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Modification of Two-Phase Flow Properties by Polymer Adsorption: Wettability Effects

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Abstract

Adsorption of water-soluble polymers in porous media is known to reduce water permeability much more than oil permeability. This effect is often referred to as "Disproportionate Permeability Reduction" (DPR) and can be used in production well treatment to reduce the water cut. This paper deals with nonionic polyacrylamide adsorption on carbonate rocks having different wettability properties. Cores were used first in their native water-wet condition, and after a wettability modification, which was obtained by aging cores saturated with a polar crude oil at 60°C for six weeks. Efficiency of the treatment was attested and quantified by strong changes in the wettability index as measured using Amott tests. Drainage and imbibition cycles were performed on these carbonate samples before and after polymer treatment. Polymer adsorption and end-point oil/water relative permeabilities were compared for both media. While the quantity of adsorbed polymer is almost the same on water-wet and on wettability-modified cores, adsorption rates, estimated from viscosity profiles of effluents, are significantly different in the two cases. All our results indicate that polyacrylamide adsorbs on the rock whatever being the wetting conditions. While disproportionate permeability reduction is always observed, DPR is greater in low-permeability cores and increases with the water-wet character of the medium.

Introduction

Excessive water production is a problem of central importance for field operators since high water cut can lead to stop production for economical reasons. Among the solutions proposed to circumvent this problem, direct injection of polymer in the surroundings of the wellbore has been shown to be an efficient one. Polymer or gel injections in producing wells are able to lower the water cut by selectively reducing water relative permeability of the rock with respect to oil relative permeability. From a physical point of view, adsorption of water-soluble polymer on pore walls is known to modify two-phase flow properties of a porous medium. Several mechanisms involved in the action of polymers or gels have been reported in the literature and put forth to explain what is often referred to as "Disproportionate Permeability Reduction" (DPR):

- segregated pathways. This hypothesis put forth by Liang et al. [1], and more recently by Nilsson et al. [2] is based on the idea that water and oil are flowing in two different pore networks, i.e. water in the smallest pores and oil in the largest ones. Consequently, since a

hydrophilic polymer flows preferentially through the water network, it is able to reduce water permeability much more than oil permeability. A similar mechanism was proposed by Thompson and Fogler [3] who observed that oil pathways are not significantly modified after gel treatment whereas water pathways are strongly reorganized after gel placement. Similarly, polymer injection was performed into rocks having different wettability properties and Nilsson et al. [2] found that DPR was greater for porous media of mixed-wettability, which is consistent with the fact that water and oil pathways are better separated in this case.

- shrinking/swelling of the adsorbed layer depending on phase flow (Menella et al. [4]). A possible explanation lies in the fact that adsorbed polymer shrinks while oil flows, and swells in the presence of water. This mechanism is consistent with the fact that the stress applied on the polymer layer by the flow is strong enough to induce a significant deformation of the layer. This mechanism was observed from direct visualization on micromodels (Dawe and Zhang [5]) but was performed with a gel and more work is required to confirm that this mechanism has also a significant effect while using a polymer. This mechanism can actually be considered as a wall effect as detailed below.

- wall effects (Zaitoun et al. [6]). Polymer adsorbs at the walls of the pores under the form of a layer, whose size is significant (typically around $0.5\ \mu\text{m}$ for high molecular weight polyacrylamide) compared to pore size (see Fig. 1). Due to the presence of this layer, the mobility of the wetting fluid (i.e. water) is much more reduced than the mobility of the non-wetting fluid (i.e. oil or gas) and this can be explained as follows. At the residual oil saturation (Fig.1a)), for which oil droplets are trapped at pore-throats entrances, water permeability is strongly reduced when the polymer layer is present since the water pathway is considerably narrowed in this case. At the irreducible water saturation (Fig. 1b)), oil is mainly flowing in the center of the pores and the oil permeability is not strongly affected by the presence or not of the adsorbed polymer layer. This mechanism is coherent with several experimental results obtained on water-wet cores of natural porous media both in drainage and imbibition conditions (Barreau et al. [7]). The phenomenological validity of this mechanism was also checked using direct pore-scale numerical simulations of oil-water flow, performed on a model-pore geometry (Barreau et al. [8], [9]). Moreover, the adsorbed polymer layer may attenuate rock tortuosity contributing to make the non-wetting phase flow easier in smoother channels.

