1. Introduction

Blast furnace or shaft furnace feed should form a permeable bed of material, permitting gas flow through it uniformly at a high rate. Iron ore concentrates in the form of fines are not suitable because, fines tend to pack into a non-permeable bed and also they are likely to be carried away as dust by the high gas flow rates. The fine ore/concentrate must therefore be agglomerated into larger particles that will improve permeability of the furnace burden, increase the rate of reduction, and reduce the amount of material blown out of the furnace as dust. Pelletizing is one of the most widely used agglomeration technique which requires preparation of green balls from fines and indurating them at higher temperatures to get the required properties. Induration of green pellets involves four different thermal treatments viz., drying, preheating, heating and cooling. In “straight-grate” machines all these four stages are completed on a travelling grate, while in “Grate-kiln” machines, drying and preheating takes place on grate, the firing process is performed in a rotary kiln before the pellets are transferred into an annular cooler. The temperature in the pelletizing process is as high as 1300–1320°C, so that a significant amount of liquid slag forms in the pellets. The main components of the slag phases are SiO$_2$, FeO, Al$_2$O$_3$, CaO and MgO in various proportions. The slag phase or silicate melt wets the solid surface and facilitates the diffusion and grain growth. After cooling, melt phase solidifies and acts as an important bonding phase in the finished pellets and influence high temperature properties, especially reduction degradation index (RDI) to a great extent.

The resistance of pellets against degradation during reduction depends on the type of bonding and increases in the order of hematite, ferrite and silicate. In this study commercial pellets of different RDI ranging from 8.5 to 14.3 were characterized. Electron and optical micro structural studies with image analysis revealed that the amount and distribution of silicate melt, alumina content of hematite phase, porosity and pore density are vital in controlling the RDI. Residual magnetite in the core of the pellets was found detrimental to RDI. Distribution of different elements in the oxide and melt phase was identified by using X-ray mapping technique and chemistry of different phases was measured using SEM-EDS analysis.

KEY WORDS: iron ore pelletizing; reduction degradation index; silicate melt; pore density; bonding phase; pellet induration.
degradation Index (RDI). Pellets of different RDI ranging from 8.5 to 14.3 were collected from a commercial iron ore pellet plant. The amount and distribution of different phases and microstructure were studied using optical microscopy and image analysis. Electron microscopy with EDS and X-ray mapping technique was used to record the chemistry and distribution of different phases.

2. Pelletizing Process

Production of iron oxide pellets from iron ore fines involves different operations like drying of ore fines to remove the moisture and grinding to get the required fineness. After mixing these ground ore fines with other additives like bentonite, limestone and coke breeze, green pellets are prepared using pelletizing discs. These green pellets are fired in straight-grate indurating machine to get the required physical, mechanical and metallurgical properties making them suitable feed to iron making units. Pellets are transported through the furnace by a traveling grate, which retains the pellets while allowing the air to flow through. The traveling grate is loaded with approximately 0.05 m height of fired pellets as hearth layer to protect the grate and then 0.50 m of green pellets. The induration process consists of three main steps; 1) drying for green pellets, 2) firing of pellets at 1250–1300°C to sinter the iron oxide particles, 3) cooling of hot pellets before discharging them on to the conveyors leading to the stockpiles. Figure 1 shows a simplified flow sheet of straight-grate indurating machine. It consists of seven different zones, viz., up-draft drying (UDD), down-draft drying (DDD), preheating (PH), firing (FZ), after-firing (AFZ), primary cooling (CZ1) and secondary cooling zone (CZ2). Process air is circulated through these different zones with the help of five interconnected process fans.

3. Experimental

Pellets with different RDI were collected from different shifts. The induration process parameters and pellet chemistry during the sample collecting shift were given in Table 1. Representative samples from the sampling lot of the respective shift were tested in the laboratory for chemical, physical, metallurgical and microstructural properties. Cracked and unfired pellets were removed from the sample lot to avoid erratic results.

