Crystallization of aluminium alloys under pressure

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Abstract. The paper presents the results of experimental studies helping to reveal the laws of the crystallization process of aluminium alloys under conditions of imposing pressure, reaching 300-400 MPa, ensuring the formation of a structure without shrinkage defects and an increase in density. Practical recommendations are given for automation of pressure overlay control and for creating metallurgical mini-productions based on a new process.

1. Introduction
Over the past 10 - 15 years the annual growth of primary aluminium production averaged about 3% in Russia. The domestic market of aluminium alloys products in 2017 amounted to 1.4 million tons, 800 thousand tons of which is due to Russian-made alloys.

The government of the Russian Federation is planning to expand the use of aluminium in the automotive industry, engineering, construction and in other industries.

Due to the impending man-made problems and the need to protect the environment a process of crystallization under pressure for the production of Al-alloys is proposed as an alternative to the well-established technologies.

Deformable high-strength alloys of aluminium have a complex of chemical and physicomechanical properties similar to those based on Cu, Fe, Ti, and their crystallization under pressure can increase the utilization rate of the liquid metal to 0.8 ... 0.9.

2. Research results
At the Department of Technological Process Automation of Vladimir State University, the crystallization process is studied using original techniques on the equipment with built-in instrumentation and control on a computer platform system [1-3].

The principal difference of the developed process scheme from injection moulding and liquid stamping is that the pressing plunger overcomes the resistance of the layer of crystallized metal under the pressure of a hydraulic press and exerts pressure on the liquid non-crystallized metal from the inside of the heat centre of the casting.

Such a scheme of applying pressure (figure 1) provides full compensation for shrinkage, and the development of crystallization from a precompressed metal has not been studied till the present moment and is of independent interest.
Contrary to the laws of physics, molten metal under the influence of pressure behaves not like an "incompressible fluid", but shows the properties of a complex rheological body. These properties include:

1 - elasticity; 2 - compressibility; 3 - the ability to "tighten" the atoms in a compact volume.

On the qualitative side, these properties are detected when deciphering the computer oscillograms presented in figures 2 and 3.

Thus, the curve 1 (figure 2) shows that the right pressing plunger (figure 1) enters the liquid metal to a depth of 100 mm exponentially, while the left plunger traced by curve 2 passes 125 mm linearly and stops, as the piston, on the rod of which the plunger is mounted, abuts against the wall of the hydraulic cylinder. Curve 3 indicates that the pressure in the cylinder cavity rises gradually according to the dependence close to linear, up to ~ 8 MPa. During the first 5 ... 6 seconds, and then it grows slowly,
while the right plunger continues to compress the crystallizing metal. The relative decrease in the volume of the liquid metal, i.e. the compressibility of the metal under pressure reaches 12.5 ... 13%, which is significantly more than the volume shrinkage, which is 7 ... 9%. It is obvious that the moment of stopping the pressing plunger corresponds to the end of crystallization. Taking into account the force developed on the pressing plunger, the maximum pressure on the metal being crystallized reaches ~ 350 ... 400 MPa.

For comparison, figure 3 shows the same dependences 1 and 2 set for the conditions of pressure imposing in the mode of pulse modulation with parameters: 10 s pressure is on, 1 s it is off.

![Figure 3. The cyclogram of the plunger movement and pressure growth in the system in the mode of pulse modulation.](image)

It was assumed that at the moment of shutdown the plunger would stop, and the result of several stops would be an incomplete “crimping” of the metal.

It turned out that at the moment of shutdown, the right pressing plunger (1) performs a “rebound”, the value of which is probably proportional to the volume of the non-crystallized metal, with an increase in the number of cycles, markedly decreases.

If we consider that for the reverse displacement of the plunger, an effort of ~ 2000 H is necessary, then we can rightly assume that the forces of mutual repulsion of atoms are large enough, therefore, the liquid metal manifests itself as an elastic body.

By the nature of the left plunger movement (2), which squeezed the metal on the opposite side, it can be seen that the movement in the direction of compression of the metal occurred with a sharp jerk. It is possible that during the stopping of the plunger in the vicinity of its end, the crystallizing metal, due to shrinkage, created a vacuum that overcame the resistance of the jumble of atoms and drastically pulled the plunger inward. This showed the ability of the crystallizing metal to "pull" atoms into a compact volume and at the same time overcome the emerging resistance. This gives rise to the idea that in the absence of external pressure, not all the nodes of the atomic-crystalline lattice are filled. The ideal in this sense is the assembly of a lattice of individual atoms. Practically, it is possible to carry out external pressure testing of atoms, creating conditions for organizing for each atom the maximum possible number of bonds with other atoms and forcing out “empty spaces” to increase the overall density of the metal.

