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Finite element analysis of the grain size effect on diffusion in polycrystalline materials

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ABSTRACT

Polycrystalline materials with refined grain size are well known to have enhanced diffusion properties compared to coarse grain materials. Due to their high grain boundary density, the macroscopic diffusivity of such materials is increased. Indeed, grain boundaries are fast diffusion channels in the material. In this paper, a numerical method to calculate the diffusivity of polycrystalline materials as a function of their grain size is proposed. A homogenization technique is applied on polycrystalline representative volume elements on which diffusion calculations are performed with a finite element method. This technique allows to extract the effective diffusivity of the material for different grain sizes. A relationship is then built between the diffusivity and the grain size. It is shown that the extracted diffusivity follows a mixture law of both diffusivities in the grains and grain boundaries, as proposed by Hart [14], but taking into account grain boundaries randomly oriented compared to the diffusion direction.

1. Introduction

In the recent years, surface nanostructured materials are of great interest to enhance mechanical parts. Regarding their mechanical properties, it has been shown that they provide improved hardness, fatigue resistance and wear resistance [1–3]. They also improve the efficiency of thermochemical treatments such as nitriding by increasing the hardness [4–8] and the nitrogen penetration depth [5,6,9]. Moreover, the nitriding temperature [5,7,8] is reduced. Likewise, the friction coefficient, the wear [4–8] and the corrosion [5] are also reduced. The main explanation given for a such efficiency improvement is that grain boundaries act like fast diffusion channels. Then, when the grain size decreases, the grain boundaries multiply, as well as the fast diffusion channels. Thermochemical treatments are usually described by the diffusion equation, which is a combination of both Fick's laws:

$$\frac{\partial c}{\partial t} = div\left(D\overrightarrow{grad}(c)\right) \tag{1}$$

where $c\left(\vec{x},t\right)$ is the concentration of the diffusing element at point $\vec{x}$ and time t, and D is its diffusivity in the material. In this formulation, no thermodynamic interactions are taken into account and one diffusing element is considered. The term D is then a macroscopic value of the diffusivity which takes into account both diffusion processes in grains and along grain boundaries. At a smaller scale, these processes can be separated in two single diffusion processes occurring in two different media with different diffusivities: D_g for the diffusion inside the grains and D_{gb} for the diffusion along the grain boundaries. It is possible to find some numerical values for diffusivities of both grains and grain boundaries in the case of self-diffusion [10–12]. Table 1 gives an example of both grain and grain boundary diffusivities for aluminum in the case of self diffusion at 400 °C. According to Harrison's classification [13], in the case of *type A diffusion kinetics*, Hart has proposed an analytic expression for the effective diffusivity [14] with grain boundaries parallels to the diffusion direction.

$$D = f_v D_{gb} + (1 - f_v) D_g \tag{2}$$

This relations is similar to the Wiener upper bound for the real effective permittivity in multicomponent media [15] and to the upper Voigt bound in elasticity [16]. f_v is the volume fraction of grain boundaries and D the effective diffusivity which can be used in the diffusion Eq. (1). The volume fraction of grain boundaries f_v can also be written

$$f_v = \beta \frac{\delta}{d} \tag{3}$$

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Table 1
Aluminum self diffusion coefficients at 400 °C for grains and grain boundaries [10].

	Brown and Ashby [11]	Gust et al. [12]
D_g (mm^2/s)	1.8×10^{-11}	2.33×10^{-11}
D_{gb} (mm^2/s)	1.84×10^{-5}	6.27×10^{-5}

where δ is the grain boundary thickness and d is the grain size. β is a numerical factor depending on the grain shape. Eq. (2) was extended to the Hart–Mortlock equation [17] which takes into account segregation at grain boundaries. Kalnins et al. [18] has also developed a model which describes diffusion in a two-phase material with segregation effects. The combination of these two equations has been performed by Belova and Murch [19] to build an analytical model of the effective diffusivity in a 2D cubic shaped polycrystal. Also, some authors studied the grain boundary diffusion effect on the effective diffusivity with numerical methods. Gryaznov et al. [20] and Bassman et al. [21] have made a finite element analysis considering two different media: grains and grain boundaries. This type of methodology has also been applied with the finite difference method [22] or by computation of the effective conductivity of two dimensional disordered random Voronoi networks [23] with Kirchhoff equations resolved by Lanczos algorithm [24]. The aim of this work is to expand Hart's equation to a 3D media where the grain boundaries are randomly oriented regarding the diffusion direction with help of an homogenization technique compatible with representative volume elements (RVE) as shown in Fig. 1. This technique enables to extract the effective diffusivity of the considered RVE. The second step of this work stands on a numerical study of flux dissociation in grains and along grain boundaries. It will lead to formulate the effective diffusivity as a weighted arithmetic average as expressed by Hart. The ponderation coefficient of this expression will be numerically measured on multiple representative volume elements and linked to the grain size.

