Investigation of the coupled conductive and radiative heat transfer of molten slag in a cylindrical enclosure based on the zonal method

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Abstract

Heat transfer of molten slag under high temperature is significant for industrial processes. Unlike other phase change materials, the phase change temperature of molten slag is always not a constant during cooling process. In this case, the basis of judging different phases could not be accurately obtained, which brings difficulties for the calculation of radiation-solidification coupled heat transfer. In present work, the coupled conductive and radiative heat transfer of molten slag during solidification is investigated by the zonal method with fixed grid and moving area. In order to determine the liquid fraction of molten slag during phase change, the enthalpy method is improved by using crystallization kinetics. In addition, ray tracing method based traversal algorithm is developed to analyze the multiple reflection among three layers. The results indicate that the diffuse radiation in multi-layer could be described by tracking the path of reflections. However, the ray tracing method is time-consuming for computation. Compared with the ray tracing method, it is more efficient to calculate the radiative heat flux by solving a system of linear equations established according to the energy balance on each surface element. In initial stage of solidification, radiation is the main way of heat transfer. After solid slag is formed, heat transfer is dominated by conduction, and the proportion of the radiative heat flux in the total heat flux is lower than 30 %.

Key words: coupled heat transfer; thermal radiation; zonal method; solidification; molten slag

1. Introduction

Molten slag plays an important role in many industrial processes, such as ironmaking and steelmaking, continuous casting, and coal gasification [1-3]. The heat transfer in molten slag undergoing high temperature has an important effect on process control and quality of product [4,5]. Many studies about the coupled conductive and radiative heat transfer have been reported, and most of them are based on numerical simulation because it is difficult to carry out experiments at high temperature [6]. Among these numerical techniques, one of the main task is to solve the Stefan moving boundary problems for phase change materials [6,7], and this problem also

exists during heat transfer of molten slag. Several methods were proposed to solve the Stefan problem, such as the moving mesh method, fixed grid front tracking method and the enthalpy method [8-10]. These methods are capable of analyzing heat transfer of materials with known melting point or solidification temperature, such as the eutectic alloy and the binary alloy. However, slag is different from metallic material, and its solidification temperature or crystallization temperature changes with cooling rate, which indicates that the temperature of the interface between the liquid layer and the solidified layer is not a constant during solidification [11]. The change of the phase change temperature makes it difficult to find an accurate criterion for judging different phases. Therefore, the moving mesh method and fixed grid front tracking method may not be applicable for the solidification of molten slag. The enthalpy method does not need to track the interface, but the value of phase change temperature is also necessary to calculate the liquid fraction [12]. Nevertheless, if the relationship between the liquid fraction and the temperature is obtained, the enthalpy method could also be used to solve the temperature field from the energy equation. Considering the difficulties in determining the phase change temperature, the kinetics of phase change is used in this paper to describe the evolution of liquid fraction during cooling process.

Compared with the conductive heat flux, the calculation of the radiative heat flux is more difficult. According to the radiative transfer equation (RTE), different methods were developed to evaluate the radiation effect, such as the discrete ordinates method, the finite volume method, the discrete transfer method, the Monte Carlo method and the zonal method [13-16]. These methods have been used to study the radiative heat transfer in different systems, such as medium with one layer and homogeneous radiative properties, medium with one layer and various radiative properties, medium with multi-layer and homogeneous radiative properties, medium with multi-layer and space-time dependent radiative properties. Generally, when molten slag is solidified, the media around the wall of mold contains three layers, including the liquid slag layer, the solid slag layer and the air gap. The radiative heat flux on each interface is influenced by the multiple reflection among the three layers. By using ray tracing method, the effect of multiple reflection among multi-layer could be investigated, and references [17-19] present clear algorithm for tracking specular reflection. However, due to the precipitation of crystals during solidification, the interface between the liquid phase and the solid phase may not be smooth enough, which indicates that the diffuse reflection is more likely to occur on the interface between the two phases. Therefore, the reflection angle and the refraction angle could be calculated accurately by the Snell's refractive law. On the other hand, the interface between the solid phase and the liquid phase is moving during solidification of molten slag, which indicates that the reflectivity of the interface is not a constant during solidification. Considering the effects of the diffuse reflection and the moving interface during solidification, the Fresnel equation and the Snell's law may not be appropriate to describe the optical properties of the interface. In order to solve this problem, a new method based traversal algorithm is proposed to track the path of diffuse reflection. Therefore, the effect of multiple reflection on distribution of the radiative heat flux could be studied.

Recently, more and more attention are paid to the development of effective method for solving the RTE. A simplified version of the plating algorithm for calculation of total exchange areas was developed to reduce CPU time for radiative heat transfer analysis [20]. Hitti *et al.* studied the transient radiation and conduction heat transfer in glass sheets, and it was found that the thin layer approximation is capable of increasing the CPU efficiency [21]. Ebrahimi *et al.* analyzed the three-dimensional radiative heat transfer in industrial furnaces, and it was found that the zonal method is an effective numerical method for modeling three-dimensional thermal performance of gas-filled enclosures [22]. These studies suggest that the zonal method is effective to analyze the radiative heat transfer. However, during the formation of the solidified layer, the position and the area of interface change continuously with time, which indicates that the solid angle obtained from the surface area needs to be calculated repeatedly for meeting the movement of the interface. Because of the importance of the computation efficiency, an approach based fixed grid and moving area is proposed in this paper to make the zonal method applicable for calculating the radiative heat transfer during solidification.

In this work, the coupled conductive and radiative heat transfer during solidification is simulated, and experiments are carried out to obtain the boundary conditions and the radiative characteristics of the solid slags. The liquid fraction of molten slag is analyzed based crystallization kinetics. Moreover, an new algorithm is developed to study the effect of multiple diffuse reflection on distribution of radiative heat flux.

Nomenclature

A	area of boundary surface	Δu	thickness of control volume
C	heat capacity of water	W	rate of cooling water
C_p	specific heat capacity of slag	x, y, z	Cartesian coordinates
d	thickness of solidified layer		
dh	length of zone in z direction	<i>Greek symbols</i>	
dw	length of zone in y direction	α	degree of transformation
E_λ	spectral emission of boundary surface	ε	emissivity
f_s	solid fraction of molten slag	ρ	density of slag
F	effective heat transfer on copper detector	κ	absorption coefficient
h	enthalpy of slag	ω	solid angle
H	height of graphite crucible	θ	angular variable
I_λ	spectral intensity of emission	λ	wavelength
k	rate constant of crystallization	φ	angle factor
l	thermal conductivity	Φ	radiative source term
L	latent heat of molten slag	σ	Stefan-Boltzman constant
n	Avrami exponent		
n_r	index of refraction	<i>Subscript</i>	
N_c	number of reflections	b	block body
q	radiative heat flux	cu	copper detector
q_t	total heat flux	f	solid slag film
Q_λ	spectral emission of surface element	g	graphite crucible
r	distance between two surface elements	i, j, m, n	index of element
R	radius of bottom of crucible	in	interface
R_a	apparent reflectivity of slag film	p	index of sublayer
R_s	reflectivity for single reflection	r, s	index of incident light
t	time	rad	radiative heat transfer
T	temperature	v	index of control volume
T_f	fluid temperature of molten slag		

through the heating elements. Meanwhile, the temperature of the molten slag can be measured by a thermocouple that is located on the bottom of the furnace. In order to simulate the solidification of molten slag, the cooling water through a flow meter is inputted into the water-cooled detector by a pump. The cooling water outputted from the detector is sent to the sink, and the output water is not returned to the detector. The temperatures of the cooling water at the inlet and the outlet are measured by two thermocouples that is located on the junction between the tube and the top surface of the detector. During testing, the temperatures of cooling water at inlet and outlet are collected by the data acquisition system, which can be used to calculate the heat flux through the molten slag during solidification.

