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A numerical model coupling diffusion and grain growth in nanocrystalline materials

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ABSTRACT

The optimization of material properties by tailoring nanoscale microstructure is garnering great attention from both material scientists and manufacturing engineers. One promising research direction is to apply conventional diffusion-based techniques such as nitriding, carburizing, coating and sintering to nanocrystalline materials to achieve unprecedented mechanical and chemical properties. Numerical methods to facilitate the experiment endeavor, however, are rare due to the extreme difficulty in dealing with the nanoscale structure evolution. In this work, a numerical scheme considering both grain growth and diffusion in the nanocrystalline structure is proposed. The diffusion along grain boundary and inner grain is modeled by a Cellular Automata (CA) method. The proposed CA method is stable, insensitive to mesh size, and most importantly numerically efficient, making it suitable for the simulation of long-time, large-scale manufacturing processes. This CA model is then integrated with a Monte Carlo (MC) algorithm to model grain growth, which typically accompanies the diffusion process. As a showcase, the integrated model is then applied to simulate the nitriding process in nanocrystalline iron. The results show that grain growth and impurities have an innegligible effect at the elevated temperature for nanocrystalline iron, consistent with experimental observations. The developed method has the potential to serve as a simulation engine for any diffusion-controlled manufacturing processing of nanostructured materials, enabling a numerical framework to establish the processing-microstructure relation.

1. Introduction

Compared with coarse-grained polycrystalline materials, nanocrystalline materials exhibit unique properties such as improved strength and hardness, higher specific heat, and superior soft magnetic properties. It is also recognized that diffusion along grain boundaries is much faster than grain interior, about 4–8 orders of magnitude higher in diffusivity [1,2]. As a result, nanocrystalline materials [3], which contain abundant grain boundaries [2], give rise to much higher diffusivity compared with their coarse-grained counterparts [2,4–8]. The unique properties of nanocrystalline materials, in conjunction with the greatly enhanced diffusivity, inspire researchers to apply diffusion-based processing techniques including sintering [9], coating [10], nitriding [11] and carburizing [12] to process nanocrystalline materials, aiming to achieve unprecedented properties with significantly improved processing efficiency. For instances, with the nanocrystalline structure formed on the surface of the aluminum alloy, the Si_3N_4 coating after the plasma electrolytic oxidation treatment is much denser, thicker and has better tribology properties compared with those using coarse-grained samples [10]; the gas nitriding temperature was substantially decreased from ~600 °C to 300 °C when using nanostructured iron [13]. Although it has been demonstrated that the idea of applying diffusion-based processing to nanocrystalline materials is surprisingly promising, the explanations are usually phenomenologically based and it is difficult to reach a quantitative relation for processing-microstructure-performance. It thus needs a simulation model to bridge the gap. The difficulty to develop such a model, however, largely lies in the multi-physics and multiscale nature of the problem, which will be detailed next.

The multi-physics nature is reflected in the fact that the diffusion process in nanocrystalline materials is usually concomitant with grain boundary migration [5]. Most grain boundary diffusion models assume static grain boundary (ignoring grain growth) [14] use a fixed grain boundary migration velocity [15]. However, with elevated temperature or external stress or strain, the grain boundary migration in crystalline material is inevitable. Even in a well-annealed polycrystalline, the grain boundary migration can still be observed in the radiotracer self-diffusion experiment [16]. When the grain size decreases to several nanometers, grain migra-

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tion exerts a more notable effect on diffusion even at room temperature [17,18]. On the other hand, it is unphysical to simply assume a constant grain migration rate. The grain growth kinetics could be parabolic (coarse grain) [19] or even more nonlinear (nanocrystalline) [20,21]. Moreover, diffusion might affect grain growth reversely [22], resulting in a challenge to isolate the two physical processes. As a consequence, many experimental studies are deliberately designed to low temperature range [5] or use thermally stable material [6–8] to avoid grain growth.

The physical processes span over a few scales from nanoscale diffusion along grain boundaries, to grain growth up to the micro scale. To gain physical insight into the mechanisms underlying the diffusion-controlled modification of nanocrystalline materials, it is urgent to develop a numerical model that is able to consider the effect of nanostructure, and meanwhile efficiently capture the large scale diffusion and grain growth. The key is to devise a numerically efficient algorithm to track grain interface. Generally speaking, interface tracking methods can be divided into two categories [23], the "sharp interface method" [23], which ignores the interface thickness, and the "smooth interface method" (nonzero interface thickness). To model diffusion in nanocrystalline materials, the grain boundary thickness needs to be considered since the grain boundary thickness (around 0.5 nm [24]) is comparable to grain size. Therefore the "smooth interface method" should be used. To model the interface with finite thickness, the mesh size needs to be finer than the interfacial width (at least 2–3 times of mesh size [23]). This could lead to an extremely fine mesh and exponentially increase the computational intensity. In our previous work, the Cellular Automata (CA) model has been proved effective in tracking the nanostructure interface [25,26]. Because of the simplified grain boundary, the mesh size can be larger than the grain boundary thickness, resulting in a substantially reduced computation time.

