ISIJ International, Vol. 32 (1992), No. 7, pp. 812–818

Effect of Reduction Rate on the Swelling Behaviour of Iron Ore Pellets

T. SHARMA, R. C. GUPTA[1)] and B. PRAKASH[1)]

Department of Fuel and Mineral Engineering, Indian School of Mines, Dhanbad-826 004, India. 1) Department of Metallurgical Engineering, Institute of Technology, Banaras Hindu University, Varanasi-221 005, India.

(*Received on November 18, 1991; accepted in final form on April 24, 1992*)

In the present investigation an attempt has been made to determine the effect of reduction rate on the swelling behaviour of iron ore pellets. For this purpose two Indian iron ore fines from Bailadila and Noamundi deposits and chemically pure iron oxide were used. The effects of parameters like, temperature, time, reducing gas partial pressure and flow rate have been studied.

From the results obtained, it has been observed that swelling of iron ore pellet is controlled by the rate of reduction of iron oxide. The growth of iron whiskers is also controlled by reduction rate.

KEY WORDS: swelling; reduction; whisker; iron ore; pellet; partial pressure; temperature; flow rate.

1. Introduction

Fired pellets are reduced to metallic iron either in the blast furnace or direct reduction furnaces. The reduction potential of the reducing atmosphere maintained inside the furnace governs the mechanism of reduction. The reduction potential of the atmosphere is dependent upon reduction temperature, nature of reducing gases and their composition. The swelling behaviour of pellet is also very much dependent on the mode of reduction. Hence, the parameters affecting rate of reduction, affect the swelling behaviour of the pellets.

Swelling and disintegration of iron ore pellets have been studied in detail by numerous investigators.[1−32)] During the study, the growth of fibrous iron or iron whiskers while reduction has been cited as major cause for the swelling and disintegration.[1−20)] The chemical composition of the pellet and parameters of pelletization and reduction have been found to affect the growth of iron whiskers and thus, catastrophic swelling. Among the reduction parameters the reduction temperature[1−3,16,21−31)] reducing gases[1−3,16,20−24)] and their partial pressures have been reported to have significant effect on swelling behaviour of the pellet. Higher swelling values have been reported where reduction was carried out with carbon monoxide gas. Fuwa *et al.*,[1)] Taniguchi *et al.*[16)] and several others[21−31)] have studied the effect of reduction temperature and have reported increase in swelling index with increase in reduction temperature upto 1 000°C and further increase in temperature beyond 1 000°C has been reported to result into decreased swelling index of the pellet. Taniguchi *et al.*[16,21−23)] have studied the effect of reduction temperature using hydrogen as reducing agent in the temperature range of 700 to 1 000°C and have observed maximum swelling at 1 000°C. They have used photographic technique to monitor the swelling index of pellets and have found an increase in porosity of the pellet with reduction. Wright and his co-workers[25)] have reported the disruptive force set up during the transformation of hematite to magnetite, as a cause of higher swelling index.

Fuwa *et al.*,[1)] Grance,[2)] and Moon *et al.*[3)] have reported that swelling took place only when reduction was carried out by carbon monoxide whereas others[5,16,21−24)] have observed abnormal swelling with hydrogen reduction and also with mixture of hydrogen and carbon monoxide. During carbon monoxide reduction, carbon deposition on the pellet has also been put forward as a possible cause of catastrophic swelling. Vom Ende *et al.*[20)] have concluded that the swelling is related to rate of reduction.

In the present investigation an attempt has been made to study the effect of reduction rate on the swelling behaviour of iron oxide pellets. For this purpose the effects of reduction temperature, partial pressure and flow rate of carbon monoxide gas have been studied. In a few experiments hydrogen has also been used as reducing agent.

2. Experimental

In the present study high purity ferric oxide and two natural iron oxide ores from Bailadila and Noamundi Mines have been taken. The chemical analyses of these oxides are given in **Table 1.** These oxides were crushed and ground to −125 μm.

The three iron oxide fines were rolled to give 12 to 13 mm diameter pellets using 10–11% moisture. The

Table 1. Chemical analysis of iron oxides.

Sl. No.	Source	Fe	SiO_2	Al_2O_3 (wt%)	CaO	Alkali
1	Pure iron oxide	69.9	Trace	Trace	Trace	Nil
2	Bailadila	68.0	0.70	1.26	Trace	Trace
3	Noamundi	64.0	2.0	3.60	Trace	Trace

green pellets were first dried at 110°C for two hours and then fired at 1 250°C for 60 min. The porosity of these pellets was measured by mercury porosimeter and pellets having porosities around 25% were stored in dessicators for swelling tests.

