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Concentration study of a specularite ore via shaking table, reverse flotation, and microwave-assisted magnetic separation

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ABSTRACT

Despite the difficulties in pelletizing specularite-type refractory iron ores, the utilization of these resources is indispensable for the steel industry due to the increasing need for iron. This study investigated Fe recovery from a refractory iron ore using gravity separation, reverse flotation, and two-stage magnetic separation. Tilt angle and particle size had a significant effect on the grade and recovery of concentrates in shaking table tests. Gravity concentration at optimum conditions resulted in an iron concentrate with 64.47% Fe grade and 90.73% Fe recovery. In the reverse flotation tests, the frother and depressant substantially affected the Fe grade of concentrates while the collector influenced the Fe recovery. A 90% Fe recovery with 64.69% Fe grade was obtained within optimum flotation conditions. The Fe grades were raised to >67.5% in products after the first magnetic separation. The tailings of the first magnetic separation were subjected to the second magnetic separation after microwave-assisted roasting to increase the magnetic susceptibility. In the second magnetic separation, a concentrate containing 66.06% Fe was separated from the microwave-roasted non-magnetic material with 82.23% Fe recovery. To the best of our knowledge, the microwave-roasting method has been applied to a specularite-type refractory iron ore for the first time.

KEYWORDS
Iron ore; shaking table; flotation; magnetic separation; upgrade

1. Introduction

Iron, which is mostly used in casting production, has an essential place in the steel industry. Since it is a relatively cheap material, iron metal is utilized in many areas from the construction sector to the automotive sector. In this respect, iron is the most commonly used metal in industry. The primary source for iron metal is iron ores (Kuskov, Kuskova, and Udovitsky 2017). The main industrial types of iron-bearing minerals are magnetite (Fe_3O_4), hematite (Fe_2O_3), goethite (FeO(OH)), limonite ($Fe_2O_3{\cdot}nH_2O$), and siderite ($FeCO_3$) (Filippov, Severov, and Filippova 2014; Tripathy et al. 2017). Hematite is the leading iron-bearing mineral for the iron and steel sectors (Filippov, Severov, and Filippova 2014). Hematite with a metal luster flake is called specularite. (Wang, Zhang, and Yang 2011). Specularite, also known as oligist, is a brittle mineral in iron black color, with a strong metal luster, and leaves red and brownish red lines on porcelain. Its Mohs hardness is 6.5 and its density is between 5.2 and 5.3 g/cm^3 (Vapur and Top 2016; Zhu et al. 2022). Specularite concentrates are of great interest due to their advantages such as high iron grade and low impurity contents (Zhu et al. 2009; Fan et al. 2012). Poor aggregation and roasting performances due to the crystal structure of specularite concentrates cause them to be considered as an unfavorable material for pellet production. Studies on the pelleting of specularite minerals to overcome these challenges has been extensively reported in the last decade (Zhu et al. 2013, 2018; Zhang et al. 2014, 2018; Pan et al. 2016; Wang et al. 2021). High-grade iron ore reserves continue to decline due to increased steel consumption, and steel industries are forced to use low-grade or lower-quality iron ores to meet their raw material needs (Yuan et al. 2022). In iron ores, gangue minerals typically include quartz and several iron-bearing silicates such as micas, amphiboles, pyroxenes, carbonates, feldspars, clays, apatites, and magnesium compounds (Araujo, Viana, and Peres 2005; Filippov, Severov, and Filippova 2014; Tripathy et al. 2017). The presence of impurities in iron ores causes various problems in the granulation and smelting processes in the blast furnace and limits the exploitation of these ores. Therefore, iron ore is enriched using different separation methods to increase the concentrate iron ratio and to remove these unwanted impurities.

According to the mineralogy and chemical composition of the ore deposit, it is necessary to select the appropriate option for the separation technique (Abdulrahman 2012). The basic beneficiation techniques for specularite mineral includes gravity separation, magnetic separation, and flotation (Wang, Zhang, and Yang 2011). Gravity separation, which depends on the difference in mineral density, is one

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Table 1. Chemical analysis of the specularite samples.

Content	*Fe*	SiO_2	*CaO*	SO_3	La_2O_3	*MnO*	*HgO*	Cr_2O_3	*CuO*	Rb_2O	MoO_3	Y_2O_3	Eu_2O_3	V_2O_5	LOI
%	57.90	8.11	8.46	0.3	0.11	0.02	0.025	0.02	0.04	0.03	0.01	0.003	0.07	0.02	4.76

of the oldest mineral separation techniques. The technique usually requires a small amount of capital for operating costs together with the absence of chemicals and excessive heating requirements. This generally makes this technique environmentally friendly. The most important advantage of the shaking table is its high selectivity with a high upgrading ratio if used correctly (Falconer 2003). The other benefit of enhanced gravity separation is its capacity to reject non-liberated particles more effectively than the flotation method (Singh and Das 2013; Filippov, Severov, and Filippova 2014). On the other hand, the flotation technique, which is widely used for metallic sulfide ores, can be applied for iron mineral separation. This method can be performed by "direct" or "reverse" techniques depending on whether the floated material is concentrated or not (Vidyadhar and Singh 2007). To separate iron minerals from quartz, both cationic and anionic collectors are used (Uwadiale 1992; Mowla, Karimi, and Ostadnezhad 2008; Ma, Marques, and Gontijo 2011; Neymayer, George, and Antonio 2013). Under the same physicochemical conditions, floating capabilities of magnetite and hematite are always lower than that of quartz (Yuhua and Jianwei 2005). Therefore, reverse cationic flotation is mostly used to obtain iron-bearing oxides (De Melo, de Araujo, and Filippov 2017; Haryono et al. 2017). In flotation, an amine or related organic compound is able to producing positively charged hydrocarbon-bearing ions (hence the name cationic collector) for the purpose of floating miscellaneous minerals, including silicate minerals (Gaudin 1957). In the solid-liquid-gas interaction system, the zeta potential values of quartz are negative. In addition, zeta potentials of iron oxide minerals are positive, especially at acidic pH values (Mesquita et al. 2001; Vidyadhar, Kumari, and Bhagat 2014). The convenient flotation reagents are added to the flotation system to change their interfacial properties of the solid-liquid-gas system (Mowla, Karimi, and Ostadnezhad 2008; Srdjan 2007).

Magnetic separators (Wang, Zhang, and Yang 2011; Tanriverdi, Sen, and Cicek 2018), spirals (Abedi et al. 2022), shaking tables (Vapur and Top 2016), and flotation cells (Li et al. 2020; Zhang et al. 2022) have been reported on specularite enrichment in the literature. Considering the particle sizes of specularite mineral, the enrichment methods can be listed as follows: gravity separation > magnetic separation ≥ flotation.

The magnetic susceptibility of minerals can be increased by microwave applications (Omran et al. 2014). By applying microwaves, not only the minerals exposed to microwaves are converted into ferromagnetic minerals, but also the total iron content of the minerals increases. The most important advantages of microwave irradiation over conventional roasting are rapid heating, non-contact heating, selective and volumetric heating (Wu et al. 2017). The recent investigations showed that microwave irradiation had a significant advantage in the processing of iron minerals such as hematite (Zhou et al. 2021), limonite (Tosun 2020), siderite (Znamenáčková et al. 2005), and goethite (Nunna et al. 2021).

With increasing consumption, iron ore reserves continue to deplete, and related industries are forced to use lower quality or previously undesirable iron ores to meet their raw material needs. Although there are studies on the beneficiation of low-grade iron ores in the literature, research on the enrichment of specularite type iron ores, especially by microwave-assisted magnetic separation, are limited. The aim of this research is to investigate the production of iron concentrate with high commercial grades (>60% Fe) from a specularite type iron ore by applying shaking table, reverse cationic flotation, and microwave-assisted magnetic separation methods. Besides that, the determination of effective separation parameters for high Fe grades and recoveries is also focused on. In this context, table tilt angle and particle size were selected as the main variable parameters in the shaking table tests, while the corn starch depressant dosage, Aero 3030 C collector dosage, X-133 frother dosage and froth collection time were selected as the main variable parameters in the reverse flotation tests. Magnetic field strength was chosen as the main variable parameter in the magnetic separation tests. Non-magnetic materials obtained as a result of the first magnetic separation were subjected to the second magnetic separation after being roasted with microwaves, and the focus was also to observe the change in their magnetic properties.

2. Experimental

2.1. Materials

The specularite sample taken from a mine deposit in Turkey was used in the experiments. The chemical composition of the sample was determined by X-ray fluorescence analysis using a Minipal-4 Panalytical device (Table 1). The sample had a significant amount of iron (57.90% Fe). The mineral phases of the sample were analyzed by using a Rigaku Minflex XRD (X-ray diffractometer) device. Phase analyses executed by PDXL and HighScore Plus software equipped with a PDF-2 database showed that the mineral phases were iron oxides (specularite (Fe_2O_3), goethite (FeO(OH)), hematite (Fe_2O_3), iron oxide (FeO)) along with quartz (SiO_2) and andradite ($Ca_3Fe_2Si_3O_{12}$)(Figure 1).

