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Prediction of dendritic micro-heterogeneity of cast steel: review of models and computer-aided analysis of problems (Part 1. Models based on thermal-physical parameters)

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The paper reviews the publications concerning dependence of dendrite arm spacings of iron-based industrial alloys from the conditions of solidification. It was noted that the used thermal parameters which characterize the conditions of dendrite formation — the rate of solidification, the temperature gradient and the cooling rate — quite often are determined with significant experimental errors, are estimated on the basis of approximate relationships and often mutually correlated. It was established that the published empirical power-type models of dendrite arm spacing for carbon and low-alloy steels are characterized by a lot of the types of the parameters- predictors and by scatters of their values, do not consider the effect of the alloys composition and slightly suitable for prediction of the dendritic structure. For objective assessment of uncertainties arising from the use of insufficiently large data sets and simplified method of estimation for model parameters the procedures of statistical analysis of the models adequacy for their correction and/or rejection were proposed.

The comparison of results of computer modeling for a steel slabs (250 mm thickness) with 0.006, 0.06 and 0.6% C are used for analysis the evolution during solidification of the rate of crystallization and the temperature gradient under various intensity of heat extraction and natural convection of the melt. It was deduced that a radical increase in the accuracy of the analysis of the conditions of formation of the dendritic structure is provided using a developed computer model of the non-equilibrium solidification of ingots and castings on the base the thermal properties of alloys, determined by means of thermodynamic modeling, with obligatory taking into account the intense convective heat transfer in the melt.

Key words: *carbon and low-alloy steel, dendritic structure, dendrite arm spacing, empirical power-type models, computer modeling, non-equilibrium crystallization.*

Setting of the problem. Multiple publications devoted to investigation of dendrite microstructure of iron-base industrial alloys [1—22 et al.], including recent review papers [11, 19, 20], contain large data about values of primary λ_1 and secondary λ_2 dendrite arm spacings as well as their relationships depending on different metallurgical and technological factors. Such attention to parameters of structural micro-heterogeneity of steel is caused by their substantial effect of forming defects of cast metal (such as dendrite segregation, gas and/or shrinkage porosity, hot cracks etc.) and by corresponding mechanical properties of deformed metal. The stream of publications describing this theme, started in 1960-ies and continuing at present time, is caused evidently not only by its importance, but also (last but not least) by exclusive complication of the observed appearances occurring during crystallization of multicomponent industrial alloys, in combination with difficulty of experimental works at increased temperatures. As a result, the analysis of the processes of dendritic crystallization does not allow yet to reveal the causes of some of the observed contradictions and to get substantiated answer on the key theoretical and practical questions in the field of quality forming of cast metal.

Confident technological forecast of dendritic microstructure in the steel with preset composition, as well as synthesis of alloys with required structure need high-quality mathematical models in combination with reliably established scientific regulations, describing comparative effect of steel components on the values of λ_1 and λ_2; this information is now rather approximate and is characterized by apparent contradiction. Local heterogeneity of dendritic structure plays an important role in forming the quality of cast metal and had not any adequate quantitative description until today. It is important to have adequate calculated relationships in

