THE ULTRASONIC WAVES EFFECTS ON OIL-WATER EMULSIFICATION, COALESCENCE, DETACHMENT, MOBILIZATION AND VISCOSITY IN POROUS MEDIA

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This thesis is dedicated to my beloved wife who has been a great source of motivation and inspiration. Also, this thesis is dedicated to my parents who have supported me all the way since the beginning of my studies.

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ABSTRACT

Ultrasonic wave technique is an unconventional EOR method, which has been of interest to researchers for more than six decades. Emulsification and demulsification are phenomena which occur at the interface of oil and water under the influence of ultrasonic waves. Therefore, the conditions in which emulsification becomes dominant over demulsification due to ultrasonic radiation in porous media should be further investigated. However, surfactants are the principal agents that enable oil and water to mix and are often the most expensive component in an emulsion. Therefore, selecting an appropriate surfactant formulation capable of mobilization of oil without significant surfactant loss due to adsorption and phase separation in the reservoir is very important. Estimation of solubilization parameters are great tools in designing economical emulsion flooding compositions. In this study, the effect of ultrasonic waves on the amount of oil and water solubilized by a unit of surfactant were investigated. It was observed that the emulsion volume and amount of oil solubilized in emulsion were increased by increasing salinity under short periods of ultrasonic wave radiation, and demulsification of the emulsion occurred after longer period of radiation. In addition, Hele-Shaw model tests were conducted to show microscopically the effect of long and short periods of ultrasonic waves' radiation at the interface of paraffin oil and surfactant solution/brine. Diffusion of phases, formation of emulsion and gas bubbles were observed after short periods of ultrasonic waves' radiation. However, demulsification and coalescence of surfactant solution/brine droplets inside emulsion was initiated after long periods of ultrasound radiation.Another objective of this study was to investigate directly the effect of ultrasonic waves on viscosity changes in three types of oil (paraffin oil, synthetic oil, and kerosene) and a brine sample. It was observed that the viscosity of all the liquids was decreased under the influence of ultrasonic waves in both uncontrolled and controlled temperature conditions. However, the reduction was found to be more significant for uncontrolled temperature condition cases. In addition, micro-model experiments were conducted to show other oil recovery mechanisms such as oil droplet coalescence, oil mobilization, and oil detachment from dead end pores under the influence of ultrasonic waves. The results revealed that these mechanisms happen in porous media under the influence of ultrasonic waves. Therefore, it was concluded that the use of ultrasonic waves could be suggested, not as a substitute for conventional EOR methods, but as an alternative or complimentary tool, which in certain instances may make conventional methods more effective and less costly.

