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THE EFFECT OF REDUCTION TIME AND SIZE DISTRIBUTION OF MIXED IRON ORE WITH COCONUT SHELL CHARCOAL ON THE PERCENTAGE OF METALLIZATION BY USING A ROTARY KILN

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ABSTRACT

Steel is an important material that is widely used and its development has occurred in conjunction with the history of mankind over the last two centuries. In general, the steelmaking process has been done through a combination of a direct reduction process and an Electric Arc Furnace (EAF) or an indirect reduction process and a Basic Oxygen Furnace (BOF). The combination of the steelmaking processes can be adapted to the specific conditions of Indonesian local iron ore. In Indonesia, the raw material reserves of iron and steel making are quite large, but spread over several islands. UPT BPML LIPI in cooperation with the Department of Metallurgical Engineering UNTIRTA conducted research to improve the economic value of the local iron ore in South Lampung Regency. The total amount of primary iron ore resources in South Lampung is estimated to be in the region of 11 million tons. South Lampung Regency iron ore is primary iron ore with a content of pure magnetite and magnetite-containing impurity silica levels ranging from 40-65% Fe in total. South Lampung Regency low-grade iron ore has the potential to be reduced by using a rotary kiln. A rotary kiln is a tool used to reduce low-grade iron ore and produce sponge iron with a high metallization. This process is in accordance with the Indonesian government policies that regulate the minimum value of percentage of sponge iron metallization for export, i.e. 85%. In this research, sponge iron is made of a mixture of Lampung iron ore pellets with coconut shell charcoal as a reduction agent. The composition of coconut shell charcoal is about 20%, which will determine the optimum amount of South Lampung iron ore pellets in the mixture. In addition, during the reduction process, the residence time of pellets in the rotary kiln is observed in order to obtain the optimal percentage of metallization. The method used in this research was the direct reduction process using a pilot-scale rotary kiln with the variables related to residence time (1, 2 and 3 hours) for the pellets and to the diameter of the pellets (-12+8mm and -20+12mm). Meanwhile, the reduction temperature was fixed, i.e. 1100^{o}C. The maximum metallization of sponge iron achieved at a residence time of 3 hours was 99.50% for the average pellet diameter of (-12+8mm).

Keywords: Coconut shell charcoal; Direct reduction; Percent metallization; Rotary kiln; Sponge iron

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1. INTRODUCTION

Steel is an important material that is widely used and its development has occured in conjunction with the history of mankind over the last two centuries. At present, the world steel production has increased from 717 million tons in 1980 to 1.237 million tons in 2006. The rate of growth projection for steel production is expected to be approximately 5% per year. Therefore, the production of steel can be predicted to reach 1.5 billion tons in 2015 and 2 billion tons in 2020, respectively (Arabinda, 2011).

The Indonesian government promulgated Regulation No.7 Year 2012, released by the Ministry of Energy and Mineral Resources (MoEMR) or *(ESDM)*, was related to Regulation No. 4, Year 2009, known as ("Minerba's Law") on Minerals and Coal Mining. MoEMR *(ESDM)* Regulation No.7 Year 2012 on the Added Value of Minerals Through Mineral Processing banned the export of unprocessed raw minerals from May 7, 2012 to January 2014. Raw materials or ore in its natural state is known as Run of Mine (ROM) or fresh ores. Furthermore, the Goverment of Indonesia issued Regulation 1 Year 2014 (GR1/2014) and the MoEMR (ESDM) published ESDM 1/2014, enacted on 11 January 2014 for ROM which contained provisions regarding obligations specifically for 11 ores, (namely 11 metals, 8 non-minerals, and 19 stones), to be exported, including iron ore and iron sand. (Winzenried et al., 2014; Lubis et al., 2014). As a result for iron and steel resources, the value-added processing of iron ore must occur using, for instance, beneficiation or reduction processes. In addition, iron ore concentrate produced by a beneficiation process must contain a minimum $\geq$ 62% Fe, while sponge iron and pig iron, which are produced from a reduction process must contain a minimum $\geq$ 75% Fe for sponge iron and $\geq$ 90% Fe for pig iron, respectively (ESDM 1/2014). Moreover, the total amount of iron ore resources, including lateritic, iron ores and iron sand in Indonesia were approximately 90 million tons in 2008 (Ishlah, 2008). The new government regulations to ban raw material exports and to domestically process all ores as well as the availability of resources presents an opportunity to explore the technologies that can improve process efficiency and also improve the added-value of iron ore.

