

WETTABILITY ALTERATION OF OIL-WET DOLOMITE USING
NANOSOLUTION IN CATIONIC SURFACTANT FOR ENHANCED OIL
RECOVERY

MOHD SHAHRIZAN BIN MOSLAN

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Specially dedicated to *Mak* and my late *Abah*
I love you *Mak* and I really miss you *Abah*
Also to *abang, akak, ude, ateh and adik*
This is for all of you

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In the name of Allah, the most beneficent and the merciful

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ABSTRACT

Reservoir wettability is depending firmly on the existence of polar components (acids or bases) which are soluble in crude oil and their interaction on the solid surfaces. Wettability alteration by chemical agents such as surfactant and nanoparticles is seen to be one of the realistic methods to modify the wettability of carbonate rock from oil-wet to water-wet conditions. An experimental study of wettability changes upon exposure to cetyltrimethyl ammonium bromide (CTAB) surfactant and nanoparticles dispersed in CTAB has been conducted. Besides that, the stability of the nanoparticles colloidal was also determined by sedimentation and transport test. In addition to that, interfacial tension (IFT) before and after addition of nanoparticles into the system was also determined. Finally, core displacement test was accomplished to determine the effectiveness of nanoparticles in displacing the remaining oil trapped in the reservoir. It was found that CTAB could alter the wettability of oil-wet dolomite to water-wet and addition of nanoparticles has enhanced the contact angle reduction. Al_2O_3 nano solution was more stable than ZrO_2 and about 55% of Al_2O_3 and 52.5% of ZrO_2 could be recovered from the transport test. The IFT of the system was 0.077 mN/m and has reduced after addition of nanoparticles into the system. The reduction of IFT due to Al_2O_3 was greater as compared to ZrO_2 . Oil recovery by water flooding was 55%, 62.7% and 56% for sand packs 1, 2, and 3 respectively. However, after injection of two pore volumes of Al_2O_3 , ZrO_2 nano solutions, and CTAB surfactant additional 20%, 16.7% and 14.1% of oil were recovered from sand packs 1, 2 and 3, respectively due to wettability alteration. As the conclusion, favourable wettability must be obtained in order to produce more oil especially in carbonate and mature reservoirs.

ABSTRAK

Kebolehbasahan reservoir bergantung pada kehadiran komponen kutub (asid atau bes) yang larut dalam minyak mentah dan interaksi masing-masing pada permukaan pepejal. Pengubahsuaian kebolehbasahan batuan oleh agen kimia misalnya surfaktan dan nanopartikel, ialah satu daripada kaedah realistik untuk mengubah suai kebolehbasahan batu karbonat dari keadaan basah minyak ke keadaan basah air. Kajian tentang perubahan kebolehbasahan apabila terdedah kepada surfaktan setiltrimetil ammonium bromida (CTAB) dan nanopartikel yang tersebar dalam CTAB telah dilaksanakan. Disamping itu, kestabilan koloid nanopartikel juga ditentukan menerusi ujian pemendapan dan ujian pengangkutan. Sebagai tambahan, tegangan antara muka (IFT) sebelum dan selepas penambahan nanopartikel turut diteliti. Akhir sekali, ujian penyesanan teras dilaksanakan bagi memerhatikan keberkesanan nanopartikel dalam menyasarkan baki minyak yang terperangkap di dalam reservoir. Hasil kajian menunjukkan bahawa CTAB boleh mengubah kebolehbasahan dari basah minyak dolomit ke basah air, dengan penambahan nanopartikel mampu mengurangkan lagi sudut sentuhan. Larutan nano Al_2O_3 lebih stabil berbanding larutan nano ZrO_2 dengan 55% Al_2O_3 dan 52.5% ZrO_2 mampu diperoleh semula menerusi ujian pengangkutan. Nilai IFT sistem ialah 0.077 mN/m dan berjaya diturunkan menerusi penambahan nanopartikel ke dalam sistem terbabit. Pengurangan IFT yang disebabkan penambahan Al_2O_3 adalah lebih besar berbanding ZrO_2 . Perolehan minyak menerusi banjiran air ialah 55%, 62.7% dan 56% bagi pek pasir 1, 2 dan 3. Walau bagaimanapun, selepas suntikan dua isi padu liang larutan nanopartikel Al_2O_3 dan ZrO_2 , serta surfaktan CTAB, tambahan masing-masing 20%, 16.7% dan 14.1% minyak baki berjaya dikeluarkan daripada pek pasir 1, 2 dan 3 berikutan berlakunya perubahan kebolehbasahan. Kesimpulannya, kebolehbasahan yang sesuai mesti diperoleh bagi mengeluarkan lebih banyak minyak terutama dari reservoir karbonat dan reservoir matang.

