HEAT TRANSFER BETWEEN MOLD AND STRAND THROUGH MOLD FLUX FILM IN CONTINUOUS CASTING OF STEEL

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Synopsis:
Laboratory experiments have been performed to make clear the heat transfer behavior in the mold of continuous casting of steel. The overall thermal resistance of parallel plate filled with the mold powder has been measured. The interfacial thermal resistance of mold-powder interface and the thermal conductivity of the mold powder are analyzed quantitatively. The interfacial thermal resistance corresponding to the air gap of 20-50μm is observed in the case of solid mold flux. When the temperature of mold surface exceeds the solidification temperature of the mold flux, the interfacial thermal resistance disappears without air gaps. The crystallization of the mold flux inhibits the radiative heat transfer that is equivalent to 20% of the total heat flux. The thermal conductivity of the mold flux depends on the size of silicate ion. Large size of silicate ion promotes the conductive heat transfer.

KEYWORDS: continuous casting, slag, contact thermal resistance, mold flux, conductive heat transfer, radiative heat transfer, air-gap formation.

1. Introduction
In the continuous casting of steel, various factors affecting slab surface quality are the cooling conditions inside a mold[1]-[10], physical properties of the mold flux[11]-[16], oscillation conditions[17]-[19], and a taper of the mold[20]. Especially many studies have been reported on the non-uniform cooling in the mold, which causes surface defects. Grill et al.[3], have explained the formation mechanism of longitudinal corner cracks, as a result of the local delay in solidification caused by the air gap formation, due to shrinkage during the δ-γ transformation of the solidified shell. Matsumiya et al.[4], studied the formation of longitudinal facial cracks with an artificially scratched mold, and pointed out the importance of uniform cooling in the mold zone.

Although casting flux plays an important role in heat transfer through the mold flux film between the solidified shell and the mold, many of uncertain factor exist. Despite the thermal conductivity being one of the most fundamental physical properties, measurements reported by Elliott et al.[21], Ogino et al.[22], Ishiguro et al.[23], and Sakuraya et al.[24], are not coincident well.

The solidified flux film on the mold has a structure of glassy and crystalline phases. Ohmiya et al.[25], performed the pioneer simulation work on the heat transfer through the mold flux film. They reported an increase in heat flux due to radiative heat transfer when the mold flux was in the transparent glass phase, while Bagha et al.[26], reported the opposite result that the heat flux density increased with the existence of the crystalline phase. Recently, the use of mold flux with high basicity and high solidification temperature is known to be effective to prevent the longitudinal facial cracks observed with the middle carbon steel[27]. However, its mechanism is not sufficiently clear.

2. Experimental method
2.1 Experimental apparatus
The thermal resistance of the mold flux was measured by the steady state method in this study. The experimental apparatus used is shown in Fig. 1. The strand was simulated by an aluminum nitride plate (AIN plate) heated with SiC heaters. The dimensions of the AIN plate were 55x55x0.63mm3. The hot pressed AIN plate sintered at 1800°C had high chemical stability to the mold powder in an N2 atmosphere over 1500°C, and
was strong enough not to deform during the experiment. Moreover, the value for thermal conductivity of 160 W/(m·K) at room temperature was high enough to keep the surface temperature of plate uniform. The temperature of the back side of the AlN plate was kept constant at 1100°C.

A stainless steel (SUS304) block of dimensions 25x30x30mm³ served as the mold, which incorporated a passage of 8mm diameter for the coolant inside the mold. The heat flux was obtained from the temperature gradient measured in the stainless steel block. The block was connected to a precision driving unit which enabled the block to be moved up and down. After complete melting of the mold flux, the block was lowered to the fixed position, the surface temperature being controlled by using air or water as the coolant. Heat flux and temperature were measured varying the surface temperature of the block from 800°C to 200°C for different mold flux thickness.

The apparatus was placed in a sealed steel chamber in order to maintain the N₂ atmosphere at a quality that would prevent the oxidation of the AlN plate.

