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Existence and uniqueness of a classical solution for the coupled heat and mass transfer during the freezing of high-water content materials

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A recent model for the coupled problem of heat and mass transfer during the solidification of high-water content materials like soils, foods, tissues and phase-change materials was developed. This model takes into account the role played by material properties and process variables on the advance of freezing and sublimation fronts, temperature and water vapour profiles and weight loss. The goal of this paper is to determine the existence of a unique local classical solution for the corresponding two-phase coupled free boundary problem in an adequate functional space. Copyright © 2011 John Wiley & Sons, Ltd.

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1. Introduction

Ice sublimation takes place from the surface of high water-content systems like moist soils, aqueous solutions, vegetable or animal tissues and foods that freeze. The rate of both phenomena (solidification and sublimation) is determined by material characteristics and cooling conditions, and the sublimation process determines fundamental features of the final quality for foods and influences the structure and usability of frozen tissues. Modelling of these simultaneous processes is very difficult because of the coupling of the heat and mass transfer balances and the existence of two moving phase-change fronts that advance at very different rates [1].

The process with only-solidification (and no sublimation) has been extensively studied [2–4]. The system has been studied both by analytical procedures [3, 5], and with numerical methods [2, 3, 6]. The process with only-sublimation of the already-frozen system has been extensively studied for freeze-drying of food and pharmaceutical materials [7, 8].

In the case of freezing with simultaneous ice sublimation, published developments are scarce and no analytical solution to the coupled problem has been developed except in [1]. Ice sublimation has been surveyed by several authors in different systems [7, 9–11]. A large bibliography on free and moving boundary problems for the heat diffusion equation is given in [12].

When high water-content materials (like foods, tissues, gels, soils, water solutions of inorganic or organic substances, held in open, permeable or loosely sealed containers) are refrigerated to below their initial solidification temperature, two simultaneous physical phenomena take place: liquid water solidifies (freeze) and surface ice sublimates. The rate and extent of these transfers is determined by different factors [1]. For the description of the freezing process, the material can be divided into three zones: unfrozen, frozen and dehydrated.

Freezing begins from the refrigerated surface. Simultaneously, ice sublimation begins at the frozen surface and a dehydration front penetrates the material, whose rate of advance is again determined by the characteristics of the material and environmental conditions. Normally, this rate is much lower than that of the freezing front [13]. A complete mathematical model has to solve both, the heat transfer (freezing) and the mass transfer (weight loss) simultaneously. Normally, uniform initial temperature T_{if} and composition are

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supposed, and convective boundary conditions at the surface for both heat and mass transfer are the usual modelling assumptions. Phase change is accounted for in the following way:

- Solidification (freezing) as a freezing front ($x = s_f(t)$) located at the point where material temperature reaches the initial freezing temperature (T_{if}), determined by material composition. For temperatures lower than T_{if} (the zone nearer to the refrigerated surface) the amount of ice formed is determined by an equilibrium line (ice content versus temperature and water content) specific to the material.
- On the dehydration front ($x = s_d(t)$) we impose Stefan-like conditions for temperature distribution and vapour concentration.

Therefore, we consider a semi-infinite material with characteristics similar to a very diluted gel (whose properties can be supposed equal to those of pure water). The system has an initial uniform temperature equal to T_{if} and an uncovered flat surface, which at time t=0 is exposed to the surrounding medium (with constant temperature T_s (lower than T_{if}) and heat and mass transfer coefficients h and K_m). We assume that $T_s < T_0(t) < T_{\mathrm{if}}$, $t > 0$ where $T_0(t)$ is the unknown sublimation temperature. In this paper the temperatures are measured in Kelvin. To calculate the evolution of temperature and water content in time, in [1] the following nonstandard, coupled two-phase free boundary problem is taken into account: Find the temperatures $T_\mathrm{d} = T_\mathrm{d}(x,t)$ (of the dehydrated region) and $T_\mathrm{f} = T_\mathrm{f}(x,t)$ (of the frozen region), the vapour concentration $C_\mathrm{v} = C_\mathrm{v}(x,t)$ (of the dehydrated region), the two free boundaries $x = s_\mathrm{d}(t)$ (sublimation front) and $x = s_\mathrm{f}(t)$ (frozen front), and the temperature $T_0 = T_0(t)$ at $x = s_\mathrm{d}(t)$, which satisfy the following differential equations and conditions:

$$\rho_\mathrm{d} c_\mathrm{d} \frac{\partial T_\mathrm{d}}{\partial t} = k_\mathrm{d} \frac{\partial^2 T_\mathrm{d}}{\partial x^2} \quad \text{in} \quad Q_{1T} \equiv \{(x,t) : 0 < x < s_\mathrm{d}(t), \quad 0 < t < T\}, \tag{1}$$

$$\varepsilon \frac{\partial C_\mathrm{v}}{\partial t} = D \frac{\partial^2 C_\mathrm{v}}{\partial x^2} \quad \text{in} \quad Q_{1T}, \tag{2}$$

$$\rho_\mathrm{f} c_\mathrm{f} \frac{\partial T_\mathrm{f}}{\partial t} = k_\mathrm{f} \frac{\partial^2 T_\mathrm{f}}{\partial x^2} \quad \text{in} \quad Q_{2T} \equiv \{(x,t) : s_\mathrm{d}(t) < x < s_\mathrm{f}(t), \quad 0 < t < T\}, \tag{3}$$

$$k_\mathrm{d} \frac{\partial T_\mathrm{d}}{\partial x}(0,t) = h_0 \left[T_\mathrm{d}(0,t) - T_\mathrm{s}\right] \quad \text{on} \quad x = 0, \quad 0 < t < T, \tag{4}$$

$$D \frac{\partial C_\mathrm{v}}{\partial x}(0,t) = k_\mathrm{m} \left[C_\mathrm{v}(0,t) - C_\mathrm{a}\right] \quad \text{on} \quad x = 0, \quad 0 < t < T, \tag{5}$$

$$T_\mathrm{d}(s_\mathrm{d}(t), t) = T_\mathrm{f}(s_\mathrm{d}(t), t) = T_0(t) \quad \text{on} \quad x = s_\mathrm{d}(t), \quad 0 < t < T, \tag{6}$$

