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The Latest Refining Technologies of Segregation Process to Produce High Purity Aluminum

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High purity aluminum is widely used in many fields and the demand is increasing year by year. There are two refining processes to produce high purity aluminum, one is trinal electrolytic process and another is segregation process. Recently the most of high purity aluminum is produced with segregation process in Japan. But the purity of high purity aluminum refined with segregation process was normally lower than that with trinal electrolytic process. Then we have been promoting the development of technology to improve Pechiney's segregation process licensed in 1982 to enhance the purity of refined aluminum up to the level of trinal electrolytic process. We reviewed and studied the refining mechanism of Pechiney's segregation process, and examined the influence of many refining factors on refining efficiency. In consequence, we could establish the advanced segregation process to achieve the same purity as that with trinal electrolytic process.

Keywords: *high purity aluminum, segregation, refining process*

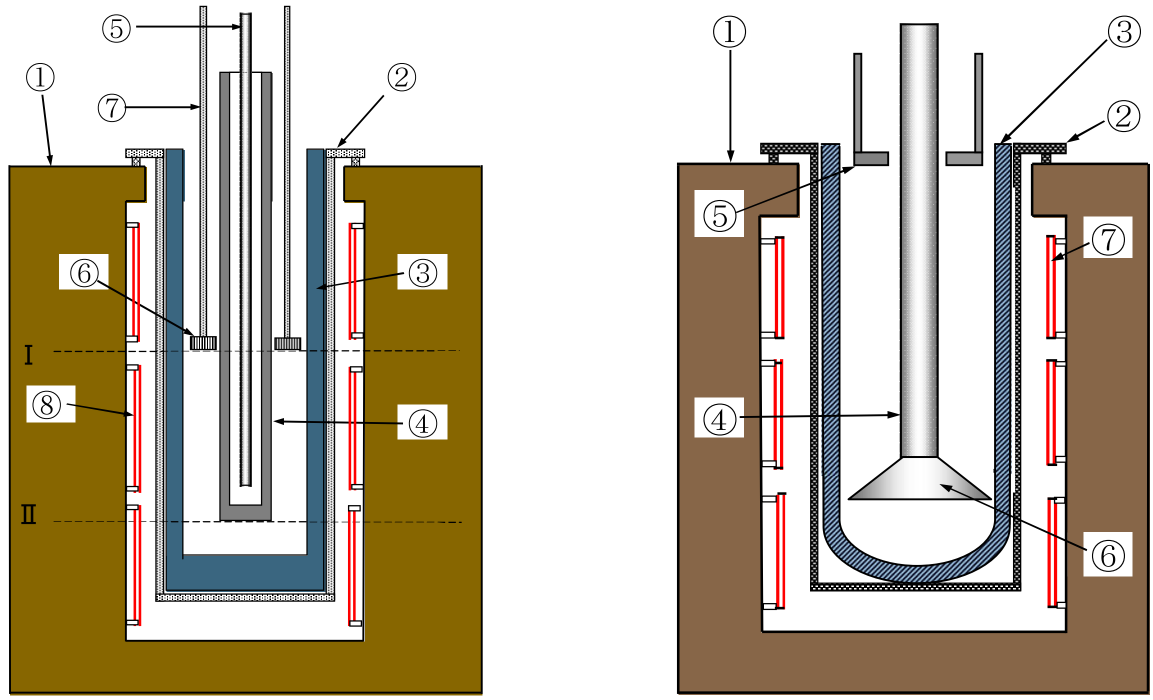
1. Introduction

High purity aluminum is applied for aluminum electrolytic capacitors, hard-disks, sputtering targets for semiconductor devices and LCDs, and others. The potential demands for these applications are increasing year by year. The principal application of high purity aluminum is aluminum electrolytic capacitor, which is used largely for home electric appliances, personal computers, industrial machines, motor vehicle and others. These applications are rapidly expanding in recent years. Moreover in near future, it is expected to develop new demands for such applications as compact self-ballasted fluorescent lamps, LED bulbs, solar power generation units and wind-power generation units. In the past, trinal electrolytic process had been used to produce all of high purity aluminum. While, in Japan segregation process started in the 1980's. Mitsui Aluminum imported the segregation technology from Pechiney in 1982, and started high purity aluminum production with the segregation process in 1983. However, the purity of aluminum refined with the segregation process was lower than that with trinal electrolytic process, then the application of high purity aluminum produced with the segregation process had been restricted.
Then Kyushu Mitsui Aluminum, who replaced Mitsui Aluminum, have been challenging to improve refining technology of the segregation process since 1992 so that the purity can achieve the same purity level as that with trinal electrolytic process.

2. Segregation Technologies

2.1 Segregation furnace