2.2 Principle of measurement
For the temperature distribution as shown in Fig.2 under the one dimensional heat flux condition in the steady state, the overall thermal resistance R_T is calculated from Eq.(1),

\[ R_T = \frac{\Delta T}{q_T} \]  
(1)

where the heat flux density q_T is obtained from Eq.(2). \( \Delta T \), the temperature difference in a parallel-sided plate, is estimated from Eqs.(3) and (4).

\[ q_T = K_{sus}(T_1 - T_2)/d_{sus} \]  
(2)

\[ T_m = T_1 + q_T d'/K_{sus} \]  
(3)

\[ T_2 = T_3 - q_T d_{AIN}/K_{AIN} \]  
(4)

3. Experimental results
3.1. Appearance of the flux film after the experiment
Four different casting fluxes were investigated. Their compositions are given in Table 1. Flux A was specially prepared for this study, its solidification temperature being extremely low. When the flux was in a molten state, it was a transparent liquid. However, it changed to an opaque solid and exhibited the crystalline phase below the solidification temperature.

Flux B is a typical flux for the continuous casting of steel. After the experiment, the film had separated into two layers, one being a light gray semi-transparent glassy phase, and the other a gray nontransparent crystalline phase.

Flux C is used for the continuous casting of middle carbon steel to reduce heat flux density and had high basicity. The solidified specimen was in the completely non-transparent crystalline phase.

Flux D is casting flux for brass, containing 75% of Na₂B₄O₇, and is characterized by its low solidification temperature. The solidified flux film was completely in the transparent glassy phase.

| Chemical compositions of the mold flux (mass-%) |
|-----------------------------------------------|
| Flux A | Flux B | Flux C | Flux D |
| SiO₂   | 14.8   | 34.4   | 31.3   |
| CaO    | 18.5   | 33.0   | 40.1   |
| Al₂O₃  | 1.1    | 6.1    | 2.08   |
| MgO    | 1.4    | 6.4    | 1.43   |
| Na₂O   | 21.7   | 13.4   | 3.98   |
| K₂O    | 22.7   | 8.1    | 11.4   |
| F      | 6.6    | 8.5    | 57.6   |
| B₂O₃   | 19.8   | 9.3    | 3.93   |
| Li₂O   | 5.8    | 2.9    | 2.0    |
| F.C.   | 0.0    | 0.0    | 0.0    |
| CaO/5SiO₂ | 1.11 | 0.96   | 1.30   |

Solidifying temperature T_s (°C) = 650

Solidifying temperature T_s (°C) = 1050

Solidifying temperature T_s (°C) = 1190

Solidifying temperature T_s (°C) = 680

Solidifying temperature T_s (°C) = 730

Table 1 Chemical compositions of the mold flux (mass-%)
3.2 Interfacial thermal resistance and thermal conductivity of the flux film

The obtained values for $R_T$ can be expressed as Eq.(5),

$$R_T = R_{INT} + \frac{d_p}{K}$$  \hspace{1cm} (5)

where $R_{INT}$ is the interfacial thermal resistance, and $K$ is apparent thermal conductivity $K_{eff}$. When $R_T$ is plotted against $d_p$ as shown in Figs.3-4, $K_{eff}$ and $R_{INT}$ can be obtained from the inverse of the slope and the intercept of the plots, respectively. The values obtained for $K_{eff}$ are shown in Table 2.

Table 2 Values obtained for effective thermal conductivity (W/(mK))

| $T_m$(°C) | Flux A | Flux B | Flux C | Flux D | Na2B4O7 [28] |
|-----------|--------|--------|--------|--------|--------------|
| 300       | 1.11   | 1.00   | 0.84   | 0.64   |               |
| 400       | 1.03   | 1.04   | 0.85   | 0.61   |               |
| 500       | 1.03   | 1.02   | 0.82   | 0.58   |               |
| 600       | 1.08   | 1.06   | 0.79   | 0.69   |               |
| 700       | 1.31   | 1.04   | 0.81   | 0.72   | 0.70          |
| 800       | 1.31   | 1.06   | 0.81   | 0.71   | 0.75          |

Figure 5 shows the relationship between interfacial thermal resistance $R_{INT}$ and mold surface temperature $T_m$. The drastic change in $R_{INT}$ can be seen for Fluxes A and D. While, the powder of high-solidification temperature fluxes B and C, show a stable $R_{INT}$ curve under these experimental conditions.

Figure 6 shows the influence of mold surface temperature $T_m$ on the effective thermal conductivity $K_{eff}$. $K_{eff}$ was almost independent of $T_m$, except for Flux A. $K_{eff}$ of Flux A increases by 22% when $T_m$ exceeds 650-700°C, which is near the solidification (650°C) temperature.

