The plasticity of highly oriented nano-layered Zr/Nb composites

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Abstract

In prior work, bulk lamellar composites of pure zirconium and niobium (Zr/Nb) were manufactured by accumulative roll bonding (ARB). After the substantial amounts of straining required to refine the layers to nanoscale dimensions, formation of highly oriented Zr crystals was observed. In this work, we employ a spatially resolved multiscale crystal plasticity based model in 3D to study the orientational stability of Zr single crystals and Zr/Nb bicrystals during rolling deformation. The analysis reveals that predominant texture components arise due to substantially reduced ratios of slip resistances among the prismatic, pyramidal I $\langle c+a \rangle$, and basal slip systems. In support, density functional theory (DFT) calculations of generalized stacking fault energy curves on these three slip systems suggest that the ratio of critical stresses to form these dislocations are within 2.5 times. This finding of reduced anisotropy in Zr at the nanoscale can provide insight into the design of nano-structuring processes for target textures, such as those containing highly oriented grains.

Keywords: Zirconium; Niobium; Crystal plasticity; Accumulative roll bonding; Interfaces

1. Introduction

When subject to bulk metal forming processes, such as rolling, polycrystalline metals develop texture [1, 2]. Texture affects both mechanical [3-5] and functional properties [6, 7]. As such, control of texture evolution during processing is an approach often used to help attain target properties and eventual performance [8-12]. For hexagonal close packed (HCP) metals, such as Zr, Mg, Ti, and Be, a preferred texture develops after some amount of straining, and if its components are orientationally stable, is maintained with further straining [13-18]. Under the severe plastic deformation process, such as accumulative roll bonding (ARB) [19], it has been shown that the characteristic rolling texture of Zr persists even up to large strains of 10 [20, 21].

Recently ARB has been applied to a two-phase Zr/Nb composite with the aim of using the biphasic interfaces to help refine the Zr phase to nanoscale dimensions [22]. Figure 1 shows micron- and nano-layered Zr/Nb composites made by ARB. Starting with $h = 1$ mm layer thickness, ARB was successfully used to reduce the layer thickness from hundreds of

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microns (Fig. 1a) to less than 100 nm (~92 nm) as seen in Fig. 1b, while keeping a continuous layered architecture, hence achieving a nanolayered Zr/Nb composite. There have been other attempts to achieve <100 nm microstructures of Zr, but these resulted in non-uniform distributions of nano to submicron grains or phase transformations [23-28].

The Zr phase maintained a conventional rolling texture characteristic of single-phase Zr until h was refined below 100 nm, at which point the texture consisted of even fewer components. For instance, orientations with their crystallographic c -axis aligned with the normal direction (ND) of the sheet are contained within the texture of rolled coarse-grained Zr, but they disappear when the Zr layers are refined below 100 nm.

Loss of orientational stability suggests that slip activity within the grains has changed, despite the fact that the same rolling deformation state is applied. When the layer thickness is reduced below 100 nm, the nano-layered structure, the layers are spanned by only one crystal and thus every Zr grain is bonded to a Nb grain. First, a change in slip activity could arise at the nanoscale because the Zr crystals must deform while at the same time meeting compatibility constraints with neighboring Nb crystals. There may be some Nb orientations for which meeting the compatibility criterion is easier than others. Second, in nanoscale grains, dislocations no longer primarily originate from the grain boundaries or the interior of the grains, but at the phase interfaces. The phase interfaces serve as nucleation sites, annihilation sites, and barriers. It is, therefore, possible that the change in slip activity occurs because the critical stresses for activating dislocations are determined by the nucleation barrier from the interfaces, not the internal crystal defects and grain boundaries. It is also likely that the critical stress to activate dislocation motion (or critical resolved shear stress, CRSS) is dominated by the nano-scale mean free path between adjacent interfaces and again, not by the dislocations that are stored within the layers. Last, the stresses that develop during rolling of such fine nanograins are much higher than those achieved in a coarse-grained polycrystalline Zr and therefore could even enable the more difficult slip modes (such as basal slip) to activate.

The aim of this work is to understand the orientation stability of nanocrystalline Zr during ARB. We use a crystal plasticity based model that allows for slip to occur on available slip modes and can directly relate texture evolution to the relative contributions of different slip modes. The analysis indicates that Zr crystals, when bonded to Nb crystals, become less orientationally stable than the same single crystal orientation under the same deformation state. We show that the “nano-preferred” texture components arise when the plastic anisotropy of Zr is substantially reduced, in which the slip resistance ratio of prismatic slip to basal slip or to pyramidal slip have decreased below 2.0. In support of this finding, we perform density functional theory (DFT) calculations of generalized stacking fault energy curves on these three slip systems and they indicate that the stresses to form these dislocations are indeed close (within 2.5 times). Although we focus this study on nanocrystalline Zr, the findings are sufficiently general to provide insight into mechanisms underlying the nanostructuring SPD processes of other HCP metals that can be refined to nano grained structures.

