Plasma treatment of aluminum dross

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ABSTRACT

In the recycling process of aluminum, a waste called aluminum dross is produced. It contains many kinds of refractory compounds like aluminum oxide and nitride, chlorides, and also heavy metals. Because arc plasma can generate high temperature, 5000-10000 ℃, it can be used to melt aluminum dross which has high melting point. Aluminum dross sample weight was 160 g, and the sample was pressed into pellets (10 x 50 x 20 mm) to be dropped into the plasma chamber easily. In the chamber the sample pellets were melted by plasma arc, and analyzed with XRD, EPMA, ICP-AES, and FL-AAS. The volume of the sample after the plasma treatment was reduced to about 40 % of that before treatment. During the plasma treatment the constituents of the sample were changed, aluminum oxide was increased from 33.1 % to 77.1 %, aluminum decreased from 23.1 % to 1.5 %, nitride from 3.0 % to 0.9 %, chloride from 7.32 % to 0.0080 % (= 80 ppm). Thermal plasma could be applied to melt the dross and the experimental results of plasma arc treatment are described.

1 INTRODUCTION

Japan uses much aluminum, 15 % in the world, and almost all of it is imported. Aluminum recycling is effective, but in the recycling process a waste called aluminum dross of 300,000 tons is produced in a year in Japan. It contains many kinds of refractory compounds like aluminum oxide and nitride, chlorides, and also heavy metals. Though at the present time it is disposed by reclaiming, environmental problems begin to occur, ammonia is produced by the reaction on aluminum nitride and water, and heavy metals flow out of reclaimed land. This is the reason why the way to establish to dispose aluminum dross is needed. Because arc plasma can generate high temperature, 5000-10000 ℃, it can be used to melt and solidify aluminum dross which has high melting point.

2 EXPERIMENTS

2.1 Experimental apparatus for generating plasma

This work was under laboratory scale. Fig. 1 shows the schematic feature of the experimental apparatus for the plasma generation. The plasma torch is D.C. transferred type. Generating power of the apparatus is 60 kW, with argon ($1.7 \times 10^{-4}\ m^3/sec$) as plasma gas. Chamber volume is 1.5 m^3. A graphite crucible is set in the chamber. Generated gas is measured with a mass spectrometer and drawn off in the air after neutralization with an absorber.

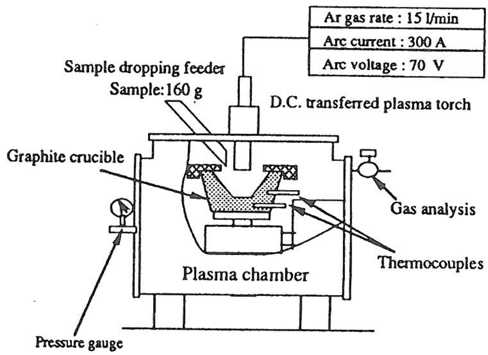


Fig.1 Arc plasma generater

2.2 Aluminum dross

Fig. 2 shows the recycling process of aluminum dross. Aluminum dross produced in aluminum refining process is called white dross. Since it contains much aluminum,

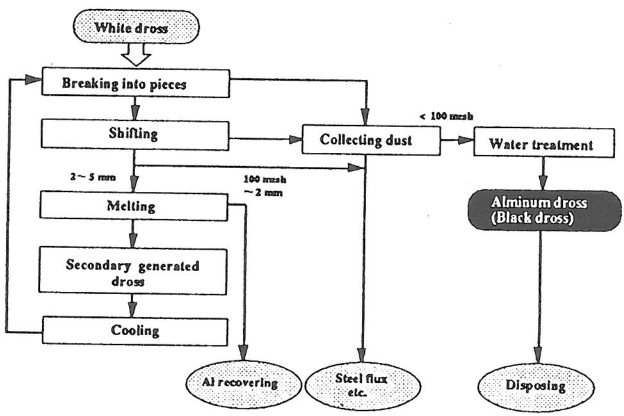


Fig.2 Recycling process of alminum dross

most of it is recovered and recycled. Aluminum dross used is just before reclaiming disposal, called black dross. White dross contains 50-70 % of aluminum, while black dross contains 20 % aluminum. It contains about 3 % nitrogen and 7 % chlorine.

