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Clay migration and entrapment in synthetic porous media*

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Water-sensitivity in sandstones is commonly ascribed to mobilization, transport and recapture of clays in downstream pore throats. This is based mainly on knowledge of the clay mineral content and its electrochemistry. This paper describes a method for directly observing water-sensitivity mechanisms under the microscope using pore networks etched in glass, with clays introduced into the networks. This allows direct identification of colloidal and transport factors, which is not possible in sandstone cores. The migration behaviour has been investigated with variations in flow rate, acceleration forces and salinity. Migration was observed to occur in two distinct modes: (1) as flocs, especially in concentrated brines and at high flow rates, (2) as deflocculated individual particles, especially in distilled water. Recapture of clays occurs by the formation of 'particle bridges'. The upstream pressure indicates that particle recapture is related to decreasing permeability.

Keywords: Water-sensitivity; Formation damage; Clays; Sandstone

Introduction

A porous medium is said to be 'water-sensitive' if its permeability is dependent on the chemistry of the flowing fluid. Permeability is, of course, a property of the porous medium and not the flowing fluid, but in water-sensitive media a change in the fluid stimulates a change in the fabric of the rock. This phenomenon has been widely recognised in the petroleum industry and is usually attributed to: (a) *in situ* swelling of clays and/or (b) migrating clays and other fine material. A third type of water-sensitivity is the reaction between acidic fluids and acid-soluble minerals, such as carbonates and iron oxides, and is relevant to removing well bore damage and reservoir stimulation techniques.

Previous work

In situ swelling of clay minerals was recognised as a cause of permeability loss by Baptist and Sweeney (1955) and has since been described by several authors (Hewitt, 1963; Land and Baptist, 1965; Veley, 1969). Sandstones with interstitial smectite (or mixed-layer clays with smectitic components) are particularly susceptible to this form of water-sensitivity. In this case, permeability is lost when the flowing fluid is changed from strong brine (say 30,000 ppm NaCl) to distilled or deionized water. This change in salinity causes increased hydration of interlamellar cations in the smectite lattices, swelling of the clay and the effective reduction or even complete blockage of smectite-lined pore throats to fluid flow. The effect of this can be seen in *Figure 1*. In this example, permeability of the sandstone remained relatively constant at approximately 24 mD when 30,000 ppm NaCl was flowed through but was reduced to < 2 mD after only a few pore volumes of 10,000 ppm NaCl. When distilled water was passed through the core plug, the permeability dropped even further. X-ray diffraction analysis of the interstitial clays revealed the presence of a Ca-smectite, and this (with the absence of fines in the effluent) suggested *in situ* swelling was the cause.

Gray and Rex (1966) defined the problem of migrating clays and there have been several subsequent

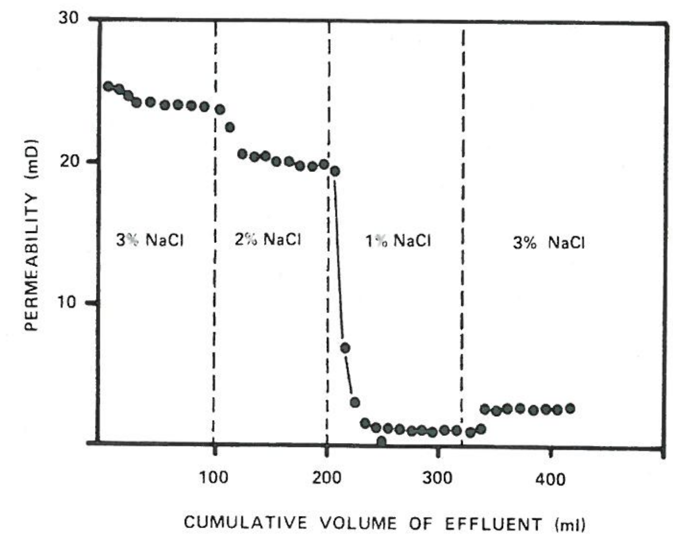

Figure 1 Water-sensitivity of the Spiney Sandstone, Moray Firth Basin. Note the large decrease in permeability when the brine is changed to 1% NaCl. The lack of suspended solids in the effluent and the presence of smectite in the core suggest *in situ* swelling is the cause of the permeability loss

