

Experimental Investigation on Asphaltene Deposition in Porous Media During Miscible Gas Injection

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ABSTRACT: *In this work the likelihood of asphaltene deposition problems during dynamic displacement of oil by natural gas in unconsolidated porous media is experimentally inspected. The two different rock materials, limestone and sandstone, are used as a representative of porous media. Dynamic flow experiments indicate that the increase of natural gas injection increases asphaltene deposition in the unconsolidated matrix. The results of the study show that increase in asphaltene deposition leads to pore plugging, porosity reduction and absolute permeability damage. Irreducible water measurements showed that natural gas-induced asphaltenes change the rock wettability to oil-wet.*

KEY WORDS: *Asphaltene deposition, Porous media, Wettability alteration, Permeability reduction, Miscible gas injection.*

INTRODUCTION

It is generally agreed that asphaltenes do not constitute a chemically identifiable class of compounds. They are the polar, polyaromatic and high molecular hydrocarbon fraction of crude oil that are soluble in light aromatic hydrocarbons but insoluble in low molecular weight paraffins such as pentane [1-3]. Asphaltenes in crude oils are as, both, colloidal dispersions (steric colloids and micelles) and dissolved states stabilized by

resins adsorbed on their surface [4]. Asphaltene deposition in reservoirs happens when asphaltene flocculate by depressurizing the oil. The second reason for asphaltene deposition is when solvents are used to displace oil in enhanced oil recovery (EOR). A variety of solvents have been used (e.g., natural gas liquids, liquefied petroleum gas, natural gas and CO₂); all lead to asphaltene flocculation and deposition [5].

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The precipitate asphaltene can reduce porosity, permeability, alter rock wettability, affect well injectivity and productivity and cause plugging of wellbores, pipes and processing equipment [6,7]. There are many factors that affect the asphaltene precipitation inside a reservoir. These may include the nature, saturation and distribution of the reservoir fluids, the mineralogy and properties of the rock, the pressure and temperature, the nature of injection fluids, the electrokinetic effects due to streaming potential generation due to reservoir fluid flow, the asphaltene and resin contents of the reservoir oil and the amount of formation brine and its composition [6-9]. The effects of some factors listed above on asphaltene deposition, have been investigated by researchers. For example *Minssioux* [7] and *Shedid and Abdulrazag* [10] showed that the increase of asphaltene content of the crude oil increases the rock damage. *Cimino et al.* [11] pointed that high asphaltene content of oil is not associated with high risk of trouble; on the contrary, relatively light oils, with a low asphaltene content (up to 4 % wt) are more prone to deposit formation.

The in vitro flocculation phenomena and asphaltene precipitation have been the topic of many papers. However, the effects of asphaltene deposition in a porous medium have just recently come under investigation.

Monger and Fu [12] used a special high-pressure/high-temperature variable-volume circulating cell (VVCC) to investigate the organic deposition process in porous media. The 0.5 μm stainless-steel filters were used as porous media. This paper deals with the effect of oil composition on CO_2 -induced organic deposition and also compares CO_2 to precipitating agents that are paraffinic. The VVCC apparatus was used later for similar experiments by other researchers [13, 14].

Kamat et al. [15] used the core holder setup to examine asphaltene deposition effects in water flooding dynamic tests. The effects of asphaltene on rock and fluid properties such as porosity, absolute and relative permeability were investigated in their work. *De Pedroza et al.* [6] reported asphaltene presence effects in some rock properties (e.g. wettability and absolute permeability) during oil injection. The effects of solvent were ignored in their study. Similar to *De Pedroza et al.* work [6], *Minssioux* [7] conducted some tests to indicate core damage from crude asphaltene deposition. CO_2 flooding and its induced asphaltene deposition was investigated by

Srivastava et al. [16]. In their work *Srivastava R.K. et al.* [16] studied asphaltene deposition during CO_2 flooding in static and dynamic tests. In dynamic tests they used some composite cores and dead oils. These tests were conducted using triaxially core holder. The oil injection effects on asphaltene deposition as well as CO_2 effects were studied in their work. *Rayes et al.* [9] carried out static and dynamic tests to determine adsorption of asphaltene from light, medium and heavy oils. The dynamic tests were performed on crushed reservoir rocks. The batch type measurements served for determination of the adsorption isotherms and kinetics of sorption. The effect of the asphaltene adsorption on wettability was also investigated by contact angle measurements on homogenous and heterogeneous rock plates. Adsorption tests were performed at room temperature and the pressure was kept constant by fixing the height of asphaltene solution above the sand pack. Many other field cases and laboratory studies have been reported on precipitation and deposition of asphaltenes in porous media during immiscible or miscible gas flooding operations [17-24].