A 45 mm inner diameter and 1 000 mm long stainless steel tube having water cooled quartz windows at both ends was employed as the reaction chamber in which the swelling behaviour of the pellet was studied. The schematic diagram of the apparatus is shown elsewhere.[32] An alumina base having small cavity was used to keep the pellets stationary in the uniform temperature zone of the furnace. The temperature of the furnace was maintained constant in the range of ± 5°C during the experiment and carbon monoxide gas was used for reduction of pellet. In a few cases hydrogen was used as reducing agent. To decrease the partial pressures of these reducing agent pure and dry argon gas was used. Carbon monoxide gas was generated by dropping formic acid on sulphuric acid kept at 250°C and the gas was purified. Hydrogen and argon gases were available in cylinders.

For determining the swelling index, a pellet of known diameter and weight was kept inside the furnace and a photograph of the pellet was taken with the help of an external light source. The pellet was heated under argon atmosphere, till the desired reduction temperature was obtained. The pellet was soaked at this temperature for 20 min and a photograph of the pellet was taken. Reducing gas was allowed to flow through the reduction tube for a certain time after which a photograph of the pellet was taken. Flow of reducing gas was then stopped and the pellet was cooled to room temperature under argon atmosphere and weight loss of the pellet was determined. The experiment was repeated for different periods of reduction and corresponding swelling values were noted. In order to ensure high accuracy of the results, the volume of pellet was calculated by projecting the pellet photograph on a screen and measuring the average diameter before and after the reduction for a given time. The swelling index of the pellet during heating and at different fractions of reduction was calculated as

$$\text{Swelling index} = \frac{V_a - V_i}{V_i} \times 100 \quad \text{..............(1)}$$

where, V_i: initial volume of pellet, and
V_a: volume of pellet after reduction.

Also the structure of the pellet was studied by scanning electron microscope. The swelling tests have been conducted at four different temperatures ranging from 800 to 1 100°C. The partial pressure of carbon monoxide and hydrogen gases were varied from 0.2 to 1.0 and flow rate from 8.44 to 41.66 cm^3/s.

Table 2. Effect of reduction temperature and flowrate of carbon monoxide on S_M values of pure iron oxide pellets.

Sl. No.	Temp. (°C)	Max. swelling index (S_M) at different flow rates 8.34	16.67	25 (cm^3/s)	33.34	41.67
1	800	61	76	92	92	93
2	900	75	93	110	111	112
3	1 000	83	102	119	118	119
4	1 100	49	60	70	72	71

The percentage volume increase in the pellet due to thermal expansion has been termed as "Thermal Swelling" and indicated by "S_T". During reduction, the volume of pellet increases with reduction time and this swelling is termed as "Reduction Swelling" and denoted by "S_R". The volume attains a maximum value after certain time and it is termed as "Maximum Swelling, "S_M". Hence,

$$S_M = S_R + S_T \quad \text{............................(2)}$$

3. Results

The effects of reduction temperature and flow rate of carbon monoxide gas on the "maximum swelling" (S_M) of pure iron oxide pellets have been presented in Table 2. From these results it is observed that the value of Maximum Swelling (S_M) is highest at 1 000°C when flow rate of carbon monoxide is 25 cm^3/s. Higher flow rate above 25 cm^3/s do not show appreciable change in S_M values.

The results showing the degree of reduction and swelling index as a function of time at four different temperatures have been plotted in **Figs. 1** and **2**. During these tests flow rates of carbon monoxide was kept constant at 25 cm^3/s. In Fig. 2 intercept at Y-axis indicates the value of Thermal Swelling "S_T". These figures reveal that with increase in temperature upto 1 000°C, the ratio of reduction as well as swelling index increases. However, further increase in temperature to 1 100°C results into increased rate of reduction but decreased swelling rate. The maximum value of swelling index at 800°C is 92% whereas 119% at 1 000°C and 70% at 1 100°C. Similar results have been observed in cases of Bailadila and Noamundi ore pellets. The results have been plotted in **Figs. 3** and **4**, whereas swelling index has been plotted as a function of percentage reduction.

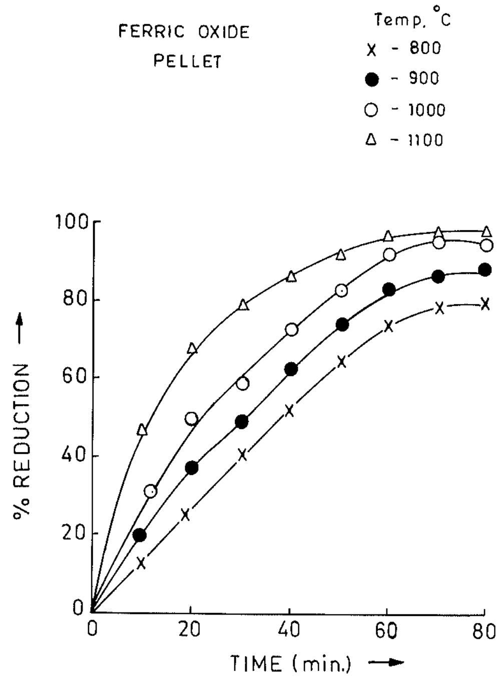

Fig. 1. Reduction *vs.* time plot for pure iron oxide pellets.

