

Aluminum Blast Furnace Process I. Prototype Furnace

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Abstract

In order to elucidate the fundamental features of an aluminum blast furnace, a small prototype furnace with a single oxygen lance has been constructed and operated. Even in coke combustion alone, a large amount of grey dusts were evolved. When bauxite was loaded, the evolution of dusts was enhanced. The dusts consisted mainly of alumina, silica, and coke powders. At the furnace shaft, these dusts coalesced cokes and, as a result, bridges formed frequently. The important observations are: (1) temperatures at other places than the combustion zone were lower than 2270 K, that is, a temperature required to reduce alumina, and (2) almost all bauxite briquettes were conveyed together with cokes to the coke/oxygen combustion zone. These features showed that bauxite was not reduced in the present furnace, but volatilized in the chemical form of SiO and Al₂O in the oxidative atmosphere inside the combustion zone. This resulted in a very low aluminum yield. The present results have suggested that a furnace should be well thermally insulated and supplied with enough heats.

I. INTRODUCTION

An aluminum blast furnace¹⁻¹²⁾ has attracted much attention in recent years especially in U. S. A. and Japan. The investigations in U. S. A.¹⁻⁹⁾ aim at producing directly aluminum-silicon alloys from clay and other minerals, whereas the main incentive of the investigations of Japan⁹⁻¹²⁾ is to seek an alternate, hopefully non-electrical, process of producing aluminum alloys.

Much work has been done to clarify the reduction of alumina by carbon at high temperatures, and revealed that aluminum alloys can be produced by carbon reduction, although it is rather difficult to produce pure aluminum metals.¹³⁻¹⁹⁾ The electrothermal process of producing aluminum alloys is however not energy-conservative when compared with the Hall-Heroult process, because reaction heats are supplied in the form of electricity. In an aluminum blast furnace, the heats are supplied by combustion of cokes. In this case, a large amount of coke should be consumed because of the extreme endothermic reaction and as a result, the corresponding amount of CO is co-generated in this process.¹⁸⁾

From the technological point of view, the followings should be investigated experimentally to establish a aluminum blast furnace process:

- (1) Can a blast furnace provide a high-temperature and reductive zone necessary to extract aluminum?
- (2) How does bauxite behave in the presence of excess coke and under a CO flow? In particular, the followings are important:
- (3) The volatilization of aluminum and silicon components may become severe under a CO flow;^{3-8,20)}
- (4) When carbides are the main products or intermediates in the presence of excess coke, it will be difficult to take the products out of a furnace;^{1,2)}
- (5) Alloys produced may be reoxidized when they are attacked by oxygen blast in the vicinity of the combustion zone.^{1,2)}

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We have investigated on the reduction of bauxite using electrical experimental furnaces and found that bauxite can be reduced even under a CO flow.²¹⁻²⁴ As a next step, we constructed an experimental furnace having no electrical sub-heater. We started with an experiment of burning cokes with oxygen and then studied the reduction of bauxite in the furnace. In the early stage of this investigation, the scale of the experimental furnace was found to be small to create a high-temperature and reductive zone. We therefore constructed larger furnaces, that is, a midsize furnace and a bench furnace. The present paper reports results of the investigation in the first prototype furnace. In the succeeding papers, results of the larger furnaces will be reported.

II. EXPERIMENTAL

In the present study, 13 experimental runs were carried out; these are designated as CR-1 to CR-13. The first five runs were for examining the coke combustion alone, the rest being for the reduction of bauxite in combination with the coke combustion.

A. Raw Materials and Briquettes.

Blast furnace coke particles (4 - 7 mm in diameter) were used as fuel, and bauxite was used as ores to be reduced. The composition of coke and bauxite is presented in Table 2. Iron powders were also used to adjust the atomic ratio of Al/Fe in some briquettes to 4.

