Exergy Evaluation on the Pellets Production and Direct Reduction Processes for the Fired and Nonfired Pellets

Tomohiro AKIYAMA, Reijiro TAKAHASHI and Jun-ichiro YAGI

Research Institute of Mineral Dressing and Metallurgy, Tohoku University, Katahira, Sendai, Miyagi-ken, 980 Japan.

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In order to make a clear evaluation of overall energy requirement for the production systems of directly reduced iron through pellets from iron ore powders, exergy analysis was applied to the two systems. The first one consists of pelletizing, firing and reduction processes, Fired pellets method, and the second one includes pellets curing process instead of firing process, Nonfired pellets method.

Being compared with enthalpy balance, exergy analysis has two advantages in that the exergy expresses the quality of energy and evaluates different kinds of energy like chemical, thermal, pressure and mixing energy by a unified measure.

As a result of exergy analysis, fired pellets lose less exergy than nonfired pellets in the shaft furnace. As the overall systems, however, exergy requirement in the nonfired pellets method is about 50 % of that in the fired pellets method. This marked difference is caused by the large exergy loss in the firing process. Furthermore, exergy loss in the production of nonfired pellets will be decreased by decreasing the amount of cement added as binder.

KEY WORDS: ironmaking; energy saving; direct reduction; iron ore; exergy; enthalpy; nonfired pellets; fired pellets; rotary kiln; shaft furnace; cement kiln.

1. Introduction

Marked attention has been recently focused on nonfired agglomerates from the point of view of energy savings^{1,2)} although fired agglomerates like sinter and pellets are usually used as raw materials to produce iron. The production process of nonfired pellets has the advantages not to produce injurious gas like NO_x or SO_x and also to accelerate reduction rate by the carbon contained in the pellets. However nonfired pellets include more amount of gangue than fired ones because several mass percent cement was added as binder. The cement addition increases the production cost. Although some reports^{3,22)} on reducibility and crushing strength of nonfired pellets have been already published, quantitative comparison of energy consumption is not yet reported between fired and nonfired pellets.

Traditional methods of thermal evaluation are mainly based on the first law of thermodynamics, which are called a heat balance and an enthalpy balance methods. In order to evaluate the quality of energy, exergy method based on the first and the second laws of thermodynamics is currently applied to many chemical industrial processes.⁴⁻¹¹⁾ In Japan, the calculation procedure for this method has been established in JIS Z9204 by Japanese Industrial Standards Committee in 1980.¹²⁾

The purpose of this paper is to estimate exergy requirement for two different production systems of directly reduced iron (DRI) from iron ore which are constituted from some processes for the production of pellets and a shaft furnace process for reduction of them. Exergy evaluation for both systems clarified

the locations of energy degradation which may lead to technological improvement. As a result, useful information on energy saving has been obtained.

2. Significance of Exergy

A heat balance and an enthalpy balance methods which are traditionally used for energy evaluation have following limitations. First, these methods only estimate the quantity of energy without considering the quality of energy. For example, mixing of 1 kg water at 0°C and 1 kg water at 80°C makes 2 kg water at 40°C within an adiabatic system.⁶⁾ A heat balance or an enthalpy balance method shows us there is no energy difference between before and after the mixing according to energy conservation principle. But, the quality of energy is undoubtedly decreased by the mixing because considering the reverse way which divides the 2 kg water at 40°C into original situation. It is impossible to rise the temperature of the water from 40 to 80°C without any heat source. Accordingly, this example indicates that thermal evaluation based on these methods gives different result from actual phenomenon.

Secondly, top pressure recovery turbine (TRT) is widely employed to recover energy of pressure at the top of a blast furnace. Energy of pressure is currently focused in other fields, *e.g.*, pressure in a cave and the method of its evaluation is increasingly important for energy storage. However, these balance methods do not estimate directly pressure energy.

Thirdly, energy is usually stored in a substance as the form of chemical energy. However, a unified measure to estimate chemical energy by these balance

methods is not proposed.

