

Modelling reustenitisation in Fe-C steels with concentration-dependent diffusivity of carbon^(*)

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Abstract A finite difference method used to model reustenitisation from a ferrite/cementite mixture in Fe-C steels is presented in this paper. Concentration-dependent carbon diffusivity in austenite is taken into account in order to generalise our earlier numerical model. We select some parameters, such as cementite dissolution time, and compare their values as calculated by different approximations available in the literature (in particular at steady state) for planar and spherical geometries. When the dependence of diffusivity on concentration or temperature is increased, the steady state approximation fails to predict correctly the above mentioned parameters and the use of numerical techniques becomes indispensable.

Keywords Steels. Austenite. Phase transformation. Diffusion. Computer simulation.

Modelización de la reustenización en aceros Fe-C con difusividad de carbono dependiente de la concentración

Resumen En este trabajo se presenta un método numérico de diferencias finitas para modelar la reustenización en aceros Fe-C a partir de una distribución inicial de ferrita y cementita. Se tiene en cuenta la dependencia de la difusividad en la austenita con la concentración de carbono, a fin de generalizar el propio modelo numérico previo. Se han seleccionado algunos parámetros, como el tiempo de disolución de la cementita, para comparar los valores obtenidos en este caso con los calculados con diferentes aproximaciones (en particular con la de estado estacionario) para los casos de geometrías plana y esférica. Los resultados obtenidos muestran que, cuando la difusividad depende fuertemente de la concentración, la aproximación de estado estacionario no predice correctamente los parámetros calculados y se hace imprescindible la aplicación de métodos numéricos.

Palabras clave Aceros. Austenita. Transformaciones de fase. Difusión. Cálculo numérico.

1. INTRODUCTION

During the last years significant progress has been made in the modelling of heat treatment processes in steels. Many studies have dealt with the prediction of the microstructural evolution of steel during cooling^[1-8], but the calculation of austenite formation during heating has been resumed only recently^[9-14]. The effect of austenite state at the end of heating on phase transition kinetics during cooling has also been considered^[15 and 16].

It used to be common to assume the diffusion coefficient to be independent of solute concentration (C in this case). However, in many systems of interest this is not the case. On the contrary, dependence may be very strong^[17] and,

when large concentration gradients are involved, this dependence cannot be neglected^[18].

Numerical and analytical approaches assuming the dependence of carbon diffusivity on its concentration have appeared in the literature^[19 and 20]. Either steady state growth^[19] or variational calculus planar geometry^[20] has been applied in these works. These assumptions allow an analytical approach for carbide dissolution in steels and are very useful for evaluating austenite formation during heating. However, those approximations are valid only when certain conditions are fulfilled: validity of the quasi-steady-state regime^[19] or planar geometry^[20].

In a recent paper^[13] we presented a numerical approach for the diffusion equation applied to

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re-austenitisation from a ferrite/cementite mixture in Fe-C steels. Planar geometry and concentration independent diffusivity of carbon in austenite were assumed in order to obtain analytical results and to compare them with our numerical method.

The present work is concerned with modelling re-austenitisation from ferrite/cementite mixtures in Fe-C steels. We develop a numerical method that allows an arbitrary dependence of carbon diffusivity on its concentration. The resulting non-linear differential equations are solved by means of a stable finite difference approach, which allows to obtain time dependent interface positions and concentration profiles of carbon in austenite during the growing process. It has the added advantage that it can be easily extended to analyse re-austenitisation during continuous heating of ferrite-cementite aggregates in Fe-C steels.

We compare our numerical results with different approximations appearing in the literature: the steady state^[19] and variational^[20] approaches. We want to remark that even when steady state conditions are well satisfied^[19] the parameters obtained by the steady state approach and the proposed numerical results may differ considerably.

The results obtained through the present model will be compared with experimental data in a future work.

2. MATHEMATICAL BACKGROUND OF THE MODEL

Re-austenitisation from ferrite-cementite austenite growth implies the simultaneous movement of the cementite-austenite and austenite-ferrite interfaces; in our first approximation we neglect the nucleation step^[9] because with very large distances between carbides, cementite particles rapidly become engulfed by austenite. Ferrite, cementite and austenite phases will be denominated α , θ and γ , respectively. Assuming that α/γ and θ/γ transformations are governed only by the solute diffusion in the γ phase, that is, that carbon diffusion into cementite (quasi-stoichiometric composition) and into ferrite (carbon almost null solubility) may be neglected, and considering the carbon diffusion coefficient as concentration dependent, the corresponding kinetics may be described by the following equation:

$$\frac{\partial C_\gamma}{\partial t} = \frac{1}{r^n} \frac{\partial}{\partial r} \left[D(C_\gamma) r^n \frac{\partial C_\gamma}{\partial r} \right] \quad (1)$$

$C_\gamma = C_\gamma(r, t)$ represents carbon concentration at a distance r from the origin at time t and $D(C_\gamma)$ is its diffusion coefficient in austenite; $n = 0$ for planar geometry and $n = 2$ for spherical geometry. The diffusion coefficient is treated as concentration dependent.

