

Modeling the deformation behavior of nanocrystalline alloy with hierarchical microstructures

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Received: 16 November 2015 / Accepted: 25 January 2016 / Published online: 8 February 2016
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Abstract A mechanism-based plasticity model based on dislocation theory is developed to describe the mechanical behavior of the hierarchical nanocrystalline alloys. The stress–strain relationship is derived by invoking the impeding effect of the intra-granular solute clusters and the inter-granular nanostructures on the dislocation movements along the sliding path. We found that the interaction between dislocations and the hierarchical microstructures contributes to the strain hardening property and greatly influence the ductility of nanocrystalline metals. The analysis indicates that the proposed model can successfully describe the enhanced strength of the nanocrystalline hierarchical alloy. Moreover, the strain hardening rate is sensitive to the volume fraction of the hierarchical microstructures. The present model provides a new perspective to design the microstructures for optimizing the mechanical properties in nanostructural metals.

Keywords Mechanism-based plasticity model · Mechanical behavior · Nanocrystalline alloys · Hierarchical nanostructures · Modeling and simulations

Introduction

Strength and ductility, as two of the most important mechanical properties of structural materials, are unfortunately mutually exclusive. This is also true for bulk nanostructured (NS) materials, which usually have high strength, but disappointingly low ductility. A high work-hardening rate is essential for good uniform elongation because it can delay the localized deformation (necking) under tensile stress. Bulk NS materials often have a very low or null work-hardening rate because of their low dislocation accumulation capability (Van Swygenhoven 2003a; Wang et al. 2002). Molecular dynamics (MD) simulations have shown that in NS metals dislocations may be emitted from grain boundaries (GBs) and disappear at the opposite sites without accumulation (Van Swygenhoven 2003a; Van Swygenhoven et al. 2004; Wu et al. 2009; Yamakov et al. 2004), providing support to the widely accepted belief that they have intrinsically diminutive strain hardening. There has been considerable effort to address the pressing need of increasing the strength and ductility of NS materials at room temperature. The strength-ductility trade-off

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can be circumvented through various approaches, for example, introduction of very small second-phase particles (Wang et al. 2002; Zhao et al. 2006), amorphous phase and nanograins (Fan et al. 2009), preexisting twins (Lu et al. 2004), bimodal grain size distribution (Zhao et al. 2008), as well as laminated structures (Wang et al. 2007). These studies provided alternative methods to obtain higher strength and ductility in the nanometer scale by adding effective microstructures to impede dislocation movements.

Recently, a super strength–ductility combination (a yield strength of 1 GPa and a total elongation to failure of 9 %) was reported in aerospace-grade 7075 Al alloy (Liddicoat et al. 2010). Systematic microstructural and concentration analyses indicate that the above super mechanical properties are resulted from the hierarchal nanostructures in the alloys: sub-nanometer intra-granular solute cluster (mean equivalent diameter $d_{\text{cluster}} = 1.6$ nm, volume fraction $f_{\text{cluster}} = 0.04$) and inter-granular nano-scale solute segregates with two different geometries ($d_{\text{nodal}} = 3.7$, $f_{\text{nodal}} = 0.001$, $d_{\text{lineal}} = 9$ nm, $f_{\text{lineal}} = 0.019$) as well as the nano-grain matrix with an average grain size of about 26 nm. The above investigations demonstrate a novel architecture which offers a design pathway towards a new generation of super-strong materials with new regimes of property–performance space. Nevertheless, the authors could not give a quantitative explanation on the super yield strength and good ductility, or identify the individual contributions to the mechanical behavior of the hierarchal nanostructures in the alloys. This certainly requires further studies. The super strong hierarchal 7075 Al alloy has four different kinds of hardening mechanisms: solute, grain refinement, precipitates or solute clusters, dislocation hardening. The non-clustered solute will provide a very limited conventional solid–solution hardening, because a solution-treated 7075 Al alloy with 100 % of the solute in the matrix has a strength of 0.145 GPa; and in fact, only about half (~ 51 %) of the solute remains dissolved in the as-prepared hierarchal 7075 alloy (Zhao et al. 2006). The hardening effect due to grain refinement could be estimated by the well-known Hall–Petch relation, and for pure Al with an average grain size of 26 nm, is ~ 0.27 GPa (Hansen 2004). The rest hardening of about 0.585 GPa could be contributed by the sub-nanometer intra-granular solute clusters and inter-granular nano-scale solute segregates with two different geometries as well as

