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Measurement of Al Freezing-Point Temperature: Effect of Initiation Process

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Abstract In CCT documents it is stated that "…for the freezing curves of the metallic fixed points, the maximum observed temperature on the plateau should be taken as the best approximation of the liquidus temperature. The fixed points should be realized with the inner and outer liquid-solid interfaces and extend past the maximum by 10 % to 20 % of the fraction frozen, to clearly establish the value of the maximum and the resolution of its determination." Also, it is accepted that "…the inner interface is essentially static. It is the temperature of the inner liquid/solid interface that is measured by the thermometer." The analysis of freezing curves obtained by the standard method of fixed-point realization shows that the parameters of the initial part of the freezing curve, the mean temperature value of which is usually taken as the liquidus temperature, depend on how the inner interface is initiated. Variations in the duration and intensity of initiation cause changes in the initial part of the freezing curve and in the resulting SPRT measurement. Moreover, the relation between the duration of the initial section of the plateau with a minor temperature change and the duration of its final section with a significant slope also depend on the initiation method used and on the furnace temperature. The effect of freezing initiation conditions on the measurement result is individual for each fixed point because of the differences in thermophysical properties of metals and in conditions of the heat transfer from the liquid–solid interface to the thermometer. Aluminum has a maximum value of the melting specific heat in comparison with other metals used in ITS-90 fixed points; in the present study, the effect of the intensity and duration of the inner liquid–solid interface initiation was investigated both experimentally and through calculation.

Keywords Fixed point · Liquidus temperature · Temperature

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1 Introduction

The methods used for the realization of freezing points of metals by initiation of the formation of the crystallization centers on the thermometer well surface have been applied since the 1960s. Renunciation of measurement of the temperature of the outer interface moving from the crucible walls is connected with peculiarities of the recalescence process, which become apparent in realization of the long-duration plateaus [1]. The process of spontaneous natural solid-phase nucleation during the supercooling of a liquid metal is poorly reproducible since disordered, irregular location of the crystallization centers on the crucible walls significantly extends the recalescence process and formation of the continuous interface. It is accompanied with slow gradual heating depending on the growth of the solid volume. As a result, the duration of the constant temperature plateau decreases to its complete elimination during a protracted process of formation of the continuous interface around the PRT [2].

The inner interface is initiated in order to receive a reproducible long-duration freezing plateau with a constant temperature and to eliminate the influence of the recalescence process.

In conformity with the ITS-90, the methods for realization of freezing points of metals shall ensure the phase equilibrium condition during the freezing process and to provide thermal equilibrium between the interface and PRT. The CCT documents [3,4] recommend to extend the freezing process up to at least 10 h in order to reach an equilibrium condition. If two interfaces are used, the maximum temperature value of the inner interface formed during several minutes is considered as the measurement result. Fulfillment of the phase equilibrium condition for an interface formed during a short period of time is not discussed here. The slowly moving outer interface, to a greater extent corresponding to the phase equilibrium condition, serves only as a thermostating screen for the inner one. The estimation of the value obtained on the basis of the measurement results of (10 to 20) % of the plateau can be only regarded as a confirmation of the thermal equilibrium between the inner interface and PRT. In the similar situation with the initiation of the TPW inner interface, it is assumed that the phase equilibrium is only reached in several days after its formation.

The recommended methods of inner interface initiation for the freezing points of metals are determined empirically for the specific fixed points and crucibles using silica glass rods and cold SPRTs kept in the thermometer well over a different period of time [4]. The question is: do the recommended initiation methods ensure formation of the continuous interface and what effect can it produce on the measurement result? If there is no continuous inner interface, the measurement result can be dependent on the process of the outer interface formation. The dynamic, intensive formation of the crystallization centers on the well surface and the subsequent marked change in the heat removal from its walls can go along with a temporary growth of the plateau temperature. Also, the measurement result can depend on the choice of the inner interface initiation moment relatively to the end of the metal supercooling process and to the moment of the change in the furnace temperature from the supercooling condition to the temperature corresponding to a long-duration plateau.

Some answers to these questions can be found in the literature on this subject; however, the conclusions are ambiguous because of a great variety of influencing factors.