The present work focuses on the investigation of asphaltenes deposition effects on porosity reduction, permeability damage and wettability alteration of reservoir rocks during natural gas flooding by dynamic flow tests. In this work two common rock types in Iranian hydrocarbon reservoirs i.e. limestone and sandstone are used as porous media rock representative. The injected oil and natural gas were taken from Iranian reservoirs.

OUTLINE OF THIS WORK

In this research an experimental system as will be discussed in next section, has been designed, constructed and installed. After that a packed system consisting of high-pressure/high-temperature cylinder, which had been packed with fine particles (a known mesh of rock gravels), was assembled as porous media. Then the porous media properties such as porosity and permeability were measured. In the next step, the dead oil was injected into water saturated porous media. Dead oil injection continued to reach the irreducible water saturation in porous media. Injection of more volumes of dead oil and simultaneously measuring the asphaltene content of the produced oil results in measuring the effect of porous media structure (the only parameter) on

asphaltene deposition. After that the miscible gas injection effects on asphaltene deposition in porous media was investigated. Finally the effects of gas-induced asphaltene on porous media properties; porosity, permeability and wettability were inspected.

EXPERIMENTAL SYSTEM

Figs. 1 and 2 show a schematic diagram and an actual depiction of corresponding high-pressure and high-temperature experimental set-up used in this work. This system was designed to investigate the asphaltene deposition effects on porous media properties in dynamic displacement of an oil sample by natural gas flooding. The major parts of this system are a positive displacement high pressure pump, a high pressure cylinder which is packed by reservoir rock (limestone or sandstone) gravels, a differential pressure transducer (DPT, Model Validyne with accuracy of $\pm 0.25\%$), two pressure gauges with accuracy of $\pm 0.5\%$ for measuring input and output pressures, a heating system consisting of heating and insulating industrial jacket and temperature controller instrument (Model CAL 9000 with accuracy of $\pm 0.1\%$), a high pressure cylinder for transferring samples (water, oil and natural gas), and a back pressure regulator and scaled sampler to gather and measure produced fluids. The rock (limestone or sandstone) gravels were crushed and passed through sieves with 40 and 45 mesh sizes. Rock particles with 350-420 micron in diameter were obtained after sieving. The high-pressure cylinder was filled with the rock gravels by simultaneously packing and shaking for two days. To prevent the exit of the fine rock particles due to fluid injection into porous media, two filters were installed on the inlet and outlet of the packed cylinder. The mesh size of the filter is about 250 micron, which is smaller than the particles size (≥ 350 micron). For suitable distribution of fluids and protection of filters the inlet and outlet of cylinder are equipped with two distributor plates (see Fig. 1-b). The packed cylinder has one inlet port for fluid injection and one outlet port for fluid production. The inlet and outlet ports of the packed cylinder are connected to the differential pressure transducer. The displacement pump with working pressure range of 0-10,000 psi and variable injection rate (2.5 cc/hr to about 580 cc/hr) was used to transfer the samples from sample cylinders to the packed system. This pump was operated in constant rate mode without

