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Metal Removal from Industrial Waste by Hydrochloric Acid

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Abstract. Leaching of metals from industrial solid waste in hydrochloric acid, HCl solution was studied. In this study, the significant of parameters such as HCl concentration, temperature, liquid to solid ratio and leaching time on the removal percentage of metal impurities were investigated using response surface methodology (RSM) with central composite design (CCD). The ANOVA study concluded that the quadratic model was fitted well to the 27 experimental runs based on p-value (<0.0001), R^2 (0.9876) and Adj-R2 (0.9731). This study also revealed that the liquid to solid ratio was the most significant factor for the leaching of metal impurities compared to HCl concentration for this specific industrial solid waste. However, temperature and leaching time showed no significant impact on the leaching rate of metal impurities. Under suitable conditions, a high removal percentage of metal (88 %) was readily achieved at condition of 6 M HCl at 70 °C with liquid to solid ratio of 10 over a leaching period of 70 mins.

1. Introduction

As a highly developing country, Malaysia produces approximately 3.2 million tonnes of industrial solid waste which consist of hazardous materials and heavy metals. This value shows that the rapid industrialization has contributed to the waste generation in which these wastes are non-biodegradable and tend to accumulate in soil. Moreover, these wastes have a long half-life that increases its probability to essentially form a cumulative toxin. This will lead to soil and water pollutions. Ojo et al. [1] stated that industrial solid waste was a serious issue in soil pollution due to its capability to leach through the soil and contaminate the underground water when it comes into contact with acid rain. The long-term exposure of metal dust will lead to Parkinson's-like disease and neurological effects. Furthermore, long term exposure to metal oxide will cause respiratory problems and reproductive effects [2]. Recycling initiatives have been receiving much attention lately. However, in Malaysia, less than 5 % of solid waste is recycled and only 42 % from the total amount of waste is incinerated or chemically treated before dispose to the environment while the remaining 43 % of waste is just dumped in landfill [3]. This kind to disposal attitude or activities has led to the excretion of leachate and methane which can pollute the waterway under the ground. According to the waste statistic generated by the World Bank in year 2010, Malaysia had produced 42.2 % of methane, 25.0 % of carbon dioxide and 10.4 % of nitrous oxide [2]. This harmful greenhouse gaseous generated from solid waste has taking concern from various Malaysian federal and states government and they started to focus on reducing the amount of greenhouse gaseous generation. Therefore, a recent challenge is to find efficient waste utilization processes that can be used to extract the precious elements or metals from industrial solid waste.

Pyrometallurgical and hydrometallurgical methods are the common and conventional methods that use to remove metal impurities or mineral from industrial waste [4]. Pyrometallurgical method is an



extraction method that involves thermal treatment that results a physical and chemical transformation in minerals which enable to remove the precious metals [5]. Whereas, hydrometallurgical method offers an interesting alternative for the recovery of metals and minerals by using solvent extraction or leaching method [6]. The most widely medium that a use in hydrometallurgical method is HCl due to it is a low-cost leaching agent and has fast dissolution between metal with Cl^- solution. Halli et al. [7] had investigated removal of heavy metal from the electric arc furnace (EAF) steel slag via HCl leaching. According to their research, 75 % of Fe was able to leach from steel slag with 3.0 M HCl at temperature of 90 °C with 1.18 mm particle size [7]. Besides that, Alafara et al. [8] also studied the leaching kinetic of Nigerian ilmenite ore by HCl acid. In their study, 77% of iron-ore was dissolved within 120 min at a stirring speed of 360 rpm. Additionally, Hernández et al [9] was studied the effect of HCL concentration towards the leaching process. Their analysis indicated that, 80 % of Ti and 85 % of Fe are dissolved as the concentration of HCl increased from 1 M to 6 M. On the other hand, an addition of NH_4Cl in HCl leach solution during leaching of laterite nickel ore was done by Li et al. [10]. In their process, it demonstrated a good leaching rate of Ni (88 %), Co (75 %), Mn (96 %) and Fe (21 %) with 2 M NH_4Cl and 3 M HCl at leaching temperature of 90 °C for 90 mins. They also proved that NH_4Cl exhibited a promising ability to activate H^+ , as evident in the simulation result [11]. Ruijing et al. [12] investigated the leaching effects of metals from electroplating sludge by HCl. They indicated that 80.6 % of Cu was leached under 1.5 mol/L HCl, liquid to solid ratio of 10:1 at 40 °C. Besides, a mixed acid solution of HCl and H_3PO_4 also provided good efficient in extracting more valuable metals such as Cu (80.6 %) and Fe (39 %) [12].

