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THE THIRD KIND OF BOUNDARY CONDITION IN NUMERICAL FREEZING CALCULATIONS*

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Abstract—Freezing of foodstuffs frequently involves boundary conditions of the third kind, which have proved to be difficult to handle and subject to error, mainly because of the requirement to calculate surface temperatures implicitly. Experimental freezing studies on model and actual biological systems have been simulated by finite difference methods with a three time level scheme and boundary conditions of the third kind, for one dimensional heat transfer. For freezing times, from onset of cooling until the centre reaches $-10°C$, experiment and prediction agreed to within $\pm 14\%$ with 99% confidence, including experimental error estimated at $\pm 4\%$.

NOMENCLATURE

C,	volumetric specific heat capacity [J/m^3 °C];
D,	slab half thickness [m];
h,	surface heat-transfer coefficient [W/m^2 °C];
H,	enthalpy [J/m^3];
k,	thermal conductivity [W/m °C];
M,	dimensionless number, $\dfrac{C(\Delta x)^2}{k\Delta t}$;
N,	dimensionless number, $\dfrac{h\Delta x}{k_s}$;
n,	number of space increments in the half slab;
No,	number of sheets of cardboard in equation (8);
Q,	surface heat flux [W/m^2];
R,	dimensionless number, $\dfrac{k_b\Delta x_c}{k_c\Delta x_b}$;
s,	thickness of one sheet of cardboard in equation (8) [m];
T,	temperature [°C];
t,	time [s];
Δt,	time step in finite difference scheme [s];
x,	displacement from the surface of the slab [m];
Δx,	space increment in finite difference scheme [m].

Superscripts

m,	defines time level in finite difference scheme;
$'$,	indicates space increment as defined by equation (13).

Subscripts

a,	ambient conditions;
b,	refers to one material in equation (7);
c,	refers to other material in equation (7);
ca,	property of the cardboard;
calc,	computation facilities required by the finite difference program;
cc,	refers to cardboard thermal contact in equation (8);
i,	nodal points in half slab;
in,	initial conditions;
p,	refers to thermal resistance of plates in equation (8);
pre,	quantities predicted by the difference scheme;
s,	surface of the half slab.

INTRODUCTION

PHASE change in solid materials is a very important phenomenon in a variety of industries. It is important in alloy solidification, preservation of foodstuffs, preservation of medical and biochemical materials, in freezing of soils and in frost damage to plants. Because of the economic importance of such processes a wide range of research into the effects of phase change, and into physical models for it has been conducted.

In spite of this extensive research a general analytical solution to the problem of a solid undergoing phase change has not been found. A variety of approaches to the problem have been tried, and these can be classified into two groups. The first group are the numerical finite difference and finite element solutions which rely on approximations to the governing partial differential equations over small time and space intervals. The second group comprise those methods where simplifying assumptions are made, leading to the solution of the partial differential equations to give either completely analytical and explicit formulae or ordinary integro-differential equations which can be evaluated numerically, to find the temperature distribution and phase change front position.

This paper is concerned with the first of these approaches, and looks at one-dimensional heat transfer in finite slabs. Two techniques have been used to take account of phase change when using finite differences. The first of these uses a heat balance over each Δx to determine the position of the phase change front at which a step change in enthalpy occurs [1, 2]. The second approach takes into account the fact that in

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*Copies of the Fortran IV computer program used in this work can be obtained from the authors.

solidifying foods and alloy mixtures phase change occurs over a range of temperature, and can be represented by a finite apparent heat capacity [3–5]. Thus the problem becomes one of solving the governing partial differential equation

$$C(T)\frac{\partial T}{\partial t} = k(T)\frac{\partial^2 T}{\partial x^2} \qquad (1)$$

with changing thermal properties subject to the appropriate boundary condition, of which four types are recognised.

