ABUAD Journal of Engineering Research and Development (AJERD)
ISSN (online): 2645-2685; ISSN (print): 2756-6811

Volume 6, Issue 2, 10-16

Synergetic Interfacial Tension Reduction Potential of Silica Nanoparticles and Enzyme

Tinuola Hannah UDOH[1] and Oyinkepreye David ORODU[2]

[1]*Department of Chemical/Petrochemical Engineering, Akwa Ibom State University, P.M.B 1167, Uyo, Nigeria*
tinuolaudoh@aksu.edu.ng

[2]*KEOT Synergy Nigeria Ltd and Robert Gordon University, Aberdeen*
preye.d.orodu@gmail.com

Corresponding Author: tinuolaudoh@aksu.edu.ng, +2348066519248
Date Submitted: 21/06/2023
Date Accepted: 25/07/2023
Date Published: 30/07/2023

Abstract: *The co-existence of multiphase fluids in the hydrocarbon reservoir rock pores plays a fundamental role in oil recovery processes because of the strong effect of interfacial forces that exist at the interface of these immiscible fluids. In this study, the effects of enzyme and silica nanoparticles on crude oil-brine interfacial tension were investigated under varied brine salinities and brine compositions. The results showed that the application of silica nanoparticles alone in brines of varied compositions and salinities does not significantly modify the crude oil-brine IFT. The use of enzyme and combined enzyme-nanoparticles however significantly reduced crude oil-brines IFT but the contribution of silica nanoparticles to the IFT reduction was not significant. The result of this study is relevant to the design and applications of enzyme and nanoparticles enhanced oil recovery processes.*

Keywords: *Interfacial tension, salinity, enzyme, silica nanoparticles, greenzyme.*

1. INTRODUCTION

Hydrocarbon reservoirs consist of different fluids such as formation brine, crude oil, and gases. The co-existence of these fluids in the rock pores plays a fundamental role in oil recovery processes because of the strong effect of interfacial forces that exist at the interface of these immiscible fluids [1]. Interfacial modifications such as interfacial tension (IFT) reduction can improve oil recovery [2]. Previous studies have showed that IFT reduction is the primary mechanism by which surface active compounds like surfactants and enzymes increase oil production [3 - 5]. This is made possible through their interfacial adsorption that resulted from their molecular composition that consists of hydrophobic and hydrophilic groups which simultaneously interact with oil and brine respectively. The resultant effect of these interactions is IFT reduction that promotes mixing of the two immiscible fluids and inviably mobilisation of residual oil saturation [6, 7].

Reduction of IFT has also been attributed to efficient nanoparticles increased oil recovery [8 - 12]. In the recent past, there has been an increased studies on the applications of nanoparticles in oil recovery processes due to the attendant advantages associated with it such as ultra-small size (1-100 nm), good strength, high surface area per unit volume, high chemical reactivity, and electrical properties [13, 14]. The enhanced oil recovery application of nanoparticles may modify the fluid properties such as IFT reduction, alter the wettability of the rock surfaces, increase the mobility of oil trapped by capillary effect and strengthened sand consolidation [15]. The enhanced oil recovery potential of different types of nanoparticles such as: SiO_2, TiO_2, Al_2O_3 have been investigated. For example, Ragab and Hannora [16] conducted a comparative study on the EOR potential of SiO_2 and Al_2O_3 nanofluids and they found SiO_2 to be more efficient than Al_2O_3. Hendraningrat *et al.* [17] also investigated the effects of SiO_2, TiO_2 and Al_2O_3 nanofluids on oil recovery and they observed the highest oil recovery with the use of TiO_2. Other study by Alomair et al. [18] has explored the EOR potential of combination of SiO_2 and Al_2O_3 nanoparticles in Berea sandstone heavy oil core flooding and observed the highest incremental oil recovery with the use of the mixture. Increased oil recovery was also observed in recent study on EOR potential of the SiO_2 nanoparticles application in core flooding experiments conducted by Udoh [19] on samples from one of the reservoirs in the Niger Delta part of Nigeria. Most of these studies however did not investigate the IFT modification potential of these nanoparticles, even though IFT reduction has been stated as one of the mechanisms by which nanoparticles enhance oil recovery.

