

# Effect of Titanium Dioxide Nanoparticles on Surfactants and Their Impact on the Interfacial Properties of the Oil–Water–Rock System

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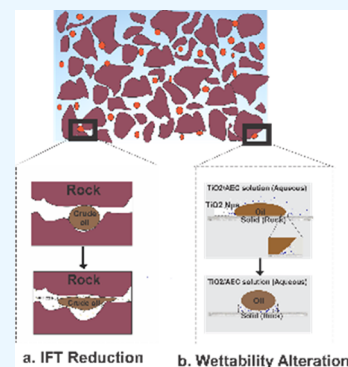
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**ABSTRACT:** The application of nanoparticles (NPs) in the oil and gas industry has received wide attention in recent years because it is increasingly being considered a promising approach to recovering trapped oil in conventional hydrocarbon reservoirs. Numerous studies have demonstrated that combining nanoparticles with a surfactant can enhance surfactant performance by changing the interfacial properties of the solution when it comes in contact with crude oil and rock surfaces. However, more information and additional experimental data are required concerning the application of titanium dioxide nanoparticles in alkyl ethoxy carboxylic surfactants. In this study, we measure the changes in interfacial tension and wettability due to the addition of titanium dioxide nanoparticles (0, 100, 250, and 500 ppm) in alkyl ethoxy carboxylic surfactant using a spinning drop tensiometer and contact angle measurements. The interfacial tension of the crude oil–water (surfactant) system decreases by approximately two orders of magnitude with an increasing titanium dioxide concentration, exhibiting a minimum value of  $5.85 \times 10^{-5}$  mN/m. Similarly, the contact angle decreases on the surface of the Berea sandstone by combining the surfactant with titanium dioxide, reaching a minimum contact angle of  $8.8^\circ$ . These results demonstrate the potential of this new approach to maximize the recovery of trapped oil and significantly improve oil production.



## 1. INTRODUCTION

Enhanced oil recovery (EOR) is a technology that involves the injection of external energy or (e.g., fluids) into subsurface formations to improve oil recovery at any stage of production and increase total recovery. Several EOR methods are frequently used to increase oil recovery,<sup>1</sup> including chemical flooding (e.g., surfactant, polymer), thermal recovery, gas injection (miscible and immiscible flooding), and microbial EOR.<sup>2,3</sup> Each method is unique, and its applicability depends on the characteristics of the rock and fluid in specific reservoirs. With this in mind, a comprehensive study regarding the selection of the EOR method is essential to minimize the risk and maximize the yield of additional oil recovery.

The use of surfactants as an EOR agent has become prevalent in recent years owing to their significant impact on additional oil recovery. Surfactant flooding is a well-established method that has been proven to successfully recover trapped oil in the laboratory scale<sup>4–6</sup> and field applications.<sup>7,8</sup> This is attributable to the numerous investigations into the mechanism of surfactants as an EOR agent, including lowering the interfacial tension (IFT) between oil and water at the interface,<sup>9,10</sup> wettability alteration<sup>11,12</sup> and microemulsion flooding.<sup>9</sup> A laboratory investigation by Haghghi et al.<sup>13</sup> showed that using a nonionic surfactant (dedocanoyl-glucosamine surfactant) can

reduce IFT from 30 to 14 mN/m, reduce the water contact angle from  $148.93^\circ$  (less water-wet) to  $65.54^\circ$  (more water-wet), and increase oil recovery by 19% after waterflooding. However, due to the complexity of subsurface formations that prevent the surfactant from working effectively (e.g., adsorption), alternative routes have been developed to enhance the performance of the surfactant.

Recently, the use of nanoparticles (NPs) has received widespread attention for its potential EOR applications.<sup>14–18</sup> NPs are particles typically ranging from 1 to 100 nm in size.<sup>19–25</sup> Several types of nanomaterials have been studied for application in EOR, including titanium dioxide,<sup>18,24,26</sup> graphene,<sup>22</sup> silica dioxide,<sup>23,27–30</sup> aluminosilicate,<sup>31</sup> zirconium dioxide,<sup>16,32,33</sup> and silica dioxide in the CO<sub>2</sub>-brine system.<sup>34,35</sup> Laboratory investigations have demonstrated that the surfactant performance can be enhanced by combining the surfactant solution with nanomaterials to improve the interfacial properties between the