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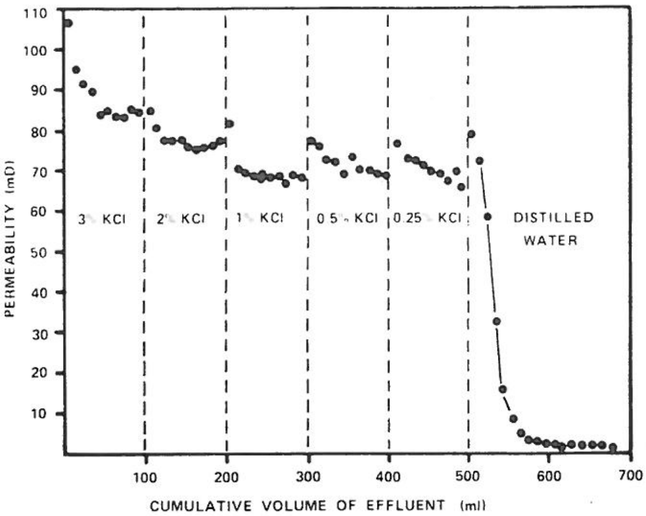

Figure 2 Water sensitivity of the Hopeman Sandstone, Moray Firth Basin. Permeability drops to less than 1 mD when distilled water is passed through the core plug. The presence of suspended solids (kaolinite, illite, quartz and lepidocracite) in the effluent is a clear indication of migrating fines (from Lever and Dawe, 1984)

examples (Reed, 1977; Khilar and Fogler, 1983; Lever and Dawe, 1984). Superficially, the effects may resemble those of *in situ* clay swelling (*Figure 2*), though the absence of smectite in the core material and the presence of suspended solids in the effluent indicates a different mechanism, wherein fines are expelled from the porous medium. In this case, changes in electrical double-layer thickness cause the loss of permeability (Aveyard and Hayden, 1973; van Olphen, 1977; Sharma *et al.*, 1985). Due to isomorphous substitution in the lattice, clay minerals generally have a net negative charge, balanced by cations held weakly at the surface by van der Waals forces. These cations are readily exchangeable, and will form an electrical double-layer when the clay is immersed in water. In this double-layer, the concentration of cations is greatest at the clay surface and decays away to that of the bulk solution at a finite distance. The double-layer will be relatively compressed in a solution of high ionic strength and expanded in those of low ionic strength. When two particles approach each other, their double-layers will interact and the particles will be mutually repelled: it therefore follows that in solutions of low ionic strength (i.e. expanded double layer) repulsion and migration will be enhanced. Therefore, a reduction in salinity from 30,000 ppm NaCl to distilled water will cause a large expansion of electrical double layers on interstitial clays, adjacent particles will repell each other, and fines will be entrained in the fluid flow, only to be recaptured downstream, so causing the blockage of pore throats and a decrease in permeability.

Unfortunately, these mechanisms of water sensitivity have not been directly observed but have been inferred to be operating within sandstone core plugs. Cation exchange processes may be verified by analysing the fluid before and after passing through the core, but the migration and recapture of the clay particles can often only be inferred by a drop in permeability and mineralogical analysis of the clays. Previous work on particle migration by Muecke (1979) used packs of unconsolidated sand and chips of carbonate material, which could be dissolved with acid once the experiment was completed. In the present study, attempts were made to observe the migration of clays through unconsolidated sand packs, though it was found that the sand migrated as much as the clay. It would be of great benefit if particle migration could be observed within a rigid medium under a standard optical microscope. The use of micromodels is one such method. The model systems permit the separation of the colloidal and transport mechanisms. Micromodels are two dimensional transparent microporous structures which attempt to simulate the basic fabric of reservoir rock. Inter-connected pores and channels of the order of 5–200 μm are formed to any required geometry. Any particle migration therein can be observed under a standard optical microscope and recorded on a video system. The aim of the work reported in this paper is therefore to reproduce the water-sensitivity results obtained within the sandstone core plugs in such a way that the nature of the particle migration is seen.