2.3 Experimental procedure

Slag of 350 gram are put into a cylindrical crucible that is made of graphite. The slag sample in the crucible is heated by a MoSi₂ furnace from room temperature to 1673 K. The slag pool in crucible is maintained at 1673 K for 10 minutes. After a homogeneous slag pool is formed, the water-cooled detector is immersed into the molten slag. The flow rate of cooling water is maintained at 0.056 kg·s⁻¹. When the top surface of the detector and the fluid level of molten slag is on the same plane, the temperatures of cooling water at inlet and outlet are recorded by computer.

As the solidification process evolves, heat of molten slag was continuously taken away by the flowing cooling water. The total heat flux through the solidified layer could be calculated according to the temperature difference of cooling water, as shown in Eq. (1). After 45 s, the water-cooled detector moves upward automatically, and the solid slag film around the detector can be obtained. The appearance of the solid slag film attached on the detector is shown in Fig. 1(b). The fluid temperature of the molten slag is determined using a high temperature microscope [23], and this temperature is considered as the temperature of interface between the liquid phase and

the solid phase.

$$q_t = \frac{W \cdot C \cdot \Delta T}{F} \quad (1)$$

In **Eq. (1)**, q_t is the total heat flux, $W \cdot m^{-2}$; W is the rate of cooling water, $m^3 \cdot s^{-1}$; ΔT is temperature difference of cooling water, K; F is the effective heat transfer area on the water-cooled detector, m^2 ; C is the heat capacity of cooling water, $J \cdot (kg \cdot K)^{-1}$.

3. Numerical Simulations

3.1. Physical model and governing equation

From the appearance of the solid slag film, the solidified layer grows evenly in the normal direction of each surface. Therefore, the conduction in the molten slag from the inner surface of crucible to the detector could be considered as transient one dimensional heat transfer. Considering the couple effect of radiation, the energy equation can be written as

$$\frac{\partial(\rho \cdot h)}{\partial t} = -\frac{\partial}{\partial x} \left(-l \frac{\partial T}{\partial x} \right) - \nabla \cdot q_{rad} \quad (2)$$

where h is the enthalpy of slag during solidification, $J \cdot kg^{-1}$; ρ is the density of slag, $kg \cdot m^{-3}$. T is temperature of molten slag, K; t is time, s; l is the thermal conductivity of slag, $W \cdot (m \cdot K)^{-1}$; q_{rad} is the radiative heat flux, $W \cdot m^{-2}$. When crystallization occurs during solidification, the value of h can be obtained from

$$h = CpT + (1 - fs)L \quad (3)$$

where Cp is the specific heat capacity of slag, $J \cdot (kg \cdot K)^{-1}$; fs is the solid fraction, and this value could be considered as the degree of transformation. L is the latent heat during phase change, $J \cdot kg^{-1}$. For molten slag or polymer, the Avrami equation is usually used to describe the degree of transformation as a function of time during solidification [24], as shown in **Eq. (4)**.

$$\alpha = 1 - \exp\left(-(k \cdot t)^n\right) \quad (4)$$

In **Eq. (4)**, parameters k and n are constants related to kinetics of phase change. The value of n is usually between 1.0 and 3.5. According to previous studies about the crystallization kinetics of molten slag, the growth type of crystals precipitated from the liquid slag is disc-like, which indicates that the value of the Avrami exponent is around 2.0 [25]. Therefore, the value of n is taken as 2.0 in this work. The advantage of using **Eq. (4)** is that the solid fraction during phase change could be obtained with unknown phase change temperature of slag. It is noted that **Eq. (4)** is derived from isothermal process. However, the Avrami equation is extended to non-isothermal processes, because non-isothermal processes could be considered as a succession of infinitesimal small isothermal steps [25]. It is reported that the Avrami equation agrees well with the experimental data during nonisothermal crystallization, which indicates that the Avrami equation could be used to describe the relationship between the solid fraction and time during nonisothermal process.

The radiative heat flux during solidification is calculated according to the zonal method with fixed grid and moving area. Description of grid and zones are shown in **Fig. 2(a)-(c)**. For the v th control volume, the energy equation can be expressed by

$$\rho Cp \frac{T_p^{m+1} - T_p^m}{\Delta t} = l \cdot \frac{T_{p+1}^m - 2T_p^m + T_{p-1}^m}{\Delta u^2} - L\rho \frac{f_{s,p}^m - f_{s,p}^{m+1}}{\Delta t} + \Phi_{rad,v}^m \quad (5)$$

where Δt is time step; Δu is thickness of control volume; In this work, the value of Δt is chosen as 0.02 s, and the value of Δx is chosen as 0.2 millimeter. T_p^m is temperature of the p th sublayer when time is m ; $\Phi_{rad,v}^m$ is radiative source term of v th control volume when time is m , which can be calculated by **Eq. (6)**.

$$\Phi_{rad,v}^m = \sum_{r=1, s=1} \left(q_{r,p+1} - |q_{r,p}| + |q_{s,p}| - |q_{s,p+1}| \right) \quad (6)$$

The radiative source term contains two parts. On the one hand, the incident radiation emitted from the inner surface of crucible transmits through the v th control volume from the $p+1$ th sublayer. On the other hand, the interface of the two phases can also emit radiation, and this energy enters into the v th control from p th sublayer, as shown in **Fig. 2(a)** and **Fig. 2(b)**. For the system studied, the radiative source term is calculated based the zonal method. When the radiative energy emitted from surface element i transfers to surface element j , the spectral intensity of emission from element i can be expressed as

$$I_{\lambda,i} = \frac{dQ_{\lambda}}{dA_i \cos\theta_1 d\omega} \quad (7)$$

where dQ_{λ} is spectral emission from element i ; θ_1 is angle between $q_{i,j}$ and normal vector of element i . ω is solid angle, which can be obtained from

$$d\omega = \frac{dA_j \cos\theta_2}{r^2} \quad (8)$$

where θ_2 is angle between q_{ij} and normal vector of element j ; r is the distance between surface element i and surface element j . The expression of $I_{\lambda,i}$ can be written as

$$I_{\lambda,i} = \frac{q_{\lambda,i}}{\pi} = \frac{dQ_{\lambda} r^2}{dA_i dA_j \cos\theta_1 \cos\theta_2} \quad (9)$$

where $q_{\lambda,i}$ is spectral heat flux of emission from element i . According to the RTE, the spectral intensity of incident radiation arriving at element j can be calculated, as shown in Eq. (10).

$$I_{\lambda,j} = I_{\lambda,i} \exp(-\kappa r) + \int_{s=0}^{\infty} \frac{\kappa n_r^2 \sigma T^4}{\pi} \exp(-\kappa(r-s)) ds \quad (10)$$

