An optimization framework for sandstone acidizing using design of experiment (DOE) and mathematical modelling

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Abstract. Fluoroboric acid (HBF₄) serve as one of the alternatives for conventional mud acid in the application of sandstone wells stimulation. Various parameters such as formation temperature and acid injection velocity would significantly affect the performance of sandstone acidizing and hence determine the success rate of well stimulation. It is therefore undeniable that a deep understanding of the effects of these major parameters are of paramount importance. However, there is a scarcity of data available in the literature regarding the use of HBF₄ in sandstone acidizing in comparison to the use of mud acid. In this work, an optimization framework is developed to study the combined effects of formation temperature and acid injection velocity to the change in porosity and pressure drop. Apart from porosity improvement, a pressure drop across the sandstone core would also give an indication to the acidizing performance. The optimization approach is achieved by using design of experiment (DOE) and response surface methodology, coupled with a mechanistic model for sandstone acidizing. The design of experiment used in this work is central composite design (CCD). Meanwhile, the mechanistic model that simulate a flow in porous media is being developed using COMSOL Multi-physics, which is a computational fluid dynamics (CFD) software that uses finite element method (FEM). In this optimization tool, a range of formation temperature was set between 41˚C and 88˚C, whereas the range of acid injection velocity was set between 1.79×10⁻⁵ m/s to 3.78×10⁻⁵ m/s. According to the results, the optimum condition studied was found out to be 88˚C and 3.78×10⁻⁵ m/s. Under such an operating condition, the favourable maximum porosity enhancement and pressure drop profile were obtained. The maximum porosity and pressure drop were up to 17% and 16.6979 kPa respectively. The porosity enhancement and pressure drop in the sandstone core showed an excellent agreement with the data predicted by the model. In general, this optimization study had proven that response surface methodology (RSM) could be applied to determine the acid performance in sandstone acidizing.

1. Introduction
Well stimulation is one of the major concerns in the petroleum engineering industry to improve the overall oil and gas recovery in a sandstone reservoir [1, 2]. Apart from hydraulic fracturing and chemical explosion well stimulation methods, matrix acidizing is another commonly applied method
in the field [3, 4]. Sandstone acidizing would greatly enhance the porosity and permeability of a sandstone formation through mineral dissolution [5, 6].

Although mud acid had been widely applied as the acidizing fluid, it had been proven to be inefficiency in stimulating high temperature wells. This is due to rapid acid spending but low porosity and permeability improvement [7]. Therefore, many other alternative acidizing fluids are developed by researchers over the past decades such as the fluoroboric acid (HBF₄) [8, 9], organic acids [10], chelates [11] and retarded acids [12]. HBF₄ is one of the acids that could enhance the porosity and permeability of the sandstone matrix at high temperature conditions [5, 13]. However, its performance could be affected by many other factors such as acid concentration, acid injection velocity, sandstone mineralogy and heterogeneity. Hence, it is important to study their effects [14]. It was found that there is insufficient understanding on the effect of various parameters that affect the performance of HBF₄, particularly the reservoir temperature and injection velocity of acid [15].

In this study, the goal is to develop a robust and reliable optimization framework to evaluate the effect of multiple factors to the overall performance and success rate of sandstone acidizing. The optimization of sandstone acidizing process is performed using the design of experiment (DOE) and response surface methodology (RSM). The first DOE is incorporated in this work by adopting the central composite design (CCD), while the latter RSM is a popular optimization method, particularly in the analytical science field [16]. This optimization framework is then integrated with the computational fluid dynamics (CFD) numerical simulation using COMSOL Multiphysics software and finite element method (FEM). Overall, this study would enhance the knowledge of sandstone acidizing. A better understanding of the coupled effects of formation temperature and injection velocity of acid on the porosity profile and pressure drop profile would be achieved. The developed optimization framework also served as a platform for further study of other parameters.

2. Mathematical Modelling

Firstly, all the governing equations are modelled, creating a complete mechanistic model that simulate the sandstone core acid flooding process. The physics that is adopted in the model is the reacting flow in porous media. Then, a 3D core scale cylindrical geometry is created to simulate a sandstone core plug. These equations are then solved using the finite element method (FEM). The following equations are the key equations used in the mathematical modelling [17].

