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International Journal of Heat and Mass Transfer 48 (2005) 1205–1213
International Journal of HEAT and MASS TRANSFER
www.elsevier.com/locate/ijhmt

Food freezing with simultaneous surface dehydration: approximate prediction of freezing time

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Received 9 September 2003
Available online 8 December 2004

Abstract

Freezing of unpackaged foods induces mass transfer in the form of surface ice sublimation, which in turn modifies heat transfer conditions. At present there are no simplified methods for predicting freezing times when surface dehydration occurs. This paper uses a previously developed model for the simulation of simultaneous heat and mass transfer during food freezing and storage to generate a complete set of predicted freezing times when dehydration occurs. Based on these data a simplified analytical method for the prediction of freezing time during freezing of unpackaged frozen foods was developed. The method accounts for product characteristics (shape, size and composition) and operating conditions (initial and refrigerant temperature, heat transfer coefficient, relative humidity). The prediction equation is very simple and results of its use—simulating usual freezing conditions for different products—shows very good accuracy when tested against the previously cited numerical model and all the available experimental data.

1. Introduction

The freezing and later storage of frozen foods are important operations in food preservation, involving millions of Tons of processed food per year alongside the world [1]. An important proportion of these foods are frozen unpackaged, like most of meat and fish hamburgers and other fresh or prepared meats, most fruits and vegetables, shrimp, etc.

The main transfer involved in freezing is heat transfer from the product to the cooling medium (in most cases air or a cryogenic gas). Notwithstanding, unpackaged foods also lose water, initially by evaporation during the cooling stage—when surface temperature is still higher than the freezing point—but mainly by sublimation of ice from the frozen surface layer as freezing goes on. Both phenomena are originated in differences in water activity between the surface and the cooling medium, that are maximal at the beginning of the process (highest surface temperature) and decrease along freezing, when surface temperature lowers. In this way a dehydrated layer is developed, whose thickness increases as time elapses [2]. If not properly managed, surface drying may produce important quality and economic losses, so its prediction and control are of the highest practical interest [3–5].

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0017-9310/$ - see front matter © 2004 Published by Elsevier Ltd.
doi:10.1016/j.ijheatmasstransfer.2004.09.030

Nomenclature

a	parameter in Eq. (12)
a_v	surface area/volume, 1/m
b	parameter in Eq. (12)
Bi	Biot number ($=hL/k_0$)
c	parameter in Eq. (12)
C	molar concentration of water, mole/m^3
Cp	specific heat, J/(kg °C)
d	parameter in Eq. (12)
D	effective diffusion coefficient, m^2/s
e	relative error, %
f_{ads}	adsorbed ice fraction, (kg of ice)/(kg of dried solid)
GI	geometric index, its value is 0 for slabs, 1 for cylinders and 2 for spheres
h	heat transfer coefficient, W/(m^2 °C)
k	thermal conductivity, W/(m °C)
k_m	mass transfer coefficient (referred to vapour water driving force), m/s
k'_m	mass transfer coefficient (referred to liquid water driving force), m/s
L	half-thickness or radius, m
L_s	sublimation heat of ice, J/kg
m_s	sublimated mass by unit volume, kg/m^3
P	pressure, Pa
P_{sat}	vapor saturation pressure, Pa
R_g	universal gas constant, 8.31 J/(mol K)
RH	relative humidity, %
t	time, s (or min)
T	temperature, °C
T_K	temperature, K
v	velocity, m/s
Vol	volume, m^3
w	ice content (kg of ice)/(kg of food)
WL	weight loss (kg of water)/(kg of food)
x	spatial coordinate, m
x_1	moving sublimation front position, m
Y_0	initial water content, (kg of water)/(kg of food)

Greek symbols

α_0	thermal diffusivity of unfrozen food, m^2/s
ρ	density, kg/m^3
ε	porosity
τ	tortuosity

Subscripts

a	air
c	thermal center
d	dehydrated
eq	equilibrium
f	freezing
f,0	freezing without dehydration
f,deh	freezing with dehydration
if	initial freezing
L	surface
0	initial
sat	saturation
v	water vapour
w	liquid water

The microscopic balance that characterizes heat transfer during food freezing and storage—even without considering any type of surface mass transfer—has no analytical solution, mainly due to the nonlinear nature of the variation of food thermophysical properties with temperature [6]. Notwithstanding, many numerical schemes have been developed to model and simulate the system. These models may accurately predict temperature profiles as well as freezing times under any operating condition of the refrigeration system. Besides, several accurate approximate methods, resulting in quite simple equations, have been developed to predict freezing and thawing times of foods without surface dehydration. Delgado and Sun [5], Cleland [7], Hung [8] and Becker and Fricke [9,10] present complete reviews on these subjects.

