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Microwave-Assisted Reduction Roasting-Magnetic Separation Studies of Two Mineralogically Different Low-Grade Iron ores

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Abstract

Microwave-assisted reduction behaviours of two low-grade iron ores having a similar Fe content of 49% but distinctly different mineralogical and liberation characteristics have been studied. Their performances in terms of the iron grade and recovery as obtained from the statistically designed microwave (MW) roasting followed by Low-intensity Magnetic Separation (LIMS) experiments have been compared. At respective optimum conditions, the titano-magnetite ore (O1) could yield an iron concentrate having a grade of 62.57% Fe with a Fe recovery of 60.01% while the goethitic ore (O2) could be upgraded to 64.4% Fe at a Fe recovery of 33.3%. Compared to the goethitic ore, the titano-magnetite ore responds better to MW heating. The characterization studies of the feed and roasted products obtained at different power and time using X-ray diffraction, optical microscopy, vibrating sample magnetometer and electron probe microanalysis explain the sequential reduction of the iron oxide phases. Finally, taking advantage of the MW absorbing character of the titano-magnetite ore, a blend of the same with the goethite-rich ore at a weight ratio of 60:40 (O2: O1) has been subjected to MW roasting resulting in a concentrate of 61.57% Fe with a Fe recovery of 64.47%.

Keyword: Microwave; Reduction roasting; Titano-magnetite ore; Goethitic ore; Magnetic separation

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1. Introduction

The coupled approach of reduction roasting followed by magnetic separation is an effective way to recover iron values from lean-grade iron ore resources that are difficult to be treated through physical beneficiation routes [1-2]. This process is not only suitable in upgrading the iron content in mineralogically complex and unliberated iron ores but also in achieving a higher Fe grade and recovery compared to complicated beneficiation circuits [3-4]. With the gradual depletion of high-grade iron ores and increasing demand for iron and steel, the research on an alternative beneficiation strategy like reduction roasting is gaining importance nowadays [5].

Microwave heating has many advantages over conventional heating, the most prominent one being rapid and material selective heating [6]. Conventional heating has a significant bottleneck in terms of its inability to stream the heat volumetrically which results in a roasted mass with "cold centres" while microwave heating has an inherent property of heating the entire volume of the material [7-8]. It is stated that microwave irradiation heats the interior of the material volume more compared to the surface [9-10]. This inverted temperature gradient can be easily observed on numerous materials, including metal oxides, which have a poor thermal conductivity [11]. Microwave-material interaction depends on many factors like real (□’) and imaginary permittivities (□’’), heat capacity, power level, geometry and mass of the sample, presence of coupling agents and occurrence of a chemical reaction or phase changes [12].

The application of microwave energy in the carbothermal reduction roasting of low-grade iron ores can be considered as a relatively new approach. Wu et al. [13] investigated the phase transformation and magnetic properties of a limonitic ore having a feed of 40.1% Fe via microwave roasting with alkali lignin as reductant and found that goethite and hematite phases present within the ore could be successively reduced to magnetite having a grade of 57.19% Fe. MW-assisted reduction roasting of a banded iron ore (37% Fe) using charcoal as the reductant was reported by Raypudi et al. [14]. The process resulted in a magnetic concentrate of grade 61.6% Fe under conditions such as an MW power of 720 W, a charcoal dosage of 9%, and time of 8 min. Other than the significant advantages such as a faster reduction rate, the minimal formation of detrimental phases like fayalite in the MW roasted mass is also reported [15]. Apart from reduction studies, the influence of microwave pre-treatment has also been investigated in the grindablity tests of some of the iron ores [16-17].

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Since several ores have different microwave heating behaviours, it is essential to explore the variation in the microwave responses of low-grade iron ores having distinctly different mineralogical characteristics and liberation pattern, which is yet to find a place in the literature. The behaviour of coal as a reductant in MW environment, which is not a very good absorber of microwaves, is also to be established as most of the studies reported so far have used high pure graphite or charcoal as the carbothermic reducing agent [14, 15, 18]. In this context, the present communication explores the microwave-assisted reduction roasting of two low-grade iron ores having almost the same iron content but distinguishably different mineralogical properties. One of them is a mine overburden rich in goethite and limonite while the other one is a titano-magnetite ore.

During the process of selective mining, the majority of the low-grade hematitic ores are rejected because of their incorporation with other mineral phases like goethite, kaolinite, and gibbsite. These phases are chemically bound with the water in the matrix resulting in high Loss on Ignition (LOI). It eventually raises high-pressure steam during induration of the pellets causing cracks inside the pellets, which leads to reduced strength. Known as overburden, these types of ores are not only fragile due to the inter-granular pore spaces and voids along the weaker bedding planes [19], but also not upgradable through physical separation processes mostly due to liberation issues [20]. The method of reduction roasting-magnetic separation has been successfully implemented for such goethetic ores [21, 22]. However, the response of such ores to microwave-assisted carbothermic reduction roasting is yet to be established. On the other hand, the titano-magnetite ores contain ilmenite and magnetite and are usually regarded as complex ores since the complete liberation of iron from the iron-oxide matrix cannot be attained even through fine grinding [23-25]. Reduction roasting, followed by magnetic separation has been successfully applied to beneficiate these kinds of ores [26-28]. Though magnetite is known to be an excellent absorber of microwave [29], the microwave response of low-grade titano-magnetite ore has not been studied to date.

The present work primarily focuses on the comparison of the microwave-based heating and reduction behaviour of a titano-magnetite ore with a goethite rich low-grade ore using high-ash coal as the reductant. The experiments have been conducted using the Taguchi based statistical design, whereas the characterization studies such as Vibrating Sample Magnetometer (VSM), X-ray Diffraction (XRD), Electron Probe Micro-analyzer (EPMA) and Optical Microscopy (OM) have been carried out to establish the difference in the MW responses as well as reduction mechanisms.

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2. Materials and methods

2.1 Samples

The titano-magnetite ore (O1) was collected from Nigeria, while the iron ore overburden sample (O2) was received from Jharkhand, India. The samples were crushed to below 10 mm and riffled to produce representative samples. The chemical analyses of the respective ore samples are listed in Table 1. A high-ash coal sample (fixed carbon: 34.57 wt.%, volatile: 29.11 wt.%, ash: 23.12 wt.%, moisture: 13.2 wt.%) used as the reluctant in this study was collected from Talcher, India. The size of the coal was reduced to below 1 mm for all the reduction experiments. The size analyses of the iron ore and coal sample are given in Table 2. In the case of O1, it is observed that the iron content decreases with the particle size while for O2, the iron values are found to be almost equally distributed across all the size fractions. In the case of the coal sample, the ash content shows a marginal increase in the ash content at finer size fractions.

Table 1 Chemical analysis of the two different types of iron ores

Iron Ore	**Fe**	**Al_2O_3**	**SiO_2**	**TiO_2**	**MgO**	**LOI**
O1	49.6	6.1	7.2	13.1	1.1	1.5
O2	49.1	10.2	7.3	-	-	12.3

Table 2 Size analyses of the iron ore and coal samples

Iron Ore					Coal		
Size, µm	O1		O2		Size, µm	Wt., %	Ash, %
	Wt., %	Fe (T)	Wt., %	Fe (T)			
-10000+5600	15.38	51.97	33.12	49.78	-1000+500	54.83	21.45
-5600+3350	14.85	51.99	23.2	49.26			
-3350+2000	11.30	50.13	13.12	49.09	-500+300	14.88	23.94
-2000+1000	18.41	50.89	12.96	48.65			
-1000+500	15.59	53.53	5.6	48.51	-300+150	14.10	24.58
-500+300	8.58	50.66	3.68	48.33			
-300+150	8.16	42.62	3.52	47.95	-150+100	4.44	25.1
-150+100	2.30	38.92	1.12	46.99			
-100	5.44	32.96	3.68	46.98	-100	11.75	27.34
Bulk	100	49.60	100	49.10	Bulk	100	23.12

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2.2 Microwave-based reduction roasting

The reduction roasting experiments were performed in a laboratory-scale microwave system supplied by VB Ceramics Consultants, Chennai. The significant features of the microwave furnace are a control panel, a power supply system, an air-cooled magnetron, a circulator with a water load, an aluminium waveguide and a multimode cavity for sample processing. The MW furnace with a frequency of 2.45 GHz is designed for a maximum power input of 10 kW and a minimum of 1 kW, allowing up to a maximum temperature of 1500ºC. The insulation zone inside the furnace is about 125 mm from all the sides of the chamber constructed with vacuum-formed ceramic fibreboard (80% Al_2O_3 and 20% SiO_2) supplied by Zircar, USA. This type of insulation helps the furnace for a fast rate of heating and long soaking hours. The temperature for all the experiments was directly measured through RAYTEK non-contact IR sensor (Pyrometer) fixed at the top of the furnace. The pyrometer can sense temperature ranging from 350 to 1500 °C, with the output being connected to a Eurotherm PID controller.