Equation (1) is the pressure equation.

$$\frac{1}{\alpha} \frac{\partial}{\partial x} \left( k \frac{\partial P}{\partial x} \right) + \frac{1}{\alpha} \frac{\partial}{\partial y} \left( k \frac{\partial P}{\partial y} \right) + \frac{1}{\alpha} \frac{\partial}{\partial z} \left( k \frac{\partial P}{\partial z} \right) = 0$$

Equation (2) is the mass balance equation for HBF₄.

$$\frac{\partial (C_{HF} \phi)}{\partial \tau} + \nabla \cdot (\rho \mu \dot{C}) = -r_h$$

Equation (3) is the mass balance equation for acids.

$$\frac{\partial (\rho \phi)}{\partial \tau} + \nabla \cdot (\rho \mu \dot{C}) = -\sum_{j=1}^{N} E_{f,i,j} S_{ij} V_j (1-\phi) C_i^{\alpha}$$

Equation (4) is the mass balance equation for minerals.
\[ \frac{\partial}{\partial t}(1-\phi)\frac{\partial j}{\partial t} = -\sum_{i=1}^{N_{di}} MW_i S_i V_i (1-\phi) \beta_j \frac{E_i}{\rho_j} C_i^{a_j} \quad j = 1, 2, 3 \]  

(4)

Equation (5) is the porosity change equation.

\[ \frac{\partial \phi}{\partial t} = -\sum_{j=1}^{N_{di}} \sum_{i=1}^{N_{si}} MW_i S_j V_j \beta_{j,i} \frac{E_{i,j}}{\rho_j} C_i \]  

(5)

Equation (6) – (8) are the boundary conditions applied.

\[
\begin{align*}
C_{HF} &= C_i^0 \quad \text{at } x = 0 \\
Q &= \text{Constant} \\
P &= P_{\text{out}} \quad \text{at } x = L \\
\frac{\partial P}{\partial r} &= 0 \quad \text{at } r = r_c
\end{align*}
\]  

(6)

(7)

(8)

Equation (9) is the initial condition applied.

\[
\begin{align*}
C_{HBF_s} &= C_{HF} = C_{H_2SiE_s} = 0 \\
V_1 &= V_1^0 \\
V_2 &= V_2^0 \quad \text{at } t = 0 \\
V_3 &= V_3^0 \\
\phi &= \phi^0
\end{align*}
\]  

(9)

3. Optimization framework

3.1. Response surface methodology (RSM)
Response surface method is applied in order to determine the optimum condition for sandstone acidizing process, in which responses such as porosity enhancement and pressure drop are affected by variables such as formation temperature and acid injection velocity. The conventional parametric study of the factors affecting the sandstone acidizing performance is conducted by altering only one variable at a time, while keeping all other parameters constant. Nevertheless, this method cannot identify the optimum condition when the whole process is also sensitive to various other factors. The RSM could empirically study the relationship between the formation temperature and acid injection velocity.

3.2. Central composite design (CCD)
The design expert software is used to carry out regression and analysis of graphical data. The optimization of porosity enhancement and pressure drop were conducted by using the central composite design (CCD), whereby it consisted of six replicates at each centre points. In present optimization investigation, the range of formation temperature was set as 41°C to 88°C, meanwhile the
range of acid injection velocity was set as $1.79 \times 10^{-5}$ m/s to $3.78 \times 10^{-5}$ m/s. The ranges of value set for each variable are shown in Table 1.