In this study, the method of exergy which overcomes the problems mentioned above is employed to evaluate two different systems for the production of DRI. Fig. 1 shows the concept of exergy. Exergy means available work, in other words, the potential, quality, of energy. Total energy at the standard condition is given by enthalpy ($H-H_0$) which consists of heat $T(S-S_0)$ and free energy ($G-G_0$). The free energy changes completely into exergy without loss. But, the heat has to obey Carnot's law therefore only the term of $T(S-S_0) \times (T-T_0)/T$ is obtained as exergy. It is found from Fig. 1 that exergy is finally expressed by $(H-H_0)-T_0(S-S_0)$ and the remaining part of the heat $T_0(S-S_0)$, which is called anergy or unavailable energy, is discarded to environment.

Table 1 shows the fundamental equation^{8,13)} of exergy for steady flow system. Atmospheric condition is selected as the standard condition which is defined as follows:

$$P_0 = 1 \text{ atm,}$$

$$T_0 = 298.15 \text{ K,}$$

$$\text{Air composition: } (75.60\% \text{N}_2 - 20.34\% \text{O}_2 - 3.12\% \text{H}_2\text{O} - 0.03\% \text{CO}_2 - 0.91\% \text{Ar}).$$

In the calculation of process exergy flow, at first, mass balances of operating data are checked. Chemical, thermal, pressure and mixing exergy are individually calculated and then are summed up by using a micro-computer. The program used for its calculation takes into account temperature dependency of average specific heat of each substance.

Table 2 shows standard chemical exergy of some substances which appear frequently in ironmaking processes. Unstable substances under the standard

condition have in general large value of standard chemical exergy and vice versa. For instance, standard chemical exergy of Fe_2O_3 is zero because it does not have any reaction potential under the standard condition. Fe_3O_4 , $\text{Fe}_{0.947}\text{O}$ and Fe are formed by the reduction from Fe_2O_3 in the ironmaking process. Iron oxide therefore increase its standard chemical exergy along with the progress of reduction.

For exergy of fuel such as oil and coal, the exergy evaluation is very difficult from the first principle due to their complicated structure therefore Rant's approximate equations¹⁴⁾ given in Table 3 are employed to evaluate exergy of fuel.

3. Exergy Evaluation for the Production Systems of DRI

3.1. Exergy Evaluation

Fig. 2 shows two systems for the production of DRI by fired pellets method and nonfired pellets method. Exergy analysis is applied to the five constituent processes which are as follows;

- 1) crushing,
- 2) cement production,
- 3) pellets curing,
- 4) pellets firing, and
- 5) pellets reduction.

Whereas pelletizing, transportation and char crushing process are not evaluated because of their relatively small energy consumption.

Before calculation of exergy, closed systems are set up by control surfaces for each process. After heat and mass balances of operating data are checked, net exergy loss is calculated by the following equation;

$$\epsilon_{\text{loss}} = \epsilon_{\text{in}} - \epsilon_{\text{out}} \dots \dots \dots (1)$$

Even if conservations of heat and mass are realized, exergy is not always conserved, because entropy always increases in the irreversible process. The net exergy loss means the decrease in quality of energy in the process. In other words, it is the net exergy loss required by the operation of the process, therefore, this value is very important for process evaluation. Net exergy losses in each constituent process are calculated and then are summed up to obtain overall exergy loss for the system considered. Energy flow and efficiency for two different systems from iron ore

Fig. 1. Concept of exergy.

Table 1. Equation of exergy.

$$\begin{aligned} \epsilon &= \sum n_i \epsilon_i^0 \\ (\text{Chemical}) & \\ &+ (\sum n_i \bar{G}_{Pi}) \{T - T_0 - T_0 \ln (T/T_0)\} \\ (\text{Thermal}) & \\ &+ (\sum n_i) RT_0 \ln \{ \sum (P_i/P_0) \} \\ (\text{Pressure}) & \\ &+ RT_0 \sum [n_i \ln \{n_i / (\sum n_i)\}] \\ (\text{Mixing}) & \end{aligned}$$

Table 3. Rant's equation to estimate exergy of fuels.¹⁴⁾

Gas	$\epsilon = 0.950 H_h$
Liquid	$\epsilon = 0.975 H_h$
Solid	$\epsilon = H_l + rw$

Table 2. Standard exergy.