One of the first publications described two interfaces at the freezing point of zinc, where the initiation was carried out by inserting a cold PRT into the well at different stages of the recalescence [1]. The thermometer well of the crucible was made of silica glass in contrast to commonly applied graphite wells. The plateau duration was about half an hour with a single interface and 1 h with two interfaces; in both cases, the freezing temperature values closely agreed within the limits of measurement accuracy. Another publication concerning the freezing point of zinc [5] described the initiation with a cold PRT in the graphite thermometer well. The plateau duration was about 2.5 h; the difference in the maximum freezing temperature values for a slow process with a single interface and two interfaces was about 2 mK. In the investigations of the initiation intensity influence on tin freezing measurements [6], the inner interface was formed by means of a bronze rod immediately after the supercooling of the metal by 1 K. The initiation time was changed from 30 s to 240 s. As a result, a significant increase in temperature was observed at the initial freezing plateau section relatively to its main part. The increase in temperature reached 0.5 mK at the beginning of the plateau and was decreasing during 0.5 h to 1.2 h, depending on its duration down to the value of the main part of the initiation plateau. The total duration of the plateau was about 4 h. Similar changes of the initial part of the freezing plateau were observed for the indium point [7] after the initiation of the inner interface with one and two rods after an active initiation of the outer interface by taking out the crucible from the thermostat for (1 to 2) min. The crucible and the thermometer well were made of fluorocarbon polymer (Teflon). The purposeful investigations of the influence of four different methods for realization of the indium fixed point on the measured freezing temperature are described in [8]. In this study, the crucible and thermometer well were made of silica glass, and a liquid thermostat was used for the freezing. The plateau duration with the constant temperature was about 1 h when freezing initiation was applied, and it was much shorter without the initiation; the measured freezing temperature values agreed within 0.1 mK. The influence of the initiation intensity of the outer interface on the measurement result of the zinc freezing point was studied in [9]. The interface initiation intensity was changed by using cold PRTs, rods made of silica glass and stainless steel having different heat capacities; their exposure in the thermometer well was changed from 1 min to 3 min. The difference between the freezing temperature values with the most intensive initiation and without initiation was 1 mK with the freezing plateau duration from 10 h to 16 h. The influence of the changes in the initiation intensity on the measurement result was estimated to be 0.5 mK.

It follows from the above results that the influence of the inner interface initiation method on the temperature measurement result was observed when the freezing processes lasted for more than 2 h. With the long-duration plateaus, the freezing temperature values were different for a single interface and two interfaces.

The purpose of this work was to estimate the influence of the inner interface initiation method on the measured freezing temperature of aluminum and to compare the maximum temperature values taken as the liquidus temperature, obtained with a single interface or two interfaces with different durations of the process.

2 Experiment Procedures

The high-purity graphite crucible with an inner diameter of 36 mm and a height of 240 mm was filled with 6N purity aluminum. The aluminum mass in the crucible was about 370 g; the inner diameter of the thermometer well was 10 mm. The crucible was put inside a silica glass tube connected with an argon source, which made it possible to control the argon pressure above the crucible. The crucible in the tube was put into a furnace with three heaters, with the temperature regulated by an automatic system. The temperature stability of the heater was within 0.02 K to 0.05 K. The temperature gradients in the thermometer well were controlled in the molten metal state before the plateau, and in the freezing period in an hour and 5 h after the beginning. The temperature gradient in the molten metal did not exceed 0.01 K throughout the metal height, and during the freezing period, the temperature gradient corresponded to the influence of hydrostatic pressure within the limits of the thermometer displacement by 4 cm from the bottom. The measurements were made by means of an SPRT and a F900 bridge.

The aluminum, molten and overheated by 5 K, was kept in the furnace for at least 12 h before the freezing. After that, the temperature in the furnace was decreased to be 5 K, lower than the freezing temperature, and the supercooling of aluminum was recorded. In our experiments, the value of the supercooling temperature was changed from 1.2 K to 1.4 K. When the metal temperature began to increase, the heater temperature rose to attain a desirable duration of the freezing plateau. The control system allowed changing the furnace temperature during (2 to 3) min.

The reproducibility of the results was confirmed by multiple experiments over 1.5 years with two Al cells Nos. 55 and 75.

3 Results

At the first stage of the investigation, the temperature was measured in the process of the freezing of aluminum with the initiation of a single outer interface, and the duration of the recalescence process and the time of continuous interface initiation were estimated at different heater temperatures. The freezing process duration increased from (5 to 26) h due to a decrease of the difference between the heater temperature and the freezing temperature from 1.5 K to 0.4 K. The temperature changes during the freezing of aluminum with a single outer interface for different heater temperatures are shown in Fig. 1.

