

Determination of Interfacial Heat Flux of Stainless Steel Solidification on Copper Substrate During the First 0.2 s

ZHANG Wei¹ (张 卫), YU Yan² (于 艳), FANG Yuan² (方 园), LI Jian-guo^{1*} (李建国)
(1. School of Materials Science and Engineering, Shanghai Jiaotong University, Shanghai 200240, China;
2. Baosteel Research Institute, Baosteel Co. Ltd, Shanghai 201900, China)

© Shanghai Jiaotong University and Springer-Verlag Berlin Heidelberg 2011

Abstract: Interfacial heat transfer is a key issue in many solidification processes. In the paper, a novel experimental apparatus has been designed and on this basis, the instantaneous interfacial heat transfer between molten steel or solidified shell and copper substrate during the first 0.2 s has been studied. The investigated parameters include melt superheat, substrate temperature and surface roughness. The results show that the peak value of the interfacial heat flux in the first stage of liquid/solid contact increases with melt superheat and changes slightly with substrate temperature and surface roughness. The interfacial heat flux in the stage of solid/solid contact has a similar trend of slow decrease in most conditions.

Key words: rapid solidification, interfacial heat transfer, heat flux

CLC number: TF 777.7 **Document code:** A

0 Introduction

It is self-evident that the interfacial heat transfer between molten metal and molds in the solidification processes is very important to casting. Most of the previous work^[1-3] concentrated on the determination of interfacial heat transfer between casting and metal or sand molds in quite a long time, for example, from seconds to minutes. However, in many rapid or near rapid solidification processes, such as strip casting, the total solidification time is less than 0.5 s. Recently, studies on the interfacial heat transfer of initial solidification in fragments of second have been paid more attentions, but the materials most used in experiments are low melting point metals and alloys, such as aluminum alloy. For the high melting point alloys with high commercial value such as steel, studies on instantaneous interfacial heat transfer in solidification have not been intensively carried out.

The solidification process of melt on metal substrate surface can be divided into two stages: liquid/solid contact and solid/solid (S/S) contact. In the first contact stage, a low interfacial thermal resistance and a high interfacial heat flux occur due to the better wettability condition between liquid and solid. In the S/S contact stage, the thermal resistance increases and the heat flux decreases owing to air gaps formed between the solidified shell and the substrate.

Accurate determination of instantaneous interfacial heat transfer can not only expand our understanding on initial solidification, but also provide boundary conditions for numerical simulation. However, there are many difficulties in this problem. The high melt temperature results in no proper thermocouples which can be placed in the melt; the short solidification time requires quick response time of the temperature measurement parts; the high value and the great change of the heat flux result in a great temperature change in the substrate. It requires proper design of the substrate and the temperature measurement to insure good resolving power for the initial interfacial heat transfer.

Various apparatuses and simulators have been developed to study this problem. In these experiments, the droplet solidification method was used most widely^[4-8]. In a typical experimental process, a melt droplet was molten and dripped onto a metal substrate surface, and then solidified to an ingot. During this period, the temperature histories of some positions in the substrate, which were used in the calculation of the interfacial heat flux or the interfacial heat transfer coefficient by solving an inverse heat conduction problem (IHCP), were determined by the embedded thermocouples. Wang and Matthys^[9] developed a set of improved droplet apparatus. In their experiment processes, the melt droplet was dropt onto a gradient substrate surface and solidified quickly to a thin strip. The interfacial heat transfer coefficient was calculated from the strip up-surface temperature measured by a pyrometer.

Strezov et al.^[10-11] carried out excellent work on the instantaneous interfacial heat transfer. In their experiments, a dipping device was designed and the heat flux in the first 50 ms was focused. During the experimental process, a copper mould as the substrate was driven to bathe into liquid steel and the temperature of substrate surface was recorded by the embedded thermocouples. The interfacial heat flux with millisecond resolution was calculated by solving an inverse heat conduction problem.

To measure the interfacial heat transfer in the first solidification stage more accurately, a novel dipping experiment is designed and the interfacial heat flux during the first 0.2 s is predominantly investigated in this paper. The effects of melt superheat, temperature and surface roughness of substrate are focused.

