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STRUCTURAL CHANGES OF POMALAA LATERITIC ORE DUE TO COAL-BASED MAGNETIZING ROASTING

PRAMUSANTO[1], NURYADI SALEH[1] and SYONI SUPRIYANTO[2]
[1]R & D Centre for Mineral and Coal Technology
Jalan Jenderal Sudirman 623 Bandung 40211,
Ph. 022 6030483, fax. 022 6003373
e-mail: pramusanto@tekmira.esdm.go.id; nuryadi@tekmira.esdm.go.id
[2]Metallurgy Department, Bandung Institute of Technology, Indonesia
Jalan Ganesha 10 Bandung 40132,
Ph. (022) 250 0935, fax. (022) 250 0935

ABSTRACT

Overburden of Indonesia's laterite ore at Pomalaa is considered as an iron cap. It performs low iron grade (41.88%) and high silica and aluminum oxide contents (18.47% and 9.46%, respectively). Around 54.74% of size distribution belong to -325 mesh fraction. Limonite iron mineral dominates in the ore in the range of 80-90% with water content of about 40%. Proven deposits of laterite iron ore are about 222 million tons. As a significant resources iron ore to be used as raw material for iron and steel industries, the iron content must be upgraded to meet the requirement of iron making industry.

Magnetizing roasting technique can be conducted to change the paramagnetic iron mineral (such as hematite, goethite, limonite or siderite) into magnetite one that has high magnetic intensity. Therefore, the changed iron mineral can be concentrated using low-intensity, magnetic separator. Coal, mixed in ore composite may also enhance the development of coal-based magnetizing roasting processes in order to reach the desired temperature. Recently, reduced iron products from many different processes have been used as the main feed mixed with steel scrap. On the other hand, iron ore resources is getting dominated by low grade lateritic iron ore with specific content of water crystal. The abundant deposits of low grade lateritic iron ore and low rank coal in Indonesia can be used as suitable resources for raw materials in the iron and steel-making industry.

Iron structural changes during magnetizing roasting process using coal as reductant agent was observed. The result showed that the non-magnetic limonite ore has been changed in to metallic iron and the iron recovery in the magnetic product depended on the coal ratio in the pellet composite. The magnetic product can be used for the development of lateritic iron ore as one of the alternatives to metallized iron feed for iron making industry.

Keyword: reduction, iron ores, limonitic ores, magnetizing roasting

INTRODUCTION

Indonesia has abundant lateritic minerals that potentially can be used as raw material for iron-making industry; however the effective utilization of such ninerals has not yet been advancing. Lateritic iron mineral, commonly containing of 30-40% Fe with metal impurities of Ni, Co and Cr, is a typical raw lateritic iron ore originally from Pomalaa, Southeast of Sulawesi. Its size is less than 74 meshes, retaining the Fe_{total} content of 43.84%, SiO_2 of 10.92%, and Al_2O_3 of 6.43%. For those specific reasons, a suitable technology for processing this mineral has begun to be studied.

It is reported by Meyer (1980) that even at a very early stage, attempts were made to beneficiate hematite-bearing ore by converting the hematite to magnetite. The process included reduction and separation process magnetite from its gangue in

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magnetic separators. Shaft furnaces, rotary kilns and fluid-bed roasters can be used for these purposes. Some previous authors (Stephenson and Larson, 1980; Stephenson, 1983; Gupta and Misra, 2001; Purwanto and Pramusanto, 2009) suggested that the use of metallized iron compound for the iron and steel making can be conducted by introducing composite materials. The purpose was to minimize the use of coke which is getting rare and expensive.

Accelerating of reduction rate and decreasing reaction temperature in iron making process are other key issues to save CO_2 emission and the energy consumption (Shimizu et al., 2007). The reduction process was found topochemically under condition of temperature and reduction agent (Winnyekyj and Ruskin, 1988; Shimizu et al., 2007, Supriyanto et al., 2009). The availability of coal with ease of supply may provide the development of coal-based direct reduction processes. Reduced iron pellets from many different processes have been used as a main feed mixed with steel scrap. Feedstock of iron ore, on the other hand, is getting dominated by low grade lateritic iron ore with specific content of water crystal as commonly found in limonitic iron ores.

