

Available online at www.synsint.com

Synthesis and Sintering

ISSN 2564-0186 (Print), ISSN 2564-0194 (Online)



Research article

Optimization of direct reduction in tunnel furnace using different resources of ferrous oxides

Sara Ahmadi ^a, Hossein Nuranian ^{b,*}^a Construction and Minerals Research Group, Technology and Engineering Research Center, Standard Research Institute, Karaj, Alborz, Iran^b Ceramic Department, Materials and Energy Research Center, Karaj, Alborz, Iran

ABSTRACT

Nowadays it is highly desired to maximize using of existing resources and recycle waste materials. The by-product of steel being hot rolled is mill scale that disposing of it as waste material has environmental effects. Therefore, the use of mill scale in iron production is economically and environmentally beneficial. In the current work, an attempt has been made to use mill scale and iron concentrate which are not applicable to pelletized, in the reduction process with tunnel kiln for iron manufacturing. Non-coking coal and limestone were utilized as reducing agents. The reducer to ferrous oxide ratio was kept constant during the reduction tests. The reduction process was carried out in a crucible at 1150 °C. The analyses of the metal Fe content in the reduced samples show that the mill scale can be used successfully in the direct reduction process to produce sponge iron. In the rolling mill scale-iron pellet, iron concentrate-iron pellet, and iron concentrate-mill scale mixtures, the compositions 70MS-30IP, 70IC-30IP, and 70IC-30MS were optimum. The result of XRD and STA results revealed that the optimal heat treatment setting for reducing utilized ferrous oxide mixtures is 1150 °C for 1 h.

© 2023 The Authors. Published by Synsint Research Group.

KEYWORDS

Direct reduction
Iron pellet
Iron mill scale
Iron concentrate
Tunnel furnace



1. Introduction

Steel production via the traditional blast furnace and basic oxygen furnace routes necessitates the use of high-quality coking coal. Since many of the hydrocarbons released during the coking process are dangerous, the production of coke is particularly troublesome for the environment [1, 2]. Furthermore, not all types of coal are suited for the manufacturing of coke. Modern direct reduction iron-making (DRI) techniques are replacing outdated blast furnace methods. Directly reduced iron, also known as sponge iron, which has a low tramp element content and a stable component as a replacement for scrap, has been proven to be an excellent feed in electric arc furnace (EAF) steelmaking [3–5]. The most valuable benefit of DRI technologies is that they do not require coke production or sintering, both of which are expensive and have negative environmental effects [6, 7].

There are two commercially accessible DR procedures based on the generation of the reducing gas: gas-based and coal/oil-based. A mixture of natural gas and off-gas from the reducing furnace is chemically reformatted to create a gas that is high in hydrogen and carbon monoxide to create the reducing gas in the gas-based DR process. However, in the coal/oil-based DR process, the reducing gas is produced in the reduction zone of the furnace, which is commonly a rotary kiln, from hydrocarbons (mainly coal, but occasionally oil and natural gas). In India and China, the coal-based DR technique is increasingly common. According to data, India is the world leader in DRI production, producing approximately 17.8 million metric tons of DRI in 2013, accounting for almost one-fourth of global DRI output [8–11].

* Corresponding author. E-mail address: h.nuranian@merc.ac.ir (H. Nuranian)

Received 25 November 2022; Received in revised form 29 January 2023; Accepted 30 January 2023.