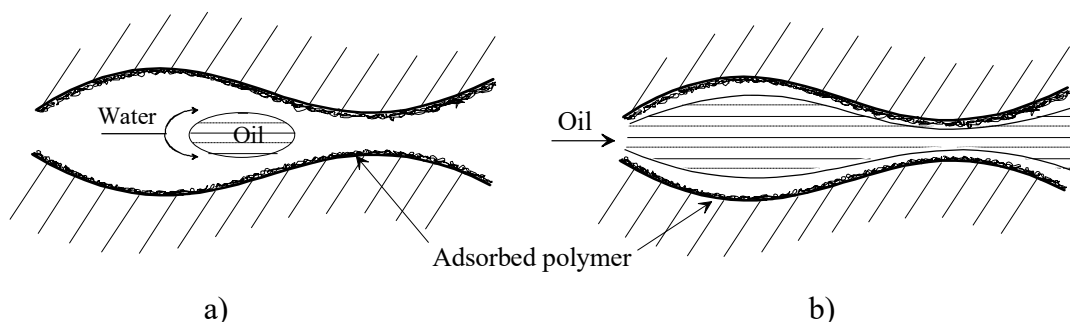


Figure 1. Pore scale end point situations

- wettability effect. Adsorption of hydrophilic polymer improves the water-wet character of the rock (Zaitoun and Kohler, [10]). As a consequence, water relative permeability may be reduced while oil relative permeability may be improved.

If all reported results highlight the DPR effect of the polymer, most of them are obtained on porous media in water-wet conditions. From a practical point of view, relative permeabilities are strongly dependent on wettability conditions of the porous medium, and these conditions can be very different from a reservoir to another one. For this reason, wettability is a parameter that must be investigated in the study of DPR by polymer.

The aim of this paper is to evaluate effects of wettability on two-phase flow modifications by polymer adsorption. Different wettability conditions of limestone core samples were obtained by saturating and aging cores with crude oil. Wettability indices to water and oil were measured, before and after crude oil treatment, in order to accurately evaluate wettability changes. Drainage and imbibition cycles were performed, before and after polymer injection in both native water-wet cores and wettability-modified cores.

Materials

Porous Media.

For each experiment, two cores were used, one in its native wettability condition (actually water-wet and referred to as W-W in the rest of the paper) and another that had been aged with crude oil (referred to as M-W in the following). Two types of carbonate rocks were used in our experiments. The first one was a St-Maximin limestone with a porosity of 0.43 and an absolute permeability to brine of $1.62 \mu\text{m}^2$. The second one was an Estailades limestone. Two different experiments, referred to as “Estail1” and “Estail2”, were carried out on this latter medium. The four Estailades cores had a porosity ranging from 0.19 to 0.236 and an absolute permeability to brine ranging from 0.136 to $0.202 \mu\text{m}^2$ respectively.

Table 1. Petrophysical properties of the cores.

Core Sample	W-W St. Maximin	M-W St. Maximin	W-W Estail1	M-W Estail1	W-W Estail2	M-W Estail2
ϕ	0.425	0.43	0.236	0.19	0.243	0.206
$K(\mu\text{m}^2)$	1.62	1.62	0.202	0.136	0.182	0.273

To estimate wettability indices, small cylindrical plugs of 4-cm in diameter and 7-cm in length were used for each type of carbonate. For displacement experiments, we used cores of 5-cm by 5-cm in cross section and 20-cm to 25-cm in length. They were coated with epoxy resin reinforced with glass fibers to ensure tightness and preserve good transparency to γ -ray.

Fluids

Synthetic high salinity brine (50g.l^{-1} KI) was used as the aqueous phase for displacement processes and polymer solvent. The potassium ion prevents clay swelling and the iodide ion improves saturation measurement accuracy while using a γ -ray attenuation technique. Lower-salinity brine (10g.l^{-1} KI) was also prepared to perform tracer propagation rate measurements in the cores. The oil-phase was a Marcol 82[®], a mineral oil having a viscosity of 24 mPa.s at 19 °C. A polar crude oil, provided by TotalFinaElf was used for wettability modification.