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oil–water–rock system.<sup>15,17,23,31,36,37</sup> Nourinia et al.<sup>15</sup> reported that adding zinc oxide/monmorillonite nanocomposites into a natural surfactant derived from cyclamen persicum reduced the IFT to a minimum of 2.5 mN/m. The authors<sup>15</sup> also observed that the water contact angle was reduced from 134° to 17.8°, indicating that the wettability was shifted to become more water-wet. Cheraghian et al.<sup>23</sup> conducted water contact angle measurements and micromodel flooding experiments in a glass micromodel using silica NPs combined with sodium dodecyl sulfate (SDS) as the surfactant. They showed that the contact angle decreased from 72° to 11°, and the recovery factor increased by 13%.<sup>23</sup> Yahya et al.<sup>31</sup> demonstrated that the combination of aluminosilicate with sulfonated alkyl ester (SAE) can reduce the contact angle from 41.9 to 40.3 and increased oil recovery from 47 to 49%. Jha et al.<sup>17</sup> showed that adding zirconia NPs to a low-salinity surfactant reduced the IFT and water contact angle on quartz surfaces. In addition, they found that the concentration ratio of divalent cations and sulfate ions significantly affected the IFT and contact angle.<sup>17</sup> Therefore, considering that one of the main purposes of the surfactant is to reduce the IFT between oil and water at the interface, adding nanomaterial to a surfactant solution can enhance the performance of surfactant to such a degree as to shift the wettability of the system (i.e., lower contact angle), thus creating more water-wet conditions due to the modification of disjoining pressure.

Disjoining pressure is the force that separates or disjoins the two interfaces,<sup>38</sup> which is related to the wetting film stability.<sup>38–40</sup> According to the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory, the disjoining pressure is equivalent to the sum of the contributions of van der Waals,<sup>41,42</sup> electrostatic, and structural interactions.<sup>38,43</sup> van der Waals interactions are based on the Hamaker theory, which assumes that interactions are attractive forces between molecules that arise from temporary fluctuations in electron distribution. These forces can influence the disjoining pressure between surfaces, particularly when the surface separation is small. As the surfaces get closer, van der Waals forces become more significant, resulting in an increase in disjoining pressure.<sup>38</sup> The next force is the electrostatic interaction that is related to the forces between the charged surface.<sup>38</sup> The electrostatic interaction of ions involves the repulsive and attractive forces<sup>44,45</sup> that arise depending on a range of parameters, namely, the temperature, solution pH, dissolved CO<sub>2</sub>,<sup>46,47</sup> mineralogy of the rock surfaces,<sup>48–50</sup> ionic strength, and composition of ions in the aqueous medium.<sup>51</sup> When two interfaces have the same charge, the interaction between them becomes repulsive and makes a positive contribution to the total disjoining pressure.<sup>39</sup> The last force is structural, where the disjoining pressure of one layer of particles is generally higher than that of a double layer.<sup>52</sup> The structural force is always repulsive, so its contribution to the total disjoining pressure is always positive.<sup>39</sup>

Notably, the application of titanium dioxide (TiO<sub>2</sub>) NPs in modifying the interfacial properties of oil–water–rock systems shows great potential for EOR. Ehtesabi et al.<sup>18</sup> demonstrated that adding TiO<sub>2</sub> NPs into NaCl brine can alter the wettability and yield, significantly increasing oil recovery from 49 to 80% in natural rock samples. However, there are limited experimental data regarding the interfacial properties of TiO<sub>2</sub> NPs combined with surfactants in the oil–water–rock system. Hence, this study aims to investigate the effect of TiO<sub>2</sub> NPs/surfactant on IFT and wettability using various NP concentrations. This combined method can help recover trapped oil in existing

reservoirs, and the results of this study help clarify the EOR mechanism of TiO<sub>2</sub> NPs combined with surfactants in oil–water–rock systems.

