

Efficiency of diffusion controlled miscible displacement in fractured porous media

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Received: 16 November 2006 / Accepted: 30 March 2007 / Published online: 3 May 2007
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Abstract Experiments were performed to study the diffusion process between matrix and fracture while there is flow in fracture. 2-inch diameter and 6-inch length Berea sandstone and Indiana limestone samples were cut cylindrically. An artificial fracture spanning between injection and production ends was created and the sample was coated with heat-shrinkable teflon tube. A miscible solvent (heptane) was injected from one end of the core saturated with oil at a constant rate. The effects of (a) oil type (mineral oil and kerosene), (b) injection rates, (c) orientation of the core, (d) matrix wettability, (e) core type (a sandstone and a limestone), and (f) amount of water in matrix on the oil recovery performance were examined. The process efficiency in terms of the time required for the recovery as well as the amount of solvent injected was also investigated. It is expected that the experimental results will be useful in deriving the matrix–fracture transfer function by diffusion that is controlled by the flow rate, matrix and fluid properties.

Keywords Naturally fractured reservoirs · Enhanced oil recovery · Miscible displacement · Matrix–fracture interaction · Molecular diffusion

1 Introduction

A large proportion of the world's proven oil reserves have been found in naturally fractured reservoirs (NFRs). During the injection of fluids that are miscible with oil for enhanced oil recovery, the transport of the injectant and the oil recovery are controlled by fracture and matrix properties in this type of reservoirs. For such systems, the transfer between matrix and fracture due to diffusion is a significant-oil recovery mechanism. Similar processes can be encountered during the sequestration of greenhouse gases, and the transport of contaminants in subsurface reservoirs. Understanding the effects of different parameters on the dynamics

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of the transfer due to diffusion is essential in modeling such processes. In fact, the description of matrix fracture interaction for dual-porosity dual-permeability models developed for NFRs is still a challenge.

Oil recovery mechanisms of fractured systems have been studied at laboratory scale since 1970's. [Thompson and Mungan \(1968\)](#) compared the displacement velocity to critical velocity (V_C) and showed its effect on recovery efficiency. Using the results of single matrix block analytical studies and multicomponent laboratory experiments, [Da Silva and Belery \(1989\)](#) confirmed the importance of molecular diffusion in NFRs and concluded that it may override the other hydrocarbon displacement mechanisms. [Firoozabadi and Markeset \(1994\)](#) showed the effect of matrix/fracture configuration and fracture aperture on first contact miscible efficiency. Matrix fracture interaction in fractured rocks for different types of fluids was investigated computationally ([Zakirov et al. 1991](#); [Jensen et al. 1992](#)) and experimentally ([Hujun et al. 2000](#); [Le Romancer et al. 1994](#)) in different studies. [Saidi \(1987\)](#) studied the diffusion/stripping process in fractured media. [Morel et al. \(1990\)](#) performed diffusion experiments with chalk and studied the effect of initial gas saturation. Analytical and numerical solutions for the diffusion process in the fracture and transport to the matrix are also available ([Hu et al. 1991](#); [Lenormand et al. 1998](#); [LaBolle et al. 2000](#); [Jamshidnezhad et al. 2004](#); [Ghorayeb and Firoozabadi 2000](#)).

More recently, static experiments were reported on the diffusion process from fracture to matrix ([Babadagli et al. 2005](#); [Rangel-German and Kovscek 2000](#); [Hatiboglu and Babadagli 2006](#)). There are also experimental methods for calculating the diffusion coefficients between two fluid systems ([Yang and Gu 2003](#); [Civan and Rasmussen 2002](#); [Raizi 1996](#); [Stubos and Poulou 1999](#)). But the matrix–fracture transfer by diffusion in fractured porous media depends on the conditions at the boundaries and fracture geometry as well as flow conditions ([Lenormand et al. 1998](#)). The oil recovery mechanism by gas injection into fractured porous media is governed by convection, dispersion and diffusion. Most mixing (dispersion) is mainly caused by adjacent rock block (rock matrix), variations in velocity due to fracture roughness, mixing at fracture intersections, variations in velocity due to differing scales of fracturing, and the variations in velocity due to variable fracture density. Recovery in fractured reservoirs requires the determination of transfer parameters between fracture and matrix, called transfer functions. The theoretical derivations of such transfer functions are available in literature ([Babadagli and Zeidani 2004](#); [Civan and Rasmussen 2005](#)).

In general four factors contribute to crossflow/mass transfer between matrix–fracture:

- Pressure drive
- Gravity drive
- Dispersion/diffusion drive
- Capillary drive

When the bypassed fluid and displacement fluids are first contact miscible (FCM), there is no capillary crossflow. When the fluids are multicontact miscible (MCM) or immiscible, there can be some capillary-driven crossflow. [Burger et al. \(Burger and Mohanty; Burger et al. 1996\)](#) found that capillary driven crossflow does not contribute significantly to mass transfer in near-miscible hydrocarbon floods. The crossflow/mass transfer between fracture and surrounding matrix block during miscible displacements needs more clarifications. The current work was done at room temperature and pressure conditions; hence the pressure drive was also out of focus.

We specifically studied the effects of flow rate, matrix, fracture, and fluid properties during the recovery of oil mainly under the influence of dispersion/diffusion drive.