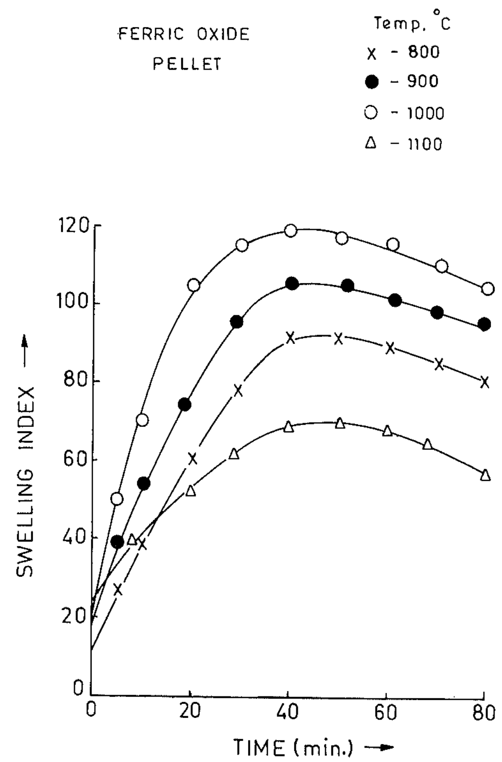

Fig. 2. Swelling index *vs.* time plot for pure iron oxide pellets.

The effect of partial pressure of carbon monoxide gas was studied by mixing argon gas in different proportion. Some typical results obtained have been plotted in **Fig. 5**, which indicate that with increase in partial pressure of carbon monoxide, swelling index is increased. This increase in maximum swelling with increase in partial pressure of CO from 0.20 to 1.0 is 40 to 119% at 1 000°C. The variation in swelling index may be due to the change in the reduction potential of the gas because

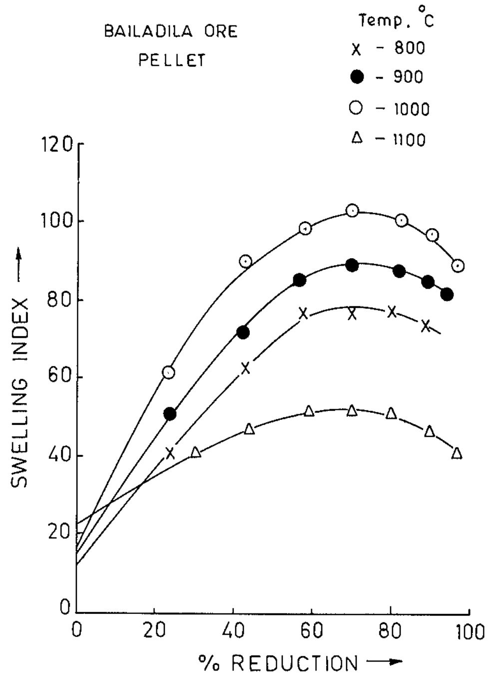

Fig. 3. Swelling index *vs.* reduction plot for Bailadila ore pellets.

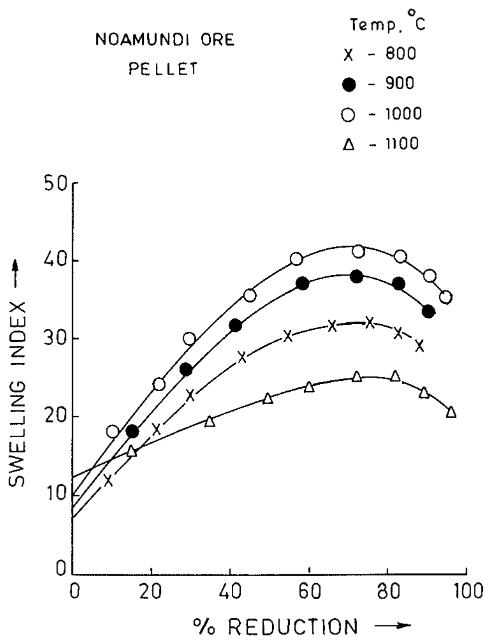

Fig. 4. Swelling index *vs.* reduction plot for Noamundi ore pellets.

of variation in concentration of carbon monoxide gas in the mixture.