Reduction had been carried out using laterite iron pellet in this project. Coal powder was mixed as a reducing agent. Experiments were initiated with coal burning to study the profiles of reducing gas.

EXPERIMENTAL

Materials

The sample used in this experiment was limonitic-type nickel laterite ore that was obtained from the overburden of Pomalaa mine, PT Aneka Tambang. Size distribution and compositions of each fraction of the sample are presented in Table 1. Average iron content (% Fe_{tot}) was calculated from the weight and iron content of each fraction (between +60 to -325 mesh). The highest Fe content (more than 40% wt) can be found in -325 mesh fraction. Mineralogical constituent of the sample is presented in Table 2. The sample was dominated by limonitic-type iron ore, a typical iron ore with high content of crystalline water. In the same time, pulverized coal as reductant in powder (-65 mesh) had also been prepared. A high volatile sub bituminous coal, consisted of 19.1% moisture content, 2.16% of ash, 40.65% of volatile matter, 38.09% of fixed carbon and 5,745 kcal/kg of caloric value, was used.

Procedure

Direct reduction of iron lateritic pellet, controled by varied temperature and air flow rates, was carried out using a rotary kiln. The mixture of pellets and fine coal were fed inside the kiln and reducing atmosphere can be reached with the various of CO/CO_2 ratios.

The ore was dried and mixed with 2% limestone to make a pellet. The average pellet diameter was 12-mm. Feed for rotary kiln reduction was prepared by re-mixing the pellets with coal fines. Before reaching the designed temperature as well as during cooling off condition to room temperature, the kiln was maintained under inert condition by controlling nitrogen flowrate at 1 l/min. The nitrogen flowrate was switched to the airflowrate of 1, 3, and 5 l/min when the reduction temperatures reached between 1,073º to 1,273° K. The heating time was one to two hours. During the process, CO and CO_2 gases evolved from heated feed, were measured and the ratio of CO/CO_2 was calculated. Figure 1 shows the kiln that was used in this experiment. A high CO/CO_2 gas ratio (>80%) was examined, indicated that reducing atmosphere in

Table 1. Chemical analyses of the laterite sample

Size, mesh	Wt, %	Fe_t	FeO	Fe_2O_3	TiO_2	SiO_2	Al_2O_3	CaO	MgO	Cr	Co	Ni	LOI
+60	13.69	37.3	0.00	53.3	0.00	26.7	7.88	0.037	0.80	1.55	0.069	0.53	8.53
-60+100	12.01	39.7	0.00	56.8	0.14	21.4	9.21	0.100	0.74	1.72	0.043	0.47	8.76
-100+140	7.14	39.1	0.64	55.2	1.00	21.0	9.68	0.065	0.74	1.64	0.037	0.45	8.59
-140+200	5.52	38.2	0.00	54.6	0.00	23.3	9.85	0.054	0.80	1.97	0.035	0.44	8.42
-200+325	6.90	39.0	0.73	54.9	0.00	23.3	9.81	0.094	0.81	1.84	0.030	0.42	8.48
-325	54.74	44.6	0.00	63.9	0.59	14.4	9.80	0.130	0.42	0.89	0.025	0.52	9.32
Total avg.	**100**	**41.88**	**0.096**	**59.84**	**0.45**	**18.47**	**9.461**	**0.102**	**0.581**	**1.26**	**0.035**	**0.49**	**8.99**

Table 2. Mineralogical analyses of the laterite sample

Minerals	Fraction, mesh					
	+60	-60+100	-100+140	-140+200	-200+325	-325
Magnetite	-	-	2.25	2.59	1.67	1.04
Hematite	1.00	0.63	-	-	-	-
Limonite	90.44	96.23	91.57	90.84	82.57	83.37
Mag-Hem.	1.00	0.96	0.74	-	1.74	0.59
Chromite	-	-	0.65	0.99	2.28	1.55
Gangue minerals	6.75	1.98	4.39	4.70	10.19	9.23

Figure 1. Laboratory scale of rotary kiln

the kiln chamber was sufficient to reduce oxygen in pellet. The reduced pellet laterites were then analyzed for their iron metallization and the percentage of the conversion.

