EVALUATION OF HYDROGEN IN LIQUID ALUMINUM BY MEANS OF THERMAL ANALYSIS

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Abstract
Thermal analysis technique may be used to predict the degree of grain refinement of hypoeutectic aluminum-silicon alloy. This technique is based upon an analysis and interpretation of cooling curve characteristics observed over the solidification of the alloy. Interestingly, it is observed that dissolved hydrogen appears to affect characteristics of the cooling curve. In order to gain insight whether such a relationship really exists, the present work investigates how the dissolved hydrogen in liquid aluminum alloy may affect the cooling curve characteristics. Casting trials were carried out using a hypoeutectic aluminum-silicon alloy. Emphasis was placed on different amounts of the dissolved hydrogen in the liquid alloy. During the trials samples of the liquid alloy were carefully controlled to minimize variations, such as composition, pouring temperature, cooling rate, which may undesirably confuse the thermal analysis. Regarding certain conditions addressed in the present study, it was found that the dissolved hydrogen appears to affect degree of undercooling of the cooling curve. Plausible mechanisms underlying this finding are noted.

1. INTRODUCTION
Thermal analysis technique may be used to predict the degree of grain refinement of hypoeutectic aluminum-silicon alloy. This technique is based upon an analysis and interpretation of cooling curve characteristics observed over the solidification of the alloy [1-4]. Nevertheless, previous studies reported that there appears to have some factors affecting thermal analysis of the cooling curve characteristics [5-6]. The present study reports experimental results addressing how the dissolved hydrogen in liquid aluminum alloy affect the cooling curve characteristics in terms of the degree of undercooling at the beginnign of primary aluminum nucleation and eutectic temperature change.

2. EXPERIMENTAL PROCEDURES

2.1 Materials
The alloy used in this present work was a hypoeutectic grade aluminum-silicon alloy, A356, one of the most widely used foundry alloys. Table 1 presents nominal composition the A356 alloy.

Table 1. Composition analysis of the A356 alloy used in this study (wt.%).

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Si</th>
<th>Mg</th>
<th>Cu</th>
<th>Mn</th>
<th>Fe</th>
<th>Zn</th>
<th>Ti</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>A356</td>
<td>6.76</td>
<td>0.20</td>
<td>0.02</td>
<td>0.06</td>
<td>0.56</td>
<td>0.11</td>
<td>0.08</td>
<td>remainder</td>
</tr>
</tbody>
</table>

2.2 Melting and degassing
In this study A356 alloy ingots were charged and melted in a silicon carbide crucible using induction furnace. The alloy was melted and further heated up and maintained not to exceed 800 °C. The molten metal was
undergone flux treatment using a foundry-grade cleaning flux and dross was carefully removed before transferring from the furnace into a controlled crucible. Subsequently, the liquid alloy was subjected to further degassing. A series of degassing was conducted using 99.99% argon gas. Different amounts of dissolved hydrogen ranging between 0.27-0.92 cc H₂ / 100 g were achieved by feeding the argon gas at an appropriate flow rate via an immersed stainless steel lance into the controlled melt with various degassing times. These times started from zero minute, i.e. no degassing, in a one-minute stepwise to six minutes. Table 2 summarizes the degassing times applied to the controlled melt.

Table 2. A summary of degassing times applied for the casting trials.

<table>
<thead>
<tr>
<th>Degassing Time (minutes)</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dissolved Hydrogen (ml / 100 g Al)</td>
<td>0.92</td>
<td>0.79</td>
<td>0.62</td>
<td>0.48</td>
<td>0.33</td>
<td>0.3</td>
<td>0.27</td>
</tr>
</tbody>
</table>

It should be noted that the melt temperature was monitored and maintained at 720-750 °C throughout the degassing cycles. During the degassing, argon gas was also purged over the controlled melt in order to prevent undesirable oxidation. This was to ensure that other factors, particularly cleanliness or inclusion content, that might confuse the test results were controlled and minimized in the present study. Hence, relatively, effects of the dissolved hydrogen amount on thermal characteristics in terms of cooling curve can be reasonably investigated. Corresponding amounts of dissolved hydrogen measured by reduce-pressure test and quantitative density analysis are also presented in Table 2. Details of the hydrogen measurement are described in the next subsection.