## 2. EXPERIMENTAL METHODOLOGY

**2.1. Materials and Sample Preparations.** **2.1.1. Rock Sample.** A Berea sandstone sample was used in this study. Table 1 provides the mineralogical composition of the sandstone

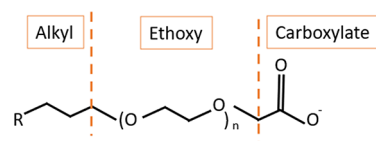
**Table 1. Mineralogical Properties of the Berea Sandstone**

mineral	concentration [wt %]
quartz	96.33
clay	1.17
pyrite	2.5

according to X-ray diffraction (SmartLab diffractometer, 3 kW, 200 VAC ± 10%, 50/60 Hz, 30 A). The main mineral in the Berea sandstone is quartz with small amounts of clay and pyrite.

Prior to conducting contact angle measurement on a surface of Berea sandstone, the rock was sliced into a thin section, approximately 1–2 mm in thickness, and then fixed on an object glass using epoxy resin. The epoxy was left to cure for at least 3 h in the oven at 30 °C. The Berea sandstone surface was then polished using 600 grit sandpaper to create a smoother surface. Finally, the thin section was cleaned by using compressed air to remove any debris.

**2.1.2. Surfactant.** An alkyl ethoxy carboxylic (AEC) anionic surfactant (Figure 1) was formulated in the Enhanced Oil



**Figure 1.** Molecular structure of the AEC surfactant. Reprinted from Herawati et al.<sup>54</sup> with permission from Elsevier.

Recovery Laboratory at the Bandung Institute of Technology, which was manufactured by PT. Rakhara Chemical Technology, Indonesia. The polar (hydrophilic) headgroup, a carboxylic molecule, has a negative charge, while the nonpolar (hydrophobic) tail consists of linear alcohol of the C<sub>10</sub>–C<sub>14</sub> carbon chains.<sup>53,54</sup>

The general formula for AEC surfactants is RO-(CH<sub>2</sub>CH<sub>2</sub>O)<sub>n</sub>CH<sub>2</sub>COO<sup>−</sup>M<sup>+</sup>, where R is an alkyl group containing C<sub>8</sub>–C<sub>18</sub>, which determines the length of the hydrocarbon chain. The variable n is the average number between 1 and 15, while n denotes the alkali metal or alkaline earth metal cations with an oxygen atom.<sup>53,54</sup> The molecular formula of the surfactant is C<sub>12</sub>H<sub>26</sub>(OCH<sub>2</sub>CH<sub>2</sub>O)<sub>7</sub>CH<sub>2</sub>COO<sup>−</sup>, with C<sub>12</sub>H<sub>26</sub> as the alkyl group, (OCH<sub>2</sub>CH<sub>2</sub>O)<sub>7</sub> as the ethoxy group, and CH<sub>2</sub>COO<sup>−</sup> as the carboxylate ion.<sup>53,54</sup>

**2.1.3. TiO<sub>2</sub> Nanoparticles.** TiO<sub>2</sub> NPs were obtained from XFNano material Tech Co, Ltd., China (≥99% mass fraction), and the detailed properties are presented in Table 2. To

**Table 2. TiO<sub>2</sub> Properties**

surface area (m <sup>2</sup> /gr)	pore diameter (nm)	pore volume (cm <sup>3</sup> /gr)	micropore area (m <sup>2</sup> /gr)
52.32	13.27	0.103	0.002

understand the morphology, transition electron microscopy (TEM; Hitachi HT7700, with a voltage of 40–120 kV and resolution of 0.204 nm, Hitachi, Japan) was performed to obtain high-resolution images of TiO<sub>2</sub>. The images show that the TiO<sub>2</sub> particles are spherical (see Figure 2).

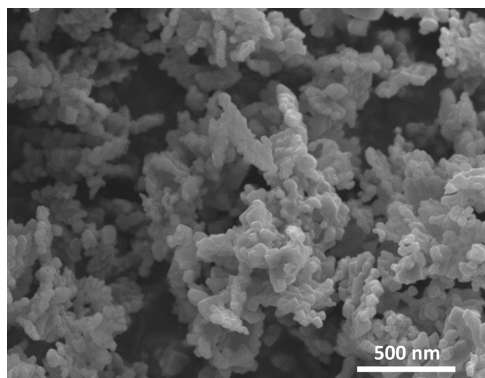


Figure 2. TEM image of TiO<sub>2</sub> nanoparticles.