The same type of prediction methods—numerical or approximate—have not been developed for foods with surface water evaporation and sublimation. Here again, the coupled microscopic balances that characterize heat and mass transfer during food freezing and storage have no analytical solution [4]. Few attempts have been made to develop physical models that take into account ice sublimation and the existence of a surface layer of dehydrated cells (in the case of meats and vegetables). Sukhwal and Aguirre-Puente [11] outlined the energy and mass balances that describe these phenomena, without reaching a quantitative (approximate or numerical) solution. Tocci and Mascheroni [12] developed a simplified model of the system, considering the dehydrated surface layer as an additional resistance to heat and mass transfer, whose value varied with that of the layer thickness. In a recent work, Campañone et al. [13] have presented a mathematical model that solves the differential equations arising from the statement of the coupled microscopic energy and mass balances during freezing and frozen storage. The model pays special attention to the generation and growth of a porous, dehydrated surface layer and to the specific characteristics of the heat and mass transfer phenomena in it. The implemented numerical solution accurately predicts freezing times and weight losses during freezing and frozen storage, growth of the dried porous layer and the behavior of products exposed to fluctuating operating conditions.

One of the main interests of design engineers and equipment users is to count on simple and accurate prediction equations for the simulation of the process they are dealing with, mainly for the calculation of process times as a function of food characteristics and operating conditions. In this respect—as previously stated—there is ample literature on prediction methods for heating, cooling, freezing and thawing without mass transfer [5,7–10,14] and some papers on the application of these methods to food dehydration [15–17]. To the best of our knowledge, there are no similar methods to predict freezing times of unpackaged foods and the related weight losses during freezing and storage.

Based on the above-mentioned needs and in the existence of an accurate numerical model for the simulation of heat and mass transfer during food freezing, the objectives of this work were

- to generate a whole set of predicted thermal histories and water content profiles for freezing of unpackaged high water-content foods, covering most product sizes and working conditions found in industrial freezing. As a consequence of these thermal histories, freezing time for each working condition could be predicted;
- to develop simple and accurate equations for freezing time prediction as a function of food characteristics and operating conditions, for the three simple regular shapes: plane plate, infinite cylinder, sphere;
- to compare freezing times predicted by these equations with those calculated with standard simplified methods;
- to validate predicted results against available experimental data, own and from literature.

In a related paper, weight loss aspects during freezing and storage are dealt with [18].

2. Theory

During the freezing and frozen storage of unwrapped foods, they lose weight due to the environmental interaction. When frozen water sublimates a porous dehydrated layer is formed on the food surface, which alters the food physical and sensory characteristics.

During the freezing, two state phase changes take place simultaneously: The free liquid water is frozen and the superficial water sublimates.

From a physical point of view, food can be considered as a combination of a solid matrix, an aqueous phase and a gaseous phase (air and water vapour). For the freezing process, the food can be divided in three zones: unfrozen, frozen and dehydrated.

A complete mathematical model has to solve the heat transfer (freezing) and the mass transfer (weight loss) simultaneously. As the industrial freezing process begins with the food at temperatures higher than T_{if} the model must consider also the weight loss during the initial refrigeration step.