Four types of bauxite briquettes used are listed in Table 1. Briquettes "BP" were natural bauxite particles (3 - 5 mm in diameter) after dried at 470 K for a night. Briquettes, designated as "BF", "BFC", and "LB", were prepared as follows; bauxite (-24 mesh), coke (-24 mesh), and iron (-12 mesh) were mixed in the mass ratio of 1000 : 300 : 62, kneaded and briquetted. Briquettes "BF" (20 - 25 mm in diameter) were used after dried at 470 K for a night. Briquettes "BFC" (25 - 30 mm in diameter) were made from "BF" coated with a 2 - 5 mm thick layer of coke powders using benzene-coal tars as paste; these were used after dried and baked at 770 - 970 K. A long briquette "LB" was moulded to hexahedron (20 x 55 x 800 mm), dried and reinforced with wood boards. Two types of slag flux, listed in Table 1, were used: Calcium/bauxite briquettes "BCa" (20 - 30 mm in diameter, CaCO₃/bauxite = 12/10 in mass ratio) were prepared from calcium carbonate (-300 mesh) and bauxite powders; the effect of magnesia addition was studied with magnesia/coke briquettes "MgC" (20 - 30 mm in diameter, MgO(-300 mesh)/coke = 40/14 in mass ratio).

B. Experimental Furnace.

Figure 1 shows an experimental furnace used. A graphite crucible (1000 mm high) was set in a water cooled vessel made of stainless steel. The inner diameter of the crucible was 150 mm for the initial four runs (CR-1 to 4), and enlarged to 275 mm afterwards (CR-5 to 13). Graphite fibers and alumina bubbles were placed as heat insulators between the crucible and the vessel. A graphite grate with many holes (10 mm in diameter) was set about 50 mm below a tuyere to help crude alloys and slags to drop smoothly.

The furnace was equipped with a single lance made of coaxial triple copper tubes; the inner tube was for oxygen path, the outer double tubes being for cooling water. The outlet diameter of the tuyere was adjusted to 5 mm or 10 mm with alumina tips.

C. Furnace Operation.

The operational characteristics of typical runs are presented in Table 1. Operation was generally made as follows: Charcoal was first loaded up to the tuyere level and ignited in oxygen blast. Oxygen was supplied from a liquid reservoir through an evaporator; the pressure and the flow rate of oxygen were controlled manually. Exhaust gases were burnt at a top opening. In the initial stage of operation, only blast furnace cokes were loaded and burnt for 3 to 4 hours to preheat the furnace. Bauxite briquettes and cokes were then loaded alternately from the top opening at intervals of 15-30 min. The height and other

features of a burden bed were inspected manually using a steel poking rod inserted from the top opening; when a bridge was formed, poking was made to destroy the bridge. Temperatures of furnace walls were measured by W-W/Re 26% thermocouples at six places indicated in Fig. 1. The coke combustion zone was visually inspected through the oxygen lance with an optical pyrometer. After each operation, a burden bed was dug up to inspect the inside.

D. Analysis.

Contents of metal elements (Al, Fe, Si, Ti, Ca) in products were determined mainly by atomic absorption method and supplementarily by X-ray fluorescence spectrometry. Carbon content was determined from the amount of CO_2 gas generated on burning samples with oxygen at 1720 K using copper oxidizer to complete the combustion especially of SiC.

To identify the chemical forms of products, X-ray diffractometric measurements were also made. When alloys were formed in products, it was difficult to derive the information of composition of alloys from X-ray diffraction patterns alone. Contents of metallic aluminum and iron were evaluated from the behavior of dissolution of finely ground samples into hydrochloric acid (1:1); aluminum and iron metals evolved H_2 on dissolution, and Al_4C_3 did CH_4 . The amounts of evolved gases were determined by gas chromatography, while the dissolved aluminum and iron were determined by atomic absorption method.

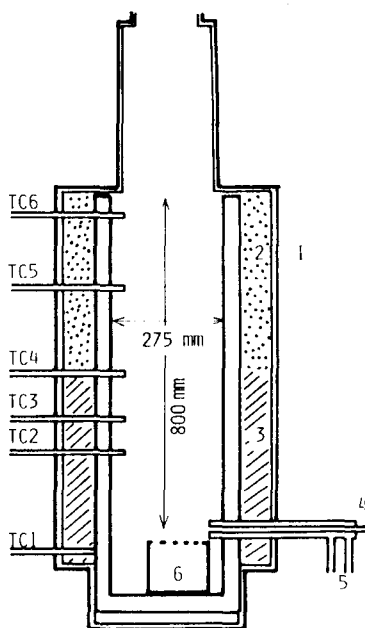


Fig. 1 Prototype furnace:
 1, Water cooled stainless steel;
 2, Alumina bubble; 3, Graphite
 fiber; 4, Oxygen lance; 5, Cooling
 water; 6, Graphite basin with
 grate (200 mm in depth).

