonded clay crystals. Slow expansion is viewed as generating insufficient local shear to overcome van der Waal and other weak forces. The differing ability of various monovalent cations to activate water sensitivity will be discussed in a separate paper.

Fraser (1964) described electrical polarization measurements of Berea sandstone that lend strong support to the concept of clay dispersion as a consequence of selective water composition changes. Electrical polarization measurements reflect the relative degree of dispersion or flocculation in claywater systems. Berea sandstone saturated with sodium chloride displayed an abrupt and pronounced polarization as the salinity of the pore water was reduced to a critical concentration of approximately 10^{-2} N. The most severe permeability decrease for Berea sandstone occurs precisely in this concentration range (Jones, 1964). The existence of a "critical concentration" is more suggestive of a surface ionization process than of a bulk fluid osmotic process. Osmotic processes depend primarily on concentration gradients instead of absolute concentration values. For these reasons we suspect that osmotic forces are not the primary cause of clay dispersion.

CONCLUSIONS

The consistent association of mixed layer mica needles and/or small kaolinite crystals with water sensitivity in the absence of montmorillonite combined with the direct evidence from high permeability sandstones strongly suggests that dispersion of mica needles and small kaolinite crystals is the principal cause of clay migration damage in the suite of sandstones studied.

REFERENCES

- DODD, C. G., CONLEY, F. R., and BARNES, P. M. (1955) Clay minerals in petroleum reservoir sands and water sensitivity effects: *Clays and Clay Minerals*, Proc. 3rd Conf., Natl. Acad. Sci.—Natl. Res. Council Pub. 395, pp. 221–38.
- FOSTER, M. D. (1955) Relation between composition and swelling in clays: Clays and Clay Minerals, Proc. 3rd Conf., Natl. Acad. Sci.—Natl. Res. Council Pub. 395, pp. 205-220.
- FRASER, D. C. (1964) Electrical properties of clay-containing sandstones—Part (B): Amer. Petrol Institute Report No. MT-64-4.
- HEWITT, C. H. (1963) Analytical techniques for recognizing water sensitive reservoir rocks: Jour. Petrol. Tech. 15, 813-8.

- JOHNSTON, N., and BEESON, C. M. (1945) Water permeability of reservoir sands: Trans. A.I.M.E. 160, 43.
- JONES, F. O. (1964) Influence of chemical composition of water on clay blocking of permeability: Jour. Petrol. Tech. 16, 441-6.
- MONAGHAN, P. H., SALATHIEL, R. E., and MORGAN, B. E. (1959) Laboratory studies of formation damage in sands containing clays: Jour. Petrol. Tech. 11, 209–15.
- MOORE, J. E. (1960) Clay mineralogy problems in oil recovery: Petroleum Engineer 32, 78-101.
- REX, R. W. (1965) Authigenic kaolinite and mica as evidence for phase equilibria at low temperatures: Clays and Clay Minerals, Proc. 13th Conf., Pergamon Press, London, pp. 95-104.
- VAN OLPHEN, H. (1963) An Introduction to Clay Colloid Chemistry: Interscience, N.Y., 301 pp.
- VON ENGELHARDT, W., and TUNN, W. C. (1954) The flow of fluids through sandstones: translated by P. Witherspoon, *Illinois State Geol. Survey Circular*, No. 194 Urbana.
- WHITE, E. J., BAPTIST, O. C., and LAND, C. S. (1962) Physical properties and clay mineral contents affecting susceptibility of oil sands to water damage, Powder River Basin, Wyoming: Rept. Invest. No. 6093, U.S. Bureau of Mines, Washington, D.C.