In Eq. (10), κ is absorption coefficient, m^{-1} ; σ is Boltzman's constant, $W \cdot m^{-2} \cdot K^{-4}$. n_r is the index of refraction. According to Eq. (9) and Eq. (10), the spectral incident radiation arriving at element j from element i is expressed as Eq. (11).

$$dQ_{\lambda,i,j} = \frac{I_{\lambda,j} dA_i dA_j \cos\theta_1 \cos\theta_2}{r^2} \quad (11)$$

The radiative heat flux arriving at element j from element i and the the radiative heat flux arriving at element i from element j can be obtained by integrating the $dQ_{\lambda,i,j}$ and $dQ_{\lambda,j,i}$ for the spectral interval, as shown in Eq. (12) and Eq. (13).

$$q_{i,j} = \frac{1}{dA_j} \sum_{\kappa} dQ_{\lambda,i,j} d\lambda \quad (12)$$

$$q_{j,i} = \frac{1}{dA_i} \sum_{\kappa} dQ_{\lambda,j,i} d\lambda \quad (13)$$

In order to obtain the radiative heat flux at each surface element, the inner surface of crucible, surfaces of detector, and the interface between the two phases are divided into a set of surface elements, as shown in Fig. 2(c). The wall of crucible is divided into 100 elements. The bottom surface has 50 elements. For the interface of the two phases, both the wide face and narrow face have 35 zones, and the narrow face is divided into 25 zones. During solidification, the position of interface moves with time. Therefore, the coordinates and the area of each surface element change with time. However, the number of zones on each surface is fixed. The point i is defined as the starting point of light, and the point j is defined as the end point of light. Both the point i and the point j is on the center of each surface element. The center of bottom surface of crucible is defined as the origin of coordinates, and the coordinates of point i on the bottom surface of crucible are determined by

$$\begin{cases} x_i = (i/10 + 0.5) \cdot dR \cdot \cos\left(\frac{2\pi}{10} (i\%10 + 0.5)\right) \\ y_i = (i/10 + 0.5) \cdot dR \cdot \sin\left(\frac{2\pi}{10} (i\%10 + 0.5)\right) \\ z_i = 0 \end{cases} \quad 0 \leq i < 50 \quad (14)$$

The coordinates of point i on the wall of crucible are determined by

$$\begin{cases} x_i = R \cdot \cos\left(\frac{2\pi}{10} ((i-50)\%10 + 0.5)\right) \\ y_i = R \cdot \sin\left(\frac{2\pi}{10} ((i-50)\%10 + 0.5)\right) \\ z_i = ((i-50)\%10 + 0.5) \cdot dH \end{cases} \quad 50 \leq i < 150 \quad (15)$$

where “/” is to obtain the integer of expression, and “%” is to obtain the remainder of expression. According to previous studies [26], the thickness of the solidified layer as a function of time is determined by

$$d = k' t^m \quad (16)$$

where d is the thickness of the solidified layer at t seconds; k' and m are constants. When the thickness of the solid film at 45 s is determined, Eq. (16) is written as

$$d = d_f \left(\frac{t}{45} \right)^k \quad (17)$$

where d_f is the thickness of the solid slag film measured by vernier caliper. The coordinates of point i on the interface are determined by

$$\begin{cases} x_i = a_x (17.5 + d_i) + b_x \cdot dw \\ y_i = a_y (10 + d_i) + b_y \cdot dn \\ z_i = 45 - d_i + a_z \cdot dh \end{cases} \quad (18)$$

where dw , dn and dh are the length of each zone in x direction, y direction and z direction, respectively. The values of a_x , b_x , a_y , b_y , a_z on each surface are listed in **Table 2**.

The **Eq. (18)** can also be used to calculate the coordinates of point j on the p th sublayer by replacing d with an integral multiple of Δu . Thus, for liquid slag, the radiative heat flux arriving at the p th sublayer from surface element i can be expressed as

$$q_{i,p} = \sum_{j(p)=150}^{304} q_{i,j(p)} \quad (19)$$

where $j(p)$ represents surface elements on the p th sublayer. For solid slag film, the radiative heat flux arriving at p th sublayer is expressed as

$$q_{i,p} = Ta \sum_{j(in)=150}^{304} q_{i,j(in)} \quad (20)$$

where $j(in)$ represents surface elements on the interface; Ta is the apparent transmittivity of slag film [21], which can be expressed as

$$Ta = \frac{(1 - Rs)^2 Ts}{1 - Rs^2 Ts^2} \quad (21)$$

where Ts is the transmittivity for single transmission. The value of Ts can be expressed as **Eq. (22)**.

$$Ts = \exp(-\kappa d) \quad (22)$$

From above analysis, radiative exchanges between the inner surface of crucible and the moving interface during solidification could be described by fixed grid and moving zones. Another problem is that the radiative heat flux of incident radiation at each surface element could not be calculated by $\epsilon_g \sigma T^4$, because multiple reflections may influence the distribution of the radiative heat flux. Assuming that light is emitted from the inner surface of crucible and the interface of two phases, energy balance on each surface element could be constructed, as shown in **Eq. (23)** and **Eq. (24)**. The radiative heat flux leaving element i contains two parts. The first part is emission of surfaces, and the second part is reflected component from all incoming directions.

$$q_{\lambda,i} = \epsilon_g \cdot E_{g,\lambda} + (1 - \epsilon_g) \sum_{j=0, j \neq i}^{j=304} q_{\lambda,j,i} \quad i < 150 \quad (23)$$

$$q_{\lambda,i} = \epsilon_f \cdot E_{in,\lambda} + Ra \sum_{j=0, j \neq i}^{j=149} q_{\lambda,j,i} \quad i > 149 \quad (24)$$

In **Eq. (23)**, $E_{g,\lambda}$ is the spectral emission of inner surface of crucible, $\text{W} \cdot \text{m}^{-2}$. ϵ_g is the emissivity of crucible; $q_{\lambda,i}$ is the spectral heat flux of radiation leaving element i . In **Eq. (24)**, $E_{in,\lambda}$ is the spectral emission of the solid slag film; ϵ_f is the emissivity of slag film. Ra' is the total reflectivity on the interface of two phases, and the value of Ra' could be obtained according to the radiative heat exchange between the slag film and the detector. The expression of Ra' is written as

$$Ra' = Ra + \frac{Ta^2}{\frac{1}{1 - \epsilon_{cu}} - Ra} \quad (25)$$

where Ra is the apparent reflectivity for a given wavelength. According to the relationship between the apparent reflectivity and the reflectivity for single reflection [21], Ra can be expressed as

$$Ra = Rs + \frac{Rs(1 - Rs)^2 Ts^2}{1 - Rs^2 Ts^2} \quad (26)$$

Despite the multiple reflections among layers, there is a certain relationship between the radiative energy emitted from different surfaces and the optical properties of slag film. Therefore, the radiative heat flux leaving each surface element could be solved from a system of linear equations, and **Eq. (23)** and **Eq. (24)** are expressed as a matrix form, as shown in **Eq. (27)**.