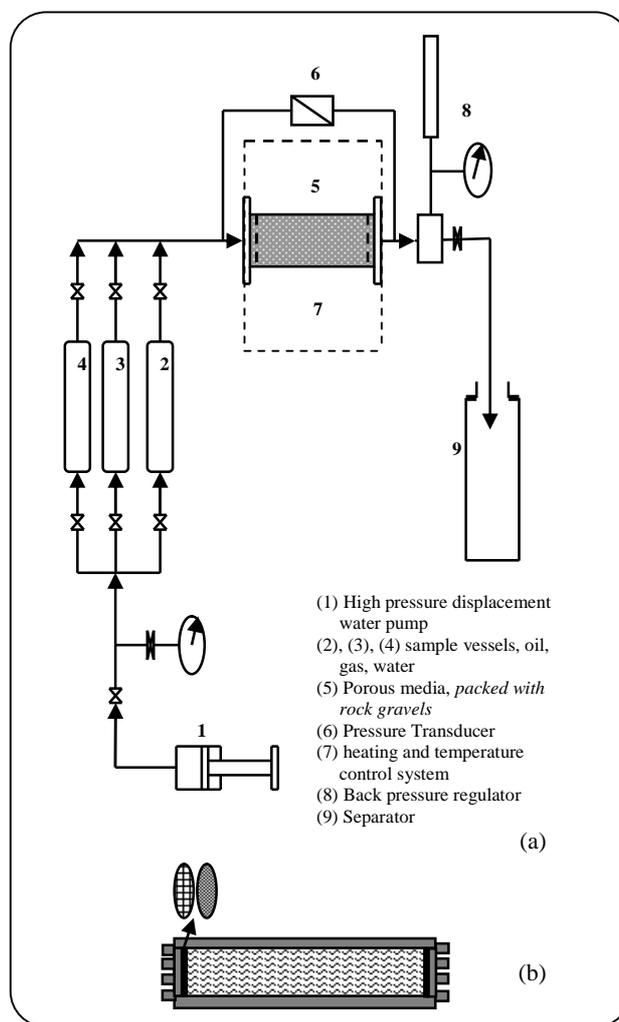


Fig. 1: Schematic of experimental set-up (a) Packed bed (b).

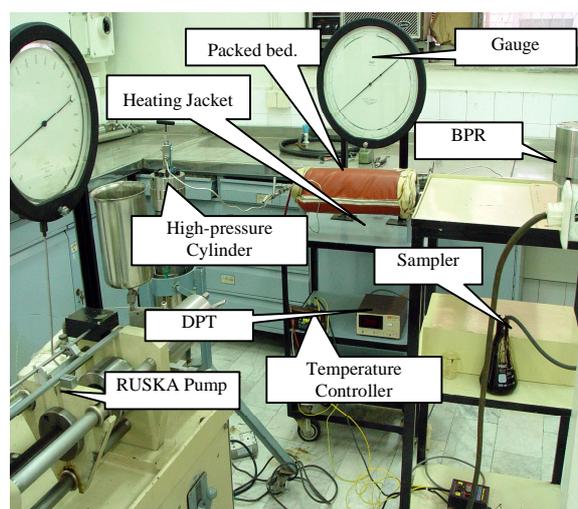


Fig. 2: The picture of experimental set-up.

any pressure fluctuations. The sample cylinder was heated up to operation temperature and then preheated fluid was injected into the packed cylinder. The temperature of the packed system was controlled automatically.

The outlet pressure of packing system was controlled by a back-pressure-regulator, (BPR). The fluids passed through BPR and then gathered in scaled sampler.

SAMPLES AND MATERIALS

The properties of crude oil sample from Iranian oil reservoir are listed in table 1. The asphaltene content of the crude oil is 6.56% weight. Table 2 shows the material type and properties of porous media. A 40 cm long stainless steel high-pressure high-temperature cylinder was used as a particle-pack holder. The limited working pressure and temperature of this system are 5000 psi and 300 °F, respectively. Table 3 gives the composition of injected natural gas from Iranian gas reservoir.

EXPERIMENTAL PROCEDURES

In order to determine the effect of the natural gas induced-asphaltene deposition on porosity of porous media the amounts of asphaltene deposition in porous media were measured. By measuring the asphaltene content of injected and produced oil samples, one can estimate the amount of deposition in porous media. The asphaltene of produced oil was measured by IP143 standard technique. Considering the initial porosity and the amount of asphaltene deposition during natural gas injection, the change in the porous media porosity can be evaluated.

We briefly explain below the procedures of all experiment stages.

1- Crushing and sieving of reservoir rock to make fine particles (rock gravels).