First kind:

$$T_s = T_a \quad \text{for} \quad t > 0 \qquad (2)$$

Second kind:

$$Q = -k\left(\frac{\partial T}{\partial x}\right)_{x=0} \quad \text{for} \quad t > 0 \qquad (3)$$

Third kind:

$$h(T_a - T_s) = -k\left(\frac{\partial T}{\partial x}\right)_{x=0} \quad \text{for} \quad t > 0 \qquad (4)$$

Fourth kind:

$$T_s = f(t) \quad \text{for} \quad t > 0. \qquad (5)$$

The second kind of boundary condition, that of constant heat flux is of no practical importance [6], and the first kind of boundary condition can be treated as a special case of either the third kind where h is infinite, or of the fourth kind where $f(t) = T_a$. The third kind of boundary condition as well as taking account of convection, can be used wherever there is a resistance to heat transfer between the surface of the body and the bulk external medium. The surface heat-transfer coefficient h, in these cases is a measure of the resistance to heat transfer from the surface of the body to the external medium. For example, in a plate freezer T_a is the temperature of the refrigerant in the plates, and h represents the resistance to heat flow of the plates themselves, any packaging, and the resistance due to imperfect thermal contact between the plates and the product.

Bonacina *et al.* [4] used a scheme suggested by Lees [7] to set up an implicit finite difference scheme which involves three time levels:

$$\frac{-2(\Delta x)^2}{3\Delta t} k^m_{i+\frac{1}{2}} T^{m+1}_{i+1} + \left(C^m_i + \frac{2(\Delta x)^2}{3\Delta t}(k^m_{i+\frac{1}{2}} + k^m_{i-\frac{1}{2}})\right) T^{m+1}_i$$
$$- \frac{2(\Delta x)^2}{3\Delta t} k^m_{i-\frac{1}{2}} T^{m+1}_{i-1}$$
$$= \frac{2(\Delta x)^2}{3\Delta t}\left(k^m_{i+\frac{1}{2}}(T^m_{i+1} - T^m_i + T^{m-1}_{i+1} - T^{m-1}_i)\right.$$
$$\left. - k^m_{i-\frac{1}{2}}(T^m_i - T^m_{i-1} + T^{m-1}_i - T^{m-1}_{i-1})\right) + C^m_i T^{m-1}_i. \qquad (6)$$

This scheme is unconditionally stable and convergent. Bonacina *et al.* [4] used it with the first and fourth kind of boundary condition. The first kind of boundary condition was checked against an analytical solution due to Neumann [8], and Bonacina *et al.* found agreement to be within 3% [9].

To check the applicability of the finite difference scheme to the fourth kind of boundary condition it is necessary to set up experimental freezing tests. For this Bonacina *et al.* [9] used as their freezing material "Karlsruhe test substance", a defined 23% methylcellulose gel ("Tylose"). This was formed into a slab which was frozen from both sides, with other edges insulated. They also froze several food materials [10]. Both surface and centre temperatures were measured, and the experimental surface temperature profile was used in the finite difference program, and the calculated and experimental centre temperatures compared.

In design situations it is rare to know a surface temperature/time profile. However often the bulk external medium temperature (ambient temperature T_a) is known, for example, air temperature in a blast freezer; and there are well known means of arriving at a surface heat-transfer coefficient h, which takes account of all resistances for heat leaving the surface of the freezing material and going into the cooling medium. This is the case of Newton's law of cooling, the third kind of boundary condition. In such cases the surface requires special consideration.

Bonacina *et al.* [11] suggest that when $i = 0$, then $T^m_{-1} = T_a$ and a pseudo-thermal conductivity is defined, $k^m_{-\frac{1}{2}} = h\Delta x$, so that a three level scheme can be used. The authors found the "Lees" scheme to be superior to others such as the work of Dusinberre [12], due to smaller truncation errors, and to better representation of thermal properties. A scheme of comparable accuracy to the "Lees" scheme is given by McAdams [13] for the interface between two dissimilar materials: the formula was applied at nodal points in the slab using thermal properties $0.5\Delta x$ on either side of the nodal point. At the surface of the slab the boundary condition is equation (4) which can be represented in the explicit finite difference scheme by the following equation:

$$T^{m+1}_0 = \frac{4N}{(M_c + RM_b)} T_a + \left(1 - \frac{4N+2}{(M_c + RM_b)}\right) T^m_0$$
$$+ \frac{2}{(M_c + RM_b)} T^m_1. \qquad (7)$$

The authors found this scheme to be of similar accuracy to the two level version of the "Lees" scheme but less versatile because it is explicit. The three level "Lees" scheme is preferred because it has several major advantages over a two level scheme [4].