$$k_\mathrm{f} \frac{\partial T_\mathrm{f}}{\partial x}(s_\mathrm{d}(t), t) - k_\mathrm{d} \frac{\partial T_\mathrm{d}}{\partial x}(s_\mathrm{d}(t), t) = L_\mathrm{s} m_\mathrm{s} \frac{\mathrm{d}s_\mathrm{d}}{\mathrm{d}t}(t) \quad \text{on} \quad x = s_\mathrm{d}(t), \quad 0 < t < T, \tag{7}$$

$$D \frac{\partial C_v}{\partial x}(s_\mathrm{d}(t), t) = m_\mathrm{s} \frac{\mathrm{d}s_\mathrm{d}}{\mathrm{d}t}(t) \quad \text{on} \quad x = s_\mathrm{d}(t), \quad 0 < t < T, \tag{8}$$

$$C_\mathrm{v}(s_\mathrm{d}(t), t) = F(T_0(t)) \quad \text{on} \quad x = s_\mathrm{d}(t), \quad 0 < t < T, \tag{9}$$

$$T_\mathrm{f}(s_\mathrm{f}(t), t) = T_\mathrm{if} \quad \text{on} \quad x = s_\mathrm{f}(t), \quad 0 < t < T, \tag{10}$$

$$k_\mathrm{f} \frac{\partial T_\mathrm{f}}{\partial x}(s_\mathrm{f}(t), t) = m_\mathrm{f} L_\mathrm{f} \frac{\mathrm{d}s_\mathrm{f}}{\mathrm{d}t}(t) \quad \text{on} \quad x = s_\mathrm{f}(t), \quad 0 < t < T, \tag{11}$$

$$s_\mathrm{d}(0) = s_{0\mathrm{d}}, \quad s_\mathrm{f}(0) = s_{0\mathrm{f}} \quad , \tag{12}$$

where all constants are defined in the nomenclature of [1] and we assume

$$H_1 \begin{cases} \rho_\mathrm{d}, c_\mathrm{d}, k_\mathrm{d}, \varepsilon, D, \rho_\mathrm{f}, c_\mathrm{f}, k_\mathrm{f}, h_0, k_\mathrm{m}, L_\mathrm{s}, M_\mathrm{s}, T_\mathrm{if}, C_\mathrm{a}, T_\mathrm{s} \text{ are positive constants} \\ T_\mathrm{s} < T_\mathrm{if}, \quad s_{0\mathrm{f}} > s_{0\mathrm{d}} > 0. \end{cases}$$

System (1)–(12) must be completed with suitable initial data for T_d, T_f and C_v. Hence, we add the following three extra conditions:

$$T_\mathrm{d}(x,0) = T_{0\mathrm{d}}(x), \quad 0 \le x \le s_{0\mathrm{d}}, \tag{13}$$

$$C_\mathrm{v}(x,0) = C_{0\mathrm{v}}(x), \quad 0 \le x \le s_{0\mathrm{d}}, \tag{14}$$

$$T_\mathrm{f}(x,0) = T_{0\mathrm{f}}(x), \quad s_{0\mathrm{d}} \le x \le s_{0\mathrm{f}}. \tag{15}$$

On the function $F(\eta)$ we stipulate the following assumption:

$$H_2 : F(\eta) \in C^3(\mathbb{R}).$$

In fact, condition (9) is a generalization condition of the one given in [1] because in the physical case we have

$$C_v(s_d(t), t) = \frac{M P_{sat}(T)}{R_g T_0(t)} = M a \frac{\exp\left(b - \frac{c}{T_0(t)}\right)}{R_g T_0(t)}$$

where $C_v(s_d(t), t)$ is the equilibrium vapour concentration at $T_0(t)$, and the saturation pressure $P_{sat}(T)$ is evaluated according to [14], that is

$$F(\eta) := \frac{M a e^{b - \frac{c}{\eta}}}{R\eta},$$

where b, M, a, R, c are constant and c is positive.

In [1], the quasi-steady heat conduction in the frozen region is assumed and system (1)–(15) is thus reduced to a set of coupled ordinary differential equations for the free boundaries $x = s_d(t)$ and $x = s_f(t)$ and the temperature $T_0 = T_0(t)$. These three values are then used to predict the temperatures $T_d(x,t)$ and $T_f(x,t)$, and the concentration C_v.

The goal of this paper is to determine in Section 2 the existence and the uniqueness of a local classical solution of the two-phase coupled free boundary problem (1)–(15) in an adequate functional space. We use the cornerstone work in [15]; other references on the subject are in [16–19].

2. Local existence and uniqueness of a classical solution

System (1)–(15) is equivalent to a new one in which the free boundary condition (8) is replaced by

$$k_f \frac{\partial T_f}{\partial x}(s_d(t), t) - k_d \frac{\partial T_d}{\partial x}(s_d(t), t) = \beta \frac{\partial C_v}{\partial x}(s_d(t), t) \text{ on } x = s_d(t),\ 0 < t < T \tag{8bis}$$

where $\beta = L_s D$; and the free boundary condition (9) is replaced by

$$C_v(s_d(t), t) = F(T_d(s_d(t), t)) \quad \text{on} \quad x = s_d(t), \quad 0 < t < T \quad . \tag{9bis}$$

We now want to rewrite system (1)–(7), (8bis), (9bis), (10)–(15) in a more convenient form. That is we want to write down a free boundary problem for a system of parabolic equations all satisfied in the same cylindrical domain. To this purpose we introduce the following change of coordinates:

$$\begin{cases} t = t \\ y = \dfrac{x}{Ax + B} \end{cases} \tag{16a}$$

and

$$\begin{cases} \theta_d(y,t) = T_d(x,t) = T_d\left(\dfrac{B(t)y}{1 - A(t)y}, t\right), \\ \theta_f(y,t) = T_f(x,t) = T_f\left(\dfrac{B(t)y}{1 - A(t)y}, t\right), \\ W(y,t) = C_v(x,t) = C_v\left(\dfrac{B(t)y}{1 - A(t)y}, t\right), \end{cases} \tag{16b}$$

where

$$A = A(t) = \frac{2s_d(t) - s_f(t)}{2(s_d(t) - s_f(t))}, \qquad B = B(t) = \frac{s_d(t)\, s_f(t)}{2(s_f(t) - s_d(t))}. \tag{16c}$$