$$\begin{bmatrix}
-1 & A_{1,0} & A_{2,0} & A_{3,0} & \boxed{?}A_{j,0} & \boxed{?}A_{304,0} \\
A_{0,1} & -1 & A_{2,1} & A_{3,1} & \boxed{?}A_{j,1} & \boxed{?}A_{304,1} \\
A_{0,2} & A_{1,2} & -1 & A_{3,2} & \boxed{?}A_{j,2} & \boxed{?}A_{304,2} \\
\boxed{?} & \boxed{?} & \boxed{?} & \boxed{?} & \boxed{?} & \boxed{?} \\
A_{0,j} & A_{1,j} & A_{2,j} & \boxed{?}A_{j-1,j} & -1 & \boxed{?}A_{304,j} \\
\boxed{?} & \boxed{?} & \boxed{?} & \boxed{?} & \boxed{?} & \boxed{?} \\
A_{0,304} & A_{1,304} & A_{2,304} & \boxed{?}A_{j,304} & \boxed{?}-1 &
\end{bmatrix}
\begin{bmatrix}
q_{\lambda,0} \\
q_{\lambda,1} \\
q_{\lambda,2} \\
q_{\lambda,j} \\
q_{\lambda,304}
\end{bmatrix}
+
\begin{bmatrix}
B_0 \\
B_1 \\
B_2 \\
B_i \\
B_{304}
\end{bmatrix}
= 0
\quad (27)$$

In Eq. (27), $A_{j,i}$ and B_i can be written as follows:

$$\begin{cases}
A_{j,j} = \frac{(-\epsilon_g) \cdot \exp(-\kappa r) \cdot dA_j \cos\theta_1 \cos\theta_2}{\pi \cdot r^2} & 0 \leq i < 150, 0 \leq j < 305, i \neq j \\
A_{j,j} = \frac{Ra' \cdot \exp(-\kappa r) \cdot dA_j \cos\theta_1 \cos\theta_2}{\pi \cdot r^2} & 150 \leq i < 305, 0 \leq j < 305, i \neq j
\end{cases}
\quad (28)$$

$$\begin{cases}
B_i = \epsilon_g E_{g,\lambda} + (-\epsilon_g) \cdot \sum_{j=0, j \neq i}^{j=304} \sum_{s=0}^{s=r} \kappa \cdot I_{\lambda,b}(r) \cdot \exp(-\kappa(r-s)) \Delta s \cdot \frac{dA_j \cos\theta_1 \cos\theta_2}{r^2} \\
\quad 0 \leq i < 150, 0 \leq j < 305, i \neq j \\
B_i = \epsilon_f E_{m,\lambda} + Ra' \cdot \sum_{j=0, j \neq i}^{j=304} \sum_{s=0}^{s=r} \kappa \cdot I_{\lambda,b}(r) \cdot \exp(-\kappa(r-s)) \Delta s \cdot \frac{dA_j \cos\theta_1 \cos\theta_2}{r^2} \\
\quad 150 \leq i < 305, 0 \leq j < 305, i \neq j
\end{cases}
\quad (29)$$

In Eq. (28) and Eq. (29), the values of $A_{j,i}$ and B_i change with time and wavelength. When time step is given, the coordinates of each node and the area of each zone can be obtained according to the thickness of the slag film obtained from Eq. (17). Because the heat absorbed by the detector is much lower than the heat taken away by the cooling water, it is assumed that the heat energy through the slag could be taken away completely by the flowing cooling water. An initial condition and two boundary conditions are necessary to solve Eq. (2). The initial condition is that the initial temperature of the molten slag is kept at 1673 K, as shown in Eq. (30).

$$T_p^0 = 1673 \quad (30)$$

The first boundary condition is that the heat flux at $x = 0$ is equal to the heat flux measured by the water-cooled detector, as shown in Eq. (31). The second boundary condition is that the bulk temperature of the molten slag is equal to the initial temperature of the molten slag, as shown in Eq. (32).

$$l \frac{T_1^m - T_0^m}{\Delta u} + \epsilon_{cu} q_{rad,0}^m = q_i^m \quad (31)$$

$$T_{50}^m = 1673 \quad (32)$$

In Eq. (31), ϵ_{cu} is emissivity of the detector; $q_{rad,0}^m$ represents the radiative heat flux through solid slag film. The value of k and k' could be determined by using the enumeration method, and Eq. (33) is used to judge whether the selected value is reasonable.

$$T_{in} = T_f \quad (33)$$

In Eq. (33), T_{in} represents the temperature of interface when time is 45 s. $q_{rad,0}^m$ is obtained according to Eq. (34).

$$q_{rad,0} = \sum_{\lambda=300}^{\lambda=2600} \sum_{i=0}^{i=149} \sum_{j=150}^{j=304} \frac{q_{\lambda,i,j}(in) Ta}{1 - (1 - \epsilon_{cu}) Ra} \cdot \Delta \lambda \quad (34)$$

It is assumed that the optical properties of slag film are dependent on wavelength, but is taken independent on temperature. The absorption coefficient of molten slag is taken as 100 m^{-1} , which is independent on wavelength and temperature. Because the thickness of the air gap between slag film and detector is very small (lower than $50 \mu\text{m}$ [4]), it is assumed that all of the radiative energy through solid slag film could arrive at surfaces of detector. In addition, the radiation emitted from the detector is not taken into consideration since the temperature of detector surface is lower than 600 K [4]. The values of parameters used in calculation are listed in Table 3. The specific heat capacity and latent heat listed in Table 3 are measured by the Differential Scanning Calorimetry. The steps of the enumeration method used to determine the values of k and k' are described as follows:

Step 1: Determine the initial value and the step length. The initial value of k is 0.001, and the initial

value of k' is 0.5; Δk is 0.0005 s⁻¹, $\Delta k'$ is 0.01.

Step 2: Calculate the temperature field of molten slag.

Step 3: Examine whether **Eq. (13)** is satisfied according to the calculated results and the experimental data. If the difference between T_{in} and T_f is lower than 1 K, the values of the two parameters will be considered reasonable. Then, go to step 6. If not, go to step 4.

Step 4: Renew the values of k and k' by using the step length. $k = k + \Delta k$, $k' = k' + \Delta k'$.

Step 5: Calculate the temperature field of molten slag by using the new values of k . Then, go to step 3.

Step 6: Output the results of temperature.

3.2. Effect of number of surface elements on temperature distribution

When the zonal method is used to calculate radiative heat flux, the surface of enclosure and the interface between two different phases need to be divided into several zones. With the increase of surface elements, the solid angles can be decomposed accurately. However, CPU time and memory space for calculation will also increase. In order to determine the numbers of zones on each surface, the effect of numbers of surface elements on temperature distribution is investigated. The parameters used in calculation are taken as follows. The total heat flux q_t at $x = 0$ is 0.5 megawatt per square meter. The temperature of interface between two different phases are 1423 K. The thickness of solid phase is 2 millimeter. Values of other parameters are described in the section 3.1 of this paper. **Fig. 3** shows that temperature distributions of molten slag under steady state for various numbers of surface elements and optical properties. From **Fig. 3**, when the number of surface elements on interface is no less than 155 and the number of surface elements on crucible is no less than 108, the number of surface elements has minor effect on temperature distributions for various optical properties. Therefore, the number of surface elements on interface is taken as 155, and the number of surface elements on inner surface of crucible is taken as 150, as shown in **Fig. 2(c)**.