Manufacture of micromodels

Micromodels were manufactured in epoxy resin using the methods of Bonet and Lenormand (1977) and Mahers *et al.* (1981). However, it is felt that the surface properties of Si-glass are likely to be closer to those of quartz-rich sandstones. The glass models were made from mirror glass, using a modified version of McKellar

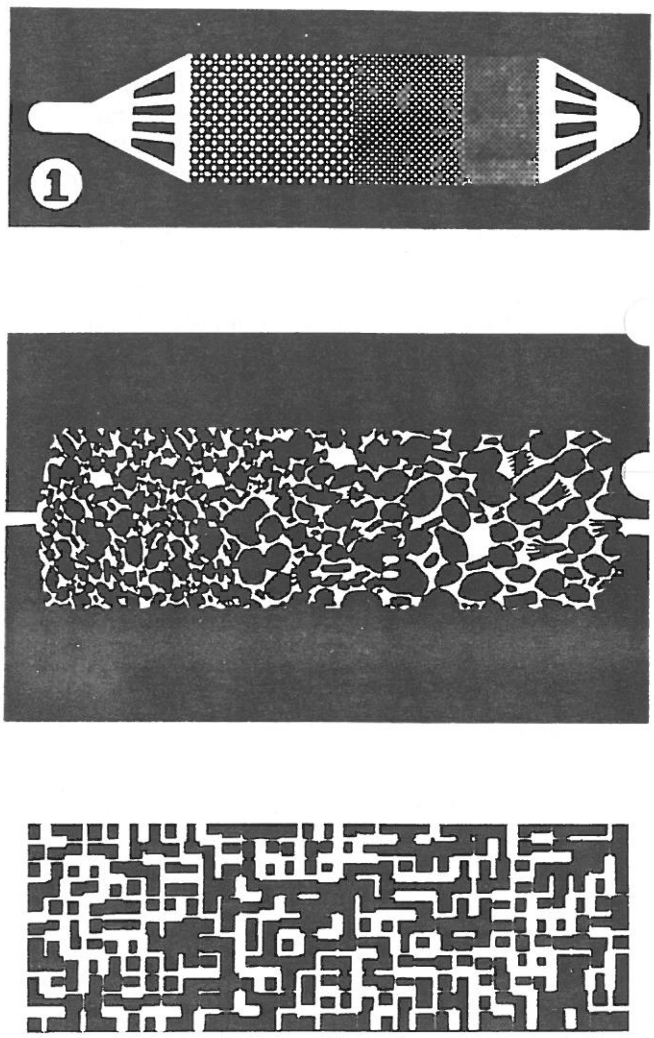

Figure 3 Some examples of network design for micromodels. The long axis of each model is approximately 3 cm