Table 1 Properties of displacing and displaced fluids

Properties	Displacing fluid	Displaced fluid	
	Heptane	Mineral oil	Kerosene
Density (g/cc)	0.69	0.83	0.81
Viscosity (cp)	0.410	33.5	2.1
Refraction Index	1.3891	1.469	1.475

Table 2 Detailed outline of experiments conducted

Case	Core type	Fracture orientation	Oil type	Aged	$S_{wi}(\%)$	Flow rate (ml/h)
BSH-3	Berea Sandstone	H	MO	No	0	3
BSH-6	Berea Sandstone	H	MO	No	0	6
BSH-9	Berea Sandstone	H	MO	No	0	9
BSH-3A	Berea Sandstone	H	MO	Yes	0	3
BSH-4.5A	Berea Sandstone	H	MO	Yes	0	4.5
BSH-6A	Berea Sandstone	H	MO	Yes	0	6
BSH-7.5A	Berea Sandstone	H	MO	Yes	0	7.5
BSH-3K	Berea Sandstone	H	Kerosene	No	0	3
BSH-6K	Berea Sandstone	H	Kerosene	No	0	6
BSV-1	Berea Sandstone	V	MO	No	0	1
BSV-3	Berea Sandstone	V	MO	No	0	3
BSV-6	Berea Sandstone	V	MO	No	0	6
BSH-3W	Berea sandstone	H	MO	No	>0	3
BSH-6W	Berea sandstone	H	MO	No	>0	6
ILH-3	Indiana Limestone	H	MO	No	0	3
ILH-6	Indiana Limestone	H	MO	No	0	6

B – Berea

S – Sandstone

I – Indiana

L – Limestone

H – Horizontal

V – Vertical

A – Aged over period of 1 month

W – Water imbibed for primary recovery

MO – Mineral Oil

2 Experimental Method

A series of experiments were conducted by injecting solvent into fractured cores. Experimental procedure is given in Tables 1 and 2. Parameters investigated in this study are as follows:

- Flow rate (injection rate) in the fracture,
- Gravity effect,
- Water saturation in the matrix,
- Rock type,
- Effect of aging (wettability),
- Effect of viscosity ratio.

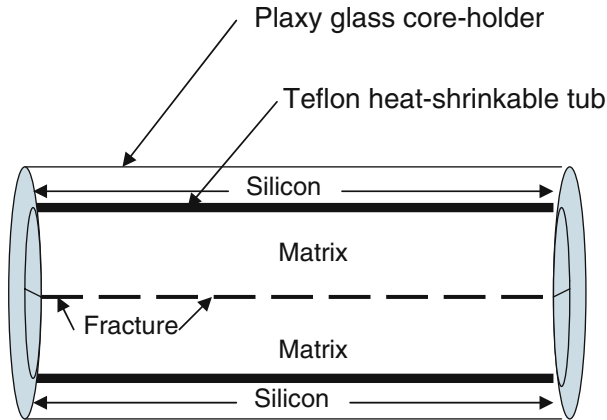


Fig. 1 Core-holder design

2.1 Porous media

Different samples representing two characteristic sedimentary rocks were used to study the effect of the rock type: (1) Berea sandstone (average permeability = 500 md and average porosity = 20%) and (2) Indiana limestone (average permeability = 15 md and average porosity = 11%). For porosity measurement, core samples were weighted and placed in a desiccator filled with oil. The desiccator was connected to a vacuum pump. The core was saturated under constant vacuum for 48–72 h. The weight of the core after saturation was measured and the porosity of the matrix was calculated from the difference between the weight of the saturated and unsaturated core. The porosity of Berea sandstone cores ranges from 19% to 21% and that of Indiana limestone cores is from 10% to 12%. The permeability of the Berea sandstone and Indiana limestone cores were calculated using the data obtained from constant rate water injection experiments and the Darcy equation. Experiments were conducted on a few samples from the same block and the average values were obtained as 500 md and 15 md for Berea sandstone and Indiana limestone, respectively.

2.2 Fluids

Two different oil types, namely light mineral oil and kerosene, were used as displaced fluids. Heptane was used as the—miscible—displacing fluid. The properties of fluids are given in Table 1.

2.3 Test conditions

All tests were performed at room-temperature (60°F) and atmospheric pressure. Solvent was injected at a constant rate while the oil was produced at atmospheric pressure.

2.4 Core preparation and test procedure

The core samples and the experimental set-up are presented in Figs. 1–3. The cores are cylindrical samples, 6 inches in length and 2 inches in diameter (Fig. 1). Cores were cut through the center in direction of longitudinal axis. They cores were weighted and fully saturated with oil

