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Optimization of selective leaching of Zn from electric arc furnace steelmaking dust using response surface methodology

Mehmet KUL, Kürşad Oğuz OSKAY, Mehmet ŞİMŞİR, Halit SÜBÜTAY, Habip KIRGEZEN

Department of Metallurgical and Materials Engineering, Engineering Faculty,
Cumhuriyet University, Sivas 58140, Turkey

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Abstract: The aim of this work is to investigate and optimize the effects of the leaching parameters on the selective leaching of zinc from electric arc furnace steelmaking dust (EAFD). The response surface method was applied on the basis of a three-level Box–Behnken experimental design method for optimization of selective leaching parameters of zinc from EAFD. The leaching recoveries of zinc (Y_{Zn}) and iron (Y_{Fe}) were taken as the response variables, where the concentration of sulphuric acid (X_1, mol/L), leaching temperature (X_2, °C), leaching time (X_3, min), and liquid/solid ratio (X_4, mL/g) were considered as the independent variables (factors). The mathematical model was proposed. Statistical ANOVA analysis and confirmation tests were applied. A maximum of 79.09% of zinc was recovered while the minimum iron recovery was 4.08% under the optimum conditions of leaching time 56.42 min, H_2SO_4 concentration 2.35 mol/L, leaching temperature 25 °C and liquid/solid ratios. By using ANOVA, the most influential factors on leaching of zinc and iron were determined as H_2SO_4 concentration and leaching temperature, respectively. The proposed model equations using response surface methodology show good agreement with the experimental data, with correlation coefficients (R^2) of 0.98 for zinc recovery and 0.97 for iron recovery.
Key words: EAFD; zinc; iron; selective leaching; optimization; Box–Behnken design

1 Introduction

Electric arc furnace dust (EAFD) is one of the most critical wastes encountered in steelmaking industries. During the meltdown of scrap, volatile components are fumed off and are collected with particulate matter in the off-gas cleaning system [1,2]. EAFD contains mainly Zn, Fe, Pb and a considerable amount of harmful elements, such as Cd, As, Cr and F. The contents of the main elements in EAF dusts may vary between: 30% of Zn, 0.3%–6% of Pb, 0.01%–0.2% of Cd, 20%–35% of Fe, 0.2%–0.7% of Cr, 1%–10% of Ca, etc [3–6]. $ZnFe_2O_4$, Fe_3O_4, $MgFe_2O_4$, $FeCr_2O_4$, $Ca_{0.15}Fe_{2.85}O_4$, MgO, Mn_3O_4, SiO_2 and ZnO phases were detected in EAFD [7].

The world generation of EAFD is estimated to be 5−7 million tons per year [7]. Zinc in the EAFD is the most valuable component due to its relatively large amount [8]. Therefore, the selective recovery of zinc from EAFD with a high percentage is an attractive option considering its low production cost.

To date, many processes have been or are being investigated worldwide to recover zinc from the EAFD [1,2,5,9−12]. For this purpose, metallurgical processing can be performed by either pyrometallurgical or hydrometallurgical routes. In pyrometallurgical processes, such as carbothermic reduction, the low-grade zinc in the residue leads to high energy consumption [6], because these processes require high heating of gangue materials. Yet, only 70% of total Zn recovery can be obtained. Given these challenges, a variety of hydrometallurgical processes such as high pressure acid leaching [5], two-stage acid leaching [13], microwave caustic leaching [10], and the use of solutions with various acids [5,9,11,12,14] or highly concentrated alkaline solution have been studied [8,13]. Also, hybridization of pyrometallurgical or hydrometallurgical routes were applied to recovering zinc from EAFD [13]. BARRERA [15] treated electric arc furnace dust (EAFD) for the recovery of zinc following pyro-hydrometallurgical method. Although there are so many studies on the leaching of EAFD, the process optimization by using RSM of the selective sulphuric acid leaching of zinc from EAFD has not been reported in literature. Hence, the present work intends to assess the effects of variables to identify the optimum

Corresponding author: Mehmet KUL; Tel: +90-346-2191010/2803; Fax: +90-346-2191179; E-mail: mkul@cumhuriyet.edu.tr
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conditions using a Box–Behnken design.

To produce electrolytic metallic zinc, an acid leaching step is required in order to lixiviate the highest possible zinc quantity. Sulphuric acid leaching is more suitable than HCl leaching because of the absence of chlorine/chloride and lower lead concentrations. The zinc recovery that can be achieved with this kind of acid liquors lies between 75% and 90% [14,16]. Once the leaching is finished, the liquor obtained is sent to the purifying stages. The first purifying stage is usually oxidation in order to remove the iron as ferric hydroxide sulphate, $Fe(OH)SO_4$. Different oxidation agents may be employed, such as hydrogen peroxide, air, manganese dioxide, or a combination of them. After the oxidation, a cementation step is usually carried out to reduce cadmium, lead and copper concentrations. In this step, zinc dust is usually employed as cementation agent [14,16]. The present study attempts to identify extraction conditions that could possibly maximize the zinc recovery but minimize the iron recovery using sulphuric acid from EAFD by optimizing the process conditions, by designing the experiments using response surface methodology (RSM). Thus, the first purifying stage of pregnant zinc leach solution could be achieved easily and economically. Although, RSM has been a common practice in searching optimal conditions in a variety of research topics, there were no reports, thus far, describing the use of the statistical experimental design approach to improve the selective sulphuric acid leaching of zinc from EAFD.