Pechiney segregation process is the way of gathering primary crystals industrially on the basis of the principle that the primary crystals are more pure than original metal. Fig.1 shows schematic drawings of segregation furnace. The left of Fig. 1 shows schematic sectioned view of Pechiney's original segregation furnace. The original furnace consists of furnace body, stainless-steel jacket, graphite crucible and plunger set comprising of inner graphite pipe, cooling pipe, graphite ring and graphite support. The original furnace was modified on account of its poor operability more than 30 years ago. Sectioned view of the modified current segregation furnace is given in right figure of Fig. 1. The operability of the segregation furnace was improved by changing mainly from plunger to tamper, piston and wiper. Additionally crucible was switched from special form to general design. But the mechanism or process of the modified furnace remained same as that of the original furnace, then the purity of the aluminum segregated with the modified furnace remained unchanged from that with the original furnace.



① Furnace body	⑤ Cooling Pipe
② Jacket for Crucible	⑥ Graphite Ring
③ Graphite Crucible	⑦ Graphite Support
④ Inner Graphite Pipe	⑧ Heater

Pechiney's original furnace

① Furnace body	⑤ Wiper
② Jacket for Crucible	⑥ Tamper
③ Graphite Crucible	⑦ Heater
④ Piston	

Modified furnace

Fig.1 Segregation Furnace [1-3]

2.2 Mechanism of Segregation

Our segregation method has four processes as shown in Fig.2. First process is for crystallization on the surface of piston. And second process is to scrape off the crystals on the piston's surface. Third process is to tamp the scraped crystals. These three processes are repeated during segregating. When the crystals deposited in the crucible attain a certain height of predetermined level, the segregation process is stopped. And high pure crystal aluminum in crucible is separated from residual molten aluminum by inclining crucibles like "(4) separating" in Fig.2.

It is very important for tamper to compress crystals deposited in crucible. The purity of crystals scraped off from piston's surface is slightly higher than that of molten aluminum. And there is impure molten aluminum in the interstitial space among the crystals. The tamper's repetitive compressing the crystals makes the impure molten aluminum pushed out from the interstitial space. The purity of the crystal block is becoming higher and higher by tamper's compressing. Furthermore, the temperature in segregation furnaces is controlled by computer to repeat a slight increase and decrease. When the temperature in furnace is slightly increasing, the crystals deposited in crucible are partially remelting and the impure molten aluminum in the interstitial space among the crystals is replaced with the remelting aluminum. When the temperature in furnace is decreasing slightly after that, the part of the molten aluminum in the interstitial space among crystals is refined by solidification. Thus the purity of the crystals deposited is becoming higher and higher by tamper's compressing and the repeat of remelting and solidifying.