**2.1.4. Crude Oil and Synthetic Brine.** The crude oil was supplied from the Tempino oil field (Table 3) located in

Table 3. Characteristic Properties of the Tempino Oil Field<sup>4</sup>

oil characteristics	value
SARA	
saturated	71.60%
aromatics	25.49%
resins	2.14%
asphaltenes	0.78%
EACN (equipment alkane carbon number)	8.29
TAN (total acid number)	1.23 mg KHO/g
viscosity	0.90 cP (66 °C)
API gravity	43.45

Indonesia. Three solution types were tested, namely, (1) pure NaCl solution with a concentration of 8000 ppm, (2) AEC surfactant solution with concentrations of 0.5–2 wt %, and (3) AEC surfactant solution mixed with TiO<sub>2</sub> NPs using concentrations from 0 to 500 ppm.

To prepare a pure NaCl solution of 8000 ppm, 8.0 g of a single reagent-grade NaCl salt ( $\geq 99\%$  mass fraction, Merck KGaA Darmstadt Germany) was dissolved in 1.0 L of demineralized (DM) water, supplied by the Chemistry Department at the Bandung Institute of Technology using reverse osmosis treatment. The solution was then stirred for 60 rpm using a magnetic stirrer (Thermo Scientific Cimarec type SP131320, with a speed range of 60–1200 rpm, surface temperature range of 5–450 °C, and voltage 220/240 V at 50/60 Hz, supplied by

PT. Indolab Utama, Indonesia) for at least 60 min to obtain the homogeneous solution.

The surfactant solutions were prepared by dissolving AEC surfactant (0.5–2 wt %) in the pure NaCl solution of (8000 ppm). The AEC surfactant was weighed using a digital balance (Fujitsu FSR-A Precision balance with a capacity of 220 g and a readability of 0.001 g, supplied by PT Interskala Mandiri, Indonesia) and stirred in a NaCl solution using a magnetic stirrer for 60 min to obtain the homogeneous solution. Ultrasonication (Krisbow ultrasonicator, 220 V, 50 W, 42 kHz, Krisbow, Indonesia) was employed to prepare the surfactant solution containing TiO<sub>2</sub> NPs and homogenize the mixed solution. All AEC surfactant solutions mixed with TiO<sub>2</sub> NPs were sonicated for 60 min and then stirred with a magnetic stirrer for another 30 min. The pH and conductivity ( $\sigma_w$ ) of all the tested solutions (Table 4) were measured using a HI98181 pH meter (accuracy of  $\pm 0.01$  pH, HANNA Instruments Co., Ltd., Korea,) and Eutech™ CON700 conductivity meter (accuracy of  $\pm 1\%$ , Thermo Fisher Scientific Co., Ltd., USA) under ambient conditions (temperature of 25 °C and pressure of 0.1 MPa).

**2.2. Methods.** **2.2.1. Interfacial Tension Measurement.** A spinning drop tensiometer with a speed and temperature accuracy of  $\pm 3$  rpm and  $\pm 0.5$  °C (TX500D, KINO, USA) was used to measure the IFT of the crude oil–water before and after adding TiO<sub>2</sub>/AEC surfactant solution at a reservoir temperature of 68.3 °C, as illustrated in Figure 3. To reduce the

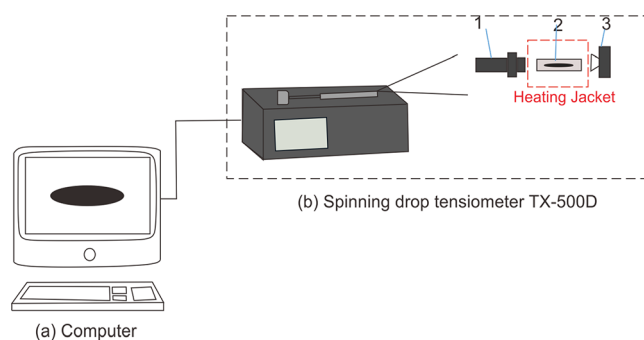


Figure 3. Experimental setup used for interfacial tension measurement. (a) Computer; (b) spinning drop tensiometer TX-500D; (#1) camera; (#2) sample; and (#3) light source.

contamination bias, the tube was cleaned with toluene, followed by acetone and DM water, and then dried before each IFT measurement. The TiO<sub>2</sub>/AEC surfactant was injected into the capillary tube until it was filled, and the crude oil was injected after that using a micro syringe with a volume of 2  $\mu$ L. The test was then run for 30 min at 3000 rpm. The IFT of the tested solution was obtained by analyzing the image of the oil in the

