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Sensitivity Analysis and Prediction of Wettability Alteration Using Response Surface Methodology and PHREEQC

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Abstract
In studying the wettability alteration of oil reservoirs in enhanced oil recovery (EOR) techniques like water injection methods, special attention is paid to the composition of injected water. However, the extent of the impact of other factors, such as the oil and rock site density, and oil and rock surface area, is not given much attention. In this study, we analyze the sensitivity of discussed factors. First, oil adhesion to the rock surface was simulated using PHREEQC software (a geochemical reaction model), then the simulation results were modeled using Design-Expert software (a statistical software package). According to the findings, oil site density had the most influence on the oil adhesion, followed by oil and rock surface area in the second place. Because oil site density indicates the amounts of carboxylate groups on the oil surface, the increase in these functional groups increases the electrostatic bonds resulting in increased oil adhesion. Furthermore, the effect of parameters increases by increasing pH, and impressment will decrease above pH 8.

Keywords: EOR, PHREEQC, Response Surface Methodology, Wettability Alteration, Sensitivity Analysis.

Introduction
The production of oil reservoirs strongly depends on the wettability of reservoir formation. Hence, engineers try to change the wettability from oil-wet to water-wet due to enhanced oil recovery [1-3]. Many factors affect the adhesion of oil to the rock surface, including pH and composition of water, rock and oil properties, and such factors. To investigate wettability alteration, the importance of these factors should be described. PHREEQC is a software that can study the oil adhesion to the rock surface [4-7]. In the rest of the manuscript, after reviewing issues related to wettability alteration, the Box-Behnken design (BBD) as a deployed method for studying the importance of effecting parameters on wettability alteration and sensitivity analysis will be discussed.
A surface complexation model (SCM) is developed to investigate complex reactions between rock and oil surfaces by Brady et al. [5]. Some studies investigated the effect of pH and brine salinity on oil adhesion using SCM [8-10]. Some researchers study the wettability alteration using bond product sum (BPS). The electrostatic force change between the oil/brine and rock/brine interfaces is explicitly represented by BPS [11-12].
In addition, utilizing chemical speciation modeling software like PHREEQC, investigations on modeling and optimizing the wettability alteration have been done [13]. Based on the initial conditions of a solution or mixture of solutions, such models may calculate ion speciation, saturation index, equilibrium conditions, and ionic strength [14].
Statistical techniques may also be used to model and optimize oil adhesion. Statistical modeling is a helpful tool for examining potential links between factors that influence a process and those that describe its efficiency. Developing such relationships allows process engineers to execute future process designs more efficiently and cost-effectively. Statistical modeling is a technique for detecting the substantial impacts of process inputs on process outputs. Further steps in such techniques might lead to the identification of an optimal operating parameter range, as well as the fitting of a model in the form of y = f(x), where y is the process output or response value, and x is a collection of process parameters or predictor variables. RSM (response surface methodology) is a well-known statistical method for this purpose that has been the subject of a few intriguing studies over the last decade [15-19]. RSM has the benefit of allowing

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researchers to comprehend the influence of changes in degrees of freedom on the optimal outcome and assisting investigators in determining which direction to alter predictor variables to maximize or reduce response variables.
Authors think that parameters, including pH, brine salinity, and water ions, could strongly affect the wettability alteration in water flooding with smart or low salt water [20-21]. This study investigates the importance of parameters that have received less attention, including oil site density, rock site density, oil surface area, and rock surface area. Forty-six simulation runs were performed using the PHREEQC to study the discussed issue. Obtained results were used to find the sensitivity of the wettability alteration regarding each of the input factors. BBD was used to understand sensitivity analysis of affecting factors. The rest of the manuscript will discuss the SCM, input parameters, response functions, and final results.

Materials and Methods

The PHREEQC program and the BBD were used to study the sensitivity analysis and prediction of the wettability alteration. The PHREEQC program simulates wettability alterations under the BBD's intended conditions at three levels and across five parameters. The bond product sum was used as the response function for this. The independent parameters of pH, oil site density, rock site density, oil surface area, and rock surface area were converted to a value between -1 and 1 using the conversion functions. 2D and 3D charts were used to investigate the effects of all of the provided parameters. The given conversion functions have the benefits of being simple to calculate and utilizing the dimensionless form of the parameters.

