

Modeling of non-stationary processes heat and mass transfer according to the cellular model of sucrose mass crystallization

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Abstract

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Introduction. The purpose of this work is to determine the influence of variable thermophysical characteristics on non-stationary diffusion mass flows of sucrose during sugar mass crystallization.

Materials and methods. In order to obtain quantitative values of nonstationary diffusion mass flows for intercrystalline sucrose solutions surrounding sugar crystals was calculated by numerical methods simultaneously for two systems of heat and mass transfer: 1) the system of 7 nonstationary thermal conductivity problems; 2) the system consisting of three nonstationary diffusion mass transfer problems.

Results and discussion. For a number of the relative sugar massecuite boiling time values 0.15-1.0 based on two systems (10 subsystems) simultaneous numerical solution of nonstationary parabolic type differential equations in partial derivatives (the first system consisted of 7 subsystems – for non-stationary thermal conductivity problems in each of the 7 areas; the second system includes three subsystems – for non-stationary diffusion mass transfer problems for 4 areas) found respectively: non-stationary temperature distributions in each of the 7 regions (4 regions with intercrystalline solutions, 2 sugar crystals and massecuite); nonstationary sucrose diffusion mass flows in each of the 4 intercrystalline sucrose solutions regions of the whole considered two cells and massecuite system. When the value of the relative sugar massecuite boiling time 0.15 when the whole cells system moving along the heating tube, the substance is first transferred from the intercrystalline sucrose solution of the smaller crystal to the intercrystalline sucrose solution of the larger crystal during 2.41–2.7 s (depending on constant or variable thermophysical characteristics). Starting from this moment of time $\tau_{k,2}$ to 3.95 s (at sugar massecuite boiling time values 0.15) when the system of cells leaves the heating tube, the situation changes to the opposite. So, in this case, only one extremum of the diffusive mass flow was clearly defined (the minimum on the graph is reached at the moment of time 1.29–1.45 s, which determines the maximum sucrose mass transfer from a smaller crystal to a larger crystal). No clear maximum has been established in this case. At the relative time 1.0 of sugar massecuite boiling two clearly defined extremes (minimum and maximum) were obtained for both constant and variable thermophysical characteristics.

Conclusions. Features of variable thermophysical characteristics influence on the non-stationary sucrose diffusion mass flows in the cellular model are shown.

Introduction

The complexity of the sucrose mass crystallization during the sugar syrup production is due to the simultaneous and mutually influencing non-stationary heat and mass exchange processes.

In this paper, by means of a numerical experiment based on the developed mathematical model [Pogoriliy, 2015 b; Pogoriliy, 2015 c; Pogoriliy, 2016 a], the peculiarities and influence of non-stationary diffusive sucrose mass flows with variable thermophysical characteristics under the cellular model of mass crystallization.

Crystallisation of sucrose is the most energy-intensive step in the industrial production of sugar (Alewijn et al., 2013; Hartel et al., 1991; Shamim et al., 2016).

The process of sugar crystallization is difficult enough to describe. This is due to the need to simultaneously take into account all non-stationary technological and all non-stationary thermophysical parameters that affect the sugar crystallization process (Duroudier, 2016; Rózsa et al., 2016; Kulinchenko et al., 2012).

The problem of crystallization of a single sugar crystal, sucrose mass crystallization, as well as related processes that directly affect these complex processes at the modern level were dealt with by a number of authors: (Bruno et al., 2019; Georgieva et al., 2003; Nagy et al., 2013; Osmanbegovic et al., 2022; Romero-Bustamante et al., 2022; Rózsa et al., 2016; Sánchez-Sánchez et al., 2017; Suarez et al., 2011; Tkachenko et al., 2020; Vetter et al., 2022).

Note that considering the proposed solutions to the sucrose mass crystallization problem in industrial conditions (Bruno et al., 2019; Georgieva et al., 2003; Rózsa et al., 2016; Sánchez-Sánchez et al., 2017; Tkachenko et al., 2020), it can conclude that there is no single generally accepted approach to this issue today. Therefore, the problem arises in creating the most universal mathematical model of the sucrose mass crystallization process in industrial conditions.

One of the main tasks that the author set before himself is that the mathematical model would describe as fully as possible the heat and mass exchange process, which occurs between the constituent parts of the multiphase system, which is a sugar massecuite.

Based on the literature review, it can also be concluded that taking into account all factors affecting the sucrose crystallization process is a rather difficult task when creating a mathematical model (Rózsa et al., 2016). Note that taking into account simultaneously all technological, thermophysical and hydrodynamic factors that affect the sucrose mass crystallization process is practically an extremely difficult or even impossible task. In view of this, when developing a mathematical model they were forced to accept a number of simplifications. Thus, the mathematical model of the mass crystallization process being developed should be classified as an idealized model.

This paper proposes one of the next steps in the mathematical model development of the sucrose mass crystallization process.

In the continuation of the works (Pogoriliy, 2015 a, b, c; 2016 a) sugar massecuite is considered from the point of view of a cellular model.

Assume that in each cell a «sugar crystal» is surrounded by a certain corresponding «intercrystalline sucrose solution amount» (Pogoriliy, 2016 a) during the entire time of boiling the sugar massecuite.

The purpose of this work is to determine the influence of variable thermophysical characteristics on non-stationary diffusion mass flows of sucrose during sugar mass crystallization.

Materials and methods

In this work, as well as in works (Pogoriliy, 2015 b, c; Pogoriliy, 2016 a), it consider a system consisting of two cells that are in contact with each other, as well as the massecuite region. Each of the considered cells includes a «sugar crystal» and an «intercrystalline sucrose solution surrounding the corresponding crystal». The considered cells are of different sizes. Therefore, one crystal and the intercrystalline solution surrounding it will have a larger size, respectively, the other will have a smaller size.

In general, we believe that hydrodynamic interactions between cells can occur only between the outer boundaries of the intercrystalline sucrose solutions regions of such cells. That is, in other words, in each cell, the region of the intercrystalline sucrose solution is as if «glued» only to its sugar crystal, which is inside the region of the intercrystalline sucrose solution during the entire time of boiling the sugar massecuite.

Areas of intercrystalline sucrose solutions of different cells can generally move relative to each other. However, in this case, we assume that in the considered system «two cells—the massecuite region» from the moment of entering the heating tube until the moment of its exit, the system components do not change the order of their location relative to each other and do not change their size. With this assumption, we fix the volume both for the crystal and for the intercrystalline sucrose solution, as well as for the area of the massecuite during the entire time τ of their stay in the heating tube.

Such an assumption was forced to be accepted, since it is not possible to take into account the dynamic changes of both crystal sizes and sizes of intercrystalline sucrose solutions surrounding the corresponding crystals, at this stage of creating a mathematical model.

We consider the non-stationary heat exchange process, which occurs both internally between the components in each cell of the entire system (that is, between the crystals and the corresponding intercrystalline sucrose solutions in each cell), and between them. At the same time, we also consider the non-stationary heat exchange between all the components of the two cells of this system and the massecuite region.

At the same time, simultaneously with the non-stationary heat exchange processes mentioned above, we consider non-stationary mass transfer processes that occur both internally between the components of each of the entire system considered cells (that is, between the crystals and the corresponding intercrystalline sucrose solutions in each cell), and between the cells themselves (we consider by determination of diffusion mass transfer processes between intercrystalline sucrose solutions of the first and second cells).

Since the simulation of non-stationary processes as heat transfer and mass transfer simultaneously for the entire system «two cells—the massecuite region» («larger sugar crystal–intercrystalline sucrose solution of a larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of a smaller crystal»–massecuite”) is quite a difficult task, so it should be carried out in several stages.

At the first stage, for the simplest geometric model case of the massecuite representation, the mathematical model described only the non-stationary temperature distribution for the system, which included only one cell («sugar crystal–intercrystalline sucrose solution») and the massecuite region (Pogoriliy, 2015 a).

Further, the non-stationary temperature distribution in the system «sugar crystal–intercrystalline sucrose solution– massecuite» was considered, taking into account the different location of the cell «sugar crystal–intercrystalline sucrose solution» in relation to the heating tube surface. Next, the case was considered for a more complex geometric model of the massecuite representation, when the mathematical model described a non-stationary

temperature distribution for a more complex system, which already included two cells of different sizes («larger crystal–intercrystalline solution of sucrose of a larger crystal»–«smaller crystal–intercrystalline sucrose solution of a smaller crystal») and the massecuite area (Pogoriliy, 2015 b).

At the second stage, the case was considered when the mathematical model simultaneously described the non-stationary temperature distribution for each component of the entire two cells system and the massecuite region (“«larger sugar crystal–intercrystalline sucrose solution of a larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of a smaller crystal»–massecuite”), and non-stationary sucrose concentrations distribution in all intercrystalline solutions regions of this system (Pogorilyy, 2015 c).

Next, we considered the case when the mathematical model simultaneously described the non-stationary temperature distribution for the each entire system component of two cells and the massecuite area (“«larger sugar crystal–intercrystalline sucrose solution of a larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of a smaller crystal»–massecuite”), and non-stationary sucrose diffusive mass fluxes in all intercrystalline solutions regions of this system (Pogorilyy, 2016 a).

We note that in the last work, several mathematical model variants were considered, when either all steels or simultaneously all variable thermophysical characteristics that took part in the calculations were taken into account.

However, for a more complete understanding of the sucrose mass crystallization process, it is worth establishing how certain factors individually affect this process.

For this purpose, it is necessary to conduct more thorough studies on solving the question of whether taking into account dynamically changing thermophysical characteristics separately for the sugar crystal, separately for the intercrystalline sucrose solution, and separately for the massecuite region affects the sucrose mass crystallization process. Provided that, if such impacts occur, establish the nature and determine the weight of such impacts.

So, in this work, we consider the system of two cells and the massecuite region (“«larger sugar crystal–intercrystalline sucrose solution of the larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of the smaller crystal»– massecuite”) inside the heating tube of the vacuum apparatus heating chamber (Figure 1).

From the 3D model of the system “«larger sugar crystal–intercrystalline sucrose solution of a larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of a smaller crystal»–massecuite” in the inside of the heating tube of the vacuum apparatus heating chamber, by projection, we will move to the 2D model, where all components of the system under consideration (Figure 2).

Finally, in the 2D model (Figure 2), by selecting the regions according to the minimum size of the overall size $b_{cr,2}$ (Figure 1) of the smaller crystal #2, which will participate in the heat and mass transfer processes, we moved to a one-dimensional formulation of such problems.

Therefore, the main task: to find the values of diffusion mass flows at the corresponding boundaries «sugar crystal–intercrystalline sucrose solution» in each cell of the considered system and between intercrystalline sucrose solutions of neighboring cells (Figure 2).

Also, at the same time, we will consider the diffusive mass transfer between intercrystalline sucrose solutions of different cells of the entire system, which consists of two cells of different sizes and massecuite “«larger sugar crystal–intercrystalline sucrose solution of a larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of a smaller crystal»–massecuite”).

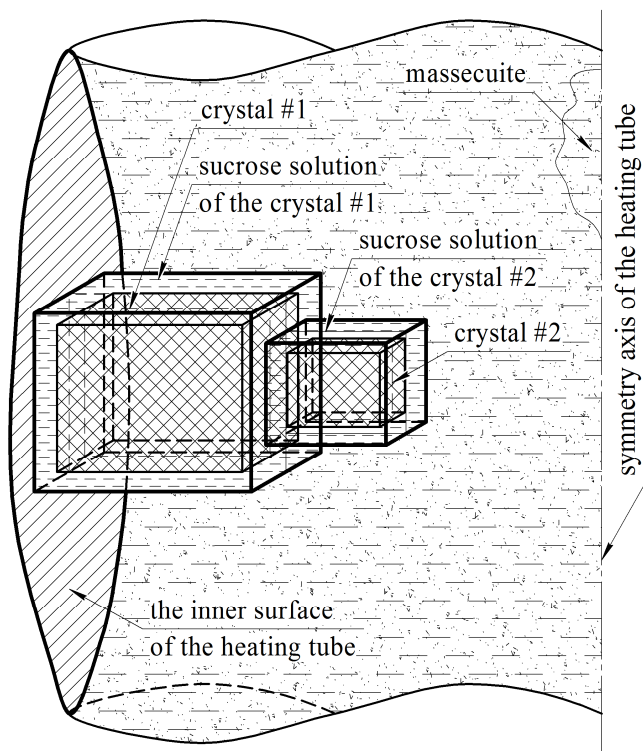


Figure 1. Three-dimensional geometric model of two cells and the massecuite area (“«larger sugar crystal #1–intercrystalline sucrose solution of larger crystal #1»–«smaller sugar crystal #2–intercrystalline sucrose solution of smaller crystal #2»–massecuite”) inside the heating tube of the vacuum apparatus heating chamber

The solution of such a complex problem is based on the simultaneous solution of a system of the non-stationary parabolic type differential equations in partial derivatives, which will describe non-stationary heat transfer problems (for seven regions: (#1), (#2), (#3), (#4), (#5), (#6) and (#7)) for the entire system «larger cell–smaller cell–massecuite» and the system of three unsteady diffusion mass transfer problems (for four regions: separately for region (#1)), separately for area (#6), and simultaneously for two areas (#3) and (#4)) for all components of the larger and smaller cells in the form of intercrystalline sucrose solutions.