It reduces our problem to the following 'rectified' form (where the $A'(t) = (dA/dt)(t)$ and $B'(t) = (dB/dt)(t)$):

$$\rho_d c_d \frac{\partial \theta_d}{\partial t}(y,t) = \frac{k_d (1 - Ay)^4}{B^2} \frac{\partial^2 \theta_d}{\partial y^2} + \frac{\partial \theta_d}{\partial y}\left[\frac{\rho_d c_d y}{B}\left(B' + y(BA' - AB')\right) - 2k_d \frac{(1 - Ay)^3 A}{B^2}\right] \text{ in } \Omega_{1T} = [0,1] \times [0,T], \tag{17}$$

$$\rho_f c_f \frac{\partial \theta_f}{\partial t}(y,t) = \frac{k_f (1-Ay)^4}{B^2} \frac{\partial^2 \theta_f}{\partial y^2} + \frac{\partial \theta_f}{\partial y}\left[\frac{\rho_f c_f y}{B}\left(B' + y(BA' - AB')\right) - 2k_f \frac{(1-Ay)^3 A}{B^2}\right] \text{ in } \Omega_{2T} = [1,2]\times[0,T], \tag{18}$$

$$\varepsilon \frac{\partial W}{\partial t}(y,t) = D\frac{(1-Ay)^4}{B^2}\frac{\partial^2 W}{\partial y^2} + \frac{\partial W}{\partial y}\left[\frac{\varepsilon y}{B}\left(B' + y(BA' - AB')\right) - 2D\frac{(1-Ay)^3}{B^2}A\right] \text{ in } \Omega_{1T}, \tag{19}$$

$$k_d \frac{\partial \theta_d}{\partial y}(0,t)\frac{1}{B} = h_0\left[\theta_d(0,t) - T_s\right], \quad 0<t<T, \tag{20}$$

$$D\frac{\partial W}{\partial y}(0,t)\frac{1}{B} = k_m\left[W(0,t) - C_a\right], \quad 0<t<T, \tag{21}$$

$$\theta_d(1,t) = \theta_f(1,t) = T_0(t), \quad 0<t<T, \tag{22}$$

$$k_f \frac{\partial \theta_f}{\partial y}(1,t) - k_d\frac{\partial \theta_d}{\partial y}(1,t) = \beta\frac{\partial W}{\partial y}(1,t), \quad 0<t<T, \tag{23}$$

$$W(1,t) = F(\theta_d(1,t)), \quad 0<t<T, \tag{24}$$

$$\theta_f(2,t) = T_{if}, \quad 0<t<T, \tag{25}$$

$$L_s m_s \frac{\mathrm{d}s_d}{\mathrm{d}t}(t) = \frac{(1-A)^2}{B}\left[k_f\frac{\partial \theta_f}{\partial y}(1,t) - k_d\frac{\partial \theta_d}{\partial y}(1,t)\right], \quad 0<t<T, \tag{26}$$

$$L_f m_f \frac{\mathrm{d}s_f}{\mathrm{d}t}(t) = \frac{(1-2A)^2}{B}k_f\frac{\partial \theta_f}{\partial y}(2,t), \quad 0<t<T, \tag{27}$$

$$s_d(0) = s_{0d}, \quad s_f(0) = s_{0f}, \tag{28}$$

$$\theta_d(y,0) = \theta_{0d}(y), \quad 0\le y\le 1, \tag{29}$$

$$\theta_f(y,0) = \theta_{0f}(y), \quad 1\le y\le 2, \tag{30}$$

$$W(y,0) = W_0(y), \quad 0\le y\le 1, \tag{31}$$

where

$$\theta_{0d}(y) = T_{0d}\left(\frac{s_{0d}s_{0f}y}{(2s_{0d}-s_{0f})y + 2(s_{0f}-s_{0d})}\right)$$
$$\theta_{0f}(y) = T_{0f}\left(\frac{s_{0d}s_{0f}y}{(2s_{0d}-s_{0f})y + 2(s_{0f}-s_{0d})}\right)$$
$$W_0(y) = C_{0v}\left(\frac{s_{0d}s_{0f}y}{(2s_{0d}-s_{0f})y + 2(s_{0f}-s_{0d})}\right).$$

Note that θ_{0d}, θ_{0f} and W_0 have the same regularity of T_{0d}, T_{0f} and C_{0v}.

Finally, to have partial differential equations that are all defined in the same domain Ω_{1T}, we introduce new unknowns

$$u_1(z,t) = \theta_d(z,t), \quad u_2(z,t) = \theta_f(2-z,t), \quad u_3(z,t) = W(z,t), \tag{32}$$

thus obtaining the following system of equation, which is equivalent to (1)–(15)

$$\rho_d c_d \frac{\partial u_1}{\partial t} = k_d \frac{(1-Az)^4}{B^2}\frac{\partial^2 u_1}{\partial z^2} + \left[\frac{\partial u_1}{\partial z}\left(\frac{z\rho_d c_d}{B^2}\left(B' + z(BA' - AB')\right) - 2k_d\frac{(1-Az)^3}{B^2}A\right)\right] \equiv \mathcal{A}_1\frac{\partial^2 u_1}{\partial z^2} + \mathcal{B}_1\frac{\partial u_1}{\partial z} \text{ in } \Omega_{1_T}, \tag{33}$$

$$\rho_f c_f \frac{\partial u_2}{\partial t} = k_f \frac{(1-A(2-z))^4}{B^2}\frac{\partial^2 u_2}{\partial z^2} - \left[\frac{\partial u_2}{\partial z}\left(\frac{\rho_f c_g (2-z)}{B}\left(B' + (2-z)(BA' - AB')\right) - 2k_f\frac{(1-A(2-z))^3}{B^2}A\right)\right] \equiv \mathcal{A}_2\frac{\partial^2 u_2}{\partial z^2} + \mathcal{B}_2\frac{\partial u_2}{\partial z} \text{ in } \Omega_{1_T}, \tag{34}$$

$$\varepsilon \frac{\partial u_3}{\partial t} = D \frac{(1-Az)^4}{B^2}\frac{\partial^2 u_3}{\partial z^2} + \left[\frac{\partial u_3}{\partial z}\left(\frac{\varepsilon z}{B}\left(B' + z(BA' - AB')\right) - 2D\frac{(1-Az)^3}{B^2}A\right)\right] \equiv \mathcal{A}_3\frac{\partial^2 u_3}{\partial z^2} + \mathcal{B}_3\frac{\partial u_3}{\partial z} \text{ in } \Omega_{1T}, \tag{35}$$