3.1. Physical and Metallurgical Testing of Pellets

Tumbler and Abrasion Index of pellets were tested as per ISO 3271 where as Cold Crushing Strength (CCS) was tested as per ISO 4700. Reduction degradation index (RDI) test procedure used in the present study is a combination of ISO 4696 (LTBT Test) and ISO 7992 (Reduction under load). Figure 2 shows a schematic diagram of experimental apparatus of RDI test with the description of test procedure. Pellets with more irregular and non-uniform shape were avoided for the microstructure study as they experience uneven heat treatment across their cross section.

3.2. Image Analysis of Microstructures

Image analysis is a technique that is used to provide an objective measurement of different phases in microstructure. Pellet samples were cut into half and hot mounted at 175°C temperature and 90 daN load for 14 min using a con-

![Fig. 1. Schematic diagram of induration process of iron ore pelletizing.](image-url)
ductive resin. Once sample has been mounted and polished, it is then placed under a microscope for examination. A black and white CCD digital camera with a maximum resolution of 756×581 pixels is mounted behind the lens of the microscope to capture the light reflected from the sample. All the images were captured at 200× magnification for the current study. At this level of magnification, the view frame on the sample surface is approximately 0.7×0.5 mm.

The signal from the camera was provided to a personal computer through a gain correction amplifier to correct the signal for optimal display. Computer software was used for interpreting the camera signal into digital image. Basically, the digital image captured from the black and white camera is represented by pixels having 256 shades of grey values (i.e. 0 to 255). The lower range of grey values represents pores and the oxide grains represent higher range values. A digitized black and white photograph is transformed into a segmented image with the specified range of grey values to different phases. Once the image has been processed, the various tools of image analyzer were used to measure the area fraction, perimeter and number of counts of different phases in the microstructure.

4. Results
4.1. Physical and Metallurgical Properties of Different RDI Pellets

Figure 3(a) shows the variation of Al$_2$O$_3$/SiO$_2$ ratio with RDI Test Conditions:

- Sample Weight: 2 Kg
- Sample Size: 12.5-16 mm & 10-12.5 mm (60% each)
- Test Temp.: 800 deg C
- Gas Flow rate: 800 L/h
- Gas Composition:
  - Gas 1: 50% CO, 35% CO & 15% H2
  - Gas 2: Change over of Gas 1 to Gas 3 at an interval of 5 minutes
  - Gas 3: 5% CO, 70% CO & 25% H2
- Gas Composition: 100% N2 (Preheating & Cooling)
- Test Time: 360 minutes.

**Fig. 2.** Schematic diagram of experimental apparatus of RDI test.

**Fig. 3.** (a) alumina/silica ratio, (b) Tumbler Index and Abrasion Index, (c) cold crushing strength and (d) reducibility of different RDI pellets.
different RDI pellets. Higher ratio in pellets makes the melt phase viscous due to the refractory nature of Al₂O₃. Pellets with high Al₂O₃/SiO₂ ratio exhibited poor RDI behavior. Figure 3(b) shows the tumbler and abrasion index of different RDI pellets. Tumbler index decreased with increased RDI of pellet, whereas abrasion index increased with increased RDI. Cold crushing strength (CCS) of pellets was found to decrease with increased RDI but is still within required specification limit of ~200 kg/pellet as shown in Fig. 3(c). Reducibility of pellets found to increase slightly with increased RDI, as shown in Fig. 3(d).

4.2. Quantitative Measures of Microstructure through Image Analysis

For image analysis studies, three pellet samples were used and 12 images were processed for each test condition. Optical microstructures of 8.5/4.1 RDI, 10.1/5.8 RDI and 14.3/9.2 RDI pellets are shown in Figs. 4, 5 and 6 respectively. In each pellet sample, four different regions viz., shell, outer mantle, inner mantle and core (see Fig. 7(f)).
were selected to represent the whole pellet cross section.

