SWELLING AND REDUCTION BEHAVIOUR OF FIRED IRON ORE PELLETS

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ABSTRACT

In this paper considered "Reduction and Swelling Behavior of Fired Iron Ore Pellets" was undertaken with a view to promote the effective utilization of iron ore and coal fines in sponge iron making. Presently, India has become the world leader in sponge iron production and the production of steel by DR-EAF route is increasing day by day. The pellets fired at 1300^0 C were processed for reduction and swelling studies in different types of coal. The degree of reduction of fired iron ore pellets increased with increase of reduction temperature and time up to the rang e studied. The extent of swelling in fired iron ore pellets during their production increased with increase of reduction time, most probably due to the structural changes and fibrous growth of iron particles. SEM images of few reduced iron ore pellets were also taken.

INTRODUCTION

As India progresses towards higher level of growth and greater and more concentrated efforts in the development of Infrastructure and manufacturing sector, the Iron and Steel industry is poised for a rapid growth in the coming years. Steel demand in the country is increasing at the rate of 10% and is likely to remain in the same range at least for the next 10-15 years. In order to meet this continuous growth of Steel demand in the country, domestic Steel producing capacity is required to be higher than 150 MT per annum by 2017.

In India due to the Sponge Iron sector the overall percentage of lump uses in steel making (approx.47%) is higher than most of the countries. India is the only country where over 30% of steel comes from the Induction Furnace sector using sponge iron. Sponge iron plants use only lumps and are located in areas in near vicinity of iron ore mines. As hard ore reserves are depleting, lump generation suitable for blast furnace operation is coming down which results in large amount of access production of fines. The world production of sponge iron has increased from 17.68 MT in 1990 to 69.95 MT in 2010. Presently, India has emerged as the largest producer of DRI (26.30 MT in 2010) in the world. Out of this 26.30 MT, the contribution of coal-based sponge iron plants was 19.10 MT and rest of gas-based plants [1] . This large difference is due to scarcity of natural gas and abundant supply of non-coking coal in India.

Presently a lot of emphasis is being given to the DR process in the country, for the utilization of lower grade coal. This change in technology for production of steel may lead to change in feed stock causing a significant shift in respective share of lumps and agglomerated iron ore for hot metal in furnace, which will enable the use of ore fines which could not be utilized earlier. The steel plants are looking towards the use of Iron ore pellets sinters and lumps as this strategy opens up the opportunity of utilization of fines and are less energy intensive.

Iron Ore Reserves of India [2] Iron ore reserve estimates for world is around 170 billion tonnes with average iron content of 47%. India has the sixth largest reserves of iron ore in the world, and these are some of the best quality iron ore reserves in the world. India along with Ukraine, Russia, China and Australia accounts for about 75% of the world reserves. India's resources of iron ore as per UNFC system as on 1.4.2010 are estimated at 28.53 billion tonnes.

Table 1: State-wise distribution of Iron Ore in India

Source: Indian Bureau of Mines, Ministry of Mines, Govt. of India

State	Hematite Reserves	Magnetite Reserves
Andhra Pradesh	381477	1463541
Assam	12600	15380
Bihar	55	2659
Chhattisgarh	3291824	0
Goa	927171	222673
Jharkhand	4596621	10542
Karnataka	2158677	7801744
Kerala	0	83435
Madhya Pradesh	231445	0
Maharashtra	283208	1360
Meghalaya	225	3800
Nagaland	0	5280
Orissa	5930233	199
Rajasthan	30561	526830
Tamil Nadu	0	507037
Utter Pradesh	38000	0
India	**17882097**	**10644061**

Direct Reduction Technique of Iron Making

The DR technique is one of the alternative methods of Iron making. Direct reduced iron is produced from direct reduction of iron ore by a reducing gas produced either from natural gas or coal. This process produces 97% pure iron which is called solid sponge iron or direct reduced iron or hot briquetted iron. The various processes of DR technique based on coal and gas are: • Coal based rotary kiln process. • Gas based shaft furnace process. • Coal/gas based rotary hearth furnace process. • Multiple hearths furnace based processes. • Coal based DR in Tunnel kilns. • Fluidised bed processes.

