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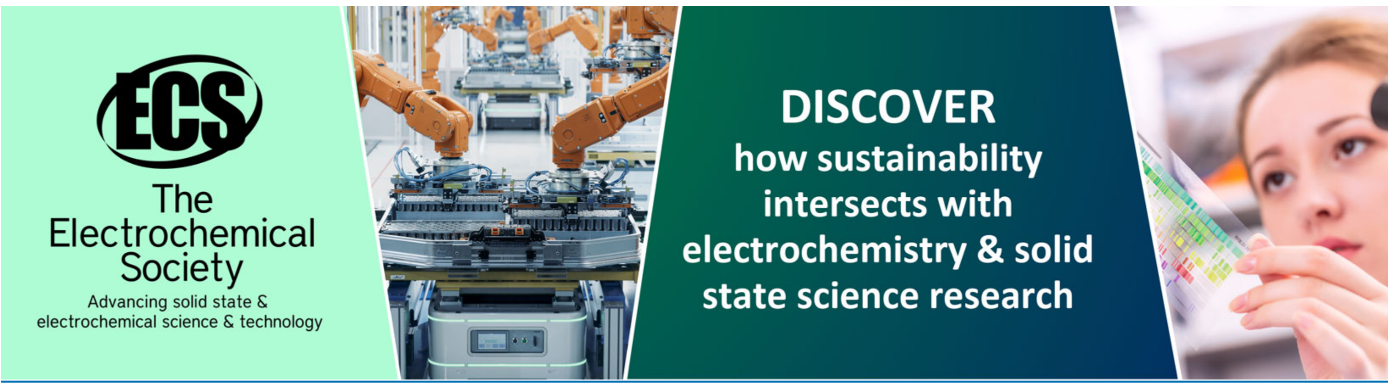

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Effect of pellet size addition on the selective reduction of limonite ore from Southeast Sulawesi

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Abstract. With the increasing demands of nickel in the world and the decreasing of nickel sulfide deposits, the processing of nickel laterite is the new challenge with selective reduction by using coal as reducing agent and Na2SO4 additives. This research aims to determine the preparation parameters of the pelletizing method before the reduction process in order to increase the content of the nickel in the reduction product. The samples were prepared by mixing limonite ore, coal, bentonite, and Na2SO4 additives, then continued by making the pellets with the variability of mass measurements of 5 grams, 10 grams and 15 grams. After the selective reduction, they were characterized by XRD to determine the formed phase, and an AAS test to determine the content after selective reduction. The test result of XRD showed the forming of magnetite, which were found in the pellets with mass size 5 grams. After all, fayalite phase was found in each size of pellet samples. The largest iron and nickel content was shown in the 5 gram pellets, 25% and 0.85%, respectively

1. Introduction

The processing of nickel laterite ore is about 40% of world total nickel production, therefore it will contribute in the scarcity of nickel sulfide in the future if it is continuously mined and exploited [1,2]. With the increasing demands of nickel in the world and the decreasing of nickel sulfide deposits, the alternative is the exploitation of nickel oxide with various methods and innovation so that the needs and demand for nickel can be maintained. One of the methods to increase the content of nickel laterite is the selective reduction method, which is the process of ore reduction with carbon reductant and addition of the additives which can increase mineral selectivity to be obtained at the reduction stage. One element added as an additive is sulfur because sulfur from some studies is considered capable of increasing the effectiveness of reduction and increasing the acquisition of nickel with other metals [3,4]. In laterite ore, the presence of sulfur accelerates the formation of FeS phase and makes ferronickel phase becomes larger so the value can be increased, as well as suppressing the growth rate of fayalite and forsterite in nickel laterite reduction process [5]. From various studies that have been done, the variables that are often used are the time of reduction, temperature, and concentration of the Na_2SO_4. In the preparation process of specimens conducted such as pellet sample preparation. The purpose of the use of pellets is that the interaction between the laterite nickel ore with the coal reducing agent and the additive progresses better so that the reduction process of nickel oxide and the reaction of iron sulfide formation takes place more effectively, thus the selective nickel reduction process laterit can be better [6]. Further research has not been done and that may affect the course of the reduction process on laterite nickel. This research aims to find the effect of pellet's variable with its physical characteristics possessed against the value of acquisition by the selective reduction method.