Non-stationary problems of heat transfer and mass transfer for the system “«larger sugar crystal–intercrystalline sucrose solution of a larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of a smaller crystal»–massecuite” were considered for the case of this entire system contact with the inner heating (boiling) tubes surface of the vacuum apparatus heating chamber (Figure 1).

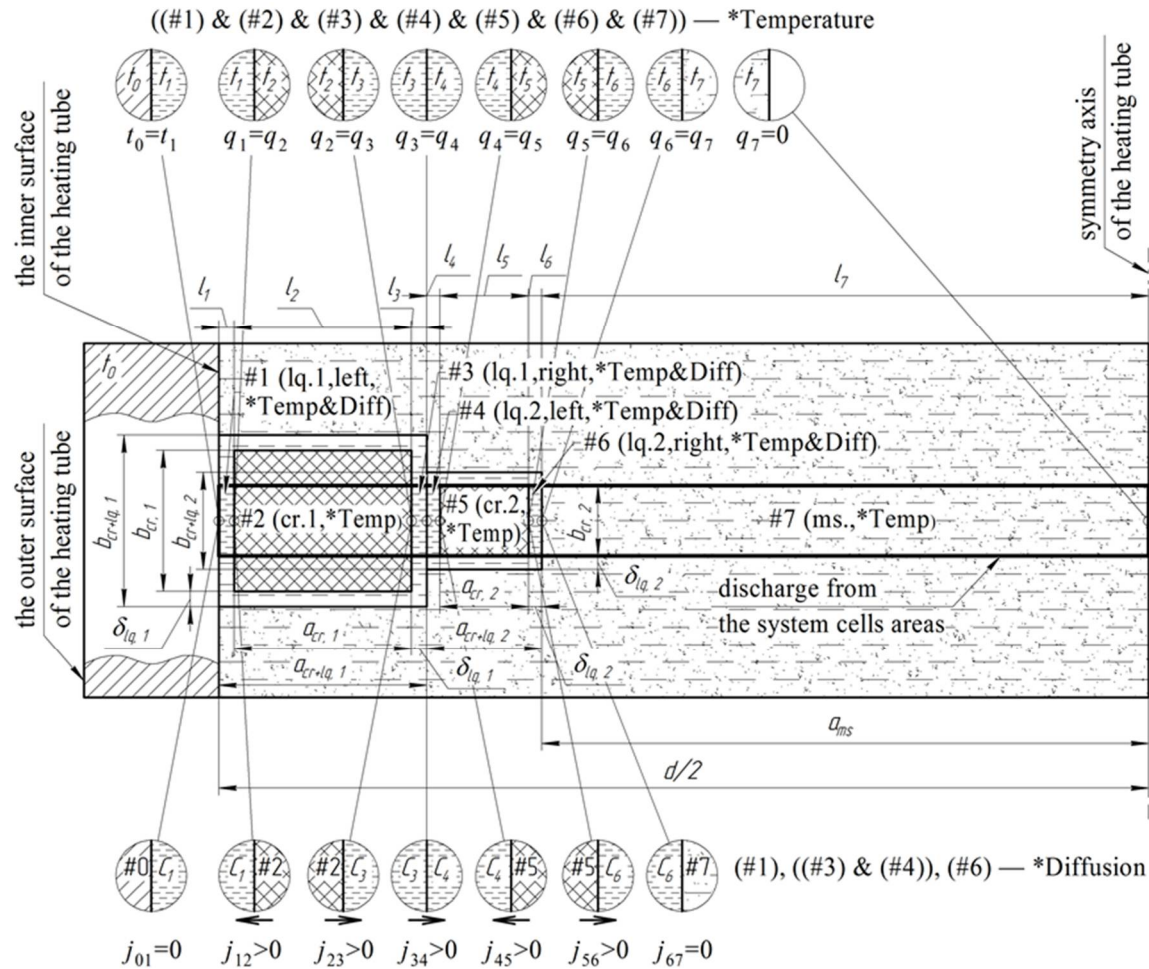


Figure 2. Selection from the 2D projection of the system “larger sugar crystal–intercrystalline sucrose solution of a larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of a smaller crystal»–massecuite” regions that simultaneously receive training in non-stationary heat exchange processes (simultaneously between regions (#1), (#2), (#3), (#4), (#5), (#6) and (#7)) and diffusive mass exchanges (separately for region (#1), separately for region (#6), and simultaneously for two regions (#3) and (#4)) for the calculation of non-stationary diffusive mass fluxes of sucrose.

Designation on Figure 2:

*) For areas:

*Temp—only the non-stationary problem of thermal conductivity was considered;

*Temp&Diff— the non-stationary thermal conductivity problem and the non-stationary diffusive mass transfer problem were considered at the same time

*) in the footnotes above for heat flows q_i , (where $i = 1..7$) boundary conditions are indicated on each separate internal boundary;

*) in the footnotes below for diffusion sucrose mass flows j_{mn} , (where the coefficients $(mn) = \{21; 23; 34; 54; 56\}$) on each separate boundary, an arrow indicates the positive direction of the sucrose mass flow accepted in the calculations (the reverse direction is negative)

It is worth noting that in this case two different concepts regarding the term "time" are considered:

a) τ/τ_c – the relative time of boiling sugar massecuite; as a rule, the complete cooking time of the sugar massecuite reaches the order of several hours; the relative time τ/τ_c varies within 0–1; where 0 corresponds to the beginning of boiling sugar massecuite; and 1 corresponds to the completion of boiling sugar massecuite; almost all technological, thermophysical indicators depend on this parameter;

b) τ_c is the residence time of the system «larger sugar crystal–intercrystalline sucrose solution of a larger crystal»–«smaller sugar crystal–intercrystalline sucrose solution of a smaller crystal»–massecuite” in the middle of the heating tube of the heating chamber; in this case, the order of magnitude is from a few seconds to a dozen seconds, and in turn, also depends on the relative time of boiling the sugar massecuite τ/τ_c .

The initial moment of time $\tau_{k,0} = 0$ of the contact of the «larger cell–smaller cell–massecuite» system with the heat carrier was taken to be the time when the entire «two cells–massecuite » system enters the lower part of the vertically located heating tube.

The final contact moment of time $\tau_{c,end}$ of the «larger cell–smaller cell–massecuite» system with the heat carrier was taken to be the moment when the entire «two cells–massecuite» system simultaneously leaves the upper part of the vertically located heating tube of the vacuum apparatus.

The issue of determining the residence time value $\tau_{c,end}$ of the «two cells– massecuite» system inside the heating tube of the vacuum apparatus heating chamber depending on the relative boiling time τ/τ_c of the sugar massecuite in industrial conditions was considered in (Pohorilyi, 2016 a).

Let us immediately note that the technological and thermophysical characteristics for each of the «two cells–massecuite» system components: for crystals, for intercrystalline solutions of sucrose and for massecuite, as well as the diffusion mass transfer coefficient only for the intercrystalline sucrose solutions regions of two cells, will depend on the relative time of boiling sugar massecuite τ/τ_c .

However, we note that due to the fact that the boiling sugar massecuite time can reach several hours, and the residence time of the «two cells–massecuite» system inside the heating tube of the heating chamber is on the order of several seconds, we consider the problem of heat and mass transfer to be quasi-stationary with respect to the variable relative boiling time τ/τ_c of sugar massecuite; and, at the same time, we consider the contact of the «two cells–massecuite» system with the inner wall of the heating tube of the vacuum apparatus heating chamber to be non-stationary with respect to the time variable. It is precisely in this sense that in this paper a study of non-stationarity in relation to heat and mass transfer processes is carried out.

As for the relative boiling time τ/τ_c , ten different values were recorded in this study from the moment of sugar crystal formation, which corresponds to $\tau/\tau_c = 0.15$, and until the end of the sugar massecuite boiling, which corresponds to $\tau/\tau_c = 1.0$, namely: $\tau/\tau_c = 0.15; 0.2; 0.3; 0.4; 0.5; 0.6; 0.7; 0.8; 0.9; 1.0$. For each of the above fixed relative time values of sugar massecuite boiling τ/τ_c , all technological parameters were found and recorded:

- a) dry substances content in the intercrystalline sucrose solution;
- b) purity of intercrystalline sucrose solution;
- c) mass content of sucrose in intercrystalline sucrose solution;
- d) mass content of crystals in massecuite.

On the basis of these fixed values, as well as depending on the temperature t , the necessary thermophysical parameters were found for intercrystalline solutions of sucrose in both cells and for massecuite.

Note that in this work, the impact of taking into account variable thermophysical characteristics coefficients on the sucrose mass crystallization process is investigated through the determination of diffusion mass flows in each of the considered cells, as well as diffusion mass exchange between the cells themselves. This is done in order to determine the influence weight of these considerations on each individual component of the «two cells–massecuite» system, and in the future to be able to apply the most optimal approach when creating further mathematical models of the sucrose mass crystallization process.

Let's explain what exactly the «fixed» coefficients and «variable» coefficients contribute to the research understanding.

«constant» coefficients means that at the beginning of the calculation of non-stationary heat and mass transfer processes, all necessary coefficients are calculated when:

- a) to the corresponding value of the relative boiling time $\tau/\tau_c = 0.15; 0.2; 0.3; 0.4; 0.5; 0.6; 0.7; 0.8; 0.9; 1.0$ (that is, all coefficients depend on the relative boiling time τ/τ_c);
- b) but each time all the necessary coefficients are calculated precisely at the initial temperature t_0 of the entire system «two cells–massecuite», which enters the lower part of the entry into the heating tube of the heating chamber.

We denote the calculation option with «constant» thermophysical coefficients by (I).

«variable» coefficients means that, just as for «constant» at the beginning of the non-stationary processes calculation of heat and mass transfer, all the necessary coefficients are calculated at the corresponding value of the relative boiling time $\tau/\tau_c = 0.15; 0.2; 0.3; 0.4; 0.5; 0.6; 0.7; 0.8; 0.9; 1.0$ (that is, all coefficients depend on the relative boiling time τ/τ_c).

At the first time step, as well as for the «constant» coefficients, those coefficients that depend on temperature are calculated at the initial temperature t_0 of the entire «two cells–massecuite» system, which enters the lower part of the entry into the heating tube of the heating chamber.

But at each subsequent step (starting from the second step) in time τ_c for the calculation of non-stationary heat and mass transfer problems, all the necessary coefficients are calculated already at the current temperature t of each component of the «two cells–massecuite» system, where the current value of the temperature t is taken from the previous step made calculations.

Note that in this work, research was conducted when «variable» thermophysical coefficients were taken into account:

- separately only for crystals (we denote this calculation version of variable thermophysical coefficients through (II a));
- separately only for intercrystalline solutions of sucrose (we denote this variant of calculating variable thermophysical coefficients through (II b));

- separately only for massecuite (we denote this version of the calculation of variable thermophysical coefficients through (II c)).

All «variable» thermophysical coefficients for the entire «two cells–massecuite» system were also taken into account at the same time (we denote this variant of the calculation of all variable thermophysical coefficients by (III)).

As mentioned earlier, the non-stationary distribution of temperatures in all seven regions (Figure 2) was determined based on the solution of one non-stationary thermal conductivity problem simultaneously for:

- #1 – the left area of the intercrystalline sucrose solution of the larger crystal 1 of the first cell;
- #2 – regions of the larger crystal 1 of the first cell;
- #3 – the right area of the intercrystalline sucrose solution of the larger crystal 1 of the first cell;
- #4 – the left area of the intercrystalline sucrose solution of smaller crystal 2 of the second cell;
- #5 – areas of the smaller crystal 2 of the second cell;
- #6 – the right area of the intercrystalline sucrose solution of smaller crystal 2 of the second cell;
- #7 – massecuite areas.

Together with the non-stationary temperature distribution, the sought non-stationary concentrations distribution in all four regions (Figure 2) of intercrystalline solutions was determined based on the simultaneous solution of three different non-stationary diffusion mass transfer problems:

- separately for area #1 (the left intercrystalline sucrose solution area of larger crystal 1 of the first cell);
- at the same time for regions #3 and #4 (intercrystalline solutions belonging to crystals of different sizes), which are in contact with each other according to the ideal law of diffusion mass transfer;
- separately for area #6 (the right area of the intercrystalline sucrose solution of smaller crystal 2 of the second cell).