Table 1. Operational characteristics of CR Runs.

condition	Run No.					
	CR-1-5	CR-6-9	CR-10	CR-11	CR-12	CR-13
Shaft diameter (mm)	150,275	275	275	275	275	275
Tuyere diameter (mm)	5, 10	10	10	10	10	10
Blast rate (dm ³ /min)	60,120	120	60	60(a)	60	50(a)
Height of bed (cm)	80	80	50	50	50	50
Coke/Briquette(kg/kg)	-	3	2	3.8	3	3.8
Briquette ^(b) (kg/type)	0	6.5/BP	5.5/LB	2.8/BFC	5.9/BF	3.2/BFC
Slag flux ^(b) (kg/type)	0	0	0	0	5/MgC	4.5/BCa
Alloys produced (kg)	0	0.6	-	0.45	0.4	1.54
State of shaft & tuyere level (-)	Heavy bridge Melt blown up	Heavy bridge Channel	Slight bridge Flooding	Slight bridge Channel	Slight bridge Flooding	Slight bridge Flooding

(a) CO₂ (8 dm³/min) was mixed in oxygen;

(b) BF, natural occurred bauxite particle (ϕ 3 - 5mm);

LB, long hexahedron briquette

(20 x 55 x 800 mm, bauxite/coke/iron = 1000/300/62 in mass ratio);

BF, ball briquette

(ϕ 20 - 25 mm, bauxite/coke/iron = 1000/300/62 in mass ratio);

BFC, ball briquette coated with coke powder

(ϕ 25 - 30 mm, bauxite/coke/iron = 1000/300/62 in mass ratio);

BCa, ball briquette (ϕ 20 - 30 mm, CaO : bauxite = 12 : 10 in mass ratio);

MgC, ball briquette (ϕ 20 - 30 mm, MgO : coke = 40 : 14 in mass ratio).

Table 2. Elemental analysis of products and burdens.
Total content of metal elements and content of the metallic state are given in mass percent.

Sample ^(a)	Total content						Metallic		X ^(b)	Y ^(c)
	Al	Fe	Si	C	Ca	Balance ^(d)	Al	Fe		
Bauxite	28.8	9.15	2.67	0.35	0.18	24.5				
Coke	1.70	0.68	2.60	87.8	0.56					
CR-7 RW Bottom	14.4	44.4	12.5	3.8	-	24.9	3.7	38.7	228	0.94
CR-11 DP	-	-	-	-	-	-	12.2	65.0	603	0.74
CR-13 DP	26.5	15.6	9.4	2.2	10.0	36.3	(11.1) ^(e)	(6.5)	36	(4.89)

(a) DP = dropped product; RW = raceway.

(b) X is the amount of evolved H₂ by HCl treatment (cm³/g).

(c) Y is the ratio of the calculated H₂ from the dissolved amount of Al and Fe into HCl to the corresponding value of evolved H₂, X.

(d) Ignition loss for bauxite; for samples RW and DP, oxygen is also included.

(e) The values of metallic contents given in parentheses indicate that CaO promoted dissolution of Al₂O₃ & FeO.

The measured amount of evolved H_2 was found to be usually larger than the amount calculated from the measured amount of dissolved aluminum and iron. This is due probably to the excess H_2 evolved from metallic silicon, although ferrosilicon in the silicon-rich region is not dissolved completely in hydrochloric acid. On contrary, when calcia was present in samples, the amount of evolved H_2 was less than that of the dissolved metals, as shown in Table 2 (CR-13); the X-ray diffraction patterns of dissolution residue confirmed that alumina and ferrous oxide were also dissolved in hydrochloric acid in such cases. Thus, there remained some uncertainty in determination of metallic aluminum and iron contents. We tentatively adopted the dissolved values as contents of 'metallic' aluminum and iron (see columns of 'Metal Al' and 'Fe' in Table 2).

III. RESULTS

The main results of the present experiments can be summarized as follows:

- (1) In the combustion of cokes, a large amount of dusts were generated. Correspondingly, bridges were frequently formed in the furnace.
- (2) When bauxite briquettes were loaded, the severe volatilization of aluminum and silicon components occurred.

Details are described below for the important features of the observations.

