DIRECT OBSERVATION AT EARLY STAGE OF SOLIDIFICATION IN CONTINUOUS CASTING WITH COPPER ALLOY AND MOLD POWDER

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Abstract
The formation of oscillation marks and the molten slag inflow from the meniscus into the flux channel between solidified shell and mold walls were directly observed through a transparent heat-resisting glass in a mold of laboratory scale caster by using molten copper and mold powder. Oscillation marks were formed by the relative movement between the solidified shell and the mold wall at meniscus. The solidified shell at meniscus was deformed towards liquid copper during the period when the mold moves downwards with a velocity larger than the casting speed (the negative strip time).
The molten slag inflow from the meniscus into the flux channel was clearly visualized and was observed mainly during the period of the negative strip time. The powder consumption with the model considering the timing of the molten slag inflow agreed well with the plant data.

INTRODUCTION
Tasks for the continuous casting of slabs include prevention of constrictive breakout during high speed casting and improvement of the surface quality of cast slabs. These tasks are closely related to the phenomenon of initial stage solidification in the mold in the vicinity of the meniscus. Therefore, in addition to optimizing the casting conditions and the characteristics of the mold powder, it is also desirable to clarify the process of initial stage solidification in continuous casting.
In the past, a considerable number of research projects have been carried out in connection with the mechanism of oscillation mark formation and the mechanism of powder inflow between the mold and cast strand with the aim of elucidating the phenomenon of initial stage solidification.[1] – [9] Recently, experiments related to the direct observation of initial stage solidification in continuous casting have also been conducted. [10], [11] Tada et al. [11] succeeded in direct observation of the formation of oscillation marks and shape of the meniscus using a
Pb-Bi alloy (melting point: 401K) as the casting metal and stearic acid (melting point: 344K) as the lubricating agent. However, the powder used in the continuous casting of steel, because it is an oxide, assumes a complex morphology comprising an unmelted layer and a molten layer, whereas stearic acid is completely molten. Therefore, doubts remain as to whether stearic acid can be used to simulate the phenomenon of initial stage solidification in continuous casting with actual equipment.

From this viewpoint, the goal of the present research was to reproduce the melting behavior of the powder and the behavior of the solidified shell in the continuous casting of molten steel, which was achieved by using molten copper and an oxide powder. The results were compared with the conventional knowledge, and the mechanism of oscillation mark formation and mechanism of powder inflow were investigated.

**EXPERIMENTAL PROCEDURE**

**Selection of Experimental System for Visualization**

Figure 1 shows the shape of the meniscus of the molten metals in Table . calculated from Bikerman's equation, \(^1\) which is given in equations (1) and (2), using the values of the physical properties of the molten metals and powders shown in the table.

\[
\begin{align*}
X &= (2a^2 - Y^2)^{0.5} + a(2^{0.5} \ln \{2^{0.5}a + (2a^2 - Y^2)^{0.5}\} - 0.3687a) \\
a &= \left\{ \frac{2}{\sqrt{S - L}} \right\}^{0.5} \cdot g
\end{align*}
\]

X: Distance from meniscus (m)
Y: Distance from wall of mold (m)
\(\sigma\): Surface tension of molten metal (N/m)
S: Density of molten metal (kg/m\(^3\))
L: Density of molten powder (kg/m\(^3\))
g: Acceleration due to gravity (m/s\(^2\))

It can be understood that the shape of the meniscus differs greatly when a low melting point metal such as Sn is used. Because the surface tension and density of a molten copper-oxide powder system are similar to those of a molten iron-oxide powder system, the shape of the meniscus is also close to that of a molten iron-oxide powder system. Therefore, a molten copper-oxide powder system was used in these experiments in order to visualize the meniscus at the surface of the molten metal in the continuous casting mold.

**Experimental Apparatus and Experimental Conditions**

Experiments were conducted using a vertical type test continuous caster with a 70 mm square cross section. The experimental apparatus is shown schematically in Figure 2. The mold is composed of an upper mold and a lower mold. In the upper mold, three sides are constructed of water-cooled copper plates, and the fourth side (observation side) is of heat-resisting glass. The maximum heat-proof temperature of the (heat-resisting) glass (96% SiO\(_2\), glass) is 1473K.
A copper alloy was used as the casting metal. The physical properties of the copper alloy and the powder used are shown in Table 1. The casting metal was melted in a graphite crucible using an electric resistance furnace and held at 1573K. The tundish was preheated with a burner until the surface temperature of the refractories become uniformly 1523K. The inner surface of the heat-resisting glass was also preheated to 873K with a dichromic wire heater to prevent the solidified shell from sticking during casting. Pouring of the molten metal began immediately after preheating of the tundish was completed. Withdrawal of the strand was begun when the molten copper had been teemed to a point 50 mm below the meniscus observation position, and mold oscillation began at the same time. After confirming that the immersion nozzle was immersed in the molten copper, powder was charged continuously at an addition rate of 0.118 kg/min. The level of the molten metal surface was controlled so as to be uniform by fine adjustment of the casting speed, which was performed manually.

