Estimation of the unknown parameters in the melt-spinning process

X. ZHANG, A. ATRENS
Department of Mining and Metallurgical Engineering, The University of Queensland, Queensland, 4072, Australia

The free-surface temperature history of the melt spinning of copper measured by Tenwick and Davies [3] is compared with those calculated using a thermokinetic model assuming different parameters. The heat-transfer coefficient, nucleation temperature and the crystal-growth kinetics were thus estimated for the melt spinning of copper at a wheel speed of $35 \, \mathrm{m \, s^{-1}}$ as follows: heat-transfer coefficient during liquid cooling stage $HL = 1.0 \times 10^7 \, \mathrm{W \, m^{-2} \, K^{-1}}$, heat-transfer coefficient after solidification finished $HS = 1.0 \times 10^5 \, \mathrm{W \, m^{-2} \, K^{-1}}$, heat-transfer coefficient during solidification $H = 1.0 \times 10^7 - 1.2 \times 10^{11} \, (t - t_n) \, (\mathrm{W \, m^{-2} \, K^{-1}})$, the nucleation temperature $T_n \approx 1233 \, \mathrm{K}$ and the crystal-growth kinetic law $V = 4.0 \times 10^{-3} \, \Delta T^{1.1} \, (\mathrm{m \, s^{-1}})$.

1. Introduction

The heat-transfer coefficient, nucleation temperature and crystal-growth kinetics have been related [1] to the free-surface temperature history in melt spinning by using a thermokinetic model [2]. These relationships can be used [1] to estimate or determine the unknown factors mentioned above by fitting the measured free-surface temperature history during melt spinning with values calculated using the thermokinetic model [2].

This paper used this method and data measured by Tenwick and Davies [3] to estimate the heat-transfer coefficient, the nucleation temperature and the crystal-growth kinetics in melt spinning.

Tenwick and Davies [3] have measured the free-surface temperature history of melt-spun copper ribbons using a high-speed photographic technique [4] at a sampling rate of 500 frames per second. Because each frame contained an image of the whole ribbon, it is possible to determine the temperatures of different points in the ribbon by the calibrated grey scale, and thus to determine the temperature history of the free-surface during melt spinning. Their measurement on copper was made under the following conditions: a wheel speed of $35 \, \mathrm{m \, s^{-1}}$, an initial melt temperature of $T_2 = 1633 \, \mathrm{K}$ (determined from the figures in [3]), an initial substrate temperature of $T_1 = 303 \, \mathrm{K}$, as no heating of the substrate was involved, and a resultant ribbon thickness of $\delta = 52 \, \mu\mathrm{m}$ as measured.

2. Estimation of unknown parameters

Using the known parameters, described above, and the thermophysical properties for copper (Table I), and assuming different heat-transfer coefficients, nucleation temperatures and crystal-growth kinetic laws, calculated free-surface-temperature-history data were produced by the thermokinetic model [2]. These com-

puted data are compared with the experimental data (determined from the figures in [3]) in Figs 1 and 2. A very long plateau was observed at the beginning of cooling ($t' = 0$ to 22.6 μs) in the experimental data because the experimentally measured data started from the back edge of the stream and went through the stream. This was taken into consideration by setting the back edge and part of the stream in the negative side of the time co-ordinate ($t = -18$ to $0 \, \mu\mathrm{s}$) in the comparison with theoretically calculated data in Figs 1 and 2. The time interval in travelling from the back edge to the start of the melt puddle, i.e. 18 μs, was determined by matching the calculated data with the experimental data during the liquid-cooling stage. This time interval represents a distance of 0.63 mm from the back edge to the middle of the stream, i.e. the start of the melt puddle.