Figure 1 shows carbon concentration profiles in the three phases.

The mass flow balances in the α/γ and γ/θ interfaces may be expressed by:

$$(C_\theta - C_\gamma^{\gamma\theta}) \frac{d r_{\theta\gamma}}{d t} = D \left(\frac{\partial C_\gamma}{\partial r} \right)_{r=r_{\theta\gamma}} \quad (2)$$

$$(C_\gamma^{\gamma\alpha} - C_\alpha) \frac{d r_{\gamma\alpha}}{d t} = D \left(\frac{\partial C_\gamma}{\partial r} \right)_{r=r_{\gamma\alpha}} \quad (3)$$

where $r_{\theta\gamma}$ and $r_{\gamma\alpha}$ represent the instantaneous positions of the cementite/austenite and ferrite/austenite interfaces, respectively, and $C_\gamma^{\gamma\theta}$ and $C_\gamma^{\gamma\alpha}$ represent the carbon concentrations at these interfaces. and represent carbon concentrations in the cementite and ferrite phases, respectively.

The initial and boundary conditions are expressed by:

$$r_{\theta\gamma}(t=0) = r_0; \quad r_{\gamma\alpha}(t=0) = r_0 \quad (4)$$

where r_0 is the initial size of the cementite particle.

$$\begin{aligned} C &= C_\theta \text{ for } 0 \leq r \leq r_{\theta\gamma} \text{ and} \\ C &= C_\alpha \text{ for } r_{\gamma\alpha} \leq r \leq \infty \end{aligned} \quad (5)$$

Local equilibrium at the interfaces is expressed by:

$$C_\gamma = C_\gamma^{\gamma\theta} \text{ at } r = r_{\theta\gamma} \text{ and } C_\gamma = C_\gamma^{\gamma\alpha} \text{ at } r = r_{\gamma\alpha} \quad (6)$$

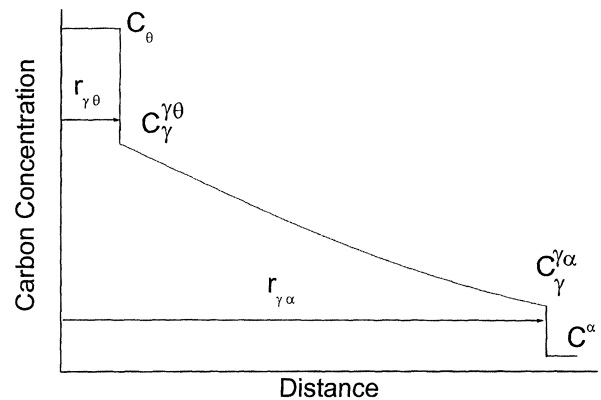


Figure 1. Scheme of the distribution of carbon concentration in ferrite, cementite and austenite phases.

Figura 1. Distribución esquemática de la concentración de carbono en las fases ferrita, cementita y austenita.

3. THE NUMERICAL SOLUTION

In this Section we present the numerical solution of Eqs. (1)-(6) by the Finite Difference approximation.

A uniform mesh with N nodes is defined between the θ/γ and γ/α interfaces. The first node corresponds to $r = r_{\theta\gamma}$ and the N^{th} to $r = r_{\gamma\alpha}$; the nodal interval is:

$$\Delta r = \frac{(r_{\gamma\alpha} - r_{\theta\gamma})}{N - 1} \quad (7)$$

At $t = 0$ there is no austenite phase ($r_{\gamma\alpha} = r_{\theta\gamma} = r_o$) but an initial mesh in this phase is necessary in order to start the Finite Difference program. This mesh is generated by allowing the system to evolve from $t = 0$ until a later time t_o . Such "initial" solution (for the Finite Difference program) is characterised by the $r_{\theta\gamma}$, $r_{\gamma\alpha}$, C^{θ} , $C^{\gamma\alpha}$, C_{θ} and r_o values. Arbitrary values of r_o and $r_{\gamma\alpha}$ may be chosen but t_o and $r_{\theta\gamma}$ will be determined by such choice. Concentration values at the interfaces have been taken from table I of Ref.^[9]. In order to estimate the movement of the θ/γ interface with respect to r_o , when the γ/α interface has moved from r_o to $r_{\gamma\alpha}$, and also the time elapsed, we resort to the stationary solution for the problem. From t_o on, the solution is calculated through the FD program.