Factors Responsible for Swelling of Fired Iron Ore Pellets

Pellets in the reduction furnace swell and hinders its operation. Two main disadvantages of swelling are: reduced strength and disintegration of compact during reduction. However an increase in volume up to 20% is tolerable and is considered as normal swelling which is the characteristics of compact. As in literature the reasons for swelling as proposed are:

Degradation of Iron grains Swelling can be as high as 130% without whisker formation, when reduced up to temperature about 900^0 C, which is explained as: carbon deposition and consequent evolution of large amount of CO/CO_2 gases, causing expansion and disintegration of iron grains [3]. However at such a high temperature about 1100^0 C, disintegration decreases and completely disappeared due to recrystallization.

Whisker or fibrous growth of iron Most of the researchers in their work claimed whisker or fibrous growth of iron grains as the major cause of swelling behavior observed during reduction [4-8]. In the recent study, the dense whiskers and plates in porous structure are formed during abnormal swelling in fired hematite compact.

Crack generated during reduction Inter-granular and trans-granular cracks are generated during reduction are responsible for the change in volume of iron ore pellets during the transformation from hematite to wustite when swelling is marked as 20-27% [9] . Cracking in the pellets is also due to the combined effect of thermal and volume strains (lattice disturbance) during the transformation from hematite to wustite [10] [6]. Growth of iron whiskers favored by cracks and voids generated.

Recrystallization of iron grains Sintering is favored by greater degree of metallization, high reducing temperature and large amount of whiskers formation which further lead to shrinkage. The newly formed iron surface is more reactive and has a greater tendency to stick together because of high energy [11] . The sticking tendency of particles is mainly due to adhesive force, area of contact, and pellet's iron content, however greater will be the size and its mass higher will be the momentum, henceforth lower will be the agglomeration [12] . Structural changes during reduction

Structural changes during reduction Sintering of iron ore pellets results and its volume change is mainly due to the changes in crystal structure during reduction. During the first stage of reduction hexagonal hematite lattice transforms into cubic magnetite lattice and results in about 25% increase in volume [12]. However lattice remains unchanged and is accompanied by a small increase (7-13%) in volume during the transformation of magnetite to wustite.

Physical Properties of Pellets Crushing strength and porosity of pellets more strongly influence its swelling characteristics than geometry [8][13] . Also with increase in crushing strength and decrease in its porosity, the swelling index of pellets decreases. Lower crushing strength and higher porosity gives more active sites for nucleation and growth of iron whiskers. The high strength of pellets is mainly due to presence of slag bonds, these whiskers hence, are not able to push mechanically the grains adjacent to it and therefore results into decrease in volume (lower swelling) [6],[14-16]. However according to some studies pellets having higher porosity indicates less swelling because more stresses can be accommodated which is produced during the course of reduction and formation of iron whiskers [17] .

Briquetting parameters Addition of gangue such as MgO, SiO_2, CaO, Al_2O_3, bentonite, molasses etc. reduces the growth of iron whiskers and hence swelling of iron ore pellets during reduction, which is further explained as addition of these constituents increases its crushing strength and hence do not allow the iron ore whiskers to grow sufficiently during reduction, as a result lower swelling is obtained [18]. Sequence of constituents, decreasing the swelling indices of fired iron ore pellets is MgO, followed by silica, lime and alumina.

Firing parameters With increase in firing temperature the swelling index of iron ore pellets decreases [19-20]. It is observed that pellets which are fired at high temperatures and for a longer time has higher crushing strength and porosity, due to formation of slag bonds and which resulted into reduced growth of iron whiskers and thus lower swelling. A decrease in number of sites for growth of iron whiskers is observed when it is fired at a high temperature, which resulted into decrease in swelling index of iron oxide compact .

Reduction Parameters Iron oxide compacts reduced with CO gas shows a gradual increase in swelling up to a maximum of 176% with rise in temperature up to 900^0 C, which further decreases with increase in temperature up to 1100^0 C [3]. Decrease in volume (lower swelling) at higher temperature was due to sintering and recrystallization of Fe grains, whereas carbon deposition and disintegration of iron grains are the main reasons for increase in volume up to 900^0 C [2] . Higher swelling in the temperature range of 900- 1000^0 C and shrinkage in the range 1100-1200^0 C is found while working on reduction of hematite and magnetite iron ore pellets containing char. In the temperature range of 700-1000^0 C swelling increases with reduction temperature and is maximum at around 900-950^0 C [3] . During reduction of iron oxide compacts by CO gas in the range 800-1100^0 C and it was found that swelling reaches a maximum value at about 900^0 C due to larger amount of whiskers at this temperature. Reducing gas containing hydrogen accelerates the rate of reduction and hence reduces the chances of whisker growth [10]. Gas-solid reaction on the iron oxide surface is inhibited due to adsorption of sulphur on it.

