Optimization and Controlling of FeCO3 Scale Deposition on the Piping System with Laminar Flow and in The Presence of Zeolite Absorbance

by Samsudi Raharjo

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Abstract. The scale of ferrous carbonate ($FeCO_3$) deposited on the pipe wall was investigated in the present study. In the experimental work, the scale-forming solution was prepared by mixing equimolar solutions of $FeCl_2$ and Na_2CO_3. Zeolite is used as an adsorbent because it has a crystal structure of alumina silica with cavities containing metal ions Fe^{2+}. It was found that in all the experiments, the mass scale $FeCO_3$ decreased with increased mass zeolite adsorbance addition. The mass scales obtained in the presence of 4000 mg zeolite was 14.2613 mg of $FeCO_3$. Similarly provided 22.1642 mg of mass scale was found in the presence of 3000 mg zeolite. The increasing mass scale 34.2652 mg was also produced at 2000 mg zeolite. Clearly the zeolite adsorbation may be used to improve the scale produced. The conductivity decreased abruptly after a certain induction period. The induction time was observed in the $FeCO_3$ system almost doubled in the presence of 2000 mg zeolite (28 min), tripled with 3000 mg zeolite (42 min). Moreover, the presence of 4000 mg zeolite (44 min) led to an induction time four times greater than that for in the absence of zeolite system (22 min). The optimum result of temperature was 44.639^0C, zeolite was 3625.068 mg variables was obtained so that the optimum the mass scale $FeCO_3$ was 14.46187 mg.

Keywords: Ferrous carbonate, Zeolite, Scale morphology

1. Introduction

Ferrous carbonate ($FeCO_3$) is a scale formed in a natural process and is a problem that is often encountered in hydraulic pipes on heavy equipment [1]. The scale is a deposit of inorganic compounds deposited and forms a crystalline heap on the surface of a substance. This is due to the presence of scale-forming elements such as alkaline, magnesium, calcium, chloride, sulphate in amounts exceeding their solubility in an equilibrium state. Corrosion is a key hindrance to the successful transportation of hydrocarbons in the oil and gas industry. Its occurrence degrades pipelines which in turn can result in major financial and environmental implications. The corrosion mechanism consists of a series of simultaneous chemical, electrochemical and transport processes [2,3].

The formation of ferrous carbonate scale ($FeCO_3$) is one of the major corrosion results caused by CO_2 carbon dioxide gas. The solid $FeCO_3$ carbonate iron will form on the steel surface if the concentration product of Ferrous ion (Fe^{2+}) and carbonate ion (CO_3^{2-}) concentration exceeds the limit of the solubility product according to the reaction [4]:

$$Fe^{2+}(aq) + CO_3^{2-} (aq) \rightarrow FeCO_3 (s) \qquad (1)$$

The scale will settle when the supersaturation value exceeds saturated, ie when the solution is saturated. This scale can precipitate and undergo strong aging attached to iron [5]. However, the rate of precipitation of ferrous carbonate can be so slow that the settling kinetics is important in determining the degree of protectivity rather than process thermodynamics [6]. The most important factors affecting the deposition of the ferrous carbonate scale are supersaturation and temperature which can increase the rate of reaction [7]. When deposition of iron carbonate on steel surfaces, it is influenced by the presence of species involved in the corrosion process [8-11].

Ferrous carbonate also forms directly on steel or iron surfaces exposed to carbon dioxide solution, forming a "iron carbonate" scale:

$$Fe + CO_2 + H_2O \rightarrow FeCO_3 + H_2 \qquad (2)$$
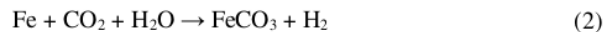
Potential scale caused by scale ferrous carbonate is owned almost all types of heavy equipment such as Heat Exchanger. Ferrous carbonates form solids or very strong deposits attached to the surface of the hydraulic pipe. So far FeCO3 is the cause of the scale on several systems such as heat exchanger [12-13]. The cause of the scale in the heat exchanger will r educe the diameter and inhibit fluid flow in the pipe system, causing problems inhibiting fluid flow. The disruption of fluid flow causes higher pressure, so the pipeline is damaged [14-16].

