Effects of Solid Fraction on the Heat Transfer Coefficient at the Casting/Mold Interface for Permanent Mold Casting of AZ91D Magnesium Alloy

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The interfacial heat transfer coefficient (HTC) between the steel mold and AZ91D magnesium (Mg) alloy that cast under various initial solid fractions were investigated in this study.

The interfacial HTC was determined by using an Inverse Method with measured temperature data and known thermo-physical properties. A Computer Aided Cooling Curve Analysis technique was used to determine the solid fraction versus temperature relationship. To comply with the requirements of the Inverse Method, a one-dimensional heat transfer system from the casting to the mold was designed for the permanent mold casting of AZ91D Mg alloy in molten and semisolid states.

Experiments were conducted with different initial solid fractions of 0, 30, and 50%. The results indicate that the HTC profile of molten AZ91D during solidification can be divided into five stages, while casting with semisolid AZ91D only into three. During each stage, the casting/mold interfacial conditions vary, which in turn causes the HTC values to vary. These data are critical for any solidification model of permanent mold casting and semisolid casting to obtain a reliable prediction of the thermal profile inside the solidifying casting and its freezing time.


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1. Introduction

Conventional casting methods are based on the pouring of a thoroughly molten metal into cavities of a die, while semisolid casting uses slurry-molten metal, which contains a certain volume fraction of solid phase. The most frequently reported benefits of semisolid forming process include energy savings, increased design flexibility of final product with low porosity, uniform microstructure, superior mechanical properties and surface finish. Since major benefits are primarily attributed to globular solid particles within the slurry, their influence is on preserving a laminar mold-filling front and causing less entrapped air and less porosity during solidification.

For conventional and semisolid permanent mold casting, due to a higher cooling rate and better surface finish as compared to sand casting, it needs better control over the heat transfer and solidification phenomena to improve the mechanical properties and soundness of the castings. If accurate temperature profiles for the whole casting period can be obtained, the solidification related defects can be alleviated and the desirable microstructure of solidified casting can be obtained through the modifications of the casting design and operation.

In recent years, the production of castings has been greatly assisted by the employment of mold filling and solidification models. The application of solidification models to simulate the thermal profiles inside the solidifying casting has also gained much popularity for analyzing casting processes. To accurately simulate solidification phenomena, these models require knowledge of the thermo-physical properties of the casting alloy and the mold as well as the heat transfer conditions between the casting and the mold. For permanent mold casting, the heat extraction from the casting is predominantly controlled by the casting/mold interface. During solidification in metallic molds, the interfacial gap formed by the interaction between the solidifying casting and the mold, creates a thermal resistance to heat flow and therefore, reduces the heat transfer and solidification rates. The heat transfer behavior of the interface is usually characterized by an interfacial heat transfer coefficient (HTC), \( h \). It is then critical for any solidification model of permanent mold casting to have an accurate data of \( h \) for obtaining a reliable prediction of the thermal profile inside the solidifying casting and its freezing time.

In the literatures, there are two methods to measure and calculate the interfacial HTC. The first method is to measure the size of the air gap (\( x \)). When the chemical composition and thermal conductivity (\( k \)) of the air/gas mixture is known, the interfacial HTC (\( h \)) between casting and mold can be simply estimated by \( h = k/x \). The major disadvantage of this method is that the accurate value of \( k \), which is the thermal conductivity of the air/gas mixture, is difficult to obtain. The second method for obtaining the interfacial HTC is to use the Inverse Method, which was developed by J. V. Beck. For this method, the temperature distribution and variation during the casting have to be measured. With the other thermo-physical properties known, the interfacial HTC can be derived by via using the Inverse Method. However, the Inverse Method can only be used for one-dimensional heat transfer systems. To comply with the requirements of the Inverse Method, a one-dimensional casting to mold heat transfer system needs to be employed.

In our previous study, the interfacial HTC, \( h \), at the interface between A356 aluminum alloy casting and metallic mold with various coatings were measured. It was found from the measurements that for all cases the variation of \( h \) versus temperature (\( h-T \)) curve can all be categorized into five...
stages. And the characteristics of the five stages and the effects of coating type and thickness as well as mold type on the variation of $h$ were discussed. Today, the heat transfer conditions between the casting and the mold for semisolid casting process are difficult to find. The comparison and detailed description of the heat transfer condition at casting/mold interface between solidification from liquids state and semisolid state are also rarely seem.