3.3. Ray tracing method

During solidification of molten slag, multiple reflections among layers make it difficult to describe the processes of radiation. Therefore, ray tracing method is introduced to analyze the radiative heat transfer among multiple layers. For the diffuse surfaces in experiment, a new method based traversal algorithm is developed to describe reflections among different layers. Combined with the zonal method, the effect of multiple reflections on distributions of the radiative heat flux on surfaces is discussed in this paper. When the incident light arrives at the surface of a medium, reflected light and transmitted light are generated. Description of the multiple reflection among three layers is described in **Fig. 4(a)-(b)**. According to the directions of propagation, the light in system could be divided into 12 categories, as shown in **Fig. 4(a)**. Initially, light is emitted from the inner surface of crucible. Evolution of light with increasing reflection times N_c is shown in **Fig. 4(b)**. From **Fig. 4(b)**, the light emitted from the inner surfaces of crucible has four branches because the radiative energy emitted from the crucible could also be absorbed by the crucible itself. The reflected light and the transmitted light always come in pairs. The data structure used to describe this process is similar to the binary tree in computer science. The number of branches increases with the increase of reflection times. The number of each node represents different kinds of light, and the value of each node can be defined as the radiative heat flux on surfaces. Obviously, the value of leaf node can be obtained according to the value of root node. Four arrays are defined to obtain the radiative heat flux under each reflection. Array A1 is used to save the indexes of root nodes and the indexes of corresponding leaf nodes. Array A2 is used to save the ratio of the radiative heat flux on leaf node and the radiative heat flux on root node. The indexes of nodes for each reflection are saved in array A3, and the radiative heat fluxes for each node are saved in array A4. The values in A1 and A2 are listed in **Table 4** and **Table 5**, respectively.

When m is 1, radiative energy is emitted from the inner surfaces of crucible. When m is 2, radiative energy is emitted from the interface of the two phases. When n is 1, incident radiation arrives at the inner surfaces of crucible. When n is 2, incident radiation arrives at the interface of the two phases. In **Table 5**, $\varphi_{m,n}$ can be obtained from

$$\varphi_{m,n} = \frac{A_m}{A_n \cdot \sum_i q_{N_c} dA_{i,m}} \sum_i \sum_j \frac{f\left(\frac{q_{N_c}}{\pi}\right) \cdot dA_{i,m} dA_{j,n} \cos\theta_1 \cos\theta_2}{r^2} \quad (35)$$

where i represents surface element on surface m , and j represents surface element on surface n . q_{N_c} is the radiative heat flux for corresponding root node when reflection time is N_c , which can be obtained from the ratio of q_{N_c} and q_{N_c+1} ; f is the radiative transfer equation, as shown in **Eq. (36)**.

$$f\left(\frac{q_{N_c}}{\pi}\right) = \frac{q_{N_c}}{\pi} \cdot \exp(-\kappa r) + \int_{s=0}^{s=r} \frac{\kappa n_r^2 \sigma T^4}{\pi} \exp(-\kappa(r-s)) ds \quad (36)$$

The data saved in array A3 and array A4 change with increasing N_c , and this process could simulate the

multiple reflections among different layers. An algorithm based level-order traversal is used to assign values for array A3 and array A4. The critical code used to implement the algorithm is written in the C language:

```
int col=0; //Index of column;
//Complete the data saved in the second column of array A3 and the data saved in the second column of array A4.
for(int j=0;A3[j][0]!=0;j++)
{
    int a=A3[j][0];
    if(A1[0][a-1]!=0)
    {
        A3[col][1]=A1[0][a-1];
        A4[col][1]=A4[j][0]*A2[A3[col][1]-1];    col++;
//The indexes of the leaf nodes are saved in the second column of array A3.
//The radiative heat fluxes for the corresponding leaf nodes are saved in the second column of array A4.
        A3[col][1]=A1[1][a-1]; //The second branch;
        A4[col][1]=A4[j][0]*A2[A3[col][1]-1];    col++;
        if(a==1) //Considering that part of the radiation could be reflected by the crucible itself.
        {
            A3[col][1]=1;A4[col][1]=A4[j][0]* $\varphi_{1,1}$ *(1- $\epsilon_g$ ); col++;
            A3[col][1]=2;A4[col][1]=A4[j][0]* $\varphi_{1,1}$ * $\epsilon_g$ ; col++;
        }
    }
}
//Renew the data saved in array A3 and array A4
for(int v=0;v<M;v++) {A4[v][0]=A4[v][1];A4[v][1]=0;
    A3[v][0]=A3[v][1];A3[v][1]=0;}
//The indexes of the root nodes are saved in the first column of array A3.
//The radiative heat fluxes for the corresponding root nodes are saved in the first column of array A4.
```

The computational steps of the ray tracing method is described as follows:

- Step 1: Determine the initial value of the reflection time N_c , the maximum value of N_c , and the initial value of $q(N_c = 1)$. For example, initially, $N_c = 1$, $q(N_c = 1) = \epsilon_g \sigma T^4$, the maximum value of N_c is 20, $A3[0][0]=1$, $A4[0][0]=\epsilon_g \sigma T^4$.
- Step 2: According to the data saved in the first column of array A3, complete the data saved in the second column of array A3 by referring the code described above.
- Step 3: For each leaf node saved in the second column of array A3, search the index of the corresponding root node according to array A1.
- Step 4: For each root node, obtain the radiative heat flux q_{N_c} from the first column of array A4.
- Step 5: Calculate the value of $\varphi_{m,n}$ by using **Eq. (35)**, and complete the data saved in the second column of array A4 by referring the code described above.
- Step 6: Examine whether the value of N_c reaches the maximum value. If the value of N_c reaches the maximum value, go to step 10. If not, go to step 7.
- Step 7: For the leaf node whose index is 5 or 10, accumulate the radiative heat flux into $q_{rad,0}$.
- Step 8: Renew the data saved in array A3 and array A4 by referring the code described above.
- Step 9: Renew the value of N_c by using $N_c = N_c + 1$, and go to step 2.
- Step 10: Output the result of $q_{rad,0}$, and the value of $q_{rad,0}$ is the radiative heat flux through the solidified layer after multiple reflections.

3.4. Effect of multiple reflection on radiative heat flux

For most of industrial furnaces, the inner surface can be considered emitting, absorbing, and reflecting. Therefore, radiative energy emitting from surfaces will reflect multiple times in different layers. In addition, a surface element will exchange radiative energy with several surface elements. In order to analyze this effect on the radiative heat flux, the ray tracing method is proposed to describe multiple diffuse reflection among multi-layer. All computation is realized in a PC with 2.60 GHz Intel Celeron CPU (G1610) and 4.0G memory. **Fig. 5** shows the results of the radiative heat flux through the solid phase changing with reflection times. For various of optical properties, it can be seen from **Fig. 5** that the value of $q_{rad,0}$ increases with increasing reflections. When the reflection time is higher than 20, the effect of multiple reflection on the radiative heat flux is very small. However, with the increase of reflection time, CPU time needed to complete calculation will also increase. The zonal method described in section 3.1 is also used to calculate $q_{rad,0}$, and results are shown in **Table 6**. From **Table 6**, results obtained from the two different methods are close to each other, but CPU time needed to calculate $q_{rad,0}$ for the zonal method is much less than time required for the ray tracing method. In addition, it can

be found that the difference of $q_{rad,0}$ between the zonal method and the ray tracing method decreases with the increase of reflection time.