Next, on the basis of the obtained solutions of three non-stationary diffusion mass transfer problems the values of $j(\tau)$, ($\text{kg}/(\text{m}^2 \cdot \text{s})$), non-stationary diffusive mass flows for intercrystalline sucrose solution regions as a function of time τ , (s). Since we have four different intercrystalline sucrose solution regions (#1, #3, #4, #6, Figure 2), in order to distinguish the diffusion mass flows for each boundary of each separate region, we denote them by $j_{mn}(\tau)$, ($(mn) = \{01; 12; 23; 34; 45; 56; 67\}$), (Figure 2), where:

- $j_{01}(\tau)$, – diffusive mass exchange between the left border of region #1 (that is, the left intercrystalline sucrose solution region of the first cell, Figure 2) and the heating wall of the vacuum apparatus heating chamber; since such a mass exchange for the system «two cells–massecuite» and an impermeable heating wall in this direction is absent due to physical considerations, the mass exchange directly of area #1 with the heating wall is also absent, therefore $j_{01}(\tau) = 0$, ($\text{kg}/(\text{m}^2 \cdot \text{s})$);
- $j_{12}(\tau)$, – diffusive mass exchange between the right border of region #1 (ie, the left intercrystalline sucrose solution region of the first larger cell, Figure 2) and the larger sugar crystal (region #2, Figure 2) of the first cell of the «two cells–massecuite»; if diffusive mass transfer occurs from the side of region #2 of the larger crystal to the side of region #1 of the intercrystalline sucrose solution, then we accept this direction

as positive, $j_{12}(\tau) > 0$, (Figure 2); if the diffusive mass transfer occurs from the side of region #1 of the intercrystalline sucrose solution in the direction of region #2 of the larger crystal of the first cell, then we take this direction of movement of sucrose as negative, $j_{12}(\tau) < 0$;

- $j_{23}(\tau)$, – diffusive mass exchange between the left border of region #3 (ie, the right intercrystalline sucrose solution region of the first larger cell, Figure 2) and the larger sugar crystal (region #2, Figure 2) of the first cell of the «two cells–massecuite» system; if diffusive mass transfer occurs from the side of region #2 of the larger crystal to the side of region #3 of the intercrystalline sucrose solution, then we accept this direction as positive, $j_{23}(\tau) > 0$, (Figure 2); if the diffusive mass transfer occurs from the side of region #3 of the intercrystalline sucrose solution in the direction of region #2 of the larger crystal of the first cell, then this direction of sucrose movement is taken as negative, $j_{23}(\tau) < 0$;
- $j_{34}(\tau)$, – diffusive mass exchange between the right border of region #3 (ie, the right region of the intercrystalline sucrose solution of the first larger cell, Figure 2), and the left border of region #4 (ie, the left region of the intercrystalline sucrose solution of the second smaller cell, Figure 2); in fact, this is the sought-after mass exchange between neighboring cells of the «two cells–massecuite» system in the recrystallization theory; if diffusive mass transfer occurs from the side of region #3 of the intercrystalline sucrose solution of the larger first cell to the side of region #4 of the intercrystalline sucrose solution of the smaller second cell, then we accept this direction as positive, $j_{34}(\tau) > 0$, (Figure 2); if the diffusion mass transfer occurs from the side of region #4 of the intercrystalline sucrose solution of the smaller crystal of the second cell in the direction of region #3 of the intercrystalline sucrose solution of the larger crystal of the first cell, then this direction of sucrose movement is taken as negative, $j_{34}(\tau) < 0$;
- $j_{45}(\tau)$, – diffusive mass exchange between the right border of region #4 (ie, the left region of the intercrystalline sucrose solution of the second smaller cell, Figure 2) and the smaller sugar crystal (region #5, Figure 2) of the second smaller cell of the «two cells–massecuite»; if diffusive mass transfer occurs from the side of the smaller crystal #5 to the side of region #4 of the intercrystalline sucrose solution, then we accept this direction as positive, $j_{45}(\tau) > 0$, (Figure 2); if the diffusive mass transfer occurs from the side of region #4 of the intercrystalline sucrose solution in the direction of region #5 of the smaller crystal of the second cell, then this direction of sucrose movement is taken as negative, $j_{45}(\tau) < 0$;
- $j_{56}(\tau)$, – diffusive mass exchange between the left border of region #6 (ie, the right region of the intercrystalline sucrose solution of the second smaller cell, Figure 2) and the smaller sugar crystal (region #5, Figure 2) of the second cell of the «two cells–massecuite»; if diffusive mass transfer occurs from the side of region #5 of the smaller sugar crystal to the side of region #6 of the intercrystalline sucrose solution, then we accept this direction as positive, $j_{56}(\tau) > 0$, (Figure 2); if the diffusive mass transfer occurs from the side of region #6 of the intercrystalline sucrose solution in the direction of region #5 of the smaller crystal of the second cell, then this direction of sucrose movement is taken as negative, $j_{56}(\tau) < 0$;

- $j_{67}(\tau)$, – diffusive mass exchange between the right border of region #6 (ie, the right region of the intercrystalline sucrose solution of the second smaller cell, Figure 2), and region #7 of the massecuite of the «two cells–massecuite» system; note that although for physical reasons such a diffusion mass exchange generally occurs, it is not possible to investigate such a diffusion mass exchange directly at this stage. Therefore, we accept the value of the diffusion mass flow between the region #6 of the intercrystalline sucrose solution and the region #7 of the massecuite $j_{67}(\tau)=0$.

Thus, on the basis of one system of equations of the non-stationary heat transfer problem and three systems of equations of the non-stationary diffusion mass transfer problems we determine the desired non-stationary diffusion mass flows $j_{mn}(\tau)$, $((mn)=\{01; 12; 23; 34; 45; 56; 67\})$.

Solve a complex system of non-stationary differential equations for the non-stationary heat transfer problem and three systems of equations of non-stationary diffusion mass transfer problems as well as finding non-stationary diffusive mass flows is a rather difficult task even under the condition of using constant thermophysical characteristics. Using analytical methods, it is difficult to solve such a system of equations even for a system that consists of three regions (in our case, there are seven of them). Therefore, the non-stationary system of problems was solved using numerical methods: the control volume method (Eymard et al., 2000; LeVeque, 2002). Diffusion mass flows were searched: for constant thermophysical characteristics (case I, accepted earlier), for alternately variable thermophysical characteristics (cases II a, II b, and II c), and for the case when the thermophysical characteristics were all variable at the same time (case III).

Note that when calculating the sucrose diffusion mass flows $j_{mn}(\tau)$, $(m=0-6, n=m+1)$, used the first and second order of their approximation. This paper presents the results of only the second order of approximation.

When calculations were carried out, the initial temperature of the components of the «two cells–massecuite» system was assumed to be the same for all areas (#1)–(#7), (Figure 2) at the same time and equal to $t_{i,0} = 75\text{ }^{\circ}\text{C}$, $(i = \overline{1,7})$.

The temperature of the inner wall t_0 of the heating tube was assumed to be constant over the entire height of this tube, i.e., during the entire time of contact with the «two cells–massecuite» system and equal to $t_0 = 100\text{ }^{\circ}\text{C}$.

When performing calculations the initial concentration for all intercrystalline sucrose solutions components of the «two cells–massecuite» system, that is, for all areas (#1), (#3)–(#4), (#6), (Figure 2), was simultaneously assumed to be the same and equal to $C_{i,0} = C_{i,0}(t_{i,0}) = 77,594\%$ (Pohorilyi, 2016 a). In this work, the initial concentration was calculated at the supersaturation factor $SS = 1$.

Based on the research conducted (Pohorilyi, 2016 a), the contact time τ_c of the «two cells–massecuite» system with the wall of the heating tube of the vacuum apparatus heating chamber depended non-linearly on the relative time of boiling sugar massecuite τ/τ_c . The following values were found:

- for the relative boiling time $\tau/\tau_c = 0,15$, the contact time is equal to $\tau_{k,\text{end}} = 3,95\text{ s}$;
- at $\tau/\tau_c = 1,0$, the contact time is equal to $\tau_{k,\text{end}} = 67,93\text{ s}$.

When using numerical methods, time discretization was $\Delta\tau_k = 0.01\text{ s}$.

In this work, the following crystal size values were adopted: $a_{cr,1} = 5,0 \cdot 10^{-4}\text{ m}$, $a_{cr,2} = 2,5 \cdot 10^{-4}\text{ m}$, (Figure 2), (Kulinchenko et al., 2012; Crestani et al., 2021; Zhong et al., 2022). On the basis of the conducted research (Pohorilyi, 2016 a), all other required dimensions of the thickness of the intercrystalline sucrose solution layer were obtained:

$\delta_{lq,1} = 4,29 \cdot 10^{-5}$ m, $\delta_{lq,2} = 3,73 \cdot 10^{-5}$ m, and the size of the massecuite region: $a_{ms} = 4,83896 \cdot 10^{-2}$ m, (Figure 2). The size of the layer thickness was calculated based on the massecuite volume in the vacuum apparatus at the moment of introducing the seed into it according to the data given in (Kulinchenko et al., 2012).

The discretization by coordinate, i.e., the partition grid, for each of the regions (#1)–(#7), (Figure 2) was adopted as follows:

- regions of intercrystalline sucrose solutions (#1), (#3), (#4), (#6), (Figure 2), the grid is uniform, the number of calculated points of division is $n_1 = n_3 = n_4 = n_6 = 10$;
- regions of sugar crystals (#2), (#5), (Figure 2), the grid is uniform, the number of calculation points of division is $n_2 = n_5 = 20$;
- the massecuite area (#7), (Figure 2), the grid is uneven, the number of calculation points of division $n_7 = 100$.

Results and discussion

On the basis of calculations carried out at the same time for:

- systems of equations of non-stationary thermal conductivity and
- three systems of non-stationary diffusive mass transfer equations for which during the entire contact time τ_c of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber, the calculations were carried out according to five different variants of the calculation (in all five cases, the technological indicators were fixed at a certain relative time value of the boiling sugar massecuite τ/τ_c , ($\tau/\tau_c = 0.15–1.0$) according to which the calculation was made):
- I) all thermophysical characteristics (coefficients) are fixed at the initial temperature $t_{i,0} = 75^\circ\text{C}$, ($i = \overline{1,7}$), and at a certain value of the relative time of boiling sugar massecuite τ/τ_c , ($\tau/\tau_c = 0.15–1.0$) according to which the calculation was carried out;
- II a) at the initial temperature $t_{i,0} = 75^\circ\text{C}$, ($i = \overline{1,7}$), and at a certain value of the relative time of boiling sugar massecuite τ/τ_c , ($\tau/\tau_c = 0.15–1.0$) only thermophysical coefficients were recorded: all for the crystal, diffusion coefficient for the intercrystalline solution of sucrose and all for the massecuite; thermophysical coefficients (thermal conductivity coefficient, heat capacity coefficient and density) for the intercrystalline sucrose solution during the entire calculation time τ_c were dependent on the current temperature $t_i(\tau)$, ($i = \overline{1,7}$), at a certain value of the relative time of boiling sugar massecuite τ/τ_c , ($\tau/\tau_c = 0.15–1.0$) according to which the calculation was carried out;
- II b) at the initial temperature $t_{i,0} = 75^\circ\text{C}$, ($i = \overline{1,7}$), and at a certain value of the relative time of boiling sugar massecuite τ/τ_c , ($\tau/\tau_c = 0.15–1.0$), only thermophysical coefficients were recorded: all for intercrystalline sucrose solution and all for massecuite; thermophysical coefficients (thermal conductivity coefficient, heat capacity coefficient and density) for the crystal during the entire calculation time τ_c were assumed to be dependent on the current temperature $t_i(\tau_c)$, ($i = \overline{1,7}$);

- II c) at the initial temperature $t_{i,0} = 75\text{ }^{\circ}\text{C}$, ($i = \overline{1,7}$), and at a certain value of the relative time of boiling sugar massecuite τ/τ_c , ($\tau/\tau_c = 0.15\text{--}1.0$), only thermophysical coefficients are fixed: all for the crystal and all for the intercrystalline solution; thermophysical coefficients (thermal conductivity coefficient, heat capacity coefficient and density) for massecuite depend on the current temperature $t_i(\tau_c)$, ($i = \overline{1,7}$), but are fixed at a certain value of the relative time of boiling sugar massecuite τ/τ_c , ($\tau/\tau_c = 0.15\text{--}1.0$);
- III) all thermophysical coefficients: for the crystal, for the intercrystalline solution, and for the massecuite depend on the current temperature $t_i(\tau_c)$, ($i = \overline{1,7}$), but are fixed at a certain value of the relative time of boiling sugar massecuite τ/τ_c , ($\tau/\tau_c = 0.15\text{--}1.0$).

Due to the limited scope of the article, this paper shows the dependences of the diffusion mass flows $j_{mn}(\tau)$, ($m=1\text{--}5$, $n=m+1$), at the boundaries of intercrystalline sucrose solutions of the «two cells–massecuite» system (Figure 2) depending on the contact time τ for only two cases of the relative time of boiling sugar massecuite τ/τ_c : at the moment of formation of the structured crystals mass ($\tau/\tau_c = 0.15$) and at the final moment of the sugar massecuite boiling time ($\tau/\tau_c = 1.0$).

