

**NON PARABOLIC INTERFACE MOTION FOR THE ONE-DIMENSIONAL STEFAN
PROBLEM: DIRICHLET BOUNDARY CONDITIONS**

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Over a finite one dimensional specimen containing two phases of a pure substance, it has been shown that the liquid-solid interface motion exhibits parabolic behavior at small time intervals. We study the interface behavior over a finite domain with homogeneous Dirichlet boundary conditions for large time intervals, where the interface motion is not parabolic due to finite size effects. Given the physical nature of the boundary conditions, we are able to predict exactly the interface position at large time values. These predictions, that to the best of our knowledge, are not found in the literature, were confirmed by using the heat balance integral method of Goodman and a non-classical finite difference scheme. Using heat transport theory, it is shown as well, that the temperature profile within the specimen is exactly linear and independent of the initial profile in the asymptotic time limit. The physics of heat transport provides a powerful tool that is used to fine tune the numerical methods. We also found that in order to capture the physical behavior of the interface, it was necessary to develop a new non classical finite difference scheme that approaches asymptotically to the predicted interface position. We offer some numerical examples where the predicted effects are illustrated, and finally we test our predictions with the heat balance integral method and the non classical finite difference scheme by studying the liquid-solid phase transition in Aluminum.

Keywords: Stefan problem, Heat Balance, Finite Difference Method

1. Introduction

The study and modeling of moving boundary problems such as liquid-solid phase transitions, implies the building of solutions as one of the most important tasks. Just for a few one dimensional problems on infinite or semi-finite regions is possible to find exact analytical solutions [1]. However, for the vast majority of moving boundary problems, the search of solutions is done by means of different strategies and approximate methods. The finite difference method offers approximate solutions and is frequently used to build numerical solutions of phase change problems with time independent boundary conditions [2, 3, 4, 5, 6, 7] and time dependent boundary conditions [8, 9, 10].

Approximate analytical solutions can be found as well for the class of problems that can not be solved exactly. One of these approaches is the Goodman's method or heat balance integral method (HBIM) [11, 12, 13, 14, 15, 16]. This method allows good results with less numerical and computational resources than other approximate methods. The liquid-solid phase transition on finite size systems has been studied by several authors where the main concern is to develop different numerical strategies to solve the same problem and compare with the few exact solutions available in the literature. For example, the exact solution on a semi-infinite region is compared with several semi-analytic and numerical methods for small time intervals [2, 3, 7, 15, 16, 17, 18] where the interface motion is approximately parabolic; or boundary conditions are chosen in such a way that parabolic motion is observed throughout the entire process [7, 17]. However, little is mentioned about the behavior for time values where the numerical solutions start to deviate significantly from the exact solution.

We will study the physical consequences of having Dirichlet boundary conditions on both sides of the specimen, where finite size effects become significant for large times. Therefore, the goal of this work is to offer a physical interpretation of one dimensional heat transport with Dirichlet boundary conditions, where a liquid-solid phase transition is taking place on a pure substance. For the solidification process of the liquid phase, it will be assumed that heat flow is low enough, so a supercooling phase is not formed. Given that the nature of the solution is highly dependent on the boundary conditions imposed on the specimen; in this work, we focus on the physical implications of homogeneous Dirichlet boundary conditions and verify our predictions with the HBIM and a new non classical finite difference scheme (NC-FDS). We found that, in order to approach asymptotically to the predicted position of the interface, it was necessary to develop this NC-FDS. The material is organized in several sections as follows: In section 2, a brief description of the problem is given and in section 3 we describe the NC-FDS proposed to solve the problem. The approximate analytical method of Goodman or HBIM is described in section 4, results from both methods are compared and tested against the exact asymptotic time limit, and an example for Aluminum is shown in section 5. Finally, the conclusions are presented in section 6.