2. Background

2.1 Stable orientations in coarse-grained Zr

The materials used in this study are high-purity Zr with the following chemistry expressed in weight parts per million (Wt. ppm) Hf = 35, Fe < 50, Al < 20, V < 50, O < 50, N < 20, C < 22 and Zr balance [29] and reactor-grade high-purity Nb (99.97% pure, ATI-Wah Chang). As we mentioned in the introduction, recently ARB was applied to 50/50 Zr/Nb, involving high-purity Zr and Nb [22, 27]. The process refined the layer thicknesses h from 1 mm to 92 nm. Figures 2 and 3 show pole figures and the $\langle 1\ 1\ \bar{2}\ 0 \rangle$ and $\langle 1\ 0\ \bar{1}\ 0 \rangle$ fibers for the Zr phase, respectively, for three layer thicknesses. From layer thicknesses $h = 1$ mm down to $h = 26\ \mu\text{m}$, the texture that developed in the Zr phase was very close to the conventional one and the $h = 106\ \mu\text{m}$ Zr/Nb composite shown in Fig. 1a, is one example. For these layer thicknesses, the layers were polycrystalline, with 10 to 20 grains spanning the thickness of the layer. The basal pole figure of the Zr phase exhibits a split peak in the TD, a characteristic feature in the texture of Zr when rolled alone. The peaks are not pronounced with intensities ranging from 4-6 multiples of random distribution (mrd) over a range of basal pole tilt from 0° to 30° from the ND.

We identify seven Zr orientations in the rolled texture of coarse-grained Zr. Usually among these preferred ones, none of them are strongly favored over the other. Along the $\langle 11\bar{2}0 \rangle$ fiber for $h = 106\ \mu\text{m}$, there are three chief Zr orientations, starting with the basal pole component aligned with the ND (Zr1) and spanning 30° from the ND (Zr2 and Zr3). Along the $\langle 1\ 0\ \bar{1}\ 0 \rangle$ fiber, there are four main orientations, including the basal pole component along the ND (Zr4) and covering orientations tilted up to 45° from the ND (Zr5-Zr7). These orientations are mapped onto a basal pole figure in Fig. 4 and listed in Table 1.

2.2 Anisotropy of Zr

In an attempt to determine the slip modes that contribute to the stable rolling texture of the Zr phase during manufacturing of the Zr/Nb composite, two types of crystal plasticity models were employed, both utilizing the same Zr hardening law [30]. The earlier attempt involved a mean-field technique and modeled Zr and Nb as separate phases, and the latter one, used a full-field model, and simulated the deformation as two phases co-deforming (compatibility across the dissimilar phases). Both models were successful at modeling texture evolution in the Zr phase in the coarse-layered Zr/Nb composite, in which the calculated texture had the basal poles spread from the ND to the TD with some weak peaks forming off ND, ranging from 10° to 35° from the ND. This calculated texture was similar to that texture of single phase Zr rolled alone [20, 31]. Both models also used the same hardening law for Zr and the set of critical resolved shear stresses (CRSS) for prismatic $\langle a \rangle$ slip, pyramidal I $\langle c+a \rangle$ slip and basal $\langle a \rangle$ slip. Under the conditions of rolling modeled here (room temperature, moderate strain rate deformation), the initial CRSS values for prismatic $\langle a \rangle$ slip, pyramidal $\langle c+a \rangle$ slip and basal $\langle a \rangle$ slip were 12 MPa, 133 MPa, and 142 MPa, respectively [32].

Both Zr/Nb plasticity models found that while all three slip modes were active during rolling, they were not equally contributing. Prismatic $\langle a \rangle$ slip dominated in accommodating the rolling strain, with pyramidal $\langle c+a \rangle$ slip contributing secondly, and basal $\langle a \rangle$ slip, contributing a minor amount. The result is consistent with the prior peak

broadening measurement on a Zr alloy [33]. Thus, it was shown that the conventional rolling texture and the Zr phase texture in the coarse-layered composites were a result of the significant plastic anisotropy of Zr, with prismatic slip being predominant.

2.3 Stable components in nanoscale Zr

An interesting transformation in texture development arises when nanolayered thicknesses in the Zr/Nb composites ($h = 92$ nm) are achieved [22]. The Zr phase texture becomes highly oriented and deviates from the conventional rolled Zr texture. To demonstrate, the fiber plots in Fig. 3 include the textures for the $h = 92$ nm and 41 nm Zr/Nb ARB composite. As shown, compared to the micron-layered composite, in the 92 nm composite, the basal components, Zr1 and Zr4, have decreased in intensity and those of Zr2, Zr3, Zr5, Zr6, and Zr7, have increased. As the layers were refined to 41 nm the basal components Zr1 and Zr4 continue to drop, while Zr2-3 and Zr6-7 continue to strengthen. This nano-scale induced transformation signifies a change in orientation stability. Orientational stability can be altered by a change in slip activity, a general term that refers to the number of active slip systems, the mode(s) involved, and the distribution of slip across the activated slip systems.

There are two possible ways worth pursuing in which slip activity in nanocrystalline Zr could have been altered. The first concerns the increased compatibility constraint arising in the nanolayered composite. When the layers refine to the nanoscale, only one crystal spans the layers, so every Zr grain becomes bonded to a Nb grain. Satisfying compatibility so that the Zr/Nb co-deform could have altered slip activity in the Zr. The second possibility is an alteration in the activation stresses for the three available slip systems, prismatic, basal, and pyramidal. Although other ways are possible, it is clear that these two prospects warrant investigation.

The goal of this work is to employ a multiscale model built upon a crystal plasticity finite element (CPFE) framework to examine these two ways in which slip could have been augmented within nanocrystalline Zr phase to drastically change its orientational stability in rolling. We first briefly review the CPFE model. In what follows, we first apply the model to investigate the orientational stability of single crystal Zr using the activation barriers for slip characteristic of coarse-grained Zr. To investigate the effects of co-deformation with Nb on the orientational stability of coarse-grained Zr crystals, we study the deformation of bicrystals of Zr/Nb. To determine whether there has been a change in activation barriers for slip, we then repeat the single crystal Zr and bicrystal Zr/Nb analysis with the resistance values for the three available slip systems, prismatic, basal, and pyramidal, altered for nanocrystalline Zr.