2.1. Determination of the heat-transfer coefficient

The heat-transfer coefficient during the liquid-cooling stage can first be determined by fitting the calculated average cooling rate for the free surface with the experimental value, because no crystallization is involved. Table II lists the average cooling rates for the calculated free surface assuming different heat-transfer coefficients and the experimental value. It can be seen that a heat-transfer coefficient of 2.1×10^6 $\mathrm{W \, m^{-2} \, K^{-1}}$ is too small – compared to the experimental value. A heat-transfer coefficient of $h = 1.0 \times 10^7$ to $1.2 \times 10^7 \, \mathrm{W \, m^{-2} \, K^{-1}}$ produced almost the same average cooling rates as the experimental value for the free surface. Therefore, the heat-transfer coefficient for the liquid-cooling stage was about $1.0 \times 10^7 \, \mathrm{W \, m^{-2} K^{-1}}$ or $1.2 \times 10^7 \, \mathrm{W \, m^{-2} \, K^{-1}}$. Judging from the overall agreement of the calculated curve with the experimental curve in Fig. 2, a heat-

TABLE I Thermophysical properties of copper [5, 6]

	T_m (K)	L (kJ kg^{-1})	C (J kg^{-1} K^{-1})	K (W m^{-1} K^{-1})	ρ (kg m^{-3})
Cu (liquid)	1353.0	204.91	501.6	168.5	8047.0
Cu (solid)	–	–	397.1	350.0	8389.3
Cu (substrate)	–	–	397.1	392.0	8728.8

TABLE II A comparison of calculated and experimental cooling rates

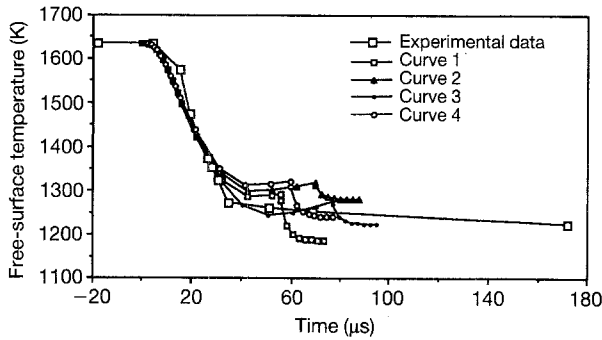| Heat-transfer coefficient h (W m^{-2} K^{-1})	2.1×10^6	5.0×10^6	1.0×10^7	1.2×10^7	Experimental
Average cooling rate for the free surface (K s^{-1})	2.38×10^6	7.0×10^6	10.67×10^6	11.71×10^6	11.82×10^6

Figure 1 A comparison between the experimental free surface temperature history (Tenwick and Davies [3]) and calculated histories assuming: Curve 1, $T_n = 1153$ K, $V = 5.0 \times 10^{-3} \Delta T^{1.25}$ m s^{-1}, HL $= 1.0 \times 10^7$ W m^{-2} K^{-1}, and HS $= 5.0 \times 10^4$ W m^{-2} K^{-1}. Curve 2, $T_n = 1153$ K, $V = 4.0 \times 10^{-3} \Delta T^{1.3}$ m s^{-1}, HL $= 1 \times 10^{-7}$ W m^{-2} K^{-1}, and HD $= 1.5 \times 10^{11}$ W m^{-2} K^{-1} s^{-1}. Curve 3, $T_n = 1233$ K, $V = 4.0 \times 10^{-3} \Delta T^{1.1}$ m s^{-1}, HL $= 1.0 \times 10^7$ W m^{-2} K^{-1}, HD $= 1.2 \times 10^{11}$ W m^{-2} K^{-1} s^{-1}, and HS $= 1.0 \times 10^5$ W m^{-2} K^{-1}. Curve 4, $T_n = 1183$ K, $V = 2.1 \times 10^{-2} \Delta T$ m s^{-1}, HL $= 1.0 \times 10^7$ W m^{-2} K^{-1}, HD $= 1.2 \times 10^{11}$ W m^{-2} K^{-1} s^{-1}, and HS $= 1.0 \times 10^5$ W m^{-2} K^{-1}. $T_2 = 1633$ K, $T_1 = 303$ K, $\delta = 52$ μm.
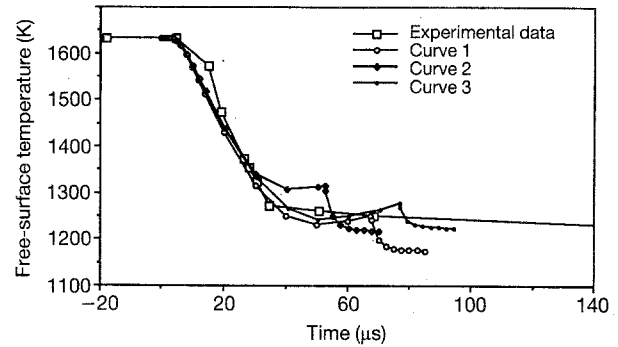