Peer review under responsibility of Synsint Research Group. This is an open access article under the CC BY license (<https://creativecommons.org/licenses/by/4.0/>).
<https://doi.org/10.53063/synsint.2023.31130>

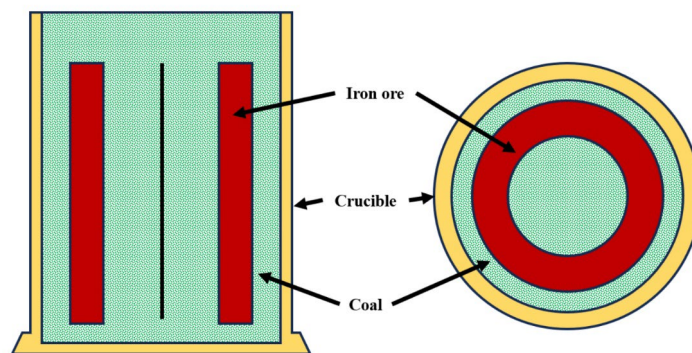


Fig. 1. Material charging in refractory crucibles in the TKDRI process.

Tunnel kiln technology, among other gas- and coal-based DRI production technologies, combines cheap material costs and minimal capital investment with simplicity, operational flexibility, and high-quality output [12, 13].

Tunnel kilns direct reduction (TKDRI) was first used for iron oxide reduction in 1908. Production of sponge iron from beneficiated iron ore fines is currently more common in China [12, 13]. A stationary bed of material moves through the preheating, reduction, and cooling stages of this process. From the feed end of the tunnel kiln, coal, and iron ore are put into a refractory crucible and packed in a tubular structure. It moves through the kiln at a set speed before emerging at the other end. This speed can be thought of as a preheating rate, reduction time, cooling rate, as well as production rate, depending on the length of the various parts of the tunnel kiln. Crucible travel speed is a crucial process parameter [14, 15]. A schematic of the tunnel kiln DRI process for material charging in crucibles is shown in Fig. 1.

The direct reduction of iron in a tunnel kiln is a volumetric process in which raw material consumption is determined by the volumetric capacity of refractory crucibles. Non-coking coal with less than 10% volatile material, up to 25% ash content, more than 65% carbon, and particle sizes that can commonly range from 0.5 mm to 7 mm is required for this procedure. It is possible to infer that these characteristics are required for coal to produce reducing agents and to provide a permeable structure for the transport of reactant and product gases during the reduction process [11, 13, 16]. Coal amount is the major parameter impacting the direct reduction process in the tunnel kiln, according to laboratory and industrial tests [16].

The most generally utilized raw materials in the manufacturing of sponge iron via the DR process include high-grade lump ore, iron ore fines concentrate, iron ore pellets, rolling mill and reductant, and limestone powder.

Coal serves as both a reductant and a fuel in the process, supplying heat to keep the appropriate temperature within the Kiln (1223–1323 K). Calcium carbonate (limestone powder), which acts as a catalyst and aids in scavenging sulfur, is combined with coal powder, and the gasification reaction is accelerated. CaCO_3 breaks down into CaO and CO_2 [9, 17].

The iron pellet is a thermally agglomerated material created by heating a varied mixture of iron ore, olivine, bentonite, dolomite, and other iron-bearing minerals in the 1250 °C to 1350 °C range. Iron ore concentration is used to make iron metal pellets. Iron concentrate is a

pulverized iron ore that cannot be employed in purification or direct reduction units due to its small size.

Steel manufacturing produces solid byproducts called rolling mill scale. It is an oxide layer of flaky, thin particles with an average thickness of less than 2 mm. It comprises of iron, iron oxides (hematite, magnetite, wusite, etc.), alkaline compounds, and traces of non-ferrous elements [18–20].

Given the non-degradability of the mill scale, disposing of it as waste has environmental consequences and incurs significant expenditures. A novel technique to benefit from the waste output of the steel manufacturing industry is the reduction of mill scale to sponge iron [21–24]. The utilization of mill scale in iron production is economically and environmentally beneficial.

The major goal of this study is to examine the effects of using various iron sources, such as rolling mills and iron concentrates, as alternatives to iron pellets on the reduction process and metallization of iron oxides. Further research has been done on heat treatment time and temperature to optimize the DR process.