Table 1. Conditions of conducting investigations and evaluations of parameters

№	Steel			Conditions of experiment		Characteristic of crystallization conditions							Analysis of the effect of chemical composition	Year of publication	Reference
							Speed V		Temperature gradient G						
	Carbon	Low-alloy	High-alloy	Industrial (continuous casting)	Laboratorial	Cooling rate R	Dendrite growth rate V_L	Rate of solidification front	Average G	In liquid phase G_L	In solid phase G_S	Local time of solidification τ_{LS}			
1		*			•	C						E	—	1968	[1]
2		*			•							E	0	1970	[2]
3		*			•	C						E	0	1972	[3]
4		*			•			E	E				+	1976	[4]
5		*			•	C		E	E	E	E	E	0	1976	[5]
6		*	*		•		E		E	E	E	C	+	1982	[6]
7		*	*		•	C						E	0	1986	[7]
8		*			•	C	E	E	E			E	0	1994	[8]
9		*		•		C		E	E	E	E	C	+	1998	[9]
10		*			•	C		E	C	C	C	C	+(C)	1999	[10]
11	*	*		•		C		C		C		C	+(C)	1999	[11]
12		*			•							C	+(C)	2008	[12]
13	*	*		•		C		C			C	E	0	2009	[13]
14	*	*	*	Liter.		E							+(C)	1977	[14]
15	*			Liter.		E	E					C	+(C)	1996	[15]
16	*	*	*	Liter.		C							+(C etc.)	2006	[16]
17	*	*		Liter.		C						E	+(C)	2001	[17]
18	*	*		Liter.		C						E	+(C)	2000	[18]
19	*	*	*	Liter.	•	C		C		C		C	+	2003	[19]
20	*	*	*	Liter.		E+C							+(C etc.)	2010	[20]

Remarks: Influence of chemical composition: (+/–) — steel components have effect / have no effect; 0 — not investigated; E — experiment; C — calculation; (C) — carbon

management of the process of dendritic crystallization; these relationships describe in general the effect of metallurgical and technological parameters on forming and development of structural and chemical microheterogeneity of cast metal that are inherited even during intensive plastic deformation. However, well-known evaluations of the input of these parameters are often not correlated among themselves, thereby essential deviation of the main calculated parameters in industrial conditions is not displayed and can't be analyzed.

The aim of current publication is discussion of the row of problematic aspects in forming dendritic heterogeneity of cast steel that were revealed as a result of comparative analysis of wide array of publications as well as revision of several discussion results of conducted investigations on the base of usage of the developed computer models.

Review of investigations of secondary dendrite arm spacings. Carbon and low-alloy steels have been chosen as the main objects of investigations because considerable array of empirical data [1–22 et al.] has been accumulated for these steels and a row of theoretical developments has been made for them.

Description of a row of investigations is presented in the **table 1**; it includes the data about conditions of their conduction and contains references to the most typical and surveying works with analysis of different aspects of the considered investigations. The data presented in these works have been obtained in different conditions — at laboratorial units using special casting forms and thermal devices as well as in industrial and semi-industrial conditions during continuous casting. Usage of laboratorial equipment allows provide the preset solidification procedure, measure temperature varia-

Table 2. Chemical composition of low-alloy steels and values of K_2 and m coefficients in $\lambda_2 = K_2\tau_{LS}^m$ formula

№	C	Si	Mn	Ni	Cr	K_2, μm/s^m	m	Year of publication	Reference
1	0.55–0.6	0.01–0.07	2.03–2.39	—	—	(29.5)	0.39	1970	[2]
2	0.59	0.03	1.1	—	—	15.8	0.44	1976	[5]
3	1.48	0.03	1.14	—	—	7.16	0.56		
4	0.1	0.08	0.4	0.05	0.05	(10.8)*	0.33	1998	[9]
5	0.08	0.86	1.48	0.01	0.04	(48.2)*			
6	0.09	0.55	1.2	0.03	0.03	(33.5)*			
7	0.63	0.21	0.67	0.03	0.03	(54.4)*			
8	0.09	—	1.36	—	—	28	0.51	1999	[10]
9	0.15	—	1.44	—	—	16.8	0.43		
10	0.55–0.56	0.24–0.3	0.75–0.85	—	—	52*	0.32	1999	[11]
11	0.47	0.37	0.73	—	—	40*	0.35		
12	0.14–0.2	0.27–0.53	1.35–1.71	—	—	15*	0.46		
13	0.11–1.01	—	—	—	—	$79C^{-0.187}$	0.38	2000	[18]
Average value and standard deviation						29.3±7	0.40±0.06		

Remarks: * — specimens received via continuous casting; K_2 — calculated on the base of formulas presented by the authors.

tion via thermocouple units, control local technological parameters and, in several cases, keep steady of vary in the preset range such parameters as cooling rate R, temperature gradient G in solidification area and/or motion rate V of solidification front, independently from each other. This equipment is used via flexible varying of size of specimens and conditions of heat transfer. In the cases of using steel continuous casting machines, variation of crystallization parameters is limited by varying of steel overheating and casting speed.

