Influence of Pellet Size on Quality and Microstructure of Iron Ore Pellets

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During the induration of pellets, temperature and oxygen partial pressure influence the formation of different phases and microstructure. With increasing the pellet size, these thermo-chemical conditions vary across its cross section. The time difference between the oxidation of the pellet surface and the core increases with increasing pellet size, i.e. the oxidation takes place under different conditions resulting in different phases and microstructure. In this study commercial pellets of different sizes (8–16 mm) were studied for their physical, mechanical and metallurgical properties. Pellets in the size range of 10–12.5 mm showed superior properties. Electron and optical micro structural studies with image analysis revealed that hematite content, pore density, amount and distribution of silicate melt of different sized pellets are different and distinctly affect the pellet quality. SEM-EDX analysis and X-ray mapping techniques revealed that Mg has a specific distribution pattern across the cross section of the pellet.

KEY WORDS: iron ore pelletisation; pore density; silicate melt; iron oxides; pellet size; FeO; MgO.

1. Introduction

The feed to a blast furnace or shaft furnace should form a permeable bed of material, permitting gas flow through it uniformly at a high rate. Powdered iron ore concentrates are not suitable in their as-produced form 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 powdered ore 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. The most common agglomeration technique is pelletisation, which requires preparation of green balls from powders and heat hardening 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 grate, where as 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 thermo-chemical conditions, e.g. the temperature, oxygen partial pressure and the amount of liquid silicate melt, change markedly during the induration process in both the pellet bed and a single pellet. As a result, the sintering intensity and the formation of slag phases differ greatly. The fired pellets often have inhomogeneous phases and structure through out their cross-section depending up on their size. The outer part of the pellet differs distinctly from the core. The time difference between the oxidation of the surface of the pellet and the core increases with increasing pellet size, i.e. the oxidation takes place under different conditions. This leads to significant microstructural differences in different zones of a pellet. The physical and metallurgical properties of these zones differ markedly. Thus pellet size has a marked effect on the formation of different phases and microstructure. Bigger the pellet, more the tendency to form different structures in different zones. A concentric duplex or triplex macrostructure has been observed to develop in the pellets. The phase composition and the micro- and macro structure determine the quality of the product pellets.

There are number of induration models for straight-grate machines and for grate-kiln machines. More recent advances in modeling of straight-grate pellet induration have been made by COREM which includes prediction of pellet metallurgical properties from green ball composition and thermal history. All these models assume that for given ore chemistry, green pellet properties and thermal history, quality of fired pellets is same. But as mentioned earlier, even keeping the above variables constant, pellet quality varies depending on their size distribution. The present study was undertaken to understand the distribution of phases and microstructure in different size pellets from a commercial set up and their influence on pellet quality. Pellets of different size ranging from 8 to 16 mm were tested for different physical, mechanical and metallurgical properties. 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. An attempt was made to correlate pellet quality to its microstructure.

2. Pelletisation Process

Pellets required for the present study were collected from a commercial straight-grate pellet plant of 3 Mt/year capacity. 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 pelleting 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

Around 1 t of fired pellets were collected from the product conveyor of the plant in one shift at an interval of 1 h. Sample collecting shift was decided in such a way that there was no production interruption in the preceding 3 shifts to ensure stable operating conditions. The induration process parameters and green pellet chemistry during the sample collecting shift were given in Table 1. The composite pellet sample was screened to get three different fractions: 8–10 mm, 10–12.5 mm and 12.5–16 mm. Cracked and unfired pellets were removed from the sample lot to avoid erratic results. Representative samples from all these fractions were tested in the laboratory for chemical, physical, metallurgical and micro structural properties.

3.1. Physical and Metallurgical Testing of Pellets

Tumbler and Abrasion Index were tested as per ISO 3271 whereas Cold Crushing Strength (CCS) was tested as per ISO 4700. Porosity of the fired pellets was tested as per ASTM E 32 test procedure. 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. For micro structural studies 3 pellets from each size fraction were collected representatively. 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 conductive resin. Once sample has been mounted and polished, it is then placed under a microscope (Zeiss-Axioplan 2) 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. A 10× eyepiece and 20× objective lens on the microscope has been selected 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. The computer software used for interpreting the camera signal into digital image was on marketing (Axiovision 4.7 Imaging System supplied by Carl Zeiss Vision). 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 analyser 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 Size Pellets

Figure 3 shows the variation of FeO content with different sized pellets. Higher FeO in pellets is as a result of insufficient oxidation during induration and is not desirable. FeO increased with increasing pellet size. Figure 4 shows the tumbler and abrasion index of different sized pellets. Tumbler index, which is a measure of resistance to generate fines handling and transportation, decreased with increasing pellet size, where as abrasion index, which is a measure of dust generating tendency, increased with increasing size. Cold crushing strength (CCS) of pellets in the size range of 8–12.5 mm is same but started decreasing beyond this
range, as shown in the Fig. 5. Reduction Degradation Index (RDI) of pellets indicates their tendency to generate fines (RDI $-6.3$ mm) and dust (RDI $-0.5$ mm) during reduction in the shaft furnace. RDI found to increase with increasing pellet size, as shown in Fig. 6.