Response surface methodology, RSM is the most common and convenient method for design of experiment which can be used to optimize a leaching or extraction process. Sushanta et. al [13] optimized the operating parameters on ash reduction from low-grade coal by using central composite design, CCD. A quadratic model was proposed to correlate the independent variables for maximum ash reduction at optimum condition. The linear effects, square effects and interactive effects of parameters were analyzed by a model plot ANOVA. Their study revealed that HF concentration was the most significant parameters with F-statistic of 933.04 which affect the large extend of ash reduction [13]. Based on their studies, second degree polynomial model was well-fitted to the experimental data at 0.003 p-value and 0.97 R^2 value. Another method from RSM, box Behnken design, BBD method was implemented by Hong et al. [14] with 5 repetition experimental runs to estimate the optimum condition of V extraction from stone-coal. From the ANOVA plot, it displayed that concentration of leach solvent was the most significant parameter with F-value of 84.21 and the model was well-fitted to the experimental data at R^2 of 0.97 [14]. D-optimal design was also one of the RSM method with 4 factors and 3 levels that employed to analyse the response on the leaching of CuS by H_2SO_4 [15]. This design technique had been applied by Soghra et al. [15] to evaluate the effect of particle size, temperature, liquid to solid ratio and redox potential parameters on the efficiency of copper extraction. Their test results indicated that the model was significant to the experimental data at a correlation coefficient, R^2 of 0.96.

In this work, the significant of parameters on the response for instance removal percentage of metal impurities was investigated. The parameter such as HCl concentration, temperature, liquid to solid ratio and time were considered as affecting parameters on the leaching process. RSM based on CCD method is performed to optimize the process parameters. An empirical model is also developed by using the design of experiment to determine the optimum leaching condition and the significance of each parameter towards the response.

2. Material and methods

2.1. Material

An untreated industrial waste was used in this study. A 106 μm of powder slag from industrial waste was prepared through grinding with a mechanical grinder (Vigor mix 700 Swing Type Dry Grinder) and sieved by using GILSON 106 μm stainless steel test sieve. The metal composition in industrial waste

was then determined by wet chemical analysis using HCl acid digestion followed by inductively coupled plasma optical emission spectrometry, ICP-OES (PerkinElmer NexION 2000 B).

2.2. Leaching experiment

In this study, leaching process with HCl acid was performed by using a round-bottomed flask (750 mL nominal capacity) equipped with a rotary evaporator and a water bath (Heidolph Hei-VAP Platinum 2 Rotary Evaporator). The agitation of the leaching content was carried out at a rotation speed of 200 rpm. The experimental leaching set-up was shown in Figure 1. After leaching process, the solid residue and liquid leachate were separated by vacuum filtration. The elemental composition of metal in liquid leachate was analyzed by ICP-OES. While, the solid residue was washed with distilled water to remove excess Cl^- and filtered in glass funnel by using Whatman filter paper (125 mm Φ) and dried in the hot air oven at 60 °C for 24 hr. Finally, the percentage removal of metal was calculated from Equation. (1).

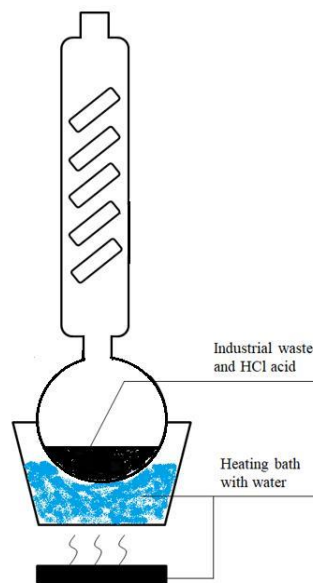


Figure 1. Experimental setup for leaching process

$$\text{Removal (\%)} = [L] / [R] \times 100 \% \quad (1)$$

where $[L]$ is the elemental concentration of metal in liquid leachate in ppm and $[R]$ is the elemental concentration of metal in raw material (industrial waste) also in ppm.

2.3. Experimental design and statistical analysis

The RSM method is a combination of numerical and statistical technique, which has been commonly used for modelling and optimization study. CCD, one of the RSM methods with 3 levels was adopted to study the main interaction of factors on the response in leaching test. In this study, 4 factors: HCl concentration, M (A), temperature, °C (B), liquid to solid ratio (C) and leaching time, mins (D) were concerned in the leaching tests. The minimum and maximum levels of each input variable were selected in accordance with previous studies [8,12,16]. The coded levels and actual values of input factors were listed in Table 1. The design generated 27 experimental runs which were comprised of 16 factorials points, 8 axial points and 3 centered points as shown in Figure 3. Statistical analysis of the experimental data was performed using Design-Expert 6.0.6 software in which response surface plots were also generated. P-values were used for the significance test of parameters and the accuracy of polynomial

model was determined from R^2 coefficient. When the P-value is smaller than the selected confidence level (e.g. 95%, $\alpha = 0.05$), it indicated that the parameter tested is statistically significant [17]. The functional relation which also known as mathematical model can be estimated by second-order polynomial (quadratic function) which generally expressed as Equation (2) [18]:

$$Y = \beta_0 + \sum \beta_i X_i + \sum_{i \neq j} \beta_{ij} X_i X_j + \sum \beta_{ii} X_i^2 + \varepsilon \quad (2)$$

For 4 input variables, the above model can be written as:

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_4 X_4 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{14} X_1 X_4 + \beta_{23} X_2 X_3 + \beta_{24} X_2 X_4 + \beta_{34} X_3 X_4 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{33} X_3^2 + \beta_{44} X_4^2 + \varepsilon \quad (3)$$

where Y is response variable, X_1 , X_2 , X_3 and X_4 were manipulated variables.

