LIGNIN FROM PAPER WASTE IN INTERFACIAL TENSION (IFT) REDUCTION FOR OIL DISPLACEMENT

VENGADESH A/L PARAMESWARAN

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ABSTRACT

This study focusses on lignin potential in reducing interfacial tension (IFT) where primary and secondary recovery only recovers minimal amount of oil in most cases and leaving behind substantial amount of oil in place unrecovered. Commercial surfactant flooding had been used to address this issue which yielded promising result at the expense of more monetary investment. The preparation of sulfonate based anionic surfactants is complex and costly. Many surfactants have been produced from edible oils, such as soybean and coconut oil, which are more expensive compared with synthetic surfactants. Some studies reported starch as a feedstock for producing surfactants but these surfactants are problematic given they would compete with food source if they became commercialized. Lignosulfonate surfactant ability in reducing the interfacial tension is yet to be tested. Lack of research regarding the effect on lignosulfonate based surfactant or co surfactant at different conditions and also whether lignosulfonate works well with alkali. Hence in case if lignin can reduce IFT then this would be one of the best method for enhanced oil recovery. Lignin extracted from paper waste is a waste product and can be procured with less cost and reduces waste at the same time. The powdered lignin based on the post-extraction treatment will be mixed with alkali to form an alkalized lignin solution. The experiment was conducted at a constant temperature of 50^0 C and ambient pressure. The temperature is representative of reservoir temperature in Malaysia with neglected pressure effect due to the known fact of IFT independency of pressure. The experiments run by using Goniometer using rising upward method. There are altogether 15 sets of run including base case with no alkalized lignin, lignin without alkalization, alkalized lignin with commercial surfactant which was to test the role of lignin as co-surfactant, lignin with varying alkali concentration and control case of only commercial surfactant. The final cases was brine with only alkali to fully understand that the IFT reduction obtained is contributed by lignin not alkali. Each measurement approximately took 2 hours to complete and gave the mixing of alkali and brine. As a matter of general comparison, there were 5 distinct results. The base case without any surfactant yield. The lignin without alkali. The critical micelle saturation (CMC) was found in this case where further lignin concentration did not cause any decrement in IFT values. The non-alkalized lignin that gives effect for the IFT reduction. Despite of producing lower IFT reduction, the significant difference in manufacturing costs of these two surfactant gave more edge to lignin as an alternative for commercial surfactant. Thus, the proven potential of lignin gave an alternative to costly surfactant and also reduce the amount of harmful waste product as well. The displacement of oil with best IFT is determined and can be applied for the oil and gas industry. Finally the potential of lignosulfonate to act as a co-surfactant also can be practiced and used widely for the benefit of the oil and gas industry.