The purpose of this study is to employ the Inverse Method to obtain the variation of interfacial HTC between fully melted and semisolid AZ91D magnesium alloy and SKD-61 mold during solidification. The present work also developed an experimental setup and procedure to achieve the well-controlled unidirectional solidification condition and less turbulence of molten metal generated during pouring. Inert gas was used to prevent the magnesium (Mg) alloy from oxidation during the heating process in this system. To make the $h$ data useful for mathematical modeling of the solidification phenomena in casting, a procedure was also developed to obtain the relation between the $h$ value and the casting temperature at the interface. The effects of initial solid fraction setups of AZ91D Mg alloy on the variation of $h$ were discussed in this study.

2. Methodology

2.1 Principles of the estimation of interfacial heat transfer coefficient with Inverse Method

The structure of Inverse Method adopted in this study was proposed by Beck. For the analysis of a heat transfer problem, an appropriate set of equations is first determined to describe the heat transfer behavior. With the boundary conditions, initial condition, and thermo-physical properties of the material known, it is possible to obtain the temperature distribution and variation of the whole system. However, if one of the thermo-physical properties of the material is not known, but the temperature information can be obtained, it is possible to calculate this unknown property by a reverse scheme, which is the basic idea of the Inverse Method.

To make use of the Inverse Method to determine $h$, temperature information has to be known. Therefore, temperature measurements are required. To make the analysis of the Inverse Method more manageable, a unidirectional heat transfer system is devised. It requires temperature measurements at least at three points. Each way as the boundary conditions of the heat transfer system and the third one as the calibration point. A detailed description of the Inverse Method to manipulate the temperature data for $h$ was shown in one of our previous studies. The algorithm and the corresponding computer program were also constructed in that study.

2.2 Principle and procedure of Computer Aided Cooling Curve Analysis (CA-CCA)

There are at least two methods to mathematically derive the quantities from the cooling curve for creating a latent heat release model as well as solid fraction ($fs$) versus temperature relationship ($fs$-$T$). One technique was developed by Argyropoulos and et al. and the other was developed by Stefanescu and et al. The principles and procedures of Stefanescu’s method were used in this study and the detail equations can be found in the reference. The assumptions of CA-CCA are: (1) Cooling behavior of the molten metal in a small crucible can be considered to be a lumped system; the Biot number ($Bi = hV/Ak$) is smaller than 0.1. (2) During cooling, the density and specific heat of the molten metal remain constant and do not vary with respect to temperature. (3) The predominant mechanism for extracting heat from the system is through convective heat transfer between the crucible and the surrounding atmosphere. Conduction is not significant and can be neglected.

A detailed description of the CA-CCA Method to manipulate the measured temperature data for obtaining the $fs$-$T$ relationship was shown in one of our previous studies. The algorithm and the corresponding computer program were also constructed in that study.

2.3 Treatment of latent heat releases of AZ91D magnesium alloy

The solidification heat transfer system considered in the analysis includes one important term which is the latent heat of the casting alloy. The latent heat ($L$) is released when the temperature of the casting metal falls in the solidification range. To extract this additional heat, the specific heat, $Cp$ is replaced by an effective specific heat, $Cp^*$, in the following manner.

$$Cp^* = Cp - \frac{\partial fs}{\partial t} L$$  \hspace{1cm} (1)

and

$$\frac{\partial fs}{\partial t} = \frac{\partial fs}{\partial T} \cdot \frac{\partial T}{\partial t}$$  \hspace{1cm} (2)

The relation of $fs$ as a function of temperature in the solidification range must be determined first and then the solidification analysis can be conducted. In this study, the $fs$ as a function of temperature was obtained from CA-CCA method described in the previous section.

3. Experimental Method

3.1 Experimental setups and procedures of CA-CCA method

In this study, the AZ91D Mg alloy was investigated. The compositions of the alloys were analyzed and shown in Table 1. The experimental apparatus used in this study included a crucible, two K-type thermocouples, a data logger, and a heating furnace with full of saturated Argon. The crucible was made out of 0.5 mm thick steel plate, with inner diameter of 30 mm and height of 40 mm.