3. Modeling

3.1 Summary of the CPFE constitutive law

In CPFE, the deformation of every material integration point in the mesh in Fig. 9 follows a constitutive law based on elastic anisotropy and rate-dependent crystallographic slip. We summarize briefly the formulation and refer the reader to [34-38] for more details.

In the finite deformation formulation, the total deformation gradient tensor $\mathbf{F}$ is decomposed into an elastic $\mathbf{F}^e$ and plastic $\mathbf{F}^p$ parts via:

$$\mathbf{F} = \mathbf{F}^e \mathbf{F}^p, \quad (1)$$

where $\mathbf{F}^e$ is comprised of deformation gradients from elastic stretching and lattice rotation. The relationship between $\mathbf{F}^e$ and stress is given by:

$$\mathbf{T}^e = \mathbf{C} \mathbf{E}^e, \quad \mathbf{T}^e = \mathbf{F}^{e^{-1}} \{ (\det \mathbf{F}^e) \boldsymbol{\sigma} \} \mathbf{F}^{e^{-T}}, \quad \mathbf{E}^e = \frac{1}{2} \{ \mathbf{F}^{eT} \mathbf{F}^e - \mathbf{I} \}, \quad (2)$$

where $\mathbf{T}^e$ and $\mathbf{E}^e$ are work conjugate stress and strain measures, $\boldsymbol{\sigma}$ is the Cauchy stress, and $\mathbf{C}$ is the fourth-order elasticity tensor.

The evolution of $\mathbf{F}^p$ is governed by crystallographic slip according to:

$$\dot{\mathbf{F}}^p = \mathbf{L}^p \mathbf{F}^p, \quad \mathbf{L}^p = \sum_s \dot{\gamma}^s \mathbf{b}_0^s \otimes \mathbf{n}_0^s, \quad \mathbf{F}^p(\tau) = \{ \mathbf{I} + \Delta t \mathbf{L}^p(\tau) \} \mathbf{F}^p(t), \quad (3)$$

where $\dot{\gamma}^s$ is the slip rate on slip system s , $\mathbf{L}^p$ is the plastic velocity gradient, $\mathbf{I}$ is the identity matrix, and $\mathbf{b}_0^s$ and $\mathbf{n}_0^s$ are the slip direction and the slip plane normal of s , respectively, in the undeformed configuration denoted by 0. The $\dot{\gamma}^s$ is related to the resolved shear stress ($\tau^s = \mathbf{T}^e \cdot \frac{1}{2} (\mathbf{b}_0^s \otimes \mathbf{n}_0^s + \mathbf{n}_0^s \otimes \mathbf{b}_0^s)$) and a characteristic shear stress (τ_c^s) for slip system s , according to a power-law relationship [39-41]:

$$\dot{\gamma}^s = \dot{\gamma}_0 \left(\frac{|\tau^s|}{\tau_c^s} \right)^{\frac{1}{m}} \text{sign}(\tau^s), \quad (4)$$

where $\dot{\gamma}_0$ is a reference slip rate (taken here as 0.001 s^{-1}). The exponent m is the strain rate sensitivity. Here it is set to 0.01, a value which applies to most metals at low homologous temperatures.

From $\mathbf{L}^p$ we can calculate the lattice spin $\mathbf{W}^e$ via

$$\mathbf{W}^e = \mathbf{W}^{app} - \mathbf{W}^p, \quad \mathbf{W}^p = \frac{1}{2} (\mathbf{L}^p - \mathbf{L}^{pT}), \quad (5)$$

where $\mathbf{W}^{app}$ is the spin applied to the polycrystal and $\mathbf{W}^p$ is the plastic spin. As shown in the right hand side of Eq. (5), $\mathbf{W}^p$ is the skew-symmetric component of $\mathbf{L}^p$. The integration of this constitutive model is performed using an iterative Newton-Raphson scheme [34].

3.2 Hardening law

For the evolution of the characteristic shear stress τ_c^s in Eq. (4) we use a dislocation density (DD) based hardening law formulation. The overall model is referred to be the DD-CPFE model. The model assumes that dislocations are stored according to thermally activated rate laws and as a consequence, the resistance stresses τ_c^s are a function of strain, temperature, and strain rate [30, 42-50]. This law is applied to every slip system s in Zr and Nb. For Zr, we make available prismatic $\{110\}\langle 11\bar{2}0 \rangle$ slip, pyramidal I $\{10\bar{1}1\}\langle \bar{1}123 \rangle$ slip and basal $\{0001\}\langle \bar{1}1\bar{2}0 \rangle$ slip [30, 51-53]. Basal slip is the hardest slip system and known to occur only at larger strains, after the easier slip systems have strain hardened [26, 54-56].

For Nb, the $\{110\}\langle 11\bar{1} \rangle$ and $\{112\}\langle \bar{1}1\bar{1} \rangle$ slip modes are made available for deformation. While we are aware that non-Schmid effects in BCC have been found to be important at certain conditions for capturing the plastic anisotropy [57-59], we neglect them in the

calculations to follow. In both metals, deformation twinning is possible, but it tends to be profuse at temperatures lower than the room temperature conditions being modeled here or under a high strain rate deformation [60-62]. Moreover, twinning was not found prevalent in prior studies of ARB Cu-Nb and Zr-Nb nanocomposites [27, 54, 63]. The selected crystal orientations studied here are much more favorably oriented for slip than for twinning with respect to the loading direction. Therefore, twinning was not considered in this work.