2- Porous media: The porous media in this work is a high pressure-high temperature stainless steel cylinder, which was packed with fine prepared gravels (see Fig. 1-b).

3- Measuring the porosity and permeability: For the porosity and permeability measurements the routine methods in packed bed properties evaluations are used. After preparation of packed bed and all necessary equipments, the experimental set-up was constructed. To measure the porosity of porous media, the packed bed

Table 1: Properties of crude oil sample.

Oil	Asphaltenes (% wt)	°API
1	6.56	21

Table 2: Properties of porous media.

Property Rock Type	Porosity (%)	Permeability (mD)	Length (cm)	Diameter (cm)
Limestone	48.54	1062.5	37.74	5.686
Sandstone	49.16	1089.6	37.74	5.686

Table 3: Injected natural gas composition.

Comp.	N2	CO2	C1	C2	C3	iC4	nC4	iC5	nC5	C6	C7+
mol %	3.84	0.7	90.9	2.89	0.8	0.21	0.27	0.12	0.08	0.1	0.2

was vacuumed and saturated with water. The porosity of made porous media can be calculated from the total volume of packed cylinder and injected water. The permeability of the porous media is obtained by flowing of water through porous media and measuring stabilized pressure drop at specific flow rate (*Darcy's law* is used to calculate permeability).

4- Measuring the irreducible water (as a measure of porous media wettability): In this step dead oil is injected into the water-saturated porous media at constant flow rate. During the dead oil injection the amount of the produced water is measured; the injection continues until the water production stops. Difference between initial water saturation and the produced water will be the amount of irreducible water. Also the asphaltene content of the produced oil is measured at each pore volume of oil injection. The oil flooding continues until the asphaltene content of the produced oil reaches to the original asphaltene amount. The amount of deposited asphaltene in the porous media was measured during the oil injection to depict the deposition effects of porous media properties.

5- Determination of gas injection effects on asphaltene precipitation: For determining natural gas - induced asphaltene precipitation, natural gas is injected into the oil-saturated porous media. The asphaltene content of the produced oil is measured to determine the effects of injected gas on asphaltene precipitation.

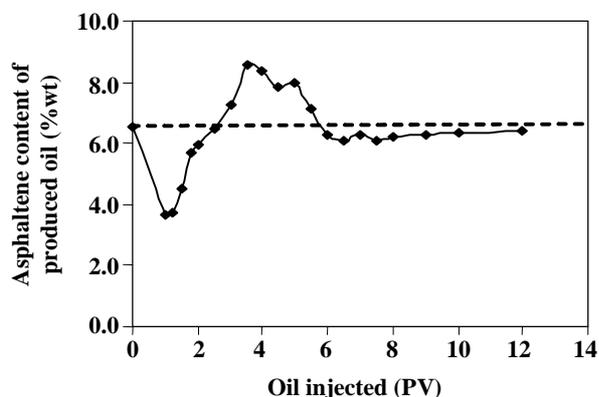


Fig. 3: Asphaltene content of oil produced during oil injection (Limestone).

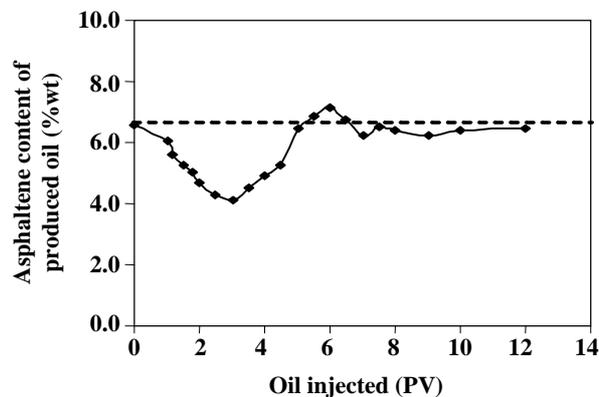


Fig. 4: Asphaltene content of oil produced during oil injection (Sandstone).

Besides, the amount of produced water was measured to evaluate the change in amount of the irreducible water due to additional asphaltene deposition.

6- Determination of deposited asphaltene effects on porous media properties: Changes of porosity, permeability and irreducible water content of the porous media (as a measure of wettability change or alteration of the porous media) indicate the asphaltene deposition effects on porous media properties.