The data presented in the table 1 testify that a row of thermo-physical parameters (R, G, V etc.) of their combinations are used for quantitative estimation of crystallization conditions. The a.m. parameters differ seriously in their techniques and possible mistake of their determination; thereby they can't always be subjected directly in mutual correspondence.

In addition to it, fuzzy character of knowledge about relationship between thermo-physical parameters and conditions of forming of dendritic structure leads to powered law models of different kinds [1–22 et al.]:

$$\lambda_1, \lambda_2 = F(R^n, V^p, G^q, ...), \quad (1)$$

often without mentioning or comparative evaluation of validity reached in this case (e.g. correlation coefficient etc.).

The most frequently used form of powered law models for secondary dendrite arm spacings of second order λ_2 includes cooling rate R and local time of solidification τ_{LS} [1–20 et al.]:

$$\lambda_1 = K_1 R^n; \lambda_2 = K_2\tau_{LS}^m, \quad (2)$$

what makes it possible to reflect influence of only thermo-physical conditions of crystallization (**table 2**); in several cases relationship between K_1 and K_2 coefficients and steel composition [4, 6, 9–20 et al.] is taken into account (it will be considered later).

The values of K_2 and m parameters presented in table 2 are varied in wide range (K_2 from 7 to 54 μm/s^{-m} at $m = 0.32–0.56$), what is typical also for other empiric expressions such as (1) and (2) [11, 19 et al.]. It doesn't allow to use them for reliable prediction of dendritic structure (**fig. 1**), but suggests searching the new possible and/or regular causes of this appearance.

Determination of parameters-predictors of dendritic structure. In order to estimate features of the used formulas, it is important to evaluate correctness of those values presented in these formulas that are used for prediction of dendritic structure, and their relations with other parameters (e.g. their mutual correlation). It is shown that thermal analysis is usually (in most words, see the table 1) applied in determination of local time of solidification τ_{LS} and temperature interval of crystallization $\Delta t_{LS} = (t_L - t_S)$, where t_L, t_S are liquidus and solidus temperatures.

Cooling rate R, that is substantially varied during solidification process, is calculated as an average value using $\overline{R} = \Delta t_{LS}/\tau_{LS}$ expression; therefore, the values of parameters in such equations as (2) are connected by the relationships $m = -n$ and $K_1 = K_2(\Delta t_{LS})^{-n}$, i.e. K_1 coefficient takes into account the direct effect of alloy composition and other values, defined the Δt_{LS} value, on λ_2, based on the equation $\lambda_2 = K_1(\Delta t_{LS})^{-n}\tau_{LS}^m$. In this case the values t_L, t_S and τ_{LS} during the investigations included thermal analysis are accepted on the base of experimental data (i.e. for non-equilibrium conditions).

However, in the cases when R, V and G are evaluated via analytical calculation [18, 21, 22 et al.], usually

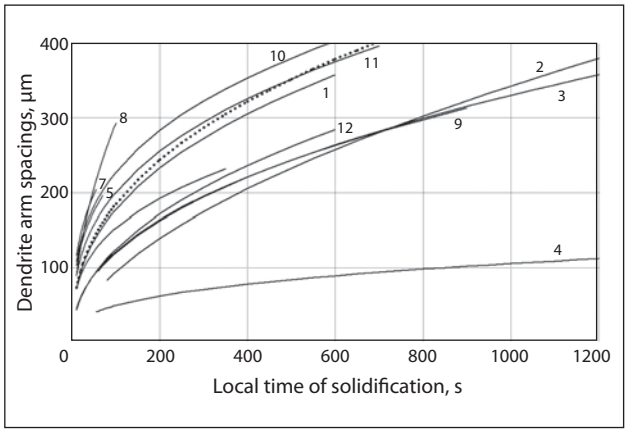

Fig. 1. Relationship of secondary dendrite arm spacings λ_2 of local time of solidification τ_{LS} according to different empiric models (numeration of coefficients 1–12 curves corresponds to table 2; dotted line corresponds to average values K_2 and m)

