



Consolidation of nanometer-sized aluminum single crystals: Microstructure and defects evolutions



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ABSTRACT

Deriving bulk materials with ultra-high mechanical strength from nanometer-sized single metallic crystals depends on the consolidation procedure. We present an accurate molecular dynamics study to quantify microstructure responses to consolidation. Aluminum single crystals with an average size up to 10.7 nm were hydrostatically compressed at temperatures up to 900 K and pressures up to 5 GPa. The consolidated material developed an average grain size that grew exponentially with the consolidation temperature, with a growth rate dependent on the starting average grain size and the consolidation pressure. The evolution of the microstructure was accompanied by a significant reduction in the concentration of defects. The ratio of vacancies to dislocation cores decreased with the average grain size and then increased after reaching a critical average grain size. The deformation mechanisms of poly-crystalline metals can be better understood in the light of the current findings.

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1. Introduction

Poly-crystalline materials with average grain sizes below 100 nm usually exhibit superior mechanical strength compared with their coarse-grained counterparts [1–5]. This fact has attracted the attention of researchers who are seeking to design a new class of materials with ultra-high mechanical strength. The major challenge in this area is how to consolidate the starting nano-crystals such that their superior, nano-scale features are preserved in the resulting bulk material. Applying conventional metallurgy consolidation and sintering methods has been found to be inadequate for this purpose and consequently some other alternative approaches are being developed [1,2].

The mechanical properties of single metallic crystals have been intensively studied [6,10–12,14–18]. The deformation behaviors of the resulting fine-grained poly-crystalline metals have been explored using both experimental [1–5] and computational [7–9,13] methods. In most of the experimental studies, the applied consolidation procedures were mostly trial-and-error in nature. The computational studies, on the other hand, focused mainly on explaining plastic deformation behavior in terms of the average grain size only, regardless of the applied consolidation method.

In fact, details of the consolidation procedures, clear descriptions of the obtained microstructures, and justifications for the chosen consolidation temperatures and pressures have rarely been reported or critically discussed.

Recently, we reported on the superior mechanical strength of nanometer-sized aluminum single crystals in the size range from 4.1 nm to 40.5 nm using multi-million molecular dynamics simulations [24]. We have shown that the ultimate mechanical strength deteriorates exponentially as the single crystal size increases. We have also found that the major mechanism behind the superiority of small crystals is their continuous ability to form vacancies and to recover them through a vacancy-dislocation interaction process. On the contrary, large-sized single crystals tend to form grains at their surfaces. Based on these results, one anticipates that it would be very beneficial to consolidate very small grains to obtain bulk materials with advanced mechanical properties. In this study, we show that this is not always the case.

Here, we present a large-scale molecular dynamics simulation study on the microstructure response to different consolidation parameters, such as the initial average grain size and the consolidation temperature and pressure. We show that in poly-crystalline aluminum, the average grain size is not the only parameter to optimize; defects play a crucial role as well. Furthermore, we show that the trade-off between the average grain size and the associated density of defects leads to the the mechanical properties of the resulting samples.

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2. Computational details

In the following, we briefly describe our molecular dynamics simulation framework. We used the Embedded Atom Method (EAM) [20,21]. The selected many-body potential was developed by Mishin et al. by fitting both experimental and *ab-initio* datasets [19]. We selected this potential from other many-body potentials based on our evaluations of the predicted structures and mechanical properties of bulk aluminum. Molecular dynamics simulations were carried out using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code [22]. The computational work was carried out on the IBM Blue Gene/P SHAHEEN supercomputer at King Abdullah University of Science and Technology (KAUST). Visualization of the resulting atomistic configurations was carried out using the ATOMEYE package [23].