$$k_d \frac{\partial u_1}{\partial z}(0,t) = B(t)\, h_0 \left[u_1(0,t) - T_s\right], \quad 0 < t < T, \tag{36}$$

$$D\frac{\partial u_3}{\partial z}(0,t) = B(t)\, k_m \left[u_3(0,t) - C_a\right], \quad 0 < t < T, \tag{37}$$

$$u_2(0,t) = T_{if}, \quad 0 < t < T, \tag{38}$$

$$u_1(1,t) = u_2(1,t), \quad 0 < t < T, \tag{39}$$

$$k_f \frac{\partial u_2}{\partial z}(1,t) + k_d\frac{\partial u_1}{\partial z}(1,t) = -\beta\frac{\partial u_3}{\partial z}(1,t), \quad 0 < t < T, \tag{40}$$

$$u_3(1,t) = F(u_1(1,t)), \quad 0 < t < T, \tag{41}$$

$$L_f m_f \frac{ds_f}{dt}(t) = -\left[\frac{(1-2A)^2}{B}k_f\frac{\partial u_2}{\partial z}\right] \equiv -\mathcal{C}\ \frac{\partial u_2}{\partial z}, \quad 0 < t < T, \tag{42}$$

$$L_s m_s \frac{ds_d}{dt}(t) = -\left[\frac{(1-A)^2}{B}\left(k_f\frac{\partial u_2}{\partial z} + k_d\frac{\partial u_1}{\partial z}\right)\right] \equiv -\mathcal{D}\left(k_f\frac{\partial u_2}{\partial z} + k_d\frac{\partial u_1}{\partial z}\right), \quad 0 < t < T, \tag{43}$$

$$s_d(0) = s_{0d}, \quad s_f(0) = s_{0f}, \tag{44}$$

$$u_1(z,0) = \theta_{0d}(z), \quad 0 \le z \le 1, \tag{45}$$

$$u_2(z,0) = \theta_{0f}(2-z), \quad 0 \le z \le 1, \tag{46}$$

$$u_3(z,0) = W_0(z), \quad 0 \le z \le 1. \tag{47}$$

with obvious meaning of the symbols $\mathcal{A}_j$, $\mathcal{B}_j$ $(j = 1,2,3)$, $\mathcal{C}$, and $\mathcal{D}$

Remark 1
Note that the original unknown $T_0(t)$ can be computed by $u_1(1,t)$ or $u_2(1,t)$.

We stipulate the following assumptions:

$$H_3 \begin{cases} T_{0d}, C_{0v} \in H^{2+\alpha}([0, s_{0d}]), & \alpha \in (0,1), \\ T_{0f} \in H^{2+\alpha}([s_{0d}, s_{0f}]), & \alpha \in (0,1), \end{cases}$$

and

H_4 : First-order compatibility conditions are satisfied when Robin type boundary conditions are imposed while second-order compatibility conditions are satisfied if we have Dirichlet boundary conditions.

Problems (33)–(47) are equivalent to problems (1)–(15) in the sense that any 'classical' solution of (33)–(47) is a classical solution of (1)–(15) and the converse is also true. For the system of equations (33)–(47) we will prove the existence of a unique classical solution provided T is chosen sufficiently small. This will be the main result of this paper. Namely we will prove

Theorem 1
Under assumptions H_1–H_4 there exists a time $\widehat{T} > 0$ such that problems (33)–(47) admit a unique classical solution in $\Omega_{1\widehat{T}}$; that is, there exists a quintuple of functions $(u_1(z,t), u_2(z,t), u_3(z,t), s_d(t), s_f(t))$ such that $u_i \in H^{2+\beta}\left(\overline{\Omega}_{1\widehat{T}}\right)$ $(i = 1, 2, 3)$, s_d, $s_f \in H^{1+\beta/2}\left(\left[0, \widehat{T}\right]\right)$, $\forall \beta < \frac{\alpha}{2}$, which satisfy problems (33)–(47).

Proof
We start by proving the existence. To this purpose we need to introduce an auxiliary problem whose solution $(u_1^h, u_2^h, u_3^h, s_d^h, s_f^h)$ will be proved to converge as h (the delay parameter) tends to zero, and for small time t, to a solution of problems (33)–(47).

The advantage of the auxiliary problem lays in the fact that it is easily proved to admit a global solution in time (thanks to an iterated step-by-step procedure and some fundamental results on parabolic systems proved by Solonnikov). This is due to the fact that the delay factor h introduced in (41)–(43) transforms the original problem in a new one, which is 'essentially' linear. Hence, for any positive constant h (the delay), we introduce the following problem for new unknowns $u_1^h, u_2^h, u_3^h, s_d^h, s_f^h$:

$$\rho_d c_d \frac{\partial u_1^h}{\partial t} = k_d\, G\left(\frac{(1-Az)^4}{B^2}\right)\frac{\partial^2 u_1^h}{\partial z^2} + \left[\frac{\partial u_1^h}{\partial z}\left(\frac{z\rho_d c_d}{B^2}\left(B' + z(BA' - AB')\right) - 2k_d \frac{(1-Az)^3}{B^2}A\right)\right] \text{ in } \Omega_{1T}, \tag{48}$$

$$\rho_f c_f \frac{\partial u_2^h}{\partial t} = k_f\, G\left(\frac{(1-A(2-z))^4}{B^2}\right)\frac{\partial^2 u_2^h}{\partial z^2} - \left[\frac{\partial u_2^h}{\partial z}\left(\frac{\rho_f c_g (2-z)}{B}\left(B' + (2-z)(BA' - AB')\right) - 2k_f \frac{(1-A(2-z))^3}{B^2}A\right)\right] \text{ in } \Omega_{1T}, \tag{49}$$

$$\varepsilon \frac{\partial u_3^h}{\partial t} = D\, G\left(\frac{(1-Az)^4}{B^2}\right)\frac{\partial^2 u_3^h}{\partial z^2} + \left[\frac{\partial u_3^h}{\partial z}\left(\frac{\varepsilon z}{B}\left(B' + z(BA' - AB')\right) - 2D \frac{(1-Az)^3}{B^2}A\right)\right] \text{ in } \Omega_{1T}. \tag{50}$$