4.2.1. Porosity and Pore Density

Low RDI (8.5/4.1) pellets have less porosity compared to high RDI (14.3/9.2) pellets. In low RDI pellets, pore size is small whereas high RDI pellets have bigger pore size. Pore density is an indication of pore size and distribution. Figure 7(a) shows the porosity and pore density (number of pores per unit area) of different RDI pellets. With increasing RDI, porosity increased, but pore density decreased. High porosity and low pore density exhibited high RDI values in pellets.

4.2.2. Silicate Melt

Amount of silicate melt is found to be higher in low RDI pellets. Silicate melt density, which is the indication of its distribution, is calculated as number of silicate melt counts per unit area. With decreased silicate melt distribution, pel-

Fig. 6. Zonewise microstructure of 14.3/9.2 RDI pellets (a) shell, (b) outer mantle, (c) inner mantle and (d) core.

Fig. 7. Quantitative distribution of different phases with respect to pellet RDI using image analysis.
Fig. 8. SEM images of 8.5/4.1 RDI pellets with EDS analysis (a) shell and (b) core.

| Component | Na₂O | MgO | Al₂O₃ | SiO₂ | P₂O₅ | CaO | TiO₂ | Fe₂O₃ |
|-----------|------|-----|-------|------|------|-----|------|-------|
| Shell     | -    | -   | 0.59  | 0.89 | -    | 0.15| -    | 98.38 |
| Core      | 0.34 | 0.76| -     | -    | -    | 0.28| -    | 98.62 |

Silicate melts:

| Component | Na₂O | MgO | Al₂O₃ | SiO₂ | K₂O | CaO | TiO₂ | FeO |
|-----------|------|-----|-------|------|-----|-----|------|-----|
| Shell     | 0.26 | 2.44| 8.55  | 45.51| 0.24| 32.54| -    | 10.46|
| Core      | 0.33 | -   | 8.66  | 45.28| 0.36| 28.46| 0.50 | 16.42|

* Analysis is average of the points mentioned.

Fig. 9. SEM images of 10.1/5.8 RDI pellets with EDS analysis (a) shell and (b) core.

| Component | Na₂O | MgO | Al₂O₃ | SiO₂ | P₂O₅ | CaO | TiO₂ | Fe₂O₃ |
|-----------|------|-----|-------|------|------|-----|------|-------|
| Shell     | -    | -   | 0.57  | 0.78 | -    | 0.19| -    | 98.45 |
| Core      | -    | -   | 1.00  | -    | -    | -   | -    | 99.00 |

Silicate melts:

| Component | Na₂O | MgO | Al₂O₃ | SiO₂ | K₂O | CaO | TiO₂ | FeO |
|-----------|------|-----|-------|------|-----|-----|------|-----|
| Shell     | 0.39 | 4.55| 7.70  | 37.03| -   | 22.33| -    | 27.98|
| Core      | 2.80 | 6.47| 32.37 | 21.63| -   | -   | -    | 36.72|

* Analysis is average of the points mentioned.

Fig. 10. SEM images of 14.3/9.2 RDI pellets with EDS analysis (a) shell and (b) core.

| Component | Na₂O | MgO | Al₂O₃ | SiO₂ | P₂O₅ | CaO | TiO₂ | Fe₂O₃ |
|-----------|------|-----|-------|------|------|-----|------|-------|
| Shell     | -    | -   | 0.57  | 0.87 | -    | -   | -    | 98.56 |
| Core      | -    | -   | 1.55  | -    | -    | -   | -    | 98.45 |

Silicate melts:

| Component | Na₂O | MgO | Al₂O₃ | SiO₂ | K₂O | CaO | TiO₂ | FeO |
|-----------|------|-----|-------|------|-----|-----|------|-----|
| Shell     | 0.76 | 2.30| 9.38  | 33.41| 0.59| 20.27| 0.35 | 32.93|
| Core      | 0.42 | 2.74| 9.59  | 41.74| 0.39| 28.58| -    | 16.54|

* Analysis is average of the points mentioned.
let RDI found to increase as indicated by silicate melt density in Fig. 7(b). Silicate melt perimeter, which is a good indicator of bonding interface between the oxide grains and bonding phase, was also measured using image analysis software. With increasing perimeter of the silicate melt, RDI of the pellet found to decrease as shown in Fig. 7(c).

