Optimization of Magnetizing Reduction and Magnetic Separation of Iron Ores by Experimental Design

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An experimental design of magnetizing reduction and magnetic separation of Egyptian low grade iron ore has been conducted using a data developed from applying the conventional single-factor experimentation in order to get the real optimum conditions of the processing. A model has been developed for magnetizing reduction to describe the relationship between the extent of reduction and the reduction temperature, time and reducing gas flow rate, whereas another model for magnetic separation process was deduced correlating the ore size and reduction time to the iron recovery in the magnetic concentrate.

KEY WORDS: experimental design; magnetizing reduction; magnetic separation; iron ores; factorial design; gaseous reduction.

1. Introduction

Investigators conducting experimental programs are most often confronted with the problem of determining the possible effect of a specific number of factors. In such situations, it is necessary to establish an acceptable procedure for choosing the conditions of each experimental run. In the traditional approach to experimentation involving a large number of factors, it is normal to vary one factor while holding all the others constant to determine the effect of that factor. The experiment is then repeated for all other factors, each time holding all others constant except the one being studied. According to this widespread single-factor experimentation, the optimum values of the different factors (calculated individually) are listed as the conditions recommended to achieve the optimum values of the goals of experimentation. On the other hand, experimental design shows the expediency of simultaneously varying all the factors under investigation. The design of an experiment is the procedure of selecting the number of trials and conditions for running them, essential and sufficient for solving the problem that has been set with the required precision. It is proposed that an investigator organizes cosecutive small series of trials, in each of which all the factors are simultaneously varied according to definite rules. The series are so organized that after mathematical processing of a preceding one it will be possible to select the conditions for conducting the following series. Thus sequentially, step by step, is the optimum reached. Some other features are minimizing the total number of trials, simultaneous variation of all the variables determining the process according to special rules called algorithms, the use of mathematical relation formalizing many actions of the experimenter and, finally, the selection of

a clear-cut strategy that helps to make substantiated decisions after each series of experiments.

In the present study, Egyptian iron ores from Bahariya Oasis are subjected to magnetizing reduction followed by magnetic separation in order to get rid of gangue minerals and produce iron ore concentrate suitable for ironmaking operations. The experiments were conducted according to the traditional single-factor experimentation followed by applying the experimental design technique in order to get the real optimum conditions of processing.

2. Previous Work

Magnetizing reduction is one of the most effective unit operations in the treatment of iron ores specially those which are poorly responsive to conventional beneficiation techniques. It involves the conversion of feably magnetic iron minerals in the ore to the strongly magnetic form by subjecting the ore to a reducing atmosphere at elevated temperatures. The resulting artificial magnetite (or γ -hematite) is separated from the non-magnetic gangue by magnetic separation. Although many investigators^{1–5)} studied the reduction of iron ores and oxides, a relatively less attention has been given to the magnetizing reduction process.^{6–8)} The following part outlines the main findings of some of the hematite–magnetite reduction work cited in the literature. El-Abdouni *et al.*⁹⁾ studied the changes in the texture during the reduction of hematite to magnetite at 650°C with CO/CO₂ as reducing gas. They found that during the reduction of α -Fe₂O₃ the structural organization of magnetite Fe₃O₄ changes and leads to a physical structure of the product which is different from the magnetite layer. These observations accounted for the kinetic

behaviour of the reduction rate in which gaseous diffusion coefficient is not constant in the magnetite layer. Adam *et al.*¹⁰⁾ reported on the cracking of hematite crystals during their reduction into magnetite, typically at 350°C with $\text{CO}/\text{CO}_2 = 20/80$ and studied the influence of temperature and of the CO/CO_2 ratio and investigated the magnitude of the stresses. They showed that the fracture mechanism is linked with the first steps of the reaction and investigated the processes which direct the nucleation frequency and the distribution of the magnetite nuclei over the hematite surface. They also found that water vapour has a sticking effects such as quasi-suppression of the cracking and of the induction period, a rather uniform distribution of small nuclei, a decreased rate of reduction for any given reacting fraction even for trace amounts of water ($p_{\text{H}_2\text{O}}$ 1 torr). El-Tabirou *et al.*¹¹⁾ in a contribution for elucidating the discrepancies among authors as regards the transition between porous and lamellar magnetites carried out a wide series of experiments with single crystals over a large range of temperatures (400–1000°C) and CO content (2–50%). They established that lamellar magnetite is favoured by high temperature and low CO content. This is explained by consideration of the conditions governing the competition between cation diffusion in the semi-coherent hematite–magnetite interface and chemical reaction rate. At low temperatures, the crystals are severely fractured, because hematite is not plastic enough, especially at a high CO_2 content. Nasr¹²⁾ studied the effect of temperature and time on the structure of magnetite produced by reduction of hematite with hydrogen at low temperature. He proposed a conceptual mechanism to elucidate both densification and porous structure development as influenced by temperature. He also made a comparative study on the effect of reduction temperature and time on the structure of hematite and magnetite.