4. Orientational stability of single crystal Zr

The model is first applied to study the orientational stability of single crystal Zr. Orientational stability depends on the initial orientation. Upon application of load, the crystal deforms elastically until the onset of slip on a combination of slip systems. Generally, the slip activity, being the number of active slip systems and distribution of shear among them, will contribute to the reorientation of the crystal from its initial orientation. As defined in Eq. (5), the total spin rate depends on the plastic spin rate and the spin rate contributed by the applied loading. The relationship between the plastic spin rate and the active slip systems is given by:

$$\mathbf{W}^p = \sum_s \dot{\gamma}^s \frac{1}{2} (\mathbf{b}_0^s \otimes \mathbf{n}_0^s - \mathbf{n}_0^s \otimes \mathbf{b}_0^s). \quad (6)$$

One indication of orientational stability would be negligible or zero total spin rate. The null result would imply that under that loading state, the slip activity that develops in this crystal orientation is such that the initial orientation is maintained under loading. Positive spin rates signify orientational instability.

Here, we will study the orientational stability of Zr crystals in PSC, an approximation of rolling deformation. In PSC, the applied spin rate is zero, and thus, it is only necessary to analyze the plastic spin rate, which is a direct consequence of the slip activity. The plastic rate spin tensor $\mathbf{W}^p$ can be represented by a scalar rotation rate angle,

$$\dot{\theta} = \sqrt{(W_{12}^p)^2 + (W_{23}^p)^2 + (W_{13}^p)^2}, \quad (7)$$

which is also called the pure spin rotating about an axis $\mathbf{n}$ i.e., $\mathbf{W}^p = \mathbf{N}\dot{\theta}$, where $\mathbf{n}$ is the dual vector of the unit anti-symmetric tensor $\mathbf{N}$.

To relate orientational stability, we calculate the reorientation rate, $\dot{\theta}$ for single crystals with a given initial orientation under PSC. The 3D $\dot{\theta}$ contour maps were calculated over the entire orientation space, taking into account the HCP crystal symmetries. These symmetry operators can be found in the appendix A. The resulting orientation space is known as the HCP fundamental zone (FZ) $FZ = \{g = (\phi_1, \Phi, \phi_2) \mid 0 \leq \phi_1 \leq 2\pi, 0 \leq \Phi \leq \frac{\pi}{2}, 0 \leq \phi_2 \leq \frac{\pi}{3}\}$ [64-67].

Figure 5a shows the $\dot{\theta}$ calculated at a total strain of 0.02. Here in the interest of space we only display the most relevant 2D sections of this FZ on which the seven Zr orientations lie (see Table 1). In Fig. 5a, it can be seen that for most of the orientation space, Zr single

crystals are not orientationally stable. The spin-rate analysis indicates that two of the seven key Zr orientations have negligible spin rates: Zr1 and Zr4. The other orientations, Zr2, Zr3, Zr5, Zr6, and Zr7 have positive spin rates, meaning that they are not as stable and would reorient away from these starting orientations. The results are consistent with the texture development in single-phase, coarse-grained Zr. In the texture that stabilizes in large-strain, room-temperature rolling of Zr, the basal poles are distributed from the ND to the TD, indicating that none of these seven components are particularly preferred over the others [54].

For more insight, Figs. 5b-d present the $\dot{\theta}$ resulting from the individual slip modes. From the analysis we find that the instability of orientations in PSC arises from the prevalence of prismatic slip. Basal slip would not lead to a significant reorientation but pyramidal slip could render some of the orientations unstable. The predominance of prismatic slip is a consequence of the initial CRSS ratios we used for prismatic : pyramidal : basal is 1 : 11.1 : 11.8, which, as mentioned, have been shown to well represent the plastic deformation of Zr [32]. The orientational instability found for over most of the orientation space is a result of the pronounced CRSS anisotropy characteristic of Zr.

5.0 Orientational stability of Zr/Nb bicrystals

The results from the spin-rate analysis, while in agreement with observations of rolled coarse-grained Zr, are, however, in significant contrast with the texture development seen in the nanolayered Zr, where the opposite result holds: the orientations covering Zr2, Zr3, Zr5, Zr6, and Zr7 are largely orientationally stable, and Zr1 and Zr4, are not.

The change in the orientational stability of a Zr crystal as it reduces from micron to nanoscale dimensions could have been induced by the increasing constraint with the neighboring Nb crystals. Across the Zr/Nb interface, the adjoining Nb and Zr nanocrystals must co-deform. Nb is much less plastically anisotropic than Zr and deforms by substantially many more slip systems. It is possible that the nanograined Zr, when made to co-deform with a neighboring nano-grained Nb, can be brought into an orientationally stable state. Whether this happens would depend on the Nb crystal orientation and hence in this way, the Nb crystal could have provided the bias toward stability of Zr2, Zr3, Zr5, Zr6, and Zr7 over the other common rolling components.

To analyze the effect of Nb co-deformation on the orientaitonal stability of Zr, we simulate the rolling of several Zr/Nb bicrystals. The preferred crystal orientations in the texture of the nanolayered Nb phase were selected for this purpose. In conventional rolling of BCC metals, two main fibers develop, the α and γ fiber [68, 69]. Figures 6 and 7 display pole figures and the plots of the two fibers for three different layer thicknesses $h = 106 \mu\text{m}$, 92 nm and 41 nm [22]. Both fibers are seen to develop in the Nb phase, provided that the layer thicknesses of Nb were above 100 nm. The fiber plot for the $h = 106$ micron composite shown in Fig. 7 provides a typical example. However, as the Nb layers refine below 100 nm from 92 nm and then to 41 nm, the α and γ fiber intensify with peaks forming around

$\{111\} < 1\bar{1}0 >$, $\{334\} < 1\bar{1}0 >$ and $\{001\} < 1\bar{1}0 >$, with a narrow $\pm 5^\circ$ deviation. The five main Nb orientations are listed on Table 1 and depicted in Fig. 8.