	Gas							Solid								
	N ₂	O ₂	H ₂ O	CO ₂	H ₂	CO	CH ₄	SiO ₂	Al ₂ O ₃	MnO	CaO	C	Fe ₂ O ₃	Fe ₃ O ₄	Fe _{0.947} O	Fe
(kJ/mol)	0.71	3.93	8.63	20.14	235.39	275.55	830.74	0.00	0.00	100.36	110.41	410.83	0.00	96.97	118.74	368.41

Fig. 2. Two different systems for directly reduced iron.

to DRI are discussed by using the net exergy loss.

3.2. Exergy Loss of the Production of Nonfired Pellets

3.2.1. Crushing of Iron Ore

Exergy of crushing is expressed by Bond's work index W_i .¹⁵⁾ The W_i is equal to the work for crushing 80 % of 1 t raw materials of infinite mass to 100 μm . For Fe_2O_3 , W_i is 73 MJ/t-Fe. This value is used in calculating exergy for crushing of iron ore in both systems.

3.2.2. Cement Production

Nonfired pellets contain several mass percent cement as binder. A process of the production of cement consists principally of three sub-processes; crushing and firing of limestones and crushing of clinker. One of the sub-processes, firing of limestones, needs large energy in comparison with other two sub-processes. A new suspension preheater (NSP) kiln shown in Fig. 3 is mainly operated for firing limestones in Japan. The kiln has four cyclones in the upper part to recover heat of output gas and one furnace to precalcinate of limestones ($\text{CaCO}_3 = \text{CaO} + \text{CO}_2$).

Operating data for the NSP kiln of 3 700 t/d is used for evaluation. As indicated by the results in Table 4, net exergy loss of this sub-process is 1 776 MJ/t-clinker. In addition, net exergy loss for crushing of limestones and clinker is 42 and 64 MJ/t, respectively. Therefore, total net exergy loss for cement production is 1 882 MJ/t-cement.

3.2.3. Pellets Curing

Yard curing method¹⁶⁾ needs little energy except solar energy. However, its method takes long time, i.e., 9 to 11 days for curing period and needs wide area, too. On the other hand, continuous rapid curing (CORAC) method¹⁷⁾ is also proposed for curing pellets. It consists of two shaft furnaces, which overcome these shortcomings mentioned above. The latter process therefore is selected for exergy evaluation in this paper.

As shown in Fig. 4, the direction of gas injection is perpendicular to solid flow in two shaft furnaces.

Fig. 3. NSP kiln process for cement.

Table 4. Exergy flow in NSP kiln process.

		Exergy (MJ/t-clinker)
(Inflow)	Coal	3 471
	Gas	1
	Raw material	8
	Dust	1
	Total	3 481
(Outflow)	Clinker	1 311
	Kiln gas	267
	Cooler gas	126
	Dust	1
	Total	1 705
Loss		1 776
Efficiency (%)		49.0

Nonfired pellets are preheated by warm air (70 to 120°C) and then are carbonated ($\text{Ca}(\text{OH})_2 + \text{CO}_2 = \text{CaCO}_3 + \text{H}_2\text{O}$) by CO_2 -containing gas (30% CO_2 -25% H_2O -45% air) in the first shaft furnace. After that they are dried out by hot gas (220°C) and are finally cooled by air in the second shaft furnace.

This process removes approximately 9 mass% water of nonfired pellets and then gives necessary crushing strength. To use this process, harmful gas such as NO_x and SO_x is suppressed to generate and industrial output gas can be used because the temperature of the gas is sufficient for this process.

