MODELING OF METAL CRYSTALLIZATION BY MEANS OF CELLULAR AUTOMATON

МОДЕЛИРОВАНИЕ КРИСТАЛЛИЗАЦИИ МЕТАЛЛОВ С ПОМОЩЬЮ КЛЕТОЧНОГО АВТОМАТА

Ass. Prof., Dr. Eng. Donii O.
National Technical University of Ukraine "Kyiv Polytechnic Institute named after Igor Sikorsky" - Kyiv, Ukraine
Email: dosha@iff.kpi.ua

Abstract: Simulation model of crystallization, which is based on combination of mathematical modeling methods and a probabilistic cellular automaton, is presented here. Such combination makes it possible to predict process of metals and alloys structure formation whiles crystallization under various cooling conditions, and also to study this process both in the course of homogeneous and heterogeneous nucleation.

KEYWORDS: *CRYSTALLIZATION, STRUCTURE OF METALS AND ALLOYS, COMPUTER MODELING, CELLULAR AUTOMATON*

1. Introduction

Traditional approach to the theoretical description of metals crystallization is based on the usage of classical thermodynamics or mathematical physics methods, which use the solution of heat equation (Stefan's problem) [1]. However, such description of the processes of solid phase's nucleuses formation and of their subsequent growth has a number of significant drawbacks. Thus, nonequilibrium conditions for the processes are not considered in equilibrium thermodynamics, while they take place in the overwhelming majority of real production technologies. Besides this, calculations, which are based on the well known formula for the critical radius of the solid phase's nucleus, that was obtained with the help of Gibbs's free energy, show that the embryo consists of a finite number of atoms (3 - 600) [2]. It means that such physical object should be described with the help of quantum mechanics. Stefan's problem is not suited for describing of crystallization of melt's volume, which was supercooled at the initial moment of time (that is typical for real, as a rule - non-equilibrium, technological processes), although its development led to creation of theory of quasi-equilibrium two-phased zone, which is based on the simultaneous solution of heat conduction and diffusion equations and satisfactorily describes processes of binary melts' mass crystallization, and is rather widely used to describe various processes of obtaining ingots and castings. However, usage of heat conduction and diffusion equations to study crystallization can not provide solutions, which describe the process of solidifying metal 's structure formation, but it is namely this process of structure formation during crystallization of metals and alloys, which is of the most interest for the practitioners. Therefore, theoretical studies of the crystallization process under various cooling conditions are of current interest.

2. Preconditions and means for resolving the problem

Computer modeling is a method, which combines theory and experiment. In this work a simulation model of crystallization and investigation of homogeneous crystallization of aluminum, which were carried out with its help, are presented. The basis of the model is combination of mathematical modeling methods and of idea of a probabilistic cellular automaton, that allows to avoid significant mathematical difficulties and predict the process of metal structure formation during crystallization under various cooling conditions, and investigate the process in the course of homogeneous and heterogeneous formation of embryos.

A cellular automaton in the model is a network of elements, which can change their state (liquid or solid) at discrete moments of time.

Change of element's state occurs according to a certain law, depending on external conditions, as well as on the state of its nearest network neighbors at the previous discrete moment of time. In this model, functioning of the cellular automaton is the subject to the external cooling conditions of the investigated system and is managed by numerical solutions of the heat conduction and diffusion equations.

3. Structure and capabilities of the model

The algorithm of the model is constructed with the usage of basic techniques of system analysis by decomposing of the investigated system into separate blocks (subsystems) and determining the connections between them. In the process of investigation of a pure metal or binary alloys crystallization, we can distinguish the basic physical processes that determine course of crystallization. First of all, it is transferring of heat, which occurs in the melt's volume under the influence of external factors. It is this process, which is crucial, since the temperature gradient is one of the most important parameters in the formation of crystal's structure. Therefore, the main unit, which determines the dynamics of the model's work, is the block, which determines the temperature field during the whole process. It is based on the Fourier's equation of thermal conductivity with boundary conditions of the third kind. Processes associated with redistribution of the second component of the alloy in the liquid due to different solubilities in the metal's liquid and solid states are the second the most significant physical phenomenon in this case. To calculate these processes, the characteristic points of the corresponding double alloy's phase diagram are introduced into the model, and liquidus and solidus lines are approximated. Fick's equation is used for description of the second component's diffusion in the liquid.