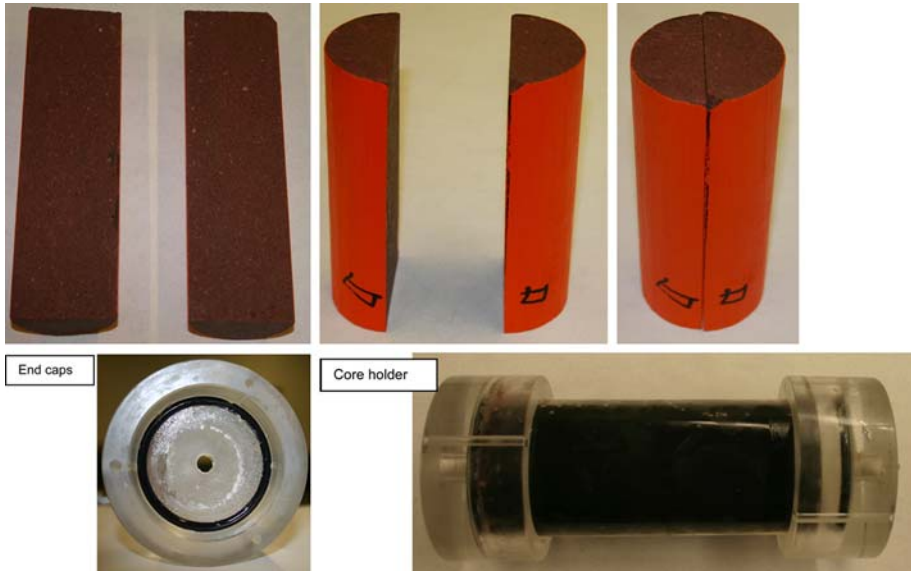


Fig. 2 Core samples cut for fracture creation and the core-holder used

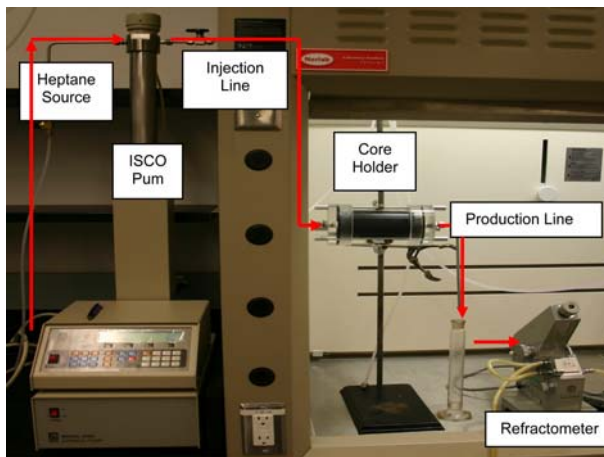
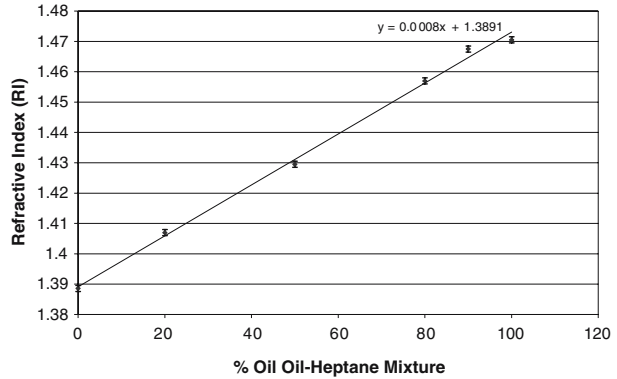


Fig. 3 Experimental set-up

under constant vacuum using vacuum pump for 48–72 h to achieve maximum saturation. The weight of the cores after saturation was measured. Porosity was obtained from the weight difference before and after saturation. The two pieces were then held together tight with heat-shrinkable rubber sleeves. The fractured core packed with rubber sleeve was placed into a Plexiglas core-holder of 6 inches length and 2.5 inch diameter. The annular area between core and core-holder was filled with silicon. This was done to ensure no flow between the core and the core-holder. The silicon used was industry-grade liquid silicon, which solidifies by adding curing agent with time.

After core preparation, the injection and producer ports were placed at the centers of both ends of the core-holder to ensure the injection through the fracture directly (Fig. 2). The

Fig. 4 Calibration chart for refractometer for mineral oil–heptane mixture



same core preparation method was used in all experiments. Note that each core was used only once to avoid the property change due to cleaning procedure. The cores are assumed to have similar properties as they were plugged out from the same block. The list of the experiments conducted in this study is given in Table 2.

Heptane was continuously injected through the fracture at a constant rate. The flow rate and the cumulative volume injected were monitored using ISCO-500 D pumps. The effluent was collected and analyzed with a refractometer (Fig. 3).

The permeability contrast between matrix and fracture was kept as high as possible to ensure that the viscous flow dominates only in the fracture and the matrix oil is recovered only by diffusion. No overburden was applied on the cores and the average value of fracture permeability on the cores was measured as around 8–10 Darcies. This value yields enough contrast between matrix and fracture permeabilities for viscous flow to be effective only in the fractures.

2.5 Measurement technique

Because of the small density difference between oil and solvent used here, the need of a different technique which does not depend on density for determining the composition of the produced liquid raised. Measurement of refractive index (RI) was used to determine the relative proportions of the mixture from the production end. The ratio of the speed of light in air to the speed of light in another media is called “refractive index” (RI). A bench-type Refractometer supplied by Fisher Scientific was used. The minimum reading of scale indicates down to 4 places of decimal of the RI values. The accuracy is ± 0.0002 refractive units. Proper care was taken to conduct all the readings at same temperature to avoid any effect of temperature change on the final reading. The RI values for the pure compounds were measured and a calibration curve was generated with the known percentage of oil mixed with heptane. This curve is presented in Fig. 4. Using this curve composition of the produced liquid was determined.

2.6 Effect of water saturation

Most of the oil reservoirs at present state are water flooded. Sometimes the percentage of water present is higher than the oil. Considering this fact, we decided to create a similar situation. Two (100%) oil-saturated Berea sandstone cores, packed in heat-shrinkable Teflon sleeve, were prepared in a similar way as the other experiments. These cores were exposed to

water in order to recover some amount of oil by spontaneous imbibition as if the reservoir was under waterflooding. The recovery was monitored against time. When oil production through imbibition reached plateau, the cores were taken out and placed into Plexiglas core-holder as discussed above, as similar to the other experiments for solvent injection. Note that this is a secondary -miscible- recovery after a primary waterflooding. The solvent injection rates applied for these water imbibed cores were 3 ml/h and 6 ml/h.

