# PILOT PLANT SCALE DEVELOPMENT OF AN INNOVATIVE IRONMAKING PROCESS FOR USAGE OF LOW GRADED RAW MATERIALS AND CO<sub>2</sub> MITIGATION<sup>1</sup>

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#### Abstract

The 'R&D and preparatory research work for the blast furnace based innovative ironmaking technologies' has started in FY2006 for usage of low graded raw materials and CO<sub>2</sub> mitigation in ironmaking process. The production processes and reaction mechanisms in a blast furnace were studied for two innovative composites with iron oxide, metallic iron and carbon to enhance the carbon and CO<sub>2</sub> reactions during FY2006 and 2008. For further developments at a pilot plant scale, a new project, 'Development of innovative ironmaking process using low cost iron ore and coal' has been started by JFE Steel Corporation, Nippon Steel Corporation, Kobe Steel, Ltd. Sumitomo Metal Industries, Ltd. and major Japanese Universities. The project has been partly supported by the New Energy and Industrial Technology Development Organization, (NEDO) in FY2009 – 2010 and by the Ministry of Economy, Trade and Industry (METI) in FY 2011 - 2012. This paper describes key technologies, the production process of the 'Ferro-coke' at a 30t/d pilot plant and experimental evaluation and simulation model development for blast furnace charging. Construction of heating and briquetting facilities has completed and standard briquetting conditions such as binder addition ratio and briquetting pressure has been established by the continuous operation. The carbonization shaft furnace has completed its construction in September, 2011 and carbonization batch tests was successfully carried out in December. Successive operation of briguetting and carbonization will be carried in the end of FY2011 and Continuous production trials and charging tests to a blast furnace is planned in FY 2012 to find process feasibility of the production process and technical subjects of blast furnace charging.

**Key words:** CO<sub>2</sub> emission; Ironmaking; Blast furnace; Ferro-coke; Ore; Coal; Coke; Carbonization; Briquetting.

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 $CO_2$  mitigation and the stable supply of raw materials are two major issues for the Japanese steel industry. The Japanese steel industry has emitted about 37% (2007) of energy source  $CO_2$  in the business sector and promoted short term developments and investments to comply with the Japan Iron and Steel Federation's voluntary action plan in parallel with developing plans fordrastic  $CO_2$  mitigation programs for the post Kyoto protocol period.

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Trends in blast furnace operation are shown in Figure1, such as productivity, reducing agent ratio (RAR) and pulverized coal injection rate (PCR) since 1965. Many R&D and operational improvements were carried out to meet demands from economic, energy and resource conditions.

Blast furnace operation changed to the low RAR and high productivity operation with oil injection in the 1970's [1] and all coke operation in mid 1980's, and to the low coke ratio operation with high pulverized coal injection [2]. From the middle of 1990, low RAR operations, waste plastic injection to a blast furnace and usage in coke ovens [3,4] and natural gas injection [5] were carried out to reduce CO<sub>2</sub> emission while keeping relatively high crude steel production.

For mid and long term CO<sub>2</sub> mitigation, the joint project, 'Preparatory research work for the blast furnace based innovative ironmaking technologies' was actively promoted by the New Energy and industrial Technology Development Organization (NEDO) from FY2006 to FY2008. The production process and reaction mechanisms in a blast furnace were studied for the two innovative composites, ferro-coke with iron oxide, metallic iron and carbon and carbon contained agglomerates (CCA).

For development at pilot plant scale, a new project, 'Development of innovative ironmaking process using low cost iron ore and coal' started in FY2009. This paper describes key technologies and recent developments of the project.



Fig.1 Trends in blast furnace operation in Japan

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### 2.1 Basic Concept and Key Technologies of the Project

The basic concept of this innovative ironmaking process for RAR reduction is demonstrated in the O/Fe and  $CO_2/(CO+CO_2)$  diagram (Rist diagram) in Figure 2. Changes in the thermal reserve temperature, Tw and O/Fe at the W point will affect the reducing agent ratio of a blast furnace. Conventional operation improvements change the operation line from (1) to (2). Innovative agglomerates are expected to lower the thermal reserve temperature by their high reactivity induced by Fe-FeO catalytic effects. As the innovative agglomerates contained about 30% of metallic iron, the metallic iron contributes to move the O/Fe downward and results in the operation point shifting from (2) to (4) and reduction of the RAR. The Ferro-coke is an innovative agglomerate with high reactivity and contributes to the enhancement of iron ore reduction and a decrease in  $CO_2$  emission in the blast furnace process.











Fig.4 Concept of Innovative ironmaking process

The target composition of innovative agglomerates is in the marked area  $\alpha$  in Figure 3. It represents a carbon matrix with iron ore or dispersed metallic iron particles. The project aims to develop the production process and evaluation in a blast furnace of the composite of metallic iron and carbon, so called 'Ferro-coke' indicated in the diagram. Figure 4 explains concept of the innovative ironmaking process. Low graded coal and iron ore are crushed and sized, mixed and blended with binder and then formed to briquette. The briquette is charged to a shaft furnace for coking and reduction. Produced ferro-coke shows sufficient strength to the blast furnace charging after coking.