2. Statement of the Problem

Consider a liquid phase in contact with a solid phase, both separated by an interface with position ξ at some fusion temperature T_f , where the total heat flow through the interface causes its displacement. Let us assume that the liquid and solid phases, have a temperature profile $T_1(x,t)$ and $T_2(x,t)$ respectively, where the temperature at any point within the liquid phase, is above the fusion temperature T_f and

within the solid phase, the temperature at any point, is below T_f . The temperature profiles have the following homogeneous Dirichlet boundary conditions

$$T_1(0,t) = T_l, \quad T_2(L,t) = T_s, \quad \text{and} \quad T_1(\xi,t) = T_2(\xi,t) = T_f \quad (1)$$

where the subindex 1 (2) represents liquid (solid) phase. The left edge of the sample in contact with the liquid is fixed at some temperature T_l , and the right edge in contact with the solid phase is fixed at some temperature T_s . We will assume temperature profiles that in general are functions of the position

$$T_i(x,0) = f_i(x), \quad i = 1, 2; \quad (2)$$

where $f(x)$ can be obtained in order to satisfy the boundary conditions given by equation (1); where $\xi(0) = B$, with $B > 0$.

The other equations that model this problem are the diffusion heat equations in mediums 1 and 2,

$$\frac{\partial T_i}{\partial t} = \alpha_i \frac{\partial^2 T_i}{\partial x^2}, \quad \text{with} \quad (i-1)\xi \leq x \leq (2-i)\xi + (i-1)L, \quad (3)$$

where $\alpha_i = k_i / \rho_i C_i$ is the heat diffusion coefficient in phase i . These diffusion constants depend on the specific heat capacity C_i , density ρ_i and thermal conductivity k_i at each phase. We will assume that these thermodynamic variables do not depend on the temperature and for the solidification case, we do not consider the formation of a supercooled liquid to drive the phase transition. With these assumptions, the velocity of the interface $d\xi / dt$, between the two materials, is given by the Stefan Condition (SC)

$$\rho_i L_f \frac{d\xi}{dt} = k_2 \left. \frac{\partial T_2}{\partial x} \right|_{x=\xi^+} - k_1 \left. \frac{\partial T_1}{\partial x} \right|_{x=\xi^-}, \quad (4)$$

where L_f is the latent heat of fusion and ρ_i is the density of the solid or liquid phase, depending on which direction is the phase transition, taking place. The problem is completely defined by equations (1)-(4). The well-posedness has been established by other authors [16, 17]; therefore, (1)-(4) has a unique classical solution, which will be approximated using the NC-FDS and the HBIM.

3. Numerical Solutions

Finite difference methods are one of the most popular numerical methods to find approximate solutions for boundary problems described by PDE's. In this method a continuous region is transformed into a finite number of points (nodes) and an approximate solution is found only at these points, which constitute a grid or mesh. For this reason, the differential operators are approximated or discretized at the mesh points.

3.1 Non Classical Finite Difference Scheme

An implicit scheme was used to find the solutions of the diffusion heat equation; that is, the partial time derivative of the temperature is expressed as a first order approximation of the backward difference in time, given by

$$\frac{\partial T_i}{\partial t} \approx \frac{{}^{m,n}T_i - {}^{m,n-1}T_i}{\Delta t}, \quad (5)$$

where Δt represents the length of the step in t . The discretization of the argument x , is represented by m , and the argument t is represented by n . Therefore, in this notation; ${}^{m,n}T_i = T_i(x_m, t_n)$. To obtain the proposed NC-FDS, we start by adding the Taylor expansions for ${}^{m+1,n}T_i$ and ${}^{m-1,n}T_i$ up to fourth order in Δx_i , which is the length of the step in x ; and keeping in mind that ${}^{m,n}T_i^{(j)} = \partial^j T_i(x_m, t_n) / \partial x^j$, we obtain

$${}^{m+1,n}T_i + {}^{m-1,n}T_i = 2 {}^{m,n}T_i + \Delta x_i^2 {}^{m,n}T_i^{(2)} + \frac{\Delta x_i^4}{12} {}^{m,n}T_i^{(4)} + \dots \quad (6)$$

Then, we apply the central difference definition to the fourth derivative as

$${}^{m,n}T_i^{(4)} = \frac{{}^{m+1,n}T_i^{(2)} - 2 {}^{m,n}T_i^{(2)} + {}^{m-1,n}T_i^{(2)}}{\Delta x_i^2} + o(\Delta x_i^2), \quad (7)$$

and substitute this expression in equation (6), to obtain the following relation

$$\frac{{}^{m+1,n}T_i - 2 {}^{m,n}T_i + {}^{m-1,n}T_i}{\Delta x_i^2} = \frac{{}^{m+1,n}T_i^{(2)} + 10 {}^{m,n}T_i^{(2)} + {}^{m-1,n}T_i^{(2)}}{12} + o(\Delta x_i^4) \quad (8)$$