On the other hand, very few investigations using experimental design have been published^{13–16)} but none of them applied this experimentation technique on magnetizing reduction and magnetic separation. Magnetizing reduction followed by magnetic separation is a compound technique for beneficiation of iron ores. Optimization of each of the reduction and separation process will have a considerable effect on improving the efficiency of this beneficiation technique. The aim of the present work is to investigate how successful experimental design can be used for optimizing such compound beneficiation process. Also, this study presents a comparative analysis of the results of conventional experimentation and those obtained when experimental design was used for optimization of magnetizing reduction and magnetic separation of an Egyptian iron ore.

3. Experimental

3.1. Chemical Analysis and Mineralogy

A sample of the Egyptian iron ores (Bahariya Oasis) used in the present study has the chemical analysis shown in **Table 1**. X-ray diffraction analysis shows that it is mainly composed of hydrogoethite, α -goethite, α -hematite, hydrated iron oxide and psilomelane as the main manganese mineral as well as some quartz, barite, halites and dolomite. Photomicrographs for the polished sections of the ore sample are shown in **Figs. 1(a)** and **1(b)**. It shows that the sample is constituted of goethite and hematite intimately mixed with different manganese minerals. Goethite generally forms botryoidal, oolitic masses or spindle-shaped crystals sometimes associated with hematite, cryptomelane and psilomelane. Hematite forms tabular, fibrous aggregates in the form of oolites and pisolites cemented with quartz and calcite. Psilomelane exhibits a colloform texture which may contain some cryptomelane and pyrolusite. Sometimes it forms botryoides, the interspaces between them are filled with calcite or quartz. Cryptomelane forms pockets and bands of needle-shaped crystals which may exhibit fan-shaped texture. Its bands intercalate with others of psilomelane, goethite and pyrolusite. Finally, pyrolusite is found as separate outer bands in the manganese botryoids and commonly occurs as stalactic

Fig. 1. Photomicrograph for the iron ore used in the study:
(a) Section showing hematite, calcite and quartz.
(b) Section showing iron minerals and pyrolusite.

Table 1. Chemical analysis of the iron ore sample.

Constituent	Fe	Mn	SiO ₂	Al ₂ O ₃	CaO	MgO	P ₂ O ₅	SO ₃	Loss on ignition	Moisture
Assay (%)	49.24	7.13	2.55	2.66	4.47	0.67	0.055	1.03	8.25	1.42

aggregates or tabular crystals forming spheroids encountered within the other manganese minerals.

3.2. Apparatus

The reduction system, as shown in Fig. 2, consists of three main parts; gas purification and controlling system, the fluidized bed reactor and absorption system for gaseous products. Two gas trains are used; one for hydrogen and the other for nitrogen, meeting at the entrance of the transparent silica reaction tube. The trains include essentially the appropriate gas purification units, flowmeters and flow regulators. The fluidized bed reactor is 70 cm long and 3.5 cm inner diameter silica tube with a sintered silica disc fixed at 40 cm from the bottom to support the solid charge. The reactor is externally heated by a tube furnace which has a constant hot zone extends 5 cm below and above the fluidized bed. The temperature is measured inside the fluidized bed by chromel–alumel thermocouple. The lower part of the silica tube is filled with silica wool to help in preheating the gas. Anhydrous calcium chloride filled U-tubes were used for collecting water vapour produced from the reaction and care was taken to prevent any condensation in the upper part of the reaction tube and the tubes leading to the U-tubes by the use of heating bands. Inside the reactor, just above the sintered disc there is a tube for blowing a high current of cold hydrogen to quench the reduced iron ore and thus stop the progress of reaction.

3.3. Reduction Procedure

Runs were conducted on 50 g ore charged at temperatures ranging between 400 and 500 °C. The particle sizes worked on were $-0.5+0.25$, $-0.25+0.125$ and -0.125 mm and the hydrogen flow rates were 2, 3 and 4 l/min at NTP. After switching on the furnace, nitrogen was allowed to pass through the apparatus until the required temperature in the reactor is obtained and

fixed to within $\pm 2^\circ\text{C}$. The progress of reduction was followed by weighing the absorption U-tubes at the pre-determined time intervals and the cumulative weight loss-time relations can be obtained. At the end of the experiment, the reduction was stopped by passing a high flow rate of cold hydrogen. The cold reduced mass was discharged into acetone to prevent reoxidation by air, then ground to $-0.125+0.057$ mm for magnetic separation.