Bonacina *et al.* [11] did not determine the surface heat-transfer coefficient independently, but rather chose a value that gave best fit of predicted temperature profiles to their experimental freezing curves in each case. The present authors have developed methods for measuring h, and used these to determine surface heat-transfer coefficients independent of the freezing process [14]. With different numbers of sheets of cardboard interposed between the slab and the external medium, cooling curves of Tylose samples were used to find h, and a regression line relating thermal resistance to number of cardboard sheets was found. From this line values of h can be obtained for use with the third kind of boundary condition. This boundary condition is especially important in freezer design, as in this case the surface temperature/time profile is unknown, but h and T_a can be found. Thus an estimate of freezing time can be made without experimental investigation.

The definition of freezing time used here is in accordance with the International Institute of Refrigeration in its recommendations for the freezing of foodstuffs [15]. Freezing is considered complete when the centre temperature reaches $-10°C$, and the freezing time is the time from the introduction of the material to the freezing environment where it is frozen from two sides, until the centre reaches $-10°C$. This definition is consequent on the release of latent heat over a range of temperature and the absence of a sharp phase change front.

EXPERIMENTAL

A plate freezer was used for all the work to ensure one dimensional heat transfer. Blocks of Karlsruhe test substance of different size were made, each surrounded by at least 0.08 m of foam insulation except on the two opposite faces from which it is frozen. The blocks were held in constant temperature rooms prior to freezing, and 10 copper/constantan thermocouples were embedded in various places in each block and connected to a 12 point recording potentiometer operating on a 30 s cycle. To vary the surface heat-transfer coefficient sheets of cardboard, waxed on both sides were placed between the plates and slab. Varying the number of sheets of cardboard altered the resistance to heat transfer from the surface of the slab, thus changing the surface heat-transfer coefficient.

$$\frac{1}{h}=\frac{1}{h_p}+No\left(\frac{1}{h_{cc}}\right)+No\left(\frac{s}{k_{ca}}\right). \tag{8}$$

The plates were lowered by an hydraulic ram to ensure good thermal contact. A 29% calcium chloride brine was circulated through the plates at approximately 1 kg/s. This was pumped from a jacketed tank, the temperature of which was controlled to ±0.5°C. The maximum temperature rise in the circulated brine under these conditions was 0.3°C.

Blocks of minced lean beef (74% water) and mashed potato (82% water) were also used to check applicability to real food materials. Surface heat-transfer coefficients were measured by methods given elsewhere [14].

RESULTS AND DISCUSSION

(A) *Thermal property data*

A total of 43 freezing experiments were conducted with Karlsruhe test substance under varying conditions of ambient temperature T_a, initial temperature T_{in}, surface heat-transfer coefficient h, and slab half thickness D. T_a was varied from $-20°C$ to $-40°C$, T_{in} from 3 to 35°C, h from 13 to 450 W/m^2 °C, and D from 0.012 to 0.050 m. Thermal conductivity data was obtained from Comini [16].

Two sets of enthalpy/temperature data are available for Karlsruhe test substance. In Fig. 1 Curve A is due to Riedel [17] and corresponds to the case where initially a true type of plateau is found in the freezing curve of a sample of Karlsruhe test substance, and Curve B from Comini's data [16] does not give this plateau although the overall enthalpy change is the same. The difference could be due to different methylcellulose types as well as experimental error. The methyl cellulose used by the present authors was found to be better represented by Curve A. Because of difficulties in incorporating this curve into a finite difference program Curve C, a close approximation was used. No appreciable changes in density were found and like Comini [16] a constant value was used.