Zeolites are alumino-silicate minerals with a three-dimensional cage structure, found in Indonesia in large quantities with almost pure shapes and low prices [17-19]. Lately many researchers have focused their attention on zeolite development so that it can be utilized in various fields of science and technology. The unique zeolite crystalline structure causes zeolite to have sorption properties, ion exchangers, "molecular sieving" and catalysts, which can be enhanced by modifying the chemical zeolite to allow zeolites to be used in various industrial processes, in managing industrial waste and in the environmental field [20-23]. Therefore, in this research, the research on the controling of scale ferrous carbonate on the pipe. This study also studied to know the influence of zeolite adsorbance and phase change of crystal morphology and growth of the mass scale of ferrous carbonate.

2. Materials and Method
2.1 Materials and equipment

The powder precursor used for the preparation of the crystal forming solution were $FeCl_2$ and Na_2CO_3 with the analytical grade. The Zeolit adsorbance sintetic grade was added in the solution in an amount of 2000 mg, 3000 mg, and 4000 mg. The $FeCO_3$ forming scale was grown using the pilot-scale equipment shown in Figure 1.

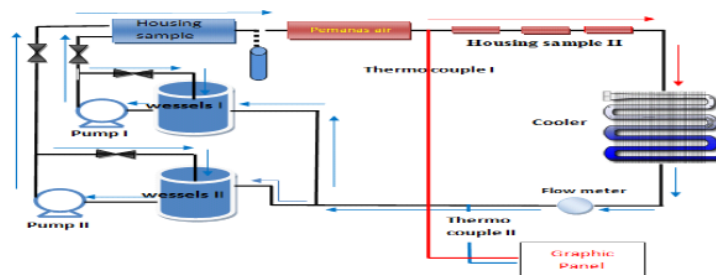**Figure 1:** Experimental setup used in the $FeCO_3$ Precipitation [22].

2.2 Experimental method of the scale formation

For the crystallization experiment, five liters of equimolar $FeCl_2$ and Na_2CO_3 were prepared in the glass beaker. This experiment was performed in the same method which was proposed by Muryanto et al., by dissolving the $FeCl_2$ and Na_2CO_3 powder with the Fe^{2+} concentration of 3000 ppm [24]. The solutions at predetermined concentrations were separately placed in the two vessels and equilibrated until the designated temperature was reached. Subsequently, the amount 2000 mg,

3000 mg, 4000 mg of zeolite adsorbance was dissolved in to a vessel containing five liters of solution $FeCl_2$, then stirred at 200 rpm until it was well mixed. The conductivity of the liquor exiting the test pipe section was continuously checked for up to 60 minutes. The scale deposited on the surface of the coupons was carefully removed and stored in vials for subsequent characterization.

3. Results and DIscussion

3.1 The influence of Zeolite Adsorbance in the Formation of $FeCO_3$ scale.

The experimental result of zeolite adsorbance addition to the mass of scale produced is presented in Figure 2. Testing variable concentrations of zeolite (2000 mg, 3000 mg, and 4000 mg) showed a proportional decrease in the mass scales with increasing Zeolite adsorbance addition.
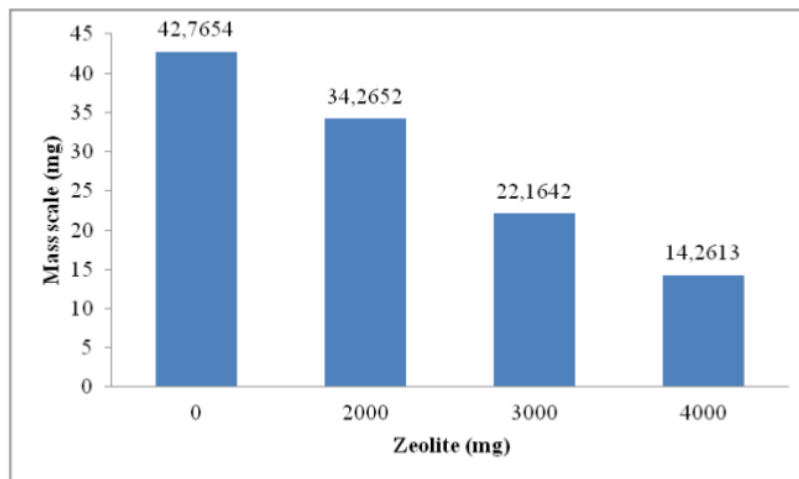

Figure 2. Effect of the Zeolite (mg) on the mass scale $FeCO_3$ produced

The mass scales obtained in the presence of 4000 mg zeolite was 14.2613 mg of $FeCO_3$. Similarly provided 22.1642 mg of mass scale was found in the presence of 3000 mg zeolite. The increasing mass scale 34.2652 mg was also produced at 2000 mg zeolite. Clearly the zeolite adsorbation may be used to improve the scale produced. Zeolites are minerals that consist of hydrated alumino silicate crystals that contain alkaline or alkaline earth cations in the framework of three dimensions so that they can function as adsorbents. Zeolites are able to absorb Fe^{2+} and CO_3^{2-} ions so that the growth of $FeCO_3$ scale becomes inhibited. The ions in the cavity or electrolyte skeleton are useful for maintaining neutral zeolites. These ions can move freely so that the ion exchange that occurs depends on the size and load as well as the type of zeolite [21,22,24].