4.2.3. Iron Oxide Phases (Hematite and Magnetite)

High RDI pellets exhibited low amount of hematite and high magnetite (see Figs. 7(d) and 7(e)). Hematite density (number of hematite grains/mm²) indicates the degree of recrystallisation. Higher hematite density means more number of unsintered individual hematite grains as a result of poor sintering conditions. Pellets with high RDI exhibited high hematite and magnetite density, i.e. poor sintering of hematite and magnetite.

4.3. SEM Study of Different RDI Pellets

Figures 8, 9 and 10 show the SEM images of 8.5/4.1 RDI, 10.1/5.8 and 14.3/9.2 RDI pellets respectively along with analysis of phases. Semi quantitative analysis of iron oxides and silicate melts in shell and core was carried out using SEM-EDS. Figure 11 shows the alumina content of hematite phase. X-ray mapping studies of the pellet samples were carried out to understand the distribution of different elements among oxide and melt phases as shown in Fig. 12.

Figure 11. Effect of Al₂O₃ in hematite phase on pellet RDI.

5. Discussion

Figure 3(a) above mentioned showed that pellets with high Al₂O₃/SiO₂ ratio exhibited high degradation during reduction. Alumina was reported to increase the viscosity of the primary melt formed during the sintering of iron ore. Studies by Saito et al.⁴ and Machida et al.⁵ revealed that alumina is very effective in increasing the viscosity of calcium ferrite slags particularly at temperatures below 1200°C. Loo et al.³ showed that increase in alumina in iron ore sinter increases the pore area and the pore shape becomes more irregular. Alumina, being dissolved in the melt phase, reduces its strength by increasing its brittleness. Pimenta et al. stated that alumina diffuses in to the hematite crystal during sintering process to form solid solution. During low temperature reduction, hematite, diffused with alumina, generates magnetite phase with distorted structure causing crack generation and propagation leading to disintegration.⁶ SEM-EDS analysis of different RDI pellets revealed that RDI increased with increased alumina content in the hematite phase, as shown in Fig. 11. Alumina content of hematite phase in the core of high RDI pellets is 1.55% against the 0.76% in low RDI pellets. This high alumina in the hematite would have resulted distorted magnetite phase, thereby leading to poor RDI.

5.1. Tumbler and Abrasion Index

Tumbler index, which is a measure of resistance to generate fines during handling and transportation, decreased with increased RDI of pellet, whereas abrasion index, which is a measure of dust generating tendency, increased with increased RDI. Amount of bonding phase (silicate melt) in the fired pellets plays a vital role in controlling their resistance to tumbling and abrasion. Low amount of silicate phase and its poor distribution, as shown in Fig. 7(b), in high RDI pellets can be attributed to poor tumble and abrasion resistance.

5.2. Pellet Strength

Cold crushing strength (CCS) is regarded as one of the principal quality criteria of fired pellets to assess their suitability as burden material for blast furnace or shaft furnace
of direct reduction process. Pellets with low strength cannot withstand the handling loads during their shipping and the load of burden in the reduction furnace. Lower CCS values of high RDI pellets can be attributed to their high porosity and low pore density, which is an indication of big interconnected pores, as shown in Fig. 7(a). Duplex structure (hematite shell and magnetite core) observed in the high RDI pellets can also be attributed to poor strength, as this kind of structure leads to concentric cracks due to the differential contraction of hematite shell and magnetite core.2–9

5.3. Reducibility

Reducibility of high RDI pellets found to be slightly higher compared to low RDI pellets. This could be attributed to the fact that high RDI pellets have more porosity and low pore density, Fig. 7(a), which is an indication of big interconnected pores thereby allowing the reducing gas easily across all the zones of pellets and hence improved reducibility.