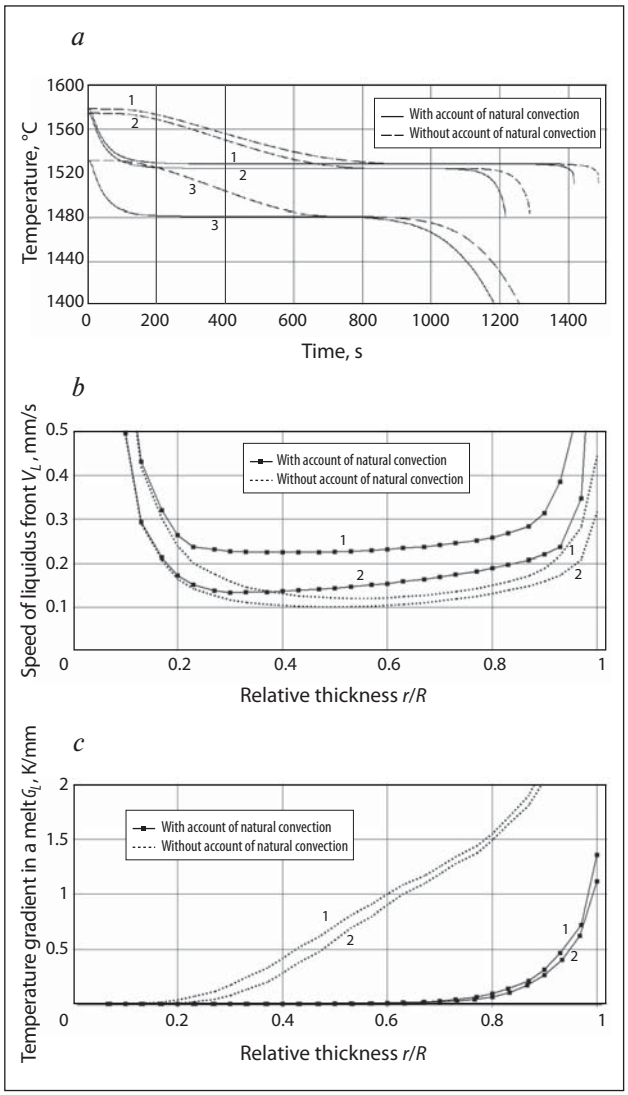

Fig. 2. Influence of natural convection on thermal curves of alloys solidification in ingot axial area (50 K overheating) (*a*), speed of liquidus front V_L in ingot cross section (*b*) and temperature gradient in a melt G_L (*c*) during solidification of ingot with thickness 240 mm of alloys Fe-C — 0.3% Si — 1% Mn with content of C, %: 0,006 (1); 0,06 (2); 0,6 (3), at α = 300 W/m²·K

equilibrium (table) values of alloy parameters are used. It is caused by difficulty of taking into account non-equilibrium features of formation of solid phase and variation of composition of liquid phase in this case; in its turn, it has effect on value of powered law parameters in the equations for λ_1 and λ_2.