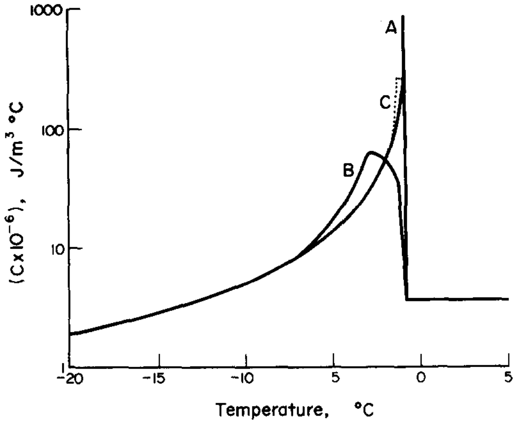

FIG. 1. Enthalpy/temperature data for Karlsruhe test substance plotted as specific heat (J/m^3 °C) vs temperature (°C). Curve A is from Riedel [17] and Curve B from Comini [16]. Curve C is an approximation to Curve A for use in finite difference calculations.

(B) *Fourth kind of boundary condition*

The experimentally determined surface temperature was used in each case with boundary conditions of the fourth kind. To obtain sufficient accuracy nine nodal points in the half slab were used with the time step Δt chosen so that there were approximately 1500 time steps for each experiment. Each calculation was stopped at the experimentally determined time for the centre of the slab to reach $-10°C$. Agreement between the computed and experimental surface temperatures is in all cases within ±1°C up until the time the centre reached $-3.0°C$. Beyond this, and until the calculation was stopped, the agreement was better than ±2°C in all except four cases where larger differences up to 3.5°C were found.

The probable cause of the larger errors is "jumping" of the latent heat peak. Where changes in temperature with respect to time and displacement are large, it is difficult to conserve changes in enthalpy due to "jumping" of the latent heat peak, and to stable oscillations that may develop in the difference scheme. Two ways of overcoming this problem have been suggested by Comini [16]. When updating the differences scheme the following can be used instead of $T_i^{m-1} \leftarrow T_i^m$

$$T_i^{m-1} \leftarrow (T_i^{m-1}+T_i^m+T_i^{m+1})/3 \tag{9}$$

and heat capacities can be found from enthalpy differences as follows:

$$C_i = [H(T_{i+\frac{1}{2}}) - H(T_{i-\frac{1}{2}})]/(T_{i+\frac{1}{2}} - T_{i-\frac{1}{2}}) \qquad (10)$$

where

$$T_{i+\frac{1}{2}} = (T_{i+1} + T_i)/2; \quad T_{i-\frac{1}{2}} = (T_i + T_{i-1})/2. \qquad (11)$$

These may not be completely satisfactory in conserving enthalpy changes and errors could occur in some cases. Reducing Δt and Δx is one way of reducing errors, but to avoid jumping completely when using Curve C, the necessary reduction is uneconomic in computation time. Hence an alternative approach was necessary.

A heat balance was used to find where jumping had occurred. In these cases Curve B, a much flatter shaped curve, was found to give much smaller errors in the heat balance. Comini *et al.* [10] showed that errors introduced by changes in the shape of the latent heat peak are small (up to 3%), and the present authors found them to be considerably smaller than the errors in the heat balance (up to 12%). Thus in these cases Curve B was considered to give more accurate results.

(C) *Third kind of boundary condition*

Bonacina *et al.* [11] suggest representing the third kind of boundary condition in the Lees scheme by using $k^m_{-\frac{1}{2}} = h\Delta x$ and $T^m_{-1} = T_a$. This scheme was used with eight Δx's in the half slab and led to the error frequency diagram shown as Curve A in Figure II when compared with the experimental freezing times. Details of the experimental conditions and experimental freezing times are given elsewhere [18]. The average error was +7.3%.