According to XRF analysis of the sample, there are quite small amount of impurities that have negative effects on sintering, blast furnace, and steelmaking. Microscopic images of the sample under the reflected light microscope (Nikon-YS100) are shown in Figure 2 ((a) −38 µm, (b) −74 µm, (c) −150 + 74 µm, (d) −500 + 150 µm, (e) −1000 + 500 µm and (f) −2000 + 1000 µm). The red, black and dark grey colored particles in the microscopic images are iron minerals such as specularite, hematite and goethite. The white colored particles are quartz. The approximate loss on ignition (LOI) values of the samples kept at 1000 °C for 1 h was 4.76%.

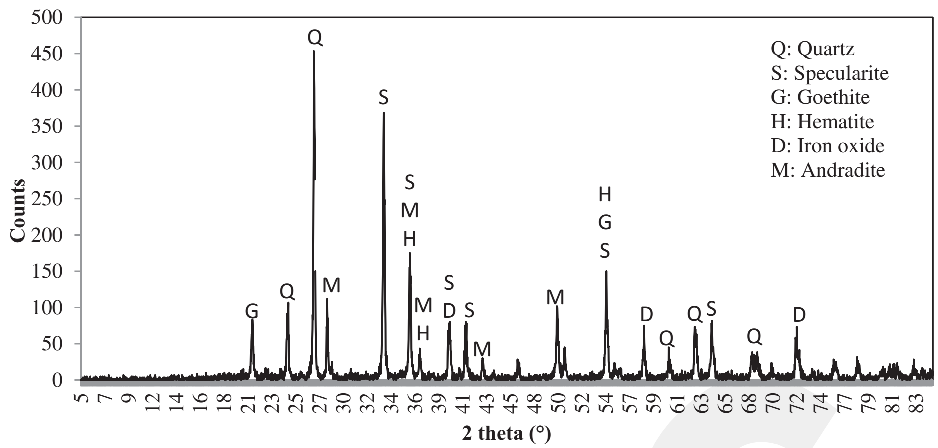

Figure 1. XRD pattern of the sample.

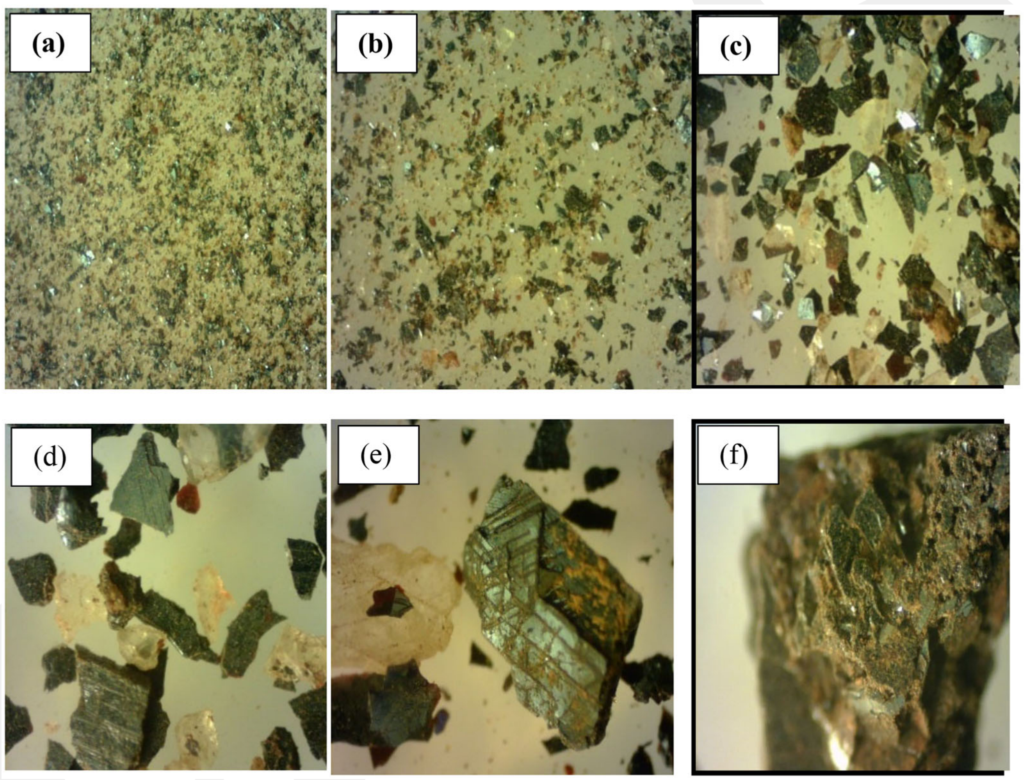

Figure 2. Microscope images of the different-sized samples.

This value shows that the material is refractory iron ore and is in line with the LOI values of refractory iron ores in the literature (Liang et al. 2019; Tang et al. 2022).

Firstly, the sample was crushed to under 10 mm in size by a laboratory-type jaw crusher. The crushed ore was ground for 12 min using a laboratory-type ball mill (the charge volume was 45% of the internal volume of the mill with 12 kg balls of various diameters) at 60 rpm drum speed under dry conditions. The samples were classified according to their particle sizes as $-4000 + 2000\,\mu m$ $-2000 + 1000\,\mu m$, $-1000 + 500\,\mu m$, $-500 + 150\,\mu m$, $-150 + 74\,\mu m$, $-74 + 38\,\mu m$, $-38\,\mu m$.

In reverse flotation tests, $-150 + 38\,\mu m$ sized fraction was used. However, before flotation, desliming was applied and the materials minus $-38\,\mu m$ particle size were rejected. The particle size distribution of the ground specularite is shown in Figure 3.

Tap water was used as the separation medium in all the separation tests. Analytical grade sulfuric acid (Merck) was used for pH adjustment in the flotation tests. Food grade corn starch was used as a depressant, commercial Aero 3030C was used as collector and OrePrep X-133 was used as frother.

2.2. Methods

All experiments were repeated at least twice to ensure verifiability. After each separation test, both concentrates and tailings were filtered, dried, weighed and sent for analysis. Fe

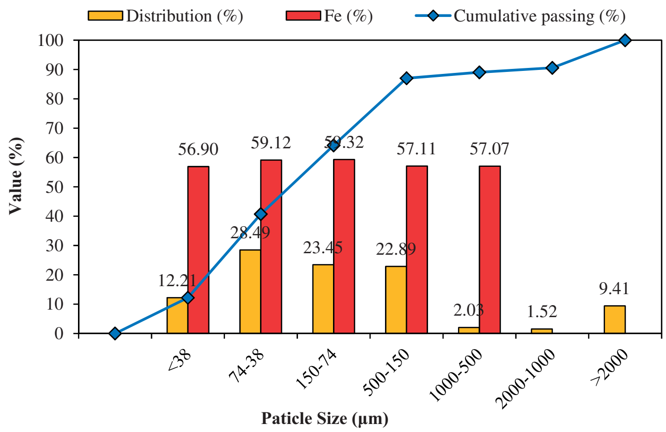

Figure 3. The particle size distribution and Fe grades of the ground sample.

recovery was calculated by using the following equation:

$$\text{Recovery }(\%) = R = \frac{C\ c}{Ff} \times 100 \tag{1}$$

Where R is the recovery of the iron ore concentrate; F is the weight of the feed solid (g), C is the weight of the concentrate (g), f is the Fe grade of the feed (%), and c is the Fe grade of the concentrate (%) (Wills and Finch 2015).

2.2.1. Shaking table experiments

Shaking table experiments were carried out using a laboratory Wilfley type shaking table (Dimensions: 50 × 100 × 35 cm (W in × L × W out)). The two fractions (−74+0µm and −150 + 74 µm) were used for the shaking table tests. The table tilt angles were applied as 3°, 4°, 5°, and 7°. All the tests were performed at 12 L/min water flow rate with 175 strokes/min shaking speed. The concentrate (heavy minerals) and tailing (lightweight minerals) were collected from the edge of the table, separately.

2.2.2. Reverse cationic flotation experiments

The reverse cationic flotation tests were carried out by a batch Denver type flotation machine with 1 L cell volume. The impeller speed was 1200 rpm. The solid/liquid ratio was 10% by weight and the pH value, which was adjusted by H_2SO_4 solution, was 3. This pH value was determined in light of preliminary experiments. After the pH was adjusted, chemicals were added. The particle size of the sample used in the flotation was −150 + 38 µm. Corn starch was used as a depressant. After conditioning of the pulp for 5 min, the starch was added to the flotation cell. The range of corn starch dosages were between 400 and 600 g/t. The cationic collector namely Aero 3030 C (2-Ethylhexanol/cationic surfactant in liquid form) was used. The collector was added directly to the flotation cell after depressant addition. The collector dosages were between 200 g/t and 400 g/t. The frother was X-133 (OrePrep X-133 Frother) and was added directly to the slurry 1 min before froth collection time. In preliminary experiments for the same conditions, X-133 frother showed better results than MIBC (methyl isobutyl carbinol) frother. Therefore, it was decided to use this frother. The addition ranges of depressant, collector, and frother dosages were selected based on the preliminary test results. The froth was collected over a period of 2 min.

