Mathematical modeling of heat and mass transfer processes in the graphite thermal unit of the crystallization apparatus for Horizontal directional solidification method

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Abstract. The design of ECU has been created, which allows in the process of growing halide crystals to create in the growth and annealing zones, respectively, the high-gradient and low-gradient temperature regions necessary for each of the processes. The mathematical study of the influence of the processes of hydrodynamics, heat and mass transfer inside ECU, between ECU and the crystallization apparatus, is considered. For numerical calculations, the Navier-Stokes equation, the Menter differential k-ω turbulence model SST, the modified Stefan-Boltzmann law, the Fourier law are used. To describe the mechanism of heat transfer and heat conduction in solids, boundary conditions of the third kind were set. The distribution of the axial temperature gradient inside the carbon-graphite heat unit was studied. The mode of flow of the refrigerant inside the cooling jacket was determined. The radiative heat transfer between the surfaces, and the mechanisms of heat transfer and heat conduction in solids were studied.

1. Introduction
The development of an energy-efficient unit is an urgent task for crystallization apparatus operating by the method of horizontal directional solidification method created by Kh.S.Bagdasarov (HDS). To improve the thermophysical characteristics of such equipment, it is necessary to create new designs of thermal units, taking into account both advanced technologies for their production and the use of high-quality materials. Promising carbon-based materials are described in the papers [1–6], relaxation processes in such materials are of particular interest [7–12]. HDS method was developed exclusively for the synthesis of oxide crystals, mainly leucosapphire in energy-consuming tungsten-molybdenum units [13, 14]. This work is devoted to the development HDS method for growing large-sized fluorides single crystals in an energy-saving carbon-graphite thermal unit (ECU) placed in a steel water-cooling jacket (SWJ) of the crystallization apparatus.

The method of constructing ECU assembly is fundamentally different from the calculation methods and design of tungsten-molybdenum assemblies [13,14]. In this regard, the authors proposed a new principle for the design and layout of ECU in Rubitek crystallization apparatus.
This unit allows you to create a high gradient region in the crystal growth zone and the low gradient temperature region in the crystal cooling zone. This technique is so important for obtaining optically perfect fluoride and other halide crystals, which have widely spread using. In particular, this type of crystals are used as scintillators for ionizing radiation detectors, laser crystals, and elements optical instruments operating in a wide spectral region from ultraviolet to medium infrared wavelength range \[15\]-\[17\].

Figure 1. a) – thermal protection element of ECU; b) – Rubitek crystallization apparatus; c) – 3D model of ECU in Rubitek crystallization apparatus;

ECU model is distinguished by its high technological design. ECU consist of individual carbon-graphite heat-insulating modules. They are easily assembled with each other according to the “LEGO” principle, forming the integral construction of ECU. Using this technique the ECU is the form of a total heat-insulating volume which allows reducing the total costs of the crystallization process by growing crystals without additional annealing \[17\].

In order to carry out theoretical studies regarding heat, mass transfer and the distribution of thermal field inhomogeneity in the construction of ECU placed in Rubitek crystallization apparatus...
apparatus (channel heat exchanger), the authors attempted to perform computer simulation and analysis of heat transfer of ECU - SWJ crystallization apparatus complex.

The real used models of the carbon-graphite thermal unit and Rubitek crystallization apparatus were adopted as objects of research [17–19].

Computer modeling of the studied objects was carried out using the SolidWorks Flow Simulation software module, which was created using the finite element method. The use of this program was explained by the fact that the Flow Simulation package in SolidWorks applies full integration and it is possible to model the geometry and perform all calculations and analyzes “in single window” [18,19]. This feature eliminates the possibility of errors of import - export of geometry through an intermediate data format (for example, .xt, step, sat, iges, etc.) [18,19]. Moreover, SolidWorks has a standard Windows graphical interface and successfully interacts with Windows applications, such as Excel, Word, etc. [18,19]. Various tasks related to issues of hydrodynamics and heat transfer in SolidWorks Flow Simulation are modeled using a system of differential equations of motion, continuity, energy, and thermal conductivity of the walls [18,19].

The SolidWorks Flow Simulation package uses the standard stages of developing a mathematical model [18–20] – these are:

- Creating a 3D model;
- Grid calculation area;
- Imposing boundary conditions;
- Visualization of temperature, pressure, etc.