2. Homogenization technique

To calculate the effective diffusivity in general cases, it is not possible to take the average of the different local diffusivities as

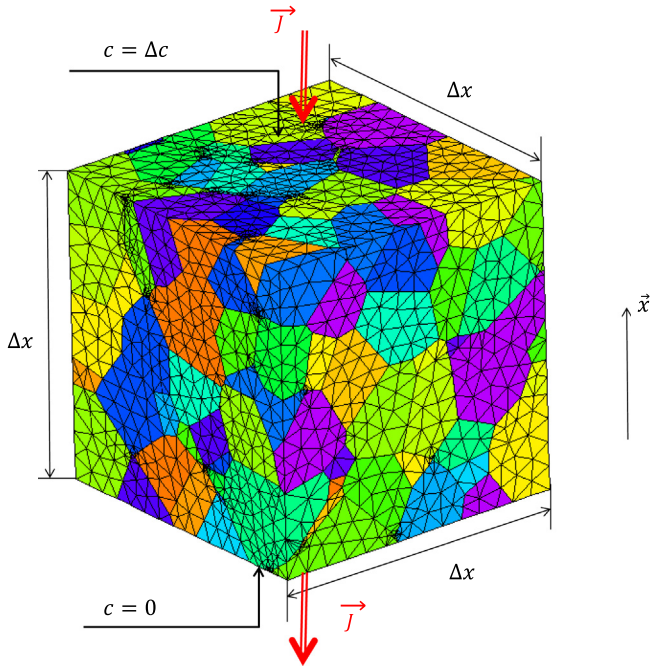

Fig. 1. Example of a representative volume element with 250 grains generated by the software Neper [25].

in perfect interfaces described by Hart's model. The homogenization technique presented here aims at giving a method to extract an effective diffusivity D from a diffusion simulation on a cubic RVE by means of the finite element method. Let us consider a cubic RVE of Δx square under a concentration difference Δc on two of its opposite faces. This RVE can be considered as a heterogeneous material constituted of grains and grain boundaries. Because the concentration is prescribed on two opposite faces of the cube, the diffusion occurs in the direction perpendicular to these surfaces. Let x designing the considered point on this direction. At a local scale, the flux density through the RVE is given by

$$\vec{J} = -D(x)\frac{\partial c(x)}{\partial x} \tag{4}$$

where $D(x)$ can be either D_g or D_{gb}. Then at steady state, considering this cube as a homogeneous of diffusivity D, the concentration gradient along the RVE can be expressed as the concentration slope

$$\frac{\partial c(x)}{\partial x} \approx \left\langle \frac{\partial c(x)}{\partial x} \right\rangle = \frac{\Delta c}{\Delta x} \tag{5}$$

where the brackets denote a global average on the whole RVE. Then, by integrating the flux density on the whole incoming or outcoming surface of the RVE, where it is assumed to be constant due to the homogenization hypothesis, the total flux Φ is given by

$$\Phi = -D\frac{\Delta c}{\Delta x}S \tag{6}$$

with $S = \Delta x^2$ is the section of the cube perpendicular to the diffusion direction. The diffusivity is then linked to the concentration difference Δc, to the side of the cube Δx and to the flux of chemical element Φ which is easily measurable from a finite element calculation. The effective diffusivity can be deduced by performing a diffusion simulation on a cubic RVE under a concentration difference at steady state and by measuring the flux on one of its two surfaces where it passes through.

$$D = \frac{-\Phi}{\Delta x \Delta c} \tag{7}$$

3. Finite elements model

3.1. Generation of representative volume elements with equiaxed grains

To generate the cubic RVEs, the software Neper [25] has been used. This software enables to generate such RVEs by adjusting the side of the cube Δx and the number of grains in the cube N, so the grain size can not be set directly. The grain shapes are obtained with a Voronoï tesselation. These RVEs are produced as a mesh of the grain and the grain boundaries, where grains are meshed with 3D elements and grain boundaries are meshed with 2D elements.

3.2. Diffusion model

To simulate the diffusion at steady state, calculations are performed with the software Sysweld® [26]. The problem is solved using Eq. (1) and two different diffusivities, D_g and D_{gb} are affected to grains and grain boundaries respectively. A thickness δ is affected to the grain boundaries in order to model the volumic diffusion along them. The grains are considered to be equiaxed, due to their generation method. Their crystallographic orientation is not taken into account.