The Model Input Parameters

A surface complexing model was used to investigate the wettability alteration with bond product sum. The input parameters of SCM are listed in Table 1. According to previous studies, the effect of -NH+ on oil adhesion was negligible [22]. and to simplify and examine the subject more accurately, this functional group was removed from the oil. The bond product sum is the electrostatic bridge between oil and rock surface species with opposite charges. This parameter was calculated using the multiplication of surface species concentration with opposite charges, and the concentrations of surface species were calculated using PHREEQC software.
All simulations are done in the constant seawater composition listed in Table 2.

Table 1 SCM input parameters.

no	reaction	Log K(at 25 c^0)
	[a] Oil	
1	$-COOH \leftrightarrow -COO^- + H^+$	-5.0
2	$-COOH + Ca^{2+} \leftrightarrow -COOCa^+ + H^+$	-3.8
3	$-COOH + Mg^{2+} \leftrightarrow -COOMg^+ + H^+$	-4.0
	[b] Calcite	
4	$>CaOH + H^+ \leftrightarrow >CaOH_2^+$	12.85
5	$>CaOH + CO_3^{2-} + H^+ \leftrightarrow >CaHCO_3 + OH^-$	10.15
6	$>CaOH + CO_3^{2-} \leftrightarrow >CaCO_3^- + OH^-$	1.55
7	$>CaOH + H^+ + SO_4^{2-} \leftrightarrow >CaOH_2SO_4^-$	14.75
8	$>CO_3H \leftrightarrow >CO_3^- + H^+$	-3.58
9	$>CO_3H + Ca^{2+} \leftrightarrow >CO_3Ca^+ + H^+$	-2.6
10	$>CO_3H + Mg^{2+} \leftrightarrow >CO_3Mg^+ + H^+$	-2.1[c]
11	$>CO_3H + H + \leftrightarrow >CO_3H_2^+$	-8.3
12	$>CaOH \leftrightarrow >CaO - + H^+$	-24.73

a,c Ref [23].
b Ref. [24].

Table 2 Seawater composition

component	Weight (g/L)
Cl^-	0.678
SO_4^{-2}	0.03477
HCO_3^-	0.00292
Mg^{+2}	0.07299
Ca^{+2}	0.0118
Na^{+2}	0.56008

Design expert version 12 was used to study the importance of effecting parameters on wettability alteration and sensitivity analysis. A quadratic design model in the Box-Behnken design type for the response surface method was implemented by us. The factors and their ranges are reported in Table 3. It should be noted that oil surface area be calculated from the oil component that participates in electrostatic linkages, such as resin or asphaltene. Designed experiments and the response calculated using PHREEQC software are reported in Table 4. According to the BBD, 46 simulation runs are required to study the response.

Table 3 Factors examined

Factor	Name	Coded parameter	Units	Type	Minimum	Maximum	Coded Low	Coded High	Mean	Std. Dev.
A	pH	X_1		Numeric	2.00	10.00	-1 ↔ 2.00	+1 ↔ 10.00	6.00	2.39
B	oil site density	X_2	site/nm^2	Numeric	0.1000	7.13	-1 ↔ 0.10	+1 ↔ 7.13	3.62	2.10
C	rock site density	X_3	site/nm^2	Numeric	2.00	4.13	-1 ↔ 2.00	+1 ↔ 4.13	3.07	0.6352
D	oil surface area	X_4	m^2/g	Numeric	0.2000	0.7682	-1 ↔ 0.20	+1 ↔ 0.77	0.4841	0.1694
E	rock surface area	X_5	m^2/g	Numeric	1.0000	3.00	-1 ↔ 1.00	+1 ↔ 3.00	2.00	0.5963

Table 4 Designed experiments.

		Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Response 1
Std	Run	A: pH	B: oil site density	C: rock site density	D: oil surface area	E: rock surface area	bond product sum
			site/nm^2	site/nm^2	m^2/g	m^2/g	(µmol/m^2)^2
14	1	10	3.61561	2	0.48409	2	3.0318E-11
30	2	6	3.61561	4.13067	0.48409	1	2.7662E-11
22	3	6	7.13122	2	0.48409	2	5.1668E-11
11	4	6	0.1	3.06534	0.48409	3	1.7602E-12
4	5	10	7.13122	3.06534	0.48409	2	9.1684E-11
12	6	6	7.13122	3.06534	0.48409	3	1.1876E-10
21	7	6	0.1	2	0.48409	2	7.6532E-13
43	8	6	3.61561	3.06534	0.48409	2	4.1066E-11
3	9	2	7.13122	3.06534	0.48409	2	5.9794E-14
2	10	10	0.1	3.06534	0.48409	2	1.2805E-12
1	11	2	0.1	3.06534	0.48409	2	8.4776E-16
19	12	6	3.61561	3.06534	0.2	3	2.5449E-11
8	13	6	3.61561	4.13067	0.768179	2	8.7791E-11
13	14	2	3.61561	2	0.48409	2	2.0294E-14
27	15	2	3.61561	3.06534	0.768179	2	4.8368E-14
26	16	10	3.61561	3.06534	0.2	2	1.9201E-11
5	17	6	3.61561	2	0.2	2	1.1072E-11
32	18	6	3.61561	4.13067	0.48409	3	8.2985E-11
34	19	10	3.61561	3.06534	0.48409	1	2.3237E-11
6	20	6	3.61561	4.13067	0.2	2	2.2857E-11
42	21	6	3.61561	3.06534	0.48409	2	4.1066E-11
40	22	6	7.13122	3.06534	0.768179	2	1.2563E-10
29	23	6	3.61561	2	0.48409	1	1.34E-11
15	24	2	3.61561	4.13067	0.48409	2	4.0635E-14
33	25	2	3.61561	3.06534	0.48409	1	1.5241E-14
46	26	6	3.61561	3.06534	0.48409	2	4.1066E-11
38	27	6	7.13122	3.06534	0.2	2	3.2709E-11
20	28	6	3.61561	3.06534	0.768179	3	9.7747E-11
16	29	10	3.61561	4.13067	0.48409	2	6.2628E-11
45	30	6	3.61561	3.06534	0.48409	2	4.1066E-11

(Continued on the next page (Table 4)

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41	31	6	3.61561	3.06534	0.48409	2	4.1066E-11
24	32	6	7.13122	4.13067	0.48409	2	1.0666E-10
44	33	6	3.61561	3.06534	0.48409	2	4.1066E-11
18	34	6	3.61561	3.06534	0.768179	1	3.2583E-11
10	35	6	7.13122	3.06534	0.48409	1	3.9586E-11
7	36	6	3.61561	2	0.768179	2	4.2526E-11
37	37	6	0.1	3.06534	0.2	2	4.848E-13
35	38	2	3.61561	3.06534	0.48409	3	4.5721E-14
28	39	10	3.61561	3.06534	0.768179	2	7.3747E-11
25	40	2	3.61561	3.06534	0.2	2	1.2593E-14
9	41	6	0.1	3.06534	0.48409	1	5.8672E-13
36	42	10	3.61561	3.06534	0.48409	3	6.9711E-11
23	43	6	0.1	4.13067	0.48409	2	1.5814E-12
17	44	6	3.61561	3.06534	0.2	1	8.4831E-12
39	45	6	0.1	3.06534	0.768179	2	1.8621E-12
31	46	6	3.61561	2	0.48409	3	4.0198E-11

BBD is one of the response surface methods (RSM) used in studies with a wide range of variables. Statistical approaches for defining the connections between a target parameter and dependent variables are referred to as RSM. The response properties are reported in Table 5. As shown in Table 5, according to the Box-Cox plot of the Design-Expert, the best lambda is 0.09, and the current lambda is 1, so a base 10 log transform was used to improve data scattering, and the current lambda changed to 0.

Results and Discussion

Many parameters, including pH, oil and rock site density, and oil and rock surface area, are involved in the wettability alteration, and some have negligible effects. The importance of input parameters is investigated using the F-values given by the design of the experiment (DOE), based on the PHREEQC results and illustrated in Figure 1. It should be noted that the p-values of mentioned parameters are less than 0.05. The importance of each factor is shown in Figure 1, and it is evident that pH sensitivity is higher than other factors. The analysis of variables (ANOVA) for the quadratic model is reported in Table 6. The interactions between parameters were ignored because of their p-values. According to Table 6, the p-value of parameter interactions is more than 0.05, so they are removed in the modeling of the response by us.

Table 5 Response properties.