Curve 1 in figure 3 shows that in the period of the on state, there is a “catch-up” of dependence to the level characteristic of continuous pressure application (figure 2). Recovery of dependence does not occur smoothly, although the pressure in the hydraulic system is restored instantly. Probably,
crystallization of a certain volume occurs within ~ 5 seconds, a vacuum is created, which draws in a pressing plunger, and when the “lag” behind the shrinkage compensation ends, the plunger continues to move under the influence of the force created by the hydraulic cylinder.

The fact that the intermittent imposition of pressure provides the effect of compensating shrinkage, which is not different from the continuous imposition of pressure, suggests that the decrease in volume at the moment of crystal formation plays a leading role in the process. When a “vacuum” has arisen in the arrangement of atoms, external pressure can fulfill the role of delivering other atoms into the vacated space. In order to form the convergence of atoms and their mutual arrangement in the crystal lattice, either a higher pressure is necessary, or this is impossible in principle. In other words, it should be recognized that the law of shrinkage compensation, i.e. the volume of metal pressed into the volume of the casting depends on the rate of crystallization. In turn, the rate of crystallization is a complex function of a number of factors, of which the most significant is the rate of cooling. There is neither mathematical model that correctly takes into account the boundary conditions, nor the dependence of the thermophysical properties on temperature and pressure. But, if the pressure on the crystallizing metal is superimposed by pulses, it is possible to eliminate the influence of such a factor as the resistance of elastically compressed non-crystallized atoms.

The oscillograms shown in figures 2 and 3 give the experimental information necessary to control the crystallization process according to a mathematical model, which is the following system of differential equations:

$$\frac{dT}{d\tau} = \left(\frac{\partial^2 T}{\partial x^2} + \cdots\right) + LV_p \cdot \rho$$

$$\frac{dp}{d\tau} = \frac{E \cdot dV}{V_0 \cdot d\tau}, \frac{dp}{d\tau} = \varepsilon \cdot E$$

$$M \frac{d^2x}{d\tau^2} = F \Delta p - \lambda \frac{dx}{d\tau} - R$$

$$\frac{dp}{d\tau} = \frac{E}{V} (K_f \Delta y_l - F \frac{dx}{d\tau})$$

$$Q = \mu y_l \sqrt{2(p_{in} - p_1)}$$

Equation (1) subject to the description of the boundary and initial conditions from experimental data can be used to determine the crystallization rate $V_{cr}$. Equation (2) in the measured relative volume change of the liquid metal under pressure is used to calculate the elastic modulus of the liquid metal $E$.

Equations (3) - (5) give an idea of the hydraulic system operation in dynamic modes.

In the system of equations (1) - (5) the following notation is used: $t$ is temperature; $\tau$ is time; $x, y, z$ are coordinates of the metal being crystallized in the volume; $L$ is the latent heat of crystallization; $\rho$ is the density of the metal; $M$ is the mass of the moving parts of the hydraulic cylinder; $r$ is movement of the plunger; $F$ is the piston area; $\lambda$ is the coefficient of friction in the hydraulic drive; $R$ is the force acting on the liquid metal; $p$ is pressure; $V$ is the volume of the liquid metal; $R$ is the force acting on the liquid metal; $K_f$ is the force ratio of the pressure regulator; $\mu$ is the coefficient of the regulator flow; $l$ is movement of the regulatory body; $p_{in}$ - pressure at the inlet to the regulator; $p_1$ is the backpressure in the hydraulic system.

This system of equations, according to well-known rules [4-6], is converted into a block diagram, and then into a control algorithm.

Based on the experimentally established dependency analysis, the following practical recommendations were developed on castings made of B95 alloy with dimensions of $\Theta 90 \times 70$ mm:
The location of the casting in space and with respect to the pressing plunger must meet the following requirement: pressure is applied from the side of the liquid metal along the crystallization front, which, moving directionally, passes the entire internal cavity of the casting.