While β_0 is a constant, β_1 , β_2 , β_3 and β_4 are linear coefficients. While β_{12} , β_{13} , β_{14} , β_{23} , β_{24} and β_{34} are cross-product coefficients, β_{11} , β_{22} , β_{33} and β_{44} are quadratic coefficients.

Table 1. The coded levels and actual values of input factors.

| Factor | | Levels | | |
|--------|-----------------------------------|-------------------|-------------|-----------|
| | | Low - α | center 0 | High A |
| A | [HCl], M | 2.0 | 4.0 | 6.0 |
| B | Temperature, °C | 30.0 | 50.0 | 70.0 |
| C | Liquid to solid ratio (unit here) | 10.0 | 20.0 | 30.0 |
| D | Leaching time, mins | 60.0 | 180.0 | 300.0 |

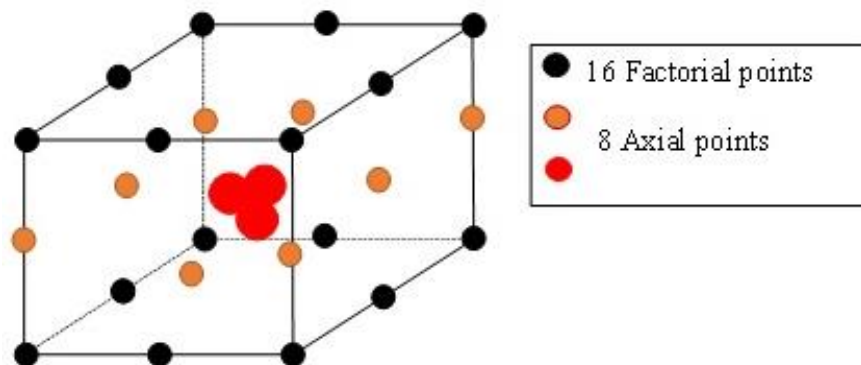


Figure 2. Location of points in the 2^4 factorials design [13]

3. Results and discussion

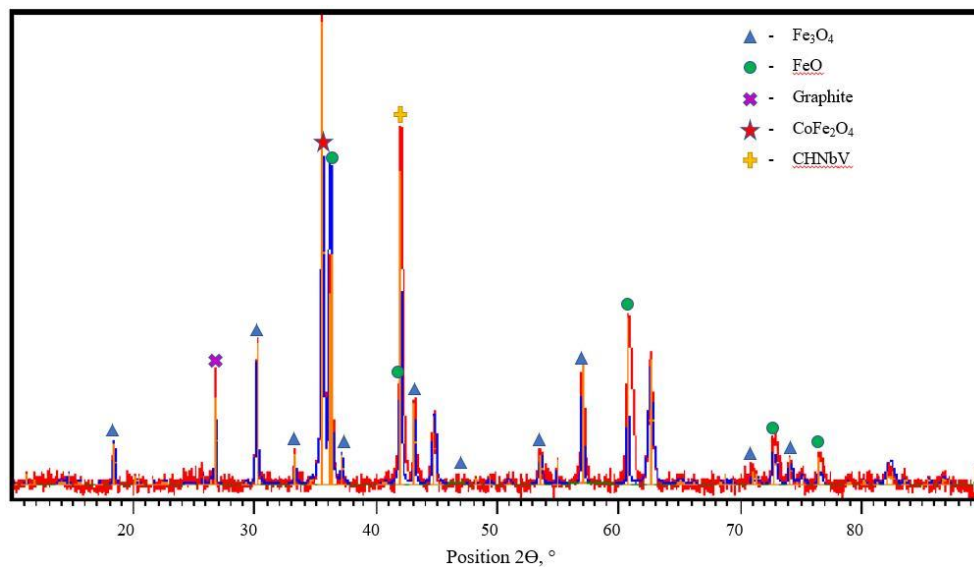
3.1. Leaching mechanism of HCl acid leaching on metal impurities

The element analysis of industrial waste revealed that more than 90 % of Fe was found in the industrial waste as refer to Table 2. The significant quantity of Fe was presented in the oxide form of the compound such as wuestite (FeO) and magnetite (Fe₃O₄) as refer to Figure 3. The results showed more than 90 % of Fe existed in industrial waste as shown in Table 2.