2. Experimental procedure

2.1. Materials and method

The three raw materials used as iron sources in this study were iron fine-grained pellets, iron concentrate, and rolling mill scale. The specifications of the raw materials used as iron sources are summarized in Table 1. Non-coking coal was used as a reductant along with limestone.

Binary iron source combinations in the form of pellets with an approximate diameter of 2 cm were buried in the crucible and reduced in a lab-scale kiln. The weight ratio of the sample to the reducing agent was 2 to 3. The reductant was composed of 15 wt% calcium carbonate and 85 wt% coal.

2.2. Characterization technique

An X-ray diffractometer (Simense, D500) with a copper cathode was used to determine the crystallinity of the reduced samples. The measurements were performed in 2θ intervals of 20–45 ° with a scanning step of 0.02 ° and detecting a time constant of 1 s in each step. Simultaneous thermal analysis (STA) was carried out using an STA-1640(PL-STA-1640) in the air. Alumina powder was used as the reference material and the heating rate was 10 °C/min.

Table 1. Characterization of used iron sources.

Material	Total Fe	Fe (metal)	MgO	FeO	Al ₂ O ₃	SiO ₂	S	CaO	P	TiO ₂
Iron pellet	65.64	-	2.04	1.64	0.58	2.75	0.051	0.69	0.037	0.07
Iron concentrate	66.7	-	2.3	27.2	0.4	2.1	0.8	0.6	0.05	0.09
Rolling mill Scale	72.05	49.23	0.032	-	0.45	0.53	0.042	0.08	0.038	0.039

The amount of metallic iron in the reduced samples was determined by a chemical method according to the Indian standard IS 15774:2007, Sponge iron/direct reduced iron (DRI) hot briquette iron (HBI) and cold briquette iron (CBI) for steelmaking-specification. The tapped density of samples was determined according to ASTM B-527, the standard test method for the tapped density of metal powders and compounds.

3. Results and discussion

3.1. Reduction temperature

Fig. 2 depicts STA curves of a combination of 40 wt% iron pellet and 60 wt% reductant. It is implied from Fig. 2 that the 10% weight loss occurs at about 750 °C. This temperature is in accordance with the endothermic peak of the DTA curve. Apparently, this temperature could be assigned to the decomposition temperature of calcium carbonate.

On the other hand, sample weight loss follows an increasing trend of up to 30% at 1150 °C. Interestingly, the slope of the DTA curve starts to change after 1000 °C. Seemingly, this effect is related to the beginning of an exothermic peak that could be related to the re-oxidation of the sample in an air atmosphere. Therefore, 1150 °C was selected as the appropriate reduction temperature.

3.2. Reduction time

It should be noted that the reduction time at the DR process in the industry scale is about 50 hours. Given the volume and size of the samples and laboratory furnaces, in order to optimize the reduction period, the samples were heated at the temperature of 1150 °C with soaking times of 1, 2, 5, and 10 h.

The sample compositions that were utilized to assess the reduction time are listed in Table 2. Iron pellet, mill scale, and iron concentrate are all denoted by the codes IP, MS, and IC, respectively.

Fig. 3 depicts the XRD pattern of single-component and two-component raw material mixtures after reduction at 1150 °C at various times. The peaks related to metal iron are signed as Fe, the other peaks in patterns are associated with different forms of iron oxides. As can be seen in the figure, increasing the soaking duration of heating declines the amount of metallic iron and causes re-oxidation of the samples. One of the most critical issues in the DR method is the reversal of the reduction reaction at high temperatures. The reverse reaction and re-oxidation are faster and easier in the lab due to the greater surface-to-volume ratio of the samples. As a result, in this study, the reduction at 1150 °C for 1 hour was selected as the optimal reduction condition for the direct reduction of the iron sources.