The initial structures were prepared as follows. Three aluminum single crystals composed of $60 \times 60 \times 60$, $80 \times 80 \times 80$, and $100 \times 100 \times 100$ unit cells were generated. The lattice constant for the employed interatomic potential was optimized to match the experimental value of 0.405 nm. The three samples were converted from the single-crystal to the poly-crystalline format by means of the Voronoi tessellation method [27]. Each sample contained 125 grains with random crystallographic orientations. We classified the five simulated samples as follows: (i) samples S1, S2, and S3 had starting average grain sizes of 6.3 nm, 8.5 nm, and 10.7 nm, respectively. These samples were consolidated at temperatures ranging from 300 K to 900 K at a fixed consolidation pressure of 1 GPa; and (ii) samples S4 and S5 both had the same starting average grain size of 6.3 nm, and both were consolidated at the same temperatures, but the consolidation pressures were different (3 GPa for S4 and 5 GPa for S5).

The simulation time step was set to 0.001 ps. Periodic boundary conditions (PBCs) were applied in the *x*, *y*, and *z* directions, to mimic an infinite bulk material. Molecular dynamics simulation of each sample comprised the following steps: (i) equilibration at the target consolidation temperature for 100 ps, employing the NVE ensemble (the number of atoms, system volume, and energy were fixed), (ii) hydrostatic compression at the target consolidation pressure for 700 ps, employing the NPT ensemble (the number of atoms, system pressure, and temperature were fixed), (iii) gradually reducing the applied pressure down to 0 GPa in 200 ps using the NPT ensemble, (iv) quenching the system temperature down to 300 K in 100 ps using the NPT ensemble, and (v) annealing the resulting structures at the room temperature for 100 ps using the NVE ensemble. It should be emphasized that no ad-hoc method to adjust pressure, such as Berendsen barostat, was used.

The microstructure of the consolidated samples was accurately analyzed as follows. First, high resolution X-ray diffraction patterns were computed from the atomistic configurations, obtained from molecular dynamics simulations, using the Debye scattering formula [25]. The crystallite size and nonuniform atomic level strain values were calculated using the XPOWDER code, employing different size-strain analysis approaches to capture the most accurate values of grain sizes. The values of grain sizes reported in this paper are the averages over the different size-strain methods.

3. Results and discussion

We begin by reporting our molecular dynamics results. In Fig. 1, we compare the final atomistic configurations obtained from consolidating samples S1 (upper panel) and S3 (lower panel) at 300 K, 700 K, 800 K, and 900 K, both at the same consolidation pressure of 1 GPa. The orange and blue spheres represent, respectively, the atoms belonging to the interior of grains (i.e., the FCC structure) and structural defects including the grain boundary regions. The

images on the upper panel have been magnified for the sake of clarity. In these images, we see that the relationship between grain growth and temperature is not linear in either sample. The effect of the starting average grain size (6.3 nm for sample S1 and 10.7 nm for sample S3) on grain growth is also obvious in this figure. In fact, grains in the sample with smaller initial grain sizes grow much faster compared to the ones in the sample with larger initial grain size. This effect is so strong that the average grain size for sample S1 (13.5 nm) consolidated at 900 K is almost as large as the one of sample S3 (14.0 nm) consolidated at the same temperature and pressure.

Fig. 2 presents a quantitative description of the evolution of the microstructures as a function of the consolidation temperature, pressure, and the starting average grain size in nanometer-sized poly-crystalline aluminum. We first consider the effect of the starting average grain size and consolidation temperature on this evolution, as reported in Fig. 2(a). This figure shows that the growth of the grain size is not a linear function of temperature. In fact, the dependence of grain size on the consolidation temperature is successfully fitted by simple exponential growth functions (the fit is shown by the blue lines in Fig. 2(a)). The rate of this exponential growth depends on the starting average grain size. In other words, starting with a larger average grain size will hamper grain growth especially at high consolidation temperatures. As a matter of fact, samples S1 and S2 could reach almost the same grain size after hydrostatic consolidation at 900 K. This means that the choice of consolidation temperature is a very crucial step in the consolidation of nano-scale single metallic crystals, particularly if we are interested in keeping the average grain size in the bulk product below a certain value.