In a previous work [13], the analysis was performed for regular unidimensional geometries (plane plate, infinite cylinder, sphere). A general formulation could be developed which accounts for each of these three geometries by means of the geometric index GI. The main characteristics of the model are as follows (the analysis is divided into two stages):

- *Stage 1*: For temperatures higher than the initial freezing point T_{if}, the following balances are valid for all the food ($0 \leqslant x < L$), where x is the axial or radial coordinate:

$$\rho Cp \frac{\partial T}{\partial t} = \frac{\partial k}{\partial x}\frac{\partial T}{\partial x} + k\frac{\partial^2 T}{\partial x^2} + \mathrm{GI}\frac{k}{x}\frac{\partial T}{\partial x} \tag{1}$$

$$\frac{\partial C_{\mathrm{w}}}{\partial t} = \frac{\partial D_{\mathrm{w}}}{\partial x}\frac{\partial C_{\mathrm{w}}}{\partial x} + D_{\mathrm{w}}\frac{\partial^2 C_{\mathrm{w}}}{\partial x^2} + \mathrm{GI}\frac{D_{\mathrm{w}}}{x}\frac{\partial C_{\mathrm{w}}}{\partial x} \tag{2}$$

with symmetry at the body centre and the following surface boundary conditions:

$$x = L, \quad -k\frac{\partial T}{\partial x} = h(T - T_{\mathrm{a}}) + L_{\mathrm{s}}k'_{\mathrm{m}}(C_{\mathrm{w}} - C_{\mathrm{eq}}) \tag{3}$$

$$x = L, \quad -D_{\mathrm{w}}\frac{\partial C_{\mathrm{w}}}{\partial x} = k'_{\mathrm{m}}(C_{\mathrm{w}} - C_{\mathrm{eq}}) \tag{4}$$

- *Stage 2*: When surface temperature T_{L} reaches T_{if}, freezing and ice sublimation begin and different zones need to be considered:
  - For the unfrozen and frozen zones ($0 \leqslant x < x_1$), Eq. (1) was applied to calculate the temperature profile, being x_1 the position of the sublimation front. This equation enables the evaluation of the freezing stage by means of the use of physical properties which are variable with temperature (the model uses apparent heat capacity that involves both sensible specific heat and the enthalpy of ice solidification).
  - For the dehydrated zone ($x_1 < x \leqslant L$) Eq. (5) was applied to calculate temperatures:

$$\rho_{\mathrm{d}} Cp_{\mathrm{d}} \frac{\partial T}{\partial t} = \frac{\partial k_{\mathrm{d}}}{\partial x}\frac{\partial T}{\partial x} + k_{\mathrm{d}}\frac{\partial^2 T}{\partial x^2} + \mathrm{GI}\frac{k_{\mathrm{d}}}{x}\frac{\partial T}{\partial x} \tag{5}$$

In this zone the model uses the physical properties corresponding to the dehydrated food.

For the energy balance, besides symmetry condition, the following boundary conditions can be stated:

$$x = x_1, \quad -k\frac{\partial T}{\partial x} = L_s m_s \frac{dx_1}{dt} - k_d \frac{\partial T}{\partial x} \tag{6}$$

$$x = L, \quad -k_d \frac{\partial T}{\partial x} = h(T - T_a) \tag{7}$$

Ice sublimation takes place within the frozen zone, leading to a dried layer with a porous structure. Thus, there is a water vapour transfer to the surrounding medium through this layer. It should be stressed, that a fraction of the ice, f_{ads}, remains adsorbed to the food matrix, its value depending on the local vapour–ice equilibrium conditions [19].

Vapour flow can be considered mainly as a diffusive flow passing throughout the dried layer, being the driving force the difference in water vapour pressure between the sublimation front and the environment. Fick's law was considered valid.

The microscopic mass balance applied to this zone ($x_1 < x \leqslant L$) was the following:

$$\varepsilon\frac{\partial C_v}{\partial t} = \frac{\partial D_v}{\partial x}\frac{\partial C_v}{\partial x} + D_v\frac{\partial^2 C_v}{\partial x^2} + \mathrm{GI}\frac{D_v}{x}\frac{\partial C_v}{\partial x} \tag{8}$$

Boundary conditions for the mass balance were, besides symmetry condition:

$$x = x_1, \quad C_v = \frac{P_{sat}(T)}{R_g T_K} \tag{9}$$

$$x = L, \quad -D_v\frac{\partial C_v}{\partial x} = k_m(C_v - C_{v,a}) \tag{10}$$

To evaluate the position of the sublimation front (and the thickness of the dehydrated layer) the model established that, at $x = x_1$:

$$-m_s\frac{dx_1}{dt} = -D_v\frac{\partial C_v}{\partial x}, \quad \text{where } m_s = \rho(w - f_{ads}(1 - Y_0)) \tag{11}$$