3.3. Composition optimization

As was already indicated, the goal of this study is to

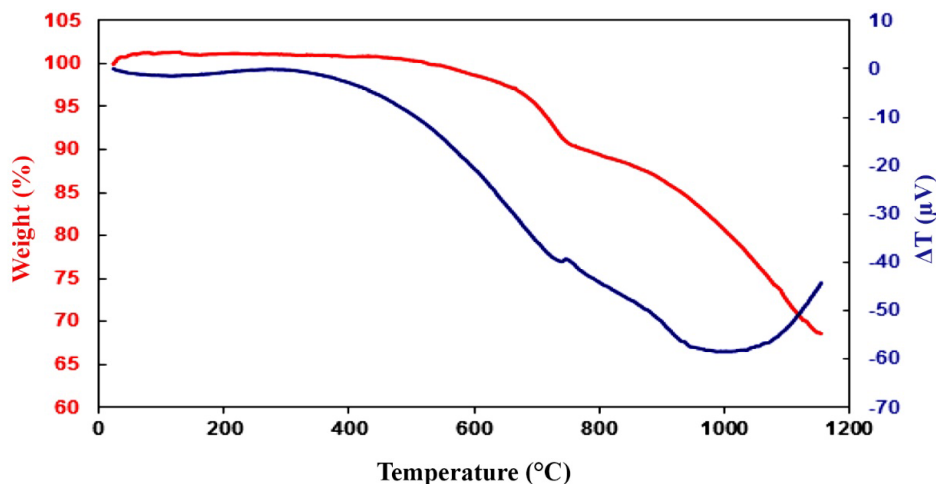


Fig. 2. The STA micrograph of the mixture of 40% iron pellets and 60% reductant.

Table 2. Sample compositions for reduction at 1150 °C.

Sample code	Iron concentrate (wt%)	Mill scale (wt%)	Iron pellet (wt%)
IP	0	0	100
MS	0	100	0
IC	100	0	0
50IP-50MS	0	50	50
50MS-50IC	50	50	0
50IC-50IP	50	0	50

use iron-containing powdered wastes that cannot be pelletized due to their flaky morphology. As a result, binary combinations of pellet, rolling mill scale, and iron concentrate were devised to optimize the composition in order to maximize the utilization of rolling mill scale and iron concentrate. Table 3 describes the reduced sample composition, the amount of metallic iron recovered and the tapped density of the samples. Fig. 4 also shows the results of the iron content for a better comparison of the reduction process. In order to safely handle the samples in real and non-laboratory conditions, the samples should have acceptable strength; otherwise, the sample will be crushed and mixed with the reduction agent. Consequently, the composition with the highest metal iron amount and appropriate density was selected as the optimal composition in each binary mixture of raw materials.

The results demonstrate that the compositions 70MS-30IP, 80IP-20IC, and 70IC-30MS are the optimal composition in the

