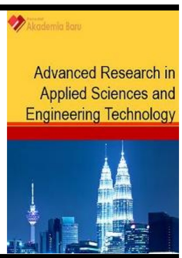
Journal of Advanced Research in Applied Sciences and Engineering Technology 22, Issue 1 (2021) 54-68

Penerbit Akademia Baru

Journal of Advanced Research in Applied Sciences and Engineering Technology

Journal homepage: www.akademiabaru.com/araset.html
ISSN: 2462-1943



Response Surface Method for Modelling the Effect of CO_2 in Brine/Waxy Oil Interfacial Tension during LSW-WAG Enhanced Oil Recovery

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ABSTRACT

Recent research on the combination of low salinity waterflood (LSW) with CO_2 water-alternating gas (WAG) has received significant attention due to its effectiveness in recovering residual oil in a mature field. The solubility of CO2 increases with decreasing brine salinity; and the presence of CO_2 in the injected water is expected to reduce the water/oil interfacial tension (IFT) leading to the release of trapped oil previously held in the rock by capillary forces. However, to date, little study has been done on the fluid/fluid interaction during the LSW-WAG process involving waxy crude oil and injected brine. In this study, two models have been developed from experimental IFT measurements that can facilitate the prediction of the CO_2 effect in the interfacial tension between oil/water interface in the presence and absence of CO_2. This objective is achieved by modelling the effect of pressure, brine salinity, and CO_2 on oil/water IFT with the response surface methodology (RSM) using the modified central composite design method (CCD) for the experimental design. Based on the developed model, the optimum values of input variables were calculated by analysis of variance (ANOVA) to obtain an acceptable model. The R-squared values demonstrate that the developed models could appropriately predict the experimental results of oil/water IFT from Dulang crude oil and 7 different brine salinity. The results of this study are expected to give insights into the fluid/fluid interaction behaviour during the LSW-WAG recovery process from a mature field with waxy crude oil.

Keywords:
Low salinity waterflood; water alternating gas; interfacial tension; response surface methodology; central composite design; enhanced oil recovery

Received: 12 February 2021 **Revised: 29 March 2021** **Accepted: 25 April 2021** **Published: 29 April 2021**

1. Introduction

Access to new energy resources from yet to explore hydrocarbon fields are getting more challenging and require significant upfront capital investment. Due to these reasons, recovering oil from existing fields through the Enhanced Oil Recovery (EOR) process is seen as a more feasible alternative as an upfront cost to develop a new field can be obviated [1]. EOR refers to the injection of chemicals, gas or steam as an external agent into the oil reservoir to promote additional oil

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https://doi.org/10.37934/araset.22.1.5468

recovery beyond what is typically realized when the reservoir produced under its natural drive or assisted by waterflooding[2]. In recent literature, it has been demonstrated that injection of brine with lower salinity than that existing in the reservoir formation, also known as low salinity waterflood (LSWF) could potentially recover more oil compared to conventional waterflood [3-6]. Many mechanisms have been proposed, with most studies showing desirable rock wettability changes as the main factor [7-10] For gas injection EOR, several studies have shown that injecting CO_2 alternating with water in WAG (water-alternating gas) process could further reduce the residual oil saturation after waterflooding process [11-13]. Nadeson Ganesan [13] evaluated the advantage of injecting immiscible water/alternating gas in a waxy crude field (Dulang) and obtained an additional oil recovery from this process. Initially, 56% recovery happened while injected water only, and a further 6.7% recovery occurred with this synergetic method [13]. In WAG, water acts to affect macroscopic sweep efficiency (the portion of reservoir rock volume swept by incoming fluid) while gas functions to improve microscopic sweep (the quantity of residual oil in the pores displaced by incoming gas)[14]. As such, combining LSWF in the water injection phase with CO_2 injection in LSW-WAG will offer the benefits of both processes and potentially result in higher oil recovery [15].

Reduction in formation brine concentration leads to lowering the interfacial tension between oil/water interface at the time low salinity water flooding [16]. Also, the contact time between the oil and water phase can affect the IFT trend [17]. Other than that, with increasing pressure, the value of IFT decreases. Alizadeh and Fatemi observed that the highest IFT reduction observed for the 10 times diluted seawater for low salinity water flooding [18]. Teklu observed that LSWAG CO_2 is an efficient EOR technique for different samples (carbonate and sandstone) [19]. Yang et al reported that there is a reduction of IFT between oil/water at room temperature and pressure to reservoir temperature and pressure [20]. Kumar et al observed that LSW with immiscible CO_2 can recover more than 65% oil [21].

In this paper, particular focus is given to IFT interaction between the oil and brine phases during LSW-WAG. In the presence of CO_2, Teklu *et al.*, [22] have shown that low salinity brine could dissolve more CO_2 in the brine phase, which leads to a reduction in oil/water IFT across the ranges of pressure and temperature in their study. Reduction in oil/water IFT will reduce the capillary forces trapping the residual oil in the pores, thus mobilizing the oil for production[23]. However, to date, a study on oil/water IFT for waxy crude oil in LSW-WAG at reservoir pressure and the temperature has received little attention. In this study, an investigation of IFT interaction for waxy crude oil from Dulang field with brine in the presence of CO_2 will be performed.

The investigation will determine the optimum fluid/fluid interaction as a function of brine concentration and pressure using the design of experiment (DOE) method. The objective of this paper is to establish a statistical model to predict the effect on the oil/water IFT in the presence and absence of CO_2 for Dulang crude oil during LSW-WAG. For this modelling, the response surface model (RSM) based on a modified central composite design has been used to achieve the proper model. The model is expected to provide insight on optimum brine concentration and pressure to achieve minimum oil/water IFT for application of LSW-WAG in the waxy crude oil reservoir.