Eqs. (1) and (2) or (1), (5), (8) and (11) led to a system of coupled nonlinear partial differential equations. The system was solved by an Implicit Finite-Differences Method (Crank-Nicolson centered method). In the dehydrated zone a variable grid was applied to overcome the deformation of the original fixed grid due to the shift of the sublimation front. Further details of the calculation procedure are presented in [4,13]. Both reports provide a complete list of the source of each equation or value used for the prediction of thermal properties, diffusion coefficients, porosity, tortuosity, ice adsorbed fraction, etc., needed by the numeric model. Heat and mass transfer coefficients at food surface have been taken from specific information from literature that accounts for the influence of food shape and size and air temperature on the values of those coefficients [20–22].

3. Simplified prediction methods

Freezing time t_f is defined as the time elapsed since the food—with initial uniform temperature T_0—begins to be refrigerated, up to the moment its thermal center reaches a prescribed final temperature T_c. In this paper $T_c = -18$ °C is assumed in all calculations, because it is the usual target value in industrial freezing of foodstuffs.

Most simplified methods for the prediction of food freezing times (t_f) are based on the original development by Plank in 1913. This author—although considering a very simplified situation with initial and final temperatures equal to freezing temperature, constant thermal properties and no freezing range—could describe adequately the influence of food size and shape and working conditions (T_a and the Biot Number Bi) on t_f. The most accurate methods take as a basis Plank's equation and correct it through modified coefficients and/or adding multiplying factors. In this way a very precise evaluation of the influence of food initial and final temperatures, food thermal properties and operating conditions on t_f is achieved [23–25].

As an example of these procedures, and also as a basis for the calculations to carry on here, the equation of Salvadori and Mascheroni [25] was chosen because it is the most simple of these approximate methods. It has the following shape:

$$t_f(\min) = a\alpha_0^{-1}(L)^2(1 + T_0)^b(1/Bi + c)(-1 - T_a)^d \tag{12}$$

Constants a, b, c and d have specific values for the three simple regular shapes (plane plate, infinite cylinder and sphere). Their values are given in Table 1.

As stated before, all these simplified methods do not consider the possibility of surface water evaporation and later ice sublimation during the freezing process. These phenomena have two opposite effects on the value of freezing time:

- a decrease in the total heat load to be extracted, due to the heat uptake by the water that evaporates or sublimates;
- a decrease in the heat transfer rate due to the building of a dehydrated surface layer that has much lower thermal conductivity than an undehydrated food.

Table 1
Parameters on Eq. (12) for the no-surface dehydration case, $t_{f,0}$

Parameter	Plane plate	Infinite cylinder	Sphere
a	1.473	0.762	0.545
b	0.096	0.037	0.073
c	0.184	0.179	0.167
d	−1.070	−1.032	−1.078

The relative weight of these factors on the value of t_f depends on food shape and size and working conditions. But—anyhow—the above-cited methods will not be accurate for prediction under surface dehydrating conditions.

To advance in the development of the approximate methods is essential to count with a comprehensive set of values for $t_{f,deh}$ and WL covering a wide range of food sizes and shapes and freezing conditions. A thorough search in literature only results in very scarce and spare experimental data [4,5]. In this work we used the results of the previously cited numerical model [13] that considers simultaneous heat and mass transfer during freezing to generate the needed data-sets. An exhaustive parametric analysis of the system had already been done [4], taking advantage of the ability of the numerical software to predict temperature and concentration profiles inside the food and water loss as a function of time.

So, the software was used to obtain a great quantity of numerical values of $t_{f,deh}$ and WL under different operating conditions (v_a, RH and T_a) and food characteristics (shape, L, T_0 and Y_0). A total number of 918 numerical runs were calculated (432 for plane plate and 243 for cylinder and sphere, respectively). The range of operating conditions studied is presented in Table 2. In all the runs of the numeric model thermal properties for minced meat, representative of high water-content foods, were used [4,13].

It was verified that RH had statistically no influence on freezing time. So, as the parameters that influence on $t_{f,deh}$ were the same as for the no-dehydration case and the characteristics of food and heat transfer mechanism were basically similar, of all the tested equations, Eq. (12) was again the best to account for the dehydration case. The new parameters for this version of the prediction method were obtained by means of a software for nonlinear regressions [26] and are presented in Table 3.

