



encit 2020



18th Brazilian Congress of Thermal Sciences and Engineering
November 16-20, 2020 (Online)

ENC-2020-0502

IMPROVED LUMPED-DIFFERENTIAL ANALYSIS OF THE FREEZING PROCESS IN A SUPERCOOLED WATER DROPLET

Emerson Barbosa dos Anjos

Carolina Palma Naveira-Cotta

Laboratory of Nano and Microfluidics and Micro-Systems - LabMEMS, Mechanical Eng. Dept - Federal University of Rio de Janeiro, Rio de Janeiro, RJ, Brazil.

emersonbanjos@coppe.ufrj.br, carolina@mecanica.coppe.ufrj.br

Renato Machado Cotta

General Directorate of Nuclear and Technological Development, DGDNTM, Brazilian Navy, Rio de Janeiro, RJ, Brazil

Laboratory of Nano and Microfluidics and Micro-Systems - LabMEMS, Mechanical Eng. Dept - Federal University of Rio de Janeiro, Rio de Janeiro, RJ, Brazil.

cotta@mecanica.coppe.ufrj.br

Igor S. Carvalho

Petrobras S.A., Rio de Janeiro, Brazil

igorscarvalho@petrobras.com.br

Manish K. Tiwari

Nanoengineered Systems Laboratory, UCL Mechanical Engineering, University College London, London, WC1E 7JE, UK

m.tiwari@ucl.ac.uk

Abstract. *This work presents a theoretical analysis of the transient freezing process of a supercooled droplet immersed in a cold air stream. The main objective is in evaluating the temperatures evolution and the time of freezing for the whole process through a proper yet simple mathematical model for the energy balance in the suspended water droplet undergoing solidification. The entire freezing process can be described by four distinct stages, namely, supercooling, recalescence, solidification, and cooling. At each stage, a model reduction methodology known as the Coupled Integral Equations Approach (CIEA) is employed, which reduces the partial differential equation for the temperature distribution within the spherical droplet into a system of coupled ordinary differential equations for temperatures and moving boundary position. The resulting lumped-differential model is expected to offer improved accuracy with respect to the classical lumped system analysis, since boundary conditions are accounted for in the averaging process through the Hermite integral approximations employed. The results of the CIEA were verified against an accurate hybrid numerical-analytical solution for the full partial differential formulation, recently advanced through the Generalized Integral Transform Technique (GITT), besides comparing against numerical and experimental results available in the literature. After verification and validation of the proposed solution, a parametric analysis was implemented, for different conditions of airflow velocity and droplet radius, highlighting that variations in these parameters cause changes in the Biot number, which has a direct effect on the accuracy of the improved lumped-differential formulation.*

Keywords: *Droplet freezing, Heat conduction, Lumped analysis, CIEA, Moving boundary, Integral transforms, GITT*

1. INTRODUCTION

The freezing phenomena within a supercooled water droplet is of interest to many engineering and environmental processes, with applications in various fields such as aerospace and aeronautics, power transmission, meteorology, refrigeration, and cryopreservation. Besides, the freezing time of supercooled water droplets is an especially important parameter in the study of surface coatings to prevent the accumulation of ice. The aeronautical industry is particularly affected due to the extreme environmental conditions in which aircrafts operate. In this sense, the mathematical modeling of the freezing mechanism is particularly useful for improved design and performance of “icephobic” surface modifications for aeronautical sensors and components.

Freezing of suspended or free water droplets has been studied extensively over the past few years by several researchers (Feuillebois et al., 1995; Hindmarsh et al., 2003; Strub et al., 2003; Tabakova and Feuillebois, 2004; Popovicheva et al., 2008; Alizadeh et al., 2012; Chaudhary and Li., 2014; Yao et al., 2019; Carvalho, 2019). The freezing process of a droplet can be described in four distinct stages. Hindmarsh et al. (2003) defined each of these stages as

follows: 1. A supercooling stage, during which the liquid droplet is cooled from an initial temperature to below the equilibrium freezing temperature until crystal nucleation occurs; 2. A recalescence stage, during which supercooling drives rapid kinetic crystal growth from the crystal nuclei. This stage ends when the supercooling is exhausted, and the droplet has reached its equilibrium freezing temperature; 3. The solidification stage, where crystal growth is governed by the heat transfer rate from the droplet to the point where the droplet liquid is completely frozen; 4. cooling or tempering stage, when the solid droplet temperature is reduced towards the ambient air temperature.

A few fairly recent research efforts have been put on developing theoretical models for droplet freezing and applying semi-analytical or numerical techniques. Hindmarsh et al. (2003) developed a simple heat balance model to predict the freezing time of water droplets and experimentally measured the temperature evolution of a suspended freezing droplet in a cold air stream. Tabakova and Feuillebois (2004) numerically solved for the freezing process of supercooled water droplets on a cold surface as a 2D one-phase Stefan problem. The authors then suggested explicit correlations to estimate the freezing time based on their numerical results. The advancement of purely numerical approaches has allowed for the computational handling of moving boundary problems in heat transfer, but has also confirmed the high computational costs involved for an accurate solution of such problems, governed by an increased number of parameters. In this sense, a robust and cost-effective hybrid numerical-analytical approach known as the Generalized Integral Transform Technique – GITT (Cotta, 1990; Cotta, 1993; Cotta and Mikhailov, 1997) has been advanced to the analysis of moving boundary heat conduction problems (Diniz et al., 1990, Ruperti Jr. et al., 1992, Cotta and Mikhailov, 1997, Sias et al., 2009, Monteiro et al., 2011). More recently, (Carvalho, 2019) employed the GITT including the adoption of a nonlinear eigenvalue problem due to the associated temperature dependent boundary conditions coefficients (Cotta et al., 2016) to obtain the temperature distributions in a freezing droplet and to accurately compute the duration of each stage in the process. This solution provides a set of reference results for this complex nonlinear partial differential system, to within moderate computational costs.

Nevertheless, simplified reduced models are expected to be particularly useful in further reducing computational costs and analytical involvement, especially in connection with computationally intensive tasks, such as in optimization, real time simulation, and inverse problem analysis, when the direct problem is required to be solved a large number of times, or when populational dynamics analysis and stochastic simulations are undertaken. Therefore, an improved lumping procedure, based on the so-called Coupled Integral Equations Approach (CIEA) (Aparecido and Cotta, 1989, Cotta et al., 1990, Cotta and Mikhailov 1997, Correa and Cotta, 1998, Sphaier et al., 2017) is here employed as a formulation simplification technique for the present heat conduction problem with moving boundaries. The resulting improved lumped-differential formulation offers substantial enhancement over classical lumping schemes in terms of accuracy, without introducing additional complexity in the corresponding final simplified differential equations to be handled. The CIEA formalism approximates integrals of the temperature and flux profiles by a linear combination of the integrand and its derivatives at the integration limits. The integral approximation formulae were originally developed in Hermite (1878) and first proposed by Mennig et al. (1983) in approximately solving initial and boundary value problems. This problem reformulation strategy has been applied to various thermal sciences and engineering problems such as in fin analysis, conjugated problems, drying, aerospace thermal protection system, membrane metals extraction, nuclear fuel rods, heat exchangers, micro-reactors for biodiesel synthesis, nanocomposites, among others (Cotta and Ramos, 1993; Scofano Neto and Cotta, 1993, Su & Cotta, 2001; Cotta et al., 2004, Dantas et al., 2007, Pontedeiro et al., 2008; Naveira-Cotta et al., 2009, Knupp et al., 2012; Cardoso et al., 2014, Dourado et al., 2017, Costa Jr. and Naveira-Cotta, 2019).