Complication of experimental determination of local value of rate V_L and temperature gradient G_L in liquid phase (at liquidus front) stipulated usage of averaged values $\overline{V}$ and $\overline{G}$ for complete width of solidification area or in solid phase (V_S, G_S) instead of V_L and G_L respectively. To calculate cooling rate R, the expression $R = GV$ is often used; it leads the equation (2) to its canonical form $\lambda_1,\lambda_2 = F(V^p G^q)$ of combined forecast equations for dendritic structure [11, 17, 19, 21 et al.]. However, $R = GV$ replacement will be true for relationship of local values $(\partial t/\partial\tau = (\partial t/\partial x)\cdot(\partial x/\partial\tau))$, and can be characterized by serious errors for the case of non-equivalent averaging of generated parameters: for complete time of crystallization (for R and V) or for width of solidification area (for G).

It was noted many times in different publications [6, 21, 23 et al.] that it is possible to provide stability or independent varying of G and/or V, therefore it will be quite correct to establish correlation between primary λ_1 and secondary λ_2 of dendrite arm spacings with such parameters and to compare them with well-known theoretical formulas such as $\lambda_1 \sim V^{-0.25} G^{-0.5}$ [21–23 et al.] and $\lambda_2 \sim GV$ [18, 21 et al.]. However, in industrial conditions and at the most experimental units, G and V values often are mutually correlated. This fact is reflected on the character of obtained results, in particular it distorts relationship of exponent factors in the expressions such as (1) formula, where this relationship depends on pair correlation coefficient value.

Fig. 2 displays comparison of calculated data about temperature variation as well as V_L and G_L values in solidification of unlimited flat ingot of alloys with different carbon content; these data were obtained via numerical solution of the task [24, 25] with the constant value of heat loss (α = 300–800 W/m²·K), based on Fourier equation:

$$c\frac{\partial t}{\partial \tau} = \nabla(\lambda \nabla t) + L\frac{\partial m}{\partial \tau}, \qquad (3)$$

where t — temperature; τ — time; λ, c and L — heat conduction, volumetric heat capacity and latent heat of solidification; m — part of solid phase. Rate (tempo) of solid phase formation for multi-component alloys is determined by the expression obtained on the base of generalized Ohnaka equation of non-equilibrium crystallization [26]:

$$\frac{dm}{dt} = \frac{1}{\sum_K \frac{p_i C_i^L (1-k_i)}{1-m(1-\sigma_i k_i)}}, \qquad (4)$$

where p_i, k_i — thermodynamic parameters (slope of liquidus surface and distribution coefficient of the i-th

component of an alloy); C_i^L — content of the *i*-th component in liquid phase, calculating with account of partial diffusion behavior in solid phase [24–25]:

$$dC_i^L = \frac{C_i^L(1-k_i)}{1-m(1-\sigma_i k_i)}dm; \quad \sigma_i = \frac{2\alpha_i}{2\alpha_i+1};\ \alpha_i = \frac{8D_i^S\tau}{\lambda_2^2}, \tag{5}$$

D_i^S — diffusion coefficient of the *i*-th component in solid phase; λ_2 value is determined by kinetics of coalescence of dendritic arms [27], that will be considered in the forthcoming parts of the review. Computer-aided analysis [24] shows that crystallization process of low-alloy steel, in the conditions of joint effect of replacing components characterized by small D_i^S value, occurred with substantial deviation from the equilibrium state in critical temperatures, rate (tempo) of solid phase formation and composition of liquid phase.

Investigated Fe-C — 0,3% Si — 1% Mn alloys differ essentially via their value of temperature interval of solidification Δt_{LS} (9, 33 and 84 K at 0.006, 0.06 и 0.60% C respectively) and thermo-physical properties (heat capacity c = 4.6–4.8 MJ/m^3·K; latent heat of solidification L =1500–1840 MJ/m^3 etc.), that were preset on the base of thermodynamic modeling of equilibrium crystallization [24, 28].