Furthermore, the potential of combined nanoparticles and surfactants has been explored in different studies in the recent past and better results have been reported [20, 21]. There are however some environmental concerns with the continuous application of surfactants due to their non-degradable nature [22]. Some biologically based surface-active compounds that are environmentally friendly such as enzyme has been identified as alternatives to surfactants and they are said to be efficient in enhance oil recovery processes [4, 5, 23 - 26]. Although different mechanisms have been associated with the observed increased oil recovery from applications of enzyme, but IFT is one of the most prominent of all. For instance, Nasiri *et al.* [25] attributed the observed increased oil recovery from the application of enzyme in Berea sandstone to wettability alteration and IFT reduction. Rahayyem *et al.* [27] also proposed IFT reduction and wettability alteration as underlying mechanisms for the increased oil recovery observed from the application of enzyme at micro-scale level in microfluidic device. Furthermore, from the experimental studies on the applications of enzyme in carbonate rock conducted by Udoh and Vinogradov [5, 23], combined effects of wettability alteration, rock dissolution and IFT reduction were attributed to the observed increased oil recovery. Also, from the study on the application of enzyme in shale formation conducted by Salahshoor *et al.* [28], wettability alteration and change in IFT were attributed to the effective enzyme application observed.

Other study by Udoh and Orodu [29] has explored the IFT modification potential of SiO_2 nanoparticles and enzyme in different salinities relevant to enhanced oil recovery processes and their results showed that the application of nanoparticles did not significantly modify oil-brine IFT, while the use of enzyme and combined enzyme-nanoparticles significantly modified oil-brine IFT. The brines investigated in their study were however limited to varied brine salinities based the formation brine in the reservoir that was used as case study. This formation brine is a multicomponent brine that comprises of different salts (NaCl, $CaCl_2$, $MgCl_2$, KCl, Na_2SO_4) and the effect of these varied salts on oil-brine interfacial interactions have not been explored. Hence, the aim of this study is to investigate the interfacial modification potentials of SiO_2 nanoparticles, enzyme and combination of SiO_2 nanoparticles and enzyme mixture at oil-brine interface using different brine compositions.

2. MATERIALS AND SAMPLE PREPARATION

The materials and method used in this study are presented in this section.

2.1 Brine Preparation

The formation brine used in this study is the same as the one used by Udoh and Orodu [29]. The compositional breakdown of the formation brine that was synthesised based the formation brine of the reservoir used as case study is presented in Table 1. The salinity of the brine is 32 g/L, with sodium chloride (NaCl) constituting 98.2%, other components are 0.8% magnesium chloride ($MgCl_2$), 0.6% calcium chloride ($CaCl_2$), and 0.2% potassium chloride (KCl) and sodium sulphate (Na_2SO_4) each. The effect of varied brine salinities on oil-brine IFT was investigated using formation brine (FMB), 50D brine (50% dilution of FMB) and 90D brine (90% dilution of FMB). The compositional breakdown of these brines and the detailed results of the study are presented in [29]. The effect of brine composition on crude oil-brine IFT was investigated with individual salt solutions of the formation brine.

Table 1: The compositional breakdown of the formation brine

Components	FMB (g/L)
NaCl	31.4240
$CaCl_2$	0.1920
$MgCl_2$	0.2560
KCl	0.0640
Na_2SO_4	0.0640

2.2 Crude Oil

The crude oil used in this study is a dead crude oil from the oilfield that was used as case study. The crude oil has density of 0.9067 g/cc, viscosity of 15.22 cp and API of 24.57^o at 25 ^oC.

2.3 Brine Nanoparticles and Enzyme

The silica nanoparticles (SiO_2) used in this study is 99.5% non-porous silica without any surface treatment supplied by Skyspring Nanomaterials Inc. Houston, USA. The SiO_2 nanoparticles have particle size of 10-20 nm. A commercial 100% concentrate greenzyme supplied by Biotech Processing Supply Dallas, Texas was the enzyme used in this study.