- Apply pressure on the liquid metal in three stages: the first is elastic compression, \( \varepsilon = 5 \ldots 7\% \), pressure on the metal \( p = 50 \ldots 70 \text{ MPa} \); the second is elastic-viscous compression, \( \varepsilon = 5 \ldots 7\% \), \( p = 100 \ldots 150 \text{ MPa} \); the third is viscous compression, compensating shrinkage, \( \varepsilon = 1 \ldots 1.5\% \), \( p = 200 \ldots 250 \text{ MPa} \) and more.

- Pressure is used in impulses; the duration of the on state and pauses should be chosen so as to ensure the relative compressibility \( \varepsilon = 12 \ldots 13.5\% \).

The use of pressure on the liquid metal at a temperature 100 ... 150 K higher than the temperature of the onset of crystallization, turns the metal as a thermodynamic system to a highly non-equilibrium state. It is not possible to determine the values of thermodynamic functions, to establish the influence of variable parameters, such as pressure, temperature, time, composition of the alloy and other factors on the structural and phase transformations.

However, if the crystallizing metal is considered as a multidimensional object, when the input has the listed controlled and controlling parameters, and the output has physical and mechanical properties, then there will be variables inside the control object. Then, analytically, using experimental data, one can trace the passage of the system along a certain trajectory in the space of variable states.

Under the influence of input parameters, the mechanism and kinetics of phase transformations change.

The microstructure of the alloy, which is formed in the process of crystallization under pressure, is shown in figure 4. Significantly that the grains, almost the same form of the main phase, are surrounded by layers of other phases. X-ray diffraction analysis and OGE spectroscopic analyzes indicate that the grains of the main phase are \( \alpha \)-solid solution, in which the concentration of Cu, Mg, Zn depends on the distance from the center of the dendritic cell to its boundary. As for the interlayers, these are excessive intermetallic phases.

![Figure 4](image)

Figure 4. The microstructure of B95 alloy, which is formed in the process of crystallization under pressure.

In the casting section with a diameter of 80 mm, no differences were found in the size of the grains and in the nature of their relative position. This suggests that the mechanism of crystallization under pressure is that in the process of elastic and subsequent elastic-plastic compression, accompanied by removal of superheat heat and latent heat of crystallization, centres of the most refractory phase appear, which, expanding, push to the boundaries of the phase with more low temperature of the beginning of crystallization. Therefore, during time crystallization develops at a rate proportional to the rate of heat removal, and in space - according to the laws of directional crystallization, but with signs of bulk crystallization. Probably, the volume compression of atoms on the number of centres and the growth rate of crystals has a stronger effect compared with the degree of supercooling. Therefore, the sizes of
dendritic cells and their shape both in the centre of the casting and in the periphery do not have significant differences.

To conduct experiments with the imposition of pressure on the crystallizing metal quite massive technological equipment is used. The wall thickness of 5HNM steel matrix is 60 mm. The temperature of the matrix before pouring the liquid metal at a temperature of 800 ... 850 ° C was 200 ... 250 ° C. The rate of cooling of the metal in the range of temperatures of decomposition of the solid solution 650 - 100 ° C was ~ 1 ... 5 K/s. It is possible that the separation or the formation of excess phases occurred precisely in this temperature range.

The casting process with crystallization under pressure, combined with heat treatment (CCP + HT) is of interest.

To do this, it is necessary to transfer the casting to the furnace immediately after opening the mold, homogenize it at a temperature of ~ 600 ° C, and then fulfill quenching and aging. The temperature and duration of soak during aging should be assigned taking into account the required strength and plastic properties.

The processes of CCP, CCP + HT as representatives of mechanic-thermo-temporary processing hybrid technologies of parts from high-strength aluminium alloys have significant advantages and high innovative potential.

The ability to respond flexibly to market conditions, combined with the high efficiency of the use of metal, create prerequisites not only for mastering new types of products, but also to increase competitiveness in the production of mass and high-volume products.

3. Conclusion
The development of technologies for the metal production from high-strength aluminium alloys based on the casting process with crystallization under pressure is of particular importance in Russia for a number of reasons, among which the main is the possibility of replacing high-temperature alloys based on Cu, Fe, Ti.

Deepening the processing of aluminium to the level of production of parts for aircraft, automobiles, gas-pneumatic-hydraulic distribution valves, etc. is necessary for changing the flow in favour of domestic consumption and extracting maximum profit from this natural resource.

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