2.3 Measurement of temperature distribution

Plasma generates high temperature 5000-10000 ℃. Since the main component of aluminum dross is aluminum oxide and it has high melting point, 2050 ℃ [1], temperature distributions of the graphite crucible and atmosphere in the chamber were measured. The relationship between generating power and temperature distribution was estimated from the results.

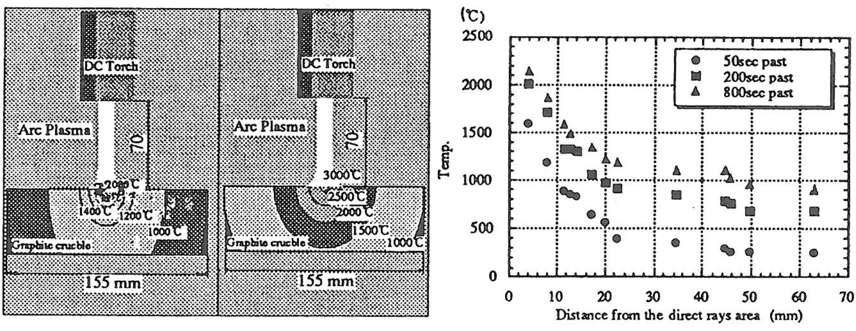
2.4 Plasma treatment of aluminum dross

Plasma treatment process of aluminum dross is described as follows. The dross sample is weighed 160 g and pressed at the shape of pellets (10 x 50 x 20 mm) to be easily fed into the plasma chamber. The samples are dropped on the graphite crucible in the chamber from sample dropping feeder, at the rate of 3 pellets/min. They are melted there with plasma arc. The gas generated during melting is measured with a mass spectrometer and neutralized with the absorber. The samples are analyzed with XDS, EPMA, ICP-AES and FL-AAS.

3 RESULTS AND DISCUSSIONS

3.1 Measurement of temperature distribution

Fig. 3(a) shows the temperature distribution of the crucible. The temperature in excess of 2000 ℃ was measured at the direct rays area of the plasma in Fig. 3(a). At the



(a) (b) (c)

Fig. 3 Temperature distribution of graphite crucible (a),(b)18 kW (c)28 kW area far from direct ray, large temperature gradient was observed by the conduction of heat of graphite. As heating time was much, the temperature increased. This phenomenon is shown in Fig. 3(c). Fig. 3(b) shows isothermal lines of the crucible at output of 28 kW. In Fig. 3(b), the temperature was over 3000 ℃. Temperature gradient was observed in the same as Fig. 3(a) and the measured temperature area in excess of 2000 ℃ was within 2 cm from direct rays area of the plasma. The temperature of the atmosphere in the chamber was also measured in the same method. The temperature of the atmosphere fell as it was far from the heating area. It was below 700 ℃ and the temperature of the chamber wall was 200 ℃ because it was water-cooled. Therefore we found that aluminum dross could be melt only near direst rays area and not with atmosphere in the chamber. Even if we raise output 28 kW and generate 3000 ℃, aluminum dross can be melted only at the area within 2 cm from direct rays. So we decided to treat it with direct rays area on 18 kW, 2000 ℃.

3.2 Plasma treatment of aluminum dross

3.2.1 Density and volume

In this process we can melt and solidify aluminum dross which is hard to melt. The density of the aluminum dross before and after the treatment and the volume ratio calculated from the density are shown in Table 1. Plasma melting increases the sample density from 1.02 g/cm^3 to 2.56 g/cm^3 and decreases the volume ratio to 39.8 % before the treatment. Therefore plasma melting is effective to decrease the volume of aluminum dross.

Table 1 Density and volume of samples

	Before plasma treatment	After plasma treatment
Density (g/cm³)	1.02	2.56
Volume ratio (%)	100	39.8

3.2.2 Qualitative analysis

The results of qualitative analysis are shown in Table 2. Dross before plasma treatment consisted of mixture of aluminum, oxide, and nitride, while dross after plasma treatment consisted of aluminum oxide and nitride, and spectrum of aluminum was not observed.