Distribution of unsteady diffusion mass flows of sucrose $j_{mn}(\tau)$, ($m=1\text{--}5$, $n=m+1$), at the boundaries of intercrystalline sucrose solutions regions (#1), (#3), (#4) and (#6), (Figure 2) depending on the contact time τ of the «two cells–massecuite» system with the inner heating tube surface of the vacuum apparatus heating chamber at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$ is presented in the following graphs below:

- the unsteady diffusion mass flow of sucrose $j_{12}(\tau)$ on the right border of the area (#1), where the movement of matter from the area (#2) of the larger crystal of the first cell to the area (#1) of the intercrystalline sucrose solution is taken as the positive direction (Figure 3);
- unsteady diffusion mass flow of sucrose $j_{23}(\tau)$ on the left border of the area (#3), where the movement of matter from the area (#2) of the larger crystal of the first cell to the area (#3) of the intercrystalline sucrose solution is taken as the positive direction (Figure 4);
- unsteady diffusion sucrose mass flow $j_{34}(\tau)$ between the right border of the region (#3) and the left border of the region (#4), where the movement of matter from the region (#3) of the intercrystalline sucrose solution of the larger crystal of the first cell to the region (#4) intercrystalline sucrose solution of the smaller crystal of the second cell (Figure 5);
- unsteady diffusion mass flow of sucrose $j_{45}(\tau)$ on the right border of the region (#4), where the movement of matter from the region (#5) of the smaller crystal of the second cell to the region (#4) of the intercrystalline sucrose solution is taken as the positive direction (Figure 6);
- unsteady diffusion mass flow of sucrose $j_{56}(\tau)$ on the left border of the area (#6), where the movement of matter from the area (#5) of the smaller crystal of the second cell to the area (#6) of the intercrystalline sucrose solution is taken as the positive direction (Figure 7).

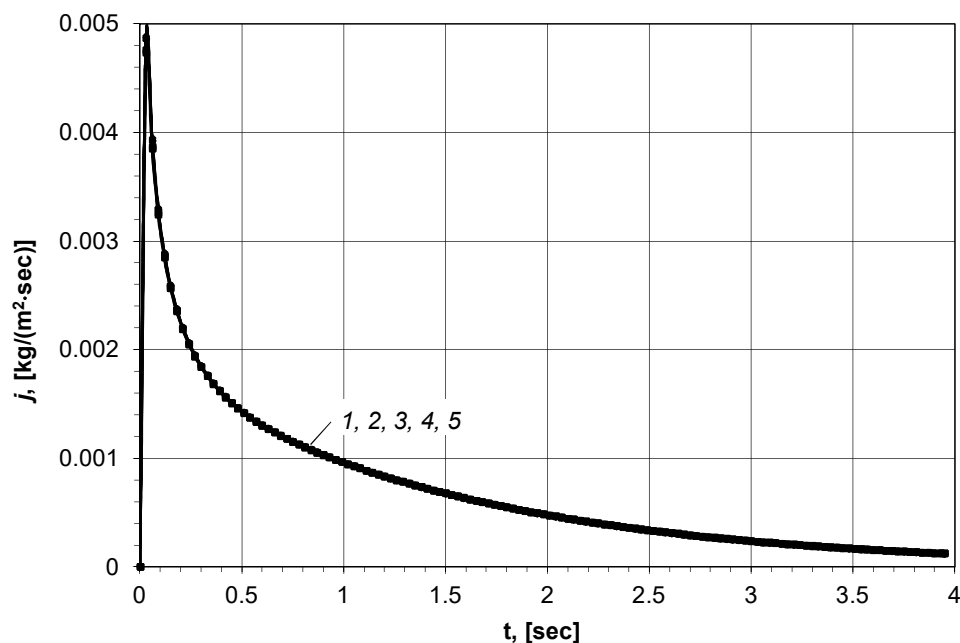


Figure 3. Dependence of the sucrose diffusion mass flow $j_{12}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber on the border (Figure 2) of area (#1), (intercrystalline sucrose solution of the first larger cell) and area (#2), (larger sugar crystal of the first cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$; [value $j_{12}(\tau) > 0$ if sucrose is transferred from region (#2) (larger sugar crystal of the first cell) to region (#1) (intercrystalline sucrose solution of the first larger cell), (Figure 2, bottom footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

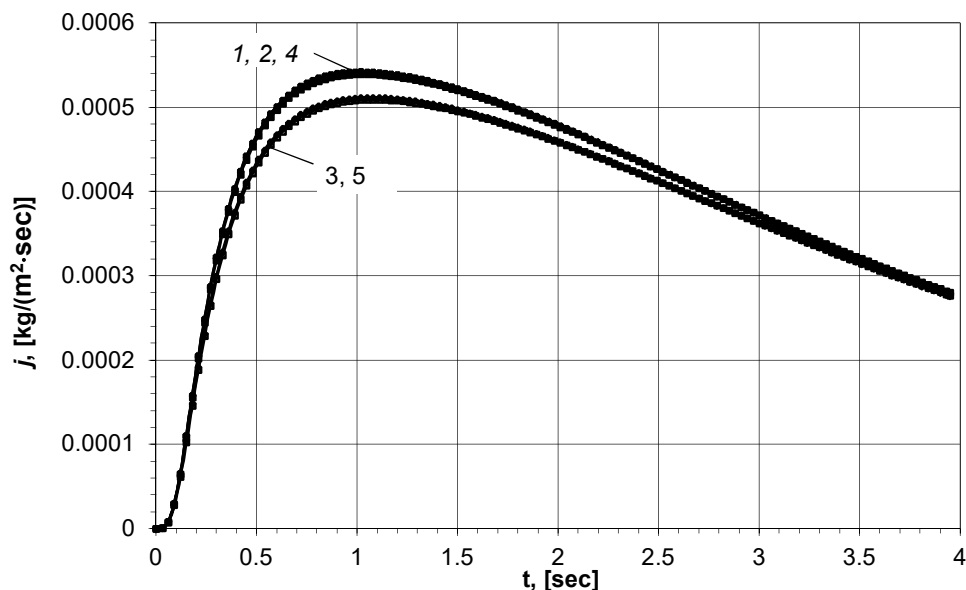


Figure 4. Dependence of the sucrose diffusion mass flow $j_{23}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber on the border (Figure 2) of region (#2), (larger sugar crystal of the first cell) and region (#3), (intercrystalline sucrose solution of the first larger cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$;
[value $j_{23}(\tau) > 0$ if sucrose is transferred from region (#2) (larger sugar crystal of the first cell) to region (#3) (intercrystalline sucrose solution of the first larger cell), (Figure 2, bottom footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

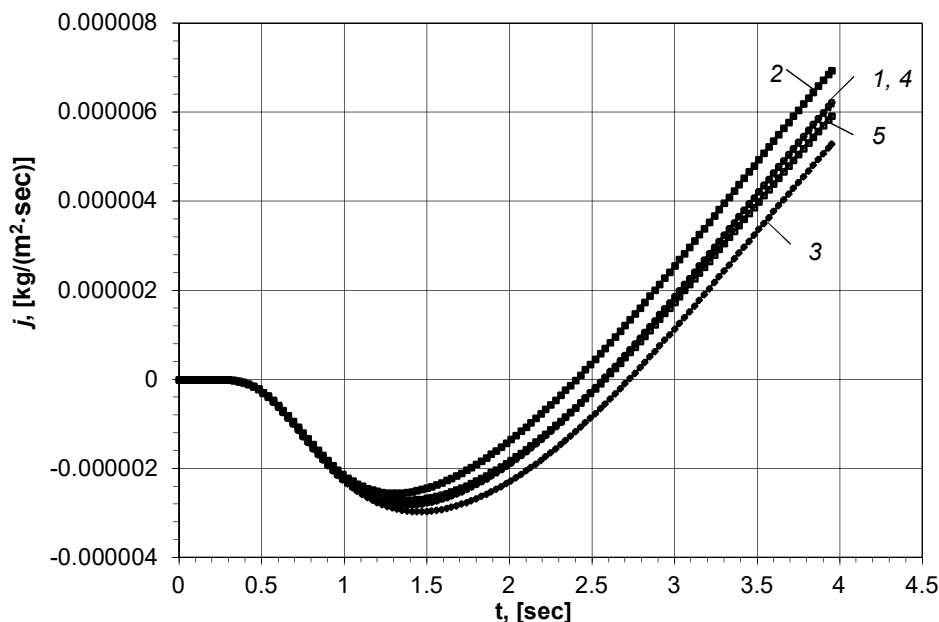


Figure 5. Dependence of the sucrose diffusion mass flow $j_{34}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber on the border (Figure 2) of region (#3), (intercrystalline sucrose solution of the first larger cell) and region (#4), (intercrystalline sucrose solution of the second smaller cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$;

[value $j_{34}(\tau) > 0$ if sucrose is transferred from region (#3) (intercrystalline sucrose solution of the first larger cell) to region (#4) (intercrystalline sucrose solution of the second smaller cell), (Figure 2, footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

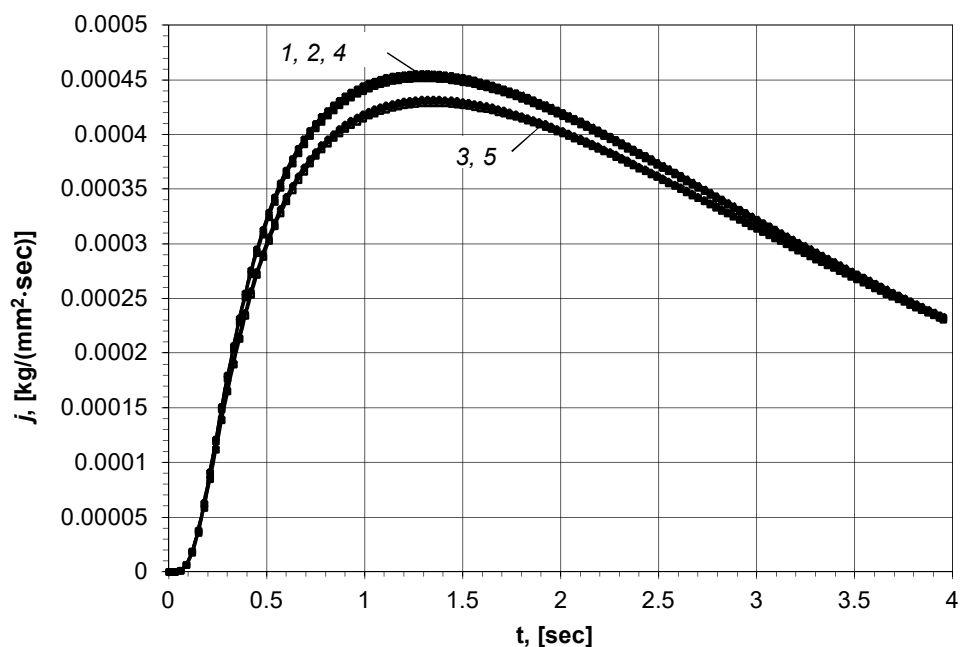


Figure 6. Dependence of the sucrose diffusion mass flow $j_{45}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber on the border (Figure 2) of area (#4), (intercrystalline sucrose solution of the second smaller cell) and area (#5), (smaller sugar crystal of the second cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$;
[value $j_{45}(\tau) > 0$ if sucrose is transferred from region (#5) (smaller sugar crystal of the second cell) to region (#4) (intercrystalline sucrose solution of the second smaller cell), (Figure 2, bottom footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

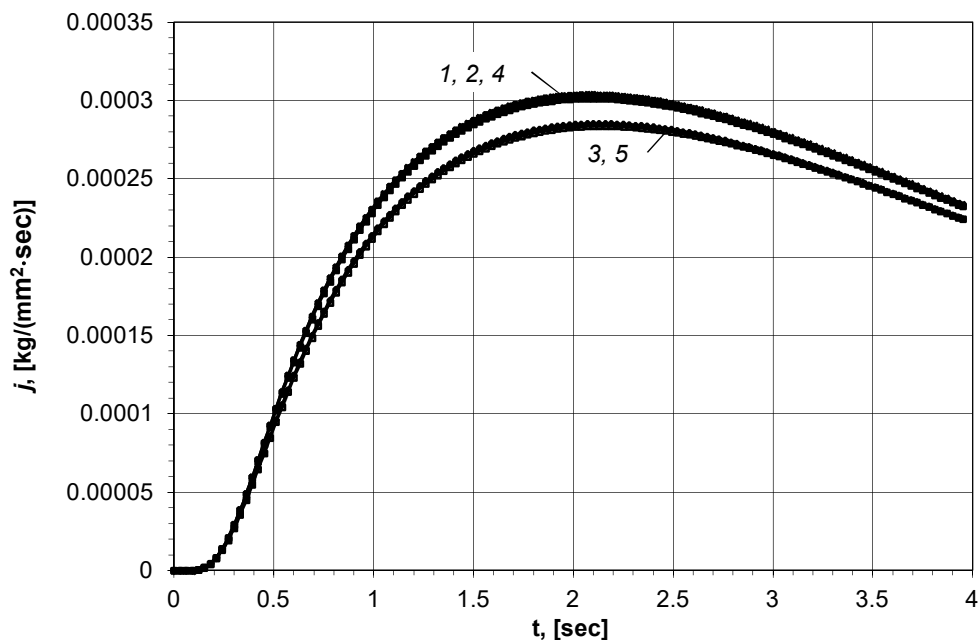


Figure 7. Dependence of the sucrose diffusion mass flow $j_{s6}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the the vacuum apparatus heating chamber on the border (Figure 2) of region (#5), (smaller sugar crystal of the second cell) and region (#6), (intercrystalline sucrose solution of the second smaller cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$;
[value $j_{s6}(\tau) > 0$ if sucrose is transferred from region (#5) (smaller sugar crystal of the second cell) to region (#6) (intercrystalline sucrose solution of the second smaller cell), (Figure 2, bottom footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

The obtained results of calculations and studies of the distribution of non-stationary diffusion mass flows of sucrose $j_{mn}(\tau)$, ($m=1-5$, $n=m+1$), at the boundaries of intercrystalline sucrose solutions regions (#1), (#3), (#4) and (#6), (Figure 2) depending on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber at eight values of the relative boiling time $\tau/\tau_c = (0.2 - 0.9)$, – it is not possible to submit due to the limited scope of this article.

We will present only the results of boiling at a relative time of $\tau/\tau_c = 1.0$.

Distribution of unsteady diffusion mass flows of sucrose $j_{mn}(\tau)$, ($m=1-5$, $n=m+1$), at the boundaries of intercrystalline sucrose solutions regions (#1), (#3), (#4) and (#6), (Figure 2) depending on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$ is presented in the following graphs below:

- unsteady diffusion mass flow of sucrose $j_{12}(\tau)$ on the right border of the area (#1), where the positive direction is taken (as in Figure 3), the movement of matter from the area (#2) of the larger crystal of the first cell to the area (#1) intercrystalline solution of sucrose, (Figure 8);
- unsteady diffusion mass flow of sucrose $j_{23}(\tau)$ on the left border of the region (#3), where the positive direction is taken (as in Figure 4) to be the movement of matter from the region (#2) of the larger crystal of the first cell to the region (#3) intercrystalline solution of sucrose, (Figure 9);
- unsteady diffusion mass flow of sucrose $j_{34}(\tau)$ between the right border of the region (#3) and the left border of the region (#4), where the positive direction is taken (as in Figure 5) to be the movement of matter from the region (#3) the intercrystalline sucrose solution of the larger crystal of the first cell into the region (#4) of the intercrystalline sucrose solution of the smaller crystal of the second cell (Figure 10);
- unsteady diffusion mass flow of sucrose $j_{45}(\tau)$ on the right border of the area (#4), where the positive direction is taken (as in Figure 6) to be the movement of matter from the area (#5) of the smaller crystal of the second cell to the area (#4) intercrystalline sucrose solution, (Figure 11);
- unsteady diffusion mass flow of sucrose $j_{56}(\tau)$ on the left border of the region (#6), where the positive direction is taken (as in Figure 7) to be the movement of matter from the region (#5) of the smaller crystal of the second cell to the region (#6) intercrystalline sucrose solution, (Figure 12).

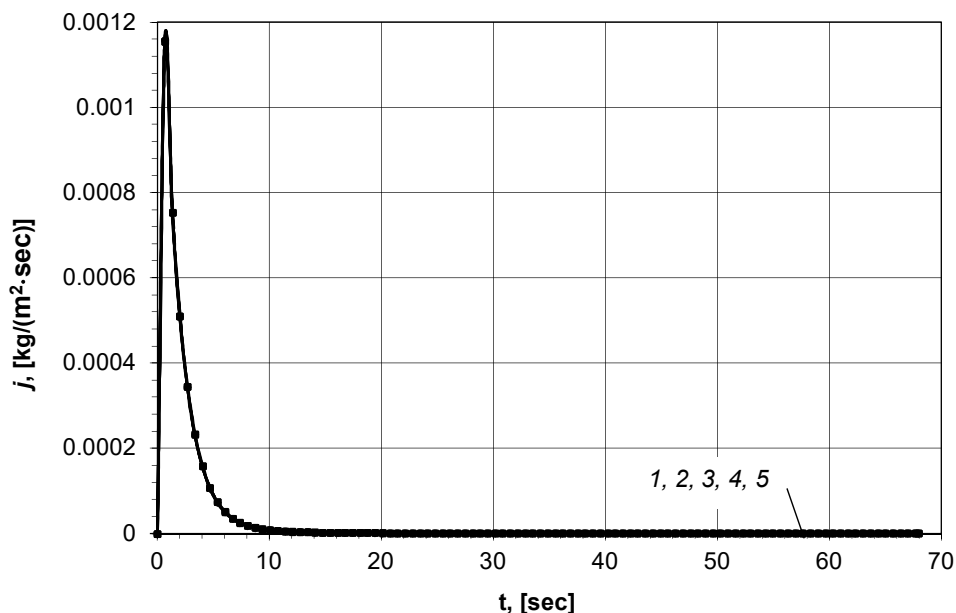


Figure 8. Dependence of the sucrose diffusion mass flow $j_{12}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber on the border (Figure 2) of area (#1), (intercrystalline sucrose solution of the first larger cell) and area (#2), (larger sugar crystal of the first cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$; [value $j_{12}(\tau) > 0$ if sucrose is transferred from region (#2) (larger sugar crystal of the first cell) to region (#1) (intercrystalline sucrose solution of the first larger cell), (Figure 2, bottom footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

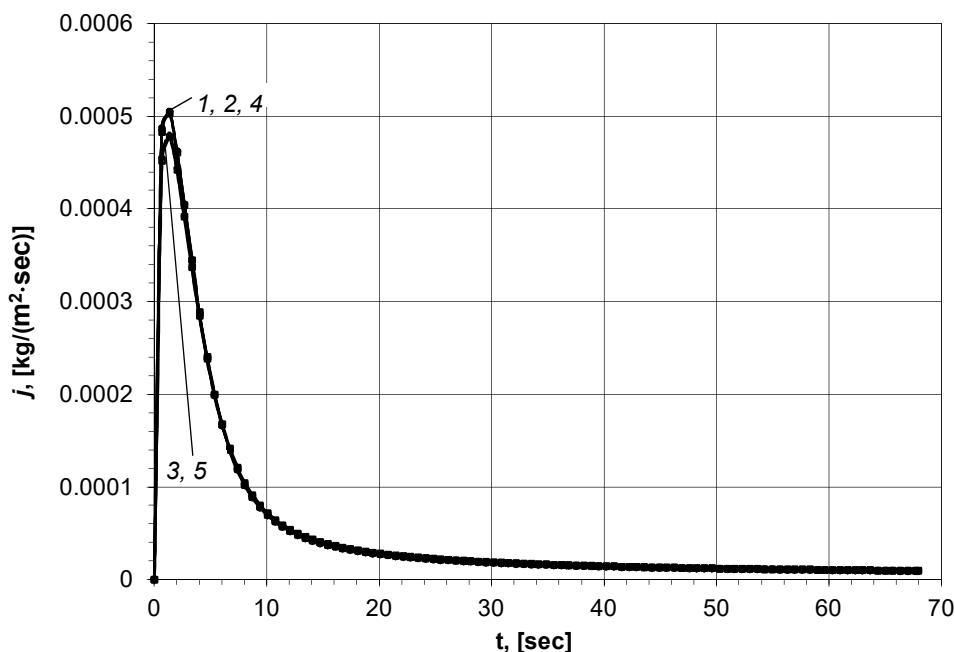


Figure 9. Dependence of the sucrose diffusion mass flow $j_{z3}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber on the border (Figure 2) of region (#2), (larger sugar crystal of the first cell) and region (#3), (intercrystalline sucrose solution of the first larger cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$;
[value $j_{z3}(\tau) > 0$ if sucrose is transferred from region (#2) (larger sugar crystal of the first cell) to region (#3) (intercrystalline sucrose solution of the first larger cell), (Figure 2, bottom footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

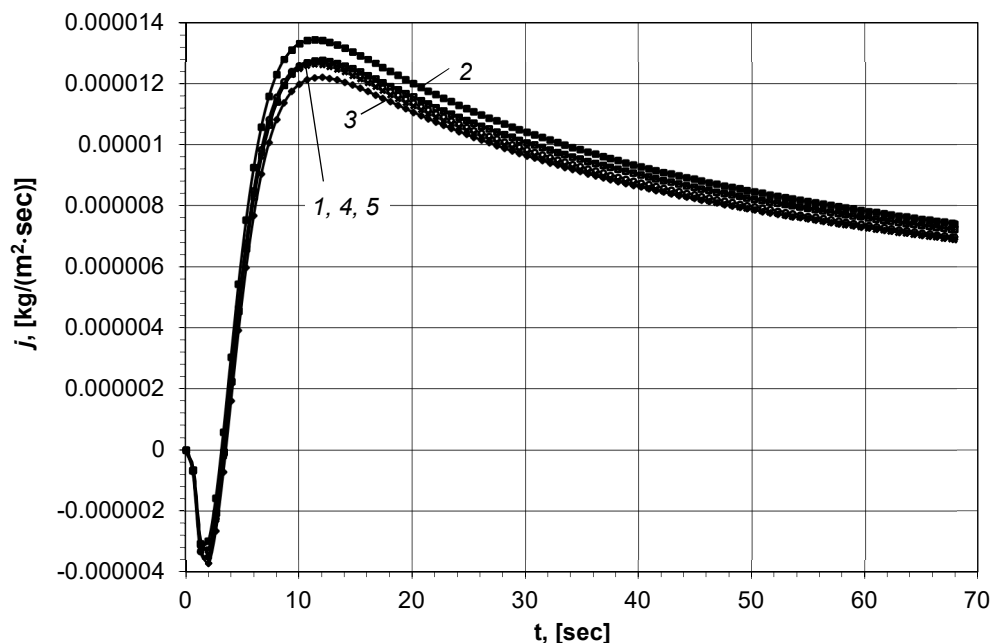


Figure 10. Dependence of the sucrose diffusion mass flow $j_{34}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber on the border (Figure 2) of region (#3), (intercrystalline sucrose solution of the first larger cell) and region (#4), (intercrystalline sucrose solution of the second smaller cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$;

[value $j_{34}(\tau) > 0$ if sucrose is transferred from region (#3) (intercrystalline sucrose solution of the first larger cell) to region (#4) (intercrystalline sucrose solution of the second smaller cell), (Figure 2, footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

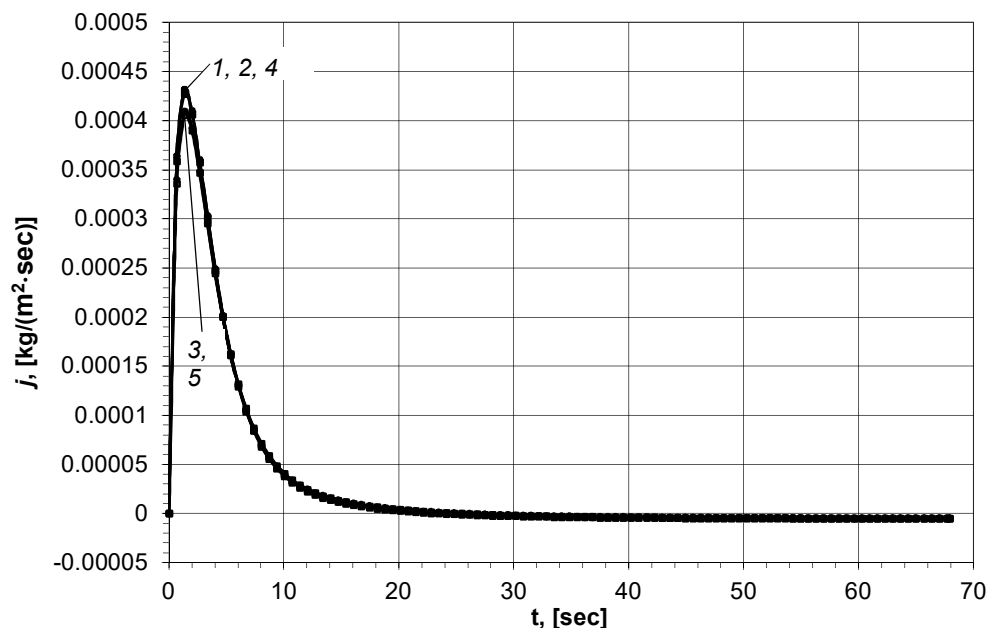
4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).



