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NUMERICAL APPROACH TO PRECIPITATE GROWTH AND DISSOLUTION CALCULATION

NUMERYCZNE OBLICZENIA WZROSTU I ROZPUSZCZANIA WYDZIELEŃ

Different aspects of precipitates formation, growth and dissolution have been thoroughly studied and many factors influencing it such as diffusion, thermodynamic properties and kinetics have been taken into consideration.

The aim of this work is to present numerical approach to precipitates growth and dissolution processes, considering them as diffusion controlled ones, described by the first Fick's law. There have been solved differential equations with two variables (time and space) by the means of finite difference method. As a result, there have been obtained dependences of precipitate radii on time in processes of coagulation and dissolution. Results calculated in such a way have been compared with the values obtained by the means of other methods.

Keywords: coagulation, growth and dissolution of precipitates, finite differential method

Tworzenia się wydzieleń było wielokrotnie analizowane, przy czym bramo pod uwagę wiele czynników mających wpływ na ten proces np. dyfuzję, kinetykę oraz właściwości termodynamiczne.

W pracy przedstawiono numeryczne podejście do procesu rozrostu i rozpuszczania się wydzieleń, przy założeniu że są to procesy kontrolowane przez dyfuzję, opisane pierwszym prawem Ficka. Rozwiązano równania różniczkowe z dwoma zmiennymi (czas i przestrzeń) metodą różnic skończonych. Uzyskano zależności promienia wydzielenia od czasu w procesie wzrostu wydzieleń i rozpuszczania. Rezultaty obliczeń porównano z wartościami uzyskanymi innymi metodami.

1. Introduction

Non metallic precipitates which may appear in a liquid metal at various stages of its elaboration or during its solidification have a strong influence on mechanical properties of a product. In order to monitor mechanical properties in relation to microstructure, the knowledge of the precipitation state is of prime importance. For these purpose various models have been developed to allow the prediction of precipitates' size and distribution using theories of nucleation and growth of precipitates [1, 2, 3, 4, 5].

2. Assumptions

As to describe the growth or dissolution of a precipitate, the following assumptions were made in order to simplify calculations [4, 6]:

- the precipitate is spherical
- each precipitate grows independent of others
- a stationary diffusion state is reached
- flux of atoms to the surface is described by the Fick's law
- growth and dissolution of precipitates were assumed to be controlled by the diffusion of the non – metallic elements of precipitates in liquid steel
- at the interface between the precipitate and the liquid steel, only the concentrations of elements forming the precipitate were considered
- the non – metallic element concentration at the interface between the precipitate and the liquid steel was accepted 0 (for the growth) and equal to the boundary dissolution value in steel (for dissolution)
- precipitates are uniformly dispersed in the volume
- precipitates grow at the expense of solute the non – metallic element
- growing leads to a steady – state size distribution of precipitates

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3. Calculations

Calculations were performed by the means of the finite difference method [7]. This method is based on approximations that enable replacing a differential equation by a finite difference equation. Approximations have an algebraic form, they combine the value of the dependent variable at the certain point with values in several neighbouring points.

Solving the problem by the finite difference method consists of three steps:

1. Division of the solution region into the mesh of nodes.
2. Approximation of a certain differential equation by the equivalent difference equation, what corresponds to the dependence of the dependent variable at the point of a solution region to its value in neigbouring points.
3. Solution of a difference equation that takes into consideration initial and/or boundary conditions.

Approximation of a difference equation by the finite differences method is as follows:

For a f(x) function it is possible to approximate its derivative (slope/tangent) at the point P by three methods (Fig. 1) 1.