Relationship between melt heat conduction coefficient λ_L and intensity of natural convection in the liquid area (with its width ΔR_L and temperature difference ΔT continuously decreased in solidification process) has been taken into account during simulation. The value λ_L has been determined on the base of calculation of convection coefficient $\varepsilon_c = \lambda_L/\lambda_0$ with assistance of the criteria equation $\varepsilon_c = 0.18(\mathrm{Gr}\cdot\mathrm{Pr})^{0.25}$ [24], where λ_0 — heat conduction coefficient without convection; $\mathrm{Gr} = g\beta\Delta T\Delta R_L^3/\nu^2$ — Grashof criterion; $\mathrm{Pr} = \nu/a$ — Prandtl criterion; g — gravity force acceleration; β, ν, a — temperature coefficient of volumetric shrinkage, kinematic viscosity and melt heat diffusivity.

In addition to known features of solidification of investigated low-carbon alloys [1, 2 et al.], thermal curves on the Fig. 2 show on substantial effect of intensity of natural convection. This effect leads to consequent decrease of initial value from $\varepsilon_c \approx 15$ to $\varepsilon_c \approx 1$ during reaching half of ingot thickness by liquidus front. As a result, variation of V_L and G_L via ingot cross section during its consequent solidification is characterized by complicated mode depending on heat loss intensity, alloy composition and conditions of melt circulation before the liquidus front (**Fig. 2** and **3**). Calculated values of V_L and G_L parameters correlate well with published results of direct measurements [4, 5, 31 et al.].

The results presented on the fig. 2 (*b*, *c*) also testify that in the case of calculated evaluation of V_L and G_L values it is very important to take into account intensive

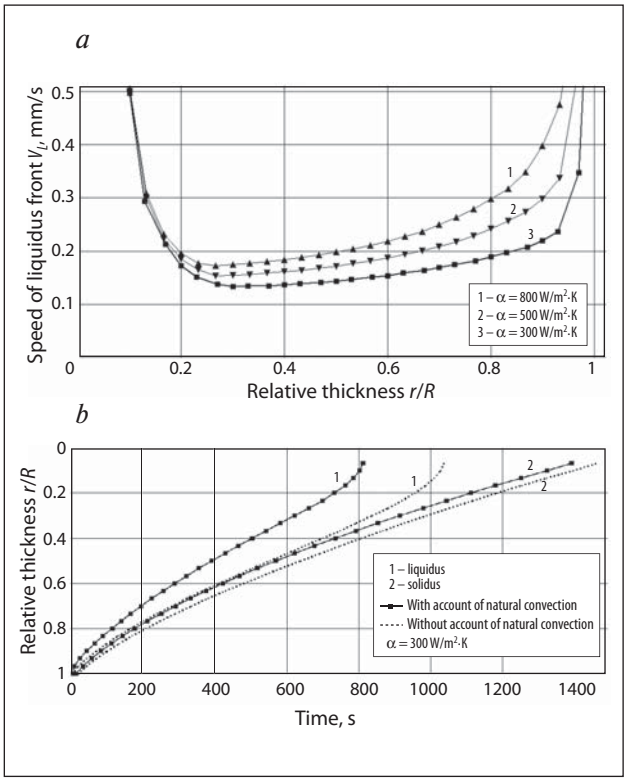

Fig. 3. Influence of heat loss intensity α on speed V_L of liquidus front (*a*) and natural convection on location of boundaries of solidification area (*b*) via cross section of an ingot with 240 mm thickness during solidification of Fe — 0.06% C — 0.3% Si — 1% Mn alloy and overheating 50 K

influence of natural and/or forced convection on their value. It can be realized as directly (via numerical hydrodynamic analysis), as well as indirectly, via introduction of efficient heat conduction ratio [24, 29 et al.], that exceeds table values of λ_L (used usually in calculations) by 10–20 times.

The results of approximate analytical calculations [18, 21, 22 et al.], where $G_L = C\cdot V_L$ value has been obtained (with evident consequences as $R = V_L\cdot G_L = C\cdot V_L^2$ equations, as well as $\lambda_2 \sim V_L^{-2n}$), have been experimentally confirmed in investigations of dendrite arm spacings for stationary conditions of solidification. However, lack of direct proportion between V_L and G_L values for the most typical and practically important conditions of flat ingot crystallization is observed; it is based on the data presented on the fig. 2 and 3, as well as revealed in [4, 5, 32].