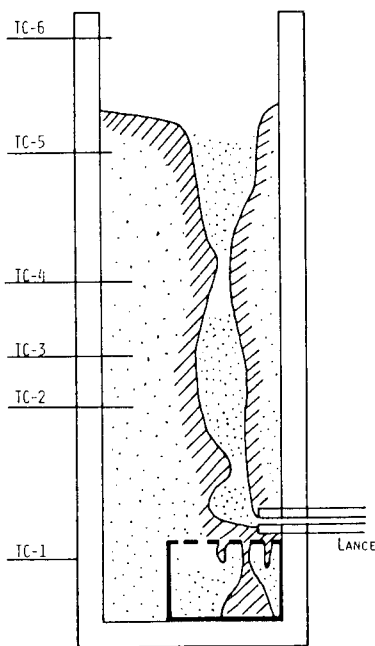


Fig. 2 Schematic dissection diagram of CR-6,7,8,11:
Hatched area, firmly coalesced part; Dotted area, loosely packed part; Blank, void.

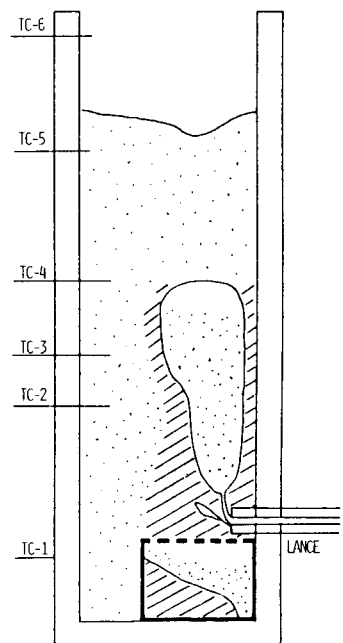


Fig. 3 Schematic dissection diagram of CR-12 and 13:
Hatched area, firmly coalesced part; Dotted area, loosely packed part; Blank, void.

A. Temperatures of Cokes and Furnace Walls.

The observed temperatures at the surface of burning cokes can be summarized as follows:

- (1) The highest temperature of burning coke surfaces was about 2670 - 2770 K when bauxite was not loaded;
- (2) During loading bauxite, the temperature decreased to 2370 - 2570 K;
- (3) The addition of CO₂ gas (CO₂/O₂ = 8/50 - 8/60 in volume ratio) also lowered the temperature to 2270 - 2470 K.

B. Dissection of Furnace.

The schematic dissection diagrams of furnace are shown in Figs. 2 and 3. In CR-6, 7, 8, 11, there remained a bird-nest-like void and loosely packed cokes in front of the tuyere (Fig. 2), where vigorously moving cokes were observed during operation. Apparently, this void was a so called "race way". Above this race way, there formed a hollow in which cokes were packed loosely. The wall of the race way and the hollow consisted of firmly coalesced cokes and slags; cokes at the inner wall of the hollow were glossy. The loaded briquettes were found only inside the hollow. In addition, outside the hollow, parts of pre-loaded charcoal remained without being burnt. This suggests that cokes and briquettes did not descend uniformly but along with the hollow. Dusts were also found only in the hollow. This indicates that gases went up through the hollow in countercurrent with descending coke and briquettes; in other words, so called "channelling" took place in the shaft.

When coke/briquette ratio was reduced (CR-9), and also when a long briquette "LB" was used (CR-10), crude alloys and slags flooded and coalesced around the tuyere; as a result, a discrete combustion zone was not formed. When slag flux was used (CR-12 and 13), the combustion zone grew up to a large dome as shown in Fig. 3.

C. Dust and Bridge.

A large amount of grey dusts were found in exhaust gases during the coke combustion. The amount of dusts increased when bauxite was introduced. The dusts were identified as a mixture of coke (graphite), alumina, and silica by X-ray diffractometry.

The dust generation was found to be closely related to the bridge formation. Dusts first coalesced cokes in the furnace shaft; once cokes started coalescing, this changed the flow route of gases containing condensable species. This in turn accelerated the formation of a firm "bridge" and "channel" as shown in Fig. 2. When a bridge was going to be formed, the oxygen blast pressure increased and the wall temperature was elevated anomalously just below the bridge, usually at the level of TC-5 in Fig. 1. Bridging was especially severe in the initial 4 runs where the shaft diameter was small (150 mm); in CR-4, burdens in the furnace were blown up finally. The furnace diameter was thus widened to 275 mm. This, in addition to keeping a burden bed height low, made bridging milder than before. Nevertheless, the destruction of bridges by poking was required to continue operation throughout all the runs.