2. Materials and Methods

2.1 Experimental Materials (Oil and Water Phase)

The crude oil used in this work was Dulang from a matured Malaysian basin with a Viscosity of (at 96°C) 0.625 cp, Pour Point 40°C and it is a waxy crude oil. Brine sample which includes formation and seawater brine taken from the field. A total of 7 samples with varying salinities has been prepared and used to achieve the objectives of this study.

Table 1
Brine compositions

Salt	Weight, g				
	LS1	LS2	LS3	LS4	LS5
$CaCl_2.2H_2O$	0.416	0.166	0.083	0.042	0.017
$MgCl_2.6H_2O$	5.136	2.055	1.027	0.514	0.205
$SrCl_2.6H_2O$	0.005	0.002	0.001	0.001	0.000
NaCl	11.859	4.744	2.372	1.186	0.474
Na_2SO_4	0.192	0.077	0.038	0.019	0.008
KCl	0.312	0.125	0.062	0.031	0.012
$NaHCO_3$	0.110	0.044	0.022	0.011	0.004

2.2 Sample Preparation

In this project, Formation brine (FB), High salinity seawater (HS), X1 times diluted seawater (LS1), X2 times diluted seawater (LS2), X3 times diluted seawater (LS3), X4 times diluted seawater (LS4), X5 times diluted seawater (LS5) were prepared with an estimated amount of salt (using electronic balance) based on composition in Table 1 and mixed with distilled water using a magnetic stirrer for 30 minutes each. As per the selected concentration, the concentrations were a range from 722-36080 ppm. This is shown in Table 2 below.

Table 2
Salinity for the brine samples

NO.	Brine Sample	Salinity (ppm)	
1	Formation Brine	FB	21400
2	High Salinity Seawater	HS	36080
3	0.5HS (2x)	LS1	18035
4	0.2HS (5x)	LS2	7214
5	0.1HS (10x)	LS3	3607
6	0.05HS (20x)	LS4	1804
7	0.02HS (50x)	LS5	722

2.3 Interfacial Tension (IFT) Measurement

At a constant temperature of 96°C, the IFT between the oil and water phases was estimated and observed. In all IFT measurements, the low salinity water was previously prepared. The heavy phase was Dulang, which defines crude oil in the reservoir. IFT 700 was used for this section of the experiment. This equipment can measure the interfacial tension between liquid-liquid and gas-liquid and high pressure and high-temperature condition using either the pendant drop or rising drop method [24]. In the case of this experiment, the IFT was measured at reservoir temperature conditions 96°C and high pressure (ranging from 200 psi to 2000 psi).

2.4 Experimental Design and ANOVA Analysis

RSM can optimize the response and predict future responses reliably by statistically calculating a regression model based on enough experimental data [25]. The RSM reduces the number of tests, which saves time and money in the experimental design process [26]. The response surface model is approximately represented by the experimenter with a model equation, while the behaviour of response variables is modelled as a function of a set of regression variables. When the model is expected satisfactory, it can be used to predict within the experimental region and to determine the operating conditions on the explanatory variables that provide the peak response but if the model is not satisfactory, more tests must be done to enhance the fit or an adjustment must be made in the mathematical form of the model [27]. The effect of salinity, pressure, and CO_2 on IFT has been determined from this model. When the model is properly developed, the accuracy of predicted IFT values was identified using ANOVA. The optimized value for oil/water interfacial tension with and without CO_2 was obtained by simultaneously minimizing and maximizing the validated model for IFT. Face-centred composite design (FCCD) is a three-level practical, experimental design in which the axial points are focused on the cubic surface rather than the sphere, and α is equal to 1. The IFT between oil/water has been optimized with the FCCCD and the operational correlation between independent factors and the response has been developed [28].

In this work design expert version, 11.1.0 has been used to get the experimental design and to generate the model. RSM modelling has many design styles, including. In this work modified central composite design was used for the design of the experiment (DOE) with CO_2 and without CO_2 gas. Concentration and pressures are the input data and IFT is the only output of this work. High and low levels were determined for the range of individual variables. The highest and lowest concentration was reported at 36080ppm and 722 ppm, with the pressure range between 200-2000 psi. The only response was assigned as interfacial tension between the oil/water phase. The experimental design matrix suggested 30 experimental runs for the experiment with CO_2 and 13 runs for the experiment without CO_2 based on the high and low levels for mentioned factors in RSM. To analyse, develop, and enhance the model parameters, the responses from the experiments were inserted into the corresponding response slots.

The statistical parameters and synergistic effects of every factor were evaluated using ANOVA. For the regression model, various suitability tests for ANOVA (lack-of-fit assessment, F-value, p-value) are recommended. Based on the agreement between the expected and observed responses, a statistical analysis using ANOVA was used to determine the degree of relevance for the chosen model Design Expert also provided 3D model graphs focused on the correlation of design variables, following the fitting of a suitable model.

3. Results and Discussions

3.1 RSM model for brine/oil IFT with CO_2

After obtaining data from the experimental run with CO_2 according to the suggested DOE were added in the predetermined slots for the response as shown in Table 3.

