



# Mechanistic understanding of chemical flooding in swelling porous media using a bio-based nonionic surfactant



Aghil Moslemizadeh <sup>a,\*</sup>, Abbas Shirmardi Dezaki <sup>a</sup>, Seyed Reza Shadizadeh <sup>b</sup>

<sup>a</sup> Department of Petroleum Engineering, Ahwaz Faculty of Petroleum Engineering, Petroleum University of Technology, Ahwaz 63134, Iran

<sup>b</sup> Department of Petroleum Engineering, Abadan Faculty of Petroleum Engineering, Petroleum University of Technology, Abadan, Iran

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## ABSTRACT

A large quantity of world's oil reserves is stored in sandstone reservoirs where clay swelling phenomenon, owing to its negative impact on reservoir quality, imposes a challenge on applicability of surfactant flooding, a subset of chemical enhanced oil recovery (EOR). Therefore, surfactant flooding in swelling porous media should be carried out with more sensitivity. In this study, a great attempt was made to mechanistically understand the performance of a bio-based nonionic surfactant named Zizyphus Spina Christi leaf extract (ZSCLE) in EOR process from swelling porous media. This purpose was achieved through determination of linear swelling, viscosity, pore plugging index, oil recovery, zeta potential, particle size as well as FT-IR analysis. Based on the results, the capability of montmorillonite (Mt, familiar swelling clay) to plug the pores was lost upon getting exposed to ZSCLE aqueous solution, a finding which is promising for surfactant flooding in swelling porous media, especially in the cases where high brine salinity is limited. In contrast to water flooding, ZSCLE was able to do a uniform sweeping with an improvement of sweep efficiency at about 10.72%. It was concluded that in addition to interfacial tension reduction and lowering of the mobility ratio, the increasing of the pore connectivity is another key parameter which is strongly effective in oil recovery improvement using ZSCLE. The adsorbed ZSCLE on Mt particles resulted in a significant decrease in the absolute magnitude of zeta potential and an increase in particle size. From FT-IR analysis, some new peaks were detected in infrared spectra of ZSCLE-modified Mt. These indications suggest the interaction between hydrophilic head of ZSCLE and oxygen atoms available on the surface of Mt. This study presents a new criterion for selecting appropriate surfactants so as to stimulate the reservoirs rich in active clays.

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## 1. Introduction

Oil recovery process is generally categorized into three different stages comprising primary, secondary and tertiary oil recovery [1]. Primary oil recovery refers to the stage in which oil production relies on the native energy of the reservoir. After this stage, the reservoir pressure reduces as it is insufficient to produce oil; thereby, it is necessary to implement the secondary oil recovery stage which includes applying an external energy (e.g. water flooding) into the reservoir for the purpose of pressure maintenance and oil displacement towards the production wells. Unfortunately, the first two stages of oil recovery are not quite efficient since a large quantity of oil always remains in reservoirs due to capillary forces as well as mobility issues. The residual oil of this stage is often the target of tertiary oil recovery or enhanced oil recovery (EOR). Actually, EOR is a general term for any technique used to increase oil production after the primary and the secondary oil recovery processes [2]. It is necessary that the extraction of the residual oil is quite

necessary to respond to the present high rate energy demand of the world. This issue necessitates the implementation of EOR techniques such as chemical flooding, gas flooding and thermal recovery [3]. The main goal of these techniques is to increase the volumetric sweep efficiency and enhance the displacement efficiency [4]. Chemical EOR processes are helpful in many reservoirs. Surfactant flooding is a chemical EOR method during which a slug of surfactant is injected into the reservoir from one or several injection wells in a special pattern [5]. In this process, surfactants decrease the existing interfacial tension (IFT) between oil and aqueous phase, leading to lower capillary forces, flowing the trapped oil bank, lower residual oil saturation and eventually higher ultimate oil recovery [6–8].