Figure 1 shows that the freezing temperature reaches its maximum value with a rather significant interval after the supercooling. It occurs in 5 h for the plateau with the duration of 26 h and in (40 to 50) min for the shortest one. The difference between the maximum values was 2 mK for the short and long plateaus.

It was quite interesting to estimate the amount of crystallized aluminum at the moment of attaining the maximum temperature. It can be obtained from the heat-balance equation for the freezing of aluminum. The heat-balance equation under the condition of a constant heat flow from the crucible to the heater during the whole phase transition can be represented by the following expression:

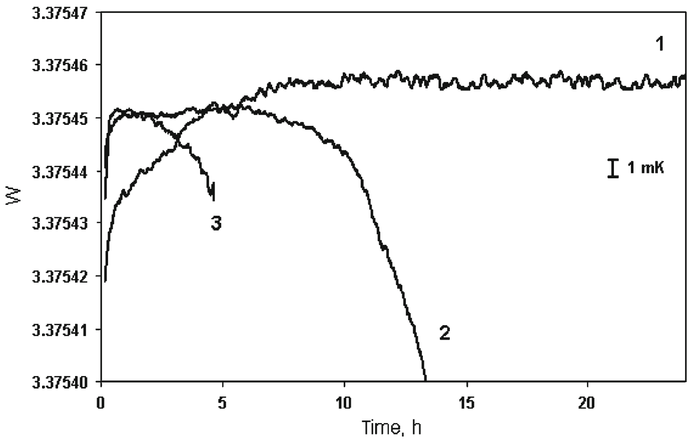

Fig. 1 Aluminum freezing curves with single outer interface: *1* $T_f - T_h = 0.4$ K, *2* $T_f - T_h = 0.7$ K, and *3* $T_f - T_h = 1.5$ K

$$mL = \alpha S(T_f - T_h)\Delta\tau \tag{1}$$

where m is the aluminum mass (kg), L is the heat of phase transition ($kJ \cdot kg^{-1}$), α is the total heat exchange coefficient between the crucible inner surface and surroundings ($W \cdot m^{-2} \cdot K^{-1}$), S is the total inner surface of the crucible (m^2), $T_f - T_h$ is the difference between the values of the aluminum freezing point and heater temperature (K), and $\Delta\tau$ is the freezing process duration (s). The values of m, L, and S are known, and they are constant for the crucible, but the value of α is unknown and is to be determined. The averaged experimental data for the four freezing plateaus were used to determine the total heat exchange coefficient, α, taking into account the uncertainty in the estimation of the complete metal freezing duration and the maintenance of the heat exchange homogeneity of the total crucible surface and heater.

The heat-balance equation shows that the freezing duration is inversely proportional to the difference between the heater temperature and the freezing temperature;

$$\Delta\tau = C(T_f - T_h)^{-1} \tag{2}$$

where $C = mL/\alpha S$.

On the basis of the data for the four aluminum freezing plateaus, we plotted (Fig. 2) the dependence of the duration process on the difference between the values of the aluminum freezing temperature and heater temperature, $\Delta\tau$ versus $(T_f - T_h)$. The trend line shown in the graph corresponds to the equation $\Delta\tau = 8.56(T_f - T_h)^{-1.08}$ with a confidence probability $R^2 = 0.96$. The resultant functional dependence confirms the equal conditions of heat exchange between the crucible surface and the heater over the whole range of the heater temperature. Accepting the mean value $C = 8.56\,K \cdot h$, the calculated value of the mean heat exchange coefficient between the crucible surface and the heater was $\alpha = 160\,W \cdot m^{-2} \cdot K^{-1}$. At first, to estimate the recalescence