RESULTS AND DISCUSSION

Reducing Atmosphere Kiln

Flow-rates of air during reduction reaction in rotary kiln may control contact retention between fresh reducing gas and pellet surface. Figure 2 shows the gas ratio of CO/CO_2 profile for individual coal burning at temperature of 1,273º K. At the beginning up to 20 minutes, the increase of gas ratio depends on the air flow rates. The gas ratio is relatively constant after equilibrium reached. Increasing the air flow rate (Q) from 1 to 5 l/minute will consequently change the kiln atmosphere to more oxidation state. These data were used to fix the experimental condition applied at 3 l/min air flow . Preliminary tests conducted at lower temperature i.e. 1,073° and 1,173º K the gas ratio was insufficient for reduction process.

Iron Recovery in Magnetic Product

The relation among the Fe content, Fe recovery and coal ratio between coal and iron ore is shown in Figure 3. As the coal composition increases from 25% to 30%, the iron content in magnetic concentrate increases from 46.3% to 55.9% and slightly decreases to 53.6% in coal composition of 40%. Using these figures, the percentage of recovery iron in each stage of coal ratio can be calculated by dividing each iron content with the Fe content that is 41.88% Fe namely the Fe content that is available in the original ore. The recovery in ferromagnetic concentrate product shows that 55.88% Fe are obtained for 25% of coal and increase up to 79.56% for 30% coal, and 81.99% for 40% coal. The iron recovery of 90% is obtained by reduction treatment and assaying about 55% Fe from 45% Fe in the original ore as previously reported by Purwanto and Pramusanto (2009). These lower recovery results can be explained due to the lower iron content and higher gangue minerals in the ores sample.

Structural Changes of Pellet

Structural analysis of iron pellet before and after reduction process was observed by optical microscope through thin sections analyses. Sequences of photomicrographs in Figure 4 show area of metallization and some unreduced remains. The unreduced part is mostly covered by metallic iron, which confirms the reaction follows unreacted core model. Some pores as void area had also been observed as a dark color that indicates a space left by oxygen during the process. Volume change was expectedly occurred during reduction.