Response	Name	Units	Observations	Analysis	Std. Dev.	Transform	Model
R1	bond product sum	$(\mu mol/m^2)^2$	46	Polynomial	3.168E-11	Base 10 Log	Reduced Quadratic

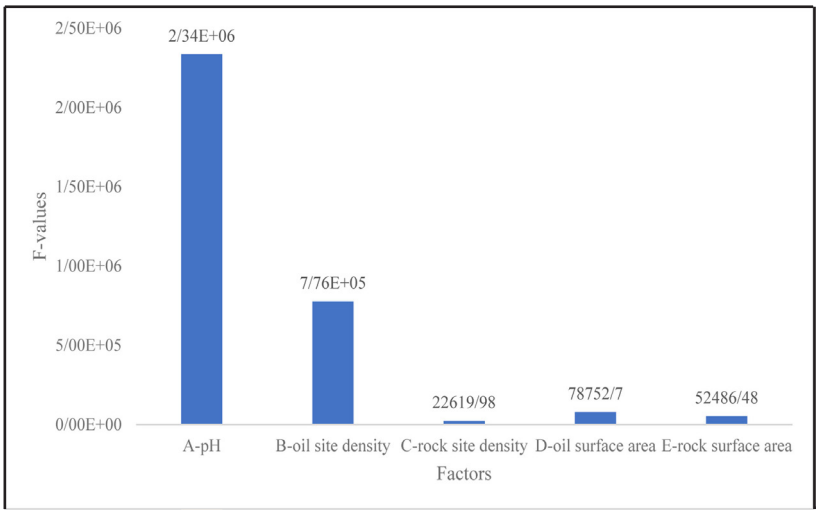

Fig. 1 Influencing level of parameters.

Table 6 ANOVA for the quadratic model.

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	81.32	20	4.07	2.344E+05	0.0001 >	significant
A-pH	40.51	1	40.51	2.335E+06	0.0001 >	
B-oil site density	13.47	1	13.47	7.762E+05	0.0001 >	
C-rock site density	0.3924	1	0.3924	22619.98	0.0001 >	
D-oil surface area	1.37	1	1.37	78752.70	0.0001 >	
E-rock surface area	0.9106	1	0.9106	52486.48	0.0001 >	
AB	0.0000	1	0.0000	0.6143	0.4405	Not significant
AC	0.0000	1	0.0000	2.64	0.1167	Not significant
AD	1.771E-11	1	1.771E-11	1.021E-06	0.9992	Not significant
AE	9.022E-11	1	9.022E-11	5.201E-06	0.9982	Not significant
BC	4.384E-08	1	4.384E-08	0.0025	0.9603	Not significant
BD	5.109E-11	1	5.109E-11	2.945E-06	0.9986	Not significant
BE	1.634E-12	1	1.634E-12	9.420E-08	0.9998	Not significant
CD	1.336E-12	1	1.336E-12	7.700E-08	0.9998	Not significant
CE	6.702E-11	1	6.702E-11	3.863E-06	0.9984	Not significant
DE	3.539E-12	1	3.539E-12	2.040E-07	0.9996	Not significant
A^2	20.65	1	20.65	1.190E+06	0.0001 >	
B^2	3.47	1	3.47	1.998E+05	0.0001 >	
C^2	0.0066	1	0.0066	380.57	0.0001 >	
D^2	0.0732	1	0.0732	4217.06	0.0001 >	
E^2	0.0339	1	0.0339	1955.23	0.0001 >	
Residual	0.0004	25	0.0000			
Lack of Fit	0.0004	20	0.0000			
Pure Error	0.0000	5	0.0000			
Cor Total	81.32	45				

The BPS is a function of surface species concentrations. The concentration of surface species changes with discussed factors, so the BPS is related to these factors and can be calculated using Equation 1.

$$\log(BPS) = 1.55148 * pH + 0.629648 * OSD + 0.295579 * RSD + 2.12695 * OSA + 0.487931 * RSA - 0.096139 * pH^2 - 0.050988 * OSD^2 - 0.024235 * RSD^2 - 1.13445 * OSA^2 - 0.062343 * RSA^2 \quad (1)$$

In Table 7, the R squares of the model are shown. The Predicted R^2 of 1.0000 is in reasonable agreement with the Adjusted R^2 of 1.0000. Adeq precision measures the signal-to-noise ratio. A ratio greater than 4 is desirable. The ratio of 2822.169 indicates an adequate signal. This model can be used to navigate the design space.

Table 7 Fit statistics of model.

Std. Dev.	0.0037	R^2	1.0000
Mean	-11.20	Adjusted R^2	1.0000
C.V. %	0.0334	Predicted R^2	1.0000
		Adeq Precision	2822.1692

According to Figure 2, the normal probability diagram shows how the residues follow a normal distribution. Even with normal data, some median distributions are expected.

