

ON A COMPUTATIONAL ALGORITHM FOR THE ANALYSIS OF NON-STATIONARY DIFFUSIONAL PHASE TRANSFORMATION *

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Abstract

Physical analysis of phase transformation of materials consisting from several (in general q substitutional and r interstitial) components, coming from the Onsager extremal thermodynamic principle, following some ideas from [2], [1] and [3], leads, as explained in [4], from the mathematical point of view, to a system of partial differential equations of evolution type, including certain integral term, with substantial differences in particular phases (α , γ) and in moving interface of finite thickness (β), in whose center the ideal liquid material behaviour can be detected. The numerical simulation of this process in MATLAB, mentioned in [5], is able to explain some phenomena (e.g. the interface velocity as a function of temperature) better than known simplified models, assuming the sharp interface and additional boundary and transfer conditions. This paper suggests an effective algorithm for the numerical analysis of the above mentioned system, applicable even for very complicated evaluations of chemical potentials.

1 Basic equations

The evolution of $q-1+r$ molar fractions c in one dimension is characterized in a Cartesian coordinate x and in time t . The coordinate x moves from the left to the right together with the interface of constant thickness h (from $x = 0$ to $x = h$); the total size of the specimen is H (in practice much greater than h), the system is assumed to be closed (with zero boundary fluxes) on the interval $\langle x_L(t), x_R(t) \rangle$. One missing molar fraction can be derived from the condition $c_1 + \dots + c_q = 1$. The resulting system of equations, starting

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from some a priori known initial values of c , reads

$$Bc' + (K + vN)c - N\frac{C}{\tau} = vNc^\diamond - N\Omega j^\diamond - N\frac{C^\times}{\tau} \quad (1)$$

where all variables are evaluated in time t , except $C^\times = C(t - \tau)$, τ denotes the time interval, referring to the implicit Euler method; (the system of differential equations can be derived from such difference ones using the limit passage $\tau \rightarrow 0$), B , K and N are square matrices of order $q - 1 + r$, B full, K and N diagonal, B and K depending on c , N dependent on x only, Ω is the constant molar volume and

$$C(x, t) = \int_0^x c(\xi, t) d\xi,$$

c^\diamond refers to molar fractions and j^\diamond to diffusive fluxes at $x = 0$ and

$$v = \frac{\Omega}{M} \sum_{i=1}^{q+r} \int_0^h c_i \mu'_i dx \quad (2)$$

for prescribed chemical potentials μ_i as complicated functions of c ; a prime symbol denotes a derivative with respect to x .

The system (1) comes (after rather long computations, performed in [4]), from the mass conservation law

$$\partial c / \partial t - vc' + \Omega j' = 0;$$

another (more evident) its consequence is

$$(C^R - C^{R^\times})/\tau - v(c^R - c^\diamond) - \Omega j^\diamond = 0, \quad (C^L - C^{L^\times})/\tau - v(c^L - c^\diamond) - \Omega j^\diamond = 0; \quad (3)$$

upper indices L and R here refer to values at x^L and x^R , respectively. As (1) generates an iterative procedure with B , K , v and x_L set by c from the preceding iteration (or time step), it is important to suggest an inexpensive solver of the system of linear algebraic equations, derived from (1), using the finite difference method. Unfortunately, such system is not triangular, thus, because of the presence of unknowns c^\diamond and j^\diamond it is not possible to express c in all nodes step-by-step; even c^\diamond and j^\diamond cannot be determined by (3) completely.

2 Computational algorithm

Let us notice that C can be computed as integrals of $c - c^a$ instead of c , using arbitrary reference constant admissible molar fractions c^a . Our problem is to find c from (1) with v inserted from (2). Clearly (1) requires discretization in x , e. g. using the finite difference method, whereas v from (2) needs numerical integration, e. g. by the Simpson rule, and

C (which can be computed from integrals of $c - c^a$ instead of c , using arbitrary reference constant admissible molar fractions c^a). should be evaluated in the corresponding way. However, for simplicity we shall explain the main idea of the computational algorithm using the original semi-discretized system (1).

Let $c^{\diamond e}$ be some estimate of c^{\diamond} (from the preceding iteration, if not available yet then from the previous time step). Let us consider $c_m^{\diamond} = \xi_m^I c_m^{\diamond e}$ and $j_m^{\diamond} = \xi_m^{II} v c_m^{\diamond e}$ for some positive real $2(q-1+r)$ factors ξ_m^I and ξ_m^{II} . We are allowed to seek for molar fractions c in form $c = c^{\diamond} + \tilde{c}$ where $\tilde{c}_m = \tilde{c}_m^O + \xi_m^I \tilde{c}_m^I + \xi_m^{II} \tilde{c}_m^{II}$. Then (1) degenerates to

$$B\tilde{c} + K\tilde{c} + vN\tilde{c} - N\frac{\tilde{C}}{\tau} = F^O + \xi_I F^I + \xi_{II} F^{II}$$

with \tilde{C} integrated from \tilde{c} , unlike C integrated from $c - c^a$ in general, and with

$$F^O = N\frac{C^{\times} - c^a x}{\tau}, \quad F^I = \left(N\frac{x}{\tau} - K\right) c^{\diamond e}, \quad F^{II} = -N\Omega v c^{\diamond e}.$$