$K_{eff}$ of the high-basicity flux, Flux C, decreases by 22% compared with that of the typical powder, Flux B. $K_{eff}$ of Flux D is close to that of Na2B4O7 [28], which reveals the validity of the experimental method.

4. Discussion

4.1 Interfacial thermal resistance between the mold and the flux film

As shown in Fig.5, $R_{INT}$ for Flux A increases abruptly when $T_m$ is lowered 650°C or less. Differential thermal analysis (DTA) have indicated the special peek of solid-liquid transformation at 660°C. Therefore, it is considered that the increase of $R_{INT}$ below 650°C is caused by the formation of air gap due to the solidification of the flux being contact with the mold. The same phenomena was observed in case of Flux D. However, the change of $R_{INT}$ occurred around 300°C. This would be correlated with the fact that Flux D is easy to supercool, namely to be glassy state.

With regard to the air gap in the continuous casting mold, Nakato et al. [20] estimated the thickness of air gap to be 10-45 μm from the heat flux measurement. Under the present experiment, $R_{INT}$ change of 0.0004-0.0008 m²/K/W corresponds to the air gap of 20-50 μm, which shows good agreement with the one of Nakato's estimation. For the surface temperature of Cu mold to be approximately 300°C under usual casting conditions, an air gap in the order of 30 μm would always exist even in the meniscus zone.

The behavior of Flux A indicates that $R_{INT}$ becomes negligible when the temperature of the interface exceeds its melting point. The temperature of strand side interface in the mold was calculated to be around 1200°C under the
4.2 Thermal conductivity of the flux film

Generally, the heat is transferred by conduction, radiation and convection. The thickness of the flux film between the mold and strand was estimated to be about 0.2 mm from its consumption rate. As convection can not develop in such the narrow space, the obtained effective thermal conductivity $K_{eff}$ would have been due to thermal conductivity by radiation and convection.

4.2.1 Radiative heat transfer

Elliott et al. [21], have observed the radiative heat transfer in molten slag had been affected by FeO addition. Furthermore, Ohmiya et al. [25], have reported that 40-50% of the total heat was transferred by the radiation. Therefore, the contribution of radiative heat transfer can not be neglected in the present work either.

As has been mentioned, Flux A was completely liquid above a temperature of 650°C, and there may have been some radiative heat transfer. Total heat flux density $q_T$ through the flux film is expressed in Eqs.(6)-(10) [29].

$$q_T = q_c + q_r \quad (6)$$

$$q_c = (T_s - T_m)/(d_p/K_c + R_{INT}) \quad (7)$$

$$q_r = a_e (T_s^4 - T_m^4) \quad (8)$$

$$a_e = n_s/(0.75 a_d d_p + e_m^{-1} + e_s^{-1}) \quad (9)$$

$$K_{eff} = K_c d_p + a_e (T_s^4 - T_m^4)/(T_s - T_m) \quad (10)$$

where $q_c$ and $q_r$ are the conductive and radiative heat flux density respectively. Substitution of Eqs.(7)-(9) into Eq.(6) gives Eq.(10). It should be noted that $R_{INT}$ is negligible as already described.

A plot of $1/R_T$ vs. $(T_s^4 - T_m^4)/(T_s - T_m)$ is shown in Fig.7. $K_c d_p$ was obtained to be 1800 W/m²K when $d_p$ was 0.58 mm. Thus, calculated $K_c$ is 1.03 W/mK. Radiative thermal conductivity $K_r$ defined by Eqs.(11) and (12) was calculated to be 0.24 W/mK.

$$q_r = K_r (T_s - T_m)/d_p \quad (11)$$

Fig. 5 Influence of mold surface temperature $T_m$ on interfacial thermal resistance $R_{INT}$ for Fluxes A-D.

Fig. 6 Influence of mold surface temperature $T_m$ on effective thermal conductivity $K_{eff}$ for Fluxes A-D.

Fig. 7 Relationship between $(T_s^4 - T_m^4)/(T_s - T_m)$ and $1/R_T$ for Flux A ($T_m=650-800 \degree C, d_p=0.58 mm$).
\[ K_r = a_r d_p (T_r^4-T_m^4)/(T_r - T_m) \]  

(12)

This value agrees well with the change of \( K_{\text{eff}} (= 0.23 \text{ W/mK}) \) at around 650°C which is illustrated in Fig. 6. This fact indicates the disappearance of radiative heat transfer below 600°C due to the formation of an opaque crystalline phase.