2.2.3. Magnetic separation experiments

Two-stage magnetic separation tests were applied. The first magnetic separation tests were performed by laboratory type high intensity wet magnetic separator (Eriez L4-20) at 7 different magnetic field intensities (0.25 T, 0.35 T, 0.45 T, 0.55 T, 0.65 T, 0.75 T, and 0.85 T) using an optimum particle sized sample (−150 + 38 µm). The particle size was selected based on the preliminary tests. The solid ratio was arranged to 20% by weight. The non-magnetic product, which was obtained from the first magnetic separation test at 0.25 T magnetic field intensity, was classified as tailings. The tailings were subjected to the microwave roasting process to increase the magnetic susceptibility of them. In the second magnetic separation tests, microwave-exposed tailings were magnetically separated under the same conditions as in the first magnetic separation.

2.2.4. Microwave roasting tests applied to non-magnetic product

In microwave roasting process, the non-magnetic product was treated in a kitchen type microwave oven (LG WAVEDOM, 2.45 GHz). To minimize the impact of the field pattern variation in the oven, the quartz pan was placed in the same central position and microwave roasting started. After roasting up, the samples were then allowed to cool to room temperature in the microwave oven. The effect of microwave exposure on magnetic separation was evaluated by The Box Behnken test design, statistically. The three input variables were microwave power (0.54–0.90 kW), and exposure time (1–3 min), and magnetic field intensity (0.25–0.85 T). The range of the microwave power and exposure time was selected based on preliminary experiments. In each test, 40 g of the non-magnetic sample was put in a quartz pan. It was approximately 10 mm in diameter and 40 mm in height.

Besides, the magnetic properties of the feeding and roasted materials were measured by a vibrating sample magnetometer (Quantum Design PPMS) at 300 K to reveal the magnetic susceptibility changes.

3. Results and discussion

3.1. Results of the shaking table experiments

The effect of the table tilt angle and particle size on the grade and recovery of iron concentrates was investigated. When −74 µm particle size sample was used, the concentrates with Fe grade of 67–69% were obtained. At 3° table tilt angle, Fe grade was at its highest value of 68.68%. It was observed that the iron recovery decreased with the increase in the tilt angle. The maximum Fe recovery was 32.42% at the tilt angle of 4°.

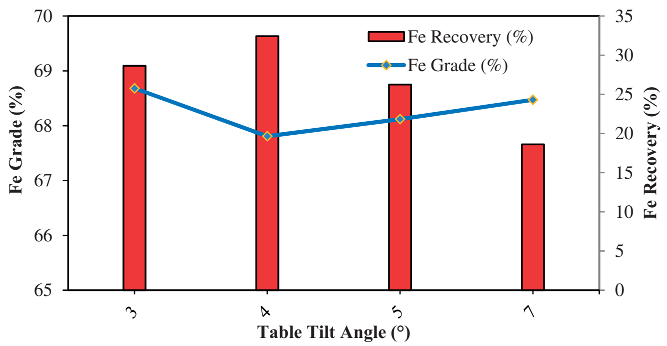

Figure 4. Effect of the table tilt angles on the grade and recovery of the concentrates for −74 μm particle size.

Figure 4 shows the effect of the table tilt angle on grades and recovery of the concentrates for −74 µm feeding material.

Figure 5 shows the effect of the table tilt angle on grade and recovery of the concentrates for −150 + 74 µm feeding material. When the table angle increased from 3° to 5°, it was seen that there was a 2% decrease in the Fe grades of the concentrates. Similar to the shaking table test results applied to the −74 micron size ore, increasing table tilt angle led to a decrease in Fe recovery.

When comparing the effect of the table tilt angle on the grade and recovery of the concentrates for −74 µm and −150 + 74 µm feeding materials, it was noted that the grades of iron concentrates were higher for −74 µm particle size fraction due to the increase of the degree of liberation. The liberated iron minerals went to the concentrate instead of the tailings due to their specific gravity. But, there was a significant loss of iron-bearing particles in the part devoted to tailings. The liberated iron minerals were expected to go into the concentrate product instead of the tailings. But the ore particles below size limits were unable to travel downstream before a particle at the top of the fluid film settled at the deck surface. When the size distribution of ore has a significant number of heavy particles (iron-bearing mineral particles) below these limit particle size, these heavies are lost during the processing (Roy 2009). Therefore, the ore particles below these size limits are invariably lost in the tailings. The problem of losing fine valuable minerals during the processing is not only in the shaking table process but in general. A significant amount of fine iron ore concentrates are lost during the processing in various conventional methods (Surkov et al. 2008). Hence, the recovery of iron concentrates were low for −74 µm compared to greater particle size fraction.

3.2. Results of reverse cationic flotation experiments

The tests were carried out to investigate the effects of collector, depressant, frother dosages and froth collection times on iron recovery and grade. Table 2 shows the results of The Box Behnken test design of reverse cationic flotation experiments. The calculations were executed by using the amount and grades of iron in the sinking part as a result of the flotation tests.

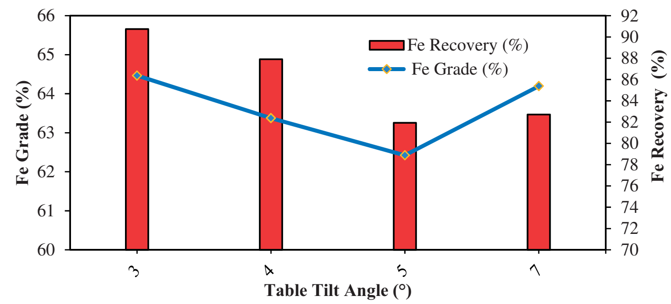

Figure 5. Effect of table tilt angles on the grade and recovery of the concentrates for −150 + 74 μm particle size.

Table 2. Results of The Box Behnken test design of reverse cationic flotation experiments.

No	Depressant (g/t) (X_1)	Collector (g/t) (X_2)	Frother (g/t) (X_3)	Time (min) (X_4)	Fe grade %	Fe recovery %	SiO_2 grade %
1	500	400	100	2	67.42	68.08	1
2	500	300	150	2	61.86	85.88	6
3	500	400	150	3	64.69	90.57	4
4	500	200	50	3	60.19	82.94	6.5
5	400	200	100	3	63.37	84.52	4.5
6	500	400	50	3	66.77	65.49	2
7	500	300	50	2	63.26	78.50	5.5
8	500	200	150	3	64.85	80.81	4
9	400	300	100	4	61.07	67.89	7.5
10	600	300	100	4	65.62	67.09	3.5
11	600	300	100	2	64.80	66.37	4
12	600	400	100	3	65.16	59.57	4
13	500	300	50	4	67.41	86.01	1
14	500	300	150	4	66.60	77.31	2
15	500	400	100	4	69.17	56.64	0.35
16	500	200	100	4	65.03	84.68	3.5
17	400	400	100	3	68.05	77.11	0.5
18	400	300	50	3	64.34	76.68	5
19	600	200	100	3	63.62	80.18	4.5
20	600	300	150	3	63.19	76.29	5.5
21	500	200	100	2	60.62	79.79	6.5
22	400	300	100	2	68.95	81.90	0.65
23	600	300	50	3	68.11	75.95	0.95
24	400	300	150	3	68.28	70.22	0.5

The effects of 400 g/t, 500 g/t, and 600 g/t depressant dosages on the Fe grades were shown in Figure 6. The highest iron grades were obtained when 400 g/t depressant was used. When the depressant dosage was applied as 500 g/t, it caused a decrease of around 5–8% in iron grades. In the experiments using 400 g/t and 500 g/t depressant, the increase in the amount of collector and frother allowed the Fe grade to increase as well. A dramatic decrease in Fe grades were observed for 600 g/t depressant usage. When the amount of depressant was applied at the highest level, the increase in the foaming ratio caused a decline in the Fe grades and an increase in collector amounts led to superior Fe grades.

Effect of the collector dosages on the Fe recovery was shown in Figure 7. At 200 g/t collector dosage, the Fe recovery was slightly increased and then began to decrease with an increase in both frother and depressant dosages. There was no particular change in Fe recoveries for 300 g/t collector dosage. At 400 g/t collector dosage, the Fe recovery was largely increased with escalating frother dosage. However, depressant additions above 500 g/t depressant caused a decrease in the Fe recovery by 10%.