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LIST OF ABBREVIATIONS

EOR	-	Enhanced oil recovery
IFT	-	Interfacial tension
CTAB	-	Cetyltrimethylammonium bromide
WAG	-	Water alternated gas
IOR	-	Improved oil recovery
CMC	-	Critical micelles concentration
EA	-	Ethoxylated alcohol
DTAB	-	Dodecyltrimethylammonium bromide
Na ₂ CO ₃	-	Sodium carbonate
SDS	-	Sodium dodecyl sulphate
MCA	-	Minimum contact angle
PZC	-	Point of zero charge
LHP	-	Lipophobic-hydrophilic polysilicon
PSNP	-	Polysilicon nanoparticles
NWP	-	Neutrally wet polysilicon
HLP	-	Hydrophobic-lipophilic polysilicon
FESEM	-	Field emission scanning electron microscope
NP	-	Nanoparticles
ppm	-	Part per million
PV	-	Pore volume
rpm	-	Revolutions per minute

LIST OF SYMBOLS

q_e	-	Adsorption capacity at equilibrium
q_t	-	Adsorption capacity at time
c_o	-	Initial concentration
c_e	-	Equilibrium concentration
v	-	Volume of solution in liter
w	-	Weight of adsorbent in gram
t	-	Time
R^2	-	Correlation coefficient
θ	-	Contact angle
σ	-	Interfacial tension
ρ	-	Density
h	-	Height
k_{ro}	-	Relative permeability to oil
k_{rw}	-	Relative permeability to water
S_{or}	-	Residual oil saturation
δ_o	-	Displacement by oil
δ_w	-	Displacement by water

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Generally, oil recovery occurs through two main processes, which are primary recovery and secondary recovery. Primary recovery commonly comes from natural drives; which are rock and liquid expansion drive, depletion drive, gas cap drive, water drive, and gravity drainage drive. Meanwhile, secondary recovery refers to the introduction of additional energy into the reservoir. One of most common technique in secondary recovery is water injection or waterflooding. Briefly, the purpose of this waterflooding is to maintain the reservoir pressure and displace the remaining oil in the reservoir. Satter *et al.* (2008) mentioned that waterflooding technique has been widely used mainly due wide availability of water, inexpensive, and lower capital investment and operating costs. Ordinarily, maximum total oil recovery after primarily and secondary stage is 40% to 60% (Farouq and Thomas, 1996). The remaining oil is trapped mostly because of the heterogeneity of reservoir, unfavorable wettability and capillary trapped. To recover this remaining oil, advanced methods, which have become known as enhanced oil recovery (EOR) methods is necessary.

About more than 60% world's oil reserves are held in carbonate reservoirs (Sheng *et al.*, 2010). It is noted that oil recovery in these reservoirs is lower ascribed by two leading factors that are fractured reservoir and oil-wet rock. Therefore, many researchers have been focusing on wettability alteration of the rock and apparently, there is an increasing interest in using chemical for this purpose. Adding chemical agents such as surfactants can modify the wettability of carbonate rock system from oil-wet to water-wet condition (Mohammadi *et al.*, 2014). Wettability alteration of rock by surfactants was studied and the usage of ionic surfactants has been considered as feasible method for the recovery of oil reservoirs by modifying the wettability of rock surface from oil-wet to water-wet.

