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**Кінетика кристалізації зони лазерного  
оплавлення металів**

**Crystallization kinetics of laser metals  
infusion zone**

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Запропоновано модель кристалізації лазерної ванни, в якій розглядаються конкуруючі механізми росту матричних кристалів від границі оплавлення (*g*-механізм) та утворення нових кристалів в рідкій частині об'єму (*ng*-механізм), а також враховується ефект викривлення форми та розмірів *ng* - кристалів внаслідок їх взаємодії з фронтом *g*-кристалів, що рухається, та вільною поверхнею оплавленої зони. Отримано кінетичні рівняння, які дозволяють визначити відносні внески  $x_g$  та  $x_{ng}$  двох механізмів перетворення в загальну частку  $x$  закристалізованого об'єму. Показано, що максимальне зниження об'ємної частки *ng*-кристалів за рахунок їх блокування граничними поверхнями складає 0,42.

Ключові слова: лазерна ванна, кінетика кристалізації, конкуруючі *g*- та *ng*-механізми, блокування *ng*-кристалів поверхнями.

The model of laser bath crystallization which examines the competing mechanisms of the matrix crystals growth from the fusion border (*g*-mechanism) and the formation of new crystals in the liquid part of the volume (*ng*-mechanism) has been proposed. The model is based on the following assumptions: *g*-type crystals acquire a columnar shape and predominant spatial orientation on the direction along the heat sink. They're advancing into the melt in the form of a flat front, parallel to the boundary melting, forming an independent structural zone and the share the crystallized volume  $x_g$ . The second mechanism of crystallization is realized in the bath areas free of columnar crystals. It lies in a homogeneous spherical nuclei formation and subsequent isotropic crystal growth. A homogeneously crystallized part of the laser bath consists of three zones: central (I) and two critical, one of which (II) abut on the free surface, and the other (III) – is located along the front of the matrix crystals (FMC). *ng*-crystals nucleating in the central zone do not reach the surfaces bounding the examined area of the bath and retain full length spherical shape in the growth process. Crystals that arise within critical zones II and III, are experiencing a blocking effect either of the free surface or of moving FMC, thereby fixed in the form of truncated spheres or spherical segments. The total contribution of *ng*-mechanism to share the converted volume is consist of three components. The shares of crystallized volume formed by two competing mechanisms are related to the fraction of liquid phase persisting in a bath. Within the limits of proponed assumptions the kinetic equations to calculate the value of  $x_g$  and  $x_{ng}$  are derived. It was shown that the maximum reduction of the *ng*-crystals volume fraction due to their blocking by boundary surfaces is 0,42.

Key words: laser bath, crystallization kinetics, competing *g*- and *ng*-mechanisms, blocking of *ng*-crystals by surfaces.

Статтю представив д.ф.-м.н., проф. Єжов С.М.

### Introduction

The design-theoretical investigations of crystallization processes of metallic materials undergoes the surface melting by laser emission are usually based on the model of matrix crystals sprouting from the melting boundary to the treatment plane [1 – 3]. Within the limits of this approach the impact of technological factors to the characteristic dimensions of the microstructure elements formed at the high-speed crystallization of the laser bath is analyzed [1, 2], as well as the conditions under which the melted surface layers transferred to the amorphous state [3].

At the same time, according to the metallographic studies data [4, 5], under the certain conditions of laser treatment in the cross sections of melted layers two distinct structural zones are revealed: the zone of columnar crystals adjacent to the not-fritted target area, or dendrites which are oriented along the heat-removing direction, and located above it the area of small equiaxed crystals. These results indicate that, in addition to the traditionally analyzed mechanism of the advancement of the front matrix crystals (FMC) in the melt, the hardening of the laser bath can be

realized by the formation of new crystals in the upper horizons of bath located above the FMC.

In works [6 – 8] the model of competitive crystallization of laser infusion zone by means of two co-occurring mechanisms that further will be denoted by the symbol "g" (the mechanism of matrix crystals growth) and "ng" (the mechanism of nucleation and growth of new crystals) has been proposed. The kinetic equations permitting to determine the relative contributions of analyzing mechanisms  $x_g$ ,  $x_{ng}$  in the total amount of crystallizing fraction  $x$  has been obtained.