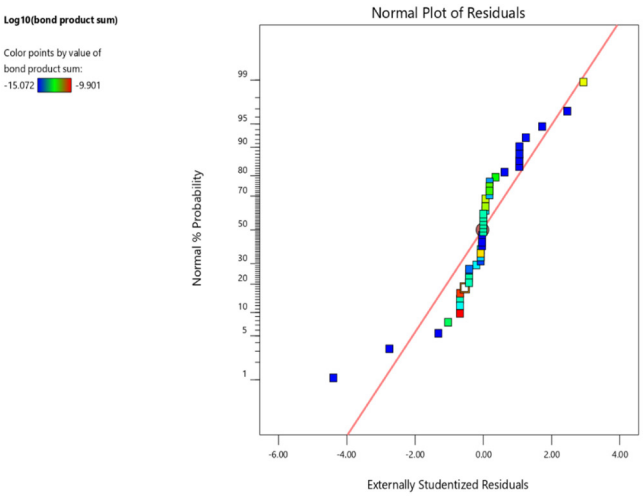

Fig. 2 The normal plot of residuals.

Figures 3 and 4 show the 3D and 2D sensitivity analysis of the effect of (a) oil site density, (b) rock site density, (c) oil surface area, and (d) rock surface area. According to the figures, the sensitivity of factors is significant in high pH, especially at a pH of 8. The effect of oil site density has consequential importance after the effect of pH, and it is due to the increase in the number of bonds with an increase in density. As the number of links increases, so does the BPS. The sensitivity of oil and rock surface area has significant importance after oil site density. This matter is consonant with Figure 1.

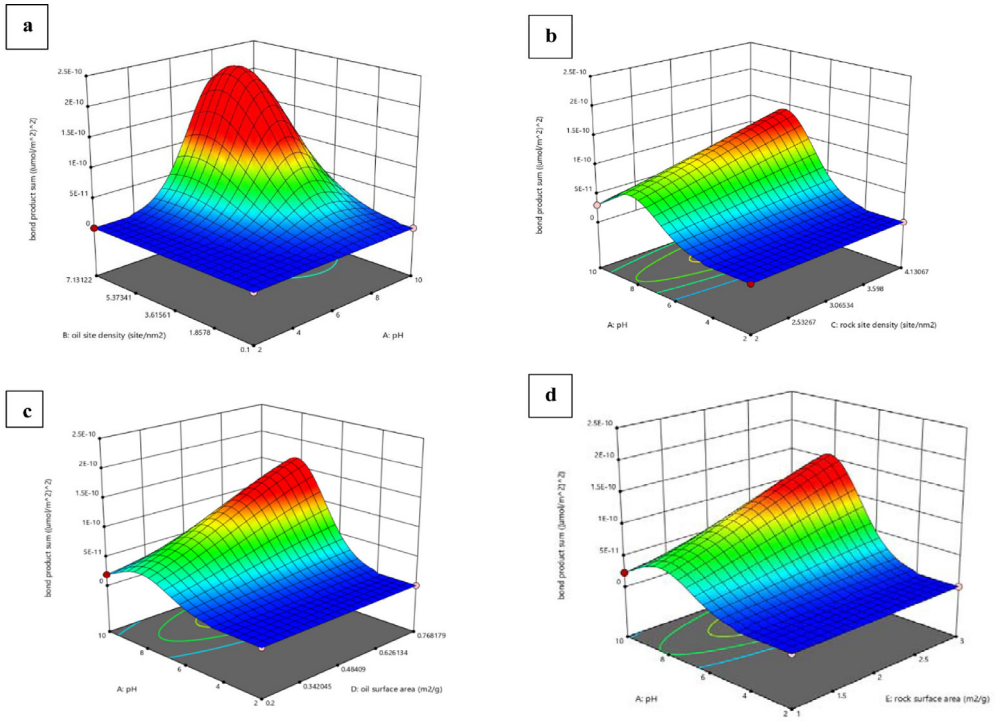

Fig. 3 Sensitivity analysis of bond product sum for (a) oil site density, (b) rock site density, (c) oil surface area, and (d) rock surface area in the presence of pH.

