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# Development of refining technology of metallic impurities in molten aluminum scrap

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## ABSTRACT

To promote aluminum scrap recycling, refining technologies of metallic impurities as silicon, iron and zinc from molten aluminum scrap were developed. Demonstration studies of vacuum distillation process in order to remove zinc, and fractional crystallization process to remove silicon were tested as 1,000t/month scale experiment through basic study from 1993 to 2002 year. It was possible to reduce zinc under 0.1% from aluminum scrap melt in 20kg/min continuous treatment by newly developed continuous agitating vacuum distillation process. And refining technology that has high silicon removal efficiency as 50% at the condition of yield rate 70% was confirmed by newly developed fractional crystallization process. This study was performed as part of the joint project "Research and Development of Technology to Promote Recycling of Non-Ferrous Metal Materials" by New Energy and Industrial Technology Development Organization (NEDO) and The Japan Research and Development Center for Metals (JRCM).

Key words: *Refining, Metallic impurities, Vacuum distillation, Fractional crystallization, Aluminum scrap, Recycling*

## 1. INTRODUCTION

Refining of metallic impurities such as iron, silicon and zinc in molten aluminum scrap is important to promote aluminum alloy scrap recycling into the wrought aluminum alloys. Unfortunately, we do not have a commercial method for decreasing such metallic impurities other than dilution with virgin Al ingot or Al scrap with low impurities. So, cascade recycling mainly for foundry alloy use is a popular system in Japan. But, in future, amount of aluminum scrap will increase beyond the amount of foundry alloy use. And product to product type recycling for wrought alloy use will be important to promote aluminum scrap recycling. Therefore refining technology to remove metallic impurities will be required. This study was performed from 1993 to 2003 as a part of the national joint project "Research and Development of Technology to Promote Recycling of Non-Ferrous Metal Materials" in Japan.

Selected target scrap is brazing sheet scrap for heat exchanger use, very difficult scrap to reuse because of clad aluminum scrap with 3000 alloy and 4000 alloy. Typical composition of molten brazing sheet scrap is estimated as Al-1-3wt%Si-0.9wt%Mn. It is difficult to remove Mn from molten aluminum alloy. So, Si and Zn are selected as metallic impurities to remove in this study. Even though there were several methods to remove metallic impurities in previous study, new economical process is necessary for refining aluminum scrap and following two technologies are proposed and developed.

## 2. VACUUM DISTILLATION PROCESS TO REMOVE Zn

### 2.1 Previous study

Vacuum distillation process, even though it is a well-known method to remove Zn and Mg from molten aluminum alloy as shown by Dimayuga et al. [1, 2, 3], is not the commercial method in Japan. Vacuum refining is advantageous because it poses no environmental threat, and Zn vapor can be reproduced as Zn-metal, which is easy to recycle.

In order to recycle aluminum scrap into the wrought Al alloy, it is desirable to keep the remaining Zn content less than 0.1wt%. Theoretical analysis indicated that was possible to keep the melt over 750°C in the vacuum pressure under 10Pa, or to keep the melt temperature over 950°C in the vacuum pressure 100Pa [4].

As indicated by Murphy [3], surface area/ volume ratio of melt is a important factor to increase Zn removal rate and/or remaining Zn content. Several methods in order to increase the surface area/volume of liquid metal were experimentally evaluated by 5kg/ch. large-scale vacuum furnace. A remarkable improvement on Zn removal behavior was obtained in mechanical stirring method. Remaining Zn content decreased with increase of stirrer rotating speed and increase of stirring time. Remaining Zn content less than 0.1wt% was obtained within 60s under the stirring condition 300rpm. Mechanical stirring method seems to be the most economical and commercial method in this experiment [5, 6].

Continuous process is proposed to reduce the running cost in vacuum distillation process. A water-model simulation shows that melt is strongly agitated with increase of rotating speed over 150rpm. This significant agitation of melt by stirring would be effective to increase the distillation rate. A continuous agitation vacuum distillation process (CAVP) was designed by using water model simulation. A CAVP pilot plant available for experiment of 1,000ton/month scale simulation as shown in Fig.1 was constructed at the Nikko works of The Furukawa Electric Co., Ltd. in spring 2000 [5, 6,7].

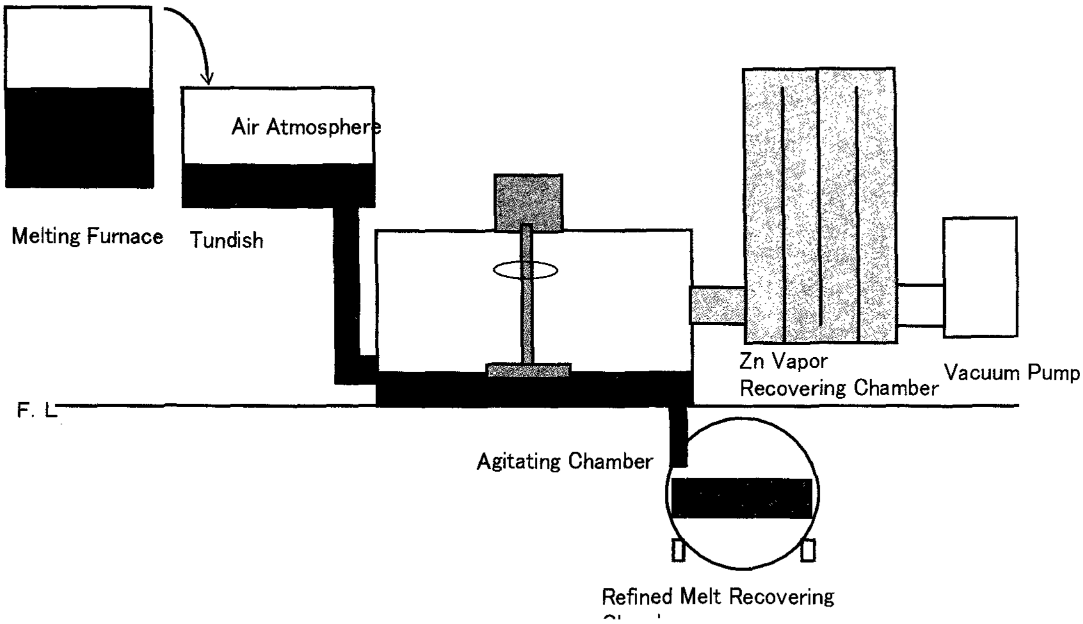


Fig. 1- Schematic Illustration of Continuous Agitation Vacuum Distillation Process

2.2 Experimental equipment

Capacity of melting furnace is 1,000kg. Scrap alloy melted in the furnace with air atmosphere is introduced continuously to pre-heated vacuum agitating chamber with rotating unit. Refined melt is also continuously discharged into the refined-melt recovering chamber throughout the hole in the bottom of the agitating chamber. Zn vapor is collected at the water-cooled plate in Zn vapor recovering chamber.

Refining characteristics of binary Al-Zn alloy and actual brazing sheet scrap in continuous flow condition of 20kg/min is investigated.

2.3 Experimental results

Melt of 1,000kg is discharged continuously from melting furnace to refined melt recovering chamber without any trouble of equipment. Zn content of refined melt is almost uniform. Effect of experimental conditions such as initial Zn content, melt temperature, initial pressure and melt flow rate is investigated by using binary Al-Zn alloys.

Example of continuous experiment in CAVP pilot plant is shown in Fig.2. Zn content after vacuum distillation decreases with increase of melt temperature and decrease of initial Zn content. Experimental conditions possible to achieve the target are made clear in 20kg/min continuous experiment. Refining characteristics of several kinds of actual aluminum scrap, including brazing sheet scrap is investigated. As shown in Fig.3 under the condition of melt temperature 950degree, flow rate 20kg/min and initial vacuum pressure <50Pa, Zn content of actual aluminum scrap after vacuum distillation is lower than that of binary Al-Zn alloy at the same initial Zn content.

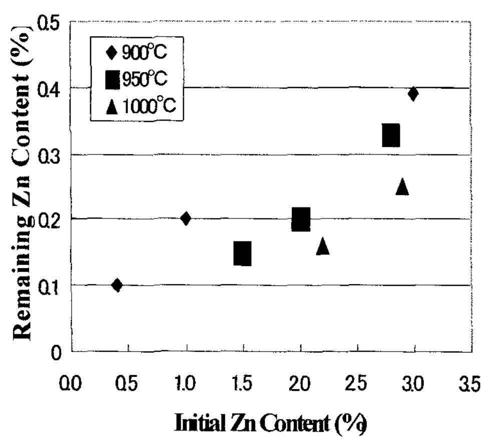


Fig.2 Effect of melt temperature in Al-Zn binary alloys

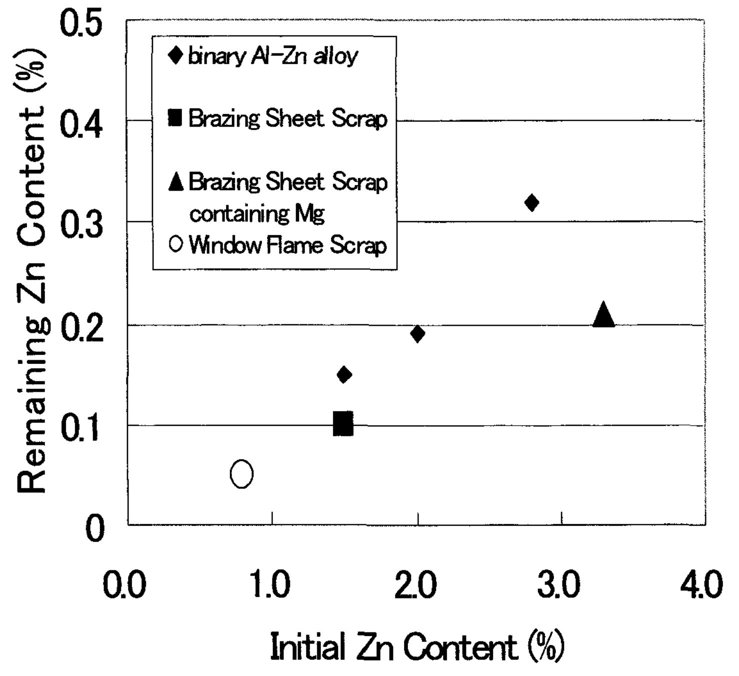


Fig.3 Experimental results in several kinds of actual aluminum scrap

## 3. FRACTIONAL CRYSTALLIZATION PROCESS TO REMOVE Si

### 3.1 Previous study

Refining by fractional crystallization is based on the equilibrium phase diagram. In case of equilibrium distribution co-efficient k<1, primary solid crystals having low impurities and remaining liquid having high impurities are co-existing in the solidification process. Refined alloy is produced by separating and collecting only the primary crystals. That technique is a well-known commercial method to produce high purity aluminum alloy. Lux and Flemings [8] have reviewed various separating methods as gravity sedimentation, centrifugal sedimentation, filtration, and filtration with compression. ALI, STUBINA and TOGURI [9] also describe an isothermal compression technique of reheated semi-solid samples with a filter. These methods are small scale reheated process and it takes a lot of time to make uniform temperature distribution in the sample. ALI [10] also describes about refining of 5XXX series aluminum alloy scrap by Alcoa fractional crystallization process.

A new fractional crystallization process was proposed. Refining characteristics was evaluated both by yield rate and impurity removal efficiency. Influence of basic experimental factors such as pressure and a temperature distribution of semi-solid material on the refining characteristics were investigated at the 8kg/charge laboratory scale experiment. Higher Si-removal efficiency was obtained by both cooling with stirring and cooling in the holding furnace than cooling in air. The design and the specification of the scale-up plant were given by this experimental result and the pilot plant available for experiment of 1,000ton/month scale simulation as shown in Figure-4 was constructed at the Nikko works of The Furukawa Electric Co., Ltd. in spring 2000 [11, 12].

### 3.2 Experimental equipment

Schematic illustration of proposed fractional crystallization process is shown in Fig.4.

Molten scrap alloy heated to 850°C in the melting furnace was poured into the pre-heated vessel. The melt in the vessel was cooled down to the experimental temperature in solid-liquid co-existing zone. In this 200-400kg/charge pilot plant experiment, primary crystals in the semi-solid materials were separated by the plunger with some holes at several pressures. Both weight and average composition of refined material were measured and refining characteristics are evaluated by Si removal efficiency and yield rate given by following equations.

$$\text{Removal efficiency}(\%) = \frac{\text{Initial composition} - \text{Refined composition}}{\text{Initial composition}} \times 100$$

$$\text{Yield rate}(\%) = \frac{\text{Weight of refined material}}{\text{Weight of initial material}} \times 100$$

### 3.3 Experimental results

Example of result in 200kg/charge experiment is shown in Fig.5. Si removal efficiency increase with increase of pressure and Si removal efficiency of 50% at yield rate of 70% is achieved at pressure of 21MPa. As height of press machine is districted in this experiment, vessel for 400kg/charge experiment has larger diameter compared with the vessel for 200kg/charge experiment. It