Polymer solution

We used a nonionic polyacrylamide (PAM provided by Floerger[®]) having a molecular weight of 10^7 Dalton, available under powder form. The solution was prepared at a concentration of 2500 ppm, by slow addition of polymer to the brine, in a vortex created by magnetic stirring. Sodium azide (NaN_3) was added to the solution at a concentration of 0.4 g.l^{-1} to prevent bacterial degradation. After complete dissolution of the powder, the solution was filtered with a set of 8, 3 and $1.2 \mu\text{m}$ Millipore[™] membranes, in order to remove microgels.

Viscosity of the polymer solution was measured for a wide range of shear rates with a Contraves LS 30[™] Couette viscometer. Viscosity versus shear rate is represented in Fig. 2 showing that a newtonian plateau at 8 mPa.s is maintained for shear rates up to 10 s^{-1} .

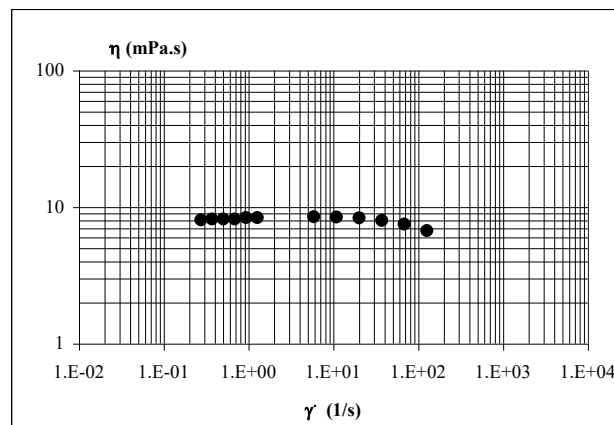


Figure 2. Polymer solution viscosity

Core test equipments

In order to estimate wettability of our cores before and after aging treatment, wettability indices were measured using Amott tests. The Amott test is based on the comparison of displaced volumes of oil and water, either by spontaneous imbibition of the cores or by forced injection. Forced injections were performed using Hassler sleeves, while spontaneous imbibitions were carried out in calibrated glass cells. As explained further on, oil and water indices are calculated from these volumes providing the global wettability index as detailed in [11].

The coreflood setup used to perform our experiments is represented in Fig. 3. Fluids were injected into the core with a Pharmacia[®] displacing pump. The core was positioned horizontally on a two-dimensional rig moving a γ -ray source and a photon counting device used to measure fluid saturation. During two-phase flow experiments, pressure drop across the core was measured with a differential pressure transducer. Cumulative production of oil and water was recorded by weighing effluents.

The estimation of the mass of adsorbed polymer can be performed from the comparison between tracer and polymer fronts. For this reason, during injections of polymer and low salinity brine, a Gilson[®] fractional collector was employed to collect effluents. All our experiments were performed at a room temperature of $19 \pm 1^\circ\text{C}$.

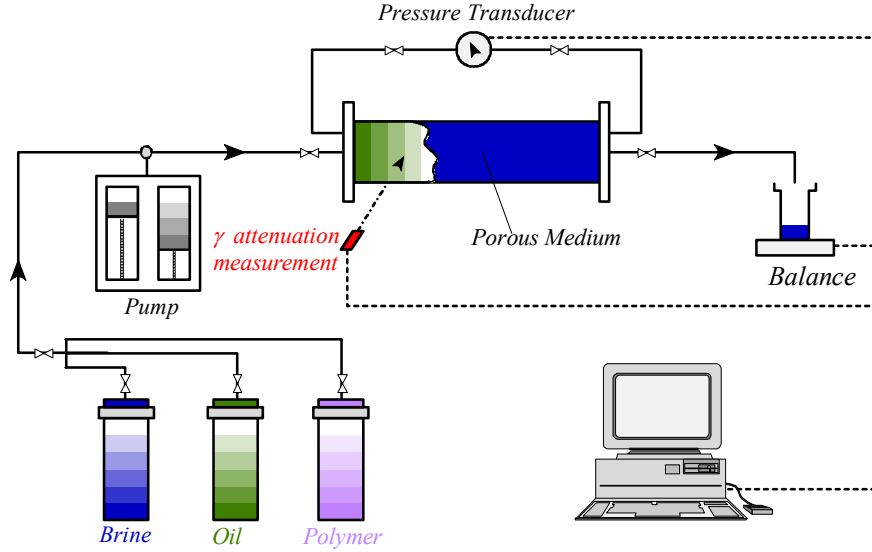


Figure 3. Experimental setup

Experimental procedures

In all our experiments, wettability modification was obtained by aging the core saturated with crude oil at irreducible water saturation, at a temperature of 60°C for 6 weeks.