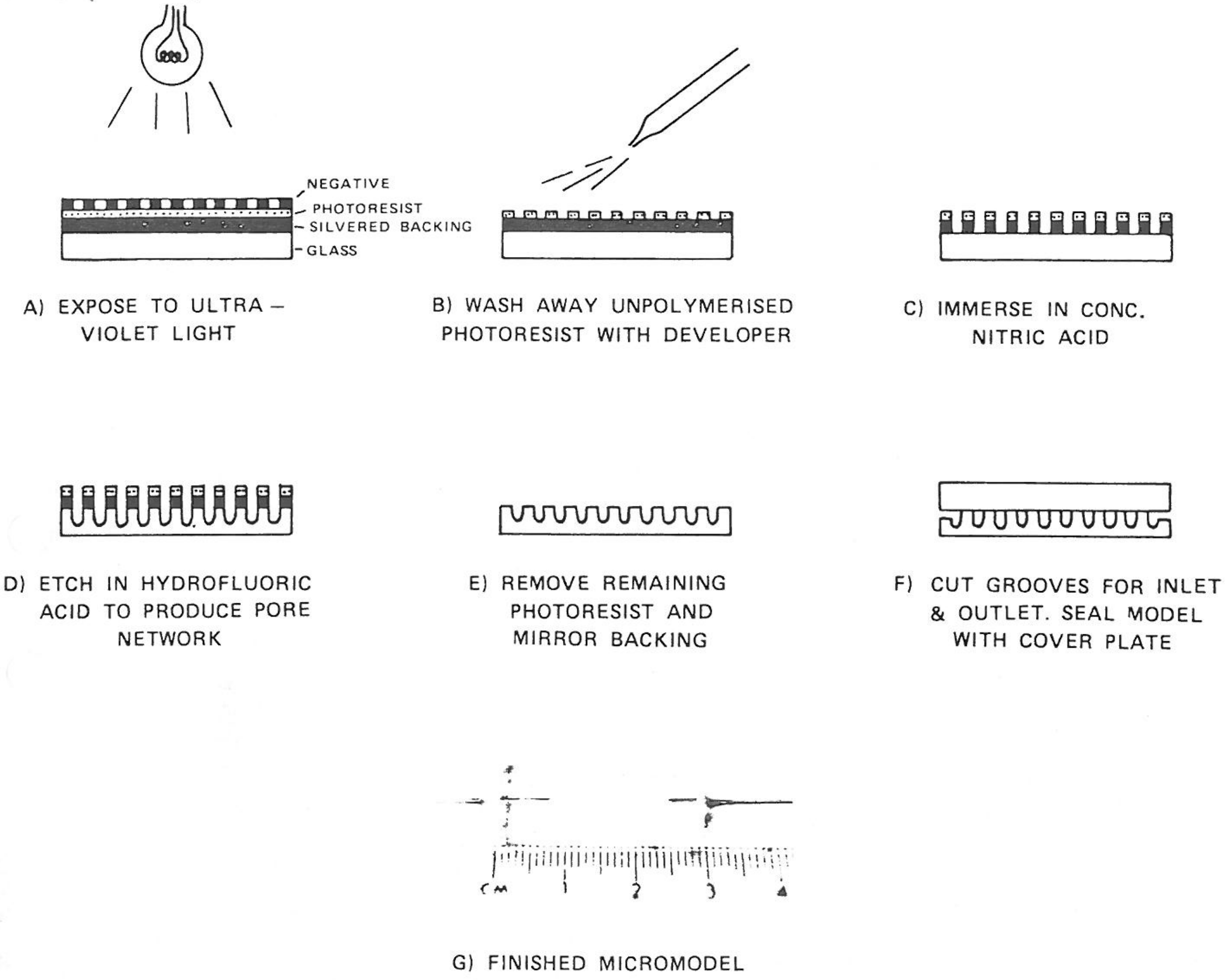

Figure 4 Summary of procedure for manufacturing micromodels in glass

a . Wardlaw's (1982) procedure. This involved essentially seven steps:

(1) Design of the network by computer graphics and/or manual drafting. The drafted image was then photographed and kept on 35 mm high contrast negative film. Some examples are shown in *Figure 3*.

(2) Removal of protective backing from the mirrored surface, by immersion of the glass in hot NaOH or cold trichloroethane, depending on the nature of the backing material.

(3) Coating of the silvered surface with photoresist. In this case, a drop of photoresist was placed on the surface and spread out by spinning the glass on a rotary motor. Thickness of the coating was thus controlled by speed of the motor.

(4) Projection of the network image onto the photoresist. This was achieved by placing the 35 mm negative on the coated surface of the glass. Good contact was maintained by using cling-film and a small vacuum frame (*Figure 4a*) and the photoresist was exposed to u.v. light.

(5) The unpolymerized photoresist was then washed away in developer, leaving a coating with 'holes' in it (*Figure 4b*) and the slide was then immersed instantaneously in concentrated (35%) HNO_3 to remove the silvered backing beneath the 'holes' (*Figure 4c*).

(6) The network was then etched in 30% HF for ~12 min (*Figure 4d*) after which the photoresist and silvered backing were removed with trichloroethane and concentrated HNO_3 respectively, to leave just the etched glass network (*Figure 4e*). Grooves were then cut in the glass to accommodate the inlet and outlet pipes.

(7) A cover plate was then sealed onto the base plate (*Figure 4f*) by heating for ~30 min at 720°C (from room temp.). A finished micromodel can be seen in *Figure 4g*.

Some experimentation is needed to suit the technique to individual cases, and many intermediate steps are needed depending on the nature of the photoresist. The general technique outlined above must be used in conjunction with the manufacturer's instructions for the photoresist, and tailored to suit individual needs.

In this study, clays were then introduced into the model as a dilute slurry in distilled water of neutral pH. Samples of Georgia kaolinite, Fithian illite and Wyoming bentonite were used. When sufficient quantities of clay had been captured in the model, the