Thus we are able to solve all \tilde{c}^O , \tilde{c}^I and \tilde{c}^{II} separately (which is very simple) and just at the end to calculate ξ^I and ξ^{II} ($q-1+r$)-times from the system of two linear algebraic equations

$$\begin{bmatrix} \tilde{C}_m^{LI}/\tau - v\tilde{c}_m^{LI} + c_m^{\diamond e} x_L/\tau & \tilde{C}_m^{LII}/\tau - v\tilde{c}_m^{LII} \\ \tilde{C}_m^{RI}/\tau - v\tilde{c}_m^{RI} + c_m^{\diamond e} x_R/\tau & \tilde{C}_m^{RII}/\tau - v\tilde{c}_m^{RII} \end{bmatrix} \cdot \begin{bmatrix} \xi_m^I \\ \xi_m^{II} \end{bmatrix} = \begin{bmatrix} -\tilde{C}_m^{LO}/\tau + v\tilde{c}_m^{LO} + C_m^{L\times}/\tau + c_m^a x_L/\tau \\ -\tilde{C}_m^{RO}/\tau + v\tilde{c}_m^{RO} + C_m^{R\times}/\tau + c_m^a x_R/\tau \end{bmatrix}. \quad (4)$$

Now let us sketch the fully discretized algorithm, generating (in each time and iterative step) a system of $2(q-1+r)$ linear algebraic equations. Using the above explained tricks with parameters ξ^I and ξ^{II} , we have a very inexpensive solver; this is very useful in situations where e.g. the algebraic expressions for evaluation of $\mu(c)$ contain thousands of instruction, preprocessed by MAPLE or toolbox *symbolic* from MATLAB environment.

We can write (1) in form

$$\begin{aligned} & \overline{B}^s \frac{c^s}{\Delta_s} + (\overline{K}^s + v\overline{N}^s) \frac{c^s}{2} - \overline{N}^s \frac{\Delta_s c^s}{2\tau} \\ & = \overline{B}^s \frac{c^{s-1}}{\Delta_s} - (\overline{K}^s + v\overline{N}^s) \frac{c^{s-1}}{2} + v\overline{N}^s c^{\diamond} - \overline{N}^s \Omega j^{\diamond} - \overline{N}^s \frac{2(C^{\times s} - C^{s-1}) - \Delta_s c^{s-1}}{2\tau} \end{aligned} \quad (5)$$

where an integer s refers to the s -th node in a sufficiently large interval, decomposed to subintervals $\langle x_{s-1}, x_s \rangle$ (values at x_L and x_R , in general not identical with any x_s , are interpolated), $\Delta_s = x_s - x_{s-1}$ and overlined s -indexed symbols refer to averaged values on

$\langle x_{s-1}, x_s \rangle$; let us remind that c^\diamond coincides with c^s always for some s . Clearly such scheme forces sufficiently small Δ_s in comparison with τ . We would like to solve $c^0, c^1, \dots, c^s, \dots$ effectively, step by step, using three versions of right-hand sides again, and complete them a posteriori, after setting ξ^I and ξ^{II} and consequently c^\diamond and j^\diamond .

The reformulation of (4) in the notation of (5) is important for the programmer; nevertheless, here can be left to the patient reader. The following simulation example makes use of this algorithm, applying the semi-empirical chemical potentials from the Montanuniversität Leoben (Austria) and from the Institute of Physics of Materials of the Czech Academy of Sciences in Brno.

3 Example of numerical simulation

For the physical interpretation of all following material characteristics let us refer to [3]. We have the purely substitutional three-component Fe-Cr-Ni system; in our notation $q = 3$ and $r = 0$, moreover Fe will be dominant. The chemical potential $\mu_i(x, c)$ can be evaluated at every point of the specimen as

$$\mu_i(x, c) = w^f(x) \mu_i^f(c),$$

making use of certain continuous weight functions $w^f(x)$, having the properties

$$\begin{aligned} w^\alpha(x) = 1, \quad w^\gamma(x) = 0 & \quad \text{if } x_L < x < h/2, \\ w^\alpha(x) = 0, \quad w^\gamma(x) = 1 & \quad \text{if } h/2 < x < x_R, \\ w^\beta(x) = 1 - w^\alpha(x) - w^\gamma(x) & \quad \text{if } x_L < x < x_R. \end{aligned}$$

The tracer diffusion coefficients D_k , $k \in \{1, 2, 3\}$, can be interpolated using the formula

$$\ln D_k = w^f \ln D_k^f,$$

thus it is sufficient to set nine values D_k^f . In general we have

$$D_k^f = D_{k0}^f \exp\left(-\frac{E^f}{RT}\right), \quad M = M_0 \exp\left(-\frac{E^*}{RT}\right).$$