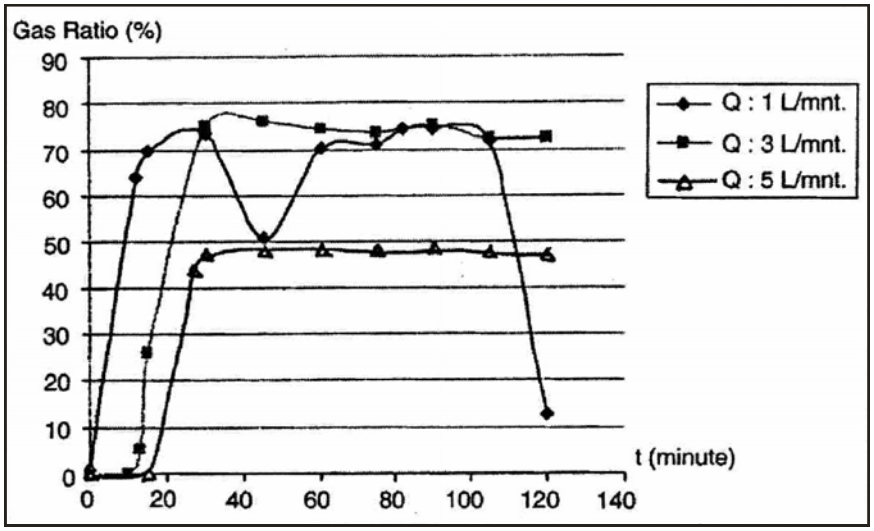

Figure 2. CO/CO$_2$ gas ratio profile during coal burning at 1,273° K

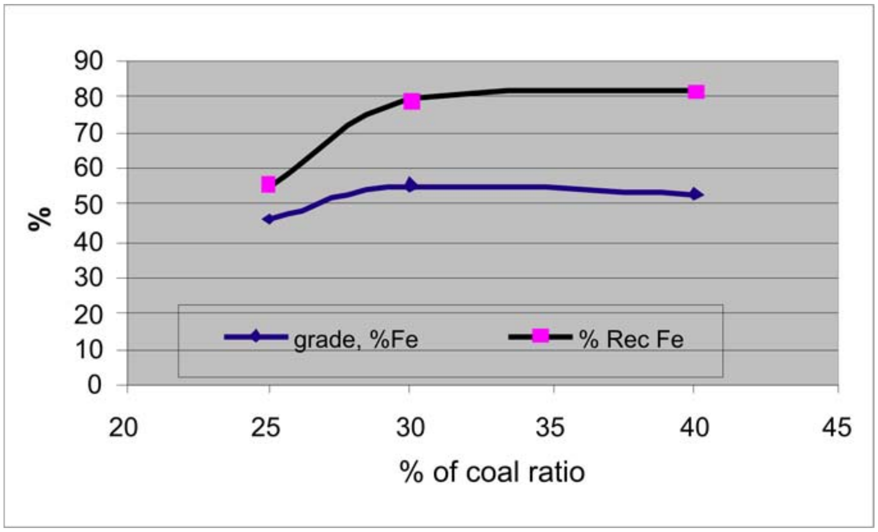

Figure 3. Effect of coal on magnetizing roast performance

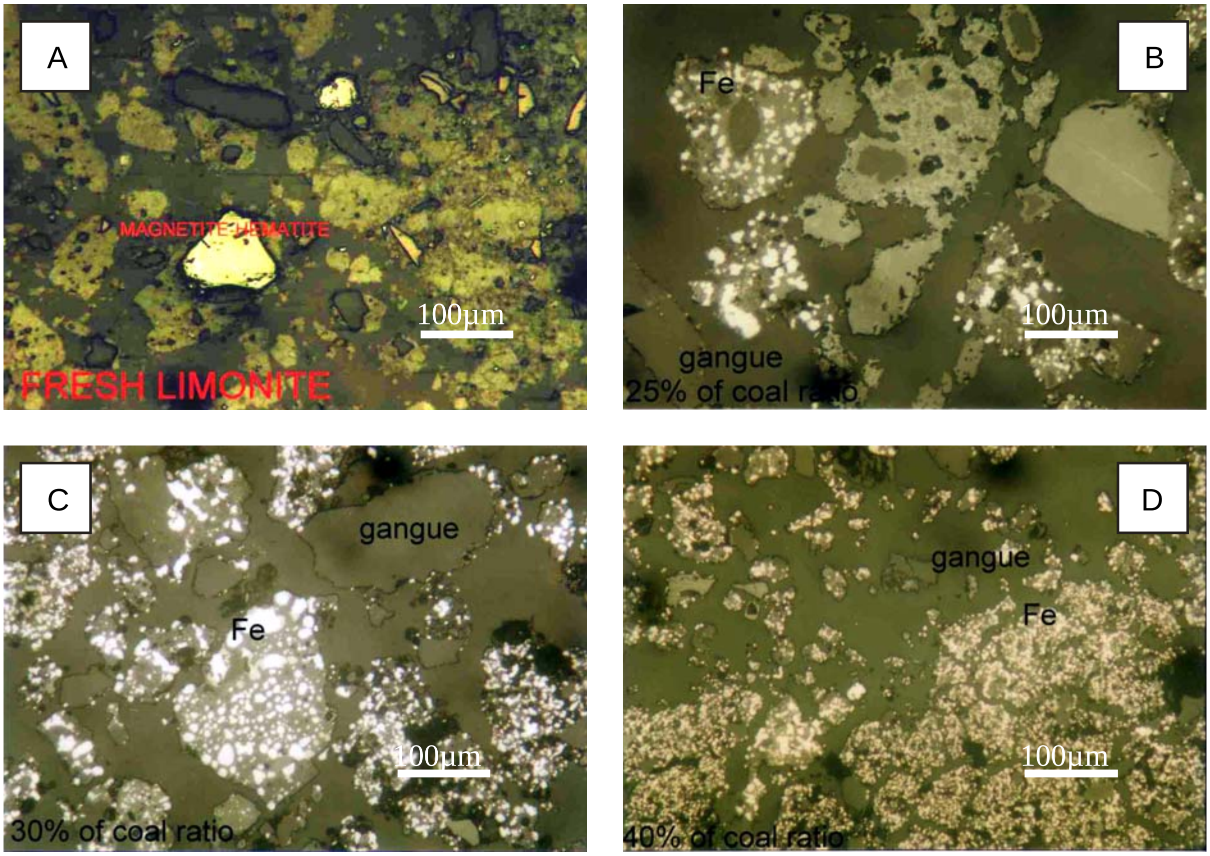

Figure 4. Photomicrograph of (A) fresh laterite mineral and the changes to magnetizing product after reduction for 90 min at 1,273° K for (B) 25%, (C) 30%, (D) 40% of coal ratio

Figure 4 A shows fresh limonite before reduction. Limonite ore appears in earthy color and gangue minerals, such as silica or aluminum oxide appear in dark gray color. Minor mineral like hematite (white) and magnetite appears in brighter color. After coal reduction, limonite changes to metallic iron as shown in Figure 4 B, C, and D. The metallic iron phase increase along with coal addition up to 30% of the coal ratio (Figure 4 B and 4 C). Using 40% of coal ratio, the rich phase of metallic iron decreases (Figure 4 D) and it grows intensively on the surface of iron ore as a thin layer. Limonitic phase alters to metallic temperature phase because temperature reduction (about 1,273° K as the actual temperature) will be higher than that of the setting one due to the excessive coal ratio that is being applied. It results in wustite over reduction to be iron metal and prevents further reduction as well. The metallic iron growth can determine the kinetic of reduction at high temperature reduction. Evidence of magnetic product during low temperature reduction such as magnetite and wustite are expectedly appeared in the reduced product.