The MW-based reduction experiments were conducted by taking the thoroughly mixed iron ore and coal sample at a scale of 500 g in a tall form quartz crucible. The mixture was kept in the MW furnace for a designated time operated at a particular power in the presence of air. On completion of each experiment, the roasted ore was quickly removed from the chamber and instantly water-quenched to inhibit re-oxidation. Consequently, the obtained roasted mass was subjected to various characterization techniques such as EPMA, XRD, VSM and OM.

2.3 Magnetic Separation

The cooled roasted mass was wet-ground in a laboratory-scale ball mill at a pulp density of 30% to a particle size below 75 µm. The slurry was then subjected to magnetic separation in a Wet LIMS unit supplied by In-Smart Systems, Hyderabad, India at a magnetic field strength of about 0.15 T. The magnetic and non-magnetic fractions were collected separately, dried and subjected to chemical analysis for determining the Fe grade and yield. The process flow diagram of the entire experimental work carried out is displayed in Fig. 1.

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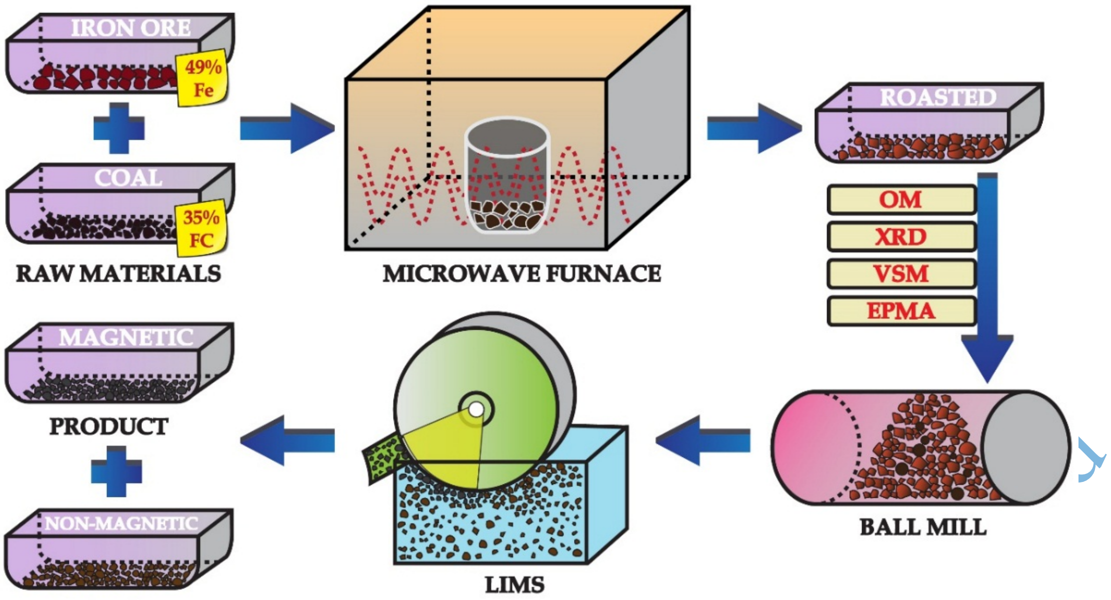

Fig. 1 Process flow diagram of the experimental work

2.4 Experimental Design

The design of the reduction experiments was done with the aid of Taguchi's L_{16} orthogonal arrays using the software MINITAB 14.0. The purpose was to find the levels of the factors responsible for maximizing the Fe grade and Fe recovery of the magnetic product. The signal to noise (S/N) ratios were determined for assessing the process response, i.e. Fe grade and recovery. The details of the factors and their respective levels in the L_{16} design are shown in Table 3.

Table 3 Assignment of levels of various factors used in the reduction

Factors	Level 1	Level 2	Level 3	Level 4
Power, kW	4	5	6	7
Time, min	10	15	20	25
Coal to ore ratio	0.05	0.10	0.15	0.20

2.5 Characterization techniques

The chemical analyses of the samples were carried out both through wet chemical and X-ray Fluorescence (Make-PANalytical, Model-Zetium) technique. The XRD studies of the representative iron ore samples and some of the products were carried out using a Philips Diffractometer (PW-1710) having automatic divergence slit, receiving slit and graphite monochromator assembly. Cu K_α radiation operating at 40 kV and 20 nA was employed for

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this work. Reflected light optical microscopic studies were conducted using a Leiz-make microscope. EPMA of the ore and some roasted samples were performed using a JEOL super probe (JXA-8200). The working voltage was maintained at 20 kV with the beam current at 40-100nA. The elemental distribution in the samples was determined using the X-ray scanning mode. The magnetic properties of the sample before and after roasting were tested using the Princeton VSM (Make-Quantum design) at room temperature. Pulverized mass was placed between the electromagnets, and then the magnetic field was increased to obtain the Saturation Magnetization (SM) value of the test sample.

3. Results and Discussion

3.1 Characterization of feed

The samples O1 and O2 were subjected to XRD and optical microscopic studies to determine the phases present in them. As evident from the XRD pattern shown in Fig. 2a, the O1 sample contains magnetite and ilmenite as the major minerals. The corresponding optical micrograph (Fig. 2b) shows that the magnetite phase is not homogeneous and contains minute intergrowth of ilmenite. Magnetite is the dominant primary oxide mineral phase occurring as irregular massive grains. It is identified by its greyish white colour with a pinkish tinge, and the non-pleochroic as well as isotropic nature, as observed under plane-polarized light. It is also observed that magnetite contains exsolution texture in the form of lamellar ilmenite. It is characterised by the development of lamellar and emulsion intergrowth of ilmenite. Isolated grains of magnetite without intergrowth is rarely seen in this sample. The fine lamellar intergrowth of ilmenite with magnetite forming "Widmanstatten texture" or "Trellis intergrowth" is noticed in the feed sample (Fig. 2b). The origin of lamellar intergrowth is generally due to unmixing or exsolution between these two phases [30].

On the other hand, as revealed from the XRD pattern given in Fig. 2c, the O2 sample is a goethite dominated ore associated with hematite, kaolinite and quartz. Goethite is usually formed under oxidizing conditions as a weathering product of iron-bearing minerals, through inorganic or organic precipitation. In this sample, alternate colloform layers of goethite are found to be well developed (Fig. 2d), with the layers having varying thickness and reflectivity. Rhythmic concentric layers of goethite in different shades are also seen. Both massive and

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colloidal forms of goethite are observed in this sample. The presence of hematite in the periphery of goethite suggests the possible dehydration of the latter leading to its formation.