The Lampung Center for the Technical Implementation of Mineral Processing (UPT BPML) LIPI, saw this opportunity. Then, (UPT BPML) LIPI built a coal-based, direct reduction technology, namely a Rotary Kiln (SL/RN) to process local iron ore. Local iron ore in South Lampung Regency is primary iron ore in the form of laterite. Primary iron ore contains pure hematite and hematite-containing impurity silica levels ranging from 40-65% Fe total. The total amount of iron ore reserves in South Lampung is about 11 million tons (Kisman, 2005). The abundant iron ore resources in South Lampung have the potential to be processed by using rotary kiln technology.

The Stelco-Lurgi/Republic Steel-National Lead (SL/RN) process was developed in the early 1960s by Outotec in collaboration with three US companies. The basic principle of the SL/RN process was used on numerous rotary kiln plants and still represents the dominant technology for the direct reduction of lump ore or pellets using coal as the reducing agent (www.outotec.com).

In the traditional SL/RN process, iron ore, coal, and dolomite or limestone, as the desulphurize agents, are charged into the rotary kiln and heated up by a countercurrent gas flow and the partial combustion of volatiles and CO released from the charge. The reduction of the iron oxides in the solid state was achieved at temperatures between 920°C and 1100°C. The reduction and coal gasification reactions occurred simultaneously during the charging steps.

The heat was generated by the controlled combustion of surplus CO leaving the charge and volatiles from the coal. The waste gas passed through a waste heat boiler for heat recovery and the generation of electrical power. The Direct-Reduced Iron (DRI) product (also called sponge iron) and the residual non-magnetic fraction are then magnetically separated (www.outotec.com).

The operational reliability of the SL/RN process has been continuously improved since the first industrial application. Numerous technical improvements have contributed to its high level of reliability and flexibility as follows:

1. SL/RN production plants are operating with the widest range of iron bearing materials, such as pellets, lump ore, beach sand and ilmenite.
2. An extensive range of reduction agents can be used, such as sub-bituminous coals and lignite, bituminous coals, and in special cases also anthracite and coke breeze.
3. The emission control meets environmental standards. (www.outotec.com)

In this research, the raw material in the rotary kiln was a mixture of. South Lampung iron ore pellets and coconut shell charcoal in two different sizes. Coconut shell charcoal served as a reducing agent. Indonesia produces 1.1 million tons/year of coconut shells which is one of the potential biomasses to be used as a reducing agent because coconut shell charcoal has a greater percentage of fixed carbon, i.e. 79.60% than sub-bituminous coal Therefore, the hypothesis of this research is coconut shell charcoal can be used to replace natural gas or coal as the reduction agent in ironmaking processes. The reducing agent in the composition of the pellets mixture is aimed to increase the rate of the reduction reaction and reduce the problem of swelling during the reduction process.

2. EXPERIMENTAL SETUP

2.1. Raw Materials

The iron ore sources used in this research were a mixture of South Lampung iron ore pellets, coconut shell charcoal, and bentonite in a respective 77:20:3 weight percentage ratio. First, the iron ore pellets and the coconut shell charcoal were analyzed by using specified ASTM standards. ASTM Vol.03, 05 E2777–69 and ASTM Vol. 03, 05 E508–73 standards were used to determine the chemical composition of iron ores, while ASTM D4422 and D4421 standards were used to consider the proximate analysis of coconut shell charcoal. The results of the chemical composition of South Lampung iron ore can be seen in Table 1. Furthermore, Table 2 shows the proximate analysis of coconut shell charcoal.

Table 1 Chemical composition of Lampung iron ores

Substitute	Fe_2O_3	SiO_2	LOI	CaO	TiO_2	MgO	Al_2O_3
[%]	86.24	7.89	3.55	0.81	0.60	0.50	0.41

Table 2 Proximate analysis of coconut shell charcoal ASTM D4422 and D4421

Subtitute	Fixed C	VM	Ash	H_2O
[%]	79.60	16.40	3.98	5.39

Second, the iron ore pellets were mixed with coconut shell charcoal and a small amount of bentonite as a binder to form iron ore pellets. Steelmaking industries generally use iron ore pellets as a raw material. Figure 1 shows the resulting iron pellets sample, which was further used for this research.