The disadvantage of this approach is that in the calculations of  $x_{ng}$  values the effects of the form and size distortion of the new crystallization due to their interaction with the FMC and the free surface of the fused zone are not taken into account. The presented below improved model of laser bath crystallization lacks this disadvantage. It allows obtaining more correct estimates of the relative contributions of g- and ng-mechanisms in the volume fraction that has undergone the transformation.

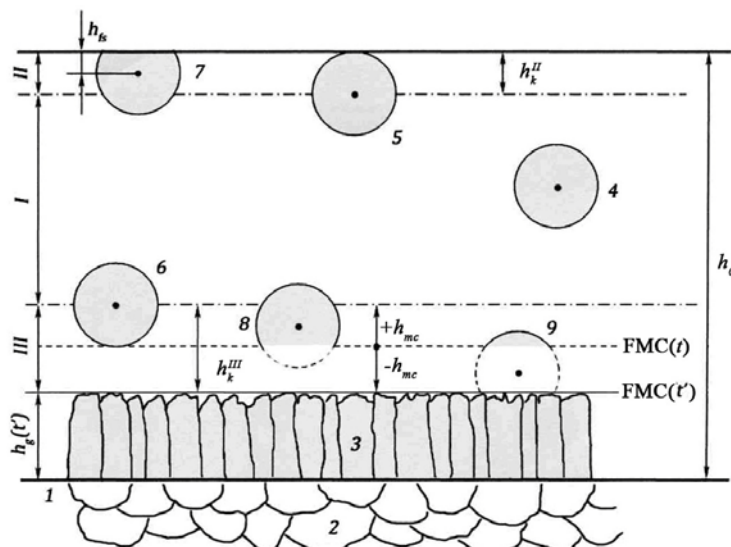


Fig. 1. The crystallization scheme of fused by laser layer: 1 – fusion boundary; 2 – partially fused target crystals; 3 – columnar crystals growing from the fusion boundary; 4 – 9 – the hypothetical spherical crystals forming in the liquid part of the zone which lies over the FMC: 4-6 – the full-sized crystals nucleated at  $t'$  in the interior (4) or at the boundaries (5, 6) of the central zone I; 7 – the truncated crystal formed in the time interval  $t' - t$  within the limits of near-surface critical zone II; 8, 9 – the crystals arising in the front critical zone III and undergoing the blocking (8) or partial absorption (9) by moving FMC.

### Physical assumptions of the model

The essence of the proposed model explains the scheme of competitive laser bath crystallization processes, which reflects the probable results of the interaction of  $ng$ -type crystals with surfaces, limiting the volume of fused zone (Fig. 1).

The constructing of the given scheme is based on the following assumptions:

1) The partially melted crystals located on the interface 1 of treated material 2 form the extensive surface where the formation of crystallization centers is facilitated, germinating in the liquid layers of a bath at cooling below the melting point  $T_m$ .

2)  $g$ -type crystals growing out of these centers acquire dendritic or columnar form and primary spatial orientation along the heat removal direction. They're moving into the melt in the form of a flat front, parallel to the fusion boundary forming the independent structural zone 3 and the crystallized volume fraction  $x_g$ .

3) The second mechanism is realized in the crystallization bath sections which are free of columnar crystals. It consists formation of the homogeneous spherical nuclei formation and subsequent isotropic crystal growth.

4) Homogeneously crystallizing part of the laser bath consists of three zones (fig. 1): the central zone (I) and two critical, one of which (II) is adjacent to the free surface, and the other (III) is located along the FMC. The crystals 4 – 6 nucleating in the central zone in the course of growth does not go beyond the surfaces bounding the considered area of baths and retain a full-length spherical form. The crystals 7 – 9 arising inside the critical zones II and III undergo the blocking action of either free surface (7) or moving FMC (8, 9), hereupon they are fixed in the form of truncated spheres (7, 8) or spherical segments (9).