The general practice for determining the important process parameters for leaching is conducted by varying one parameter and keeping the others at a constant level. This is the one-variable-at-a-time technique. The major disadvantage of this technique is that it does not include interactive effects among the variables and, eventually, it does not depict the complete effects of various parameters on the process. In order to overcome this problem, optimization studies can be carried out using the RSM. The basic theoretical and fundamental aspects of RSM have been described in the related literature [17,18]. RSM is the most popular technique used to find the optimal conditions by using quadratic polynomial model and is applied as a consequence of a screening or diagnostic experiment [18,19]. RSM reduces the number of experimental trials needed to evaluate multiple parameters and their interactions; therefore, it is less laborious and time-consuming than other approaches. So, the experimental and analytical methods using RSM are more advanced than one-variable-one-time method. RSM has been applied to modelling and optimization in leaching processing [19–22].

Although there are so many studies on the leaching of EAFD, the process optimization using RSM for the selective sulphuric acid leaching of zinc from EAFD has not been reported in literature. Hence, the present work intends to assess the effects of variables such as sulphuric acid concentration, leaching time and temperature, and liquid/solid ratio to identify the optimum conditions using a Box–Behnken design. Moreover, the interactions among various factors may not be ignored, hence the chance of approaching a true optimum is very likely. The characteristics of sample are assessed using the analytical instruments such as X-ray diffraction (XRD) and atomic absorption spectrometry (AAS).

2 Experimental

2.1 Materials and apparatus

The chemical composition of EAFD was determined by AAS (GBC Sigma model AAS) and gravimetric & volumetric analysis methods. These results are presented in Table 1. In order to determine the compounds (phases) in EAFD, XRD analysis was performed, and the result is shown in Fig. 1. According to the XRD pattern, ZnO, $(MgO_{0.26}Mn_{0.397}Fe_{0.571}Zn_{0.006})(Mg_{0.449}Ti_{0.002}Mn_{0.0049}Fe_{1.497})O_4$ and $(Zn_{0.06}Fe_{0.04})(Fe_{0.98}\text{-}Zn_{1.02})O_4$ phases were present in the EAFD. The original shape of EAFD sample was agglomerated sphere and the size was 1−3 mm in diameter. But when the sample was added to leaching solution, it would separate and turn to very fine powder particles in the leaching solution. Therefore, it was not ground for fineness.

Table 1 Chemical composition of EAFD (mass fraction, %)

Zn	Fe	Pb	Cd	SiO_2	CaO	Al_2O_3
26.95	27.39	3.75	0.12	3.53	3.49	1.47

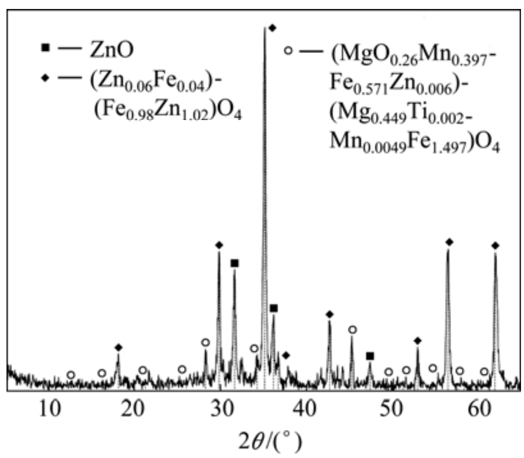

Fig. 1 XRD pattern of EAFD

2.2 Experimental methods

The leaching solution was prepared by mixing analytical grade acid with distilled water. The solution was put into a four-necked 250 mL glass reactor and then it was heated to a bit lower than desired temperature using a digital and thermostatic magnetic stirrer. After that, weighted EAFD sample was added to the leaching solution, followed by raising the temperature of mixing to approximately desired temperature due to exothermic dissolution reactions. The agitation speed was kept constant at 750 r/min in all experiments, so as to keep the contents of the reaction well stirred and suspended. The contents of reactor were filtered, upon completion of the experiment, and the filtrate was analyzed for the zinc and iron contents using AAS. The amounts of zinc and iron leached were estimated using Eq. (1):

$$\eta=(m_1/m_0)\times 100\% \tag{1}$$

where η is the leaching recovery, m_0 and m_1 correspond to zinc or iron contents of sample before and after leaching.

2.3 Experimental design

In this research, a Box–Behnken design, that is widely used form of RSM, was employed for optimization of selective leaching of zinc from EAFD. RSM contains three steps: 1) design and experiments, 2) response surface modeling through regression and 3) optimization. In this work, the main objective of RSM was the maximizing the leaching of zinc but the minimizing the leaching of iron in EAFD waste. The leaching recoveries of zinc (Y_{Zn}) and iron (Y_{Fe}) were taken as the response variable, where the concentration of sulphuric acid (X_1, mol/L), leaching temperature (X_2, °C), leaching time (X_3, min), and liquid/solid ratio (X_4, mL/g) were considered as the independent variables (factors). Pre- experiments were carried out in order to determine the upper and lower limits of the independent variables. According to the results of these previous studies, levels and actual values of independent factors were listed in Table 2. The results of the 27 experimental runs were used to estimate the response variable. The RSM design of experiment was carried out using Design Expert trial software for the analysis and testing parts of this work. For statistical calculations, the relation between the coded values and actual values are described as Eq. (2) [21,22]

