Analyses of the melt cooling rate in the melt-spinning process

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Received 19.02.2012; published in revised form 01.04.2012

ABSTRACT

Purpose: Rapid solidification (RS) of metallic melts is important for the development of the advance metallic materials, because enables the production of new alloys with superior properties according to conventionally treated alloys. In practice it turned out, that single roll melt spinning process has one of the highest melt cooling rates among all continuous casting processes. But, because very short solidification time and movement of the melt and substrate, melt cooling rate is very difficult to measure with confidence. Primary goal of our work was to determine the limits of cooling rate over the ribbon thickness and to outline, which property or typical feature of the process has the greatest influence on cooling rate of the melt.

Design/methodology/approach: On the basis of developed mathematical model, a computer program was made and used for melt cooling rate calculation in the melt-spinning process.

Findings: The calculations show that distance from the contact surface in relation to the thermal properties of the melt, chilling wheel material and contact resistance between metal melt and chilling wheel have the greatest influence on melt/ribbon cooling rate. In the case of continuous casting, significant “long term” surface temperature increase may take place, if the wheel is not internally cooled.

Research limitations/implications: Influence of the melt physical properties, chill wheel material, contact resistance and cooling mode of the chill wheel on melt cooling rate are outlined.

Practical implications: Practical limits of melt cooling rate over ribbon thickness are outlined and directions for the chill wheel cooling system design are indicated.

Originality/value: Comparison between cooling rates calculated at various thermal resistance assumptions of particular constituents is outlined. New method for determining contact resistance through variable heat transfer coefficient is introduced which takes into account physical properties of the casting material, process parameters and contact time/length between metal melt/ribbon and substrate and enables cooling rate prediction before the experiment execution. In the case of continuous casting, heat balance of the melt-spinning process is calculated and influence of the chill wheel cooling mode on cooling rate of metallic ribbon is analyzed.

Keywords: Modelling; Rapid solidification; Metallic materials; Heat transfer; Heat transfer coefficient

1. Introduction

Rapid solidification (RS) of metallic melts is important for the development of the advance metallic materials, because enables the production of new alloys with superior properties according to conventionally treated alloys [1-3]. What is the speed of solidification, which divides between classical procedures of casting and rapid solidification, is not exactly
specification. Differences in interpretation refer to the development of final microstructure, but the characteristic of all rapid solidification techniques is a final non-equilibrium state of the microstructure [4,5], which can be seen as: refined grains, extended solubility in metal matrix, quantitative and qualitative change of microstructural constituents, reduced micro segregation, etc.

In general, rapid solidification of melts could be achieved by two approaches:

- with high undercooling of the melt prior solidification, by reduction or even elimination of heterogeneous nucleation sites in so called containerless processes (atomization, droplet emulsion technique, drop tube technique, magnetic levitation, etc.),
- with rapid cooling to bypass heterogeneous nucleation kinetically (melt-spinning, splat quenching, remelting of surface layer, etc.) [6-8]

Single roll melt spinning (Figure 1) is one of the most commonly used processes for rapid solidification. Its major advantage is the possibility of continuous production of rapidly solidified material in the form of thin ribbons or foils, even on large industrial scale [9-11]. In this technique, a thin stream of melt is directed onto a circumferential surface of a rotating wheel, where so called melt puddle forms (Figure 2).

Ribbon is then dragged out from the puddle by relative motion of the wheel. Microstructure of the ribbon can be completely crystalline, amorphous or combined and depends upon the contact resistance between the melt and substrate, latent heat of crystallization of casting material, heat transfer in the melt and the wheel, and nucleation and crystal growth characteristics of the particular casting material [13,14].