Conditions (36)–(40) are satisfied by u_i^h, i=1,2,3, $\forall t \in [0,T]$. We will label these conditions (51)–(55). Conditions (45)–(47) are assumed to be satisfied $\forall\,(z,t) \in [0,1] \times [-h, 0]$. We will label these conditions (56)–(58). In addition, we assume the following four boundary conditions:

$$u_3^h(1,t) = F(\theta_{0d}(1)) + F'(\theta_{0d}(1))\left(u_1^h(1,t-h) - \theta_{0d}(1)\right) - \int_{\theta_{0d}(1)}^{u_1^h(1,t-h)} \left(s - u_1^h(1,t-h)\right) F''(s)\, ds, \quad t \in [0,T], \tag{59}$$

$$L_f m_f \frac{ds_f^h}{dt}(t) = -\left[\frac{(1-2A)^2}{B} k_f \frac{\partial u_2^h}{\partial z}\right](0, t-h), \quad t \in [0,T], \tag{60}$$

$$L_s m_s \frac{ds_d^h}{dt}(t) = -\left[\frac{(1-A)^2}{B}\left(k_f\frac{\partial u_2^h}{\partial z} + k_d\frac{\partial u_1^h}{\partial z}\right)\right](1, t-h), \quad t \in [0, T], \tag{61}$$

$$s_d^h(t) = s_{0d}, \quad s_f^h(t) = s_{0f}, \quad \forall t \in [-h, 0]. \tag{62}$$

Note that, if $h \equiv 0$, (59) is equivalent to (41) because the right-hand side of (59) is the Taylor expansion of the right-hand side of (41) centred at $\theta_{0d}(1)$. Also note that the terms in square brackets in (60) and (61) are evaluated at $(z, t-h)$, that is they are subjected to a delay in time. The function $G(\eta)$, appearing in the left-hand side of (48)–(50), is defined as follows:

$$G(\eta) = \begin{cases} \frac{1}{k} & \text{if } \eta \le \frac{1}{k} \\ \eta & \text{if } \frac{1}{k} \le \eta \le k \\ k & \text{if } \eta \ge k \end{cases} \tag{63}$$

with $0 < k < \infty$ defined as follows:

$$k = 2 \max_{[0,1]}\left(\frac{(1-A(0)z)^4}{B(0)^2}, \frac{(1-A(0)(2-z))^4}{B(0)^2}, \frac{B(0)^2}{(1-A(0)z)^4}, \frac{B(0)^2}{(1-A(0)(2-z))^4}\right) > 0.$$

We note that the function $G(\eta)$ is introduced to guarantee the boundedness of the coefficients of the principal parts of the parabolic equations (48)–(50) and their uniform parabolicity.

Problems (48)–(62) can be solved step by step, where any time step has a width h. In fact, if $s_d^h(t)$, $s_f^h(t)$ and $u_i^h(x,t)$ are known in $[(n-1)h, nh]$, then $s_d^h(t)$, $s_f^h(t)$ can be determined in $[nh, (n+1)h,]$ thanks to (60)–(61) and belong to the class $H^{1+\alpha/2+1/2}([nh, (n+1)h,])$. Finally, $u_i^h(z,t)$ can also be found in $[nh, (n+1)h,]$ solving the linear system of equations (48)–(59).

The solvability of the system of equations (48)–(59) in $H^{2+\alpha, 1+\alpha/2}\left(\overline{\Omega}_{1\widehat{T}}\right)$, in turn, is proved using Sobolev's technique as found in [15, Section 10, Chapter VII, Theorem 10.1, p. 616]. Note that determining u_i in $[(n-1)h, nh]$ provides the initial data for problems (48)–(55), (59) in the new time interval $[nh, (n+1)h,]$. The applicability of Theorem 10.1 follows from the fact that the system is parabolic. In fact, following the notation of [15], we see that Definition 4 [15, p. 601] is satisfied with $b = 1, r = 3, s_k = 2, t_k = 0, r_j = 1$ while the complementary condition for the initial data, as found in [15, p. 614], is straightforwardly satisfied with $\rho_\alpha \equiv 0$. The previous two conditions are easily proved to be satisfied thanks to the fact that the parabolic equations are uncoupled. The coupling, in fact, is realized only through the boundary conditions at $x = 1$. For this reason the complementary condition of [15, p. 611] is much more difficult to be verified; nevertheless, it holds true, taking $(G_1, G_2, G_3) \equiv (0, 0, 1)$, ($G_3$ is associated to boundary condition (55) and G_1, G_2 to boundary conditions (54), (59)). Using [15, Theorem 10.1, p. 616] and iterating the previous procedure step by step we prove (in a finite number of steps) the solvability of the system (48)–(62) in the afore quoted function spaces.

We now set

$$M(T) \equiv \left(1 + \sum_{i=1}^{3}\left|u_i^h(z,t)\right|_{\Omega_T}^{(2+\beta)} + \left|s_d^h\right|_{[0,T]}^{(1+\beta/2)} + \left|s_f^h\right|_{[0,T]}^{(1+\beta/2)}\right), \tag{64}$$

with $\beta < \frac{\alpha}{2}$ and $T < 1$. We remark that, thanks to the results of the Appendix, $M(T)$ is a continuous function of T.

Because problems (48)–(62) are solvable in the whole $[0, T]$ we can apply [15, Theorem 10.1, p. 616] to problems (48)–(59), regarding the function G in the Equations (48)–(50) and the term in square brackets (as well as the integral term in (59)) as known terms. Setting $U^h = (u_1^h, u_2^h, u_3^h)$, $S^h = (s_d^h, s_f^h)$ we get

$$\left|U^h\right|_{\Omega_T}^{(2+\beta)} \le F_1\left(\left|S^h\right|_{[0,T]}^{\left(1+\beta/2\right)}\right)\left[1 + T\left|U^h\right|_{\Omega_T}^{(2+\beta)} + \left(T^{1/2}\left|U^h\right|_{\Omega_T}^{(2+\beta)}\right)^2\right] \tag{65}$$

where F_1 and all the functions F_i used below are positive, increasing functions of their entries, which do not depend on h. To get (65) we have also made use of the fact that the integral on the right-hand side of (59) has the $H^{1+(\beta/2)}$-norm controlled in terms of known quantities and $T\left(|\partial u_1/\partial t|^{(1+(\beta/2))}\right)^2$. On the other hand, using (60) and (61) we get that

