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Particle deposition and deformation from high speed impaction of Ag nanoparticles

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ABSTRACT

The impaction of a single Ag nanoparticle onto an (001) Ag substrate was studied as a function of particle diameter (2–9 nm) and impaction velocity (10–1500 m/sec) using molecular dynamics simulations. The final crystallographic structures were observed to transition from a polycrystalline to an epitaxial morphology as impaction velocity was increased and the velocity required to achieve epitaxy increased with particle size. To understand how the crystallographic structures evolved to their final state, the deformation mechanisms were then studied over a range of time scales, beginning immediately upon impaction. The observed mechanisms included disordering of the atoms and the initiation and propagation of partial dislocations. Deformation increased with impaction velocity due to increases in the degree of disordering and the partial dislocation density. At longer time scales, relaxation of the disordered particles produced epitaxial morphologies, whereas polycrystalline morphologies were observed following incomplete disordering. These results suggest that the microstructures of thick films produced by high speed impaction of nanoparticle aerosols are strongly influenced by processing parameters.

1. Introduction

A number of high-velocity aerosol processes have been developed over the past two decades for producing thick (1–100 μm) metallic and ceramic films. One of the more commonly used methods is cold-spray deposition [1,2] where a powder is injected into a high-pressure gas and accelerated to high velocity through a nozzle. Polycrystalline films result when the particles impact onto a substrate. The aerosol deposition method is an alternative method for producing thick films [3–6] that utilizes particles that are accelerated from atmospheric pressure through a nozzle into vacuum. The impaction velocities for both processes are similar, ranging from 400 to 1200 m/s. However, the particle sizes are quite different: 100–400 nm for the aerosol deposition method and 5–25 μm for the cold-spray process. A third aerosol deposition process for producing thick films, the Laser Ablation of Microparticle Aerosols (LAMA) [7–9] process, utilizes similar impaction velocities as both the cold spray and the aerosol deposition method, but employs even smaller nanoparticles (2–40 nm) in the aerosol.

Despite the similarities in the impaction velocities between all of these aerosol processes, the final microstructures are dramatically different. For example, impaction of micron-sized particles results in thick films that are nearly 100% dense and whose grains are heavily deformed along the impact direction. In contrast, films produced using the LAMA process are ≈70% dense and the grains are equiaxed [10]. This suggests that there are significant differences in the deformation mechanisms as the particle size is reduced from the micron-scale to the nano-scale.

TEM studies [11] and finite element simulations [12] have been used to understand the deformation and film formation mechanisms that occur when micron-sized particles impact a surface at high velocity, but there has been comparatively little work on understanding the deformation mechanisms that occur for smaller particles. The large local strain gradients, rapid cooling, and lack of constitutive property data for particles in this size range make it challenging to study nanoparticles (NPs) using experimental methods or continuum-based numerical modeling. Instead, molecular dynamics (MD) simulations have been utilized to study the final states after impaction of NPs. For example, MD simulations have been used by Haberland et al. [13] to study the porosity of thin nanostructured films of molybdenum for a range of impaction

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energies. Meinander and Nordlund [14] studied the density dependence of copper films on the impacting particle's size and energy. They also studied the effects of these variables on the morphology after impaction of a single nanoparticle onto a substrate [15]. However, only the final film morphologies were emphasized, and there has been little consideration of the particle size- and velocity-dependent mechanisms of deformation that occur during and after impaction.

Ogawa [16] studied the influence of particle velocity on single particle impaction of 10 nm monoclinic zirconia particles over a range of velocities from 250 to 2000 m/sec using MD simulations. He observed no significant particle deformation for impaction velocities below 1000 m/sec, but above this velocity significant deformation occurred. At this velocity, discrete planar defects were observed that were hypothesized to be {100} ⟨110⟩ dislocations. At higher velocities, crystalline regions were separated by regions of more complex defect structures were observed that exhibited lower degrees of crystallinity. Thus, Ogawa's study demonstrated that there are significant effects of particle velocity on deformation, but a detailed study of the nature of these defect structures and how they form were beyond the scope of his paper.

In this paper, we study the deformation mechanisms that occur when a single silver nanoparticle impacts onto a silver substrate, and seek to understand how these mechanisms influence the final nanostructure of the particle. We use classical MD simulations to study a range of NP sizes and impaction velocities which match the experimentally accessible range of variables from the LAMA experiments. In the first part of the analysis, the final microstructure of the nanoparticles and the conditions that lead to them are analyzed. In the latter part, the deformation and annealing mechanisms that lead to the observed final morphologies are studied.