3.4. Magnetic Separation

Magnetic separation was carried out using Davis-tube tester as a magnetic separator under the following conditions: 230 stroke/min, 1 ampere current, 0.3 l/min water flow rate and 5 min time of agitation. Cleaning was carried out for the non-magnetic fraction using the same conditions but the time for agitation was only 3 min.

4. Results and Discussion

4.1. Magnetizing Reduction

Iron ore samples of the size $-0.5+0.25$ mm were subjected to reduction with H_2 at a flow rate of 3 l/min at 400, 450 and 500 °C for 30 min. The magnetic fractions of the reduced samples were separated and subjected to chemical analysis to determine the iron and manganese contents; the results are given in Figs. 3 and 4. The reproducibility of weight loss and Fe recovery are in the range of 2 %, each run was repeated three times and the results given in the figures represent the mean values. It is clear that at a given reduction temperature iron recovery in the magnetic fraction exhibits a sharp increase at the onset of reduction (1–2 min) and keeps its high value ($>90\%$) for 10–11 min then slowly declines. Figure 3 also indicates that manganese content is greatly affected by the magnetizing reduction treatment where it decreases from 7 % in the ore to 2.6–1.7 % in the magnetic

Fig. 2. Schematic diagram for the reduction system.

Fig. 3. Effect of reduction temperature on Fe and Mn contents in magnetic fraction.

Fig. 5. Effect of ore particle size on Fe and Mn contents in magnetic fraction for samples reduced at 450°C.

Fig. 4. Effect of reduction temperature on Fe recovery and weight loss during magnetizing reduction.

Fig. 6. Effect of ore particle size on Fe recovery and weight loss during magnetizing reduction at 450°C.

fraction. For samples reduced at 400 and 450°C manganese content shows a slight increase after a reduction lapse of 2 min then decreases at longer periods of reduction, whereas at 500°C manganese content starts to decrease directly at the onset of reduction. Bearing in mind that one of the main aims of the ore treatment is to remove as much as possible of manganese from the ore and, at the same time, not to lose iron in the non-magnetic fraction, it is obvious that we are looking for the magnetizing reduction conditions which result in high values for both iron recovery and content as well as low value of manganese content in the magnetic fraction. According to traditional experimentation, Fig. 4 shows that the highest value of iron recovery (93%) is achieved for samples reduced at 400°C for 4 min, at 450°C for 3 min and at 500°C for 2 min. Figure 4 also shows that the highest values of weight loss are achieved for samples reduced at 450–500°C for 25–30 min. It is clear that with the conventional experimentation method it is difficult to determine the process conditions which make weight loss and Fe recovery optimum. On the other hand, the highest value of iron content (64%) is

obtained for samples reduced at 450°C for 20 or 30 min whereas samples reduced at 500°C for 20 or 30 min have the lowest value of manganese content (1.7%). Although the above results are valid only when the ore size and reducing gas flow rate are kept constant at $-0.5 + 0.25$ mm and 3 l/min respectively, they show a more or less wide discrepancy. The temperature of reduction lies in the range 400–500°C while the time varied from 2 to 30 min. Accordingly, no optimum specific values of reduction temperature or duration can be concluded out of the above mentioned results.

In order to investigate the effect of iron ore particle size on the magnetizing reduction treatment, reduction was carried out at 450°C using H_2 at a flow rate of 3 l/min for samples having the following size ranges $-0.5 + 0.25$, $-0.25 + 0.125$ and -0.125 mm. The results of these runs are given in Figs. 5 and 6. It indicates that the highest iron recovery (93%) is obtained for the largest size fraction reduced for 3 min and decreases for lower sizes which produce higher values of weight loss. On the other hand, the lowest manganese content (1.3%) can

Table 2. Phases identified by X-ray diffraction of reduced samples.