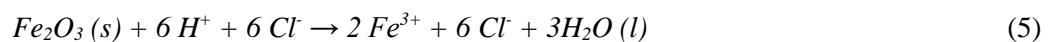
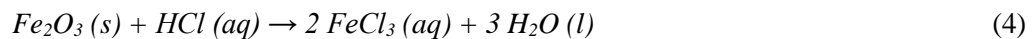
Table 2. Metal composition of industrial waste sample used in the present study.

| Element | Cr | Mn | Ni | B | Fe |
|-------------|------|------|------|------|-------|
| Content (%) | 2.44 | 2.72 | 2.21 | 1.41 | 91.22 |

X-ray diffraction, XRD analysis was used for mineralogy study of industrial waste. The major mineral of the Fe element was reported as magnetite (Fe_3O_4) and wuestite (FeO) as illustrated in Figure 3.

**Figure 3.** XRD pattern of industrial waste

The leaching process mechanism occurred by the HCl acid diffusion, which acid reacted to the metal impurities and dissolves almost the entire Cr, Mn, Ni, B and Fe components. Acid concentration, temperature, liquid to solid ratio and time were the main parameters that influenced the dissolution behavior of metal during the leaching process. Compared to other chemical solvents (i.e. H_2SO_4 , HNO_3), HCl acid was one of the most efficient and well-known reagents that provided a greater potential of elution in industrial applications [19]. This is because HCl provides a high degree of dissolution where high affinity of Cl^- towards the metal in the industrial waste [11]. The major metal (Fe) in waste was reacted with HCl acid during the leaching and formed soluble chloride compounds such as FeCl_3 as shown in Equations. (4), (5) and (6) [20].



3.2. Development of model

The leaching experimental data through RSM was evaluated via ANOVA to determine the significance of model [21]. For this leaching study, the required experimental runs with coded level and actual level are given in Table 3, which showed the design of experiment and experimental result. As the output

proposed from RSM, the quadratic model was aliased. The empirical model in terms of code factor for removal of metal impurities, (Y , %) was shown in Eq. (7):

$$Y = 62.22 + 6.63A + 0.69B - 15.44C + 0.041D - 0.19A^2 + 2.29B^2 - 0.19C^2 - 0.19D^2 - 1.67AB - 0.64AC - 2.24AD - 1.55BC - 1.32BD - 2.35CD \quad (7)$$

Table 3. Experimental factors in coded and actual units and experimental response.

| Exp run | Independent factor in coded form | | | | Independent factor in actual form | | | | Metal removal (%) |
|---------|----------------------------------|----|----|----|-----------------------------------|--------|----|----------|-------------------|
| | A | B | C | D | A (M) | B (°C) | C | D (mins) | |
| 1 | +1 | -1 | +1 | -1 | 6 | 30 | 30 | 60 | 60.60 |
| 2 | +1 | 0 | 0 | 0 | 6 | 50 | 20 | 180 | 70.35 |
| 3 | -1 | -1 | +1 | +1 | 2 | 30 | 30 | 300 | 42.10 |
| 4 | -1 | -1 | -1 | +1 | 2 | 30 | 10 | 300 | 76.10 |
| 5 | +1 | -1 | -1 | -1 | 6 | 30 | 10 | 60 | 85.04 |
| 6 | 0 | 0 | +1 | 0 | 4 | 50 | 30 | 180 | 48.28 |
| 7 | 0 | 0 | 0 | 0 | 4 | 50 | 20 | 180 | 60.30 |
| 8 | +1 | +1 | -1 | +1 | 6 | 70 | 10 | 300 | 85.94 |
| 9 | +1 | +1 | -1 | -1 | 6 | 70 | 10 | 60 | 88.20 |
| 10 | 0 | +1 | 0 | 0 | 4 | 70 | 20 | 180 | 69.10 |
| 11 | -1 | +1 | -1 | -1 | 2 | 70 | 10 | 60 | 72.59 |
| 12 | -1 | -1 | +1 | -1 | 2 | 30 | 30 | 60 | 40.89 |
| 13 | +1 | +1 | +1 | -1 | 6 | 70 | 30 | 60 | 56.19 |
| 14 | -1 | 0 | 0 | 0 | 2 | 50 | 20 | 180 | 56.48 |
| 15 | 0 | -1 | 0 | 0 | 4 | 30 | 20 | 180 | 62.70 |
| 16 | +1 | -1 | +1 | +1 | 6 | 30 | 30 | 300 | 54.24 |
| 17 | 0 | 0 | 0 | +1 | 4 | 50 | 20 | 300 | 63.15 |
| 18 | 0 | 0 | 0 | 0 | 4 | 50 | 20 | 180 | 60.30 |
| 19 | -1 | -1 | -1 | -1 | 2 | 30 | 10 | 60 | 61.40 |
| 20 | 0 | 0 | 0 | 0 | 4 | 50 | 20 | 180 | 57.70 |
| 21 | 0 | 0 | 0 | -1 | 4 | 50 | 20 | 60 | 63.68 |
| 22 | +1 | +1 | +1 | +1 | 6 | 70 | 30 | 300 | 45.90 |
| 23 | +1 | -1 | -1 | +1 | 6 | 30 | 10 | 300 | 86.70 |
| 24 | -1 | +1 | +1 | +1 | 2 | 70 | 30 | 300 | 41.79 |
| 25 | 0 | 0 | -1 | 0 | 4 | 50 | 10 | 180 | 78.55 |
| 26 | -1 | +1 | -1 | +1 | 2 | 70 | 10 | 300 | 77.90 |
| 27 | -1 | +1 | +1 | -1 | 2 | 70 | 30 | 60 | 44.50 |