2.7 Effect of aging

Aging the rock with oil over a period of time may change the wettability of the rock type and may cause considerable effect on oil recovery process. Though there is no water in the cores, aging over longer period of time changes the affinity as well as the contact angle of oil to the rock. To study this effect, four Berea sandstone cores were aged in mineral oil over one month. The experimental results as well as the imbibition tests reported in other papers (Babadagli and Ershaghi 1993; Babadagli 2000; Trivedi and Babadagli 2006) indicated that the longer contact time altered the wettability of the rock to mineral oil. Although the polar components are not as high as in crude oils, the mineral oil we used showed change in the affinity to oil after aging. Solvent was injected into these aged samples at the flow rates of 3, 4.5, 6 and 9 ml/h.

2.8 Effect of viscosity ratio/density

In order to analyze the effects of viscosity and density of the non-wetting phase on the matrix–fracture diffusion process, we conducted some experiments with kerosene in addition to mineral oil. Cores were saturated with kerosene under vacuum and prepared in a similar way as the other experiments.

2.9 Effect of gravity

Gravity also contributes to recovery, mostly when the fracture orientation is vertical. In our work, we studied the recovery from three samples with vertical orientation of fracture. After preparing the cores following the same procedure, we mounted them vertically and injected solvent at three different rates of 1, 3 and 6 ml/h from the top of the sample downward to the bottom. For these experiments, we used only mineral oil as the displaced phase.

2.10 Results and analysis

The recovery curves for the above mentioned experiments are presented in Figs. 5–8 as pore volume injected vs. pore volume recovered. Here, recovery of oil was used as *y*-axis. In a sense, this is the solvent concentration in the system. We used heptane as solvent to mimic miscible CO₂ displacement in fractured oil reservoirs. As our preliminary target was oil recovery, we preferred this type of representation in *y*-axis rather than solvent concentration.

With capillary drive and pressure drive not affecting the recovery and gravity forces minimized as the orientation of the cores was horizontal, the only mechanism governing the process could be diffusion/displacement drive. The diffusion dominant process yields slower recovery of oil with a delayed solvent breakthrough when the solvent injection rate was low, but it results in higher ultimate recovery for the horizontal mineral oil-Berea sandstone cases (Fig. 5). When comparing the recovery trends for lower (BSH-3) and higher (BSH-6) rates, BSH-6 shows faster initial recovery compared to BSH-3. But after certain period of time, the BSH-3 case overpasses the production as an evidence of diffusion controlling mechanism.

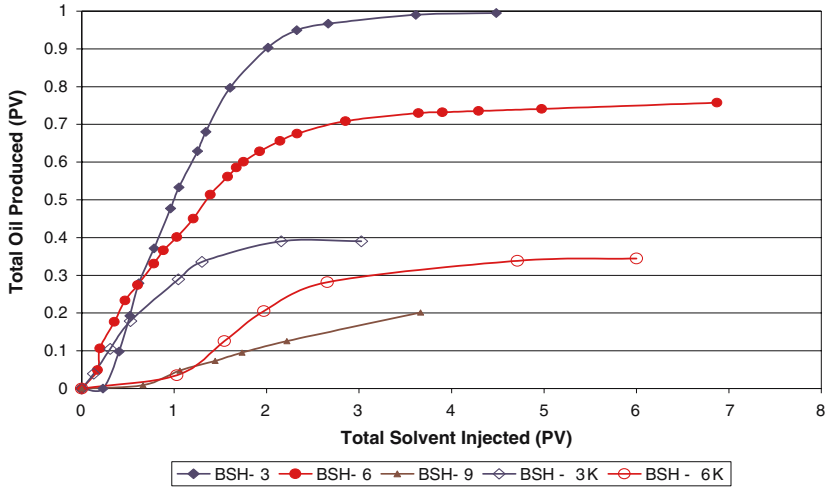


Fig. 5 Cumulative production with solvent injected (PV) for Berea sandstone horizontal orientation

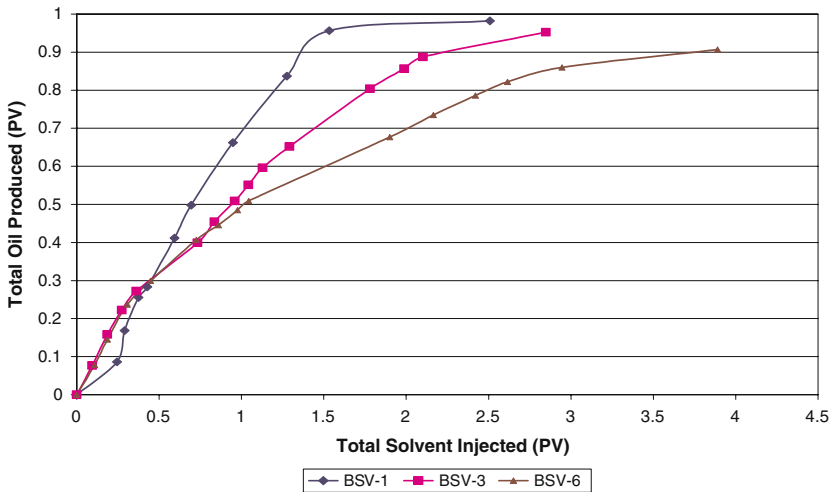


Fig. 6 Cumulative production with solvent injected (PV) for Berea sandstone vertical orientation

It is also evident that during the initial phase of the recovery, the displacement was driven by diffusion between fracture and matrix for lower rates rather than viscous flow in fracture. Hence, 3 ml/h flow rate overrides the recovery of 6 ml/h flow rate. Performance of kerosene was also included in this plot. The recovery rate and ultimate recovery for the kerosene case is remarkably lower than those of mineral oil cases for the same injection rate. The effect of viscosity and oil type on this process needs more detailed study and this part of research is on-going.