Wettability modification

The Amott test consists in the following sequences:

- 1) Spontaneous imbibition in brine and measurement of displaced oil volume $V_o^{(1)}$.
- 2) Waterflood at a flow-rate of $3 \cdot 10^{-5} \text{ m}^3/\text{h}$ and measurement of displaced oil volume $V_o^{(2)}$.
- 3) Spontaneous imbibition in oil and measurement of displaced water volume $V_w^{(1)}$.
- 4) Oilflood at a flow-rate of $3 \cdot 10^{-5} \text{ m}^3/\text{h}$ and measurement of displaced water volume $V_w^{(2)}$.

Wettability indices to oil and water are respectively given by the following relationships

$$I_w = V_o^{(1)} / (V_o^{(1)} + V_o^{(2)}) \quad (1)$$

$$I_o = V_w^{(1)} / (V_w^{(1)} + V_w^{(2)}) \quad (2)$$

while the global wettability index of the rock can be written as

$$WI = I_w - I_o \quad (3)$$

Wettability indices I_w and I_o underline the relative importance of spontaneous imbibition versus forced injection for each phase. For a water-wet medium, I_w is close to 1. For an oil-wet medium I_o is close to 1. The global index, WI , is ranging between -1 and $+1$. It is close to -1 for a strongly oil-wet medium and to $+1$ for a strongly water-wet medium.

Table 2. Wettability indices

St Maximin Limestone			Estailades Limestone		
	Before Aging	After Aging		Before Aging	After Aging
I_w	1.	0.04	I_w	0.85	0.04
I_o	0.	0.35	I_o	0.	0.
WI	1.	-0.31	WI	0.85	-0.04

In Table 2, we have reported results obtained on the two types of limestones investigated in this work. Wettability indices indicate that the aging process induced strong changes of the wettability. Both limestones were initially strongly water-wet, the Estailades limestone being less water-wet ($I_w=0.848$) than the St-Maximin limestone ($I_w=1$). Crude oil treatment was found to inverse the wettability of St-Maximin limestone (WI decreased from 1 to -0.316) while on Estailades limestone, the wettability index to water I_w was reduced making the core less water-wet, but not really oil-wet. This brought us to the conclusion that the aging process was an efficient way to modify wettability of these two limestones.

Polymer injection experiments

For each limestone, two sets of experiments were performed, the first one with a W-W core and the second one with a M-W core. For the water-wet cores, imbibition and drainage cycles were run with the following sequences.

- 1) Saturation of the core with brine. Measurement of porosity, ϕ , and absolute permeability to brine, K .
- 2) First oilflood. Measurement of $S_{wi}^{(1)}$ and $K_{ro}^{(1)}@S_{wi}^{(1)}$.
- 3) First waterflood. Measurement of $S_{or}^{(1)}$ and $K_{rw}^{(1)}@S_{or}^{(1)}$.
- 4) Low-salinity brine injection. Measurement of tracer front using γ -ray attenuation on collected effluents.
- 5) Polymer injection. Evaluation of polymer adsorption by measuring effluent viscosity and delay with respect to the tracer.
- 6) Waterflood to displace non-adsorbed polymer from the core.
- 7) Second oilflood. Measurement of $S_{wi}^{(2)}$ and $K_{ro}^{(2)}@S_{wi}^{(2)}$.
- 8) Second waterflood. Measurement of $S_{or}^{(2)}$ and $K_{rw}^{(2)}@S_{or}^{(2)}$.

In the same way, flow tests were performed on M-W samples. The complete experimental procedure, including wettability modification step, was:

- 1) Saturation of the core with brine. Measurement of porosity, ϕ , and absolute permeability to brine, K .
- 2) Drainage with the mineral oil to obtain irreducible water saturation.
- 3) Crude oil injection and aging for 6 weeks at 60 °C.
- 4) Oilflood to displace crude oil.
- 5) First waterflood. Measurement of $S_{or}^{(1)}$ and $K_{rw}^{(1)}@S_{or}^{(1)}$.
- 6) First oilflood. Measurement of $S_{wi}^{(1)}$ and $K_{ro}^{(1)}@S_{wi}^{(1)}$.