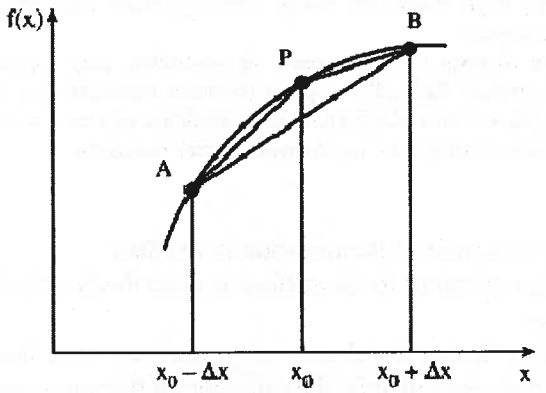

Fig. 1. Approximation of a function's derivatives at the point x_0, three methods

1. By the slope of PB arc described as a difference of functions:

$$f'(x_0) \cong \frac{f(x_0+\Delta x)-f(x_0)}{\Delta x}. \tag{1}$$

2. By the slope of AP arc described as a reverse difference of functions:

$$f'(x_0) \cong \frac{f(x_0)-f(x_0-\Delta x)}{\Delta x}. \tag{2}$$

3. By the slope of AB arc given as a central difference of functions:

$$f'(x_0) \cong \frac{f(x_0+\Delta x)-f(x_0-\Delta x)}{2\Delta x}. \tag{3}$$

The second derivative f'(x) at point P can be approximated as:

$$f''(x_0) \cong \frac{f'(x_0+\Delta x/2)-f'(x_0-\Delta x/2)}{\Delta x} = \\ = \frac{1}{\Delta x}\left(\frac{f(x_0+\Delta x)-f(x_0)}{\Delta x} - \frac{f(x_0)-f(x_0-\Delta x)}{\Delta x}\right). \tag{4}$$

That is:

$$f''(x_0) \cong \left(\frac{f(x_0+\Delta x)-2f(x_0)+f(x_0-\Delta x)}{\Delta x^2}\right). \tag{5}$$

The above interpretation of finite differences results from a function's expansion into Taylor series:

$$f(x+\Delta x) = f(x)+f'(x)+f''(x)/2+\ldots = \sum_{n=0}^{\infty}\frac{f^n(x)}{n!}(\Delta x)^n. \tag{6}$$

Where elements of higher order derivatives are omitted. As to find Φ (x,t) function, the solution region in the $x-t$ plane is divided into rectangles of Δx and Δt sides – the region of calculation is discretized by mesh nodes with two coordinates – time and space [8]. By reducing the space to one dimension, the space of calculations time – distance is obtained (Fig.2).

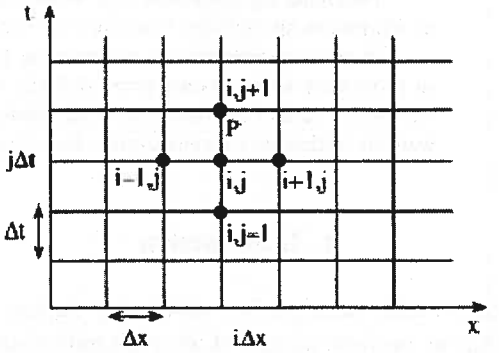

Fig. 2. Time and space discretization

The f(x) function values at mesh nodes for t_{n+1} calculation step are determined on the basis of earlier determined values for t_n calculation step. Therefore, it is necessary to know the mesh values for the initial time t_0, so called initial conditions. Moreover, for some mesh nodes the function fulfils boundary conditions (essential or/and natural), which means that the function's values or its derivatives are fixed and known. Sometimes, (for example in this paper), nodes' coordinates may be changed during calculations.

It has been assumed, that the processes of coagulation and dissolution are described by the first Fick's law:

$$J = -D\nabla C, \tag{7}$$

where:
J – flow of atoms (or molecules),
D – the diffusion coefficient,
∇C – the gradient of concentration.

Diffusion is regarded as steady, when:

$$\frac{\partial C}{\partial t} = -D\frac{\partial C}{\partial x}. \tag{8}$$

Or not steady, when:

$$\frac{\partial C}{\partial t} = -D\frac{\partial^2 C}{\partial x^2}. \tag{9}$$

It has been assumed that diffusion is steady.