2. Materials and method

2.1. Materials

The material used is limonite nickel from South Sulawesi, Indonesia. initial sample of limonite used in this research was initially characterized with the test of XRD, XRF, and AAS. According to Zevgolis et al. [7] there are three types of nickel laterite deposits that can economically be processed and utilized, i.e. limonite, intermediate and saprolite (Garnierite). The three types of nickel mineral deposits have different mineral constituent phases. These three types of nickel mineral deposit have a different phase mineral constituent, as in the type of saprolitic deposit containing Garnieritic $(Ni, Mg)_6Si_4O_{10}(OH)_8$ with high magnesia content but low iron content. Limonite deposits contain iron oxide as a major constituent of such mineral bonds, such elements as goethite (FeO(OH), hematite (Fe_2O_3) or magnetite (Fe_3O_4).

In the figure 1 below, showing the available phases of the laterite nickel XRD data of the sample before the selective reduction process was carried out containing goethite, quartz (SiO_2), kaolinite ($Al_2Si_2O_5(OH)_4$), and magnetite, so it can be said that the mineral used in this research is limonite.

In table 1 the following is the result of the initial XRF test of limonite. Beside the limonite, coal as a reductor was also tested with proximate and the ultimate test, the proximate and ultimate analysis aims to determine the physical parameters and chemical content in the coal [8]. The test results can be seen in table 2.

Table 1. XRF analysis of iron and nickel oxide content of limonite ore.

Oxide	Content (%)
Fe_2O_3	75.3
NiO	1.69

Table 2. Proximate and Ultimate Analysis of Coal.

Parameter	Content (% adb)
Proximate Analysis	
Moisture in air dried sample	7.26
Ash	15.87
Volatile Matter	16.44
Fixed Carbon	60.43
Ultimate Analysis	
Carbon	64.02
Hydrogen	3.11
Nitrogen	0.60
Oxygen	16.32

2.2. Experiment
The research method is done through several stages, starting with the initial characterization of limonite and coal such as XRD test, XRF test, and AAS test. Furthermore, the preparation of lateritic nickel ore, Na_2SO_4, and coal sample will be done. The ratio of the pellet mixture between limonite and coal is 100:6, then mixing with Na_2SO_4 additive is about 10% of the total mixing weight between the limonite and coal which will then be processed into pellet form manually with the addition of water gradually. To increase the binding power between the pellet components, 3% of bentonite was added from the total mixture of three previous materials. The pellets have been varied by 5 grams, 10 grams, and 15 grams in size. After were dried for 2 hours in the oven at a temperature of 100° C, they were proceeded with the reduction process at a temperature of 800°C for 1 hour in the furnace with a gradual rise in temperature. The first stage was at 480 ° C at a rate of 10°C per minute, after reaching the temperature and held for 10 minutes, then the furnace temperature will rise to the target temperature which was then held for 1 hour. After the reduction process is completed, then the cooling process was done in the furnace. The reduction pellet is weighed after the reduction process, then crushed to prepare for the AAS characterization, XRD.