In order to fit the experimental data to the model, the significance of the regression model with individual model coefficient and lack of fit was tested through ANOVA analysis. These significant factors were normally ranked based on the F-values, p-values, R^2 and Adj- R^2 [21]. In Table 4, the ANOVA for the polynomial model is presented. High significant of the corresponding coefficient was shown as in high F-value and low p-value (Prob. > F) [22]. In the leaching experiment, the F-value of 68.17 and p-value of <0.0001 indicated that the model was significant since both values were at a confidence level of 99.99 %.

In the analysis, the contribution of each factor and their interactions are importance. It helps to understand the role and impact of different process conditions on output response. The individual model terms are significant when the p-values were less than 0.05 [18]. Among the individual terms, acid concentration (A) and liquid to solid ratio (C) were ranked as the highest significant with the p-value of

<0.0001 respectively. Whereas, leaching temperature and time were considered as the non-significant terms with the p-value of 0.2449 and 0.9435 respectively. This analysis indicated that the metal removal rate in leaching test was mainly influenced by acid concentration and liquid to solid ratio. Meanwhile, the interaction terms such as AB, AD, BC, BD and CD were also highly significant with the p-values less than 0.05 and provided a large contribution in the F-value and the total sum of squares.

The fitting of the model is further verified by R^2 and Adj- R^2 . The value of R^2 beyond 0.90 defined the model has good agreement between the experimental and the predicted value (metal removal)[16]. The values of R^2 and Adj- R^2 for this fitted model were 0.98 and 0.97, respectively. The value of R^2 indicated that 98.8 % of the total variation of each factor in leaching test can be explained by these linear, interaction and quadratic effects [20,23]. Therefore, it concluded that the model was well fitted to experiment data and highly significant as shown in Figure 4. Adequate Precision is also the main analysis which used to measure the signal-to-noise ratio [13,18]. An adequate precision ratio greater than 4 indicates the model is highly precise [24,25]. From the analysis, the adequate precision ratio was displayed 27.04 in which indicated an adequate signal. Finally, the diagnostic plot illustrated in Fig. 4 showed that there was no outlier presented in the data and the selected model provided good prediction of the response variables.

Table 4. ANOVA results for response surface quadratic model for removal of metal impurities.

| Source | Sum of square | Degree of freedom | Mean square | F value | Prob. > F (P value) | Remarks |
|--------------------|---------------|-------------------|-------------|-----------|---------------------|-----------------|
| Model | 5400.05 | 14 | 385.72 | 68.17 | < 0.0001 | Significant |
| A | 792.15 | 1 | 792.15 | 139.99 | < 0.0001 | Significant |
| B | 8.16 | 1 | 8.16 | 1.50 | 0.2449 | Not significant |
| C | 4291.39 | 1 | 4291.39 | 758.40 | < 0.0001 | Significant |
| D | 0.030 | 1 | 0.030 | 5.232E-03 | 0.9435 | Not significant |
| A ² | 0.096 | 1 | 0.096 | 0.017 | 0.8986 | Not significant |
| B ² | 13.51 | 1 | 13.51 | 2.39 | 0.1483 | Not significant |
| C ² | 0.096 | 1 | 0.096 | 0.017 | 0.8986 | Not significant |
| D ² | 0.096 | 1 | 0.096 | 0.017 | 0.8986 | Not significant |
| AB | 44.36 | 1 | 44.36 | 7.884 | 0.0161 | Significant |
| AC | 6.55 | 1 | 6.55 | 1.16 | 0.3030 | Not significant |
| AD | 79.92 | 1 | 79.92 | 14.12 | 0.0027 | Significant |
| BC | 38.56 | 1 | 38.56 | 6.82 | 0.0228 | Significant |
| BD | 27.98 | 1 | 27.98 | 4.95 | 0.0461 | Significant |
| CD | 88.17 | 1 | 88.17 | 15.58 | 0.0019 | Significant |
| Residual | 67.90 | 12 | 5.66 | | | |
| Lack of fit | 63.39 | 10 | 6.345 | 2.81 | 0.2906 | Not significant |
| Pure error | 4.51 | 2 | 2.25 | | | |
| Total | 5467.95 | | | | | |
| R^2 | 0.9876 | | | | | |
| Adj- R^2 | 0.9731 | | | | | |
| Adequate precision | 27.039 | | | | | |