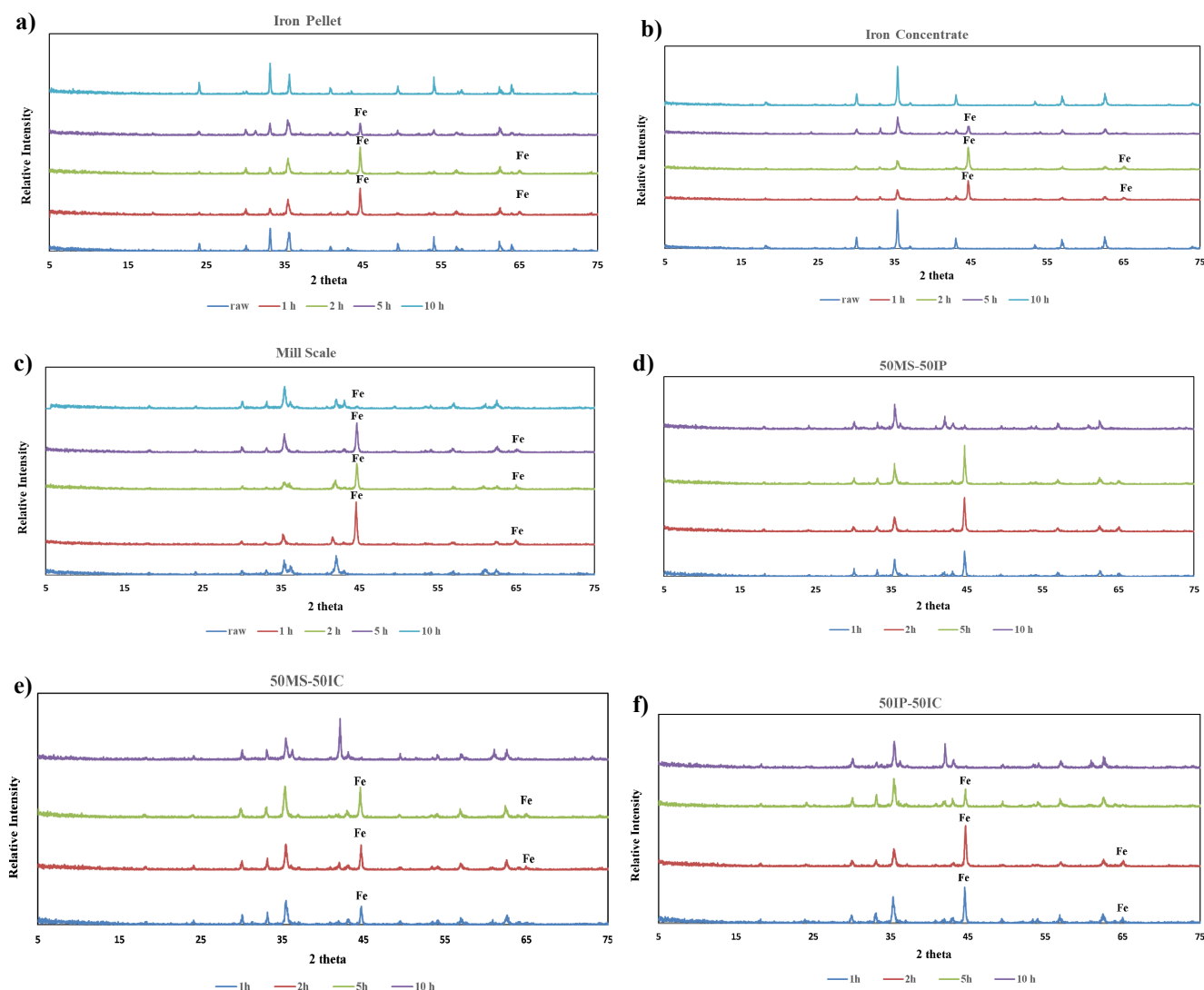


Fig. 3. XRD patterns of single-component and two-component mixtures of iron sources after reduction at 1150 °C for different times.

Table 3. The amount of metal Fe and bulk density in the single and two-component mixtures of iron sources.

Sample code	Mill scale (wt%)	Iron pellet (wt%)	Iron concentrate (wt%)	Fe-metal (wt%)	Tapped density (g/cm ³)
MS	100	0	0	62.5	2.83
90MS-10IP	90	10	0	37.47	2.78
80 MS-20IP	80	20	0	37.5	2.81
70 MS-30IP	70	30	0	73.49	2.81
60 MS-40IP	60	40	0	36.8	2.77
50 MS-50IP	50	50	0	43.24	2.79
40 MS-60IP	40	60	0	43.24	2.81
30 MS-70IP	30	70	0	42.74	2.73
20 MS-80IP	20	80	0	42.98	2.77
10 MS-90IP	10	90	0	42.15	2.75
IP	0	100	0	71.71	2.74
90IP-10IC	0	90	10	67.57	2.68
80IP-20IC	0	80	20	73.78	2.85
70IP-30IC	0	70	30	61.05	2.93
60IP-40IC	0	60	40	54.97	2.93
50IP-50IC	0	50	50	68.29	3.03
40IP-60IC	0	40	60	54.49	3.03
30IP-70IC	0	30	70	66.58	3.04
20IP-80IC	0	20	80	63.67	3.07
10IP-90IC	0	10	90	62.32	3.09
IC	0	0	100	72.56	2.97
90IC-10MS	10	0	90	46.28	2.98
80IC-20MS	20	0	80	44.4	3.00
70IC-30MS	30	0	70	68.43	3.01
60IC-40MS	40	0	60	10.27	2.97
50IC-50MS	50	0	50	66.27	2.99
40IC-60MS	60	0	40	53.89	2.93
30IC-70MS	70	0	30	50.34	2.92
20IC-80MS	80	0	20	34.42	2.89
10IC-90MS	90	0	10	22.52	2.87
MS	100	0	0	62.5	2.83