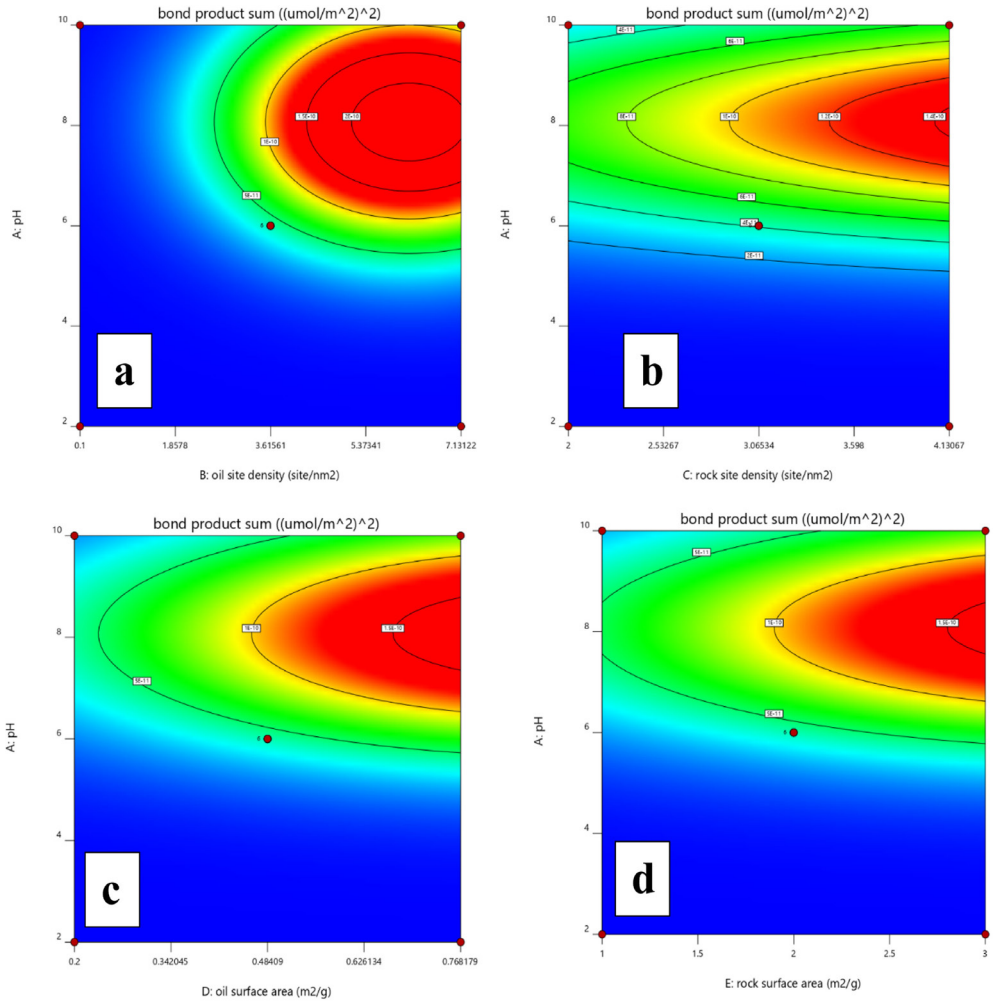

Fig. 4 Contour plot of sensitivity analysis of bond product sum for (a) oil site density, (b) rock site density, (c) oil surface area, and (d) rock surface area according to pH..

Validation of Proposed Models

The obtained response function discussed above has a high R^2 and readjusted R^2. As a result, they are tailored to the input data. Some data points were chosen to verify the consistency between PHREEQC software outputs and BBD outcomes to assess the validity of suggested response function relations. Data points used for the validation are shown in Table 8, representing that the results of the PHREEQC software and response functions of the BPS have distinguished consistency.

Table 8 Actual and predicted values.