Table 3
Actual design matrix

	Factor 1	Factor 2	Response 1
Run	A:Concentration	B:pressure	IFT
	ppm	psi	N-m
1	722	200	17.79
2	722	2000	17.47
16	722	200	18.99
17	722	2000	16.5
26	722	200	18.69
27	722	2000	17.04

18	1804	1000	12.6
28	1804	1000	10.78
4	3608	200	23.21
5	3608	1800	13.7
19	3608	200	20.21
20	3608	1800	13.25
29	3608	200	22.5
30	3608	1800	13.25
6	7216	600	17.2
7	7216	1600	12
21	7216	600	18.175
22	7216	1600	11.2
3	18040	1000	10
8	18040	200	18.1
23	18040	200	19.21
24	18040	1000	12.21
9	21400	2000	11.49
25	21400	2000	11.95
10	36080	200	15.96
11	36080	1000	13.5
12	36080	1800	14.21
13	36080	1800	13.92
14	36080	200	16.31
15	36080	1000	14.5

After analysing all the parameters and response, a model has been suggested and optimized to get the desired value of interfacial tension.

Table 4
Model summary statistics for IFT between oil/water interface with CO_2

Source	Sequential p-value	Lack of Fit p-value	Adjusted R^2	Predicted R^2	
Linear	< 0.0001	< 0.0001	0.4493	0.3919	
2FI	0.5816	< 0.0001	0.4359	0.3685	
Quadratic	< 0.0001	< 0.0001	0.7015	0.6175	
Cubic	**< 0.0001**	**0.0641**	**0.9296**	**0.9067**	**Suggested**
Quartic	0.0641		0.9439	0.9042	Aliased

Table 4 represents, the suggested model for interfacial tension between the oil/water phase in presence of CO_2. The regular coefficient of determination (R^2), probability (Prob > F), adjusted coefficient of determination (Adj. R^2), and predicted coefficient of determination (Pred. R^2) are used to validate the suitability of regression models for Interfacial tension. In Table 4, the predicted R^2 value and adjusted R^2 value are highest for the cubic model respectively the values are 0.9067 and

0.9296. The sequential p-value is less than 0.0001, which implies that all the model terms are significant [29]. Based on all the coefficients the suggested model is the cubic model where the quartic model is aliased.

3.1.1 Model analysis

The cubic model (with CO_2 gas) is then subjected to an ANOVA analysis as the model is selected by the software. Table 5 represents the ANOVA for the cubic model (in presence of CO_2) and the first column represents all the parameters for the model where A is the concentration of brine (722-36080 ppm) and B was system pressure(200-2000psi).

In this model, the F-value is 46.50 which indicates the model is significant. Here, P-value is not more than 0.0001 which implies all the model terms are significant. In this case, A, B, AB, A^2, B^2, A^2B, AB^2, A^3, B^3 are significant model terms. For the significant model terms, the values should be less than 0.0001. if there several insignificant model terms are present, the reduction of these terms can improve the model. The Lack of Fit F-value of 2.86 implies there is a 6.41% chance that a Lack of Fit F-value this large could occur due to noise. Significant Lack of fit is bad. This relatively low probability (<10%) is troubling. Based on the sum of squares, mean square, F-value, P-value the software suggested the cubic model is significant. Moreover, an insignificant lack of fit ensures a good fit for the model.

Table 5
ANOVA for Cubic model with CO_2

Source	**Sum of Squares**	**Mean Square**	**F-value**	**p-value**	
Model	383.44	42.60	46.50	< 0.0001	significant
A-Concentration	22.13	22.13	24.16	< 0.0001	
B-pressure	48.33	48.33	52.75	< 0.0001	
AB	5.23	5.23	5.71	0.0258	
A^2	8.58	8.58	9.37	0.0057	
B^2	99.38	99.38	108.47	< 0.0001	
A^2B	15.62	15.62	17.04	0.0004	
AB^2	16.58	16.58	18.09	0.0003	
A^3	29.86	29.86	32.59	< 0.0001	
B^3	14.93	14.93	16.30	0.0006	
Residual	20.16	0.9162			
Lack of Fit	6.27	2.09	2.86	0.0641	not significant

Since the models have several negligible terms, they have been decreased and manually simplified by eliminating insignificant terms. After eliminating actual factors, the final empirical models can be expressed as follows,

$$IFT= 9.93-6.40A-7.34B+O.7610\ AB+1.53\ A^2+5.82B^2+2.50A^2B-2.79AB^2+8.38A^3+ 4.42B^3 \quad (1)$$

Here, in equation (1) A, B, AB, A^2, B^2, A^2B, AB^2, A^3, B^3 all these factors are significant so that these factors are used to generate the final empirical equation for determining interfacial tension between oil/water in presence of CO_2 gas.

Figure 1(a) indicates the normal probability vs externally studentized residual plot. The residual points (differences between the expected values and the test response values) on the straight line are used to ensure the regular distribution of the IFT model. Fig. 1(b) represents the expected and actual values of the IFT with CO_2 model are in place. Similarly, Fig. 1(c) displays the residual plot concerning increased expected response values. The random distribution of residuals within the graph's red limits demonstrates the precision and predictability of the model. Fig 1(d) indicates the residuals vs the run. In other words, the expected variables of the model do not show any clear increase or decrease [30,31]

