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Effect of flux addition and reductant type in smelting process of Indonesian limonite ore in electric arc furnace

R Andika[1], W Astuti[2], Syafriadi[1], and F Nurjaman[2]

[1]Department of Physics, Lampung University
[2]Research Unit for Mineral Technology, Indonesian Institute of Sciences (LIPI)
Jl. Ir. Sutami Km. 15, Tanjung Bintang, Lampung Selatan, Lampung Province

Email: widi004@lipi.go.id

Abstract. Nickel in the form of nickel laterite ore is one of the most prominent types of mining products in Southeast Sulawesi, Indonesia. One of the utilization of laterite nickel ore is as raw material for ferronickel and NPI production. In this research, the NPI was produced from laterite nickel ore originating from Sulawesi Island in electric arc furnace (EAF). This study aims to determine the effect of the amount of flux (limestone) and type of reducing agent (coal, coke, and shell charcoal) on the content of Ni and Fe in the NPI product and metal yield. Laterite nickel ore used in this study is a type of limonite from Pomaala (Sulawesi Island). The reducing agents used are coke, coal, and coconut shell charcoal. Limestone was used as a flux to adjust the basicity of material during the smelting process of lateritic nickel ore. Reduction and melting process of limonite nickel ore to produce NPI were conducted in small EAF with capacity of 30 kg/heat. The dried pellets are reduced and melted in the EAF with added reducing agents and limestone. Based on the results of the study it can be concluded that the results of the flux addition effect obtained that the optimum Ni and Fe values were obtained on basicity of 1. The results of the reductant type effect showed that coke was the greatest reductant in this study due to the highest Ni content produced when coke used as reductant. In addition, the XRD results showed that the FeNi phase has been formed.

1. Introduction

Indonesia has a nickel resource of around 2,633 million tons with nickel reserves of 577 million tons [1]. Nickel is one of the most prominent types of mining products in Sulawesi Island, Indonesia. In addition, the largest export commodity in Indonesia is nickel with an export value of around 36.57 million dollars or 53.30 percent of the total export value [2].

Nickel is one of the most important metals and has many applications in industry. Many alloy materials are made based on nickel because they have structural strength in the creep, fatigue, and surface stability (corrosion oxidation) process [2]. As much as 62% of nickel is used as raw material for stainless steel production. Around 18% nickel is also used as raw material for the manufacture of alloy steels and non-ferrous alloys [3].

Nickel ores are classified into two namely laterite nickel ores and nickel sulfide ores. Laterite nickel ores are produced from weathering of ultramafic rocks with high iron and magnesium content and are usually found in tropical and sub-tropical regions [4]. Nickel sulfide ores are formed by volcanic or hydrothermal processes and contain lots of copper and cobalt and contain few precious metals such as gold, platinum, palladium, and rhodium [5]. Nickel sulfide ores are found in Russia and Canada [6]. During this time, as much as 60% of the need for nickel is commercially supplied from sulfide ores. In fact, around 70% of the world's nickel reserves are trapped in the form of laterite. Laterite ores have a low nickel content, so they are rarely used as the main source of nickel because special treatment is needed to increase nickel contents in these ores [3]. Basically, laterite rocks can be divided into two layers, namely limonite and saprolite layers. In each layer, there are various types of minerals. The main minerals that compose the limonite layer are goethite [FeO(OH)] and hematite

(Fe_2O_3), whereas the saprolite layer is mainly composed of serpentine minerals (($(Ni,Mg)SiO_3.nH_2O$)] [7].

One of the utilization of laterite nickel ore is as raw material for ferronickel production. Nickel content in ferronickel ranges from 20-40%. Ferronickel is commonly used to make stainless steel. Besides ferronickel, laterite nickel ore is also used in making nickel pig iron (NPI) that usually contains 1.5-25% Ni [8]. Research on the production of NPI from laterite nickel ore has been carried out by several researchers [8] that used coal as reductant. Reductions were carried out at temperatures of 900^{o}C, 950^{o}C, 1000^{o}C, and 1100^{o}C for 1 hour. The results showed that the NPI obtained had Ni content of around 1% and Fe content of around 11%.