3. Result and discussion
In Figure 1 below explains the XRD graph of pellets before reduction to pellet after reduction with various mass sizes. The graph before the selective reduction process was carried out containing goethite, quartz (SiO_2), kaolinite ($Al_2Si_2O_5(OH)_4$), and magnetite, so it can be said that the mineral used in this research is limonite. After the selective reduction process in the temperature of 800 °C for 1 hour, a phase change in each pellet causes a distinct reducibility of each pellet by various test weight sizes. In pellets weighing 5 grams, the phases formed are fayalite ($Fe_2Si_2O_4$), magnetite, hematite and quartz. In a selective reduction of 5 grams of pellets, the goethite and kaolinite phases are dehydroxylated. Meanwhile, the quartz and magnetite phases do not appear to be noticeable changes in the XRD pattern. When goethite decomposition was occurred, nickel which is in the different solid solution with goethite structure [(FeNi)O.OH-n.H_2O] will be released from its bonds, and become reduced. This mechanism is the reinforced by the results of AAS analysis in table 3, wherein the pellet reduction of 5gr higher amount of Fe and Ni than other pellets size that has been detected. This further strengthens the notion that in 5g pellet size there are more goethite phase changes being hematite at the time of the reduction process. An increase in Ni content of 5 gram reduction results indicates that in the pellet condition there is release of the Nickel bond from oxide to metallic Nickel. There was a decrease content of Fe for 5 grams compared to iron contents in the initial sample, it is because it has not completely decomposed into Fe, it is a desirable condition because it can increase the NI selectivity [9]. This result is similar to the research done by Johny et al [10] where the hematite phase is transformed into a magnetite phase in saprolite reduction using coal as a reducing agent.

Dehydroxilation phase of kaolinite from hematite occurred in the temperature of 700 °C so that it formed the phase of meta kaolinite, however this form of phase is not stable so it could not be seen in the peak XRD of 5 grams pellets reduction results.

At the reduced temperature, the fayalite phase formation occurred, whose intensity is not very significant in the 5 gram pellets. That phase should be avoided as it covers the iron oxide grains. In the presence of a Fayalite phase on a 5-grams pellet it could kinetically inhibit the pellet reduction process. Fayalite phase formation occurred because of several minerals belonging to the serpentine minerals species such as chrysolite, lizardite, and antigorite.