rolling mill scale-iron pellet, iron pellet-iron concentrate and iron concentrate-rolling mill scale mixture respectively, due to the highest amount of metal Fe after reduction.

The process of direct reduction of iron oxides in the tunnel furnace and the production of sponge iron depends on the rate of reactions and heat transfer, which in turn depends on the particle size and morphology of the raw materials, permeability, density, and activity of the materials as well as the heat treatment condition of the process. Furthermore, the type and size of the furnace, the furnace atmosphere, the pressure of

gases inside the pores, and the composition of gases, particularly the amount of CO gas between the pores, are other influential factors in the reduction of iron oxide.

It is clear that the reduction of iron oxide is a challenging process that is simultaneously controlled by several factors. According to the morphology and particle size of the iron sources, it appears that the permeability of the mixture controls the amount of reduction and the ideal composition in the binary combination of the raw materials.

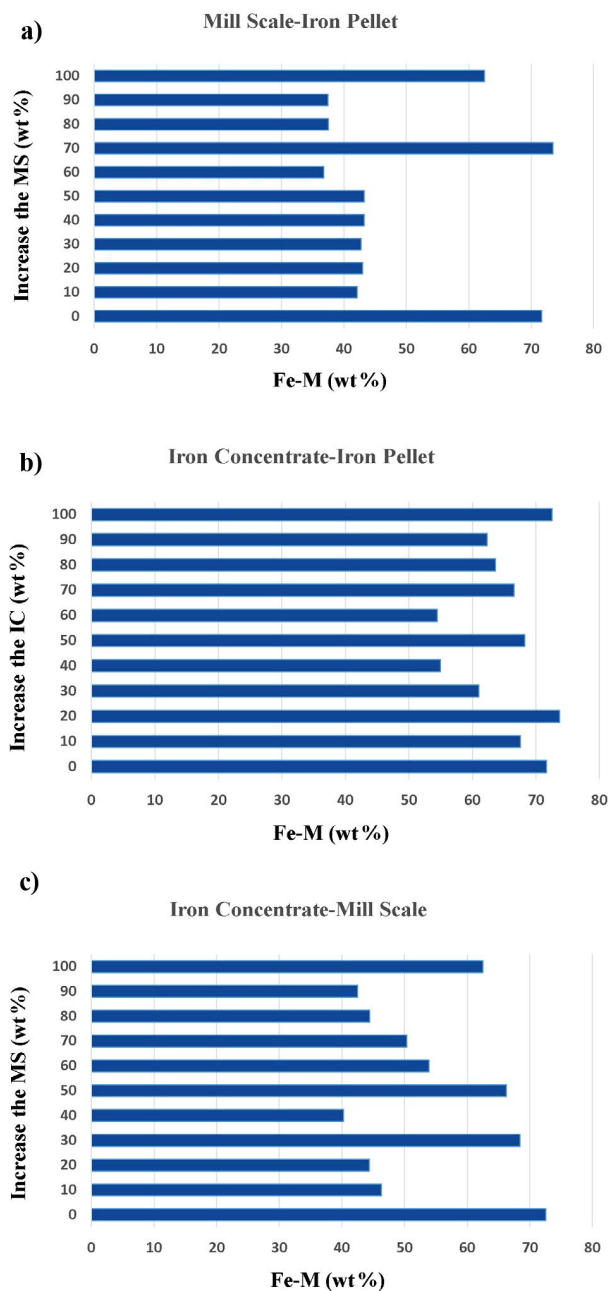


Fig. 4. The amount of metal Fe in the two-component mixtures of iron sources a) mill scale-iron pellet, b) iron pellet-iron concentrate, and c) iron concentrate-mill scale.

4. Conclusions

The findings of this study demonstrate that the reduction and manufacturing of sponge iron can be successfully carried out using powdered ferrous oxide wastes that cannot be pelletized. The optimum temperature and time for the DR process in the laboratory condition were determined at 1150 °C for one hour. Additionally, the optimal amount of rolling mill scale and iron concentrate as a substitute for iron pellets was obtained based on the highest amount of metallic iron that was determined based on the chemical method.

CRediT authorship contribution statement

Sara Ahmadi: Visualization, Writing – original draft, Writing – review & editing.

Hossein Nuranian: Conceptualization, Methodology, Project administration, Formal Analysis.

Data availability

The data underlying this article will be shared on reasonable request to the corresponding author.

Declaration of competing interest

The authors declare no competing interests.

Funding and acknowledgment

The authors acknowledge with grateful appreciation the assistance and financial support provided by Golgozar Mining and Industrial Company, under project No. 97/3580.