Table I. Values of dissolution times of cementite as a function of temperature for planar geometry and linear dependence of diffusivity (see text)

Tabla I. Valores de los tiempos de disolución de la cementita en función de la temperatura para geometría plana y con una dependencia lineal de la difusividad (ver texto)

T (°C)	"Dissolution time" (s)			
	FD	Dav	Steady case	Difference (%)
750	454	456	499.6	8.7
775	163.3	163.7	217	24.6
800	79.2	81.2	134.4	35.6
825	43	45	100.8	55.4
850	24.7	26.9	90.4	70.2
875	15	16.55	162.4	89.8
900	9.65	10.6	223.2	95.2

Following usual finite difference numerical methods^[21] and denoting the discrete time increment by Δt , we define,

$$C_i^j = C(i\Delta r, j\Delta t) \quad (8)$$

$$\frac{C_i^{j+1} - C_i^j}{\Delta t} = n \frac{1}{r_{\theta\gamma} + i\Delta r} a_{pi} \frac{(C_{i+1}^{j+1} - C_{i-1}^j)}{2\Delta r} + \frac{1}{\Delta r^2} \left\{ a_{i+1} (C_{i+1}^{j+1} - C_i^{j+1}) - a_{i+1} (C_i^{j+1} - C_i^{j+1}) \right\} \quad (9)$$

where,

$$\begin{aligned} a_i [C] &= D \left[\frac{C_i + C_{i-1}}{2} \right] \\ a_{i+1} [C] &= \left[D \frac{C_{i+1} + C_i}{2} \right] \\ a_{pi} [C] &= D [C_i] \end{aligned} \quad (10)$$

In order to take into account the increasing of the nodal intervals with time we use the standard expression^[22]:

$$\frac{\partial C}{\partial t} \Big|_{i=\text{cst}} = \frac{dC}{dt} = \frac{\partial C}{\partial r} \cdot \frac{dr}{dt} + \frac{\partial C}{\partial t} \Big|_{r=\text{cst}} \quad (11)$$

where,

$$\frac{\partial C}{\partial r} \cdot \frac{dr}{dt} = \frac{(C_{i+1}^{j+1} - C_{i-1}^j)}{2 \cdot \Delta r} V_i \quad (12)$$

Here V_i is related to the interfaces velocities and the corresponding distances to the interfaces positions by:

$$V_i = \frac{r_{\gamma\alpha} - r}{r_{\gamma\alpha} - r_{\theta\gamma}} \frac{dr_{\theta\gamma}}{dt} + \frac{r - r_{\theta\gamma}}{r_{\gamma\alpha} - r_{\theta\gamma}} \frac{dr_{\gamma\alpha}}{dt} \quad (13)$$

Using σ and $(1-\sigma)$, $0 \leq \sigma \leq 1$, as the weight corresponding to times $(j+1) \Delta t$ and $j\Delta t$, respectively and defining $\gamma = \frac{\Delta t}{\Delta r^2}$, we obtain:

$$\begin{aligned}
 & C_i^{j+1} [1 + \sigma \gamma (a_{i+1} + a_i)] + \\
 & + C_{i-1}^{j+1} \left[V_i \frac{\sigma \Delta t}{2 \Delta r} + \sigma \gamma \left(\frac{n}{2} \cdot \frac{a_{pi}}{r_{\theta\gamma} + i \Delta r} - a_i \right) \right] + \\
 & + C_{i+1}^{j+1} \left[-V_i \frac{\sigma \Delta t}{2 \Delta r} - \sigma \gamma \left(\frac{n}{2} \cdot \frac{a_{pi}}{r_{\theta\gamma} + i \Delta r} + a_{i+1} \right) \right] = \\
 & = C_i^j [1 - (1 - \sigma) \gamma (a_{i+1} + a_i)] + C_{i-1}^j (1 - \sigma) \cdot \\
 & \left[-V_i \frac{\sigma \Delta t}{2 \Delta r} - \gamma \left(-\frac{n}{2} \cdot \frac{a_{pi}}{r_{\theta\gamma} + i \Delta r} + a_i \right) \right] + \quad (14) \\
 & + C_{i+1}^j (1 - \sigma) \left[V_i \frac{\sigma \Delta t}{2 \Delta r} + \gamma \left(\frac{n}{2} \cdot \frac{a_{pi}}{r_{\theta\gamma} + i \Delta r} + a_{i+1} \right) \right]
 \end{aligned}$$