### 2. Heat transfer calculation

For calculation of temperature distribution inside the melt puddle and chill wheel, we used explicit finite difference method (FDM) with cylindrical coordinate system [15-17]. Thermal properties of the melt and wheel material (λ(Τ), c(T)) are temperature dependent and calculated for each iteration step with linear interpolation from tabulated values of particular casting material. Density in solidification interval of the casting material is changing linearly, whereas density of the wheel material is approximated as constant. Because melt puddle is thin compare to its width and length we made an assumption of two-dimensional (2D) transient heat transfer. A schematic diagram of the idealized melt puddle geometry used in mathematical model is shown in Figure 2. Assuming 2D transient heat transfer with variable thermal properties and internal heat generation (latent heat of crystallization), general partial differential equation for the melt is reduced to:

\[
\frac{1}{r} \left( \lambda \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial r} \left( \lambda \frac{\partial T}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial \varphi} \left( \lambda \frac{\partial T}{\partial \varphi} \right) + q = \rho \cdot c \cdot \frac{\partial T}{\partial t} \tag{1}
\]

And for chill wheel, where no latent heat is released:

\[
\frac{1}{r} \left( \lambda \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial r} \left( \lambda \frac{\partial T}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial \varphi} \left( \lambda \frac{\partial T}{\partial \varphi} \right) = \rho \cdot c \cdot \frac{\partial T}{\partial t} \tag{2}
\]

where \(r, \varphi\) represent spatial coordinates in cylindrical coordinate system [m; rad], \(T\) temperature [K], \(\lambda\) density [kgm\(^{-3}\)], \(c\) thermal conductivity [Wm\(^{-1}\)K\(^{-1}\)], \(\varphi\) specific heat of the melt or chill wheel material [Jkg\(^{-1}\)K\(^{-1}\)], and \(q\) volumetric heat generation rate [Wm\(^{-3}\)].

![Fig. 1. Laboratory melt-spinning apparatus - Faculty of Natural Science and Engineering, University of Ljubljana (left), Componets assembly in the vacuum chamber (right)]

![Fig. 2. Close-up speed frame camera snap shot of the melt puddle (tin casting) on the circumferential surface of the chill wheel, free jet melt-spinning [12]]

![Fig. 3. Idealized geometry of the melt puddle used in mathematical model]
Ribbon thickness is assumed to be proportional to circumferential velocity of the chill wheel \( (u_w) \) and the pressure in the crucible \( (p) \), as predicted by continuity and Bernoulli equations. Temperature of the melt in the puddle direct under the impinging jet stays equal to casting temperature, because strong turbulences in that region. To ascertain the influence of the casting material thermal properties, chill wheel material, inner and external cooling of the chilling wheel, and contact resistance on cooling rate of the ribbon, comparison between calculated cooling rates at various assumptions was made: ideal isothermal wheel versus copper wheel, influence of various contact resistance, and influence of solidified ribbon thickness [18].

3. Results and discussion

Figure 4 represents calculated cooling curves of contact and free surface of cast ribbons in the case of an assumption of ideal isothermal cooling wheel and Figure 5 calculated cooling curves as a function of the distance from the contact surface.

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because real materials can’t absorb so much energy without heating up significantly.

Fig. 7. Cooling rate as a function of a distance from the contact surface (Copper wheel, ideal contact)

Among thermal properties of the melt, volumetric heat of crystallization have major influence on melt cooling rate across the transverse cross-section of the cast ribbon. Figure 8 represent contact and free surface cooling curves of Al ribbon in dependence of the amount of the released crystallization heat. If we assume 50% lower crystallization heat, solidification time for free surface of 60 µm thick ribbon will decrease for 30%, and when amorphous solidification is presumed, 50% shorter is the time of solidification.

In situations where a detailed description of thermal physics is very complicated, such as in melt-spinning process, combined modes of heat exchange are usually taken in to account with the overall heat transfer coefficient (a), which includes conduction and radiation of heat, as well as any convective effects. Ideal contact between the melt and wheel surface is not physically realistic, because there are no metallic atomic bonds between atoms of the melt and atoms of the surface of the wheel. By applying different values of heat transfer coefficient into calculations of heat transfer between melt and copper wheel surface, influence of contact resistance on cooling rate (Figure 9) could be estimated. In comparison to ideal contact (Figures 6, 7), cooling rate of the ribbon and cooling rate as a function of a distance from the contact surface (Figure 9) are relatively much lower when some contact resistance is considered, although the contact resistance value is very low (10⁻⁶ (m²·K)/W). Value for constant contact resistance for aluminum was obtained with subsequent microstructure analyses and reported by other authors [19]. Integral method calculation of the heat transfer coefficient gives the most logical results for entire duration of the contact. Calculated solidification time is practically the same to those obtained by overall (constant) heat transfer coefficient, but final temperature of the ribbon at the detachment point is much greater, especially for longer contact time. Constant contact resistance approximation (10⁻⁹ (m²·K)/W) for longer contact time predicts even lower ribbon temperature then ideal contact calculation, which is physically unlikely. The local heat transfer coefficient a(x) is calculated with the integral method for liquid metals flow over flat plate [18,20]. The equation for local heat transfer coefficient a(x) calculation, included in the numerical scheme is:

\[
\alpha(x) = \frac{3 \cdot \lambda}{2 \cdot \sqrt{8}} \cdot \frac{u_w}{a \cdot x}
\]

where \(u_w\) represent circumferential velocity of the wheel [ms⁻¹], \(\lambda\) thermal conductivity of the casting material [Wm⁻¹K⁻¹], \(a\) temperature diffusivity of the casting material [m²s⁻¹], and \(x\) distance from the initial contact point to the actual calculation point [m].

Fig. 8. Hypothetical calculated cooling curves of free and contact surface of Al ribbon as a function of different assumptions of latent heat of crystallization (ribbon thickness 60 µm, contact time 0.923 msec, ideal contact)

Fig. 9. Cooling curves of free and contact surface of Al ribbon and contact surface of the copper wheel as a function of different contact resistance (ribbon thickness 60 µm, contact time 0.923 msec)
Among thermal properties of the melt, volumetric heat of fusion is a key factor in determining the solidification rate. During the melt-spinning process, very complicated heat transfer processes occur, such as in the melt-spinning process, where the ribbon undergoes a rapid solidification. The latent heat of crystallization plays a significant role in this process, with a ribbon thickness of 60 µm and a contact time of 0.923 msec, considered ideal for achieving high cooling rates.

Figure 8 represents the calculated cooling curves for free and contact surfaces of an Al ribbon in the transverse cross-section of the cast ribbon. For the free surface, when amorphous solidification is presumed, the solidification time is 50% shorter than for the contact surface. This difference is due to the lower crystallization heat and the solidification rate for the contact surface, which is influenced by the contact resistance.

Figure 9 shows the cooling curves for the free and contact surfaces of an Al ribbon. It is observed that, even for longer contact times, the cooling rate decreases, with contact resistance values being very low (10^-6 m²K/W). The local heat transfer coefficient, which includes conduction, radiation, and convection, is calculated using the integral method for liquid metals flow. The circumferential velocity of the wheel is an essential factor in determining the cooling rate.

Figure 10 illustrates the cooling rate as a function of the distance from the contact surface for different cast materials. The integral method is used for these calculations. The local heat transfer coefficient, which is physically unlikely, is calculated using this approach. The contact resistance value is very low (10^-6 m²K/W), and for longer contact times, the contact resistance value decreases even further.

Figure 11 shows the calculated temperature profiles in steel or copper wheels during the melt-spinning process. It is evident that the wheel surface temperature increases as a function of contact time, with the copper wheel showing a more significant temperature increase than the steel wheel. This is because the copper wheel has a lower thermal diffusivity than steel, allowing the heat to penetrate deeper into the wheel material.

When materials with higher melting points are cast, the wheel surface temperature increases, indicating the importance of wheel material selection. Pure deoxidized copper has the highest thermal diffusivity among commercially useful materials, making it the best choice for the wheel material. For steel wheels, which have lower thermal conductivity, the wheel surface temperature increase is more pronounced.

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Fig. 10. Cooling rate as a function of a distance from the contact surface for different cast materials (α(x) = integral method)

Fig. 11. Wheel temperature increase as a function of contact time and wheel material (Al casting)

Irrespective of contact resistance approximation, when aluminum melt is cast, the wheel surface temperature reaches its maximum decrease, although it is in contact with the hotter ribbon. This seems unlikely, but if we consider the wheel as a whole, its enthalpy is constantly rising, since temperature more than 0.3 mm under the surface is increasing with the entire contact time. Namely, conduction heat transfer rate in the wheel is faster than the heat transfer rate across ribbon/wheel interface and through solidified ribbon.

During continuous casting processes, significant “long term” effects can be observed. In these processes, surface temperature increase may take place, influencing the wheel's performance. Therefore, it is crucial to consider the wheel material selection and its impact on the overall process efficiency.
For first ten to hundred revolutions surface temperature increase may indeed not be significant, but when continuous casting is performed, especially when materials with high melting point are cast, surface temperature of the wheel can increase in such an extent, that formation of the ribbon will be disturbed, because of decreased cooling and solidifying rate in the melt puddle.