1 Experimental

1.1 Apparatus and Process

The schematic of the experimental apparatus is shown in Fig. 1. It is composed of an induction furnace, a substrate driver, a data collector and a computer. The substrate, on which the melt solidifies, is fixed on the driver and dives into melt in vertical direction, in order to simulate the solidification process of strip casting and wipe off the gravity effect. The speed of the substrate driver can be adjusted from 0.1 to 1 m/s. During the experimental process, a material sample of 2 kg is heated and melted by the induction furnace at a nitrogen atmosphere. The melt temperature is measured by S-type thermocouples protected by a quartz tube. When the melt reaches the preset temperature, the substrate is driven to dive into the molten steel, keeping a short time and then returning to its original position. During this period, the temperature data of the substrate are determined by the thermocouples and transformed automatically to temperature measurements by the data collector and stored in the computer. The data collection frequency is 1 kHz. The temperature data must be filtered to wipe off noise before storing.

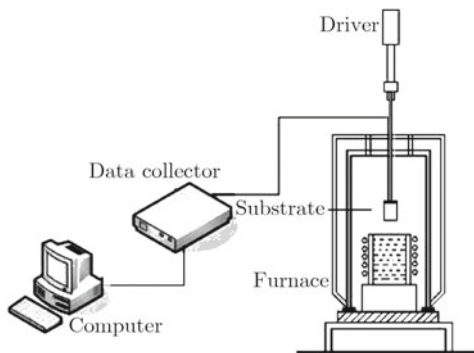


Fig. 1 Schematic of the apparatus

In the measurement of interfacial heat transfer, the key issues are the design of the substrate and the assembly of the thermocouples. A hole-drilling design is widely used in many researchers' devices mentioned above. This sort of design would bring many problems, for instance, the influence on the response speed of the thermocouples. In the present apparatus, a novel design is used. The substrate consists of two copper plates, whose size is 40 mm×40 mm×1.5 mm, as shown in Fig. 2. One or more 0.25 mm K-type thermocouples are welded on the center position of the copper plate inner surface. Finally, two copper plates are assembled symmetrically and fixed on the driver. Compared with traditional devices, the substrate design in this paper has the following advantages: ① its structure is simple, so the reliability of the measurement can be enhanced; ② without drilling in the substrate, the substrate temperature field is not disturbed; ③ welding on the substrate surface can ensure a rapid response speed of the thermocouples; ④ due to the substrate size, the heat transfer can be treated as a one-dimension model; ⑤ because of the symmetrical assembly, the boundary condition of the substrate inner surface is strictly adiabatic, which can benefit the interfacial heat flux calculation.

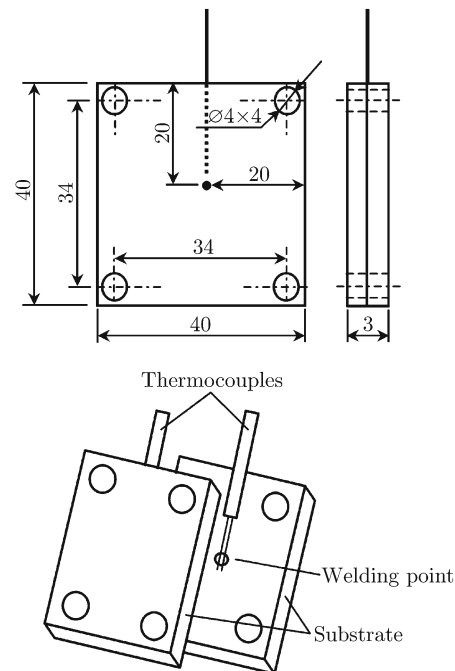


Fig. 2 Schematic of the substrate (mm)

During the experiment processes, the melt and the substrate temperatures are set to 1460 °C and room temperature respectively unless specified. The surface roughness is made by sand papers and determined by a roughness tester. In the studies of the effects of melt and substrate temperatures, the surface roughness R_a is made to about 2.0 μm by 1200# sand paper. The driver

speed is 0.6 m/s and the time of the substrate keeping in the melt is 0.3 ms in all cases. The experiment material is AISI304 stainless steel and the composition is shown in Table 1. Two or more experiments are carried out to ensure the reliability of the results in each experimental condition.