2. Experimental part

The studied object is ECU which consists of heaters, protective shield of various materials and a graphite crucible with a grown single crystal. The temperature field in this design is determined by the processes of thermal conductivity and re-radiation between the surfaces. The volume in which carbon-graphite thermal units are placed is filled with vacuum. Outside, this design is placed in the channel heat exchanger (“water-cooling jacket”), through which distilled water circulates as the heat carrier.

The crystal with a CaF$_2$ fluorite structure, known as an optical material with a wide transparency window from 0.15 to 7.5 µm and which is the matrix for numerical laser crystals, was chosen as a target crystal [21–23]. The main thermophysical characteristics of the CaF$_2$ single crystal are shown in Table 1 [24].

During the experiment, the temperature on the heaters was set based on the particularities of the CaF$_2$ crystal growing method; for example, the temperature for HDS method was set at 1550 °C on the lower heater and 1650 °C on the upper one (to control the angle of the vertical temperature gradient) [13,15–17]. The temperature of the coolant at the inlet was 20 °C at the pressure of 202.6 KPa and head speeds of 2 m/s. The ambient temperature acting on the heat exchanger was also equal to 20 °C at 101.3 KPa.

| Table 1. Basic thermophysical characteristics of CaF$_2$ single crystal |
|-----------------|-----------------|
| **Density**     | 3.18 g/cm$^3$   |
| **The melting temperature** | 1418 °C        |
| **Coefficient of thermal conductivity** | 9.71 W/(m/K)   |
| **Coefficient of thermal expansion** | 18.5 x 10$^{-6}$/ |
| **Knoop hardness** | 158.3 kg/mm$^2$ (100) |
| **Specific heat** | 854 watt second |
| **Emissivity (degree of blackness)** | 0.45-0.70 (on carbon) |
The geometric model of ECU and Rubitek crystallization apparatus was built on the basis of the drawings and structural layout schemes, which are presented in the public domain [15–17,25]. Fig 2 shows the design of the ECU which consists of three main modules. The first one is the heating block alias the central module that includes the crystal growth zone and after growth annealing. The second one is loading zone alias the loading module, and the third one is the unloading zone alias the receiving module. These modules include thermal screen systems and peripheral thermal insulation. The heating unit also contains a system of heaters. The system of heat shields, consisting of interchangeable, easily replaceable elements, makes it easy to pump the heat unit to a high vacuum of $10^{-5}$ mm Hg. The heating unit includes two heating elements - upper and lower heaters, peripheral thermal insulation serves as additional thermal protection for entire SWJ crystallization unit. For the manufacture of removable heat shields and heaters, fine-grained isostatic graphite of the German company SGL Carbon of the R4550 brand was used, and soft graphite felt (GV) for vacuum furnaces was used as heat-insulating material. The heating unit also includes both additional modules. The first one (left-wing) is the horizontal mine of the crucible loading module which carries the charge for the growing crystal. And the second one (right-wing) is the horizontal mine of the crucible receiving module which carries growing crystal. Also this mine (right-wing) has a function of annealing a growing crystal at the same time. The heating module has removable thermal diaphragms which create the necessary axial temperature gradients in a melt crucible in the time when the crucible passing under them. Also the heating module provides the necessary shape of the isotherm of the crystallization front of the growing crystal. Basic requirements for the design of the thermal unit:

- evacuation of the installation to a pressure of $5 \times 10^{-5}$ mm Hg
- maximum temperature on heaters up to 1800$^\circ$C
- values of direct currents on heaters no more than 800 A
- values of constant voltage on heaters no more than 30 V
- current density on heaters no more than 2 A/mm$^2$

The list of thermophysical parameters of the basic materials used in the calculations is given in Table 2 [26].