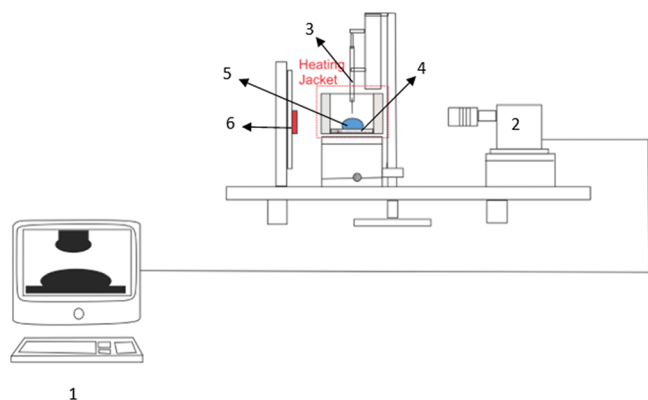
Table 4. PH and Conductivity Values for all Tested Experimental Conditions<sup>4</sup>

solution	TiO <sub>2</sub> concentration (ppm)	pH value	$\sigma_w$ (mS/m)
NaCl brine 8000 ppm		6.28 $\pm$ 0.1	14.25 $\pm$ 0.01
AEC surfactant 1.25 wt %	0	4.69 $\pm$ 0.1	7.37 $\pm$ 0.01
	100	4.76 $\pm$ 0.1	7.62 $\pm$ 0.01
	250	4.73 $\pm$ 0.1	7.74 $\pm$ 0.01
	500	4.75 $\pm$ 0.1	8.03 $\pm$ 0.01

<sup>4</sup>The reported uncertainties of pH and  $\sigma_w$  are based on the measurement repeatability.

TiO<sub>2</sub> NPs/AEC surfactant using Interfacial Tension TX500D software.

**2.2.2. Contact Angle Measurement.** A Theta One Attention Optical Tensiometer (Accuracy of  $\pm 0.1^\circ$ , Biolin Scientific, UK) was used to measure the contact angle, as depicted in Figure 4.



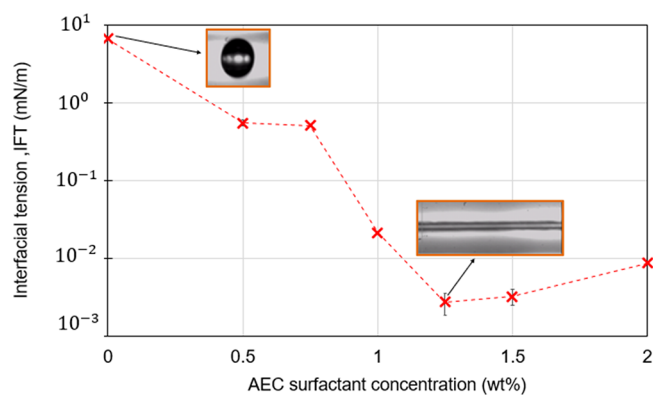
**Figure 4.** Schematic of the experimental setup used for the contact angle measurement; (#1) Computer with drop image program; (#2) camera; (#3) micro syringe; (#4) thin section of rock sample; (#5) substrate; and (#6) light source.

Prior to conducting the measurement, the contact angle cell was heated at  $68.3^\circ\text{C}$  (reservoir temperature) for at least one hour to ensure a constant temperature during the experiment. The thin section of Berea and the micro syringe filled with the tested solution were placed into the heated contact angle cell for another hour to condition the temperature of the thin section and the tested solution. A TiO<sub>2</sub> NPs/AEC surfactant solution droplet was then dispensed on top of the thin Berea section using the micro syringe, and all images of the process were recorded using One Attention software. The contact angle was obtained by analyzing the captured image of the droplet on the rock surface. To ensure no contaminants bias, the micro syringe and thin section of Berea were replaced with a new clean syringe and a new uncontaminated thin section of Berea after each measurement. In addition, the contact angle measurement was repeated at least two times to determine the experimental repeatability.

