Numerical modelling of thick-interface multi-component diffusional phase transformation

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ABSTRACT

Diffusion in multi-component systems can be characterized by three attributes: (i) the vacancy mechanism of diffusion for "slowly" diffusing substitutional components, (ii) the process of non-uniform generation and annihilation of vacancies, (iii) the "quick" diffusion of atoms of interstitial components. To simulate diffusional phase transformation, it is necessary to solve the coupled problem of diffusion and interface migration, incorporating (i), (ii), (iii) in all phases. However, in most computational model the interface is assumed to be sharp and some artificial interface contact conditions, as local equilibrium with partitioning, local equilibrium with negligible partitioning, para-equilibrium, etc., by [1], are applied at the interface. Moreover, for multi-component systems (compound from 3 or more components) the reliable theory of existence of (classical or weak) solutions of corresponding mathematical initial and boundary value problems and of convergence of sequences (e. g. of Rothe) of approximate solutions, like that from [2] for binary alloys, is missing or insufficient, containing many open problems.

A new multi-component model, taking into account a real migrating interface of finite thickness h, based on the balance for the total Gibbs energy in the interface, has been derived in [3]. Following the same idea, coming from the Onsager thermodynamic extremal principle, [5] introduces a one-dimensional two-phase system, containing two phases α and γ (the interface is considered as an additional phase β) with a finite number r of substitutional components, in which the $\gamma \rightarrow \alpha$ transformation occurs; the MATLAB-supported software simulations are used to the quantitative analysis of the steady-state massive phase transformation in a Fe-Cr-Ni system and also to the diffusion-induced grain boundary motion. Nevertheless, this approach needs generalizations in several directions, namely: (a) including some finite number s of interstitial components, (b) respecting the existence of (non-ideal) sources and sinks for vacancies, following the attribute (ii), (c) development of methods, algorithms and software for non-stationary simulations, (d) extension to two- and three-dimensional geometrical configurations, handling also curved interfaces. Some of these generalizations will be discussed in this contribution.

The concentration of particular components at every location x can be characterized by r molar fractions c_k with $k \in \{1, \ldots, r\}$, corresponding to substitutional components, satisfying an additional condition $c_1 + \ldots + c_r = 1$, and s molar fractions c_k with $k \in \{r + 1, \ldots, r + s\}$, corresponding to interstitial components; briefly $c = (c_1, \ldots, c_{r+s})$. Further variables in the system are 3 components of a finite

velocity $v = (v_1, v_2, v_3)$ and also $3(r + s)$ components of diffusion fluxes j_{kp}, $k \in \{1, \ldots, r + s\}$, $p \in \{1, 2, 3\}$. Let Ω be the molar volume. In the moving reference frame with dot symbols denoting partial time derivatives then the mass conservation law for an arbitrary component k reads

$$\dot{c}_k - v_p \nabla_p c_k = -\Omega \nabla_p j_{kp} \, ;$$

here p are sum indices from $\{1, 2, 3\}$ following the Einstein summation rule, similarly we shall later apply indices k, l from $\{1, \ldots, r + s\}$. The thermodynamic extremal principle yields

$$j_{kp} = -\rho_{kl} \nabla_p \mu_l(c)$$

with certain material characteristics ρ_{kl} and chemical potentials μ_l. Now we have $4(r + s)$ equations for $4(r + s) + 3$ unknowns. To complete the system, we need to evaluate velocity components v_p as functions of c, evidently incorporating $\mu_l(c)$ again. Unfortunately, this is in general rather complicated: namely [4] demonstrates that such relations have to include, in addition to the classical terms with $\nabla_p \mu_l(c)$, other (divergence-free) ones, covering the vacancy mechanism by (ii). Let us also notice that only $r + s - 1$ components of c are independent; this enables us to reduce the number of equations by one (which is the well-known trick in the simplified theory of binary substitutional alloys where, after all substitutions, only one final equations for c_1 remains).

For illustration, let us show some formally simple results for a one-dimensional system: we can remove an index 1 and write a prime symbol instead of ∇_1, α-phase is located from x_L to 0, γ-phase from h to x_R, $\max(-x_L, x_R - h) \ll h$. Ignoring all complicated improvements coming from (ii) and assuming that v, for some prescribed interface mobility M, is an interface velocity, considered as a function of t only (not of x), we can write

$$\dot{c} - vc' = -\Omega j' \, , \qquad v = \frac{\Omega}{M} \int_0^h c_k \mu_k'(c) \, \mathrm{d}x$$

and, denoting by C an integral from x_L to x, applied to $\dot{c}$, consequently

$$C - v(c - c(x_L)) = -\Omega j \, , \qquad C(x_R) - v(c(x_R) - c(x_L)) = 0$$

because in a closed system (although x_L and x_R are moving in time due to $v \neq 0$) we are allowed to set $j(x_L) = j(x_R) = 0$. Moreover (after long calculations) we are able to express j as

$$N(x)j = -B(c)c' - K(c)c$$

where a diagonal matrix N contains characteristics of diffusion; a full square matrix B (in practice close to a unit one) and a diagonal matrix K (non-zero only inside β) are complicated functions of $r + s - 1$ independent components of c (1 substitutional component has been removed), identified by experimental research of chemical potentials. The system can be then formulated (at any time) in form

$$B(c)c' + K(c)c + vN(x)c = vN(x)c_L + N(x)C \, ,$$

solvable numerically using an iterative MATLAB-supported FDM-based procedure.

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