1. Introduction
The pig iron nugget process was developed by Kobe Steel as an environmentally friendlier, economical alternative to the traditional blast furnace process. It aims to produce pig iron nuggets which have similar chemical and physical properties to blast furnace pig iron. Throughout this single step process, self reducing–fluxing dried greenballs were reduced, carburized and smelted. The self reducing–fluxing dried greenballs were produced by agglomeration of iron ore concentrate (magnetite), flux (limestone), binder (bentonite), and reducing–carburizing agent (coal). The comparison of the basic process properties of blast furnace process and pig iron nugget process is shown in Table 1.

The objectives of the pig iron nugget process can be summarized as follows:

i. High Degree of Metallization, High Iron Yield
The degree of metallization calculated according to Eq. (1), of liquid iron (pig iron) produced by blast furnace ranges between 94–98% with approximate iron content of 90–95.5% depending on the reducibility properties of the ore, and ash content of the coke. Thus, the pig iron nuggets produced were intended to have similar or higher degrees of metallization and iron content values in order to be competitive.

\[
\%M = \frac{\%Fe_n \cdot M_n}{\%Fe_{dgh} \cdot M_{dgh}}
\] ..........................(1)

Where:
\%M : the percent metallization,
M_n : the weight of the pig iron nugget (or pig iron),
M_{dgh} : the weight of the dried greenball (or pellet, sinter),
\%Fe_n : the percent iron concentration of the pig iron nugget (or pig iron), and
\%Fe_{dgh} : the percent iron content of the dried greenball (or pellet, sinter).

ii. Reduced Energy Losses in Iron Oxide Feed Preparation
The blast furnace process utilizes indurated pellets and sinters as iron oxide feed stock. The indurated pellets are produced by agglomerating iron ore concentrate and binder in balling drums and then heat hardening them in a kiln. The sinters are produced by heating up a granulated mix of iron ore concentrate to slightly above the softening temperature of 1 200°C. Throughout the sintering process coke breeze is utilized as main energy source to give them solid state bonding by inter-diffusion of contacting particles. After these indurated pellets or sinters are produced they are cooled for easy transportation to the blast furnace site.
where they are reheated in the blast furnace.

The pig iron nugget process aims to save energy by making it possible to produce pig iron from self-reducing–fluxing dried greenballs with just one heating step. Self-reducing–fluxing dried greenballs will be heat treated directly without any induration or sintering steps. This will enable the elimination of the energy used by heating, cooling and reheating of the pellets and sinters.

iii. Reduced Residence Time in the Reactor

For the pig iron nugget process the kinetics of the reduction and carburization reactions are enhanced by agglomeration of an iron ore source, flux, and reducing–carburizing agent together. The close contact of the reacting materials and availability of a large number of reacting sites enhances the solid–solid reduction and slag forming reactions. Also, the internal gas generation, and small diffusion distances in the sample enhances the solid–gas reduction reactions, reducing gas regeneration, slag forming and carburization reactions.10–15

iv. Impurity (Gangue) Separation with Formation of Fusible Slag

For the blast furnace process and pig iron nugget process, the impurities are separated from the metal by the formation of slag. Flux materials such as limestone, dolomite, and magnesia are added to lower fusion temperatures to enable the slag formation. The slag can only be separated from the metal when metal and slag are in liquid state. This requires metal (pure iron fusion temperature of 1 535°C) and slag (slag fusion temperature around 1 200°C) to be heated above their fusion temperatures. The blast furnace process operates at temperatures above the fusion temperature of iron and produces liquid metal (pig iron) and slag. On the other hand, throughout the pig iron nugget process, it is intended to have carburization of pure iron, lowering its fusion temperature (this effect takes place until the eutectic carbon composition is reached). This should allow the pig iron nugget process to operate at lower temperatures than the fusion temperature of iron and still obtain slag separation.

v. Cost Competitive Process, Raw Iron Oxide and Reducing–Carburizing Agent Utilization

The major cost components of the traditional blast furnace iron making process are shown in Fig. 1.16,17 To make the processes more cost competitive the critical cost components (iron oxide feed, energy, and capital costs) should be reduced. Blast furnace operation is a capital intensive operation due to ancillary plant and equipment requirements. Primary cost cuts can be made by eliminating the need for high quality coke and pellet/sinter heating. The pig iron nugget process utilizes dried greenballs instead of indurated pellets and sinter as iron oxide feed material. In addition, it utilizes widely available coal instead of metallurgical coke. This thereby eliminates coke ovens, sinter plants, and induration (heat hardening) kilns.