Bicrystals were created by considering all combinations of these five Nb orientations and the seven Zr orientations in Table 1. Figure 9 shows the model set up of the Zr/Nb bicrystal and the mesh used. The FE mesh consists of 3300 C3D8 (continuum three-dimensional eight-nodal) elements, which are divided into two equal sections representing individual single crystals of Zr and Nb. We use periodic boundary conditions [70], wherein the deformation of each pair of boundary faces (top/bottom, front/back and left/right) is equal and as a result the stress tensors on each pair are opposite in sign. The PSC boundary conditions are prescribed by specifying the displacement along the negative z-direction, while enabling the model to expand in the positive x-direction and keeping the y-dimension of the model constant during the deformation. These represent mixed boundary conditions that permit a non-zero rigid spin, which are more accurate, particularly for coupled bicrystals, than pure plane strain compression boundary conditions that would overconstrained the system by enforcing zero rigid spin.

The same thermally activated dislocation density based hardening law used for Zr was employed for Nb, with, however, the specific slip modes and material parameters pertaining to Nb. The material parameters for $\{110\} < 1\bar{1}1 >$ and $\{112\} < 11\bar{1} >$ slip in Nb were characterized previously in [32] and these are the ones used here.

For assessing orientational stability, the Zr/Nb bicrystal simulations are used to analyze the c-axis lattice reorientation during rolling and 3D reorientation contours after 50% reduction. To establish a connection with the underlying deformation mechanisms, we then study the relative contributions of slip associated with each case.

For all bicrystal combinations, Figure 10 presents the reorientation pathways taken by the c-axis of the Zr crystal on basal pole figures as the bicrystal is deformed up to 50% rolling reduction (approximately 60% strain). The pathway shown is the average over all points in the crystal. The darker colored points are the starting orientation and the lighter color points follow the reorientation path with strain. The color shades are divided into strain increments of 0.2. In nearly all the cases, the Zr crystals reorient away from their initial orientation toward the TD. They appear to be even less orientationally stable when constrained to deform with the Nb crystal. The only exceptions are the Zr1/Nb1 and Zr4/Nb1 bicrystal combinations; however, these two Zr orientations were not predominant in the nanolayered composites. Therefore, the compatibility constraint imposed by co-deforming with the Nb crystal neither stabilizes the Zr crystal nor does it choose to co-deform in a stable manner with a particular Zr component.

For more detail, Figure 11 compares the reorientation contours of the bicrystals at 0.6 strain. Again we see that the Zr4/Nb1 is stable with low total reorientation. The Zr1/Nb1 bicrystal, although undergoing minimal average reorientation, develops orientation gradients. Even more outstanding finding from these contours is that most of the Nb crystals remain relatively stable, experiencing little reorientation. The orientational stability of Nb is influenced negligibly by Zr, an outcome that likely is a consequence of its

large number of slip systems and the relatively low differences in the CRSS values of {110} and {112} slip. Nonetheless, despite the stability of Nb and the kinematic constraint enforced by co-deformation between the Zr and Nb crystals, the Zr is still orientationally unstable. Thus, the predominance of the Zr orientations, Zr2, Zr3, Zr5, Zr6, and Zr7, has little to do with its co-deformation with Nb.

To relate texture evolution with the relative amounts of slip, we calculate from the model the relative activities of the three slip modes, basal, prismatic, and pyramidal slip. At an integration point at a given time (or strain) increment, we define the relative activity (RA^α) as:

$$RA^\alpha = \frac{\sum_\alpha \dot{\gamma}^s}{\sum_s^{Nsl} \dot{\gamma}^s} . \quad (8)$$

For analysis, the relative activity RA^α for a particular mode is then averaged over all integration points for every element.

Figure 12 shows the relative slip activities of the three modes in the Zr crystal during deformation. In nearly all cases, prismatic slip prevails, particularly near the end of deformation. The only exceptions lay in the two more orientationally stable bicrystals, Zr1/Nb1 and Zr4/Nb1, where pyramidal slip dominates. Basal slip contributes very little. Broadly, these results can be expected from the highly anisotropic CRSS ratios among these three slip modes (1:11.1:11.8), characteristic of Zr.

7.0 Reduced plastic anisotropy: Orientational stability of Zr and Zr/Nb bicrystals based on the nano-grained CRSS ratio

We consider next the second possibility that the highly oriented texture arises from a change in the slip activity resulting from a change in the activation of slip within nanocrystalline Zr. The prior set of CRSS values was based on the relative differences in activating and propagating prismatic, pyramidal, and basal slip dislocations in coarse-grained polycrystalline Zr. In these cases, the CRSS values are controlled by defects lying within the crystal interiors. In the present case, the Zr crystals are bounded by Zr/Nb interfaces and substantially less grain boundaries or surfaces. The Zr/Nb interfaces will then serve as the primary sites for nucleation, annihilation sites, and barriers for slip. The change in slip activity could occur because the critical stresses for activating the various types of dislocations from the interfaces are not as disparate. Once nucleated, dislocations can glide unhindered to the other interface. On this basis, it is also possible that the critical resistance to dislocation motion is dominated by the nano-scale mean free path between adjacent interfaces. Consequently the differences in CRSS values among the three slip modes could have been reduced.

To relate a reduction in the plastic anisotropy of Zr to the evolution of crystal orientation in rolling, we adjust the initial CRSS ratios among prismatic : pyramidal : basal slip to be nearly unity to 1 : 1.8 : 1.12 in the hardening law. Although these CRSS ratios differ substantially from that characterized previously for coarse-grained Zr, it is fundamental to note, however, that prismatic slip still remains the easiest slip mode.

7.1. Spin-rate analysis

To determine the effect of reduced anisotropy on orientational stability, we first repeat the plastic rate analysis on single crystal Zr with the reduced CRSS ratios. Figure 13 shows the corresponding $\hat{\theta}$ contour map. The striking difference from the analysis with conventional CRSS values is that all seven Zr crystal orientations are stable. This is a direct outcome of the substantially reduced anisotropy among the CRSS values. Thus less CRSS anisotropy, means more orientational stability.

7.2 Bicrystal deformation with reduced anisotropy

The spin-rate analysis with reduced plastic anisotropy in Zr shows that the orientational stability of the seven Zr single crystals has substantially improved. When co-deforming with a Nb crystal, the orientational stability can change. To better understand their development in the Zr/Nb nanolayered composites, we repeat the bicrystal simulations with reduced plastic anisotropy in Zr. We again consider a bicrystal consisting of all combinations of Zr and Nb orientations. Figure 14 shows the average c-axis reorientation pathways for Zr on basal pole figures. The most important finding is that by this metric the predominant components Zr2 and Zr3 in the nanolayered composites are stable with all five Nb crystals. This result is a significant deviation from the behavior of Zr/Nb bicrystals in which Zr deformed using the conventional set of activation stresses. Most importantly it is consistent with the highly oriented Zr textures that develop in the nanolayered Zr/Nb composites, where it was seen that Zr2 and Zr3 should prevail over the other common rolling components in rolling.

The paths taken by the Zr orientations, Zr5, Zr6, and Zr7, are shown in Fig. 14 columns 5, 6 and 7. Again, unlike the behavior of the Zr/Nb bicrystals with conventional CRSS ratios for slip, the bicrystals with Zr6 and Zr7 are orientationally stable. Zr5 is less so, but Zr5 reorients a small degree from its initial position toward Zr6. In contrast to conventional anisotropic Zr and consistent with experiment, for reduced anisotropic Zr, orientations Zr6 and Zr7 are orientationally stable.

Last, we remark on the calculated reorientation paths for those Zr orientations not seen in the nanolayered Zr textures, Zr1 and Zr4, which are shown in Fig. 14, columns 1 and 4. We observe that the usually stable components in rolled single phase Zr, Zr1 and Zr4 are no longer stable when co-deforming with Nb and the anisotropy of Zr has been reduced. Not only are they unstable, but they are found to reorient towards Zr2 and Zr3, the preferred components seen in the highly oriented texture that develops in the nanolayered Zr/Nb composites.

Figure 14 displays the c-axis reorientation only. The average total reorientation angles for anisotropic and reduced anisotropic Zr are compared in Table 2. We see that reduced anisotropy in Zr has the most profound influence on stabilizing Zr2 and Zr3 in rolling,

which strongly indicates that the unusual, highly oriented texture that emerges in the rolled nanolayered Zr phase is a consequence of reduced CRSS anisotropy.

For a more refined analysis of orientationally stability, Figure 15 presents the 3D reorientation contours of the Zr/Nb bicrystals after a certain amount of strain with the plastically low anisotropic Zr. As seen before in Zr/Nb bicrystals with conventional Zr CRSS values, the Nb remains stable during rolling. However, the compelling results lie in the behavior of the Zr crystal, which has changed substantially. First, compared to the single Zr crystal with the same reduced anisotropic CRSS values and orientation, the Zr crystal is less orientationally stable when joined to the Nb crystal. Thus, being bonded to Nb makes the Zr crystal less, not more, stable. Second, when bonded to the same Nb crystal, the Zr crystal with reduced anisotropy in slip reorients much less than the same Zr crystal with higher anisotropy. Last, when considering all five Nb crystals, bicrystals with Zr2 and Zr3 are the most stable and Zr1 and Zr4 are the least. Zr6 and Zr7 are orientationally stable for some Nb crystals, but not for others. In particular, when bonded to Nb3, Nb4, and Nb5, Zr orientation Zr7 develops orientation gradients. Thus, once again, consistent with the nanolayered Zr textures, these more highly resolved model calculations reveal that Zr2 and Zr3 are the most stable and should prevail over the other common rolling components in PSC, when co-deforming with Nb and the plastic anisotropy of Zr has been reduced.

7.3 Relative activities

With the model, we can directly link texture evolution with the underlying slip activity. With reduced anisotropy and remarkable changes in which orientations prevail as stable, we cannot expect the slip patterns to be the same as those seen in conventional Zr.

Figure 16 presents the relative activities associated with bicrystals for all Nb and Zr with reduced CRSS anisotropy. A common feature of the more orientationally stable cases, Zr2 and Zr3, when joined with most of the Nb orientations (Nb2-Nb5), is that they deform by similar amounts of slip from all three slip modes. The same collaborative slip pattern is also seen in the other outstanding stable orientation, Zr6, when joined with Nb2-Nb5. Conversely, those lesser stable bicrystals, such as those including Zr1 and Zr4, the deformation is dominated by pyramidal slip. It is important to note that for every starting bicrystal orientation, the CRSS ratios for Zr were reduced, but only in a few orientations does this reduction result in orientational stability in rolling. Most significantly, these few orientations are those seen experimentally in the highly oriented nanolayered Zr/Nb composites.