Chabert *et al.* (2010) perceived 80 % of carbonate reservoir classified as neutral to oil-wet due to the positive zeta potential of the rock surface. The adsorption of polar organic component presents in crude oil has been identified to be the main factor for the wettability alteration of carbonate surfaces to oil wet condition (Mahvash *et al.*, 2015). Cationic surfactant was observed to perform better than anionic surfactant in changing wettability of carbonate rock from oil-wet to more water-wet. This is due to the formation of ion pairs between the cationic heads of the surfactants molecules and the acidic components of the crude oil absorbed on the surface of carbonate rock (Austad *et al.*, 1997). However, according to Salehi *et al.* (2008) anionic surfactant molecules form a monolayer on the surface of carbonate rock through hydrophobic interaction of the tails of the surfactants molecules with the absorbed crude oil components on the rock surfaces. In addition, the hydrophobic interactions are much weaker than ion pair interaction, this explains why cationic surfactants have better performance than anionic surfactants in altering carbonate rock's wettability to a more water-wet state.

Nanotechnology is developing in oil and gas industry due to its potential to solve or manage several problems in the petroleum industries. EOR is one of the potential areas of nanotechnology application. Nanoparticles have the ability to alter certain factors in the formations and oil properties and this can be advantageous to enhance oil recovery (Ogolo *et al.*, 2012). The authors also reported that the wettability of a formation could be changed by applications of nanoparticles. Several studies have

been conducted and the applications of nanoparticles to change rock wettability and its subsequent effects on oil recovery have been documented by several authors (Karimi *et al.*, 2012; Ogolo *et al.*, 2012; and Hendraningrat *et al.*, 2013). Nanoparticles can enhance the recovery through two major mechanisms, via interfacial tension reduction or wettability alterations (Roustaei *et al.*, 2012). In 2013, Hendraningrat *et al.* stated that the displacement mechanism by nanoparticles is called as structural disjoining pressure mechanism. Brownian motion and electrostatic repulsion between the nanoparticles are the energies that drive this mechanism in which smaller size nanoparticles would cause a bigger repulsion force. Structural disjoining pressure is associated to the fluids ability to spread along the surface of a substrate due to the imbalance interfacial forces among solid, oil phase and aqueous phase, which is the nanofluids. The interfacial forces will cause the nanofluids contact angle to decrease and the result is called as wedge film and this wedge film will act to separate formation fluids such as oil from the formation surface (Liu *et al.*, 2012).

Nanoparticles have high energy surface and therefore the adsorption of nanoparticles on a solid surface can change the surface energy and subsequently change the wettability of the rocks. In this research, the introduction of nanofluids to alter the wettability of carbonate rock is studied by experimental approach in which the experiment is also covering the stability of nanofluids and the effect of nanofluids on IFT.

1.2 Problem Statement

Current world oil production mostly comes from mature fields and over the past decades, oil-based companies are maximizing the oil production from these fields. However, stagnant oil production and unimpressive recovery by primary and secondary recovery caused the situation to be more precarious (Roustaei *et al.*, 2012). Enhanced oil recovery (EOR) technologies are necessary to meet the energy demand in many more years to come.

There is a huge interest to enhance recovery from the carbonate rock system by considering the remaining amount of oil in place. The addition of some chemical agents such as surfactants and nanoparticles into the injecting water can modify the wettability of the rock surface. In spite of all reported studies, the performance of nanoparticles in cationic surfactant i.e cetyltrimethylammonium bromide (CTAB) to alter wettability of oil-wet dolomite to water-wet condition has not yet well studied. Even though CTAB surfactant is expensive and toxic, this research is still in the preliminary study and further study need to be conducted in order to be implemented in the real application.

Therefore, this research is carried out to investigate and identify the types of nanoparticles that are effective as wettability modifiers. The challenging part in this study is to understand the mechanisms involved in this alteration. Parameters such as nanofluids concentrations, and stability of the colloids are very important and is investigated in order to achieve an optimized condition in this study.

1.3 Objectives of Study

The objectives of this research are as follows:

- I. To study the stability of nanoparticles in cationic surfactant i.e CTAB and the quantities of nanoparticles deposition during the transport through dolomite rock.
- II. To investigate the wettability alteration of dolomite rock system after introduction of nano solution.
- III. To study the effect of wettability alteration on the oil recovery

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