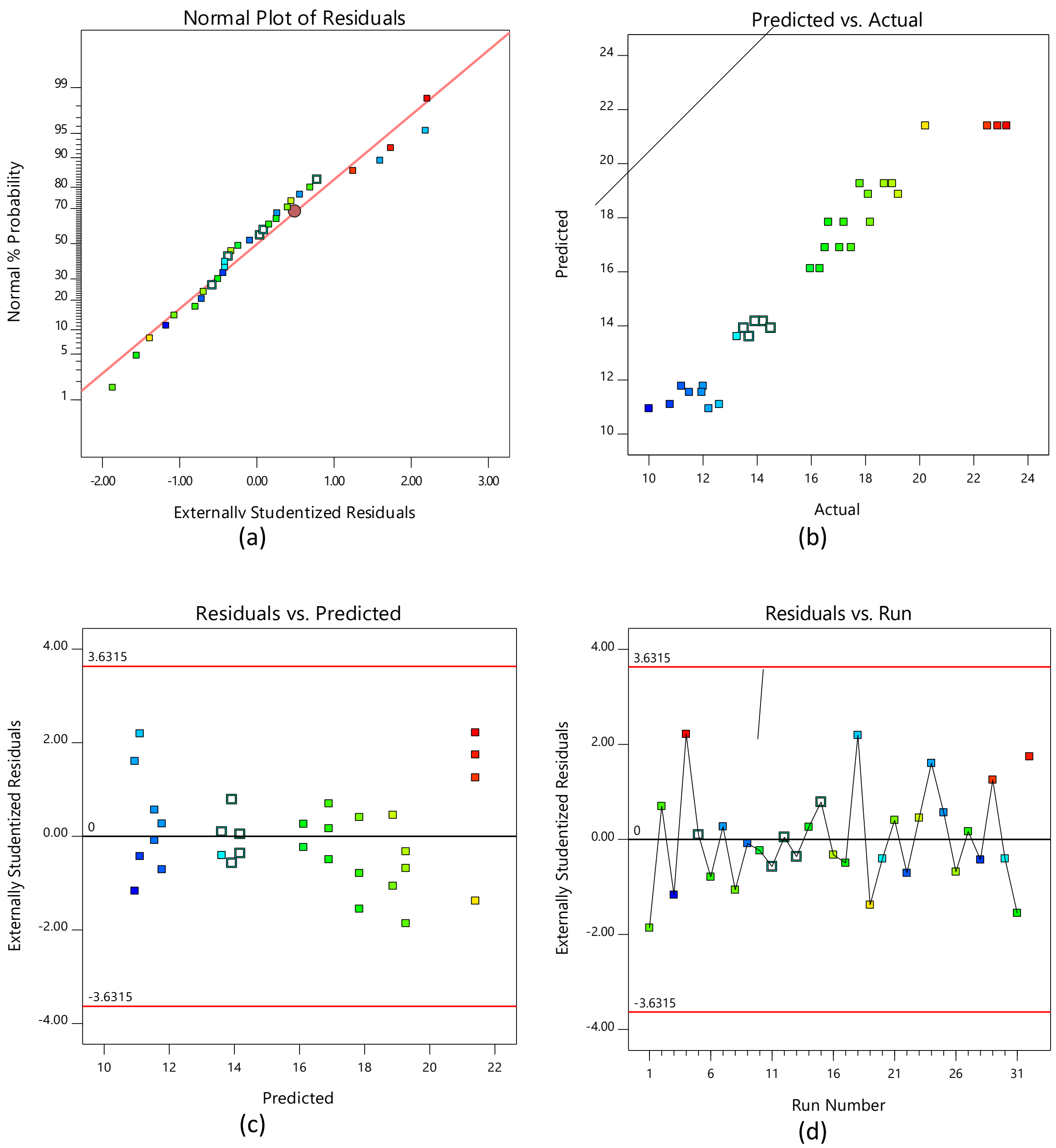


Fig. 1. Model diagrams for IFT with CO_2 gas: (a) Normal probability vs. residuals (b) Predicted vs. Actual (c) Residuals vs. predicted (d) Residuals vs. Run

From Figure 2(a) it has been seen that the value of IFT is dependent on the concentration of brine when the oil phase is Dulang (waxy crude) and the gas phase is CO_2. The IFT value is highest for the highest concentration of brine and the trend changes to its lowest value. At 722ppm the value of IFT was lower but at 26800 ppm the value of IFT was lowest. fig 2(b) indicates that when the pressure increases the value of IFT decreases with it. The highest value of IFT is at 200psi pressure and the lowest value of IFT is at 1515.89 psi. After 1515.89 psi with the increasing pressure, the value of IFT increases. So, 1515.89 was the pressure in which we can get the lowest IFT value in presence of CO_2 gas.