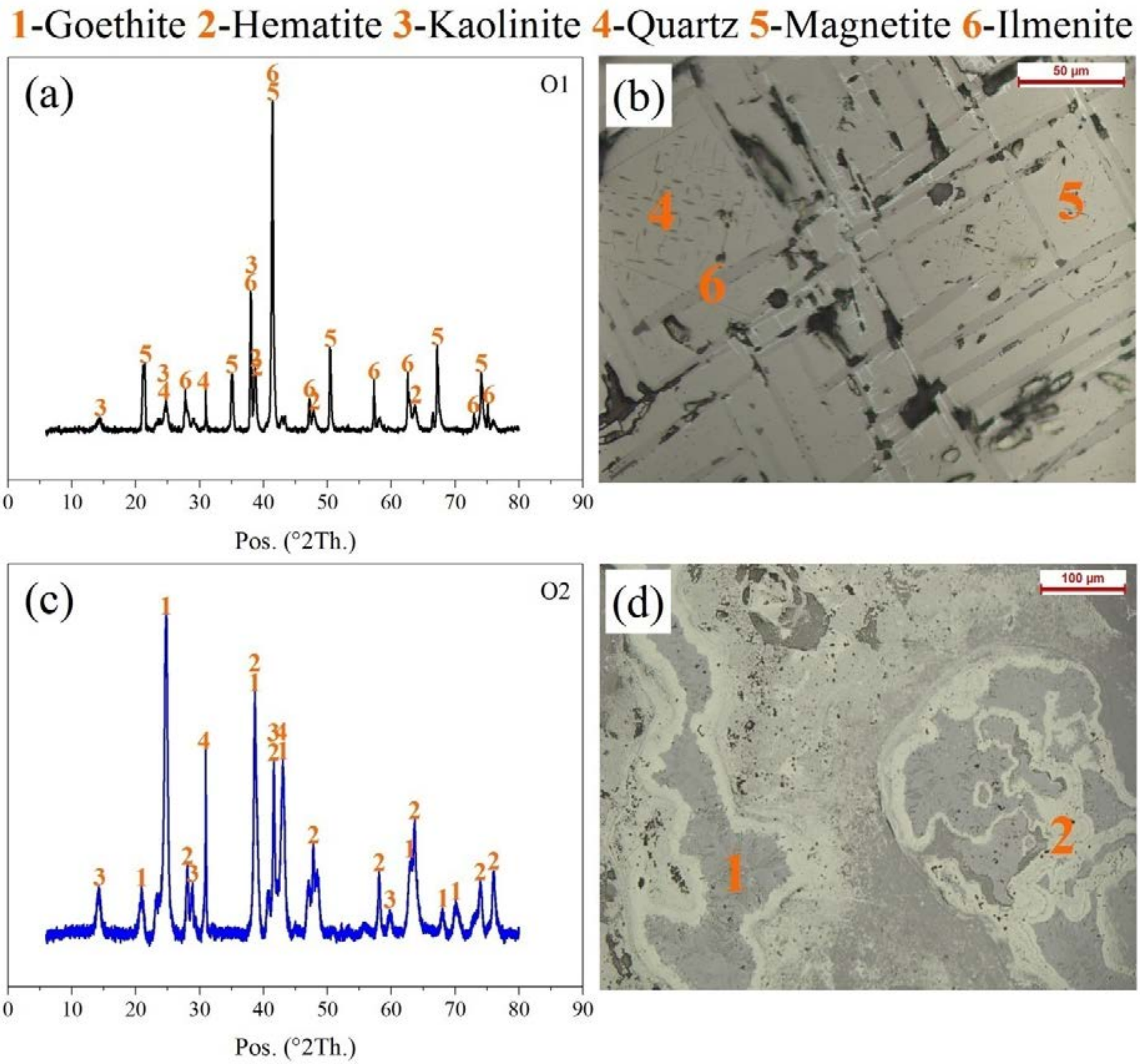

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Fig. 2 XRD patterns and optical micrographs of the feed ores (a and b) O1 and (c and d) O2

3.2 Microwave-based reduction roasting of O1 and O2 ores

The reduction roasting-magnetic separation experiments were conducted as per the L_{16} design, and the corresponding results in terms of the Fe grade and recovery of the magnetic products are given in Table 4. On comparing the grade and recovery for both the ores, it is understood that O1 responded reasonably better compared to O2 in the microwave-assisted reduction roasting followed by the magnetic separation process. The results indicate that ore O1 could be upgraded to 62.88% Fe with a Fe recovery of 62.19%. Though experiments were resulting in higher Fe recovery, the conditions were not adequate to yield Fe content in the range of 63-64%. However, in the case of O2, the grades obtained were not very impressive since the best concentrate was obtained with only 60.2% Fe and a Fe recovery of 66.32%. It is also noticeable that the ore O2 needed more MW power and residence time to achieve a Fe grade of 60.2%. Further, the results were subjected to statistical analyses to comprehend the roles of different factors in the reducing behaviour of both the ores.

Table 4 Iron grade and recovery response from statistically designed experiments

Exp. No.	**Experimental Conditions**			**O1+Coal**		**O2+Coal**	
	Power, kW	**Time, min**	**Coal to ore ratio**	**Grade, %**	**Recovery, %**	**Grade, %**	**Recovery, %**
1	4	10	0.05	61.59	72.59	59.57	6.48
2	4	15	0.1	60.14	77.40	59.51	11.33
3	4	20	0.15	61.31	58.97	58.97	36.89
4	4	25	0.2	60.47	66.36	57.24	24.13
5	5	10	0.1	58.97	74.30	58.73	21.57
6	5	15	0.05	61.08	55.44	59.89	20.36
7	5	20	0.2	62.88	62.19	58.36	80.16
8	5	25	0.15	66.85	51.62	59.17	61.96
9	6	10	0.15	62.24	66.61	58.06	52.11
10	6	15	0.2	65.34	54.00	59.14	48.50
11	6	20	0.05	64.11	40.82	58.41	78.64
12	6	25	0.1	63.22	59.10	58.35	66.66
13	7	10	0.2	59.67	65.48	58.32	46.00
14	7	15	0.15	70.55	43.01	57.78	62.86
15	7	20	0.1	66.59	29.77	59.32	76.47
16	7	25	0.05	65.25	45.08	60.2	66.32

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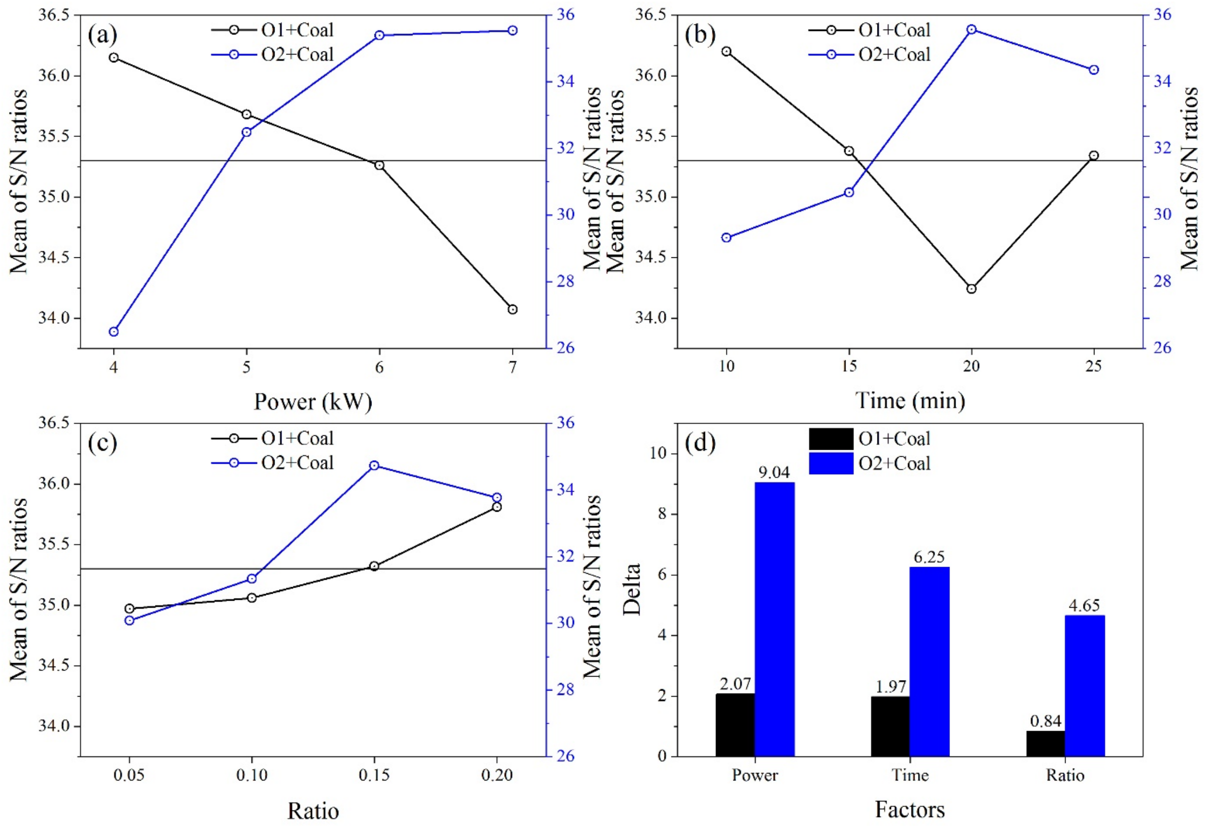