In fact, this CORAC process produces at present Si-Mn pellets containing carbonaceous materials on 100 t/d scale.¹⁸⁾ Exergy calculation is conducted with the data predicted on the basis of this operation. Net exergy loss in the CORAC process is 138 MJ/t-Fe as listed in Table 5.

3.3. Exergy Loss of the Production of Fired Pellets

Fig. 5 shows a rotary kiln process with a traveling grate which is on 9 000 t/d scale. Green pellets are preheated to remove adhered and combined water on the traveling grate. After that, pellets are heated up to 1 300°C by combustion of coal in the rotary kiln.

Fig. 4. Continuous rapid curing process.

Fig. 5. Grate rotary kiln process for firing pellets.

Table 5. Exergy flow in continuous rapid curing process.

	Inflow	Outflow (MJ/t-Fe)
Pellets	2 768	2 772
Preheating gas	55	2
Carbonating gas	69	62
Drying gas	145	49
Cooling gas	0	14
Total	3 037	2 899
Exergy loss	138	—
Efficiency (%)	95.5	—

Table 6. Exergy flow in grate rotary kiln process.

	Traveling grate	Rotary kiln	Annular cooler (MJ/t-Fe)
(Inflow)			
Pellets	88	699	1 114
Coal	0	1 022	0
Gas	1 026	888	2
Electric power	—	6	—
Total	1 114	2 615	1 116
(Outflow)			
Pellets	699	1 114	0
Gas	0	1 026	486
Total	699	2 140	486
Loss	415	475	630
Efficiency (%)	62.7	81.8	43.5

Finally, sensible heat of pellets is recovered by blowing air from the bottom of an annular cooler. Net exergy loss in this process listed in Table 6 amounts to 1 520 MJ/t-Fe.

3.4. Exergy Loss of the Reduction of Pellets

Reduction experiments of the pellets were carried out by using an experimental apparatus shown in Fig. 6, which produced DRI at the nominal rate of 1 t/d. Since this apparatus has already explained in the previous papers,¹⁹⁻²²⁾ detail explanation is not reproduced here. As raw materials, three kinds of pellets, N and W fired pellets and SC nonfired pellets including char of which chemical composition are

Fig. 6. Schematic diagram of experimental apparatus.

listed in Table 7 were charged into the shaft furnace. Each pellet has 12 to 14 mm diameter. SC pellets have higher gangue composition such as CaO because they included 8 mass% cement.

Experimental conditions used here are shown in Table 8. Conditions of temperature and pressure at inlet gas were almost same for three Runs. Runs F-1, F-2 and C-1 used N, W fired pellets and SC nonfired pellets, respectively. Reducing gas is usually produced by steam reforming of natural gas. It is therefore multi-component gas (H_2 -CO- H_2O -CO₂-CH₄). In the experiments the multi-component gas which was produced by mixing some kinds of pure gas from cylinders was used in Runs F-1 and C-1, while pure hydrogen gas was chosen in Run F-2 to examine the influence of hydrogen reduction in comparison with carbon monoxide reduction. A process for the production of reducing gas was omitted from the exergy analysis in this study because it is common in both systems for the production of DRI.

Exergy balances of three Runs are shown in Table 9 and Fig. 7. Enthalpy balances are also shown in Fig. 8. Comparing Fig. 7 with Fig. 8, enthalpy is larger than exergy. It is because enthalpy is composed of exergy and anergy. The relation among enthalpy, exergy and anergy in Run F-1 is shown in

Table 7. Chemical composition of the pellets. (wt%)

Pellet	T.Fe	FeO	CaO	SiO ₂	Al ₂ O ₃	MgO	MnO
N	64.43	0.26	2.62	2.53	0.48	0.06	—
SC*	62.90	0.15	6.43	2.13	0.51	0.04	—
W	64.60	0.26	0.26	3.71	2.18	0.14	0.66

* Except char (5.0 wt%)

Table 8. Experimental conditions for the shaft furnace operations.