2. Procedure

LAMMPS (the Large-scale Atomic/Molecular Massively Parallel Simulator) was used to perform MD simulations of the impaction of the Ag nanoparticles [17]. The LAMMPS simulations were run in parallel by simultaneously solving the equations of motion of the atoms that interact with a pairwise potential on the Lonestar Linux cluster at the Texas Advanced Computing Center (TACC) at the University of Texas at Austin [18]. Two visualization packages, OVITO [19] and AtomEYE [20], were used to visualize atomic positions along planes of interest.

Nanoparticles were simulated in the size range of 2–9 nm (diameter) and the impaction velocities ranged from 10 to 1500 m/s. These ranges of sizes and velocities were selected because they represent the experimentally accessible ranges for nanoparticles produced and impacted using the LAMA process [21]. The simulation volume was 60 × 60 lattice constants in the x and y directions (the lattice constant for silver is 0.409 nm), and 80 lattice constants in the nanoparticle impaction (z) direction. The boundary conditions in all 3 directions were periodic. For smaller nanoparticles, it was possible to simulate smaller volumes with a concomitant reduction in simulation times. However, a standard volume was adopted to accommodate the impaction of the largest nanoparticles to be analyzed. The substrate was located at the bottom of the simulation volume and extended 40 lattice constants in the z direction. The substrate was oriented such that the (100) and (010) faces were parallel to the simulation box, and the z-axis was normal to the (001) face of the substrate. To consider a general case where the nanoparticle is misaligned relative to the substrate when the nanoparticle impacts, the nanoparticle was rotated by 45° about the z-axis with respect to the substrate orientation prior to initiating motion of the NP. Within the substrate, the z-direction momentum in the bottom 5 atomic rows was fixed at zero during the simulation so that atoms would not move out of the simulation volume as a result of the momentum transfer from the impaction. There was no thermal boundary condition set for the substrate base, and the substrate interface acted as a vacuum interface with respect to the periodic boundary condition imposed in the z direction.

For the simulations, the Ag atom interactions were described using the embedded-atom method (EAM) potential [22,23] obtained from the NIST Interatomic Potentials Repository Project [24]. To initiate the simulation, a spherical nanoparticle was placed near the center of the simulation volume at a height of 20–25 lattice constants above the substrate surface. Although Wulff polyhedra [25] are the equilibrium shapes for nanoparticles, nanoparticles produced by LAMA are generally spherical, and thus spherical nanoparticles were selected for these simulations [8]. The silver atoms in the substrate and the nanoparticle were then thermalized at room temperature (300 K, to emulate the experimental conditions for LAMA) for 30–40 ps, depending on the nanoparticle size. The time integration and equilibration of the system to 300 K were performed with a time step of 0.001 ps (1 fs) using Nose-Hoover style, non-Hamiltonian equations of motion on the isothermal-isobaric (npt) ensemble [26], which generated positions and velocities of the atoms at every time step. This integrator was configured to incorporate a thermostat to a 300 K bath. The time constant for temperature dissipation to the bath was selected to give reasonable cooling times (~500 ps) for the substrate, which was much longer than the NP thermalization times of less than 50 ps.

Following the thermalization, a constant velocity was imparted to all atoms in the NP. During this stage of the simulation, the angular momentum for all atoms in the nanoparticle were zeroed at every time step in order to prevent rotation of the particle during flight. Upon impaction, the atomic positions were output to a file for subsequent analysis at time steps of every 0.5–1 ps for the first 5–20 ps after impaction and then every 20–30 ps for the remaining 300–1000 ps. The relaxation time for all nanoparticles to reach their final states ranged from 300 to 1000 ps, depending on the size of the NP.

For some simulations, the atomic positions were post-processed [27] to find the bond order parameter, q6 [28], in order to obtain information about the local atomic order and to identify atomic-scale defects. For figures in which the q6 parameter is displayed, the magnitude of q6 is quantified by the color of individual atoms. Representations of the bond order parameter, q6, were obtained for the short times only. For reference, an ideal FCC crystal has a q6 value of 0.575, an ideal hcp crystal has a value of 0.485, and disordered atoms/amorphous materials have lower q6 values of between 0.1 and 0.2 [29].

We initially conducted MD simulations of a single NP impacting the Ag (001) surface at a range of velocities and NP diameters in order to study how these parameters influence the resulting film morphologies. A summary of the NP sizes and their impaction velocities that were used is presented in Table 1.

3. Results

3.1. Final morphologies

The final morphologies that were observed after impaction of an Ag NP onto a substrate at the ranges of particle sizes and impaction velocities that were studied can be broadly classified into three categories: