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Grain-resolved kinetics and rotation during grain growth of nanocrystalline Aluminium by molecular dynamics

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ABSTRACT

Grain growth in nanocrystalline Al was studied by means of molecular dynamics simulations. The novelty of this study results from the utilization of an algorithm to resolve per-grain kinetics and orientation change from molecular dynamics data sets. To this aim, a highly efficient algorithm for the identification and reconstruction of crystallites from molecular dynamics data sets of FCC materials was developed. This method is capable of calculating specific attributes of grains, namely, volume, center of mass, average orientation and orientation spread. In addition, it provides a mapping method to track grains during time-row data sets. In the present contribution, we describe and validate the algorithm, which is then used to analyze grain growth in polycrystalline Al with a weak texture. For the conditions tested, the algorithm was able to find all of the input orientations and reconstruct the grains according to their crystallographic orientation. With the help of the developed algorithms, we studied grain growth kinetics and grain rotation. The results of the simulations showed slightly slowed-down kinetics in particular in the initial stages of grain growth and marginal rotation of the grains.

1. Introduction

1.1. Grain growth in nanocrystalline polycrystals

Nanomaterials have become an important source for the development of new technologies due to their extraordinary properties that allow their application in fields where conventional materials fail. Among other important technological fields, nanomaterials have been utilized in medicine (drug delivery), electronics (circuit miniaturization), aerospace industry (improved fatigue-resistant materials) and many other applications. Nevertheless, due to the high density of crystal defects per unit volume in polycrystalline nanomaterials, they are in such a state of thermodynamic non-equilibrium that provides, even at room temperature, a tremendous driving force for the elimination of the crystal defects and thus, for the transition from nano- to micro-sized grained materials. Hence, the thermal stability of nanomaterials is of utmost importance for their design and development.

The thermal stability of nanocrystalline materials is still a scientific conundrum owing to the intrinsic difficulties of investigating opaque materials with nanoscaled grains and even smaller microstructural features. Nanomaterials are prone to microstructural change because they own a very high free energy ΔG stemming from their large density of crystal defects in particular grain boundaries. Nevertheless, a polycrystal is not exclusively composed of grain boundaries. The junctions that connect grain boundaries, namely triple lines and quadruple junctions, also form part of the polycrystalline structure and affect the microstructural evolution under certain conditions. For mesoscopic grain sizes, the volume occupied by grain boundaries exceeds by orders of magnitude that of other structural elements. However, this difference becomes insignificant in nano-sized materials as triple lines, being the most frequent topological element, can occupy as much volume as grain boundaries in a microstructure. The contribution of these components to the free energy is so high that most rapid grain growth is expected and also usually observed [1–3].

Different approaches have been suggested for the stabilization of the microstructure in nanomaterials [4–6]. There are basically two approaches to achieve thermal stability in nanocrystalline materials, namely, the *kinetic* and the *thermodynamic approach* [5]. In the former, it is sought to hinder grain boundaries by incorporating obstacles for their migration [7–24]. In this context, there is also available literature that seems to suggest that intrinsic phenomena to the grain boundary structure might aid thermal stability during the normal development of the microstructure [25–29]. The second approach relies on the decrease of the free energy

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caused by the segregation of solute atoms to grain boundaries [30–33].

1.2. Simulation of grain growth in nanocrystalline polycrystals

Different computer models have been utilized in the past to simulate grain growth in nanocrystalline materials. Most of these simulations have been performed by mesoscopic models [34–37,7–9,38,10–12,39–41]. These models rely on the properties of the structural elements of a microstructure, namely, the grain boundaries, the triple lines and the quadruple junctions. Any effect acting on these elements can be separately incorporated to the models. The advantage of these models is that they allow the simulation of relatively large representative volume elements (RVE) that can be composed of thousand to million grains and thus, the RVEs are statistically representative. The disadvantage of mesoscopic models is that the effects and physical mechanisms are predefined and pre-implemented. This is useful to test theories by isolating particular effects that are expected to affect considerably the evolution of a system but for obvious reasons, it is impossible to resolve the mechanisms of phenomena acting at the atomic scale. Nevertheless, a combination with experiments or atomistic simulation is most useful to complement the models and/or understand particular aspects of the experiments.