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. Microstructures of 10 mm, 12 mm and 16 mm size pellets are shown in Figs. 7, 8 and 9 respectively. In each size fraction, four different regions viz., shell, outer mantle, inner mantle and core (see Fig. 15), were selected to represent the whole pellet cross section.

4.2.1. Porosity and Pore Density

In 10 mm pellets, pore size is small, and most of the pores are interconnected resulting more open porosity. In 12 mm pellets, pores are round in shape and pore assimilation is not yet taken place where as in 16 mm pellets all small pores are assimilated to form bigger pores. But these bigger pores are not round in shape, as in 12 mm pellets. Figure 10(a) shows the porosity and pore density (number of pores) of different size pellets. Porosity decreased with increasing pellet size, but pore density found to be higher in 10–12.5 mm pellets.

4.2.2. Silicate Melt

Amount of silicate melt is found to be higher in 10–12.5 mm pellets. Silicate melt density, which is the indication of its distribution, is calculated as number of silicate melt counts per unit area. Melt distribution decreased with increasing the pellet size as indicated by silicate melt density in Fig. 10(b). High pore density is associated with the presence of high amount of silicate melt as shown in Fig. 10(c). 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. Perimeter of melt in 8–10 mm and 10–12.5 mm size is almost equal and higher compared to 12.5–16 mm pellets (Fig. 10(d)).

4.2.3. Iron Oxide Phases (Hematite and Magnetite)

With increasing pellet size, amount of hematite de-
creased but magnetite increased (see Figs. 10(e) and 10(f)). Hematite density (number of hematite grains/mm$^2$) indicates the degree of recrystallisation. Higher hematite density means more number of unsintered individual hematite grains as a result of poor sintering conditions. With increasing size, hematite density found to be increased and magnetite density decreased.

4.3. SEM Study of 12 mm and 16 mm Size Pellets

Figures 11 and 12 show the SEM images of 12 mm and 16 mm pellets along with analysis of phases. Semi quantitative analysis of iron oxides and silicate melts in shell, outer mantle (OM), inner mantle (IM) and core was carried out using SEM-EDS. It was observed from the results, that MgO follows a specific distribution pattern among different zones of pellet. MgO content of the silicate melts is increasing from the core of the pellet to the shell, as shown in Fig. 13. This trend is observed in both 12 mm and 16 mm pellets. X-ray mapping studies of the pellet sample also confirmed this type of Mg distribution, as shown in Fig. 14.
5. Discussion

Figure 3 above mentioned showed that FeO content of the pellet increases with increasing its size. Studies by other workers Kotrmann et al. also confirmed this observation with the pellet imported to Germany from Canada, Brazil and Scandinavia. The origin of FeO in the pellets from the hematite pellet feed is the added coke breeze in the green pellets. Low oxygen partial pressure inside the bigger size pellet, especially in the inner mantle and core, during pellet firing creates reducing conditions and results in the formation of magnetite.

5.1. Pellet Strength

Figure 4 above mentioned showed the strength of pellets as a function of their size. The strength (CCS) dropped drastically with increasing size more than 12.5 mm. With increasing the pellet size, thermo-chemical conditions like temperature and oxygen partial pressure changes across its cross section. As a result, a duplex structure is formed across the cross section of pellet with hematite predominantly in the shell and outer mantle and unoxidised magnetite in the inner mantle and core as shown in the above Fig. 9. The duplex microstructure is distinctly visible in the broken or cut pellets with the naked eye also as shown in Fig. 15(a). Bond and lattice strains caused due to the differential contraction of these phases reduce the pellet strength in bigger size pellets. And also, increasing the size increases the propensity towards the large shrinkage gradients in the radial direction from pellet surface to the core and result in the defective pellet texture. A physical model proposed Wynnyckyz et al. (1974) about the formation of the concentric shells during sintering, under non-steady state temperature conditions is shown in Fig. 15(b) along with the micrograph of 14 mm size pellet with concentric cracks. Formation of these cracks as a result of differential shrinkage also reduces the pellet strength. Regarding the Tumbler and Abrasion Index of pellets also, similar arguments apply.