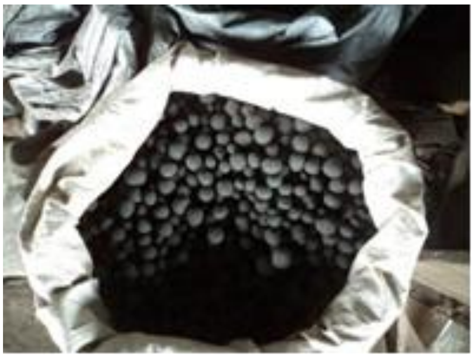


Figure 1 South Lampung iron ore pellets, which are mixed with coconut shell charcoal and bentonite

2.2. Reduction Process and Analysis

After having analyzed the sample, the South Lampung iron ore pellets were inserted into the rotary kiln (pilot scale, Figure 2) to be reduced to become sponge iron at a temperature of 1100°C. The variation in pellet diameter used in this research was (-12+8mm), and (20+12mm), respectively, while the retention of reduction times in the rotary kiln was 1, 2 and 3 hours, respectively. Sponge iron was produced from a rotary kiln and it was cooled in a cooler tube. Finally, it was analyzed by using ASTM Vol.03, 05 E2777-69, and ASTM Vol. 03, 05 E508-73 standards. The sponge iron produced in this research is shown in Figure 3.

(a) Side view

(b) Back view

Figure 2 Rotary Kiln at UPT BPML LIPI Pilot Plant

Figure 3 South Lampung Sponge Iron

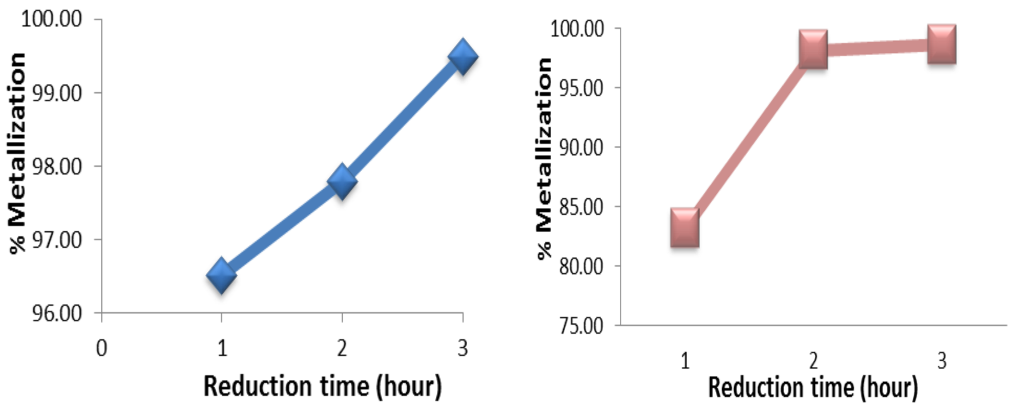
3. RESULTS

3.1. Effect of Time Reduction in a Rotary Kiln based on a Percentage of Metallization of Sponge Iron

Figure 4 shows the effect of reduction time based on a percentage of metallization (removal of oxygen) of sponge iron with variations in the pellet diameter, i.e. Figures 4a and 4b for pellet diameter of (-12+8mm) and (-20+12mm), respectively. Table 3 shows the remaining carbon content in the sponge iron.

Table 3 Carbon content in sponge iron

Reduction Time (hour)	Remain C in sponge iron diameter (-12+8 mm)	Remain Carbon in sponge iron diameter (-20+12 mm)
1	0.03	0.73
2	0.33	0.06
3	0.02	0.03



(a) -12+8mm (b) -20+12 mm

Figure 4 Effect of reduction time on a percentage of metallization of sponge iron

3.2. Effect of Diameter Pellet on a Percentage of Metallization of Sponge Iron

Figure 5 shows the effect of the diameter pellet on a percentage of metallization of sponge iron that was produced during the reduction process. The reduction process was conducted in three retention time variations during the reduction process, i.e. 1, 2, and 3 hours, respectively.