dislocation hardening effects. The remaining question is how much the above three hardening effects could contribute to the strength respectively? And which hardening effect is dominant or overwhelming? To answer the above questions, we developed a quantitative continuum plasticity model based on dislocation theory to describe the mechanical behavior of the hierarchal nanocrystalline 7075 Al alloys. We found the interaction between the hierarchal nanostructures and dislocations contributes to the strain hardening property, and greatly influences the ductility of nanocrystalline alloys.

Quantitative continuum plasticity model development

The grain refinement hardening, originated from dislocation accumulation at GBs, increases with decreasing grain size. Owing to extremely profuse defects and nonhomogeneous inter-atomic spacings compared with the regular lattice in the grain interior (GI), the GB regions and its significant volume fraction plays a significant role in the mechanical responses of NS materials especially as grain size decreases to nanoscale. GB regions were considered to be the sources of dislocations and showed a faster rate of hardening than that of GI. It is further described that the plastic flow firstly took place in the GB regions as a result of the GB dislocation accumulation. According to the experimental observations from hierarchical 7075 alloy (Liddicoat et al. 2010), it is revealed that a high density of dislocations ($\sim 3.2 \times 10^6 \text{m}^{-2}$) are located in GI and GBs. Thus, the grain boundary-affected zone (GBAZ) (Schwaiger et al. 2003) nearby the GBs, in which the strain gradients are involved, is introduced in our model. The high density of intra-granular solute clusters (comprising ~ 38 % of the total solute (Liddicoat et al. 2010)) can contribute to significant cluster strengthening by means of providing effective resistance to slip dislocations, and therefore, increase to the strain hardening and ductility by increasing the dislocation storage capability. The other major architectural features of this solid solution are the inter-granular solute nanostructures at GBs, which are expected to provide nucleation sites of precipitates, stabilize nanograin growth, strengthen interface bond cohesion, and resist embrittlement and defect generation. Therefore, according to the reported

intra-/inter-granular solute structures in literature, we generated our model as shown in Fig. 1.

As there exhibits the strain gradient nearby the GBs, the theory of mechanism-based strain gradient plasticity (Huang et al. 2004) is utilized to calculate the stress–strain relation of the hierarchical alloy. The flow stress is based on the Taylor model.

$$\sigma_{\text{flow}} = \sigma_0 + M\alpha Gb\sqrt{\rho} \tag{1}$$

here, σ_0 is the lattice friction stress, M , α , G and b is the Taylor factor, the empirical constant, the shear modulus, and the Burger constant, respectively. ρ denotes the total dislocation density. To identify the flow stress based on the Taylor model in Eq. (1), the dislocation density must be addressed in the hierarchical alloy. The total dislocation density includes the

geometrically necessary dislocation (GND) density in the GBAZ ρ_{GB} due to the local and non-uniform deformation, and the statistically stored dislocation (SSD) density due to the uniform deformation in the interior crystal ρ_{GI} , as well as the dislocation density arising from the dislocations accumulation at nano-scale solute clusters ρ_{cluster} . Thus, $\rho = \rho_{\text{GI}} + \rho_{\text{GB}} + \rho_{\text{cluster}}$

The density of dislocations in the crystal interior obeys the evolution law with plastic strain according to Kocks and Mecking’s model (Kocks and Mecking 2003).

$$\frac{\partial \rho_{\text{GI}}}{\partial \epsilon^p} = M(k + k_1\sqrt{\rho_{\text{GI}}} - k_2\rho_{\text{GI}}) \tag{2}$$

where M is the Taylor factor, k is an extra term utilized to account for the static dislocation storage and