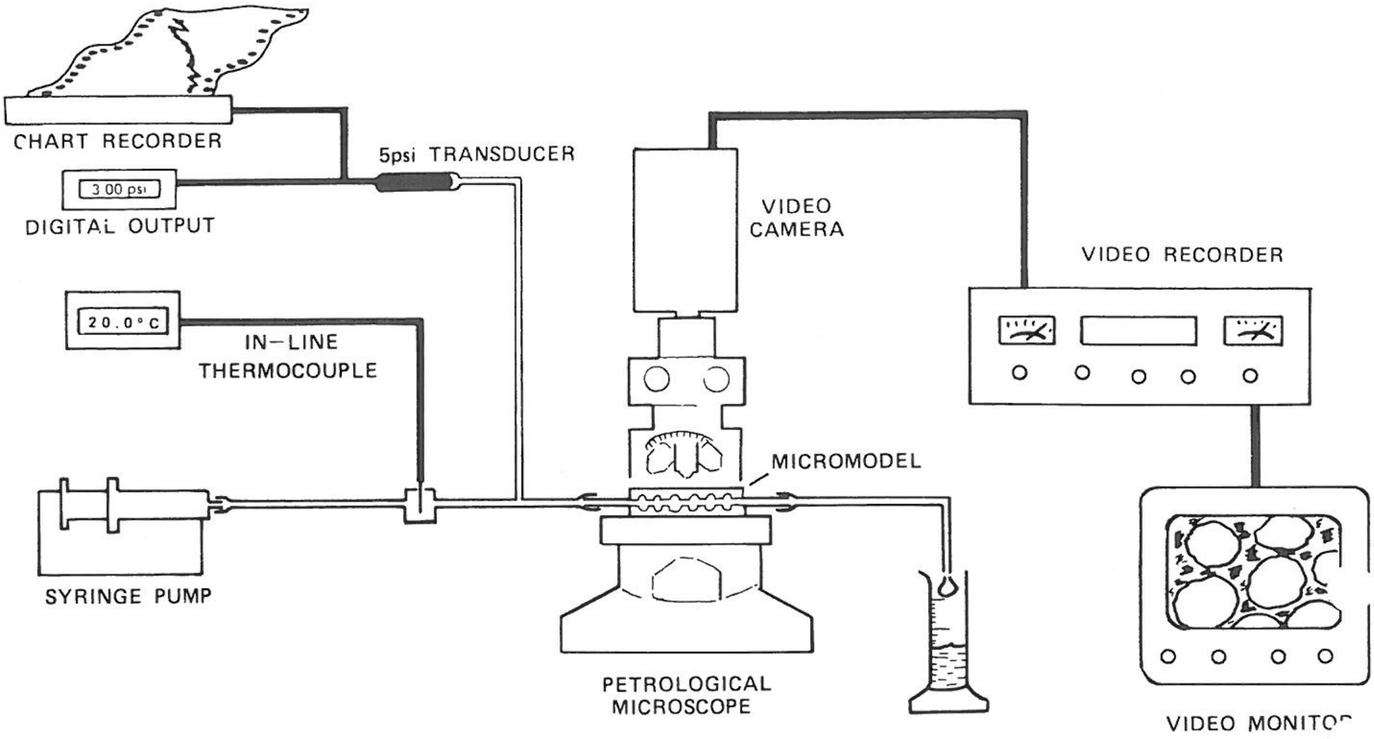

Figure 5 Experimental set-up for micromodel studies. Micromodels are mounted on the stage of a microscope and fluid is pumped through using a syringe pump. Upstream temperature and pressure are monitored and clay migration is recorded on video

flow of slurry was ceased and the water-sensitivity tests began without allowing the model to dry out.

Water-sensitivity tests

In the water-sensitivity tests, fluid was pumped through the model using a syringe pump at a variable flow rate. The temperature of the flowing fluid was measured with an in-line thermo-couple and upstream pressure was measured with a 5 psig pressure transducer. From these parameters, a measure of the permeability of the system could be continuously monitored. Movement of particles was observed with the microscope and recorded on video or 35 mm film using an SLR camera. The effects of flow rate and salinity change on particle migration were investigated. The experimental apparatus is shown in *Figure 5*.

The scope of this technique to identify mechanisms will be shown by a number of examples. We have carried out a study of colloidal (salinity changes in this case) and transport (hydrodynamic) effects due to velocity change and clay particle migration.

Results

In-situ swelling

The experiments to induce *in situ* swelling of Wyoming bentonite in micromodels were successful. In two cases (one with a glass micromodel, one with a resin micromodel), flooding the model with NaCl brine, then distilled water, caused a decrease in permeability. The resin micromodel became virtually impermeable and the permeability profile of the glass micromodel is shown in *Figure 6*. As can be seen, the switch to distilled water caused a permeability drop which was investigated until there was a distinct danger of over-pressuring the pressure transducer. The *in situ* bentonite showed no difference before and after the introduction of distilled water under the optical microscope, so the swelling effect remains speculative.

Migration of clays

Early tests failed to identify any appreciable migration of the clays and it became obvious that, once the clay had been captured from the slurry (in the introduction of the clay into the micromodel), it had probably obtained a stable configuration and was unlikely move again. For this reason, having flushed the model with the first fluid (e.g. 30,000 ppm NaCl), the clays were redispersed within the model by 15–30 min ultrasonics. Only after this redispersion did the particles migrate.