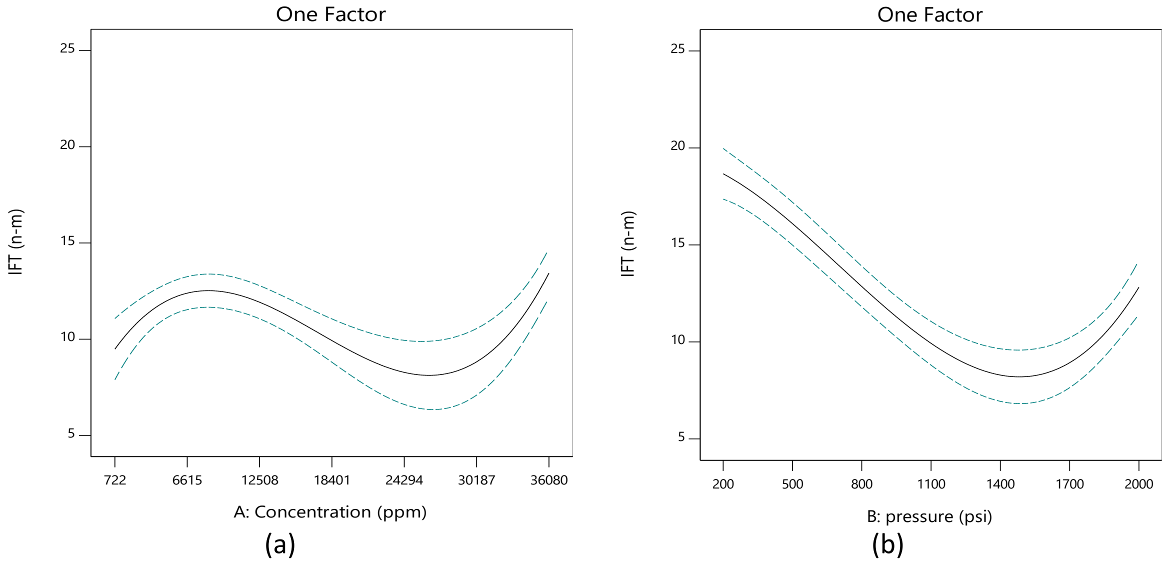


Fig. 2. IFT vs concentration and pressure (with CO_2) (a) IFT vs. Concentration (b) IFT vs. Pressure

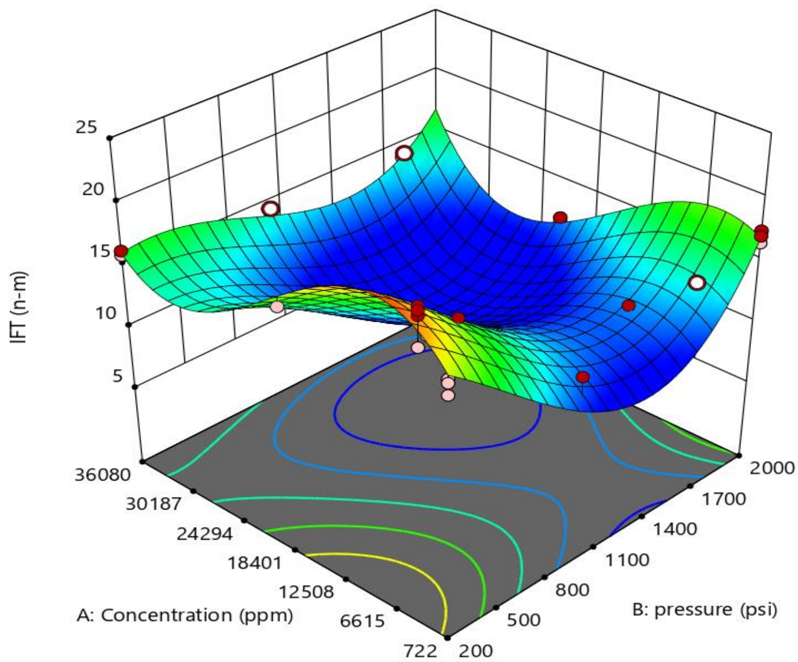


Fig. 3. Synergistic effects of factors on IFT (with CO_2)

This 3D graph from Figure 3 can be used to determine the synergetic effect of pressure, concentration, and CO_2 gas on the interfacial tension. Interfacial tension was observed to increase with rising concentration and to decrease with increasing strain. The effect of CO_2 is more prominent in the IFT with CO_2 than the IFT without CO_2. For example, when the IFT is lowest, the concentration of brine is 26800 ppm, and the pressure is 1515.89 psi.

3.2 RSM Model for Brine/oil IFT without CO_2

After obtaining data from the experimental run without CO_2 according to the suggested DOE were added in the predetermined slots for the response as shown in Table 6.

Table 6
Actual design matrix

	Factor 1	Factor 2	Response 1
Run	A:Concentration	B:pressure	IFT
	ppm	psi	n-m
1	722	200	12.9
5	722	2000	13.49
6	722	2000	15.49
2	3608	200	12.66
3	3608	200	12.66
4	3608	1800	10.25
7	7216	600	6.22
8	7216	1600	7.79
9	18040	200	7.91
10	21400	2000	9.339
11	36080	200	11.27
12	36080	1000	8.28
13	36080	1800	14.36

According to Table 7, the predicted R^2 value and adjusted R^2 value are highest for the quadratic model respectively the values are 0.7338 and 0.8985 but for other models, the values of R^2 are not acceptable due to their negative value. The sequential P-value is less than 0.0001 only for the quadratic model that means the confidence level more than 95% and the model is significant. Finally, the quadratic model is suggested by the software where the quartic model is aliased.

Table 7
Model summary statistics for IFT between oil/water interface without CO_2

Source	**Sequential p-value**	**Lack of Fit p-value**	**Adjusted R^2**	**Predicted R^2**	
Linear	0.6873	0.0833	-0.1133	-0.5539	
2FI	0.7803	0.0739	-0.2258	-1.5378	
Quadratic	**< 0.0001**	**0.6419**	**0.8985**	**0.7338**	**Suggested**
Cubic	0.4762	0.6111	0.9060	-23.1841	
Quartic	0.6111		0.8803		Aliased

3.2.1 Model analysis

In the absence of CO_2 gas, the model is suggested quadratic model and after that, the model is subjected to an ANOVA analysis where the first column represents the parameters for the model. A is the concentration of brine (722-36080ppm) and B is pressure (200-2000psi).