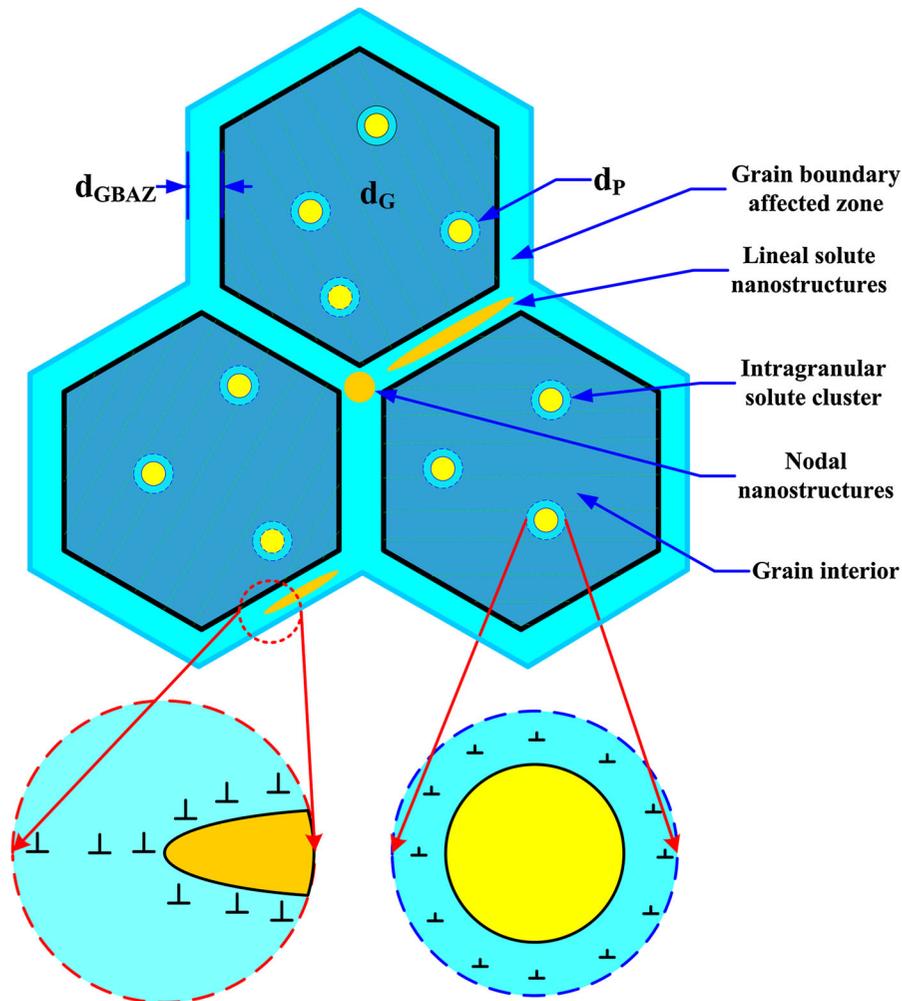


Fig. 1 Schematic drawings of microstructure of nanostructured hierarchical 7075 Al for the proposed model

$k = 1/d_G b$, where d_G is the size of an unit cell considered to be the grain size; k_1 is a constant accounting for the dislocation storage, which is an athermal process and $k_1 = \psi/b$, where ψ is a proportionality factor, $k_2 = k_{20}(\dot{\epsilon}^p/\dot{\epsilon}_0)^{-1/n}$ represents the thermally activated dynamic recovery process, k_{20} and $\dot{\epsilon}_0$ are the constants, and n is inversely proportional to the temperature. In the GBAZ, dislocation-based activities in GBs are assumed to be the predominant sources of plastic deformation in the hierarchical alloy. Thus, it is supposed that the strain gradients are all stored in GBAZs and the plastic strain in the GBAZ is assumed to increase gradually from interior to the GBs. Different from the uniform lattice in GI, the heterogeneous plastic deformation of GB region, with distorted atomic structure and large inter-atomic spacing, is realized with respect to activities of GNDs. Due to the combination effects of flow stress σ_{flow} and the different atomic arrangements within GBAZ, the plastic deformation strain generated by the dislocations in the GBAZ is denoted as γ^{GB} , which can be described by the following equation $\gamma^{\text{GB}} = \phi n^{\text{GB}} b/d_G$, where ϕ is a geometrical factor, n^{GB} is the number of dislocations around the GBs. The local mean strain gradient created by the dislocations in the GBAZ can be defined as