5) The growth rate of columnar and the newly formed spherical crystals is characterized by the effective parameter  $u^{eff}(t)$  which is proportional to the volume fraction of the parent phase in the zones I – III lying over the FMC, and is calculated using the expression [6–8]:

$$u^{eff}(t) = \frac{1 - x_g(t) - x_{ng}(t)}{1 - x_g(t)} \cdot u(t), \quad (1)$$

where  $u(t)$  is the physical rate of crystal growth depending on the process mechanism and of thermal conditions at the interface.

6) The width of the critical zone II is specified by condition that the crystals nucleated at its boundary at  $t'$  (crystal 5 in Fig. 1), in another control moment of time  $t > t'$  reach the free surface of the bath, i.e.:

$$h_k^{II}(t', t) = R_k(t') + \int_{t'}^t u^{eff}(t'') dt'', \quad (2)$$

where  $R_k(t')$  is the radius of the critical nucleus.

The width of the zone III includes two segments (fig. 1), one of which is equal to the radius of the spherical crystal 6 growing during the time interval  $t' - t$ , and a the second is equal to the distance which overcomes the FMC for the same period of time.

It is assumed that these distances are equal and, consequently:

$$h_k^{III}(t', t) = 2h_k^{II}(t', t). \quad (3)$$

7) The total contribution of  $x_{ng}$  mechanism of new crystals formation in the transformed volume fraction is composed of three components:

$$x_{ng} = x_{ng}^I + x_{ng}^{II} + x_{ng}^{III}, \quad (4)$$

where  $x_{ng}^I$ ,  $x_{ng}^{II}$ ,  $x_{ng}^{III}$  - a volume fractions of crystals nucleated within the zones I, II and III, respectively.

8) Crystallized volume fractions  $x_g$ ,  $x_{ng}$  forming by means of two competing mechanisms are related with the fraction of liquid phase  $x_L$  remaining in the bath by relation:

$$x_g + x_{ng} + x_L = 1. \quad (5)$$

### The influence of boundary surfaces of a laser bath on the size and form of $ng$ -type crystals

Within the bounds of considered above assumptions, we derive the kinetic equations  $x_g(t)$ ,  $x_{ng}(t)$  taking into account the effects of the interaction of  $ng$ -type crystals with a free surface of a laser bath and a moving FMC. Suppose that for some moment of time  $t_m < t' < t_k$  (where  $t_m$  is the time of the melting temperature achievement  $T_m$ ;  $t_k$  – time of the crystallization completion) crystals growing from the boundary fusion penetrated deep into the melt at a distance  $h_g(t')$  and occupied a volume  $V_g(t')$ . The increment of volume fraction  $dx_g(t')$  occupied by growing matrix crystals, for a small period of time from  $t'$  to  $t' + dt'$  is:

$$\begin{aligned} dx_g(t') &= (1/V_0) \cdot S_g(t') dh_g(t') = \\ &= (1/V_0) \cdot S_g(t') \cdot u_g^{eff}(t') dt', \end{aligned} \quad (6)$$

where  $V_0$  is initial volume of the bath;  $S_g(t')$  is the area of columnar crystals front.

Assuming that the value  $S_g(t') = S_g$  is constant, by integration of the expression (6) we obtain:

$$\begin{aligned} x_g(t) &= (1/h_0) \int_{t_m}^t \left[ \frac{1 - x_g(t') - x_{ng}(t')}{1 - x_g(t')} \right] \times \\ &\times u_g(t') dt' \end{aligned} \quad (7)$$

where  $h_0 = V_0 / S_g$  is an initial depth of laser bath.

Passing to the analysis of the kinetics of  $ng$ -type crystals formation, we define the number of crystal nuclei which are formed spontaneously in the liquid part of the observable volume for the elementary time interval  $dt'$ :

$$dN(t') = V_0 \cdot x_L(t') \cdot I(t') dt', \quad (8)$$

where  $I(t')$  is the nucleation rate.