Figure 2 A comparison between the experimental free-surface temperature history (Tenwick and Davies [3]) and calculated histories assuming: HD $= 1.2 \times 10^{11}$ W m^{-2} K^{-1} s^{-1} and HS $= 1.0 \times 10^5$ W m^{-2} K^{-1}. Curve 1, $T_n = 1233$ K, $V = 4.0 \times 10^{-3} \Delta T^{1.1}$ m s^{-1}, and HL $= 1.2 \times 10^7$ W m^{-2} K^{-1}. Curve 2, $T_n = 1213$ K, $V = 2.1 \times 10^{-2} \Delta T$ m s^{-1}, and HL $= 1.2 \times 10^7$ W m^{-2} K^{-1}. Curve 3, $T_n = 1233$ K, $V = 4.0 \times 10^{-3} \Delta T^{1.1}$ m s^{-1}, and HL $= 1.0 \times 10^7$ W m^{-2} K^{-1} s^{-1}. $T = 1633$ K, $T_1 = 303$ K, $\delta = 52$ μm.

transfer coefficient of about 1.0×10^7 W m^{-2} K^{-1} is more likely.

Assuming different nucleation temperatures and different crystal-growth kinetic laws, the free-surface temperature histories were calculated using the thermokinetic model and compared with the experimental value in Figs 1 and 2. If the heat-transfer coefficient during solidification is taken to be the same as the value before solidification starts, there is a big step at the finish of solidification (see Curve 1 in Fig. 1) which was not observed in the experimental measurement. This suggests that the heat-transfer coefficient kept changing during solidification. This suggestion is reasonable as the degree contact between the ribbon and the substrate wheel surface can be lessened by the stress developed from the contraction during solidification.

Assuming that the heat-transfer coefficient decreases with time linearly during solidification, the heat-transfer coefficient at a time t during solidification is calculated by

$$H(t) = \text{HL} - \text{HD}(t - t_n) \qquad (1)$$

where HL is the heat-transfer coefficient for the liquid-cooling stage before solidification starts, HD is the heat-transfer-coefficient decrease per second and t_n is the time at the onset of nucleation. Assuming HD $= 1.2$–1.5×10^{11} W m^{-2} K^{-1} s^{-1}, the calculated results are presented in Fig. 1. The step at the finish of solidification was reduced to quite a small value (see Curves 2 and 3 in Fig. 1), which may not be detectable by the photography method using a calibrated grey scale.

By a comparison between the calculated cooling gradient and the experimental gradient during the solid-cooling stage, the heat-transfer coefficient during the solid-cooling stage after solidification finishes was determined to be about 1.0×10^5 W m^{-2} K^{-1}.

2.2. Estimation of the kinetic factors

Figs 1 and 2 compare the experimental free-surface temperature history with the calculated values assuming different nucleation temperatures, different crystal-growth kinetic laws and different heat-transfer-coefficient decrease constants (HD). It can be seen that Curve 3 (calculated with the parameters HL $= 1.0 \times 10^7$ W m^{-2} K^{-1}, HD $= 1.2 \times 10^{11}$ W m^{-2} K^{-1} s^{-1},

HS $= 1.0 \times 10^5$, $T_n = 1233$ K and $V = 4.0 \times 10^{-3} \Delta T^{1.1}$ (m s^{-1}), has the best agreement with the experimental value.

