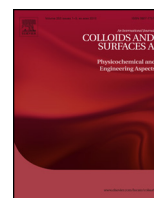




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# Colloids and Surfaces A: Physicochemical and Engineering Aspects

journal homepage: [www.elsevier.com/locate/colsurfa](http://www.elsevier.com/locate/colsurfa)



## The synergistic effects of anionic and cationic chemical surfactants, and bacterial solution on wettability alteration of carbonate rock: An experimental investigation

Farzad Hajibagheri<sup>a</sup>, Mostafa Lashkarbolooki<sup>b,\*</sup>, Shahab Ayatollahi<sup>c</sup>,  
Abdolnabi Hashemi<sup>d</sup>

<sup>a</sup> Enhanced Oil Recovery (EOR) Research Centre, School of Chemical and Petroleum Engineering, Shiraz University, Shiraz, Iran

<sup>b</sup> School of Chemical Engineering, Babol University of Technology, Babol, Iran

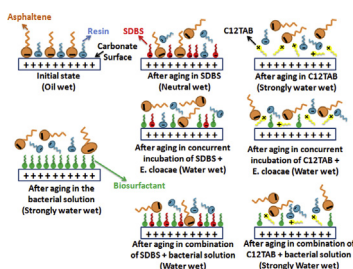
<sup>c</sup> School of Chemical and Petroleum Engineering, Sharif University of Technology, Tehran, Iran

<sup>d</sup> Department of Petroleum Engineering, Petroleum University of Technology, Ahwaz, Iran

### HIGHLIGHTS

- The synergistic effects of chemical surfactants and *E. cloacae* are investigated.
- Wettability alteration based on contact angle measurement is performed.
- The bacterial solution desorbs asphaltene and resin from the carbonate rock surface.
- The presence of the bacterial solution not play out only as a sacrificial agent.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 27 September 2016  
Received in revised form 27 October 2016  
Accepted 3 November 2016  
Available online xxx

#### Keywords:

SDBS  
C12TAB  
Biosurfactant  
Contact angle  
MEOR

### ABSTRACT

Changing the wettability of reservoir rock towards strongly water-wet state is effective way to enhance oil recovery from fractured carbonate reservoirs which are typically oil-wet. Regarding this fact, the injection of surfactant and the bacterial solution as EOR agents is proposed in the current work as a potential method to alter the wettability of rock surface reservoir. Nevertheless, there is a definite lack of experimental data regarding this method and the synergistic effect of both chemical and bacterial solutions on this process. In this study, the sole and combined effects of the bacterial solution using an *Enterobacter cloacae* strain as a biosurfactant-producer are compared with sodium dodecylbenzenesulfonate (SDBS) and dodecyltrimethylammonium bromide (C12TAB) as anionic and cationic surfactants, respectively. For this purpose, static contact angle measurement are utilized to investigate the wettability alteration. The obtained results revealed that bacterial solution can alter the wettability of calcite surfaces to the same extent of C12TAB and more than SDBS. In addition, base on the obtained results it can be concluded that the combination of bacterial solution and chemical surfactant are not proposed for in-situ biosurfactant techniques, since the *Enterobacter cloacae* uses the chemical surfactant as the carbon source concomitantly a reduction in the biosurfactant production occurs.

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## 1. Introduction

During the recent decades, different methods are proposed to mobilize and produce the trapped oil in the reservoir called

\* Corresponding author.

E-mail addresses: [m.lashkarbolooki@nit.ac.ir](mailto:m.lashkarbolooki@nit.ac.ir), [lashkar\\_80@yahoo.com](mailto:lashkar_80@yahoo.com) (M. Lashkarbolooki).

<http://dx.doi.org/10.1016/j.colsurfa.2016.11.010>  
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enhanced oil recovery (EOR) processes [1–4] such as surfactant flooding [5–7] and microbial enhanced oil recovery (MEOR) [8–11]. MEOR is a biologically based technology utilizes bio-products to recover the trapped oil. Biological methods have some advantages over physical-chemical methods. First of all, MEOR as low cost process offer in-situ biodegradation of the petroleum hydrocarbons using microorganisms. In addition, slight water solubility of petroleum hydrocarbons introduce the potential to limit the capacity of bacteria to access and degrade the substrate. For this reason, bacteria often produce biosurfactant to increase the bioavailability of hydrocarbons and to facilitate their uptake that lead to MEOR as more environmentally friendly method [11,12].

Biosurfactants, known as one of the most important bioproducts, can reduce the capillary forces in the reservoir rock by the reduction of oil/water interfacial tension and wettability alteration similar to chemical surfactant [13–21]. For the case of IFT reduction, for efficient oil recovery, it needs to reach to ultra-low value ( $<10^{-2}$  mN/m) which is difficult and costly for the both surfactants and biosurfactants [22–24]. Besides the capability of IFT reduction, it has been accepted that surfactants regardless of their source which can be chemical or bio can change the wettability of carbonate rock surfaces as another effective mechanism of using surfactants or biosurfactants [24–32]. In other words, despite the IFT reduction that was observed using MEOR [11,24], the effect of wettability alteration on the total microbial oil recovery was found to be more significant [2,4,10,21,33].