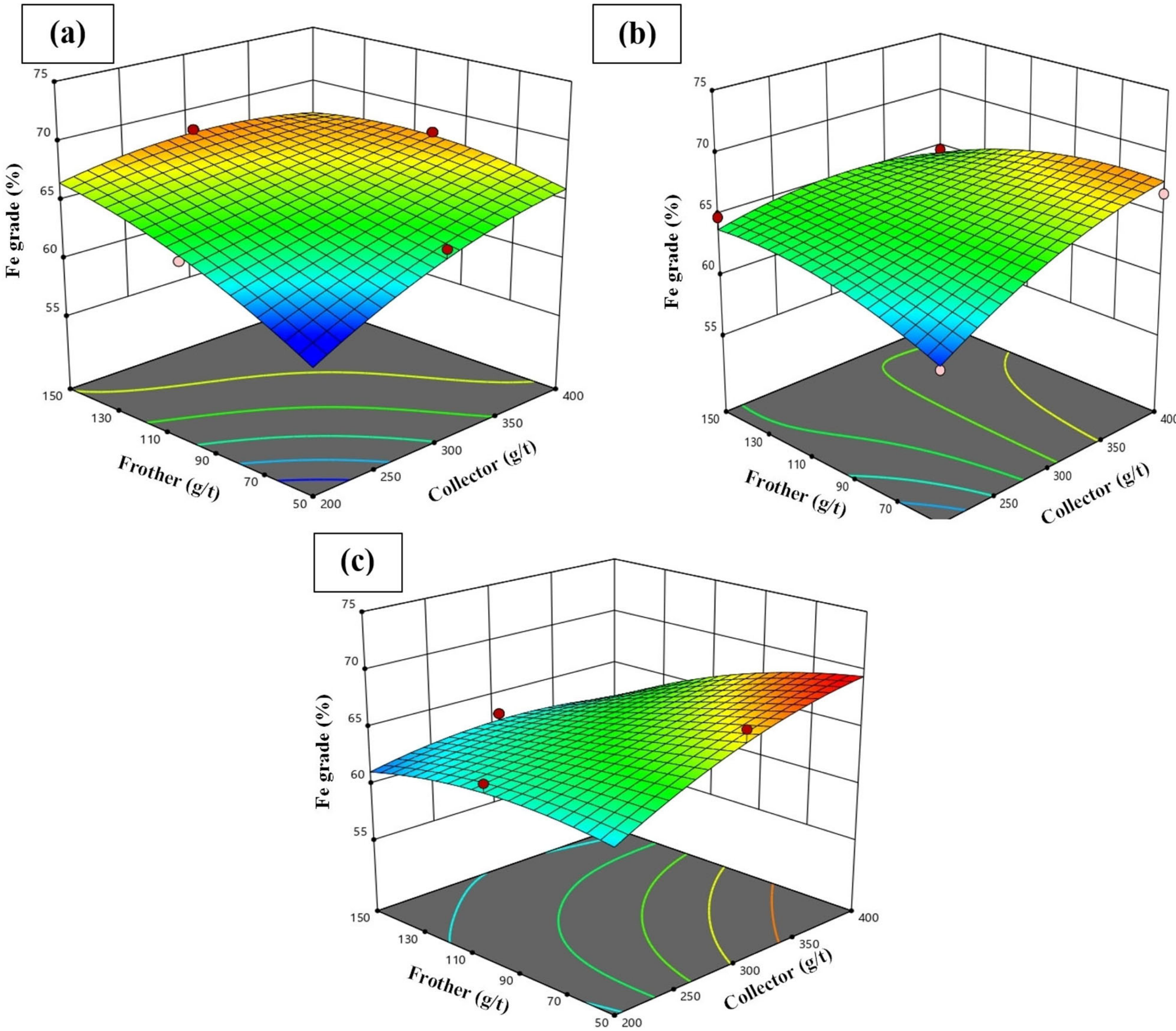

Figure 6. Effect of depressant dosages on the Fe grade; 400 g/t (a), 500 g/t (b), and 600 g/t (c).

The frother affects the particle bubble adhesion kinetics, rendering the relaxation time shorter than the contact time. Under the mixed usage of frother and collector, the collision time is longer than the time required for thinning and rupture of the lamella surrounding the bubble (Araujo, Viana, and Peres 2005). The presence of the frother works to raise the efficiency of the bubble, to maintain and increase the collision time, which in turn works to collect and take as many undesirable materials (silica and calcium oxide) from the ore and thus increase the grades and recovery of iron concentrate. Effects of 50 g/t, 100 g/t, and 150 g/t frother dosages on the Fe grades were shown in Figure 8. It has been determined that the collector has a significant effect on the Fe grade in the experiments where the frother has been used in low quantities (50 g/t). Similarly, in the experiments using a high amount of frother (150 g/t), the negative effect of the depressant on the Fe grade is striking. In the experiments, in which 100 g/t frother has been used, the change in Fe grades is very slight. Although it is aimed to float impurities other than iron in reverse flotation tests, it is understood that the collector also floats the iron content to some extent. For this reason, it was observed that the Fe recovery was higher in the experiments where the collector was used less. It was found that the depressant had a low effect in terms of Fe recovery.

In the reverse cationic flotation tests, 400 g/t depressant dosage, 300 g/t collector dosage, 100 g/t frother dosage, and 2 min froth collection time were the optimum conditions with the other fixed conditions (impeller speed, pH, and particle size). At these conditions, the Fe grade and Fe recovery of the concentrate were 68.95% and 81.90%, respectively. In addition, silica content was 0.61% SiO_2 which does not negatively affect the downstream processing for sintering, blast furnace, and steelmaking industries (<5% SiO_2) (Clout and Manuel 2015).

According to ANOVA analysis for reduced quadratic model, the correlation coefficients (R^2) for the Fe grades (Tables 3 and 4) and recoveries were calculated as 0.91 (p-value <0.0001) and 0.75 (p-value = 0.0007), respectively. The statistical results showed that the generated model matched with the obtained experimental results for the Fe grades. However, it is seen that the R^2 value of the created

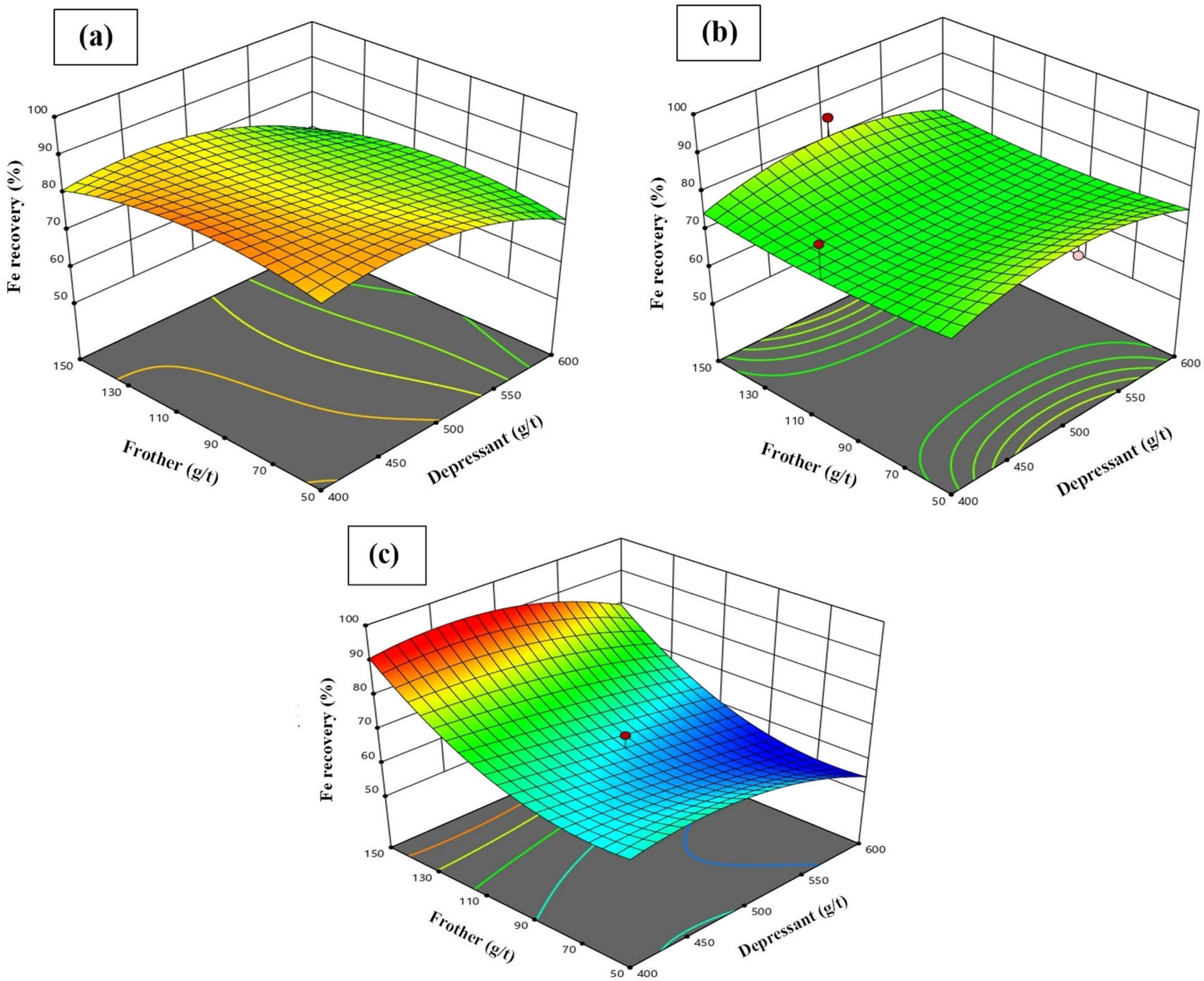

Figure 7. Effect of collector dosages on the Fe recovery; 200 g/t (a), 300 g/t (b), and 400 g/t (c).