Run Order	Log Actual Value Using PHREEQC	Log Predicted Value Using RSM	Residual	Internally Studentized Residuals	Externally Studentized Residuals	Influence on Fitted Value DFFITS	Standard Order
1	-10.52	-10.52	-0.0007	-0.207	-0.204	-0.118	14
2	-10.56	-10.56	0.0002	0.061	0.060	0.035	30
3	-10.29	-10.28	-0.0033	-1.031	-1.032	-0.596	22
4	-11.75	-11.76	0.0034	1.057	1.059	0.611	11
5	-10.04	-10.05	0.0086	2.658	2.933	1.693(1)	4
6	-9.93	-9.92	-0.0022	-0.685	-0.680	-0.393	12
7	-12.12	-12.12	0.0020	0.628	0.623	0.360	21
8	-10.39	-10.39	0.0000	0.000	0.000	0.000	43
9	-13.22	-13.23	0.0054	1.678	1.725	0.996	3
10	-11.89	-11.88	-0.0115	-3.561	-4.396	-2.538	2
11	-15.07	-15.06	-0.0082	-2.527	-2.754	-1.590	1
12	-10.59	-10.59	-0.0003	-0.078	-0.077	-0.044	19
13	-10.06	-10.06	0.0002	0.064	0.063	0.036	8
14	-13.69	-13.70	0.0075	2.301	2.461	1.421	13
15	-13.32	-13.32	-0.0001	-0.038	-0.038	-0.022	27
16	-10.72	-10.72	0.0006	0.189	0.187	0.108	26
17	-10.96	-10.95	-0.0014	-0.423	-0.417	-0.241	5
18	-10.08	-10.08	0.0002	0.061	0.060	0.035	32
19	-10.63	-10.63	0.0006	0.184	0.182	0.105	34
20	-10.64	-10.64	0.0002	0.064	0.063	0.036	6
21	-10.39	-10.39	0.0000	0.000	0.000	0.000	42
22	-9.90	-9.90	-0.0022	-0.691	-0.686	-0.396	40
23	-10.87	-10.87	-0.0014	-0.420	-0.415	-0.240	29
24	-13.39	-13.39	-0.0042	-1.304	-1.318	-0.761	15
25	-13.82	-13.82	-0.0001	-0.034	-0.033	-0.019	33
26	-10.39	-10.39	0.0000	0.000	0.000	0.000	46
27	-10.49	-10.48	-0.0022	-0.690	-0.685	-0.395	38
28	-10.01	-10.01	-0.0003	-0.077	-0.076	-0.044	20
29	-10.20	-10.20	0.0012	0.364	0.360	0.208	16
30	-10.39	-10.39	0.0000	0.000	0.000	0.000	45
31	-10.39	-10.39	0.0000	0.000	0.000	0.000	41
32	-9.97	-9.97	-0.0018	-0.549	-0.543	-0.314	24
33	-10.39	-10.39	0.0000	0.000	0.000	0.000	44
34	-10.49	-10.49	-0.0002	-0.076	-0.075	-0.043	18
35	-10.40	-10.40	-0.0022	-0.689	-0.684	-0.395	10
36	-10.37	-10.37	-0.0014	-0.423	-0.418	-0.241	7
37	-12.31	-12.32	0.0034	1.054	1.055	0.609	37
38	-13.34	-13.34	-0.0001	-0.038	-0.037	-0.022	35
39	-10.13	-10.13	0.0006	0.186	0.183	0.106	28
40	-13.90	-13.90	-0.0001	-0.038	-0.037	-0.021	25
41	-12.23	-12.23	0.0034	1.053	1.054	0.609	9
42	-10.16	-10.16	0.0006	0.186	0.183	0.106	36
43	-11.80	-11.80	0.0040	1.239	1.249	0.721	23
44	-11.07	-11.07	-0.0003	-0.078	-0.077	-0.044	17
45	-11.73	-11.73	0.0034	1.057	1.059	0.611	39
46	-10.40	-10.39	-0.0014	-0.425	-0.420	-0.243	31

Conclusions

The simulation results show that all factors, including pH, oil site density, rock site density, oil surface area, and rock surface area, influence the wettability alteration. The two most influencing parameters in the BPS prediction are pH and oil site density because oil surface area indicates carboxylate groups participating in electrostatic linkages. The pH leads to hydrolyzing of these functional groups, which changes the electrostatic bonds and wettability. Several factors, including pH, oil and rock surface area, and oil and rock site density, are known to affect wettability alteration, but pH and oil site density are more influential. According to the findings, the BPS of oil site density, rock site density, oil surface area, and rock surface area are 2.27e-10, 1.4e-10, 1.65e-10, and 1.51e-10, respectively, at constant pH of 8.2. It shows oil site density had the most BPS and affected wettability alteration more than other parameters. The sensitivity of discussed factors increases with an increase in pH. After the pH of 8, sensitivity partially decreases.

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Nomenclature

BPS	Bond product sum $((mol/m^2)^2)$
OSD	Oil site density $(site/nm^2)$
RSD	Rock site density $(site/nm^2)$
OSA	Oil surface area (m^2/g)
RSA	Rock surface area (m^2/g)

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