$$\left|S^h\right|_{[0,T]}^{\left(1+1/2+\beta/2\right)} \le F_2\left(|S|_{[0,T]}^{(1)}\right)\left[\left|U^h\right|_{\Omega_T}^{(2+\beta)} + 1\right] \tag{66}$$

and hence, using (65)

$$\left|S^h\right|_{[0,T]}^{\left(1+1/2+\beta/2\right)} \le F_3\left(\left|S^h\right|_{[0,T]}^{\left(1+\beta/2\right)}\right)\left[1 + T\left|U^h\right|_{\Omega_T}^{(2+\beta)} + \left(T^{1/2}\left|U^h\right|_{\Omega_T}^{(2+\beta)}\right)^2\right] \tag{67}$$

where F_2 and F_3 are two functions with the same properties of function F_1. Moreover, inserting (66) in (65) we get

$$\begin{aligned}\left|U^h\right|^{(2+\beta)}_{\Omega_T} &\le F_4\left(F_2\left(\left|S^h\right|^{(1)}_{[0,T]}\right)\left[\left|U^h\right|^{(2+\beta)}_{\Omega_T}+1\right]T^{1/2}\right)\left(1+T\left|U^h\right|^{(2+\beta)}_{\Omega_T}+\left(T^{1/2}\left|U^h\right|^{(2+\beta)}_{\Omega_T}\right)^2\right)\\ &\le F_4\left(F_5(M)\,T^{1/2}\right)\left(1+\left(T^{1/2}\left|U^h\right|^{(2+\beta)}_{\Omega_T}\right)^2+T\left|U^h\right|^{(2+\beta)}_{\Omega_T}\right)\\ &\le F_4\left(F_5(M)\,T^{1/2}\right)\left(1+\left(T^{1/2}M\right)^2+TM\right)\le F_6\left(F_5(M)\,T^{1/2}\right)\left(1+TM^2\right)\end{aligned} \tag{68}$$

and (66) implies

$$\left|S^h\right|^{\left(1+\beta/2\right)}_{[0,T]}\le\left|S^h(0)\right|+\left|\frac{\mathrm{d}S^h}{\mathrm{d}t}(0)\right|+3T^{1/2}F_2(M)(1+M), \tag{69}$$

where $\left|S^h(0)\right|+\left|(\mathrm{d}S^h/\mathrm{d}t)(0)\right|$ does not depend on h because it can be computed using the initial data for the free boundaries (62) and the Equations (60) and (61). We will denote this quantity with the symbol L.

Adding (68) and (69) we get

$$M(T)\le F_6\left(F_5(M)\,T^{1/2}\right)\left(1+TM^2\right)+L+3T^{1/2}F_2(M)(1+M). \tag{70}$$

We now define

$$K=2F_6(1)+L+4$$
$$\widehat{T}=\min\left((F_5(K))^{-2},\,M^{-2},\,(F_2(K)(1+K))^{-2}\right).$$

In this regard it is important to remember that all the functions F_i $(i=1,\dots,6)$ are positive, increasing functions that do not depend on h. Hence, the quantities K and $\widehat{T}$ are also independent of h.

We claim that

$$M(T)\le K \text{ in } \Omega_{\widehat{T}}. \tag{71}$$

This last assertion can be easily proved taking into account that $M(0)<K$ and using that $M(T)$ is continuous (this continuity can be obtained using the results of the Appendix, keeping in mind that $u_1(z,t)$, $u_2(z,t)$, $u_3(z,t)\in H^{2+\alpha,1+\alpha/2}(\Omega_T)$ and $s_{\mathrm{d}}(t)$, $s_{\mathrm{f}}(t)\in H^{1+\alpha/2}([0,T])$ with $\alpha>2\beta$).

Inequality (71) implies the existence of a fixed domain $\Omega_{\widehat{T}}$ in which a classical *a priori* estimate (independent of h) holds. In turn, this allows us to pass the limit as h tends to zero (at least passing to subsequences; however, the uniqueness result proved below guarantees the convergence as h tends to zero) thus obtaining a solution of problems (48)–(62) with $h\equiv 0$. On the other hand, choosing, if necessary, a smaller $\widehat{T}$ we can be sure that the argument of the function G, in (48)–(50), is in between $1/k$ and k; hence, $G(\eta)\equiv\eta$. Consequently, problems (48)–(62) are equivalent to (33)–(47). Because problems (33)–(47) are equivalent to (1)–(15) the previous considerations conclude the existence of the proof .

We will prove uniqueness for problems (33)–(47) with (41) replaced by (72) below; as previously explained this problem is equivalent to (1)–(15) (the original one),

$$\begin{aligned}u_3(1,t)&=F(\theta_{0\mathrm{d}}(1))+F'(\theta_{0\mathrm{d}}(1))(u_1(1,t)-\theta_{0\mathrm{d}}(1))\\ &\quad-\int_{\theta_{0\mathrm{d}}(1)}^{u_1(1,t)}(s-u_1(1,t))F''(s)\,\mathrm{d}s,\quad t\in[0,T],\end{aligned} \tag{72}$$

To this purpose we assume that two different solutions of problems (33)–(40), (72) and (42)–(47) exist and do not coincide in any initial time interval. We write down the system of equations satisfied by their difference,

$$\begin{aligned}&(U_1(z,t),\,U_2(z,t),\,U_3(z,t),\,S_{\mathrm{d}}(t),\,S_{\mathrm{f}}(t))\\ &=\left(u_1^2(z,t)-u_1^1(z,t),\,u_2^2(z,t)-u_2^1(z,t),\,u_3^2(z,t)-u_3^1(z,t),\,s_{\mathrm{d}}^2(t)-s_{\mathrm{d}}^1(t),\,s_{\mathrm{f}}^2(t)-s_{\mathrm{f}}^1(t)\right)(t)\end{aligned}$$