### 3. RESULTS AND DISCUSSION

**3.1. Interfacial tension.** Various concentrations of the AEC surfactant were tested from 0.5 to 2 wt % to determine the surfactant's critical micelle concentration (CMC; Figure 5). The CMC of the AEC surfactant was obtained at the concentration of 1.25 wt %, exhibiting the smallest measured IFT value of  $2.7 \times 10^{-3}$  mN/m. To enhance the effect of TiO<sub>2</sub> NPs on the IFT, the AEC surfactant with the CMC (1.25 wt %) was then mixed with various concentrations of TiO<sub>2</sub> NPs (Table 5 and Figure 6).

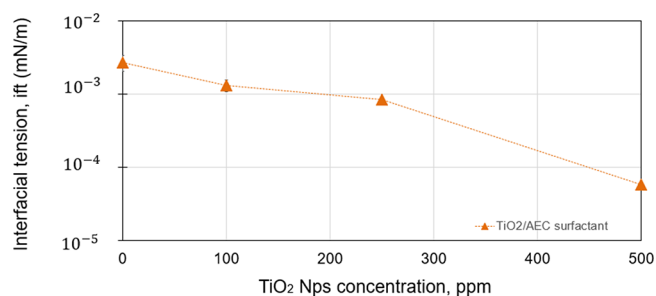
Figure 6 shows that the IFT of the AEC surfactant combined with TiO<sub>2</sub> NPs decreases with increasing TiO<sub>2</sub> concentration. Thus, the presence of TiO<sub>2</sub> NPs enhances the ability of the AEC surfactant to reduce IFT in the oil–water system. This can be attributed to the adsorption of the surfactant onto the NPs. The reduction of IFT may be explained by considering that NPs become more hydrophobic after adsorbing positively charged of surfactant molecules, consistent with the argument from the previous studies of Al-Anssari et al.<sup>28</sup> and Moghadam and Azizian.<sup>55</sup> The reduction of IFT in the oil–water system also



**Figure 5.** IFT of the oil–water system with AEC surfactant concentrations in the range of 0.5–2 wt %.

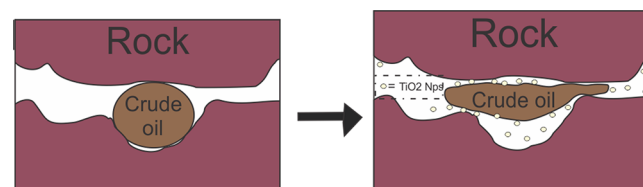
**Table 5. Summary of IFT Measurements for Various Concentrations of TiO<sub>2</sub> NPs Combined with 1.25 wt % AEC Surfactant Solution**

surfactant	nano particles	concentration of NPs (%)	IFT (mN/m)
AEC 1.25 wt %	TiO <sub>2</sub>	0	$2.7 \times 10^{-3}$
		100	$1.31 \times 10^{-3}$
		250	$8.50 \times 10^{-4}$
		500	$5.85 \times 10^{-5}$



**Figure 6.** Interfacial tension of the oil–water system at different TiO<sub>2</sub> NPs concentrations mixed with the AEC surfactant 1.25 wt %.

impacts the ability of oil to easily flow through the small pores and yield additional oil recovery (Figure 7).

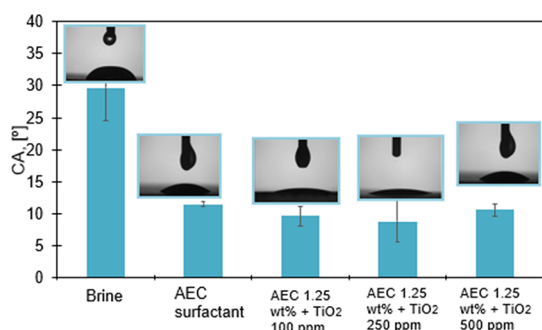


**Figure 7.** Illustration of crude oil in porous media and the effect of IFT reduction using the TiO<sub>2</sub> NPs/AEC surfactant.