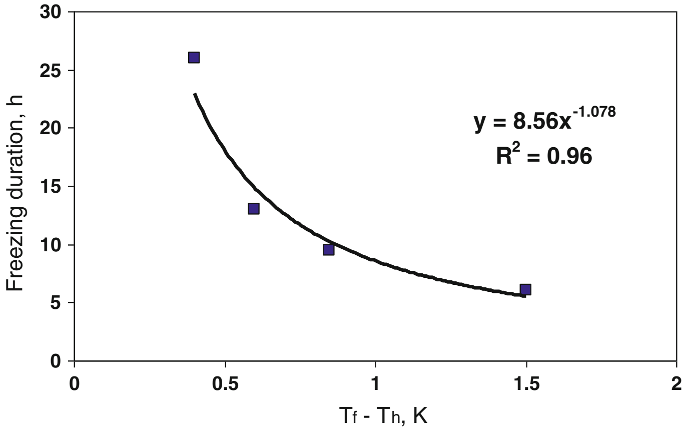

Fig. 2 Dependence of freezing plateau duration on the difference between the values of the aluminum freezing point and heater temperature

process accompanied by a progressive temperature rise, the amount of solid fraction required for the increase of the aluminum temperature from supercooling conditions to the freezing temperature, was calculated. The constant heat dissipation from the crucible to the surroundings was taken into account in the calculation.

The quantity of heat requred to increase the temperature of the aluminum and thermometer well by 1.4 K from the supercooling state, taking into account their specific heat, is about 500 J. The heat quantity transferred from the crucible to the surroundings after the supercooling in the process of the change of the heater temperature with the average difference in their temperature of 2.5 K, is about 1.6 kJ; the total quantity of heat necessary to reach the freezing temperature during this period is about 2.1 kJ. This quantity of heat is released during crystallization of a very insignificant amount of aluminum of only 5.4 g. If we assume that the formed aluminum crystallization centers and the growing solid phase are uniformly distributed over the crucible surface, the thickness of the formed solid aluminum layer will be about 0.08 mm, which is comparable to the roughness of the crucible surface. Probably, this thin, irregular surface film of the growing solid phase does not form a continuous outer interface, and as a result, the PRT records a continuing increase of the aluminum temperature caused by the growth of the solid aluminum surface and a decrease of the heater temperature influence.

The time required to reach the maximum temperature values at the long plateaus agrees well with the calculation of the time interval necessary to form the solid phase layer on the crucible wall having a thickness of 0.8 mm to 1.0 mm. The formation of such a layer that required a heat of about 20 kJ to 25 kJ was removed from the crucible walls. The maximum freezing temperature values and the continuous interface on the crucible walls were attained in about 1.8 h and 5 h after the supercooling for the 10 h and 26 h plateaus, respectively. The initiation period of the continuous interface only depends on the heat dissipation speed, i.e., on the heater temperature, and it can be reduced to several minutes with the relevant values of the heater temperature.

It is important to notice that about (10 to 15) % of the aluminum mass was solidified by the time of attaining the maximum freezing temperature with a single outer interface. This circumstance is not considered in the analysis of freezing curves made to define the quantity of impurities [10]. The influence of solidified metal at the maximum freezing temperature will change the inclination of curves $\mathrm{d}W/\mathrm{d}(1/F)$ [10].

At the second stage of experiment, the effectiveness of the inner interface initiation was estimated when using ceramic and silica glass rods with different exposure times in the thermometer well of the crucible. The effectiveness of the procedure was estimated by the results of the calculation of the aluminum quantity crystallized on the thermometer well surface and the thickness of the formed layer. The quantity of the heat removed by the rod inserted into the thermometer well was determined on the basis of the time variation of its temperature in the process of insertion and the conditions of its heat exchange inside the well.

The temperature of the rod, as well as any other body inserted into the medium with a constant temperature, varies under the law similar to the exponential one. Hence, the heat flow between the rod and metal in the crucible is not constant and varies according to the variation of the difference between the temperature of the rod and the aluminum freezing temperature. The quantity of heat removed by the rod from the molten aluminum can be calculated by two methods: first, by the formula,

$$Q = \alpha s \int_{0}^{\tau_1} \Delta t(\tau) \mathrm{d}\tau \tag{3}$$

where α is the total heat-transfer coefficient between the rod and aluminum, s is the rod surface, $\Delta t(\tau)$ is the function of the variation of the difference between the aluminum temperature and the rod temperature with time, τ is the time, τ_1 is the time of the rod exposure in the thermometer well; and second, on the basis of the total heat capacity of the inserted rod and its temperature rise in the well. The heat flow variation is plotted in Fig. 3.