vi. Environmentally Friendlier Process

Coking is the major contributor of environmental emissions in iron making processes. The amount of coke required for the blast furnace process can be reduced by using pre-reduced sinters and pellets and higher temperatures in the furnace. However, the reduction of amount of coke utilized in the blast furnace is not enough, due to increasing environmental regulations on coke making and increasing operational costs. On the other hand, pig iron nugget process utilizes widely available coal as the reducing and carburizing agent, eliminating the need for coke.

vii. Integration of the Process with Existing Works

The blast furnace pig iron can be used in the basic oxygen furnaces and electric arc furnaces (in the granulated form) for steel production. On the other hand, the pig iron nuggets are aimed to be beneficiated in (i) the electric arc furnaces for steel production, (ii) the electric arc furnaces, foundry cupolas and basic oxygen furnaces to lower residuals, replacing scrap and direct reduced iron, (iii) as a coolant in electric arc furnaces, foundry cupolas and basic oxygen furnaces, and (iv) in ferrous foundry operations as iron unit charge.

viii. Flexible Process

Optimum, economically viable high production rates and high degrees (85–90%) of heat utilization can be achieved for the blast furnace operation when operating at high capacities.41 This requires a continuous supply of high quality raw materials such as iron ore pellets, sinter and coke. Since blast furnaces can not be shut down and restarted easily this continuous high capacity production takes place even under depressed market conditions. Conversely, since the pig iron nugget process is designed as a single step process it is easier to adjust and control its operation with the changing market conditions. It is designed in such a way that it can be shut down and restarted easier than the blast furnace process, and its operational capacity can also be incrementally increased.

ix. Reduced Shipping, Transportation, and Handling Costs and Efforts

It can be seen from Fig. 2 that for the traditional blast furnace process, iron ore pellets are transported from the mineral processing plant to the blast furnace site. These pellets contain only 60–65 wt% iron. Then pig iron is produced at the blast furnace site and transported to steel plants. On the other hand, it can be seen from Fig. 3 that if the pig iron nugget process is integrated with the existing mineral processing operation, pig iron nuggets can be produced and transported directly to steel plants. This would reduce the transportation costs.
Hence, the objective of this work was to (i) produce pig iron nuggets at laboratory conditions, (ii) physically and chemically characterize the pig iron nuggets produced and (iii) compare them with blast furnace pig iron. The physical and chemical characterization of the pig iron nuggets was accomplished by the investigation of their chemical composition, degree of metallization, degree of slag separation, apparent density, and microstructure.

2. Experimental

The experiments involved firing of the self reducing–fluxing dried greenballs made out of magnetite ore, coal, limestone and bentonite in a laboratory scale resistance box furnace at furnace temperatures of 1450°C and 1475°C and furnace residence times of 22 and 28 min. These furnace temperatures and residence times for pig iron nugget production were decided according to previous work.7,8) Pig iron nuggets were characterized utilizing apparent density measurements, optical microscopy, scanning electron microscopy with energy dispersive spectroscopy, and bulk chemical analysis.

2.1. Raw Materials

The self reducing–fluxing greenballs to be fired contained 71.84% dry wt. magnetite ore as an iron source, 0.66% dry wt. bentonite clay as binder, 7.5% dry wt. limestone as flux, and 20% dry wt. high volatile bituminous coal as reducing and carburizing agent. They were made in a laboratory scale balling drum shown in Fig. 4.

Greenball diameter ranged between 12.7 and 11.2 mm (1/2 to 7/16 inch). The density of greenballs was 2.5 g/cm³ and their chemical composition is shown in Table 2.

The magnetite concentrate utilized as an iron source was 80% passing 25 microns (500 mesh). The X-ray diffraction pattern of the magnetite concentrate utilized is shown in Fig. 5. It can be seen from Fig. 5 that only small quantities of silicate gangue was present in the magnetite concentrate.

The bentonite clay utilized as a binder was a Na montmorillonite based clay and was 85% passing 75 microns (200 mesh).

Limestone was utilized as flux. The X-ray diffraction of the limestone utilized is shown in Fig. 6. It can be seen...
from Fig. 6 that the limestone was pure calcium carbonate.