Generally, the variable x means a three – element vector of coordinates in three – dimensional space. But sometimes, due to symmetry, equation /8/ may have a one – dimension form, (cartesian, cylindrical or spherical):
In 1D

$$\text{Cartesian} \qquad D\frac{\partial^2 C}{\partial x^2} = 0 \tag{10}$$

$$\text{Cylindrical} \qquad \frac{D}{r}\frac{\partial}{\partial r}\left[r\frac{\partial C}{\partial r}\right] = 0 \tag{11}$$

$$\text{Spherical} \qquad \frac{D}{r^2}\frac{\partial}{\partial r}\left[r^2\frac{\partial C}{\partial r}\right] = 0. \tag{12}$$

In the paper, there were accepted spherical coordinates. In the formula /8/ derivatives of time and space, (dC/dt, dC/dx) were discretized

$$\frac{\partial C}{\partial t} \cong \frac{C_i^{j+1} - C_i^j}{\Delta t} \tag{13}$$

and

$$\frac{\partial C}{\partial x} \cong \frac{C_{i+1}^j - C_{i-1}^j}{2\Delta x}. \tag{14}$$

Finally, the following formula was obtained:

$$\frac{C_i^{j+1} - C_i^j}{\Delta t} \cong -D\frac{C_{i+1}^j - C_{i-1}^j}{2\Delta x}. \tag{15}$$

That enables determining concentrations values in the net i, in the time step $j + 1$: C_i^{j+1} on the basis of values determined in the step j:

$$C_i^{j+1} \cong C_i^j - D\Delta t\frac{C_{i+1}^j - C_{i-1}^j}{2\Delta x}. \tag{16}$$

During calculations, the stability criterion has to be fulfilled:

$$\Delta t \leqslant \frac{(\Delta x)^2}{2D}. \tag{17}$$

4. Results

In this paper, the growth of Al_2O_3 and Ti_3O_5 precipitates in steel at 1873K and the dissolution of Al_2O_3 precipitates in steel at 2200K were calculated.

Calculations were carried out by the use of the finite difference method and all assumptions presented above were taken into consideration.

The area around an precipitate was divided into spheres of radii r_i and surface S_i. (Fig. 3). In this area a process of one precipitate's coagulation or dissolution take place. The radius of this area was calculated on the basis of number of precipitates per volume.

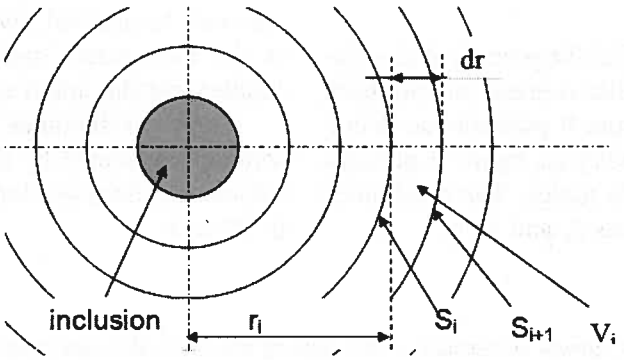

Fig. 3. Discretized area around an inclusion

Values of standard free energies of reactions, solubility of products and diffusion coefficient of oxygen were calculated from data presented in [6], whereas the amount of oxygen taking part in a certain reaction was accepted from [4]. The radii of areas, in which coagulations take place are:
For Al_2O_3 : $r_i = 0.64*10^{-4}$m for Ti_3O_5: $r_i = 1.14*10^{-5}$m

The number of precipitates in $1m^3$ is:
For Al_2O_3 : $1.17*10^{12}$ for Ti_3O_5: $6.7*10^{14}$

The area around precipitate was divided into the fixed number of elements limited by spheres, (S_i, S_{i+1}), see figure 3, and during calculations, when the precipitate's dimension was changing (it was increasing for coagulation and decreasing for dissolution), the distances between spheres (d_r), were changing as well (they decreased for coagulation and increased for dissolution).