3.2 Determination of induction time during the precipitation of ferrous carbonate.

The addition of Zeolite adsorbation (2000 mg, 3000 mg, and 4000 mg) resulted in the increase of induction time of crystallization (Fig. 3). The induction time was observed in the $FeCO_3$ system almost doubled in the presence of 2000 mg zeolite (28 min), tripled with 3000 mg zeolite (42 min). Moreover, the presence of 4000 mg zeolite (44 min) led to an induction time four times greater than that for in the absence of zeolit e system (22 min). Figure 3. it can be seen that the greater the Zeolite mass added, the slower the induction time that occurs, the greater the induction time means that the slower the crystal nucleus is formed [24]. Addition of Zeolite can reduce the formation of $FeCO_3$ scale in the spying process and its growth because Zeolites have the ability as adsorbents [25].
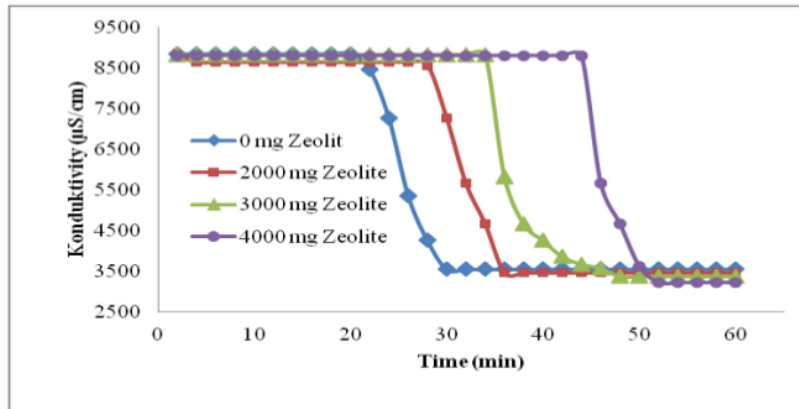
Figure 3. Induction time measured on the $FeCO_3$ precipitation

3.3 Optimation Zeolite Adsorbance and temperature on the mass scale $FeCO_3$

Optimization of the variables used by the statistical method that is by RSM (Response Surface Method), using the software Statistica 6. With this method will get the proper response value, equations mathematical model that fits the data obtained from the experiments and the independent variables optimal conditions. In this study, there is a change of variables, such as zeolite (mg) and temperature (^{0}C) ions with a mass response is the mass scales $FeCO_3$. Limitation of changing variables shown in Table 1 .

Table 1. Experimental Running

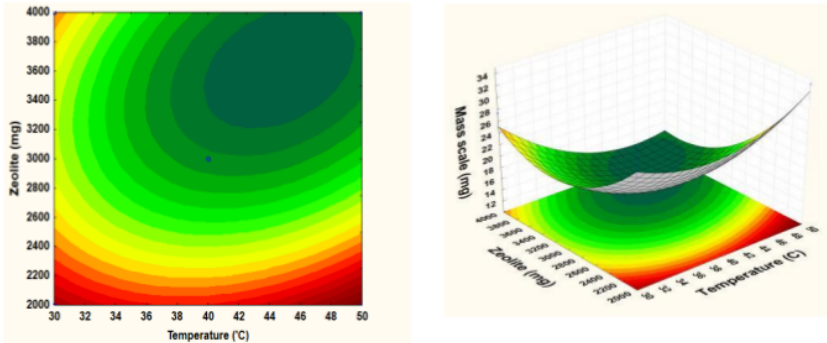| Run	Temperature (^{0}C)	Zeolite (mg)	Mass scale (mg)
1	30.00000	2000.000	33.6268
2	30.00000	4000.000	27.4080
3	50.00000	2000.000	34.1600
4	50.00000	4000.000	14.2600
5	25.85786	3000.000	23.1436
6	54.14214	3000.000	21.3667
7	40.00000	1585.786	32.7383
8	40.00000	4414.214	18.1686
9	40.00000	3000.000	15.3258
10	40.00000	3000.000	15.3258