These values are of the same order than those given in Table 1 for the no-dehydration case. This shows the high analogy between the physical mechanisms involved in both cases of heat transfer, which implies equivalent weights of the different operating variables on freezing time.

Table 2
Range of operating conditions of the numerical runs

Parameter	Plane plate	Infinite cylinder	Sphere
L	0.01–0.04	0.02–0.04	0.02–0.04
T_0	0–10	0–10	0–10
v_a	0.1–5	1–5	1–5
T_a	−20 to −40	−20 to −40	−20 to −40

Table 3
Parameters and statistics for Eq. (12) for the surface dehydration case

Parameter	Plane plate	Infinite cylinder	Sphere
a	3.556	1.020	0.552
b	0.037	0.032	0.033
c	0.245	0.319	0.259
d	−1.298	−1.213	−1.180
R^2	0.999	0.999	0.999
e (%)			
Average	0.23	0.49	−0.62
Upper limit	5.52	5.53	4.45
Lower limit	−4.39	−5.12	−6.36

4. Results and discussion

4.1. Validation against numerical data

Fig. 1(a)–(c) shows the predicted freezing times (by means Eq. (12)), vs. the numerical values for the three regular geometries studied. To evaluate the accuracy of the prediction equation the average percent error of each value was calculated. It is defined as

$$e_{t_f}\ (\%) = 100\frac{\text{Predicted } t_f - \text{Numerical } t_f}{\text{Numerical } t_f} \tag{13}$$

The upper and lower limits and the average value of the relative errors are also shown in Table 3. The evaluation of data from Table 3 and Fig. 1 shows the very good agreement between predicted and numerical values.

4.2. Relationships between t_f for the different shapes

In the measurement and prediction of cooling times (no phase change) and freezing times without dehydration it has been proven that constant relationships hold among process times for the different shapes, when equal operating conditions and food characteristics are considered [7,23].

These relationships are inversely proportional to the surface area/volume ratio (a_v) for each shape and also depend, to a very minor extent, on the Biot number. Table 4 presents a brief report on these data.

For the cases under study in this work, the Bi range tested was between 0.715 and 2.519, typical of food freezing operations. Within this range, as also shown in Table 4, there was little variation for the relationships $t_{f,deh\ shape}/t_{f,deh\ sphere}$, with average values of 4.3136 for plane plate and 1.7584 for infinite cylinder. These values have important differences—they are much higher in the case of plate—from those of the no-dehydration case. We consider that these differences are due to the higher thickness of the dehydrated surface layers for plates respect to that of cylinders and spheres, as originated in