The experimental procedures were to first put the AZ91D alloy into the crucible. And the crucible was placed in the furnace with Argon atmosphere and heated the alloy to 615°C. The crucible was then immediately pulled up by steel

<table>
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<tr>
<th>C</th>
<th>Mn</th>
<th>Si</th>
<th>Cr</th>
<th>Mo</th>
<th>V</th>
<th>Al</th>
<th>Zn</th>
<th>Mg</th>
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<tbody>
<tr>
<td>AZ91D</td>
<td>0.25</td>
<td>0.013</td>
<td>—</td>
<td>—</td>
<td>8.31</td>
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wires not to contact with the wall of furnace. And then, shutting down the heating plates and increasing of Argon flow rate were conducted to cool down the alloy. Two thermocouples were inserted into the crucible; one at the center position and the other near the wall. The illustration of the experimental setup and procedures for CA-CCA method are shown in Fig. 1. The cooling curve was recorded by the data logger at a rate of 1 Hz. After solidification, the collected cooling curve data were then processed in various ways using a spreadsheet. They included different smoothing and curve fitting techniques. The average at every five points for smoothing was proved to be almost identical to five-point central smoothing. The calculations were performed with the temperature data from the central. Finally, the CA-CCA method was employed to calculate the relationship between solid fraction and temperature.

3.2 Experimental setups and procedures of $h$ measurement

To meet the requirements of the Inverse Method, a one-dimensional heat transfer from the casting to the mold needs to be devised. Figure 2 is a schematic illustration of the unidirectional heat transfer measure system used in this study and Fig. 3 is its photograph. The main part of this system is a SKD-61 steel cylindrical container. Argon was introduced into the cylindrical container from the top to protect the Mg alloy from oxidizing during the heating process. A pre-produced AZ91D Mg alloy ingot (with non-dendritic structure) was put into the cylinder container. The mold material SKD-61 was on the bottom of the cylindrical container and in contact with the AZ91D ingot, on top of a copper plate water cooling system. The cylindrical container was covered with heat insulating material and a heating wire heater. Five K-type thermocouples were inserted into this system. Thermocouples #1, #2, #4, and #5 was inserted into the casting and #3 into the SKD-61. The data read from thermocouples #1, #2, and #3 was as input for the Inverse Program to calculate the heat transfer coefficient between the casting and the mold. Thermocouple #4 and #5 were used to ensure that the system was a one-dimensional heat transfer system.

In order to understand the $h$ variations with different initial solid fractions, the following procedural steps were carried out:

1. The AZ91D Mg alloy with non-dendritic microstructure was put into the cylindrical container.
2. The heater heated the casting to the desired temperature (the temperature was used to control the initial solid fraction of the casting).
3. The heaters were turned off.
4. The cooling system was activated and the temperatures were recorded (at a scanning rate of 5 Hz) until the casting had solidified to a certain degree.
5. The temperature readings were used as input for the Inverse Program to calculate the values of $h$. 

Fig. 1 Schematic illustration of the experimental measurement for CA-CCA method.

Fig. 2 Schematic illustration of the unidirectional heat transfer system.

Fig. 3 Photograph of the unidirectional heat transfer system.
4. Results and Discussions

4.1 Measurement of the $f_s$-$T$ relationship of AZ91D alloy by CA-CCA method

The cooling curve of AZ91D alloy during solidification is shown in Fig. 4. It shows that the temperature at which the AZ91D alloy starts to change state from liquid to solid is nearly 583°C and around 409°C the casting is completely solidified. Figure 5 shows the $dT/dt$ curve and null curve for AZ91D alloy. The thick line is the NLSF optimized null curve obtained from CA-CCA theory. By substituting the cooling rate curve and null curve data into the CA-CCA program, the $f_s$-$T$ relationship of AZ91D alloy was obtained, as shown in Fig. 6. It can be easily obtained from the figure that the alloy start to solidify (solid phase start to form) is located at nearly 583°C, the temperature that the alloy completely solidified is located at nearly 409°C, and temperatures in the mushy zone with solid contents of 30, 40 and 50% are located at nearly 569.7, 563.7 and 556.5°C, respectively. The data is useful for the setting of initial temperature for $h$ measurements under different semisolid states. And the $f_s$-$T$ curve was put into the inverse program as the database for the latent heat release module. In this study, we treated that the rate of latent heat release was proportional to the increasing rate of solid fraction as described in section 2.3.