From the total number of crystallization centers we distinguish three parts proportional to the relative volumes of zones I, II, III in which they arise, and differing by final shape of crystals growing out of them. According to fig. 1 the relative volume of the central zone I is as follows:

$$\begin{aligned} K_I(t', t) &= (h_0 - h_g(t') - h_k^{II}(t', t) - \\ &- h_k^{III}(t', t)) / (h_0 - h_g(t')) = 1 - 3H(t', t). \end{aligned} \quad (9)$$

where  $h_k^{II}$  and  $h_k^{III}$  is the width of near-surface critical zones II and III defined by the relations (2) and (3);

$$H(t', t) = h_k^{II}(t', t) / (h_0 - h_g(t')). \quad (10)$$

Then the number of crystallization centres arising within the zone I during the time interval  $dt'$  can be found using the expression:

$$dN_I(t', t) = K_I(t', t) dN(t'). \quad (11)$$

As agreed above, considered centers in the growth process retains the shape of spherical particles, radius of which  $R(t', t)$  is numerically equal to the right side of expression (2). Hence it follows that the increment of crystallized volume caused by the formation of new crystals into the zone I during the interval  $t' - t$  is:

$$dV_{ng}^I(t', t) = \frac{4}{3} \pi R^3(t', t) dN_I(t', t). \quad (12)$$

By integrating (12) taking into account the relations (8) – (11) we obtain the volume fraction occupied by the  $ng$ -crystals crystals in the central zone at a given time moment  $t$ :

$$\begin{aligned} x_{ng}^I(t) &= \frac{4}{3} \pi \int_{t_m}^t [1 - 3H(t', t)] x_L(t') \times \\ &\times I(t') \cdot R^3(t', t) dt' \end{aligned} \quad (13)$$

Next, we consider the second part of the crystals which are formed in the critical zone II with relative volume:

$$K_{II}(t', t) = h_k^{II}(t', t) / (h_0 - h_g(t')) = H(t', t) \quad (14)$$

At a certain time between  $t'$  and  $t$  these crystals reach the free surface of the fused layer which causes a reduction of their final volume. Quantitatively, this effect can be estimated using the model of a crystal in the form of a truncated sphere (crystal 7 in fig. 1) and the corresponding formula for its volume:

$$V_{ng}^{II}(t', t) = \frac{\pi}{3} \times \left( 2R^3(t', t) + 3R^2(t', t)h_{fs}(t', t) - h_{fs}^3(t', t) \right) \quad (15)$$

where  $h_{fs}(t', t)$  is the distance from the free surface of the laser bath to the center of crystal, which nucleated at the moment  $t'$  in the zone II ( $0 \leq h_{fs}(t', t) < h_k^{II}(t', t)$ ).

Consequently, the increase of the converted volume per elementary time interval  $dt'$  related with the formation of truncated spherical crystals in the zone II is determined by the expression (16):

$$dV_{ng}^{II}(t', t) = V_{ng}^{II}(t', t) \cdot K_{II}(t', t) dN(t'). \quad (16)$$

Hence we obtain the expression for the calculation of the crystals contribution of II zone in a fraction of converted volume  $x_{ng}(t)$ :

$$x_{ng}^{II}(t) = \frac{1}{3} \pi \int_{t_m}^t H(t', t) \cdot x_L(t') \cdot I(t') \times \left[ 2R^3(t', t) + 3R^2(t', t)h_{fs}(t', t) - h_{fs}^3(t', t) \right] dt' \quad (17)$$

Last, the third part of  $ng$ -type crystals nucleates in the critical zone III which is located near the moving front of matrix crystals. As can be seen from fig. 1 the relative volume of this zone is:

$$K_{III}(t', t) = 2h_k^{III}(t', t) / (h_0 - h_g(t')) = 2H(t', t) \quad (18)$$

Number of nuclei formed in the considered zone for the elementary time interval  $dt'$  we find from the expression:

$$dN_{III}(t', t) = K_{III}(t', t) dN(t'). \quad (19)$$

The resulting crystal form generating in the zone III is specified by the plane, which corresponds to the position on the FMC in time moment  $t$  (fig. 1). During transformation the crystallization centers can arise on either side of this plane.

The crystals nucleating from the side of the moving front are more or less (not less than 50% by volume) absorbed by  $g$ -type crystals increasing from the fusion boundary and at the control time moment  $t$  they gain the form of a spherical segment projecting over the FMC (crystal 9 on fig.1). The crystals nucleating from the opposite side of the plane FMC ( $t$ ) with further growth are blocked by moving opposite to front with the result that they acquire the form of truncated sphere (crystal 8 in fig. 1).

