Removal of lead from scrap brass

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Method for lead removal from molten brass was investigated to reduce the concentration of lead in scrap brass. Granular Ca-Si compound was added to the molten brass to form large Ca-Pb-Si compounds. The Ca-Pb-Si compounds were then skimmed off from the molten brass. After casting into a metal mold, the brass ingot was characterized by EPMA and fluorescence X-ray analysis. The effects of grain size of Ca-Si compound, the method of adding Ca-Si compound, agitation time, holding time after agitation and compound removal temperature on the percentage of lead removal were discussed. It was confirmed based on the results that the compound method that uses granular Ca-Si compound to form Ca-Pb-Si compounds which are skimmed off from the molten metal enables the reduction of lead concentration.

Keywords: Lead removal, Brass, Compound method, CaSi₂

I. INTRODUCTION

In recent years, an unleaded alternative to steel, aluminum alloy and so on has been in demand.¹ ² Brasses that are used as alloys for faucets contain Pb. The elution limit for Pb tends to become severe. As one of the countermeasures, Pb-free brasses that are made of pure Cu, pure Zn and other alternative elements to Pb have been developed.³ ⁴ However, as a result, many problems such as high cost and depletion of resources arise. On the other hand, recycling is of major importance from the viewpoint of LCA. As a way of discovering an unleaded alternative and increasing recycling efforts at the same time, the method of removing Pb from scrap brass is suitable. As the methods for Pb removal, so far conventional vaporization and oxidation methods have been applied.¹ ² However, they may not be adopted today due to high cost, environmental pollution and so on. The method of forming Ca-Pb compounds by the addition of Ca followed by filtration using a pressurized filter was also attempted.⁵ Nevertheless, the percentage of Pb removal was low (29%) because the grain size of the compounds was small (several μm). However, the percentage of Pb removal may be increased by making the compounds larger. In the present study, the possibility of Pb removal by this compound method is examined.

II. EXPERIMENTAL PROCEDURE

As test material, leaded brass (JIS: CAC203, Cu: 58.1 mass%, Zn: 38.58 mass%, Pb: 2.15 mass%) was used. Figure 1 shows a SEM image of as-received brass. Pb remains undissolved within the matrix and is dispersed as fine particles of about 1 μm in diameter. In order to make the Pb compounds to float to the surface of a molten metal, granular Ca-Si compound consisting mainly of
CaSi$_2$ (Ca: 31.2 mass%, Si: 62.2 mass%, C: 0.63 mass%) was used as a promising additive.

![SEM image of microstructure of brass used.](image)

The reasons for choosing Ca-Si compound are as follows: (1) The melting point of Ca-Si compound is high (980°C), (2) Ca-Si compound may be maintained in a solid state at a molten temperature lower than 980°C, (3) Compounds formed easily float to the surface of a molten metal as a result of their low densities.

Brass was melted at 950°C using a high-frequency induction furnace. Granular Ca-Si compound was added to the molten metal in nitrogen gas atmosphere, followed by agitation. Formed compounds floated to the surface of the molten metal with time. The compounds were skimmed off from the molten metal. Thereafter, the molten metal was poured into a metal mold. Experimental conditions are listed in Table 1. Grain size corresponds to the size of the eyes of a sieve. Microstructures of the skimmed compounds and ingots were examined by EPMA.

### Table 1. Experimental conditions.

<table>
<thead>
<tr>
<th>Grain size of Ca-Si compound (mm)</th>
<th>Number of addition (times)</th>
<th>Agitation time (min)</th>
<th>Holding time after agitation (min)</th>
<th>Compound removal temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5–1.0</td>
<td>1</td>
<td>3.5</td>
<td>3</td>
<td>930</td>
</tr>
<tr>
<td>1.0–1.4</td>
<td>4</td>
<td>7</td>
<td>6</td>
<td>960</td>
</tr>
<tr>
<td>1.4–1.8</td>
<td>14</td>
<td>14</td>
<td>12</td>
<td>1006</td>
</tr>
</tbody>
</table>

Quantitative analysis of Pb in ingot was carried out by fluorescence X-ray analysis.