The boundary conditions in this case assume absence of exchange by substances at the boundaries of the system. Taking into account the big difference of diffusion's coefficients in liquid and solid states, the redistribution of elements along the grain (dendrites), is not considered. Temperature and concentration at each melt's point determine the size of the local supercooling at this point, which is calculated as difference between the liquidus temperature for this point and its temperature at a given moment of time.

It should be noted that calculation of the second component's concentration in the liquid is not performed in the model, when crystallization of pure metal is modeled. Local supercooling in this case is defined as the difference between the metal's temperature of crystallization and given point's temperature at a given time.

In case of supercooling, conditions for crystal's formation or for growth of an already existing embryo appear in the liquid state of the melt. It is known [3] that growth of the crystal occurs under smaller supercoolings, that is, these processes differ from each other in terms of energy. Taking into account the fluctuating nature of solid-state's nuclei formation and growth of crystals in accordance with normal mechanism, which is characteristic for metals [4], in model these processes are separated and an element of randomness is built into them. For the element in which transition from liquid to a solid state is possible, the states of the nearest cells are checked. If there is at least one cell in the solid state, then it is considered that the growth of the already existing embryo is possible, otherwise the emergence of a new center is possible. To emphasize the energetical differences of the processes of crystals' formation and growth, conditional "barriers" are introduced, the size of which is determined by subcooling at a given point and is set at the level between 0 and 1.

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The cellular automato n, which implements the above mentioned scheme, is part of the general algorithm of the simulation model. Its flowchart is shown in Figure 1. The operation of the cellular automaton begins with examination of the nearest neighbors of the cell, in which supercooling exists and phase transformation is possible (block 1). After this, a decision is taken: either a new crystallization center appears, or an already existing crystal grows,

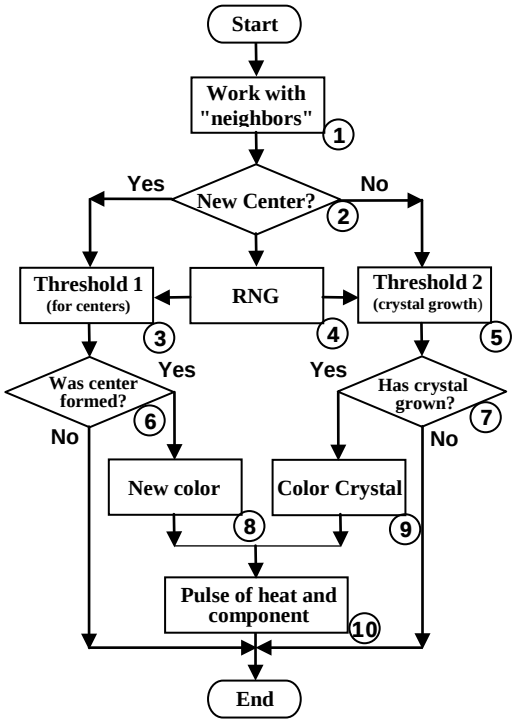

Fig.1.Fragment of algorithm of the probabilistic cellular automaton of the simulation model of crystallization.

or nothing happens (block 2). In blocks 3 and 4 conditional thresholds of transformation for a new center or for an existing crystal are determined, the values of which depend on the supercooling. After the appropriate threshold is calculated, a random number from the random number generator (RNG, block 5) enters blocks 3 or 4, and if it exceeds this threshold, a phase transformation occurs, that is, the transition of this point from liquid to solid state. Usage of RNG (Block 5) is a characteristic feature of the simulation model and allows to emphasize the random nature of the process [4].

Differentiation of colors is used to display the emerging structure of solid phase's growing grains on the computer screen. If a new embryo is formed, it is colored with a new randomly selected color. The growth of the existing crystal is supported by the color of the "solid neighbor". Black-and-white version of the image also exists, here grains' boundaries are digitally fixed according to a specially designed algorithm. Phase transition is accompanied by release of crystallization's heat and,for melts, also by second component's change in concentration at given point's region. Formation of the corresponding "impulses" is taken into account when calculating the thermal field and concentration of the second component of the alloy.