Besides the radiative heat flux through the solid phase, the temperature distributions under steady state for various optical properties and the distributions of the radiative heat flux on the inner surface of crucible are calculated by the two approaches. **Fig. 6** shows comparisons for temperature distributions under steady state. In **Fig. 6**, points represent results obtained from the method described in section 3.1, and solid lines represent results obtained from the ray tracing method. It can be observed from **Fig. 6** that the two approaches give very similar results. The results calculated from the two approaches indicate that the optical properties of the solid phase influence temperature distribution obviously. When the solidified layer has high reflectivity and low transmittivity, the radiative heat flux through the solidified layer is low, as shown in **Fig. 5**. In this situation, conduction dominates the heat transfer from the crucible to the detector, and high temperature gradient is observed in the solidified layer on the detector side. In addition, intense reflections of interface may decrease the radiative energy inputted into the solidified layer. In this case, the value of radiative source term for each control volume is much lower than the value of conductive source term. As a result, for the solidified layer with high reflectivity and low transmittivity, a linear relationship between temperature and thickness of slag film is observed when x is lower than 2.0 millimeter, as shown in **Fig. 6**.

Distributions of the radiative heat flux on the inner surface of crucible are also calculated by the two different approaches, and the values of R_a and T_a are chosen as 0.1 and 0.6, respectively. **Fig. 7(a)-(b)** gives the results obtained from the method described in section 3.1. The results obtained by the ray tracing method are shown in **Fig. 7(c)-(f)**. **Fig. 7(c)** gives the distribution of the radiative heat flux on the bottom when the reflection time is taken as 24. The distributions of the radiative heat flux on the inner surface of crucible for different reflection times are shown in **Fig. 7(d)-(f)**. For the bottom of the crucible, it can be observed from **Fig. 7(a)** and **Fig. 7(c)** that the heat flux of the incident radiation on the inner regions is higher than that on the outer regions. This phenomenon may be caused by the multiple reflection from the interface between two phases. Similarly, the edge of the bottom surface may receive more radiation due to the reflection from the wall of crucible. From **Fig. 7(a)** and **Fig. 7(c)**, when R/R_c is higher than 2.0, the radiative heat flux increases with the increase of R .

For the radiative heat flux on the inner wall, the incident light is reflected between the narrow surface of the detector and the wall of crucible. As a result, when h/h_c is between 0.5 and 0.6, the heat flux of the incident radiation is higher than the heat flux on other regions. Due to the reflection from the bottom of the crucible, two regions with strong incident radiation are observed when h/h_c is between 0.25 and 0.30. In addition, it can be observed from **Fig. 7(d)-(f)** that the heat flux of the incident radiation on the inner wall increases with increasing reflection time, which suggests that the multiple reflection could influence the distribution of heat flux. Furthermore, the heat flux calculated by the ray tracing method is lower than results calculated by the zonal method. However, the differences of results between the two method decrease with increase of reflection time.

Whether for the distributions of the radiative heat flux or the value of heat flux through the solidified layer, both the two approaches could give similar results. The ray tracing method is conducive to understanding the distribution of the radiative heat flux because this method is capable of describing process of radiation. However, compared with the ray tracing method, it is more efficient to calculate the radiative heat flux on each surface element by solving a system of linear equations established according to the energy balance between the incident radiation and the reflected radiation.

4. Results and Discussion

In this part, firstly, the results calculated according to the method described in section 3.1 are presented, including the variation of temperature field during cooling process and the distribution of the incident radiation on each surface. Then, the transient heat transfer process during solidification of molten slag is analyzed based on the calculated results. Finally, the differences between the crystalline slag and the glassy slag are discussed from the perspective of the radiative heat flux through the solid slag film.

4.1 Total heat flux

The variation of total heat flux measured by the water-cooled detector is shown in **Fig. 8**. It can be obtained from **Fig. 8** that the total heat flux first increases and then decrease with time. Before 6 s, the heat flux increases sharply with time from about 0.2 megawatt per square meter to 0.9 megawatt per square meter. After a solidified layer is formed, part of the radiative energy is reflected by the solidified layer. Meanwhile, the thermal resistance begins to increase due to the increase of the slag film thickness and the formation of air gap, which leads to the sharp decrease of heat flux after 6 s. Because the crystallization could increase the interface thermal resistance by increasing the surface roughness of the solid slag film, the total heat flux of crystalline slag is lower than that of glassy slag, as shown in **Fig. 8**.

4.2. Optical properties

The two slag films obtained from the detector have different optical properties. The apparent reflectivity and the apparent transmittivity as a function of wavelength are shown in **Fig. 9(a)** and **Fig. 9(b)** respectively. From **Fig. 9**, it can be observed that the optical properties of the slag film changes with the wavelength from 300 nanometer to 2600 nanometer. The optical properties of the crystalline slag film change slightly with the wavelength. Unlike the crystalline slag film, both the apparent reflectivity and the apparent transmittivity

increase with the increase of the wavelength. In addition, the apparent transmittivity of the crystalline slag film is lower than that of glassy slag film, as shown in **Fig. 9(b)**.

4.3. Temperature distribution

The temperature fields of slag 1 is calculated to analyze the solidification process, as shown in **Fig. 10**. Before the heat flux reaches the maximum value, the temperature of molten slag at $x = 0$ sharply decreases with time, and the maximum cooling rate exceeds 50 K/s. After 6 s, the solidified layer began to form, and the air gap between the solidified layer and the detector increases the heat resistance of conduction. As a result, the cooling rate of slag on the detector side decreases after the total heat flux reaches the maximum, and temperature distribution at 6 s is similar to that at 10 s, as shown in **Fig. 10(a)** and **Fig. 10(b)**. After 10 s, the thickness of the solidified layer increases with time, and the exchanges of the radiative energy occur between the interface of solid-liquid layer and the inner surface of crucible. On the one hand, part of incident radiation energy arriving at the interface of two phases will be reflected, which indicates that the radiative energy through the solidified layer is decreased. On the other hand, the interface of two phases moves slowly during the latter stage of solidification, which indicates that the heat resistance of conduction changes slightly with time after 10 s. Therefore, the cooling rate of molten slag on the detector side remains stable.

4.4. Distribution of incident radiation on the detector

Both the shape of the enclosure and the optical properties of the solidified layer will influence the distribution of the radiative heat flux. The distributions of the incident radiation during solidification of slag 1 are shown in **Fig. 11(a)-(i)**. The heat flux distributions on each surface at different moment for slag 2 are shown in **Fig. 12(a)-(i)**.

From **Fig. 11** and **Fig. 12**, the radiative energy is not evenly distributed on each surface, and the value of the radiative heat flux on each zone changes with time and position during solidification of molten slag. Furthermore, the crystalline slag film has higher reflectivity and lower transmittivity. Therefore, the radiative heat flux through the crystalline slag film is lower than that of glassy slag film. As solidification continues, the thickness of solidified layer will also increase, which leads to the decrease of transmittivity. As a result, it is observed that the heat flux of the incident radiation on each surface decreases with increase of time.

