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A Comparison Study of Nickel Laterite Reduction using Coal and Coconut Shell Charcoal: A FactSage Simulation

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Abstract. Replacing metallurgical coke with greener material is a long-term challenge in pyrometallurgy. With the fact of biomass abundance in Indonesia, the usage of bio-char as a replacement for coke resulted from coal has been an intensive study. This study compared the performance between anthracite coal and coconut shell charcoal for nickel laterite reduction using the Equilibrium module on FactSage software simulation. The simulation was done by inputting a 4:4:1:20 weight ratio of nickel laterite ore, reductant (coal or coconut shell charcoal), lime and air, respectively. The temperature studied was 1300°C to 1700°C, and the pressure was atmospheric. The result shows that coconut shell charcoal has the potential to substitute anthracite coal as a reductant material in blast furnace processes. Anthracite coal consistently gives better results on the metal phase product with higher liquid metal yield and higher nickel and iron content. However, the coconut shell charcoal posseses the potential as a substitute material with temperatures above 1400°C regarding to the higher nickel concentration compared to anthracite coal. The resulting nickel and iron concentrations can reach above 2% and 96%, respectively, which is the required characteristics of a nickel pig iron product.

Keywords: FactSage; Nickel; Pyrometallurgy

1. Introduction

Nickel could be a transition component that has properties of ferrous and nonferrous metal properties (Kim *et al.*, 2010). Nickel store is affiliated with the press (nickel laterite) or sulfur (nickel sulfide). Almost 58% of nickel request is provided by sulfide metals, in spite of the fact that 78% of nickel stored lies in laterite minerals (Petrus *et al.*, 2019). However,

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as sulphidic ores were continuously exploited in recent years, the sources began to become scarce, and underground mining has been introduced. Consequently, the exploitation cost was rising, especially the labor cost. On the contrary, the mining activity of laterite deposits is considerably shallow (usually less than 50 meters). So, much concern has been concentrated on using low-grade nickel ore (especially those containing <2.0 wt.% nickel), such as laterites (Petrus *et al.*, 2019).

About 12% of the world's nickel laterite resources are stored in Indonesia, mostly in the form of laterites (Rasyid and Petrus, 2016). There are two kinds of laterite, namely limonite and saprolite. Limonite is low-nickel content laterite (around 0.8-1.5% Ni-mass), and saprolite is a rich-nickel content (more than 1.5-3% Ni-mass) (Zhou *et al.*, 2017). Both hydrometallurgical and pyrometallurgical processes can be used to extract nickel from the laterites. However, due to its high nickel content, saprolite ore is better processed by pyrometallurgy (Minister of Energy and Mineral Resources Republic of Indonesia, 2013; Li, Wang, and Wei, 2011). In the pyrometallurgical process there are usually three unit operations, namely roasting, smelting, and converting. The reduction process consumes carbon-based reductants, usually coke, and produces a huge amount of carbon dioxide. This process is highly energy-consuming and not environmentally friendly. Concerning carbon dioxide emission, replacing coke using bio-reductant has been an interesting issue to be studied.

The studies conducted to explore the possibility of using bio-reductants in the process, while paying attention to certain parameters, have been limited. Chen *et al.* (2015) suggested that bio-coal reductants can be used to reduce the major phase in the limonitic laterite ore ($Fe_{1.833}(OH)_{0.5}O_{2.5}$, and Fe_2SiO_4) into a metal phase, such as Fe, $Fe_{0.64}Ni_{0.36}$. Yunus *et al.* (2014) suggested that bio-char derived from empty fruit bunches can enhance the magnetic properties of goethite-rich iron ore via a temperature-dependent sequential reduction process involving hematite (Fe2O3), magnetite (Fe3O4), and wustite (FeO). Wanta *et al.* studied the nickel extraction from nickel laterite ore using citric acid leaching (Wanta *et al.*, 2022), but the big-scale metallurgical process still prefers high-temperature slag treatment (Chen *et al.*, 2019). Petrus *et al.* studied the kinetics and mechanism of saprolitic nickel laterite reduction using palm kernel shell charcoal under 1000°C and shows the promising result of the bio-based charcoal utilization in the process (Petrus *et al.*, 2022).