Figure 2. The design of ECU; 1) - the heating block alias the central module; 2) - the crucible loading module (left-wing); 3) - the crucible receiving module (right-wing);
Table 2. The list of thermophysical parameters of the basic materials used in the calculations

| Material                       | Thermal conductivity $\lambda$ [W/m $\times$ K] | Heat capacity $\rho$ [watt second/kg $\times$ K] | Density $\rho$ [kg/m$^3$] | Emissivity factor, $\varepsilon$ |
|--------------------------------|-------------------------------------------------|-------------------------------------------------|---------------------------|-------------------------------|
| Steel, 5140 1040               | 46                                              | 460                                             | 7874                      | 0.4                           |
| Steel, A 659 CS Type 1020 A 794 CS Type 1020 | 55                                              | 460                                             | 7874                      | 0.4                           |
| SGL Carbon R4550               | 105                                             | 840                                             | 1830                      | 0.81                          |
| Carbon, PG-8,7                 | 95                                              | 840                                             | 1850                      | 0.8                           |
| Carbon felt, GV                | 0.3                                             | 1000                                            | 97                        | 0.9                           |
| SGL Carbon SIGRAFLEX           | 5                                               | 900                                             | 700                       | 0.115                         |
| SGL Carbon SIGRABOND           | 13                                              | 800                                             | 1450                      | 0.8                           |
| Tungsten                       | 129                                             | 129                                             | 19300                     | 0.04                          |
| Copper                         | 386.1                                           | 400                                             | 8950                      | 0.7                           |
| Molybdenum                     | 138                                             | 195                                             | 10220                     | 0.3                           |

3. Results and discussion
3.1. Computational mesh
The curvilinear block-structured base mesh was formed after constructing the three-dimensional model of the crystallization apparatus. This curvilinear block-structured base mesh divides the physical volume into a number of isolated blocks. The resolution of the local grid was selected based on the characteristics of the studied area for each isolated block. Fig. 3 shows the computational mesh for the crystallization apparatus including ECU.

![Figure 3. Computational mesh for the crystallization apparatus including ECU](image-url)

The total number of cells of the base mesh used was 4721500, including a cell in a fluid medium 1036504, cells in a solid medium 3684996 and cells at the interface between a solid and
a fluid 1036504. The dimension of the base grid \( N_x = 36, N_y = 30, N_z = 70 \). The resolution of the local grid in the selected region was (in this calculation, the region of the crystal with the crucible) 446727 cells.

### 3.2. Numerical procedure

The full-size model of ECU placed in the Rubitek crystallization apparatus was considered. The bodies were assumed to be opaque, and the “gray body” approximation was used in radiation heat transfer in this model. Heat from heated graphite heaters is absorbed by the mass of graphite and steel. The central module experiences maximum heating, in the vicinity of which thermal energy is released corresponding to its heating to melt the charge. From the heated central module, heat is transferred to the peripheral parts to the loading and receiving module. In the ECU design in solid elements, heat transfer is carried out due to the thermal conductivity of the material, and is also radiated due to radiative heat transfer and is absorbed by open surfaces, taking into account their mutual visibility. The main heat removal from ECU design is made due to the fit of ECU elements to SWJ of the crystallization apparatus.

The main problem in creating a model for thermal analysis is the calculation of the velocity field of the refrigerant. Rubitek crystallization apparatus has SWJ (duct heat exchanger), the flow region of which has a complex geometric shape. The fluid flow (distilled water) was considered as viscous, incompressible, isothermal inside SWR in the area of refrigerant flow. To calculate the velocity field \( V \), the Navier-Stokes equations [26, 27] were solved, which are written in vector form as follows:

\[
\rho \frac{D \vec{w}}{D \tau} = -\grad p + (\nu \Delta \vec{v} + \vec{g})\rho
\]

(1)

The left side of the equation represents the inertia forces of the fluid volume element, written in general form. The right side represents the sum of the forces acting on the fluid volume element from the parts surrounding it: force \( \vec{P} = -\grad p \), viscous friction force \( \nu \Delta \vec{w} \) and the force of weight \( \vec{g} p \), there is \( \vec{w} \) is the velocity vector at a given point; \( \frac{D \vec{w}}{D \tau} \) is the total acceleration of the liquid element under consideration when it moves in space [26, 27]:

\[
\frac{D \vec{w}}{D \tau} = \frac{\partial \vec{w}}{\partial \tau} + (\vec{g}, \grad) \vec{w},
\]

(2)