RESULTS AND DISCUSSION

Porous media effects

The porous media was first saturated with water at 205 °F and 2500 psi. Dead oil was injected into porous media at a rate 9.72 cc/min. The asphaltene content of the produced oil was measured at different steps during oil injection. The asphaltene content of injected oil was 6.56% weight. Figs. 3 and 4 depict the porous media effects on asphaltene deposition during oil injection for limestone and sandstone packed beds, respectively. Because of asphaltene adsorption on the clean surface of porous media, the asphaltene content of the produced oil at first pore volume of oil injection was sharply reduced (from 6.56 to 3.69 %wt for limestone and from 6.56 to 4.11 %wt for sandstone). Oil injection continued until water production stopped (at 2.5 PV oil injection for limestone and at 5 PV for sandstone). During this process the asphaltene content of the produced oil started to increase but remained below the initial value, because the outflow of water in porous media leads to creation of the new sites, which are available for additional deposition of asphaltenes. Once water production stopped, the asphaltene content of the produced oil reached a value

slightly over its original value (up to 8.59 %wt for limestone and 7.11 % wt for sandstone). This increase over the asphaltene content of the original oil indicates the possible re-dissolution of adsorbed asphaltene on the porous media surface. The fluctuation of asphaltene content of the produced oil around the initial value (6.56% wt) with additional oil injection was due to slight redissolution and tacking-off of the adsorbed asphaltene. This fluctuation stopped (after about 8 PV oil injected for limestone and 9 PV oil injected for sandstone) and the asphaltene content of the produced oil almost reached near to its initial value. This state may be assumed as the *steady state* condition. The steady state indicates that the ability of porous media in adsorption of asphaltenes has come to end and the natural gas flooding effects on additional asphaltene deposition can be tested.

Figs. 3 and 4 indicate that the presence of water film on rock surface can reduce or delay the asphaltene deposition process. In strongly water-wet rock (such as sandstones) the presence of water shields more surfaces from direct interaction with asphaltene. As a result the amount of asphaltene deposition onto more water-wet surfaces will be less than the amount deposited on less water-wet surfaces (such as some limestones). Besides, these figures show that the asphaltene deposition rate on the sandstone surface is less than the deposition rate on limestone media i.e. in comparison with the limestone medium it takes more time for the oil in sandstone media to displace water and reach its initial asphaltene content.

Figs. 5 and 6 show the absolute permeability variations during dead oil injection into the porous media. From these figures both the adsorption and desorption of asphaltenes through porous media can be seen.

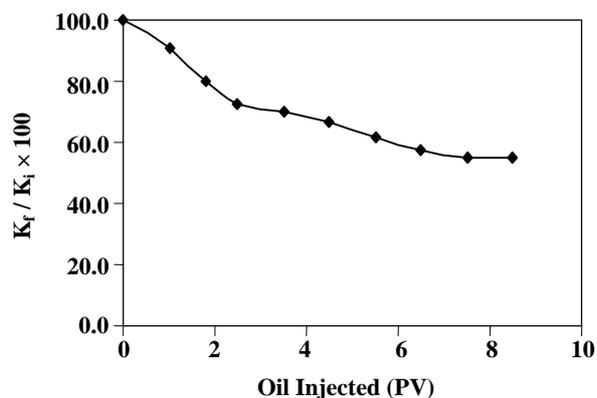


Fig. 5: Permeability variation during oil injection (Limestone) (K_i : initial permeability; K_f : current permeability).

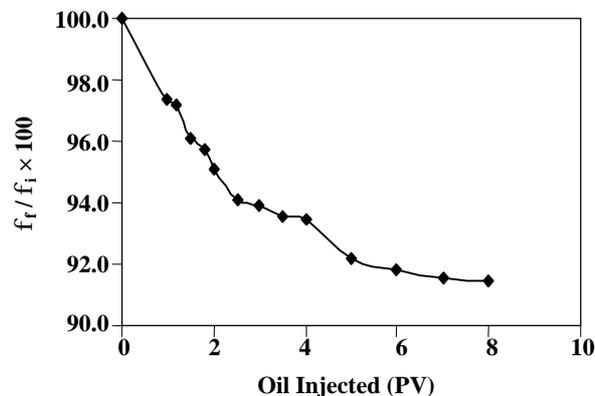


Fig. 7: Porosity reduction during oil injection by asphaltene deposition (limestone) (f_i : initial porosity; f_t : current porosity).