Therefore, the nucleation temperature for the experiment for melt-spun copper was approximately 1233 K, which represents an initial undercooling of about $\Delta T = T_m - T_n = 1353-1233$ K $= 120$ K. The crystal-growth kinetic law is approximately $V = 4.0 \times 10^{-3} \Delta T^{1.1}$ (m s^{-1}). This kinetic law has a slightly larger power index than the linear Wilson–Frenkel equation but the pre-basial constant is about one fifth of the value of that in the Wilson–Frenkel equation for copper [7]. The heat-transfer coefficient is about 1.0×10^7 W m^{-2} K^{-1} before the onset of solidification, about 1.0×10^5 W m^{-2} K^{-1} after solidification finishes, and about $H(t) = 1.0 \times 10^7 - 1.2 \times 10^{11} (t - t_n)$ (W m^{-2} K^{-1}) during solidification.

Using the parameters determined above, the process characteristics in Tenwick and Davies' experiment [3] can be simulated by the thermokinetic model [2]. Fig. 3a shows the interface-temperature variation with the interface position. It can be seen that the interface temperature increased at first, and then it decreased, and later it increased again towards the end of solidification. This behaviour is characteristic of the linear-growth kinetics [8], and it was due to the almost-linear-growth kinetics determined. Fig. 3b depicts the interface velocity at different interface positions. The maximum velocity is only about 0.8 m s^{-1} at the beginning of solidification. The interface velocity increased slightly at first and then decreased to a relatively steady value before it decreased further towards the end of solidification. The interface velocity is quite low in this cooling regime. This was due to the almost-linear-growth kinetics with a much smaller pre-basial constant than the Wilson–Frenkel kinetic law [7].

Fig. 3c and d show the temperature histories of different layers in the ribbon and the temperature distributions across the ribbon at different times, respectively. The recalescence effect was not so strong because the interface velocity was relatively low and the heat-transfer coefficient was relatively large.