Considered above types of  $ng$ -crystals interaction with FMK are influenced their final volume which can be defined by the expression:

$$V_{ng}^{III}(t', t) = \frac{\pi}{3} \times \left[ 2R^3(t', t) + 3R^2(t', t)h_{mc}(t', t) - h_{mc}^3(t', t) \right] \quad (20)$$

where  $h_{mc}(t', t)$  is the distance from the nucleation point to the finishing position of the front matrix crystals FMC( $t$ ) ( $-R(t', t) \leq h_{mc}(t', t) \leq R(t', t)$ ).

When changing  $h_{mc}(t', t)$  from  $-R(t', t)$  to 0 the formula (20) gives the segment of sphere volume and at symmetrical change in positive values  $h_{mc}$  it gives the volume of a truncated sphere. Knowing the number of crystallization centers arising in the zone III for a small period of time  $dt'$ , as well as the volume of crystals growing out of them, by the relations analogous to (16) and (17) we can determine the corresponding increment of the crystallized volume  $dV_{ng}^{III}(t', t)$  and the magnitude of contribution of considered crystals  $x_{ng}^{III}(t)$  in a total fraction of transformed volume:

$$dV_{ng}^{III}(t',t) = v_{ng}^{III}(t',t) \cdot K_{III}(t',t) dN(t') \quad (21)$$

$$x_{ng}^{III}(t) = \frac{2}{3} \pi \int_{t_m}^t H(t',t) \cdot x_L(t') \cdot I(t') \times \\ \times \left[ 2R^3(t',t) + 3R^2(t',t)h_{mc}(t',t) - h_{mc}^3(t',t) \right] dt' \quad (22)$$

As it can be seen the obtained kinetic equations (16) and (21) contain in the integrands undefined variables  $h_{fs}(t',t)$  and  $h_{mc}(t',t)$  which have the sense of the distances from the free surface (*fs*) or the front of matrix crystals (*mc*) to the center of crystals nucleating in critical zones II and III. Under the proposed model we'll assume that the changes in the considered parameters in time  $t'$  are described by the following linear relations:

$$h_{fs}(t',t) = R(t',t) \cdot (t' - t_m) / (t - t_m), \quad (23)$$

$$h_{mc}(t',t) = R(t',t) \cdot (2t' - t_m - t) / (t - t_m). \quad (24)$$

Equations (23) and (24) provide the passage by parameters  $h_{fs}(t',t)$  and  $h_{mc}(t',t)$  specified by the model of value intervals  $0 \leq h_{fs}(t',t) \leq R(t',t)$  and  $-R(t',t) \leq h_{mc}(t',t) \leq R(t',t)$  when the  $t'$  changes from  $t_m$  to  $t$ . From the physical point of view, the use of dependencies (23), (24) means that in initial time moments the crystals are nucleated on the free surface of the fused layer and at the boundary zone of matrix crystals, and at the later stages of process the nucleation points displace deep into the melt.

Taking into account (23) and (24) the equations (17) and (22) are transformed to the form:

$$x_{ng}^{II}(t) = \frac{1}{3} \pi \int_{t_m}^t H(t',t) \cdot A(t',t) \cdot x_L(t') I(t') \times \\ \times R^3(t',t) dt', \quad (25)$$

$$x_{ng}^{III}(t) = \frac{2}{3} \pi \int_{t_m}^t H(t',t) \cdot B(t',t) \cdot x_L(t') I(t') \times \\ \times R^3(t',t) dt', \quad (26)$$

where

$$A(t',t) = \\ = 2 + 3(t' - t_m) / (t - t_m) - \left( (t' - t_m) / (t - t_m) \right)^3, \quad (27)$$

$$B(t',t) = \\ = 2 + 3(2t' - t_m - t) / (t - t_m) - \left( (2t' - t_m - t) / (t - t_m) \right)^3. \quad (28)$$