- 7) Low salinity brine injection. Analysis of tracer propagation front using γ -ray attenuation.
- 8) Polymer injection. Evaluation of polymer adsorption from the delay of polymer front with respect to the tracer.
- 9) Waterflood to eliminate non-adsorbed polymer from the core.
- 10) Second oilflood. Measurement of $S_{wi}^{(2)}$ and $K_{ro}^{(2)}@S_{wi}^{(2)}$.
- 11) Second waterflood. Measurement of $S_{or}^{(2)}$ and $K_{rw}^{(2)}@S_{or}^{(2)}$.

Since the γ -ray attenuation of a solute depends on its salt concentration, we used this technique on effluents collected during steps 4 and 7 of the above procedures in order to obtain a tracer front profile. The breakthrough of this profile was compared to the polymer front breakthrough providing the possibility to quantify polymer adsorption in the core. Polymer concentration fronts were determined from viscosity measurements performed on effluents collected in steps 5 and 8 of the above procedures. As mentioned above, two sets of experiments, "Estail1" and "Estail2", were performed on Estailades limestones. In the first set ("Estail1"), only one polymer injection was carried out, while in the second set ("Estail2"), two polymer injections were performed, each one followed by a waterflood. This last procedure was used to take into account the excluded pore volume and hence provide a better estimation of the mass of adsorbed polymer (Broseta et al. [12]).

Oilflood and waterflood flow-rates were $1.5 \cdot 10^{-5} \text{ m}^3/\text{h}$ for St-Maximin limestone and $5 \cdot 10^{-6} \text{ m}^3/\text{h}$ for Estailades limestone. These values were chosen in order to be coherent with a field velocity of 20 cm/day. Polymer flow-rates were $1.5 \cdot 10^{-5} \text{ m}^3/\text{h}$ for St-Maximin limestone and $5 \cdot 10^{-6} \text{ m}^3/\text{h}$ for Estailades limestone. Waterflood after polymer injections was run at a high flow rate ($2 \cdot 10^{-5} \text{ m}^3/\text{h}$) in order to clean the core from unabsorbed polymer molecules.

Results and discussion

Polymer adsorption

All our experiments clearly show that polymer adsorption occurs in the different cores, whatever being the wettability conditions. This can be easily seen in Fig. 4 to 9 where a delay of the polymer front with respect to the tracer front systematically occurs. The use of a second polymer front definitely indicates that this delay cannot be attributed to an excluded pore-volume effect only (see Fig. 8 and 9). In fact, when two polymer fronts are performed, a delay of the first polymer front with respect to the second one is always observed. This is indicative of an adsorption process and not of a retention due to mechanical entrapment for instance since this would produce a delay of the second front with respect to the first one.

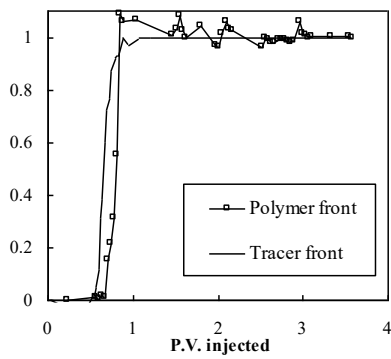


Fig. 4. Tracer and polymer front in W-W St. Maximin limestone

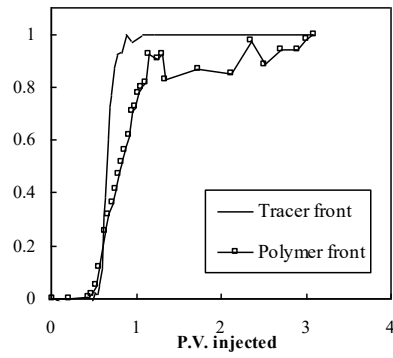


Fig. 5. Tracer and polymer front in M-W St. Maximin limestone

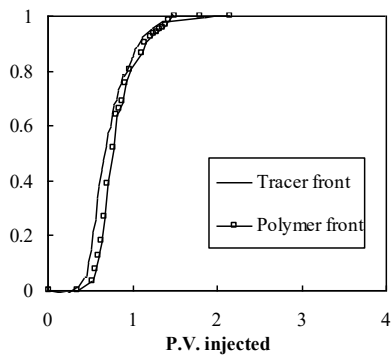


Fig. 6. Tracer and polymer front in W-W Estailades limestone ("Estail1")