Thus, the present work aims at advancing a lumped-differential model for the solidification of a supercooled droplet immersed in a cold air stream, subject to the three main transport phenomena at the interface between the droplet and the surroundings: convective heat transfer, convective mass transfer, and thermal radiation. The CIEA is adopted to transform the partial differential equations (PDE) formulation of the energy balance, into an ordinary differential equations (ODEs) system for the boundary temperatures and moving boundary position. The resulting reduced model is then solved using a code developed on the *Mathematica* platform, Wolfram (2019). The transient temperature distributions for each stage of the process are obtained and analyzed in different scenarios. In addition, a critical analysis of the results via CIEA is undertaken through a comparison with the reference results obtained via GITT and numerical/experimental results from the literature.

2. PROBLEM FORMULATION

In formulating the energy balance for the freezing of a suspended water droplet, the following assumptions are considered: i) The droplet is at rest and immersed in air, subjected to heat convection; ii) The droplet continues with the same volume and spherical shape all along the process; iii) The heat transfer process is assumed to be one-dimensional in the radial direction; iv) Ice and water are isotropic and homogeneous, with constant properties; v) Density changes in the interface liquid/ice are disregarded; vi) In the recalescence stage, the droplet temperature will be considered uniform and equal to the equilibrium temperature for freezing (T_f); vii) In the solidification stage, the liquid phase temperature is considered constant and equal to T_f . Under these assumptions, the mathematical model is written as, Carvalho (2019):

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 k_i \frac{\partial T_{i,j}(r,t)}{\partial r} \right) = \rho_i c_i \frac{\partial T_{i,j}(r,t)}{\partial t} \quad \text{for} \begin{cases} 0 < r < R, \quad t > t_{j-1} \text{ (1st and 4th stage)} \\ s(t) < r < R, \quad t > t_2 \text{ (3th stage)} \end{cases} \quad (1)$$

$$T_{l,l}(r, t_0) = T_{ini} ; T_{ice,3}(r, t_2) = T_f ; T_{ice,4}(r, t_3) = T_d(r) ; s(t_2) = R_{ini} \quad (2a-d)$$

$$\left. \frac{\partial T_{l,l}(r,t)}{\partial r} \right|_{r=0} = 0 ; \left. \frac{\partial T_{ice,4}(r,t)}{\partial r} \right|_{r=0} = 0 \quad (3a,b)$$

$$-k_i \left. \frac{\partial T_{i,j}(r,t)}{\partial r} \right|_{r=R} = h [T_{i,j}(R,t) - T_\infty] + h_m L_k [\rho_{vi,s}(T_{i,j}(R,t)) - \rho_{vi,a}] + \varepsilon \sigma [T_{i,j}^4(R,t) - T_a^4] \quad (4)$$

$$L \rho_{ice} \frac{\partial s(t)}{\partial t} = k_{ice} \left. \frac{\partial T_{ice,3}(r,t)}{\partial r} \right|_{r=s(t)} ; T_{ice,3}(s(t), t) = T_f \quad (5a,b)$$

where i stands for the physical state of water, liquid (l) or ice, $T_{i,j}$ [K] is the temperature of phase “i” in the “j” stage, k_i [W/mK] is the thermal conductivity of phase “i”, c_i [J/kgK] is the specific heat, T_{ini} [K] is the initial temperature, T_a [K] is the air temperature, T_f [K] is the equilibrium freezing temperature, $T_d(r)$ is the temperature profile at the end of the freezing stage, h [W/m²K] is the convective heat transfer coefficient, R_{ini} [m] is the initial interface position, h_m [m/s] is the mass transfer coefficient, $\rho_{vi,s}$ [kg/m³] is the vapor density at the droplet surface, $\rho_{vi,a}$ [kg/m³] is the vapor density in the air, ρ_i is the water density of phase “i”, $s(t)$ [m] is the freezing front position, ε is the emissivity of the droplet surface, σ [W/m²K⁴] is the Stefan-Boltzmann constant, r [m] is the droplet radius, L_k [J/kg] is the evaporation latent heat (L_e) for the 1st stage and latent heat of sublimation (L_s) for the 3rd and 4th stages, and L is the latent heat of water fusion, t [s] is time, j is the index of the respective stage, i.e., supercooling (1), recalescence (2), solidification (3), or cooling (4), $t_0 = 0$, $t_2 \cong t_1$, and Eqs. (2a-d) represent the initial conditions.

Murphy and Kopp (2005) present a literature review of correlations for saturated vapor pressure in terms of temperature. Among these, the correlation of Bohren and Albrechet (1998) was chosen in the present work. Thus, the equations for $\rho_{v,i,s}$ and $\rho_{v,i,a}$ for water in liquid and solid states are shown below, where RH is the relative humidity in the air:

$$\rho_{v,l,s}(T_l(R,t)) = \frac{1.323}{T_{l,l}(R,t)} \exp \left(19.83 - \frac{5417}{T_{l,l}(R,t)} \right) ; \quad \rho_{v,l,a}(T_i(R,t)) = RH \left[\frac{1.323}{T_a} \exp \left(19.83 - \frac{5417}{T_a} \right) \right] \quad (6a,b)$$

$$\rho_{v,ice,s}(T_{ice}(R,t)) = \frac{1.323}{T_{ice,j}(R,t)} \exp \left(22.49 - \frac{6141}{T_{ice,j}(R,t)} \right) ; \quad \rho_{v,ice,a}(T_i(R,t)) = RH \left[\frac{1.323}{T_a} \exp \left(22.49 - \frac{6141}{T_a} \right) \right] \quad (7a,b)$$

For the calculation of the convective transfer coefficients for heat (h) and mass (h_m), correlations for the Nusselt and Sherwood numbers were taken from Beard (1976). Thus, h and h_m were calculated using the following correlations:

$$Nu = 1.56 + 0.616(Re^{1/2})(Pr^{1/3}) \quad (8)$$

$$Sh = 1.56 + 0.616(Re^{1/2})(Sc^{1/3}) \quad (9)$$

where Nu is Nusselt number, Re is Reynolds number, Pr is Prandtl number, Sh is Sherwood number, and Sc is Schmidt number, and the parameters refer to the air (ρ_a, D_a, μ_a).

For the recalescence (2nd) stage, differential equations are not required in the present model. Once nucleation occurs, it is necessary to locate the ice crystals initially formed. Two hypotheses were formulated by Hindmarsh et al. (2003), the first is when the nucleation initially occurs at the outer surface of the droplet, being colder than the inside of the droplet and thus first reaches the nucleation temperature. This leads to the formation of a spherical shell of ice which propagates inward over time. The second hypothesis, on the other hand, considers that nucleation occurs homogeneously, with crystals dispersed uniformly throughout the droplet, forming a liquid-solid mixture with an opaque appearance. The recalescence model is based on the premise that the heat required to raise the droplet temperature from T_n (nucleation temperature) to T_f , must be equal to the latent heat released to form the volume of ice produced by the nucleation. The equation proposed by Hindmarsh et al. (2003) is represented as:

$$V_{ice} = \frac{V_{dp} c_l \rho_l (T_f - T_n)}{L \rho_{ice}} \quad (10)$$

where, V_{ice} [m³] is the volume of ice, V_{dp} [m³] is the volume of the droplet.

As mentioned above, V_{ice} can be treated by two hypotheses. For the hypothesis of a spherical shell on the surface, the initial position of the interface (R_{ini} , Eq. 2d), is obtained from the formula to calculate a spherical volume. For the second hypothesis, a water-ice mixture can be considered as an uniform phase, and there is a homogeneous distribution of the ice formed throughout the droplet; however, the latent heat L should be substituted by a new value relative to the water-ice mixture, L_m , given by:

$$L_m = L \left(1 - c_l \rho_l (T_f - T_n) / L \rho_{ice} \right) \quad (11)$$

In the solidification stage, Eq. (5a) refers to the differential equation for the position of the freezing front and will depend on the hypothesis adopted to locate the ice formed in the recalescence (2nd) stage. Additional information about this subject can be found in Carvalho (2019) and Tabakova et al. (2010).

The CIEA reformulation methodology is then applied to the dimensionless form of equations (1-5). The dimensionless parameters adopted are shown in Table 1. Different levels of approximation in such mixed lumped-differential formulations can be used, starting from the plain and classical lumped system analysis, towards improved formulations, obtained through Hermite-type approximations for integrals based on the values of the integrand and its derivatives at the integration limits. In the present work, we consider just the two more usual approximations, $H_{0,0}$ and $H_{1,1}$, given by:

$$H_{0,0} \rightarrow \int_0^h y(x) dx \cong \frac{h}{2} [y(0) + y(h)] \quad (12)$$

$$H_{1,1} \rightarrow \int_0^h y(x) dx \cong \frac{h}{2} [y(0) + y(h)] + \frac{h^2}{12} [y'(0) - y'(h)] \quad (13)$$

which correspond to the classical trapezoidal and corrected trapezoidal integration rules. The final expressions for the reduced model obtained from the CIEA implementation are consolidated in Table 2.

Table 1. Dimensionless parameters for each phase of the freezing process.

Supercooling Stage (1 st)	Solidification Stage (3 rd)	Cooling Stage (4 th)	Position of the Freezing Front
$\theta_{l,1} = \frac{T_{l,1}}{T_a}$ (14)	$\theta_{ice,3} = \frac{T_{ice,3} - T_f}{T_a - T_f}$ (22)	$\theta_{ice,4} = \frac{T_{ice,4}}{T_a}$ (30)	$v(\tau) = \frac{s(\tau)}{R}$ (37)
$\theta_1^* = x \theta_{l,1}$ (15)	$\theta_3^* = x \theta_{ice,3}$ (23)	$\theta_4^* = x \theta_{ice,4}$ (31)	$n(\tau) = 1 - v(\tau)$ (38)
$x = \frac{r}{R}$ (16)	$x = 1 - \frac{r}{R}$ (24)	$x = \frac{r}{R}$ (32)	$St = c_{ice} \frac{T_f - T_a}{L}$ (39)
$\tau = \frac{k_l t}{c_l \rho_l R^2}$ (17)	$\tau = \frac{k_{ice} t}{c_{ice} \rho_{ice} R^2}$ (25)	$\tau = \frac{k_{ice} t}{c_{ice} \rho_{ice} R^2}$ (33)	
$Bi_1 = \frac{hR}{k_l}$ (18)	$Bi_3 = \frac{hR}{k_{ice}}$ (26)	$Bi_4 = \frac{hR}{k_{ice}}$ (34)	
$Bim_1 = \frac{h_m L_e R \rho_{v,o}}{k_l}$ (19)	$Bim_3 = \frac{h_m L_s R \rho_{v,o}}{k_{ice} (T_a - T_f)}$ (27)	$Bim_4 = \frac{h_m L_s R \rho_{v,o}}{k_{ice} T_a}$ (35)	
$Bir_1 = \frac{\sigma \varepsilon R T_a^3}{k_l}$ (20)	$Bir_3 = \frac{\sigma \varepsilon R T_f^3}{k_{ice}}$ (28)	$Bir_4 = \frac{\sigma \varepsilon R T_a^3}{k_{ice}}$ (36)	
$\theta_{l,0} = x \frac{T_0}{T_a}$ (21)	$\beta = \frac{T_a - T_f}{T_f}$ (29)		

It should be pointed out that the dimensionless temperatures ($\theta_{i,j}$) are defined by Eqs. (14, 22 and 30), however, the auxiliary dimensionless temperature variables (θ_j^*), as defined in Eqs. (15, 23 and 31) of Table 1, incorporate the dependent variable transformation involving the space variable x , for simplification of the differential operators in spherical coordinates into a similar representation as for rectangular coordinates.

Table 2. Reformulated governing equations obtained from the CIEA implementation.