Eq. (14) is solved by the Gauss-Seidel method^[23] assuming $\sigma = 0.5$. For any time step j , a sufficient number of iterations were made until the relative variation between the concentration values was less than 10^{-4} for each i . This assures the stability and accuracy of our numerical method.

4. COMPARISON

In this section we present the main results of this paper, which consist in applications of the approximation described above, to the planar and spherical cases.

In order to show the importance of using numerical methods, we select some parameters of interest and compare their values as calculated by different approximations.

The dependence on concentration of the diffusion coefficient of carbon in austenite is taken from ref.^[17] (linear function) and ref.^[24] (exponential function):

$$D = (0.07 + 0.06 \% C) \cdot \text{Exp} \left(-\frac{32000}{RT} \right) \text{ with } \% C : \text{ wt } \% ; \quad (15)$$

$$D = 0.5 \text{Exp} \left(\frac{38000}{RT} \right) \text{Exp} \left[\left(\frac{150000}{RT} - 30 \right) C \right] \text{ with } C : \text{ mole fraction}; \quad (16)$$

$$[D] = \frac{\text{cm}^2}{\text{s}} \quad [T] = {}^\circ\text{K} \quad R = 8.314 \frac{\text{J}}{\text{K} \cdot \text{mol}}$$

The computation of cementite dissolution times was made assuming that both interfaces

move without limits since analytical and steady state solutions are unable to take into account that the cementite particle is embedded in a ferrite matrix of finite volume that stops γ/α interface advance when ferrite volume is exhausted. This situation could also occur when re-austenitisation is carried out within the intercritical range ($\text{Ac1} < T < \text{Ac3}$) the final state of which is a mixture of ferrite and austenite, in proportions given by the lever rule. The above restrictions may be easily avoided with the use of our numerical method, but, in order to compare the results obtained by different techniques, we assume no limitations in interface movement in the numerical method as well.

4.1. Planar geometry

When planar geometry is considered an exact analytical solution can be obtained for a constant (concentration-independent) diffusion coefficient^[9 and 13]; in this case both interfaces positions obey a square root law with time:

$$r_{\theta\gamma}(t) = \eta_{\theta\gamma} t^{-1/2} + r_0 \quad (17)$$

$$r_{\gamma\alpha}(t) = \eta_{\gamma\alpha} t^{-1/2} + r_0$$

The coefficients of $t^{1/2}$ for $r_{\theta\gamma}(t)$ and $r_{\gamma\alpha}(t)$ have opposite signs ($\eta_{\theta\gamma} < 0$, $\eta_{\gamma\alpha} > 0$) because, as can be seen from Eqs.(2) and (3), $\frac{dr_{\gamma\alpha}}{dt} > 0$ and $\frac{dr_{\theta\gamma}}{dt} < 0$

reflecting the fact that the position of the cementite/austenite (austenite/ferrite) interface is a decreasing (increasing) function of time.

It has been common practice (see Ref.^[9] and references therein), to assume a constant diffusivity to be in the form of a weighted average defined as:

$$D_{av} = \frac{\int_{C_\gamma^{\gamma\alpha}}^{C_\gamma^{\theta\gamma}} D(C) dC}{(C_\gamma^{\theta\gamma} - C_\gamma^{\gamma\alpha})} \quad (18)$$

In figure 2 we compare the values of $\eta_{\theta\gamma}$ and $\eta_{\gamma\alpha}$ as calculated through the present finite difference method with a linear dependence of the diffusion coefficient (Eq. 15) and by the analytical solution with the diffusivity $D = D_{av}$