Instead of substituting the Taylor expansions in the heat equation; in this higher order scheme, the discretized heat equation is substituted in equation (8) to obtain the following model of six points or nodes, which we have defined as NC-FDS

$$\beta_i^{(1)} {}^{m-1,n}T_i + \beta_i^{(2)} {}^{m,n}T_i + \beta_i^{(1)} {}^{m+1,n}T_i - {}^{m+1,n-1}T_i - 10 {}^{m,n-1}T_i - {}^{m-1,n-1}T_i = 0, \quad (9)$$

with $\beta_i^{(1)} = (1 - 12\lambda_i)$, $\beta_i^{(2)} = (10 + 24\lambda_i)$ and $\lambda_i = \alpha_i \Delta t / \Delta x_i^2$, for $i = 1, 2$. The derivatives that appear in the SC, equation (4), are also obtained by using a fourth order approximation as

$${}^{m,n}T_i^{(j)} = \frac{j}{12\Delta x_i} \left(-25 {}^{m,n}T_i + 48 {}^{m+j,n}T_i - 36 {}^{m+2j,n}T_i + 16 {}^{m+3j,n}T_i - 3 {}^{m+4j,n}T_i \right), \quad (10)$$

where $j = -1$ for $i = 1$ (medium 1), and $j = 1$ for $i = 2$ (medium 2).

4. Approximate Analytical Solution

An approximate analytical method was proposed by Goodman [11], to seek solutions for moving boundary problems, where the equations that govern transport phenomena are the heat-diffusion equations, and the dynamics of the moving boundary is governed by the balance in the heat flux. Temperature profiles will not only be assumed constant along each medium, we will also consider cases with parabolic profiles in the position; however, other profile shapes can be considered since, as we will show later, the physical behavior of the system at large time values is completely independent of the initial profile. In this framework, the HBIM [12] suggests representing the temperature profiles as

$$T_i(x, t) = a_i (\xi - x) + b_i (\xi - x)^2, \quad \text{with } (i-1)\xi \leq x \leq (2-i)\xi + (i-1)L, \quad (11)$$

where a_i and b_i with $i = 1, 2$ for mediums 1 and 2, are functions of time. Equation (11) obeys the Dirichlet boundary conditions given by equation (1).

The constants a_i and b_i at $t = 0$ s with $i = 1, 2$, are determined in order to satisfy the boundary conditions. Some extra conditions are needed to determine the initial values of a_i and b_i which can be obtained

from the initial temperature profile. After applying the Dirichlet boundary conditions to these parabolic profiles, the following relations between the functions a_i and b_i are obtained for the liquid and solid phases as

$$a_1\xi + b_1\xi^2 = T_l, \quad \text{and} \quad a_2(L - \xi) + b_2(L - \xi)^2 = T_s. \quad (12)$$

Once the boundary conditions have been applied to the temperature profiles, the dynamics of the moving boundary can be obtained by substitution of equation (12) in the SC, equation (4). The resulting equation is an ordinary differential equation in time (ODE), for the position ξ of the interface

$$L_f \frac{d\xi}{dt} = k_1 a_1 + k_2 a_2. \quad (13)$$

The key element of this method is to average the diffusion equations over the position variable, by integrating the diffusion equation in medium 1, from $x = 0$ to $x = \xi$, and in a similar manner, integrating the diffusion equation in medium 2, from $x = \xi$ to $x = L$. After averaging in the liquid phase (medium 1), it is obtained an ODE in time, given by

$$\frac{1}{3} \frac{db_1}{dt} \xi^2 + \frac{d\xi}{dt} (b_1 \xi + a_1) + \frac{1}{2} \frac{da_1}{dt} \xi - 2\alpha_1 b_1 = 0. \quad (14)$$

Similarly for the solid phase, the ODE equation obtained after averaging over the position is given by

$$\frac{1}{3} \frac{db_2}{dt} (L - \xi)^2 - \frac{d\xi}{dt} b_2 (L - \xi) + \frac{1}{2} \frac{da_2}{dt} (L - \xi) - a_2 \frac{d\xi}{dt} - 2\alpha_2 b_2 = 0. \quad (15)$$

Solving for a_1 and a_2 from equation (12) and substituting in equations (13), (14) and (15); a set of three ODE in time is obtained, and solved with the initial boundary conditions.