In the current study, the NPI was produced from laterite nickel ore originating from Sulawesi Island in electric arc furnace (EAF). This study aims to determine the effect of the amount of flux (limestone) and type of reducing agent (coal, coke, and shell charcoal) on the content of Ni and Fe in the NPI product and metal yield.

2. Materials and method

2.1. Materials

Laterite nickel ore used in this study is a type of limonite from Pomalaa (Sulawesi Island). The reducing agents used are coke, coal, and coconut shell charcoal. The lateritic nickel ores were refined and sieved until the size of <74 μm was obtained. These samples, then, were analyzed using X-Ray Fluorescence (XRF, Epsilon 3XLE, PANalaytical, Netherland) and X-Ray Diffraction (XRD, X'Pert 3 Powder, PANalytical, Netherland). XRF analysis aims to determine the chemical elements contained in the sample and XRD analysis was required to determine the dominant phase in laterite nickel ore. Reductants were analyzed using a proximate method to determine the carbon, ash, and volatile matter content of each reducing agent. The ore and reducing agents were ground to 100% passing screen aperture of 74 μm to increase the contact between them before they were mixed and agglomerated into pellets with diameter 10-20 mm. Limestone was used as a flux to adjust the basicity of material during the smelting process of lateritic nickel ore.

2.2. Experimental method

Reduction and melting process of limonite nickel ore to produce NPI were conducted in small EAF with capacity of 30 kg/heat. The dried pellets are reduced and melted in the EAF with added reducing agents and limestone as flux. The addition of limestone is used to separate metal and smelting slag. The amount of limestone is calculated based on the basicity used. The effect of basicity on the recovery of nickel and iron was conducted by adjusting the composition of limestone in the raw material with the basicity used in this experiment is 0.8; 1; and 1.4 that are equal with the limestone amount of 0, 0.9, and 3.12 kg. The effect of reductant types was also studied by varying of coke, coal, and coconut shell charcoal as reducing agents. The composition of raw material in this smelting process is listed in Table 1. The furnace used is an electric arc furnace (EAF) with proceesing temperature at around 1400^{o}C for 30-40 minutes. The pouring temperature of slag and hot metal was 1246-1578^{o}C. Metal that produced from the experiments were analyzed using optical emission spectrometry (OES, Shimadzu, Japan) to get their chemical composition. Recovery of metal particularly iron (Fe) and nickel (Ni) as well as the yield of metal can be calculated from the iron and nickel content in the NPI produced.

Table 1. The composition of raw material

Trial No.	Laterite ore	Reducing agents	Reductant amount (kg)	Limestone (kg)	Basicity
1	30	Coke	7	-	0.85
2	30	Coke	7	0.90	1

3	30	Coke	7	3.12	1.4
4	30	Coal	7	-	0.85
5	30	Coconut Shell Charcoal	7	-	0.85

3. Result and Discussion

3.1. Characterization of limonite ore and reducing agents

Laterite nickel ore was analyzed using XRF to find out the elements contained therein, particularly the Ni and Fe contents that was shown in Table 2. The results of the analysis showed that laterite nickel ore contained 1.26% Ni which was a type of low-grade laterite nickel [9], while Fe content was 43.1%. Laterite nickel ores are known to have quite high Fe elements. To investigate the dominant phase in laterite nickel ore, analysis using XRD was carried out and the results can be seen in Figure 1. Figure 1 shows that the dominant compounds found in laterite nickel ore are goethite (FeOOH). This is consistent with previous research [10] which shows that mineral phase in laterite nickel ore are dominated by goethite (FeOOH). In addition, there is another main mineral contained, namely quartz (SiO2). Nickel is generally associated with goethite so that the XRD patterns do not show the presence of nickel compounds as an independent compound [11].