The effect of consolidation pressure on grain growth is shown in Fig. 2(b), where we start from the same average grain size (6.3 nm) to determine the effect of consolidation pressure on the grain growth process. In agreement with Fig. 2(a), all the curves are fitted by exponential growth functions (shown by the blue lines). It is apparent that increasing the consolidation pressure suppresses the grain growth rate. In fact, applying hydrostatic pressure of 1 GPa at 900 K leads to an average grain size of 13.5 nm, while applying pressure of 5 GPa at the same temperature leads to an average grain size of 11.2 nm. This behavior of pressure is expected, since increasing the hydrostatic pressure would increase the melting point of the nano-crystalline aluminum system [26] and therefore would reduce the grain growth rate at any given temperature.

When optimizing the consolidation process of metal nano-structures, the microstructure is not the only aspect to consider and optimize. The situation is made complicated by the presence of structural defects, such as vacancies and dislocation cores, which are very important in determining the mechanical properties of the resulting material. We now focus on the evolution of the structural defects as a function of consolidation temperature, pressure, and the starting average grain size in nanometer-sized poly-crystalline aluminum. Fig. 3 reports a quantitative description of this evolution. Vacancies and dislocation cores are structurally defined as aluminum atoms having coordination numbers of 11 and 13, respectively. Fig. 3(a) and (b) show, respectively, the concentration of vacancies and dislocation cores as functions of the initial average grain size as well as the consolidation temperature and pressure.

From Fig. 3(a) and (b), it is clear that the grain growth caused by the consolidation process is accompanied by opposite behaviors by the structural defects. It is seen that the concentrations of vacancies and dislocation cores both decrease with increasing initial average grain size and consolidation temperature. It is also clear that the effect of increasing the consolidation pressure (shown as empty circles in Fig. 3(a) and (b)) is to increase the concentration of vacancies and dislocation cores in the consolidated material. It

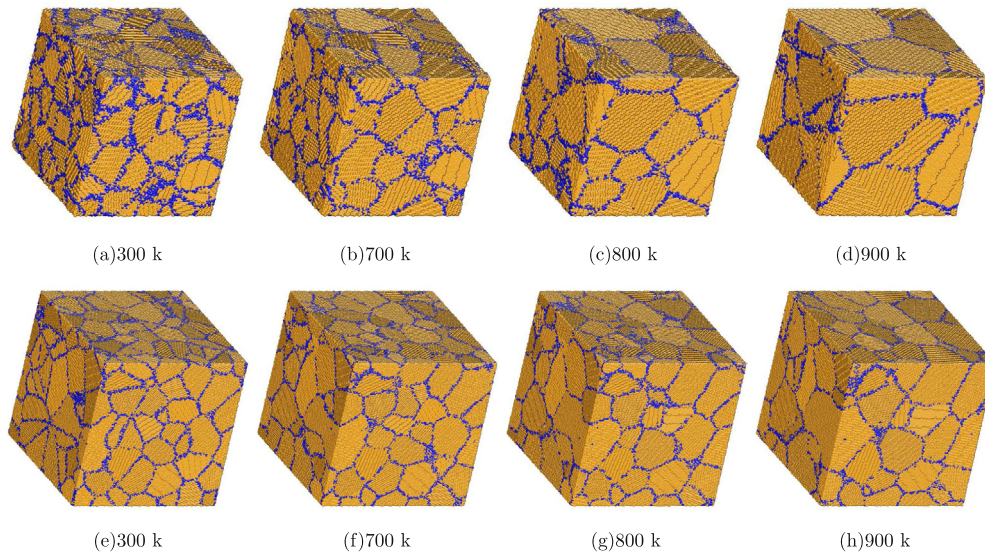


Fig. 1. Atomistic configurations resulting from hydrostatic consolidation of aluminum single crystals with initial grain sizes of 6.3 nm (upper panel) and 10.7 nm (lower panel) at different temperatures. The consolidation pressure was fixed at 1 GPa for all temperatures. The orange and blue spheres represent, respectively, the atoms belonging to the interior of grains (i.e., the FCC structure) and structural defects including the grain boundary regions. Images in the upper panel were magnified for the sake of clarity. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