4.2 Data from temperature measurements for calculating $h$

Experiments were conducted for the casting of AZ91D Mg alloy under molten and semisolid states employing the experimental setup and procedures described in the previous section. The density of AZ91D Mg alloy and SKD-61 were treated as constant values of 1810 and 7760 kg/m$^3$. The thermal conductivity and the specific heat of the casting and the mold used in this study are listed in Figs. 7 and 8. Figure 9 shows a temperature-time curve from an experiment when the casting was heated up to a temperature above liquidus and then cooled down. It should be noted that thermocouples #2, #4 and #5 were aligned in a straight line parallel to the casting/mold interface. This temperature data was used to check if the unidirectional heat transfer condition was maintained throughout the experiment. It can be seen from Fig. 9 that the temperature variation of the thermocouple #4 coincides with that of the thermocouples #2 and #5. This was also the case in the other experiments. It is therefore
clear that the design of the apparatus for this study fulfilled the requirement of unidirectional solidification heat transfer. Experiments were also conducted for the semisolid casting of AZ91D Mg alloys with two sets of initial solid fractions, which include solid contents of 30% solid and 50%, and SKD-61 mold material. A pre-produced AZ91D Mg alloy ingot with non-dendritic structure was used as the raw materials in order to express the real semisolid casting process. The thermal histories with five thermocouples as shown in Figs. 10, and 11 and the cooling curves show similar tendency under different initial solid fraction setups. The temperature profiles of semisolid casting show unapparent phase transformation feature in the mushy zone as compared with casting with molten alloy.

### 4.3 Interfacial heat transfer coefficients for casting with molten alloy

The Inverse program was then used to calculate the $h$ values from the temperature data. From that calculation, the relationship between $h$ values and time; the so called $h$-$t$ curves could be derived. However, it should be noted that the $h$ data can be useful for solidification modeling only when the relation of its value with the casting temperature near the interface; the so called $h$-$T$ curve, is known. To comply with this need, the $h$-$t$ curves were then transformed to $h$-$T$ curves with the temperature-time relations already calculated during the inverse program analyses. The relationships between the interfacial heat transfer coefficient and temperature at the casting surface as well as the fitted $h$-$T$ curve was obtained as shown in Fig. 12. And the relationships between $h$, temperature and the related solid fraction at the interface and inside of casting for the casting of AZ91D Mg alloy in fully melted condition is shown in Fig. 13.

From the figure, it can be found that the $h$-$T$ curve can be divided into five stages. As the cooling water turns on, a thin solidified layer and a gap suddenly are appeared and therefore result in a sharp decrease of $h$ to a value of 1681 W m$^{-2}$K$^{-1}$ at casting surface temperature around 588°C in the first stage. And then, it takes a little while for the inside molten metal to bring pressure to the solidified thin layer to get in good contact with the mold wall again. During this period, the $h$ value starts from 1681 W m$^{-2}$K$^{-1}$ at casting surface temperature around 588°C and then increases to a peak value of around 2931 W m$^{-2}$K$^{-1}$ at a casting surface temperature around 562°C. Comparing with the solid fraction conditions...
of casting at the interface, it can be observed that $fs$ increases quickly at this stage until a solid fraction of 30% and the inner part of casting is still in liquid state.

While the value of $h$ reaches its peak, massive heat is extracted from the casting to the mold. During the third stage, solid skin starts to form and air gap is gradually generated between the casting metal and the mold. As the gap is formed, it presents a higher resistance to the heat transfer between the casting metal and the mold. As the gap is solidified, the un-melted metal inside casting keeps pushing the solid skin towards the mold. Therefore, the value of $h$ is seen to drop to 2216 W m$^{-2}$ K$^{-1}$ rather quickly until the casting surface temperature reaches around 533°C. It can be seen from the figure that the solid fraction at the casting surface still increases quickly to a value of 60% but slows down its tendency and the inner part of casting starts to solidify.

Following the gap formation, the hydrostatic pressure of the un-melted metal inside casting keeps pushing the solid shell towards the mold. As a consequence, the value of $h$ increases slightly between 2216−2577 W m$^{-2}$ K$^{-1}$ at a casting surface temperature cools from 533 to 430°C. This constitutes the fourth stage. In this stage, the increasing of solid fraction at the casting surface slows down and the solidification speed of casting inside increases quickly. The difference of solid fraction between casting surface and inside is getting smaller. It is also the evidence that heat coming from casting inside offers steady heat source and therefore inhibits the solidification speed at interface in this stage.

As more molten metal solidifies, solid shell grows with gradually precipitated Mg$_2$Al$_{12}$ phase and becomes stronger enough to resist the hydrostatic pressure coming from the un-melted metal. At this stage: the last stage, shrinkage of casting metal due to solidification and cooling gradually enlarge the gap. Naturally, the value of $h$ decreases accordingly.

Here, the $h$-$T$ curve characteristic is similar to that in previous researches on A356 aluminum alloy$^{8}$ and Inconel-600, SKD-61, SKT-4 alloy. During the solidification process, all alloys present similar tendency of $h$-$T$ curves. It means the mechanism of interface situation between the casting and the mold is similar.