Table 1 shows the casting conditions. Mold oscillation was performed using a sine wave form or a non-sine wave form. Condition A is a sine wave form and condition B a non-sine one, in which the downward velocity of the mold is large compared with the upward velocity. Condition C is also a non-sine wave form, but has the inverse condition to B, the upward velocity of the mold being large compared with the downward velocity.

In direct observation of the meniscus, after continuously photographing the area of the upper mold indicated by the box in Figure 2 with a CCD camera, the level of the molten metal surface, the shape of the meniscus, and the condition of mold powder inflow were observed from the recorded images at intervals of 0.03 second.

In addition, the pitch and depth of the oscillation marks and the solidification structure were investigated from the cast metal.

**EXPERIMENTAL RESULTS**

**Results of Direct Observation**

**Formation of Oscillation Marks**

Stable casting was possible for approximately 60 seconds, during which the withdrawal behavior of the solidified shell at the meniscus, the process of oscillation mark formation, and the inflow behavior of the powder between the mold and the cast strand could be observed.

An example of a photograph of the meniscus during casting is shown in Figure 4. Accumulation of the mold powder on the molten metal was observed. In contrast to the black color of the unmelted layer, the melted layer appears as white to gray, making it possible to distinguish the state of melting of the mold powder. Moreover, depending on how the light struck the material, the solidified shell could be observed to shine only at the leading end, which made it possible to designate the position of the end. It might also be mentioned that sound growth of the lower solidified shell was confirmed by stopping withdrawal during
casting and observing the strand.

A schematic diagram of the area around an oscillation mark is shown in Figure 5, and a schematic diagram showing the relationship among casting displacement, x, the velocity of mold oscillation, V_M (mm/sec), the casting speed, V_C (mm/sec), and the shape of the solidified shell is presented in Figure 6. It was confirmed that the formation of oscillation marks occurs one time in one cycle. Collapse of the solidified shell toward the molten copper side occurred during the negative strip period, when the relationship between the velocity of mold oscillation, V_M, and the casting speed, V_C (upward direction: positive) is (V_M - V_C) > 0, as shown in Figure 6, stages c-d. During the positive strip period, when (V_M - V_C) < 0, it was observed that the solidified shell was pulled back to the mold side. A bending point, corresponding to the valley (D in Figure 5) in the oscillation mark, was formed when (V_M - V_C) becomes positive (Figure 6, stages d-a). The final point of the depression in the oscillation mark (A in Figure 5) was formed by the mold and the solidified shell coming together again at the position of maximum upward velocity of the mold (Figure 6, stage a). From the results of observation described above, it was found that the formation of oscillation marks begins during the negative strip period and is completed in the time until one-half of the positive strip period, when the upward velocity of the mold reaches its maximum.

It might also be noted that the length of the flat part between oscillation marks (C in Figure 5) was virtually the same as the casting distance during one-half of the positive strip time, t_p (sec).

**Condition of Powder Inflow**

Acceleration of the inflow of powder between the mold and the cast strand in synchronization with the period of oscillation mark formation was observed.

The existence of slag rim was not clearly observed during these experiments. The slag rim, however, exists during casting judging from the fact that, in direct observation, the mold powder at the expected slag rim position became black, and that a firmly adhering layer of powder approximately 1 mm thick was found on the mold walls after casting.

**Results of Investigation of Cast Metals**

Figure 7 shows the relationship between the pitch of the oscillation marks on the cast metal and the ratio, V_C/f, of the casting speed, V_C, to mold oscillation frequency, f. The pitch of the oscillation marks shows linear relation with V_C/f. These results were also in agreement with previous studies. [4],[11]

The relationship between the negative strip time and the depth of oscillation marks is shown in Figure 8. The depth of oscillation marks has a tendency to be proportional to negative strip time, and the coefficient (value) of the relation is closer to that of steel casting than to low melting point metal casting. [11],[13]
A form resembling a break ring could be observed on the cast slab in the part corresponding to the glass surface. To explain this, in contrast to the other three surfaces, the existence of this form corresponds to the fact that solidification at the center of the glass surface begins in the lower mold. Judging from the position, the thickness of the solidified shell at the lower edge of the upper mold was considered to be approximately 8 mm.