Kinetics of Pellet Reduction

The reduction kinetic (Sohn and Wadsworth, 1979) at 1,273º K was determined by reducing the time for 45, 90 and 135 minutes, then the metallization results were calculated using equation of reduction rate of iron pellet with respect to topochemical and the three elementary processes of iron reduction. The rate equation was proposed as eq.1) (Supriyanto et al., 2009):

$$t\text{-}t_f = A\times A_1\{3F^2-2F^3+\frac{A_2}{A_1}F\} \quad \text{........................ (eq.1)}$$

Figure 5 was made by plotting the F function, $\{3F^2-2F^3+\frac{A_2}{A_1}F\}$ versus time reaction from the experimental data reported elsewhere. The F function depends on three conditions of $\frac{A_2}{A_1}$, namely $\frac{A_2}{A_1}$ >1; =1 ; H" 0. The optimum condition was reached at condition of H" 0, at the best fit value of R^2 = 0.6624 compare to another condition. So the coefficient of gas diffusion was 1.6209 $mm^2.minute^{-1}$.

where:

$A = \rho_0\, \tau_0/(Cb - C^*)$; $A_1 = \rho_0/(6De)$;
$A_2 = K/Kc(1+K)$; $t^* = t - t_f$

Based on the equation, reduction was controlled by CO/CO_2 diffusion through a solid product layer. At 1,273° K; metallization increases from 50.67% for 45 minutes reaction and exceeds to 52.61% for 135 minutes but it does not significantly increase for further reaction time because of the resistance to reducing gas diffusion. This results also explained the growth of metallic iron as shown in Figure 4 as previously reported (Supriyanto et al., 2009).