The calculations were carried on in the Mathematica program. There was calculated the amount of the non – metallic element moving from one mesh node to the other (due to diffusion) and finally reaching the precipitate (for coagulation), which resulted in the growth of precipitate. Moreover, the increments of precipitates' masses (Fig. 4 and 5) and changes of precipitates' radii

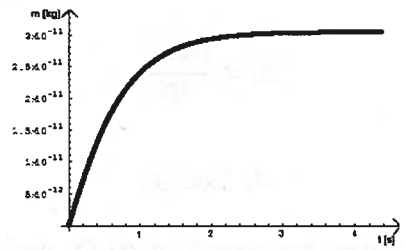

Fig. 4. Increments of Al_2O_3 inclusion's mass at 1873K

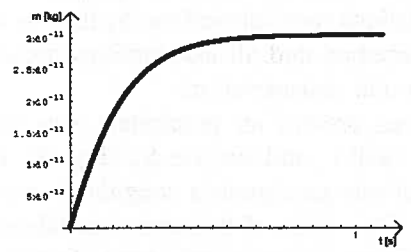

Fig. 5. Increments of Ti_3O_5 inclusion's mass at 1873K

(Fig. 6 and 7) were calculated. For the process of dissolution, the amount of non - metallic element moving from precipitate was calculated. Figure 8 presents the decrements of precipitate's mass, whereas figure 9 presents the decrease of the precipitate's radius. The mesh node 'i' was situated between surfaces S_i and S_{i+1}.

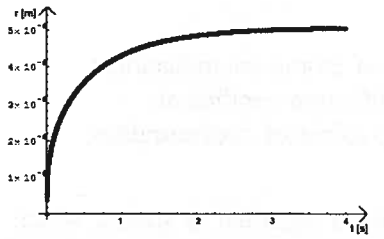

Fig. 6. The dependence of Al_2O_3 inclusion radii of time at 1873K

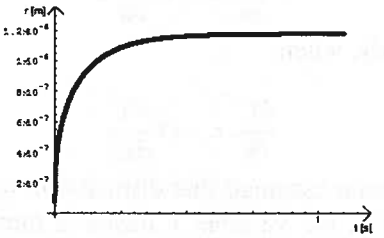

Fig. 7. The dependence of Ti_3O_5 inclusion radii of time at 1873K

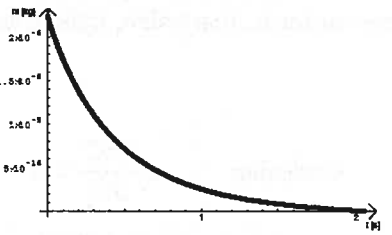

Fig. 8. Decrements of Al_2O_3 inclusion's mass at 2200K

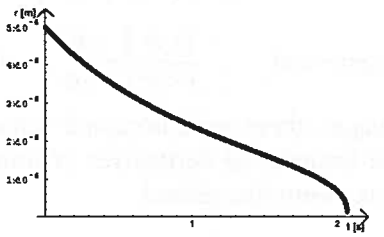

Fig. 9. The dependence of Al_2O_3 inclusion radii of time at 2200K

Calculations were finished when all oxygen in the area was 'consumed' by an precipitate (for coagulation) or the precipitate's radius became one hundred times smaller that the initial one (for dissolution).

Values of the times of precipitations' growth or dissolution calculated by the finite difference method and values calculated by Hong and DebRoy [6] are presented in Table 1.

TABLE

Values of the times of precipitations' growth or dissolution calculated by the finite difference method and values calculated by Hong and DebRoy [6]

precipitation	time [s] calculated	time [s] literature data [6]
Al_2O_3 (growth)	4	4
Al_2O_3 (dissolution)	2	2.5
Ti_3O_5 (growth)	1	1.1

5. Summary

This paper presents a numerical approach to the growth of and dissolution of precipitates calculations. The results obtained in such a way are in good agreement with values presented by Hong and DebRoy [6], that is 4 seconds for the growth, 2.5 seconds for the dissolution of Al_2O_3 precipitates and 1.1 second for the growth of Ti_3O_5 precipitates. Therefore, the method proposed in this paper, although quite simple and based only on well known physical and chemical values may be a promising one and is worth further investigations.

At the end, I would like to express my gratitude to my colleagues from the Rzeszów University for the help in operating the Mathematica program.

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