The recovery rate behavior for the cases of solvent injection with vertical orientation of Berea sandstone cores is very similar to that of the horizontal case. But the ultimate recoveries for all vertical orientated cores turned out to be the same despite different solvent injection rates (Fig. 6), unlike the horizontal orientation. The gravity drainage mechanism might have

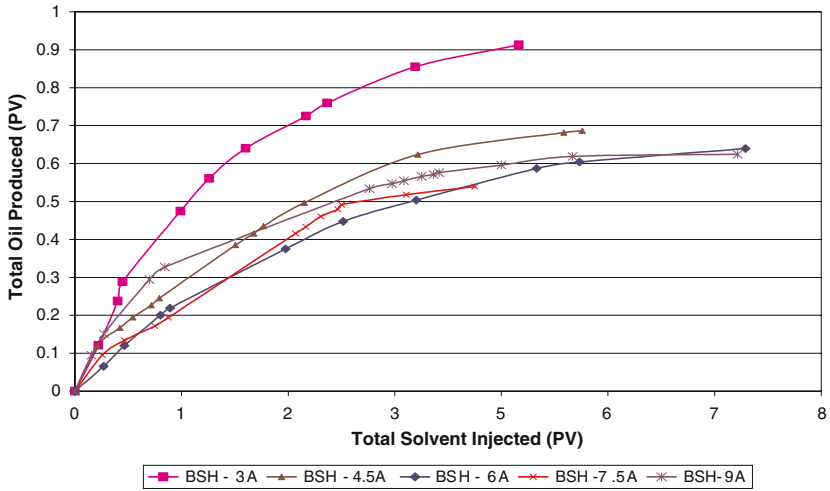


Fig. 7 Cumulative production with solvent injected (PV) for aged Berea sandstone horizontal orientation

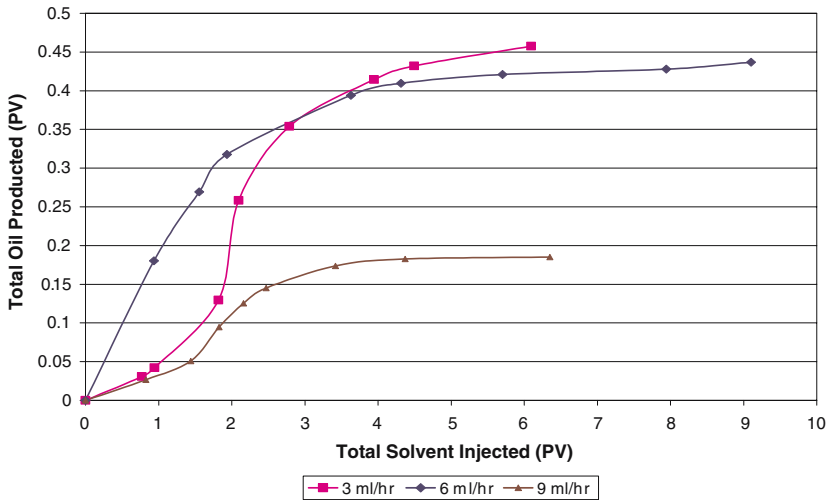


Fig. 8 Cumulative production with solvent injected (PV) for Indiana limestone horizontal orientation

played a role in the oil recovery process. Similar to the horizontal Berea sandstone cases, the recovery curve at lower solvent injection rate overpasses the one at higher solvent injection rate after a certain period of time for the cores with vertical orientation, i.e., BSV-1, BSV-3 and BSV-6. Initial difference in the recoveries is not much compared to the difference between the two equivalent horizontal cases (BSH-3 and BSH-6). Still at a certain point, the recovery of BSV-1 curve overpasses the recovery curves of higher solvent injection rate cases (BSV-3 and BSV-6). The location of this crossover point occurs almost at the same time as noticed during horizontal experiments.

In the case of limestone experiments, the recovery trends with 3 ml/h and 6 ml/h injection rates are almost similar to the Berea sandstone cases (Fig. 8). But the ultimate recoveries are very low being almost half to that of Berea sandstone experiments.