Fig. 3 Main effects plot showing the variation in S/N ratio as a function of different factors (a) MW power (b)time (c) coal/ore ratio and (d) delta values as calculated for the L16 microwave reduction roasting experiments for O1+Coal and O2+Coal

Fig. 3 shows the impact of the factors on the grade and recovery of both the ore types, i.e., O1 and O2. The main effects, as well as the delta plot, indicate that the microwave power and exposure time are the most significant parameters, while the ore to coal ratio has a relatively less substantial influence. It is evident from the main effects plot that the mean S/N ratio for O1, which was calculated considering both Fe grade and recovery as the responses, decreases as the microwave power increases from 4 to 7 kW. In contrast, in the case of O2, the mean S/N ratio rises with the same increase in microwave power. This variation in effect may be attributed to the difference in mineralogical characteristics of O1 and O2. Although a similar trend can also be seen in the case of the exposure time of microwave radiation, there is an increase in the mean S/N ratio for O1 at a high time of 25 min. This trend is observed because of the rise in iron grade with a marginal effect on the recovery at an exposure time of 25 min (Table 3). The mean S/N ratios obtained at different coal to ore ratios for O1 do not vary significantly, and hence, the factor ratio has less impact on the iron grade and recovery as compared to microwave power and time. However, the rise in mean S/N ratios at high ore to coal ratios, i.e., 0.15 and 0.20 for O2, suggests that ore O2 is not as amenable as O1 to

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microwave radiation. Thus, microwave heating profiles of both the ore types were studied further to comprehend the above-discussed results.

3.3 Microwave heating studies of ores with response to coal addition

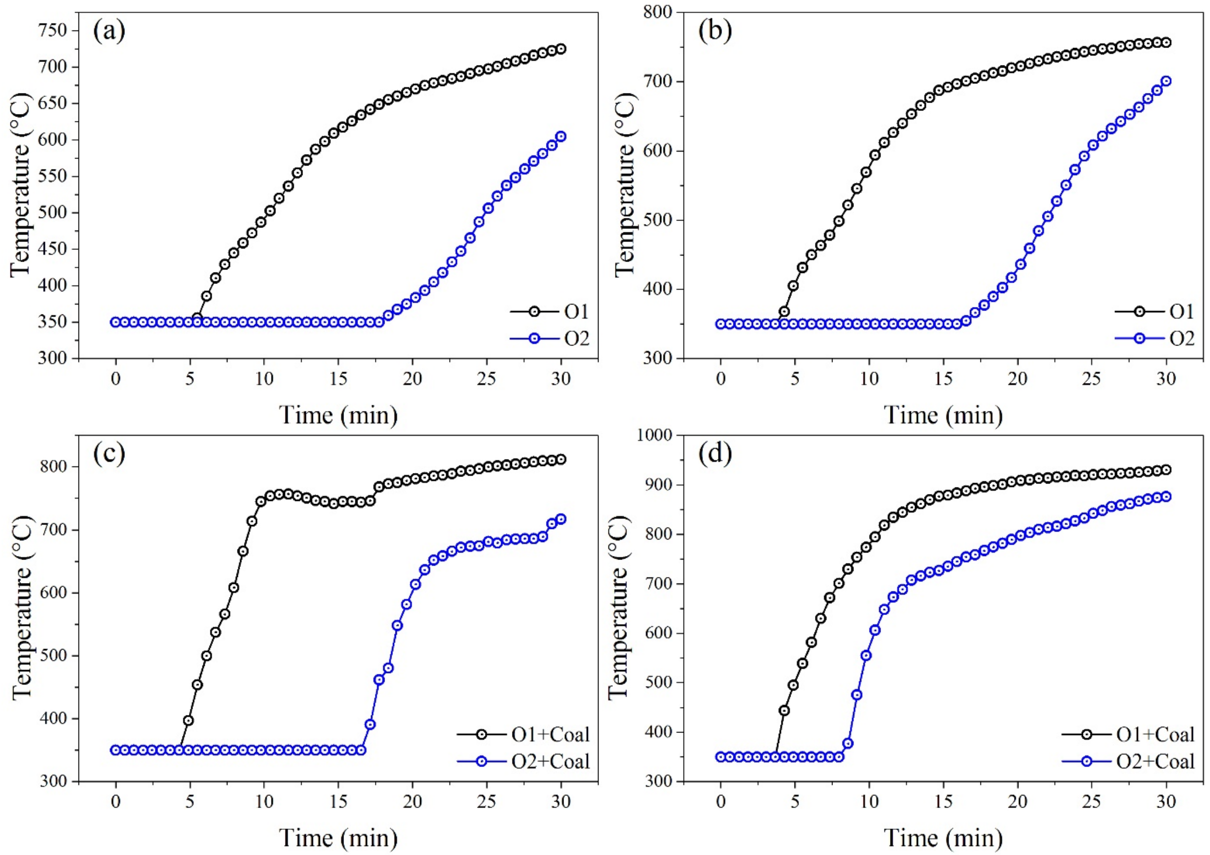

Fig. 4 Microwave heating behaviour of raw ores O1 and O2 at an MW power of (a) 5 kW and (b) 7 kW and roasted with addition of coal (0.2 coal/ore) at an MW power of (c) 5 kW and (d) 7 kW

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The temperatures of the O1 and O2 ores versus microwave heating time are shown in Fig. 4. Though both the ores heated well in response to the microwave treatment, O1 heated significantly better than O2. For example, at a microwave power of 5 kW, the ore O1 could reach a maximum temperature of about 727 °C after 30 min heating time while the ore O2 behaved utterly different with the maximum temperature around 600 °C. In the same way, the ore O1 showed better heating characteristics than O2 at a higher microwave power of 7 kW. It can also be seen that the ore O1 had a higher initial heating rate, which declined gradually with an increase in microwave exposure time. It is this heating characteristic of O1, which was the main reason for better reduction even at low microwave power as well as time. It can be easily inferred from Fig. 4 that the temperature for O1, when it was roasted along with coal, almost reached 800 °C at a microwave power of 5 kW and time of 20 min. In contrast, the poor heating characteristic of O2 led to a maximum temperature of about 700 °C when it was roasted with coal. Therefore, sufficient conversion of the iron oxides, such as the goethite present in O2, to magnetite couldn't occur, and poor iron grade and recovery resulted under these conditions. However, at a high microwave power of 7 kW, the ore O2 showed better results since the temperature reached around 850 °C after 25 min of microwave exposure. Meanwhile, the temperature for O1 crossed around 900 °C after 15 min of roasting with coal, and thus, facilitated the formation of phases like wustite and iron metal. A detailed study of the reduction sequence and formation of different phases, using several characterization techniques, was therefore undertaken and being presented in the following section.

3.4 Characterization of the roasted samples

The characterization studies were done for the samples O1 and O2, roasted at two sets of conditions, are given in Table 5 along with the corresponding Fe grade and Fe recovery obtained as a result of the LIMS of the respective roasted mass. For the studies of optical microscopy, EPMA and VSM, the roasted mass was used as the test sample while magnetic and non-magnetic samples separated through LIMS were subjected to XRD analysis.

Table 5 Experimental conditions to generate roasted samples for characterization studies with the corresponding results of LIMS in terms of Fe grade and recovery (Coal to ore ratio is fixed at 0.2)

	Experimental conditions		**Grade, %**	**Recovery, %**
	Power, kW	**Time, min**		
O1+Coal	5	15	62.57	60.01
	7	25	67.9	44.34
O2+Coal	5	15	59.02	16.86
	7	25	64.4	33.3

The relative XRD patterns of the magnetic and non-magnetic fractions are shown in Fig. 5. The magnetic fraction of O1 roasted at MW conditions of power: 5 kW, time: 15 min and coal/ore: 0.2 shows magnetite as the significant phase along with a little amount of wustite and metallic iron (Fig. 5a). The corresponding non-magnetic fraction primarily contains ilmenite and quartz with traces of wustite (Fig. 5b). It suggests that the hematite present in the feed O1 has converted to magnetite, whereas a part of the magnetite already existing in the feed has reduced to wustite and metallic iron. Even though wustite is feebly magnetic, it was reported in the magnetic fraction owing to its possible close association with magnetite. The

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presence of metallic iron and wustite in the product indicates the corresponding reducing conditions to be more than sufficient for the magnetization of the hematite phases present in the feed O1. The Fe grade and Fe recovery values of 62.57% and 60.01%, respectively, obtained from O1 at these conditions validate the findings from XRD.

