



## Emulsion flooding for enhanced oil recovery: Interactive optimization of phase behavior, microvisual and core-flood experiments



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### ABSTRACT

Emulsion flooding has been proved to be an effective chemical enhanced oil recovery (CEOR) method. The performance of this technique is strongly dependant on the formulation of the emulsion slug. To achieve the best formulation, different approaches have been introduced in the literature. Nevertheless, a systematic workflow containing the method(s) of the design of experiments (DOEs) has not been reported as yet. In this paper, we developed a workflow which is comprised mainly of three stages. In the first place, the phase behavior experiments of sodium dodecyl sulfate (SDS)/water/diesel (as an efficient and economic hydrocarbon phase)/salt system were carried out using response surface methodology (RSM) to model and optimize the emulsification process. The second stage was followed by the characterization of optimum formulation in terms of rheological behavior and particle size distribution. Finally, microvisual and core-flood displacement tests were performed to evaluate the efficacy of emulsion flooding to recover the residual oil bypassed or trapped after water flooding. This paper presents the results of experiments done in different stages of the proposed workflow. The results demonstrate the high potential of emulsion formulated systematically by DOE approach to increase oil recovery factor.

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### Introduction

In conventional petroleum reservoirs, maximum oil production by natural-drive (primary) mechanisms and consequently pressure maintenance (secondary) methods is merely 20–60% original oil in place (OOIP); in other words, nearly  $2.0 \times 10^{12}$  barrels of conventional oil, which is the main objective of various enhanced oil recovery (EOR) methods [1–4]. Poor sweep efficiency as one of the most underlying reasons of this issue contributes to bypassing the substantial amount of oil in reservoirs. Weak macroscopic displacement efficiency contributes to unfavorable mobility ratio between displacing and displaced fluids as well as reservoir heterogeneity.

From pore-scale standpoint, capillary forces are the factor most responsible to trap oil ganglia in pore space which causes inefficient microscopic displacement efficiency and consequently

entraps an immense amount of oil in the pay zone. Capillary forces become dominant when the pores constrain the water–oil interface to a high degree of curvature. By contrast, viscous forces, i.e. drag forces produced by the fluid flow of continuous phase, behave against capillary forces so that they can overcome them and displace trapped oil. The ratio of viscous to capillary forces which is known as capillary number determines how strongly trapped oil is within a specific porous medium [5].

To mobilize the oil bypassed and/or trapped in the reservoir, effective mobility ratio of displacing fluid and/or significant reduction of interfacial tension between residual oil and displacing fluid are required. For this purpose, injection of emulsions has been recognized as a potentially efficient chemical EOR process. Emulsion flooding refers to a slug comprised of stable solution of hydrocarbon, brine and one or more surfactants which is injected into the reservoir to mobilize or solubilize the remaining oil [6–9].

Emulsions (macroemulsions) are dispersion of one immiscible liquid inside another continuous liquid phase. They are kinetically stable and are not formed spontaneously; that is to say they

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require external energy to be produced [10–12]. According to the concept of capillary number, emulsion flooding may be a unique chemical EOR approach since both favorable microscopic and macroscopic displacement efficiencies are provided simultaneously. In other words, reduction of interfacial energy induced by emulsion phase as well as non-Newtonian behavior of displacing front are available concurrently. Therefore, emulsion flooding has become one of the promising chemical EOR techniques in recent years [13–15].

Selection of hydrocarbon phase has been one of the major challenges of emulsion/microemulsion flooding. It is of vital importance as a large volume of emulsion slug is necessary to be injected into the reservoir. Accordingly, to select and also provide the hydrocarbon phase, it is imperative that different criteria be considered such as: stability at reservoir conditions, high efficiency, prevention of formation damage through appropriate particle size distribution, availability, cost effectiveness and environmental compatibility as well. Regarding these factors, different sources have been proposed in the literature: CO<sub>2</sub> [16] and supercritical CO<sub>2</sub> [17], gear oil [18], used engine oil [19], palm kernel oil [20], and residual oil in place [9].

In this work, diesel has been suggested as the hydrocarbon phase with outstanding performance. The objective of current study is to investigate the EOR efficiency of emulsion flooding prepared by diesel. For this purpose, a 3-stage workflow was used: first of all, phase behavior tests were systematically planned using design of experiments (DOEs). Then, optimum formulation of emulsion was selected according to statistical analysis of the responses of designed experiments. Following this, it was characterized as the second stage in terms of rheological behavior and particle size distribution. Finally, the optimum mixtures were injected into a micromodel fabricated using a developed compositional method as well as a carbonate core in order to evaluate the efficiency of emulsion flooding for chemical EOR.