The Ferro-coke, the third burden material after sintered ore and coke, decreases the reducing agent ratio of a blast furnace and increases usage of low grade iron ore and coke. Four Japanese integrated steel mills and major Japanese universities such as Tohoku University and Osaka University have joined the three years project from FY2009. JFE Steel Corp. has engaged in the production process development of the ferro-coke with a pilot plant of 30 ton per day [6-9]. Nippon Steel Corp. and Sumitomo Metals have studied burden distribution technology and behavior of the Ferro-coke in a blast furnace.

An innovative ironmaking process consists of three key technologies, optimization of structure and compositions of agglomerates, carbonization and reduction process, and a blast furnace operation using the ferro-coke as shown in Figure 5. The first step is to establish optimized structure and composition of the ferro-coke by coal and ore blending, innovative binder development and optimization of briquetting process to obtain high strength products. Green briquette is charged to a shaft furnace directly heated by the circulating gas. Carbonization and reduction of the briquette are carried out in the furnace to produce the ferro-coke with a strength and reduction degree high enough for the blast furnace usage.

The ferro-coke is then charged to a blast furnace mixed with iron ore. The third technology consists of burden distribution control to place the ferro-coke at the optimum position in an ore layer and simulation model development to access overall performance of the blast furnace operation.





Fig.5 Development of innovative ironmaking process

### 2.2 Carbonization and Reduction in a Shaft Furnace

The coking and reduction temperature in the shaft furnace is the most important factor to attain both reduction ratio and strength. Green briquettes were heated at a heating rate of 5°C/min up to the coking temperature and held for 1 to 3 h. The reduction ratio and strength were measured after cooling in a N2 atmosphere. The reduction degree mainly depends on the coking temperature in the range of  $800 - 1,000^{\circ}$ C while the holding time slightly affects it as shown in Figure 6.

The heating pattern of a bench scale plant was decided based on similar experiments for various blending and coking conditions. The bench scale plant of a production capacity 0.5 t/day shown in Figure 7 was designed to realize the heating pattern by means of an electric heater. Continuous experiments for more than 24 h have confirmed the process reliability at a bench scale.



Fig.6 Effect of coking temperature and holding time on the reduction degree



Fig.7 The bench scale plant of 0.5 t/d and its products.

## 2.3 Process Flow and Recent Trials of The Pilot Plant of Ferro-Coke

Process flow of the pilot plant is shown in Figure 8. The pilot plant is designed with a capacity of 30 t/d and has been built at Keihin Region, JFE Steel Corp. Iron ore and coal are crushed and dried, then mixed to form briquettes. Green briquettes are transported to a shaft furnace with the gas heating system for carbonization and reduction.

Recent over view of our pilot plant is shown in Figure 9. From the left hand side, the plant consists of coal and ore receiving hopper, crushing plant of coal, drying and heating rotary kiln, mixing and briquetting machine, carbonization shaft furnace, gas recycling and processing plant and stock yard of product.

In March, 2011, the plant construction has completed up to the briquetting machine. Commissioning operation and trials were conducted to establish standard preprocessing and briquetting operation. In September, 2011, all pilot plant facilities have finished their construction and started commissioning operations. During the trial, we have completed batch trials of carbonization. 6<sup>th</sup> International Congress on the Science and Technology of Ironmaking - ICSTI 42<sup>th</sup> Ironmaking and Raw Materials Seminari 42<sup>st</sup> Seminário de Redução de Minério de Ferro e Matérias-primas 13<sup>th</sup> Brazilian Symposium on Iron Ore / 13<sup>st</sup> Seminário Brasileiro de Minério de Ferro



Fig.8 Process flow of a pilot plant of 30t/d for the ferro-coke production



Fig.9 Overview of a pilot plant of 30t/d for the ferro-coke production





Ferro-coke Coke

Metal:-50 μ m



Fig.10 Briquetting operations at the pilot plant of ferro-coke

Figure 10 shows a typical briquetting operation at the pilot plant. After mixing more than two minutes, the binder, SOP was added and the mixture was discharged to the briquetting machine. Briquetting power started increasing soon after the discharge and was controlled to the target range, 160 - 185t, 4 - 4.5 t/cm. Produced green briquettes are shown in Figure 10. Although, there remain some minor problems in the green briquette quality, such as relatively thick briquette edge and variations briquetting pressure, the successive operation up to briquetting has completed in a pilot plant scale. Further process optimization will be tried for quality improvements, cost reduction and stable operations.

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During the commissioning operation, carbonization trials are carried out at batch charging of green briquette, 400 kg. We have produced ferro-coke particles in a relatively good shape without adhesion of briquette as shown in Figure 11. We need some modification and improvement in our carbonization furnace. Successive operation of briquetting and carbonization will be completed in this fiscal year and continuous operation is planned in FY 2012 and charging of the ferro-coke to a blast furnace will be carried out to find operational subjects of blast furnace usage.