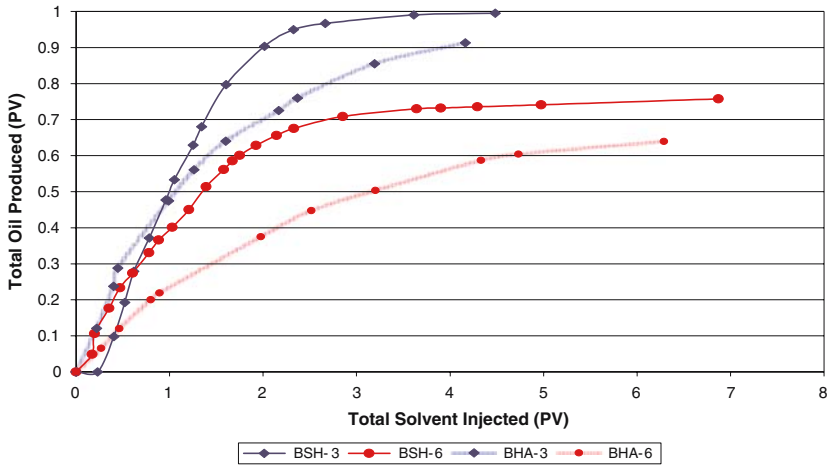


Fig. 9 Cumulative production with solvent injected (PV) comparing aged and unaged Berea sandstone horizontal orientation

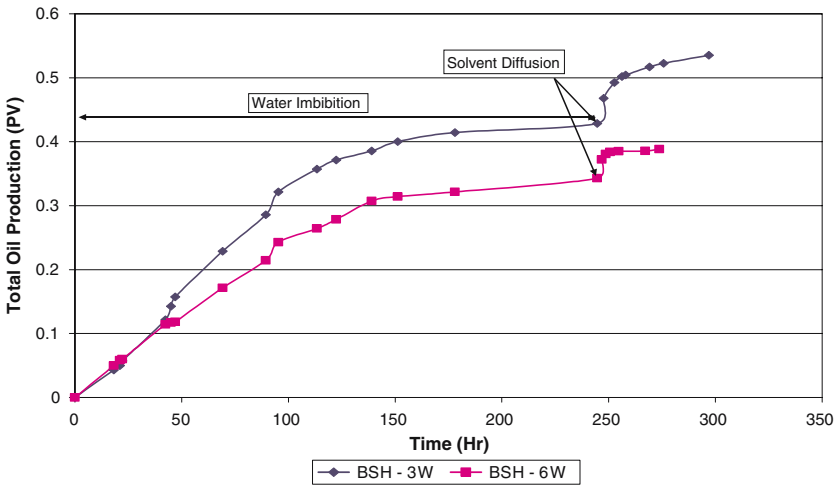


Fig. 10 Cumulative production with time for water imbibed cores

With the aged samples of Berea sandstone cores, the recovery is lower than the same cores without aging (Fig. 9). For the cases of BSH-3A and BSH-6A, aging has a negative effect on miscible displacement efficiency compared to cases BSH-3 and BSH-6 (Fig. 7). It is notable that slowest solvent injection rate (3 ml/h) yielded the highest ultimate recovery. Similar recovery trends and ultimate recoveries were obtained from all other four rates.

The recovery curves for the previously depleted matrix cases by water imbibition are shown in Fig. 10. The BSH-3W case showed higher ultimate recovery than BSH-6W. The ultimate recovery after solvent injection into previously water flooded cores were only 55%, much lower than the one without water flooded (95%). This indicates that starting the process by solvent injection (diffusion) results in much higher ultimate recovery than injecting solvent into already waterflooded cases for water-wet samples.

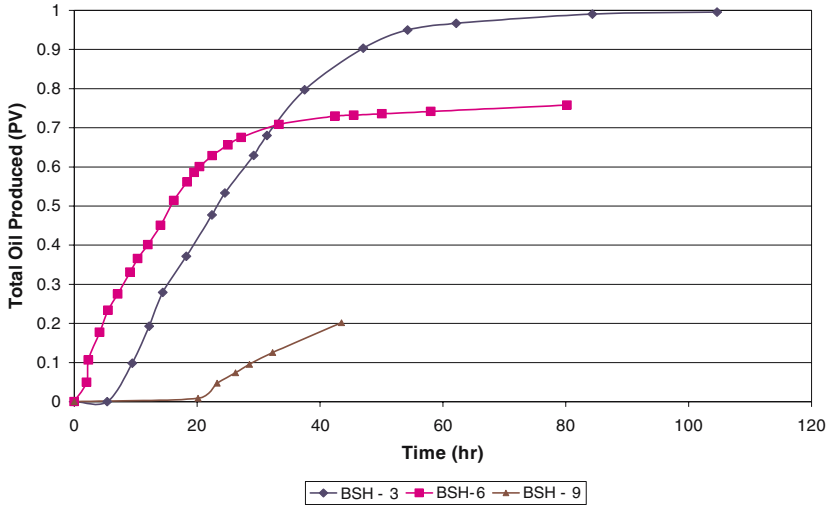


Fig. 11 Cumulative production with time for Berea sandstone horizontal orientation

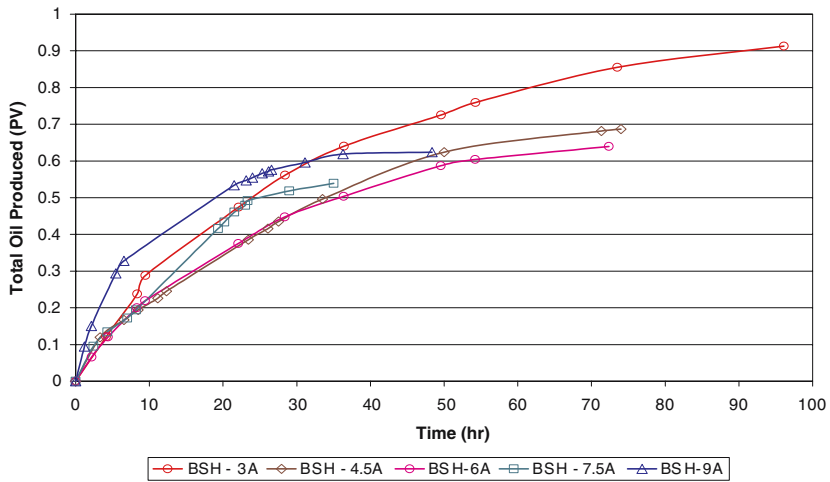


Fig. 12 Cumulative production with time for aged Berea sandstone horizontal orientation