ABSTRAK

Teknik gelombang ultrasonik adalah kaedah EOR bukan konvensional yang telah menarik minat ramai penyelidik sejak lebih dari enam dekad. Emulsifikasi dan demulsifikasi adalah fenomena yang terbentuk pada antaramuka diantara minyak dan air di bawah pengaruh gelombang ultrasonik. Dengan itu, keadaan di mana emulsifikasi menjadi dominan berbanding demulsifikasi akibat sinaran ultrasonik dalam media berliang harus dikaji selanjutnya. Namun begitu, surfaktan adalah agen penting yang membolehkan minyak dan air bercampur dan umumnya merupakan komponen yang paling mahal dalam emulsi. Oleh demikian, memilh formulasi surfaktan yang sesuai bagi membolehkan mobilisasi minyak tanpa kehilangan surfaktan yang signifikan disebabkan jerapan dan pemisahan fasa dalam reservoir adalah sangat penting. Menganggar parameter pemelarutan adalah alatan penting dalam merekabentuk komposisi banjiran emulsi secara ekonomik. Dalam kajian ini, kesan gelombang ultrasonik ke atas jumlah minyak dan air terlarut oleh satu unit surfaktan adalah dikaji. Hasil cerapan didapati isipadu emulsi dan jumlah minyak terlarut dalam emulsi adalah meningkat dengan peningkatan kemasinan di bawah radiasi gelombang ultrasonik dalam jangkamasa pendik. Selain itu, ujian secara mikroskopik menggunakan model Hele-Shaw menunjukkan yang kesan radiasi gelombang ultrasonik pada masa jangka masa yang panjang dan pendik pada antaramuka minyak parafin dan larutan/air garam surfaktan. Penyebaran fasa, pembentukan emulsi dan buih-buih gas dapat diperhatikan selepas radiasi gelombang ultrasonik dalam jangka masa pendik. Namun begitu, demulsifikasi dan pegabungan titisan larutan/air garam surfaktan di dalam emulsi telah terjadi selepas radiasi ultrabunyi pada jangka masa panjang. Objektif seterusnya bagi kajian ini adalah untuk mengkaji secara langsung kesan gelombang ultrasonik ke atas perubahan kelikatan ke atas tiga jenis minyak (minyak parafin, minyak sintetik, dan kerosen) dan satu sampel air garam. Didapati kelikatan bagi semua cecair adalah berkurang dengan pengaruh gelombang ultrasonik bagi kedua-dua keadaan suhu sama ada suhu terkawal atau tanpa kawalan. Sebagai tambahan, eksperimen mikro-model telah dijalankan bagi menilai mekanisme perolehan minyak yang lain seperti pegabungan titisan minyak, mobilisasi minyak dan pengenyahan minyak dari hujung liang di bawah pengaruh gelombang ultrasonik. Dengan itu, dapat dibuat kesimpulan yang penggunaan gelombang ultrasonik boleh dicadangkan, bukan sebagai pengganti bagi kaedah EOR konvensional, tetapi sebagai satu pilihan atau alatan sampingan, yang mana dalam keadaan tertentu boleh membuat kaedah konvensional lebih berkesan dan dengan kos yang rendah.

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AOS - Alpha Olefin Sulfonate

C - Circumference of ring

Ca - Capillary number

CMC - Critical Micelle Concentration

D - Density of water at 25°C

d - Density of test specimen at 25°C

EOR - Enhanced Oil Recovery

f - Frequency

IFT - Interfacial Tension

L - Capillary tube length

n - Power-Law fluid index

NUS - Non-Ultrasound

OW - Oil Wet

P - Scale reading

PO - Paraffin oil

ΔP_s - External pressure gradient

ΔP - Differential pressure

Q - Flow rate

R^{right} - Radius of the right meniscus

R^{left} - Radius of the left meniscus

R - Radius of ring

r - Radius of wire of ring

$TCMS$ - Trichloromethylsilane

US - Ultrasound

V_o - Amount of oil in microemulsion

V_w - Amount of water in microemulsion

WW - Water Wet

μ - Viscosity

θ - Contact angle

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CHAPTER 1

INTRODUCTION

1.1 Background

In the oil industry, the reduction of oil production is of major concern as world necessity for oil increases. Therefore, developing and applying new techniques to mobilize residual oil left in the reservoir and make best of the original oil in place (OOIP) is very crucial.

As the world's human populationgrowing, decreasing of production in oil recovery processes is of major concern. Crude oil production and development of a petroleum reservoir is divided into three distinct stages such as primary, secondary and tertiary or Enhanced Oil Recovery (EOR). During primary recovery, the natural pressure of the reservoir, combined with pumping equipment, brings oil to the surface. Primary recovery is the easiest and cheapest way to extract oil from the ground. However, this method of production typically produces only about 10 percent of a reservoir'sOOIP reserve. In the secondary recovery phase, water or gas is injected to displace oil, making it much easier to drive it to a production wellbore. This technique generally results in the recovery of 20 to 40 percent of the OOIP. Consequently, the oil left in the reservoir is the goal of tertiary recovery (EOR) process. In addition to maintaining reservoir pressure, this type of recovery seeks to alter the properties of the oil in ways that facilitate additional production. The three major types of tertiary recovery are chemical flooding, thermal recovery (such as a steamflood) and miscible displacement involving carbon dioxide (CO_2), hydrocarbon or nitrogen injection. All of the conventional EOR methods include some limitations.Some of them are expensive to use, need a wide range of surface apparatus, generate dangerous environmental results, and have technical limitations(Xiao *et al.,* 2004). Therefore, some unconventional methods have been proposed to EOR.