4. Discussion

It has been reported by many authors[1-20] that the swelling index of pellet is due to change in structure of the pellet and/or growth of iron whiskers during reduction. The nucleation and growth of iron whiskers take place at selected sites like edges, corners *etc.* The freshly reduced iron migrates towards these nuclei and hence, growing whiskers pushes the adjacent grains resulting into increase in volume of the pellet. It has been reported that the reduction rate is enhanced

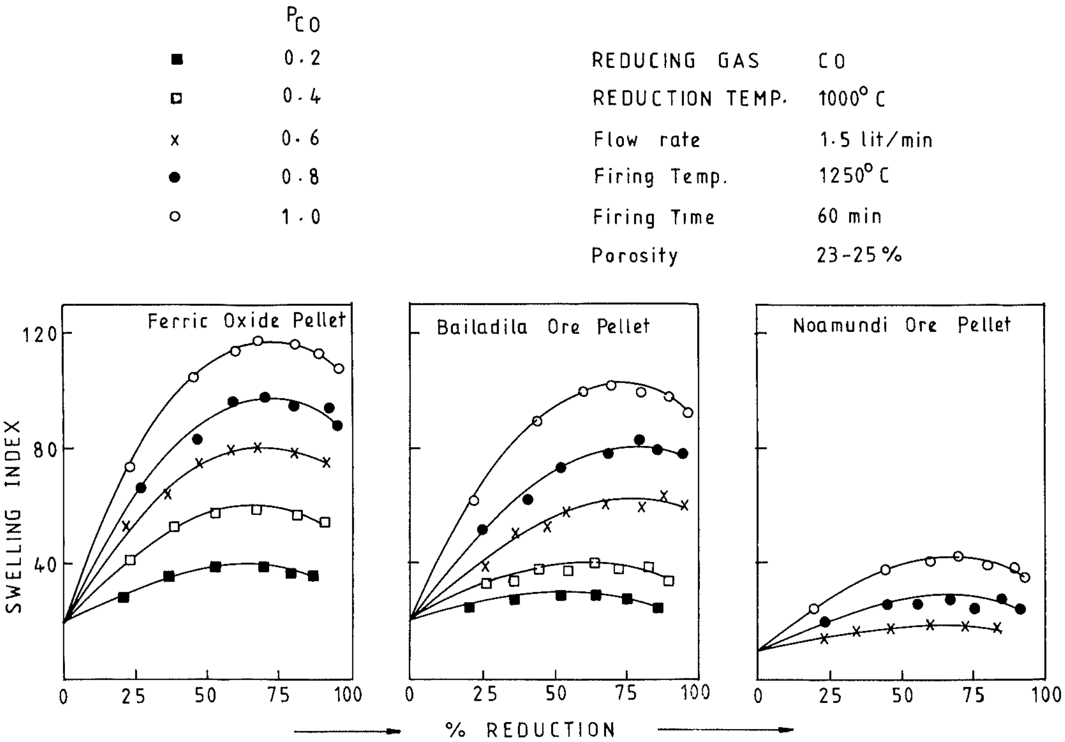

Fig. 5. Effect of P_{CO} on the swelling behaviour of iron oxide pellet.

with increase in reduction temperature and thus, the nucleation and growth rate of iron whiskers. At lower temperatures the reduction rate is so slow that the morphology of nucleation and growth of metallic iron whiskers is changed. With increase in temperature the reduction rate is enhanced causing growth of iron whiskers leading to higher swelling. The swelling index is highest at 1 000°C. With further increase in reduction temperature, the reduction rate is enhanced and swelling should have been higher due to increased rate of whisker growth but, instead, a decrease in swelling index is observed. This may be because of the physical changes *e.g.*, sintering of reduced iron and collapse of pores. At higher temperatures iron whiskers become soft and get bent, may be due to sagging under their own weight or because of growth in a restricted place where porosity is getting decreased as a result of sintering of particles.[33] Due to this sintering of whiskers is observed in pellets reduced at 1 100°C, which can be seen from the scanning electron micrographs shown in **Fig. 6**. In the micrographs longer whiskers have been observed in pellets reduced at 1 000°C.

It has been shown earlier[32] and can be seen from Figs. 3 to 5 that a maximum value of swelling index (S_M) is obtained in between 60 and 70% reduction. Therefore, the rate of reduction at different temperatures, flow rates and partial pressures have been calculated for 60% reduction. The value of $(dR/dt)_{\alpha=0.60}$ have been plotted as function of temperature, PCO, and flowrate of CO in **Fig. 7**. From this figure it is observed that the rate of reduction increases with increase in temperature from 800 to 1 100°C. The swelling index also increases with increase in temperature upto 1 000°C. However, it is decreased at 1 100°C. In this case it can be said that highest swelling index is observed at a reduction rate of 1.75%/min which is obtained at 1 000°C in this case. It seems, at this reduction rate the growth of iron whiskers is highest. If the reduction rate is above 1.75%/min the whiskers grown are larger in number but smaller in size. When the reduction rate is lower than this value the growth of iron whisker is lower and hence lower swelling is observed in both the cases. The reduction rate increases linearly with increase in partial pressure of carbon monoxide from 0.20 to 1.0. The swelling index also increases with increase in partial pressure. With increase in flow rate of carbon monoxide from 8.35 to 25 cm^3/s, the rate of reduction increases. However, further increase in flow rate does not have significant effect on the reduction rate and it remains almost constant. A maximum reduction rate of 1.75%/min is obtained at a partial pressure of 1.0 and flow rate of 25 cm^3/s and under these conditions swelling index is highest. Thus, it can be said that highest swelling index is obtained when the reduction rate is 1.75%/min for 60% reduction.