References

- [1] A. Singh, P. Jain, A study on the accretion formation in DRI kilns and possible ways for its reduction, *IOSR J. Mech. Civ. Eng.* 12 (2015) 98–103. <https://doi.org/10.9790/1684-126598103>.
- [2] M. Kumar, H. Baghel, S.K. Patel, Reduction and Swelling of Fired Hematite Iron Ore Pellets by Non-coking Coal Fines for Application in Sponge Ironmaking, *Miner. Process. Extr. Metall. Rev.* 34 (2013) 249–267. <https://doi.org/10.1080/08827508.2012.656776>.
- [3] J. Yang, T. Mori, M. Kuwabara, Mechanism of Carbothermic Reduction of Hematite–Carbon Composite Pellets, *ISIJ Int.* 47 (2007) 1394–1400. <https://doi.org/10.2355/isijinternational.47.1394>.
- [4] J. Aota, L. Morin, Q. Zhuang, B. Clements, Direct reduced iron production using cold bonded carbon bearing pellets Part 1 – Laboratory metallization, *Ironmak. Steelmak.* 33 (2006) 426–428. <https://doi.org/10.1179/174328106X118053>.
- [5] D. Zhu, V. Mendes, T. Chun, J. Pan, Q. Li, et al., Direct Reduction Behaviors of Composite Binder Magnetite Pellets in Coal-based Grate-rotary Kiln Process, *ISIJ Int.* 51 (2011) 214–219. <https://doi.org/10.2355/isijinternational.51.214>.
- [6] O.P. Chukwuleke, J. Cai, S. Chukwujekwu, S. Xiao, Shift from coke to coal using direct reduction method and challenges, *J. Iron Steel Res. Int.* 16 (2009) 1–5. [https://doi.org/10.1016/S1006-706X\(09\)60018-2](https://doi.org/10.1016/S1006-706X(09)60018-2).
- [7] A. Sarangi, B. Sarangi, *Sponge Iron Production in Rotary Kiln*, New Delhi PHI Learning, Pvt. Ltd. (2011).
- [8] L. Lu, J. Pan, D. Zhu, Quality requirements of iron ore for iron production, *Iron Ore: Mineralogy, Processing and Environmental Sustainability*, Woodhead Publishing. (2015) 475–504. <https://doi.org/10.1016/B978-1-78242-156-6.00016-2>.
- [9] R. Sen, S. Dehiya, U. Pandel, M.K. Banerjee, Utilization of low-grade core for direct reduction of mill scale to obtain of sponge iron: effect of reduction time and particle size, *Procedia Earth Planet. Sci.* 11 (2015) 8–14. <https://doi.org/10.1016/j.proeps.2015.06.003>.
- [10] K.K. Prasad, H. Shankar Ray, Advances in Rotary Kiln Sponge Iron Plant, *New Age International.* (2009).
- [11] A. Chatterjee, *Sponge Iron Production by Direct Reduction of Iron Oxide*, Prentice-Hall Of India Pvt Ltd. (2010).
- [12] S.C. Khattai, G.G. Roy, Sponge iron production from ore – coal composite pellets in tunnel kiln, *India Iron Production*, Project. (2013).