In addition to the above factors, distribution of pores could also influence the strength of pellets. Large pores with irregular shape and low pore density as observed in 16 mm size pellets (Fig. 9), could also be contributed to-
wards poor strength compared to small pores with nearly round shape and high pore density in 12 mm pellets (Fig. 8).

5.2. Reduction Degradation Index (RDI)

The reduction degradation of pellets is an undesirable phenomenon that occurs at low temperatures in the reduc-

![Fig. 11. SEM images of 12 mm size pellet with EDS analysis.](image)

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![Fig. 12. SEM images of 16 mm size pellet with EDS analysis.](image)

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![Fig. 13. Distribution of MgO in different zones of pellet.](image)

![Fig. 14. Distribution of Fe, Si and Mg in 16 mm size pellet (a) shell and (b) inner mantle.](image)
tion shaft of blast furnace or shaft furnace of any direct reduction unit. The primary cause of low temperature disintegration is thought to be crystalline transformation from rhombohedral hematite to cubic magnetite. 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 two main groups: Iron oxides bonds (hematite, magnetite), silicate bonds and local bonds 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.

From the quantitative image analysis, it was observed that the amount and distribution of hematite, magnetite and silicate melt is different in different size pellets as shown in Figs. 10(b) to 10(f). It is due to the fact that with increasing the size, thermo-chemical conditions changes across its cross section. The time difference between the oxidation of the pellet surface and the core increases with increasing pellet size, i.e. the oxidation takes place under different conditions resulting in different phases and microstructure. Hematite dominates the oxidized shell where as magnetite in the poorly oxidized core. The chemical composition and amount of silicate melt also change markedly across the cross section. Melting of calcium ferrites that takes place within a relatively narrow temperature range has a great effect on the volume and composition of the silicate melt. As the amount of core increases with increasing pellet size, distribution of different phases also change accordingly.

Figure 6 above mentioned showed a drastic increase in RDI of pellets of more than 12.5 mm size. Superior RDI values in pellets less than 12.5 mm in size can be attributed to high amount of silicate melt (Fig. 10(d)). The melt wets the solid particles and facilitates the diffusion and grain growth. The liquid between the pores exerts pressure to pull them together due to interfacial forces. Smaller pores are absorbed in to large ones and as a result shape and size of the pores change.

Considering the overall quality parameters, it is obvious that the desired properties of pellets are superior in the size range of 8–12.5 mm and deteriorates with further increase in size. So the pelletising plants should tune their balling and screening circuits to minimize the fraction over 12.5 mm in size in the product pellets.

5.3. MgO Distribution

EDS analysis revealed that MgO follows a specific distribution pattern among different zones of pellet. MgO content of the silicate melt was increasing from the core of the pellet to the shell, as shown in Fig. 13 above mentioned. During induration, MgO may join either slag phase or iron oxide grains. In the oxidized hematite shell, all the released MgO is distributed between silicate slag and magnesioferrite. Hematite contains only traces of magnesium because the crystal lattice of hematite does not allow any substantial amount of magnesium to enter. Mg$^{2+}$ ions can not replace Fe$^{3+}$ ions in the hematite lattice. In the magnetite core, bulk of released magnesium is concentrated in the magnetite grains and a minor part in the slag phase. Magnetite contains both Fe$^{2+}$ and Fe$^{3+}$ ions. Mg$^{2+}$ ions can easily substitute a part of the Fe$^{2+}$ and form Mg-magnetite.

6. Conclusions

During the induration of pellets, temperature and oxygen partial pressure influence the formation of different phases and microstructure. Under similar firing conditions, with increasing the pellet size, these thermo-chemical conditions vary across its cross section resulting in different phases and microstructure. Pellets of different size 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. An attempt was made to correlate the pellet quality to its phases and microstructure.

The following conclusions can be drawn from this work:

1. Increasing the size of pellet increases its FeO content. A drastic increase was observed beyond 12.5 mm size.
Low oxygen partial pressure inside the bigger size pellet results in reducing conditions thereby increasing the FeO content.

(2) CCS of pellets remarkably drops with increasing their size beyond 12.5 mm. Duplex microstructure in these pellets with hematite in the shell and magnetite in the core causes severe lattice and bond strains lowering the strength. Concentric cracks as a result of large shrinkage gradients in the radial direction can also be attributed to low strength of bigger size pellets.

(3) Superior RDI of the pellets under 12.5 mm in size can be attributed to high amount and better distribution of silicate melt. Good bonding interface was observed between the iron oxides grains and bonding phases in the form of high silicate melt perimeter in this size fraction of pellets.

(4) To improve the quality of pellets, pelletising plants should tune their balling and screening circuits to minimize the fraction over 12.5 mm in size in the product pellets.

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