One of the unconventional methods is the application of wave energy for enhancing oil recovery in reservoirs. This method has been of interest for more than six decades.There are numerous investigations to show the effect of earthquakes on increasing oil recovery (Steinbrugge and Moran, 1954; Voytov *et al.*, 1972; Simkin and Lopukhov, 1989).An earthquake is the result of a sudden release of energy in the Earth'scrust that creates seismic waves. However, the question is that how long one should wait until an earthquake happening. Therefore, the waves were generated artificially. The wave energy can be sent to an oil reservoir by using seismic method, and in-situ sonication (ultrasonic wave method). The seismic waves can be applied into the reservoir by surface vibrators or explosives. The method utilizes low frequency compressional waves. The in-situ sonication method uses an acoustic transducer, which is delivered into the bottom of the hole and producing ultrasonic waves with high frequency and high intensity. Therefore, the ultrasonic waves move within the porous media and stimulate the fluids mechanically (Hamida, 2006).

There are some advantages of using this method compared to the other conventional techniques that can be reviewed here:

(i) In the methods using fluids, hydraulic force is in charge of delivering the driving force in the conventional methods and fluids always choose the least resistance pathway and suffer from bypassing effects. Therefore, numerous EOR techniques are unsuccessful in heterogeneous formations and running off great oil pools unaffected. However, in the methods using waves, the energy is distributed in all direction and is unchanged by permeability of medium or pore network. Therefore, it is easy to affect every point in the reservoir at the same time (Beresnev *et al.,* 2005).

(ii) The need for chemical stimulation (acid, solvents, and etc.) is replaced by using waves. Because chemical stimulation is not compatible in some cases with the reservoir rock or fluid (Beresnev *et al.,* 2005).

(iii) The waves can be applied to the reservoir while the well is producing (Beresnev *et al.,* 2005).

On the other side, the main limitation of using wave energy is the quick attenuation in porous media particularly at high frequencies (20 kHz up to several gigahertz)(Dunn, 1986). Therefore, the application of ultrasonic method is restricted to near-wellbore area because of its high attenuation in porous media. By studying Biot's theory, one recognizes that the length of attenuation of ultrasonic waves with frequency about 20 kHz is 2 to 10 cm (Biot, 1956b; Biot, 1962). Therefore, a great number of researches have been performedusing low frequency waves that are able to propagate in reservoir several kilometers. On the other hand, it is recognized that ultrasonic waves exist in the reservoir after applying seismic waves (low frequency waves) to the reservoir because low frequency waves dispersion generate ultrasonic noise (highfrequency waves) in porous media (Nikolaevskii and Stepanova, 2005). Ultrasonic waves have short wavelengths and considerably play an important role in mechanical perturbation at the pore scale. Consequently, it is supposed that the effect of ultrasonic waves (high frequency waves) is more significant compare to low frequency waves at pore scale (Duhon and Campbell, 1965; Beresnev *et al.*, 2005; Xiaoyan *et al.*, 2007). Nevertheless, the mechanisms caused by ultrasonic waves in porous media are not well recognized yet and requires deep investigations to disclose the physics and mechanisms process included in recovery of oil.

1.2 Statement of Problem

The interest in using ultrasonic waves as an unconventional method for stimulation of oil reservoirs dates back to 1950's. Most of the studies that have been performed over these years are limited to macromodel studies such as measuring oil recovery after applying ultrasonic waves. In addition, the oil recovery mechanisms mentioned in the previous studiesare theoretical and lacks fundamental researches(microscopic or pore scale studies) (Naderi, 2008). Studying the mechanisms that lead to oil mobilization by ultrasonic waves is very important and is essential for field purposes. However, in spite of many studies, questions about the effective mechanisms causing increase in oil recovery still existed. Therefore, it is very crucial to perform basic experiments (pore scale) to achieve a good knowledge and deep insight into the mechanisms.