5.4. Reduction Degradation Index (RDI)

Reduction degradation of pellets is an undesirable phenomenon that occurs at low temperatures during their reduction in blast furnace or shaft furnace of direct reduction process. The primary cause of low temperature disintegration is thought to be crystalline transformation from rhombohedral hematite to cubic magnetite.10) The anisotropic dimensional change due to the transformation leads to severe stresses in certain planes, resulting cracks in brittle matrix. The effect is particularly severe in the grain boundary. It is very clear that iron oxide in the indurated pellets is mainly in the form of hematite, therefore, generation of internal stress is in principle is unavoidable. The remedy for disintegration is essentially to increase the amount and better distribution of stable bonding phases, which are less brittle at lower temperatures. Bonding which forms during induration, in principle can be divided in to three main groups: Iron oxides bonds (hematite, magnetite), silicate bonds and local bonds (calcium ferrite, magnesioferrite) that are close to particular mineral phases. Hematite bonds are common and strong, but they are not stable during reduction. But when hematite bonds are reduced to magnetite, silicate bonds remain unaltered. They soften and melt later.11)

From the quantitative image analysis, it was observed that the amount and distribution of silicate melt, hematite and magnetite fraction is different in different RDI pellets as shown in Figs. 7(b) to 7(e). Low RDI values can be attributed to low amount and better distribution of silicate melt in these pellets, as shown in Fig. 7(b). Silicate melt interface found to effectively reduce pellet RDI. Better distribution of silicate melt observed in low RDI pellets can be attributed to high firing temperature and longer residence time (as indicated by slow machine speed) as shown in Table 1. High alumina content in the hematite crystal, as shown in Fig. 11, is also another reason for high RDI of pellets. During reduction, alumina enriched hematite results in distorted magnetite phase, causing crack generation and propagation leading to high reduction disintegration. High pore density in low RDI pellets can be attributed to high amount of silicate melt that wets the solid particles and facilitates the diffusion and grain growth. The liquid between the particles exerts pressure to pull them together due to interfacial forces leading to smaller pores and hence high pore density. Small rounded pores offer more resistance to crack propagation compared to long elongated pores.

To ensure proper silicate melt formation, firing temperature should be properly maintained during the pellet induration. As shown in Table 1, comparatively high firing temperature during the shift of low RDI pellets could be attributed high amount and better distribution of silicate melt in these pellets.

6. Conclusions

The resistance of pellets against degradation during reduction depends on the type of bonding and increases in the order of hematite, ferrite and silicate. The firing temperature during pellet induration is as high as 1 300–1 320°C, so that a significant amount of liquid slag forms in the pellets. After cooling, the slag/melt phase solidifies and acts as important bonding phase in the finished pellets and influence the reduction degradation index (RDI) to a great extent. In this study, pellets of different RDI values from a commercial pellet plant were tested for their mechanical and metallurgical properties. Amount and distribution of different phases were determined using quantitative image analysis and SEM-EDS. An attempt was made to correlate the pellet RDI to its phases and microstructure.

The following conclusions can be drawn from the present work:

1) Porosity and pore density, amount and distribution of melt phase and alumina content of hematite phase influences the RDI of pellets.
2) Lower RDI values are associated with low porosity and fine pores as indicated by high pore density.
3) Finely distributed silicate melt with more oxide-melt interface found to effectively reduce pellet RDI.
4) High amount of diffused alumina in hematite phase found to increase RDI of pellets.
5) Presence of high amount of residual magnetite is detrimental to RDI.
6) Small pores and homogeneously distributed silicate melt also improves pellet crushing strength and tumbler and abrasion resistance.

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