Reduction temp., °C	Reduction time, min	Phases
450	2	Fe ₂ O ₃ , Fe ₃ O ₄ , Mn ₂ O ₃ , MnO, SiO ₂
450	5	Fe ₂ O ₃ , Fe ₃ O ₄ , MnO, SiO ₂
450	10	Fe ₂ O ₃ , Fe ₃ O ₄ , MnO, SiO ₂
450	20	Fe ₂ O ₃ , Fe ₃ O ₄ , MnO, SiO ₂
450	30	Fe ₃ O ₄ , MnO, SiO ₂ , Fe
500	2	Fe ₂ O ₃ , Fe ₃ O ₄ , MnO, SiO ₂
500	5	Fe ₂ O ₃ , Fe ₃ O ₄ , MnO, SiO ₂
500	10	Fe ₂ O ₃ , Fe ₃ O ₄ , MnO, SiO ₂
500	20	Fe ₃ O ₄ , MnO, SiO ₂ , Fe
500	30	Fe ₃ O ₄ , MnO, SiO ₂ , FeO, Fe

be observed for the fine ore (−0.125 mm) reduced for 30 min. It is clear that the experimental conditions which result in highest iron recovery is different from those producing lowest manganese content in the magnetic fraction. Again determining the optimum magnetizing reduction conditions is getting more ambiguous, since we can not decide which ore size can be selected and for how long should it be reduced out of the results given in Figs. 5 and 6 for reduction at 450°C using 3 l/min hydrogen. The above results indicate the difficulty of giving the process conditions which make reduction ratio and Fe recovery optimum by means of conventional experimentation method.

The problem is getting more complicated when a range of reducing gas flow rates is also considered. It is conceived that high reduction temperature as well as small iron ore size and, to a certain extent, higher rate of reducing gas flow lead to a larger reduction extent. This is favourable as long as hematite is being reduced to magnetite and that the magnetite–wustite conversion step has not been reached. **Table 2** indicates the phases present in the samples reduced at 450 and 500°C for different periods. It can be easily noticed that at 450°C, some hematite grains are still unreduced even for a reduction time of 20 min whereas at 30 min all hematite has been converted to magnetite. On the other hand, MnO₂ in the ore samples has been converted quickly to the non-magnetic phases Mn₂O₃ and MnO, while quartz is kept unaffected. As reduction temperature increases to 500°C, hematite is totally converted to magnetite in samples reduced for 20 min. In the samples reduced for 30 min, Table 2 shows that the non-magnetic ferrous oxide (wustite) phase has been developed which means that over-reduction of hematite took place with the consequent lowering of iron recovery in the magnetic phase.

The above results show that the magnetizing reduction process has been physically carried out, and we are confronted with the task of optimizing it.

4.2. Experimental Design

Among the many possible ways of finding the optimal conditions, the present study shall consider one that

has been named Box–Wilson method or the method of steepest ascent. This method allows for obtaining statistical mathematical models of the process using factorial design, regression analysis and movement along the gradient. Few assumptions have to be made beforehand, such as, the problem permits one optimization parameter to be selected, a set of determining factors will be selected such that each of the factors is controllable, the results of the trials are reproducible and finally the mathematical model of the process is not known. In the present work, two processes will be optimized, these are magnetizing reduction and magnetic separation. Optimization of magnetizing reduction gives the optimum experimental conditions for converting hematite in the iron ore to magnetite which in turn is to be separated magnetically. Therefore, reduction ratio is a good optimization parameter for measuring the efficiency of conversion in the magnetizing reduction process. On the other hand, when reduced samples are subjected to magnetic separation and Fe recovery is chosen as optimization parameter, both the degree of reduction and the efficiency of magnetic separation technique will affect the process. That is because optimization of magnetic separation process will, in one way or another, be affected by the efficiency of the magnetic separation technique. In case of using highly efficient magnetic separation technique, the optimization of magnetic separation will give conditions more or less similar to the optimized conditions for magnetizing reduction. On the other hand, if there is a wide discrepancy between the optimized conditions for magnetizing reduction and those for magnetic separation, this will mean that the magnetic separation technique is not operating well or not suitable for the process.

4.3. Optimization of Magnetizing Reduction Conditions

The specific objective of magnetizing reduction is to convert the feebly magnetic iron oxide phase hematite to the strongly magnetic one magnetite. Therefore, the weight loss percent (Y), which is a sum of O₂ removed during reduction of iron and manganese oxides together with moisture and loss on ignition with respect to total reducible O₂ in the sample, was selected as the optimization parameter given that the reduction of hematite is confined to magnetite. In this domain, weight loss percent is a good optimization parameter since it effectively reflects the response to the actions of the experimental factors, besides that it is a simple number that can be easily calculated. Four experimental variables (factors) namely, reduction temperature (X_1), reduction time (X_2), gas flow rate (X_3) and ore particle size (X_4) were allowed to vary simultaneously, and the other conditions including quantity of the ore and type of reducing gas were kept constant. The basic levels of these factors were chosen as follows:

- reduction temperature $X_1^\circ = 450^\circ\text{C}$
- time of reduction $X_2^\circ = 20\text{ min}$
- gas flow rate $X_3^\circ = 3\text{ l/min}$
- ore particle size $X_4^\circ = 0.25\text{--}0.125\text{ mm}$

In order to avoid over-reduction of magnetite to wustite, relatively narrow boundaries of the regions of determination of the factors were experimentally considered. Accordingly, the upper and lower levels of the factors are:

$$\begin{array}{ll} X_1 & 400, 500^\circ\text{C} \\ X_2 & 10, 30 \text{ min} \\ X_3 & 2, 4 \text{ l/min} \\ X_4 & 0.5-0.125, -0.125 \text{ mm} \end{array}$$

That is, the variation intervals of the factors are:

$$\begin{array}{ll} \Delta X_1 = 50^\circ\text{C} & \Delta X_2 = 10 \text{ min} \\ \Delta X_3 = 1 \text{ l/min} & \Delta X_4 = 0.125 \text{ mm} \end{array}$$

(where the mean diameters were used as the representative diameters of X_3 and X_4)

It is clear that the selection of the experimental region of factor space was directly connected with the detailed analysis of the experimental results of the already performed runs. In the light of the above outlined design, the number of trials will be equal to the number of levels of the factors, i.e. 2, raised to a power equals to the number of factors, i.e. 4, that comes up with 16 trials. The different trials together with the values of factors and the corresponding optimization parameters are listed in Table 3. As one of the main advantages of experimental design, the interaction effects between two, three and four factors are considered as follows:

– 6 two-factor interactions:

$$\begin{array}{lll} X_5 = X_1 X_2 & X_6 = X_1 X_3 & X_7 = X_1 X_4 \\ X_8 = X_2 X_3 & X_9 = X_2 X_4 & X_{10} = X_3 X_4 \end{array}$$

– 4 three-factor interactions:

$$\begin{array}{ll} X_{11} = X_1 X_2 X_3 & X_{12} = X_1 X_2 X_4 \\ X_{13} = X_1 X_3 X_4 & X_{14} = X_2 X_3 X_4 \end{array}$$

– 1 four-factor interaction:

$$X_{15} = X_1 X_2 X_3 X_4$$

For movement toward the optimum point we need a model that can accurately describe the process, this model is linear if the factors are independent and non-linear when factors interactions are considered. In the latter case, relatively too many coefficients have to be calculated. A design matrix can be developed, as shown in Table 4, in which the number of rows is equal to the number of trials, N , and the number of columns is equal to the number of both the single and multiple factors, k , plus one more column. The extra column (denoted X_0 in Table 4) is augmented to the matrix in order to be used for calculating the independent factor in the model. The total number of columns in the design matrix sums up to $k+1$ which is equal to the number of model coefficients. Each cell in the design matrix contains either

Table 3. Values of experimental factors and the corresponding optimization parameter for magnetizing reduction.

Trial no.	Experimental factors				
	X_1	X_2	X_3	X_4	Y
1	500	10	2	0.125	23
2	500	10	2	0.5-0.125	27
3	500	10	4	0.125	31
4	500	10	4	0.5-0.125	35
5	500	30	2	0.125	45
6	500	30	2	0.5-0.125	49
7	500	30	4	0.125	51
8	500	30	4	0.5-0.125	55
9	400	10	2	0.125	20
10	400	10	2	0.5-0.125	23
11	400	10	4	0.125	23
12	400	10	4	0.5-0.125	27
13	400	30	2	0.125	33
14	400	30	2	0.5-0.125	37
15	400	30	4	0.125	45
16	400	30	4	0.5-0.125	49

Table 4. Design matrix for magnetizing reduction.