High volatile bituminous coal utilized as reducing and carburizing agent was 100% passing 850 microns (20 mesh). The proximate and ultimate analysis of the coal is shown in Table 3.

### Table 3. The proximate and ultimate analysis of the coal utilized as reducing and carburizing agent.

<table>
<thead>
<tr>
<th></th>
<th>As Received</th>
<th>Moisture Free</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture %</td>
<td>6.47</td>
<td></td>
</tr>
<tr>
<td>Ash %</td>
<td>7.34</td>
<td>7.85</td>
</tr>
<tr>
<td>Sulfur %</td>
<td>2.11</td>
<td>2.25</td>
</tr>
<tr>
<td>Mj/kg</td>
<td>30.575</td>
<td>32.690</td>
</tr>
<tr>
<td>Fixed Carbon %</td>
<td>55.46</td>
<td>59.29</td>
</tr>
</tbody>
</table>

### 3. Results

#### 3.1. Pig Iron Nuggets

Pig iron nuggets produced had a pebble shape structure and high apparent density. The slag accompanied with the nuggets was easily separated by physical means leaving a slag free product.7,8)

The pig iron nuggets accompanied with their slag produced at furnace temperature of 1450°C and residence time...
Fig. 8. Pig iron nuggets produced at furnace temperature of 1450°C and furnace residence time of 22 min furnace.

Table 4. The physical and chemical analysis techniques utilized for pig iron nugget characterization.

<table>
<thead>
<tr>
<th>Analysis Technique</th>
<th>Objective</th>
</tr>
</thead>
<tbody>
<tr>
<td>Apparent Density Measurements (a)</td>
<td>Evaluation of degree of metallization, porosity, and slag separation</td>
</tr>
<tr>
<td>Optical Microscopy (20,22)</td>
<td>Phase and microconstituent identification, microstructural characterization and relating the pig iron nugget structure to known microstructures</td>
</tr>
<tr>
<td>Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDS) (20,22)</td>
<td>Characterization of the microstructure, identification of the phases and microconstituents</td>
</tr>
<tr>
<td>X-ray Diffraction (XRD)</td>
<td>Phase identification</td>
</tr>
<tr>
<td>X-ray Fluorescence Spectroscopy (XRF)</td>
<td>Obtaining bulk chemical analysis which can be compared with the blast furnace pig iron</td>
</tr>
</tbody>
</table>

Table 5. A comparison of the chemical compositions of the blast furnace pig iron and pig iron nuggets. (An average chemical composition of the pig iron nuggets produced at furnace temperatures of 1450°C and 1475°C and furnace residence times of 22 and 28 min are shown.)

<table>
<thead>
<tr>
<th>Element</th>
<th>Blast Furnace Pig Iron wt % (b)</th>
<th>Pig Iron Nuggets wt %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>90 – 95.5</td>
<td>95 – 97</td>
</tr>
<tr>
<td>C</td>
<td>4 – 5</td>
<td>1.7 – 3.5</td>
</tr>
<tr>
<td>Si</td>
<td>0.3 – 1</td>
<td>0.8</td>
</tr>
<tr>
<td>S</td>
<td>0.03</td>
<td>0.2 – 0.8</td>
</tr>
<tr>
<td>P</td>
<td>&lt;1</td>
<td>0.12</td>
</tr>
<tr>
<td>Mn</td>
<td>0.1 – 2.5</td>
<td>0.12</td>
</tr>
</tbody>
</table>

of 22 min are shown in Fig. 8. Molten pig iron nuggets and molten slag were observed after the heat treatment. The formation of these molten phases led the slag to separate from the metal due to immiscibility and density difference (see Fig. 8).7,8)

3.2. Chemical Analysis of the Pig iron Nuggets

The chemical analysis of the pig iron nuggets was investigated in order to be able to better define and compare them with blast furnace pig iron. The average chemical analysis of the pig iron nuggets produced at furnace temperatures of 1450 and 1475°C and residence times of 22 and 28 min along with blast furnace pig iron is shown in Table 5.

It can be seen from Table 5 that pig iron nuggets contain equivalent or more % iron than blast furnace pig iron. Pig iron nuggets contain an equivalent amount of carbon when compared with white cast iron. It should be noted that pig iron nuggets were produced under controlled laboratory conditions.