model for Fe recovery is lower. The created model meets the experimental results at a rate of 75%. This result showed that although a depressant was used, the ferrous contents also floated. Besides, collector dosage had the greatest effect on grade and recovery of the concentrate than froth collection time while the effect of the depressant and frother dosages on the grade and recovery of the concentrate was not significant. The equations obtained from the models were as follows:

$$\begin{aligned}\text{Fegrade}\,(\%) = {} & 65.30 - 0.495\,X_1 + 1.96\,X_2 + 2.22\,X_4 \\ & -2.21\,X_1X_3 - 1.68\,X_2X_3 - 3.99\,X_1^2X_4 \\ & -1.68\,X_1X_4^2 - 0.68\,X_2^2X_4 - 1.17\,X_2^2X_3^2\end{aligned} \tag{2}$$

$$\begin{aligned}\text{Ferecovery}\,(\%) = {} & 72.82 - 8.47\,X_2 + 6.8\,X_2X_3 + 8.12\,X_3^2 \\ & -5.47\,X_1X_2^2 + 5.74\,X_2^2X_3 + 6.55\,X_2X_3^2 \\ & -6.15\,X_1^2X_3^2\end{aligned} \tag{3}$$

Where X_1 is the depressant dosage (g/t), X_2 is the collector dosage (g/t), X_3 is the frother dosage (g/t), and X_4 is the froth collection time (min).

In some previous studies, Vidyadhar, Kumari, and Bhagat (2012), and Lu et al. (2017) showed that the use of mixtures containing cationic and anionic collectors can provide increased flotation selectivity and recovery compared to each separate reagent. The reverse flotation of specularite mineral using cationic and anionic collector mixtures was planned for future studies.

3.3. Results of first magnetic separation experiments

The beneficiation experiments were carried out to explore the effect of magnetic field intensity on the specularite upgrading (Figure 9).

The results showed that the magnetic field intensity had a significant effect on both the grade and the yield of iron concentrate. When the magnetic field intensity was reduced, iron concentrate grade gradually increased and reached to the highest value of 68.74% at the magnetic field of 0.25 T. The recovery yield of iron concentrates increased with the increasing magnetic field intensity due to the attraction of quartz-iron or andradite-iron bearing particles. Therefore, the Fe grade decreased.

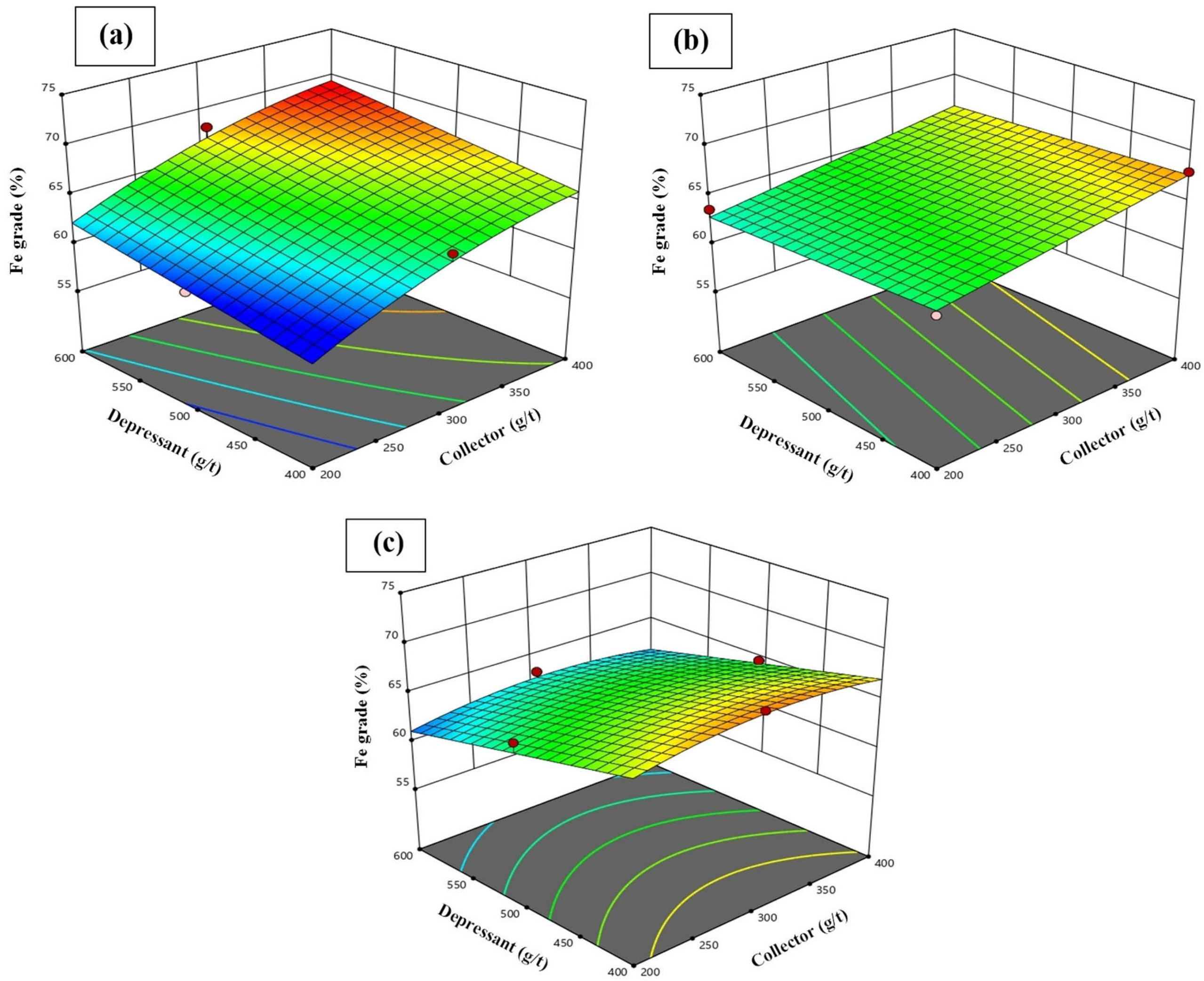

Figure 8. Effect of frother dosages on the Fe grade; 50 g/t (a), 100 g/t (b), and 150 g/t (c).

Table 3. ANOVA for reduced quadratic model (For Fe grade).

Source	Sum of squares	*df*	Mean square	*F*-value	*p*-Value
Model	295.97	9	32.89	15.27	<0.0001*
X_1-Depressant	3.98	1	3.98	1.85	0.1957
X_2-Collector	94.58	1	94.58	43.92	<0.0001
X_4-Froth collection time	19.40	1	19.40	9.01	0.0095
$X_1 X_3$	40.13	1	40.13	18.64	0.0007
$X_2 X_3$	23.28	1	23.28	10.81	0.0054
$X_1^2 X_4$	44.70	1	44.70	20.76	0.0004
$X_1 X_4^2$	15.42	1	15.42	7.16	0.0181
$X_3^2 X_4$	2.00	1	2.00	0.9287	0.3516
$X_2^2 X_3^2$	9.38	1	9.38	4.36	0.0557
Residual	30.15	14	2.15		
Cor total	326.12	23			

*Significant.

Table 4. Fit statistics (For Fe grade).

Std. Dev.	1.47	R^2	0.9075
Mean	93.09	Adjusted R^2	0.8481
C.V. %	1.58	Predicted R^2	0.7319
		Adeq precision	13.2301

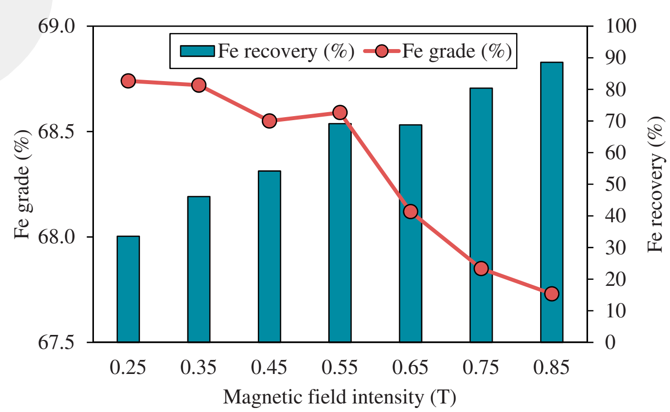

Figure 9. Fe recoveries and Fe grades of the concentrates as a function of magnetic field intensity.