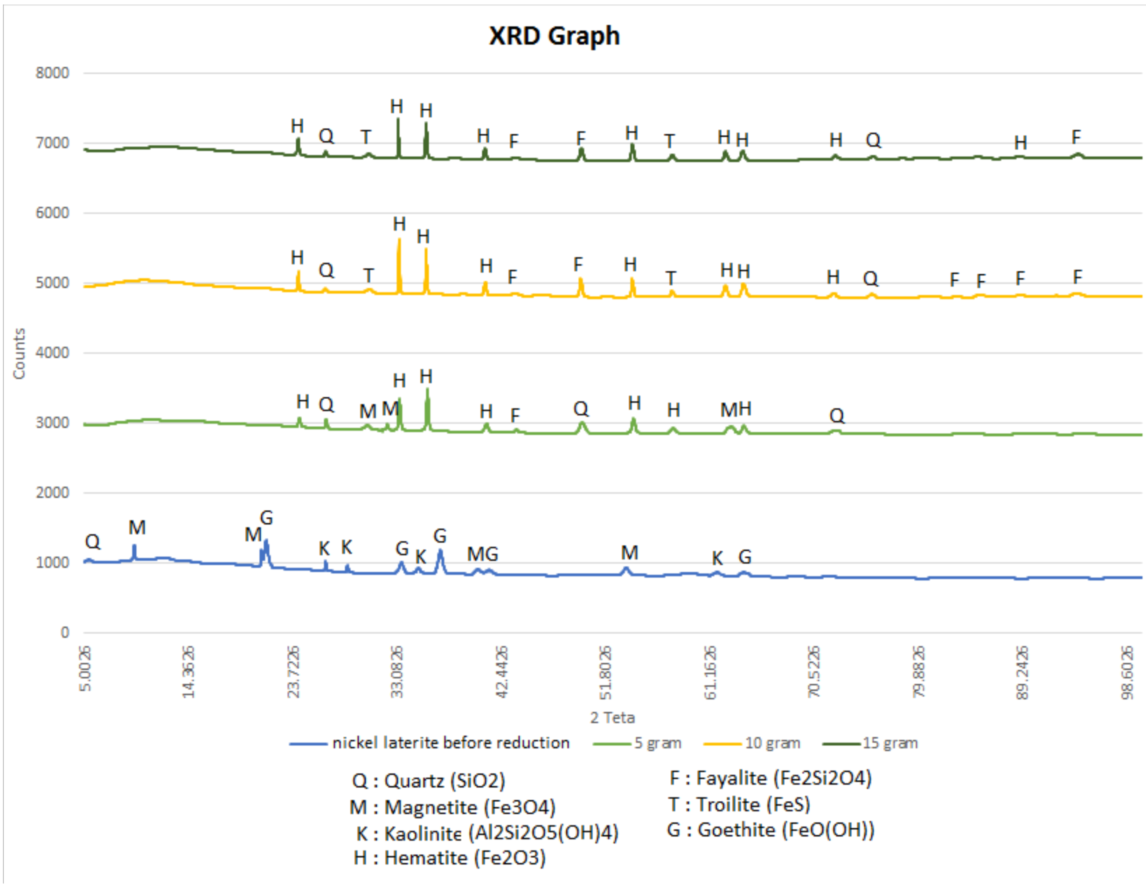

Figure 1. XRD Pattern of initial limonite and limonite after selective reduction with pellet size variations

At a temperature of 700 - 800°C the mineral will decompose and crystallize into olivine, fayalite and enstatite. The phase formation occurs when the reduction rate runs longer in the furnace, although in this study the reduction process has been added with the Na_2SO_4 additive with a 10% additiion of the fayalite phase remaining, the conclusion could mean that the amount of the content has not been able to break the silica bond with the oxide at the pellet and the difference of heat transfer due to the difference of temperature gradient present in each pellet tested.

At the same time the quartz phase is still available in the pellets with 5 grams weight, because decomposition of the quartz phase occured in the temperature of 1000°C. However, in XRD graph of 5 grams pellets occured the growth of the quartz phase, it is possible because the dehydroxylation of serpentine which produce fayalite and quartz. The forming process of that phase will increase along with the increase of temperature, which used to reduce nickel laterite.

In the 10 grams and 15 gram pellets there was a phase transformation into fayalite, hematite and quartz. The formation of the fayalite and hematite phases in both pellets indicated a longer reduction rate than the 5 grams pellet due to the magnitude of the temperature gradient to convert the phase into a metallic phase. In the phase transformation of both pellets, troilite phase was occurred [2]. The Troilite phase acted as an activating agent to accelerate the formation of phase shake and increase the rate of particle aggregation. The existence of such phases will facilitate the separation magnetically. Based on the

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table below, indicates a decrease in the iron content of each pellet along with the increase in the size of the pellet mass, but at the nickel content, there is an increase in the content of pellet's weighing 5 grams, and decreasing content in pellet's size 10 grams and 15 grams.

Table 3. Comparison of nickel and iron content measurement of limonite between before and after reduction by Atomic Absorption Spectroscopy.

Element	Before reduction	After reduction			Unit
		Pellet size 5 gr	Pellet size 10 gr	Pellet size 15 gr	
Fe	41.46	30.57	16.67	7.61	% w/w
Ni	1.16	1.18	0.82	0.56	% w/w

4. Conclusion
Based on the research which has been done to the limonite with various variations of pellets mass with 10 wt% Na_2SO_4 as additives and coal reductor in the temperature of 800 °C, it could be concluded that,
1. Pellets with the weight of 5 grams have Magnetite phase more than other size pellets.
2. There was a decrease content of Fe for 5 grams compared to iron contents in the initial sample it is because it has not completely decomposed into Fe.
3. Fayalite phase in the Pellets with the weight of 5 grams formed not too significantly, it was proven in the intensity of peaks in the graphic of XRD results.
4. Pellets with the weight of 5 grams have gained the higher content than other size pellets, even though the reduction degree of each pellets with various sizes was lower than the reduction degree of initial limonite.

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