### 4.4 Interfacial heat transfer coefficients for casting under semisolid state

According to the results of CA-CCA experiment, the ingot with globular grain microstructure was heated up to 569°C in order to produce semisolid casting with 30% solid fraction at the interface. A shown in Fig. 14, $h$ variations can be divided into three stages. In the beginning, $h$ drops sharply as the molten metal solidifies, and then the decreasing tendency slows down and reaches a value around 1400 W m$^{-2}$ K$^{-1}$. After the temperature of casting surface decreases to 409°C, the $h$ starts to descend smoothly.

From the figure, it also can be seen that the smoother curve is obtained compared with Fig. 12 and it means better numerical stability for calculating of $h$-$T$ curve. It would be the reason that the network of solidified part between the globular grains at the interface or inside the casting can be a restriction to prohibit the movement of un-solidified phase and also the interface. Especially as the casting surface becomes stronger after contacting with the cooled mold in the beginning stage. It also can be explained that the variation of gap formation during semi-solid casting is more uniform than casting with fully melted alloy.

According to the result of CA-CCA experiment, the ingot with globular grain microstructure was heated up to 556°C in order to produce semi-solid casting with 50% solid fraction at the interface. The $h$-$T$ curve showed in Fig. 15 is similar with that casting with 30% solid fraction. At the beginning,
The decreasing of \( h \) slowed down and became stable or even increased slightly at a casting surface temperature range around 533 to 430°C. The \( h \) data for cast with fully melted one had higher value than cast under semisolid state. This indicates that casting with semisolid state has a solid network microstructure in the beginning then can solidify uniformly. This solid network inside the casting and at the casting surface is strong enough to keep a stable condition at the interface and not to further reduce the air gap. It is also interesting that both \( h-T \) curves that cast under semisolid states decrease to a certain value (1400 W m\(^{-2}\) K\(^{-1}\)) at a casting surface temperature around 430°C.

Finally, as the casting kept solidifying, decreasing tendency of \( h \) values for casting with fully melted one was quicker than the others. It was due to the reason that obvious directional solidification procedures from the casting/wall interface toward inside kept steady shrinkage and therefore enlarged the gap. But for casting with semisolid state ones, due to the lower casting temperature and the network of solidified phase will let the casting solidify uniformly and cause fewer shrink percentage than casting with fully melted alloy. After 430°C the \( h-T \) curves for the semisolid ones started to separate again and it might due to the reason that higher solid fraction means less volumetric shrink during solidification. The air gap was smaller hence the \( h \) value was larger.

5. Conclusion

In this study, a one-dimensional heat transfer system from the casting to the mold needs has been designed and fabricated for the permanent mold casting of AZ91D Mg alloy in molten and semisolid states. Three different initial solid fractions of 0, 30, and 50% for casting were conducted. Temperature measurements were then analyzed with the Inverse Method to estimate for the interfacial HTC, \( h \). Then the variation of \( h \) with respect casting surface temperature (\( h-T \) curve) was obtained. The characteristics of the \( h-T \) curves and the effects of initial solid fraction on the variation of \( h \) can be summarized as follows.

1. The \( h-T \) curve obtained from casting with fully melted alloy had different values and trend than the others that casting under semisolid states. It is critical for any solidification model of permanent mold casting as well as semisolid casting for obtaining a reliable prediction of the thermal profile inside the solidifying casting and its freezing time.

2. The \( h-T \) curve obtained from casting with fully melted alloy could be distinguished in five stages. During each stage, the casting/mold interfacial conditions varied, which in turn caused the \( h \) values to vary. The \( h-T \) curve characteristic was similar to that in previous works on A356 aluminum alloy and Inconel-600, SKD-61, SKT-4 alloy.\(^{13}\)
The $h$-$T$ curves obtained from casting with semisolid alloy could only be distinguished into three stages. It lacked of the first and the second stage of $h$-$T$ variation obtained from casting with fully melted alloy. It is because that their intrinsic solidified network inside the casting and at the casting surface in the beginning and during solidification will let the casting solidify uniformly and will prevent the hydraulic pressure coming from the un-solidified phase.

Semisolid casting with 30% solid fraction shows higher $h$ in the beginning stage. And then the values converge toward similar value in the second stage until 430°C. After the temperature, the $h$-$T$ curve started to separate again and it might due to the reason that higher solid fraction means less volumetric shrink during solidification.

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