**DISCUSSION**

**Mechanism of Formation of Oscillation Marks**

Considering the relationship between the mold oscillation period and the formation of oscillation marks in these experiments, as shown in Figure 6, collapse of the solidified shell occurred during the negative strip period, and pulling back of the shell toward the mold wall was substantially completed at the point of maximum upward velocity of the mold. The fact that collapse of the solidified shell occurs during the negative strip period agrees with the result calculated by Takeuchi et al. [4] using a mathematical model of the pressure in the powder between the mold and the strand. Therefore, it can be said that the formation of oscillation marks is caused by changes in the pressure of the molten powder at the meniscus, which in turn are caused by changes in the relative velocity of the mold and solidified shell.

To summarize the observation results, the mechanism by which oscillation marks are formed can be explained as follows, referring to Figure 6.

Stage c: The downward velocity of the mold becomes larger than the casting speed, and as a result, molten powder is dragged into vicinity of the mold and flows downward in the slit. At this time, the solidified shell is subject to the pressure toward the molten metal, and is pushed and bent toward the molten metal side.

Stage d: The relative velocity of the mold and the solidified shell becomes zero, and collapse of the solidified shell toward the molten metal is completed. At this point, the amount of collapse reaches its maximum. Powder inflow is virtually complete.

Stage e: At the point when the relative velocity, \(V_M - V_C\), of the upward velocity of the mold, \(V_M\), and the velocity of the solidified shell, \(V_C\) (downward direction: positive) becomes positive, a bending point is generated in the solidified shell. Because of the presence of powder which had flowed into the slit during stages c and d, bending occurs in the upper shell, where the strength is weak.

Stage a: The upward velocity of the mold is at its maximum, and the slit between the mold and the solidified shell reaches its minimum width. The solidified shell is formed, corresponding to the end point of the depression of the oscillation mark. Because the amount of inflow into the slit is small, the molten powder in the vicinity of the mold shows its greatest amount of rise.

Stage b: The slit between the mold and the solidified shell remains unchanged at its minimum width, and the molten powder becomes to a virtually static condition.
Mechanism of Powder Inflow

Two theories have been proposed regarding the period when powder flows into the slit between the mold and the strand.

In the first theory, powder inflow occurs during the negative strip period. This explanation is represented mainly by the theory proposed by Nakato et al.\textsuperscript{[7]} that inflow occurs due to a "pumping" mechanism, in which the powder is pushed by the slag rim which has formed at the meniscus and as a result, flows into the flow channel, and the theoretical analysis by Anzai et al.,\textsuperscript{[9]} who solved a 2-dimensional Navier-Stokes equation by numerical calculation. In the analysis by Anzai, et al., it was reported that the pressure and the amount of molten powder inflow change in a sine wave form during one cycle of mold oscillation, with pressure and powder inflow reaching their maximum when the downward velocity of the mold is largest and showing their minimum when the upward velocity is largest.

In the second theory, powder flows in predominantly during the positive strip period.\textsuperscript{[8]} During the negative strip period, powder inflow is difficult because the molten powder flow channel is shut off by semi-molten powder and the solidified shell, and the flow channel is formed during the positive strip period. The results of measurements which show that powder consumption is proportional to positive strip time support this view.

Up to the present, it has not been possible to conclude which of these theories is correct.

In the present work, powder inflow behavior corresponds to the period of oscillation mark formation, and the larger part of inflow occurs during the negative strip period. This result is in good agreement with the mechanism of powder inflow proposed by Nakato et al.\textsuperscript{[7]} and the results of the theoretical analysis by Anzai et al.\textsuperscript{[9]} It is therefore considered that the inflow of powder is related to changes in pressure in the slit.

Influence of Oscillation Wave Form

In order to investigate the influence of the mold oscillation wave form on the formation of oscillation marks and powder inflow, a comparative study was carried out using the distance, $X_S$ (mm) between the mold wall and the leading edge of the solidified shell obtained from the results of observation. Figure 9 shows the relationship between the amount of collapse of the solidified shell and the mold oscillation velocity per oscillation cycle under various oscillation conditions. From this investigation, it became clear that collapse of the solidified shell with both the sine wave form (condition A) and the non-sine wave forms (conditions B and C) began for the most part during the negative strip period, and the end point of shell pull-back toward the mold wall corresponded to one-half of the positive strip period, when the upward velocity of the mold reached its maximum.