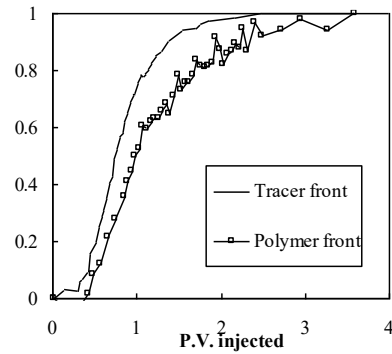


Fig. 7. Tracer and polymer front in M-W Estailades limestone ("Estail1")

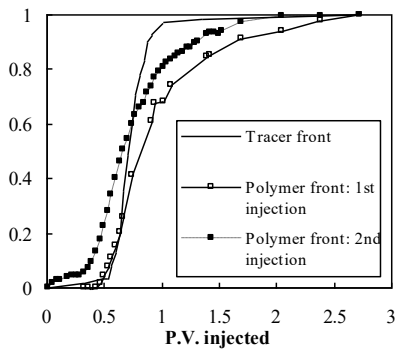


Fig. 8. Tracer and polymer fronts in W-W Estailades limestone ("Estail2")

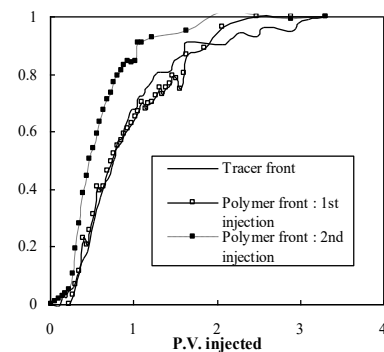


Fig. 9. Tracer and polymer fronts in M-W Estailades limestone ("Estail1")

Another important observation deals with adsorption kinetics, which seems much slower on wettability-modified cores than on water-wet cores. For example, while total adsorption was roughly obtained after injection of two pore volume of polymer solution on the W-W "Estail1" medium (Fig. 6), it was necessary to inject almost four pore volumes of polymer solution into the corresponding M-W medium in order to achieve complete

adsorption (Fig. 7). Although some heterogeneity present in the sample used for wettability modification might also be responsible of such a behavior, it is reasonable to think that the adsorption process is affected by the wettability modification and this is supported by the following. For all wettability-modified cores, additional oil recovery was observed during polymer injection. This phenomenon was not observed for water-wet samples, even though cores were always at residual oil saturation before injecting the polymer solution. This behavior was already observed by Barreau et al.[7] while comparing the injection of a polymer solution in an artificial strongly oil-wet medium obtained by silane treatment on the one hand and in the same medium under its native wettability condition on the other hand. From these observations, different scenarios can be proposed to give a plausible physical interpretation. The first one lies in the fact that polymer adsorption can be made possible by oil desorption giving rise to a small oil production increment. The second one is based on the idea that polymer actually adsorbs directly on oil layers. The third one is based on the idea that only strongly oil-wet local spots at the microscale are responsible of the wettability modification at the macroscale, leaving enough hydrophilic sites for polymer molecules to adsorb and actually form a layer. In all cases, it seems clear that adsorption of hydrophilic polymer that did occur in our oil or intermediate wet limestones, restores part of the water-wet character of the medium. From our data, PAM adsorption on “Estail2” limestone is 42 $\mu\text{g/g}$ for the W-W medium and 69 $\mu\text{g/g}$ for the M-W medium. This result indicates that polymer adsorption seems to increase with a decreasing wettability index. However, more experimental work is required to confirm this result.

Relatives permeabilities

For each core, the absolute permeability, K , to brine was the reference value used in the determination of relative permeabilities. End point data before and after polymer injection are reported in Tables 3, 4 and 5. From these data, water and oil permeability reductions, R_{kw} and R_{ko} , were calculated at the same saturation by assuming that permeabilities are linear functions of the water saturation. These values are also reported in the following tables.