In accordance with the above experimental results can be made for the optimization , resulting equation as below :

$$\text{Mass scale (Y)} = 123.7099 - 2.6366X_1 + 0.04342X_1^2 - 0.027806X_2 + 0.000005941X_2^2 - 0.00034203X_1X_2$$

Where : X_1= Temperature (^{0}C); X_2 = Zeolite (mg)

Analysis of Variance (ANOVA) is a tool for analysis of the mass of the mass scale as shown in Table 2. The influence of the significance of a factor seen from Table F and p value . p-value is the probability of rejecting the null hypothesis of the study when the null hypothesis was in fact true, the value of p-value less than 0.05 with an accuracy of 95% indicates that variable significant effect. F - value is the ratio between MSF (Mean Squares of Factor) of the MSE (Mean Squares of Error). A

factor is said to have a significant influence when the F -value is greater than F – table.


Figure 4. 2D and 3D Graph Variables Influence of temperature (^{0}C), zeolite (mg) on the mass scale FeCO$_3$.

Table 2. ANOVA (*Analysis of Variance*) optimization

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Source	Sum of Squares	Degree of Freedom	Mean Square	F-value	F-table	R^2
S.S. Regression	595.8307	5	595.8307	57.864058	4.74	0.976
S.S. Error	41.1883	4	10.2971			
S.S. Total	637.0190	9				

The suitability of the model equations for the mass scales response can be tested by multiple criteria. Analysis of Variance (ANOVA) contained in Table 2. to the mass scales response. The coefficient of determination obtained R^2 = 0.976 showed only 2.4% of the total variation does not fit the model of equality, suitability of the model equations with mass scale response tested with static Fisher (F) . The value of the F -value models compared with the F-table, found in Table 3, F -value (57.864058) each effect and for the F - table = 4.74. Based on results for the response the mass scale ANOVA, F -value is greater than F-table. This proves a significant effect on the response variable the mass scales. The same thing can be seen in Chart Pareto, as shown in Figure 5.
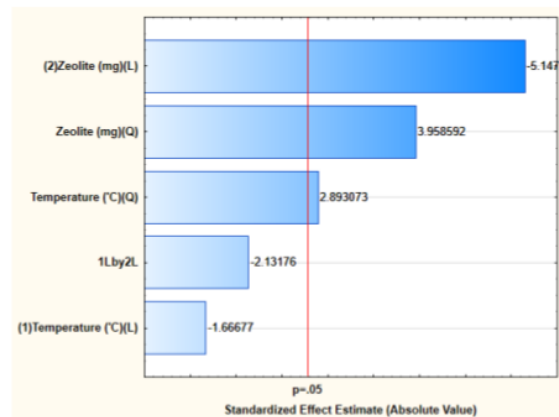
Figure 5. Graph of Pareto optimization variables on the response of the mass scales (mg)

To determine the optimum mass scales, done by entering values into the equation the optimum ratio of the mass response variable optimization scale. Massa optimum scale is shown in Table 3.

Table 3. Optimum mass scale $FeCO_3$

Factor	optimum condition	Mass scale optimum (mg)
Temperature (^{0}C)	44.639	14.46187
Zeolite (mg)	3625.068	

Based on optimum results of temperature,concentration Fe(2+) and zeolite (mg) variables on the mass scale $FeCO_3$. The optimum result of temperature was 44.639^0C, zeolite was 3625.068 mg variables was obtained so that the optimum the mass scale $FeCO_3$ was 14.46187 mg.

4. Conclusions

It was found that in all the experiments, the mass scale $FeCO_3$ decreased with increased mass zeolite adsorbance addition. The mass scales obtained in the presence of 4000 mg zeolite was 14.2613 mg of $FeCO_3$. Similarly provided 22.1642 mg of mass scale was found in the presence of 3000 mg zeolite. The increasing mass scale 34.2652 mg was also produced at 2000 mg zeolite. Clearly the zeolite adsorbation may be used to improve the scale produced. The conductivity decreased abruptly after a certain induction period. The induction time was observed in the $FeCO_3$ system almost doubled in the presence of 2000 mg zeolite (28 min), tripled with 3000 mg zeolite (42 min). Moreover, the presence of 4000 mg zeolite (44 min) led to an induction time four times greater than that for in the absence of zeolite system (22 min). The optimum result of temperature was 44.639^0C, zeolite was 3625.068 mg variables was obtained so that the optimum the mass scale $FeCO_3$ was 14.46187 mg.

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