$$\eta^{\text{GB}} = \frac{\gamma^{\text{GB}}}{d_{\text{GBAZ}}} \quad (3)$$

Here, d_{GBAZ} is the thickness of the GBAZ. Thus, the dislocation density of the GBAZ can be formulated as follows:

$$\rho_{\text{GB}} = \frac{n^{\text{GB}} \lambda^{\text{GB}}}{V_{\text{cell}}} \quad (4)$$

where $\lambda^{\text{GB}} = \pi d_G$ denotes the average length of dislocation loops in the GBAZ and $V_{\text{cell}} (= \pi d_G^3/6)$ is the volume of the unit cell. Due to the high number of dislocations stored along the GBs, the local strain gradients in the GBAZ can be introduced to obtain the density of dislocation in the GBAZ. Sequentially, this dislocation density can be expressed by

$$\rho_{\text{GB}} = k^{\text{GB}} \frac{\eta^{\text{GB}}}{b} \quad (5)$$

where $k^{\text{GB}} = 6d_{\text{GBAZ}}/\phi^{\text{GB}} d_G$, in which ϕ^{GB} is a constant (=1). It can be found that the flow stress

increases with the inverse of the square root of grain size, which is consistent with the Hall–Petch relation.

During the plastic deformation process of the hierarchical alloy, the nodal and lineal solute nanostructures will retard dislocations nucleated at the GBs as shown in Fig. 1. The lineal solute nanostructures on GBs will be available to immobilize GB dislocations and strengthen the alloy. The stress develops once dislocations accumulate at GBs forming pileups, impeding the movements of similar dislocations and finally inducing the strain hardening of materials. In the present model, the two geometries of nano-scale inter-granular solute structures, i.e., nodal and lineal solute nanostructures locating at the GBs, can be treated as the obstacles hindering the GB dislocation movements. The dislocation accumulation will be intensified due to the existence of the GB solute arrangements distributed in the normal GBs. A kinetic evolution for dislocation accumulation has been proposed by Proudhon et al. (2008). The number of dislocation loops N stored around inter-granular solute clusters is related to the plastic strain through an expression that leads to the saturation of the dislocation loops number N_0 at solute clusters.

$$\frac{dN}{d\varepsilon_{\text{cluster}}} = \frac{2r}{b} \left(1 - \frac{N}{N_0} \right) \quad (6)$$

where $\varepsilon_{\text{cluster}}$, r , N_0 denote the plastic strain, average radius of inter-granular solute cluster, the maximum number of dislocation loops around inter-granular solute clusters, respectively. This equation has been used to describe the effect of grain size on work hardening (Sinclair et al. 2006), as well as the precipitate contribution to kinematic hardening (Bouaziz and Brechet 2009). A simple interpretation of this equation is that there are only N_0 spaces available for dislocation storage so that the storage will have a probability effectively equal to $(1 - N/N_0)$. The following expression for N as a function of plastic strain is thus obtained $N = N_0(1 - \exp(-d_{\text{cluster}}/bN_0))$. The dislocation density stored around inter-granular solute clusters ρ_{cluster} can be expressed as the product of the number of loops efficiently stored around one cluster $n\phi$, the cluster density $3f/4\pi r^3$ and the length of a loop around a cluster.

$$\rho_{\text{cluster}} = N\phi \cdot \frac{3f}{4\pi r^3} \cdot 2\pi r = N\phi \frac{3f}{2r^2} \quad (7)$$

where φ is a constant (0.04), f is the volume fraction of inter-granular solute clusters, r is the mean equivalent average radius of nano-scale solute clusters.