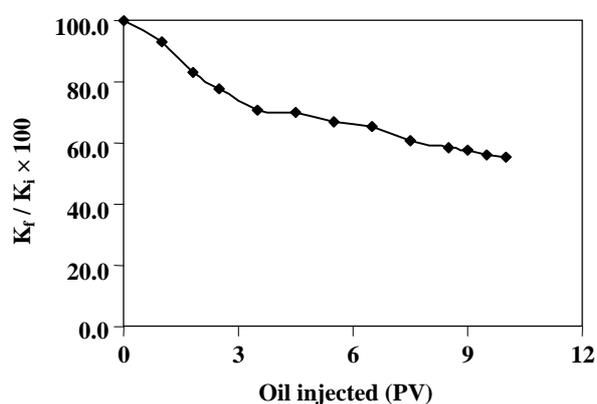


Fig. 6: Permeability variation during oil injection (Sandstone).

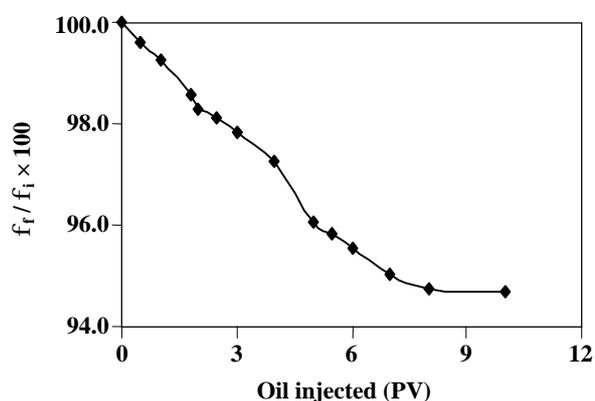


Fig. 8: Porosity reduction during oil injection by asphaltene deposition (Sandstone).

At first the high rate of asphaltene adsorption on porous media causes high permeability and pressure drop reduction, then after a certain amount of dead oil flooding (almost 2.5 and 4.5 PV for limestone and sandstone, respectively) the desorption process starts and the slope of the permeability reduction is decreased.

The porosity reduction due to asphaltene deposition during oil injection is shown in Figs. 7 and 8. It can be seen that the higher the asphaltene deposition on porous media greater is the reduction in porosity.

Miscible natural gas flooding effects

Miscible natural gas was injected into oil-saturated porous media to determine its effects on asphaltene deposition. The injection rate was 0.1737 cc/min. When the injected gas contacted the oil, it changed the composition of the oil and caused asphaltene instability and its eventual deposition. Figs. 9 and 10 show the variation in the asphaltene content of the produced oil

with PV of injected miscible natural gas.

The IP143 analysis on the produced oil indicated that the asphaltene content of the produced oil did not change until about 0.6 PV of natural gas injection. After that the asphaltene content of the oil produced was reduced. This reduction was due to an increase in asphaltene deposition in porous media through natural gas injection. Additional natural gas injection caused a sharp reduction in the asphaltene content of the produced oil. These results show that additional asphaltene precipitation/adsorption occurs in porous media during miscible natural gas injection.

Porosity and permeability reduction

Additional asphaltene deposition in porous media can cause further porosity reduction. Figs. 11 and 12 show the porosity variations of the porous media. The porosity values after asphaltene deposition have been depicted versus the amount of injected gas. Porosity reduction in

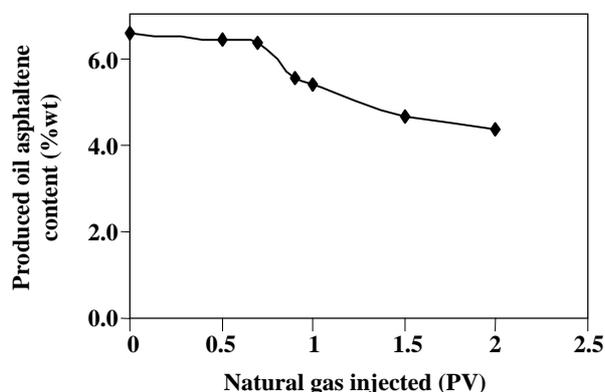


Fig. 9: Asphaltene content of oil produced during natural gas injection (Limestone).