3. EXPERIMENTAL METHOD

The experimental study was conducted in two phases that comprises of four sequences each. In the first phase, the effect of varied salinities on oil-brine interfacial interactions was investigated; the details of the experiment and the results are presented in [29]. In the second phase, the effect of varied brine composition on oil-brine interfacial interactions was investigated in four sequences. In the first sequence, the IFT of crude oil and each salt solution (NaCl, $MgCl_2$, $CaCl_2$, KCl and Na_2SO_4) were measured without the presence of either nanoparticles or enzyme. This was aimed at investigating the effect of each salt component on brine-oil interfacial interaction. The results of this phase were used to analyse the efficiency of SiO_2 nanoparticles and enzyme in their respective solutions. In the second and third sequences, the effect of nanoparticles and enzyme on these brines and crude oil interactions was investigated respectively. In the fourth sequence, the effect of combine SiO_2 nanoparticles and enzyme on these brines and crude oil interactions was investigated. All the tests were conducted with Sigma 703D tensiometer using the Du Nouy ring method. A fixed concentration of 1 g/L of nanoparticles and enzyme was used for all the tests and the experiments were conducted at ambient temperature of 25 ^oC. The choice of this concentration is based on past studies on successful EOR applications of the nanoparticles and enzyme at this concentration [5, 19]. A ratio of one-to-one nanoparticles to enzyme was used for the investigation of the effect of combined nanoparticles and enzyme.

4. RESULTS AND DISCUSSION

Figure 1 shows a comparison between the results of the IFT modifications of crude oil-water and crude oil-brines interactions using nanoparticles, enzyme, and combination of both. From the results, two distinct IFT regimes characterised by high and low IFT were observed. The IFT of the crude oil-water/brines interactions without nanoparticles and enzyme as well as crude oil-water/brines interactions with nanoparticles constitute the high IFT regime. This shows that the application of SiO_2 nanoparticles does not significantly influence the IFT of crude oil-water/brines interactions and hence, the results lay in the high regime and signifies no impact. The applications of enzyme and combine nanoparticles-enzyme however fall into the low regime characterised by low IFT due to their capacities to reduce IFT of the crude oil-water and crude oil-brines. This signifies a positive impact that can be explored for additional oil recovery.

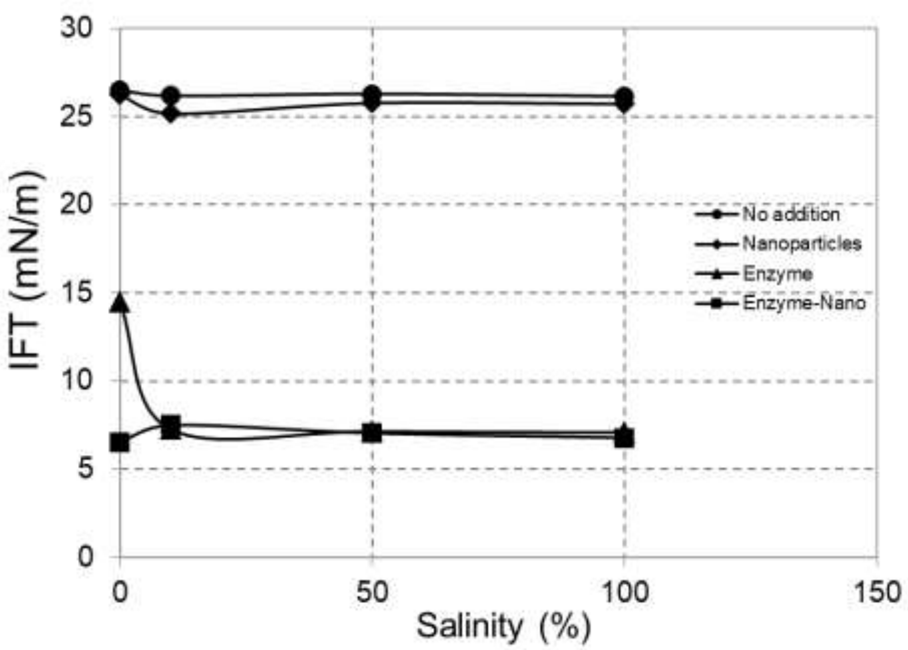

Figure 1: Comparison between the effects of SiO_2 nanoparticles, enzyme and SiO_2 nanoparticles-enzyme on crude oil-water and crude oil-brines interactions.