2.3 Hydrogen measurement

The hydrogen measurement was performed using enhanced reduce-pressure test equipment as illustrated in Figure 1. The pressure level in the vacuum chamber of this equipment can be adjusted to suit solidification characteristics of the aluminum alloy. The equipment has a built-in thermocouple in order to properly monitor and control the liquid metal temperature prior to applying a reduced pressure level of concern [7]. The reduced-pressure test samples, whose weights were approximately 200 grams, were cast into a cup-shape mold made of stainless steel. The aluminum cast samples were further evaluated in order to assess hydrogen content employing using quantitative density analysis.

Calculation of hydrogen content in the cast samples was conducted in a newly developed Hydrogen/Porosity Analysis System based on Archimedes’ principle as depicted. The system comprises of a balance, with high accuracy level, i.e. 3-digit scale, a density-measurement kit, and a
computer program exclusively developed for quantitative reduced-pressure test analysis of cast aluminum. This computer program takes into account of important casting parameters such as pouring temperature and pressure level during solidification. The computer program features a calculation of hydrogen content in terms of porosity level in a solidified aluminum sample can be expected to thermodynamically yield improved result [7].

In addition to the quantitative density analysis, all reduced pressures test samples were also subject to visual inspection. The samples were sectioned vertically into two halves and subsequently hydrogen-induced porosities inspected.

2.4 Thermal analysis

Figure 2 shows schematic of thermal data acquisition. Once the liquid alloy in the controlled crucible was degassed at a pre-defined time duration, each representative melt was taken into a stainless steel cup. Cooling curve of each melt was plotted from its acquired thermal data. All cooling curves representing different amounts of dissolved hydrogen were analyzed in order to determine thermal characteristics during solidification of the melts featuring different hydrogen content. These included degree of undercooling at the beginning of primary aluminum nucleation, eutectic temperature and time. Finally, comparisons between dissolved hydrogen amounts and such thermal characteristics were investigated and discussed.

3. RESULTS AND DISCUSSION

Figures 3 is plot between dissolved hydrogen amount vs. undercooling. It was found that the degree of undercooling of primary aluminium nucleation increased as the dissolved hydrogen amount increased. The undercooling was 0.92 °C at the hydrogen level 0.27 ml / 100 g Al. And the undercooling was 1.58 °C at the hydrogen level 0.92 ml / 100 g Al. In fact, solubility of hydrogen decreases considerably as liquid aluminium undergoes solidification [4]. Such an increase in undercooling could therefore be affected by evolution of hydrogen gas taking place together with primary aluminium nucleation. Simultaneously, additional heat associated with more formation of hydrogen gas, as a result of higher dissolved hydrogen in the melt, could lead to greater degree of undercooling.
Figure 3  Plot between dissolved hydrogen amount vs. undercooling temperature.

Figure 4 shows another change of the thermal characteristics in terms of eutectic temperature. It was found that the eutectic temperature decreased as the dissolved hydrogen amount increased. Mechanisms underlying this effect of dissolved hydrogen on the eutectic temperature during solidification of aluminium-silicon alloy are yet to be investigated further in the next study.

Figure 4  Plot between dissolved hydrogen amount vs. eutectic temperature.

Figure 5 displays visual inspection of some reduce-pressure test specimens. Also noted herewith are dissolved hydrogen content and degree of undercooling. Obviously, as the dissolved hydrogen in the liquid alloy were higher, the solidified specimen became more porous.
4. CONCLUSIONS

Based on the series of casting trials carried out in the present study, it was found that dissolved hydrogen appeared to affect cooling curve or thermal characteristics of the hypoeutectic aluminium-silicon alloy. It was observed that the degree of undercooling at the beginning of the nucleation of primary aluminium phase became smaller as the amount of dissolved hydrogen reduced. This could be associated with evolution of hydrogen gas taking place simultaneously at the beginning of the nucleation. Further investigation will however be carried out in order to gain better insight concerning this thermodynamic phenomena. More detailed thermal analysis, such as eutectic temperature, eutectic time, will also be examined based on statistical concern. It is expected that useful empirical relationships will then be established.

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