Table 1 Composition of experiment material

Sample	w/%	Sample	w/%
C	0.006	S	0.012
Si	0.63	Cr	17.08
Mn	0.70	Ni	8.36
P	0.024	Fe	Balance

1.2 Heat Flux Calculation

For the substrate, the size in the thickness direction is quite smaller than the other two directions. Thus the substrate temperature θ can be obtained by solving a one-dimension heat transfer model. The thermal conduction equation and the boundary conditions can be written as

$$\rho c \frac{\partial \theta}{\partial t} = \lambda \frac{\partial^2 \theta}{\partial x^2}, \quad (1)$$

$$\lambda \frac{\partial \theta}{\partial x} \Big|_{x=0} = q, \quad (2)$$

$$\lambda \frac{\partial \theta}{\partial x} \Big|_{x=d} = 0, \quad (3)$$

$$\theta_{x=d} = \theta_s, \quad (4)$$

where, ρ , c and λ are the density, heat capacity and thermal conductivity of the copper substrate, respectively; x is position, θ_x is the temperature at the x position; q is the interfacial heat flux between the melt and the substrate; d is the thickness of copper plates; and θ_s is the temperature of the substrate inner surface. Since the temperature of the substrate inner surface is known and the interfacial heat flux needs to be solved, this problem is a typical inverse heat conduction problem (IHCP). For IHCP, Beck's nonlinear estimation method was used widely^[12], of course no exception in this paper. A brief description is given as follows.

Define the sensitivity coefficient Z as

$$Z(x, t) \equiv \partial \theta(x, t) / \partial q(t). \quad (5)$$

Assume that the heat flux in an individual time interval τ is a constant, thus the q_τ in this interval can be written as

$$q_\tau^n = q_\tau^{n-1} + (\theta_x^m - \theta_x^{n-1}) / Z_{x,\tau}, \quad (6)$$

where n is the iteration number, $Z_{x,\tau}$ is the sensitivity coefficient in x position and τ time, and θ_x^m is the measured temperature at the x position. The calculated temperature θ_x^{n-1} can be gotten by solving the thermal

conduction equation with q_τ^{n-1} as the boundary condition. Equation (6) is used iteratively until the following convergence condition is met:

$$(q_\tau^n - q_\tau^{n-1}) / q_\tau^{n-1} < 0.005. \quad (7)$$

2 Results and Discussion

2.1 Typical Interfacial Heat Flux

Figure 3 shows a typical temperature history of the copper plate inner surface and the corresponding interfacial heat flux between the melt or the solidified shell and the substrate during the melt cooling and solidification (melt temperature $\theta_m = 1460^\circ\text{C}$, substrate initial temperature: $\theta_i = 20^\circ\text{C}$, surface roughness $R_a = 2.0\ \mu\text{m}$). We can see that, once the substrate in room temperature (20°C) contacts with the liquid steel, its inner surface temperature increases quickly and subsequently an obvious inflexion occurs in the temperature history at about 20 ms after the contact beginning. At 0.2s, the temperature reaches about 300°C . For the heat flux curve, the heat transfer behavior during the solidification process shows more evidently. In the first about 100 ms, an obvious peak occurs in the heat flux curve and the maximum value is about $16\ \text{MW}/\text{m}^2$. Keeping for a very short time, the heat flux decreases rapidly to a slow change stage, waving from 6 to $4\ \text{MW}/\text{m}^2$. Comparing with the solidification process, the first stage with high heat flux can be considered as the liquid/solid contact and the following as the solidified shell developing stage. Verified by experimental results, the interfacial heat flux measurement in the initial solidification stage is achieved by