5. Results and Discussion

In this part of the results we show the asymptotic limits found for this problem and a few numerical experiments that validate the NC-FDS and the HBIM. For these experiments, we will set the thermodynamic variables of density and specific heat equal to one. Therefore, the diffusivity is reduced to $\alpha_1(\alpha_2) = k_1(k_2)$ for the liquid (solid) phase. The heat equation in each medium is simplified correspondingly, and the SC, equation (4) is reduced to

$$L_f \frac{d\xi(t)}{dt} = k_2 \left. \frac{\partial T_2(x,t)}{\partial x} \right|_{x=\xi(t)^+} - k_1 \left. \frac{\partial T_1(x,t)}{\partial x} \right|_{x=\xi(t)^-}, \quad (16)$$

since we are considering phases with the same density $\rho_i = 1$. The following examples show a comparison between the NC-FDS and HBIM, indicating also, the maximum deviation between both types of approaches in each example as $|\xi_{\text{HBIM}} - \xi_{\text{NC-FDS}}|_{\text{max}}$ ($|T_{\text{HBIM}} - T_{\text{NC-FDS}}|_{\text{max}}$) for the interface (temperature) history. For the finite difference simulations, the results presented in these figures, use a fine mesh and a value of $\Delta t = 2.5 \times 10^{-5}$.

Figure 1 is an example where the motion of the interface is initially set at $\xi(0) = 0.20$. The figure shows the solution obtained with the NC-FDS and the HBIM. A latent heat of $L_f = 2.0$ is used, and the

temperatures at the boundaries are $T_l = 1.0$ and $T_s = -0.4$. The diffusivities used are $k_1 = 1.5$ and $k_2 = 2.0$. The time interval where the motion of the interface was studied is $t_{max} = 2.0$ with $N_t = 8.0 \times 10^4$ time partitions, and the spatial mesh in this example had, $N_1 = 41$ and $N_2 = 81$ nodes. Figure 1 also shows the time evolution of the temperature at $x = 0.40$. All remaining figures indicate the limiting value for the interface and temperature, and as will be shown in section 5.1, these values are obtained by studying the proper physical behavior of the interface and temperature profile for large values of t .

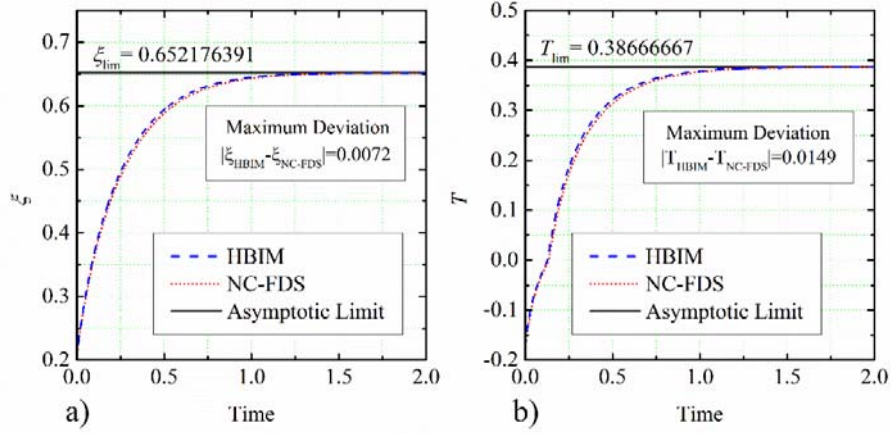


Fig. 1. a) Interface movement and b) temperature history at $x = 0.4$

5.1 Large Time Limit of the Solution

In this section we will offer a physical insight to the obtained solutions according to the boundary conditions considered. The physical phenomena is described by the phase transition between the two phases and heat transport within each phase, given that the specimen is subject to Dirichlet boundary conditions. These boundary conditions imply that the specimen is constantly heated from the left edge by a heat source, keeping the temperature at this boundary fixed, at some value above the fusion point of the substance. On the other side of the specimen, heat is being drained by a cold reservoir, keeping the right edge at some temperature below the fusion point.