Stages: 1 st (j = 1) or 4 th (j = 4) CIEA H _{0,0} /H _{0,0}		Stages: 1 st (j = 1) or 4 th (j = 4) CIEA H _{1,1} /H _{0,0}	
$\frac{\partial \theta_j^*(1, \tau)}{\partial \tau} = -4\theta_j^*(1, \tau) + 4[H_j - B_j(1, \tau)\theta_j^*(1, \tau)] \quad (40)$		$\frac{\partial \theta_j^*(1, \tau)}{\partial \tau} + \frac{B_j(1, \tau)}{4} \frac{\partial \theta_j^*(1, \tau)}{\partial \tau} + \frac{\theta_j^*(1, \tau)}{4} \frac{\partial B_j(1, \tau)}{\partial \tau} = -3[\theta_j^*(1, \tau) - H_j + B_j(1, \tau)\theta_j^*(1, \tau)] \quad (41)$	
Dimensionless Boundary Temperatures			
$\theta_{i,j}(1, \tau) = \theta_j^*(1, \tau) \quad (42)$		$\theta_{i,j}(0, \tau) = 2\theta_j^*(1, \tau) - H_j + B_j(1, \tau)\theta_j^*(1, \tau) \quad (43)$	
Stage: 3 rd (j = 3) CIEA H _{0,0} /H _{0,0}		Stage: 3 rd (j = 3) CIEA H _{1,1} /H _{0,0}	
$n(\tau) \frac{\partial \theta_3^*(0, \tau)}{\partial \tau} + \theta_3^*(0, \tau) \frac{\partial n(\tau)}{\partial \tau} = -4 \left[\frac{\theta_3^*(0, \tau)}{n(\tau)} + B_3(0, \tau)\theta_3^*(0, \tau) - H_3 \right] \quad (44)$		$\left[4n(\tau) + B_3(0, \tau)n(\tau)^2 \right] \frac{\partial \theta_3^*(0, \tau)}{\partial \tau} + \theta_3^*(0, \tau) \frac{\partial}{\partial \tau} \left[4n(\tau) + B_3(0, \tau)n(\tau)^2 \right] - 2n(\tau)H_3 \frac{\partial n(\tau)}{\partial \tau} = -12 \left[\frac{\theta_3^*(0, \tau)}{n(\tau)} + B_3(0, \tau)\theta_3^*(0, \tau) - H_1 \right] \quad (45)$	
Dimensionless Boundary Temperature and Dimensionless Position of the Freezing Front			
$\theta_{ice,3}(0, \tau) = \theta_3^*(0, \tau) \quad (46)$		$\frac{\partial n(\tau)}{\partial \tau} = -\frac{L}{L_m} St \left[\frac{-2\theta_3^*(0, \tau) - B_3(0, \tau)}{1 - n(\tau)} \right] \quad (47)$	
Initial Conditions			
$\theta_{l,1}(x, 0) = \frac{T_0}{T_a} \quad (48)$	$\theta_{ice,3}(x, 0) = 0 \quad (49)$	$n(0) = 0 \quad (50)$	$\theta_{ice,4}(x, 0) = \frac{T_{ice,3}(x, t_3)}{T_a} \quad (51)$
Parameters			
$H_j = Bi_j + Bir_j + \frac{\rho_{v,\infty} Bim_j}{\rho_{v,\infty}} \quad (52)$		$B_j(1, \tau) = Bi_j + Bir\theta_{i,j}^3(1, \tau) + \frac{\rho_{v,i,s,j}(\theta_{i,j}(1, \tau))}{\rho_{v,\infty}} Bim_j - 1 \quad (53)$	
$B_3(0, \tau) = Bi_3 + 4Bir_3 \left[1 + \beta\theta_{ice,3}(0, \tau) + \frac{\beta^2(\theta_{ice,3}(0, \tau))^2}{2} \right] \left[1 + \frac{\beta\theta_{ice,3}(0, \tau)}{2} \right] + \frac{\rho_{v,ice,s}(\theta_{ice,3}(0, \tau))}{\rho_{v,ice,a}} Bim_3 - 1 \quad (54)$			
$\rho_{v,l,s,1}(\theta_{l,1}(1, \tau)) = \frac{1.323}{T_a\theta_{l,1}(1, \tau)} \exp\left(19.83 - \frac{5417}{T_a\theta_{l,1}(1, \tau)}\right) \quad (55)$		$\rho_{v,l,ice,4}(\theta_{ice,4}(1, \tau)) = \frac{1.323}{T_a\theta_{ice,4}(1, \tau)} \exp\left(22.49 - \frac{6141}{T_a\theta_{ice,4}(1, \tau)}\right) \quad (56)$	
$\rho_{v,ice,s,3}(\theta_{ice,3}(0, \tau)) = \frac{1.323}{[(T_a - T_c)\theta_{ice,3}(0, \tau) + T_c]} \exp\left(22.49 - \frac{6141}{(T_a - T_c)\theta_{ice,3}(0, \tau) + T_c}\right) \quad (57)$			

3. RESULTS AND DISCUSSION

The ODEs of the reduced model obtained by the CIEA (Table 2) are numerically solved through a symbolic-numerical code built on the *Mathematica* platform, Wolfram (2019). Initially, a comparison for different values of the Biot number (Bi) is presented showing the solutions via GITT (PDE model, Eqs. (1-5)), CIEA with H_{1,1}/H_{0,0}, and CIEA with H_{0,0}/H_{0,0} approximations. Subsequently, a comparison of CIEA and GITT results for boundary temperatures is provided. For the GITT, the truncation order was taken as M = 40 for all the presented results. Then, the present approach is verified and validated against theoretical and experimental results in the literature and a parametric analysis is performed, varying the radius of the droplet and the airflow velocity, which affect the Biot number value.

3.1 Biot number variation

The Biot number represents a measure of the ratio of the conductive and convective thermal resistances at the droplet surface, being an important governing parameter for this application. A few values of Bi were chosen to explore the limit of applicability and to demonstrate the accuracy of the CIEA. To achieve this objective, results obtained by CIEA are compared against those for the full partial differential model, Eqs. (1-5), obtained by the GITT hybrid approach, as described in Carvalho (2019). Figs. 1(a)-(c) present a comparison of the dimensionless boundary temperature $\theta_{l,1}(1, \tau)$, Eq. (42), evolution, for increasing values of Biot number at the surface of the water droplet, $Bi = 0.1, 1.0, 10.0$, as computed from the improved lumped-differential formulations ($H_{0,0}/H_{0,0}$ and $H_{1,1}/H_{0,0}$) and from the full model by GITT ($M = 20$), in a range of τ from 0 to 2 for the supercooling (1^{st}) stage. For the lower Biot number, represented by Fig. 1(a), $Bi = 0.1$, the two lumped formulations do not show a marked difference between them, and both are fairly accurate approximations to the partial differential formulation results (GITT). For low Biot numbers (e.g. $Bi = 0.1$) the water droplet has approximately uniform temperature fields along its radius, favoring the application of the $H_{0,0}/H_{0,0}$ and $H_{1,1}/H_{0,0}$ lumping schemes. The classical lumped system analysis essentially equates the boundary and average temperature values. The CIEA seeks to obtain an improved relation between $\theta_{l,1}(1, \tau)$ and $\theta_{av}(\tau)$, through the application of Eqs. (12-13) into Eqs. (1-4), and the greater the order of the formulation ($H_{1,1}/H_{0,0}$ is of higher order than $H_{0,0}/H_{0,0}$), the more accurate the results obtained, as seen in Figs.1. As the Biot number increases, the deviations between the $H_{1,1}/H_{0,0}$, $H_{0,0}/H_{0,0}$ formulations become more evident, and their respective deviations to the GITT becomes clearer. For the case with $Bi = 1.0$ (Fig. 1(b)), the $H_{0,0}/H_{0,0}$ is still quite reasonable, while $H_{1,1}/H_{0,0}$ remains accurate in relation to GITT. In the last case, when $Bi = 10.0$, Fig. 1(c), the fact that $H_{0,0}/H_{0,0}$ is indeed less accurate, especially at shorter times, is noticeable. On the other hand, the $H_{1,1}/H_{0,0}$ model remains with good accuracy in relation to GITT and can be used as a reliable reduced model for this problem. It is clear from Eqs. (40) and (41) that the formulation $H_{1,1}/H_{0,0}$ carries more information about the problem than the $H_{0,0}/H_{0,0}$ approximation.

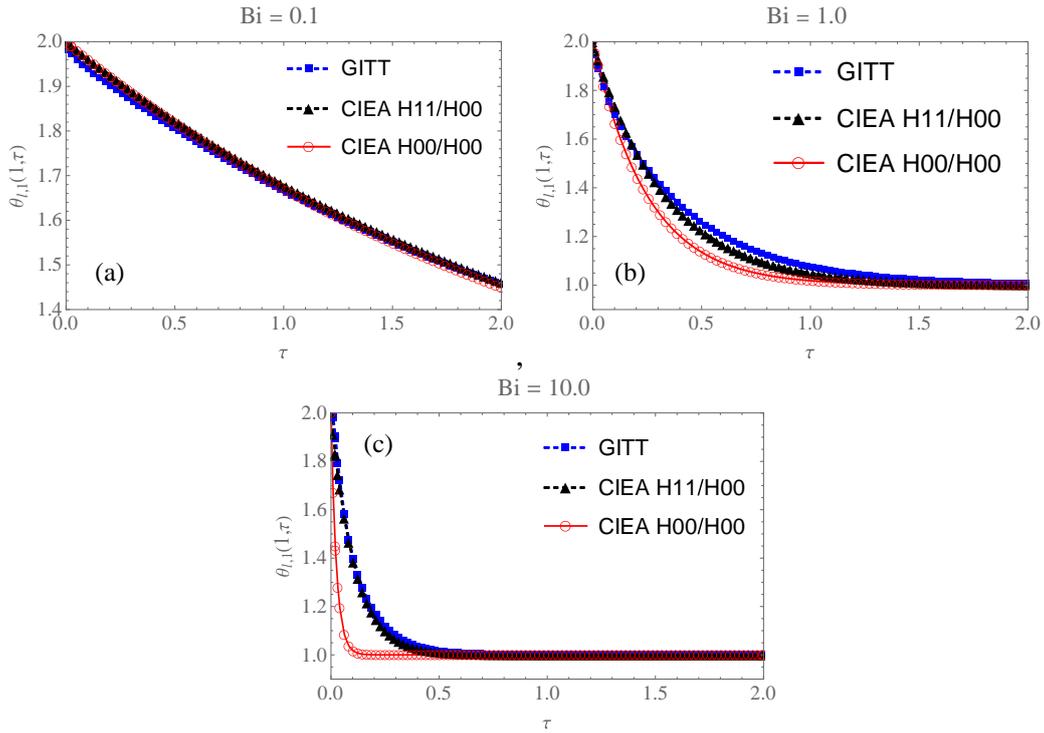


Figure 1. Transient behavior of the dimensionless temperature for the supercooling(1st) stage at three different values of Biot number: (a) $Bi = 0.1$, (b) $Bi = 1.0$, (c) $Bi = 10.0$, $Bim_1=Bir_1=0$, $T_0=2T_a$, computed by GITT ($M= 40$), CIEA $H_{0,0}/H_{0,0}$, and CIEA $H_{1,1}/H_{0,0}$.

Figures 2(c-d) and 2(e-f) show the behavior of the dimensionless position of the freezing front and the evolution of the dimensionless boundary temperature, $\theta_{ice,3}(0, \tau)$, when $Bi = 0.1$ to $Bi = 10.0$ for three different Stefan number (St) values ($St=0.11, 0.15, 0.20$), respectively. The Stefan number (Eq. 39) is an important parameter for problems with phase change, defined as the ratio between the sensible and latent heats released during the phase change process. Comparing the results for the Biot number values, as Bi is increased, one can observe an increasing deviation on the prediction of the freezing front, as the end of the solidification process is approached, among the approximate formulations and the reference GITT solution of the full partial differential model, and this deviation is still present regardless of the Stefan number. It can be noticed that the freezing front velocity, represented by the time derivative of the front position in Figs.2,

increases markedly as the end of the solidification is approached. On the other hand, according to Eqs. (47), this velocity is directly proportional to the temperature spatial derivative at the interface, which is in fact approximated by the lumped-differential formulation. Therefore, even a relatively small error in this quantity may induce a significant variation on the prediction of the solidification ending time, as can be observed in Figs.2. (c, e), though less noticeable in the dimensionless temperature predictions. As the end of the solidification (3rd) stage is approached, temperature predictions by different solution techniques reach their final values at different values of τ , as can particularly be observed in Figs. 2.(d, f). As expected, the freezing process transient duration is noticeably linked to the Biot number and Stefan number values, with much faster processes for larger Bi and St.

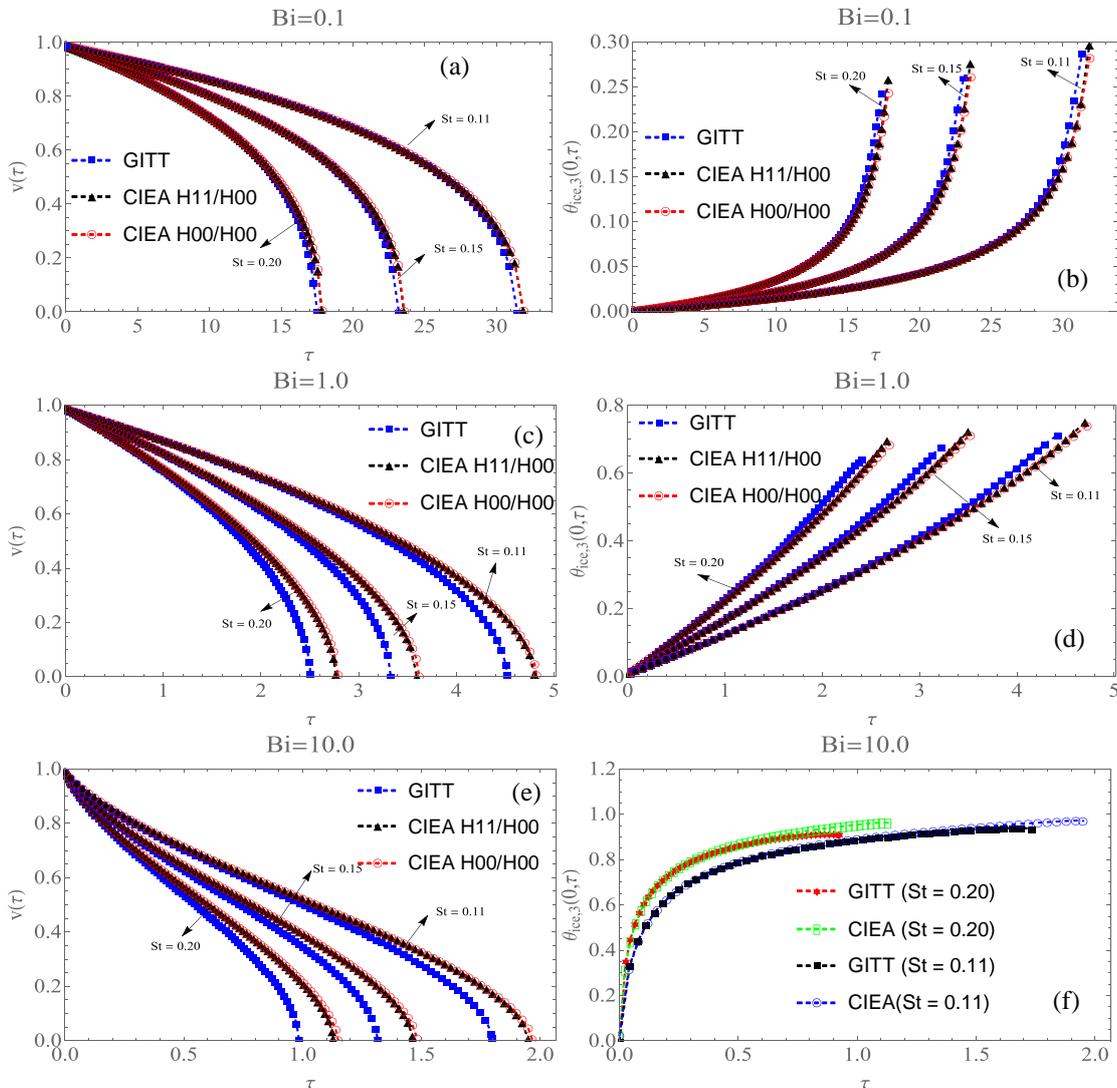


Figure 2. Comparison of GITT solution and CIEA lumped-differential formulations for the dimensionless position of the freezing front and for the dimensionless boundary temperature in the solidification stage: (a) and (b) $Bi = 0.1$, (c) and (d) $Bi = 1.0$, and (e) and (f) $Bi = 10.0$, $Bim_3 = Bir_3 = 0$.

3.2 Validation and Parametric Analysis

The present methodology is now employed in a typical situation of supercooled droplet freezing, which corresponds to a previously reported experimental and theoretical work by Hindmarsh et al. (2003). Table 3 summarizes the input data here employed. Figure 3 shows a comparison of the results from Hindmarsh et al. (2003) with the present study for the evolution of the dimensionless boundary temperature throughout the super-cooling (1st) (Fig. 3a) and cooling (4th) (Fig. 3b) stages. The results of Hindmarsh et al. (2003) agree fairly well with those obtained via both the CIEA $H_{1,1}/H_{0,0}$ formulation and the GITT benchmark. Variations in the airflow velocity (“ v ”) and in the water droplet radius (“ R ”) directly affect the Biot number values, and typical situations are here analyzed. Figs. 4(a-d) show the influence of the parameters v and R in the solidification (3rd) stage, through the behaviors of the dimensionless boundary temperature and the dimensionless position of the freezing front, $v(\tau)$. Increasing the airflow velocity from 0.42 to 0.97 m/s corresponds to a significant increase in Biot number (up to 33%). For the radius variation, an increase from 0.49 mm to 0.78 mm increases the Biot value by up to 38%, and an increase of 60% in the dimensionless evaporative cooling term. Thus, based

on these graphical results, it is concluded that droplets under higher airflow velocities and smaller radius, freeze more quickly. In the typical range of parameters for this application, the CIEA results for the solidification stage provide excellent predictions of dimensionless boundary temperatures and of the dimensionless position of the freezing front in comparison to the GITT reference results.

Table 3. Input data from Hindmarsh *et al.* (2003). (water and ice properties at 273.13K, and for air at 254.13K).

Variable	Values	Variable	Values	Variable	Values
D_a [m ² /s]	2.060×10^{-5}	ρ_l [kg/m ³]	1000	L [J/kg]	3.33×10^5
c_l [J/kgK]	4217	ρ_{ice} [kg/m ³]	920	L_e [J/kg]	2.502×10^6
c_{ice} [J/kgK]	2040	$\rho_{v,o}$ [kg/m ³]	4.8473×10^{-3}	L_s [J/kg]	2.838×10^6
k_a [J/msK]	0.0234	ρ_a [kg/m ³]	1.3317	ϵ	0.96
k_l [J/msK]	0.569	σ	5.670×10^{-8}	μ_a [Ns/m ²]	1.663×10^{-5}
k_{ice} [J/msK]	1.88	v [m/s]	0.42		

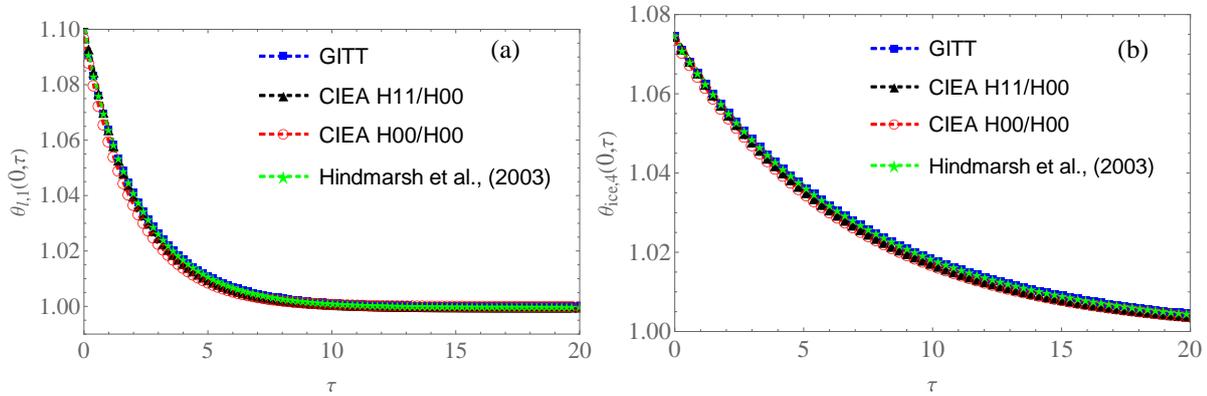


Figure 3. Comparison of the dimensionless boundary temperature at a water droplet during (a) the super-cooling (1st) and (b) the cooling (4th) stages, for the proposed approaches (GITT and CIEA) and data from Hindmarsh *et al.* (2003).