- [13] H.A. Mashhadi, A. Rastgoo, J.V. Khaki, An investigation on the reduction of iron ore pellets in fixed bed of domestic non-coking coals, *Int. J. Iron Steel Soc. Iran.* 5 (2008) 8–14.
- [14] N. Toloue Farrokh, M. Askari, T. Fabritius, Behavior of three non-coking coals from Iranian's deposits in simulated thermal regime of tunnel kiln direct reduction of iron, *J. Anal. Appl. Pyrolysis.* 123 (2017) 395–401. <https://doi.org/10.1016/j.jaap.2016.10.028>.
- [15] G.S. Upadhyaya, *Powder Metallurgy Technology*, Cambridge International Science Publishing, Cambridge. (1997).
- [16] M.J. Keyser, M. Conradie, M. Coertzen, J.C. Van Dyk, Effect of coal particle size distribution on packed bed pressure drop and gas flow distribution, *Fuel.* 85 (2006) 1439–1445. <https://doi.org/10.1016/j.fuel.2005.12.012>.
- [17] K. Jouhari, P. Datta, *Proceedings of the seminar on experimental approaches in pyrometallurgical research*, chapter- Effect of Calcium Carbonate on degree of Reduction in Mill Scale by Coal, Allied Publishers. (2000).
- [18] M.I. Martin, F.A. Lopez, M.E. Rabanal, J.M. Torralba, Production of Sponge Iron Powder by Reduction of a by-product of Steel making Industry, PM2010 World congress-Waer Atomized Powders. (2010).
- [19] M.I. Martin, F.A. Lopez, M.E. Rabanal, J.M. Torralba, Obtainment of Sponge Iron by Reduction of a by-product, National Conference on Advances Materials Recycling and Eco-Energy. (2009).
- [20] O. Benchiheb, S. Mechachti, S. Serri, M.G. Khalifa, Elaboration of Iron powder from Mill Scale, *J. Mater. Environ. Sci.* 1 (2010) 267–276.
- [21] L. Camci, S. Aydin, C. Arslan, Reduction of iron oxides in solid wastes generated by steelworks, *Turk. J. Environ. Eng. Sci.* 26 (2002) 37–44.
- [22] J.-W. Park, J.-C. Ahn, H. Song, K. Park, H. Shin, J.-s. Ahn, Reduction characteristics of oily hot rolling mill sludge by direct reduced iron method, *Resour. Conserv. Recycl.* 34 (2002) 129–140. [https://doi.org/10.1016/S0921-3449\(01\)00098-2](https://doi.org/10.1016/S0921-3449(01)00098-2).
- [23] A.D. Pelton, C.W. Bale, *Direct Reduced Iron: Technology and Economics of Production and Use*. Iron & Steel Society, Warrendale. (1999).
- [24] V.B. Akimenko, I.A. Gulyaev, M.A. Sekachev, O.Y. Kalashnikova, Reduced Iron powder: Manufacturing problems and Prospects, *Steel Transl.* 41 (2011) 622–626. <https://doi.org/10.3103/S0967091211070023>.