Figure 2 shows the results of the first sequence of the second phase in which the effect of brines of different compositions on oil-brine IFT modification was investigated in the absence of SiO_2 nanoparticles and enzyme. It is worth noting that in this phase of experiments, the concentration of each salt in the formation brine was used. This showcases the contribution of each salt composition to the interfacial performance of the formation brine. The results of these tests were compared with that of the deionised water and the formation brine (FMB) which serve as the base cases for no salt composition and combined salt composition. When only salt ions were present in the aqueous solution, the NaCl brine that constitute 98% of the formation brine effected better IFT reduction while the other salt solutions were characterised by insignificant IFT reduction that lies within similar range. This observed IFT reduction from 26.24 mN/m to 13.95 mN/m with the use NaCl solution is attributable to interfacial adsorption of the salt ions at the oil-brine interface. This is consistent with the previous study by Udoh and Vinogradov [7] that showed that surface activity of NaCl is higher in high salinity brine. All the other salt solutions were however characterised by high IFT. This invariably impacted the surface active of the FMB as evident by its high IFT which is attributable to screening effects of the other salt ions.

The results of the second sequence in which SiO_2 nanoparticles were added to the brines of varied compositions are presented in Figure 3. The addition of the SiO_2 nanoparticles to all the brines did not modify the IFT within experimental errors except for NaCl brine. This shows insignificant effect of the SiO_2 nanoparticles on the interfacial activity of these brine compositions. An increase in oil-NaCl solution IFT from 19.94 mN/m to 22.07 mN/m was however observed with the addition of SiO_2 nanoparticle to the brine. This shows the significant effect of SiO_2 nanoparticles interfacial adsorption. When nanoparticles adsorb at the oil-brine interface, the salt ions concentration at the interface depletes thereby resulting in dominant effect of nanoparticles. This is evident in the observed increase in IFT with the addition of nanoparticles to NaCl brine. This is consistent with the result of the molecular dynamics studies on the effect of salts and silica nanoparticles on oil-brine interfacial properties conducted by Wenhui *et al.* [30]. In which they observed interfacial adsorption of nanoparticles, but depletion of salt ions from the interface in brines. They also said that nanoparticles make water interface to be less orderly but have insignificant effect on oil-brine IFT. The results of these tests further showed that successful EOR application of the SiO_2 nanoparticles cannot be attributed to oil-brine IFT reduction because from all indications the nanoparticles have not demonstrated this potential.

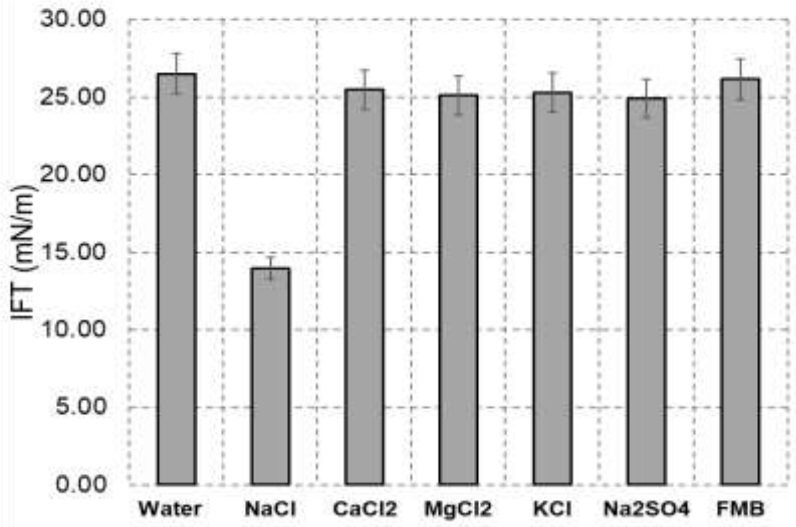

Figure 2: The effect of varied brine compositions on crude oil-brines interaction.