3. Discussion

The maximum discrepancy of about 30 K between the experimental data and the calculated temperature of the free surface was observed near the stream at the beginning of cooling. This probably resulted from the slightly thicker melt near the stream impinging on the wheel surface. This discrepancy may be reduced by considering a thicker melt near the impingement point of the stream. The maximum discrepancy during solidification is about 20 K at the finish of solidification. The agreement is very good considering that the accuracy of the photography method is not so high and the exposure time of 2 ms was about 25 times larger than the time at the finish of solidification. This means that the measured data were the average of about 25 runs of the same process. The discrepancy of 20 K under such measuring conditions is reasonably good.
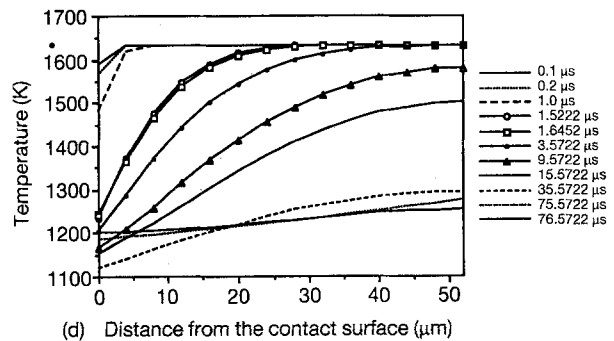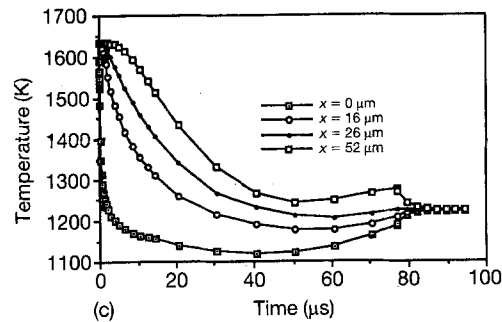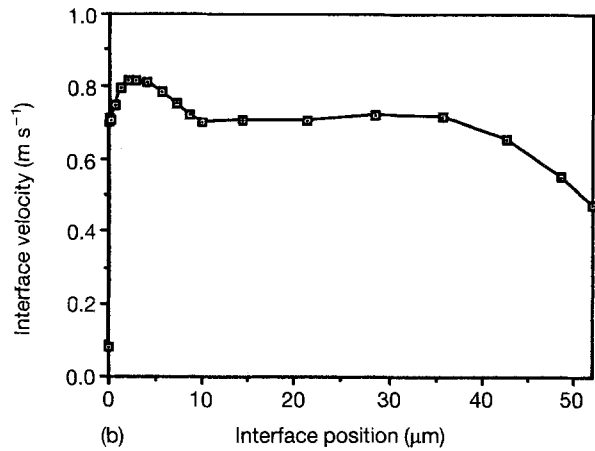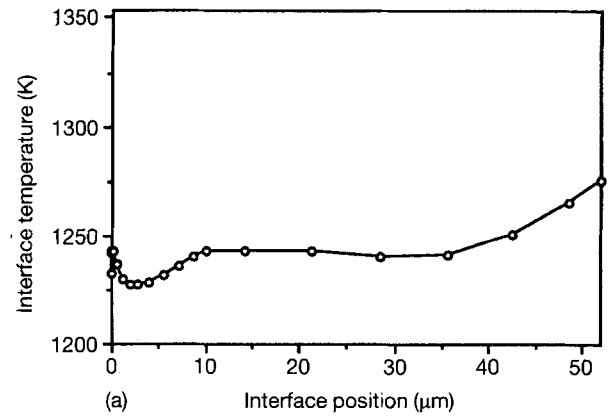
Figure 3 (a) Interface temperature versus interface position curve. HL $= 1.0 \times 10^7$ W m^{-2} K^{-1}, HD $= 1.2 \times 10^{11}$ W m^{-2} K^{-1} s^{-1}, HS $= 1.0 \times 10^5$ W m^{-2} K^{-1}, $\delta = 52$ μm, $T_2 = 1633$ K, $T_1 = 303$ K, $T_n = 1233$ K, $V = 4.0 \times 10^{-3} \Delta T^{1.1}$ m s^{-1}. (b) Interface velocity versus interface position curve. (c) Temperature histories of different layers as indicated. (d) Temperature distribution across the thickness at different times as indicated. HL $= 1.0 \times 10^7$ W m^{-2} K^{-1}, HD $= 1.2 \times 10^{11}$ W m^{-2} K^{-1} s^{-1}, HS $= 1.0 \times 10^5$ W m^{-2} K^{-1}, $\delta = 52$ μm, $T_2 = 1633$ K, $T_1 = 303$ K, $T_n = 1233$ K, $V = 4.0 \times 10^{-3} \Delta T^{1.1}$ m s^{-1}.

The above determination of the heat-transfer coefficient is different from the rough estimations in [3]. These estimations were $HL = 2.1 \times 10^6$ W m^{-2} K^{-1} before solidification finished, and $HS = 7.4 \times 10^4$ W m^{-2} K^{-1} after solidification finished. While their estimation for the heat-transfer coefficient after solidification finished was only slightly smaller than the present determination by modelling, the estimation before solidification finished was too low in comparison with the present determination by theoretical calculation. This was because timing started from the back edge of the stream in their estimation instead of starting from the centre of the stream. The average cooling rate so estimated in [3] was about 5 $\times 10^6$ K s^{-1} at the early stage of cooling instead of the 11.82×10^6 K s^{-1} estimated by beginning the timing from the centre of impingement of the stream on the wheel surface.