$$x_i=\frac{(X_i-X_0)}{\Delta X_i} \tag{2}$$

where x_i is a coded value of the variable, X_i is the actual value of variable, X_0 is the actual value of the X_i at the center point, and ΔX_i is the step change of variable. These actual levels of variables were determined according to preliminary test results as mentioned previously. The mathematical relationship between the four independent variables and the response can be approximated by the second order polynomial:

$$\begin{aligned}Y=&\beta_0+\beta_1x_1+\beta_2x_2+\beta_3x_3+\beta_4x_4+\beta_{11}x_1^2+\beta_{22}x_2^2+\\&\beta_{33}x_3^2+\beta_{44}x_4^2+\beta_{12}x_1x_2+\beta_{13}x_1x_3+\beta_{14}x_1x_4+\\&\beta_{23}x_2x_3+\beta_{24}x_2x_4+\beta_{34}x_3x_4\end{aligned} \tag{3}$$

where Y is the predicted response; β_0 is the model constant; x_1, x_2, x_3 and x_4 are independent variables; β_1, β_2, β_3 and β_4 are linear coefficients; β_{12}, β_{13}, β_{14}, β_{23}, β_{24}, and β_{34} are cross product coefficients and β_{11}, β_{22}, β_{33} and β_{44} are the quadratic coefficients [21–23]. The coefficients, i.e., the main effect (β_i) and two factor interactions (β_{ij}) have been estimated from the experimental results using the Design Expert trial software package.

Table 2 Process factors and design levels used

Factor	Symbol		Coded level		
			Low	Center	High
	Uncoded	Coded	−1	0	+1
Acid concentration/(mol·L^{-1})	X_1	x_1	0.1	1.6	3.1
Temperature/°C	X_2	x_2	25	55	85
Time/min	X_3	x_3	10	50	90
Liquid/solid ratio/(mL·g^{-1})	X_4	x_4	5	17.5	30

3 Results and discussion

Using the Box–Behnken experimental design method, 27 sets of tests with appropriate combinations of acid concentration (x_1), leaching temperature (x_2), leaching time (x_3) and liquid/solid ratio (x_4) were conducted. Box–Behnken design with coded/actual values and results were given in Table 3. Each run was performed in duplicate and thus the values of leaching recoveries of zinc and iron given in Table 3 were the mean of two experiments, while the predicted values of response (leaching recoveries of zinc and iron) were obtained from quadratic model equations using the mathematical software package. Leaching reactions of the main species in the dust sample with sulfuric acid were described by HAVLIK et al [5], the reactions of the main species occurring in the EAFD and their stoichiometry can be stated as follows:

$$ZnO+H_2SO_4\rightarrow Zn^{2+}+SO_4^{2-} \tag{4}$$

$$ZnFe_2O_4+4H_2SO_4\rightarrow Zn^{2+}+SO_4^{2-}+Fe_2(SO_4)_3+4H_2O \tag{5}$$

$$ZnFe_2O_4+4H_2SO_4\rightarrow Zn^{2+}+SO_4^{2-}+Fe_2O_3+H_2O \tag{6}$$

$$CaCO_3+H_2SO_4\rightarrow CaSO_4+CO_2+H_2O \tag{7}$$

$$CaO+H_2SO_4\rightarrow CaSO_4+H_2O \tag{8}$$

$$Fe_2O_3+3H_2SO_4\rightarrow Fe_2(SO_4)_3+3H_2O \tag{9}$$

Table 3 Box–Behnken experimental design and response value

Run No.	Coded level of variables				Actual level of variables				Observed recovery/%		Predicted recovery/%	
	x_1	x_2	x_3	x_4	X_1/(mol·L^{-1})	X_2/°C	X_3/min	X_4/(mL·g^{-1})	Zn	Fe	Zn	Fe
1	−1	−1	0	0	0.1	25	50	17.5	33	0.2	31	2
2	1	−1	0	0	3.1	25	50	17.5	71	14	73	11
3	−1	1	0	0	0.1	85	50	17.5	44	1	41	4
4	1	1	0	0	3.1	85	50	17.5	83	82	84	81
5	0	0	1	−1	1.6	55	90	5	84	29	80	27
6	0	0	1	1	1.6	55	90	30	77	35	77	32
7	0	0	−1	−1	1.6	55	10	5	62	21	61	16
8	0	0	0	0	1.6	55	50	17.5	76	32	78	24
9	−1	0	0	−1	0.1	55	50	5	29	0.6	31	2
10	1	0	1	0	3.1	55	90	17.5	74	54	74	58
11	−1	0	−1	0	0.1	55	10	17.5	19	0.5	22	6
12	1	0	0	1	3.1	55	50	30	83	48	80	52
13	0	−1	0	−1	1.6	25	50	5	69	3	70	3
14	0	1	1	0	1.6	85	90	17.5	79	57	80	55
15	0	−1	−1	0	1.6	25	10	17.5	63	11	60	11
16	0	1	−1	0	1.6	85	10	17.5	73	35	73	34
17	−1	0	0	1	0.1	55	50	30	50	0.4	46	1
18	1	0	−1	0	3.1	55	10	17.5	72	32	70	38
19	−1	0	1	0	0.1	55	90	17.5	32	0.8	37	4
20	1	0	0	−1	3.1	55	50	5	80	32	82	37
21	0	−1	1	0	1.6	25	90	17.5	73	7	72	7
22	0	1	0	−1	1.6	85	50	5	84	35	84	35
23	0	−1	0	1	1.6	25	50	30	79	5	81	7
24	0	1	0	1	1.6	85	50	30	86	45	87	47
25	0	0	−1	1	1.6	55	10	30	73	31	77	26
26	0	0	0	0	1.6	55	50	17.5	78	28	78	24
27	0	0	0	0	1.6	55	50	17.5	79	28	78	24

Reaction (5) occurs slowly at room temperature, but runs at a high rate at elevated temperatures.