To confirm the above observations few experiments were conducted at 1 000°C using different partial pressure of hydrogen. The results obtained have been plotted in **Fig. 8**, where Reduction Swelling (S_R) has been plotted as a function of partial pressure of hydrogen as well as carbon monoxide. From Fig. 8, it is observed that the S_R values increases with increase in partial pressure of carbon monoxide but it decreases with increase in partial pressure of hydrogen. The maximum value of S_R has been obtained at 0.20 partial pressure of hydrogen and 1.0 partial pressure of carbon monoxide. The rate of reduction at 0.20 partial pressures of hydrogen is around 1.70%/min, which is almost equal to the rate of reduction obtained in case of 1.0 partial pressure of carbon monoxide. The rate of

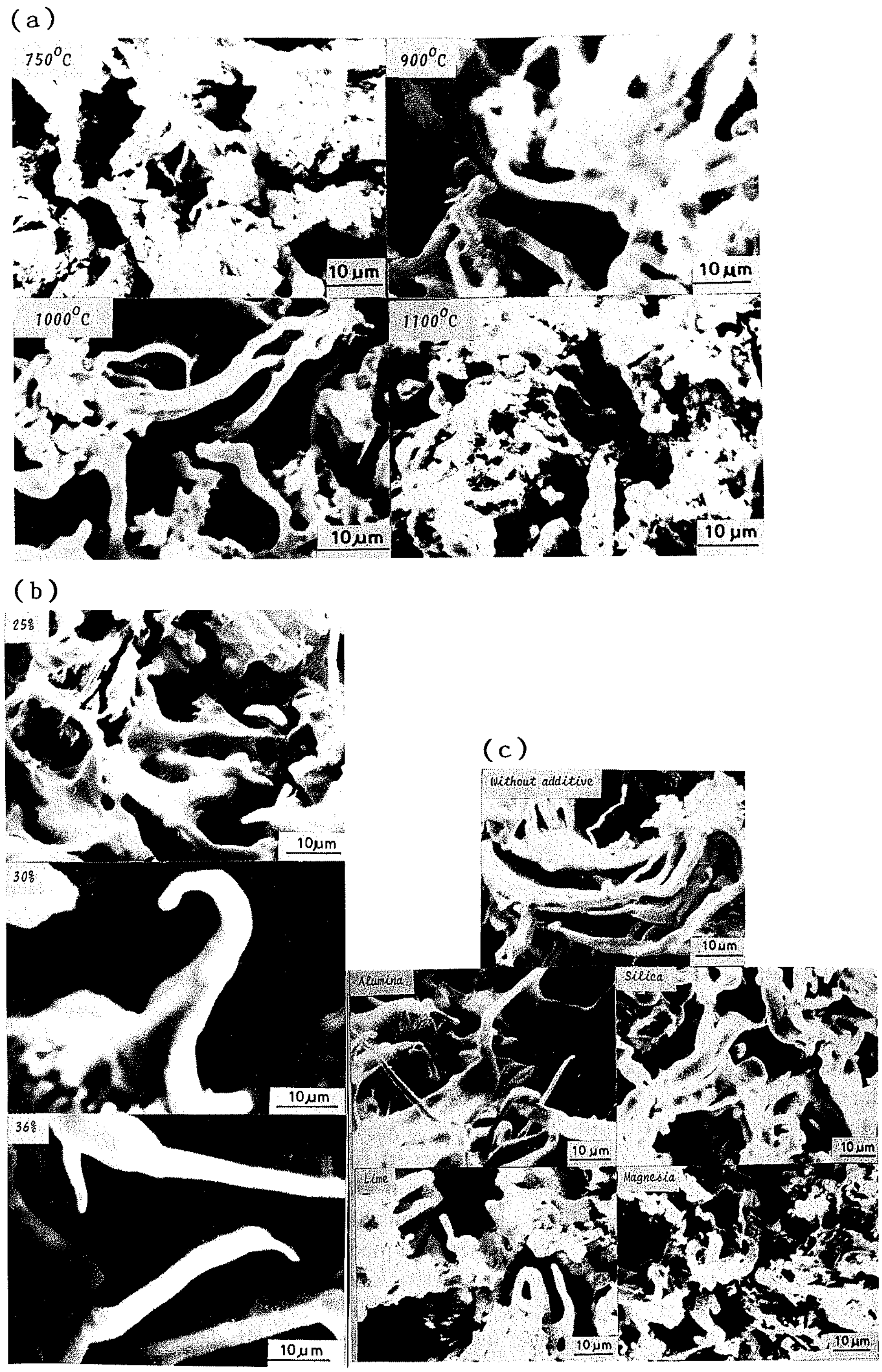

Fig. 6.
Scanning Electron Micrograph of fractured surface of ferric oxide pellet produced under different conditions.