In the traditional solid solution strengthening mechanism in coarse-grained alloys, solute atoms serve as obstacles to dislocation motion, influencing the elastic energy of a dislocation due to both local size and modulus changes. When the grain size is reduced to nanoscale, GB dislocation activity will dominate plastic deformation. It is widely accepted that the strength of nanocrystalline materials is limited by the mechanism that a full dislocation (or a complete set of partial dislocations) being emitted from the GB and traversing a grain. Here, the scaling of Fleischer model still works, but the dislocations bow between pinning sites within the GBs, such as GB ledges. Under such circumstances, the dislocation obstacle spacing is close to the grain size diameter and GB pinning points will serve as strong obstacles to the dislocation movements. An enhanced solid solution effects on the strength of nanocrystalline alloys by incorporating the effect of traditional solid solution strengthening mechanistic model of local dislocation pinning with nano-scale strengthening term arises by considering GBs as pinning points for dislocation motion in NS alloy (Rupert et al. 2011). The additive contributions to the total strengthening from solute addition can be given by

$$\sigma_{ss} = M(AG_{\text{solvent}}\epsilon_s^{3/2}c^{1/2} + \frac{G_{\text{solvent}}b_{\text{solvent}}}{d}\epsilon_{\text{nc}}c) \quad (8)$$

The first term in the parentheses is the Fleischer equation which has been widely used to predict the traditional solid solution strengthening; the second term denotes nano-scale strengthening term predicting the nanocrystalline solution pinning effect.

$$\epsilon_s = \left| \frac{\frac{1}{G_{\text{solvent}}}\frac{\partial G}{\partial c}}{1 + \frac{1}{2}\left|\frac{1}{G_{\text{solvent}}}\frac{\partial G}{\partial c}\right|} - 3\frac{1}{b_{\text{solvent}}}\frac{\partial b}{\partial c} \right| \quad \text{and} \quad \epsilon_{\text{nc}} = \frac{1}{G_{\text{solvent}}}\frac{\partial G}{\partial c} + \frac{1}{b_{\text{solvent}}}\frac{\partial b}{\partial c}$$

are the interaction parameter for the Fleischer model and the nanocrystalline solution pinning model.

The architecture of the investigated nanostructure hierarchical 7075 Al alloy is featured with sub-nanometer intra-granular solute clusters and two geometries of nanometer-scale inter-granular solute structures (Liddicoat et al. 2010). Orowan strengthening describes improvements in strength in the

matrix material based on the resistance of small particles against the motion of dislocations (Kim et al. 2013). Since the Orowan-type of strengthening is observed when the reinforcement particles are sufficiently small, it is proposed that the strength increases are only effective for the particles with sizes under 1 μm (Miller and Humphreys 1991). The Ashby–Orowan formulation ($\sigma_{\text{orowan}} = AGb/2\pi\lambda \ln(d_p/r)$) is an advanced revision which has been widely used in predicting the strength improvements. Among variations of the general form of the Ashby-Orowan equation, one of the most widely used formula to estimate the Orowan-type contribution to the yield strength improvement is shown as follows (GOH et al. 2007; Habibnejad-Korayem et al. 2009; Sun et al. 2012):

$$\sigma_{\text{orowan}} = \frac{0.13Gb}{\lambda} \ln\left(\frac{d}{2b}\right) \quad (9)$$

where d is average diameter of solute clusters ($=2r$). To simplify the mathematical calculations, the particles are assumed to be spherical and randomly distributed in the matrix; thus the inter-solute-clusters mean free path $\lambda = d\left[\left(1/(2V)\right)^{1/3}-1\right]$, where $V (= \pi d^3/6)$ is the volume of solute cluster.

Then it comes to the calculation of the total flow stress, the problem that how to superimpose of these different strengthening contributions arises. In most cases, contributions from these different mechanisms are assumed to act independently (Sabirov et al. 2013), thus the total flow stress of NS alloys can be expressed as

$$\sigma_{\text{flow}} = \sigma_0 + M\alpha Gb\sqrt{\rho} + \sigma_{ss} + \sigma_{\text{orowan}} \quad (10)$$

where σ_0 is lattice friction stress, σ_{ss} enhanced nano-scale solid solution strengthening, σ_{orowan} Orowan strengthening.