On the other hand, the magnetic fraction obtained from sample O2 roasted at the same conditions shows the presence of hematite and maghemite (Fig. 5a). It is to be noted that maghemite is an intermediate phase formed between hematite to magnetite conversion, and its presence in the product infers that the roasting conditions were not adequate for producing magnetite. This is evident from a Fe grade of 59.02% and a poor Fe recovery of 16.86% for the magnetic product obtained from the MW roasting-magnetic separation of O2 at these conditions. Moreover, the presence of hematite, goethite and kaolinite in the corresponding non-magnetic fraction (Fig. 5b) suggests the conditions were not appropriate even for the complete dehydration of the phases. In the XRD pattern of the magnetic fraction of O1 roasted at conditions such as power: 7 kW, time: 25 min and coal/ore: 0.2 (Fig. 5c), metallic iron appears to be the major phase along with magnetite. It is in tune with the relevant magnetic product with a high Fe grade of 67.9% and 44.34% Fe recovery obtained at these conditions. The magnetic fraction also contains a minor amount of corundum (Al_2O_3), which forms when kaolinite fragments at high temperatures. The XRD of the concerned non-magnetic fraction, as shown in Fig. 5d shows similar peaks as Fig. 5b with hercynite ($FeAl_2O_4$) being an addition. Hercynite is a phase formed as a result of the reaction between iron oxide and kaolinite at the higher reducing conditions. The XRD of the magnetic product of O2 roasted at the same condition mostly includes metallic iron, magnetite along with wustite and fayalite (Fe_2SiO_4). This magnetic product has a grade of 64.4% Fe with 33.3% Fe recovery, which is higher than the previous conditions (power: 5 kW, time: 15 min and coal/ore: 0.2). Though fayalite is paramagnetic, its presence in the magnetic part is possibly due to its intimate association in the ferromagnetic rich area. Quartz, wustite, hercynite, and fayalite are found to be present in the non-magnetic portion of the ore O2 (Fig. 5d). The XRD studies give a picture of the formation of several magnetic and non-magnetic minerals at two different sets of MW roasting conditions, thereby validating the corresponding Fe grade and recovery of the products obtained.

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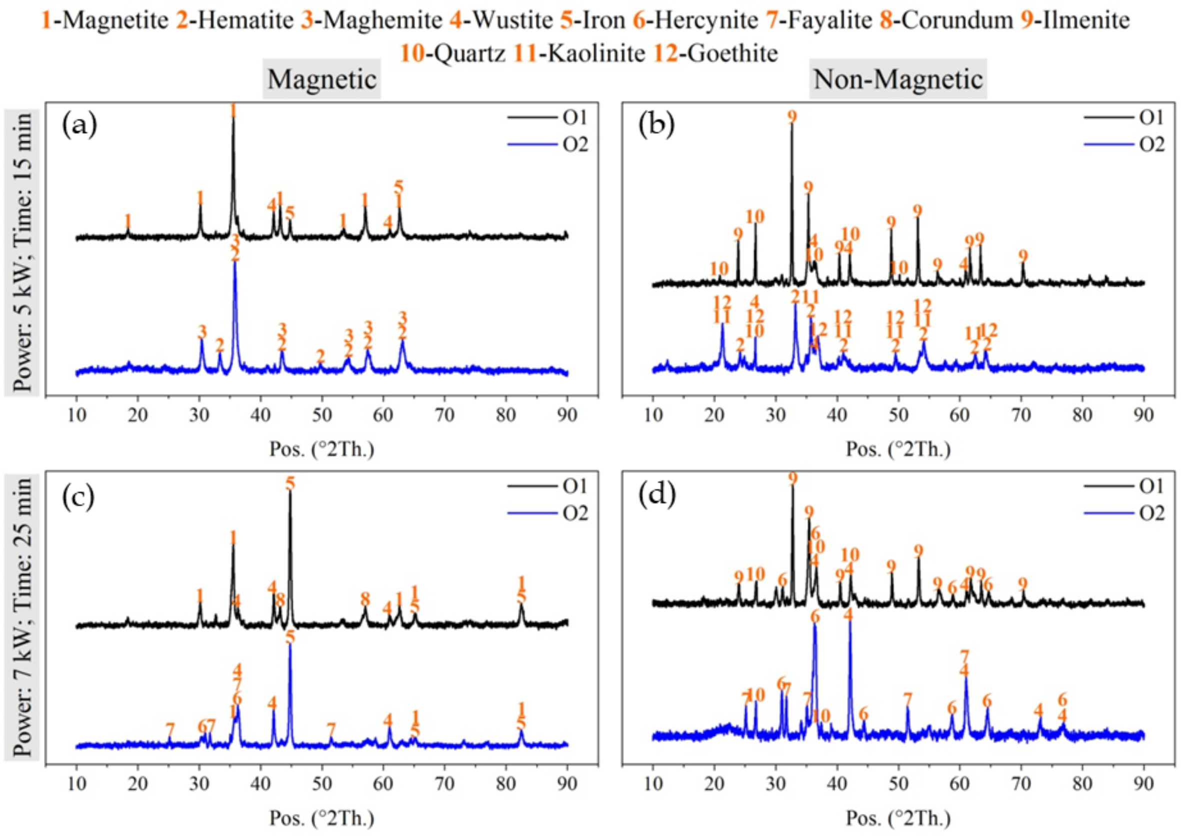

Fig. 5 XRD patterns of magnetic and non-magnetic concentrates obtained from the reduction roasting of O1 and O2 operated at conditions (a and b) Power: 5 kW, Time: 15 min (c and d) Power: 7 kW, Time: 25 min

To further validate the formation of the phases as well as understand their association with each other, reflected light microscopic studies of the roasted products obtained at the same conditions were undertaken, and the corresponding micrographs are displayed in Fig. 6.

In sample O1, most of the grains are partially or wholly martitized mainly along the grain boundaries and the weak planes of ilmenite. Magnetite shows pitted appearance having a lot of silicate inclusions (Fig. 6a). Perfect euhedral to subhedral crystals of ilmenite exhibiting mutual boundary texture with magnetite is noticed. With the increase in power and time, the concentration of iron-rich phase is increased to form wustite or metallic iron (Fig. 6c) along with the formation of iron silicates both in the contact boundaries and the fracture planes of ilmenite and metallic phase as well as within the open spaces between metallic phases.

In sample O2, at 5 kW power with 15 min, mostly massive goethite phases are seen together with the needle-shaped fine-grained hematite with some clay patches in between (Fig. 6b). With the increase in power and time, the hematite and goethite are all found to be converted to magnetite, and some silicates (Fe/Al-silicates) are observed as the fillings (Fig. 6d). The

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presence of the metallic phase is almost negligible when compared to O1 roasted at the same conditions.

