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Nanocrystalline gradient engineering: Grain evolution and grain boundary networks

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ABSTRACT

We study materials with spatial gradients in nanoscale grain size (5–120 nm), and quantitatively examine the effect of spatial gradient on microstructure evolution and thermal stability using mesoscale Monte Carlo modeling and statistical analysis. The spatial grain size gradient weakens and the grain size distribution widens at elevated temperatures, accompanied by grain rounding and movement of grains along the gradient direction. Introducing heterogeneous grain boundary networks into gradient materials leads to better preservation of the spatial grain size gradient but less equiaxed grains. Coarsening in small grain regions is accompanied by an increase in the local fraction of low-energy grain boundaries, as these are competing mechanisms for reducing total energy, and spatial gradients in grain boundary character distribution and triple junction character distribution develop in the material. We further compare concave, linear, and convex gradient materials with increasing grain size gradient for small grains. Grains in convex gradient materials have the highest grain growth rate compared to grains of the same size in linear and concave gradient materials. The accelerated grain growth in the presence of a steeper grain size gradient is attributed to a change in local grain neighbor environment that promotes grain boundary curvature (and pressure) and enhances the driving force for grain growth.

1. Introduction

Nanocrystalline materials usually exhibit high yield strength and hardness but limited tensile ductility [1–4]. There is rarely a single grain size that simultaneously optimizes strength and ductility. Recently, nanocrystalline materials with bimodal grain size distributions [5] and with spatial grain size gradients [6–9] have been explored, with the aim to optimize competing properties. Surface nanocrystalline materials [6,7,9], in which the grain size increases from nanocrystalline (e.g., 20 nm [7]) at the surface to microcrystalline at the core (e.g., 10 μm beyond a depth of 200 μm [7]), have been fabricated by various surface deformation techniques [9–15]. Synergistic strengthening in graded regions [16], extensive strain hardening [17], good tensile ductility [18–21] and fatigue resistance [22–24] have been demonstrated. Patterned nanocrystalline materials, such as Ni-W electrodeposits with grain size gradually decreasing from 70 nm to 20 nm over a distance of 100 μm, multilayered structures with alternating 10-μm-thick layers of 70 nm and 7 nm grain sizes, as well as nanocrystalline-amorphous composites of Al-Mn [25,26], have been fabricated by modulating solute concentration during electrodeposition [8,25]. Nanoindentation of a Ni-W sample with grain size graded from 90 nm at the top to 20 nm at the bottom revealed higher pile-up than alloys of either grain size, possibly due to the yield strength gradient that led to larger plastic strains near the indentation impression [27,28]. Spatial grain size gradients were also introduced into shape memory alloys for functional grading as martensitic transformation temperature and stress are grain size dependent, often by nonuniform grain growth under surface laser annealing of nanocrystalline NiTi films [29].

Nanocrystalline materials are generally susceptible to microstructure evolution due to the high density of grain boundaries in them [30–34]. For example, the grain size in nanocrystalline Cu increases from 43 nm to 70 nm after 5 h annealing at 100 °C and to 278 nm at 500 °C [35]. Grain growth in nanocrystalline materials may occur by grain boundary migration [36] (and also possibly grain rotation [37,38] and grain boundary sliding [39]), and depends on grain boundary energy γ [40,41] and mobility M [42]. Grain growth has also been observed in nanocrystalline materials during deformation, such as during tension [43–46], compression [44,47,48], fatigue [49,50], torsion [51–53], indentation [54–56] and creep [57–59].

In graded nanocrystalline materials, the grain growth behavior depends on the grain size distribution as well as the spatial grain

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size gradient. Hunderi et al. derived the growth rate of a bubble with size d_i [60]:

$$\dot{d}_i = \frac{\partial d_i}{\partial t} \propto \frac{M\gamma}{d_i^2}\sum_j A_{ij}\left(\frac{1}{d_j}-\frac{1}{d_i}\right) \quad (1)$$

where Σ sums over its neighbors, and A_{ij} is the contact area between bubbles i and j. When all neighbors have equal d_j, Eq. (1) is reduced to a form similar to the Hillert equation for grain growth, Eq. (2) [61,62]:

$$\dot{d}_i = \alpha M\gamma\left(\frac{1}{d_c}-\frac{1}{d_i}\right) \quad (2)$$

where α is a geometrical factor equal to 2 and 4 for two- and three-dimensions, respectively, and d_c can be viewed as a critical (or average) grain size. If d_i is below/above d_c, grain i tends to shrink/grow. A system with one [63] or several large grain(s) [64] embedded in a matrix of small grains is commonly studied, and grain boundaries in the system are usually assumed to have the same properties. In this case, the size of the initially large grain i will approach those of the matrix grains after grain growth, and $\partial[d_i(t)/d_c(t)]/\partial t$ depends on the grain size distribution. Benson and Wert compared the evolution of grain structures of different initial grain size distributions characterized by d_{max}/d_c, where d_{max} is the largest grain size, and $f_{d>2d_c}$, the fraction of grains with $d > 2d_c$, and concluded that higher d_{max}/d_c and $f_{d>2d_c}$ enhance the initial $\dot{d}_c$ of the system [64]. Moreover, the Grain Boundary Character Distribution (GBCD) may affect the evolution of the spatial grain size gradient, and vice versa. A fundamental understanding of concomitant nanocrystalline grain size gradient evolution and GBCD evolution is currently lacking.