**Figure 11. Dependence of the sucrose diffusion mass flow $j_{45}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber on the border (Figure 2) of area (#4), (intercrystalline sucrose solution of the second smaller cell) and area (#5), (smaller sugar crystal of the second cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$;
[value $j_{45}(\tau) > 0$ if sucrose is transferred from region (#5) (smaller sugar crystal of the second cell) to region (#4) (intercrystalline sucrose solution of the second smaller cell), (Figure 2, bottom footnote)].**

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

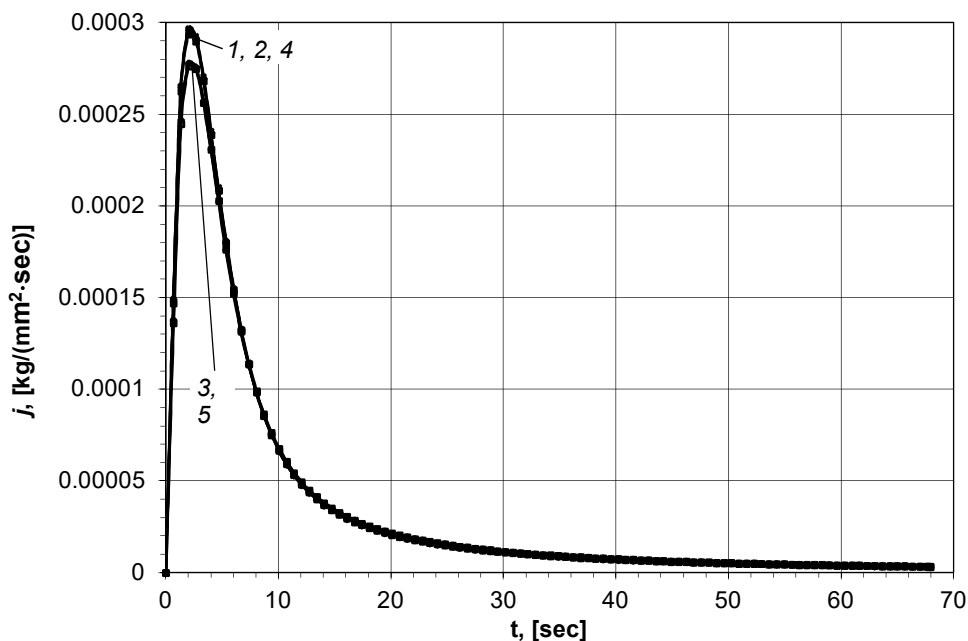


Figure 12. Dependence of the sucrose diffusion mass flow $j_{s6}(\tau)$ on the contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the the vacuum apparatus heating chamber on the border (Figure 2) of region (#5), (smaller sugar crystal of the second cell) and region (#6), (intercrystalline sucrose solution of the second smaller cell) with the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$;
 [value $j_{s6}(\tau) > 0$ if sucrose is transferred from region (#5) (smaller sugar crystal of the second cell) to region (#6) (intercrystalline sucrose solution of the second smaller cell), (Figure 2, bottom footnote)].

*** Designations:**

1 – all thermophysical characteristics (coefficients): for the crystal, for the intercrystalline sucrose solution and for the massecuite there are constant values (option I);

2 – thermophysical characteristics (coefficients):

all for the crystal, diffusion coefficient for the intercrystalline sucrose solution and all for the massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the intercrystalline sucrose solution, are variable values (option II, a);

3 – thermophysical characteristics (coefficients):

all for intercrystalline sucrose solution and all for massecuite are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for the crystal are variable quantities (option II, b);

4 – thermophysical characteristics (coefficients):

all for the crystal, all for the intercrystalline solution are constant values;

thermal conductivity coefficient, heat capacity coefficient and density for massecuite, are variable quantities (option II, c);

5 – all thermophysical characteristics (coefficients):

for the crystal, for the intercrystalline solution, and for the massecuite are variable values (option III).

The graphs (Figures 3, 4, 8, 9) show that the larger crystal of the first cell is dissolving. As can be seen from the obtained graphs (Figs. 3, 4), the nature of the change in diffusion mass flows from one side of the larger crystal of the first cell (Figs. 1, 2), which is closer to the heating wall of the vacuum apparatus heating chamber (Figure 3), is not quite coincides with the nature of the change in diffusion mass flows from the other side (Figure 4) at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$. But we get a completely different situation when $\tau/\tau_c = 1.0$, where the nature of the change in the form of diffusion mass flows is almost the same (Figs. 8, 9). It is clear that the closer the cell is to the heating wall of the vacuum apparatus heating chamber, the faster the process of heating the components of this cell of the «two cells–massecuite» system occurs, and therefore the diffusion mass flow reaches its maximum in time earlier than from the other side of the crystal.

As for the second, smaller cell of the «two cells–massecuite» system, which is located further from the heating wall (Figs. 1, 2), the nature of the change in diffusion mass flows from one and the other side of the smaller crystal of the second cell (Figure 6, 7), practically coincide with each other at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$, and at $\tau/\tau_c = 1.0$. Although, again, the graphs show that the intercrystalline solution on the side of the heating wall heats up faster, and as a result, the diffusive mass flows reach their maximum faster. The decline of diffusive mass flows after reaching their maximum can be explained by the fact that a saturated state is reached in each region of the intercrystalline sucrose solution.

The uneven movement nature of diffusion mass flows between cells (Figs. 5, 10), that is, between regions of intercrystalline sucrose solutions, can also be explained by the fact that the first cell warms up faster than the second. As a result, at the first stage, the intercrystalline sucrose solution of the first cell moves from the saturated state to the unsaturated state faster than the second cell. This leads to the dissolution of the first crystal faster than the second. But the rate of the crystal dissolution does not cover the need to reach a saturated state of the intercrystalline sucrose solution in the first cell, as a result of which sucrose is transferred from the second cell to the first (Figure 5, 10).

At the second stage, the saturated state of the first cell is equalized and at the same time the second cell has time to warm up sufficiently. At the same time, the saturation coefficient also decreases in its intercrystalline sucrose solution. Then the flow of sucrose already begins to occur from the intercrystalline sucrose solution of the first cell to the intercrystalline sucrose solution of the second cell.

All this plays an indicative role in the recrystallization process. It is clear that predicting the nature of the sucrose movement within one cell, and even more so between the cells itself, was quite a difficult task before conducting research. But now we have obtained the main indicators of diffusional flow of sucrose between cells and inside cells, albeit on the basis of an idealized mathematical model.

In order to compare the effect of taking into account variable thermophysical coefficients on the diffusion mass transfer process in an intercrystalline solution with constant thermophysical coefficients, a study was conducted and the maximum and minimum values of the relative deviations of the diffusion mass flows values $j_{mn}(\tau)$, ($m = 1-5$, $n = m + 1$), were found, (Figure 2).

Let us denote the diffusive mass flows, which were calculated at constant thermophysical coefficients by $j_{mn, \text{const}}(\tau)$, ($m = 1-5$, $n = m + 1$). Similarly, let's denote diffusion mass flows, which were calculated with variable thermophysical coefficients through $j_{mn, \text{var}}(\tau)$, ($m = 1-5$, $n = m + 1$).

The relative change of diffusion mass flows will be carried out in two cases:

- during the entire stay of the «two cells–massecuite» system (Figures 1, 2) in the heating tube of the vacuum apparatus heating chamber;
- at the time of exit of the «two cells–massecuite» system from the heating tube.

For comparison, we will find the largest deviation of diffusion mass flows $j_{mn,var}(\tau)$, ($m = 1-5$, $n = m + 1$; $0 \leq \tau \leq \tau_{c,end}$), from $j_{mn,const}(\tau)$, ($m = 1-5$, $n = m + 1$; $0 \leq \tau \leq \tau_{c,end}$). And then already on the basis of these data we will be able to draw a conclusion: which method should be used for further mathematical modeling. For this, it is necessary to find the maximum and minimum deviation of the values of diffusion mass flows $j_{mn,var}(\tau)$, ($m = 1-5$, $n = m + 1$; $0 \leq \tau \leq \tau_{c,end}$), from $j_{mn,const}(\tau)$, ($m = 1-5$, $n = m + 1$; $0 \leq \tau \leq \tau_{c,end}$).

Relative changes in diffusion mass flows $j_{mn}(\tau)$, ($m = 1-5$, $n = m + 1$; $0 \leq \tau \leq \tau_{c,end}$), at the boundaries of the corresponding regions of intercrystalline sucrose solutions (#1), (#3), (#4) and (#6), (Figure 2), calculated during the entire contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$ given in Table 1.

Table 1
Relative changes in diffusion mass flows $j_{mn}(\tau)$, ($m = 1-5$, $n = m + 1$; $0 \leq \tau \leq \tau_{c,end}$), calculated during the entire contact time τ at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$

$\tau_c = (0.0-3.95) \text{ c}$ ($\tau/\tau_c = 0.15$) (Min–Max)	II, a(lq.var) / $I_{(all.const)}, \%$	II, b(cr.var) / $I_{(all.const)}, \%$	II, c(ms.var) / $I_{(all.const)}, \%$	III(all.var) / $I_{(all.const)}, \%$
$(j_{12,var}(\tau) / j_{12,const}(\tau) - 1) _{min} \cdot 100\%$	– 2.5 ($\tau = 0.03 \text{ s}$)	0.0 $\tau = 0.39-3.95 \text{ s}$	0.0 $\tau = 0.00-3.95 \text{ s}$	– 2.1 ($\tau = 0.03 \text{ s}$)
$(j_{12,var}(\tau) / j_{12,const}(\tau) - 1) _{max} \cdot 100\%$	0.0 $\tau = 0.69-3.95 \text{ s}$	+ 0.5 $\tau = 0.03-0.06 \text{ s}$	0.0 $\tau = 0.00-3.95 \text{ s}$	0.0 $\tau = 0.51-3.95 \text{ s}$
$(j_{23,var}(\tau) / j_{23,const}(\tau) - 1) _{min} \cdot 100\%$	– 0.8 $\tau = 0.36-0.48 \text{ s}$	– 6.2 $\tau = 0.6-0.72 \text{ s}$	0.0 $(\tau = 0.0-2.91 \text{ s})$	– 6.8 $\tau = 0.57-0.69 \text{ s}$
$(j_{23,var}(\tau) / j_{23,const}(\tau) - 1) _{max} \cdot 100\%$	0.1 $\tau = 3.18-3.95 \text{ s}$	0.0 $(\tau = 0.0-0.06 \text{ s})$	0.1 $\tau = 3.03-3.95 \text{ s}$	0.0 $\tau = 0.0-0.03 \text{ s}$
$(j_{34,var}(\tau) / j_{34,const}(\tau) - 1) _{min} \cdot 100\%$	0.0 $(\tau = 0.0-0.48 \text{ s})$	– 10.3 $\tau = 3.93-3.95 \text{ s}$	– 0.5 $\tau = 3.84-3.95 \text{ s}$	– 3.3 $\tau = 2.93-2.95 \text{ s}$
$(j_{34,var}(\tau) / j_{34,const}(\tau) - 1) _{max} \cdot 100\%$	8.1 $\tau = 3.87-3.95 \text{ s}$	0.5 $\tau = 0.69-0.84 \text{ s}$	0.0 $(\tau = 0.0-1.8 \text{ s})$	1.3 $\tau = 0.99-1.26 \text{ s}$
$(j_{45,var}(\tau) / j_{45,const}(\tau) - 1) _{min} \cdot 100\%$	– 1.0 $(\tau = 0.6-0.96 \text{ s})$	– 5.7 $\tau = 0.75-0.96 \text{ s}$	0.0 $(\tau = 0.0-2.25 \text{ s})$	– 6.6 $\tau = 0.68-0.9 \text{ s}$
$(j_{45,var}(\tau) / j_{45,const}(\tau) - 1) _{max} \cdot 100\%$	0.0 $(\tau = 0.0-0.09 \text{ s})$	0.0 $(\tau = 0.0-0.06 \text{ s})$	0.1 $\tau = 2.28-3.95 \text{ s}$	0.0 $\tau = 0.0-0.06 \text{ s}$
$(j_{56,var}(\tau) / j_{56,const}(\tau) - 1) _{min} \cdot 100\%$	– 0.8 $\tau = 0.87-1.86 \text{ s}$	– 6.4 $\tau = 1.44-1.74 \text{ s}$	0.0 $(\tau = 0.0-1.41 \text{ s})$	– 7.1 $\tau = 1.53-1.56 \text{ s}$
$(j_{56,var}(\tau) / j_{56,const}(\tau) - 1) _{max} \cdot 100\%$	0.0 $(\tau = 0.0-0.21 \text{ s})$	0.0 $(\tau = 0.0-0.15 \text{ s})$	0.2 $\tau = 2.61-3.95 \text{ s}$	0.0 $\tau = 0.0-0.15 \text{ s}$

Relative changes in diffusion mass fluxes $j_{mn}(\tau_{c,end})$, ($m=1-5, n=m+1$), at the boundaries of the corresponding regions of intercrystalline sucrose solutions (#1), (#3), (#4) and (#6), (Figure 2), calculated at the moment of time $\tau_{c,end}$ of the exit of the entire system «two cells–massecuite» from the heating tube of the vacuum apparatus heating chamber at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$ given in Table 2.

Relative changes in diffusion mass flows $j_{mn}(\tau)$, ($m = 1-5$, $n = m + 1$; $0 \leq \tau \leq \tau_{c,end}$), at the boundaries of the corresponding regions of intercrystalline sucrose solutions (#1), (#3), (#4) and (#6), (Figure 2), calculated during the entire contact time τ of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$ given in Table 3.

Relative changes in diffusion mass fluxes $j_{mn}(\tau_{c,end})$, ($m=1-5$, $n=m+1$), at the boundaries of the corresponding regions of intercrystalline sucrose solutions (#1), (#3), (#4) and (#6), (Figure 2), calculated at the moment of time $\tau_{c,end}$ of the exit of the entire system «two cells–massecuite» from the heating tube of the vacuum apparatus heating chamber at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$ given in Table 4.