### III. RESULTS AND DISCUSSION

Figure 2 shows the relationship between the percentage of Pb removal and compound removal temperature. The percentage of Pb removal is very high at 930°C, but decreases with increasing compound removal temperature. Pb is hardly removed at 1006°C higher than the melting temperature of Ca-Si compound.

![Graph showing relationship between Pb removal and temperature](image)

Figure 3 shows results of the EPMA analysis of the compound skimmed off from the molten metal at 930°C. The grain size of this compound is very large. Based on results of the area analysis, the distribution of Pb is similar to that of Ca. On the other hand, Si exists around the compound. This suggests that the compound is Ca-Pb-Si compound. Thus, when the compound removal temperature is lower than the melting point of Ca-Si compound, Ca-Pb-Si compound may be readily skimmed off from the molten metal because of floating to the surface of the molten metal with as-solidified condition. The percentage of Pb removal under this condition is 43.7%. Thus the elution limit for Pb (0.01mg/L) may be achieved by the reduction of Pb content due to this compound method.
Figure 4 shows SEM images of the microstructures of ingots. At a compound removal temperature of 930°C, Ca-Pb compounds of several μm in diameter are observed. At 1006°C, fine-grained Ca-Pb compounds of nearly 1μm in diameter are dispersed. The grain size of these compounds is similar to that of Pb in as-received brass. This suggests that fine-grained Ca-Pb compounds are formed by in situ reaction. When the compound removal temperature is higher than the melting point of Ca-Si compound, added Ca-Si compound liquefies. Fine-grained Ca-Pb compounds, which are formed by in situ reaction, do not float to the surface of the molten metal, resulting in a low percentage of Pb removal.

Figure 5(a) shows the relationship between the percentage of Pb removal and grain size of Ca-Si compound. The percentage of Pb removal increases with an increase in grain size of Ca-Si compound. This is because the grain size of Ca-Pb-Si compound increases with that of Ca-Si compound, activating the rise of the Ca-Pb-Si compound to the surface of the molten metal.

Figure 5(b) shows the relationship between the percentage of Pb removal and the method of adding Ca-Si compound. The percentage of Pb removal decreases with an increase in the number of additions of Ca-Si compound. This suggests that most of the Ca-Pb-Si reaction occurs during the first addition of Ca-Si compound and the contribution of subsequent additions of Ca-Si compound is relatively insignificant.

Figure 6 shows the relationship between the percentage of Pb removal and agitation time. The percentage of Pb removal reaches a maximum value at 6 minutes, and decreases with a further increase in agitation time. The increase in the percentage of Pb removal at the early stage may be caused by an increase in opportunity for contact between Ca-Si compound and Pb. However, with further agitation after formation of Ca-Pb-Si compounds, it is likely that small Ca-Pb compound particles separate from
the large compounds particles and become suspended in the molten metal.

Ca-Pb-Si compounds are formed during agitation and then the compounds float to the surface of the molten metal during holding after agitation. The compound rises in a short period of time as its grain size is large. In Fig.7, the high percentage of Pb removal at 3.5 minutes is considered to be due to this. However, the percentage of Pb removal decreases as holding time is a little too large. This is caused by the separation of small particles from large compound particles as mentioned above.

Thus, the results reveal the optimum agitation time and holding time after agitation for Pb removal.

IV. CONCLUSIONS

1. The compound method that uses Ca-Si compound to from Ca-Pb-Si compounds which are skimmed off from the molten metal enables the reduction of Pb concentration in brass.
2. The percentage of Pb removal increases with an increase in grain size of Ca-Si compound.
3. It is most effective to add all of Ca-Si compound at one time.
4. There are the optimum agitation time and holding time after agitation for Pb removal.

REFERENCES

1 H.Nakae, J. Japan Foundry Engineering Society 73, 128(2001)
3 R.Mannheim, E.Ortiz and O.Bustos, Metall 51, Jahrgang, Nr. 4/97,190 (1997)

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