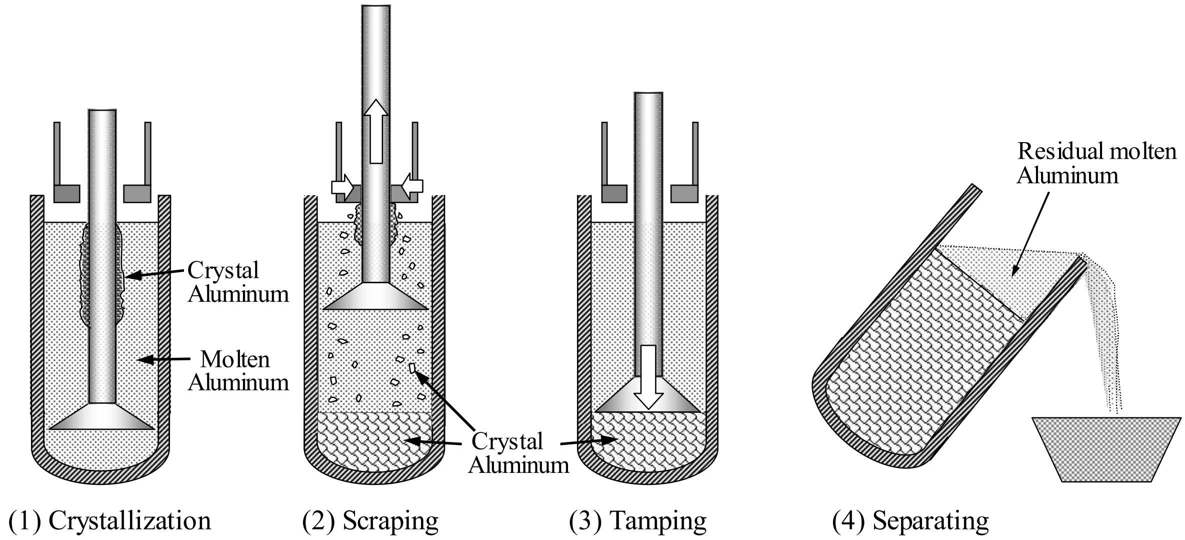


Fig.2 Mechanism of Segregation Process [2,3]

2.3 Characteristics of Pechiney's Segregation Process

High purity aluminum blocks produced with Pechiney's segregation process have its own characteristic concentration distributions of impurities. Fig. 3 shows Fe and Si distributions in a high purity aluminum block that were taken out of a crucible after segregated with a modified furnace. The contents of Fe and Si in a block are increasing toward the upper part and toward the center of a block. In the contrast, their contents are decreasing toward the lower part and the surface of a block. The similar results are also confirmed on the concentration distributions of other eutectic impurity elements.

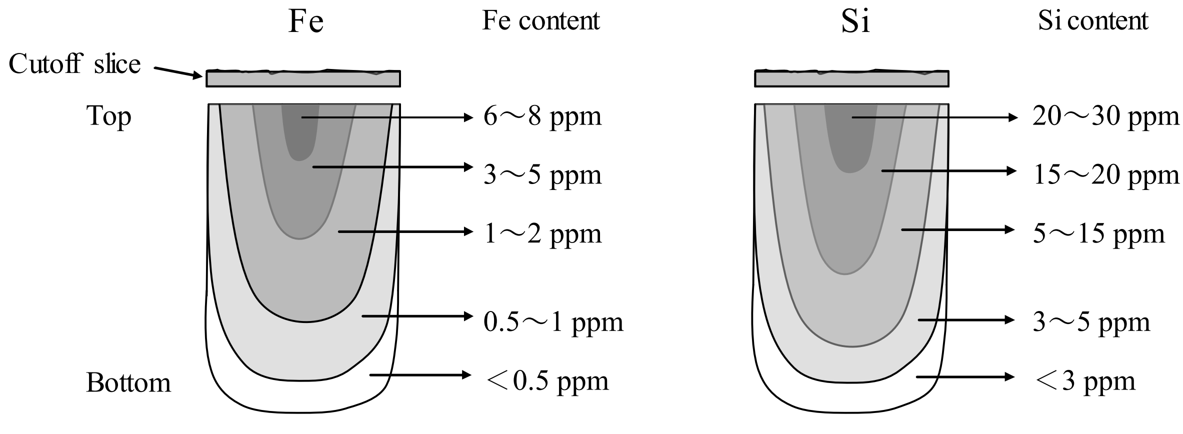


Fig. 3 Typical Fe and Si distributions in high purity aluminum blocks produced with Pechiney's segregation process [2,3]

The content of eutectic impurity elements is decreasing toward the lower part of blocks, which is caused by two reasons. First reason is that initial crystals starting to form deposit in crucible was solidified at first stage when the molten aluminum has still lower content of eutectic impurity elements. This leads to higher purity of initial crystals. Second reason is as follows. The content of eutectic elements in the crystal is decreasing by the repeat of remelting and solidifying during processing. The crystals at the bottom are exposed to a number of cycles of remelting and solidifying until end of segregation process. As a result, the crystals at the bottom become more and more high pure.