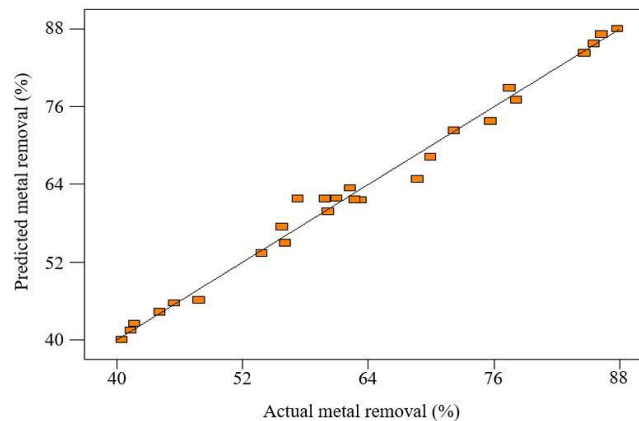


Figure 4. Comparison between the model prediction and experimental data.

3.3. Individual effect of variables

Figure 5 shows the effect of HCl concentration on metal removal in the range of 2 to 6 M for 150 mins. The effect of HCl concentration against metal removal percentage was monitored every 30 mins at the constant temperature of 70 °C and solid to liquid ratio of 1:10. As can be seen, there is a significant increase of metal removal percentage from 30 to 60 mins which the increment of HCl concentration from 2 to 6 M. This prevision can be discussed according to the reaction mechanism in Eqs. (4) and (5) in which Cl^- is generated from HCl acid that acts as a strong activator to dissolve the metals from the industrial waste and it is strongly affecting the dissolution rate of metal [26]. Therefore, it can be stated that high concentration of HCl will increase the amount of Cl^- as activator and react with metal impurities to form soluble compounds, Fe_2Cl_3 . These soluble compounds can be easily removed through filtration.

Figure 6 shows the effect of liquid to solid ratio on metal removal in the range of 1:10 to 1:30 M for 150 mins. The effect of liquid to solid ratio against metal removal percentage was monitored every 30 mins at the constant temperature of 70 °C and 6M HCl. As can be seen, there is an increment of metal removal percentage with the reduction of liquid to solid ratio from 1:30 to 1:10. As the liquid to solid ratio reduces, the metal impurities can be dispersed effectively and reacted with acid thereby enhancing the percentage of metal removal. Additionally, the high concentration of HCl assists in reducing the viscosity of the solution which enhances the availability of Cl^- to react with metal impurities since more surface contact between metal impurities and acid are provided. Similar profile of metal removal percentage was found in the studies conducted by [12,27].

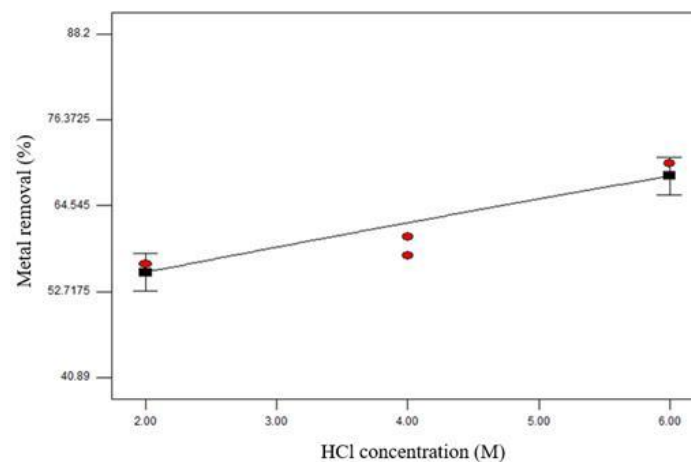


Figure 5. Effect on HCl concentration on metal removal.

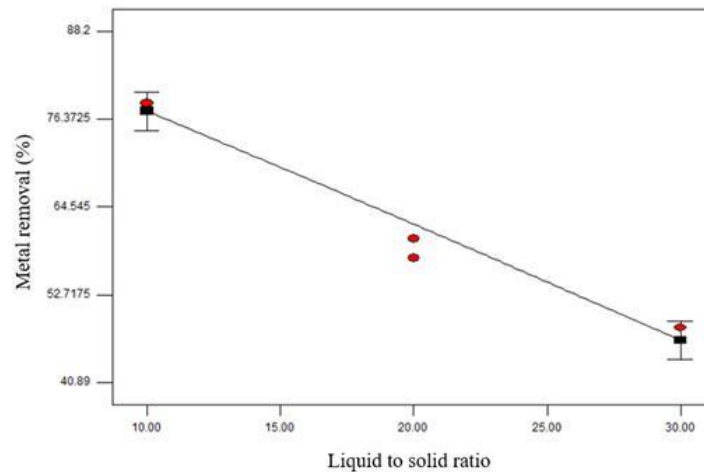


Figure 6. Effect on liquid to solid ratio on metal removal.

3.4. Interaction on parameters

Figure 7 shows the mutual effect of HCl concentration and temperature on metal removal. As can be seen, the metal removal increases with increasing of HCl concentration. In the meanwhile, there is no noticeable change for the metal removal at the entire temperature range (30 °C to 70 °C). At 6 M HCl, higher metal removal has been reported at higher HCl concentrations since more Cl^- ion is generated which enhance the reaction rate.