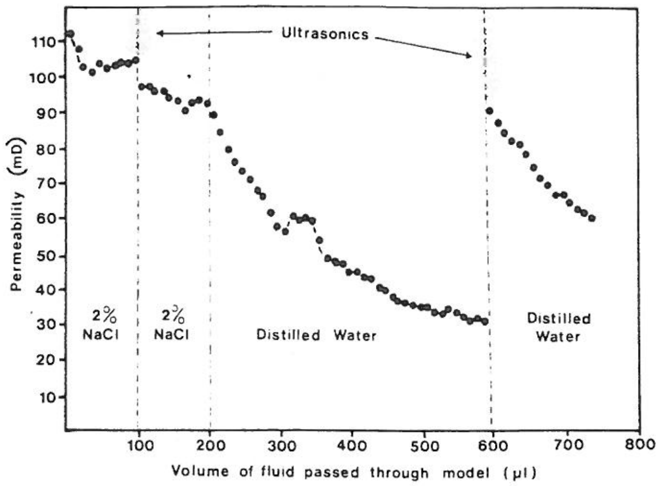

Figure 6 Water-sensitivity of a glass micromodel containing Wyoming bentonite. Permeability is lost when distilled water is passed through. Re-arranging the clays by ultrasonic treatment temporarily restores permeability

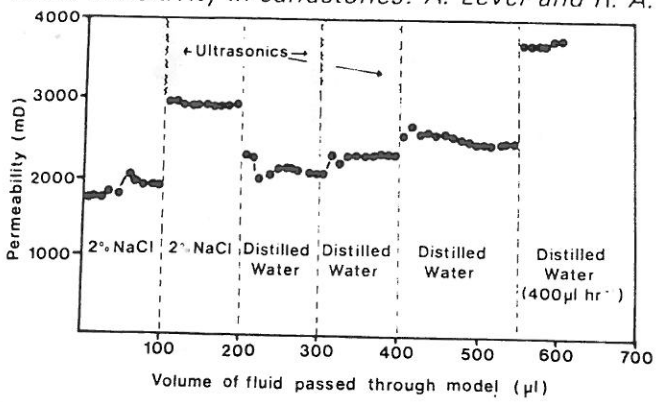

Figure 7 Permeability profile for glass micromodel containing Georgia kaolinite. Despite the migration of some of the clay, permeability remains high, even after redistribution of the clay by ultrasonics

The micromodel was subjected to brine flow tests following the sequence often used with water sensitivity sandstone core plug tests. We varied the flow rate and salinity of the flowing fluids to identify the conditions conducive to migration and recapture.

Migration appeared to occur in two distinct modes:

(1) As medium to larger-sized flocs ($\leqslant 100$ μm) of clay. This appeared to occur on the initiation of flow, or after a disturbance of the system, and in strong brine or distilled water. It is therefore thought to be independent of salinity.

(2) As fine ($<5\mu$m) individual particles. This was most clearly seen in dead-end pores and is best described as like the 'swarming' of insects. It occurred only when distilled water was the flowing fluid and was therefore salinity-dependent. Individual particles could be seen to migrate slowly away from the 'swarming' mass and into the path of fluid flow. This effect is interpreted as the Brownian motion of deflocculating clay particles. The process probably occurs in the main flow channels as well, but is not seen, as the particles are quickly swept away.

At present, the size of the channels and pore throats in our glass models is some 10–50 μm. This is probably ger than those in most sandstone cores. Therefore, it is not unexpected that the permeability loss experienced with sandstone core plugs was not seen in the micromodel tests. As *Figure 7* illustrates, no dramatic decrease in permeability occurred, and if anything, the trend was slightly upward. The large-scale migration of flocs did not seem to have an adverse effect on the permeability, and any permeability loss due to deflocculation in distilled water was minimal, even though the recapture of the migrating fines was reproduced to some effect. In some cases the mobile flocs were seen to collide with captured flocs, which caused the aggregates to mutually adhere, whilst in other cases, collision caused the break-up of flocs into smaller units. However, with further improvements and manufacturing procedures, as discussed later, this aspect of water sensitivity can be further studied.