Trial no.	X_0	X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8	X_9	X_{10}	X_{11}	X_{12}	X_{13}	X_{14}	X_{15}	Y
1	+	+	-	-	-	-	-	-	+	+	+	+	+	+	-	-	23
2	+	+	-	-	+	-	-	+	+	-	-	+	-	-	+	+	27
3	+	+	-	+	-	-	+	-	-	+	-	-	+	-	+	+	31
4	+	+	-	+	+	-	+	+	-	-	+	-	-	+	-	-	35
5	+	+	+	-	-	+	-	-	-	-	+	-	-	+	+	+	45
6	+	+	+	-	+	+	-	+	-	+	-	-	+	-	-	-	49
7	+	+	+	+	-	+	+	-	+	-	-	+	-	-	-	-	51
8	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	55
9	+	-	-	-	-	+	+	+	+	+	+	-	-	-	-	+	20
10	+	-	-	-	+	+	+	-	+	-	-	-	+	+	+	-	23
11	+	-	-	+	-	+	-	+	-	+	-	+	-	+	+	-	23
12	+	-	-	+	+	+	-	-	-	-	+	+	+	-	-	+	27
13	+	-	+	-	-	-	+	+	-	-	+	+	+	-	+	-	33
14	+	-	+	-	+	-	+	-	-	+	-	+	-	+	-	+	37
15	+	-	+	+	-	-	-	+	+	-	-	-	+	+	-	+	45
16	+	-	+	+	+	-	-	-	+	+	+	-	-	-	+	-	49

+ or - which means the upper or lower level of the factor respectively. The matrix design helps in calculating the mathematical model coefficients according to the following formula:

$$a_{j-1} = \left\{ \sum_{i=1}^N X_{ij} Y_i \right\} / N, \quad j = 1, 2, \dots, k+1 \dots (1)$$

where, a : coefficient value,
 i and j : number of row and column respectively in the design matrix.

The regression model equation can be written as follows:

$$Y = a_0 + a_1 X_1 + a_2 X_2 + \dots + a_k X_k \dots (2)$$

The values of the calculated coefficients are:

$$\begin{array}{llll} a_0 = 35.81 & a_1 = 3.69 & a_2 = 9.69 & a_3 = 3.69 \\ a_4 = 1.94 & a_5 = 0.81 & a_6 = -0.19 & a_7 = 0.06 \\ a_8 = 0.81 & a_9 = 0.06 & a_{10} = 0.06 & a_{11} = -1.31 \\ a_{12} = -0.06 & a_{13} = -0.06 & a_{14} = -0.06 & a_{15} = 0.06 \end{array}$$

4.4. Testing the Significance of Coefficients

The significance of each coefficient is tested separately. The statistical "Student's" t method at 5% significance level is used in the present study. It is first of all necessary to find the variance of the regression coefficient v_c^2 . In our case it is determined by the formula

$$v_c^2 = v_r^2 / N \dots (3)$$

where v_r^2 is the variance of reproducibility with its own number of degrees of freedom. The formula clearly indicates that the variances of all coefficients are equal to one another since they depend only on the error of the experiments and the number of trials. The confidence interval Δb_j is calculated as follows;

$$\Delta b_j = \pm t v_c \dots (4)$$

where t is the tabulated value of "Student's" criterion for the number of degrees of freedom with which v_r^2 was determined and at the selected significance level. In the present magnetizing reduction study, $v_r^2 = 25$ and $N = 16$, accordingly $v_c = 1.25$. From "Student's" t tables at 5% significance level,¹⁷⁾ $t = 2.12$, therefore $\Delta b_j = \pm 2.65$. A coefficient is significant if its absolute value is greater than the confidence interval, accordingly, the coefficients a_4 through a_{15} are insignificant. The model is then reduced to the following formula;

$$Y = 35.81 + 3.69 X_1 + 9.69 X_2 + 3.69 X_3 \dots (5)$$

4.5. Testing Adequacy of the Model

This step is performed for the purpose of checking how fit is the model to the experimental results. In this context the residual sum of squares is quite suitable for characterizing the mean scatter relative to the line of regression. In order to remove the effect of the number of trials, the residual sum of squares is divided by the number of degrees of freedom and termed hereinafter as the variance of adequacy (v_{ad}^2)

$$v_{ad}^2 = \left\{ \sum_{i=1}^N \Delta Y_i^2 \right\} / f \dots (6)$$

where f is the number of degrees of freedom and equals $N - (k + 1)$, where k is the number of variables worked on. A ratio has been developed in statistics that is very convenient for testing a hypothesis on the adequacy of the model. It is called Fisher ratio (or F ratio) and is determined by the following formula;

$$F = v_{ad}^2 / v_r^2 \dots (7)$$

The convenience of using the F ratio consists in that the testing of a hypothesis can be reduced to comparison with a tabulated value.¹⁷⁾ The calculated value of F , i.e. F_{cal} , can be derived from the following formula;

$$v_{ad}^2 = \left\{ \sum_{i=1}^N Y_i^2 - N \sum_{j=0}^k a_j^2 \right\} / f v_r^2 \dots (8)$$

Accordingly, $F_{cal} = 0.4327$. On the other hand, from the table of F ratio values at 1% significance level, $F_{tab} = 3.94$. Since the calculated value of the F ratio does not exceed the tabulated one, then with the corresponding confidence probability the model is considered adequate.