It is clear that the injection rate is a critical parameter as well as matrix and fluid properties on the process. Higher injection rates resulted in earlier breakthrough and less residing time in order for the matrix–fracture interaction due to diffusion to take place. This was more significant for some cases and it even affected the ultimate recovery from the matrix. Therefore, it is essential to define a critical rate as a function of matrix and fluid properties. This can be done in two different ways depending on what type of efficiency is considered. For example, if the critical parameter in the efficiency of the process is the amount of oil produced per amount of solvent injected, one can define a critical rate (Babadagli and Ershaghi 1993). If the critical parameter is the time to complete the process rather than the amount of injected fluid, then one can define an optimal injection rate as suggested by a previous study for water imbibition transfer between matrix and fracture (Babadagli 2000).

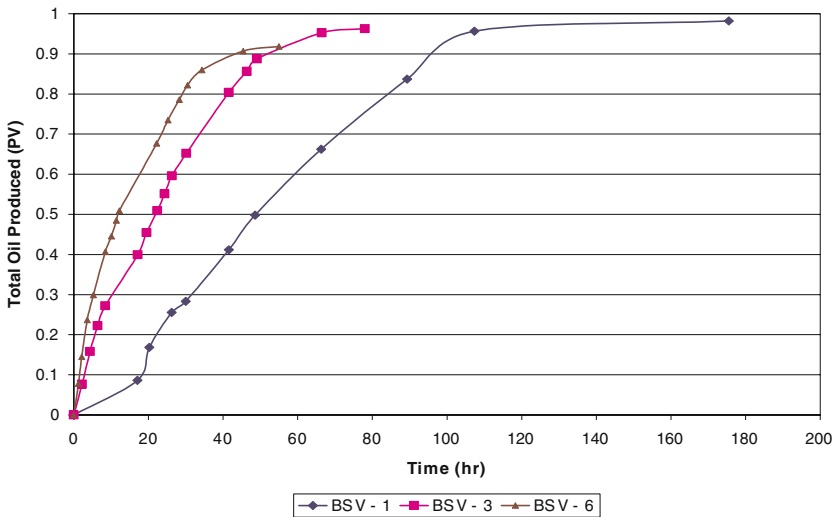


Fig. 13 Cumulative production with time for Berea sandstone vertical orientation

2.11 Efficiency analysis

In many enhanced oil recovery processes including miscible processes, the amount of solvent injected per oil recovery is the critical parameter. However, oil recovery rate, i.e., process time required to reach the ultimate recovery could also be important because the time value of money might offset the cost of extra injected solvent. Therefore, the time to reach the ultimate recovery could be equally important parameter with the amount of solvent injected in certain circumstances, especially when the oil prices are high. Hence, the efficiency of the process was analyzed with respect to the diffusion time as well. For this exercise, the amount of oil produced was plotted against time instead of total amount of solvent injected. The plots are given in Figs. 10–13. Using those plots, the amount of total oil produced (PV) was plotted against the flow rate at different times. Figures 14–16 show the optimal rate for maximum oil recovery at different diffusion time scale. The optimal rate was observed to be around 6 ml/h for the sandstone and limestone horizontal experiments without aging (Figs. 14 and 15). At the end of the process after 40–50 h, the slower rate (3 ml/h) overpasses the production obtained from the 6 ml/h case. This indicates the dominance of the diffusional flow (matrix–fracture interaction) compared to the viscous flow (in fracture). One can conclude from these results that, for faster and higher ultimate recovery, the process should be operated at 6 ml/h at the start and should be switched to 3 ml/h. Figure 16 suggests that there is no optimal rate for the aged sample as they all followed a similar recovery trend as given in Fig. 12 regardless the rate. Interestingly enough, the ultimate recoveries of aged samples vary significantly compared to that obtained from unaged samples.

3 Conclusions

Dominance of the phase diffusion into matrix through fracture over viscous flow in the fracture was shown in this study. Though the higher solvent injection rate yields high production rate of oil in the initial period of the project life, considering the long run, the low rate solvent

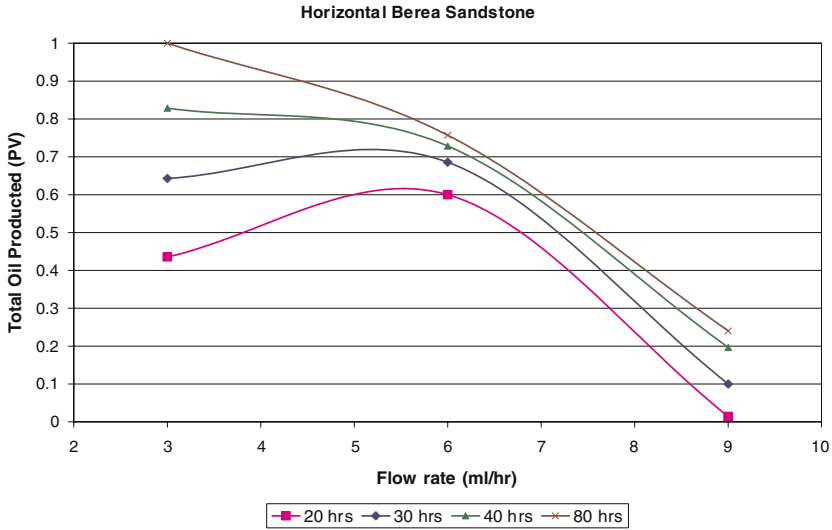


Fig. 14 Cumulative production at different flow rate and time axis for Berea sandstone horizontal orientation