ABSTRAK

Kajian ini menfokuskan pada potensi lignin dalam mengurangkan ketegangan antara permukaan (IFT) di mana pemulihan primer dan sekunder hanya memperoleh jumlah minyak yang minimum dalam kebanyakan kes dan meninggalkan sejumlah besar minyak di tempat yang tidak ditemui. Banjir surfaktan komersial telah digunakan untuk mengatasi masalah ini yang memberikan hasil yang memuaskan dengan mengeluarkan lebih banyak pelaburan wang. Penyediaan surfaktan anionik berasaskan sulfonat adalah kompleks dan mahal. Banyak surfaktan telah dihasilkan dari minyak yang boleh dimakan, seperti kacang soya dan minyak kelapa, yang lebih mahal jika dibandingkan dengan surfaktan sintetik. Beberapa kajian melaporkan pati sebagai bahan makanan untuk menghasilkan surfaktan tetapi surfaktan ini bermasalah kerana mereka akan bersaing dengan sumber makanan jika mereka dikomersialkan. Keupayaan surfaktan lignosulfonate dalam mengurangkan ketegangan antara permukaan masih belum diuji. Kurangnya penyelidikan mengenai kesan terhadap surfaktan berasaskan lignosulfonat atau surfaktan bersama pada keadaan yang berbeza dan juga sama ada lignosulfonat berfungsi dengan baik dengan alkali. Oleh itu sekiranya lignin dapat mengurangkan ketegangan maka ini akan menjadi salah satu kaedah terbaik untuk pemulihan minyak yang dipertingkatkan. Lignin yang diekstrak dari sisa kertas adalah produk buangan dan dapat diperoleh dengan kos yang lebih rendah dan mengurangkan sisa pada masa yang sama. Lignin serbuk berdasarkan rawatan pasca pengekstrakan akan dicampurkan dengan alkali untuk membentuk larutan lignin beralkali. Eksperimen ini dijalankan pada suhu tetap 500 C dan tekanan persekitaran. Suhu mewakili suhu takungan di Malaysia dengan kesan tekanan yang diabaikan kerana fakta yang diketahui mengenai kebebasan tekanan IFT. Eksperimen dijalankan dengan menggunakan Goniometer menggunakan kaedah menaik ke atas. Terdapat kesemuanya 15 set run termasuk kes asas tanpa lignin alkali, lignin tanpa alkalisasi, lignin beralkali dengan surfaktan komersial yang bertujuan untuk menguji peranan lignin sebagai co-surfaktan, lignin dengan kepekatan alkali yang berbeza dan kes kawalan hanya surfaktan komersial. Kes terakhir adalah air garam dengan hanya alkali untuk memahami sepenuhnya bahawa pengurangan IFT yang diperoleh disumbang oleh lignin bukan alkali. Setiap pengukuran kira-kira mengambil masa 2 jam untuk diselesaikan dan memberikan pencampuran alkali dan air garam. Sebagai perbandingan umum, terdapat 5 hasil yang berbeza. Sarung asas tanpa hasil surfaktan. Lignin tanpa alkali. Ketepuan misel kritikal (CMC) dijumpai dalam kes ini di mana kepekatan lignin selanjutnya tidak menyebabkan penurunan nilai IFT. Lignin tanpa alkali yang memberi kesan kepada pengurangan IFT. Walaupun menghasilkan pengurangan IFT yang lebih rendah, perbezaan ketara dalam kos pembuatan kedua surfaktan ini memberi kelebihan kepada lignin sebagai alternatif untuk surfaktan komersial. Oleh itu, potensi lignin yang terbukti memberikan alternatif kepada surfaktan yang mahal dan juga mengurangkan jumlah produk sisa berbahaya. Perubahan minyak dengan IFT terbaik ditentukan dan boleh digunakan untuk industri minyak dan gas. Akhirnya potensi lignosulfonate untuk bertindak sebagai surfaktan bersama juga dapat diamalkan dan digunakan secara meluas untuk kepentingan industri minyak dan gas.

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CHAPTER 1

INTRODUCTION

1.0 Background Study

By nature crude oil is a limited resource. Thus, the amount of crude oil available has to meet the current worldwide demands. From time to time, oil production has been intentionally reduced, and this has resulted in serious oil crises accompanied by a general increase in the oil price. Hence, this turn has forced the oil industry to recover oil from more complicated areas in the world, where the oil is less accessible meaning where the recovery techniques are constantly advanced. This has contributed to the development of techniques for enhanced oil recovery, (EOR), which while used today, also constantly undergo further advancement and development. Up to two third of the world crude oil remains trapped in the reservoirs after the primary and secondary recovery in an average of an oil reservoir, (Rosen et al., 2005).

EOR is then required to optimize the depletion, as the remaining oil is trapped in the pore structure inside the reservoir. EOR covers several different advanced recovery techniques, which will be introduced in this chapter. The focus for this thesis has been on the phase behavior properties inside the reservoir in connection towards the surfactant flooding and oil/ brine systems. The phase behavior in the surfactant system is overall the most important factor determining the success of a chemical flood [Skauge and Fotland, 1990]. Currently, there are no adequate models (such as equations of state) to describe phase behavior in such systems.