Table 2 Analyzed chemical species

Before plasma treatment	After plasma treatment
Al	AlN
AlN	Al_3O_3N
Al_2O_3	$Al_{8/3+x/3}O_{4-x}N_x$
KCl	
NaCl	
$MgAl_2O_4$	

3.2.3 Quantitative analysis

Table 3 Quantitative analytical result

	Before plasma treatment	After plasma treatment	Boiling point (℃)		Boiling point (℃)
Al (%)	23.0	1.5	2470		
Al_2O_3	33.1	77.1	2980		
SiO_2	6.7	11.4	1880		
Fe_2O_3	< 0.1	< 0.1	-		
Na_2O	16.6	< 0.1	1275	NaCl	1413
MgO	6.8	1.6	3600	$MgCl_2$	1412
K_2O	5.5	< 0.1	350 (Decomposition)	KCl	1500 (Sublimation)
CaO	< 0.1	0.2	2850	$CaCl_2$	>1600
N	3	0.9	-		
Cl	7.3	0.008	-		
Cr (ppm)	123	31	2670		
Pb	28	0.9	1740		
Cd	6.2	< D.L.	765		
Cyano compound	1.3	< D.L.	-		

D.L. : Detection Limit

(1) Aluminum

The result of quantitative analysis are shown in Table 3. Plasma treatment decreased the concentration of aluminum from 23.0 % to 1.5 %, whereas increased that of aluminum oxide from 33.1 % to 77.1 %. The increment of 44.0 % of aluminum oxide was almost equal to the value of 40.6 % estimated that aluminum is oxidized during the treatment. This accordance shows that aluminum was oxidized, not vaporized, because of the air getting into the chamber from the sample dropping feeder.

(2)Nitrogen

Plasma treatment decreases the concentration of nitrogen from 3.0 % to 0.9 %. Nitrogen almost seems to exist as AlN in the dross. Though AlN melts at 2200 ℃, it starts to sublime at 2000 ℃ [2]. So the concentration seems to decrease because of sublimation or decomposition with plasma. AlN powder produces ammonia by the reaction to water. While sintered aluminum does not react [2]. Plasma treatment not only decreases nitrogen but restrains the generation of ammonia by sintering.

(3)Chlorine

Plasma treatment decreases the concentration of chlorine from 7.32 % to 0.0080 % (= 80 ppm) and removes 99.9 % of chlorine out of the dross. Chlorine is casted as flux in the secondary refining process of the dross and forms chlorides of metal. Those chlorides seem to vaporize or decompose by high temperature of plasma because of their low boiling points. Aluminum dross is reported that in its addition to cement it can not degrade the quality of cement when the concentration of chlorine is below 500 ppm [3]. As dross after plasma treatment is low concentration of 80 ppm, it can be used for recycling as a cement material.

(4)Other metal element

Plasma treatment greatly decreased such metal elements as sodium, magnesium, potassium, calcium and iron from ~16.6 % to ~1.6 %. Table 3 show the boiling points of these oxides and chlorides. Though the boiling points of oxides are not necessarily lower than the melting point of aluminum, those of chlorides are lower than that of aluminum (2050 ℃) by 500 ℃. Such low boiling points compounds seem to vaporize at the temperature at which aluminum dross melts.

(5)Cyanide

Aluminum dross contains a little cyanide. The cyanide is reported to be produced by the reaction between nitrogen in the air and carbon of crucible [4]. 1.3 ppm cyan was also contained at the sample used. It seems to decompose by the heat of plasma because it was not detected after plasma treatment.

4 CONCLUSION

The results of this work are described as follows. This process can melt aluminum dross which is difficult to melt and decrease the volume to 39.8 % before the plasma treatment. This process can also remove 70 % nitrogen and 99.9 % chlorine out of the dross. Removal of nitrogen and increase of sintered aluminum are effective to restrain ammonia generating. Removal of chlorine will enable recycling of aluminum dross for cement material.

5 REFERENCES

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