Following Indonesia's target to export a minimum nickel content of 4.0%, developing a nickel laterite processing plant in the country is necessary. To support the idea, a study confirms that Ni content in Fe-Ni alloy from lateritic sources can reach 4.5% (Citrawati, Dwiwandono, and Firmansyah, 2020). The latest research concerning the phase transformation and kinetics study uses coconut shell and lamtoro charcoal as reductants and proves that biomass-based charcoal could be a good substitute for conventional coal in the roasting process of nickel laterite. Both studies yield magnetite (Fe_3O_4) with identical kinetics parameters to conventional coal, leading to a good step for biomass-based charcoal to substitute conventional coal (Petrus *et al.*, 2017; Putera *et al.*, 2017). It has been widely known that carbothermic reduction of nickel laterite ore for nickel pig iron production is an exceptionally dirty process as it requires a vast energy and emits huge amounts of carbon to the atmosphere. Blast furnace smelting is the oldest method that is later used to process nickel laterite to produce 2-5% Ni. Rotary-kiln electric furnace (RKEF) is a much better process that harnesses the electric current to melt the nickel laterite relatively quicker that harnesses the electric current to melt the nickel laterite in a relatively quick time and produces 20-40% Ni in the final product. The krupp-Renn process is addressed to the low-grade iron ore and able to produce approximately 23% Ni. All current nickel laterite processes, including blast furnace, RKEF, and Krupp-Renn, harness high temperatures. In addition, the blast furnace and Krupp-Renn process use metallurgical coke, anthracite, and limestone (Rao *et al.*, 2013). Therefore, this research studies the potential of an eco-friendly material, charcoal from coconut shells, to substitute the conventional coke commonly used in the mentioned processes.

The current work aims to provide a thermodynamical analysis using FactSage 7.2 of nickel laterite reduction process using conventional coal and coconut shell charcoal. The software offers a benefit to predicting the result of high temperature experiments through complex thermodynamical database calculation (Islam, 2015). Coconut shell charcoal is selected due to its unique characteristics, such as being rich in carbon, environmentally friendly, and cheap (Purnomo *et al.*, 2017). The observed yield of the process will be iron and nickel concentrations. The current study is a continuation of the previous study by Petrus *et al.* (2019), which was limited to 1000°C.

2. Methods

2.1. Raw material characterization

The type of nickel laterite used in this research is limonitic laterite. The reductants are anthracite coal collected from *Lembaga Ilmu Pengetahuan Indonesia (LIPI)* and coconut shell charcoal collected from a local market. The composition of the limonitic laterite, anthracite coal and coconut shell charcoal has been studied by Petrus *et al.*, as shown in Tables 1 to 3.

Table 1 Limonitic laterite composition. Source: (Petrus *et al.*, 2019)

Compound	Percentage (%wt.)
SiO_2	5.2
Al_2O_3	14.96
Fe_2O_3	61.31
TiO_2	0.36
K_2O	0.006
CaO	0.074
MnO	0.13
MgO	0.57
P_2O_3	0.035
NiO	0.72
Cr_2O_3	1.66
Others	14.98

Table 2 Reductant material compositions (Petrus *et al.*, 2019)

Composition %wt_Reductant	Moisture	Volatile Matter	Ash	Fixed Carbon
Anthracite coal	2.3	7.4	2.5	87.9
Coconut shell charcoal	6	13.2	3.7	77.2

Table 3 Ash composition for different reductant types (Petrus *et al.*, 2019)

Ash	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3
Anthracite coal	50.9	34.9	3.5	2.4	1.4	0.2	0.3	0.4
Coconut shell charcoal	6.8	ND	0.7	9.4	10.7	13.4	40.8	14.1