Model of Powder Inflow

A schematic diagram of the shape of the meniscus is shown in Figure 10. In this model,
powder inflow was considered to be composed of the following two elements.

(A) Element which flows intermittently into a valley part during formation of the oscillation mark

(B) Element which flows continuously into the molten flux channel between the mold and the cast strand

From this, the amount of powder consumption, $Q_M$, can be represented by the sum of consumption due to (A), $Q_A$, and consumption due to (B), $Q_B$.

$$Q_M = Q_A + Q_B \quad (kg/m^2)$$  \hspace{1cm} (3)

$Q_A$ can be determined from the shape of the oscillation mark. If the mark is regarded as having the shape shown in Figure 10, then the terms (A) and (B) can be represented by equations (4) and (5), respectively.

$$Q_A = \frac{(L \cdot d_{os} \cdot d_X \cdot f)}{2 \cdot V_C} \quad (kg/m^2)$$  \hspace{1cm} (4)

$$Q_B = L \cdot d_f \quad (kg/m^2)$$  \hspace{1cm} (5)

$d_{os}$: Depth of oscillation mark (m)

$d_X$: Length of oscillation mark (m)

$f$: Frequency of mold oscillation (1/sec)

$V_C$: Casting speed (m/sec)

$L$: Density of molten flux (kg/m$^3$)

$d_f$: Thickness of molten flux layer in slit (m)

As shown in Figure 8, the depth of oscillation marks, $d_{os}$, is considered to be proportional to the negative strip time, $t_N$ (sec), and is expressed by equation (6)

$$d_{os} = K \cdot t_N \quad (m)$$  \hspace{1cm} (6)

In contrast to this result, $K$ was regarded as equal to 0.002 from Figure 8.

If the length of an oscillation mark, $d_X$, is considered to correspond to the amount of the solidified shell which is withdrawn during the powder inflow period, the length can be expressed by equation (7) using the powder inflow time, $t_N + t_p/2$, which was obtained by direct observation.

$$d_X = (t_N + t_p/2) \cdot V_C \quad (m)$$  \hspace{1cm} (7)

From the results of direct observation, $d_f$ was of the following order:

$$d_f = 0.0001 \quad (m)$$

Although the thickness of the molten flux layer, $d_f$, is regarded as varying depending on the powder used and the steel type, for convenience, it is considered as constant in the present work.

From the above, the amount of powder inflow, $Q_M$, can be expressed by equation (8).

$$Q_M = L \cdot K \cdot t_N \cdot (f \cdot t_N + 1) + L \cdot d_f \quad (kg/m^2)$$  \hspace{1cm} (8)

Figure 11 shows the relationship between the measured value of powder consumption in an actual machine, $Q_{M, \text{meas.}}$, and the values calculated with the equation (8), $Q_{M, \text{calc.}}$. Because the
inferred equation for powder consumption, which was derived based on the powder inflow behavior observed in the visualization experiments, is substantially in agreement with the powder consumption in the actual caster, it is also possible to explain the condition of powder inflow in the continuous casting of steel with this model.

CONCLUSION

In order to clarify the mechanism of oscillation mark formation in the initial stage of solidification during continuous casting and the mechanism of powder inflow, the authors succeeded in direct observation of the meniscus in a 70 mm square test continuous caster using a copper alloy-oxide powder system. The following results were obtained.

(1) Oscillation marks are formed by a process in which collapse of the solidified shell begins during the negative strip period, and pull-back toward the mold wall occurs until one-half of the positive strip period, when the upward velocity of the mold reaches its maximum.

(2) Powder inflow occurs in synchronization with the oscillation mark formation, and accelerates from the start of the negative strip period until one-half of the positive strip period, when the upward velocity of the mold reaches its maximum.

(3) A model for powder inflow was proposed based on direct observation. The calculated results of the amount of powder inflow by this model and the measured values from an actual continuous caster showed good agreement.

(Symbols)

- $d_{os}$: Depth of oscillation mark [m], [mm]
- $d_X$: Length of oscillation mark (casting direction) [m], [mm]
- $d_f$: Thickness of molten layer in flux channel [m], [mm]
- $f$: Mold oscillation frequency: [cpm] ([min$^{-1}$]), [sec$^{-1}$]
- $g$: Acceleration due to gravity (= 9.8 [m/s$^2$])
- $K$: Constant
- $Q_{M}$, $Q_{M, \text{calc.}}$, $Q_{M, \text{meas.}}$, $Q_A$, $Q_B$: Powder consumption [kg/m$^2$]
- $S$: Mold oscillation stroke [mm]
- $t_N$: Negative strip time [sec]
- $t_P$: Positive strip time [sec]
- $T_{m,L}$: Melting point of mold powder [K]
- $T_{m,S}$: Melting point of metal [K]
- $V_C$: Casting speed [m/min], [m/sec], [mm/sec]
- $V_M$: Mold oscillation velocity [mm/sec]
- $X$: Mold displacement [m]
- $X_s$: The amount of collapse of the solidified shell [mm]
- $\gamma$: Wave form distortion factor
.L: Density of fluid (flux) [kg/m³]
.s: Density of molten metal [kg/m³]
.: Surface tension of molten metal [N/m]