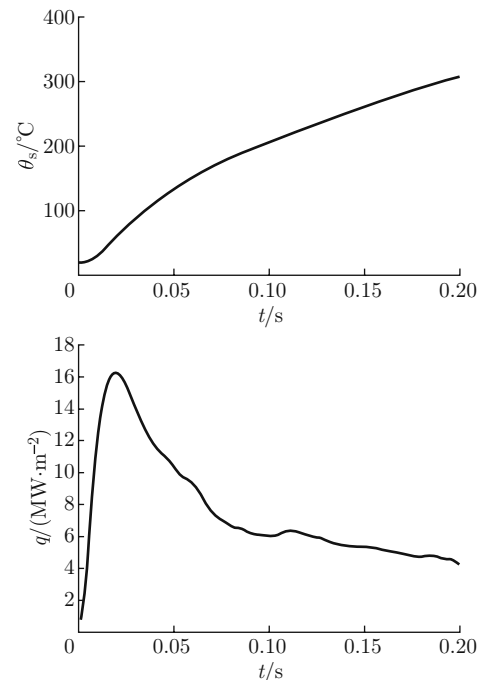


Fig. 3 Typical temperature and interfacial heat flux history results

using the novel apparatus, whose resolution can reach millisecond level.

2.2 Effect of Melt Temperature

Figure 4 shows the effects of melt superheat on the substrate temperature and heat flux histories. From Fig. 4(a) we can see, with different superheat, the temperature histories have a similar shape. The different is that the temperature histories are more gradient in higher superheat. At 0.2 s after the solidification beginning, the temperature of the substrate in 1590 °C melt can reach about 600 °C. In Fig. 4(b), the relation between the peak heat flux and the superheat shows more clearly. The peak heat flux increases rapidly with the superheat increase, but the heat flux in the shell developing stage does not change evidently with superheat. The curves are slightly higher in high superheat. Figure 5 shows the effect of melt temperature on the peak heat flux. The maximum of heat flux can reach 70 MW/m² when the melt temperature is 1590 °C. Similar results that interfacial heat flux increases with superheat have been reported in Refs. [4, 13] except Ref. [10]. In their experiment the peak heat flux decreases with superheat increase.

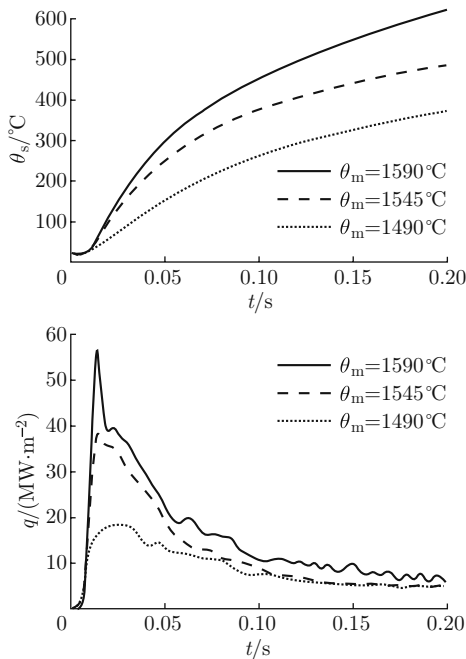


Fig. 4 Effects of melt superheat on the substrate temperature and heat flux histories

The interfacial heat transfer is dramatically affected by the wettability between melt and substrate. In general, melt surface tension decreases with melt superheat increase. The decrease of surface tension will make a better wettability between solid and liquid. In addition, the melt superheat increases the temperature difference between melt and substrate. Therefore, the superheat increase increases the peak heat flux.