The slag separated from these nuggets contained between 13–20 wt% iron. This corresponded to approximately 3–4% of the iron introduced with the self reducing fluxing dried greenballs. Approximately 30% of this iron was magnetic and 70% was non magnetic.7)

The degrees of metallization for pig iron nuggets were found to be 94–97% which was equivalent to degrees of metallization for blast furnace pig iron.

3.3. Pig Iron Nugget Microstructure

i. Optical Microscopy

The optical micrographs of the pig iron nuggets produced at furnace temperature of 1475°C and furnace residence times of 22 and 28 min are shown in Figs. 9 and 10 respectively.

The pig iron nugget microstructures shown in Figs. 9 and 10 are composed of (i) a dendritic pattern of fine pearlite (dark grey areas), (ii) an interdendritic eutectic mixture of cementite (light areas) and pearlite (dark grey areas), and (iii) cementite which is formed in the solid state (light areas). These microstructures are very similar to the suggested characteristic of white cast iron structure.7,8,24–31)

ii. Interpretation of Pig Iron Nugget Microstructures

The metastable iron carbon phase diagram shown in Fig. 11 was utilized for the interpretation of the pig iron nugget microstructures. As an example, consider a molten pig iron nugget containing 3.2% wt carbon containing molten pig iron nugget cooling will be discussed. It can be seen from Fig 11, (Point B) that during cooling austenite dendrites (dark grey areas), (ii) an interdendritic eutectic mixture of cementite (light areas) and pearlite (dark grey areas), and (iii) cementite which is formed in the solid state (light areas). These microstructures are very similar to the suggested characteristic of white cast iron structure.7,8,24–31)
and eutectic iron carbide (cementite) \((\text{Fe}_3\text{C})\) (6.67% C) as shown in Point E. Fraction of austenite (2.08% C) and iron carbide (6.67% C) is shown in Table 6.

As the cooling proceeds in the solid state below the eutectic temperature the fraction of iron carbide increases as the solubility of carbon in the austenite decreases. This increase is shown by the fraction of iron carbide values for point F and G at Table 6.
It can be seen from Figs. 9 and 10 that this increasing amount of iron carbide in the solid state appears to have penetrated into the austenite.24–31) When Point H (just before the eutectoid reaction) is reached the fraction of iron carbide and austenite is 0.41 and 0.59 respectively (see Fig. 11).

Finally, when the eutectoid temperature of 723°C is reached the austenite transforms to Pearlite which is a micro-constituent with alternating layers of α ferrite and iron carbide (cementite) as shown in Point I. The total fraction of iron carbide at this point is shown on Table 6.

The final microstructure attained by this slow cooling scheme described in Fig. 10 is similar to the reported white cast iron structure reported in the literature to the and pig iron nugget microstructures shown in Figs. 9 and 10.24–31)

### Table 6. Fractions of austenite, iron carbide and liquid present in the system as the pig iron nugget cools (see Fig. 11). These values were calculated utilizing the lever rule.26,27)

<table>
<thead>
<tr>
<th>Point</th>
<th>Fraction of Austenite</th>
<th>Fraction of Liquid</th>
<th>Fraction of Iron Carbide</th>
<th>Fraction of Pearlite</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>0.25</td>
<td>0.75</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>D</td>
<td>0.5</td>
<td>0.5</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>E</td>
<td>0.76</td>
<td>0.26</td>
<td>-</td>
<td>0.24</td>
</tr>
<tr>
<td>F</td>
<td>0.68</td>
<td></td>
<td>0.32</td>
<td>-</td>
</tr>
<tr>
<td>G</td>
<td>0.62</td>
<td></td>
<td>0.58</td>
<td>-</td>
</tr>
<tr>
<td>H</td>
<td>0.59</td>
<td></td>
<td>0.41</td>
<td>-</td>
</tr>
<tr>
<td>I</td>
<td>-</td>
<td></td>
<td>0.48</td>
<td>0.52</td>
</tr>
</tbody>
</table>

Fig. 12. A scanning electron microscopy secondary image of the austenite dendrites present in the porosity of the pig iron nugget made at furnace temperature of 1,450°C and residence time of 22 min.