The function $\Delta t(\tau)$ of the variation of the difference between the aluminum temperature and the ceramic rod temperature with time and the thermal inertia constant of the rod, ε, were determined experimentally. With this purpose, the ceramic rod had a built-in type K thermocouple and its temperature variation during insertion into the well was recorded. During the process of the insertion into the well, the rod temperature increased by 20 K. The thermal inertia constant of the rod, ε, was determined on the basis of the transition curve of its temperature variation; it was equal to 1.5 min. The temperature variation of the rod after its insertion into the well can be calculated by the formula,

$$\Delta T(\tau) = 620[1 - \exp(-\tau/\varepsilon)] \tag{4}$$

The calculated value of the volumetric heat capacity of the ceramic rod was $23\,\mathrm{J \cdot K^{-1}}$.

As a result, the quantity of heat removed by the rod during 1 min is about $Q = 6.5\,\mathrm{kJ}$, which entails the crystallization of about 16 g of aluminum. This amount of aluminum is enough for initiation of a solid phase layer on the thermometer well surface

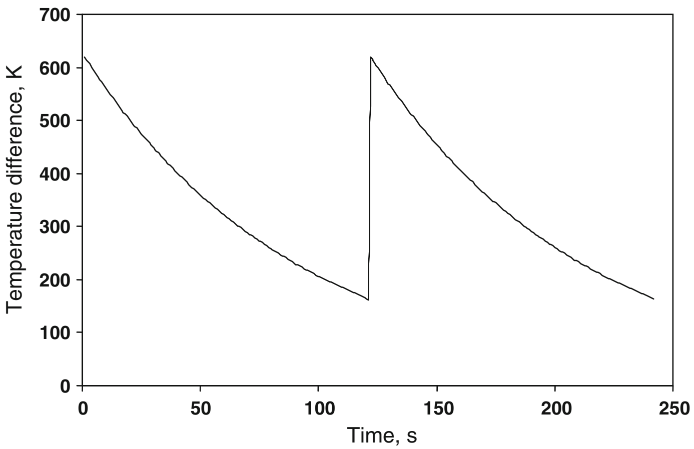

Fig. 3 Character of heat flow time variations after the initiation of the inner interface by two rods during 2 min

with a thickness of about 0.5 mm under the condition of a uniform metal distribution over the surface. After a rod exposure of 2 min, $Q = 10.5$ kJ and the solid aluminum layer thickness is 0.8 mm. Since most of the heat flow is withdrawn from aluminum at the moment of inserting the rod, it could be more productive to increase the number of rods and to decrease their exposure time in the well in order to increase the heat dissipation. The three rods inserted into the well, each one for 1 min, remove about 27 kJ and a layer with a thickness of about 2 mm is formed. By a rough estimation, the cold thermometer inserted for measurement can increase the thickness of frozen aluminum by 0.1 mm to 0.2 mm.

The volumetric heat capacity of a silica glass rod is two times lower than that of a ceramic rod. One can assume that the conditions of its heat exchange in the well are similar to those for the ceramic rod, then its thermal inertia constant is $\varepsilon = 1$ min. As a result, if it is kept in the well for 2 min, the solid aluminum layer thickness will be about 0.9 mm.

At the next stage of investigation, we measured the aluminum freezing temperature with the inner interface initiation using a ceramic rod at different heater temperatures ($T_f - T_h = 0.4$ K, 0.7 K, 0.85 K, and1.5 K). Different configurations were applied to initiate the inner interface: one rod during 1 min, two rods during 2 min, and three rods during 1.5 min each. The inner interface initiation was started simultaneously with the beginning of the outer interface initiation, immediately after recording the beginning of the metal temperature rise after the supercooling of aluminum. At the same time, the heater temperature was changed from $T_f - T_h = 5$ K to a new value chosen for the realization of the freezing plateau; the interval between the beginning of the temperature rise after supercooling and the beginning of the initiation did not exceed 3 min.