REFERENCES

Figure 1: Calculated results of meniscus shape.
Figure 2: Schematic view of experimental apparatus.

Figure 3: Schematic view of mold for visualization.

Figure 4: Example of meniscus periphery visualized by direct observation.

Figure 5: Schematic diagram of oscillation mark.
Figure 6: Relationship between mold movement and meniscus shape (A sine wave form).

Figure 7: Relation between ratio of casting speed to frequency, $V_C/f$, and pitch of oscillation marks.

Figure 8: Relationship between negative strip time and depth of oscillation marks.
Figure 9: Relationship between oscillation conditions and collapse of solidified shell (A: sine wave form, B: non-sine wave form, C: inverse sine wave form).

Figure 10: Schematic diagram of powder inflow model.

Figure 11: Relationship between calculated values of powder consumption and measured values in actual machine.

Table 1: Physical properties of metals and molten fluxes.

<table>
<thead>
<tr>
<th></th>
<th>Melting point $T_m$ (K)</th>
<th>Density of metal $\rho_S$ (kg/m$^3$)</th>
<th>Surface tension $\sigma$ (N/m)</th>
<th>Density of powder $\rho_L$ (kg/m$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>1360</td>
<td>7900</td>
<td>1.285</td>
<td>2300 (Cixtle powder)</td>
</tr>
<tr>
<td>Fe</td>
<td>1839</td>
<td>7200</td>
<td>1.700</td>
<td>2800 (Cixtle powder)</td>
</tr>
<tr>
<td>Sn</td>
<td>564.9</td>
<td>7000</td>
<td>0.544</td>
<td>940 (Stearic acid)</td>
</tr>
</tbody>
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Table 2: Properties of metals and molten fluxes.

<table>
<thead>
<tr>
<th>Metal</th>
<th>Copper alloy</th>
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<tr>
<td>Melting point</td>
<td>1273K</td>
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<tr>
<td>Density</td>
<td>8320 kg/m³</td>
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</tbody>
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Mold powder:

<table>
<thead>
<tr>
<th>Composition</th>
</tr>
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<tbody>
<tr>
<td>TC (wt%)</td>
</tr>
<tr>
<td>SiO₂</td>
</tr>
<tr>
<td>CaO</td>
</tr>
<tr>
<td>Al₂O₃</td>
</tr>
<tr>
<td>Na₂O</td>
</tr>
<tr>
<td>Fe</td>
</tr>
<tr>
<td>BaO</td>
</tr>
<tr>
<td>MgO</td>
</tr>
<tr>
<td>B₂O₃</td>
</tr>
<tr>
<td>CaO/Al₂O₃</td>
</tr>
<tr>
<td>Softening point</td>
</tr>
<tr>
<td>Freezing point</td>
</tr>
<tr>
<td>Viscosity @ 1473K</td>
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</table>

Table 3: Experimental conditions.

<table>
<thead>
<tr>
<th>Casting condition</th>
</tr>
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<tbody>
<tr>
<td>Casting weight (Casting length)</td>
</tr>
<tr>
<td>Superheat</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>Powder feeding rate</td>
</tr>
<tr>
<td>Casting speed</td>
</tr>
</tbody>
</table>

Oscillation condition

<table>
<thead>
<tr>
<th>Condition</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oscillation mode</td>
<td>Sin</td>
<td>Non-sin</td>
<td>Non-sin</td>
</tr>
<tr>
<td>Frequency</td>
<td>60 rpm</td>
<td>64 rpm</td>
<td>95 rpm</td>
</tr>
<tr>
<td>α</td>
<td>- 42%</td>
<td>45%</td>
<td></td>
</tr>
<tr>
<td>Stroke</td>
<td>8mm</td>
<td>8mm</td>
<td>8mm</td>
</tr>
</tbody>
</table>

Modification ratio:

\[ \alpha(\%) = \frac{t_{non-sin} \cdot \sin \theta}{t_{sin}} \times 100 \]