Results of simulation are as following:

- files in which the following data are recorded during the program's work: temperature in the central point at each step of the calculation, the number of centers of crystallization which have appeared and speed of their formation, the amount of solid phase and its growth rate, magnitude of temperaturesэ gradient along the system's cross section;
- picture, which is modeled by a cellular automaton, is an output window that eventually is filled with colored dots, at that color indicates belonging to a certain generation of transformed cells;
- quantitative evaluation of grain's size;
- presence of supercooling in the system and distribution of the second element's concentration along system's cross section when modeling the crystallization of the alloy.

Adequacy of the simulation model of crystallization is proved by comparing the data obtained in real and in computer experiments. Figure 2 shows results of modeling of the process of formation of structure of aluminum during crystallization. In this case, coefficients of thermal transfer are set equal on three sides and one

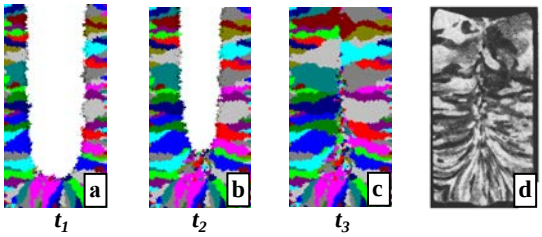

Fig. 2. Structure (a, b, c), which has been modeled in crystallization of aluminum at the moments of time (t1 <t2 <t3) and real structure of pure aluminum (d) [5].

coefficient from the top is absent.

It can be seen that front of crystallization has not flat, but diffusion character, which is typical for the normal type of crystallization. At the beginning of the process, relatively small grains appear in the corners and on the walls, and during further growth they begin to interact with each other, and some of them stop growing, while others turn into long grains directed toward the center. At the end of crystallization, small disoriented grains appear in the center of the casting. The real structure of pure aluminum is shown for comparison (Fig. 2 d).

4. Results and discussion

Before computational experiment with the simulation model of crystallization is carried out, the model has to be adjusted for modelling of the particular metal's crystallization. To do this, at first experiment with the real metal is performed, and as a result one can obtain macro structure of the metal and recorded cooling curve of the thermal analysis. Then the model's parameters are selected in such a way that the calculated results of modeling of the structure and of cooling curve coincide with the experimental ones. This is achieved by adjusting of the model's parameters, which are the following: initial temperature and temperature of calculation's end; environment's temperature (left, right, bottom, top); coefficients of heat transfer from all sides (left, right, bottom, top); specific thermal capacity; specific latent heat of crystallization; coefficient of thermal conductivity; density; geometric dimensions of the system and time interval of calculations; temperature of pure metal's crystallization and parameters of the alloy state's diagram; amount (or absence) of the modifier. There are also heuristic parameters: metastability's interval for crystallization centers' formation; maximum supercooling for crystallization centers' formation; metastability's interval for crystals' rate of growth; maximum supercooling for crystals' rate of growth. The paper presents studies of cooling rate's (V_c) influence on the process of formation of structure of pure aluminium during its homogeneous crystallization. It should be noted that in this case an ideal system is being studied, in which there are no impurities at all. During the experiments, coefficients of heat transfer were chosen to be the same from all directions: 5; 10; 20; 50; 100; 150 and 200 W/(m^2·s·K). At that, for the system with geometric parameters 0.03×0.03 m^2, the cooling rate, which was calculated from the simulated cooling curve, acquired the following values: 0.17; 0.33; 0.67; 1.67; 3.33; 5.00 and 6.67 degrees/s. The modifier in the form of already solid particles was absent. Metastability interval for crystallization centers' formation in this case was chosen to be 5 degrees, and metastability interval for growth of crystallization was 1 degree. As it can be seen from Figure 3, shape of the cooling curve begins to change as the cooling rate increases. For Vox in the range from 0.17 to 1.67 deg/s (Fig. 3 a, b, c), thermograms differ very slightly from one another and have a classical shape. With a stronger cooling (V_c = 3.33, 5.00 and 6.67 deg/s), the shapes of curves are changed significantly. Initially, a peak appears at the end of crystallization (Fig. 3 d), and

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then the recalescence of the temperature disappears at the beginning of the process and curves take on a shape with the region of almost constant temperature, which is below the temperature of crystallization (Fig. 3 e, f).