(a)	Reduction temp.	: 750, 900, 1 000, 1 100°C
	Reduction time	: 40 min
	Reduction gas	: 100% CO
	Flow rate	: 25 cm³/s
	Porosity	: 25%
(b)	Reduction temp.	: 900°C
	Porosity	: 25, 30, 36%
	Reduction rate	: 1.75%/min for pellet with 36% porosity
	Reducing gas	: 100% CO
	Flow rate	: 25 cm³/s
(c)	Reduction temp.	: 1 000°C
	Flow rate	: 25 cm³/s of 100% CO
	Additives	: Rate of Reduction
	Nil	: 1.75%/min
	2% alumina	: 1.70%/min
	2% lime	: 1.68%/min
	2% silica	: 1.62%/min
	2% magnesia	: 1.60%/min

reduction has been found to increase with partial pressure of hydrogen and the swelling index is found to be decreased. Hence, it can be said that maximum swelling index is obtained at a critical value of reduction rate irrespective of reducing agent. This phenomena is clearly shown in **Fig. 9**, where S_R values have been plotted against reduction rate, showing highest value of swelling index at a critical rate of reduction (1.75%/min). In this figure reduction rate (dR/dt) calculated for CO and H_2 under various conditions have been plotted against S_R values and it is observed that in case of carbon monoxide the S_R value increases with increase in reduction rate upto 1.75%/min and it decreases with further increase in reduction rate either by CO and H_2.

The reduction data obtained during the present investigation have been analysed and it has been found that the chemical reaction between ironoxide and carbon monoxide/hydrogen is the rate controlling step. It has also been reported by Nicolle and Rist that larger whiskers are observed during chemical control.[35]

From the results and Fig. 9, it is observed that the S_R value maintains a linear relationship with reduction rate. The curve on its extrapolation passes through origin and follows the relationship

$$Y=mX$$

In the present case

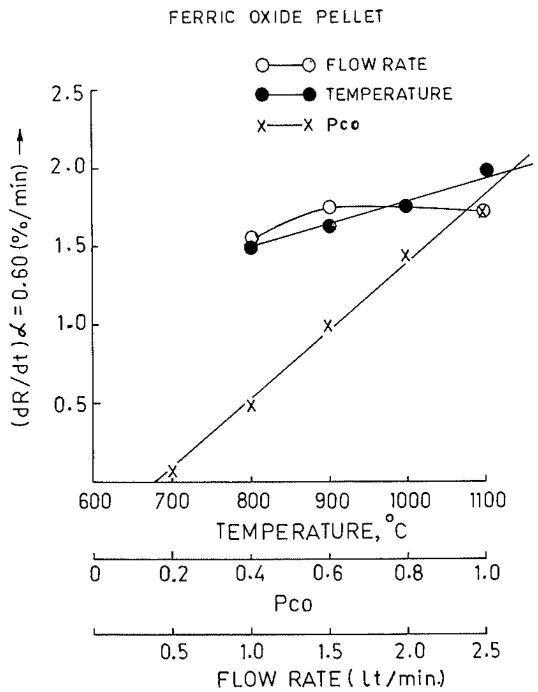

Fig. 7. Effect of temperature, P_{CO} and flow rate on $(dR/dt)=0.60$. (Effect of flow rate and P_{CO} was studied at 1 000°C. Effect of temperature and P_{CO} was studied at a flow rate of 25 cm^3/min.)