Figure 8 shows the combined effect of HCl concentration and time on metal removal. As illustrated, leaching time results in increasing the metal removal at 2 M HCl while an inverse trend is observed at 6 M HCl. This is due to less activator (Cl^-) is generated at low acid concentration and more time is required for Cl^- to fully dissolved with metals from industrial waste. However, at high HCl concentration, high amount of Cl^- is formed and this ion can dissolve the metals in short period. As the leaching time goes on, the soluble metal chloride compounds will start precipitate and form solid compound which lead to decrease in the metal removal.

Figure 9 shows the interaction effect between temperature and time on metal removal. As can be seen the leaching behavior illustrated in Fig. 9 is similar to Fig. 8. The metal removal is increasing with time at 30 °C and vice versa at 70 °C. At constant leaching time (60 mins), temperature enhances the rate of reaction since the Fe complexation is decreased at higher temperature at refer to reaction Eq. (4). However, as the leaching time goes on, some bubbles will be found in the solution and start forming a yellow precipitation. These bubbles and precipitations reduce the penetration between acid with metal and hence the leaching rate is reduced.

Figure 10 shows the mutual effect of temperature and liquid to solid ratio metal removal. As can be easily seen, at higher liquid to solid ratio, the metal removal is reduced with increased in amount of solid (industrial waste). Generally, the metal removal increases with reducing the solid densities and viscosity since high amount of leaching acid is added to low content of solid. Besides, no noticeable change is observed between temperature and metal removal with constant liquid to solid ratio.

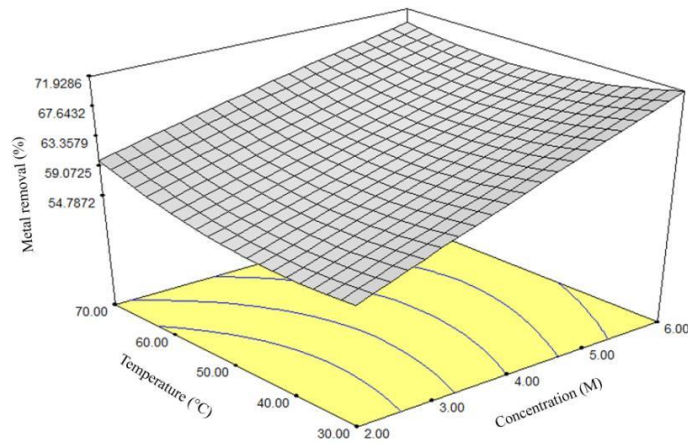


Figure 7. Combined effect of concentration and temperature on metal removal

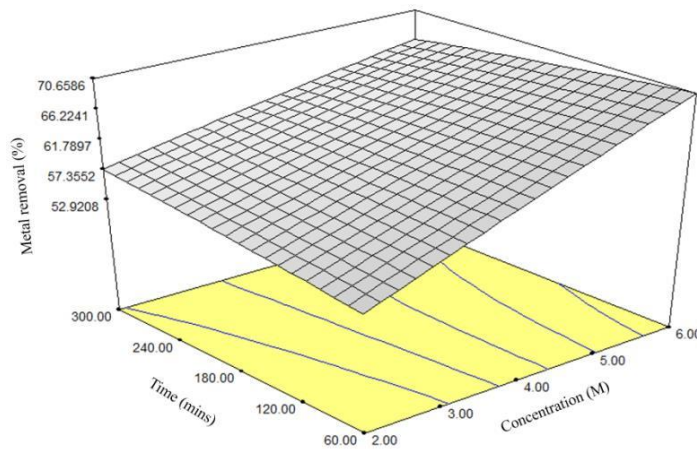


Figure 8. Combined effect of concentration and time on metal removal.

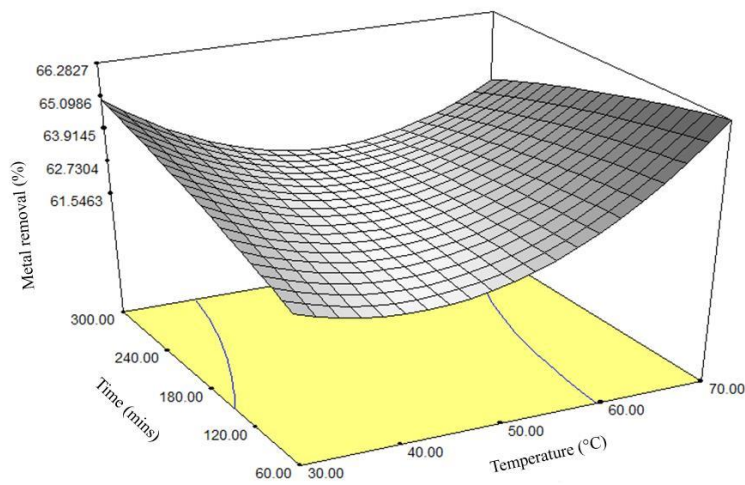


Figure 9. Combined effect of temperature and time on metal removal.