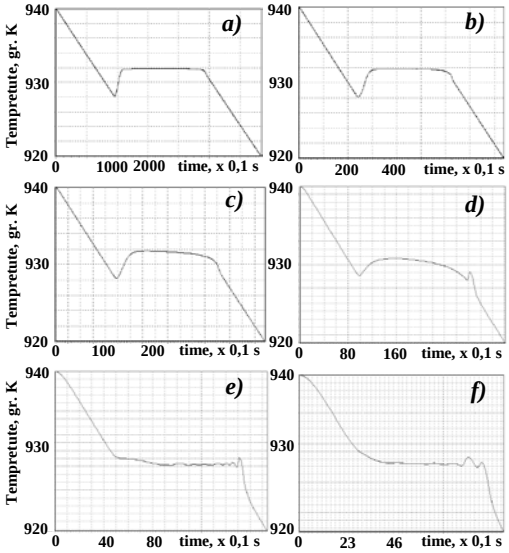

Fig. 3. Cooling curves of pure aluminum, simulated at different cooling rates : a) - 0.17 deg./s; b) - 0.67 deg./s; c) - 1,67 deg./s; d) - 3,33 deg./s; e) - 5,00 deg./s; f) - 6,67 deg./s

Temperatures' fluctuations, which are observed in thermograms (Fig. 3 d, e, f), occur also in real experiments, but in much smaller ranges. This can be explained by inertia of a real thermocouple, which has protection elements. Explainations of cooling curves shapes' changes could be done by analyzing dependence of the number of crystallization centers (Fig. 4) and the rate of their formation (Fig. 5) on time in the process of crystallization.

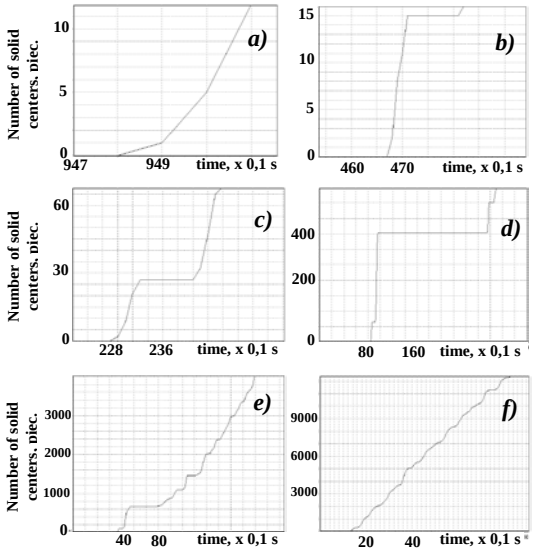

Fig. 4. Number of solids in the solid formed by homogeneous crystallization of aluminum at different cooling rates: a) - 0.17 deg./s; b) - 0.67 deg./s; c) - 1,67 deg./s; d) - 3,33 deg./s; e) - 5,00 deg./s; f) - 6,67 deg./s

At a low cooling rate of the liquid metal V_{ox} = 0.17 deg/s, the curve of the dependence of the formed crystallization centers on the time $N = f(t)$ is smoothly increasing (Fig. 4 a), the rate of their formation has one extremum (Fig. 5 a). All embryos appeared during a short interval of time (0.3 s). After centers' appearance, supercooling disappears, recalescence appears in the thermogram and then a gradual growth of crystals takes place, to which rectilinear portion of the cooling curve is corresponding ("a shelf"). The structure of a solid metal in this case consists of large almost non-oriented grains (Fig. 6 a).