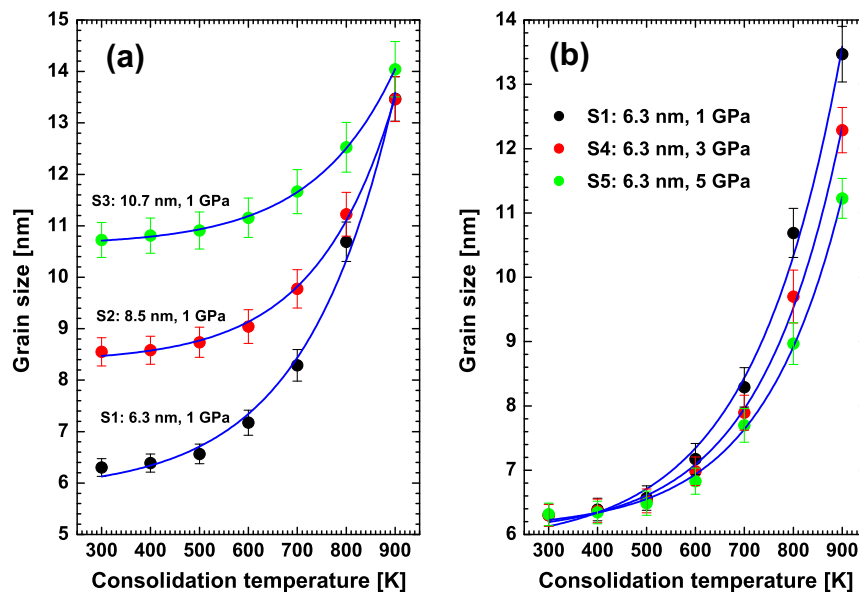


Fig. 2. The dependence of grain growth on consolidation temperature, pressure, and the starting average grain size in nanometer-sized poly-crystalline aluminum: (a) consolidation at different temperatures starting from different average grain sizes; and (b) consolidation at different temperatures and pressures. Grain size values (solid circles) were computed from theoretical X-ray diffraction patterns. These values are the averages over different microstructure analysis methods. The blue lines signify the exponential growth fitting of the different curves. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

is important to note, however, that the structural defects are mostly present in the grain boundary regions, but they are also present in the crystalline structure to a certain extent.

We make a relative comparison between the reductions in vacancies and dislocation cores during the consolidation process. In Fig. 3(c), we report the dependence of the vacancies-to-dislocation-core-concentration ratio on the average grain size. This figure shows that this ratio decreases as the average grain size increases until it reaches a critical average grain size value. It then increases again. Note that this critical average grain size depends strongly on the starting average grain size. The critical average grain sizes for samples S1, S2, and S3 are 7.2 nm, 9.8 nm, and 11.7 nm, respectively. We see that increasing the consolidation pressure (shown

by the empty circles in Fig. 3(c)) from 1 GPa to 5 GPa has no significant influence on this result.

We now discuss these results in the light of the relevant literature. In a previous study on the mechanical properties of aluminum single crystals in the size range from 4.1 nm to 40.5 nm [24], it was concluded that a single crystal shows greater mechanical strength as its size decreases. Since the overall target in this research field is to derive high-performance mechanical bulk materials starting from metal nano-crystals, the most relevant question is how we choose our starting fine grains from size point of view. In our previous study, we speculated that it is better to start from small grains to get better mechanical strength. The current results show that this speculation may not always be true. In a