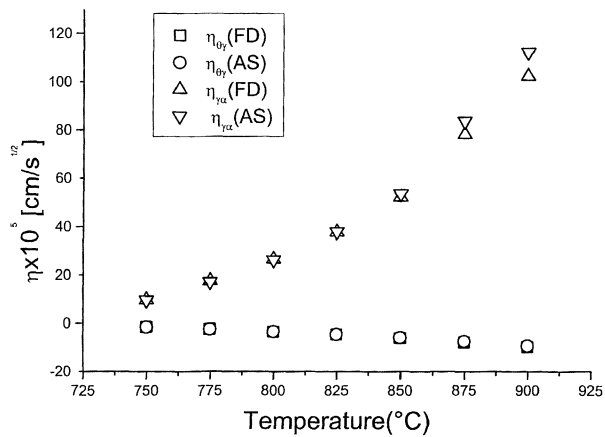


Figure 2. Values of $\eta_{\theta\gamma}$ and $\eta_{\gamma\alpha}$ calculated by the proposed finite difference method with a linear dependence of the diffusion coefficient (Eq. 15) and its analytical expression with $D = D_{av}$.

Figura 2. Valores de $\eta_{\theta\gamma}$ y $\eta_{\gamma\alpha}$ calculados a través del método propuesto de diferencias finitas con una dependencia lineal del coeficiente de difusión (Ec. 15) y su expresión analítica con $D = D_{av}$.

The differences in the values of these parameters, as obtained through the approximate analytical method ($D = D_{av}$) and through the numerical method (FD), proved to increase with temperature, a phenomenon already observed by Atkinson *et al.* [20].

In figure 3 we show the same parameters as in figure 2, but assuming an exponential dependence of diffusivity with concentration (Eq. 16). We observe an increasing difference between these parameters.

Table I shows the values of dissolution time of cementite as a function of temperature for planar geometry. Dissolution times calculated by the proposed numerical method with linear and constant ($D = D_{av}$) concentration-dependent diffusivity are presented in the first and second column, respectively. The third column shows dissolution time calculated by steady state approximation. Percentual differences between values of first and third column appear in the last column. Table II takes into account an exponential dependence of diffusivity and shows in its columns the corresponding dissolution times as in table I.

The steady state approximation failed to predict correctly dissolution time values as well as $\eta_{\theta\gamma}$ and $\eta_{\gamma\alpha}$ yielding greater deviations when the dependence of diffusivity on concentration is stronger. On the other hand, the numerical method yields correct results even when using a constant diffusivity ($D = D_{av}$).

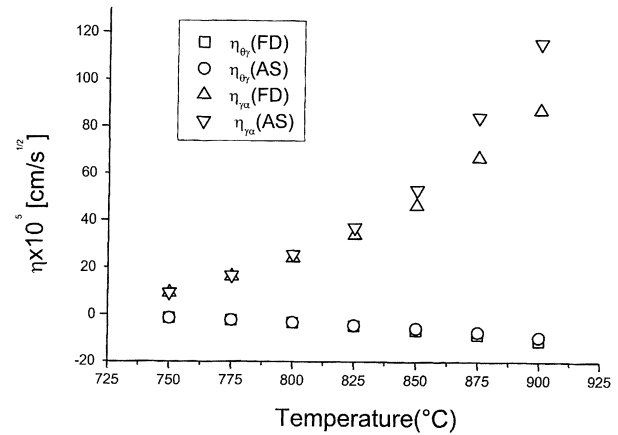


Figure 3. Values of $\eta_{\theta\gamma}$ and $\eta_{\gamma\alpha}$ calculated by the proposed finite difference method with an exponential dependence of the diffusion coefficient (Eq. 16) and its analytical expression with $D = D_{av}$.

Figura 3. Valores de $\eta_{\theta\gamma}$ y $\eta_{\gamma\alpha}$ calculados a través del método propuesto de diferencias finitas con una dependencia exponencial del coeficiente de difusión (Ec. 16) y su expresión analítica con $D = D_{av}$.

Table II. Values of dissolution times of cementite as a function of temperature for planar geometry and exponential dependence of diffusivity (see text)

Tabla II. Valores de los tiempos de disolución de la cementita en función de la temperatura para geometría plana y con una dependencia exponencial de la difusividad (ver texto)

T (°C)	"Dissolution time" (s)			
	FD	Dav	Steady case	Difference (%)
750	470	476	556	14.4
775	169	176.27	252.3	30.4
800	79.9	86.33	157.7	45.2
825	41.7	47.11	117	59.7
850	23	27.3	102.9	75
875	12.7	16.4	181.9	91
900	7.6	9.97	234.88	96.8

4.2. Spherical geometry

No exact analytical solution is known for the spherical case and, so, we must appeal to approximation methods. The most used of them is the steady state approximation [19]. In tables III and IV we assumed spherical geometry of carbide and compare its dissolution time values for different temperatures when the diffusion coefficient of