It should be noticed that wettability alteration towards the preferred state vary from non-fractured and fractured rocks [21]. In non-fractured rocks, the change in wettability from extremely wet conditions to neutral wettability is a favorable modification based on obtaining higher oil recovery efficiency [24]. While, wettability alteration from strongly oil wet state to strongly water wet state is preferred in fractured reservoirs to promote water imbibition for more oil recovery [21]. Capillary forces will change from negative towards positive value due to this wettability alteration as a consequence of spontaneous water imbibition into the matrices followed by countercurrent displacement of oil [21,34]. The worth mentioning point is that there is no need of large shift in capillary pressure for EOR agents to have an effective influence through the systems deals with displacements by viscous forces. So, it seems that a relatively slight change in wettability alteration by chemicals can be quietly effective for non-fractured carbonates [35]. It is well understood that natural fractures and low permeability matrixes are the two important characteristics of the most carbonate reservoir rock in the world [36,37]. If the matrix blocks are oil-wet, low oil recovery efficiency is achieved because of ineffective spontaneous imbibition of water due to the small or negative capillary pressure. In this case, the injected water will follow the fractures from the injection well to the production well without displacing a significant amount of oil. The result is an early water breakthrough and poor sweep efficiency leads to entrapment of large volume of oil in the complex pore structures [38]. In this regard, any wettability alteration of carbonate rock towards more water wetness changes the capillary forces from negative to positive condition that consequently leads to spontaneous imbibition of water into the matrices followed by countercurrent displacement of crude oil [21,39]. Surface-active chemicals [40,41] during chemical injection and bio-products from MEOR process [4,10,11,21] have been utilized to investigate the possible change of the reservoir rock wettability. However, few researchers have investigated the behavior of chemical surfactant and microorganisms conjunction. Al-Sulaimani et al. [33] performed core flooding test to investigate the potential of a biosurfactant produced by a *Bacillus subtilis* strain for its potential in enhancing oil recovery on Berea sandstone cores. It was found that the performance of

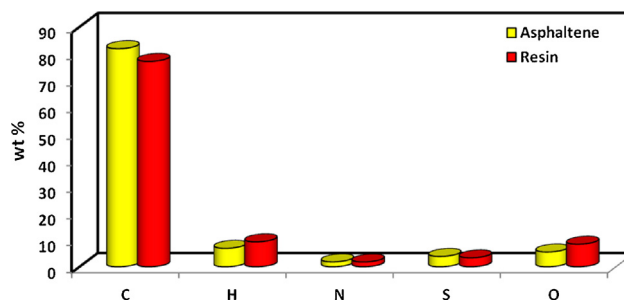


Fig. 1. The results of elemental analysis.

the biosurfactant was increased when it is mixed with chemical surfactants ethoxylated sulfonates [33].

In addition, Daoshan et al. [24] proposed the biosurfactant-rhamnolipid-fermentation liquor (RH) as sacrificing agent to decrease surfactant adsorption on the reservoir rock during ASP flooding process. Static adsorption tests indicated that the adsorption loss of alkylbenzene sulfonate (ORS) on sandstone was reduced by 25–30%, when RH was mixed with ORS or as pre-adsorbed case [24]. According to previous studies on the capability of MEOR enriched with surfactant, there is a definite lack of experimental data can be used to compare the capability of these methods on wettability alteration of carbonate rock surfaces. In addition, to the best of our knowledge, there is no report on the synergistic effect of chemical and bio surfactants combination on the wettability alteration of carbonate rock and the adsorption reducing character of biosurfactant in contiguity of chemical surfactant. In this regard, the main purpose of this work is to investigate the performance of surfactants and biosurfactants on the wettability alteration of carbonate rocks, which could be summarized as below:

- compare the potential of the two different categories of chemical surfactants including SDBS (anionic) and C12TAB (cationic) with a bacterial solution on the wettability alteration from oil-wet toward water or neutral wet conditions.
- investigate the synergistic effect of the used common surfactants and bacterial solution. In this regard, the combination of chemical surfactants and incubated bacterial solution (first scenario) and concurrent incubation of the chemical surfactants and *E. cloacae* (second scenario) are also investigated.

## 2. Experimental and methodology

### 2.1. Materials

#### 2.1.1. Crude oil

The model crude oil with 24.46 API° was supplied from one of the southern Iranian oil reservoir (ABCO). The results of SARA (saturated-aromatic-resin-asphaltene) analysis show that the ABCO contains 42.68, 40.69, 7.63, and 9.00 wt.% of saturated, aromatic, resin, and asphaltene, respectively. Due to the importance of asphaltene and resin structure, the elemental analysis (Carbon, Hydrogen, Nitrogen, Sulfur, Oxygen (CHNSO) test) of the extracted asphaltene and resin of the crude oils was performed which revealed possible empirical formula of the  $C_{507}H_{511}N_{10}S_9O_{26}$  and  $C_{246}H_{356}N_5S_4O_{20}$  for asphaltene and resin, respectively (see Fig. 1).

#### 2.1.2. Brine

The synthetic formation brine (FB) (see Table 1) with high total dissolved solid (TDS) i.e. 202072 ppm and electrical conductivity of 164.3 mS/cm was prepared to mimic the reservoir brine by addition of different amount of ions into deionized water with elec-

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