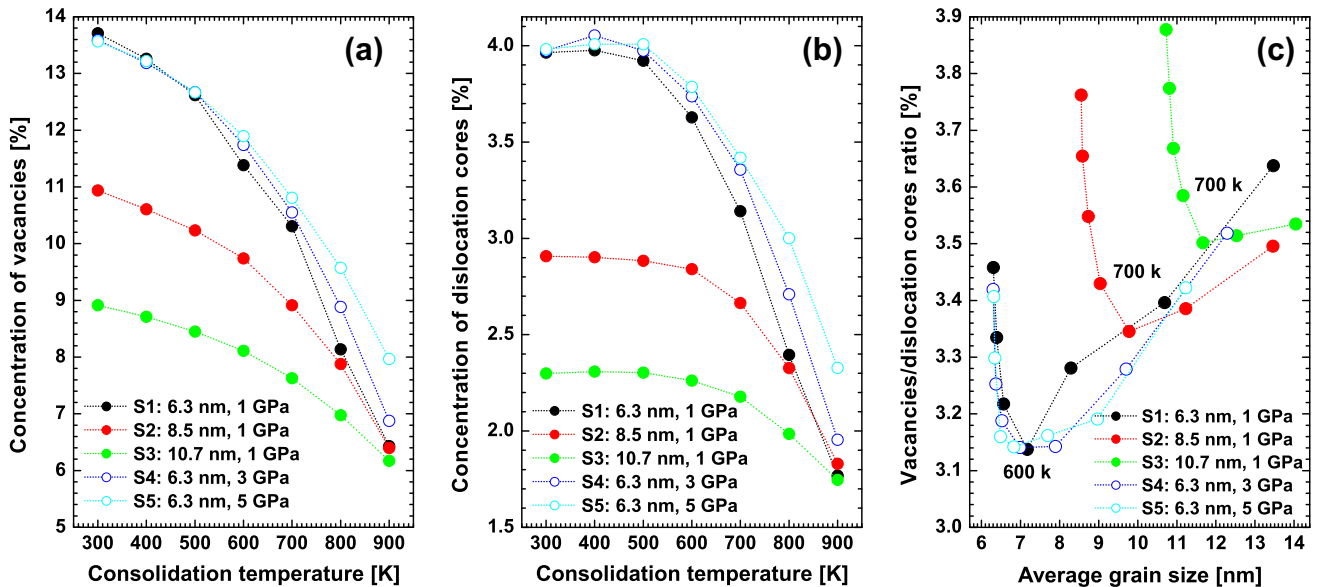


Fig. 3. The dependence of the concentration of structural defects on the initial average grain size as well as the consolidation temperature and pressure in nanometer-sized poly-crystalline aluminum: (a) concentration of vacancies (represented by the 11-coordinated aluminum atoms), (b) concentration of dislocation cores (represented by the 13-coordinated aluminum atoms), and (c) the vacancy-to-dislocation-core-concentration ratio. The solid circles indicate consolidation at different temperatures starting from different average grain sizes, while the empty circles represent consolidation at different temperatures and pressures.

future communication, we will describe the major key factors that control the mechanical strength of consolidated nano-crystalline metals in the light of the current findings.

The largest molecular dynamics simulation study on plastic deformation of nano-crystalline copper in the poly-crystalline format was carried out by Schiøtz and Jacobsen [8]. They studied an average grain size ranging from 5.0 nm to 50.0 nm. They found that the ultimate mechanical strength of the poly-crystalline system increases by increasing the average grain size, reaching a maximum at an average grain size between 10 and 15 nm, and then decreasing again. They attributed this behavior to a shift in the microscopic deformation mechanism from dislocation-mediated plasticity dominant in the samples with large average grain sizes to a grain boundary dominant in samples with small average grain sizes. From our point of view, these results can be explained by the competition between the effects of average grain size and levels of structural defects at a given strain rate.

4. Conclusion

We have presented a quantitative description of the evolution of microstructures and structural defects due to different consolidation parameters. We have shown that the average grain size increases exponentially by increasing the consolidation temperature. This growth is suppressed by increasing the starting average grain size and consolidation pressure. We have also shown that concentration of structural defects, such as vacancies and dislocation cores, evolves in the opposite direction, compared with the microstructure. Our quantitative analysis of defects has shown that there exists a critical average grain size beyond which the vacancy-to-dislocation-core ratio increases. Based on this study, we believe that more sophisticated consolidation procedures must be found to allow for the production of bulk materials with small average grain sizes and acceptable densities of structural defects. In a future communication, we will explain why dislocations-assisted

and grain boundaries-assisted deformation mechanisms dominate in samples with small and large grain size, respectively. A correlation between the mechanical strength of consolidated nano-crystalline bulk material and properties of the starting nano-structures will be highlighted.