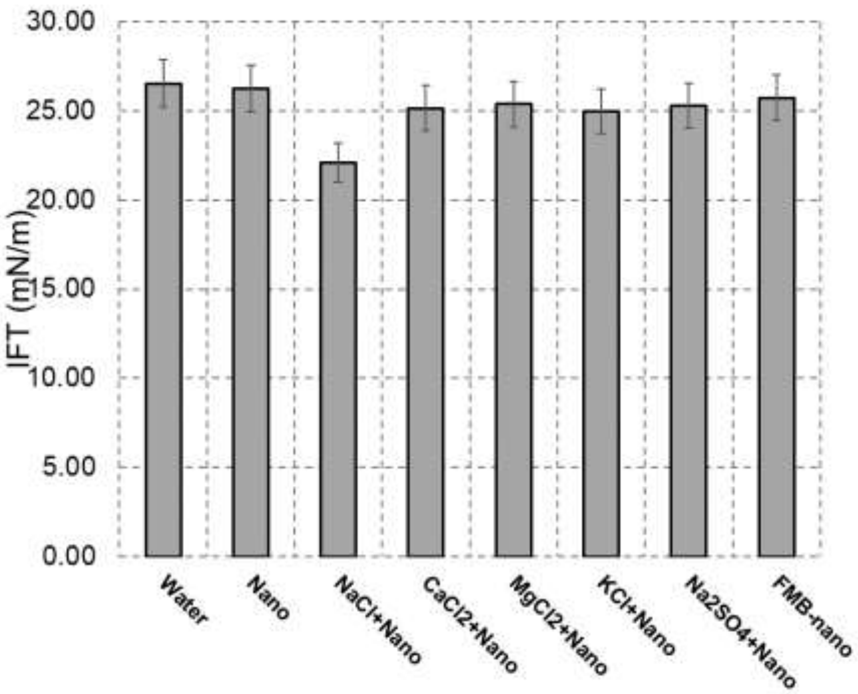

Figure 3: The effect of varied brine compositions and SiO_2 nanoparticles on crude oil-brines interaction.

Figure 4 shows the results of the third sequence of the second phase experiments in which the effect of enzyme on oil-brine IFT was investigated with brine of varied compositions. Generally, IFT reduction was observed with the addition of enzyme to all the brines, although at varied degrees. Better IFT reduction was achieved in divalent brines ($CaCl_2$ and $MgCl_2$), multicomponent brine (FMB) and high salinity monovalent brine (NaCl). This is consistent with previous study that showed that this enzyme has better stability and surface effect in high salinity and divalent brines than monovalent brines [7]. This probably explains why higher IFT was observed with KCl and Na_2SO_4 brines since their concentrations were very small. This result further shows that IFT reduction is one of the mechanisms underlying successful EOR applications of this enzyme as reported by other studies such as [4, 5, 23].

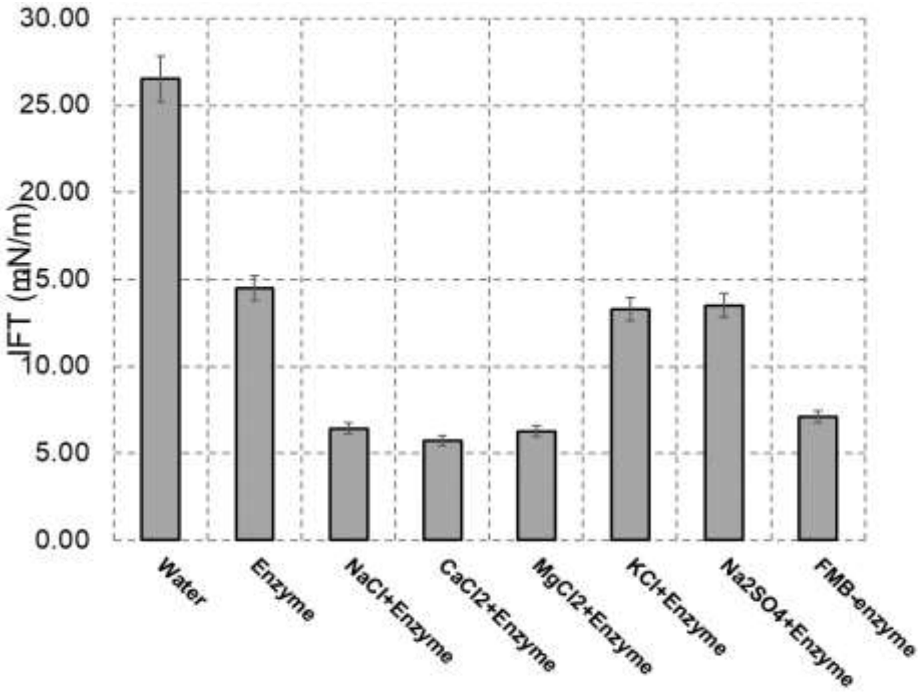

Figure 4: The effect of varied brine compositions and enzyme on crude oil-brines interaction.