The aluminum freezing temperature curves in the process with initiation were overlapped with the curves obtained with a single outer interface, the heater temperature being the same. The origin of the curves without initiation corresponds to the beginning

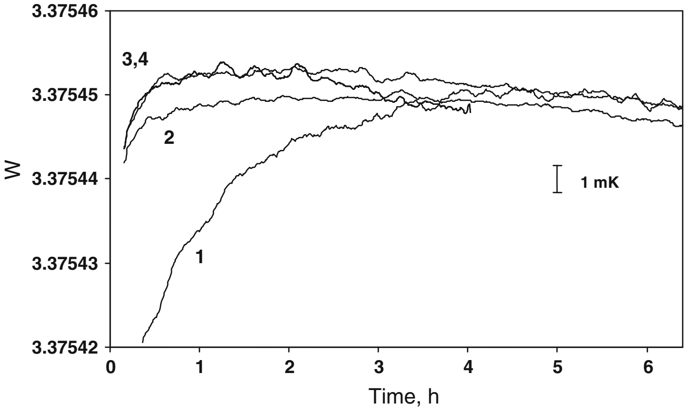

Fig. 4 Aluminum freezing curves at a difference of 0.7 K between the values of the aluminum freezing point and heater temperature: *1* freezing temperature with a single outer interface, *2* after the initiation of the inner interface by one rod during 1 min, *3* after the initiation by two rods with exposure in the well of 2 min each, and *4* after initiation with three rods with exposure of 1.5 min each

of the temperature rise after the supercooling, while for the curves with the inner interface, it corresponds to the stabilization of the thermometer readings after initiation of the inner interface. The phase shift of the temperature curves does not exceed 10 min. Overlapping of the curves makes it possible to compare and analyze the synchronous temperature variations of the outer and inner interfaces.

The first series of experiments for the investigation of the initiation influence on the maximum temperature value during the freezing were conducted at $T_f - T_h = 0.7$ K. At this set value, freezing continues for about 10 h which agrees with the recommendations of the document [4]. Figure 4 shows the aluminum freezing curves obtained with a single outer interface and after different initiations of the inner interface. Curve 1 corresponds to the freezing temperature with a single outer interface, curve 2—the freezing temperature after the initiation of the inner interface with one rod during 1 min, curve 3—after the initiation by two rods during 2 min each, and curve 4—after the initiation by three rods during 1.5 min each. The maximum values of curves 3 and 4 are practically the same; they exceed the maximum temperature value obtained for a single interface by 0.5 mK, and for curve 2 by 1 mK. It follows from the results that the initiation with one rod during 1 min does not form a continuous interface, and the measurement result is influenced by the unfinished process of outer interface formation, and two or three rods ensure the continuous interface. An example of the process of gradual formation of the outer interface during zinc freezing is clearly visible in the autoradiographs in [5]. It is seen from the autoradiographs that the process was completed in 60 min.

Figure 5 shows three curves obtained at the temperature difference of $T_f - T_h = 1.5$ K. Curve 1 corresponds to the freezing temperature with a single outer interface, curve 2—the temperature after initiation of the inner interface by two rods during 2 min, curve 3—after initiation by three rods during 1.5 min each. In order to avoid the influence of “noise” on the curves, the mean W values were calculated for the flat

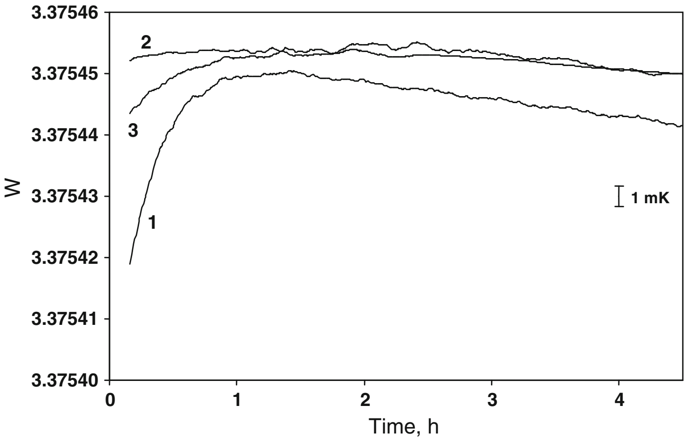

Fig. 5 Aluminum freezing curves at a difference of 1.5 K between the values of the aluminum freezing point and the heater temperature: *1* freezing temperature with a single outer interface, *2* after initiation of the inner interface by two rods with exposure in the well of 2 min each, and *3* after initiation by three rods with exposure of 1.5 min each