Modeling is often performed to gain quantitative insights on microstructure evolution. Molecular Dynamics simulations have been performed to study grain evolution in nanoscale systems under thermal [65,66] or mechanical [59,67] stimuli. There are also several common mesoscale methods for simulating microstructure evolution at larger length and time scales, such as the vertex model [68], the phase field model [69], the cellular automata model [70,71], the Monte Carlo Potts model [72–76], and hybrid models combining two of these approaches [77,78]. Among them, the Monte Carlo model, which is a discrete stochastic simulation method [79,80], has been widely used to study microstructure evolution in nanocrystalline materials [81]. For example, it was modified to incorporate the effect of triple junctions [82,83] or to study the dominant grain growth mechanism [84]; it was also used to model grain growth in dual-phase nanocrystalline materials [85].

Although microstructure evolution in nanocrystalline materials with a uniform grain size has been studied experimentally and computationally, nanocrystalline grain size gradient effects and evolution in graded nanocrystalline materials are not quantitatively or mechanistically understood. In this paper, we study thermally-driven microstructure evolution in nanocrystalline metals with spatial grain size gradients using the Monte Carlo modeling method, aiming to elucidate the influence of spatial grain size gradient and GBCD on the concurrent evolution of grain gradient and GBCD at the nanoscale.

2. Simulation procedures

2.1. Generating graded nanocrystalline grain structures

Our modeling systems are two-dimensional (2D) nanocrystalline materials with various types of spatial grain size gradients along the y direction, which resemble gradient regions in surface nanocrystalline materials. For simplicity, other microstructural gradient (such as dislocation density) that might develop during processing is not considered. As illustrated in Fig. 1(a), in the 600 nm by 1200 nm systems, the gradient region is sandwiched between two layers with uniform grain sizes of d_1 = 5 nm at the top and d_2 = 120 nm at the bottom. Grain structures are generated using Voronoi tessellation [86], where each grain is constructed based on one seed such that every location in the grain is closer to this seed than any other seeding points. The grain seeds are generated using a Monte Carlo procedure while imposing a minimum distance, d_{min}, between seeds to avoid small grain outliers and improve grain uniformity in the x direction [87]. $d_{min}(y)$ is set as $0.6d_g(y)$, where $d_g(y)$ is the desired grain size at a given y location. $d_g = d_2$ for $0 \leqslant y < 300$ nm; $d_g = d_1$ for $1100 < y \leqslant 1200$ nm; in the gradient region ($300 \leqslant y \leqslant 1100$ nm),

$$d_g(y) = \begin{cases} d_2 - 2\cdot\exp[a(y-300)] + 2 & \text{convex gradient} \\ \frac{d_1-d_2}{1100-300}(y-300)+d_2 & \text{linear gradient} \\ d_1 + \exp[b(1100-y)] - 1 & \text{concave gradient} \end{cases} \quad (3)$$

where the constants $a = 0.005086$ and $b = 0.005942$. In each of the Monte Carlo steps, a seed with random x and y coordinates is generated; the seed is kept only if its acceptance probability, $\lambda(y) = [d_1/d_g(y)]^2$, is greater than a newly generated uniform random number between 0 and 1 and its distance to the nearest seed exceeds d_{min}. λ is lower at small y, resulting in fewer seeds and larger grains at the bottom of the system. The Monte Carlo procedure is repeated until the desired spatial grain size distribution is achieved. Fig. 1(b) shows an example area in the gradient region where the "+" symbols are grain seeds.

The modeling systems are mapped to triangular lattices with a node spacing of 0.5 nm. A triangular lattice symmetry is chosen to provide a large number of neighboring nodes (i.e., six neighbors) in order to mitigate lattice effects [73]. Nodes belonging to a specific grain are assigned a unique index (see Fig. 1(c)). Grain boundaries are identified as interfaces between nearest-neighbor nodes with different grain indices (grain boundary width therefore may be considered 0.5 nm). We determine the size d of a grain from its area A, $d = \sqrt{4A/\pi}$. The average grain size at a specific y, d_y, is evaluated by performing an area average of grains with any part falling within a 10 nm-thick layer centered at y (analogous to moving average). Fig. 1(d) plots d_y at each y location in the three types of gradient structures generated using the above procedure. The convex structure exhibits the steepest spatial grain size gradient, $\partial d_y/\partial y$, at the top of the gradient region, while the concave structure has large spatial gradient near the bottom of the gradient region. $\partial d_y/\partial y$ is nearly constant in the linear gradient structure. For comparison purposes, we also run simulations on a 600 nm by 600 nm system with a uniform grain structure (i.e., without notable spatial grain size gradient) and with an initial grain size of 5 nm.

We first examine the behavior of systems where all grain boundaries are assigned the same properties. The grain boundary mobility M is set as 0.77×10^{-17} m/(s Pa), a reasonable value for metals at our simulation temperature T = 473 K [42]; the grain boundary energy γ is set as 1.6×10^{-20} J/bond (2.45 kT, where k is the Boltzmann constant). Later, we will introduce GBCD, and study systems in which grain boundaries are classified into general boundaries with $\gamma_g = 1.6 \times 10^{-20}$ J/bond (2.45 kT) and special boundaries with $\gamma_s = 1.2 \times 10^{-20}$ J/bond (1.84 kT) or 0.9×10^{-20} J/bond (1.38 kT); for simplicity, M is still assumed the same for all boundaries. The orientation of each grain is assigned by an in-plane rotation by a random angle between zero and Φ_{max}. A grain boundary is considered special if its misorientation angle is less than a threshold $\theta_t = 15°$, and otherwise is general. This binary grain boundary classification may be considered a simplification for the more general