Table 8
ANOVA for Quadratic model without CO_2

Source	Sum of Squares	Mean Square	F-value	p-value	
Model	94.32	18.86	22.24	0.0004	significant
A-Concentration	0.0102	0.0102	0.0121	0.9156	
B-pressure	12.19	12.19	14.37	0.0068	
AB	6.49	6.49	7.66	0.0278	
A^2	49.09	49.09	57.88	0.0001	
B^2	38.12	38.12	44.94	0.0003	
Residual	5.94	0.8482			
Lack of Fit	3.94	0.7874	0.7874	0.6419	not significant

In Table 8, the F-value of this model is 22.24 which indicates the model is significant. Here, P-value is not more than 0.0001 which implies all the model terms are significant. In this case, A, B, AB, A^2, B^2 are significant model terms. For the significant model terms, the values should be less than 0.0001. if there several insignificant model terms are present, the reduction of these terms can improve the model. The Lack of Fit F-value of 0.79 implies there is a 64.19% chance that a Lack of Fit F-value this large could occur due to noise. Significant Lack of fit is bad. This relatively low probability (<10%) is troubling. Based on the sum of squares, mean square, F-value, P-value the software suggested the quadratic model is significant.

Final empirical models can be expressed as follows,

$$IFT = 3.09-0.0382A+1.29B+1.13AB+5.52A^2+5.47B^2 \quad (2)$$

Here, in equation 2 A, B, AB, A^2, B^2 all these factors were significant so that these factors are used to generate the final empirical equation for determining interfacial tension between oil/water without CO_2 gas. The equation is useful for identifying the relative impact of the factors by comparing the factor coefficients. This equation can be used to make the response predictions for a given level of each factor. In Figure 4 all the diagnostic plots are in the range so that it is clear that the model will predict all the responses accurately like the previous model.

In Figure 5(a) the IFT value decreases with the decreasing concentration and IFT is highest at the highest concentration. At 18504 ppm concentration, the IFT value is the lowest, and the trend moves upward with decreasing concentration. In Figure 5(b) the IFT value decreases with increasing pressure. At 1100psi the lowest value is detected. And after that, the value of IFT increased with the increasing pressure. The trend of IFT concerning pressure and concentration was likely similar. There is no significant change occurred like the IFT-pressure and IFT-concentration graphs with CO_2 gas.

This 3D graph in Figure 6 represents the synergetic effect of pressure, concentration on the interfacial tension when there is no presence of CO_2. Interfacial tension is observed to decrease with rising concentration and pressure. For example, the IFT value is lowest when the concentration of brine is 18400 ppm, and the pressure is 1100 psi.