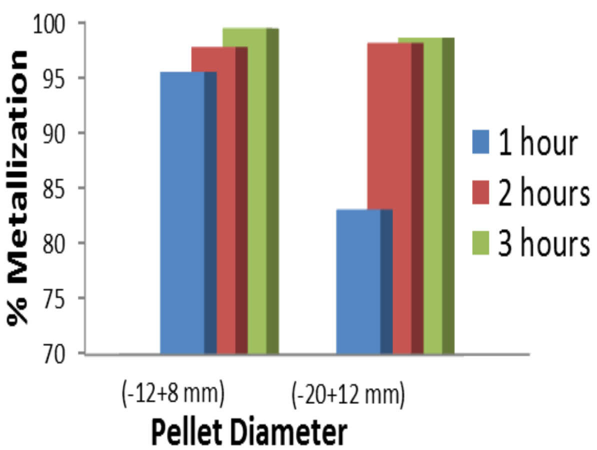


Figure 5 Effect of pellet diameter on a percentage of metallization of sponge iron

4. DISCUSSION

4.1. Effect of Reduction Time in Rotary Kiln on a Percentage of Metallization of Sponge Iron

This research focused on sponge iron made by a direct reduction process using a pilot-scale rotary kiln with a variation in the retention of reduction time and the size of pellet diameter. The aims of this research were mainly focused on the effect of those variables on the percent reduction and the percentage of metallization of iron sponge. The percent reduction of sponge iron is the amount of oxygen production which results from the iron oxide during the reduction process. The percent reduction of sponge iron shows how successful the iron ore reduction process through a direct reduction process was in its implementation.

Besides percent reduction, the quality of sponge iron also can be determined by using the percentage of metallization of sponge iron, i.e. the ratio of Fe metal and the Fe total in sponge iron. The percentage of metallization of sponge iron shows the amount of iron oxide reduced to metallic iron in the direct reduction process. The equation of percent metallization can be seen in Equation 1 (Soesaptri, 2009):

$$\text{Percent of sponge iron metallization } (\%\text{Mtz}) = \frac{\%\text{ Fe Metal}}{\%\text{ Fe Total}} \times 100 \quad (1)$$

As previously mentioned in the discussion of the results, the effect of reduction time on a percentage of metallization of sponge iron with the size of pellet (-12 + 8 mm) and (-20+12 mm), respectively produced from the research can be seen in Figures 4a and 4b. It could be said, that the percent metallization of sponge iron from pellets with a diameter of (-12+8 mm) is about 96.52 %.

In addition, this value has exceeded the ISO standards for the percentage of metallization of sponge iron, which is 85% (SNI 07-0942-1989) in just a one-hour reduction process. It was caused when the pellets were inserted into the rotary kiln at a temperature of 1100^{o}C. At that temperature, the heat transfer from the air through the kiln wall towards the pellets was accelerated, then the reduction reaction of the hematite in the pellets started immediately. This result was supported by the research of Stunley Shuye Sun (Sun, 1997). On the other hand, pellet size of (-20+12mm) needed a reduction time longer than that was needed for the pellet size of (-12+8mm) to produce the percentage of metallization of sponge iron that meets ISO standards, as can be seen in Figure 4b. This was due to the size of iron ore pellets that affected the value of percentage of metallization of sponge iron. The smaller the pellet size, the larger the cross section is, then the diffusion of carbon dioxide gas occurs more easily and automatically increases the percentage of metallization of sponge iron.