The applied constants are for Cr (corresponding to $k = 1$) $D_{10}^\alpha = 0.00032 \text{ m}^2 \text{ s}^{-2}$, $D_{10}^\beta = 0.00022 \text{ m}^2 \text{ s}^{-2}$, $D_{10}^\gamma = 0.00035 \text{ m}^2 \text{ s}^{-2}$, for Ni ($k = 2$) $D_{20}^\alpha = 0.000048 \text{ m}^2 \text{ s}^{-2}$, $D_{20}^\beta = 0.000022 \text{ m}^2 \text{ s}^{-2}$, $D_{20}^\gamma = 0.000035 \text{ m}^2 \text{ s}^{-2}$, for Fe ($k = 3$) $D_{30}^\alpha = 0.00016 \text{ m}^2 \text{ s}^{-2}$, $D_{30}^\beta = 0.00011 \text{ m}^2 \text{ s}^{-2}$, $D_{30}^\gamma = 0.00007 \text{ m}^2 \text{ s}^{-2}$, and for all components $E^\alpha = 240000 \text{ J mol}^{-1}$, $E^\beta = 155000 \text{ J mol}^{-1}$, $E^\gamma = 286000 \text{ J mol}^{-1}$, $E^* = 140000 \text{ J mol}^{-1}$; it remains to set only $M_0 = 0.00041 \text{ m}^2 \text{ s kg}^{-1}$.

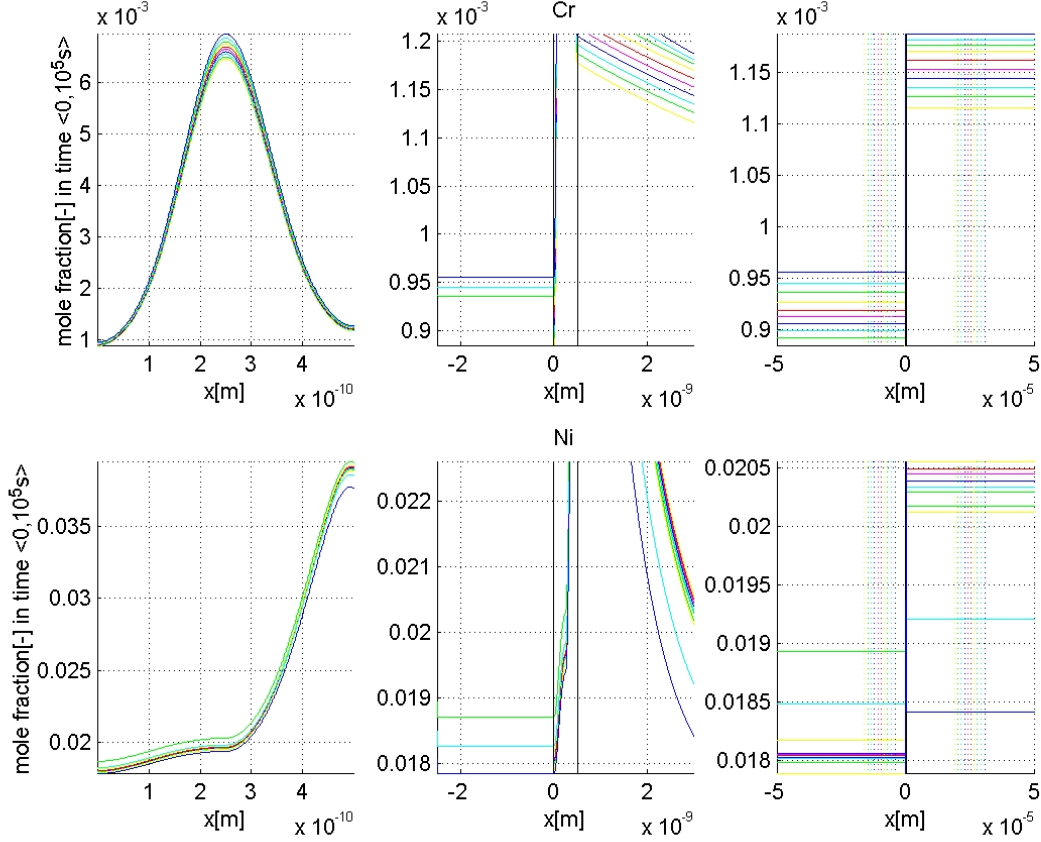


Figure 1: Distributions of Cr and Ni molar fractions in Fe-Cr-Ni system

The figure shows three couples of time-variable distributions of c_1 and c_2 . The interface thickness is $h = 5 \cdot 10^{-10}$ m, the specimen length $H = 10^{-4}$ m. From the originally constant mole fractions $c_1 = 0.001$ and $c_2 = 0.019$ (consequently $c_3 = 0.980$) in all phases due to the phase transformation driven by changes in chemical potentials, the time development leads to qualitative new distributions. The left figures show mole fractions inside the interface, the central ones document different behaviour of Cr and Ni components near the interface, the right ones show larger-scale quasi-constant distribution with seemingly sharp interface, whose physically transparent macroscopic description is not available.

Several directions of possible generalizations, even of types not included in this paper, are discussed in [4], p. 85. However, [4] contains only one example of modelling of non-stationary redistribution of molar fractions, moreover only near the initial time; a more extensive study is being prepared for [6].

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