The heat from the hot reservoir will flow to the interface at constant rate $k_1(T_l - T_f) / \xi$, and at the same time, heat will be removed from the interface by the cold reservoir at a constant rate $k_2(T_f - T_s) / (L - \xi)$.

Therefore the net flux through the interface, determined by these two quantities, will dictate how much solid melts or how much liquid is solidified. At some time, which depends on the diffusivities at each phase and the latent heat of fusion, the net flux through the interface will be equal to zero. When the interface reaches a specific position, where the net flux is zero, there is no energy left to sustain the phase transition, and the liquid-solid interface will stop moving. The exact value of the position for the interface, where this happens, can be obtained through this analysis; therefore, this position is given by

$$\xi_{lim} = \frac{k_1(T_l - T_f)L}{k_2(T_f - T_s) + k_1(T_l - T_f)}, \quad (17)$$

which will happen at some time $t \gg 0$, depending on the starting position of the interface. According to this argument, any solution for the interface position, must approach asymptotically to this value. This limit is shown in figure 1, and is compared with the value obtained by using the NC-FDS and HBIM. During the numerical simulations it was observed that the interface motion practically stopped at $t_{max} = 2$, so the exact value given by equation (17) is compared with the corresponding numerical solutions, as shown in figure 1.

For large values of t , the net flux through any point x within the specimen is also zero. We can use the flux equation at any position, and obtain an asymptotic value for the temperature T_{lim} at that position as well. By setting the incoming and outgoing flux equal to each other at a given position x , the temperature of a point within the liquid and solid phase for large values of t is given by

$$T_{1_{lim}}(x) = T_l \left(\frac{\xi_{lim} - x}{\xi_{lim}} \right) + T_f \frac{x}{\xi_{lim}} \text{ and } T_{2_{lim}}(x) = T_f \left(\frac{L - x}{L - \xi_{lim}} \right) + T_s \left(\frac{x - \xi_{lim}}{L - \xi_{lim}} \right), \quad (18)$$

respectively. Here ξ_{lim} is given by equation (17), L is the length of the specimen and x is the position of any point where the asymptotic value for the temperature is to be determined. In figure 1, the asymptotic temperature at $x = 0.4$ is shown as well, and compared with the temperatures obtained from the NC-FDS and HBIM at $t_{max} = 2$.

The predicting power of equation (17) also lies in the fact, that we are able to calculate the amount of liquid or solid that will remain in the specimen, independently of the initial position of the interface. In figure 2, the interface is initially placed to the right side of the limiting position given by equation (17). In this case, the liquid close to the interface will be transformed into solid phase, approaching the asymptotic value predicted by equation (17). The temperatures at the boundaries and thermodynamic variables of latent heat and diffusivities from the first example were used in this example as well, only that the interface is intentionally placed at $\xi(0) = 0.80$, so it moves to the left, as predicted by equation (17). All this is shown in figure 2, where it is also evident that the behavior of the interface motion is not parabolic as it approaches the asymptotic position.

A fine mesh with $N_1 = 81$ and $N_2 = 41$ was used, for mediums 1 and 2 respectively. The whole time interval studied was $t_{max} = 2.0$, and $N_t = 8 \times 10^4$ time partitions were used as in the previous example. As expected, the NC-FDS and HBIM capture the motion of the interface with a small observable difference between them. Also in this case, both solutions approach asymptotically to the limiting value, which according to equation (17) is: $\xi_{lim} = 0.65217391$. Figure 2, also shows the temperature history at $x = 0.70$, where both solutions approach the asymptotic value for the temperature at this position $T_{lim} = -0.055$, according to equation (18).