Numerical results and discussion

In this paper, we take the HPT-processed aluminum alloy (Liddicoat et al. 2010) containing hierarchical nanostructures and possessing mechanical properties that expand known performance boundaries as an example to explore the interplay between the mechanical properties and microstructures. Using the parameters appropriate for 7075 aluminum alloy in the prior

literatures (Fribourg et al. 2011; Ma et al. 2014), $G = 28.4$ GPa, $\nu = 0.33$, $b = 0.286$ nm, $M = 3.06$, $\alpha = 0.2$, and $d_{GBAZ} = 1$ nm, we plot the true stress–strain of the hierarchical 7075 Al (converted from the engineering stress–strain curve using standard formula) curves and the theoretical calculated curves in Fig. 2. In order to acquire a clear understanding of contributions from different deformation mechanisms, contributions from dislocation strengthening mechanism with/without density of dislocations around nano-scale solute structures, enhanced nano-scale solid solution strengthening mechanism, Orowan strengthening mechanism are plotted in Fig. 2. Note that the three deformation mechanisms contribute to the total strength of hierarchical 7075 Al in a descending order: Orowan strengthening (603 MPa), dislocation strengthening (519 MPa), enhanced nano-scale solid solution strengthening (105 MPa). As shown in Fig. 2, the dislocation activities in GI and GB act only on the yield strength and has nearly no contribution to strain hardening. It also can be found that the strain hardening is mainly associated with the dislocation activities around the nano-scale intra-granular solute clusters and inter-granular solute structures. This is because dislocation cells formed by the dislocations stopped around the nano-scale solute clusters will impede the movement of similar dislocations by a ‘back stress’ effect. Interaction of dislocations with

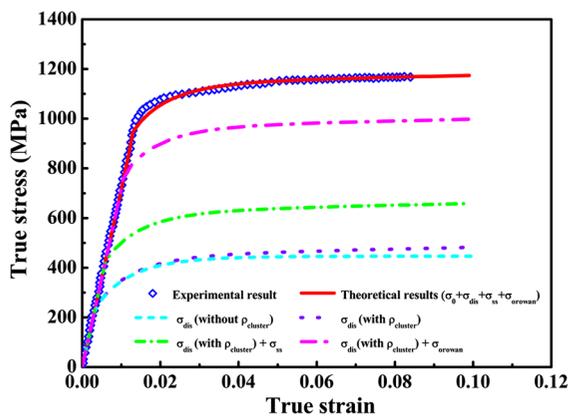


Fig. 2 Comparison of the true stress–strain relationship between theoretical and experimental results. The separate strengthening contributions are associated with Orowan strengthening σ_{Orowan} , enhanced nanoscale solid solution strengthening σ_{ss} , and dislocations strengthening σ_{dis} with/without dislocations around nanoscale solute clusters ρ_{cluster} , respectively

nano-scale solute clusters and structures will lead to the dislocation accumulation resulting in increasing strain hardening and, therefore, enhancing ductility.

It is supposed that any improvements of the strain hardening rate will be beneficial to enhancing the homogeneous plastic deformation of nanocrystalline metal (Van Swygenhoven 2003b; Wang et al. 2002). Considering the strain hardening effect can be mainly attributed to the existence of the dislocation accumulation around the nano-scale solute clusters, the contribution from dislocation strengthening of theoretical results with different volume fractions (0–0.08) and mean diameters (2–6 nm) of nano-scale solute clusters is plotted in Figs. 3, 4, respectively. Meanwhile, the corresponding strain hardening rate as a function of plastic strain of each curve is also calculated and plotted. It is worth noting that appropriate simplification has been made in the theoretical calculation. The nano-scale intra-granular solute clusters and inter-granular solute structures are equivalent to be spherical with the equivalent volume ($\pi d_{\text{ns}}^3/6$) and volume fraction. Clearly, the density of dislocations around the nano-scale solute clusters is a function of the volume fraction and mean diameter, and the improved strain hardening rate, as a consequence, is sensitive to the volume fraction of the nano-scale solute clusters. In the existence of the nano-scale solute clusters, the strain hardening rate monotonically increases with the decreasing volume fraction, as depicted in Fig. 3. According to the atomic composition of the 7075 Al, the total solute volume fraction is about 12.5 % ($1 - f_{\text{Al}}, f_{\text{Al}}$) is the volume fraction of