Table III. Values of dissolution times of cementite as a function of temperature for spherical geometry and linear dependence of diffusivity (see text)

Tabla III. Valores de los tiempos de disolución de la cementita en función de la temperatura para geometría esférica y con una dependencia lineal de la difusividad (ver texto)

T (°C)	"Dissolution time" (s)			
	FD	Dav	Steady case	Difference (%)
750	37.36	37.58	32.72	12.4
775	14.16	14.34	11.22	20.7
800	7.30	7.46	5.36	26.6
825	4.24	4.35	2.96	30.4
850	2.62	2.69	1.78	32
875	1.66	1.72	1.18	29
900	1.11	1.18	0.82	26

Table IV. Values of dissolution times of cementite as a function of temperature for spherical geometry and exponential dependence of diffusivity (see text)

Tabla IV. Valores de los tiempos de disolución de la cementita en función de la temperatura para geometría esférica y con una dependencia exponencial de la difusividad (ver texto)

T (°C)	"Dissolution time" (s)			
	FD	Dav	Steady case	Difference (%)
750	38.59	39.15	28.02	27.4
775	14.74	15.24	8.41	43.0
800	7.43	7.87	3.58	52.0
825	4.17	4.52	1.79	57.0
850	2.47	2.75	1.00	60.0
875	1.49	1.73	0.63	58.0
900	0.94	1.12	0.42	55.0

carbon in austenite has linear and exponential dependence on concentration, respectively.

As in the planar case, the difference between dissolution time values calculated with the finite difference method and the steady state approximation increases as the diffusion coefficient dependence on concentration gets stronger.

We also calculate the interface positions as a function of time by using the proposed finite difference method and compare them with the

steady state approximation. Figure 4 shows both results.

5. CONCLUSIONS

A finite difference numerical method to account for carbon diffusivity dependence on concentration in re-austenitisation from ferrite/cementite mixtures has been developed in this work. Considering that analytical solutions to this problem are generally unknown, numerical modelling becomes indispensable. Even when approximate analytical expressions are currently used to describe re-austenitisation, their validity must be tested through other methods. Moreover, when no analytical expressions are available^[18] for the diffusion coefficient, the use of numerical methods cannot be avoided.

We may assure that for planar geometry, the steady state approximation shows no satisfactory behaviour of certain parameters like dissolution time, as such time should decrease monotonously as isothermal reaction temperature grows. This fact arises from the fact that austenite formation energy and diffusion velocity increase with temperature. This, however, is not the behaviour predicted from the steady state solution (Tables I and II).

On the other hand, the analytical solution with an average constant diffusion coefficient ($D=D_{av}$), as defined in section 4, notably yields values of the mentioned parameters quite similar to those obtained by numerical methods.

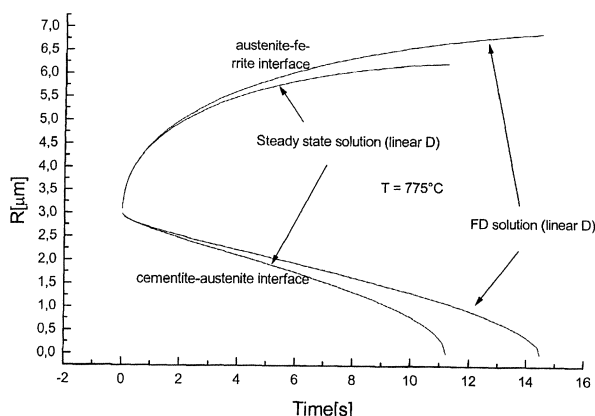


Figure 4. Interfaces position as function of time calculated by the proposed finite difference method and by the steady state approximation for spherical geometry.

Figura 4. Posición de las interfaces en función del tiempo calculadas a través del método propuesto de diferencias finitas y por la aproximación de estado estacionario para geometría esférica.

In the case of spherical carbides analysed above, we see that, even when quasi-stationary approaches are valid^[19], the dissolution time values as calculated with this approximation result in error (Tables III y IV).

Austenite nucleation at carbide-ferrite interfaces and carbide size initial distribution as well as the presence of ternary alloying must be taken into account to compare our results with experimental procedures^[19]. We are currently working to incorporate the above conditions in our numerical program.

Finally, it is worth mentioning that numerical calculation must be used when the physical model is improved to account for more complex situations, as, for example, when the system is heated or cooled continuously. Work on this line is currently in progress.

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