\( \frac{D \vec{w}}{D \tau} \) – local acceleration at a fixed point associated with the dependence of speed on time; \( (\vec{w}, \grad) \vec{w} \) – convective acceleration associated with the dependence of the velocity on the position along the coordinate axis (convective derivative). For this model, equation (1) in a rectangular system finally takes the form (for example, on the \( x \) axis) [20, 26, 27]:

\[
\begin{align*}
\frac{\partial w_x}{\partial \tau} + w_x \frac{\partial w_x}{\partial x} + w_y \frac{\partial w_x}{\partial y} + w_z \frac{\partial w_x}{\partial z} = -\frac{1}{\rho} \frac{\partial P}{\partial x} + v \left( \frac{\partial^2 w_x}{\partial x^2} + \frac{\partial^2 w_x}{\partial y^2} + \frac{\partial^2 w_x}{\partial z^2} \right) + g_x.
\end{align*}
\]

(3)

The differential model was used to simulate turbulence. \( k-\omega \) Menter’s model SST

\[
\rho \frac{D \vec{w}}{D \tau} = \nabla \left( (\mu + \sigma_\omega \mu_T) \nabla \omega \right) + \frac{\omega}{K} P_h - \rho \beta \omega^2 + \sigma_d \frac{\rho}{\omega} (\nabla k)(\nabla \omega).
\]

(4)

On the outer walls of crystallization plant, the equation of convective heat transfer with the environment was solved, which uses Newton’s law, introducing the convective heat transfer coefficient \( \alpha(W/m^2 \times K) \) [20, 26, 27]:

\[
\alpha = -\frac{\lambda_f \grad T}{\Delta T} \bigg|_{n \to 0},
\]

(5)
and
\[ q = \alpha \Delta T. \]  

(6)

There is \( \lambda_f \) – thermal conductivity, \( \Delta T \) – temperature difference between the surface and the environment, \( \alpha \) heat flux density \( q(W/m^2) \), referred to a unit temperature difference \( \Delta T \) (K).

In the internal volume of the crystallization apparatus on the surface of ECU and SWJ of the crystallization apparatus surrounding them, more complex heat transfer conditions were set taking into account two mechanisms of surface heat transfer. These include: radiative heat transfer with “visible” surrounding surfaces and energy transfer (heat transfer) from the warmer parts of the assembly to less heated parts.

To calculate the rate of heat transfer due to radiation between a completely black body and the environment for real surfaces, the Stefan-Boltzmann law was subjected to some changes. For non-completely black surfaces, the spectral intensity of the radiation does not obey the Planck distribution and the emitted radiation has a preferred direction for emission.

The modified Stefan-Boltzmann law for a not absolutely black body is given by the relation \[ Q = \varepsilon \times \delta \times A \times (T_s^4 - T_a^4) \]

There is \( \varepsilon \) – the emissivity of the emitting surface, defined as the ratio of the emission power of such a surface to the emission power of a completely black body at the same temperature, \( \delta \) – Stefan-Boltzmann constant, \( T_s^4 \) – absolute temperature of a black body, \( T_a^4 \) – ambient temperature, \( A \) – radiated surface area.

In those cases when the radiating bodies partially exchanged the radiation, the concept of the coefficient of visibility of radiation \( (F) \) was introduced. The coefficient of visibility of the radiation of surface \( i \) with respect to surface \( j \) is defined as the ratio of the energy emitted from surface \( i \) and directly reaching surface \( j \) to the total amount of energy leaving surface \( i \). Bearing in mind this definition, we find that the net radiation exchange between a surface of area \( A_i \) and temperature \( T_i \) and some surface \( A_j \) with temperature \( T_j \) is given by \[ Q_{\text{radiation}} = \varepsilon_i \times \delta \times A_i \times (T_s^4 - T_a^4) \times F_{ij}. \]

There is \( F_{ij} \) – the configuration factor of surface \( i \) relative to surface \( j \), \( \varepsilon_i \) is the emissivity of surface \( i \).

In cases where the surfaces had different emission coefficients \( \varepsilon_i \) and \( \varepsilon_j \), the residual radiation heat transfer was specified by the following formula \[ Q_{\text{radiation}} = \delta \times (T_s^4 - T_a^4) \times \left( \frac{1 - \varepsilon_i}{A_i \varepsilon_i} + \frac{1}{A_i F_{ij}} \right) + \left( \frac{1 - \varepsilon_j}{A_j \varepsilon_j} \right). \]

Heat transfer by conductivity obeys the Fourier law, which establishes that the rate of heat conduction \( Q_{\text{conductivity}} \) proportional to the area of heat transfer \( (A) \) and the temperature gradient \( dT/dx \) or \[ Q_{\text{conductivity}} = -K \times A \times \frac{dT}{dx}. \]

There is \( K \) – thermal conductivity, measures the ability of a material to conduct heat.