In general, the energy of irradiated light is absorbed as lattice vibrations resonated with the wavelength of light in an optically non-transparent crystalline material. It is known that radiative heat transfer is absorbed, especially in ionic crystals, for the same reason \[30\]. The film of Flux D was glassy and transparent, so that no absorption change was observed. The films of Fluxes B and C always involved crystalline layers under the experimental conditions, so that radiation was absorbed in the whole temperature range.

For the radiative heat flux in the liquid phase of Flux A, the contribution of radiation would have been 20%. This is lower than the figure of Ohmiya et al.\[25\]. Radiative thermal conductivity \( K_r \) is proportional to the third power of the strand surface temperature. Therefore, the value for \( T_s \) affects the radiative heat transfer considerably. \( T_s \) of the Ohmiya's experiment was 1400°C compared with 1100°C in the present experiments, which would have caused the difference in contribution radiative heat transfer.

4.2.2 Conductive heat transfer

Most casting fluxes contain several tens of percent of SiO\(_2\). The thermal conductivity of molten flux containing SiO\(_2\) is comparable to that of a solid because of the network structure of silicate ions -(SiO\(_2\))\(_n\). The large size of silicate ion promotes conductive heat transfer. Mills et al.\[31\] have proposed an index to represent the size of silicate ions by the mole ratio of non-bridging oxygen (NBO) to the atoms of the network former as Si and Al. They recognized that the index had a linear relationship with the thermal conductivity of molten SiO\(_2\)-CaO-Al\(_2\)O\(_3\) systems.

Because the casting powder in this study is more complex systems, the index was modified as follows.

1. Not only the oxygen of CaO but also the oxygen of the alkali and alkaline earth oxide could be taken into consideration.
2. B is regarded as the atoms of the network former, besides Si and Al.

Fluorine is known as a network modifier. However, the addition of CaF\(_2\) has been reported to increase the thermal conductivity of slag\[32\]. Because of the discrepancy in the effect of fluorine on thermal conductivity, the effect of fluorine is neglected in this analysis.

The results of this relationship are shown in Fig. 8. Data from the present experiments are in good agreement with previous investigations\[24\],\[33\],\[34\]. It should be noted that the thermal conductivity at temperatures below 650°C was adopted for Flux A in order to eliminate the influence of radiation.

Flux C is utilized in the continuous casting of middle carbon steel to prevent longitudinal facial cracks. The use of Flux C reduced the mold heat flux by 10% in plant operation. This can be explained by the formation of micro-cracks with crystallization \[16\] or by the increased fraction of solid in the flux film.\[27\] The present experiments make it clear that the thermal conductivity of a high-basicity powder as Flux C was decreased due to a structural change in the silicate network, which contributed to a reduction of heat flux to some extent.

5. Conclusions

From the investigation of the effect of the mold surface temperature and physical properties of the mold flux on heat transfer through the flux film, following knowledge have been obtained.

1. When the temperature of the mold surface exceeded the solidification temperature of the mold flux, the interfacial thermal resistance disappeared together with disappearance of air gaps.
2. An interfacial thermal resistance corresponding to an air gap of 20-50 \( \mu \text{m} \) was observed in the case of the solid mold flux.
3. Crystallization of the mold flux inhibited radiative heat transfer which was equivalent to 20% of the total heat flux.
4. The thermal conductivity of the mold flux was related to the size of silicate ions. An increase in CaO/SiO\(_2\) ratio decreased the phonon thermal conductivity.
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APPENDIX

\( a_p \): absorption coefficient
\( \delta_f \): thickness of flux film
\( K_{ef} \): effective thermal conductivity
\( K_c \): conductive thermal conductivity
\( K_r \): radiative thermal conductivity
\( n \): index of refraction
\( q_t \): total heat flux density
\( q_r \): radiative heat flux density
\( q_c \): conductive heat flux density
\( T_s \): surface temperature of the strand
\( T_m \): surface temperature of the mold
\( T_s \): solidification temperature of the flux
\( \alpha_r \): radiative heat transfer coefficient
\( \varepsilon_m \): emissivity of the mold surface
\( \varepsilon_s \): emissivity of the strand surface
\( \sigma \): Stefan-Boltzmann constant \( 5.67051 \times 10^{-8} \) (W/(m\(^2\)·K\(^4\))}