3.1 Construction of model equation and adequacy checking

The experimental results in Table 3 were fitted to a full quadratic second order model equation by applying multiple regression analysis for leaching recoveries of zinc and iron using the software mentioned. The model equations representing Y_{Zn} and Y_{Fe} were expressed as a function of acid concentration (x_1), leaching temperature (x_2), leaching time (x_3), liquid/solid ratio (x_4) for coded unit as follows:

$$Y_{Zn}=77.67+21.33x_1+5.08x_2+4.75x_3+3.33x_4-20.58x_1^2+0.29x_2^2-6.71x_3^2+2.67x_4^2-4.50x_1x_4-1.00x_2x_3-2.00x_2x_4-4.50x_3x_4 \quad (10)$$

$$Y_{Fe}=24.33+21.54x_1+17.90x_2+4.36x_3+3.65x_4+1.68x_1x_2+5.43x_1x_3+4.05x_1x_4+6.50x_2x_3+2.00x_2x_4-1.00x_3x_4-0.038x_2^2+2.22x_3^2-1.31x_4^2 \quad (11)$$

The adequacy or accuracy of fit of the regression model for Y_{Zn} and Y_{Fe} (Eqs. (10) and (11)) was analyzed by ANOVA at 5% significance level, and the results are summarized in Tables 4 and 5. The high F and low P (P<0.05) values of the regression model and each variable term (linear, square, and interaction) in the model indicated that they were statistically significant. ANOVA results in Tables 4 and 5 denoted that the quadratic model was significant at 95% confidence level (P<0.05). The same statistical analysis also indicated that the model parameters and their interactions were significant (P<0.05).

Table 4 ANOVA results of regression model for Y_{Zn} (Eq. (10))

Source	SS	df	MSS	F value	Prob>F	Significance	Contribution/%
Model	9374.99	12	781.25	64.94	<0.0001	Significant	
X_1	5461.33	1	5461.33	453.99	<0.0001	Significant	57.226
X_2	310.08	1	310.08	25.78	0.0002	Significant	3.249
X_3	270.75	1	270.75	22.51	0.0003	Significant	2.837
X_4	133.33	1	133.33	11.08	0.0050	Significant	1.397
$X_1.X_4$	81.00	1	81.00	6.73	0.0212	Significant	0.848
$X_2.X_3$	4.00	1	4.00	0.33	0.5733		0.041
$X_2.X_4$	16.00	1	16.00	1.33	0.2681		0.167
$X_3.X_4$	81.00	1	81.00	6.73	0.0212	Significant	0.848
X_1^2	2259.59	1	2259.59	187.83	<0.0001	Significant	23.677
X_2^2	0.45	1	0.45	0.038	0.8488		0.004
X_3^2	240.01	1	240.01	19.95	0.0005	Significant	2.514
X_4^2	37.93	1	37.93	3.15	0.0975		0.397
Residual	168.42	12	12,03				1.445
Lack of fit	163.75	10	13.65	5.85	0.1552	Not significant	1.396
Pure error	4.67	2	2.33				0.048
Cor total	9543.41	26					100
R^2	0.9824						
Adjusted R^2	0.9672						
Predicted R^2	0.9305						
Adequate precision	26.073						

SS: Sum of squares; df: Degree of freedom; MSS: Mean sum of squares

Table 5 ANOVA results of regression model for Y_{Fe} (Eq. (11))

Source	SS	df	MSS	F value	Prob>F	Significance	Contribution/%
Model	11354.91	12	946.24	41.21	<0.0001	Significant	
X_1	5568.52	1	5568.52	242.52	<0.0001	Significant	47.690
X_2	3844.92	1	3844.92	167.45	<0.0001	Significant	32.927
X_3	227.94	1	227.94	9.93	0.0071	Significant	1.952
X_4	159.87	1	159.87	6.96	0.0195	Significant	1.369
$X_1.X_2$	1128.96	1	1128.96	49.17	<0.0001	Significant	9.668
$X_1.X_3$	117.72	1	117.72	5.13	0.0400	Significant	1.008
$X_1.X_4$	65.61	1	65.61	2.86	0.1131		0.561
$X_2.X_3$	169.00	1	169.00	7.36	0.0168	Significant	1.447
$X_2.X_4$	16.00	1	16.00	0.70	0.4179		0.137
$X_3.X_4$	4.00	1	4.00	0.17	0.6827		0.034
X_3^2	31.95	1	31.95	1.39	0.2578		0.273
X_4^2	10.87	1	10.87	0.47	0.5027		0.093
Residual	321.45	14	22.96				2.753
Lack of fit	310.79	12	25.90	4.86	0.1833	Not significant	2.661
Pure error	10.67	2	5.33				0.091
Cor total	11676.37	26					99.997
R^2	0.9725						
Adjusted R^2	0.9489						
Predicted R^2	0.8978						
Adequate precision	23.906						

The low F and high P (P>0.05) values of lack of fit show that the models are adequate for predicting Y_{Zn} and Y_{Fe} within the range of variables studied. The R^2 values of the models obtained are 0.982 and 0.972 (Tables 4 and 5). These also imply that 98.2% and 97.2% (for Y_{Zn} and Y_{Fe}, respectively) of the sample variation are explained by the models. The small deviation between the R^2 and R^2 (adjusted (adj)) values, i.e., 1.52% and 2.36% in Tables 4 and 5, respectively, implies that there is less chance for the inclusion of any insignificant terms in the model and the models are highly significant [18]. The predicted Y_{Zn} and Y_{Fe} confidence levels were compared with the experimental Y_{Zn} and Y_{Fe} (Figs. 2 and 3). The high value of R^2 indicates that the quadratic equations are capable of representing the system under the given experimental domain. These are also evident from the plots of predicted versus observed values for Y_{Zn} and Y_{Fe} in Figs. 2 and 3.