The approach above provides a means to determine the unknown thermal and kinetic factors, that is, the heat-transfer coefficient, the nucleation temperature and the crystal-growth kinetic law, by fitting the measured free-surface temperature history with the calculated values via the thermokinetic model. The accuracy for such a determination depends not only on the accuracy of the measurement for the free-surface temperature history during melt spinning, but also on the thickness of the ribbon.

The relevance of the thermal and kinetic factors to the free-surface temperature history is stronger for a thinner melt than for a thicker one, as expected. A slight change in the thermal and kinetic factors can cause an obvious change in the free-surface temperature history for a melt thickness of $\delta = 30$ μm or less, as has been shown [1], while the change may not be so obvious [1] for a melt thickness of $\delta = 50$ μm, or more, for the same slight change in the factors. For example [1], an initial undercooling of 180 K resulted in a deep valley in the temperature history of the layer at a thickness of $\delta = 30$ μm and a shallow valley at a thickness of $\delta = 40$ μm, while it produced only a downhill temperature profile for the thickness $\delta = 50$ μm. Therefore, the accuracy of the determination of the unknown thermal and kinetic factors by fitting the experimental free-surface temperature history with the calculated values, assuming different values of the factors, depends not only on the accuracy in the measurement of the free-surface temperature history

but also on the thickness of the ribbon. The thinner the ribbon, the higher is the accuracy in the determination of the factors. For ribbons thicker than about 50 μm, caution should be taken in the determination to ensure that a certain amount of certainty is reached, otherwise an estimation within a certain range should be given instead of an exact determination.

4. Conclusions

The free-surface temperature history in the melt spinning of copper measured by Tenwick and Davies [3] was compared with those values calculated by a thermokinetic model [2] for rapid solidification. The unknown parameters were thus estimated to be as follows: heat-transfer coefficient during the liquid-cooling stage $HL = 1.0 \times 10^7$ W m^{-2} K^{-1}, heat-transfer coefficient after solidification finished $HS = 1.0 \times 10^5$ W m^{-2} K^{-1}, heat-transfer coefficient during solidification $H = 1.0 \times 10^7$–1.2×10^{11} $(t - t_n)$ (W m^{-2} K^{-1}), the nucleation temperature $T_n \approx 1233$ K and the crystal-growth kinetic law is $V \approx 4.0 \times 10^{-3} \Delta T^{1.1}$ (m s^{-1}). This approach is expected to give good estimations of the unknown parameter for thinner ribbons.

References

1. X. ZHANG and A. ATRENS, *J. Mater. Sci.*, accepted.
2. *Idem. Int. J. Rapid Solidification.* 7 (1992) 83–107.
3. M. J. TENWICK and H. A. DAVIES, in "Rapidly quenched metals V", Proceedings of the Fifth International Conference on Rapidly quenched Metals, Warzburg, Amsterdam edited by S. Steeb and H. Warlimont, (Elsevier Science, Amsterdam, 1985) pp. 67–70.
4. D. H. WARRINGTON, H. A. DAVIES and N. SHOHOJI, in "Rapidly quenched metals IV", Proceedings of the Fourth International Conference on Rapidly Quenched Metals, Sendai, Japan, 1981, edited by T. Masumoto and K. Suzuki (The Japanese Institute of Metals, Sendai (J), 1982) pp. 69–72
5. C. J. SMITHELLS, "Metals reference book", 5th Edn (Butterworths, 1976)
6. Y. S. TOULOUKIAN *et al.*, "Thermophysical properties of matter, the TPRC Data Series, Vol. 1 and Vol. 4, (Plenum, USA, 1970).
7. T. W. CLYNE, *Metallurgical Trans. B* **15** (1984) 369–381.
8. X. ZHANG and A. ATRENS, *Mater. Sci. Engng A* **159** (1992) 243–251.