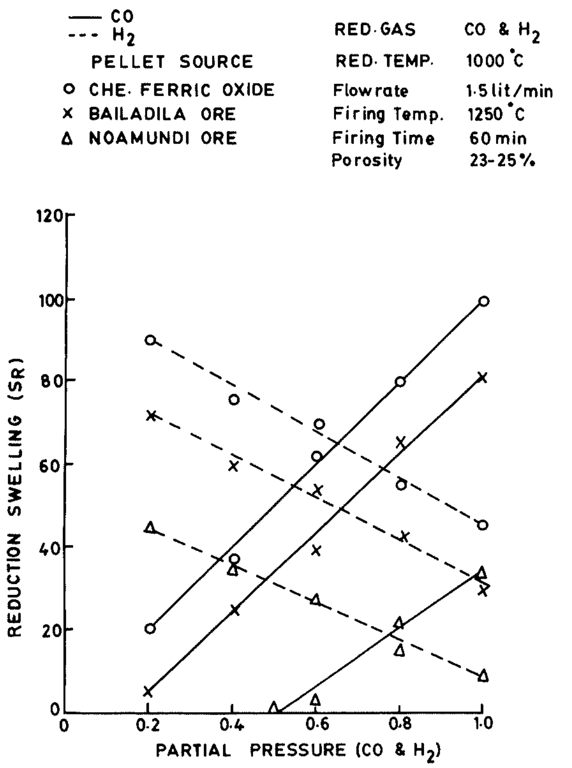

Fig. 8. Effect of P_{CO} and P_{H_2} on reduction swelling (S_R) of iron oxide pellet.

$$S_R = m\left(\frac{dR}{dt}\right)_{\alpha=0.60}$$

In this case the value of $m=57.25$. Hence,

$$S_R = 57.25\left(\frac{dR}{dt}\right)_{\alpha=0.60} \quad \text{......................(3)}$$

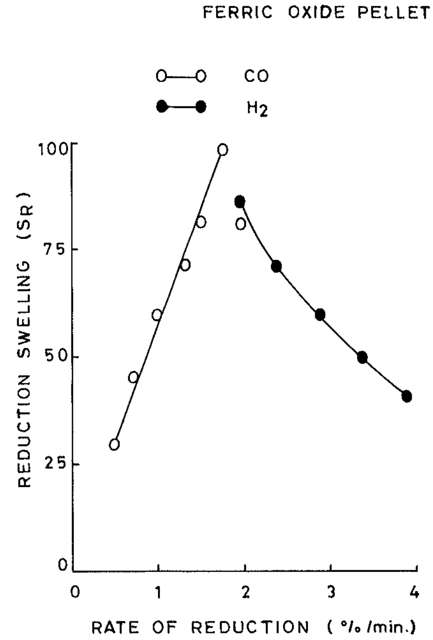

Fig. 9. Effect of rate of reduction swelling (S_R).

Hence, we can say that the reduction swelling (S_R) is proportional to the rate of reduction upto 1.75% per min. This critical rate of reduction is dependent upon the combined effect of all the three parameters *viz.* reduction temperature, the partial pressure and flow rate of the reducing gases. And the equation for maximum swelling (S_M) will be

$$S_M = S_T + 57.25\left(\frac{dR}{dt}\right)_{\alpha=0.60} \quad \text{................(4)}$$

where, S_T: thermal swelling.

The above Eqs. (3) and (4) are empirical relation derived on the basis of limited data obtained during the investigation and need further verification for different ores, ingredients and porosity. The equations are only applicable for pure iron oxide upto 1 000°C. Its application for different ores will be subject to modification of the equation.

From the present investigation and the observation of various other workers, it seems that the swelling index of pellet is dependent upon the growth of iron whiskers during reduction, which is very much dependent upon the rate of reduction, number of nucleation sites and strength of the pellet. These whiskers have been observed to grow on the wustite surface in pores at the edges and corners, either originally present in the pellet or created during reduction as reported by Al Kahtany *et al.*[34] When the bonding strength of the pellet is less, these whiskers push the adjacent ore grains of iron or other oxides and hence, result in increase in volume and sometimes, cracking and disintegration of pellets take place. But when the bonding strength of the pellet is higher either due to stronger bridging of iron oxides or due to presence of slag bonds, these whiskers are not able to push the adjacent surface mechanically and in this case lower swelling is observed. The growth of iron whiskers in the present case is related to .the non-stoichiometric composition of wustite. It is related to the Wagner's mechanism of reduction of non-stoichiometric compounds. It is combined effect of chemical reaction at the surface and iron transport which results into accumulation of iron on selected sites and formation of iron whiskers.

The rate of whisker growth is a resultant of two processes *viz.* rate of reduction and rate of sintering of iron while the rate of reduction is controlled by reduction parameters, like reducing gas, reduction temperature, flow rate of reducing gas *etc.*, the rate of sintering of iron is controlled by temperature. The rate of sintering of iron is rapid above 1 000°C and due to this reason, lower swelling has been observed at 1 100°C, though the reduction rate is faster.

There is difference in swelling index of the three pellets (pure iron oxide, Bailadila ore and Noamundi ore). It is because of difference in gangue content of the pellet. The effect of gangue content has already been reported by the authors.[32,36] The gangue content of Bailadila ore is around 2% and Noamundi ore is 5.6% whereas gangue content of pure iron oxide is almost nil.

In order to verify the linear relationship of Reduction rate with "Reduction Swelling" random experiments were carried out with pure iron oxide pellets/briquettes. The condition for reduction were changed to get different reduction rate. It was found that the $(dR/dt)_{\alpha=0.6}$ maintains a linear relationship with reduction swelling. The relation was found to be applicable even in the case of cylindrical shaped briquettes.