For the wide surface of the detector, part of radiative energy emitted from the top of crucible transfers to environment, and these rays will not reach the detector. It can be observed from **Fig. 11(a)-(c)** and **Fig. 12(a)-(c)** that the radiative heat flux on the bottom of the wide surface is higher than that on the top of the wide surface. The distribution of heat flux on the narrow surface is different from that on the wide surface. It can be seen from **Fig. 11(d)-(f)** and **Fig. 12(d)-(f)** that the heat flux on the top side is similar to the heat flux on the bottom side. Compared with the wide surface, the narrow surface is closer to the wall of crucible, which indicates that strong reflection may occur between the narrow surface and the wall of cylinder. Consequently, the incident heat flux on the narrow surface is larger than that on the wide surface. This phenomenon is more obvious when y/n is around 0.5. The heat flux of the incident radiation is evenly distributed on the bottom surface of the detector. However, the corner of the bottom surface could receive more energy from different directions, as shown in **Fig. 11(g)-(i)** and **Fig. 12(g)-(i)**.

Besides the radiation, the conduction is also important for heat transfer during solidification. In order to investigate the main way of heat transfer during solidification, the total heat flux measured in experiment is separated into two parts, including the conductive heat flux and the radiative heat flux. **Fig. 13(a)** shows the conductive heat flux at $x = 0$ for the two slags. **Fig. 13(b)** shows the radiative heat flux at $x = 0$ for glassy slag film and crystalline slag film. The change of the conductive heat flux before 6 s is different from that of the radiative heat flux. It can be found from **Fig. 13(a)** that the conductive heat flux at $x = 0$ sharply increases with time during initial stage of solidification. After the conductive heat flux reaches the maximum, the conductive heat flux decreases slightly with time. Before 4 s, the conductive heat flux is lower than 0.45 megawatt per square meter. This value is close to that of the radiative heat flux at the same moment, which indicates that the heat transfer of solidification during initial stage is dominated by radiation. After 5 s, the radiative heat flux through the solid slag film decreases with time due to the reflection between the solid-liquid interface and the inner surface of the crucible. For the two kinds of slag films with different microstructure, the proportion of the radiative heat flux in the total heat flux is different. On the one hand, the crystallization of molten slag during cooling process may lead to the increase of the reflectivity and the decrease of the transmittivity. On the other hand, the thermal conductivity of the crystalline slag film is higher than that of glassy slag film. As a result, the Planck number (conduction-to-radiation ratio) of the crystalline slag film is higher than that of glassy slag film after 10 s, as shown in **Fig. 13(c)**. In the whole process of solidification, the conductive heat flux for glassy slag film is similar to that for the crystalline slag film. However, because the transmittivity of the crystalline slag film is lower than that of the glassy slag film, it is found from **Fig. 13(b)** that the radiative heat flux through the crystalline slag film is lower than that through the glassy slag film. After the solid slag film is formed, the proportion of the radiative heat flux in the total heat flux is lower than 30 %, which suggests that conduction is the main way of heat transfer in the latter stage of solidification.

5. Conclusions

In this work, solidification of molten slag is conducted in laboratory, and the boundary conditions are obtained

by experimental methods. Combined with the crystallization kinetics, the enthalpy method is used to solve the phase change problem of molten slag with unfixed phase change temperature. The zonal method based on the fixed grid and moving area are developed to assess the radiation effect during solidification. Some conclusions can be summarized as follows:

- (1) The effect of multiple reflections on the distribution of the radiative heat flux could be analyzed by using the ray tracing method. Compared with the ray tracing method, it is more efficient to calculate the radiative heat flux on each surface element by solving a system of linear equations established according to the energy balance between the incident radiation and the reflected radiation. When the reflection time is higher than 20, the distributions of the radiative heat flux obtained by the ray tracing method remain nearly constant.
- (2) The radiative heat flux is not distributed evenly on each surface. The calculated results indicate that the corner and edge of a surface can receive more radiation due to the multiple reflections from different directions. Results obtained by the ray tracing method indicates that the radiative heat flux increases with the increase of reflections. However, when the number of reflections is big enough, the reflection time has minor effect on the radiative heat flux.
- (3) The radiative heat flux through the crystalline slag film is lower than that through the glassy slag film, because the formation of crystals may lead to the increase of the reflectivity and the decrease of the transmittivity. In the whole process of solidification, the conductive heat flux through the glassy slag film is close to the heat flux through the crystalline slag film. The heat transfer during initial stage of solidification is dominated by radiative cooling. After 5 s, the proportion of the radiative heat flux in the total heat flux is lower than 30 %, which indicates that conduction becomes the main way of heat transfer in latter stage of solidification.

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Figure captions

Figure 1 Experimental apparatus used to measure heat flux: (a) schematic of apparatus, (b) appearance of the solid slag film, (c) cross section of the furnace.

Description: The total heat flux through solidified layer can be measured by the copper detector immersed into the molten slag, according to the temperature difference of cooling water.

Figure 2 Descriptions of grid and zones: (a) front view of the crucible, (b) top view of the crucible, (c) zones on each surface.

Description: In Figure 2, p is the index of interface of each sublayer, and v is index of each control volume. $\mathbf{q}$ is vector of radiative heat flux. dw is the length of element on wide surface in the direction of x axis. dn is the length of element on wide surface in the direction of y axis. dh is the length of element on wide surface in the direction of z axis. dA is the area of surface element, and this value changes with time.

Figure 3 Effect of number of surface elements on temperature distribution.

Description: When the number of elements is big enough, the number of surface elements has minor effect on temperature distributions for various optical properties.

Figure 4 Description of multiple reflection among three layers. (a) category of light in the three layers, (b) data structure used to describe the multiple reflection.

Description: The symbol 'g' represents the graphite crucible, and the symbol 'cu' represents the copper detector. The light in system can be divided into 12 categories. The evolution of light in system is similar to a binary tree in computer science. The value of root node is defined as the index of light leaving interface. The value of leaf node is defined as the index of light arriving at interface.

Figure 5 Radiative heat flux through the solid slag film.

Description: When the number of reflection time is higher than 20, the effect of multiple reflection on radiative heat flux is very small.

Figure 6 Results of temperature fields obtained by two different approaches.

Description: For different optical properties of slag film, the zonal method and the ray tracing method give very similar results.

Figure 7 Distribution of radiative heat flux on the inner surface of crucible: (a) bottom, zonal method, (b) inner wall, zonal method, (c) bottom, $N_c = 24$, (d) inner wall, $N_c = 1$, (e) inner wall, $N_c = 5$, (f) inner wall, $N_c = 24$.

Description: The heat flux of incident radiation on red regions is higher than heat flux on blue regions. The radiative heat flux on inner wall increases with increase of reflections.

Figure 8 The variation of total heat flux changing with time.

Description: The total heat flux first increases, and then decreases with time. At 6 s, the total heat flux reaches the maximum. After 40 s, the heat flux changes slightly with time, which indicates that the heat transfer reaches steady state.

Figure 9 Optical properties of the two slag films: (a) apparent reflectivity, (b) apparent transmittivity.

Description: The apparent reflectivity of crystalline slag film is higher than that of glassy slag film. The apparent transmittivity of crystalline slag film is lower than that of glassy slag film.

Figure 10 Temperature fields of slag 1 during solidification: (a) $t = 6$ s, (b) $t = 10$ s, (c) $t = 45$ s.

Description: The temperature of slag on the detector side is lower than that on the crucible side. The cooling rate of molten

slag on the detector side reaches maximum at about 6 s.