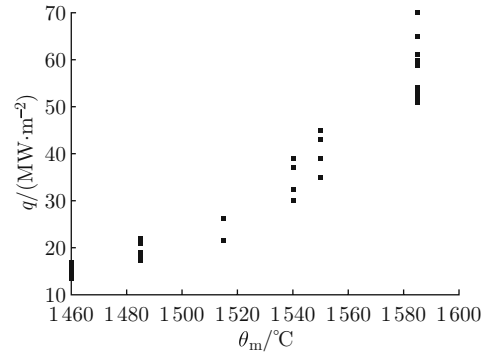


Fig. 5 Effect of melt temperature on the peak heat flux

2.3 Effect of Surface Roughness

Figure 6 shows the substrate inner surface temperature and the corresponding heat flux histories with different substrate surface roughnesses. Figure 7 shows the effect of surface roughness on the peak heat flux. From these figures we can see that surface roughness has no significant impact on the peak heat flux, except the polished surface. On very smooth substrate surface, the peak heat flux has a higher value than that on rough surface. The similar results have been reported in some researchers' works^[4-5,9]. The interfacial heat flux in the second stage (solidified shell developing stage) is almost the same. The possible reasons for the effect of surface roughness on interfacial heat flux can be explained as follows. Surface roughness increases the true surface area and rough surface produces more gaps between melt and substrate in the first contact stage, due

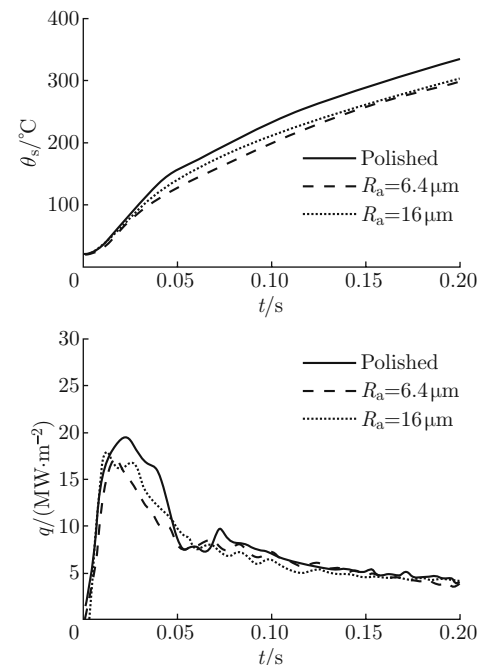


Fig. 6 Surface temperature and heat flux histories with different substrate surface roughnesses

to the melt surface tension and the gas in the micro-valley. Therefore surface roughness has no significant effect on the interfacial heat transfer.

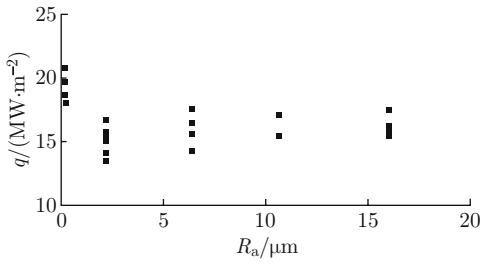


Fig. 7 Effect of surface roughness on the peak heat flux

2.4 Effect of Substrate Initial Temperature

Figure 8 shows the substrate inner surface temperature and the corresponding heat flux histories on the substrate with different initial temperatures. Figure 9 shows the effect of substrate initial temperature on the peak heat flux. It can be seen that the peak heat flux increases first and then decreases with the substrate temperature increase. The maximum peak value, about 28 MW/m², appears when the substrate temperature is 160 °C. Like surface roughness, substrate initial temperature has no evident influence on the heat flux in the second stage. Actually, the wetting process of liquid on solid surface is a process of liquid displacing the adsorbed gas on the solid surface. It is well known, when substrate temperature rises, the ad-

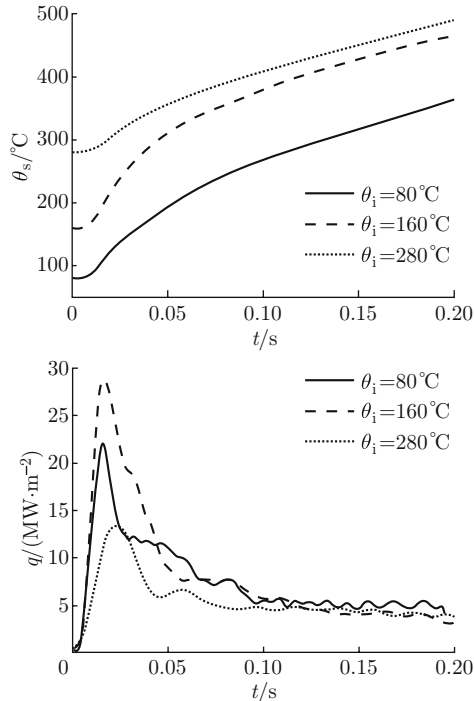


Fig. 8 Surface temperature and heat flux histories on the substrate with different initial temperatures