The results of the final sequence of the phase two experiments in which the effect of combined nanoparticles-enzyme on oil-brine IFT was investigated are shown in Figure 5. The observed effect is like that of enzyme application alone (Figure 4) for all the salt compositions within experimental errors expect for NaCl that has a more pronounced increased IFT. This shows that combination of SiO_2 nanoparticles with enzyme did not make any positive effect on oil-brine IFT irrespective of the brine composition; hence, there is no need to combine them for IFT reduction. This however does not mean that their combination will not increase oil recovery because other factors may influence the enhanced oil recovery process. Previous studies have shown that SiO_2 nanoparticles can increase oil recovery and other mechanisms such disjoining pressure, wettability alteration, Brownian motion in addition to IFT reduction have been proposed as mechanisms underlying the increased oil recovery [14, 19, 31, 32].

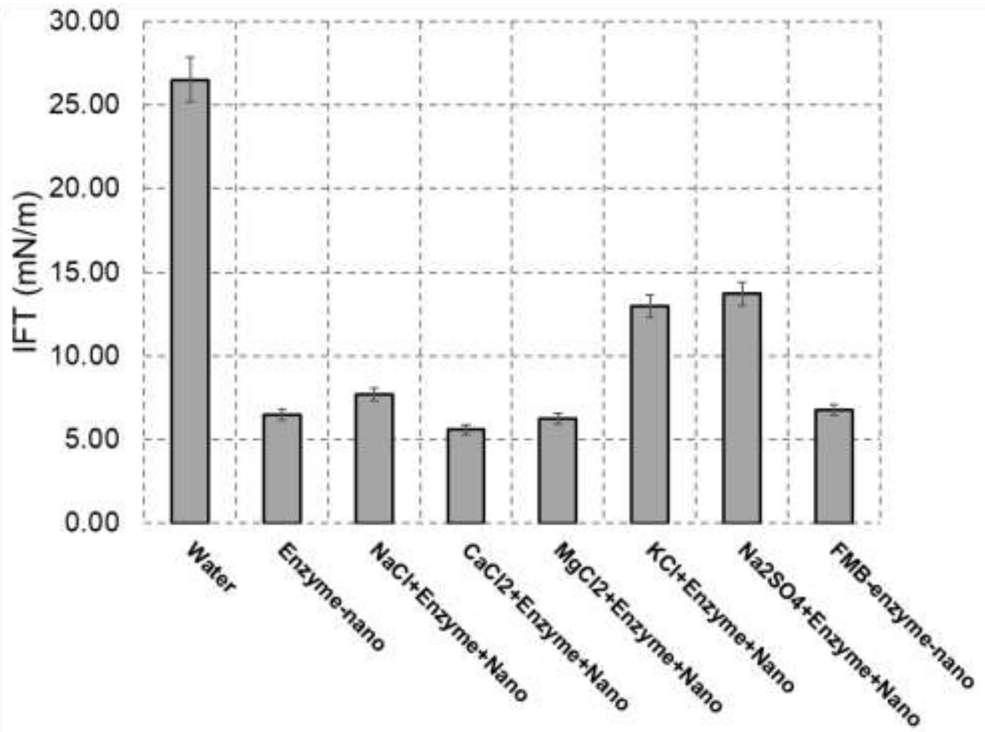

Figure 5: The effect of varied brine compositions, enzyme and SiO_2 nanoparticles on crude oil-brines interaction.

5. CONCLUSION

In this study, experimental investigations of the IFT modification potential of silica (SiO_2) nanoparticles, enzyme, and the combination of both were conducted under similar conditions and the results show that:

- Crude oil-water and crude oil-brines interactions were characterised by high IFT.
- The used silica nanoparticles as an IFT modification agent did not significantly modify the crude oil-water and crude oil-brines interactions.
- The application of enzyme as an IFT modification agent significantly influence crude oil-water and crude oil-brines interactions by reducing their respective IFT.
- Combination of nanoparticles and enzyme effected good IFT modification of crude oil-water interaction in the absence of any salt but makes no significant effect in oil-brine interactions.
- Brine salinity and composition do not significantly influence silica nanoparticles interfacial interaction effect.

ACKNOWLEDGMENT

The authors appreciate Biotech Processing Supply Dallas, Texas for the supply of the enzyme used in this study.