The effect of flow rate increase on the migration of flocs was investigated in an experiment where flow rate was increased at progressively faster rates. As before, the migration effects were recorded on video. In this experiment, flow was initiated with 30,000 ppm NaCl at 7.66×10^{-3} μl s^{-1}, and flow rate was doubled in 40 s at each successive step, reaching an ultimate flow rate of 0.2 μl s^{-1}. Migration of particles was searched for at each flow/pressure increase, but none was observed until a flow rate increase (d^2V/dt^2) of 0.0153 μl s^{-1}/40 s (3.83×10^{-4} μl s^{-2}), and the phenomenon was only seen on a large scale upon flow rate increases of 7.66×10^{-4} μl s^{-2}. With progressively faster flow rate increases, progressively larger volumes of flocculated material were moved (*Figure 8*). After flow had been increased to 0.2 μl s^{-1}, the main flow channels had been cleared of clay, as if the clay flocs had 'sandblasted' a clear channel through.

Recapture of clays

In one experiment, the recapture of clay flocs was reproduced. In this case, the build-up of 'particle bridges' across a pore throat coincided with the increase in upstream pressure (*Figure 9*) and therefore decrease in permeability. After flowing 100 μl of fluid through the model, flow was reversed. However, the particle 'bridge' had attained a stable configuration and remained intact, even after very large increases in flow rate. As a result, permeability remained low.

Discussion

The objective of this paper is to indicate a possible method of directly observing the water sensitivity mechanisms that occurs in sandstone rock. At this stage, the sandstone water sensitivity results are only partially reproduced, though improvements in the manufacture of micromodels may help. With regard to this, a few points need to be made.

(1) The micromodels produced tend to have permeabilities in the 1–4 Darcy range (i.e. an order of magnitude higher than those of the sandstone core plugs) and it is therefore unlikely that any migration of clay would have a large effect on permeability. Manufacture of 'tighter' micromodels may solve this problem but technical difficulties can occur (see 2).

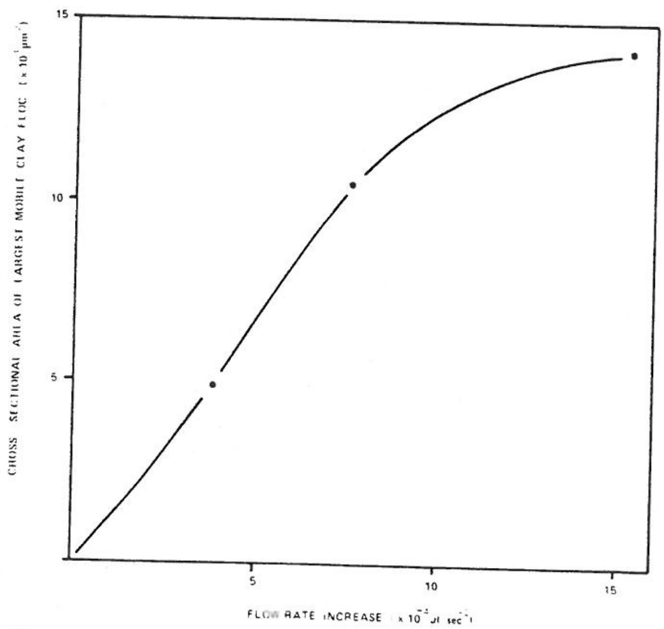

Figure 8 Relationship between flow rate increase and size of migrating kaolinite flocs