Clay swelling phenomenon which comes from the interaction of active clays with aqueous phase can strongly influence the efficacy of chemical flooding by its adverse impact on reservoir quality and ultimately oil recovery [9,10]. This phenomenon is more acute in reservoirs with smaller porosity and higher clay content [11]. Hence, it is imperative to consider the issue of formation damage due to clay swelling in chemical EOR scheme. In much simpler words, all the fluids or chemicals that are injected into the clay containing reservoirs must be checked for compatibility. One of the relevant studies

\* Corresponding author at: Ahwaz Faculty of Petroleum Engineering, Petroleum University of Technology, P. O. BOX 63134, Ahwaz, Iran.

E-mail address: [a.moslemizadeh.put@gmail.com](mailto:a.moslemizadeh.put@gmail.com) (A. Moslemizadeh).

in this area is the work of Kazempour et al. [12] who investigated the role of active clays on alkaline-surfactant-polymer (ASP) performance. They stated that the performance of ASP flooding is significantly affected by active clays such as smectite. The evaluation of fluid-rock interaction has strongly been recommended in their work. Most recently, Moslemizadeh et al. [13] assessed the performance of Mulberry leaves extract for improving oil recovery from reservoirs rich in active clays. Despite showing the high capability of active clays in oil entrapment, they concluded that clay swelling cannot be problematic in the case of this surfactant because it can potentially inhibit the swelling of active clays such as smectite.

The major restrictions of synthetic surfactants are their cost and environmental concerns which limit their use. An alternative for synthetic surfactants is bio-based ones which are both environmentally friendly and cost effective. Zizyphus Spina Christi leaf extract (ZSCLE) is a bio-based surfactant which has attracted increasing attention in petroleum industry both in EOR and drilling areas [14–24]. Nevertheless, the performance of this surfactant on oil recovery from swelling porous media has not yet been investigated in literature. This article assesses for the first time the performance and mechanism of ZSCLE in surfactant flooding process in reservoirs rich in montmorillonite (Mt, familiar swelling clay). To this end, first, the tests including linear swelling, viscosity measurements and pore plugging are carried out to examine the interaction between ZSCLE aqueous solution and Mt. Second, water flooding with and without ZSCLE into the heterogeneous swelling porous media is performed to evaluate the performance of ZSCLE on oil recovery factor. As the final step, the tests comprising particle size distribution, zeta potential measurements and FT-IR analysis are conducted to clarify the interaction mechanism between ZSCLE and Mt. The results obtained from this study are presented and discussed in greater details throughout the paper. This investigation is quite instructive because it offers a new criterion for selecting appropriate surfactants in stimulation of the reservoirs rich in active clays.

## 2. Experimental section

### 2.1. Materials

#### 2.1.1. Surfactant

Surfactants, surface active agents, are usually organic compounds that are containing both hydrophobic (tails) and hydrophilic (heads) groups. Therefore, a surfactant molecule contains both water insoluble and water soluble components. Based on the nature of the hydrophilic head, surfactants are classified into four groups including anionic, cationic, zwitterionic, and nonionic [25]. Anionic and cationic surfactants bear a negative and a positive charge on the hydrophilic heads, respectively. On the other hand, zwitterionic surfactants have both a negative and a positive charge. Nonionic surfactants bear no apparent ionic charge; however, the hydrophilic part is soluble in water because of polar groups. These groups can be hydroxyl (OH) or polyethylene oxides  $(OCH_2CH_2)_2$  [26]. Nonionic surfactants have several fascinating aspects compared to other surfactants including inexpensively, compatibility with most other chemicals, better control properties [27], less toxicity, and higher biodegradation potential [28]. Therefore, more effective surfactants with much broader applicability can be obtained from nonionic surfactants alone or by combining them with other surfactants [27].