Table 2. Chemical composition of limonite nickel ore from Pomalaa (Sulawesi Island, Indonesia)

Elements	Si	Ni	Fe	Mg	Ca	Al
wt. %	3.23	1.26	43.10	6.29	0.34	2.83

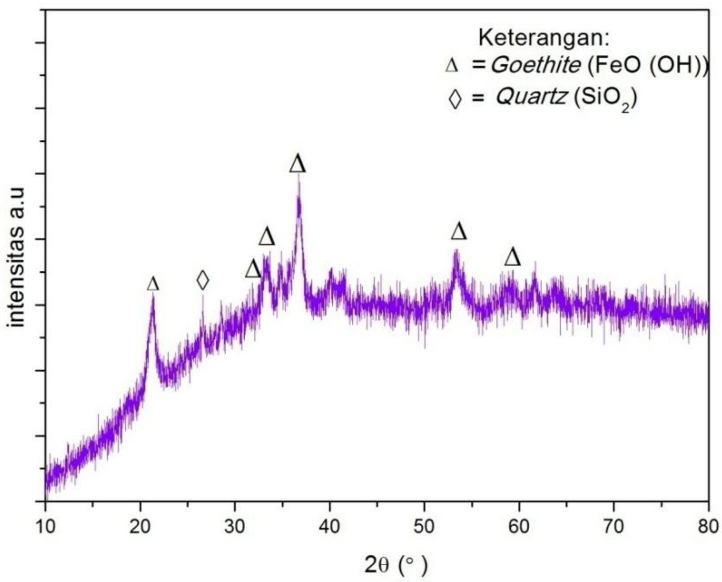

Figure 1. X-ray diffraction pattern of limonite nickel ore from Pomalaa (Sulawesi Island, Indonesia)

The reducing agents used in this study were coke, coal, and coconut-shell charcoal. Each reducing agent has a different carbon content and other compositions. To determine the carbon content of each reducing agent, a proximate analysis was conducted that can be seen in Table 3. It is known that coke has the highest carbon value compared to other types of reducing agents. The coke should be a good reductant because of its high fixed carbon and volatile matter, low ash, and relatively low harmful element (S, P) content. The coke is common reducing agent in the reduction process of laterite ore. However, another important thing is to discuss the feasibility of the other reductant to be used as a substitute in the reduction process.

Table 3. Proximate analysis of reducing agents

Parameter	Moisture (wt. %)	Volatile matter (wt. %)	Ash (wt. %)	Fixed carbon (wt. %)
Coke	0.20	9.65	27.18	62.97
Coal	0.25	48.5	14.00	37.25
Coconut Shell Charcoal	0.15	18.62	26.21	55.02

3.2. The effect of flux additions

The flux is usually used to separate metal and smelting slag. Limestone (CaO) is usually applied as a flux. As for nickel laterite, which consists largely of the magnesium silicate, the influence of CaO on generation of liquid phase was calculated thermodynamically. CaO was added to modify the quaternary basicity-{CaO+MgO)/(Al_2O_3+SiO_2)}. The influence of the basicity on the NPI products was studied in this study using basicities of 0.85, 1, and 1.4.

Basicity value is arranged by adding limestone to the material where the amount of limestone added is calculated according to the Equation 1 and based on the elemental composition of laterite nickel ore.

$$B = \frac{CaO + MgO}{SiO_2 + Al_2O_3} \quad (1)$$

In this experiment, limestone was applied as a flux. The impurities contained in laterite nickel ore will be discharged into the slag. Every experiment in this section used as much as 7 kg of coke as reducing agent. With the basicity of 0.85, no limestone was added where the basicity value was only calculated based on the content of SiO_2, MgO, CaO, and Al_2O_3 in the laterite nickel ore. Whereas on basicity of 1 and 1.4, limestone added was 0.9 and 3.12 kg. The results are shown in Figure 2, in which CaO could definitely enhance the formation of low melting temperature components and generate more liquid phase, particularly before 1300°C. The increase of basicity can significantly reduce the viscosity of molten slag above 1400°C, which is beneficial for the separation between slag and metal.