References

- [1] H. Salem, A. Sadek, *J. Mater. Eng. Perform.* 19 (2010) 356.
- [2] H.G. Salem, S. El-Eskandarany, A. Kandil, H.A. Fattah, *J. Nanomater.* 2009 (2009) 479185.
- [3] E. Bonetti, L. Pasquini, E. Sampaloesi, *Nanostruct. Mater.* 9 (1997) 611.
- [4] X. Sun, H. Cong, M. Yang, M. Sun, *Metall. Mater. Trans.* 31 (2000) 1017.
- [5] A.S. Khan, Y.S. Suh, X. Chen, L. Takacs, H. Zhang, *Int. J. Plast.* 22 (2006) 195.
- [6] V. Bulatov, F.F. Abraham, L. Kubin, B. Devincere, S. Yip, *Nature* 391 (1998) 669.
- [7] V. Yamakov, D. Wolf, S.R. Phillpot, A.K. Mukherjee, H. Gleiter, *Nat. Mater.* 1 (2002) 45.
- [8] J. Schiøtz, K.W. Jacobsen, *Science* 301 (2003) 1357.
- [9] Z. Shan, E. Stach, J. Wieszorek, J. Knapp, D. Follstaedt, S. Mao, *Science* 305 (2004) 654.
- [10] E. Bringa, K. Rosolankova, R. Rudd, B. Remington, J. Wark, M. Duchaineau, D. Kalantar, J. Hawreliak, J. Belak, *Nat. Mater.* 5 (2006) 805.
- [11] L.E. Levine, B.C. Larson, W. Yang, M.E. Kassner, J.Z. Tischler, M.A. Delos-Reyes, R.J. Fields, W. Liu, *Nat. Mater.* 5 (2006) 619.
- [12] F.F. Csikor, C. Motz, D. Weygand, M. Zaiser, S. Zapperi, *Science* 318 (2007) 251.
- [13] J. Rajagopalan, J.H. Han, M.T.A. Saif, *Science* 315 (2007) 1831.
- [14] Z. Shan, R.K. Mishra, S.S. Asif, O.L. Warren, A.M. Minor, *Nat. Mater.* 7 (2007) 115.
- [15] S.H. Oh, M. Legros, D. Kiener, G. Dehm, *Nat. Mater.* 8 (2009) 95.
- [16] R. Komanduri, N. Chandrasekaran, L. Raff, *Int. J. Mech. Sci.* 43 (2001) 2237.
- [17] L. Yuan, D. Shan, B. Guo, *J. Mater. Process. Technol.* 184 (2007) 1.
- [18] X. Li, Y. Wei, W. Yang, H. Gao, *Proc. Natl. Acad. Sci. USA* 106 (2009) 16108.
- [19] Y. Mishin, D. Farkas, M. Mehl, D. Papaconstantopoulos, *Phys. Rev.* 59 (1999) 3393.
- [20] M.S. Daw, M.I. Baskes, *Phys. Rev.* 29 (1984) 6443.
- [21] M.W. Finnis, J.E. Sinclair, *Philos. Mag.* 50 (1984) 45.
- [22] S. Plimpton, *J. Comput. Phys.* 117 (1995) 1.
- [23] J. Li, *Modell. Simul. Mater. Sci. Eng.* 11 (2003) 173.
- [24] N.D. Afffy, H.G. Salem, A. Yavari, T. El Sayed, *Comput. Mater. Sci.* 78 (2013) 34.
- [25] B.E. Warren, *X-Ray Diffraction*, Addison-Wesley Publishing Co., 1969.
- [26] Y. Zou, L. Chen, *Phys. Status Solidi* 2412 (2005) 242.
- [27] A. Leonardi, P. Scardi, M. Leoni, *Philos. Mag.* (2012) 1.