REFERENCES

[1] Liu, S. (2008). Alkaline Surfactant Polymer enhanced oil recovery process, PhD Thesis, Rice University, Houston, Texas, United State. https://hdl.handle.net/1911/22224.

[2] Towler, B., Lehr, H., Austin, S., Bowthorpe, B., Feldman, J., Forbis, S., Germack, D. & Firouzi, M. (2017). Spontaneous imbibition experiments of enhanced oil recovery with surfactants and complex nano-fluids, *Journal of Surfactants Detergent*, 20, 367-377.

[3] Johannessen, A. M. & Spildo, K. (2013). Enhanced oil recovery (EOR) by combining surfactant with low salinity injection, *Energy & Fuels*, 27(20), 5738–5749.

[4] Udoh, T., Akanji, L. & Vinogradov, J. (2018). Experimental Investigation of Potential of Combined Controlled Salinity and Bio-Surfactant CSBS in Enhanced Oil Recovery EOR Processes," in *Proceeding of SPE Nigeria Annual International Conference and Exhibition. Society of Petroleum Engineers, Lagos, Nigeria, 6-8 August, 2018, Paper* 193388, 1-22. doi:10.2118/193388-MS.

[5] Udoh, T. & Vinogradov, J. (2019). A Synergy between Controlled Salinity Brine and Biosurfactant Flooding for Improved Oil Recovery: An Experimental Investigation Based on Zeta Potential and Interfacial Tension Measurements, *International Journal of Geophysics*, 2019, 1-15.

[6] Rosen, M. (2004). Adsorption of surface-active agents at interfaces: the electrical double layer, *Surfactants Interfacial Phenomena*, 3, 34-104.

[7] Udoh, T. & Vinogradov, J. (2019). Experimental Investigations of Behaviour of Biosurfactants in Brine Solutions Relevant to Hydrocarbon Reservoirs, *Colloids Interfaces*, 3(24), 1-15.

[8] Karimi, A., Fakhroueian, Z., Bahramian, A. Pour Khiabani, N., Darabad, J., Azin, R. & Arya, S. (2012). Wettability alteration in carbonates using zirconium oxide nanofluids: EOR implications, *Energy Fuels*, 26, 1028-1036.

[9] Ogolo, N., Olafuyi, O. & Onyekonwu, M. (2012). Enhanced oil recovery using nanoparticles, *In Proceedings of the SPE Saudi Arabia Section Technical Symposium and Exhibition, Al-Khobar, Al-Khobar, Saudi Arabia, 8-11 April 2012, Paper 160847*.

[10] Suleimanov, B., Ismalov, F. & Veliyev, E. (2011). Nanofluid for enhanced oil recovery, *Journal of Petroleum Science and Engineering*, 78(2), 431-437.

[11] Roustaei, A. & Saffarzadeh, S. M. M. (2013). An evaluation of modified silica nanoparticles' efficiency in enhancing oil recovery of light and intermediate oil reservoirs, *Egyptian Journal of Petroleum*, 22, 427-433.

[12] Sajjad, M., Arezou, J. & Soheila, J. (2019). Temperature effect on performance of nanoparticle/surfactant flooding in enhanced heavy oil recovery, *Petroleum Science*, 16, 1387-1402.

[13] Engeset, B. (2012). The Potential of Hydrophilic Silica Nanoparticles for EOR Purposes: A Literature Review and an Experimental Study, MSc. Thesis, Norwegian University of Science and Technology, Trondheim.

[14] Udoh, T.(2021). Improved insight on the application of nanoparticles in enhanced oil recovery process, *Scientific African*, 13, e00873, 1-20.

[15] Cheraghian, G. & Khalili, N. S.(2015). Experimental Investigation of Polymer Solutions Used in Enhanced Oil Recovery: Thermal properties Improved by Nanoclay, in *Proceedings of 77th EAGE Conference and Exhibition, Madrid, Spain 1-4 June, 2015*.

[16] Ragab, A. & Hannora, A. A. (2015). Comparative investigation of nano particle effects for improved oil recovery–experimental work, *in Proceedings of SPE Kuwait Oil and Gas Show and Conference, Mishref, Kuwait, 11–14 October, 2015*.