In Tables 1–4, the notation of various variants of the performed calculations is used:

- option I_(all.const)– constant thermophysical coefficients;
- option (II, a)_(liq.var)– variable thermophysical coefficients only for regions of intercrystalline sucrose solutions (#1), (#3), (#4) and (#6), (Figure 2);
- (II, b)_(cr.var)– variable thermophysical coefficients only for regions of sugar crystals (#2) and (#5), (Figure 2);
- (II, c)_(ms.var)– variable thermophysical coefficients only for the massecuite region (#7), (Figure 2);
- (III)_(all.var)– variable thermophysical coefficients for all regions (#1)–(#7), (Figure 2), simultaneously.

From Tables 1 and 2, it can be concluded that taking into account the influence of variable thermophysical coefficients on the non-stationary process of diffusion mass transfer is clearly non-linear and non-uniform in nature.

For an example in Table 1, the relative change in diffusion mass flow $j_{34}(\tau)$ in the case of variable thermophysical coefficients only for sugar crystals for the case $\tau/\tau_u = 0.15$, with contact time $\tau = 0.69-0.84$ s, is 0.5%, and already at the contact time $\tau = 3.93-3.95$ s, it is – 10.3%. Similarly, according to Table 3, the relative change in the diffusion mass flow $j_{23}(\tau)$ in the case of all simultaneously changing thermophysical coefficients for the case of $\tau/\tau_c = 1.0$, with a contact time of $\tau = 0.67$ s, is – 6.2%, and already at the contact time $\tau = 7.37-11.39$ s, it is 0.3%.

That is, the deviation of diffusive mass flows with variable thermophysical coefficients from constant ones during the entire contact time of the "two cell-heater" system with the heating tube is positive at one point in time, and negative at another point in time (that is, it has a clearly expressed variable character). It is clear that in such cases it is possible to find moments of time τ where diffusive mass flows with variable thermophysical coefficients will completely coincide with diffusive mass flows with constant thermophysical coefficients.

Let's examine in more detail the weight of the impact of taking into account variable thermophysical coefficients on the process of diffusion mass transfer in comparison with constant coefficients. For this purpose, we will find the maximum relative deviations of diffusive mass flows with variables, where the case with constant coefficients was taken as the basic values.

Diffusion mass flows with variable coefficients only in the region of the intercrystalline sucrose solution (option II-a) differ the most from diffusion mass flows with constant coefficients (I) by 8.1% at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$, and by 4.7% at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$ (in both cases for $j_{34}(\tau)$).

Table 2

Relative changes in diffusion mass fluxes $j_{mn}(\tau_{c, \text{end}})$, ($m=1-5$, $n=m+1$), calculated at the moment of time $\tau_{c, \text{end}}$ at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$

$\tau_c = \tau_{c, \text{end}} = 3.95 \text{ c}$ ($\tau/\tau_c = 0.15$)	II a (lq.var) / I (all.const), %	II b (cr.var) / I (all.const), %	II c (ms.var) / I (all.const), %	III (all.var) / I (all.const), %
$(j_{12, \text{var}}(\tau) / j_{12, \text{const}}(\tau) - 1) _{\text{end}} \cdot 100\%$	0.028	- 0.010	0.001	0.018
$(j_{23, \text{var}}(\tau) / j_{23, \text{const}}(\tau) - 1) _{\text{end}} \cdot 100\%$	0.131	- 0.683	0.065	- 0.517
$(j_{34, \text{var}}(\tau) / j_{34, \text{const}}(\tau) - 1) _{\text{end}} \cdot 100\%$	8.065	- 10.299	- 0.473	- 3.298
$(j_{45, \text{var}}(\tau) / j_{45, \text{const}}(\tau) - 1) _{\text{end}} \cdot 100\%$	- 0.173	- 0.413	0.092	- 0.509
$(j_{56, \text{var}}(\tau) / j_{56, \text{const}}(\tau) - 1) _{\text{end}} \cdot 100\%$	- 0.254	- 2.870	0.191	- 2.949

Table 3

Relative changes in diffusion mass flows $j_{mn}(\tau)$, ($m = 1-5$, $n = m + 1$; $0 \leq \tau \leq \tau_{c, \text{end}}$), calculated during the entire contact time τ at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$

$\tau_c = (0.0-67.93) \text{ c}$ ($\tau/\tau_c = 1.0$) (Min–Max)	II a (lq.var) / I (all.const), %	II b (cr.var) / I (all.const), %	II c (ms.var) / I (all.const), %	III (all.var) / I (all.const), %
$(j_{12, \text{var}}(\tau) / j_{12, \text{const}}(\tau) - 1) _{\text{min}} \cdot 100\%$	- 0.3 ($\tau = 0.67 \text{ s}$)	0.0 ($\tau = 1.34-67.93 \text{ s}$)	0.0 ($\tau = 0.0-67.93 \text{ s}$)	- 0.2 ($\tau = 0.67 \text{ s}$)
$(j_{12, \text{var}}(\tau) / j_{12, \text{const}}(\tau) - 1) _{\text{max}} \cdot 100\%$	0.1 ($\tau = 2.01-7.37 \text{ s}$)	0.1 ($\tau = 0.67 \text{ s}$)	0.0 ($\tau = 0.0-67.93 \text{ s}$)	0.1 ($\tau = 2.01-6.03 \text{ s}$)
$(j_{23, \text{var}}(\tau) / j_{23, \text{const}}(\tau) - 1) _{\text{min}} \cdot 100\%$	- 0.8 ($\tau = 0.67 \text{ s}$)	- 6.2 ($\tau = 0.67 \text{ s}$)	0.0 ($\tau = 0.0-2.01 \text{ s}$) ($\tau = 9.38-67.93 \text{ s}$)	- 6.9 ($\tau = 0.67 \text{ s}$)
$(j_{23, \text{var}}(\tau) / j_{23, \text{const}}(\tau) - 1) _{\text{max}} \cdot 100\%$	0.2 $\tau = 4.69-17.42 \text{ s}$	0.3 $\tau = 7.37-11.39 \text{ s}$	0.1 ($\tau = 2.68-8.71 \text{ s}$)	0.6 ($\tau = 7.37-8.71 \text{ s}$)
$(j_{34, \text{var}}(\tau) / j_{34, \text{const}}(\tau) - 1) _{\text{min}} \cdot 100\%$	0.1 ($\tau = 0.67 \text{ s}$)	- 5.0 ($\tau = 6.03 \text{ s}$)	- 0.7 ($\tau = 8.04-47.55 \text{ s}$)	- 1.2 ($\tau = 6.03-6.7 \text{ s}$)
$(j_{34, \text{var}}(\tau) / j_{34, \text{const}}(\tau) - 1) _{\text{max}} \cdot 100\%$	4.7 ($\tau = 4.02-6.03 \text{ s}$)	0.2 ($\tau = 0.67 \text{ s}$)	0.0 ($\tau = 0.0-1.34 \text{ s}$)	1.6 $\tau = 32.16-57.62 \text{ s}$
$(j_{45, \text{var}}(\tau) / j_{45, \text{const}}(\tau) - 1) _{\text{min}} \cdot 100\%$	- 1.1 ($\tau = 0.67 \text{ s}$)	- 5.4 ($\tau = 0.67 \text{ s}$)	0.0 ($\tau = 0.0-1.34 \text{ s}$) $\tau = 14.74-67.93 \text{ s}$	- 6.4 ($\tau = 0.67 \text{ s}$)
$(j_{45, \text{var}}(\tau) / j_{45, \text{const}}(\tau) - 1) _{\text{max}} \cdot 100\%$	0.0 ($\tau = 0.0 \text{ s}$)	0.6 $\tau = 7.37-8.04 \text{ s}$	0.2 $\tau = 4.02-6.03 \text{ s}$	0.6 $\tau = 7.37-8.71 \text{ s}$
$(j_{56, \text{var}}(\tau) / j_{56, \text{const}}(\tau) - 1) _{\text{min}} \cdot 100\%$	- 1.0 ($\tau = 1.34 \text{ s}$)	- 6.2 ($\tau = 1.34-2.01 \text{ s}$)	0.0 ($\tau = 0.0-0.67 \text{ s}$) $\tau = 14.07-67.93 \text{ s}$	- 7.0 ($\tau = 1.34 \text{ s}$)
$(j_{56, \text{var}}(\tau) / j_{56, \text{const}}(\tau) - 1) _{\text{max}} \cdot 100\%$	0.1 $\tau = 8.71-14.07 \text{ s}$	0.4 $\tau = 11.39-14.07 \text{ s}$	0.3 ($\tau = 2.68-6.03 \text{ s}$)	0.5 $\tau = 10.05-14.07 \text{ s}$

Table 4

Relative changes in diffusion mass fluxes $j_{mn}(\tau_{c,end})$, ($m=1-5$, $n=m+1$), calculated at the moment of time $\tau_{c,end}$ at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$

$\tau_c = \tau_{c,end} = 67.93 \text{ c}$ ($\tau/\tau_c = 1.0$)	II a (lq.var) / I (all.const), %	II b (cr.var) / I (all.const), %	II c (ms.var) / I (all.const), %	III (all.var) / I (all.const), %
$(j_{12,var}(\tau) / j_{12,const}(\tau) - 1) \cdot 100\%$	0.001	0.000	0.000	0.001
$(j_{23,var}(\tau) / j_{23,const}(\tau) - 1) \cdot 100\%$	0.089	0.021	-0.024	0.085
$(j_{34,var}(\tau) / j_{34,const}(\tau) - 1) \cdot 100\%$	2.665	-0.487	-0.606	1.512
$(j_{45,var}(\tau) / j_{45,const}(\tau) - 1) \cdot 100\%$	-0.094	0.055	0.018	-0.019
$(j_{56,var}(\tau) / j_{56,const}(\tau) - 1) \cdot 100\%$	0.009	0.074	-0.010	0.073

Diffusion mass flows with variable coefficients only in the region of the crystal (II-b) differ the most from diffusion mass flows with constant coefficients (I) by - 10.3% at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$ (the case for $j_{34}(\tau)$), and by - 6.2% at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$ (cases for $j_{23}(\tau)$ and $j_{56}(\tau)$).

Diffusion mass flows with variable coefficients only in the region of massecuite (II-c) differ the most from diffusion mass flows with constant coefficients (I) by - 0.5% at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$, and by - 0.7% at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$ (in both cases for $j_{34}(\tau)$).

Diffusion mass flows with simultaneously all variable coefficients (III) differ the most from diffusion mass flows with constant coefficients (I) by - 7.1% at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$, and by - 7.0% at the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$ (in both cases for $j_{56}(\tau)$).

As can be seen from Table 1, the closest to the obtained results of sucrose diffusion mass flows in intercrystalline solutions with constant thermophysical coefficients (option I) is the result with variable thermophysical coefficients only for the massecuite region (option II-c), where the maximum relative deviation is - 0.7%.

In second place in terms of the relative deviation from the obtained results of sucrose diffusion mass flows in intercrystalline solutions with constant thermophysical coefficients (option I) is the variant of diffusion mass flows with all simultaneously variable thermophysical coefficients (option III), where the maximum relative deviation is - 7.1%.

In third place in terms of the relative deviation from the obtained results of sucrose diffusion mass flows in intercrystalline solutions with constant thermophysical coefficients (option I) is the variant of diffusion mass flows with variable coefficients only for intercrystalline sucrose solutions (option II-a), where the maximum relative deviation is 8.1%.

Finally, in the last fourth place in terms of relative deviation from the obtained results of sucrose diffusion mass flows in intercrystalline solutions with constant thermophysical coefficients (option I) there is a variant of diffusion mass flows with variable coefficients only for crystals (option II-b), where the maximum relative deviation is - 10.3%.

That is, in the last, fourth case, we get the maximum relative deviations of diffusive mass transfers from the case obtained with constant thermophysical coefficients.

It is worth noting that the thermophysical coefficients of the massecuite are a certain combination of the corresponding thermophysical coefficients for the crystal and the intercrystalline solution. As we can see from the conducted studies, the option only for

intercrystalline solutions of sucrose (option II-a) has the maximum impact with a «+» sign (8.1%). While the variant for sugar crystals only (variant II-b) has the maximum impact with a «-» sign (- 10.3%). Their simultaneous influence was expected to be obtained as the sum of such two influences. However, in reality, we got a slightly different result (- 0.5%), which does not match the numerical value with the expected $8.1\% - 10.3\% = - 2.2\%$ by almost 4.4 times. The only explanation the author can offer is that the process of sugar crystallization is quite complex to understand. Note that in such a complex process as sugar crystallization, the values of certain thermophysical characteristics for massecuite (for example, density, thermal conductivity coefficient) are not calculated based on simple ratios of the thermophysical characteristics of massecuite components (i.e., crystal, intercrystalline sucrose solution).

Before starting the study of the dependence of non-stationary diffusion mass flows on the method used to calculate the process (with constant or variable thermophysical coefficients; alternately or all at once), it was expected to obtain a result that would confirm that the simultaneous consideration of all variable thermophysical characteristics makes the biggest impact.