8. Discussion

In this work, deformation texture evolution is used as a signature for inferring the types of slip patterns that dominate the plastic deformation of nanocrystalline Zr. With crystal plasticity, we predict that only when the CRSS ratios among prismatic, pyramidal and basal reduce substantially to values below 2, do the predominant Zr2 and Zr3 orientations seen in nanocrystalline Zr become stable in rolling while the other conventional rolling components Zr1 and Zr4 turn unstable. The reason for the reduce anisotropy, however, has

yet to be clarified. It has been proposed in [71] that the reduction in CRSS differences among distinct slip modes occurs because in nanoscale crystals the CRSS has increased substantially above the friction stress for all slip systems. As one possibility, the enhancement could arise from a significant reduction in the mean free path for dislocation glide as layers attain nanoscale dimensions. In a recent study on the ARB Cu/Nb nanolayered system, a continual strengthening with increased reduction in h down to at least $h = 15$ nm was reported and shown to follow a Hall-Petch-like law [72]. On this basis, we can consider hypothetically the situation in which the CRSS enhancement is due to the Hall-Petch strengthening only, $\tau_{c-ng}^{\alpha} = \tau_{c-cg}^{\alpha} + K_{\alpha}/\sqrt{h}$, where τ_{c-cg}^{α} are the established CRSS values giving the macroscopic yield strength for coarse-grained Zr [32]. For $h = 100$ nm, the reduced anisotropic CRSS ratio of 1:1.8:1.1 can be estimated using $K_{prism} = 230 \text{ MPa } \mu\text{m}^{1/2}$, $K_{pyr} = 385 \text{ MPa } \mu\text{m}^{1/2}$ and $K_{bas} = 220 \text{ MPa } \mu\text{m}^{1/2}$. The resulting nano-grained, τ_{c-ng}^{α} values are 750 MPa, 1350 MPa, and 840 MPa for prismatic, pyramidal and basal. These values produce a material yield stress that is consistent with the hardness value of 4.5 GPa measured for this nanocomposite.

It may also be the case that the CRSS enhancement arises because the CRSS is associated with the barrier for nucleating dislocations from interfaces rather than the resistance to motion across a forest of dislocations accumulated the crystal. To obtain the ideal values for producing and moving dislocations, we calculated using first principles DFT the generalized stacking fault energy (GSFE) curves on the prismatic, basal, and pyramidal I slip system.

The DFT calculations were performed using the VASP code [73, 74]. In these calculations, a pseudopotential is employed to model the interaction between the valence electrons and ionic cores. For the exchange correlation functional in DFT, we adopted the generalized gradient approximation (GGA) with the Perdew-Becke-Erzenhof (PBE) parameterization [75]. The number of valence electrons in our Zr pseudopotential is four. To obtain the (GSFE) curves for a given slip (x-z) plane, the upper half of the crystal is shifted rigidly with respect to the lower half in the z-direction. Atoms are free to relax in the x and y directions. All the calculations were performed with periodic boundary conditions and with an energy cutoff of 400 eV for the plane wave expansion within the projector augmented wave (PAW) formalism [76, 77]. We used the gamma centered Monkhorst-Pack [78, 79] k -point mesh with a Gaussian smearing for the Brillouin zone integrations. The k -point mesh was large enough that the convergence of total energy was less than 1 meV per atom with respect to change in k -point mesh size. An optimized structure was obtained when the force on each atom is smaller than 0.01 eV/Å. The calculated $\langle a \rangle$ and $\langle c \rangle$ lattice parameters for the Zr primitive unit cell are 3.231 Å and 5.174 Å, respectively, which agrees well with experimental measurements [80] and earlier calculations [81].

Figure 17 shows the calculated GSFE curve for the prismatic slip system. This energy profile shows a very weak local minimum so dissociation of full dislocation into the partials is not stable and thus, unlikely. The peak value, or the unstable stacking fault, γ_{usf} , can be related to the barrier to produce the dislocation. The lattice resistance can be also

estimated from the γ surface by the maximum value of $(1/b)d\gamma/du$ [82]. For the prismatic dislocation, $\gamma_{\text{usf}} = 202 \text{ mJ/m}^2$ and the lattice resistance is 2.4 GPa.

For comparison, Fig. 17 also shows the GSFE curve for the basal plane $\{0001\}$ in the direction of the Burgers vector for a full dislocation $1/3 \langle 11\bar{2}0 \rangle$. For the full basal dislocation, $\gamma_{\text{usf}} = 273 \text{ mJ/m}^2$ and the lattice resistance, 3.5 GPa, which are higher than those for prismatic.

Last, the GSFE curve for pyramidal type I slip is provided in Fig. 17. Two local minima develop, suggesting that the dislocation can reduce its energy by dissociating into partial dislocations. The γ_{usf} and lattice resistance from the maximum of $(1/b)d\gamma/du$ are taken from the sense of direction where both are lower (from right to left in Fig. 17) and these are 479 mJ/m^2 and 5.2 GPa, respectively.

Taken together, we find that the ratios of the γ_{usf} from prismatic : pyramidal : basal are 1:2.37:1.35 and for the resistance they are 1:2.17:1.45., notably comparable to the CRSS ratios (1:1.8:1.12) found via crystal plasticity for the nanolayered Zr material and substantially reduced from the CRSS ratios characteristic of coarse-grained Zr (1:11.1:11.8).