Table 3. St. Maximim end point data

		S_{wi}	k_{ro}	S_{or}	k_{rw}	R_{kw}	R_{ko}
W-W	Before polymer	0.482	0.636	0.19	0.209	2.3	1.4
	After Polymer	0.490	0.454	0.187	0.093		
M-W	Before polymer	0.450	0.386	0.190	0.108	1.5	1.
	After Polymer	0.480	0.339	0.2	0.069		

Table 4. “Estail1” end point data

		S_{wi}	k_{ro}	S_{or}	k_{rw}	R_{kw}	R_{ko}
W-W	Before polymer	0.426	0.851	0.29	0.059	3.	1.6
	After Polymer	0.400	0.633	0.300	0.020		
M-W	Before polymer	0.476	0.816	0.148	0.051	1.75	1.3
	After Polymer	0.600	0.470	0.090	0.051		

Table 5. “Estail2” end point data

		S_{wi}	k_{ro}	S_{or}	k_{rw}	R_{kw}	R_{ko}
W-W	Before polymer	0.51	0.818	0.262	0.071	5.2	1.3
	After Polymer	0.52	0.601	0.232	0.011		
M-W	Before polymer	0.57	0.33	0.27	0.066	2.6	1.3
	After Polymer	0.555	0.252	0.262	0.025		

Important results are obtained from these experiments.

First, a selective effect of adsorbed polymer is observed on all the cores, whatever the wettability conditions. In fact, the adsorbed polymer affects much more water permeability than oil permeability since R_{K_o} values are ranging from 1. to 1.6 while R_{kw} ranges between 1.5 and 5.2. This first observation is consistent with the wall effect interpretation in combination with the fact that polymer adsorption restores part of the water-wet character of M-W media.

Second, while R_{K_w}/R_{K_o} is 1.4 for the W-W St Maximin limestone for which absolute permeability is $1.62\mu\text{m}^2$, it is around 4 for W-W “Estail2” limestone which is roughly 9 times less permeable, confirming that the less the absolute permeability to brine, the more significant the DPR effect. This result is also in perfect agreement with the physical interpretation of the DPR by the wall effect (Barreau et al. [9]).

Finally, our results clearly indicate that, when wettability of the core is modified, making the core less water-wet, water permeability reduction decreases slightly while oil permeability reduction keeps constant. In that way, DPR effect obtained by polymer adsorption seems less pronounced when rock wettability to water decreases. Since significant modification of the pore structure by aging is most unlikely, this last behavior can not be directly attributed to the wall effect. For this reason, a scenario of polymer invasion associated to oil and water distribution conditioned by the wettability of the medium is proposed in the following.

Two-phase occupancy at the pore level.

For sake of clarity and without any particular restriction, we illustrate our interpretation on the basis of a porous medium having a pore size distribution that obeys a gaussian law (Fig. 10). For simplicity, we distinguish three main classes of pores on this distribution: small, intermediate and large ones. In a water-wet medium, initially saturated with water and flooded by oil (drainage process), irreducible water remains in small pores while oil invades all the other ones. If the medium is now water-flooded (imbibition process), residual oil saturation mainly results from trapping in the large pores. The situation can easily be transposed in the case of a strongly oil-wet medium i.e. S_{wi} due to water trapping in large pores and S_{or} to oil trapping in small ones (Anderson, [13]).

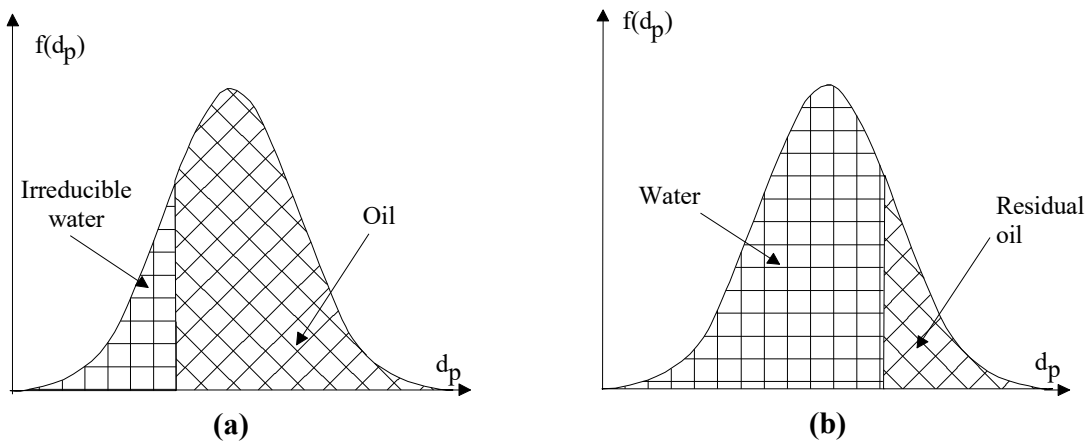


Fig.10. Pore occupancy of a water-wet porous medium (a) after drainage; (b) after imbibition

If the porous medium is neither strongly water- nor oil-wet but of intermediate, heterogeneous or fractional wettability, the two-phase pore occupancy is quite different. In fact, in this case, irreducible water and residual oil are distributed among all the pore population (Fig. 11) depending on local wettability.