The non-magnetic product, which was obtained at 0.25 T magnetic field intensity, was treated by microwaves to obtain high grade and yield after the second magnetic separation. The Fe grade of the non-magnetic product was 56.39% Fe while the content of the SiO_2 and CaO were 11.2% and 7.6%, respectively. The SEM images of the non-magnetic product are shown in Figure 10.

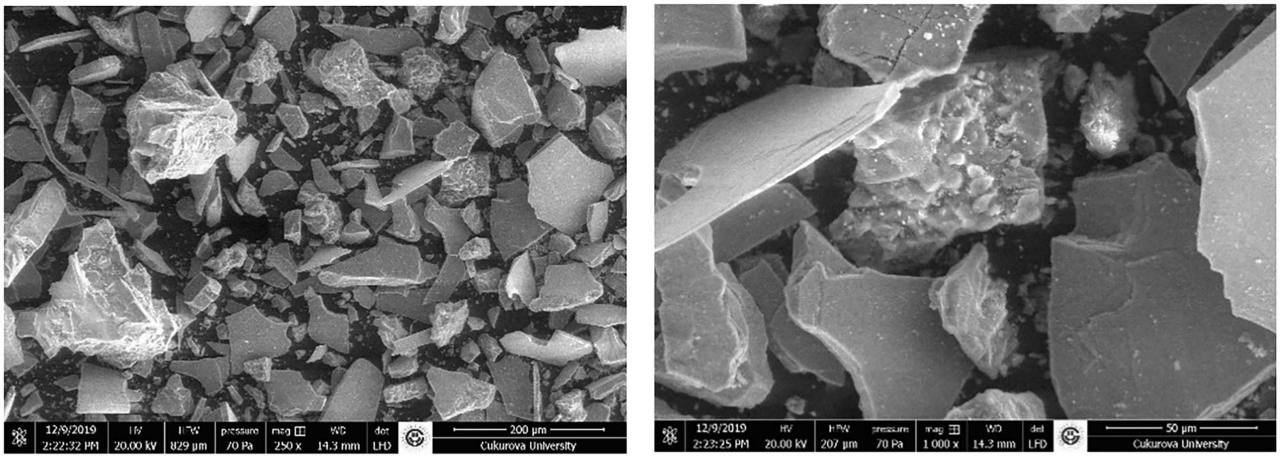

Figure 10. SEM images of the non-magnetic product.

Table 5. Results of the Box-Behnken test design for microwave roasting followed by magnetic separation.

Runs	A: microwave power (kW) (X_1)	B: exposure time (min) (X_2)	C: Magnetic field intensity (T) (X_3)	Fe grade (%)	Recovery (%)
1	0.90	1	0.55	67.80	61.42
2	0.90	2	0.85	65.80	79.91
3	0.72	2	0.55	67.25	70.76
4	0.90	3	0.55	66.72	75.81
5	0.72	3	0.85	66.06	82.23
6	0.72	2	0.55	67.56	65.25
7	0.72	3	0.25	68.80	31.22
8	0.54	2	0.25	66.53	68.02
9	0.90	2	0.25	68.90	27.28
10	0.72	1	0.25	68.90	22.63
11	0.54	1	0.55	65.89	64.47
12	0.72	1	0.85	66.23	72.98
13	0.72	2	0.55	66.71	78.16
14	0.54	2	0.85	66.64	75.79
15	0.54	3	0.55	68.07	59.83

3.4. Results of second magnetic separation applied to the microwave-roasted non-magnetic products

Table 5 shows the Box-Behnken test design for microwave roasting followed by magnetic separation. The Fe grade and recovery of the magnetic products were taken as responses for the statistical analysis.

The Fe grades of concentrates obtained in all experiments were increased from 56.39% to over 65%. The concentrate recoveries vary between 21% and 83%. When the microwave power is applied at low levels, it is seen that the exposure time has no effect on the Fe recovery yields. As the microwave power increased along with the exposure time, the Fe recovery efficiencies increased (Figure 11(a)). It is seen in Figure 11(b) that there is an increase of up to 40% in the Fe recovery efficiencies, especially at low magnetic field intensities after increasing the microwave power. Regardless of the applied microwave exposure time, the increase in magnetic field strength led to the higher Fe recovery (Figure 11(c)).

The recoveries above 72% were obtained at 0.85 T magnetic field intensity. The Fe recovery reached the highest value of 82.23% at 0.72 kW microwave power and 3 min exposure time. A concentrate containing 66.06% Fe was obtained from the non-magnetic material with 56.39% Fe grade under these conditions: 0.85 T magnetic field intensity, 0.72 kW microwave power, 3 min exposure time, $-150+38\,\mu m$ particle size, and 20% solid to liquid ratio. The recovery yield was 82.23%. The 2FI model, which has p value of <0.05, was chosen as the appropriate model with a 76% R^2 value (Table 6).

The equation for the proposed model was formulated in Equation 4.

$$\text{Ferecovery}\,(\%) = 62.38 - 2.96\,X_1 + 3.45\,X_2 + 20.22\,X_3 + 4.76\,X_1X_2 + 11.21\,X_1X_3 + 0.1650\,X_2X_3 \quad (4)$$

The M-H curves of non-magnetic material and microwave-roasted samples are shown in Figure 12. An increase of 0.2 emu/g has been observed in magnetic susceptibilities after microwave exposure.

4. Conclusion

The depletion of iron ore reserves has directed the attention of most sectors, especially the iron and steel industry, to iron minerals such as specularite, which has difficulties in pelleting and sintering. At the same time, specularite concentrates are of great importance because of high iron grades and low impurity values. The focus of this paper was to increase Fe grades of the concentrates to above the

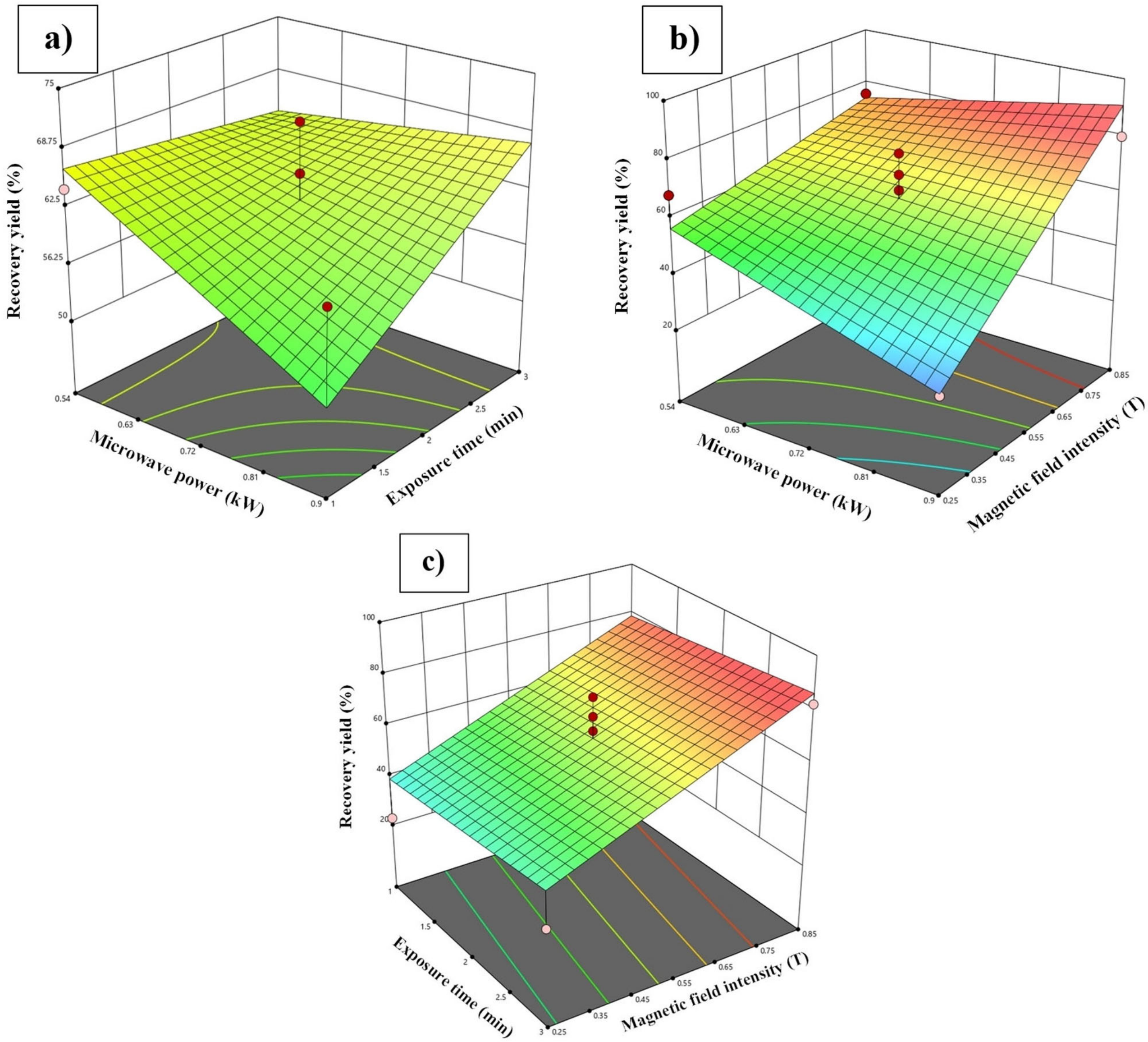

Figure 11. Effect of microwave-assisted magnetic separation on the Fe recovery (a: 0.55 T magnetic field intensity, b: 2 min exposure time, c: 0.54 kW microwave power).