In the industry , extraction of crude oil is segregated into 3 stages which are primary recovery, secondary recovery and tertiary recovery or enhanced oil recovery. Primary recovery depends on natural pressure drive to transport the oil to the surface and secondary recovery is done with the aid of water and gas injection to produce the oil since the natural pressure drive is no longer sufficient. According to Farouq and Stahl

(1970), huge amount of petroleum resource cannot be recovered by conventional methods, the primary recovery recovers up to 10 % of the Oil Originally In Place (OOIP) and secondary recovery recovers around 25% of the OOIP. New strategies are needed to supply the vastly growing energy industry when the conventional method are no longer reliable and due to decrease in production rate. Enhanced Oil Recovery (EOR) is the usage of various techniques to increase the amount of crude oil that can be produced . EOR is done to remove remaining oil from complex reservoir when the primary and secondary recovery are no longer feasible.

The aspiration of operators of maturing waterflooding fields is to maximize the profitable recovery of the remaining oil after waterflooding at the lowest investment cost possible. This requires both the optimization of the reservoir sweep efficiency and to put physical, chemical, or thermal mechanisms at work that will improve the overall microscopic oil displacement. Enhanced oil recovery (EOR) methods can be divided into thermal methods (e.g., steam methods) and nonthermal methods. Nonthermal methods include in chemical methods (e.g., designer water, polymer flooding, alkali/surfactant/polymer (ASP) flooding, surfactant flooding) and nonchemical methods (e.g., miscible or immiscible gasflooding). Shale oil are unconventional oil produced from oil shale rock fragment which are rocks containing kerogen which are extracted through several processes such as pyrolysis , hydrogenation or thermal dissolution . Basically shale rocks have complicated pore structures and has low permeability and porosity . This is whereby surfactant flooding technique is being implemented.

To define EOR methods in a better physical context, we can recall that hydrocarbons are trapped in the pores either by an unfavorable viscosity ratio or by the capillary forces acting on different scales. For instance, gasflooding or wasterflooding (CO_2, N_2, etc.) with a high oil viscosity leads to an unfavorable mobility ratio between the displacing and displaced fluid. A large fraction of the oil is not contacted by the injected fluid[1] and the oil that is contacted is poorly displaced. The rock/fluid interactions are responsible for the adhesion of fluids to the porous medium, which can be oil-wet, water-wet, or mixed-wet. In mixed- or oil-wet formations, oil is retained in the pores due to the affinity of the oil to the rock. Capillarity makes it difficult to mobilize oil blobs so that they can be displaced over macroscopic distances (Figure 1.1).Waterflooding in light oil reservoir can have a high reservoir sweep efficiency. The sweep efficiency deteriorates as the viscosity of the oil increases.

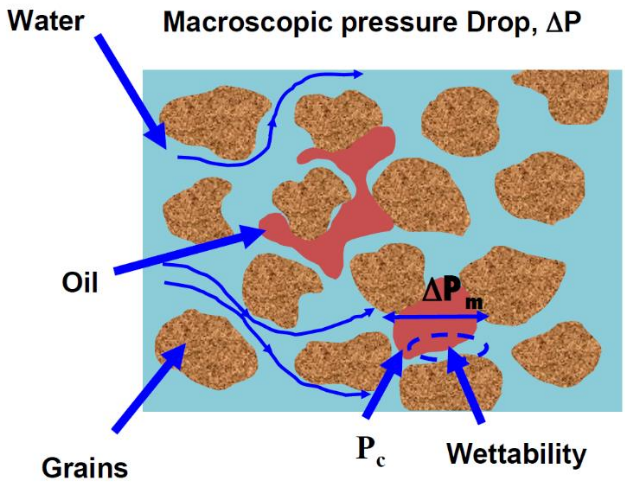

Figure 1.1 - Microscopic trapping of oil in rocks.