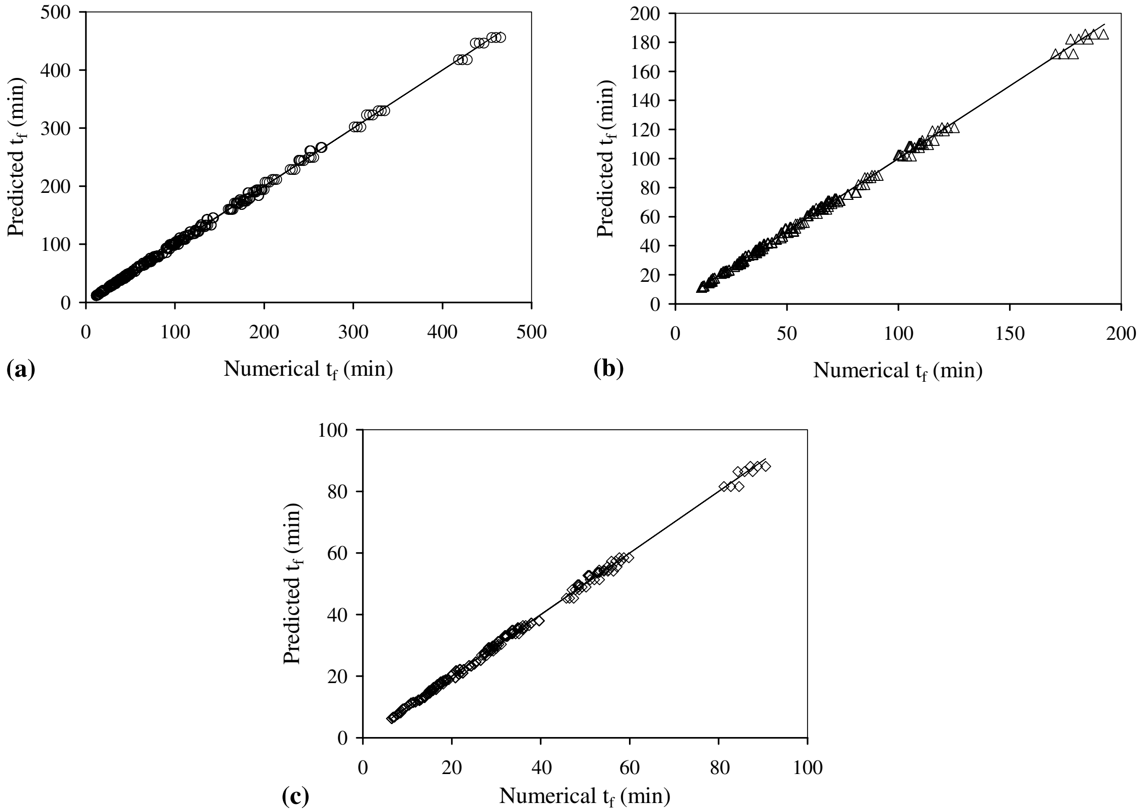

Fig. 1. (a) Predicted (○) vs. numerical t_f for plane plates. (b) Predicted (△) vs. numerical t_f for infinite cylinders. (c) Predicted (◇) vs. numerical t_f for spheres.

Table 4
Relationship between process times for different shapes

Shape		Plane plate	Infinite cylinder	Sphere
a_v (m^{-1})		$1/L$[a]	$2/L$[a]	$3/L$[a]
$\frac{\text{Cooling time shape}}{\text{Cooling time sphere}}$	$Bi \to 0$	3	1.5000	1
	$Bi \to \infty$	4	1.7064	1
$\frac{\text{Freezing time } t_{f,0} \text{ shape}}{\text{Freezing time } t_{f,0} \text{ sphere}}$		3	1.5000	1
$\frac{\text{Freezing time } t_{f,deh} \text{ shape}}{\text{Freezing time } t_{f,deh} \text{ sphere}}$	Average	4.3136	1.7584	1
	Lower *Bi*	4.5553	1.6926	1
	Higher *Bi*	4.0933	1.8301	1

[a] L = characteristic length: half thickness for plate and radius for cylinder and sphere.

the differences in a_v between shapes. This factor is further discussed in later paragraphs.

4.3. Comparison against predicted t_f without surface dehydration

It is important to quantify the calculation error that is being committed each time that literature formulas for predicting t_f under no-dehydrating conditions are used for freezing of unpackaged foods. For performing the comparisons, Eq. (12) was used in its original shape (parameters given in Table 1) to predict freezing times without weight loss, named $t_{f,0}$. In the same sense Eq. (12), with parameters given in Table 3, was used for predictions under surface dehydrating conditions. These freezing times are termed $t_{f,deh}$.