The weight of the NPI metal produced during the smelting process at each basicity value is 8.6 kg, 6.3 kg and 7.8 kg respectively. On the basicity of 0.85, the highest weight of the metal was obtained. The Ni contents of the NPI products that have been analyzed using OES results were 3.64%, 4%, and 3.86%, respectively for basicity of 0.85, 1, and 1.4. Iron (Fe) is also the main element in the NPI sample. The values of Fe in metals were 82.81%, 89.00%, and 87.3%, respectively for basicity of 0.85, 1, and 1.4. The highest Ni and Fe levels in metal NPI produced were obtained from experiments using basicity 1 or flux addition of 0.9 kg. From the results of this experiment, it was seen that with the flux addition of 0.9 kg, although the weight of the metal obtained was the least, but the NPI metal produced contained the greatest content of Fe and Ni. The relationship between flux addition and recovery values can be seen in Table 4.

Table 4. Effect of flux addition on recovery of Ni and Fe

No	Flux addition (kg)	Metal produced (kg)	Ni in metal (%)	*Recovery of* Ni (%)	*Metal yield* (%)	Fe in raw (%)	Fe in metal (%)	*Recovery* of Fe (%)
1	0	8.6	3.64	82.81	28.66	43.10	82.81	55.06

2	0.9	6.3	4.00	66.66	21.00	43.10	89.00	43.35
3	3.12	7.8	3.86	79.65	26.00	43.10	87.30	52.65

The addition of flux can promote the reduction of nickel and iron and optimize the dynamic condition of laterite nickel ore. The dynamic condition will affect the aggregation and the expansion of nickel and iron as well as the separation of slag and metal. It creates the increase of nickel and iron grades. However, when the flux addition was over than the optimum, there was so much liquid phase generated, resulting in the decrease of the contact area between ore and reductant. Nickel and iron particles will be difficult to aggregate and grow up that lead the entrance of more fine particles into the slag and increase the difficulty of separation. Finally, grades and recoveries of the nickel and iron declined as a result.

3.3. The effect of reducing agent types

Another important thing is to discuss the feasibility of the reductant to be used as a substitute in the reduction roasting process. Experiments to study the effect of reducing agents, namely coke, coal, and coconut shell charcoal are presented in Figure 2. Each smelting process uses each reducing agent with a weight of 7 kg and without flux. Metal NPI produced from the smelting process are 8.6 kg, 8 kg and 9 kg respectively when coke, coal and coconut sheel charcoal was used with the Ni compositions were 3.64%, 3.57%, and 2.37%. The NPI using coconut-shell charcoal as a reductant has the lowest Ni content, while the highest Ni content is obtained in coke reducing agents. Iron contained in the metal were 82.81%, 89.40%, and 89.99%, and with the reductants of coal, coke, and coconut-shell charcoal respectively.