Limonitic laterite is rich in iron (61.31%) and has several valuable metals for alloy making, such as nickel (0.72%) and chromium (1.66%). In high-temperature treatment, silica, alumina, magnesia, and lime are expected to form slag phases. Although coconut shell charcoal's carbon content is not as high as anthracite coal's, it still has a considerably high fixed carbon content (77.2%). Carbon is the essential element in a reductant because it will form carbon monoxide, reducing valuable metals such as iron, nickel, and chromium. To further explain this, Figure 1 illustrates the Ellingham diagram for carbon, iron, nickel, and chromium, which was created using FactSage. As shown in Figure 1, carbon monoxide can reduce nickel oxide and hematite (Fe_2O_3) at around 700°C and 1400°C, respectively. Additionally, chromium can be reduced above 2000°C.

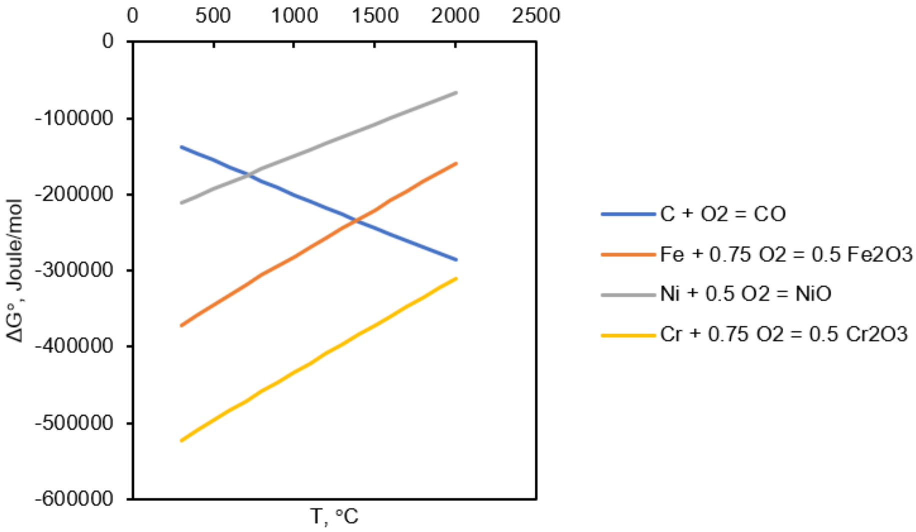

Figure 1 Ellingham diagram of selected elements (C, Fe, Ni, and Cr)

2.2. Process simulation

The process of pig iron making is illustrated in Figure 2. Laterite, reductant, coke, and slag are fed into the blast furnace. Silica and alumina are naturally present in the laterite and must be removed in the slag phase, which commonly uses lime (CaO). This study's selected mass ratio is 4:4:1 for the laterite ore, reductant, and lime. A 1:1 ratio between the ore and coal is assumed to be enough for the reduction process, whereas 4:1 ratio between the coal and lime is the generally accepted practice in the industry (Chakraborty, 2014). In addition, excess hot air (5 times of laterite mass) is fed to the system.