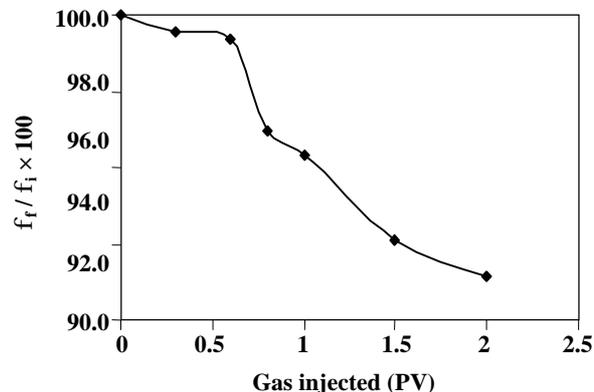


Fig. 11: Porosity reduction during gas injection by asphaltene deposition (Limestone).

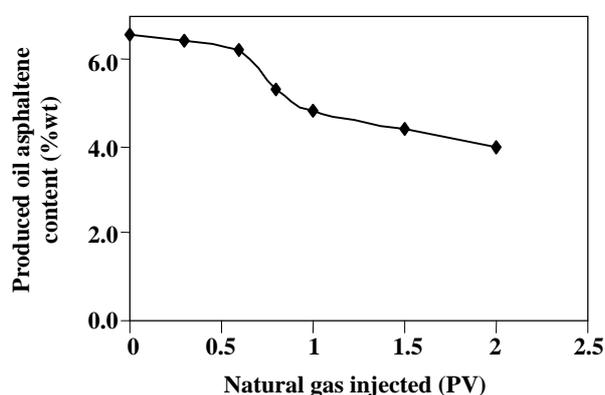


Fig. 10: Asphaltene content of oil produced during natural gas injection (Sandstone).

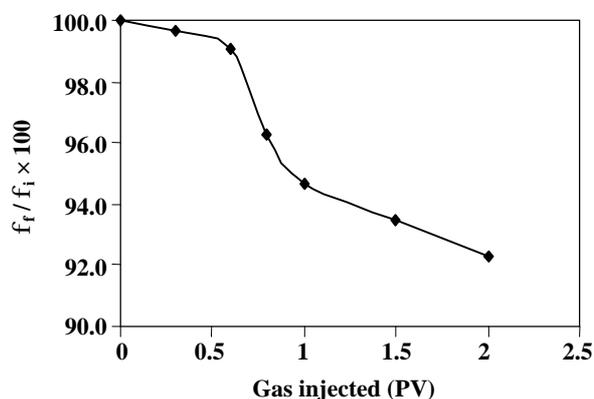


Fig. 12: Porosity reduction during gas injection by asphaltene deposition (Sandstone).

the natural gas injection process was greater than porosity reduction during saturating porous media by oil injection; these variations indicate that most of asphaltene depositions occur after gas injection.

Table 4 gives the ratio of absolute permeability of the porous media after asphaltene deposition to the original absolute permeability before and after gas injection. These data indicate that the permeability reduction has increased after gas injection.

The contributions of oil and gas injection processes in porosity and permeability reductions are also shown by Figs. 13 and 14, respectively. Indeed these figures compare the effects of two mentioned processes on rocks properties.

Fig. 13 depicts that after injection of about 2 PV of the natural gas, the porosity of sandstone and limestone type rock has reduced about 1.9 and 1.7% respectively (crosshatched areas).

Fig. 14 shows the natural gas injection effects on permeability reduction of two porous mediums.

As it can be seen injection of about 2 PV of natural gas causes 3.5 and 3.2 % reduction in sandstone and limestone rock types respectively (crosshatched areas).