Increase of the cooling rate (V_c = 0,67 deg/s) leads to appearance of a "stair" in the curve $N = f(t)$ (Fig. 4 b), and the rate of their

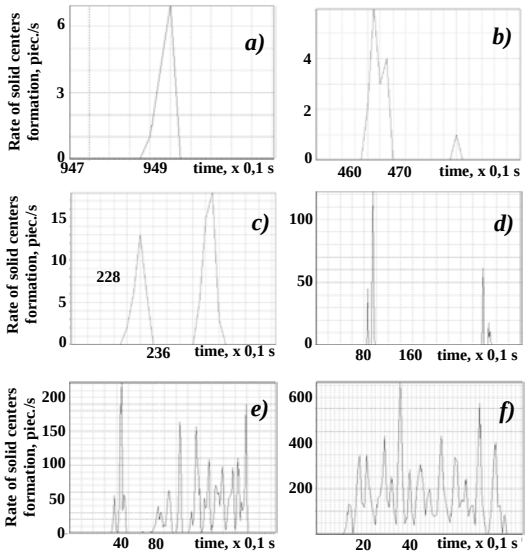

Fig. 5. Rate of homogeneous formation of solids in crystallization of aluminum at different cooling rates: a) - 0.17 deg./s; b) - 0.67 deg./s; c) - 1,67 deg./s; d) - 3,33 deg./s; e) - 5,00 deg./s; f) - 6,67 deg./s

formation has already two peaks (Fig. 5 b). That is, centers of crystallization arise in two stages: at the beginning of the process as well as its end. At that, changes in the cooling curve are insignificant. The temperature of the "shelf" decreases slightly (by a tenth of a degree) and, before the end of crystallization, temperature decreases in a slightly slower rate (Fig. 3 b). The structure of the solid metal remains practically unchanged (Fig. 6 b), although there are almost invisible small undirected grains in the center.

With a subsequent increase of V_c up to 1.67 deg/s, the total number of formed centers solid phase increases, and the "stair" on the curve of their time dependence becomes well noticed (Fig. 4 c). The rate

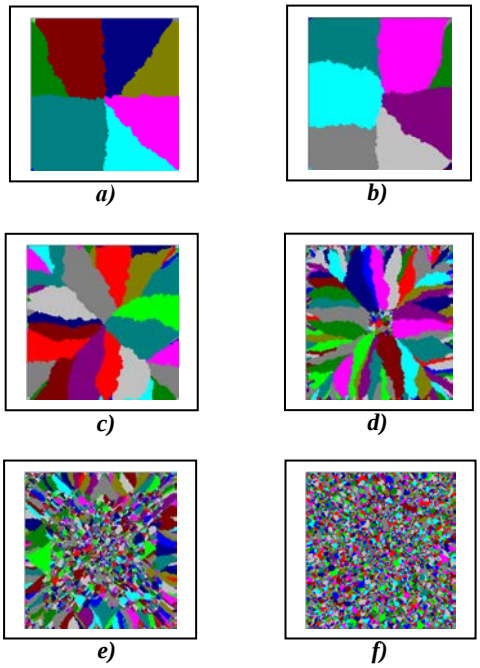

Fig. 6. Macrostructures of pure aluminum, modeled at different cooling rates: a)- 0.16 deg./s; b) - 0.67 deg./s; c) - 1,67 deg./s; d) - 3,33 deg./s; e) - 5,00 deg./s; f) - 6,667 deg./s

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of centers' formation has two brightly visible peaks (Fig. 5 c). Changes in the cooling curve become more noticeable. Thus, the temperature of the "shelf" decreases before the end of crystallization (Fig. 3 c). The structure of the solid metal is substantially crushed (Fig. 6 c) and the grains become more elongated toward the center of the system.

Further growth of the cooling rate (V_c = 3,33 deg/s) leads to the appearance of several "stairs" on the curve $N = f(t)$ (Fig. 4 d). Two peaks that were observed in the previous case on the curve of dependence of rate of embryos' formation begin to split up, and we can see already four extrema (Fig. 5 d). Thus, the process of formation of crystallization centers becomes multistage. Significant changes are observed inn the cooling curve: the value of recalescence decreases, the temperature's "shelf" practically disappears, the temperature before the end of crystallization decreases even faster. The structure of the solid metal becomes three- zoned (Fig. 6 d). Small grains are observed on the form's conditional walls (at the edges of the system), some of them are suppressed by other crystallites. Then a zone of crystals, which are elongated and oriented to the center, is formed. There is a small zone of non-oriented grains in the center.