Run No.	T_g (K)	P_i (MPa)	$G \times 10^2$ (m ³ (STP)/s)	$W \times 10^3$ (kg(Fe)/s)	G/W (m ³ (STP)/kg(Fe))	Y_{H_2} (%)	Y_{CO} (%)	Y_{H_2O} (%)	Y_{CO_2} (%)	Y_{CH_4} (%)	Y_{N_2} (%)
F-1	1112	0.142	0.793	3.01	2.63	74.71	14.84	1.684	4.136	1.530	3.38
F-2	1180	0.116	1.44	6.24	2.30	100	—	—	—	—	—
C-1	1133	0.147	0.799	2.58	3.10	79.66	13.10	0.718	4.708	0.226	1.64

Table 9. Enthalpy and exergy in the shaft furnace. (MJ/t-Fe)

		F-1		F-2		C-1		NSC ⁽²³⁾	
		Enthalpy	Exergy	Enthalpy	Exergy	Enthalpy	Exergy	Enthalpy	Exergy
(Inflow)	Gas	35 271	28 982	31 830	25 436	36 092	29 342	27 605	22 874
	Pellets	138	88	50	33	2 751	2 734	59	42
	Total	35 409	29 070	31 880	25 469	38 843	32 076	27 664	22 916
(Outflow)	Gas	27 525	22 706	23 811	19 114	26 763	21 818	19 109	15 965
	Pellets	7 055	5 996	6 988	5 937	10 723	9 576	7 767	6 695
	Total	34 580	28 702	30 799	25 051	37 486	31 394	26 876	22 660
Loss		829	368	1 081	418	1 357	682	788	256
Efficiency (%)		97.7	98.7	96.6	98.4	96.5	97.9	92.7	98.9

Fig. 7.
Exergy balance in the shaft furnace.

Fig. 9. Inflow and outflow of enthalpy are equal, however, exergy is decreased in the shaft furnace due to increase in entropy. Net exergy and enthalpy loss in Run F-1, multi-component gas, are smaller than in Run F-2, pure hydrogen. This is caused by heat generation in the reduction of iron oxide by carbon monoxide.

Net exergy loss in Run C-1, nonfired pellets, is larger than in Run F-1, fired pellets. This result is interpreted by the following three reasons. First, dehydration of adhered and combined water contained in the nonfired pellets consumes exergy. Secondly,

extra exergy is necessary to heat up useless gangue. Finally, as nonfired pellets contain 5 mass% char to accelerate reduction, exergy is lost by endothermic gasification reaction of char ($C + H_2O = CO + H_2$, $C + CO_2 = 2CO$). However, exergy loss due to the gasification reaction is not so large in the experiments because weight loss of the char is very small amount.

Larger inflow exergy by charged pellets in Run C-1 in comparison with other runs, is caused by higher standard chemical exergy of carbon (See Table 2). To examine the scale effect, the result of exergy analysis on 500 t/d demonstration plant of Nippon Steel

Fig. 8. Enthalpy balance in the shaft furnace.

Fig. 9. Enthalpy, exergy and energy flows in the shaft furnace.

Fig. 10. Overall net exergy consumption for production of directly reduced iron.

Corp.²³⁾ are also listed.

Net exergy losses in the reduction process are 368 MJ/t-Fe for fired pellets and 682 MJ/t-Fe for nonfired pellets. Therefore, in the reduction process alone, exergy loss for fired pellets is smaller than for nonfired pellets.

3.5. Overall Evaluation of Two Systems

Fig. 10 shows overall net exergy losses for both systems. The fired pellets system consumes only half exergy of nonfired pellets system in the reduction process. However, as net exergy loss of the rotary kiln process is ten times larger than that of CORAC process, it is concluded that overall net exergy loss of fired pellets system is almost two times larger than that of nonfired pellets system. These calculated values are not definitely fixed. It may be affected by operating condition, operating technique, scale of the plant. Even if these effects are considered, the difference in exergy consumption between both systems is so large that the reverse result will not be obtained.