Zizyphus Spina Christi commonly known as Christ's Thorn Jujube is a deciduous tree with light-grey and very cracked bark. Tropical and subtropical regions are the most suitable areas for growing this tree [29]. It is typically found in Jordan, Iran, Iraq and Egypt [30]. In Iran, different regions, especially south regions, are dedicated for cultivating this tree. Humans have employed the leaves of this plant, which are locally known as "Sedr" and "Konar", for washing the hair and body. It has been reported that three cyclopeptide alkaloids, four Saponin glycosides, and several avonoids can be extracted from the leaves of Zizyphus

Spina Christi [31]. Saponins are bio-surfactants mainly produced by plants and less frequently by marine organism and insects [32]. These bio-surfactants could present in >500 plant species [33,34]. The leaves of Zizyphus Spina Christi are rich in Saponins [30]. It is important to consider that molecules of Saponins (Fig. 1) contain both hydrophobic and hydrophilic parts much the same as the synthetic surfactants. The hydrophobic part contains a triterpenoid, steroid or steroid backbone, while the hydrophilic part consists of several saccharide residues attached to the hydrophobic scaffold via glycoside bonds [35].

For the purpose of this study, leaves of the Zizyphus Spina Christi were hand-picked from the cultivated trees in Petroleum University of Technology campus, Ahwaz, southern Iran. Then, Saponin which is a biodegradable nonionic surfactant was extracted from the collected leaves by spray dryer method [36–38]. The extracted powder, Zizyphus Spina Christi leaf extract (ZSCLE), was utilized in this study. The properties of ZSCLE are given in Table 1.

#### 2.1.2. Swelling clay

Typical smectite clay, montmorillonite (Mt), with cation exchange capacity of 70.5 meq/100 g characterized by methylene blue test was utilized in this study. The raw sample was characterized by X-ray diffraction (XRD). The test analysis revealed that Mt content is about 65.5%, while other minerals are quartz 12%, cristobalite 10%, muscovite 0.5%, gypsum 0.5%, anorthite 11.5%.

#### 2.1.3. Oil

In the present study, the oil sample was taken from one of the Lavan's oil fields, Lavan Island, southern of Iran. The general properties of the utilized oil are given in Table 2.

## 2.2. Methods

### 2.2.1. Preparation of surfactant solutions

In this study, homogeneous surfactant aqueous solution was prepared by mixing a certain amount of ZSCLE in deionized water using magnetic stirrer. The powder of ZSCLE needs to be slowly added to vortex of deionized water. After dissolving the whole of ZSCLE, the stirrer speed was retarded and solution was stirred for 2 h. In the cases where various concentrations of ZSCLE aqueous solution are required, master ZSCLE solution was first prepared and then lower concentrations were achieved by appropriate dilution of it.

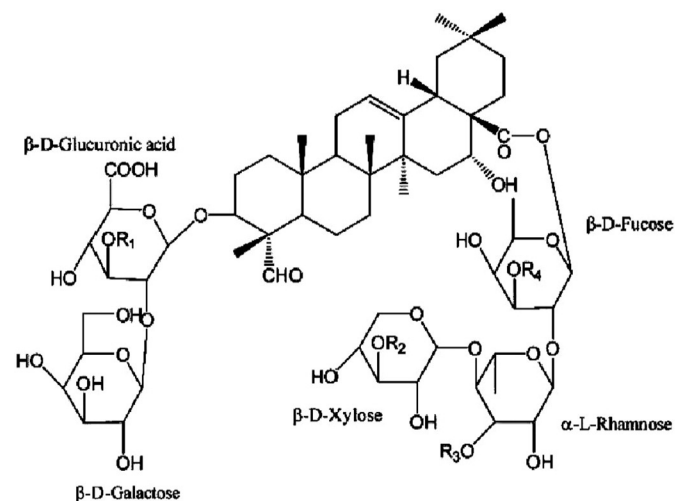


Fig. 1. General chemical structure of saponins. The notation R1–R4 represents either H or various sugar groups [16].

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