To describe the mechanism of heat transfer and heat conduction in solids of ECU and the surrounding SWJ, boundary conditions of the third kind were set for single-layer flat and cylindrical walls. And it was solved by the equation \[ q = \frac{T_{f_2} - T_{f_1}}{R} = K_\delta \Delta T. \]

There are \( T_{f_2}, T_{f_1} \) – the values of the ambient temperature, \( R \) – entire thermal resistance, \( \lambda_f \) – heat transfer coefficient through a flat wall, under \( \lambda_w = \text{const.} \).
3.3. Results of numerical calculations

Corresponding calculations were performed according to the methodology for calculating the coupled heat transfer using the SolidWorks Premium CFD code in the Flow Simulation package.

In Fig. 4 shows the velocities and current vectors of the flow of the refrigerant when purging in SWJ with an inlet head velocity of 2 m/s. The figure gives an idea of the complexity of the flow of refrigerant inside the SWJ.

![Image of flow simulation](image)

**Figure 4.** Velocities and current vectors of the flow of the refrigerant when blowing in SWJ with an inlet velocity of 2 m/s.

Fig. 5 gives an idea of the temperature distribution of a solid on the outer walls of SWJ crystallization apparatus.

According to the calculated data presented in Fig 6, one can trace the influence of the velocity and current vectors of the flow of the refrigerant on the temperature distribution of the fluid.

Fig. 7 (a-d) shows the distribution of the temperature of a solid over sections on the walls of SWJ of the crystallization apparatus and ECU placed in it. From Fig. 7 (d) follows that the site design and the high thermal conductivity of graphite make it possible to create the high gradient region in the crystal growth zone and the low gradient temperature region in the annealing zone. The step of changing the temperature in the crystal growth zone is 80 – 70 °C/cm and 20 – 40 °C/cm in the crystal annealing zone, respectively.

From Fig. 8 also follows that ECU allows you to create the symmetric temperature field necessary to create the morphologically stable crystallization front. Also, as shown in Fig. 8...
4. Conclusion

In this paper, we consider the main method of construction of ECU placed in SWJ crystallization unit. For the construction of ECU the mathematical model of heat transfer processes is proposed: convective, radiative in the gray-body approximation, and conductive mechanisms. This model describes the processes of mass and heat transfer in geometrically diverse, heterogeneous, and heterogeneous regions in terms of their thermophysical properties. The finite element method is used for the numerical solution of such a problem.

As the result of the calculations, the conjugate problem of temperature fields in the thermal unit and in SWJ was solved. The features of the flow of refrigerant inside SWJ and its influence
Figure 7. The temperature distribution of the solid state of ECU and SWJ crystallization apparatus; a) – in longitudinal section (a side view); b) – in longitudinal section (a top view); c) – in cross section; d) is the isotherm of temperature distribution along from the beginning of the loading zone to the end of the annealing zone;

on the degree of cooling of SWJ are established. The complete picture of the temperature distribution in solids of ECU and SWJ is established, as well as a picture of the temperature distribution in a fluid.

The picture of the temperature distribution of the solid on a surface along the working space
Figure 8. The temperature distribution of the solid body of a CaF\(_2\) crystal, which clearly demonstrates the symmetric temperature field and the slightly convex isotherm of the crystal growth front of ECU (Fig. 7, d) is represented by a graphical dependence and is shown in Fig. 9. Also, Fig. 9 shows the graphical dependence of the temperature distribution pattern in a CaF\(_2\) crystal along the horizontal mine (from the beginning of the loading zone to the end of the crystal annealing zone).

Figure 9. The picture of the graphic dependence of the distribution of solid temperature (the figure below shows a general view of the shaft where the temperature was taken, from the crucible loading module to the crucible receiving module); a) – on the surface along the working space of ECU from the beginning of the loading zone to the end the crystal annealing zone; b) – on the surface along the CaF\(_2\) crystal
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