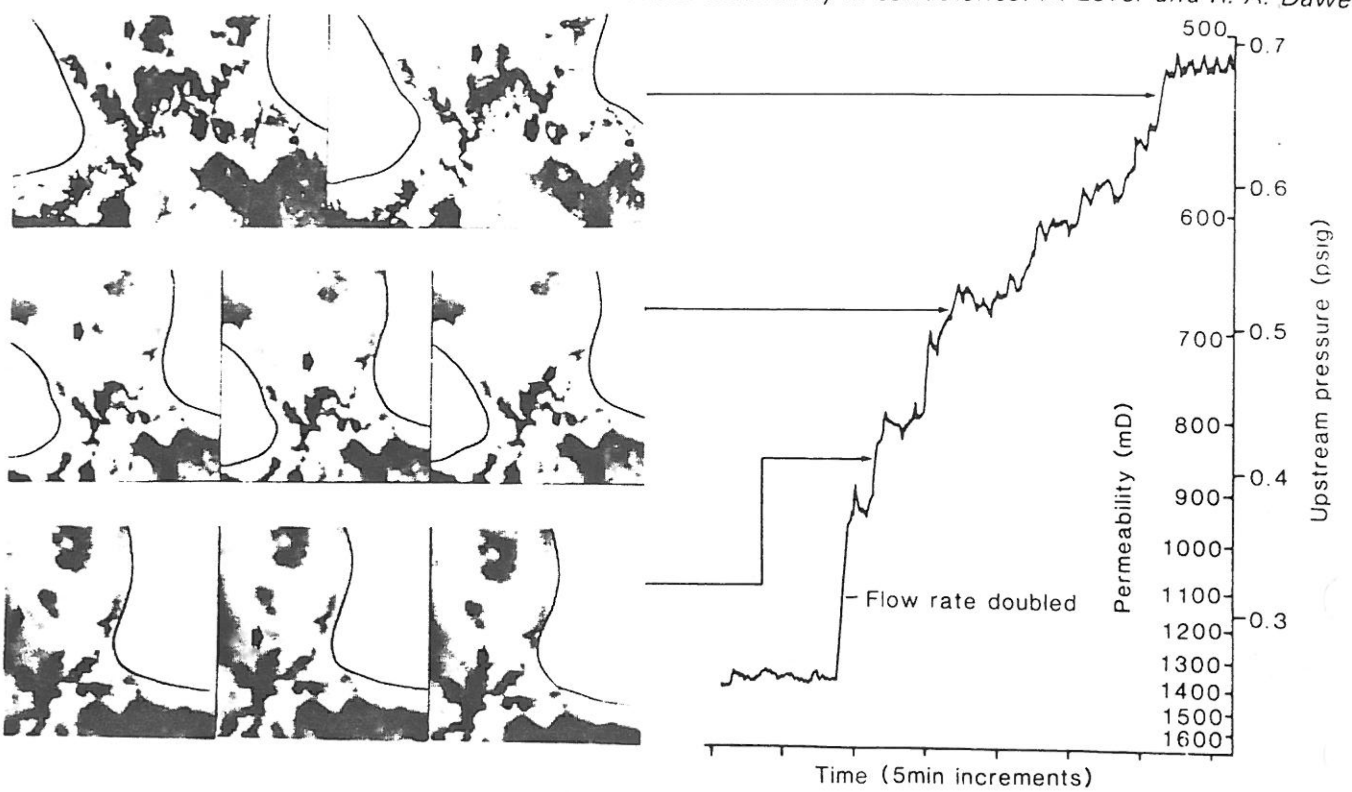

Figure 9 Effect of clay recapture on micromodel permeability. As the clay 'bridge' develops across the pore throat, permeability decreases. Increases in upstream pressure can be correlated with the recapture of individual flocs

(2) In our current etching processes, the HF tends to etch as wide as it etches deep and it is therefore difficult to etch the long, narrow channels of a 'tight' micromodel. However, by suitable sintering processes, we can overcome this difficulty to some extent.

(3) In the sealing process, the time and temperature at which the glass is sintered is crucial. A difference of ± 5 min or ± 5°C may make the difference between an unsealed model and a lump of molten glass! Furthermore, in the sealing process, some areas can sometimes seal before others and extra sealing is needed to achieve a seal over all of the model. Even if shallow, narrow channels could be etched, therefore, it can be difficult to achieve a good, even seal.

(4) The pressure differential needed to produce flow rates of 100–200 μl h^{-1} is very small (~0.01 atm.) due to the high permeability and small overall dimensions of the micromodels used. The pressure measurement probably will have a large experimental error under these conditions. Larger models, some ten times larger in area, can now be made, but sealing becomes an even greater problem. The pressure drops across the models will be some 5–10 times higher.

(5) It must also be remembered that by flowing the clay into the model as a slurry and then redistributing it by ultrasonics, one has absolutely no control on the distribution of clay. In particular, clay may be entrapped in the inlet or outlet pipes, in which case a change in permeability may occur although the reason may not be apparent. This is possibly the cause of the permeability decrease in *Figure 6*.

Conclusions

The simulation of sandstone water-sensitivity in transparent glass micromodels is a useful technique in identifying the mechanisms occurring within the rock-fluid system. Preliminary results are promising, and suggest that clay may migrate through a porous medium as: (a) Individual particles, due to deflocculation in low ionic strength solutions, or (b) as flocs when flow rate is increased too quickly. Recaptu of clay causes a decrease in permeability. Improvements in micromodel manufacture and increasing their size are planned before further experiments are carried out.

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