By contrast, atomistic simulations allow the determination of the mechanisms of the phenomena involved. In the case, of molecular dynamics simulations empirical interatomic potentials are utilized to describe atomic interactions so far with great success. The disadvantages of atomistic simulations is that the volume that can be simulated in a reasonable time is very limited compared to mesoscopic simulations. This results occasionally in insufficient statistical representativeness and undesired effects of the size of the simulated volume. Regarding grain growth, molecular dynamics have been already employed to simulate this phenomenon. In particular, the seminal work by Farkas et al. [42–47], Haslam and Yamakov [48–50], Holm and Foiles [27] and Srolovitz et al. [51,26,52–54] have substantially advanced our knowledge on the mechanisms of grain growth in nanocrystalline materials. The purpose of the present manuscript is to identify particular mechanisms of grain growth in nanocrystalline materials. For this purpose, we utilized molecular dynamics simulations and developed novel characterization methods for the evaluation of computational microstructures from molecular dynamics data sets.

1.3. Post-processing of molecular dynamics

In molecular dynamics (MD) simulations, the analysis and interpretation of the data remain a substantial and difficult part. Certain tools are required in order to reduce the amount of information and obtain useful quantities. In several situations, information on the evolution of 3D features is necessary, for example, grains in polycrystalline aggregates. This information is, however, not delivered by most of the available methods for the characterization of MD data sets. This task remains difficult because it involves identifying crystallographically the environment of the atoms to assign an orientation respective to a coordinate system and to reconstruct from this information the volume with identical orientation i.e., the grains. Due to the mathematical complexity of rotations and reconstruction algorithms, these calculations are slow and costly for their execution during runtime and even during post-processing if large data sets are involved. Nevertheless, several methods which enable the identification of crystal structures from atomic-based data have been already developed. The most straightforward method to distinguish between some of these is to compute the coordination number, i.e. the number of nearest neighbors (NNs) or next-nearest neighbors (NNNs) of the atoms. While this is a fast tool and also easy to implement, it obviously cannot distinguish between different atomic structures exhibiting the same coordination number. Another technique which allows a much more reliable identification of crystalline structures was first introduced by Honeycutt and Andersen [55]. This method analyzes the extent and connectivity properties of atom-diagrams which are comprised of nearest neighbors to two adjacent atoms. Based on this principle, also known as Common Neighbor Analysis or CNA, several algorithms have been implemented [56–58] and successfully performed to MD simulation data. Similarly, the centro-symmetry parameter, energy filters, bond order and angle and Voronoi analysis can also be used to discriminate atoms arranged in specific crystal structures [56]. Nevertheless, none of these methods can distinguish between groups of atoms with the same crystallographic orientation. To differentiate between differently oriented crystals, the definition of order parameters has been utilized. The drawbacks of this approach are that the orientations ought to be known beforehand and that complex rotations of the crystals might out-range the scope of the parameter. Nevertheless, this approach has been successfully utilized in numerous simulations to track, for instance, grain boundaries [59,52,53,60,61] and grain rotation [62–66].

The motivation of the present contribution is to introduce a method that allows the determination of the relative orientation of groups of atoms and reconstruct the grain from only this information. The method is able to generate a space-resolved three dimensional grain decomposition of atom-position data sets for FCC materials, such as those generated by MD-simulations. The first step of the method calculates an orientation for each atom by taking into account the positions of the nearest-neighbor atoms. In a second step, the method generates grain entities from atoms with similar orientations by collecting them via nearest-neighbor paths. The method is capable of calculating grain specific properties, namely volume, center of mass, average orientation and orientation spread. The method additionally provides a mapping method in order to track grains during a time-row data set. The method is introduced in order to be able to track the grain growth evolution of individual grains during MD-simulations, additionally enabling a grain-resolved visualization of polycrystalline FCC data sets. Whereas the algorithms here presented were independently developed, they are admittedly similar to the method introduced by Panzarino et al. [67,68]. However, we emphasize that in contrast to these previous contributions, we developed our algorithms for high-performance post-processing and that we offer the code to the community as a totally open-source project [69].

2. Methods

To identify the grains from a data set containing atomic positions, it is necessary to determine first the orientation of the atoms as a per-atom attribute from the local neighborhood. Once an atom (referred to as *central atom*) is selected arbitrarily, the calculation of the orientation proceeds in the following way:

1. Identify all the nearest neighbors and calculate the relative positions of the neighbors to the corresponding central atom.
2. Determine the affine 3×3 transformation matrix M from the reference (non-rotated case) to the current crystal-configuration.
3. Determine the rotation that best fits M as a least square solution.

All of these steps have an inherent mathematical and/or programmatic complexity. In the present section, we will discuss the algorithms that were utilized to solve each of these problems. In