In fact, we found that taking into account all variable thermophysical coefficients is not the first place, but only the second place. The largest relative deviations were obtained when variable coefficients for crystals were taken into account separately, as well as when variable coefficients were taken into account for intercrystalline sucrose solutions.

Again, this can be explained only by the fact that the sucrose mass crystallization process is very complex and has a pronounced nonlinear character. Moreover, the crystallization process investigated in this work is non-stationary.

Summing up, it can be concluded that the influence of variable thermophysical coefficients on the relative changes in the values of diffusive mass flows in comparison with constant thermophysical coefficients during the entire time of contact of the «two cells–massecuite» system with the inner wall of the heating tube of the vacuum apparatus heating chamber is within the range of - 10.3% to + 8.1%.

With regard to the final results at the final moment of time $\tau = \tau_{c, \text{end}}$ of the «two cells–massecuite» system contact at the exit from the heating tube the deviations are as follows:

- at $\tau/\tau_c = 0.15$, the largest relative deviations were obtained when taking into account variable thermophysical coefficients only for sugar crystals (- 10.3%), then variable thermophysical coefficients only for intercrystalline sucrose solutions (+ 8.1%); then with all variable coefficients at the same time (- 3.3%); and finally, taking into account variable coefficients only for massecuite (- 0.5%);
- at $\tau/\tau_c = 1.0$, the largest relative deviations were obtained when taking into account variable thermophysical coefficients only for intercrystalline solutions of sucrose (+ 2.7%); then with all variable coefficients at the same time (+ 1.5%); further, taking into account variable coefficients only for massecuite (- 0.6%); and finally, taking into account variable coefficients only for sugar crystals (- 0.5%).

Summing up, it can be concluded that the influence of variable thermophysical coefficients on the relative changes in the values of diffusive mass flows in comparison with constant thermophysical coefficients at the output of the «two cells–massecuite» system from the heating tube of the vacuum apparatus heating chamber is within the range of - 10.3% up to + 8.1%.

Also, it was established that at the value of the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$, when the entire system of cells moves along the heating tube, the substance is first transferred from the region of the intercrystalline sucrose solution of a smaller crystal to the region

of the intercrystalline sucrose solution of a larger crystal during $\tau_{c,2} = 2.41\text{--}2.7$ s depending on constant or variable thermophysical characteristics (Figure 5). Starting from the moment of time $\tau_{c,2}$ to the moment of time $\tau_{c,\text{end}} = 3.95$ s (at $\tau/\tau_c = 0.15$) when the system of cells leaves the heating tube, the situation changes to the opposite (Figure 5). So, in this case, only one extreme of the diffusion mass flow was clearly expressed (the minimum on the graph, which is reached at the moment of time $\tau_{c,1} = 1.29\text{--}1.45$ s, determines the maximum mass transfer of sucrose from a smaller crystal to a larger crystal). There is no clear maximum in this case.

At the relative time of boiling sugar syrup $\tau/\tau_c = 1.0$, two extremes were clearly expressed (Figure 10):

- minimum at $\tau_{c,2} = 1.34\text{--}2.01$ s and
- maximum at $\tau_{c,2} = 11.39\text{--}12.06$ s, depending on constant or variable thermophysical characteristics.

Conclusions

The effect of taking into account variable thermophysical coefficients on the results of calculations of unsteady diffusion mass flows of sucrose for the system «two cells–massecuite» (Figure 1, 2) was investigated, as for the case of their alternate change (only separately for the intercrystalline solution of sucrose, only separately for sugar crystals and only separately for utfel), as well as for the case of simultaneous change of all thermophysical coefficients.

The distribution of non-stationary diffusive mass flows was obtained on the basis of the simultaneous solution of a large system of partial differential equations consisting of a non-stationary heat conduction problems system and simultaneously three different systems for non-stationary diffusive mass transfers problems. Solving such a complex system of problems was carried out using numerical methods.

For a deeper understanding, cases were considered for a certain series of relative time of boiling sugar massecuite $\tau/\tau_c = 0.15; -1.0$.

This paper presents the distribution of non-stationary diffusion mass flows of sucrose only for the case of relative boiling time: $\tau/\tau_c = 0.15$ and $\tau/\tau_c = 1.0$.

To determine the influence of variable thermophysical coefficients on the calculations, the obtained unsteady diffusion mass flows of sucrose at variable thermophysical coefficients were compared with the unsteady diffusion mass flows of sucrose obtained at constant thermophysical coefficients (subject to the same other conditions).

The comparison was made for two cases:

- a) during the entire time of contact of the «two cells–massecuite» system with the inner surface of the heating tube of the vacuum apparatus heating chamber ($0 \leq \tau \leq \tau_{c,\text{end}}$);
- b) at the moment of time when the «two cells–massecuite» system leaves this heating tube ($\tau = \tau_{c,\text{end}}$).

For option (a), the following studies were conducted:

- to determine the weight of the impact of taking into account variable thermophysical coefficients on the obtained result of diffusion mass flows of sucrose; they give reasons to accept the calculations made with constant coefficients close to the calculations made with variable coefficients only for massecuite; the maximum relative deviation in this case is: -0.5% at $\tau/\tau_c = 0.15$, and -0.7% at $\tau/\tau_c = 1.0$;

- unsteady diffusive mass flows with variable thermophysical coefficients only in the region of the crystal differ from the calculation option with constant coefficients by – 10.3% at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$, and by – 6.2% at to the relative time of boiling sugar syrup $\tau/\tau_c = 1.0$;
- non-stationary diffusive mass flows with variable thermophysical coefficients only in the region of the intercrystalline sucrose solution differ from the calculation option with constant coefficients by 8.1% at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$, and by 4.7% at to the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$;
- finally, the conducted studies of non-stationary diffusion mass flows with all simultaneously variable thermophysical coefficients differ from the calculation option with constant coefficients by – 7.1% at the relative time of boiling sugar massecuite $\tau/\tau_c = 0.15$, and – 7.0% with the relative time of boiling sugar massecuite $\tau/\tau_c = 1.0$.

Summarizing the research carried out for option (b), it can be concluded that the influence of variable thermophysical coefficients on the relative changes in the values of diffusion mass flows in comparison with constant thermophysical coefficients at the output of the «two cells–massecuite» system from the heating tube of the vacuum apparatus heating chamber is ranges from – 10.3% to + 8.1%.

A general conclusion can be made as follows. Taking into account variable thermophysical coefficients affects the non-stationary process of diffusion mass transfer for intercrystalline solutions of sucrose in the components of the «two cells–massecuite» system. The limits of this influence are from – 10.3% to + 8.1% of relative units.

If the researcher is satisfied with deviations of this order, he can further use thermophysical coefficients for calculations of steel (note that the calculation time of the entire system of problems in this case is significantly reduced by many times). Otherwise, it is worth using a more accurate model in which variable thermophysical coefficients will be taken into account, and all components of the «two cells–massecuite» system at the same time (maximum relative deviations in this case will be 7.1%).

References

- Alewijn W.F., Honig P. (2013), Chapter 9. Technology of sugar crystallization, *Crystallization*, Elsevier, pp. 318–370.
- Bruno J. C. de Castro, Melécio Marciniuk Junior, Marco Giulietti, André Bernardo (2019), Sucrose crystallization: modeling and evaluation of production responses to typical process fluctuations, *Brazilian Journal of Chemical Engineering*, 36(3), pp. 1237–1253, dx.doi.org/10.1590/0104-6632.20190363s20180240.
- Crestani C. E., Bernardo A., Costa C.B.B., Giulietti M. (2021), An artificial neural network model applied to convert sucrose chord length distributions into particle size distributions, *Powder Technology*, 384, pp. 186–194, <https://doi.org/10.1016/j.powtec.2021.01.075>
- Duroudier J.P. (2016), *Crystallization and Crystallizers*, Industrial Equipment for Chemical Engineering Set, London.
- Eymard R. Gallouët, T. R. Herbin R. (2000), *The finite volume method. Handbook of Numerical Analysis*, VII, pp. 713–1020.

- Gakhovych N., Savhenko I. (2022), Strategic priorities of Ukraine's economy recovery based on the sustainable development principles, *Selected papers of IV International Conference on European Dimensions of Sustainable Development, October 20–21, 2022, Kyiv, NUFT, Kyiv*.
- Georgieva P., Feye de Azevedo S., Goncalves M.J., Ho P. (2003). Modeling of sugar crystallization through knowledge integration, *Engineering in Life Sciences*, 3, pp. 146–153, <http://doi.org/10.1002/elsc.200390019>
- Hartel R.W., Shastry A.V. (1991), Sugar crystallization in food products, *Critical Reviews in Food Science and Nutrition*, 30(1), pp. 49–112. <https://doi.org/10.1080/10408399109527541>
- Kulinchenko V., Mironchuk V. (2012), *Industrial Crystallization of Sugary Substances*, NUFT, Kyiv.
- LeVeque R. (2002), *Finite Volume Methods for Hyperbolic Problems*, Cambridge University Press.
- Nagy Z.K., Fevotte G., Kramer H., Simon L.L. (2013), Recent advances in the monitoring, modelling and control of crystallization systems, *Chemical Engineering Research and Design*, 91(10), pp. 1903–1922.
- Osmanbegovic N., Alopaeus V., Han B., Vuorinen V., Louhi-Kultanen M. (2022), Experimental and CFD study on influence of viscosity on layer melt crystallization, *Separation and Purification Technology*, 284, pp. 1–8, <https://doi.org/10.1016/j.seppur.2021.120170>
- Pogoriliy T. (2015a), The distribution of temperatures in the sucrose solution–sugar crystal–sucrose solution–massecuite cells depending on the boiling sugar massecuite time, *Ukrainian Journal of Food Science*, 3(1), pp. 139–148.
- Pogoriliy T. (2015b), Temperatures distribution in the «larger sugar crystal–larger crystal sucrose solution–less crystal sugar sucrose solution–smaller sugar crystal–massecuite» cells system depending on the boiling sugar massecuite time, *Ukrainian Food Journal*, 4(4), pp. 648–661.
- Pogoriliy T. (2015c) Simultaneous unsteady calculation of temperature distribution in the «larger sugar crystal–larger sugar crystal sucrose solution–less sugar crystal sucrose solution–smaller sugar crystal–massecuite» system cells and sucrose solutions cells concentrations in the same system depending on the boiling sugar massecuite time, *Ukrainian Journal of Food Science*, 3(2), pp. 322–341.
- Pogoriliy T. (2016a), Non-stationary sucrose diffusion mass flow calculation for sucrose solution cells from the «larger sugar crystal–larger sugar crystal sucrose solution–less sugar crystal sucrose solution–smaller sugar crystal–massecuite» system cells depending on the boiling sugar massecuite time, *Ukrainian Food Journal*, 5(2), pp. 350–367.
- Romero-Bustamante J.A., Velazquez-Camilo O., Garcia-Hernandez A., Rivera V.M., Hernandez-Martinez E., (2022), Monitoring of cane sugar crystallization process by multiscale time-series analysis, *Chaos, Solitons & Fractals*, 156, p. 111848, <https://doi.org/10.1016/j.chaos.2022.111848>.
- Rózsa L., Rózsa J., Kilpinen S. (2016), Crystal growth and crystallization control tactics in industrial sugar crystallizers Part 1. Crystal growth, *International Sugar Journal*, 118, pp. 746–755.
- Sánchez-Sánchez K.B., Bolaños-Reynoso E., Urrea-García, G.R. (2017), Analysis of operating conditions for cane sugar batch crystallization based on MSZW coupled with mechanistic kinetic models, *Revista Mexicana de Ingeniería Química*, 16(3), pp. 1029–1052, Available at: <https://www.redalyc.org/pdf/620/62053304028.pdf>

- Shamim F., Hernández R., Paulen R., Engell S.A. (2016), A hierarchical coordination approach to the optimal operation of a sugar crystallization process, *Computer Aided Chemical Engineering*, 38, pp. 703–708, DOI; 10.1016/B978-0-444-63428-3.50122-3.
- Suarez L.A.P., Georgieva P., de Azevedo S.F. (2011), Model Predictive Control Strategies for Batch Sugar Crystallization Process, In (Ed.), *Advanced Model Predictive Control, IntechOpen*, Available at: <https://www.intechopen.com/chapters/16065>
- Tkachenko S.V., Sheiko T.V, Petrenko V.V., Anisimova O.M., Kuznietsova I.V., Khomichak L.M., Obodovych O.M. (2020), Influence of crystallizing agent on sugar quality, *Acta Scientiarum Polonorum-Technologia Alimentaria*, 19(4), pp. 457–465. <http://doi.org/10.17306/J.AFS.2020.0867>
- Vetter F.L., Strube J. (2022), Enabling total process digital twin in sugar refining through the integration of secondary crystallization influences, *Processes*, 10(373), <https://doi.org/10.3390/pr10020373>.
- Zhong X., Huang C., Chen L., Yang Q., Huang Y. (2022), Effect of ultrasound on the kinetics of anti-solvent crystallization of sucrose, *Ultrasonics Sonochemistry*, 82(105886), pp. 1–10, <https://doi.org/10.1016/j.ultsonch.2021.105886>