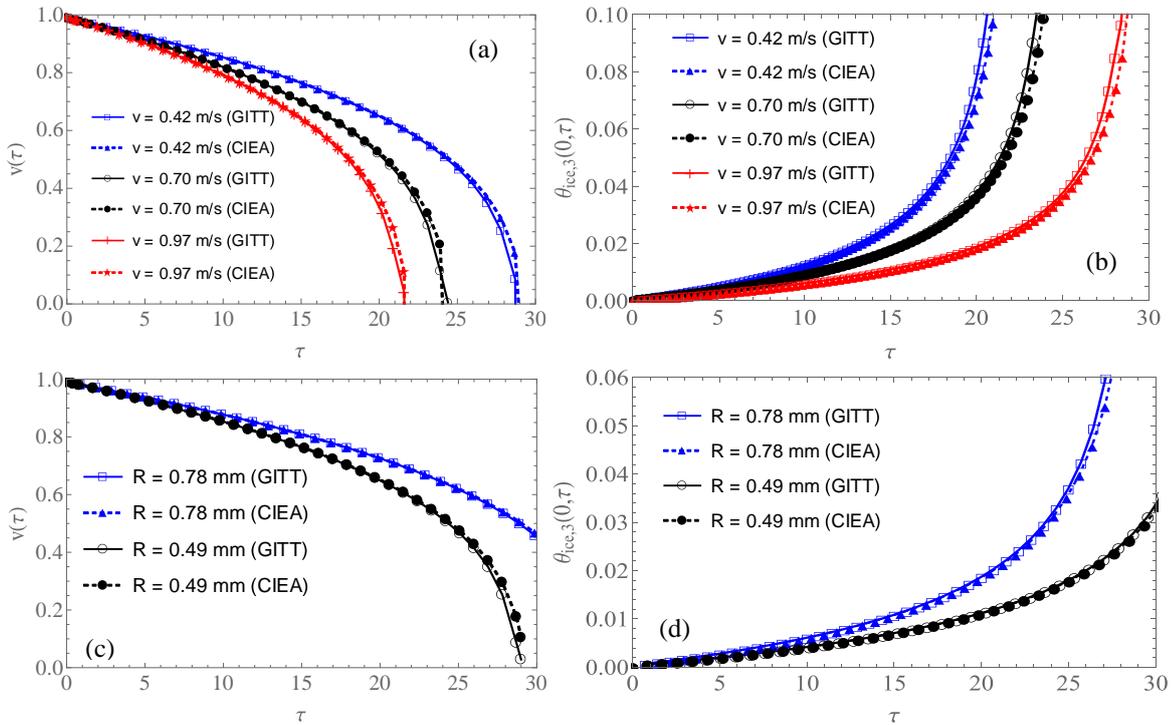


Figure 4. Influence of airflow velocity and droplet radius parameters in the position of the of dimensionless position of the freezing front and on the dimensionless droplet temperature: GITT ($M = 40$) and CIEA ($H_{1,1}/H_{0,0}$) results, $St = 0.11$.

4. CONCLUSION

A theoretical analysis was performed on the freezing of a water droplet, including all the stages of the process (supercooling, recalescence, solidification, and cooling). The energy balance for each stage was reformulated by the Coupled Integral Equations Approach (CIEA), which is a tool for generating improved lumped-differential formulations in diffusion and convection-diffusion problems. Thus, the original partial differential model for heat transfer with phase change was reduced to an ordinary differential model for the dimensionless surface temperature at the droplet and the dimensionless position of the freezing front. The numerical solution of two CIEA formulations of different accuracy orders ($H_{1,1}/H_{0,0}$ and $H_{0,0}/H_{0,0}$ approximations), were critically compared with a precision-controlled hybrid numerical-analytical solution of the full partial differential model, based on the Generalized Integral Transform Technique (GITT). Results for the local boundary temperature and freezing front position along the transient process were then analyzed in terms of the Biot number, exploring the limiting range for the lumping procedure here proposed. Besides, such approximations are validated against results in the literature for an actual application of water droplet freezing, with excellent agreement. Finally, a parametric analysis was performed for the freezing stage, varying the values of airflow velocity and droplet radius, within the typical ranges of the related application. This analysis showed that the proposed Coupled Integral Equations Approach can be utilized as an effective model reduction tool to simulate the transient behavior along the freezing of a supercooled spherical droplet. The present methodology may also be extended in reformulating more involved problems in droplets freezing, including deformed droplets along the flow, space variable boundary coefficients, and irregularly shaped droplets sitting on a surface.

5. REFERENCES

- Alizadeh, A., Yamada, M.L., Shang, W., Otta, S, et al., 2012. "Dynamics of ice nucleation on water repellent surfaces". *Langmuir*, Vol. 28 No. 6, pp. 3180-3186.
- Aparecido, J.B., and Cotta, R.M. 1989, "Improved One-Dimensional Fin Solutions", *Heat Transf. Eng.*, Vol. 11, no. 1, pp. 49-59
- Beard, K. V. 1976. "Terminal velocity and shape of cloud and precipitation drops aloft". *J. of the Atm. Sci.*, Vol. 33, No. 5, 851-864.
- Bohren, C., and Albrecht, B. 1998. "Atmospheric Thermodynamics". *Oxford University Press*, New York, USA.
- Cardoso, I. A., Macêdo, E. N., and Quaresma., J. N. N. 2014. "Improved lumped solutions for mass transfer analysis in membrane separation process of metals". *International Journal of Heat and Mass Transfer*, Vol. 68, pp. 599-611.
- Carvalho, I, S. 2019. "Análise Híbrida Numérico-Analítica da Solidificação de Gotículas Super-Resfriadas". MSc thesis, Universidade Federal do Rio de Janeiro, Rio de Janeiro, Brasil.
- Chaudhary, G., and Li, R. 2014. "Freezing of Water Droplets on Solid Surfaces: An Experimental and Numerical Study." *Experimental Thermal and Fluid Science*, Vol. 57, pp.86-93
- Correa, E.J., and Cotta, R.M., 1998, "Enhanced Lumped-Differential Formulations of Diffusion Problems", *Appl. Math*, Vol. 22, pp. 137-152.
- Costa Junior J.M., and Naveira-Cotta, C.P., 2019. "Estimation of Kinetic Constants in Micro-Reactors for Biodiesel Synthesis: Bayesian Inference with Reduced Mass Transfer Model", *Chemical Engineering Research and Design*, Vol. 141, pp.550-565.
- Cotta, R.M., 1990. "Hybrid numerical-analytical approach to nonlinear diffusion problems", *Num. Heat Transfer, Part B*, Vol. 127, pp. 217-226.
- Cotta, R.M., Ozisik, M.N., and Mennig, J. 1990. "Coupled Integral Equation Approach for Phase-Change Problem in Two-Regions Finite Slab", *J. of the Franklin Institute*, Vol. 327, No.2, pp. 225-234.
- Cotta, R.M., 1993. *Integral Transforms in Computational Heat and Fluid Flow*, *CRC Press*, Boca Raton, USA.
- Cotta, R.M., and Ramos, R. 1993. "Error Analysis and Improved Formulations for Extended Surfaces", *NATO Advanced Study Institute on Cooling of Electronic Systems*, NATO ASI Series E: Applied Sciences, Vol. 258, pp. 753-787.
- Cotta, R.M., Ruperti Jr., N.J., Falkenberg, C.V., and Su, J. 2004. "Engineering Analysis of Ablative Thermal Protection for Atmospheric Reentry:- Improved Lumped Formulations and Symbolic-Numerical Computation", *Heat Transfer Engng.*, Vol.25, No.6, pp.1-12.
- Cotta, R.M., Naveira-Cotta, C.P., and Knupp, D.C. 2016. "Nonlinear Eigenvalue Problem in the Integral Transforms Solution of Convection-Diffusion with Nonlinear Boundary Conditions", *Int. J. Num. Meth. Heat & Fluid Flow*, Invited Paper, 25th Anniversary Special Issue, Vol.26, nos.3&4, pp.767-789.
- Cotta, R.M. and Mikhailov, M.D. 1997, "Heat Conduction: Lumped Analysis, Integral Transforms, Symbolic Computation", *Wiley-Interscience*, New York.
- Dantas, L.B., Orlande, H.R.B., and Cotta, R.M. 2007. "Improved Lumped-Differential Formulations and Hybrid Solution Methods for Drying in Porous Media", *Int. J. Thermal Sciences*, Vol.46, No.9, pp.878-889.