List of symbols:

$t - t_f$ = reduction time in iron pellet rate of reduction equation (min)
t_i = reduction time (min)
t^* = $t - t_f$ (min)
ρ_0 = specific density of pellet ($g.mm^{-3}$)
τ_0 = radius of iron oxide before reduction (mm)
K = equilibrium reduction constant of reaction ‹FeO› + (CO) 1 ⇔ ‹Fe› + (CO_2)
F = fraction of conversion $[1-(1-f)^{S!}]$
f = metallization degree
De = coefficient effective diffusivity of CO/CO_2 gas in pellet ($mm^2.minute^{-1}$)
Cb = concentration of gas reductor at bulk layer ($mol.mm^{-3}$)
C^* = concentration of gas redactor at surface of iron oxide, Fe_2O_3 ($mol.mm^{-3}$)

CONCLUSIONS

A very low magnetic limonite mineral in laterite iron ore can be changed to ferromagnetic product using coal as a reductant in roasting reduction process, in a rotary kiln. The roasted product can be concentrated using low intensity magnetic. The process conditions include:

- grade and recovery of iron in magnetic concentrate in 20% and 30% coal that increase from 46.3% to 55.9% Fe, as well as iron recovery from 55.88% to79.56%;
- in 40% of coal ratio, the iron grade decrease to 53.6% but iron recovery is reasonably high 81.99%;

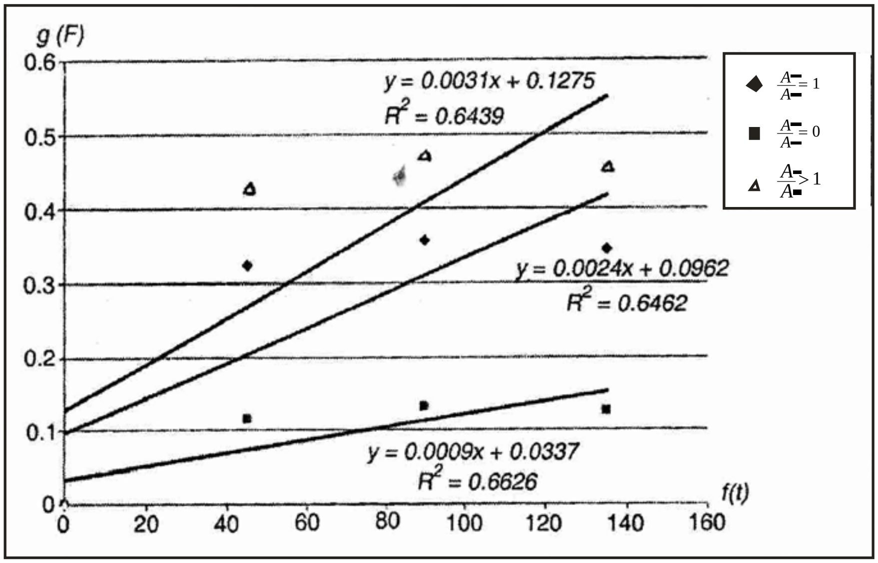

Figure 5. Relation between the conversion factor g(F) and time of controlling factor for the kinetic reduction of coal and iron lateritic pellet at 1,273º K

- the chage of limonite to metallic iron that performed at temperature of reduction of about 1,273° K suggests that is temperature condition is beyond the magnetite phase stability regions;

- diffusion of gas CO/CO_2 through a solid product layer governs reduction rate of lateritic iron pellet;

- further reduction after 135 minutes does not significantly increase the metallization as the resistance to reducing gas diffusion.

- The optimum condition is reached at 30% of coal ratio with iron recovery of 76.56%.

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