For estimation comparing of relative amounts of *ng*-type crystals forming in different zones of laser baths and for the approximate determination of the total volume fraction of these crystals  $x_{ng}(t)$  we substitute the functions  $A(t',t)$  and  $B(t',t)$  for their average variation interval  $t_m \leq t' \leq t$  values:  $\bar{A} = 3$ ,  $\bar{B} = 2$ . Then:

$$x_{ng}^{II}(t) \approx \pi \int_{t_m}^t H(t',t) \cdot x_L(t') \cdot I(t') \times \\ \times R^3(t',t) dt', \quad (29)$$

$$x_{ng}^{III}(t) \approx \frac{4}{3} \pi \int_{t_m}^t H(t',t) \cdot x_L(t') \cdot I(t') \times \\ \times R^3(t',t) dt', \quad (30)$$

$$x_{ng}(t) = x_{ng}^I(t) + x_{ng}^{II}(t) + x_{ng}^{III}(t) \approx \\ \approx \frac{4}{3} \pi \int_{t_m}^t \left[ 1 - \frac{4}{5} H(t',t) \right] \cdot x_L(t') \cdot I(t') \times \\ \times R^3(t',t) dt'. \quad (31)$$

### Analysis of the kinetic varieties of *ng*-process

The volume fraction of *ng*-type crystals, defining by equation (31), essentially depends on the relative volume of critical zones II and III within the limits of spontaneously crystallizing laser bath. This value is specified by relation  $3h_k^{II}(t',t) / (h_0 - h_g(t')) = 3H(t',t)$  and changes in the

values range from 0 to 1. Taking into account the above mentioned, we consider the possible variants of dependencies  $x_{ng}(t)$ .

1. If the time-averaged values of the critical zones thickness is much smaller than the corresponding linear size of the area in which the mechanism of  $ng$ -crystallization is realized, i.e.  $\bar{h}_k / (h_0 - \bar{h}_g) \rightarrow 0$  or  $\bar{H} \rightarrow 0$ , the interaction of the newly formed crystals with surfaces limited by crystallized volume of the laser bath, can be neglected. In this case, the formation of  $ng$ -type crystals have a purely volume character, that provides the maximum  $x_{ng}^v$  contribution of this crystallization mechanism in the total share of the transformed volume:

$$x_{ng}^v(t) \approx \frac{4}{3} \pi \int_{t_m}^t x_L(t') \cdot I(t') \cdot R^3(t', t) dt'. \quad (32)$$

2. If the total thickness of the critical zones is commensurate with the length of spontaneously crystallized area, i.e.  $3\bar{h}_k \rightarrow (h_0 - \bar{h}_g)$  or  $3\bar{H} \rightarrow 1$  then at this conditions all  $ng$ -crystals are blocked by boundary surfaces, so that their total volume, and therefore the contribution to the total value  $x_{ng}$  is reduced to the minimum of  $x_{ng}^s$ . Corresponding kinetic equation acquires the form:

$$x_{ng}^s(t) \approx \frac{7}{9} \pi \int_{t_m}^t x_L(t') \cdot I(t') \cdot R^3(t', t) dt'. \quad (33)$$

As can be seen from a comparison of equations (32) and (33), the maximum relative reduction of the spontaneously crystallizing volume fraction due to the effects of interaction of crystals with the free surface of fused layer and FMC is:

$$(x_{ng}^v(t) - x_{ng}^s(t)) / x_{ng}^v(t) = 5/12 \approx 0,42. \quad (34)$$

### Summary

1. The improved model of laser bath competitive crystallization has been presented. In this model the distortions of crystals size and form spontaneously generating due to their blocking by the free surface of the fused layer and matrix crystals front moving from the fusion border are estimated.

2. Within the developed model the kinetic equations permitting to determine the relative contributions to the total crystallized volume fraction by  $g$ - and  $ng$ -mechanisms of transformation at different ratios of the volumes of the central and critical zones in formation  $ng$ -crystals regions are derived.

3. It has been shown that the maximum reduction of the  $ng$ -crystals volume fraction owing to the loss of full-dimensional spherical form due to their interaction with the boundary surfaces is 0,42. It is significantly higher than the inaccuracy of the model calculations and argues about the reasonability of using the proposed model to derive the correct numerical estimates of analyzed parameters  $x_g$  and  $x_{ng}$ .

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