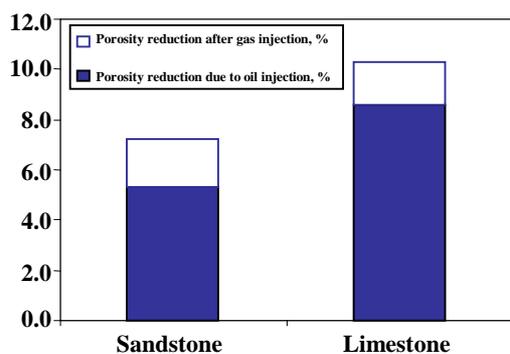
Wettability alteration

In this work the change in the amount of irreducible water in porous media was used as an indication of porous media wettability alteration. It has been demonstrated that oil wet formations have much lower irreducible water than water-wet ones. Table 5 gives the values of irreducible water in porous media after natural gas injection. Water-wet limestone reservoir rock was used in this work. The amount of irreducible water in media can be evaluated by measurement of the initial water content of the porous media and the cumulative produced water (when there is no more water production).

Table 4: Permeability data before and after gas injection.

Permeability reduction / Rock type	$K_f/K_i \times 100^*$, (%) (Before gas injection)	$K_f/K_i \times 100^*$, (%) (After gas injection)
Limestone	54.79	51.58
Sandstone	55.71	52.17

* K_i : initial permeability K_f : permeability after or before gas injection

**Fig. 13: Comparison of porosity reduction during oil and gas injection processes.**

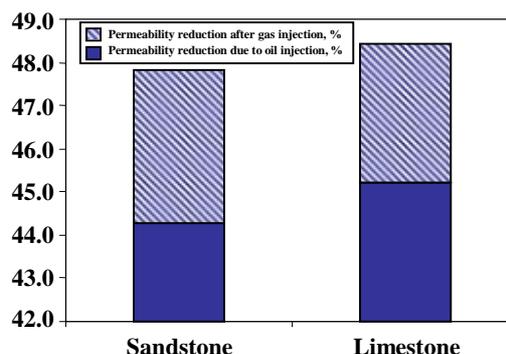
The amount of water production was also measured during natural gas injection; this value was used to determine the amount of irreducible water after natural gas injection. As it is shown in table 5 the irreducible water in porous media was reduced after natural gas injection. These results indicate that asphaltene deposition caused changes in the wettability of porous media toward oil-wet.

CONCLUSIONS

Investigation of asphaltene deposition in porous media especially in miscible gas injection processes requires significant experimental and theoretical efforts. Depending on the rock type; medium acidity (petroleum fluid) and medium nature the adsorption of flocculated asphaltene on rock surface can cause great rock porosity and permeability reductions. Also in miscible gas injection processes (EOR) the deposited asphaltene interact with rock surface. This interaction leads to change in the physical properties of the rock. In fluid-rock systems, asphaltene deposition affects common properties such as permeability and wettability. Change in rock wettability due to gas-induced asphaltene deposition (in EOR processes) can cause severe problems in oil production.

Table 5: Wettability alteration data during gas injection.

Irreducible water / Rock type	Irreducible water (Before gas injection), %PV	Irreducible water (After gas injection), %PV
Limestone	26.5	10.7
Sandstone	31	16.5

**Fig. 14: Comparison of permeability reduction during oil and gas injection processes.**

The results of this work depict that due to asphaltene depositions on the rock surface, the porosity and permeability of the rock are reduced and its wettability is altered from water-wetting to oil-wetting. Changes in content of the irreducible water in porous media have been used as a criterion for wettability alteration.

The dynamics flow experiments in this work, showed that rock type (porous media materials) affects the asphaltene deposition on matrix surface, considerably. The results for two types of the rock used in this work (i.e. limestone and sandstone) indicate that the sandstone rock material is originally more water-wet than limestone. In other words, the amount of deposited asphaltene on sandstone rock will be less than the amount deposited on limestone rock.

Based on the experimental observations in this work, it seems that the asphaltene deposition rate on the sandstone surface is less than the depositions rate on limestone media i.e. in comparison with the limestone medium it takes more time for the produced oil in sandstone media to reach its initial asphaltene content.

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