Emulsification is one of the oil recovery mechanisms happening in porous media under the influence of ultrasonic waves. Numerous macromodel studies have demonstrated that the emulsion has been generated at the interface of two immiscible fluids under the influence of ultrasonic waves (Wood and Loomis, 1927; Richards, 1929; Bondy and Sollner, 1935; Campbell and Long, 1949; Neduzhii, 1962; Li and Fogler, 1978; Cucheval and Chow, 2008; Ramisetty and Shyamsunder,2011; Mohammadian *et al.,* 2012). In addition, chemical flooding involves injection of a surfactant solution, which can cause the oil/aqueous interfacial tension to drop and allowing emulsification and displacement of the oil. Surfactants are the principal agents that enable oil and water to mix and are often the most expensive component in an emulsion. Estimation of solubilization parameters is a great tool in designing the economical emulsion flooding compositions. In EOR, one of the most important designing factors for chemical flood is to select an appropriate surfactant formulation capable of mobilization oil without significant surfactant losses due to adsorption and phase separation in the reservoir.An optimum condition for the oil recovery is observed when the middle phase contains the added surfactant and equal amounts of oil and water (Reed and Healy, 1977). Therefore, the effect of ultrasonic waves on the amounts of oil and water solubilized by a unit of surfactant should be investigated and the results must be compared with the case using no ultrasonic waves.

In addition, the majority of the studies on the effect of ultrasonic waves on emulsification are macroscopic and no work (microscopic) to show exactly what happens at the interface of two immiscible liquids. Therefore, it is necessary to study the effect of ultrasonic waves at the liquid-liquid interface to show the phenomena happening there by using Hele-Shaw models. Hele-Shaw experiments very accurately show what happens at the liquid-liquid interface, without the additional complexity arising from a porous (channeled) structure.

Another mechanism through which ultrasonic waves improves the recovery of oil from porous media is viscosity reduction (Duhon and Campbell, 1965; Xiao *et al.*2004; Naderi, 2008; Mohammadian *et al.*, 2012). In all of the previous studies, the viscosity was measured using either indirect methods i.e. calculating the viscosity from temperature changes or it was measured in a static condition.In other words, in spite of numerous studies, it is not yet clear that whether the viscosity reduction in ultrasonic stimulated fluids is due to the thermal effect of waves or due to other reasons. For example, Poesio *et al.* (2002) convincingly demonstrated that, the only reason for reduction of viscosity is temperature increase in the media. Mohammadian *et al.* (2012) considered viscosity reduction as one of the possible contributing mechanisms in the recovery. They further concluded that viscosity of brine and oil are reduced as a result of sonication. They also inferred that the reduction in viscosity of fluids is not solely due to heat generated as a result of sonication. Moreover, in previous researches the effects of power of waves, as an independently factor, has not been discussed on viscosity. In the other word, viscosity reduction was reported as a side effect of ultrasonic waves radiation. The area therefore could be explored further.

There are also some other oil recovery mechanisms under the influence of ultrasonic wavesin porous media such as oil coalescence, mobilization and detachment. Unfortunately, the mechanisms mentioned are almost theoretical or speculative. For example, there is no micromodel study under microscope to show oil droplets coalescence due to the Bjerknes forces (forces between the vibrating oil droplets under the influence of ultrasonic wavesthat lead to their attractions)or there is no micromodel study to show oil detachment from dead end pores (Naderi, 2008). Therefore, it is crucial to demonstrate these mechanisms in 2D glass micromodels and prove that if ultrasonic waves can cause oil droplet coalescence and detachment. If these questions answered then it is possible to improve oil recovery techniques in the field and interpret the laboratory experiments more confidently.

1.3 Research Objectives

The objective of this research can be subdivided into four (4) groups as following:

(i) To investigate the effect of ultrasonic waves on the volume of emulsion and amount of oil and water solubilized in emulsion by a unit of surfactant.