Evaluation and adjustment of the parameters of λ_2 ***(R, τ_{LS}) empiric models.*** Essential errors of evaluation of such differential parameters as R, V and G and their mutual inconsistency that were mentioned above, determine (together with other below described factors) substantial deviation of empiric parameters (n, m, p, q, ...) of the formulas of (1) type. The values n and m (table 2) for steel are usually located in the range (0.32–0.56) [19]. It is important to mention that their

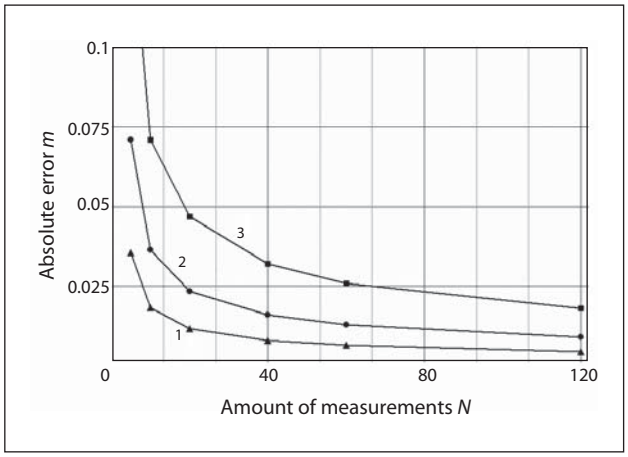

Fig. 4. Dependence of probable error Δm in determination of m parameter on amount of measurements N and s/s_x relationship. Value s/s_x: 0.025 (1); 0.05 (2); 0.1 (3)

lower boundary corresponds to theoretically substantiated λ_2 relationship from local time of diffusive coalescence [27] $\lambda_2^3(\tau) - \lambda_2^3(0) = k\tau$, that at $\lambda_2(\tau) >> \lambda_2(0)$ displays widely used final relationship $\lambda_2 \approx (k\tau_{LS})^{0,33}$. Based on this reason, in some cases the values of K_1 and K_2 ratios in empiric formulas of (2) type are estimated at preset values $n = m = 1/3$ [9, 18, 20 et al.], with acceptance of the fact that their deviation of 1/3 value is connected with errors of determination (they are usually not mentioned in publications).

Evaluated values of experimentally obtained K_2 and m parameters (table 2) should be assessed (with corrections of possible effect of alloys composition) taking into account the two important factors: used technique of τ_{LS} determination (calculation or measurement) and volume of used array N of experimental data through structure (not presented in the main part of publications). These two circumstances determine calculation error for empiric parameters.

Firstly, error occurs in linearization of (2) expression; it is connected with the fact that evaluation of K_2 and m parameters, usually obtained via the least square method, minimize the sum of deviation squares of transformed variables ($\log\lambda_2$, $\log\tau_{LS}$), but not the initial values (λ_2, τ_{LS}), what leads sometimes to necessity of corresponding corrections. Secondly, the error of measurements and calculations is rarely evaluated in publications, while the value of empiric parameters n and m, as well as K_1 and K_2 varies essentially depending on amount of primary measurements and obtained accuracy of averaged experimental data, as well as on number of conducted experiments.

It is shown in publications [33, 34] that it is possible to decrease mean square error of the model $\lambda_2 = F_1(\tau_{LS})$ compared with linearized model $\lg\lambda_2 = F_2(\lg\tau_{LS})$, via modification of the technique of statistical processing of experimental data (introduction of normalizing factor in the canonic system of equations); it is also possible to obtain substantial variation of K_2 and m parameters, what is especially important for small amount of experimental data and their non-uniform grouping.