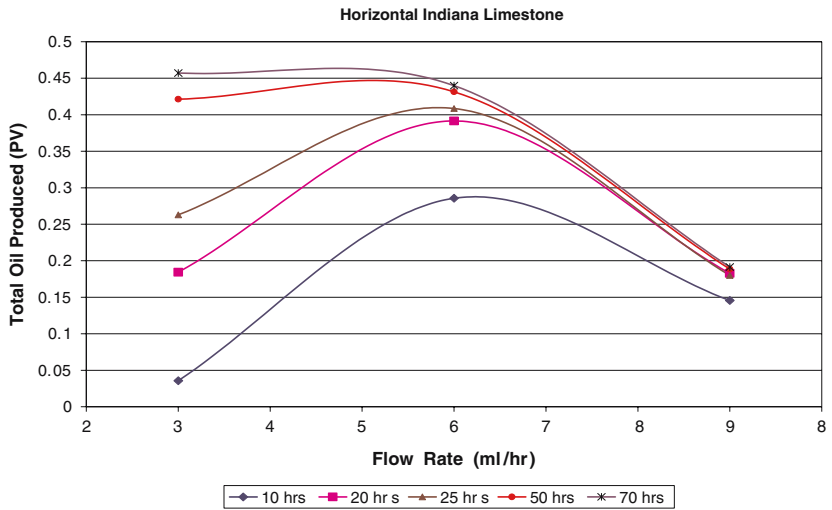


Fig. 15 Cumulative production at different flow rate and time axis for Indiana limestone horizontal orientation

injection strategy is the best with most of the production contribution from matrix through the diffusion process. Solvent injected at lower rates has more time to diffuse into matrix in transverse direction before it breaks through. This results in higher ultimate recovery than that of higher rate solvent injection.

With some water existing in the rock from previous waterflooding, one can still obtain oil recovery from matrix by the diffusion process. Note that the ultimate recovery obtained from the waterflooding followed by the solvent diffusion process would be much lower than the cases with solvent diffusion only for water-wet rocks.

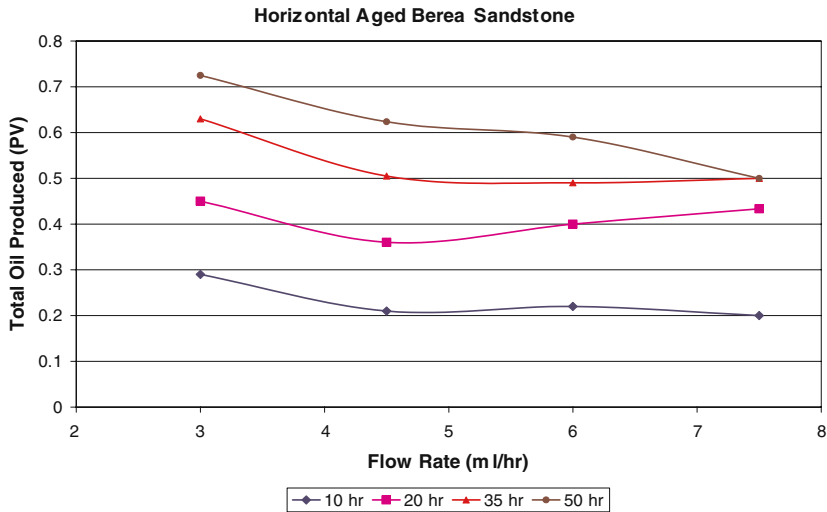


Fig. 16 Cumulative production at different flow rate and time axis for aged Berea sandstone horizontal orientation

The efficiency of the process was investigated using two different parameters, oil recovery against the amount of solvent injected and time. When the amount of oil produced per time is the critical-efficiency parameter, a rate of 6 ml/h was found to be the optimal rate for almost all the cases except the aged samples. This approach for efficiency analysis is useful for enhanced oil recovery applications.

In this study, we have not varied two critical parameters; the size of the matrix (length or width) and oil type (mainly for viscosity variation). Further attempts were made in subsequent studies where the process has been theoretically modeled using the experimental observations and considering those parameters (Trivedi and Babadagli 2006, 2007). Also, this study focused on first contact miscibility. The multicontact miscible diffusion at reservoir temperature and pressure is more complex. This case requires more effort and could be critical especially in CO₂ injection for EOR and sequestration.

Acknowledgements This study was partly funded by an NSERC Grant (No: G121210595) and FSIDA (University of Alberta, Fund for Support of International Development Activities) Grant (#04-140). The funds for the equipment used in the experiments were obtained from the Canadian Foundation for Innovation (CFI, Project #7566) and the University of Alberta. We also would like to thank Sean Watt for providing his technical expertise in preparing core-holder design. This article is the revised and improved version of SPE 100411 presented at the 2006 SPE Western Regional/AAPG Pacific Section/GSA Cordilleran Section Joint Meeting held in Anchorage, AL, 8–10 May.

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