(ii) To investigate changes in the viscosity of various liquids exposed to radiation of ultrasonic waves of various power outputs and constant frequency.

(iii) To study the effect of ultrasonic waves at the liquid-liquid interface microscopically to show the mechanisms happening there using Hele-Shaw models.

(iv) To show the effect of ultrasonic waves on oil droplet coalescence, mobilization, and detachment from dead end pores, in porous media.

1.4 Scope of Study

To investigate the effect of ultrasonic waves on oil recovery mechanisms, three series of experiments were conducted.

Firstly, emulsiontests were performed to investigate the effect of ultrasonic waves (40 kHz and 500 W) on the volume of emulsion and amounts of paraffin oil and aqueous solution (a solution of the surfactant (1000 ppm AOS) and sodium chloride solutions at varying concentrations (5000, 10000, 15000, 20000, 25000, 30000 ppm) in de-ionized water) in emulsion using test tubes.All the experiments were conducted under 60 mins mechanical agitation (50 RPM) of the test tubesbyRotospin-rotary mixer inside the ultrasonic bath (40 kHz and 500 W) under three periods of ultrasonic waves radiation (0 (NUS), 15, 60 mins).

Secondly, a smooth capillary tube was employed for investigating the viscosity changes under the influence of ultrasonic waves (constant frequency of 40 kHz). Some parameters were changed such as type of fluids (synthetic oil, paraffin oil, kerosene, and brine), and ultrasonic waves power outputs (100, 250, 500 W).Pouiselle's equation was taken into account for the calculation of the viscosity. In these experiments, the process was examined for different conditionssuch as controlled and uncontrolled temperature conditions.

As a third attempt, to have a better insight into the oil recovery mechanisms a series of experiments were conducted at pore scale in Hele-Shaw and 2D glass micromodels. For the experiments using Hele-Shaw models,the emulsification mechanism at liquid-liquid interface under the influence of ultrasonic waves (40 kHz and 500 W) was investigated under microscope using two different etched thickness (500 and 26 µm) models. Some experiments were performed to show more oil recovery mechanisms under the influence of ultrasonic waves such as oil droplet coalescence, oil mobilization, and oil detachment from dead end pores under microscope in two 2Dglassmicromodels with triangular and circle patterns.The experiments were performed in different wettability conditions (oil-wet and water-wet) and flow conditions (static and dynamic).

1.5 Significance of the Study

In enhanced oil recovery (EOR) using surfactant, low interfacial tension at low surfactant concentrations, and acceptable adsorption levels are considered to be important design parameters in optimizing chemical systems for recovering trapped oil from petroleum reservoirs.In addition, surfactants are the principal agents that enable oil and water to mix and are often the most expensive component in an emulsion. Therefore, attempts should be made to increase the volume of emulsion and prevent from phase separation for a specific concentration of surfactant in order to have an economical surfactant flooding. On the other hand, emulsification is one of the oil recovery mechanisms under the effect of ultrasonic waves. Therefore, in this study the effect of ultrasonic waves on phase behavior of surfactant-brine-oil system was investigated, which is an important step in optimizing performance of emulsion systems for enhanced oil recovery.

The viscosity reduction is another oil recovery mechanismhappening under influence of ultrasonic waves. In all of the previous studies, the viscosity was measured using either indirect methods i.e. calculating the viscosity from temperature changes or it was measured in a static condition. In this study, the viscosity changes of the fluids in controlled (constant) and uncontrolled temperature conditions under influence of ultrasonic waves was investigated.

There are also some other oil recovery mechanisms under the influence of ultrasonic waves in porous media such as oil coalescence, mobilization and detachment. However, the mechanisms mentioned are almost theoretical or speculative. Therefore, in this study these mechanisms were demonstrated under influence of ultrasonic waves usingHele-Shaw models and micromodels.

In conclusion, this study contributes:

(i) To makeclarifying the oil recovery mechanisms under the influence of ultrasonic waves and

(ii) To find the factors or circumstances in which ultrasonic waves are effective for the purpose of increase in oil recovery.

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