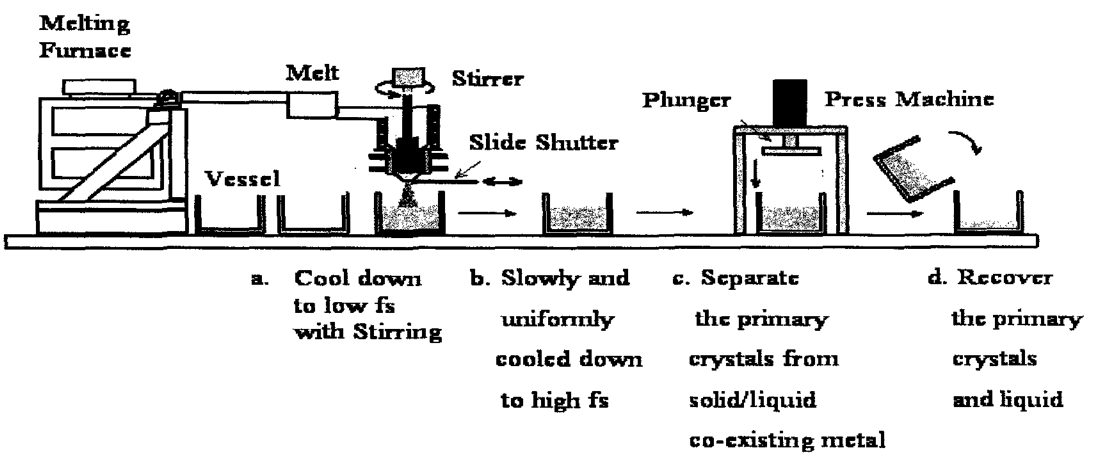


Fig.4 Schematic illustration of proposed fractional crystallization process

seems that large and/or long size of vessel results in un-uniformity of temperature distribution in solid-liquid co-existing metal in vessel. And pressure applied to metal in large vessel becomes smaller at the same press machine, compared with that in small vessel. In this 400kg/charge experiment, pressure applied to metal in vessel is estimated at 17MPa. So, Si removal efficiency may decrease with increase of vessel diameter.

Example of result in 400kg/charge experiment is shown in fig.6. Theoretical line which is calculated by using the thermo-calc, mathematical calculation software of equilibrium diagram, shows that Si removal efficiency decreases gradually with increase of yield rate. Experimental values show the same tendency and Si removal efficiency of experimental value is almost 80% of theoretical value at each yield rate. In this experiment, effect of cooling condition on Si removal efficiency is small and Si removal efficiency of 50% at yield rate of 70% is achieved.

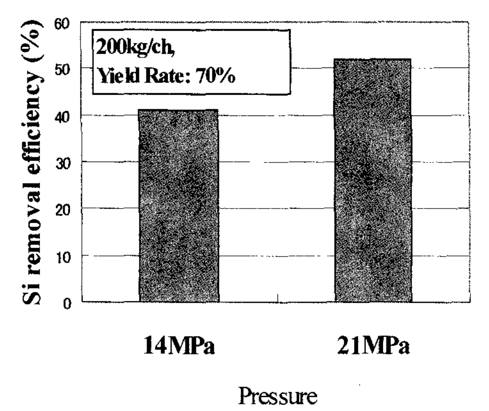


Fig.5 Example of the result in 200kg/charge experiment

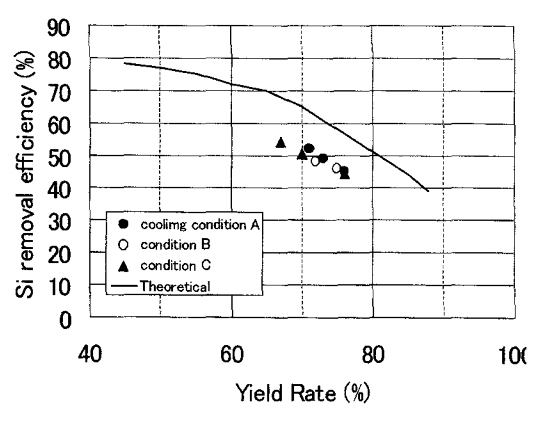


Fig.6 Example of the result in 400kg/charge experiment

## 4. CONCLUSION

To promote aluminum scrap recycling, refining technologies of metallic impurities as Si and Zn from molten aluminum scrap were developed. Brazing sheet alloy scrap is selected as target scrap. Demonstration studies of vacuum distillation process in order to remove Zn, and fractional crystallization process to remove Si were investigated by using the pilot plant available for 1,000t/month scale experiment. It was possible to reduce zinc under 0.1% from aluminum scrap melt in 20kg/min continuous treatment by newly developed continuous agitating vacuum distillation process (CAVP). And refining technology that has high Si removal efficiency of 50% at yield rate of 70% was confirmed in not only the 200kg/charge experiment but also the 400kg/charge one by newly developed fractional crystallization process.

It is possible to reduce metallic impurities in molten aluminum scrap not only at the laboratory scale experiment but also at the pilot plant scale experiment.

These developed technologies would be practical process and product to product type recycling would be popular in future.

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This study was performed as part of the joint project "Research and Development of Technology to Promote Recycling of Non-Ferrous Metal Materials" by New Energy and Industrial Technology Development Organization (NEDO) and The Japan Research and Development Center for Metals (JRCM).

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