Denoting A_i, B_i, $\mathcal{A}^i_j$, $\mathcal{B}^i_j$ $\mathcal{C}^i$, $\mathcal{D}^i$ the functions A, B, $\mathcal{A}_j$, $\mathcal{B}_j$, $\mathcal{C}$, $\mathcal{D}$ evaluated at $s^i_{\mathrm{d}}(t)$ and $s^i_{\mathrm{f}}(t)$, we get

$$\begin{aligned}\rho_{\mathrm{d}}c_{\mathrm{d}}\frac{\partial U_1}{\partial t}&=k_{\mathrm{d}}\frac{(1-A_1z)^4}{(B_1)^2}\frac{\partial^2U_1}{\partial z^2}\\ &\quad+\left[\frac{\partial U_1}{\partial z}\left(\frac{z\rho_{\mathrm{d}}c_{\mathrm{d}}}{B_1^2}\left(B_1'+z\left(B_1A_1'-A_1B_1'\right)\right)-2k_{\mathrm{d}}\frac{(1-A_1z)^3}{B_1^2}A_1\right)\right]+R_1(z,t)\text{ in }\Omega_{1T},\end{aligned} \tag{73}$$

$$\rho_f c_f \frac{\partial U_2}{\partial t} = k_f \frac{(1-A_1(2-z))^4}{B_1^2}\frac{\partial^2 U_2}{\partial z^2} - \left[\frac{\partial U_2}{\partial z}\left(\frac{\rho_f c_g (2-z)}{B_1}\left(B_1' + (2-z)\left(B_1A_1' - A_1B_1'\right)\right) - 2k_f\frac{(1-A_1(2-z))^3}{B_1^2}A_1\right)\right] + R_2(z,t) \quad \text{in } \Omega_{1_T}, \tag{74}$$

$$\varepsilon\frac{\partial U_3}{\partial t} = D\frac{(1-A_1z)^4}{B_1^2}\frac{\partial^2 U_3}{\partial z^2} + \left[\frac{\partial U_3}{\partial z}\left(\frac{\varepsilon z}{B_1}\left(B_1' + z\left(B_1A_1' - A_1B_1'\right)\right) - 2D\frac{(1-A_1z)^3}{B_1^2}A_1\right)\right] + R_3(z,t) \quad \text{in } \Omega_{1T}, \tag{75}$$

$$k_d\frac{\partial U_1}{\partial z}(0,t) = B_1(t)\,h_0U_1(0,t) + H_1(t), \quad 0<t<T, \tag{76}$$

$$D\frac{\partial U_3}{\partial z}(0,t) = B_1(t)\,k_mU_3(0,t) + H_3(t), \quad 0<t<T, \tag{77}$$

$$U_2(0,t) = 0, \quad 0<t<T, \tag{78}$$

$$U_1(1,t) = U_2(1,t), \quad 0<t<T, \tag{79}$$

$$k_f\frac{\partial U_2}{\partial z}(1,t) + k_d\frac{\partial U_1}{\partial z}(1,t) = -\beta\frac{\partial U_3}{\partial z}(1,t), \quad 0<t<T, \tag{80}$$

$$U_3(1,t) - F'\left(u_1^1(1,t)\right)U_1(1,t) = -\int_{u_1^1(1,t)}^{u_1^2(1,t)}\left(s - u_1^2(1,t)\right)F''(s)\,ds \equiv I(t), \quad t\in[0,T], \tag{81}$$

$$L_fm_f\frac{dS_f}{dt}(t) = -\left[\frac{(1-2A_1)^2}{B_1}k_f\frac{\partial U_2}{\partial z}\right] + J_f(t), \qquad t\in[0,T], \tag{82}$$

$$L_sm_s\frac{dS_d}{dt}(t) = -\left[\frac{(1-A_1)^2}{B_1}\left(k_f\frac{\partial U_2}{\partial z} + k_d\frac{\partial U_1}{\partial z}\right)\right] + J_d(t), \quad t\in[0,T], \tag{83}$$

$$S_d(0) = 0, \quad S_f(0) = 0, \tag{84}$$

$$U_1(z,0) = 0, \quad 0\le z\le 1, \tag{85}$$

$$U_2(z,0) = 0, \quad 0\le z\le 1, \tag{86}$$

$$U_3(z,0) = 0, \quad 0\le z\le 1, \tag{87}$$

where

$$R_j = u_{jzz}^2\left(\mathcal{A}_j^2 - \mathcal{A}_j^1\right) + u_{jz}^2\left(\mathcal{B}_j^2 - \mathcal{B}_j^1\right)$$
$$J_f = -u_{2z}^2\left(\mathcal{C}^2 - \mathcal{C}^1\right)$$
$$J_d = -\left(k_fu_{2z}^2 + k_du_{1z}^2\right)\left(\mathcal{D}^2 - \mathcal{D}^1\right)$$
$$H_1 = h_0[u_1^2(0,t) - T_s]\,(\mathcal{B}_2 - \mathcal{B}_1)$$
$$H_3 = k_m[u_3^2(0,t) - C_a]\,(\mathcal{B}_2 - \mathcal{B}_1)\,.$$

Using (84)–(87) it is easy to prove that (and this is the key point of the uniqueness proof)

$$\begin{aligned}
&|R_j|^{(\beta)}_{\Omega_T}, |H_1|^{\left(\frac{1+\beta}{2}\right)}_{[0,T]}, |H_3|^{\left(\frac{1+\beta}{2}\right)}_{[0,T]}, \ |J_{\rm f}|^{(\beta/2)}_{[0,T]}, \ |J_{\rm d}|^{(\beta/2)}_{[0,T]}, \ |I|^{(1+\beta/2)}_{[0,T]} \\
&\leq C_0T^r\left(\sum_{i=1}^{2}\left(\sum_{j=1}^{3}\left|u^i_j(z,t)\right|^{(2+\beta)}_{\Omega_T}+\left|s^i_{\rm d}\right|^{(1+\beta/2)}_{[0,T]}+\left|s^i_{\rm f}\right|^{(1+\beta/2)}_{[0,T]}\right)\right)\left(|U_1|^{(2+\beta)}_{\Omega_T}+|S_{\rm d}|^{\left(1+\frac{\beta}{2}\right)}_{[0,T]}+|S_{\rm f}|^{\left(1+\frac{\beta}{2}\right)}_{[0,T]}\right) \\
&\leq 2KC_0T^r\left(|U_1|^{(2+\beta)}_{\Omega_T}+|S_{\rm d}|^{\left(1+\frac{\beta}{2}\right)}_{[0,T]}+|S_{\rm f}|^{\left(1+\frac{\beta}{2}\right)}_{[0,T]}\right)
\end{aligned} \tag{88}$$

for a suitable positive r and K as in (71).