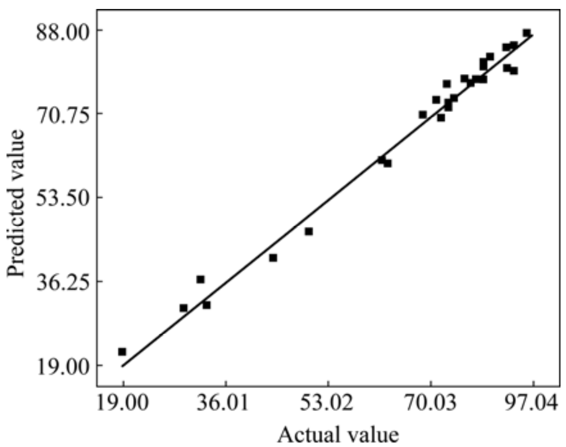

Fig. 2 Relationship between observed and predicted Y_{Zn} values

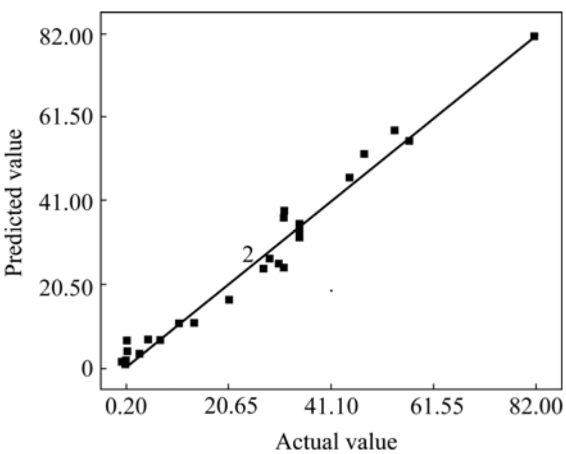

Fig. 3 Relationship between observed and predicted Y_{Fe} values

Results of ANOVA of the regression model for Y_{Zn} and Y_{Fe} (Tables 4 and 5): the model F-values of 64.94 and 41.21 for Y_{Zn} and Y_{Fe}, respectively, imply that the models are significant. There is only a 0.01% chance that a "model F-value" this large could occur due to noise. Values of "Prob>F" less than 0.0500 indicate that the model terms are significant. In this case, acid concentration (x_1), leach temperature (x_2), leaching time (x_3), liquid/solid ratio (x_4), x_1x_4, x_3x_4, x_1 and x_3 are significant model terms for Y_{Zn}, and x_1, x_2, x_3, x_4, x_1x_2, x_1x_3 and x_2x_3 are significant model terms for Y_{Fe}. Values of "Prob>F" greater than 0.1000 indicate that the model terms are not significant. The "Lack of fit F-values" of 5.85 and 4.86 for Y_{Zn} and Y_{Fe} respectively imply the lack of fits is not significant relative to the pure error. There are 15.81% and 18.33% chance for Y_{Zn} and Y_{Fe} respectively that a "lack of fit F-values" these large could occur due to noise. Non-significant lack of fit is good for the models to fit. The "Predicted R^2" of 0.9305 for Y_{Zn} and of 0.8978 for Y_{Fe} are in reasonable agreement with the "Adjusted R^2" of 0.9672 for Y_{Zn} and of 0.9489 for Y_{Fe}, so the models are significant. "Adequate Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. Our ratios of 26.073 for Y_{Zn} and of 23.906 for Y_{Fe} indicate an adequate signal. These models can be used to navigate the design space. All of these results show that the constructed models for zinc and iron leaching recoveries from EAFD are significant.

Contribution of the individual factors and their interactions within the model are important to understand the role and influence of each of them and to control and optimize the selective leaching of zinc from EAFD. With the purpose to determine the factors that have the greatest influence over the system response, ANOVA was used to calculate the contribution of each factor and the results are shown in Tables 4 and 5. Considering that most of the factors are statistically significant at 95% confidence limit, the contribution for each individual factor was calculated by the ratio of adjusted sum of squares of each factor to the total sum of squares. Among all the factors considered on Y_{Zn}, the individual factor acid concentration (X_1) and quadratic factors of X_1 were the most influential within the model, accounting for 57.2% and 23.7% respectively, and also the most ineffective variables on Y_{Zn} were the interaction factors between the independent variables as shown in Table 4. The most effective parameters on Y_{Fe} were acid concentration (X_1) and leaching temperature as seen in Table 5. By studying the main effects and contribution of each factor, the process could be characterized, thus the level of factor to produce the best results could be predicted [24].