[17] Hendraningrat, L. & Torsaeter, O. (2014). Unlocking the potential of metal oxides nanoparticles to enhance the oil recovery, in *Proceedings Offshore Technology Conference-Asia, Kuala Lumpur, Malaysia, 25-28 March, 2014*.

[18] Alomair, O. M. K. & Alsaeed, Y. (2014). Nanofluids application for heavy oil recovery, in *Proceedings* of *SPE Asia Pacific Oil & Gas Conference and Exhibition*, Adelaide, Australia.

[19] Udoh, T. (2021). Enhanced Oil Recovery Application of Nanoparticles in Niger Delta Water-Wet Reservoir Rock, in *Proceedings* of *Nigeria Annual International Conference and Exhibition*, *Lagos, Nigeria, 2-4 August, 2021*, paper 207150.

[20] Mohajeri, M., Hemmati, M. & SadatShekarabi, A. (). An experimental study on using a nanosurfactant in an EOR process of heavy oil in a fractured micro- model," *J. Petrol. Sci. Eng.*, 126, 162–173.

[21] Zargartalebi, M., Kharrat, R. & Barati, N. (2015). Enhancement of surfactant flooding performance by the use of silica nanoparticles," *Fuel*, 143, 21–27.

[22] Hamme, J. D. V., Singh, A. & Ward, O. P. (2006). Physiological aspects. Part 1 in a series of papers devoted to surfactants in microbiology and biotechnology, *Biotechnology Advances*, 24(6), 604–620.

[23] Udoh, T. & Vinogradov, J. (2019). Effects of Temperature on Crude-Oil-Rock-Brine Interactions During Controlled Salinity Biosurfactant Flooding, in *Proceedings of Nigeria Annual International Conference and Exhibition*, *Lagos, Nigeria, 2019, Paper 198761*.

[24] Feng, Q., Ni, F., Shao, D., Ma, X. Qin, B. & Zhou, L. C. (2007). Eor pilot tests with modified enzyme, in *Proceedings of EUROPEC/EAGE Conference and Exhibition Society of Petroleum Engineers*, China.

[25] Nasiri, H., Spildo, K. & Skauge, A. (2009). Use of enzymes to improve waterflood performance, in *Proceedings of International Symposium of the Society of Core Analysts*, Noordwijk, Netherlands.

[26] Wang, Y. K. A., Li, B., Li, Z. Wang, Q. & Zhao, M. (2008). New agent for formation-damage mitigation in heavy-oil reservoir: Mechanism and application, in *Proceedings of SPE International Symposium and Exhibition on Formation Damage Control*.

[27] Rahayyem, M., Mostaghimi, P., Alzahid, Y. A., Halim, A. Evangelista, L. & Armstrong, R. T. (2019). Enzyme enhanced oil recovery eeor: A microfluidics approach, in *Proceedings of SPE Middle East Oil and Gas Show and Conference.*.

[28] Salahshoor, S., Gomez, S. & Fahes, M. (2019). Experimental investigation on the application of biological enzymes for eor in shale formations, in *Proceedings of Unconventional Resources Technology Conference* , Denver, Colorado, USA.

[29] Udoh, T. H. & Orodu, O. D. (2022). Experimental Investigation on Effect of Enzyme and Nanoparticles on Oil-Brine Interfacial Tension, in *Proceedings of SPE Nigeria Annual International Conference and Exhibition*, Lagos, Nigeria 1 - 3 August 2022.

[30] Wenhui, L. Y. N., Qing, Y., Qichao, X. & Zhehui, J. (2020). Effects of salts and silica nanoparticles on oil-brine interfacial properties under hydrocarbon reservoir conditions: A molecular dynamics simulation study, *Journal of Molecular Liquids*, 305, 112860. https://doi.org/10.1016/j.molliq.2020.112860

[31] Li, S., Hendraningrat, L. & Torsæter, O. (2013). Improved oil recovery by hydrophilic nanoparticles suspension: 2-phase flow experimental studies in *Proceedings of International Petroleum Technology Conference* , Beijing, China.

[32] Moghaddam, R., Bahramian, A., Fakhroueian, Z. K. A. & Arya, S. (2015). Comparative study of using nanoparticles for enhanced oil recovery: wettability alteration of carbonate rocks, *Energy Fuels*, 29(14), 2111–2119.