A significant increase of the cooling rate (V_c = 5.00 deg/s) leads to the appearance of many "stairs" in the curve $N = f(t)$ (Fig. 4 e). Actually this dependence becomes "wavy" by nature. This is clearly seen also in the curve of dependence of nuclei rate's formation (Fig. 5e). There are a lot of formed peaks and the process of crystallization centers' formation becomes almost periodic. Although at the beginning of crystallization, formation of centers has, as before, an explosive character. The shape of the cooling curve changes in principle: recalescence disappears, the temperature's "shelf" appears again, but its temperature decreases and is actually equal to the temperature of crystallization minus the interval of metastability for embryos' formation (Fig. 3 e). The structure of the solid metal becomes grained differently (Fig. 6 e). Larger grains are observed near to the mold's conditional walls, and there are smaller ones in the center of the casting.

The next increase of the cooling rate V_c to 6.67 deg/s emphasizes the effects observed in the previous case. Dependence $N = f(t)$ has a "wavy" character (Fig. 4 f). The process of crystallization centers' formation becomes quasi-periodic and continuous in time (Fig. 5 f). The shape of the thermogram has an almost straight lane corresponding to the crystallization of the liquid metal. But at the beginning of crystallization the form of the cooling curve becomes smoother, which is explained by the effect of the temperature gradient across the cross section of the system at already sufficiently high cooling rate (Fig. 3 f). The structure of the solid metal becomes finely granular (Fig. 6 f).

The obtained results of the changes in the cooling curves, dependences of the number of crystallization centers and of their formation's rate on time, and the structure of the solid metal, which are observed in the computational experiment, can be explained as follows. In the case of slow cooling, nucleus of the solid phase appear after the metal's temperature becomes lower than the temperature of crystallizationby (T_0) by value of interval of metastability (ΔT). At that, the heat of crystallization is released. The rate at which this heat is released significantly exceeds the rate of heat transfer, that results in system's temperature increase almost to a temperature, which is below T_0 by the value of metastability's interval for crystals' growth ΔT, which is close to zero (i.e., the supercooling value is about ~ 0.1 degrees). With this supercooling new crystallization centers do not appear and only already formed crystals grow. After the end of the phase transformation, the metal is simply cooled and its temperature decreases gradually.

In the case of faster cooling, temperature in the whole system or in its parts decreases after appearance of the first formations of the solid phase, in spite of the released heat of crystallization, due to sufficiently rapid cooling. After a certain time, supercooling occurs and conditions appear for the formation of new nuclei of the solid phase. Their appearance and corresponding releasing of crystallization's heat reduces the supercooling and growth of already existing crystals continues. Such processes can occur either one or several times, after that crystallization ends and the metal simply is cooled. Formation of the classic three-zoned casting's structure can be explained by formation of the nucleation of the solid phase in several stages (Fig. 6 d). Centers of crystallization appear at two stages: at the beginning of crystallization and at the stage which corresponds to formation of the central zone of equilibrium crystals.

When the system is cooled at high speed, the rapid heat removal does not give the metal's temperature to rise despite release of crystallization's heat. Conditions are created for the simultaneous formation of crystallization's centers and for the growth of already existing crystals, as Figures 4 f and 5 f demonstrate.

5. Conclusion

1. The simulation model of crystallization is presented, which is based on the combination of cellular automataton with the classical problems of heat conduction and diffusion. This model allows us to investigate the process of the metal structure formation during crystallization and the dynamics of this process under various cooling conditions.

2. Computer experiments which were carried out on modeling of homogeneous crystallization of aluminum showed:

- formation of all the centers of the solid phase at a cooling rate of 0.17 degrees/s occurs in a short time interval (about 1 s) and then centers' further growth occurs;
- with an increase of the cooling rate, the centers of the solid phase are formed in two stages - at the beginning of crystallization and in the second half of the process; this leads to the formation of three-zoned structure;
- at a cooling rate of 5 degrees/s and higher, the solid phase's centers are formed over a long period of time, their growth proceeds in parallel, and the structure of the solid metal becomes finely granular.

6. Literature

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