The finite difference boundary condition is derived from a heat balance over the surface space increment which extends in $0.5\Delta x$ from the surface. This is shown in Fig. 3.

$$\frac{k_{+\frac{1}{2}}}{\Delta x}(T_1 - T_0) = h(T_0 - T_a) + C(T_0)\frac{\partial T_0}{\partial t}. \qquad (12)$$

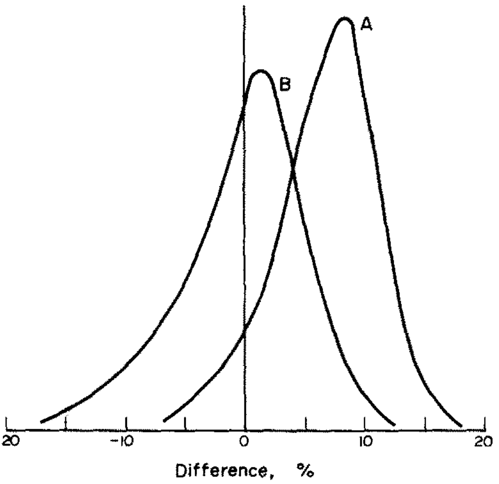

FIG. 2. Error frequency diagram for set of 43 freezing experiments with Karlsruhe test substance. Calculated freezing times are compared to experimentally determined values and differences expressed as a percentage. (A) difference scheme as suggested by Bonacina *et al.* [11]. (B) authors' modification.

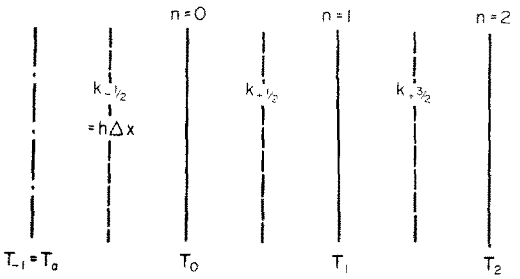

FIG. 3. Diagrammatic representation of the finite difference scheme at the surface of the half slab.

In reality the surface nodal point has the heat capacity of only $0.5\Delta x$'s associated with it. This means that the boundary equation is such that too much heat is removed from the slab by a factor of $(n+\frac{1}{2})/n$. When $n = 8$ as in our case, the finite difference scheme will necessitate the removal of 6.7% too much heat in the overall balance to arrive at any new temperature distribution.

An obvious way to reduce the errors is to increase n, the number of space increments. The error is $1/2n$ and halves each time n is doubled, but each time this is done the computation time also doubles. Table 1 shows the effect of increasing n. As can be seen the increase in computation time makes this an uneconomic approach.

Table 1. Calculated freezing times for $D = 0.0125$ m; $h = 51.9$ W/m² °C; $T_a = -40.0$°C; $T_{in} = 30.0$°C with varying numbers of space increments. (Experimental freezing time = 2300 ± 150 s.) Calculated freezing times t_{pre}, and computation time t_{calc}, are both in seconds

n	t_{pre}	t_{calc}	$\frac{100}{2n}$ %
8	2550	38	6.7
15	2490	70	3.3
30	2460	136	1.7

To avoid this problem a useful approach is to define the space increment as follows:

$$\Delta x' = D/(n+\tfrac{1}{2}) \qquad (13)$$

and then to calculate the freezing time of a slab of half thickness $n\Delta x'$. This slab is $\frac{1}{2}\Delta x'$ smaller, but the finite difference scheme will now remove the correct total amount of enthalpy for the original slab. The centre temperature/time profile is valid but the calculated surface temperature is not because of the difference in position of the nodal temperature T^m_0. In many cases the difference in surface temperature calculated for slab half thicknesses of $n\Delta x$ and $n\Delta x'$ is less than 0.5°C, with the surface temperature of the smaller slab always higher. This latter profile agreed within ±1°C of the experimental surface temperature initially but towards the end of some runs the calculated surface temperature was up to 3°C higher than the experimental one. Extrapolation of the calculated temperature profiles to a point $0.5\Delta x'$ beyond T_0^m in some cases gave a better estimate of the actual surface temperature, but as this was not always the case no advantages could be seen in this procedure. While the calculated surface temperatures tracked the experimental ones, the calculated centre temperature followed the measured centre temperature within ±1°C. Once the calculated surface temperature started to deviate from the experimental results so did the calculated centre temperature within the same levels of tolerance as the surface temperature.