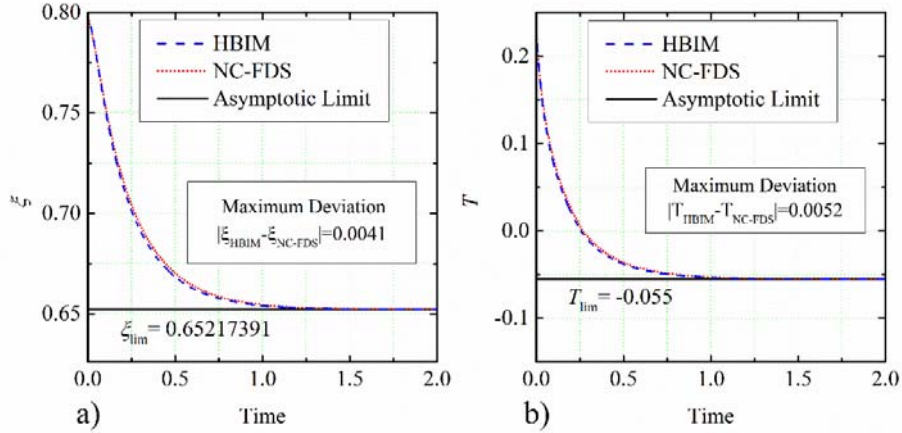


Fig. 2. a) Interface movement and b) temperature history at $x = 0.70$.

5.2 Interface motion in Aluminum

In this part of the results we will discuss the consequences of equations (17)-(18) for the phase transition in pure Aluminum. The thermodynamic variables are obtained from [7], and assumed to be constant. For this part of the discussion, we use the SC as shown in equation (4), and the diffusion equations must take into account the density and specific heat capacity of each phase in the diffusivities $\alpha_1 = k_1 / \rho_1 C_1$ and $\alpha_2 = k_2 / \rho_2 C_2$. The thermodynamic variables taken from [7] are: $\rho_1 = 2380 \text{ kg/m}^3$, $\rho_2 = 2545 \text{ kg/m}^3$, $C_1 = 1130 \text{ J} \cdot \text{kg}^{-1} \text{ K}^{-1}$, $C_2 = 1016 \text{ J} \cdot \text{kg}^{-1} \text{ K}^{-1}$, $k_1 = 215 \text{ W} \cdot \text{m}^{-1} \text{ K}^{-1}$, $k_2 = 225.5 \text{ W} \cdot \text{m}^{-1} \text{ K}^{-1}$, $L_f = 396 \times 10^3 \text{ J/kg}$, and we use same temperatures at the boundaries; $T_l = 1073 \text{ K}$ and $T_s = 573 \text{ K}$. As in section 5.1, we will consider two cases; a) one, where the interface will be placed initially at $\xi(0 \text{ s}) = 0.10 \text{ m}$, and b) when $\xi(0 \text{ s}) = 0.90 \text{ m}$.

In figure 3, we show the interface position as a function of time; obtained from the NC-FDS and HBIM, by assuming initially, a temperature profile of the form discussed in section 4. In case a), the interface moves to the right of its initial position according to equation (17), so in order to obtain a physically viable solution, we must use the density of the solid ρ_2 in the SC, equation (4). In case b), we must use the density of the liquid phase ρ_1 in the SC, since formation of solid is expected according to equation (17). Substituting the thermal conductivities of Aluminum and temperatures at the edges of the specimen in equation (17), we can predict the asymptotic position of the interface ($\xi_{lim} = 0.26947 \text{ m}$). In order to obtain the proper behavior with the finite difference method, we found that a second order approximation in the space variable, overestimated the predicted value, and we developed a NC-FDS in order to approach asymptotically to the expected value. Even though, the NC-FDS uses a fourth order approximation, a very fine mesh was needed. In the example shown in figure 3, we used a mesh with $N_1 = 160$ and $N_2 = 360$ nodes in a) and a mesh with $N_1 = 360$ and $N_2 = 160$ nodes in b). The time step was $\Delta t = 1/3 \text{ s}$ for $t_{max} = 50 \times 10^3 \text{ s}$ of simulation.