The nonfired pellets used in this study contained 8 mass% cement. However, according to the recent report,²⁴⁾ the nonfired pellets have sufficient crushing strength by addition of only 3 to 4 mass% cement. Based on this report, net exergy loss for addition of cement will be decrease to half value. CORAC process was supported as an energy saving process for curing pellets. However, if yard curing process is

employed, net exergy loss for curing pellets will be decreased. Consequently, overall net exergy loss in nonfired pellets system would be less than in fired pellets system.

4. Discussion

Exergy loss is caused by the following five reasons;

- 1) Heat loss through the wall,
- 2) Irreversible chemical reaction,
- 3) Heating and cooling,
- 4) Pressure drop, and
- 5) Mixing.

Exergy loss through heat loss is caused largely by imperfect thermal insulation, a generation of crack, discontinuous operation and heat transportation by water coolant. In general, heat loss of large scale process is recognized to be smaller than that of small scale process because surface area per inner volume is small in a large scale plant. In this study, exergy analysis was conducted for two different scales of shaft furnaces, which were 1 and 500 t/d, however, there is no distinct difference between both net exergy losses. This means the thermal insulation is excellent in the 1 t/d scale experimental apparatus.

One of typical examples of exergy loss caused by chemical reaction is combustion. It is noteworthy that two processes of the production of cement and the firing of pellets which include combustion of carbon consume a large quantity of exergy. This phe-

nomenon indicates that combustion control is important. When theoretically required amount of air is only consumed for combustion, exergy loss will be least. However, in fact, as many industrial processes do not need such high temperature of 2 200°C directly, excess air is introduced. This introduction of air causes needless exergy loss.

To decrease exergy loss caused by heating and cooling of charged materials, it is important that unnecessary materials is removed before heating. In the shaft furnace, as exergy is wasted by heating gangue and char added, appropriate addition weight of char and cement should be examined. Exergy losses caused by pressure drop and by mixing occurs from defective airtight condition and diffusion, respectively. In this study, the rate of these exergy losses was very small quantity and then negligible in every process.

A heat balance or an enthalpy balance method can only evaluate heat loss. Although heat and enthalpy are conserved, the phenomena from 2) to 5) undoubtedly degrade energy. It is noteworthy that outflow of exergy from each process was very large in comparison with net exergy loss. Therefore, it is desirable to develop high efficiency technique for exergy recovery.

5. Conclusions

(1) Overall net exergy loss in nonfired pellets system is approximately half as small as in fired pellets system. The overall net exergy loss may decrease further by less addition of cement with keeping strength of the pellets.

(2) Net exergy loss in the production process of cement is not small and occupies 40 % of net exergy loss in the production of nonfired pellets. Whereas, net exergy loss of CORAC process is considerably smaller than that of the rotary kiln process and the process is operated at high exergy efficiency.

(3) In the reduction process, nonfired pellets system is more disadvantageous in exergy consumption than fired pellets system due to included water, cement and char.

(4) The method of exergy analysis is attractive for the energy evaluation of ironmaking systems where not only heat transfer, but also separation of materials and chemical reactions take place simultaneously because it can evaluate both amount and quality of energy.

Nomenclature

- C_p : average specific heat (kJ/molK)
 G : free energy (kJ)
 G_g : gas flow rate of inlet gas (m³/s)
 H : enthalpy (kJ)
 Hh : high calorific value (kJ)
 Hl : low calorific value (kJ)
 n : mole (mol)
 r : heat of evaporation of water (kJ/kg)
 P : pressure (MPa)

- S : entropy (kJ/K)
 T : temperature (K)
 w : water content of fuel (—)
 W_s : mass flow rate (kg/s)
 W_i : grinding work index (kJ)
 Y : gas composition (—)

Greek symbols

- ε : exergy (kJ)
 ε° : standard chemical exergy (kJ/mol)

Subscripts

- i : i component
 0 : standard condition
 g : gas
 s : solid

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