The modified approach led to Curve B on Fig. 2. The average percentage error is +0.7% and the standard deviation 4.7%. The error in the mean could arise from two possible causes. One possibility is errors in thermal properties, an error which would be consistent over all runs. The other is a changing surface heat-transfer coefficient. The slab is held between plates with sheets of cardboard inserted between the plates and the slab to give thermal resistance. When the slab freezes there is slight expansion, and the slab is pressed more tightly against the plates. This could lead to decreasing thermal contact resistances, and hence to an increasing overall surface heat-transfer coefficient. Thus the final experimental surface temperature would be lower than the calculated one (as was found), and the freezing process would take less time than predicted. A 1% error in freezing time corresponds to a change in contact resistance of about 10% which is quite feasible. Either of these reasons or both could explain the offset mean, but it was not possible to differentiate.

(D) *Errors in the finite difference scheme*

The deviation around the mean is due to a number of factors. Firstly there is experimental error which was approximately ±4% for each freezing experiment. This includes ±0.5°C error in temperature measurement and control, 0.3 mm error in slab thickness and an estimated 2% error in surface heat-transfer coefficient. This does not explain the variations that were found of ±12%. Other possible sources of error are (a) rounding errors (b) truncation errors in the finite difference scheme (c) truncation errors in the boundary equation (d) errors due to piece-wise approximation of continuous functions $k(T)$ and $C(T)$ which must be assumed constant over time Δt and distance Δx.

(a) Rounding errors on a modern computer are small and can be neglected.

(b) Truncation errors in the difference scheme are

$$-\frac{(\Delta x)^2}{12}k(T)\frac{\partial^4 T}{\partial x^4}. \tag{14}$$

$$-\frac{(\Delta t)^2}{6}C(T)\frac{\partial^3 T}{\partial t^3}. \tag{15}$$

The truncation error in the space term should be small despite a threefold increase in $k(T)$. If the time step is halved twice as many are needed per calculation so that the sum of the truncation errors with respect to time is still proportional to Δt. Also $C(T)$ increases by a factor of more than 50 in the phase change region, and errors will be largest here. This will affect the rate at which any calculated temperature moves off the freezing plateau, and is the more likely source of error.

(c) The third kind of boundary condition, equation (4) is represented by equation (12) which contains a difference approximation with the following truncation error:

$$-\frac{(\frac{1}{2}\Delta x)^2}{6}k_{+\frac{1}{2}}(T)\frac{\partial^3 T}{\partial x^3} \tag{16}$$

$\partial T/\partial x$ is greatest near the surface and other derivatives will also be large in this region so that these truncation errors may add significantly to the others.

(d) It is assumed that C_i^m represents $C(T)$ over the range $C_i^{m-\frac{1}{2}}$ to $C_i^{m+\frac{1}{2}}$ which is only true if it changes linearly. It is assumed that $k(T)$ can be represented by

$$k(T_{i+\frac{1}{2}}) \simeq k[(T_i + T_{i+1})/2] \tag{17}$$

$$k(T_{i-\frac{1}{2}}) \simeq k[(T_i + T_{i-1})/2]. \tag{18}$$

These add further small errors. Although each error in itself is small it is repeated $(n+1)$ times for each time step, and for a large number of time steps. This could lead to errors of the order indicated above, but these are difficult to estimate.