4.6. Interpretation of the Model

Up to now the model, given by Eq. (5), has been derived and the significance of coefficients as well as the adequacy of the model has been tested using an abstract mathematical language. Translation of the model into an experimental language includes the estimation of the magnitude and direction of the influence of separate factors and their interactions. The value of the regression coefficient is a quantitative measure of its influence on the optimization parameter. The higher the value of the coefficient, the greater is the influence of a factor. The nature of the influence of the factors is shown by the signs of the coefficients. Accordingly, the derived model indicates that, in the ranges of factors studied, reduction time has a greater positive influence on the reduction percent than both reduction temperature and gas flow rate whereas, ore particle size has a relatively insignificant effect. It is important to emphasize that the above findings are not general but rather valid only in the range of the predetermined levels of factors. It is also worth mentioning that the absolute values of the regression coefficients grow with an increase in the intervals whereas their signs remain invariant to the change in the intervals. Multiplying the coefficients of the model (Eq. (5)) and the relevant intervals presents the values by which variation in the factors values will keep the movement just along the gradient of the optimum. If the values are so big that they may increase the probability of skipping the region of the optimum, their absolute values can all be multiplied by any appropriate fraction in order to reduce the values of the movement steps and at the same time keep the relevant ratios of the original values. This property can be used in searching for the "optimum" using the steepest ascent method.

The calculation of the multiplication of the regression coefficient and the relevant interval for magnetizing reduction experiments is as follows:

- Temperature, $\Delta T = 184.5^\circ\text{C}$

- Time, $\Delta t = 96.9$ min
- Flow rate, $\Delta F = 3.69$ l/min
- Size, $\Delta S = 0.2425$ mm

It is clear that the above figures are so big as movement steps that adopting them may result in missing the optimum point. Taking a movement step of 20°C for the temperature factor seems to be a quite proper choice. This means that ΔT has been reduced by a factor of $20/184.5 = 0.108$ which can be rounded off to 0.1. By multiplying ΔT , Δt , ΔF and ΔS by 0.1, the values of displacements for the different factors are reduced to:

$$\begin{aligned}\Delta T &= 18.5^\circ\text{C} & \Delta t &= 9.7 \text{ min} \\ \Delta F &= 0.37 \text{ l/min} & \Delta S &= 0.02 \text{ mm}\end{aligned}$$

Accordingly, the recommended values of the main effective factors or rather the optimum conditions are:

temperature 470°C , time 30 min and
gas flow rate 3.5 l/min

Consulting these conditions with the “*priori*” information obtained from the results of the sixteen magnetizing reduction runs indicates that the conditions are convenient, since they are less powerful than those producing over-reduction to wustite and more powerful than the conditions produced magnetite. An experiment performed with the above mentioned optimum conditions showed a weight loss of 54% without over-reduction of hematite to wustite. This is a very good result if we compare it with the calculated theoretical weight loss value of 55.9%.

4.7. Optimization of Magnetic Separation

The reduced samples were subjected to magnetic separation in order to get an iron rich concentrate. Experimental design has been exercised on the experimental results using three factors; reduction temperature, reduction time and ore particle size while recovery of iron in the magnetic fraction was taken as the optimization parameter. The basic levels of the factors were chosen as follows:

- reduction temperature, $Z_1 = 450^\circ\text{C}$
- time of reduction, $Z_2 = 10$ min
- ore particle size, $Z_3 = 0.25\text{--}0.125$ mm

The upper and lower levels of the factors are:

Z_1 400, 500°C Z_2 5, 15 min Z_3 0.5–0.125, –0.125 mm

Accordingly the variation intervals are:

$$\Delta Z_1 = 50^\circ\text{C}, \quad \Delta Z_2 = 5 \text{ min} \quad \text{and} \quad \Delta Z_3 = 0.125 \text{ mm}$$

Table 5 shows the experimental conditions of the eight runs whose data were used for experimental design. The design matrix is given in **Table 6** where the following factors interactions are considered:

$$\begin{aligned}Z_4 &= Z_1 Z_2 & Z_5 &= Z_1 Z_3 \\ Z_6 &= Z_2 Z_3 & Z_7 &= Z_1 Z_2 Z_3\end{aligned}$$

The regression model equation can be written as follows:

$$\psi = c_0 + c_1 Z_1 + c_2 Z_2 + \dots + c_{m-1} Z_{m-1} \dots \dots \dots (9)$$

The coefficients of the model are calculated using the

Table 5. Values of experimental factors and the corresponding optimization parameters for magnetic separation.