Figure 11 Distribution of incident radiation on the detector during solidification of slag 1 : (a) $t = 1$, wide surface, (b) $t = 10$, wide surface, (c) $t = 45$, wide surface, (d) $t = 1$, narrow surface, (e) $t = 10$, narrow surface, (f) $t = 45$, narrow surface, (g) $t = 1$, bottom, (h) $t = 10$, bottom, (i) $t = 45$, bottom.

Description: The radiative heat flux on the edge and the bottom of the wide surface is higher than that on the inner regions. Intense radiation is observed on the middle regions of narrow surfaces. For the bottom surface of the detector, the radiative heat flux on the corner is higher than that on the inner regions. Furthermore, as solidification continuous, the heat flux of incident radiation decreases.

Figure 12 Distribution of incident radiation on the detector during solidification of slag 2 : (a) $t = 1$, wide surface, (b) $t = 10$, wide surface, (c) $t = 45$, wide surface, (d) $t = 1$, narrow surface, (e) $t = 10$, narrow surface, (f) $t = 45$, narrow surface, (g) $t = 1$, bottom, (h) $t = 10$, bottom, (i) $t = 45$, bottom.

Description: The radiative heat flux on the edge and the bottom of wide surface is higher than that on the inner regions. The radiative heat flux through the glassy slag film is higher than that through the crystalline slag film. It can be observed that the radiative heat flux decreases as the solidified layer evolves.

Figure 13 Variation of conductive heat flux and radiative heat flux through slag films: (a) conductive heat flux, (b) radiative heat flux, (c) conduction to radiation ration.

Description: The conductive heat flux at $d = 0$ sharply increases with time before 4 s, and the radiative heat flux is higher than the conductive heat flux. After 5 s, the heat flux of conduction is much larger than that of radiation, which indicates that conduction is the main way of heat transfer in latter stage.

Figures

Fig.1. Experimental apparatus used to measure heat flux: (a) schematic of apparatus, (b) appearance of the solid slag film, (c) cross section of the furnace.

Fig.2. Descriptions of grid and zones: (a) front view of the crucible, (b) top view of the crucible, (c) zones on each surface.

Fig.3. Effect of number of surface elements on temperature distribution.

Fig.4. Description of multiple reflection among three layers. (a) category of light in the three layers, (b) data structure used to describe the multiple reflection.

Fig.5. Radiative heat flux through the solid slag film.

Fig.6. Results of temperature fields obtained by two different approaches.

Fig.7. Distribution of radiative heat flux on the inner surface of crucible: (a) bottom, zonal method, (b) inner wall, zonal method, (c) bottom, $N_c = 24$, (d) inner wall, $N_c = 1$, (e) inner wall, $N_c = 5$, (f) inner wall, $N_c = 24$.

Fig.8. The variation of total heat flux changing with time.

Fig.9. Optical properties of the two slag films: (a) apparent reflectivity, (b) apparent transmittivity.

Fig.10. Temperature fields of slag 1 during solidification: (a) $t = 6$ s, (b) $t = 10$ s, (c) $t = 45$ s.

Fig.11. Distribution of incident radiation on the detector during solidification of slag 1 : (a) $t = 1$, wide surface, (b) $t = 10$, wide surface, (c) $t = 45$, wide surface, (d) $t = 1$, narrow surface, (e) $t = 10$, narrow surface, (f) $t = 45$, narrow surface, (g) $t = 1$, bottom, (h) $t = 10$, bottom, (i) $t = 45$, bottom.

Fig.12. Distribution of incident radiation on the detector during solidification of slag 2 : (a) $t = 1$, wide surface, (b) $t = 10$, wide surface, (c) $t = 45$, wide surface, (d) $t = 1$, narrow surface, (e) $t = 10$, narrow surface, (f) $t = 45$, narrow surface, (g) $t = 1$, bottom, (h) $t = 10$, bottom, (i) $t = 45$, bottom.

Fig. 13. Variation of conductive heat flux and radiative heat flux through slag films: (a) conductive heat flux, (b) radiative heat flux, (c) conduction to radiation ratio.

Tables

Table 1 Compositions of slags used in experiment

NO.	SiO ₂	CaO	F	Na ₂ O	Al ₂ O ₃	MgO	Li ₂ O	Total
1	13.8	41.2	9.0	10.0	24.0	1.0	1.0	100.0
2	36.0	40.0	8.0	8.0	7.0	1.0	0.0	100.0

Table 2 Values of a_x , b_x , a_y , b_y , a_z and corresponding range of i

position	a_x	b_x	a_y	b_y	a_z	i
west	-1	0	-1	$(i-150)\%5+0.5$	$(i-150)\%5+0.5$	$150 \leq i < 175$
south	-1	$(i-175)\%7+0.5$	-1	0	$(i-175)\%7+0.5$	$175 \leq i < 210$
east	1	0	-1	$(i-210)\%5+0.5$	$(i-210)\%5+0.5$	$210 \leq i < 235$
north	-1	$(i-235)\%7+0.5$	1	0	$(i-235)\%7+0.5$	$235 \leq i < 270$
bottom	-1	$(i-270)\%7+0.5$	-1	$(i-270)\%7+0.5$	0	$270 \leq i < 305$

Table 3 Values of parameters used in calculation

Emissivity	copper detector	0.4
	graphite crucible	0.8
Thickness of solid slag film (10^{-3} m)	slag 1	2.2
	slag 2	2.4
Fluid temperature (K)	slag 1	1451
	slag 2	1461
Thermal conductivity[27,28] ($W \cdot (m \cdot K)^{-1}$)	$1523 < T < 1673$	0.50
	$1523 < T < 1273$	0.80
	$T < 1273$	1.65
specific heat capacity C_{ps} ($J \cdot (g \cdot K)^{-1}$)	slag 1	$5.125 - 0.00749T + 3.4375 \times 10^{-6}T^2$
	slag 2	$2.937 - 0.004T + 1.875 \times 10^{-6}T^2$
Latent heat ($J \cdot g^{-1}$)	slag 1	265

Table 4 Data in Array A1

root node	1	2	3	4	5	6	7	8	9	10	11	12
leaf node 1	3	0	5	1	7	11	0	9	11	7	1	5
leaf node 2	4	0	6	2	8	12	0	10	12	8	2	6

Table 5 Data in Array A2

index	1	2	3	4	5	6
q_{Nc+1}/q_{Nc}	$\varphi_{2,1}(1-\varepsilon_g)$	$\varphi_{2,1}\varepsilon_g$	$\varphi_{1,2}(1-R_s)$	$\varphi_{1,2}R_s$	$T_s(1-R_s)$	T_sR_s
index	7	8	9	10	11	12
q_{Nc+1}/q_{Nc}	ε_{cu}	$1-\varepsilon_{cu}$	$1-R_s$	R_s	$T_s(1-R_s)$	T_sR_s

Table 6 Comparison of results between the ray tracing method and the zonal method

		Ray tracing method					Zonal method
Reflection times Nc		8	12	16	18	24	
$q_{rad,0}$ 10^6W/m^2	$Ra = 0.1, Ta = 0.6$	0.2858	0.2862	0.2865	0.2866	0.2870	0.3021
	$Ra = 0.3, Ta = 0.4$	0.2219	0.2232	0.2235	0.2236	0.2240	0.2441
	$Ra = 0.5, Ta = 0.2$	0.1152	0.1324	0.1323	0.1328	0.1331	0.1452
CPU time (s)		299	459	616	695	931	20