### Table 1. Summary of factors.

| Factor | A                  | B                  |
|--------|--------------------|--------------------|
| Name   | Temperature        | Acid Injection Velocity |
| Units  | °C                 | m/s                |
| Type   | Numeric            | Numeric            |
| Minimum| 25.00              | $1.11 \times 10^{-5}$ |
| Maximum| 105.00             | $4.46 \times 10^{-5}$ |
| Coded Low | $-1 \leftrightarrow 41.22$ | $-1 \leftrightarrow 1.78904 \times 10^{-5}$ |
| Coded High | $+1 \leftrightarrow 88.78$ | $+1 \leftrightarrow 3.78096 \times 10^{-5}$ |
| Mean   | 65.00              | $2.78 \times 10^{-5}$ |
| Std. Dev. | 20.16             | $8.444 \times 10^{-6}$ |

After that, the designed simulation sets are run using the COMSOL Multiphysics, CFD software. The porosity and pressure drop are then taken as the DOE. Table 2 showed the summary of responses obtained in this study.

### Table 2. Summary of responses.

| Response | R1                   | R2                   |
|----------|----------------------|----------------------|
| Name     | Porosity             | Pressure drop        |
| Units    | [1]                  | Pa                   |
| Analysis | Polynomial           | Polynomial           |
| Minimum  | 0.12397              | 1270.99              |
| Maximum  | 0.172131             | 16697.9              |
| Mean     | 0.1398               | 6432.13              |
| Std. Dev. | 0.0136              | 3828.80              |
| Ratio    | 1.39                 | 13.14                |
| Transform | None               | None                 |
| Model    | Quadratic            | Quadratic            |

### 4. Results and discussion

#### 4.1. Porosity response

The porosity change is one of the most reliable indication for the performance and effectiveness of a sandstone acidizing process. The change in porosity indicated that the amount of pore space in the sandstone core matrix had been increased due to dissolution of minerals presented in it. The initial porosity of the sandstone core is 0.12. After acid stimulation, the porosity is increased. Table 3 represented the fit summary for porosity response.

### Table 3. Fit summary for porosity response.

| Source   | Sequential p-value | Adjusted $R^2$ | Predicted $R^2$ |
|----------|--------------------|----------------|-----------------|
| Linear   | < 0.0001           | 0.8730         | 0.8051          |
| 2FI      | 0.0007             | 0.9562         | 0.9313          |
| Quadratic| < 0.0001           | 0.9959         | 0.9828 Suggested |
| Cubic    | < 0.0001           | 1.0000         | 0.9970 Aliased   |
Based on Table 3, a quadratic model is selected for the prediction of porosity. The sequential p-value of the quadratic model was less than 0.05, indicating that the model terms are significant. Therefore, the model terms of formation temperature and acid injection velocity are significant. If the value is greater than 0.10, then the model terms are not significant. Meanwhile, the predicted $R^2$ value of 0.9828 is in reasonably good agreement with the adjusted $R^2$ value of 0.9959, showing a difference of less than 0.2. Figure 1 shows the predicted vs actual plot for porosity response. The results indicated that the results of model prediction are well matched with the actual simulation data, hence proven the reliability of the optimization model.

![Figure 1. Predicted vs Actual plot for porosity response.](image1)

Figures 2 and 3 show the contour plot and 3D surface plot for porosity responses respectively. According to the results obtained, it was evident that the temperature had a significant effect on the porosity enhancement of the sandstone. A higher porosity increment is observed at higher temperature. This is because the hydrolysis rate of HBF$_4$ at high temperature also increases, thus dissolving the minerals more effectively. Besides, the increment in acid injection velocity also positively increase the porosity of sandstone matrix. However, it is clearly observable that the effect of acid injection velocity is not as remarkable as the effect of formation temperature.

![Figure 2. Contour plot for porosity response.](image2)
4.2. Pressure drop response

The pressure drop is an evident when the porosity across the sandstone core plug is increased. Therefore, a pressure drop is a useful response to determine the efficiency of sandstone acidizing. When acid is being injected into the core, it begins to react and dissolve the minerals, hence resulting in immediate pressure drop. Table 4 shows the fit summary for pressure drop response.

| Source     | Sequential p-value | Adjusted $R^2$ | Predicted $R^2$ |
|------------|--------------------|----------------|-----------------|
| Linear     | < 0.0001           | 0.9051         | 0.8511          |
| 2FI        | < 0.0001           | 0.9824         | 0.9610          |
| Quadratic  | 0.0090             | 0.9924         | 0.9670          |
| Suggested  | 1.0000             | 0.9987         | 0.9987          |

As shown in Table 4, a quadratic model is also suggested for the prediction of pressure drop. Since the sequential p-value of the quadratic model is less than 0.05, the model terms are significant. Thereafter, the model terms of formation temperature and acid injection velocity are significant. Moreover, the predicted $R^2$ value of 0.9670 is in reasonably good agreement with the adjusted $R^2$ value of 0.9924, showing a difference of less than 0.2. Figure 4 depicted the predicted vs actual plot for pressure drop response. The results proved that the results of model prediction are nicely matched with the actual simulation data, hence showing that the optimization model is reliable.