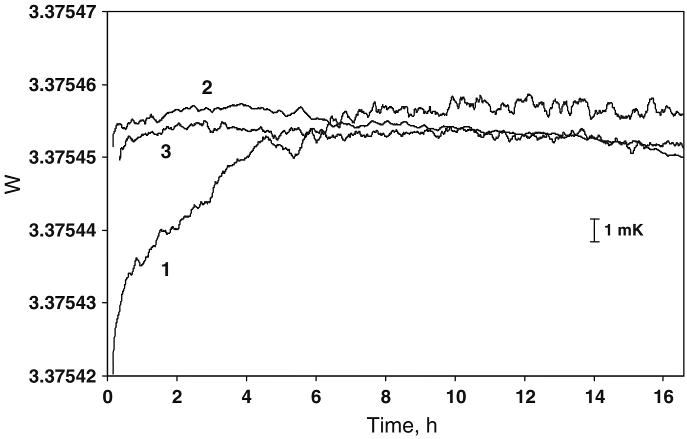

Fig. 6 Aluminum freezing curves at a difference of 0.4 K between the values of the aluminum freezing point and heater temperature: *1* freezing temperature with a single outer interface, *2* after initiation of the inner interface by two rods with exposure in the well of 2 min each, and *3* after initiation by three rods with exposure of 1.5 min each

part of each freezing plateau near the curve maximum. The highest maximum temperature value corresponds to the process with the initiation by three rods. It exceeds the maximum values of the curve without initiation and the curve with initiation by two rods by 1 mK and 0.3 mK, respectively. The moments of attaining the maximum values for the three freezing modes coincide quite closely.

The curves obtained in the process of the freezing at the difference between the freezing temperature and the heater temperature of 0.4 K are shown in Fig. 6. In the slow processes, the maximum freezing temperature value corresponds to the process

Table 1 Maximum values of W after initiation by 2 rods during 2 min

Date	$T_f - T_h$ (K)	W_{max}
25.03.09	0.7	3.375 453 6
27.04.09	0.7	3.375 454 1
08.05.09	1.5	3.375 454 6
16.05.09	1.5	3.375 455 1
23.05.09	0.85	3.375 455 2
09.09.09	0.7	3.375 453 7
15.09.09	0.7	3.375 454 0
24.09.09	0.4	3.375 455 7
	Mean value of W	3.375 454 5

with a single interface (curve 1). It is higher by 1.5 mK than the maximum freezing temperature with the initiation by three rods during 1.5 min and by 1 mK than the freezing temperature with the initiation by two rods during 2 min.

Table 1 gives the maximum temperature values obtained from the freezing curves with the initiation by two rods during 2 min at different heater temperatures. The standard uncertainty of the maximum temperature values of these curves is 0.3 mK.

The temperature curves with initiation (Figs. 4, 5, 6) immediately after the supercooling demonstrate a small increase in temperature within 0.1 mK to 0.2 mK at the initial section. In one experiment with the initiation by two rods during 2 min with the temperature difference $T_f - T_h = 0.85$ K, the heater setting was changed in 10 min after the supercooling, i.e., the condition for formation of the continuous outer interface was created. The maximum temperature value is observed on the resultant freezing temperature curve with the initiation (Fig. 7, curve 1) immediately after the stabilization of the thermometer readings. After that, the freezing temperature is decreasing during 3 h to the temperature corresponding to freezing with a single interface. The difference between the maximum values is 1.2 mK.

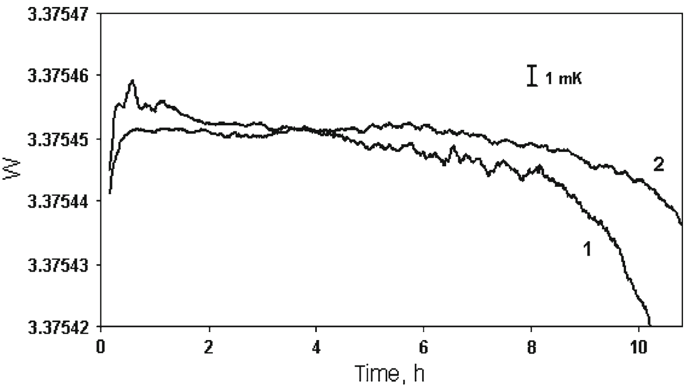

Fig. 7 Aluminum freezing curves at a difference of 0.7 K between the values of the aluminum freezing point and heater temperature: *1* after the initiation of inner interface in the presence of continuous outer interface and *2* without initiation of inner interface

The results of the present investigation are indicative of the fact that the maximum freezing temperature value with the initiation of the inner interface was higher than in the case of a single interface (with the exception of the process with the duration of 26 h).