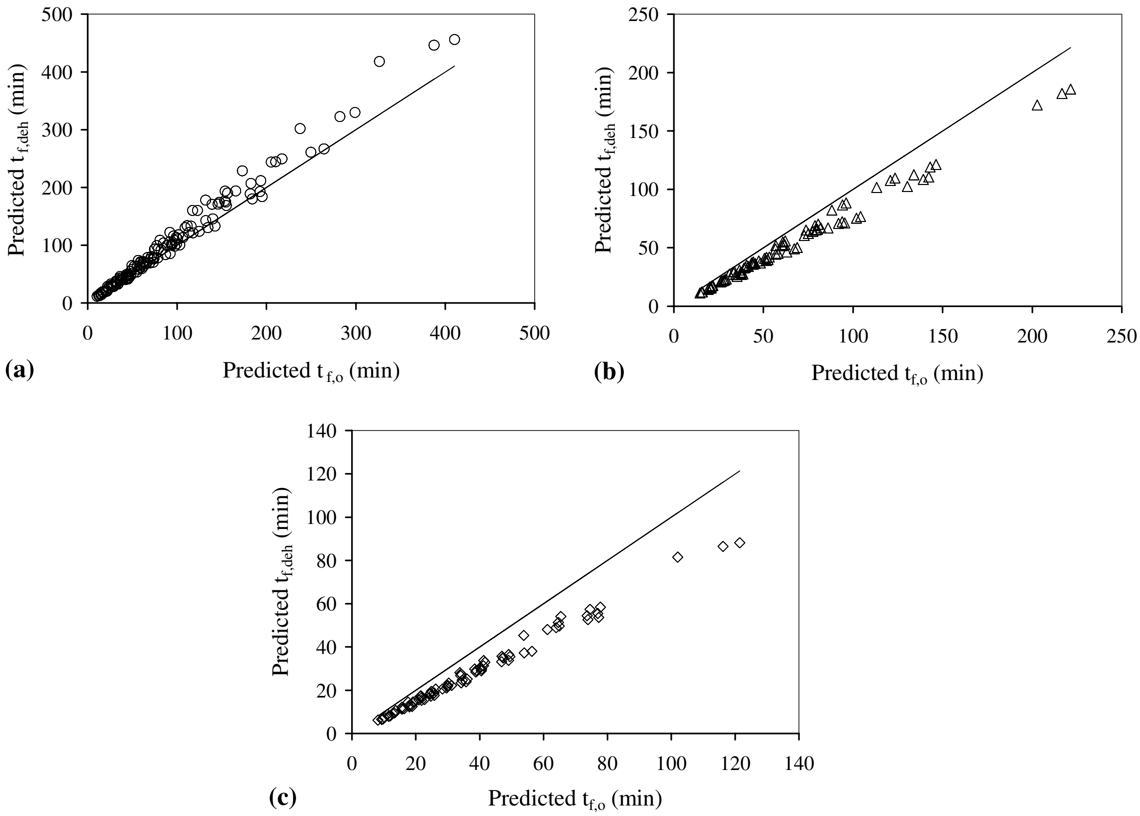

Fig. 2. (a) Predicted $t_{f,deh}$ (○) vs. $t_{f,0}$ for plane plates. (b) Predicted $t_{f,deh}$ (△) vs. $t_{f,0}$ for infinite cylinders. (c) Predicted $t_{f,deh}$ (◇) vs. $t_{f,0}$ for spheres.

Fig. 2(a)–(c) presents the comparison of $t_{f,deh}$ with $t_{f,0}$ for the three shapes. As seen in the figures, the different shapes have contrasting behaviours. So, $t_{f,deh}$ for slabs are in average 1.120 times higher than the respective $t_{f,0}$, values ranging between 0.944 and 1.363 times. For cylinders and spheres $t_{f,deh}$ are lower, averaging 0.815 and 0.742 times, respectively, their $t_{f,0}$ values. The respective ranges are 0.719 to 0.932 for cylinders and 0.664 to 0.844 for spheres. These differences show that errors of up to 50% can be incurred when using unappropriate prediction formulas.

The trends in the relations $t_{f,deh}/t_{f,0}$ are in accordance with what was presented in previous paragraphs when relating $t_{f,deh}$ for the different shapes. The common origin of these features is—as studied by using the prediction software developed in [4]—that, for equal working conditions, the thickness of the dehydrated layer is much higher for plates than for spheres, being values for cylinders closer to those of spheres. This thicker dehydrated layer lowers heat transfer rate. This effect compensates and most times exceeds the increase of freezing rate due to surface ice sublimation. The result is a $t_{f,deh}$ higher than the respective $t_{f,0}$. Contrary sense, the resistance of the thinner dehydrated layer for spheres and cylinders is not sufficient to counter-balance the increase of freezing rate due to surface ice sublimation. This leads to $t_{f,deh}$ lower than $t_{f,0}$ for those shapes.