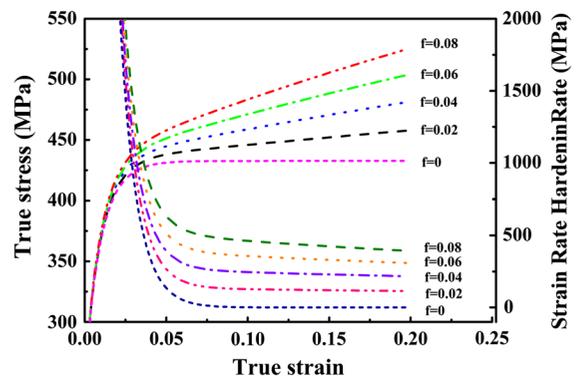


Fig. 3 Stress–strain response from the contribution of dislocation strengthening and the corresponding strain hardening rate with different volume fractions of the nanoscale solute structures

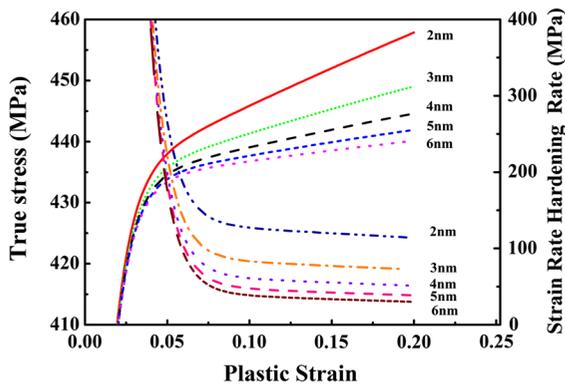


Fig. 4 Stress–strain response from the contribution of dislocation strengthening and the corresponding strain hardening rate with different mean diameters of the nanoscale solute clusters

Al). Thus, there exhibits a critical volume fraction at which the dislocation strengthening operates the greatest contribution to the flow stress of hierarchical 7075 Al. Note that, with the same volume fraction, the decrement of the diameter of the nano-scale solute clusters can enhance strain hardening rate remarkably, as shown in Fig. 4. It is also interesting to find that the strain hardening rate becomes more apparent with decreasing diameters of the nano-scale solute clusters. This is because that the density of nano-scale solute clusters increases with the decrement of diameters, inducing the increment of the dislocation densities in Eq. (7). In each case of a given volume fraction, the hierarchical NS 7075 Al can approach the maximum strain hardening rate by tuning the volume fraction and diameter of nano-scale solute clusters. These results suggest that the maximal strength in the hierarchically NS alloy can be achieved by controlling the solid solution structure parameters.

Conclusions

To summarize, a quantitative continuum plasticity model predicting the strength in hierarchical NS alloy has been presented. Quantitative contributions were calculated for different strengthening mechanisms in the nanocrystalline hierarchical structured 7075 Al alloy, including dislocation-grain size-cluster strengthening, enhanced nanocrystalline solid-solution strengthening, Orowan strengthening. This model shows good agreement with the experimental results,

suggesting that the hierarchical architecture has an important influence on aluminum alloy strengthening. The model will provide a theoretical basis for the development of a new generation of advanced alloys with new regimes of property-performance space by controlling the solute architecture as an alternative approach.

Acknowledgments The authors gratefully acknowledge the financial supports of Key Project of Chinese Ministry of Education (211061), Program for New Century Excellent Talents in University, National Natural Science Foundation of China (10502025, 10872087, 11272143, 51225102 and 2012CB932203), and Natural Science Foundation of Hubei Province (Q20111501).

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