3.2 Three-dimensional (3D) response surface plots

In order to gain a better understanding of the interaction effects of variables on Y_{Zn} and Y_{Fe}, three-dimensional (3D) plots for the measured responses were formed based on the model equations (Eqs. (10) and (11)). Also, the relationship between the variables and responses can be further understood by these plots. Since each model had four variables, two variables were held constant at the center level for each plot; therefore, a total of 12 response surface plots could be produced for the responses. Figures 4(a–f) show the 3D response surface plots for the relationship between two variables when the other two variables were held at their center levels for Y_{Zn}. As shown in Fig. 4(a, b, c), sulphuric acid concentration is a dominant factor on zinc recovery. When these graphs are examined, Y_{Zn} increases linearly with increasing acid concentration at low acid concentrations, but the increment decreases slightly after acid concentration reaches approximately 1.6 mol/L. So, Y_{Zn} reaches a pick value at approximately 2.35 mol/L acid concentration; however, it decreases slowly with increasing acid concentration after the pick value. Also, Y_{Zn} increases exponentially with increasing both of L/S ratio and leaching temperature at the same time as shown in Fig. 4(e). Similar result can be attained for Y_{Fe} from Fig. 5(a). In other words, Y_{Fe} increases exponentially with increasing both acid concentration and leaching temperature at the same time. The purpose of study was

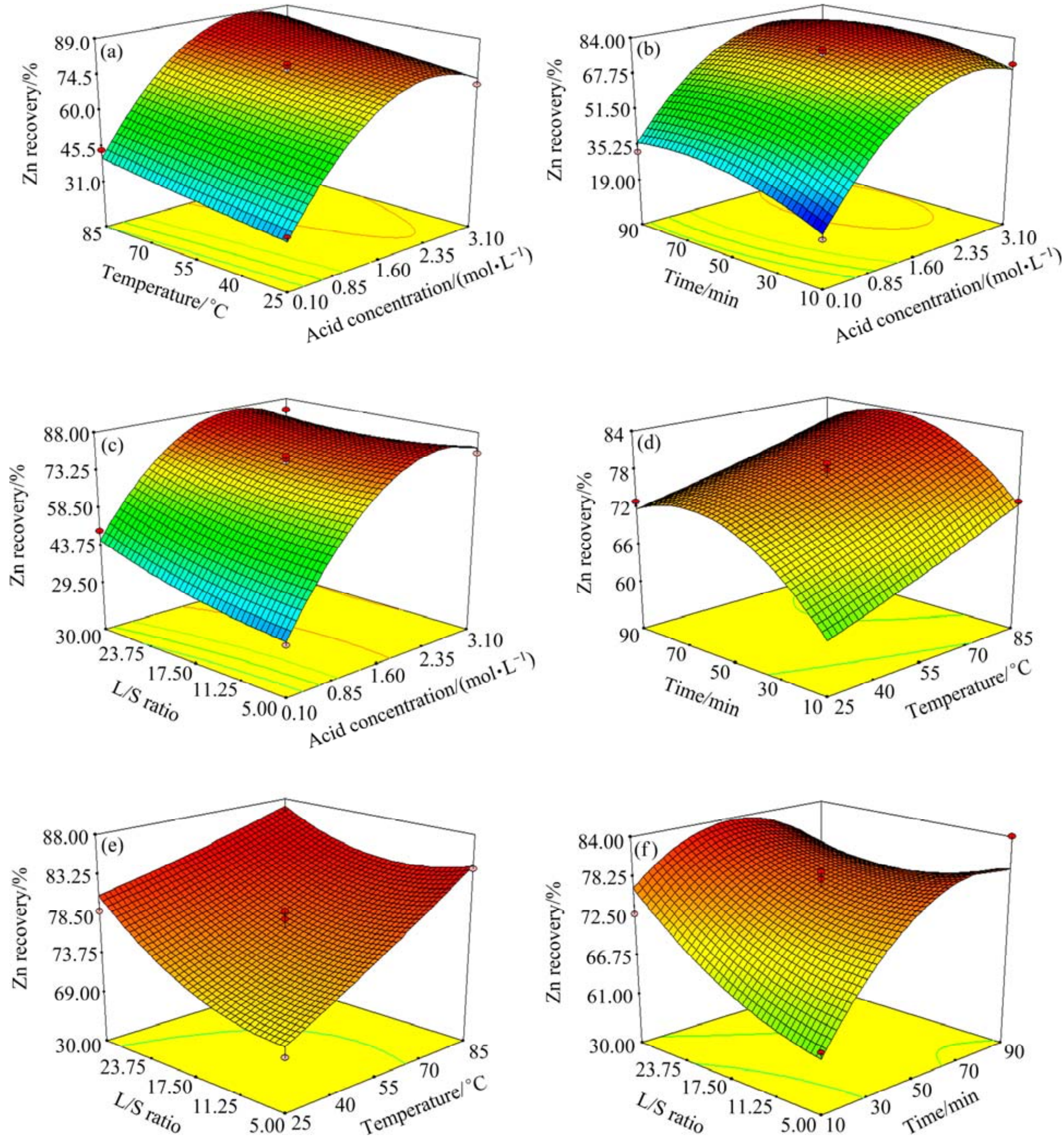

Fig. 4 Response surface plots showing effect of two variables on Zn recovery (Other two variables are held at center level): (a) Temperature and acid concentration; (b) Time and acid concentration; (c) L/S ratio and acid concentration; (d) Time and temperature; (e) L/S ratio and temperature; (f) L/S ratio and time

the identification of optimum sulphuric acid leaching parameters which maximized the Y_{Zn} and minimize the Y_{Fe}. As apparently seen in Figs. 4 and 5 and according to previous explanations, the optimization of leaching variables has to be done in order to achieve the purpose.