Applying [15, Section 10, Chapter VII, Theorem 10.1, p. 616], using (88) we get

$$\left(\sum_{j=1}^{3}|U_j|^{(2+\beta)}_{\Omega_T}+|S_{\rm d}|^{(1+\beta/2)}_{[0,T]}+|S_{\rm f}|^{(1+\beta/2)}_{[0,T]}\right)\leq C_1T^r\left(|U_1|^{(2+\beta)}_{\Omega_T}+|S_{\rm d}|^{(1+\beta/2)}_{[0,T]}+|S_{\rm f}|^{(1+\beta/2)}_{[0,T]}\right). \tag{89}$$

Inequality (89) implies that, taking a sufficiently small T

$$\left(\sum_{j=1}^{3}|U_j|^{(2+\beta)}_{\Omega_T}+|S_{\rm d}|^{(1+\beta/2)}_{[0,T]}+|S_{\rm f}|^{(1+\beta/2)}_{[0,T]}\right)\equiv 0, \tag{90}$$

which implies uniqueness in any time interval $[0, T]$. □

Appendix

We now prove some fundamental inequalities that have been used in Section 2 (throughout this Appendix we use the notations in [15]). Namely, we prove

Lemma 1

If $u \in H^{\alpha,\alpha/2}(\Omega_T)$ and $\beta < \frac{\alpha}{2}$ then

$$|u|^{(\beta)}_{\Omega_T} \leq |u(x,0)|^{(\beta)}_{\Omega} + 8\,|u|^{(\alpha)}_{\Omega_T}\,T^{\alpha/2-\beta}. \tag{A1}$$

Proof

As a first step we assume that $u(x, 0) \equiv 0$, and, for the sake of simplicity, $T \leq 1$.

Clearly we have

$$|u|^{(0)}_{\Omega_T} \leq |u|^{(\alpha)}_{\Omega_T}\,T^{\alpha/2}, \tag{A2}$$

$$\langle u\rangle^{(\beta/2)}_{t,\Omega_T} \leq |u|^{(\alpha)}_{\Omega_T}\,T^{\frac{\alpha-\beta}{2}}, \tag{A3}$$

(and (A3) holds true even if $u(x, 0) \neq 0$).

Remember that [15]

$$\begin{aligned}
|u|^{(0)}_{\Omega_T} &= \sup_{(x,t)\in\Omega_T}|u(x,t)| \\
|u|^{(\alpha)}_{\Omega_T} &= |u|^{(0)}_{\Omega_T}+\langle u\rangle^{(\alpha)}_{x,\Omega_T}+\langle u\rangle^{(\alpha/2)}_{t,\Omega_T} \\
\langle u\rangle^{(\alpha)}_{x,\Omega_T} &= \sup_{\substack{(x_1,t)\in\Omega_T\\(x_2,t)\in\Omega_T}}\frac{|u(x_1,t)-u(x_2,t)|}{|x_1-x_2|^{\alpha}} \\
\langle u\rangle^{(\alpha/2)}_{t,\Omega_T} &= \sup_{\substack{(x,t_1)\in\Omega_T\\(x,t_2)\in\Omega_T}}\frac{|u(x,t_1)-u(x,t_2)|}{|t_1-t_2|^{(\alpha/2)}}.
\end{aligned}$$

On the other hand,

$$\langle u\rangle^{(\beta)}_{x,\Omega_T} \equiv \sup_{\substack{x,x'\in\Omega\\t\in[0,T]}}\frac{\left|u(x,t)-u(x',t)\right|}{|x-x'|^{\beta}} = \sup_{\substack{x,x'\in\Omega\\t\in[0,T]}} I(x,x',t). \tag{A4}$$

However,

$$I(x,x',t) \le \frac{|u(x,t)| + |u(x',t)|}{|x-x'|^{\beta}},$$

at this point using the Holder continuity in time of order $\alpha/2$ of the function u we get

$$I(x,x',t) \le \frac{2\,|u|^{(\alpha)}_{\Omega_T}\, t^{\alpha/2}}{|x-x'|^{\beta}}.$$

If $|x-x'| \ge t$ we get

$$I(x,x',t) \le 2\,|u|^{(\alpha)}_{\Omega_T}\, t^{\frac{\alpha-2\beta}{2}} \le 2\,|u|^{(\alpha)}_{\Omega_T}\, T^{\alpha/2-\beta}. \tag{A5}$$

Also, using the Holder continuity in space of order α of the function u, we get

$$I(x,x',t) \le |u|^{(\alpha)}_{\Omega_T}\, |x-x'|^{\alpha-\beta}.$$

And, if $|x-x'| < t$,

$$I(x,x',t) \le |u|^{(\alpha)}_{\Omega_T}\, t^{\alpha-\beta} \le |u|^{(\alpha)}_{\Omega_T}\, T^{\alpha-\beta}. \tag{A6}$$

Putting together (A5) with (A6) we, finally, obtain

$$\langle u\rangle^{(\beta)}_{x,\Omega_T} \equiv \sup_{\substack{x,x'\in\Omega\\ t\in[0,T]}} \frac{|u(x,t)-u(x',t)|}{|x-x'|^{\beta}} \le 2\,|u|^{(\alpha)}_{\Omega_T}\, T^{\alpha/2-\beta}. \tag{A7}$$

Using (A2), (A3) and (A7) we get

$$|u|^{(\beta)}_{\Omega_T} \le 4\,|u|^{(\alpha)}_{\Omega_T}\, T^{\alpha/2-\beta}. \tag{A8}$$

If $u(x,0) \ne 0$ we proceed as follows:

$$\begin{aligned} |u-u(x,0)|^{(\beta)}_{\Omega_T} &\le 4\,|u-u(x,0)|^{(\alpha)}_{\Omega_T}\, T^{\alpha/2-\beta} \le 4\left(|u|^{(\alpha)}_{\Omega_T} + |u(x,0)|^{(\alpha)}_{\Omega_T}\right) T^{\alpha/2-\beta}\\ &\le 8\,|u|^{(\alpha)}_{\Omega_T}\, T^{\alpha/2-\beta}, \end{aligned}$$

which implies (A1). □

Remark 2
Note that, using (A1), it is not difficult to prove that, if $u \in H^{\alpha,\alpha/2}(\Omega_T)$ and $\beta < (\alpha/2)$, then the Holder norm $|u|^{(\beta)}_{\Omega_T}$ is continuous with respect to the variable T.

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