4.4. Validation against experimental data

Theoretical predictions must be validated against experimental data, when available. In the case of food freezing with simultaneous surface dehydration this type of information is very poor in quantity and quality [4,5] and most of the data lacks of precisions about experimental conditions or food properties. All the available literature data for freezing times of unpackaged foods for which product characteristics and operating conditions were clearly determined, and whose freezing parameters were within the range given in Table 2 were used for tests. In Table 5 the type, quantity and origin of the data used for validation are presented. As can be seen, it includes only four sets of data from [27] and two sets of data from our laboratory. The specific values for the thermal properties of each food were used, the

Table 5
Experimental data used for the validation of the proposed prediction method

Product	Shape	Number of data	Thermophysical properties [4,29,30]		Average error (%)
			k	α	
Tylose [27]	Slab	22	0.55	1.50×10^{-7}	5.47
Mashed potato [27]	Slab	8	0.53	1.45×10^{-7}	−1.39
Lean beef [27]	Slab	7	0.48	1.31×10^{-7}	4.83
Carp [27]	Slab	7	0.48	1.34×10^{-7}	8.64
Minced beef [28]	Sphere	48	$0.4373 + 3.5365 \times 10^{-4}T$	1.33×10^{-7}	−9.84
Minced beef [4]	Sphere	9	$0.0866 + 0.501\,Y_0 + 5.0521 \times 10^{-4}\,Y_0 T$	$k_0/(1053 \times 3474.86)$	−4.56

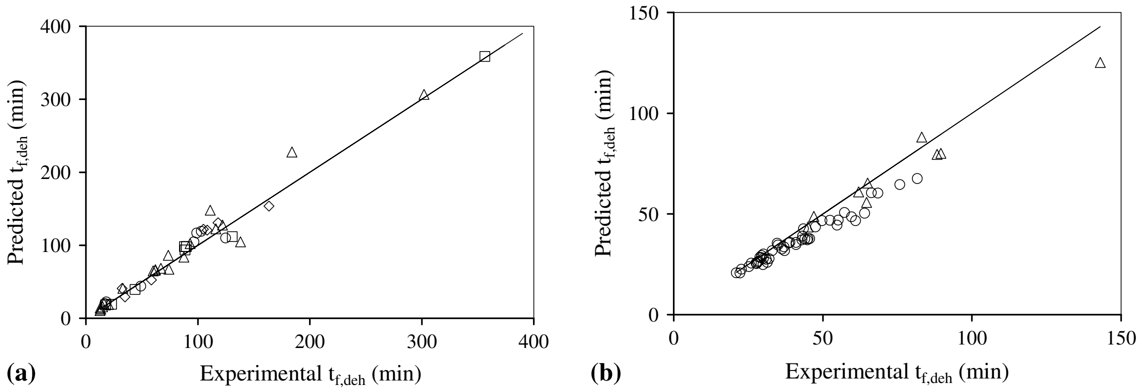

Fig. 3. (a) Predicted vs. experimental $t_{f,deh}$ for different foods (plates): (◇) lean beef, (○) carp, (△) tylose, (□) mashed potatoes. Experimental data are from Ref. [27]. (b) Predicted vs. experimental $t_{f,deh}$ for meat balls (spheres): (△) Ref. [4], (○) Ref. [28].

values needed for the heat transfer coefficient were taken from the original data. Those values are included in Table 5.

A total of 101 experimental data were used, 44 for slabs and 57 for spheres. Fig. 3(a) and (b) presents—for slabs and spheres, respectively—the comparison between all predicted and experimental $t_{f,deh}$. The whole set of predicted $t_{f,deh}$ had an average error of −3.01%, with averages of 4.62% and −9.01% for plates and spheres, respectively. As shown in Table 5, each of the sets had average errors lower than 10%. In view of these values and considering the differences in food types, composition and structure, working conditions and also the inherent uncertainties of experimental data, we consider that the proposed prediction method is very accurate.

5. Conclusions

A very simple method was developed for the prediction of food freezing times with surface dehydration, valid for the three simple regular geometries: plane plate, infinite cylinder and sphere.

The method accurately reproduces freezing time values predicted by a proven numerical software, covering a wide range of food sizes and working conditions.

Model predictions were validated against all the available experimental data of freezing times with surface dehydration, showing high accuracy for all the data sets tested.

Acknowledgments

Authors Salvadori and Mascheroni are Scientific Researchers and author Campañone is a Fellow, all from the National Research Council (CONICET) of Argentina. This research was supported by grants from CONICET, ANPCyT and Universidad Nacional de La Plata from Argentina.

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