The simplest for the nonthermal oil recovery is designer of the water; i.e., the manipulation of the mineral composition of the drive water to achieve a high microscopic displacement efficiency. In the early days of this technique, low-salinity field trials were reported by BP (Seccombe et al. 2008), and since then study of the designer water has grown to be high interest to the industry. Low salinity implies that low ionic strength, especially the divalent ions. Designer water may mean increase in concentration of certain ions and reduction in composition of other ions. The fundamental mechanisms for the improvement of the oil displacement are far from well understood, and there has been debate about the true benefits. It can be said there is sufficient evidence that can assume the modification of the rock surface for it to be a responsible factor for the improved microscopic displacement. There are many research, in the combining surface chemistry and rock plus fluid interactions, which are needed in detail to know the mechanism and assert whether further gain can be obtained from the application of the designer water.

The term surfactant comes from surface-active agent, and surfactants are chemical compounds utilized to reduce the IFT between two different phases by adsorbing on a surface or a fluid–fluid interface. Surfactants are extensively used chemicals having various EOR applications due to their significance in IFTs reduction and their capability in changing wetting properties (Green and Willhite 1998). Surfactants can be known as amphiphilic or also amphipathic molecules which contain a polar (hydrophilic) portion and also a nonpolar (hydrophobic or hydrocarbon loving) portion. The origin of the term amphiphilic comes from the Greek word “amphi,” meaning “both,” and this describes the fact that all surfactant molecules have at least two parts, the hydrophilic part which is soluble in a certain specific fluid, example such as water, and the hydrophobic part which is insoluble in water (Tadros 2014).

According to some research the nature of the hydrophilic head group, surfactants can be classified into different types, and this classification of surfactants are made based on the charges of the polar head group of the surfactant molecule. Surfactants can be divided into few classes called anionics (negative charge), cationics (positive charge), nonionics (no charge), and zwitterionics (negative and positive charge) (Bera and Belhaj 2016; Tadros 2014).

It is well accepted that oil recovery efficiency can be improved by obtaining ultralow interfacial tension between oil and water by adsorption at the interface. The flow of trapped oil droplets or ganglia through the narrow necks of pores is illustrated schematically in Figure 1.2 (Donaldson et al., 1989)

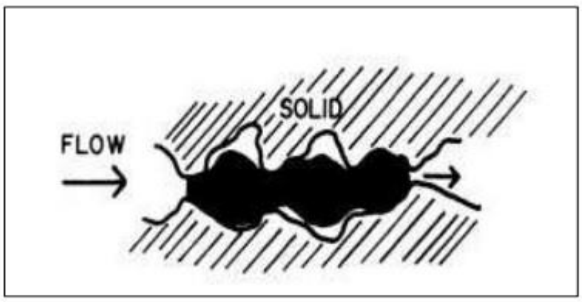

Figure 1.2: The effect of IFT on the movement of oil ganglia through the narrow necks of pores (Donaldson et al., 1989)

An ultra-low interfacial tension (often less than 10-3 dyne/cm) between oil and water phases is required for easy the flow of trapped oil drops since it reduces the deformation work needed for oil ganglia to move through the narrow necks of pore 9 channels (Donaldson et al., 1989). Foster (1973) and Hill, Reisberg and Stegemeier (1973) observed that relatively small concentrations of petroleum sulfonates can produce such low interfacial tension between oil and water. Researchers found that the IFT of an oil–water–surfactant system is a function of salinity, oil composition, surfactant type and concentration, cosurfactant, electrolytes, temperature, and the phase behavior of the system (Adkins et al., 2012).

• Influence of salinity on IFT

Winsor (1954) recognized three types of phase equilibria in microemulsion phase as type I, type II, and type III. Healy and Reed (1974) explained how the Winsor-type behavior describes the change in phase behavior, solubilization of oil and water and IFT as a function of salinity for anionic surfactants. The oil–water–surfactant system is strongly affected by the water salinity. This phase behavior is represented by a ternary diagram as shown in the Figure 1.3:

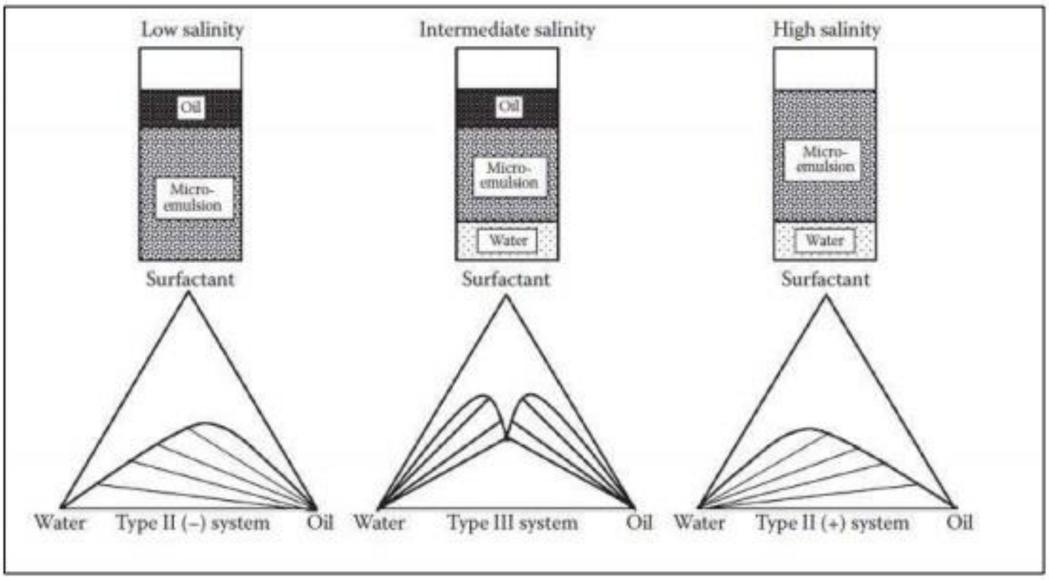

Figure 1.3: Three types of microemulsions and the effect of salinity on phase behavior (Healy and Reed, 1974)

The surfactant flood exhibit good aqueous phase solubility and poor oleic phase solubility in case of low brine salinities, thus forming type I phase behavior. In type I system, an oil-in-water microemulsion is formed, and the surfactant remains in the aqueous phase (Schramm et al., 2000). This system is referred to as the lower phase microemulsion or type II (-) system, where II means no more than two phases can form and (-) means that the tie-lines have negative slope. This phase behavior is not favorable to achieve ultralow IFT. A water-in-oil microemulsion with an excess oil phase is defined as the upper phase microemulsion or type II (+). This behavior leads to the retention of surfactants in the oil phase and is not favored in EOR. In a type III microemulsion, the surfactant forms a microemulsion in a separate phase between the oil and aqueous phases. This phase forms a continuous layer containing surfactant, water and dissolved hydrocarbons. Usually, type III provides low IFT especially when equal volumes of water and oil are solubilized in the microemulsion. This condition is defined as optimal salinity, which exhibits the lowest IFT between the water and the oil. In addition, optimal salinity can be expressed as the midpoint salinity where IFT between microemulsion and water and that between microemulsion and oil are more or less the same. Type III system is desirable for EOR processes (Aoudia et al., 1995).

1.2 Problem Statement

Several parameters would affect the interfacial tension such as the temperature, pressure, salinity. Although surfactants are mostly used in industry, they also have some negative role. For example, the preparation of sulfonate based anionic surfactants is complex and costly. Adding on, there are some applications have very specific requirements that can be difficult to achieve with currently available surfactants. In short can be said, surfactants must be tailored according to the properties of the oil reservoir, which varies based on their fractures, matrix permeability, dead-end pore volume, hardness, salinity, and mineral heterogeneity for enhanced oil recovery (M. Mushtaq, I. Tan, M. Sagir,2014). Due to their different structures and properties, several reaction pathways and feedstocks can be used for producing more environmentally friendly surfactant for replacing these oil-based surfactants.(J.Sheng,2010)