EXPERIMENTAL DETAILS

Sample Preparation The Iron ore fines -100 mesh size approx. 84%, -16+25 mesh size approx. 16% were thoroughly mixed with the addition of concentrated sugarcane juice as binder with varying amount as 2%, 4 %, 6% and little amount of water in it. Pellets were then made by Hand Rolling method. The pellets were dried in electric oven at 110 ^{0} C for more than 5 hours. The dried pellets were fired by heating them in muffle furnace from room temperature to 1300^0 C at a rate of about 40 C/min and soaking at this temperature for 1 hour, followed by furnace cooling.

Reduction and Swelling Behaviour Separate reductions were carried out in coal fines of -4+6 mesh size. In the present investigation, single pellet type reduction experiments, on the weighed fired iron ore pellets which were embedded centrally inside the packed bed of coal particles in each of the stainless steel reactors (size: 75mm height x 40 mm diameter), have been carried out by heating the reactors from room temperature to the predetermined temperature of 950^0 C, 1000^0 C at a rate of about 40 C/min. Each reactor was tightly closed with an air tight cover having an outlet for the release of gas. The temperature was controlled within ± 5 0 C. After soaking for predetermined period of time the reactors were taken out of the furnace after an interval of 15 minutes and cooled to room temperature in air. The reduced pellets were weighed and the degree of reduction was calculated by the wt. % of oxygen removed from each of them. Using Vernier Calliper the diameter of the pellet before and after reduction were measured three times each and averaged for determination of volumes. The swelling/shrinkage at different intervals of reduction was calculated by using the formula:

Swelling index (%) = $\{(V_f - V_i)/V_i\} \times 100$

Where, V_i − Initial Volume of the Pellet, and
V_f – Final Volume of the Pellet after reduction for a given time.
Weight losses in pellets were recorded by an electronic balance to calculate to calculate the Degree of Reduction.

Degree of Reduction was calculated by following formula:

Degree of Reduction = (Weight loss in pellets / total oxygen content in the pellets) x 100

Scanning Electron Microscope Observation In order to assess the surface characteristics and structural changes in some of the reduced iron ore pellets JEOL scanning electron Microscope (6480 LV model) is used. In this study, fractured surfaces of reduced pellets were gold coated to produce a conductive path and examined at magnifications increasing to 2000.

Results and Discussion

Reduction and Swelling Behaviour of Fired Hematite Iron Ore Pellets Data on the degree of reduction versus time for fired Sakaruddin hematite iron ore pellets, reduced in Indian (Ananta), Indonesian, South African, and Australian Coal (size: -4+6 mesh size) at temperatures of 950 and 1000^0 C, have been presented graphically in figure 1 and 2

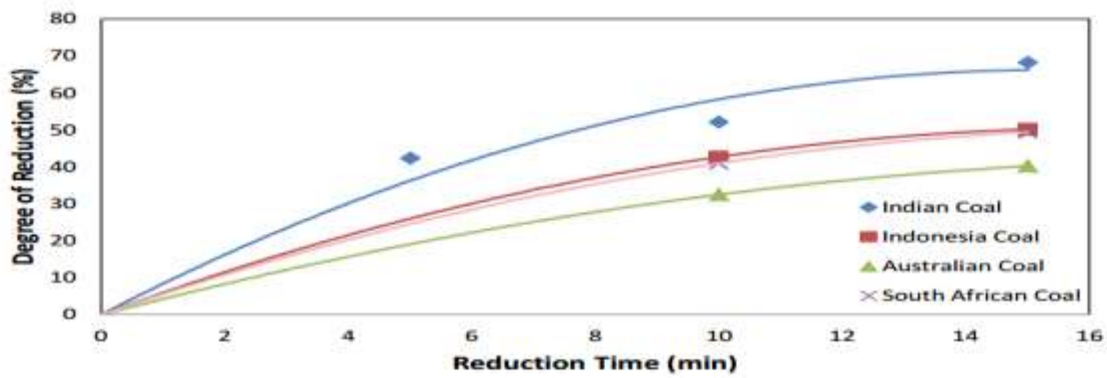

Figure 1: Degree of Reduction vs. Time Plots for the reduction of fired Sakaruddin Hematite Iron Ore Pellets fired at 1300^0 C and reduced in coal (-4+6 mesh size) at a temperature of 950^0 C