sorbed gas decreases. Therefore the displacing process is easy and the peak heat flux increases with the substrate temperature increase. However, when the initial temperature reaches 280 °C, the peak heat flux has an obvious decrease. The possible reason is that an oxide layer appears on the substrate surface in high temperature. Fukumoto et al.^[14] in their experiments found that on the substrate with lower temperature, gas holes appeared in the solidified shell surface, which indicated the effect of the adsorbed gas on the melt solidification.

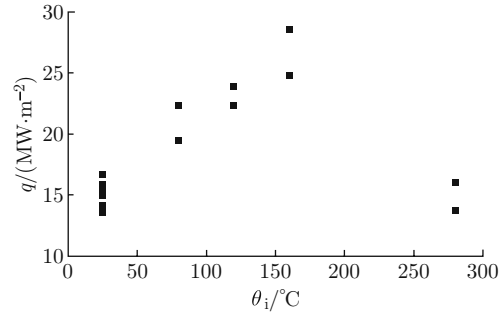


Fig. 9 Effect of substrate initial temperature on the peak heat flux

2.5 Model of Interfacial Heat Transfer

The thermal resistance between melt and substrate can be written as

$$\frac{1}{R} = \frac{1}{R_{cond}} + \frac{1}{R_{conv}} + \frac{1}{R_{rad}}, \quad (8)$$

where R is the total thermal resistance, R_{cond} is the thermal resistance of conduction, R_{conv} is the thermal resistance of convection, and R_{rad} is the thermal resistance of radiation. Convection and radiation work mainly through gap between melt and substrate while conduction works mainly through the direct contact. In general, R_{cond} is far lower than the thermal resistance of convection and radiation. Therefore, the greater the contact area between melt and substrate is, the lower total thermal resistance will exist. In order to study the relationship between the actual contact area and the various parameters, a simple heat transfer model is built. The model is based on the most universal situation of the contact between melt and substrate. The actual substrate surface is simplified as a sawtooth texture, as shown in Fig. 10.

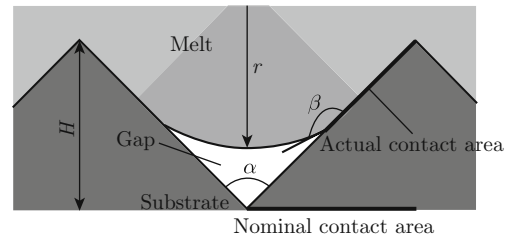


Fig. 10 Model of the melt contacting with the substrate

From the geometric relationship in Fig. 10, a relation between the actual contact area and the nominal contact area can be obtained by

$$S_a/S_0 = \frac{1}{\sin \frac{\alpha}{2}} + \frac{\cos \frac{\alpha}{2} \cos \left(\beta - \frac{\alpha}{2} \right) r}{\sin^2 \frac{\alpha}{2} H}, \quad (9)$$

where S_a is the actual contact area, S_0 is the nominal contact area, α is the valley angle as shown in Fig. 10, β is wetting angle between the melt and the substrate, H is the ridge height meaning the surface roughness, and r is the radius of curving surface of the pressed liquid steel. The radius of curving surface of the pressed liquid steel is determined by the melt surface tension σ_{LG} and the static pressure p according to Laplace equation:

$$r = \sigma_{LG}/p. \quad (10)$$

From Eq. (9) we can find that wetting angle is a sensitive factor to the actual contact area. As the wetting angle decreases, the actual contact area increases dramatically. This is the reason for the effect of melt superheat and substrate temperature on the interfacial heat flux. The change of ridge height has no notable effect on the actual contact area in a certain range. Therefore, the effect of the surface roughness on the interfacial heat transfer is not as good as the effects of the melt and substrate temperatures.