**3.2. Contact Angle.** The contact angle indicates the wettability and the extent to which it is altered. In this study, a contact angle test was performed to determine the effect of adding TiO<sub>2</sub> NPs to altering the wettability of the surfactant (Table 6 and Figure 8). Figure 8 shows that the contact angle slightly decreases with the addition of TiO<sub>2</sub> NPs in the AEC surfactant solution. Note that increasing the TiO<sub>2</sub> concentration in the AEC surfactant does not significantly alter the contact angle but the value remains lower than that for the AEC

**Table 6. Summary of Contact Angle Results of TiO<sub>2</sub> NP-Surfactant Flooding Experiment**

solution	TiO <sub>2</sub> concentration (ppm)	CA,°
8000 ppm brine	0	29.7
AEC surfactant	0	11.5
AEC surfactant 1.25 wt % + TiO <sub>2</sub>	100	9.7
	250	8.8
	500	10.6

**Figure 8.** Contact angle measurements of AEC surfactant 1.25 wt % mixed with TiO<sub>2</sub> NPs using various concentrations on the surface of Berea sandstone. The error bars represent the experimental uncertainty and the measurement repeatability.

surfactant alone. The presence of TiO<sub>2</sub> NPs on the wedge film also made the Berea sandstone more water-wet as a result of the layering and structuring of the TiO<sub>2</sub> NPs, which increased the structural disjoining pressure. The structural disjoining pressure gradient or film tension gradient (Figure 9) directed toward the wedge from the bulk solution is the driving force for the spreading of the nanofluid; thus, the film tension is high toward the vertex owing to the nanoparticle structuring in the wedge confinement. As the film tension increases toward the wedge's vertex, the nanofluid spreads toward the wedge tip, improving the dynamic spreading characteristic of the nanofluid and causing the oil droplet to be released from the pore surface of the rock. This argument is also consistent with the study reported by Wasan et al.<sup>52</sup>

#### 4. CONCLUSIONS

The addition of TiO<sub>2</sub> NPs to the AEC surfactant was investigated herein. The results and conclusions can be summarized as follows:

1. TiO<sub>2</sub> NPs improve the ability of the AEC surfactant to reduce the IFT between the oil and water system by two-

order magnitude, from  $1.31 \times 10^{-3}$  to  $5.85 \times 10^{-5}$  mN/m, which later can be translated to reducing the capillary number and yield to additional oil recovery.

2. The TiO<sub>2</sub> NPs reduce the contact angle of the AEC surfactant solution on the Berea sandstone, from 29.7° to 8.8°. This result indicates that adding TiO<sub>2</sub> NPs into the AEC surfactant can shift the wetting state of the system to be more water-wet, which preferentially is a better condition to gain additional oil recovery.
3. The addition of TiO<sub>2</sub> NPs to the AEC surfactant has the potential to be applied in EOR technology and increase oil production by removing more trapped oil in subsurface oil reservoirs.

Future experimental work involves testing the addition of TiO<sub>2</sub> NPs in the AEC surfactant using an intact natural Berea sandstone in a one-dimensional displacement coreflooding apparatus. This experiment can help estimate the amount of trapped oil that can be recovered from natural porous media using the combination of TiO<sub>2</sub> NPs and the AEC surfactant. Moreover, additional data must be obtained for higher ionic strength and complex brine compositions to determine the effect of divalent ions and ionic strength on the interfacial properties of the TiO<sub>2</sub>/AEC surfactant in the oil–water–rock system.

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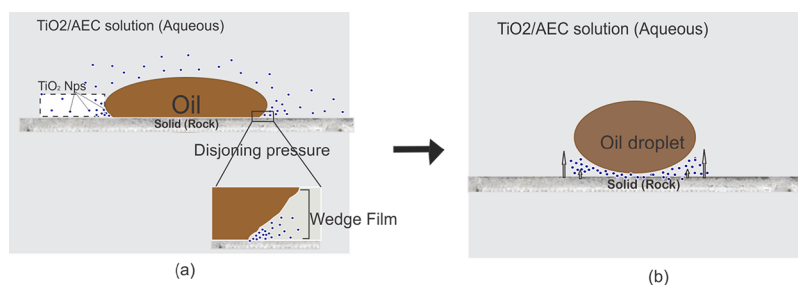
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**Figure 9.** (a) Illustration of structural disjoining pressure gradient mechanism among crude oil and TiO<sub>2</sub> NPs/AEC surfactant solution in the aqueous phase due to nanoparticle structuring in the wedge-film, adapted from Wasan et al.<sup>52</sup> with permission from Elsevier. (b) Illustration of an oil droplet to be released from the pore surface of the rock due to disjoining pressure.

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### Notes

The authors declare no competing financial interest.

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