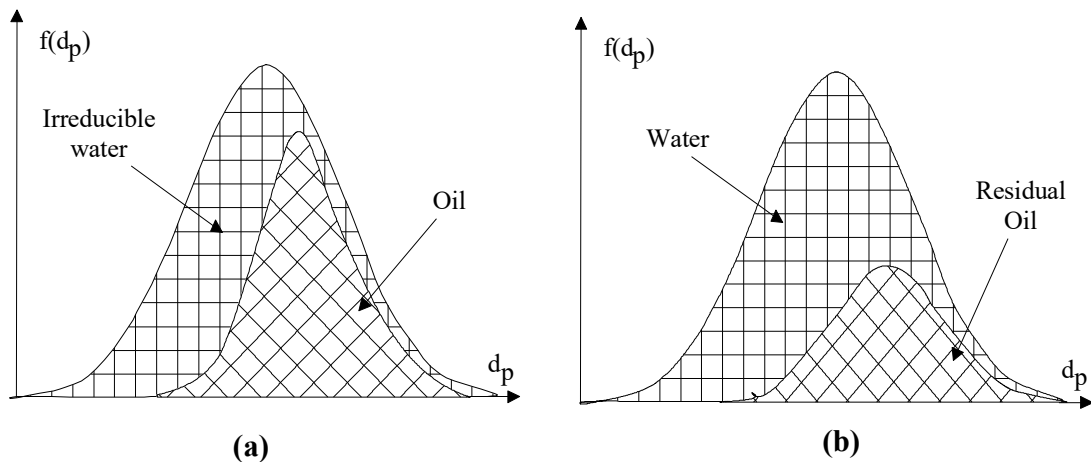


Fig.11. Pore occupancy of an intermediate-wet porous medium: (a) after water flooding; (b) after oil flooding

In our particular case, two situations are to be compared: initial W-W media and similar M-W media resulting from aging.

In all experiments, polymer was injected in the presence of residual oil saturation and we assume that polymer preferentially flows in connected pores occupied by water. For W-W media, the polymer solution invades pores of small and intermediate size, and adsorption does not occur in the larger ones occupied by oil (Fig. 10 (b)). For M-W media, the situation is quite different. Because aging was performed on initial water-wet cores at their irreducible water saturation, smaller pores are preserved from wettability modification. After aging, waterflood leaves residual oil trapped in intermediate and larger pores (Fig. 11 (b)) and water is present in almost all the pore-size range: in the smaller from the original stage as well as in the intermediate and larger ones due to wettability modification. Under these circumstances, polymer adsorption occurs in almost all pores and in particular in large ones that are not accessible to the polymer solution in the W-W case. In other words, adsorption occurs in larger pores as the global wettability index decreases. Combining this scenario with the fact that permeability reduction decreases with increasing permeability -i.e. for larger pores- provides a plausible physical interpretation of the trend observed for the three types of cores which experimental data are reported in Tables 3, 4 and 5.

Conclusions

Effects of wettability on two-phase flow modifications by polymer adsorption were investigated in this work from an experimental point of view. Our results lead to the following conclusion:

- 1) crude oil aging was found to modify wettability of water-wet limestone cores. Native water-wet Estailades limestone became less water-wet, while a wettability inversion was observed for St-Maximin limestone. Reservoir wettability conditions could be reasonably reproduced by using such a treatment.
- 2) adsorption of the hydrophilic polymer in partially oil-wet cores seems to indicate that the polymer restores part of the initial water wettability that was reduced by crude oil aging treatment.
- 3) polymer injection in limestone under different wettability conditions, from strongly water-wet to weakly oil-wet, resulted in a DPR effect. This effect increased when the absolute permeability of the rock decreases.
- 4) the DPR effect was less pronounced in mixed wet conditions than in strongly water-wet media. This behavior can be consistently explained by taking into account phase distribution at the pore-scale according to the wettability of the medium.

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