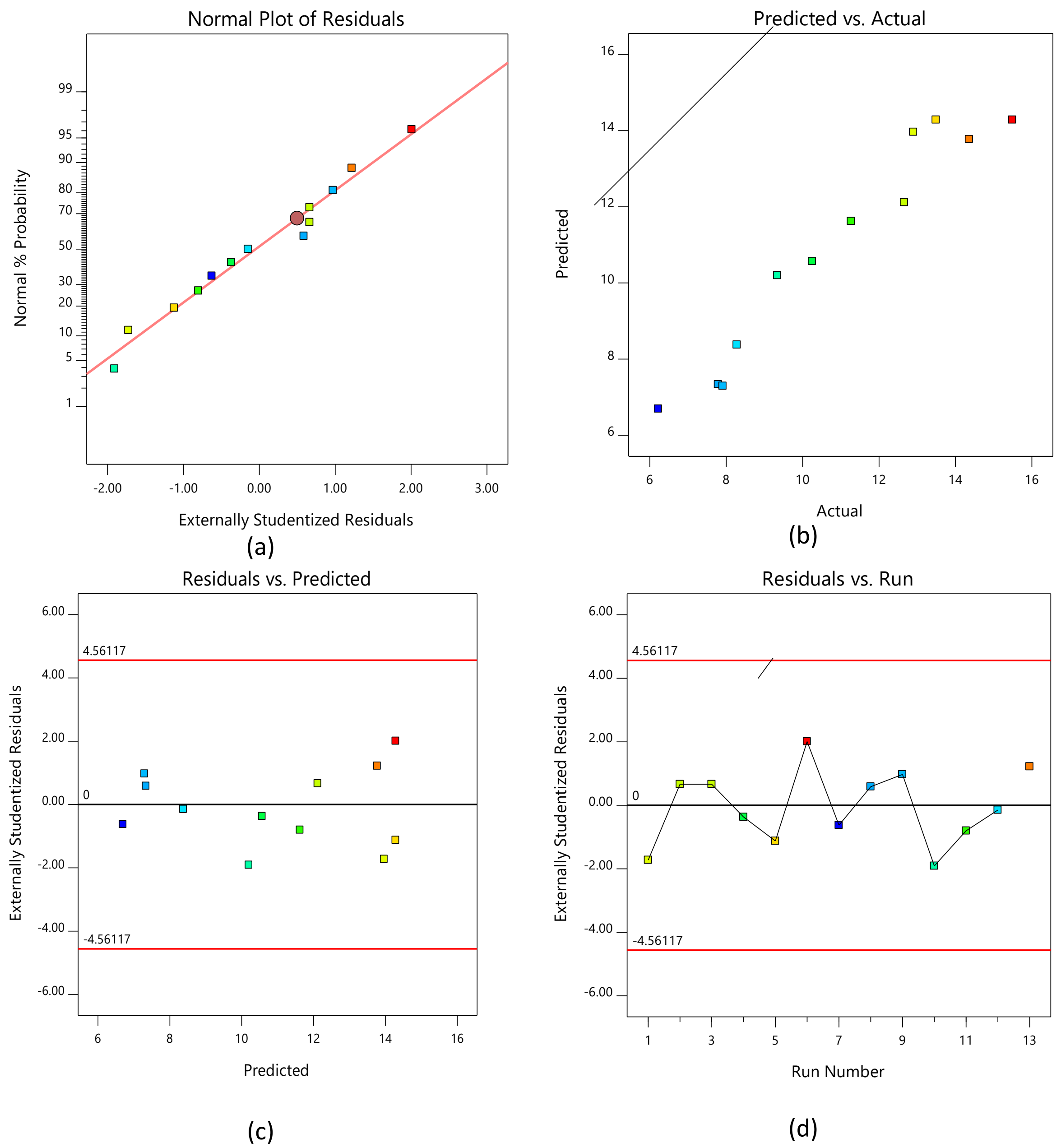


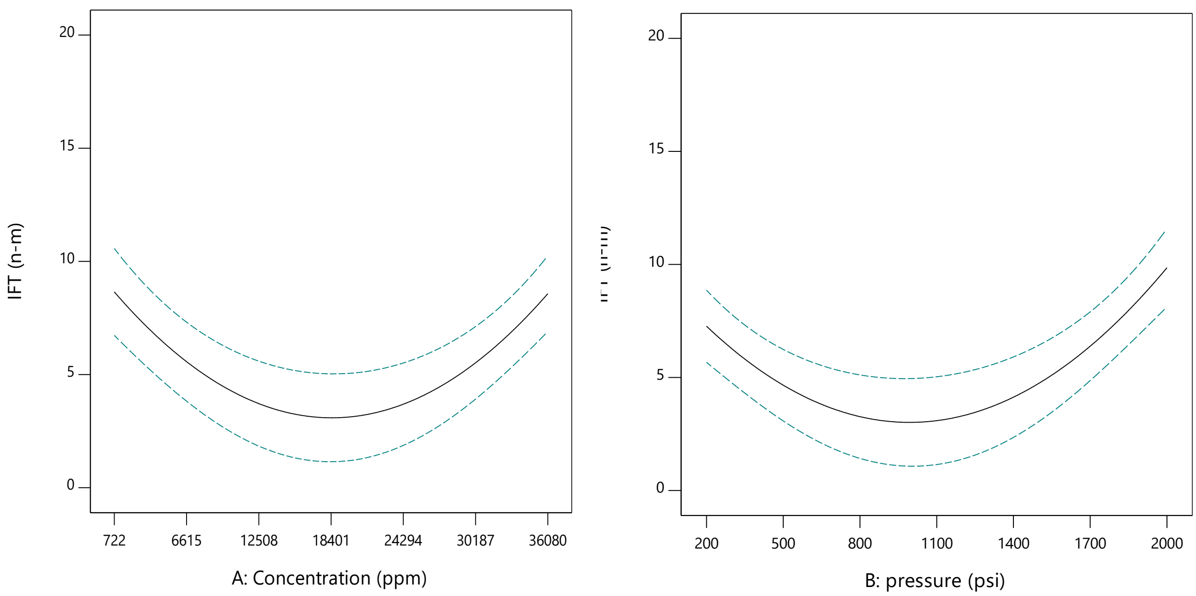
Fig. 4. Model diagrams for IFT without CO_2 gas. (a) Normal probability vs. residuals (b) Predicted vs. Actual (c) Residuals vs. predicted (d) Residuals vs. Run

3.3 Optimization using RSM

Based on the models obtained in the previous section, we further determined the optimum brine concentration and pressure that would lead to minimum oil/water IFT for both CO_2 and without CO_2 cases. RSM is a very effective technique to predict interfacial tension values in presence of CO_2 and the absence of CO_2 too.

Figure 7 provides a ramp presentation of the optimization and also represents brief optimization data of the IFT modelling with CO_2. Here, for optimization the concentration was in a target (21400 ppm), the pressure was in a target (600 psi) and the IFT value was minimized because when oil/water IFT is minimum, it will reduce the capillary force that held the oil, which further helps in mobilization and transportation of oil. The value of IFT is 11.3825 N-m for the concentration of 21400 ppm and the pressure 600 psi. The desirability of the predicted IFT value is 0.907. that means it is 90.7% acceptable.

For optimization, in Figure 8 the concentration is in a target (722), the pressure is in a target (800 psi) and the IFT value is minimized because when the IFT value is lowest, it is better for fluid movement held in the rock. the value of IFT is 11.3825 N-m for the concentration 722ppm and the pressure 800psi. Figure 8 also represents the desirability of the predicted IFT value is 0.946. that means it is 94.6% acceptable.



(a) (b)

Fig.5. IFT vs concentration and pressure (without CO_2) (a) IFT vs. Concentration (b) IFT vs. Pressure

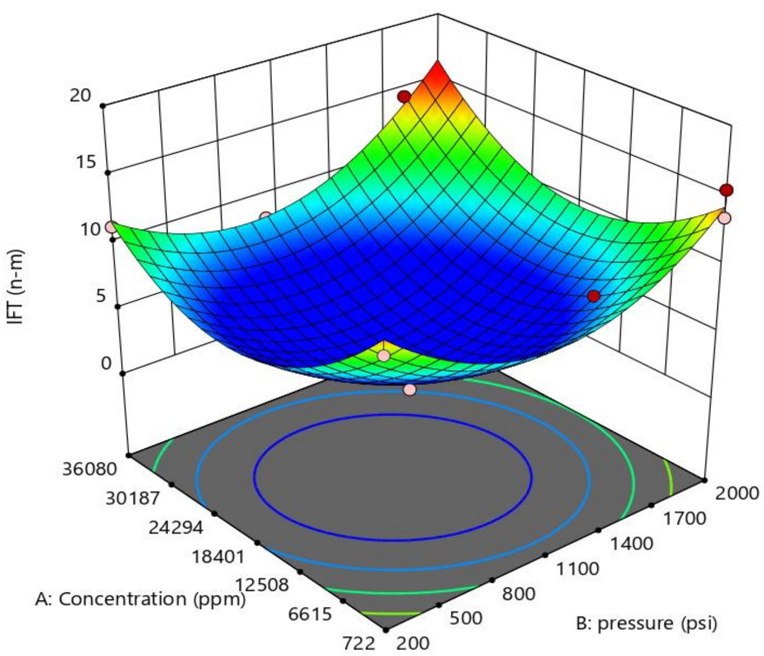


Fig.6. Synergistic effects of factors on IFT (without CO_2)