3.3 Optimization studies and confirmation tests

Equations ((10) and (11)) were optimized using quadratic programming of the mathematical software package (Design Expert) to maximize Y_{Zn} and minimize Y_{Fe} within the experimental range studied. The optimum

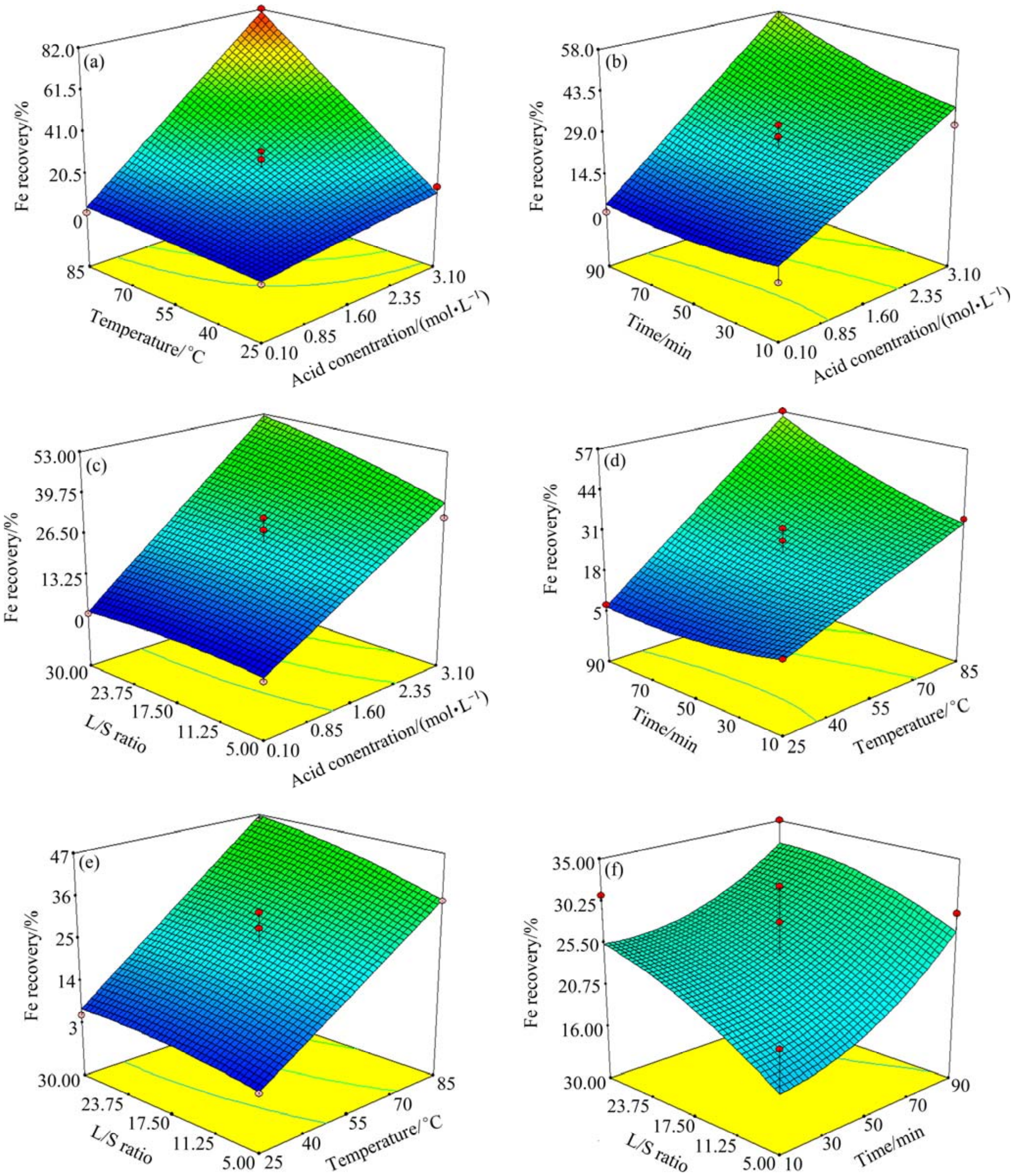

Fig. 5 Response surface plots showing effect of two variables on Fe recovery (Other two variables are held at center level): (a) Temperature and acid concentration; (b) Time and acid concentration; (c) L/S ratio and acid concentration; (d) Time and temperature; (e) L/S ratio and temperature; (f) L/S ratio and time

Table 6 Optimum leaching conditions for maximizing Y_{Zn} and minimizing Y_{Fe}

Test No.	Acid concentration/ ($mol \cdot L^{-1}$)	Temperature/ °C	Time/ min	L/S ratio	Zn recovery/%	Fe recovery/%	Desirability
1	2.35	25	56.42	5	79.09	4.08	0.941123
2	2.33	25	56.41	5	79.03	4.06	0.941092
3	2.27	25	58.01	5	79.05	4.08	0.940998
4	1.02	25	75.41	30	70.22	0.93	0.94091
5	1.01	25	75.51	30	69.90	0.80	0.940908
6	2.34	25.03	55.21	5	78.86	4.03	0.940847
7	2.47	25	52.79	5	78.69	3.98	0.940834
8	2.23	25	54.32	5	78.24	3.87	0.940458
9	1.1	25	70.96	30	72.32	1.80	0.940307
10	2.46	25	49.01	5	77.88	3.78	0.94023