In this work, a grain growth model based on Monte Carlo (MC) algorithm [19] will be integrated with the CA diffusion model [25,26]. This model, for the first time, considers grain growth and diffusion in nanocrystalline materials. In addition, the impurities, which are immobile and have zero diffusivity of solute, are considered as well. The nitrogen diffusion in nanocrystalline iron is then studied and the effects of grain growth and impurity on diffusion are discussed based on the simulation results. This work provides a numerical strategy to study microstructure evolution in nanocrystalline materials.

2. Model description

2.1. Grain structure discretization

The first step of the simulation is to discretize the nanocrystalline structure and couple the two physical processes, diffusion and grain growth, on the discretized structure. One inherent issue is that grain growth and diffusion occur at two scales. The thickness of the elementary channel for diffusion, grain boundary, is around 0.5 nm, whereas the typical grain boundary migration distance can be a few orders of magnitude larger. For example, the grain size of nanocrystalline Cu during a tensile test at room temperature can be increased from 30 nm to 600 nm [27]. With the vast size gap, a numerical scheme to efficiently capture the small scale diffusion is essential for the coupling. To circumvent the issue, a simplified grain boundary, as shown in Fig. 1, will be applied to bridge the scale gap [25,26]. According to the Fisher's model [24], the grain boundary is treated as a thin slab embedded in the grain mesh with a predefined thickness. The grain structure is obtained from the Voronoi diagram [28] shown in Fig. 1. In our work, the grain boundary is always placed at the center of the mesh and the shapes of grain boundary could be straight or zigzag. One can notice that the mesh size is actually larger than the thickness of the grain boundary. The treatment of grain boundaries as center-placed slabs will significantly improve the computational efficiency, making it possible to integrate diffusion with grain growth. The validity of this simplification will be carefully investigated in this work.

For grain growth, the polycrystalline structure is represented as a set of discretizing cells with different orientations. The cells in one grain will share the same orientation. The grain boundary lies between cells with different orientations. Considering that we place grain boundary at the center of diffusion, it is thus natural to shift a half mesh size between grain growth mesh and diffusion mesh as shown in Fig. 2.

In addition to grain boundary, there is another type of interface, i.e., the interface between impurities, which is considered as immobile and have zero diffusivity of solute in this work, and the substrate matrix. The impurities are randomly generated in the simulation zone and their orientations are labeled as negative values. The treatment of impurities interface is the same with the grain boundary discretization. With this hybrid discretization scheme, the coupling of grain growth and diffusion calculation can be realized.

2.2. Diffusion model

In this work, as grain boundaries play important roles in the diffusion of nanocrystalline material, the CA model is introduced to track the location of grain boundaries. Finite volume method is combined with the CA model to calculate the diffusion. The CA method has been widely used as a robust and stable numerical tool for multi-physics modeling [29–35]. A typical CA model consists of three fundamental elements: cells, neighborhood relation, and evolution rules. Cells in CA method refer to the discretized mesh, which could have multiple "states" to represent physical attributes like temperature, concentration, grain orientation and etc. In this work, three kinds of states are defined as follows.

(1) Cell structure, a variable to describe the local microstructure of a cell. It has three possible states: matrix (M), grain boundary (GB) and impurity. The impurity cells are randomly generated at the beginning of the simulation with a predefined density. For the rest cells, the GB states will be attributed to the cells with grain boundaries across them, otherwise, the M states.
(2) Average solute concentration in this cell;
(3) Temperature.

The second element in the CA method is the neighborhood relation, which is the channel to connect these discrete cells together. The cell state in the next time step is a function of the states of its neighbor cells. Two of the most widely used neighborhood relations, Moore neighborhood and von Neumann neighborhood [36], are shown in Fig. 3. The diamond-shaped von Neumann neighborhood is used in the diffusion calculation in our previous work [25,26], while the Moore neighborhood is widely used in grain growth model [19,37,38].

The third element of the CA model is evolution rule. A number of evolution rules will be established based on the physical processes. After defining the neighborhood relation, the states of cells will evolve to a new state at the next time step based on the states of its neighbor cells and the evolution rules. Recall that we have already simplified the grain boundary to be either straight or zigzag. Based on the simplification, there are only six possible cases between any center cell and its neighbor cells (except the impurity cell). The six cases are illustrated in Fig. 4(a)–(f). The black slab in