It can be seen from Figs. 9 and 10 that this increasing amount of iron carbide in the solid state appears to have penetrated into the austenite.24–31)

### Table 7. The phases and constituents present in the pig iron nugget microstructures.

<table>
<thead>
<tr>
<th>Phases present in the pig iron nugget microstructures</th>
<th>Constituents present in the pig iron nugget microstructures</th>
</tr>
</thead>
<tbody>
<tr>
<td>α ferrite (iron)</td>
<td>Eutectic cementite</td>
</tr>
<tr>
<td>Iron carbide (cementite)</td>
<td>Pearlite (former austenite dendrites)</td>
</tr>
<tr>
<td>Iron sulfide</td>
<td>Pearlite (former eutectic austenite)</td>
</tr>
<tr>
<td>Iron carbide (cementite) solid state formation</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 13. SEM secondary image of the pig iron nugget made at furnace temperature of 1,475°C and furnace residence time of 22 min.

### 3.4. Identification of the Phases and Constituents Present in the Pig Iron Nugget Microstructures—Scanning Electron Microscopy and X-ray Diffraction

The phases and constituents present in the pig iron nugget microstructure are shown in Table 7. These phases and constituents were identified utilizing scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) and X-ray diffraction analysis.

Secondary SEM image for the pig iron nugget produced at furnace temperature of 1,475°C and residence time of 22 min is shown in Fig. 13. The phases, iron sulfide, iron carbide, and α ferrite identified utilizing local chemical analysis (EDS) and their approximate chemical compositions are given in Table 8. Utilization of high magnification images enabled the determination and identification of presence of iron sulfide phase as shown in Fig. 13.

The X-ray diffraction pattern of the pig iron nuggets produced at furnace temperature of 1,450°C and residence time of 22 min is shown in Fig. 14. It can be seen from Fig. 14 that only iron (α ferrite) and iron carbide phases were identified from the pattern.
3.5. Apparent Density Measurements

The apparent density measurements of the pig iron nuggets produced at furnace temperature of 1450°C and 1475°C and residence times of 22 and 28 min utilizing water displacement method are shown in Table 9.

Apparent density values were used for the evaluation of the following material properties:

Degree of Metallization: Apparent density measurements were utilized as an indication of degrees of metallization. As the degrees of metallization increased, the apparent density values increased. The real density values of selected ferrous materials are shown in Table 10. It can be seen from Tables 9 and 10 that pig iron nuggets produced had comparable apparent density values (i.e. degrees of metallization) with blast furnace pig iron and white cast iron.

Porosity: The actual density of the nuggets was not determined because they were porous and pores were not continuous. No measurements were performed to determine the percent porosity. However, it was observed that the porosity did decrease with increasing furnace temperature and/or residence time as indicated microstructurally and by lowered standard deviation values for apparent density measurements. This allowed apparent densities and real densities to be more comparable as shown in Tables 9 and 10.

Slag Separation: The high apparent density values of the pig iron nuggets indicate that the low density slag (approximate slag density 2.5–3 g/cm³) was successfully separated from the metal (iron density 7.87 g/cm³).

It can be seen from Tables 9 and 10 that the apparent density of the pig iron nuggets produced was comparable with the blast furnace pig iron. This is due to complete separation of low density slag and high degrees of metallization.

3.6. Comparison of Pig Iron Nuggets with Blast Furnace Pig Iron

Pig iron nuggets produced at laboratory scale conditions had comparable physical and chemical properties with the blast furnace pig iron as shown in Table 11.

4. Conclusions

Pig iron nuggets which have comparable physical and chemical properties with blast furnace pig iron were produced at furnace temperatures of 1450 and 1475°C and residence times of 22 and 28 min.

Pig iron nuggets produced had comparable high apparent densities, high iron concentration and high degrees of metallization with blast furnace pig iron. Similar to blast furnace process through out the pig iron process the gangue minerals contained in the feed was fused into slag phase. This slag phase was separated from the metal by obtaining a metal melt. However, unlike blast furnace process the melting of the metal was accomplished by the carburization and subsequent melting temperature decrease.

In addition to these, the microstructure of the pig iron...
nuggets was investigated, the presence of following phases: iron (α ferrite), iron carbide, and iron sulfide and; microconstituents: eutectic cementite, pearlite, and iron carbide was determined. The pig iron nuggets had similar microstructures with low carbon, low silicon white cast iron microstructure, which is essentially the same as pig iron from a blast furnace.

REFERENCES