Table 6. The results of ANOVA analysis for 2FI model (for Fe recovery).

Source	Sum of squares	*df*	Mean square	*F*-value	*p*-Value	
Model	4029.839	6	671.6399	4.139134	0.034344	Significant
A-Microwave power	70.15201	1	70.15201	0.432328	0.529309	
B-Exposure time	95.15101	1	95.15101	0.58639	0.465809	
C-Magnetic Field Intensity	3270.787	1	3270.787	20.15697	0.00203	
AB	90.53523	1	90.53523	0.557944	0.476467	
AC	503.1049	1	503.1049	3.100498	0.116301	
BC	0.1089	1	0.1089	0.000671	0.979967	
Residual	1298.127	8	162.2658			
Lack of fit	1214.197	6	202.3662	4.822296	0.181693	Not significant
Pure error	83.9294	2	41.9647			
Cor total	5327.966	14				

commercial grades. The separation techniques including shaking table, magnetic separator, and reverse flotation were proposed to recover Fe from specularite-type refractory iron ore.

In the shaking table separation tests, table tilt angle and particle size had a significant effect on the grades and recovery of the concentrates. The recovery yields of iron concentrates were low for $-74\,\mu m$ particle size fraction compared to higher particle size fraction. A concentrate with 64.67% Fe grade and 90.17% Fe recovery yield was obtained under optimum conditions: particle size of $-150+74\,\mu m$, tilt angle of 3°, shaking speed of 175 strokes/min, and water flow rate of 12 L/min.

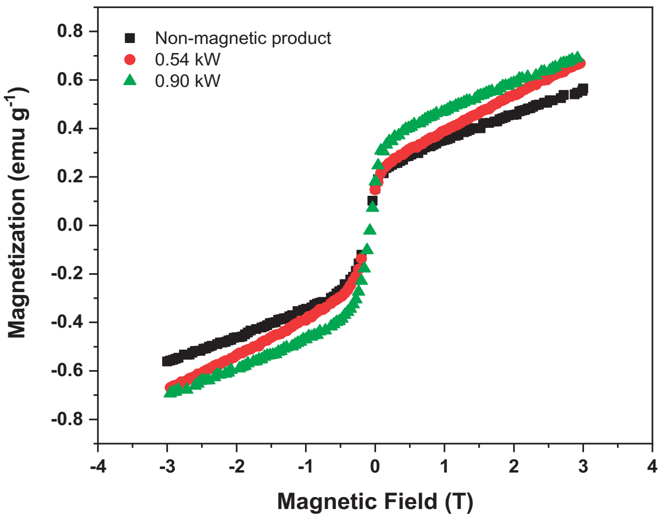

Figure 12. M–H curves of the non-magnetic and microwave-roasted samples (exposure time: 2 min).

In the reverse cationic flotation tests, effects of the corn starch depressant, Aero 3030 C collector, X-133 frother, and froth collection time were investigated by using the Box Behnken test design. The statistical models were created according to the test results. The flotation tests showed that frother and depressant additions increased the floatability of impurities and the higher Fe grades were obtained from the sinking part. In addition, it was determined that the collector had a significant effect on Fe recovery. The optimum reverse flotation test conditions, in which a concentrate with 68.95% Fe grade and 81.9% Fe recovery was obtained, were determined as 400 g/t depressant, 300 g/t collector, 100 g/t frother dosages, 2 min froth collection time, and $-150+38\,\mu m$ particle size at pH 3.

In magnetic separation tests, a concentrate with 67.73% Fe could be produced with 88.57% recovery by applying 0.85 T magnetic field strength at 20% solid to liquid ratio for $-150+38\,\mu m$ particle size. The non-magnetic product, which was obtained at 0.25 T magnetic field intensity, $-150+38\,\mu m$ particle size, and 20% solid/liquid ratio, was treated by microwaves to increase its magnetic susceptibility. Then, the microwave-roasted materials were enriched by the second magnetic separation. To the best of our knowledge, microwave-roasting method was applied to a specularite-type refractory iron ore for the first time. The M-H curves proved that the magnetic susceptibilities of the microwave-roasted materials increased. As a result of the experiments carried out in accordance with the the Box Behnken test design, a concentrate containing 66.06% Fe was obtained from the non-magnetic material with 56.39% Fe grade under these conditions: 0.85 T magnetic field intensity, 0.72 kW microwave power, 3 min exposure time, $-150+38\,\mu m$ particle size, and 20% solid to liquid ratio. The Fe recovery was 82.23%.

Disclosure statement

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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References

Abdulrahman, A. S. 2012. Susceptibility of Agbaja iron ore deposit in north central Nigeria to magnetic separation. *Journal of Modern Manufacturing Technology* 4 (2):87–95.

Abedi, A., A. Bahrami, S. Chehreghani, M. Ghadri, and F. Kazemi. 2022. Mapping specular hematite ore beneficiation routes to industrial application standards. *Rudarsko-geološko-naftni zbornik* 37 (1): 1–9. doi:10.17794/rgn.2022.1.1.

Araujo, A. C., P. R. M. Viana, and A. E. C. Peres. 2005. Reagents in iron ores flotation. *Minerals Engineering* 18 (2):219–24. doi:10.1016/j.mineng.2004.08.023.

Clout, J. M. F., and J. R. Manuel. 2015. Mineralogical, chemical, and physical characteristics of iron ore. In *Iron Ore*, ed. L. Lu, 1st ed., 45–84. Cambridge, UK: Woodhead Publishing.

De Melo, C. H., A. C. de Araujo, and L. Filippov. 2017. Reverse cationic flotation of iron ores with complex silicate gangue minerals. Paper presented at Iron Ore Conference, Perth, Australia, July 24–26.

Falconer, A. 2003. Gravity separation: Old technique/new methods. *Physical Separation in Science and Engineering* 12 (1):31–48. doi:10.1080/1478647031000104293.

Fan, J.-J., G.-Z. Qiu, T. Jiang, Y.-F. Guo, H.-Z. Hao, and Y.-B. Yang. 2012. Mechanism of high pressure roll grinding on compression strength of oxidized hematite pellets. *Journal of Central South University* 19 (9):2611–9. doi:10.1007/s11771-012-1318-5.

Filippov, L. O., V. V. Severov, and I. V. Filippova. 2014. An overview of the beneficiation of iron ores via reverse cationic flotation. *International Journal of Mineral Processing* 127:62–9. doi:10.1016/j.minpro.2014.01.002.

Gaudin, A. M. 1957. *Flotation*. 2nd ed. New York: McGraw-Hill Book Co.

Haryono, D., H. Nugraha, M. Al Huda, W. P. Taruno, and S. Harjanto. 2017. The effect of flotation reagents addition (MIBC and PAX) on the relative permittivity value using 2-electrode capacitance sensor. *Procedia Engineering* 170 (1):369–72. doi:10.1016/j.proeng.2017.03.059.

Kuskov, V., Y. Kuskova, and V. Udovitsky. 2017. Effective processing of the iron ores. *E3S Web of Conferences* 21:02010. doi:10.1051/e3sconf/20172102010.

Li, M., J. Liu, Y. Hu, X. Gao, Q. Yuan, and F. Zhao. 2020. Investigation of the specularite/chlorite separation using chitosan as a novel depressant by direct flotation. *Carbohydrate Polymers* 240: 116334. doi:10.1016/j.carbpol.2020.116334.

Liang, Z., L. Yi, Z. Huang, B. Huang, and H. Han. 2019. A novel and green metallurgical technique of highly efficient iron recovery from refractory low-grade iron ores. *ACS Sustainable Chemistry & Engineering* 7 (22):18726–37. doi:10.1021/acssuschemeng.9b05423.

Lu, D., Y. Hu, Y. Li, T. Jiang, W. Sun, and Y. Wang. 2017. Reverse flotation of ultrafine magnetic concentrate by using mixed anionic/cationic collectors. *Physicochemical Problems of Mineral Processing* 53 (2):724–36. doi:10.5277/ppmp170204.

Ma, X., M. Marques, and C. Gontijo. 2011. Comparative studies of reverse cationic/anionic flotation of Vale iron ore. *International Journal of Mineral Processing* 100 (3–4):179–83. doi:10.1016/j.minpro.2011.07.001.