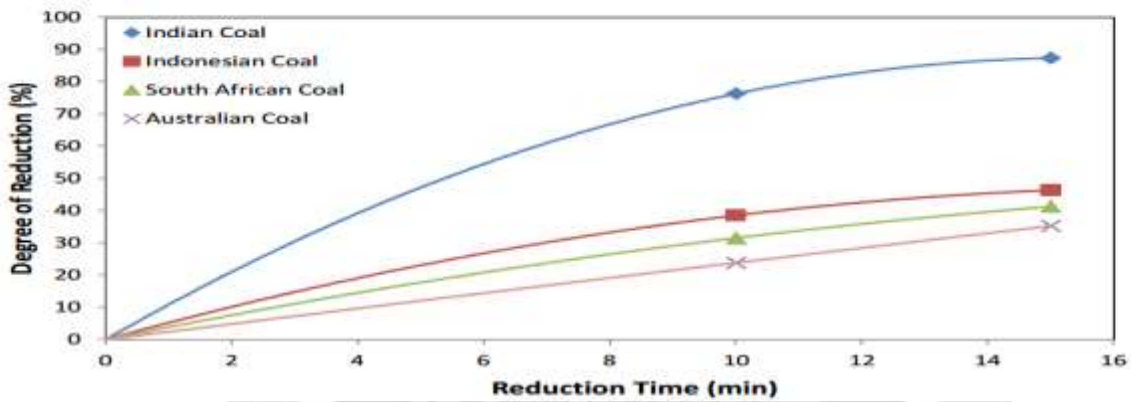

Figure 2: Degree of Reduction vs. Time Plots for the reduction of fired Sakaruddin Hematite Iron Ore Pellets fired at 1300^0 C and reduced in coal (-4+6 mesh size) at a temperature of 1000^0 C

The results (figure 1 and 2) established that in all the studied fired iron ore pellets, the reduction rate improved greatly with increase of temperature up to (950^0 C, 1000^0 C). Asshown in this figure, the degree of reduction also increased with time at all the studied temperatures.

Effects of Temperature and Time As depicted in the figure 3 and 4, swelling is found to be strongly dependent on reduction temperature and time. As shown in figure 3 and 4, pellets were reduced at 950 and 1000^0 C and swelling is slightly higher at 950^0 C. On the basis of researches, literature available and observations made in the scanning electron micrographs (SEMs), all the volume changes at reduction temperatures of 950 and 1000^0 C appear to be due to the combined effects of creation of cracks and voids, formation of iron whiskers and their growth, and phase transformation in the reduced products. They clearly indicate the presence of cracks / voids and iron whiskers in the reduced structures. More carbon deposition (through thermal decomposition of CO gas) and evolution of large amounts of CO / CO2 gases from inner zones of the pellets, reduced at temperatures of 950 and 1000^0 C, are also expected to contribute to their swelling. Thus, the availability of more porous structure and higher carbon deposition appear to be the most probable reasons for slightly higher swelling in the pellets reduced at 950^0 C.

As can be seen in figure 3 and 4, all the fired pellets reduced at temperatures of 950 and 1000^0 C exhibit shrinkage (being more at 1000^0 C) and the extent of this shrinkage, in general, increased with the progress of reduction time / degree of reduction. The higher degree of sintering of iron fibres / grains and their densification may be held responsible for shrinkage in the pellets reduced at temperatures of 950 and 1000^0 C.

A reduction temperature of 1000^0 C greatly increases the ability of iron whiskers/grains to sinter and re-crystallize – a matter which would increase the shrinkage. However, it must be emphasized that a higher amount of carbon (through increased mobility and migration from coal) gets dissolved in the dense iron layers, which in turn causes distortion in iron lattice and thus increase in volume.