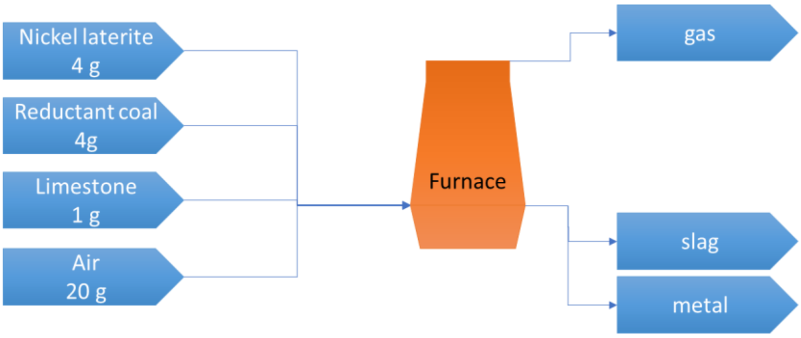

Figure 2 Process illustration

Tables 2 and 3 clearly show that alumina and silica are present in the system. The addition of CaO has the potential to reduce slag's melting point. From the explanation above, the major concentration for the slag will be the $CaO-SiO_2-Al_2O_3$ system, and the slag's composition will be $53\%CaO-14\%SiO_2-33\%Al_2O_3$ (%wt) and $56\%CaO-11\%SiO_2-33\%Al_2O_3$ (% wt) for the system with anthracite coal and coconut shell charcoal, respectively. Before the temperature of the simulation is determined, the minimum melting point of the slag must be predicted first.

Figure 3 shows the phase diagram of $CaO-SiO_2-Al_2O_3$ system at 1500°C and 1 atm, which was done using the 'phase diagram' module in FactSage. As illustrated below, at the mentioned composition, the system is fully in the liquid phase at 1500°C. The phase diagram shows that both slag conditions are partially in the liquidus region, indicating that the slag-metal reaction can startat this temperature.

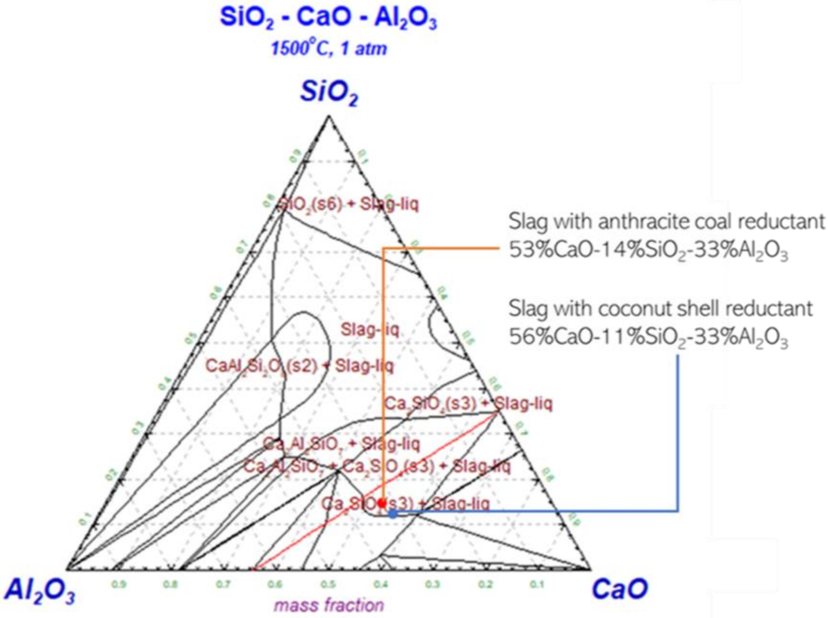

Figure 3 Phase diagram for $CaO-SiO_2-Al_2O_3$ system at 1500°C and 1 atm

2.3. FactSage simulation input

Table 4 Compound input of the simulation process

Compound	Mass, gram	
	Nickel laterite + coal	Nickel laterite + coconut shell charcoal
SiO_2	0.2589	0.2080
Al_2O_3	0.6333	0.5984
Fe_2O_3	2.4524	2.4524
TiO_2	0.0144	0.0144
K_2O	0.0002	0.0606
CaO	1.0030	1.0169
MnO	0.0052	0.0052
MgO	0.0228	0.0386
P_2O_3	0.0014	0.0014
NiO	0.0288	0.0288
Cr_2O_3	0.0664	0.0664
H_2O	0.0920	0.2400
C	3.5160	3.0880
CH_4	0.2960	0.5280
Na_2O	0.0000	0.0198
SO_3	0.0000	0.0209
O_2	4.5400	4.5400
N2	15.5400	15.5400

FactSage is a simulation software introduced in 2001, which is the combination of FACT-Win and ChemSage thermochemical packages that were founded 25 years earlier. In this study, FactSage version 7.2 is used. FactSage consists of information, database and calculation modules most beneficial to chemical and physical metallurgists, chemical engineers, corrosion engineers, etc. FactSage is widely known for its ability to simulate complex chemical equilibria and process simulation, also used in this study (Islam, 2015).