### 2.4 Development of Innovative Binder for Ferro-Coke

In the ferro-coke production, two conventional binders are used, coal-tar pitch from coke oven, SOP, and pitch from petro-chemical industry, asphalt pitch, to improve green briquette and ferro-coke strength respectively as shown in Figure 12. As both materials are by-products, production capacities and properties are limited by the production of main streams. Objectives in the project are to develop a new binder customized to the ferro-coke based on solvent extraction of coal, so-called Hypercoal Process.

The process consists of slurry make-up, extraction with two-ring aromatic compounds, solid liquid separation, solvent recovery, and briquetting.

Advantages of the Hypercoal process are as follows:

- Solvent recycle.
- No hydrogen is needed.
- Mild reaction condition less than 2 Mpa and 400°C.
- High yield of the extracts.

However, the properties at this stage are optimized for coke oven additives and the properties should be customized to the requirement of the ferro-coke.

Two modifications for the ferro-coke application were tested by changing the heat treatment and extraction temperature. As shown in the Figure 13, high temperature extraction of 410°C enhances the fluidity of binder itself and the mixture of non-coking coal and binder. The fluidity by a Gieseler plastometer was suppressed by the heat treatment after extraction as shown in Figure 14. By increasing the heat treatment temperature to 450, 470°C, the Gieseler fluidity curves shift to a higher temperature and peak height is suppressed.

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Fig.12 Development of innovative binder for Ferro-coke

Solvent to coal ratio: 1; Residence time: 30 min





Fig.13 Effects of the extraction temperature on the fluidity of the binder

Fig.14 Effects of the heat treatment temperature on the Gieselar fluidity curves of the binder

### 2.5 Effect of Layer Structure on Ore Reduction

The thermal reserve temperature was measured in the BIS furnace by Nomura et al.[10] with the Fe contained formed coke, ferro-coke. Performance of the ferro-coke has been studied with blast furnace inner reaction simulator (BIS) at sinter layer thickness ranged between 30 and 40 mm. At the replacement ratio of carbon to ferro-coke, 30%, five mixing conditions of ferro-coke were tested, base condition, homogeneously mixed in sinter layer, segregated at the upper sinter layer, homogenously mixed in coke layer and segregated of conventional coke at the upper layer for references. The highest shaft efficiency is observed at the upper layer blending of ferro-coke and hematite based gas utilization ratio reaches the highest value of 55.5%. The thermal reserve zone temperature is reduced to 959°C. They demonstrates that ferro-coke in sinter is better in lowering the thermal reserve zone temperature compared with in coke.

As reduction gas composition will change in the sinter and coke layer, we have carried out at larger layer thickness in plant condition. Test procedure in large layer thickness is shown in Figure 15. The reaction tube, 65 mm ID and two 330 mm test sections are heated with electric heater at a constant heating rate. The gas composition was changed to simulate blast furnace condition. Four sets of mixing conditions in the ore layer were tested, homogenously mixed, lower layer mixed, upper layer mixed and upper layer mixed with mild segregation. Reduction degree of sinter is the highest in the case of homogenously mixed or segregated mixed in the lower part of sinter layer in Figure 16. The highest carbon consumption was observed at the homogenously mixed condition. Two results in different equipment indicate that homogenously mixed in ore layer is the best arrangement of Ferro-coke.

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Next step of research is how to charging ore and ferro-coke with bell-less top charging system to obtain the homogenously mixed condition.



### 2.6 Blast Furnace Operation Using the Ferro-Coke

Sumitomo Metals and Kyushu University has been developing a numerical blast furnace model to evaluate the effect of ferro-coke on various in-furnace reactions. In a blast furnace, ore, coke and ferro-coke are charged at top. These three materials are tracked separately during heating and reaction amongst these phases in a virtual blast furnace as shown in Figure 17. Rate constants of ferro-coke are measured by Kyushu University and modeled in simple rate equations.

Preliminary calculation was carried out to evaluate the effect of ferro-coke charging, 100 kg/tp, on in-furnace state by introducing rate constants measured by Kyushu University.

Decrease in the thermal reserve zone temperature are clearly found in Figure 18, however, further analytical examinations will be needed to simulate changes in thermal reserve zone temperature and gas utilization rates observed in the BIS furnace and fixed bed furnace for various mixing conditions.

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Fig.18 Effect of Ferro-coke (100kg/pt) on in-furnace state. Sinter : JIS-RI=68.4%, Normal coke : CRI=30.0%, Ferrocoke : (Fix.C=59.9%, T.Fe=26.5%)

The innovative ironmaking process development project reaches the final year of four years project in FY2012. R&D objectives in this fiscal year are as follows:

• Optimization of production conditions of an innovative binder from Hypercoal process.

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- Successive operation of briquetting and carbonization will be carried in the end of FY2011 and Continuous production trials and charging tests to a blast furnace is planned in FY 2012 to find process feasibility of the production process and technical subjects of blast furnace charging.
- Evaluation of blast furnace operation with ferro-coke.
- Development of charging condition for the optimum layer structure.

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