5. Conclusions

From the present investigation we can conclude that

(1) the swelling index of pellet is affected by reduction temperature, partial pressure and flow rate of reducing gases.

(2) maximum swelling value is obtained at a critical rate of reduction 1.75%/min irrespective of reducing gases,

(3) swelling index maintains a linear relationship with $(dR/dt)_{\alpha=0.60}$ and the relation is represented by

$$S_R = 57.25\left(\frac{dR}{dt}\right)_{\alpha=0.60}$$

and

$$S_M = S_T + 57.25\left(\frac{dR}{dt}\right)_{\alpha=0.60}$$

REFERENCES

1) T. Fuwa and S. Ban-ya: *Trans. Iron Steel Inst. Jpn.*, **9** (1969), 137.
2) L. Grance: Proc. ICS TIS Tokyo, *Suppl. Trans. Iron Steel Inst. Jpn.*, **11** (1971), 45.
3) J. T. Moon and R. D. Walker: *Ironmaking Steelmaking*, **2** (1975), 30.
4) W. Wenzel and H. W. Gudenau: *Trans. Soc. Min. Engrs. AIME*, **252** (1972), 281.
5) J. K. Wright: *Proc. Aust. Inst. Min. Met.*, **265** (1978), 1.
6) W-K. Lu: Ironmaking Proc., Atlantic City, **33** (1974), 61.
7) H. Vom Ende *et al.*: *Stahl Eisen*, **90** (1970), 586.
8) W-K. Lu: *Scand. J. Metall.*, **3** (1974), 49.
9) W-K. Lu: *Scand. J. Metall.*, **2** (1973), 273.
10) W-K. Lu: *Scand. J. Metall.*, **2** (1973), 169.
11) W-K. Lu: *Scand. J. Metall.*, **2** (1973), 65.
12) M. Sasaki and T. Nakajawa: *Trans. Iron Steel Inst. Jpn.*, **10** (1970), 464.
13) K. Kaneko and A. Okura: *Trans. Iron Steel Inst. Jpn.*, **11** (1971), 294.
14) M. Amatatsu *et al.*: *Trans. Iron Steel Inst. Jpn.*, **15** (1975), 417.
15) H. Itaya *et al.*: *Trans. Iron Steel Inst. Jpn.*, **15** (1975), 429.
16) S. Taniguchi: *Trans. Iron Steel Inst. Jpn.*, **18** (1978), 633.
17) R. L. Bleifuss: *Trans. Soc. Min. Engrs. AIME*, **247** (1970), 225.
18) F. Nakiboglu *et al.*: Proc. Aust./Japan Ext. Met. Symp. Sydney 16–18, July, (1980), 157.
19) S. Sayama *et al.*: *Trans. Iron Steel Inst. Jpn.*, **19** (1979), 521.
20) H. VomEnde *et al.*: *Stahl Eisen*, **91** (1971), 815.
21) S. Taniguchi and M. Ohmi: *Trans. Jpn. Inst. Met.*, **21** (1980), 433.
22) S. Taniguchi and M. Ohmi: *Trans. Jpn. Inst. Met.*, **19** (1978), 581.
23) S. Taniguchi and M. Ohmi: *Trans. Iron Steel Inst. Jpn.*, **20** (1980), 433.
24) J. K. Wright: *Trans. Iron Steel Inst. Jpn.*, **17** (1977), 726.
25) J. K. Wright and A. L. Morrison: Proc. Aust/Japan Extractive Metallurgy Symp., Sydney, 16–18, July, (1980).
26) G. Thaning: *Ironmaking Steelmaking*, **3** (1976), 57.
27) T. Y. Malysheva *et al.*: *Steel USSR*, **2** (1972), 418.
28) I. Shigaki *et al.*: *Trans. Iron Steel Inst. Jpn.*, **22** (1982), 838.
29) A. N. Spektor, and A. N. Pyrikov: *Steel USSR*, **2** (1974), 4.
30) S. Taniguchi *et al.*: *Trans. Iron Steel Inst. Jpn.*, **20** (1980), 753.
31) T. Sharma *et al.*: Swelling Behaviour of Iron Ore Pellet, presented in 36th ATM of IIM (1982).
32) T. Sharma *et al.*: *Mineral Engineering*, **3** (1990), 509.
33) D. G. Khokhlov: Stal in English, (1970), 761.
34) M. M. Al-Kahtany and Y. K. Rao: *Ironmaking Steelmaking*, **7** (1980), 49.
35) R. Nicolle and A. Rist: *Metall. Trans. B*, **10B** (1979), 429.
36) T. Sharma *et al.*: *ISIJ Int.*, **31** (1991), 312.