- Diniz, A.J., Aparecido, J.B., and Cotta, R.M. 1990. "Heat Conduction with Ablation in a Finite Slab", *Int. J. Heat & Technology*, Vol. 8, pp. 30-43.
- Dourado, E.R.G., Cotta, R.M., and Jian, S., 2017. "Transient Heat Conduction Analysis in a Simplified PWR Model by an Improved Lumped Parameter Approach", *2017 International Nuclear Atlantic Conference - INAC 2017*, pp.1-10, Associação Brasileira de Energia Nuclear, ABEN, Belo Horizonte, MG, Brazil, October 22-27th.
- Feuillebois, F., Lasek, A., Creismas, P., Pigeonneau, F., and Szaniawski, A. 1995. "Freezing of a Subcooled Liquid Droplet." *Journal of Colloid And Interface Science*, Vol. 169, No. 1, pp. 90-102.
- Hermite, M Ch. 1878. "Sur la formule d'interpolation de Lagrange." *J Crelle*, Vol.84, pp.70-79
- Hindmarsh, J. P., Russell, A. B., Chen, X. D. 2003. "Experimental and Numerical Analysis of the Temperature Transition of a Suspended Freezing Water Droplet." *Int. J. of Heat and Mass T.*, Vol. 46 No.7, pp. 1199-1213.
- Knupp, D. C., Naveira-Cotta, C. P., Ayres, J. V., Cotta, R. M., and Orlande, H. R. 2012. "Theoretical-experimental analysis of heat transfer in nonhomogeneous solids via improved lumped formulation, integral transforms and infrared thermography". *Int. J. of Thermal Sciences*, Vol.62, pp. 71-84
- Mennig, J., Auerbach, T., and Halg. W. 1983. "Two Point Hermite Approximations for the Solution of Linear Initial Value and Boundary Value Problems." *Computer Methods in Applied Mechanics and Eng.* Vol. 39, No. 2: pp.199-224.
- Monteiro, E.R., Quaresma, J.N.N., and Cotta, R.M. 2011 "Integral Transformation of Multidimensional Phase Change Problems: Computational and Physical Analysis", *21st International Congress of Mechanical Engineering, COBEM-2011*, Paper # COB06391, pp.1-10, ABCM, Natal, RN, Brazil, October.
- Murphy, D. M., and Koop, T. 2005. "Review of the vapour pressures of ice and supercooled water for atmospheric applications". *Q. J. R. Meteorol. Soc.*, 131. 1539-1565.
- Naveira-Cotta, C.P., Lachi, M., Cotta, R. M., and Padet, J., 2009. "Hybrid Formulation and Solution for Transient Conjugated Conduction-External Convection", *Int. J. Heat & Mass Transfer*, Vol. 52, Issues 1-2, pp. 112-123.
- Pontedeiro, A. C., Cotta, R. M., and Su, J. 2008. "Improved lumped model for thermal analysis of high burn-up nuclear fuel rods". *Progress in Nuclear Energy*, Vol. 50, No. 7, pp. 767-773.
- Popovicheva, O., Kireeva, E., Persiantseva, N., Khokhlova, T., Shonija, N., Tishkova, V., and Demirdjian, B. 2008. "Effect of soot on immersion freezing of water and possible atmospheric implications". *Atmospheric Research*, Vol. 90, pp. 49-59.
- Ruperti Jr., N.J., Zapparoli, E.L., and Cotta, R.M. 1992. "Hybrid Solution for Phase Change Problems in Multiregion Media", *30th Eurotherm Seminar - Heat Transfer in Phase-Change Processes*, pp. 181-184, Orsay, France, October.
- Scofano Neto, F., and Cotta, R.M. 1993. "Improved Hybrid Lumped-Differential Formulation for Double-Pipe Heat Exchangers Analysis", *J. Heat Transfer*, Vol. 115, pp. 921-927.
- Sias, D.F., Ruperti Jr., N.J., and Cotta, R.M., 2009. "Enhanced Convergence of Integral Transform Solution of Ablation Problems", *High Temperatures-High Pressures, Int. J. of Thermophysical Properties Research*, Vol.38, no.2, pp.81-96.
- Sphaier, L.A., J. Su, J., and Cotta, R.M., 2017. "Macroscopic Heat Conduction Formulation", In: *Handbook of Thermal Science and Engineering*, Chapter 4, Francis A. Kulacki et al., Eds., Springer International Publishing.
- Strub, M., Jabbour, O., Strub F., and Bedecarrats J. P. 2003. "Experimental study and modelling of the crystallization of a water droplet". *Int. J. Refrig.* Vol. 26, No. 1, pp. 59-68.
- Su, J., and R. M. Cotta, R.M. 2001. "Improved Lumped Parameter Formulation for Simplified LWR Thermohydraulic Analysis", *Annals of Nuclear Energy*, Vol.28, pp.1019-1031.
- Tabakova, S., and Feuillebois, F. 2004. "On the Solidification of a Supercooled Liquid Droplet Lying on a Surface." *Journal of Colloid and Interface Science*, Vol. 272 No. 1, pp. 225-34.
- Tabakova, S., Feuillebois, F., and Radev S. 2010. "Freezing of a Supercooled Spherical Droplet with Mixed Boundary Conditions." *Proc. of the Royal Soc. A: Mathematical, Physical and Eng. Sci.*, Vol. 466, No. 2116, pp. 1117-34.
- Wolfram, S., 2019. *Mathematica v.12* – A system for doing mathematics by computer, Wolfram Research Inc., IL.
- Yao, Y., Yang, R., Li, C., Tao, Z., and Zhang, H. 2019. "Investigation of the freezing process of water droplets based on average and local initial ice fraction". *Exp. Heat Transfer*, Vol.33, No.3, pp.197-209.

6. RESPONSIBILITY NOTICE

The authors are the only responsible for the printed material included in this paper.