When analyzing the obtained differences between the maximum temperature values, the conditions of formation and growth of aluminum crystals on the outer wall and on the thermometer well were compared. The resultant maximum aluminum freezing temperature discrepancies can be explained, in the first place, by differing conditions of formation and growth of aluminum crystals on the outer wall and on the thermometer well, and second, by the thermometer position with respect to the interface. In the case of a single interface, there is an additional thermal resistance of the liquid aluminum layer between the outer interface and thermometer. However, this thermal resistance is small in comparison with that of the thermometer well walls and the gap between the well and thermometer, and it does not affect the result of measurement.

The heat flow densities during the formation of the solid phase were quite different. The heat flow density on the crucible walls varied from $64\,\mathrm{W \cdot m^{-2}}$ to $240\,\mathrm{W \cdot m^{-2}}$ depending on the time of freezing. Under initiation by a ceramic rod, the heat flow density on the well surface at the initial moment was $3 \times 10^4\,\mathrm{W \cdot m^{-2}}$, and its average value during 2 min could be estimated as $7 \times 10^3\,\mathrm{W \cdot m^{-2}}$. As a result, the maximum temperature value on the outer wall corresponds to the temperature of the boundary of the layer with a thickness of 0.8 mm to 1.0 mm formed with a velocity of $0.025\,\mathrm{mm \cdot min^{-1}}$ and about $0.003\,\mathrm{mm \cdot min^{-1}}$ for the process lasting 5 h and for the plateau of 26 h, respectively. When using the initiation, the maximum value corresponds to the temperature of the boundary of the inner interface with a thickness of about 0.8 mm formed with a velocity of $0.2\,\mathrm{mm \cdot min^{-1}}$.

The velocity of the interface formation also determines the character of impurity segregation during freezing. Considerable changes in the velocity of the interface formation alter considerably the impurity segregation character, and, consequently, the temperature of the interface. The significant difference in the velocities of the formation of outer and inner interfaces is probably the reason for discrepancies between the recorded maximum temperature values. In the case of short freezing plateaus without the initiation, the velocities of formation of the inner and outer interfaces are practically the same, and, as is demonstrated in [8], the maximum freezing temperature values with and without initiation coincide.

4 Conclusions

As a result of investigations of aluminum crystallization with a single interface, it was found that at the moment of attaining the maximum temperature value, about (10 to 15) % of the metal froze in the crucible in the process lasting from 5 h to 26 h. The maximum value corresponded to the formation of a continuous outer interface with a thickness of 0.8 mm to 1.0 mm.

The initiation of the inner interface with two ceramic rods with an exposure in the thermometer well of 2 min each entails a reproducible flat part of the freezing plateau being (10 to 20) % of the plateau total duration. In this case, the standard uncertainty of the maximum temperature values is within 0.3 mK. The initiation with one rod during 1 min does not form a continuous interface around the well, and the measurement result is influenced by the interphase boundary on the crucible walls having a lower temperature. It is established that the choice of the initiation moment of the inner interface can influence the freezing plateau. The initiation after the formation of an outer interface on the crucible walls may lead to an increase in temperature and to variations at the initial part of the plateau of up to 1.2 mK, which was also shown in [6]. When realizing the procedure of inner interface formation, it should be taken into account that the completion of the continuous outer interface depends on the velocity of the heater temperature change from the metal supercooling condition to the temperature of the plateau realization.

During the investigation, it was discovered that the discrepancies in the maximum temperature values with a single interface or two interfaces were about 1 mK to 1.5 mK, the crystallization lasting from 5 h to 26 h. These discrepancies could be plausibly explained by the changes in impurity segregation due to differing freezing velocities of the interface on the crucible walls and in the well surface. The maximum freezing temperature value on the outer interface corresponds to the temperature of the boundary of the interface formed with a velocity of $0.003\ \mathrm{mm \cdot min^{-1}}$ to $0.025\ \mathrm{mm \cdot min^{-1}}$ and its dependence on the duration of freezing; in the case of the initiation with a variable heat flow—the maximum freezing temperature value corresponds to the temperature of the interface formed with a velocity of $0.2\ \mathrm{mm \cdot min^{-1}}$.

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