The simulation of chemical equilibria in this study will be done in the 'Equilib' module in FactSage. The module's function is Gibbs energy minimization, where the concentrations of chemical species will be calculated when specified elements or compounds react or partially react in a state of chemical equilibrium (Islam, 2015). The reactants input, following the information from raw material characterization, is presented in Table 4. For simplicity, volatile matter in the reductant is assumed as methane, CH_4, and the moisture is H_2O. The process will be conducted at 1300 to 1700°C and 1 atm. The database selected for this simulation is FactPS for a pure substance, FToxid for oxides, and FTstel for iron-bearing metal.

3. Results and Discussion

As explained, nickel laterite with lime will produce two main products in high-temperature treatment: (1) liquid metal and (2) slag phases. Both phases' mass and composition will vary depending on the applied temperature. In this case, the expected result is a high-yield metal phase and high composition of iron and nickel. Generally, blast furnace processes will produce 2-5% of Ni content in the pig iron, and the nickel recovery ranges from 50 to 90% (Rao *et al.,* 2013). Figure 4 shows the nickel recovery of nickel laterite roasting with the reductant from 1300 to 1700°C. In this case, nickel recovery is the amount of nickel that is still incorporated in the metal phase. For example, at 1500°C, the nickel recovery for a metal-coal sample is 78.53%, meaning that 20.7% of the nickel was removed to the slag phase. As illustrated in the line chart, the nickel recovery of nickel laterite using anthracite coal consistently outperforms the recovery achieved with coconut shell charcoal. However, both reductants can recover more than 78% in most cases. Moreover, the highest nickel recovery is obtained at 1500°C for both reductants.

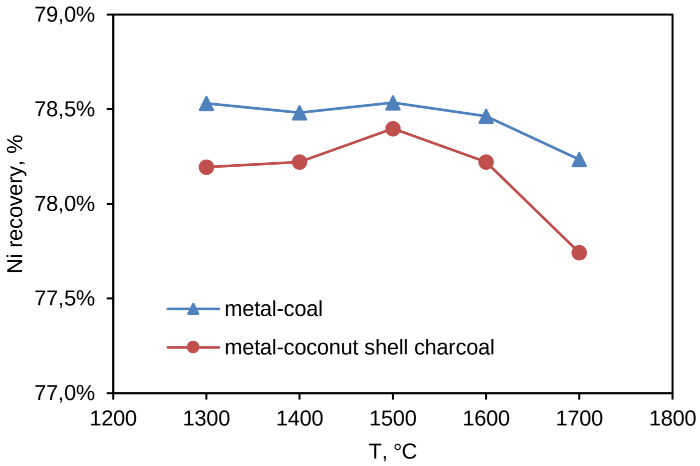

Figure 4 Nickel recovery with a different reductant

The liquid metal produced from the simulation is presented in Figure 5. From initially 4 g of total nickel laterite ore, the lowest metal mass-produced is obtained at the temperature of 1300°C, around 0.2 g. Additionally, both samples with anthracite coal and coconut shell charcoal as the reductants yield their maximum liquid metals at the temperature of 1500°C. However, the laterite that was smelted with the anthracite produced significantly more metal than the laterite that was smelted with the coconut shell charcoal. The metal mass-produced for the system with anthracite coal and coconut shell charcoal are 1.5566 g and 1.0778 g, respectively.