Moreover, the heat absorbed by the pellets increases the temperature of the pellets. In the first 5 minutes, the pellet temperature reached 800^{o}C. According to the Glasner-Boudoard diagram, it could be said that if the reduction temperature is about 800^{o}C, the production of CO gas is higher, i.e. by 80% in order to reduce the hematite, Fe_2O_3 to magnetite, Fe_3O_4. Then, CO gas reduces magnetite to wustite, FeO. Finally, wustite reduces to iron metal by CO gas at temperatures ranging from 870-1200^{o}C (Ross, 1980). This showed that in the first 1-hour period, the Fe metal was formed and the percentage of metallization of sponge iron automatically increased. The equations of reduction iron ores by CO gas can be seen as follows in Equations 2, 3, and 4 (Pelton, 2000; Rosenqvist, 1983):

$$3Fe_2O_3 + CO \rightleftharpoons 2Fe_3O_4 + CO_2,\ \Delta G^{o}_{1273} = -211{,}292 \text{ KJ/mol} \quad (2)$$

$$Fe_3O_4 + CO \rightleftharpoons 3Fe_3O + CO_2,\ \Delta G^{o}_{1273} = -35{,}564 \text{ KJ/mol} \tag{3}$$

$$FeO + CO \rightleftharpoons Fe + CO_2,\ \Delta G^{o}_{1273} = -344,\ 917 \text{ KJ/mol} \tag{4}$$

In addition, according to Figure 4 it could be said that percentage of metallization of sponge iron had risen significantly with an increasing time reduction up to 2 hours when it became constant. This showed that all the heat has been absorbed by the pellets in a 1-hour time reduction. Furthermore, the absorbed heat used for making carbon gasification reaction produced 100% of the CO gas. Therefore, the iron ores were reduced quickly. After a 2-hour reduction, the coconut shell charcoal mixed with the iron ore pellets started to run out because the amount of the carbon source in the coconut shell charcoal reacted with the CO_2 to form CO gas during the 2-hour reduction process. This was evidenced by the significant difference in the residual carbon of the sponge iron pellet with a -20+12mm diameter between the 1- and 2-hour reduction periods. The remains of the carbon content in the sponge iron can be seen in Table 3.

4.2. Effect of Pellet Diameter on the Percentage of Metallization of Sponge

The reduction process occurred due to diffusion of CO gas through the pores of the pellets. The pores of the pellets reflect the porosity of the pellet. A high porosity pellet means the pellet has a large amount of pores, and vice versa, i.e. a low porosity pellet has a small amount of pores. If the pellets have a large amount of pores, the diffusion of CO gas into the ore becomes easier and a large amount of iron ore will be reduced into iron metal.

In addition, the reduction of iron ore pellets depends on the size of the iron ore itself. A large diameter of iron ore pellets has a smaller surface area, therefore the diffusion of CO gas will be decreased. The effect of pellet diameter on the percentage of metallization of sponge iron in this research can be seen in Figure 5.

At 1-hour and 3-hour intervals, the pellets with a diameter (-12+8mm) had a percentage of metallization greater than the pellets with a diameter of (-20+12mm). However, at a 2- hour interval during the reduction process, the percentage of metallization of pellets with a diameter of (-20+12mm) were greater than pellets with a diameter of (-12+8mm). This was due to the changing of porosity during the reduction process (Deqing, 2010). In the first 40 minutes, the porosity of the pellets increased significantly, due to the loss of oxygen on the surface of the pellets. The increasing value of the percentage of porosity in the pellets was provided in the space of the pellet diameter of (-20+12mm). Therefore, the contact between CO gas with iron oxide occurred more extensively. This led to an improvement in the amount of iron reduced to metallic iron.

Meanwhile, the differences in initial porosity values for both pellets also affected the maximum swelling of the pellets during the reduction process. Pellets with a small diameter have a high porosity. High porosity means low swelling due to uniform distribution of stresses during the reduction process. A small value of porous pellets with a diameter (-20+12 mm) are able to withstand expansion because each pellet has a narrow cavity, which also resulted in a large swelling and low speed reduction (Taein, 2007).

5. CONCLUSION

Based on this research, it can be concluded that: (1) The percentage of metallization of sponge iron is affected by the diameter of the pellets in which pellets with a (-12+8 mm) diameter produced the maximum percentage of metallization of sponge iron, i.e. 99.50%; (2) For all pellet diameters, the maximum percentage of metallization of sponge iron occurred in the three-hour reduction time interval, such as 99.50% and 98.63% for pellets with a diameter of (-

12+8mm) and (-20+12mm), respectively; (3) The sponge iron produced in this research exceeded the ISO standards for the percetage of metallization of sponge iron, which is 85 % (SNI 07-0942-1989) within just a one-hour reduction process.

6. ACKNOWLEDGEMENT

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