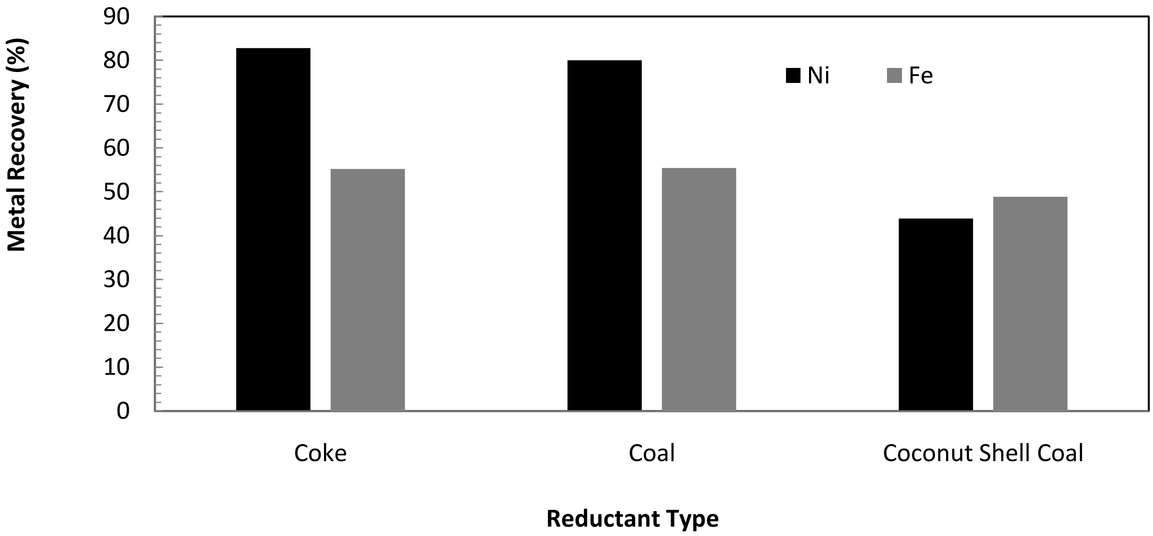

Figure 2. Effect of reductant type on the recovery of Ni and Fe

Figure 2 gives the comparation between using coconut shell charcoal and anthracite coal. Between two reductants, there are no significant differences in terms of magnetite, hematite, olivine, and quartz formation. Therefore, coconut crust charcoal is a potential substance that can be used as a substitute in the process. Mentioning the fact that the final condition of both reductants are mineralogically similat, it can be seen that the effect of the ash content the reductants (see table 3) is insignificant due to the lower concentration of ash in both reductants (see table 2).

From the results, the type of reducing agent used in Ni recovery calculation obtained 82.81%, 75.55% and 56.42% with the highest recovery value in coke. On the contrary, from the calculation of Fe recovery, 55.06%, 55.3%, and 62.63% were obtained with the highest Fe recovery in coconut shell charcoal. The results of the calculation of metal yield were obtained by 28.66%, 26.66%, and 30%, with the lowest metal yield obtained in coal, while the highest metal yield was in charcoal shells.

3.4. Characerization of NPI product

Phase identification from the results of the laterite nickel ore smelting process was carried out using XRD. NPI metal testing using XRD will provide qualitative results for each compound contained in the NPI metal. The test was carried out with a 2θ angle position from 20^o to 80^o and using a wavelength of 1.54056 Å. The test results that can be seen in Figure 3shows that the dominant phase in the metal product of smelting process is Fe and FeNi. It can be concluded that using electric arc furnace, Indonesian nickel laterite ore could be treated by pyrometallurgical method to produce high grade NPI that contained iron and iron nickel.

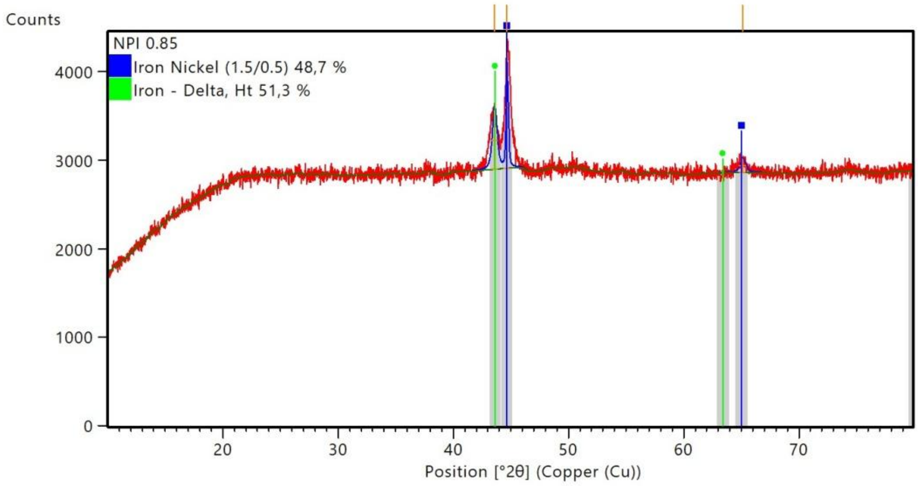

Figure 3. XRD pattern of NPI metal

4. Conclusion

Based on the results of the study it can be concluded that the optimum condition that produced NPI with the greatest Ni (4%) and Fe (89%) was the basicity of 1 and coke as reductant. Using electric arc furnace, Indonesian nickel laterite ore could be treated by pyrometallurgical method to produce high grade NPI that contained iron and iron nickel.

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