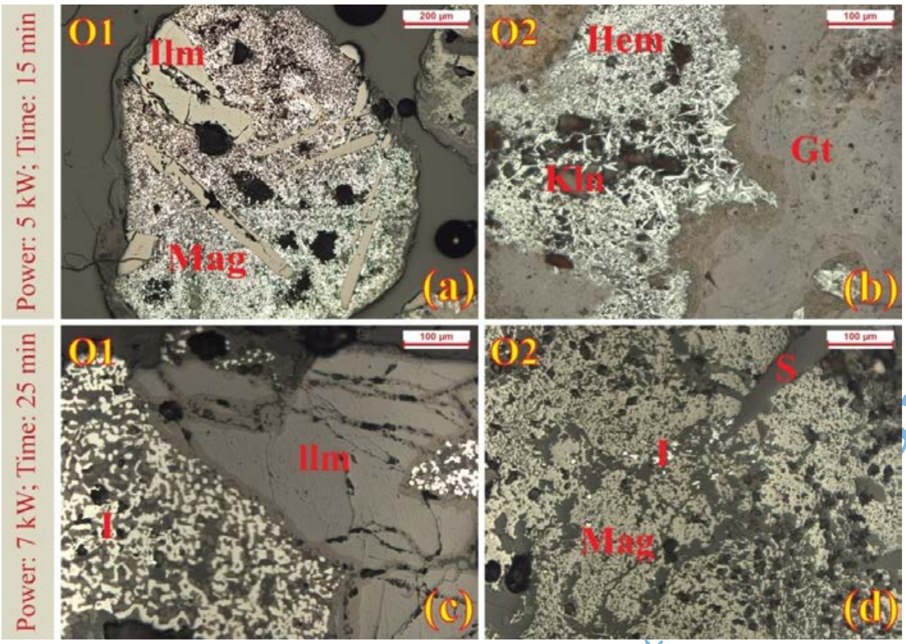

Fig. 6 Mineral phases identified by optical microscopy for the reduced iron ore sample of both O1 (a and c) and O2 (b and d) at two different reducing conditions. (Gt: Goethite, Kln: Kaolinite, H: Hematite, I: Ilmenite, S: Silicatesand Mag: Magnetite)

The elemental distribution maps of the feed O1 and O2 accompanied by their roasted products are illustrated in Figs. 7 and 8, respectively. The Back Scattered Electron (BSE) image with Fe, Ti, Al-mapping of O1 sample explains the presence of some Al as impurities dispersed within the iron phase. Ti and Fe-bearing phases are uniformly distributed among themselves, which typically represent the ilmenite phase (Fig. 7a). However, the electron mapping of the roasted product shown in Fig. 7b explains the dispersion of iron particles in the groundmass containing laths of ilmenite. Some Al-minerals are also found to be adsorbed within the iron phases. With the increase of time and power, the formation of metallic iron is clear from the Fe-map of Fig. 7c.

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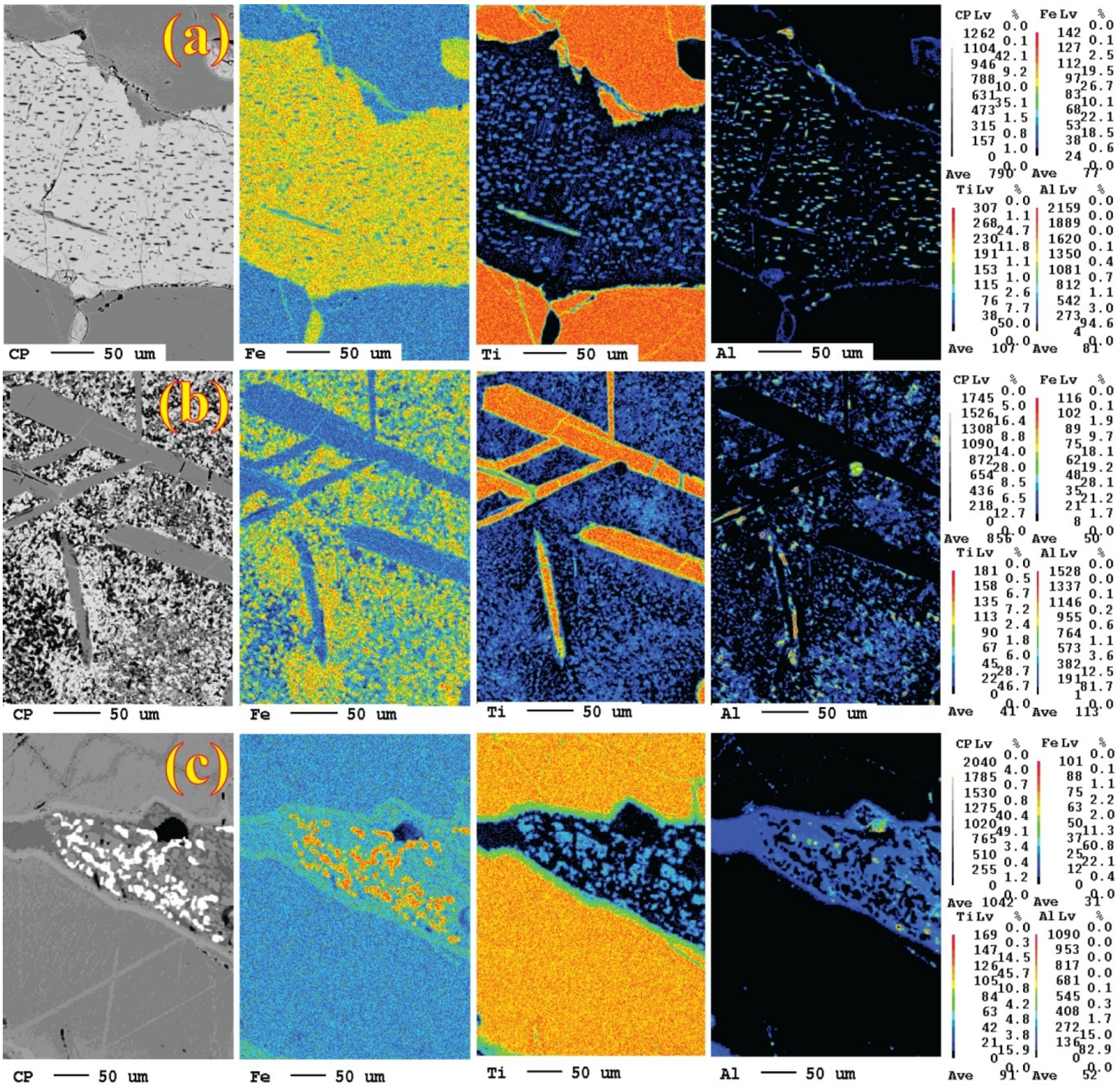

Fig. 7 EPMA based elemental mapping of ore sample O1 (a) feed ore and roasted at MW conditions such as (b) 5 kW and 15 min (c) 7 kW and 25 min with coal/ore of 0.2

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The elemental mapping of the O2 sample describes the concentration of Fe, Al and Si present in the feed as well as the roasted products. Fig. 8a shows a typical colloidal band of goethite, which mostly comprises Fe. The presence of Al-phase within the goethite band is an indication of the close association of kaolinite with goethite. At conditions such as 5 kW power and 15 min time, dehydration starts with the formation of iron oxides and Al-silicate as marked in the sample (Fig. 8b). The formation of iron-rich phases like magnetite and metallic iron/wustite is marked in the sample roasted at 7 kW power and 25 min period (Fig. 8c). The red dots in the Fe-map of Fig. 8c confirms the little presence of the metallic phase in this condition. On comparing the elemental mapping of the roasted products of O1 and O2, it is evident that O2 has less metallic Fe and more Al-silicate phases embedded in the magnetite matrix explaining why the magnetic separation in the case of O2 could not yield a high iron recovery like O1.