![Figure 3. 3D surface plot for porosity response.](image)

![Figure 4. Predicted vs Actual plot for pressure drop response.](image)
Figures 5 and 6 show the contour plot and 3D surface plot for pressure drop responses respectively. According to the trend of plots obtained, it was clearly shown that both the temperature and acid injection velocity had a significant effect on the pressure drop across the sandstone. A higher pressure drop is observed at both higher temperature and higher injection velocity. This is because the rate of HBF$_4$ hydrolysis at high temperature increases, thus creating more reacting surface between the acid and the minerals. Furthermore, the higher injection velocity would increase the rate of acid penetration into the sandstone core, resulting in higher efficiency of mineral dissolution. Therefore, it is noted that a high temperature and high acid injection velocity would favour the overall pressure drop response.

**Figure 5.** Contour plot for pressure drop response.

**Figure 6.** 3D surface plot for pressure drop response.

5. **Conclusion and recommendation**
An optimization approach using RSM had been integrated with the CFD mathematical modelling to study the coupled effect of formation temperature with the velocity of acid injection to the performance of sandstone matrix acidizing. After being simulated using COMSOL Multiphysics, the responses of porosity enhancement and pressure drop across the sandstone core sample were obtained
and analysed. In present optimization investigation, the ranges of formation temperature and acid injection velocity are set as 41°C to 88°C and 1.79×10^{-5} m/s to 3.78×10^{-5} m/s respectively.

The optimization results indicated that both the factors, temperature and injection velocity are significant in predicting the porosity and pressure drop response of sandstone matrix acidizing. Furthermore, the predicted results from the optimization model showed good agreement with the actual data from the simulation. The optimum condition determined from the optimization model is 88°C and 3.78×10^{-5} m/s because this operating condition would favour the sandstone acidizing performance. A maximum porosity improvement as well as pressure drop were observed. The maximum porosity is 17% whereas the maximum pressure drop is 16.6979 kPa.

Overall, this study had shown that RSM is a reliable and robust method for optimizing the operating condition of sandstone matrix acidizing. In the future, it is also recommended to apply this optimization tool to predict other sandstone acidizing responses such as permeability, acid consumption, mineral dissolution and precipitations scenarios. In addition, more parameters like acid concentration and mineral contents could be added into this optimization framework to investigate their effects to sandstone acid stimulation performance.

6. List of variables
All the variables used in this paper have been tabulated in Table 5.

| Symbol | Description | Unit |
|--------|-------------|------|
| \(i\) | Type of variables | - |
| \(j\) | Type of variables | - |
| \(P\) | Pressure | Pa |
| \(C_j\) | Concentration of HBF\(_4\) | mol/m\(^3\) |
| \(t\) | Time | min |
| \(\phi\) | Porosity | l |
| \(\bar{u}\) | Vector velocity | m/s |
| \(r_h\) | Hydrolysis rate of HBF\(_4\) | mol/m\(^3\)/s |
| \(C_i\) | Concentration of acid | mol/m\(^3\) |
| \(V_j\) | Volume fraction of mineral | l |
| \(N_m\) | Total number of minerals | l |
| \(E_{f,i,j}\) | Acid-Mineral Reaction rate | m/s |
| \(S_j\) | Reaction surface of mineral | l/m |
| \(MW_i\) | Molecular weight of acid \(i\) | g/mol |
| \(\rho_j\) | Density of mineral \(j\) | kg/m\(^3\) |
| \(\beta_{i,j}\) | Dissolving power of mineral | l |

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