Phase behavior study of surfactant/hydrocarbon/brine is a set of tests to screen and optimize the formulation of emulsion slug and then to interpret the amount of oil recovered by optimized emulsion flooding. It is the single most critical factor assuring the success of any chemical flooding containing surface active components [21–23]. Generally, it is controlled by several parameters such as the properties of the surfactant as well as the oil phase [24–27], addition of electrolyte [28,29] especially divalent ions [25], presence of co-surfactant or co-solvent [25,30], water/oil ratio (WOR) [31], temperature [32–34], pH [35], and pressure [36–38]. A systematic change in effective parameters during formulation can result in the most stable emulsion (optimum mixture) being injected into candidate reservoir.

Of the vast number of investigations on the surfactant phase behavior, only a few researches have been focused on the systematic interaction of parameters. In this study, response surface methodology (RSM) has been proposed to study the phase behavior of sodium dodecyl sulfate (SDS)/brine/diesel system based on the interaction of selected parameters in addition to their single effects. RSM consists of a collection of mathematical and statistical techniques. It can be well utilized when a response or a set of responses of interest are influenced by several associated variables. As a statistically designed experimental protocol, it is used for designing experiments, developing functional relationship between a response and a number of related factors through analyzing their interactions and optimizing the levels of these factors to attain the best process performance [39–42].

## Background of emulsion flooding

McAuliffe was apparently the first person to propose dilute and stable emulsions as mobility control agents for enhanced oil

recovery (EOR) and from then on many studies have been conducted, especially in recent years to achieve a better description of this process [43]. Abdul and Farouq Ali combined an emulsion slug as blockage agent and a polymer solution as mobility control agent to modify the drawbacks of water flooding in reservoirs suffering from water leg problems [44]. Cobos et al. carried out the flow experiments of oil-in-water emulsions in quartz micro-capillary tubes. The pore-throat mechanism of blockage was observed in experiments, and therefore changes in local fluid mobility would explain additional oil recovery obtained by emulsion flooding [45]. Mandal et al. characterized the oil-water emulsions produced by gear oil as hydrocarbon phase using physicochemical properties and size distribution of dispersed oil in water phase [18].

Rocha De Farias et al. investigated the potential of oil-water emulsions for EOR applications of viscous oils. The hydrocarbon phase was the residual oil in place. They compared emulsion flooding with both water flooding and surfactant flooding [9]. Fu and Mamora studied EOR application of emulsions generated by engine oil as hydrocarbon phase [19]. Guillen et al. carried out pore scale flow visualization experiments as well as core flooding tests in parallel sandstone cores having different permeabilities to understand mechanism(s) of incremental oil recovery achieved by emulsion injection. The results showed emulsion flooding offers multi-scale mechanisms which are microscopic displacement efficiency and conformance sweep efficiency [13]. Guillen et al. introduced a selective mobility control mechanism of emulsion droplets driven by capillary forces so that strong interfacial forces (low capillary number) have significant effect in lowering down the mobility of emulsion containing larger drops compared to the diameter of throats. On the other hand, displacing fluid mobility is only slightly influenced by emulsion drops in high capillary numbers [46]. Moradi et al. evaluated the efficiency of emulsion injection to improve oil recovery. Furthermore, single phase flow experiments were carried for better understanding of emulsion flow in pore level. They showed blockage-release mechanism for pore scale dynamics which is dependent on the emulsion drop size as well as local capillary number [15].

## Experimental

### Materials

In general, there are at least four compositional variables in an emulsion flood: water, salt, hydrocarbon as well as emulsifying agent. Distilled water was provided by reverse osmosis method (New Human Power I). The brine was prepared by dissolution of sodium chloride (NaCl, Merck Co.) salt in distilled water. Diesel was proposed as hydrocarbon phase. At ambient temperature (about 25 °C), its density was 0.8394 g/ml and its viscosity (DBRobinson electro-magnetic viscometer) was measured as 5.15 cP.

An anionic surfactant, sodium dodecyl sulfate (SDS, purity > 85%) was used for emulsification process. It was received from Merck Co. with molecular weight of 288.38 g. Critical micelle concentration (CMC) of this surfactant was reported as  $8 \times 10^{-3}$  M [47]. Its surface tension at CMC and ambient temperature (about 25 °C) was measured as 34.6 mN/m using Du Noüy ring method (Krüss K10). Emulsion slugs of flooding experiments were prepared according to the optimum formulation resulted from RSM.

Crude oil for dynamic displacement experiments was prepared from an Iranian offshore oilfield in the Persian Gulf. Its properties were °API of 30.9, density of 0.8675 g/ml, and viscosity of 19.0 cP at ambient temperature (about 25 °C). The basic fluids for water flooding in microvisual experiments and core-flood test were distilled water and synthetic brine (distilled water containing

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