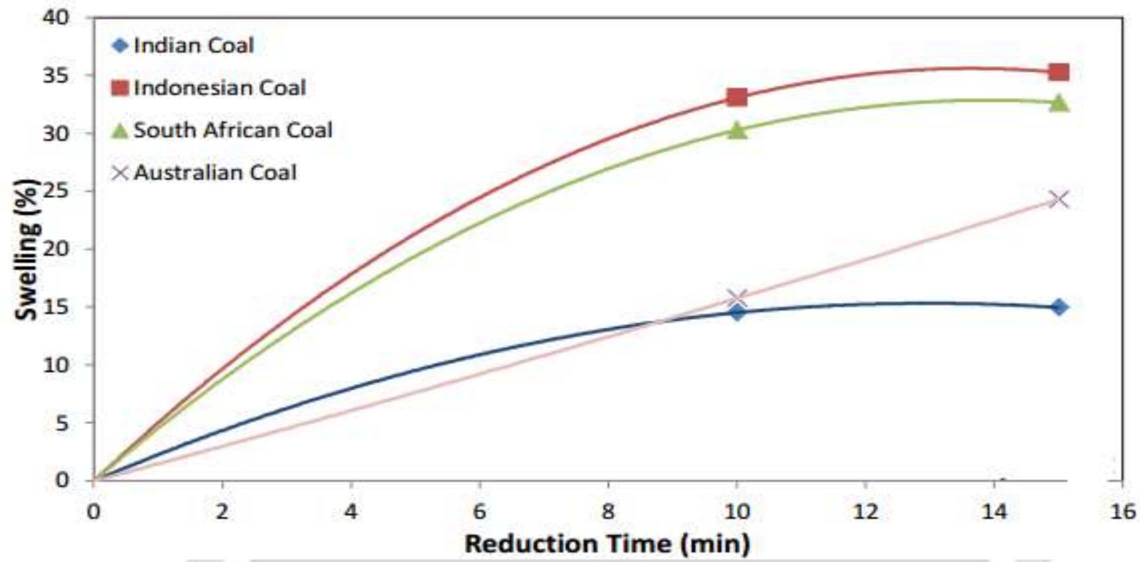

Figure 3: Swelling vs. Time Plots for the reduction of fired Sakaruddin Hematite Iron Ore pellets fired at 1300^0 C and reduced in coal (-4+6 mesh size) at a temperature of 950^0 C.

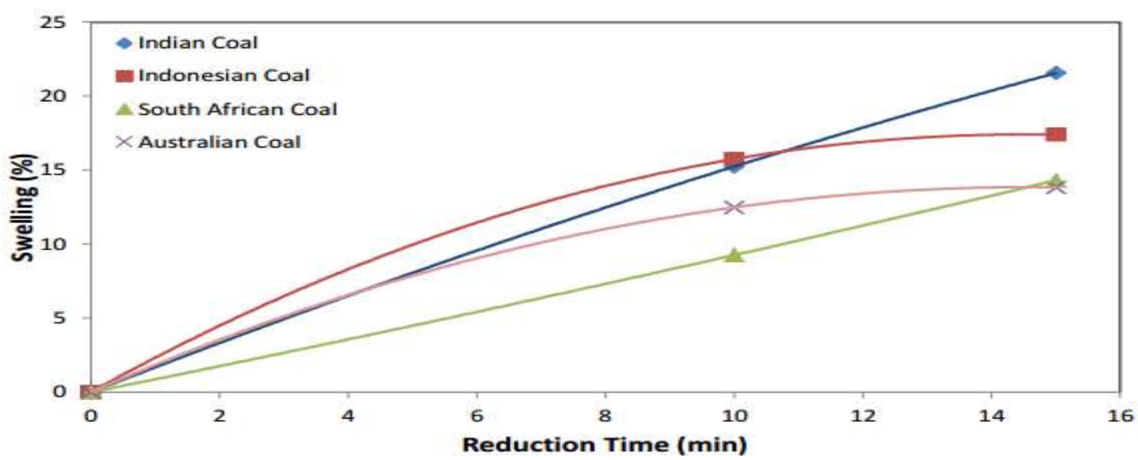

Figure 4: Swelling vs. Time Plots for the reduction of fired Sakaruddin Hematite Iron Ore pellets fired at 1300^0 C and reduced in coal (-4+6 mesh size) at a temperature of 1000^0 C

CONCLUSIONS

I. The crushing strength and drop number of the fired iron ore pellets decrease with the addition of concentrated sugarcane juice binder from 2% to 6%.
II. The degree of reduction increases with increase in temperature in the studied temperature range (950 – 1000^0 C).
III. The degree of reduction of pellets increases with increase in reduction time up to the range studied.
IV. The degree of reduction of fired pellets increased with increase in the reactivity of the coal.
V. The swelling in the fired iron ore pellets increased with increase in reduction time.

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