Figure 15 shows calculated temperature profiles in inner water cooled wheel \( (R_w = 0.2 \, \text{m}) \) with different casing thickness. From the outside, wheel is convectively cooled with surrounding atmosphere, and from the inside with water stream. For simplicity of the mathematical model, convective heat transfer coefficients are taken as constants \( (\alpha_{\text{ext}} = 5000 \, \text{W}/(\text{m}^2\cdot\text{K}) \) and \( \alpha_{\text{int}} = 50 \, \text{W}/(\text{m}^2\cdot\text{K}) \) \) and represent average values, calculated from forced convection correlation equations. No radiation from the crucible is taking into account. To ascertain influence of external cooling, we also make an assumption of exaggerated value for convective heat transfer coefficient \( (\alpha_{\text{ext}} = 1000 \, \text{W}/(\text{m}^2\cdot\text{K})) \).

Each saw tooth spike corresponds to the temperature of the wheel surface being underneath the puddle. As we can see, internally water cooled wheel will reach the periodic steady state after few revolutions. Conducting of heat into the wheel and cooling of the inner casing surface with water stream is much faster than external convective cooling with surrounding atmosphere. If we reduce wheel casing thickness up to 2 mm, internal water cooling will be more effective, and wheel surface temperature that melt will effectively “feel” at the beginning of the next pass of the wheel under the puddle, will be practically the same as at the first revolution (Figure 15c), even if high melting temperature materials are cast.

But if we reduce wheel casing even further, beneath the heat penetration depth under the melt puddle, convective heat resistance on the inner side (wheel - water interface) becomes significant (Figure 16). Even if we assume heat transfer coefficient value on inner side of a casing as high as 100000 \( \text{W}/(\text{m}^2\cdot\text{K}) \), which can be reached only with high pressure impingement water jets [21], heat removal from the melt will still be slower as in the case of full or internally water cooled wheel with 10 mm thick casing (Figure 15a). Reducing the thickness of the wheel casing is unsuitable, from rapid solidification and from steadiness point of view.

Fig. 14. Cooling curve of iron ribbon as a function of its thickness \( (\alpha(x) = \text{integral method}) \)

Fig. 15. Surface temperature of the internally water cooled wheel as a function of copper casing thickness a) 10 mm b) 5 mm c) 2 mm (aluminum casting, wheel radius 0.2 m)
Conclusions

In our numerical model (FDM), new method for determining contact resistance through variable heat transfer coefficient is introduced which takes into account physical properties of the casting material, process parameters and contact time/length between metallic melt or metallic ribbon and substrate respectively, and enables cooling and solidifying rate prediction before the experiment execution. Comparison of the results, acquired at different assumptions of heat transfer resistances shows, that heat contact resistance between the melt and the chill wheel, and finite thermal diffusivity of the wheel material have great influence on cooling rate of the ribbon and must not be neglected in heat transfer calculations. The fact that the wheel surface temperature is increasing significantly under the melt puddle, selection of the wheel material is not trivial. Because copper has the highest thermal diffusivity between all commercially useful materials, we propose deoxidized copper for a wheel material, and in the case of the need for increased strength, oxide dispersive strengthened (ODS) copper alloys.

For continuous casting, internally cooled wheel is preferable, but only in the case, when wheel casing thickness is correctly selected. When too thick casing is applied, water cooling will not have adequate influence on wheel surface temperature increase. When the casing of the wheel is too thin, thermal resistance on the cooling side becomes the limiting factor, which reduces the heat transfer from the melt and consequently its cooling and solidifying rate.

Acknowledgements

The authors want to thank professor Ladislav Kosec (University of Ljubljana) for mentorship at study of rapid solidification and development of advanced shape memory materials, professor Mirko Gojić (University of Zagreb) for scientific cooperation at synthesis and characterization of rapidly solidified alloys, and professor Mirko Soković (University of Ljubljana) for technical informations and discussions.

References


Fig. 16. Temperature profile in aluminium ribbon and surface temperature of the internally water cooled wheel as a function of copper casing thickness (0.1 mm and 2 mm thick casing)