3 Conclusion

An experimental simulator has been developed for studying the instantaneous interfacial heat transfer between the solidifying stainless steel and the copper substrate in the first 0.2 s. Verified by experiments, the design for interfacial heat flux measurement in this paper is successful, especially within the first stage of solidification. Its resolution can reach millisecond level. Peak heat flux increases with melt superheat increase. No significant relation between peak heat flux and surface roughness has been observed, except the higher peak heat flux on the polished substrate surface. Peak heat flux increases first and then decreases with the substrate temperature increase.

Acknowledgement The authors wish to acknowledge the help of LIANG Gao-fei and WANG Cheng-quan and the financial aid from Baosteel.

References

- [1] FERREIRA I, SPINELLI J E, PIRES J C, et al. The effect of melt temperature profile on the transient metal/mold heat transfer coefficient during solidification [J]. *Materials Science and Engineering A*, 2005, **408**(1-2): 317-325.
- [2] LEE J, KIM H S, WON C W, et al. Effect of the gap distance on the cooling behavior and the microstructure of indirect squeeze cast and gravity die cast 5083 wrought Al alloy [J]. *Materials Science and Engineering A*, 2002, **338**(1-2): 182-190.
- [3] NARAYAN P K, RAVISHANKAR B. Effect of modification melt treatment on casting/chill interfacial heat transfer and electrical conductivity of Al-13% Si alloy [J]. *Materials Science and Engineering A*, 2003, **360**(1-2): 293-298.
- [4] LOULOU T, ARTYUKHIN E, BARDON J. Estimation of thermal contact resistance during the first stages of metal solidification process. II. Experimental setup and results [J]. *International Journal of Heat and Mass Transfer*, 1999, **42**(12): 2129-2142.
- [5] BOUCHARD D, LEBOEUF S, NADEAU J P, et al. Dynamic wetting and heat transfer at the initiation of aluminum solidification on copper substrates [J]. *Journal of Materials Science*, 2009, **44**(8): 1923-1933.
- [6] NOLLI P, CRAMB A. Interaction between iron droplets and H₂S during solidification: Effects on heat transfer, surface tension and composition [J]. *ISIJ International*, 2007, **47**(9): 1284-1293.
- [7] YU Y, CRAMB A W, HEARD R, et al. The effect of oxygen partial pressure on heat transfer and solidification [J]. *ISIJ International*, 2006, **46**(10): 1427-1431.
- [8] TODOROKI H, PHINICHKA N. Heat transfer behavior of molten iron and nickel during the first 0.2 seconds of solidification [J]. *ISIJ International*, 2009, **49**(9): 1347-1355.
- [9] WANG G, MATTHYS E. Experimental determination of the interfacial heat transfer during cooling and solidification of molten metal droplets impacting on a metallic substrate: Effect of roughness and superheat [J]. *International Journal of Heat and Mass Transfer*, 2002, **45**(25): 4967-4981.
- [10] STREZOV L, HERBERTSON J. Experimental studies of interfacial heat transfer and initial solidification pertinent to strip casting [J]. *ISIJ International*, 1998, **38**(9): 959-966.
- [11] STREZOV L, HERBERTSON J, BELTON G. Mechanisms of initial melt/substrate heat transfer pertinent to strip casting [J]. *Metallurgical and Materials Transactions B*, 2000, **31**(5): 1023-1030.
- [12] BECK J, WOODBURY K. Inverse problems and parameter estimation: Integration of measurements and analysis [J]. *Measurement Science and Technology*, 1998, **9**: 839-847.
- [13] NETTO P, TAVARES R P, ISAC M, et al. A technique for the evaluation of instantaneous heat fluxes for the horizontal strip casting of aluminum alloys [J]. *ISIJ International*, 2001, **41**(11): 1340-1349.
- [14] FUKUMOTO M, NISHIOKA E, MATSUBARA T. Flattening and solidification behavior of a metal droplet on a flat substrate surface held at various temperatures [J]. *Surface and Coatings Technology*, 1999, **120**: 131-137.