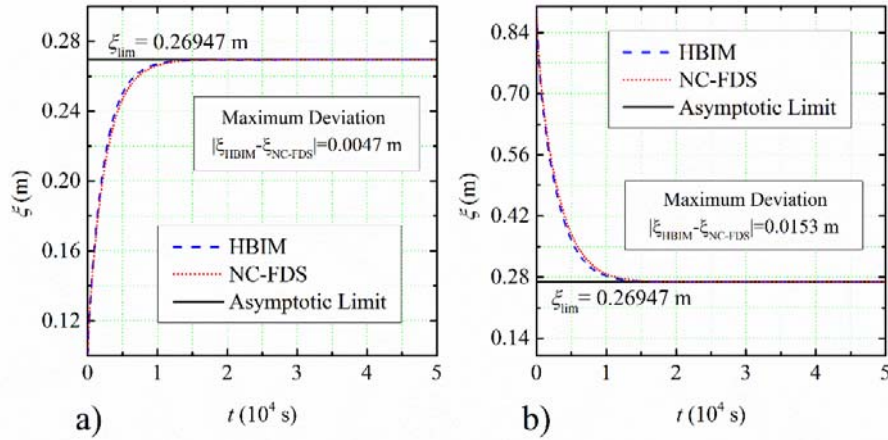


Fig.3. Interface motion for Aluminum.

By using equation (18), we can also predict the temperature profile within the specimen in this limit. Equation (18) predicts different linear temperature profile within the liquid and solid at large time values. These asymptotic profiles only depend on the boundary conditions, fusion temperature, specimen's size and thermal conductivities. Since they must not depend on the initial interface position and initial temperature profile, we also test this prediction in figure 4, where the time evolution of the profile within the specimen is shown for each case. The solution was obtained with the NC-FDS, and compared with the exact value in the asymptotic limit. We also use two different initial profiles in each case, in order to illustrate the generality of the result predicted by equation (18). The time evolution of the profile is shown for an initial parabolic and step function-like profiles, reaching the same asymptotic temperature in both situations, independently of the initial profile or interface position.

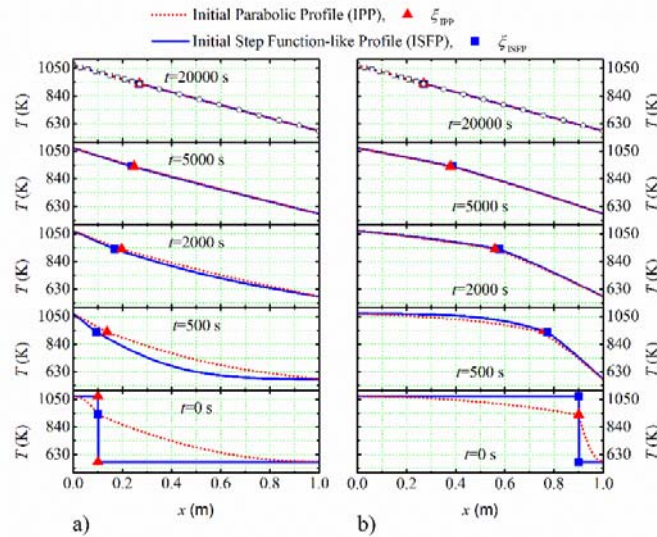


Fig. 4. Time evolution of two different temperature profiles in Aluminum obtained with the NC-FDS. These solutions correspond to the boundary conditions and initial interface positions used in figure 3. White squares and white circles belong to the exact solution predicted by equation (18)

6. Conclusions

Under homogeneous Dirichlet boundary conditions applied over a one dimensional sample with a liquid-solid phase transition taking place, we have found several results that, to the best of our knowledge, are not mentioned anywhere in the literature:

- Non parabolic motion of the interface could be explained by using heat transport theory,
- The nature of the boundary conditions imposed on the specimen, imply an asymptotic behavior that can be predicted, and enables to find the amount of liquid and solid that will remain on the sample,
- The net flux of energy through the interface will determine its position at large time values, according to equation (17), independently of the initial amount of liquid or solid,
- For large time values, the temperature profile in each phase is exactly linear according to equation (18),
- Equation (18), predicts an asymptotic temperature profile within the liquid and solid phase, that is independent of the initial profile and initial position of the interface,
- The NC-FDS and HBIM capture the physics predicted by equations (17)-(18),
- Given that a second order finite difference scheme overestimates the position of the interface at large time values, a new NC-FDS was developed, in order to obtain the proper asymptotic behavior,
- The general results presented in this work provide a deeper understanding of heat transport phenomena in pure substances, and experiments ought to be planned in order to validate these results.

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