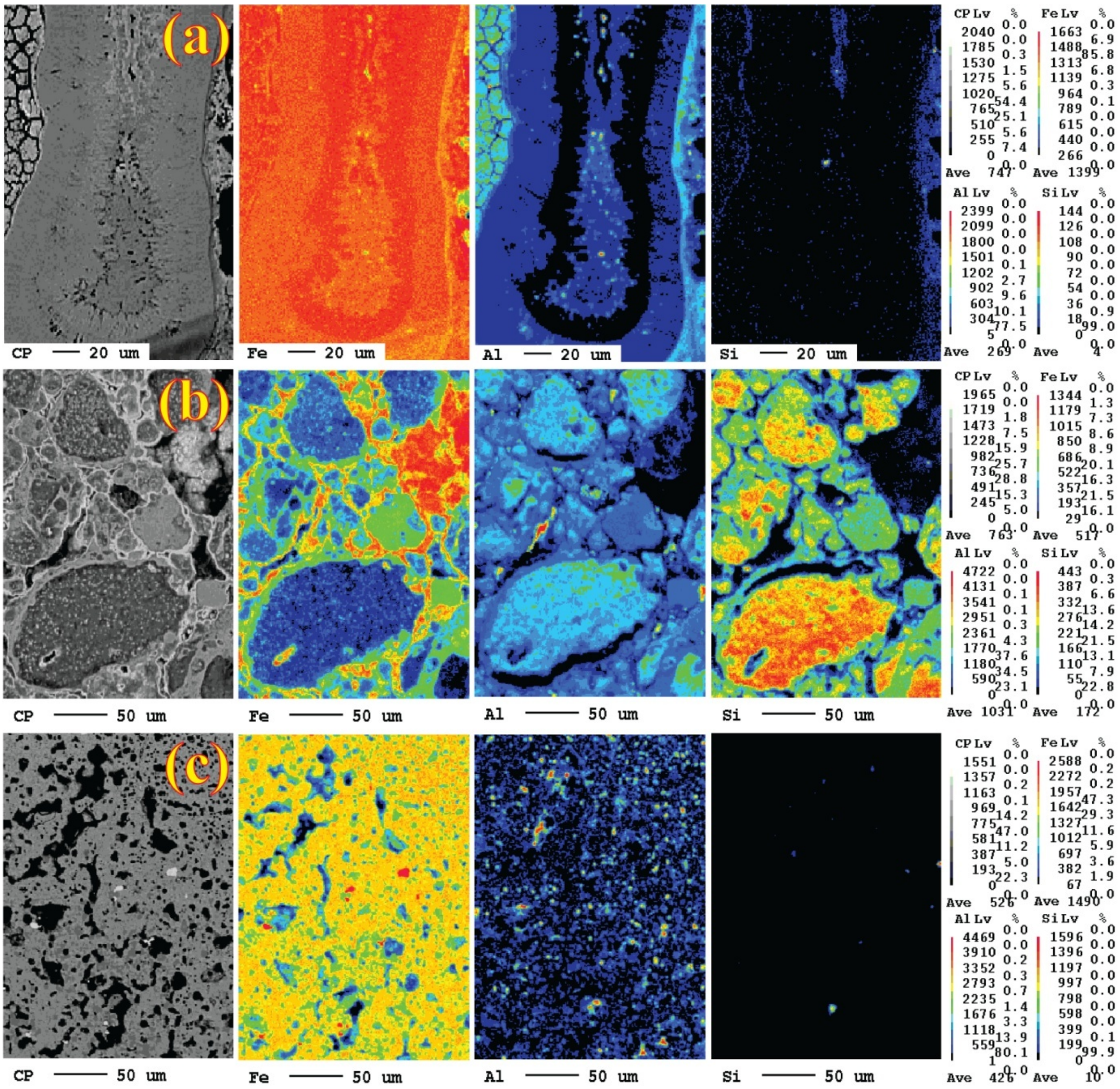

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Fig. 8 EPMA based elemental mapping of ore sample O2 (a) feed ore and roasted at MW conditions such as (b) 5 kW and 15 min (c) 7 kW and 25 min with coal/ore of 0.2

3.5 Magnetic properties of roasted samples

The Saturation Magnetization (SM) is considered to be an essential parameter that determines to which extent a substance can respond to magnetic separation. Therefore, the O1 and O2 samples were subjected to VSM to understand the effect of microwave heating with and without the reductant. Fig. 9 shows the SM values of the ores treated at different MW exposure periods from 10 to 25 min at a fixed power of 5 kW. As the sample O1 has a majority of its iron oxides in the form of magnetite, the SM value was found to be 27.34

emu/g. When treated in MW without any reductant, there is a gradual decrease in the saturation magnetization value to 21.66 emu/g (Fig. 9a). This is mainly attributed to the partial oxidation of the magnetite present in the ore to hematite ($2Fe_3O_4 + O_2 \rightarrow 3Fe_2O_3$), which has less magnetic susceptibility compared to the former. In contrast, when O2 was MW-treated at four different time intervals, the SM is found to be increased to a maximum of 5.31 emu/g. This increase in magnetization values is basically due to the thermal decomposition of goethite into hematite ($2FeOOH \rightarrow Fe_2O_3 + H_2O$), which is considered as more magnetic in comparison to the former.

The SM values of the products roasted using coal, as a function of time is depicted in Fig. 9b. It is seen that the addition of coal has enhanced the SM of both the roasted samples. Understandably, the improved values were mostly due to the phase transformation of the feeble magnetic minerals into higher magnetic susceptible products as already confirmed through the XRD and microscopic studies. In the case of O1, the saturation magnetism is found to be slightly increasing, resulting in up to a maximum of about 31.7 emu/g for a period of 25 min. On the contrary, the goethite-rich O2 ore could witness a maximum saturation magnetism of 22.94 emu/g even at a period of 25 min. These results further confirm that the ore O2 needs higher MW exposure time to attend sufficient magnetization for being separated into a useful magnetic concentrate.

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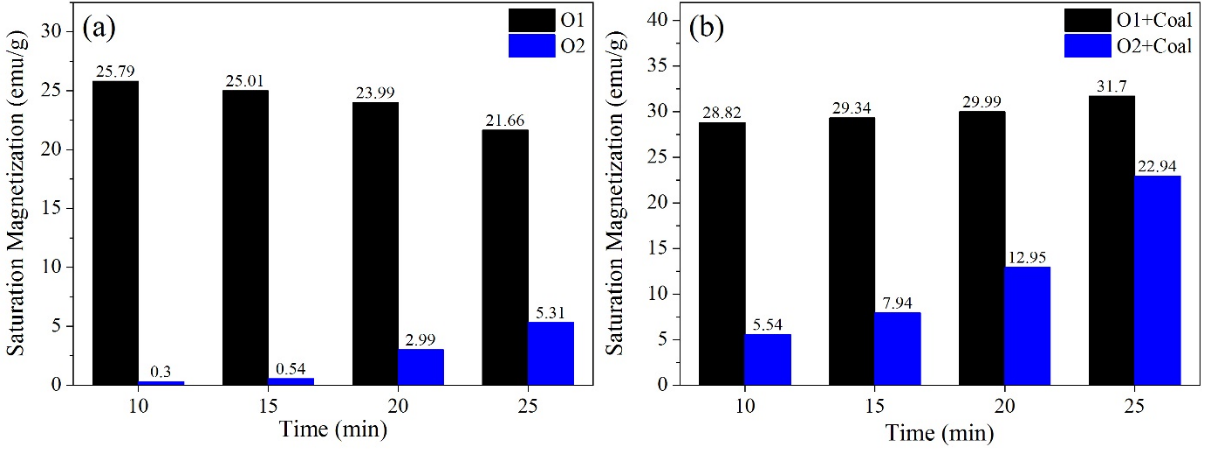

Fig. 9 Effect of MW exposure on saturation magnetisation of (a) feed ore O1 and O2 (b) feed ore O1 and O2 with 0.2 coal/ore at a power of 5 kW

3.6 Microwave-based reduction roasting of ore mixtures

The results discussed so far suggest that using MW-assisted reduction roasting followed by magnetic separation, it is relatively easier to obtain a suitable iron ore concentrate from O1 in comparison to O2. The primary reason is the presence of magnetite and ilmenite in the former, which are MW absorbing minerals [31]. In the case of O2, where goethite (α-FeOOH) is mostly present, the MW heating leads to its decomposition to hematite (α-Fe_2O_3) and water. Therefore, during the initial stage of thermal transformation, the real and imaginary permittivity values are very low, but with an increasing temperature, the rate of water removal increases as the flux of migrating hydroxyl unit increases, resulting in the improvement of microwave absorbing characteristics of the ore [32-34]. It explains why the MW-assisted heating or reduction in the case of the goethite-rich O2 is lesser compared to O1.