levels of variables were found to be sulphuric acid concentration 2.35 mol/L, leaching temperature 25 °C, leaching duration 56.42 min and L/S ratio 5 for maximizing Y_{Zn} and minimizing Y_{Fe}. The predicted values of Y_{Zn} and Y_{Fe} are 79.09% and 4.08%, respectively, at the determined optimum levels of variables. Once the optimal levels of the control factors were selected, the final step was to verify the improvement of leaching performance using these optimal levels. In order to demonstrate the predictive capacity of optimization study, three more leaching tests were also conducted under these optimum conditions. Mean values obtained from the verification experiment for Y_{Zn} and Y_{Fe} were 76.29% and 3.75%, respectively. The difference between Y_{Zn} and Y_{Fe} was 75.01% which was higher than those obtained in the initial 27 tests in Table 3. The maximum difference between Y_{Zn} and Y_{Fe} was 66.7% according to data in Table 3 which was smaller than 75.01% that obtained as a result of optimization. The mathematical software package suggests such alternative optimum levels of variables for maximizing Y_{Zn} and minimizing Y_{Fe} as seen in Table 6. The lowest value of Y_{Fe} could be achieved by using the fifth suggestion in Table 6, but the desirability

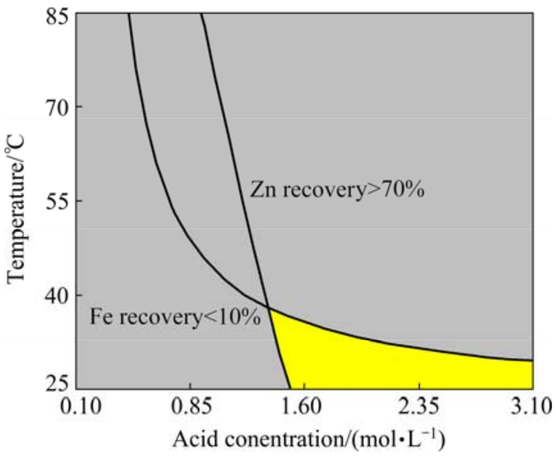

Fig. 6 Overlay plot for optimal region

decreased in this situation. In other words, if this suggestion was realized, Y_{Zn} would be decreased from 79.09% to 69.09%, which was not desirable.

Also, the overlaying contour plot was developed to determine the range of optimal acid concentration and leaching temperature, leading to the best response values when considering all two responses simultaneously. The colored areas on the overlay plots for leaching duration of 56.42 min and L/S ratio of 5, as shown in Fig. 6, are the regions that meet the proposed criteria in which Zn recovery is greater than 70% and iron recovery is lower than 10%.

4 Conclusions

1) The present study was aimed to explore the effects of various leaching parameters on the leaching of zinc and iron from EAFD and to optimize the process conditions using RSM. For this purpose, a three-level Box–Behnken design was employed for modelling and optimizing leaching parameters for selective leaching of zinc from EAFD. Four variables including acid concentration, L/S ratio, leaching temperature and time were investigated in this study. The mathematical model equation was derived for maximizing Y_{Zn} and minimizing Y_{Fe} by using sets of experimental data. Predicted values obtained using the model equations were in very good agreement with the observed values. The model equations were optimized individually using quadratic programming to maximize Y_{Zn} and minimize Y_{Fe} within the experimental range studied.

2) The most influential factors on leaching of zinc were determined as acid concentration and quadratic factors of acid concentration by using ANOVA. In addition, acid concentration and leaching temperature were the most effective parameters on iron leach recovery.

3) The optimum conditions to maximize Y_{Zn} and

minimize Y_{Fe} were determined to be sulphuric acid concentration 2.35 mol/L, leaching temperature 25 °C, leaching duration 56.42 min and 5 L/S ratio with a predicted Y_{Zn} and Y_{Fe} of 79.09% and 4.08%, respectively.

4) This study demonstrates that the response surface methodology (RSM) can be successfully used for the determination of optimum selective leaching parameters of zinc from EAFD.

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响应面法优化电炉炼钢粉尘中 Zn 的选择性浸出

Mehmet KUL, Kürşad Oğuz OSKAY, Mehmet ŞİMŞİR, Halit SÜBÜTAY, Habip KIRGEZEN

Department of Metallurgical and Materials Engineering, Engineering Faculty, Cumhuriyet University, Sivas 58140, Turkey

摘　要：研究浸出参数对电炉炼钢粉尘灰中选择浸出性 Zn 的影响，以 Zn 和 Fe 的浸出率为响应变量，以硫酸浓度、浸出温度、浸出时间和液固比为独立变量，采用基于三水平 Box−Behnken 的响应面法对浸出参数进行优化。对试验结果进行 ANOVA 分析和验证。在硫酸浓度为 2.35 mol/L，浸出温度为 25℃，浸出时间为 56.42 min，液固比为 5 的条件下，可得到 Zn 的最大浸出率为 79.09%, Fe 的最小浸出率为 4.08%。通过 ANOVA 分析表明，对 Zn 和 Fe 浸出率影响最大的因素为硫酸浓度和浸出温度。基于响应面法的模型与试验数据具有很好的一致性，Zn 和 Fe 浸出率的相关系数分别为 0.98 和 0.97。

关键词：电炉炼钢粉尘；锌；铁；选择性浸出；优化；Box-Behnken 设计

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