In traverse direction, the crystals in the near-surface of blocks are close to the heater of the furnace and shall be heated quickly in the case of heat up. Therefore, the crystals in the near-surface of blocks are exposed to a number of cycles of remelting and solidifying, and then become more and more high pure. On the contrary, the crystals in the center-part of blocks are not heated quickly and are not exposed to many repeats of remelting and solidifying. So the crystals in the center-part of blocks do not become more high pure. On account of these reasons, high purity aluminum block shows the concentration distributions of eutectic impurity elements as seen in Fig.3.

3. Development of new segregation technology

The main factors influencing on refining efficiency for Pechiney segregation process are in below. The optimum condition of each factor and the matching combination of these ten factors were investigated with many experiments, and new refining conditions bringing much higher refining efficiency were established.

(1) Rate of crystal rising
(2) Tamping pressure
(3) Cooling condition for pistons
(4) Final crystal's height in crucible
(5) Outside diameter of piston
(6) Cut off volume of blocks
(7) Tamping cycle
(8) Temperature in furnace
(9) Heating capacity of furnace
(10) Dimensions of crucible

The typical analysis results are shown in table 1 for high purity aluminum with new segregation process established under new refining conditions and trinal electrolytic processes. Previously, the purity of refined aluminum with traditional Pechiney's segregation process was lower than that with trinal electrolytic process, then it was believed that the trinal electrolytic process was suitable to produce higher purity aluminum. However the new segregation process enabled us to produce almost the same high purity aluminum as that with trinal electrolytic process as shown in Table 1.

Table 1 Typical analysis results of high purity aluminum produced with new segregation process and trinal electrolytic processes [2]

(ppm)

Process	Cu	Fe	Si	Mg	Zn	Ga
New Segregation Process	0.1 | 0.3	1 | 3	3 | 5	0.3 | 0.5	3 | 6	4 | 6
Trinal Electrolytic Process (sample A)	5	5	6	5	0.5	0.5
Trinal Electrolytic Process (sample B)	30	5	2	15	0.5	0.5

In particular, the new segregation process has much higher refining efficiencies to eliminate Fe and Cu than trinal electrolytic process. For Pechiney's segregation process, aluminum of purity over 99.90 % was used as feed metals. From this result, it would appear that the new segregation process has possibility to use aluminum less than 99.90% as feed metal. But the new segregation process will be required to have more high refining efficiencies for Zn and Ga as the segregation process has less refining efficiencies for these elements than trinal electrolytic process. This is the future task to enhance new segregation process.

4. Aluminum refining process in future

The demand of high purity aluminum will increase in future because the high purity aluminum is essential material in the electronics fields recently developing. Electronic parts are required to be more miniaturized, more weight saving, more reliable, highly efficient and further price-reduction without limit. For high purity aluminum, improvement and stabilization for quality, and price-reduction are strongly demanded. The high purity aluminum with both processes will meet their requirements for the quality. But to meet further cost-reduction demand, the trinal electrolytic process will become to make it more difficult to produce 99.99% up high purity aluminum especially in the countries having high electric power cost. Additionally, the submittal for CO_2 emission's data in the processes becomes to be requested from customers in recent years. CO_2 emission amount for electric energy with the new segregation process is about a tenth to a twentieth of that with the trinal electrolytic process. On account of cost-reduction and energy-saving, the segregation process will globally become a major process to produce high purity aluminum in future.

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