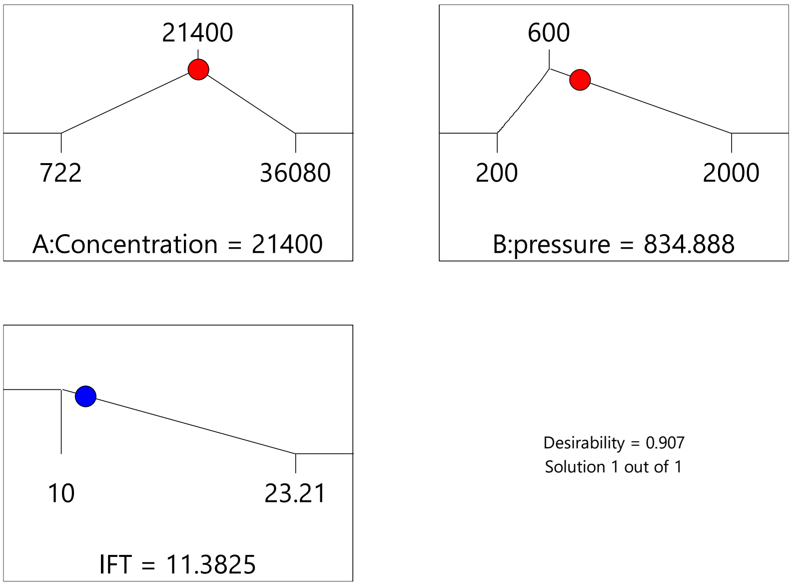


Fig. 7. Optimization ramp for IFT with CO_2

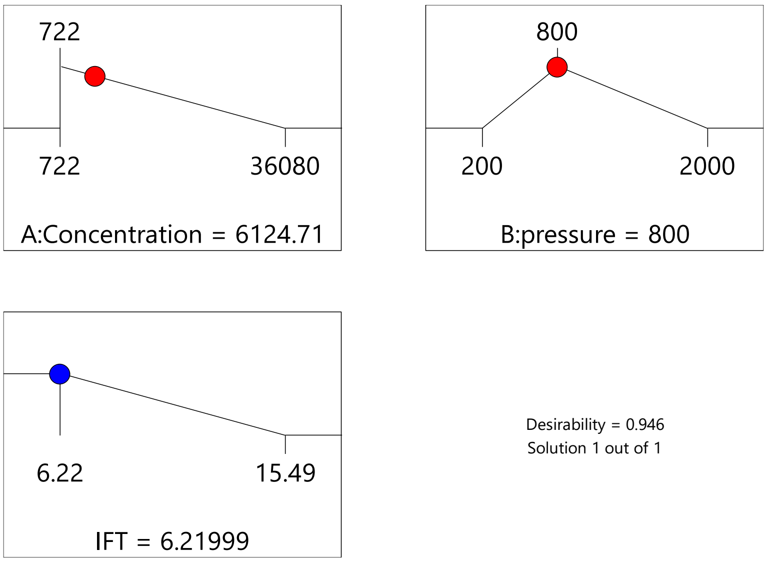


Fig. 8. Optimization ramp for IFT without CO_2

4. Conclusion

In this analysis, RSM has been used for modelling the interfacial tension between oil and water phases in the presence and absence of CO_2 to determine the impact of CO_2 gas on IFT. Using RSM physical characterization, statistical analysis, modelling, and interfacial tension optimization were investigated. To obtain an acceptable model, the optimum values of input variables were determined using analysis of variance (ANOVA) based on the established model. R-squared value shows the accuracy of the predicted value with the experimental value. It represents the accuracy of the model to predict all the test results of oil/water interfacial tension. These established models are capable to determine the interfacial tension between the oil/water phase for any pressure and brine concentration within the measured experimental values. The cubic model was suggested for the IFT with CO_2 and the quadratic model was suggested for the IFT without CO_2. By applying response data and manually excluding insignificant terms, the models were improved. The modelling was found to be reliable and accurate in both statistical and graphical terms. IFT with CO_2 the suggested model is cubic and without CO_2 the suggested model is quadratic. The effect of CO_2 on interfacial tension values is discussed based on the graphs and it is clear that CO_2 has a great effect on interfacial tension value. For the concentration of brine 26400 ppm and pressure 1515.89 psi, the lowest IFT value in presence of CO_2 gas was found. Also, the IFT value is the lowest when the concentration of brine is 18400 ppm, and the pressure is 1100 psi.

Acknowledgement
The author would like to acknowledge support from the Graduate Assistantship scheme for her MSc study. This research was funded by Yayasan UTP Fundamental Research Grant 015LC0-157. Technical assistance in the laboratory by Nurul Nadia Izwani Reepei is also deeply acknowledged.

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