Mesquita, L. M. S., M. L. Torem, C. A. Lima, and F. A. F. Lins. 2001. Bioflotation of hematite and quartz - fundamental studies. Paper presented at National Meeting on Ore Treatment and Extractive Metallurgy, Rio De Janeiro, Brasil.

Mowla, D., G. Karimi, and K. Ostadnezhad. 2008. Removal of hematite from silica sand ore by reverse flotation technique. *Separation and Purification Technology* 58 (3):419–23. doi:10.1016/j.seppur.2007.08.023.

Neymayer, P. L., E. S. V. George, and E. C. P. Antonio. 2013. Effect of amine and starch dosage on the reverse cationic flotation of an iron ore. *Minerals Engineering* 45:180–4. doi:10.1016/j.mineng.2013.03.001.

Nunna, V., S. Hapugoda, M. I. Pownceby, and G. J. Sparrow. 2021. Beneficiation of low-grade, goethite-rich iron ore using microwave-assisted magnetizing roasting. *Minerals Engineering* 166:106826. doi:10.1016/j.mineng.2021.106826.

Omran, M., T. Fabritius, A. M. Elmahdy, N. A. Abdel-Khalek, M. El-Aref, and A. E. H. Elmanawi. 2014. Effect of microwave pre-treatment on the magnetic properties of iron ore and its implications on magnetic separation. *Separation and Purification Technology* 136:223–32. doi:10.1016/j.seppur.2014.09.011.

Pan, J., B. Shi, D. Zhu, and Y. Mo. 2016. Improving sintering performance of specularite concentrates by pre-briquetting process. *ISIJ International* 56 (5):777–85. doi:10.2355/isijinternational.ISIJINT-2015-578.

Roy, S. 2009. Recovery improvement of fine iron ore particles by multi gravity separation. *The Open Mineral Processing Journal* 2 (1):17–30. doi:10.2174/1874841400902010017.

Singh, R. K., and A. Das. 2013. Analysis of separation response of Kelsey centrifugal jig in processing fine coal. *Fuel Processing Technology* 115:71–8. doi:10.1016/j.fuproc.2013.04.005.

Srdjan, M. B. 2007. *Handbook of flotation reagents: Chemistry, theory and practice*. Oxford: Elsevier.

Surkov, A. V., E. V. Samykina, L. V. Eppelbaum, and S. V. Semenov. 2008. The main reason for mineral loss in gravity dressing. *The Open Mineral Processing Journal* 1 (1):37–44. doi:10.2174/1874841400801010037.

Tang, Z., H. Xiao, Y. Sun, P. Gao, and Y. Zhang. 2022. Exploration of hydrogen-based suspension magnetization roasting for refractory iron ore towards a carbon-neutral future: A pilot-scale study. *International Journal of Hydrogen Energy* 47 (33):15074–83. doi:10.1016/j.ijhydene.2022.02.219.

Tanriverdi, M., S. Sen, and T. Cicek. 2018. Micaceous iron oxide production by application of magnetic separation. *Physicochemical Problems of Mineral Processing* 54:35–44. doi:10.5277/ppmp1845.

Tosun, Y. İ. 2020. Concentration and microwave radiated reduction of Southeastern Anatolian hematite and limonite ores—reduced iron ore production. In *Iron Ores*, ed. V., Shatokha, 1–20. London: IntechOpen.

Tripathy, S. K., V. Singh, Y. R. Murthy, P. K. Banerjee, and N. Suresh. 2017. Influence of process parameters of dry high intensity magnetic separators on separation of hematite. *International Journal of Mineral Processing* 160:16–31. doi:10.1016/j.minpro.2017.01.007.

Uwadiale, G. G. O. O. 1992. Flotation of iron oxides and quartz—a review. *Mineral Processing and Extractive Metallurgy Review* 11 (3):129–61. doi:10.1080/08827509208914209.

Vapur, H, and S. Top. 2016. Spekülarit cevherinin kalite özelliklerinin iyileştirilmesi [Improving of quality properties of the specularite ore]. *Çukurova University Journal of the Faculty of Engineering and Architecture* 31 (1):293–300. doi:10.21605/cukurovaummfd.317835.

Vidyadhar, A., N. Kumari, and R. P. Bhagat. 2012. Adsorption mechanism of mixed collector systems on hematite flotation. *Minerals Engineering* 26:102–4. doi:10.1016/j.mineng.2011.11.005.

Vidyadhar, A., N. Kumari, and R. P. Bhagat. 2014. Adsorption mechanism of mixed cationic/anionic collectors in quartz–hematite flotation system. *Mineral Processing and Extractive Metallurgy Review* 35 (2):117–25. doi:10.1080/08827508.2012.723649.

Vidyadhar, A., and R. Singh. 2007. Froth flotation and its application to concentration of low grade iron ores. In *Processing of iron ore*, 103–114. Perth: Australasian Institute of Mining and Metallurgy Publication.

Wang, S., Y. Jiang, Y. Guo, F. Chen, and L. Yang. 2021. Effects of specularite on the preheating and roasting characteristics of fluorine-bearing iron concentrate pellets. *Crystals* 11 (11):1319. doi:10.3390/cryst11111319.

Wang, W. Z., J. R. Zhang, and C. G. Yang. 2011. Experimental research on beneficiation process for a specularite ore. *Advanced Materials Research* 304:387–90. doi:10.4028/www.scientific.net/AMR.304.387.

Wills, B. A., and J. Finch. 2015. *Wills' mineral processing technology: An introduction to the practical aspects of ore treatment and mineral recovery*. 8th ed. Oxford, UK: Butterworth-Heinemann.

Wu, F., Z. Cao, S. Wang, and H. Zhong. 2017. Phase transformation of iron in limonite ore by microwave roasting with addition of alkali lignin and its effects on magnetic separation. *Journal of Alloys and Compounds* 722:651–61. doi:10.1016/j.jallcom.2017.06.142.

Yuan, S., H. Xiao, R. Wang, Y. Li, and P. Gao. 2022. Improved iron recovery from low-grade iron ore by efficient suspension magnetization roasting and magnetic separation. *Minerals Engineering* 186:107761. doi:10.1016/j.mineng.2022.107761.

Yuhua, W., and R. Jianwei. 2005. The flotation of quartz from iron minerals with a combined quaternary ammonium salt. *International Journal of Mineral Processing* 77 (2):116–22. doi:10.1016/j.minpro.2005.03.001.

Zhang, X., J. Deng, M. Huangfu, Y. Wang, B. Wu, S. Li, Z. Pang, and H. Mei. 2022. Novel insights into the influence of ferric ion as a surface modifier to enhance the floatability of specularite. *Powder Technology* 398:117141. doi:10.1016/j.powtec.2022.117141.

Zhang, Y., Y. Zhou, B. Liu, G. Li, and T. Jiang. 2014. Roasting characteristics of specularite pellets with modified humic acid based (MHA) binder under different oxygen atmospheres. *Powder Technology* 261:279–87. doi:10.1016/j.powtec.2014.04.053.

Zhang, F., D. Zhu, J. Pan, Y. Mo, and Z. Guo. 2018. Improving the sintering performance of blends containing Canadian specularite concentrate by modifying the binding medium. *International Journal of Minerals, Metallurgy, and Materials* 25 (6):598–608. doi:10.1007/s12613-018-1607-6.

Zhou, W., Y. Sun, Y. Han, P. Gao, and Y. Li. 2021. Recycling iron from oolitic hematite via microwave fluidization roasting and magnetic separation. *Minerals Engineering* 164:106851. doi:10.1016/j.mineng.2021.106851.

Zhu, D., T. Chun, J. Pan, and J. Zhang. 2013. Influence of basicity and MgO content on metallurgical performances of Brazilian specularite pellets. *International Journal of Mineral Processing* 125:51–60. doi:10.1016/j.minpro.2013.09.008.

Zhu, X., Y. Han, Y. Cao, Y. Sun, and Y. Li. 2022. Magnetization roasting of specularite ore: Phase transformation, magnetism and kinetics. *Mineral Processing and Extractive Metallurgy Review. Advance Online Publication*. doi:10.1080/08827508.2022.2068010.

Zhu, D., B. Shi, J. Pan, and F. Zhang. 2018. Effect of pre-briquetting on the granulation of sinter mixture containing high proportion of specularite concentrate. *Powder Technology* 331:250–7. doi:10.1016/j.powtec.2018.03.015.

Zhu, D. Q., Y. Y. Tang, V. Mendes, J. Pan, and Y. Zhai. 2009. Improvement in pelletization of Brazilian specularite by high-pressure roller grinding. *Chinese Journal of Engineering* 31 (1):30–5. doi:10.13374/j.issn1001-053x.2009.01.003.

Znamenáčková, I., M. Lovás, A. Mockovčiaková, Š. Jakabský, and J. Briančin. 2005. Modification of magnetic properties of siderite ore by microwave energy. *Separation and Purification Technology* 43 (2):169–74. doi:10.1016/j.seppur.2004.11.002.