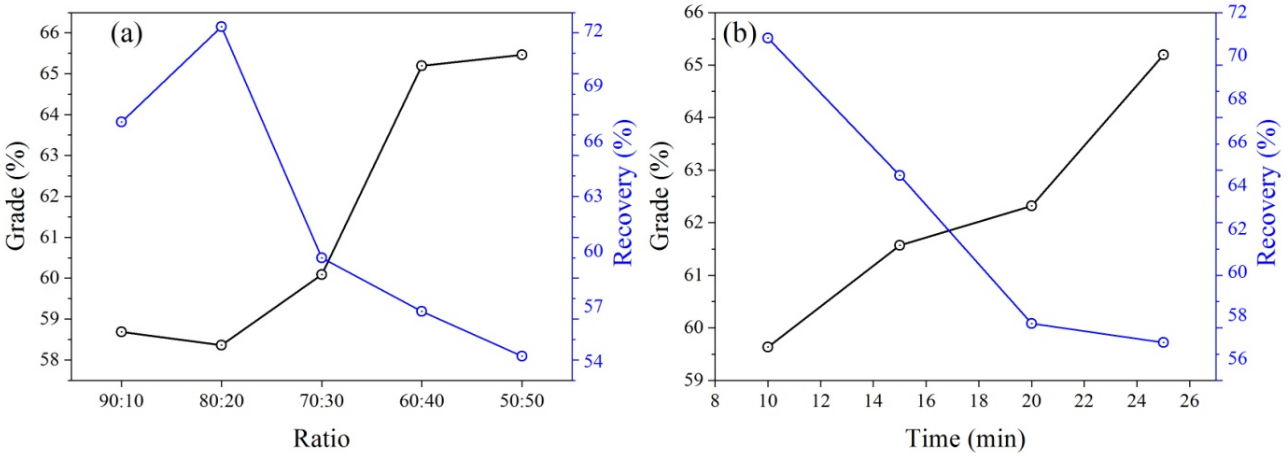

Fig. 10 Variation of grade and recovery in (a) different ratios of O2 and O1 (O2:O1) and (b) different level of time with a constant mixture of 60:40

Further attempts were made to blend these samples and subject to MW roasting followed by magnetic separation. Fig. 10a shows the response of the ore-mixtures in terms of Fe grade and recovery, which were conducted at a power of 5 kW with coal/ore of 0.2 for a time period of 25 min. As can be seen in Fig. 10a, the iron grade decreases and the recovery rises with an increasing amount of O2 in the mixture. In consideration with both iron grade and recovery, the mixture of 60:40 (O2: O1) corresponding to a grade of 65.19% Fe at a total Fe recovery of 56.75% was chosen as the optimum level. Furthermore, this ore mixture was subjected to different microwave exposure times at 5 kW power. The impact of time on the iron grade and recovery is shown in Fig. 10b. It is observed that at an exposure time of 15 min, a magnetite concentrate of 61.57% Fe with 64.47% iron recovery could be obtained. However, on increasing the time beyond 15 min, the recovery drops even though the Fe grade rises. Therefore, an exposure time of 15 min at 5 kW microwave power and a ratio of 60:40

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were found to be ideal. Overall, it can be inferred that O2 ore, which could be enriched to a maximum of 64.4% Fe with only 33.3% iron recovery at a power of 7 kW can now be upgraded to a higher recovery of 64.47% with a slight drop in grade to 61.57% Fe by blending with O1.

To summarise, the grade versus recovery plot for ores O1, O2 and ore mixture at different microwave process parameters is shown in Fig. 11. The plot reviews the grade-recovery results listed in Table 5 and that obtained with the optimum ratio of O1 and O2. The ore O1 gives a high Fe grade along with a low Fe recovery at a microwave power of 7 kW and a period of 25 min. At same conditions, O2 shows a lower grade as well as recovery. At a microwave power of 5 kW and a period of 15 min, the ore O1, in comparison to O2, displays a better response in terms of grade and recovery. Nevertheless, it is essential to note that the highest recovery of 64.47% with 61.57% Fe could only be achieved with the 60:40 (O2:O1) ore mixture at 5 kW and 15 min. It is also to be noted that, at these MW conditions, the energy consumption was 1.2 kWh. The grade-recovery plot hints that, while dealing with goethite–rich ores like O2 in microwave-aided reduction roasting process, it is always advisable to blend them with magnetite-rich ores (such as O1 in this case) to obtain the perfect combination of iron grade and recovery.

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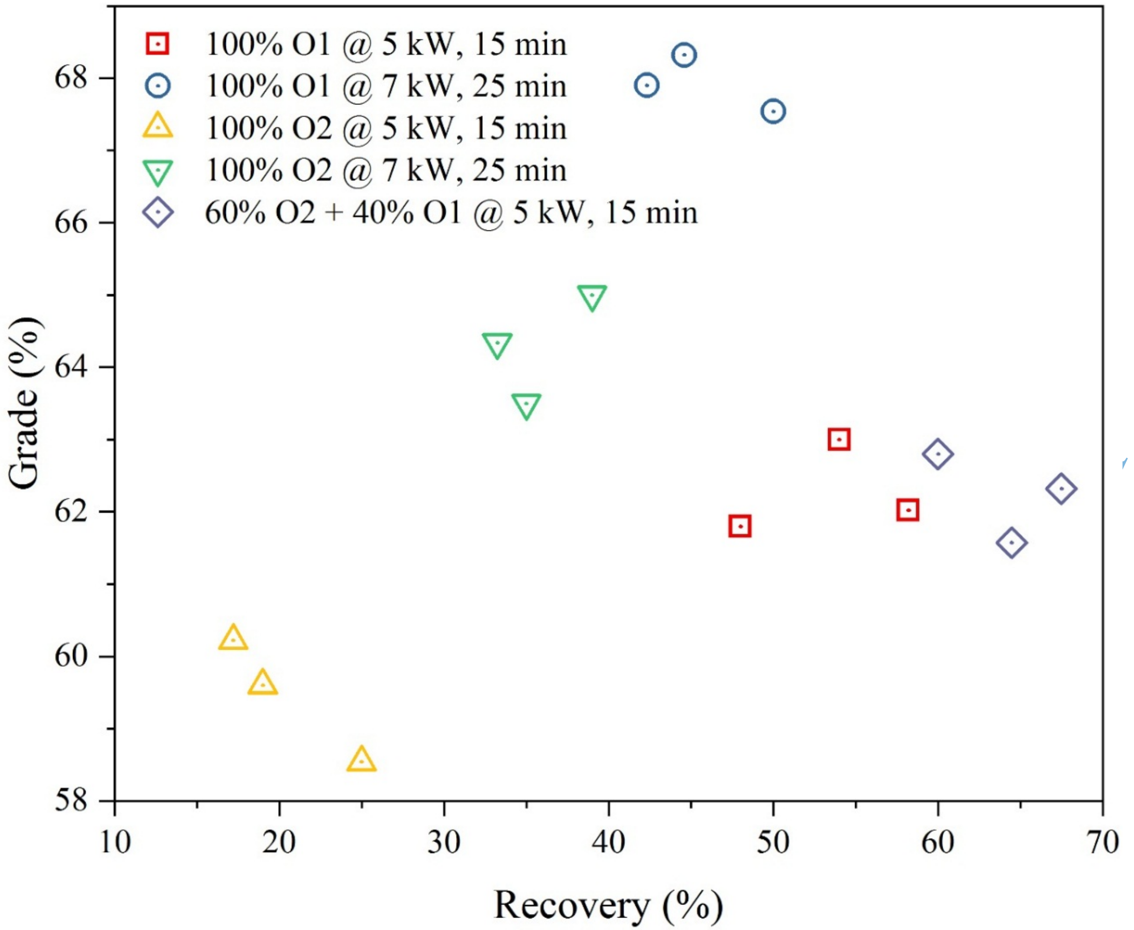

Fig. 11 Grade-Recovery plot for O1 and O2 at different MW reducing conditions

4. Conclusion

The up-gradation in the iron content of two low-grade iron ores was attempted via microwave-assisted reduction roasting followed by magnetic separation process. The research work addressed the issues and complications related to the development of a MW process for iron ores with mineralogical variations. The key excerpts of the work are listed below.

i. The titano-magnetite ore was upgraded to 62.57% Fe at a power of 5 kW, coal/ore ratio of 0.2 and time of 15 min in contrast to the goethite ore which yielded a recovery of 16.8% at the same conditions. This indicated that mineralogy plays a crucial role in MW heating.

ii. The VSM studies revealed that it is challenging to magnetize the goethite-rich sample as it took around 25 min at 7 kW MW power to reach an SM value of 22.94 emu/g, which was even lesser than the SM value of the untreated titano-magnetite ore.

iii. The blend of the two ores prepared by mixing 60% of the goethite rich ore with 40% of the titano-magnetite ore could be upgraded to 61.57% Fe at a MW power of 5 kW and time of 15 min with coal to ore ratio of 0.2. This inferred that the blending of a low-grade magnetite rich ore could improve the results of the MW-assisted magnetizing roasting process of low-grade ores that do not absorb microwave very well.

iv. Reduction roasting followed by magnetic separation is one of the most effective way to upgrade the iron content of poorly liberated ores. In this regard, the present work can be considered as a preliminary endeavour to introduce microwave energy in the reduction-roasting of such ores. It would take more efforts to establish the process on a bigger scale and calculate the energy consumption, and viability for commercial exploitation.

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