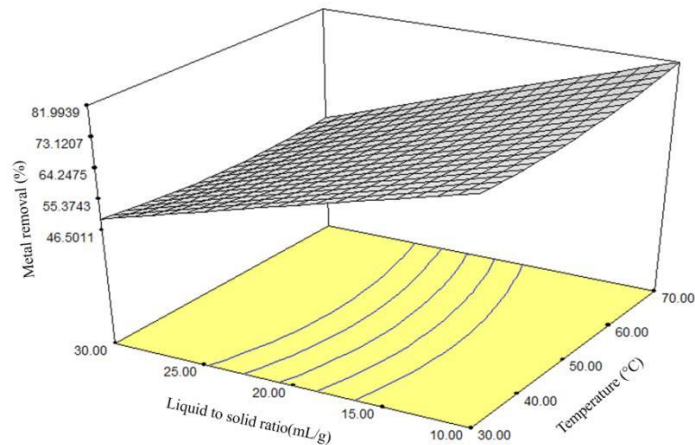


Figure 10. Combined effect of temperature and liquid to solid ratio on metal removal.

3.5. Optimization study

In this study, numerical optimization in CCD design is adopted to determine the optimum leaching condition at maximum of metal removal. In the numerical optimization, the desired goal for each factor and response was chosen. In optimization goals, in range level for each factors and maximum level for response were selected are summarized in Figure 11. The selected optimum operating condition for metal removal from industrial waste via leaching method is at 5.94 M HCl, reaction temperature of 69.47 °C, liquid to solid ratio of 1:10 and leaching time 152.30 mins. This optimum condition was able to obtain 85.4 %. A validation between predicted response and experimental data was calculated by relative standard error, RSE delineated in Eq. (8). The result indicated the error was 2.9 % as shown in Table 6. Thus, it can be concluded that, most of the metal impurities were effectively removed under suggested optimum condition.

$$RSE = \frac{P-E}{P} \times 100 \% \quad (8)$$

Where P and E is the predicted value and experimental value respectively.

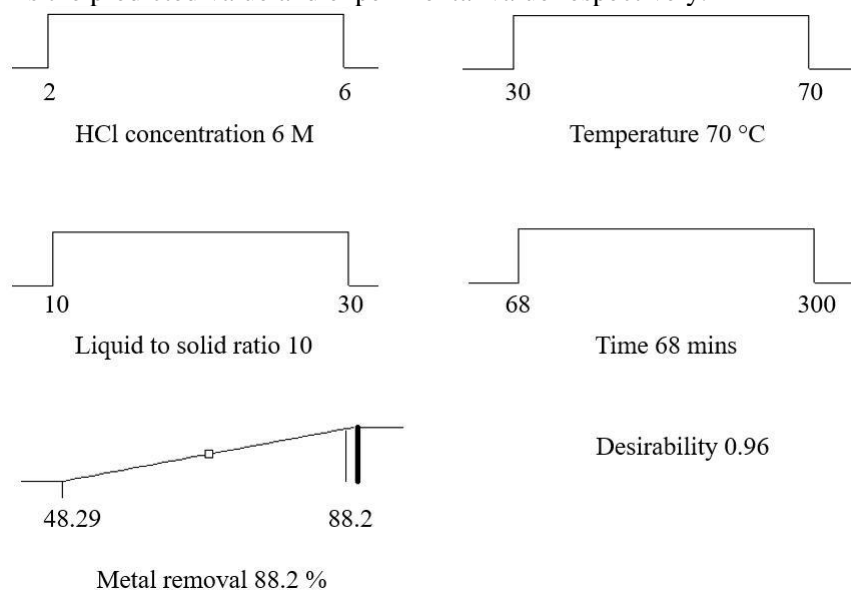


Figure 11. Optimum condition of experiment.

Table 5. Reliability results of optimum conditions.

| Predicted value of metal removal (%) | Actual value of metal removal (%) |
|--------------------------------------|-----------------------------------|
| 88.2 | 85.4 |

4. Conclusion

Conclusively, HCl leaching method is a efficiency method for metal removal from industrial waste. RSM with CCD method was carried out to investigate the significant parameters with their interaction on metal removal percentage from industrial waste. Based on the ANOVA analysis the model were found as statistically significant as well with the R^2 and Adj- R^2 values were observed to be 0.98 and 0.97 respectively. Besides, liquid to solid ratio and HCl concentration were identified to be the most significant parameters for the leaching process with p-value of <0.0001. The metal removal linearly increased with the HCl acid concentration and reduction of liquid to solid ratio. The optimum condition was found to be 6 M HCl, 70.00 °C reaction temperature, liquid to solid ratio of 1:10 and 68 mins of leaching time. The predicted and experimental percentages of metal removal were 88.32 % and 85.0 % respectively with the RSE error of 2.9 %.

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