D. Products and Analysis.

Alloys and slags were found below the graphite grate and at the bottom of the combustion zone. The X-ray diffractometric results of these crude alloys and slags indicated the presence of FeO, Fe₂O₃, mullite, α -Al₂O₃, Al₄C₃, graphite, β -SiC, and FeSi_x. In addition, there were strong diffraction lines at $2\theta(\text{Cu}) = 44 - 45^\circ$. This suggested the production of Al-Fe-Si alloys, because the lines disappeared after treatment with hydrochloric acid in which H₂ gas was evolved, and because these lines cannot be ascribed to FeSi_x nor Fe. When a calcia slag flux "BCa" was used in CR-13, aluminum calcium double oxides were found by X-ray diffractometry. When a magnesia slag flux was used, no magnesium component was detected in the crude alloys neither by X-ray diffractometry nor by elemental analysis (CR-12).

In CR-7 and 8, the total aluminum contents in the crude alloys (about 14 %) were much less than the initial aluminum content in bauxite (29 %), whereas the silicon and iron contents increased from 2.6 % and 8.9 % in bauxite to 10 - 12 % and 44 - 55 % in products, respectively (see Table 2). The content of metallic aluminum was only about 4 % in the products. In CR-9 and 10 where briquettes melted and flooded around the tuyere, no metallic product was detected. In CR-11, aluminum metal content increased to 12 % because of coating briquettes with cokes and of lowering combustion temperatures by mixing CO₂ in oxygen blast. In CR-12, the addition of a magnesia slag flux showed no appreciable effect on reduction enhancement. In CR-13 where a calcia slag flux was used under a similar condition to CR-11, the formation of stable aluminum-calcium double oxides resulted in the incomplete reduction of alumina.

IV. DISCUSSION

The present study has revealed that a large amount of dusts were produced even with the coke combustion alone; similar results were also obtained in an earlier study on the coke combustion with highly oxygen enriched air in an iron blast furnace.²⁵⁾ It is most likely that this dust generation will be intrinsic to the pure oxygen blast because of the high combustion temperature. With increasing the combustion temperature, the aluminum and silicon components in coke ashes tend to volatilize in the form of Al₂O(g) and SiO(g). On ascending in the furnace, these gaseous species tend to condensate at a low temperature zone and to form a bridge. There may be two possible ways of preventing the frequent formation of bridges. The first one is to decrease the volatilization of aluminum and silicon components. One of aims of the addition of calcia was to form slags in the furnace and to extract ashes to this slags. However, this did not work well in the present experiments. The second way of preventing the bridge formation is to prevent the condensation of the gaseous species in the furnace. The present operations suggested that the following methods may work well:

- (1) reducing the CO gas flow rate per furnace cross section area;
- (2) lowering a burden bed height to blow off dusts with the exhaust gas.

When bauxite was loaded in the present furnace, the followings were observed:

- (1) Bauxite moved directly to the combustion zone;
- (2) The major parts of the aluminum component in bauxite volatilized and caused the frequent bridge formation;
- (3) A small part of the aluminum component was reduced to dilute iron alloys and dropped below grate;
- (4) Other parts of the aluminum component in bauxite as well as cokes remained in raceway walls in the chemical form of dilute iron alloys or of carbides.

These features are apparently related with the formation of a channel. As shown in the dissection diagrams given in Figs. 2 and 3, the size of the combustion zone is relatively small and the hot CO gas ascended straightly in the channel. This determined the movement and chemical behavior of bauxite described above. For example, the typical content (about 4 %) of metallic aluminum in the present products is much smaller than the value of 35 - 39% obtained under a fast CO gas flow.²⁰⁾ This shows that the present main reaction of bauxite took place, not at a CO atmosphere, but at a rather oxidative atmosphere, that is, some area in the combustion zone. This is confirmed by the fact that plate-like metal lumps, found in front of the tuyere, had tracks of oxygen blast on their surface. To improve these features, several attempts have been made in the present study; however, the scale of the present furnace is too small to obtain constructive ideas except for coating briquettes which imply some improvement.

V. CONCLUSION

From the present investigation, it has been concluded that the heat supply and the thermal insulation in the present furnace were insufficient to elevate the temperatures of a burden bed up to higher than 2270 K. This is the reason for the low yield of metallic aluminum. Further investigation will be required with an experimental furnace with improved heat supply and thermal insulation.

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