However, lignin is a chemically complex biopolymer, which makes its utilization challenging.(A. Jönsson, A. Nordin, O. Wallberg,2008) The variation and complexity in chemical structure of lignin may impact its conversion to other value-added products.(A. Ragauskas, G. Beckham, et al,2014) For these reasons, lignin currently has a limited commercial value. In the past, lignin has been mainly used as a fuel source, but recent studies have shown that it could be converted to many value-added derivatives. (K. Hazarika, S. Gogoi,2014). Hence lignin can be extracted from paper waste and be used to reduce the interfacial tension in the enhanced oil recovery process. There a lot of paper mills factories that are just throwing away the waste without knowing the benefits of it. Thus, with a proper and efficient study will ensure the benefits of lignin from this paper waste to be used widely in the oil and gas industry.

Many studies have investigated the potential use of natural surfactants. It is challenging, however, to produce low-cost, multifunctional, biodegradable and effective surfactants from biomass.(A.Olajire,2014) In this case, many surfactants have been produced from edible oils, such as soybean and coconut oils, which are more expensive compared with synthetic surfactants.(K.A. Elraies, I.M. Tan,2012) Some studies reported starch as a feedstock for producing surfactants,(W.S. Perkins,1998) but these surfactants are problematic given they would compete with food source if they became commercialized.(J. Merta, P. Stenius,1999) Lignin, however, has been suggested to be a potentially desirable material for surfactant production due to its ready availability and low costs. (K. Askvik,2001).Lignin is ranked second after cellulose in availbility as a natural polymer on earth and can be utilized to produce value-added products. (D. Watkins, M. Nuruddin, M. Hosur, A. Tcherbi-Narteh, S. Jeelani,2015) Using lignin as a feedstock for producing value-added products has some advantages and disadvantages.(L. Hu, H. Pan, Y. Zhou, M. Zhang,2011) In its favor, lignin is not used as food by human and it has an aromatic hydrophobic ring. (A. Tejado, C. Peña, et.al,2007)

Lignosulfonate surfactant ability in reducing the interfacial tension is yet to be tested. The usage of surfactant in a reservoir has some economic constraint since the cost of surfactants are high. There are also lack of research regarding the effect on lignosulfonate based surfactant or co surfactant at different conditions and also whether lignosulfonate works well with alkali .

1.5 Hypothesis

1. Lignin will be able to reduce IFT.
2. The performance of lignin can also be increased with the addition of alkali.
3. Lignin acts as a good co-surfactatnt to reduce IFT.
4. Higher concentration of lignosulfonate gives a good oil displacement.

1.6 Objectives

The main objective are :

1. To extract the lignin compound from paper waste and characterize it.

2. To determine the critical micelle concentration (CMC) of the lignosulfonate concentration.

3. To determine the recovery of oil with different concentration of lignosulfonate compared to the currently being used surfactant.

4. To investigate the potential of lignosulfonate compound from paper waste to reduce the interfacial tension.

5. To determine the best oil displacement.

1.7 Scope of Study

1. Preparing and characterizing lignin compound from paper waste industry to be used in reducing the interfacial tension later on in this experiment.

2. Measuring the interfacial tension using different alkali concentration to determine whether it gives a positive outcome on the IFT or not.

3. Identifying the CMC point and plot the graph to see the best interfacial tension point among all the concentrations of substance being used.

4. Preparing the artificial sandpack by using the almost same size sand to find the displacement of oil using different substance that the IFT have been tested.

5. Observing the reaction of lignosulphate as co-surfactant with Sodium Dodecyl Sulfate (SDS) which is a commercial surfactant to see whether it gives a better outcome on the interfacial tension or not.

CHAPTER 2

LITERATURE REVIEW

2.1 Enhanced Oil Recovery

Enhanced oil recovery (EOR) is actually increasing for the usage in the oil industry and several different technologies have also emerged during, the last decades in order to optimize oil recovery after certain recovery methods have been applied. Surfactant flooding is an EOR technique in which the phase behavior inside the reservoir can be manipulated by the injection of surfactants and co-surfactants, creating advantageous conditions in order to mobilize trapped oil. Correctly designed surfactant systems

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