Trial no.	Z_1	Z_2	Z_3	ψ
1	400	5	0.125	79
2	400	5	0.5–0.125	92
3	400	15	0.125	77
4	400	15	0.5–0.125	87
5	500	5	0.125	81
6	500	5	0.5–0.125	98
7	500	15	0.125	78
8	500	15	0.5–0.125	80

Table 6. Design matrix for magnetic separation.

Trial no.	Z_0	Z_1	Z_2	Z_3	Z_4	Z_5	Z_6	Z_7	ψ
1	+	–	–	–	+	+	+	–	79
2	+	–	–	+	+	–	–	+	92
3	+	–	+	–	–	+	–	+	77
4	+	–	+	+	–	–	+	–	87
5	+	+	–	–	–	–	+	+	81
6	+	+	–	+	–	+	–	–	98
7	+	+	+	–	+	–	–	–	78
8	+	+	+	+	+	+	+	+	80

formula previously given in Eq. (1) to get the following values:

$$\begin{aligned}c_0 &= 84 & c_1 &= 0.25 & c_2 &= -3.5 & c_3 &= 5.25 \\ c_4 &= -1.75 & c_5 &= -0.5 & c_6 &= -2.25 & c_7 &= -1.5\end{aligned}$$

Testing the significance of the coefficients is performed for a variance of reproducibility $\nu_r^2 = 12$ and $N = 8$ to get a variance of the regression coefficients (according to Eq. (3)) $\nu_c^2 = 1.5$. From “Student’s” t table the corresponding value of t equals 2.306. The confidence interval can be calculated from Eq. (4), then $\Delta b_j = \pm 3.25$. Accordingly, the regression coefficients c_1 , c_4 , c_5 , c_6 and c_7 are insignificant. The model is then reduced to:

$$\psi = 84 - 3.5 Z_2 + 5.25 Z_3 \dots \dots \dots (10)$$

The procedure of testing the adequacy of the model (Eqs. (6)–(8)) has been performed to get the values of F_{cal} and F_{tab} as 1.43 and 6.3 respectively. Therefore, the model is proved to be adequate. The displacements of the factors’ levels to move toward the optimum are:

$$\Delta t = -17.5 \text{ min} \quad \text{and} \quad \Delta S = 0.6563 \text{ mm}$$

In order to get values relevant to the basic level values of the factors, 0.2 of the above values are considered. Accordingly, $\Delta t = -3.5$ min and $\Delta S = 0.13$ mm. The recovery of iron in the magnetic fraction reaches 94%.

A look at Table 5 indicates that run number 6 for samples of a size $-0.5 + 0.125$ mm reduced at 500°C for 5 min resulted in a relatively unprecedented iron recovery of 98%. From the statistical point of view, this abrupt high value of iron recovery is an odd single value compared to the iron recovery values obtained for runs performed at neighbouring conditions. Nevertheless, the model points out that, with relevance to the basic levels of factors ($t = 10$ min, $S = 0.25\text{--}0.125$ mm), decreasing

time and increasing particle size are favourable and lead to better iron recovery values. This is a movement of the experimental conditions towards the optimum which is at the same time a displacement of the factors's values to be closer to conditions of run number 6, a matter which manifests the efficiency of the model.

5. Conclusions

(1) The quality of Egyptian iron ores can be greatly improved by being subjected to magnetizing reduction followed by magnetic separation technique. A great deal of impurities are removed and the iron content is pronouncingly increased.

(2) Most of manganese in the iron ore is directed to the non-magnetic fraction. However, it was noticed that getting a higher removal of manganese is on the expense of a relatively lower iron recovery in the magnetic fraction.

(3) Experimental design is efficiently used in the magnetizing reduction and magnetic separation processes in order to study the simultaneous effect of variables such as ore particle size, reduction temperature, reduction time and gas flow rate on both the reducibility of the ore and the iron recovery in the magnetic fraction.

(4) One more advantage of the experimental design procedure over the traditional experimental technique is getting a model which correlates the different experimental condition values to the optimization parameter. A model has been developed correlating the reduction temperature, time and gas flow rate to the reduction extent whereas another model was deduced correlating

the ore size and reduction time to the iron recovery in the magnetic fraction.

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