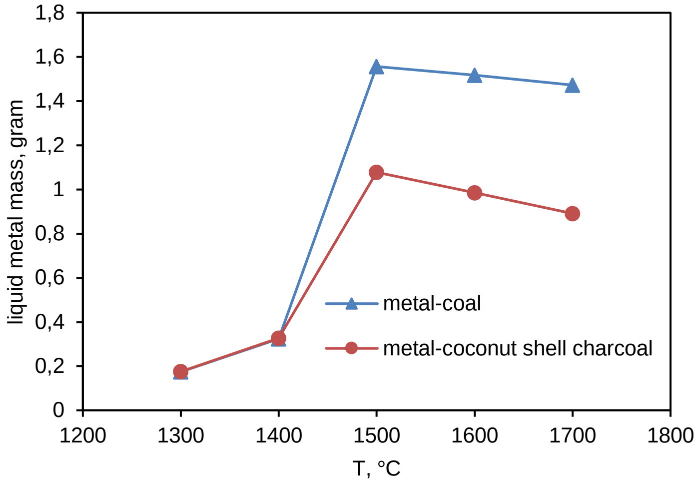

Figure 5 Liquid metal mass-produced with a different reductant

Iron and nickel concentration in the liquid metal phase, using different reductant systems, is presented in Figure 6. As the line chart provides, higher temperatures yield higher iron concentrations and lower nickel concentrations. In this study, the process appears to be favorable at a temperature around 1400°C because the iron concentration can reach above 96% for both reductant systems, and the nickel concentration is still about 4%, which is in the mentioned target range for blast furnace smelting (Rao *et al.*, 2013). In addition, coconut shell charcoal yields satisfactory results as a bio-reductant material in the simulation and may have the potential to substitute conventional coal. While the prices of metallurgical coke and biomass-based charcoal are similar and sometimes overlap, depending on the region (Campos and Assis, 2021), the utilization of coconut shell charcoal can potentially retard extensive mining activity and promote above-ground carbon cycle.

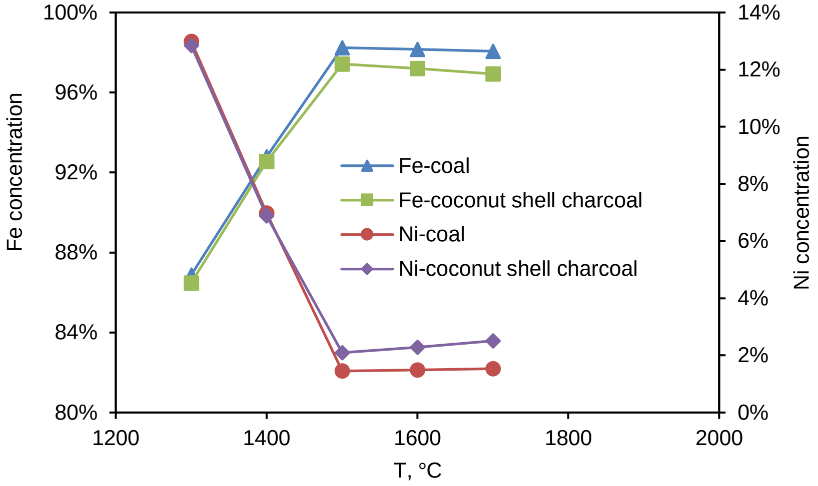

Figure 6 Iron and nickel concentrations in liquid metal with different reductant

4. Conclusions

The simulation of the blast furnace process of nickel laterite using anthracite coal and coconut shell charcoal as reductant, lime, SAAand the air is successfully done with FacSage software version 7.2, using FactPS, FToxid, and FSstel databases. The result of this simulation is often perceived as the ideal condition that cannot be achieved in real experimental work. However, the simulation gives us an insight of the upcoming further research results. In addition, software simulation can also save time, energy, and cost of the real experiment. The simulation shows that anthracite coal consistently gives AA results than coconut shell charcoal in liquid metal mass-produced and iron concentration. However, coconut shell charcoal may have the potential to substitute the conventional coal used in the blast furnace processes because the result is similar to anthracite coal. In addition, it can still yield nickel concentration above 2% and iron concentration above 96% at a temperature higher than 1400°C. Based on this simulation result, further experimental work can be targeted at temperatures above 1400°C.

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