Evaluation of precision of publishing experimental parameters of linearized equation $\lg\lambda = \lg K + m\lg\tau$ depends on amount N of measurements $\lambda_i(\tau_i)$ and error s of obtained model, that together define confidence limits of probable error Δm and $\Delta(\lg K)$ of obtained empiric values m and K [35]:

$$\Delta(\lg K) = \pm t(\alpha, N-2)\frac{s}{s_x\sqrt{N}}\sqrt{\frac{1}{N}\sum_n x_i^2};$$

$$\Delta m = \pm t(\alpha, N-2)\frac{s}{s_x\sqrt{N}},$$

where $t(\alpha, N-2)$ — table value of Student distribution quantile; α — accepted level of signification; N and s_x — total number of experimental points with known values $x_i = \lg\tau_i$ and their standard deviation $s_x = \sqrt{\sum_N (x_i - \overline{x})^2/N}$ related to the mean value of total data array $\overline{x} = \sum_N x_i/N$; s — standard error of the linear model;

$$s^2 = \frac{1}{N-2}\sum_N (\lg\lambda_i - \lg K - m\lg\tau_i)^2.$$

Fig. 4 shows variation of absolute Δm error of calculation of model m parameter depending on amount of measurements and total accuracy of approximation of experimental data, taking into account by s/s_x relationship. The less is deviation of the points relating to regression line (s parameter) and the more is expansion of investigated x values (s_x parameter), the less number of measurements is required for obtaining satisfactory results (fig. 4). Acceptable level of model error at $\alpha = 0.05$ can be evaluated by $\Delta m = \pm 0.02$, what is provided at $N \approx 20$ for typical value $s/s_x \approx 0.025$; in the case of lesser accuracy of experimental data ($s/s_x \approx 0.06$–0.008) the required amount of experimental data increased to $N > 40$–50.

Review of a row of publications shows that required parameters K_2 and m of powered law models are calculated sometimes on the base of data containing 4–6 experimental points, though for the most reliable evaluations they used arrays containing up to 70 values obtaining via averaging of essential amount (up to 150) of primary measurements [1, 21 et al.]. Statistical evaluation of m parameter in the aggregate of the data that generally characterize published powered law models of different alloys is varying for the model (2) in the range 0.17–0.48 with average value 0.34 ± 0.04 [21].

Based on calculation of the error of model parameters during its linearization and evaluation of its accuracy, it is possible to compare objectively the results of experiments of different scales and to exclude doubtful data, as well as to achieve better correlation between experimental and calculated data. To improve quality of empiric models, numerical modeling of solidification of experimental castings and ingots [11, 12, 16, 17, 20, 21, 24, 29, 30] in combination with multi-factor statistical

analysis of experimental data on their dendritic structure can be considered as the most efficient tool.

* * *

The review and comparative evaluation of published empirical powered law models for secondary dendrite arm spacings of carbon and low-alloy steels display that they are forming using a row of thermal-physical parameters-predictors (defined with essential experimental errors), are evaluated on the base of approximate relationships and are often mutually correlated. As a result, the mentioned models don't account effect of alloys composition, are not characterized in the most cases by quantitative (and/or insufficient) statistical validity and are hardly suitable for reliable predicting of dendriric structure.

Expedience of determination of reliable values of different parameters of solidification process (R, V_L, G_L and τ_{LS}) on the base of numerical simulation of thermal processes is noted as the main methodological conclusion caused by quality estimation of the published empiric models, describing relationship between parameters of dendritic structure of cast steel and the a.m. parameters. These thermal processes may be reliably evaluated at obligatory conditions of usage of parameters of investigated alloys, defined on the base of thermodynamic modeling of phase transformations during their crystallization, taking into account the effect of convective circulation of overheated melt in non-solidified part of a casting (ingot).

In the case of conventional formation of empiric models of dendritic structure, it is required to accept steps evident in their importance and aimed on forming rather complete arrays of averaged experimental data and on improvement of remedies for their statistical analysis, with publishing necessary quality evaluations of initial information and obtained models.

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