The overall error range (±12%) minus the experimental error (±4%) indicates the error of the finite difference scheme. This is approximately ±8% and appears to represent a practical limit to the accuracy of finite difference calculations with the third kind of boundary condition for predicting food freezing times. Decreasing Δt and Δx improves accuracy but at the expense of large amounts of computation time. The smaller errors associated with the fourth kind of boundary condition are due to there being no extra truncation errors from the boundary equation. Temperatures within the slab follow the surface temperature so that if the surface temperature is incorrectly calculated all other calculated temperatures will also be wrong. Use of the fourth kind of boundary condition avoids errors in the surface temperature, and hence decreases errors in the calculated centre temperature, whereas when the third kind of boundary condition is applied errors in the surface temperature are uncorrected, and hence are reflected in all other calculated temperatures.

(E) *Application to foodstuffs*

When the scheme was applied to the freezing of mashed potato and minced lean beef the range of errors over 12 freezing experiments was −7.7 to +11.0% with a mean of +1.8% and a standard deviation of 5.4%. This slightly higher standard deviation could be due to heterogeneity in food materials and errors in thermal property approximations.

CONCLUSION

Finite difference calculations on freezing systems have been applied using the third kind of boundary condition: these are less accurate, but more related to practice than calculations where the fourth kind of boundary condition is applied. However, the accuracy should be adequate for many applications.

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LA CONDITION A LA LIMITE DE TROISIEME ESPECE DANS LES CALCULS NUMERIQUE DU GEL

Résumé—La congélation des aliments est fréquemment décrite à partir des conditions de troisième espèce qui sont difficiles à traiter et qui sont sujettes à des erreurs, principalement à cause de la nécessité de calculer implicitement les températures de surface. Des études expérimentales de congélation sur modèle et sur des systèmes biologiques réels ont été simulées par des méthodes aux différences finies avec un schéma à trois niveaux de temps et des conditions de troisième espèce, pour le transfert thermique à une seule dimension. Pour des temps de congélation allant depuis le début du refroidissement jusqu'à ce que le centre atteigne −10°C, l'expérience et le calcul s'accordent à ±14% près avec une confiance à 99%, en incluant l'erreur expérimentale estimée à ±4%.

NUMERISCHE BERECHNUNG DES GEFRIERENS MIT DER RANDBEDINGUNG 3. ART

Zusammenfassung—Beim Gefrieren von Lebensmitteln trifft man häufig die Randbedingung 3. Art an; da hierbei die Oberflächentemperaturen implizit errechnet werden müssen, ist dieses Problem schwierig zu behandeln und gibt zu Fehlern Anlaß. Es wurde das Gefrieren von Modellsubstanzen und biologischem Material experimentell untersucht. Die numerische Berechnung für eindimensionalen Wärmestrom und für die Randbedingung 3. Art erfolgte mit Hilfe eines Differenzenverfahrens. Die Versuchswerte und die Rechenwerte für die Gefrierzeiten von Beginn der Abkühlung bis zu einer Kerntemperatur von −10°C stimmten zu 99% mit Abweichungen von ±14% überein, wobei der Meßfehler zu ±4% angesetzt wurde.

ГРАНИЧНОЕ УСЛОВИЕ ТРЕТЬЕГО РОДА ПРИ ЧИСЛЕННЫХ МЕТОДАХ ИССЛЕДОВАНИЯ ПРОЦЕССА ЗАМОРАЖИВАНИЯ

Аннотация — Задачи по замораживанию пищевых продуктов часто включают граничные условия третьего рода, рассмотрение которых представляет некоторые трудности и может приводить к погрешностям главным образом в силу того, что температуру поверхности приходится рассчитывать в неявном виде. Опыты по замораживанию модельных и реальных биологических систем моделировались с помощью конечно-разностных методов с использованием трехслойной схемы и граничных условий третьего рода для одномерного процесса теплопереноса. Для отрезка времени от начала процесса охлаждения до момента, когда температура центра образца достигает −10°C, экспериментальные и расчетные данные совпали с точностью ±14% при достоверности 99% и погрешности эксперимента ±4%.