9. Conclusions

In this work, we use a 3D multiscale crystal plasticity based model to study the orientational stability of nanocrystalline Zr in rolling. The material studied is nano-layered Zr/Nb composites made by accumulative roll bonding. It was reported that once the layers reduced below 100 nm, the texture became unusually highly oriented in a few special orientations. Thus, the nanolayered Zr phase provided the opportunity to study potentially unconventional aspects of plastic deformation of nanocrystalline hcp Zr as well as the effects of co-deformation between the Zr/Nb nanocrystals. Other SPD methods on single-phase Zr cause phase transformations or are unable to refine hcp Zr to very fine nano-grain dimensions.

The rolling deformation simulations are carried out on both single crystal Zr and Zr/Nb bicrystals. The analysis finds that Zr crystals, when bonded to Nb crystals become less orientationally stable than the same single crystal orientation under the same deformation state. We show that conventional plastic anisotropy characteristic of coarse-grained Zr leads to stable texture components that deviate from those that emerge in the nanolayered composites. A substantial reduction in the CRSS ratios among the slip modes below 2 causes the few observed texture composites in nanolayered to become stable at the expense of the instability of the more common components. The few special Zr orientations that prevail under reduced anisotropy involve nearly equal contributions of all three slip modes. This slip pattern is shown to be significantly different than that operating in coarse-grained Zr. These results thus strongly suggest that the differences in activation stresses for slip in nanograined Zr are significantly lower than that of coarse-grained Zr. The near equalization of the critical stresses for activating slip is attributed to a transition from

crystal-controlled to interface-controlled dislocation nucleation and propagation. Using DFT, we show that the reduced CRSS ratios among the three slip systems are consistent with the stresses required to form these dislocations. Although we focus this study on nanocrystalline Zr, the findings are sufficiently general to provide insight into mechanisms underlying the nanostructuring SPD processes of other hcp metals.

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Appendix A.

Table A1 shows 12 equivalent crystal orientations within the rotational point symmetry group associated with HCP crystals.

Appendix B. Supplementary data

The following are the supplementary data to this article:

1. All three poles for Zr5 with all the 5 Nb orientations with the conventional and reduced anisotropy CRSS values at a strain of 0.6 and
2. All three poles for Nb1 and Nb2 with all the 7 Zr orientations for the reduced anisotropy CRSS at a strain of 0.6.

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Fig. 1. Lamellar microstructures in the Zr/Nb composite produced by the ARB process: (a) optical micrograph showing the structure when the average layer thicknesses approximately 106 μm at a strain level of approximately 2.9 and (b) transmission electron microscopy diffraction contrast image showing the structure when the average layer thickness is approximately 92 nm at a strain level of approximately 10.

Fig. 2. Pole figures showing the deformation texture in the Zr phase of the rolled Zr/Nb composite to (a) 106 μ m, (b) 92 nm and (c) 41 nm layer thickness.

Fig. 3. $\langle 1\ 1\ \bar{2}\ 0 \rangle$ and $\langle 1\ 0\ \bar{1}\ 0 \rangle$ fiber plots for the Zr phase in the Zr/Nb ARB composites with nominal 106 μm, 92 nm and 41 nm Zr layer thickness.

Fig. 4. Basal pole figures showing location of the seven Zr components whose stability under rolling is studied in this work.

Fig. 5. Spin rate contours in selected cross-sections of orientation space containing all Zr components at 0.02 strain for the following slip resistance ratio: prismatic : pyramidal : basal 1 : 11.1 : 11.8.

Fig. 6. Pole figures showing the deformation texture in the Nb phase of the rolled Zr/Nb composite to (a) 106 μm , (b) 92 nm and (c) 41 nm layer thickness.

Fig. 7. Fiber plots for the Nb phase in the Zr/Nb ARB composites with nominal at 106 μm , 92 nm and 41 nm layer thickness.

Fig. 8. {111} pole figure showing location of the five Nb components studied in this work.

Fig. 9. FE model consisted of 3300 C3D8 elements used in the DD-CPFE bicrystal simulations.

Fig. 10. Basal pole figures for all 7 Zr and all 5 Nb orientations calculated based on the coarse-grained Zr slip resistance ratio: prismatic : pyramidal : basal = 1 : 11.1 : 11.8.

Fig. 11. 3D reorientation contours for all 7 Zr and all 5 Nb orientations calculated based on the coarse-grained Zr slip resistance ratio: prismatic : pyramidal : basal = 1 : 11.1 : 11.8.

Fig. 12. Relative activities associated with bicrystals for all 7 Zr and all 5 Nb orientations based on the coarse-grained Zr slip resistance ratio: prismatic : pyramidal : basal = 1 : 11.1 : 11.8.

Fig. 13. Spin rate contours in Zr calculated based on prismatic : pyramidal : basal slip ratio of 1 : 1.8 : 1.12.

Fig. 14. Basal pole figures for all seven Zr and all five Nb orientations based on the nano-grained (reduced anisotropy) Zr slip resistance ratio: prismatic : pyramidal : basal slip ratio of 1 : 1.8 : 1.12.

Fig. 15. 3D reorientation contours for all seven Zr and all five Nb orientations based on the nano-grained (reduced anisotropy) Zr slip resistance ratio: prismatic : pyramidal : basal slip ratio of 1 : 1.8 : 1.12.

Fig. 16. Relative activities associated with bicrystals for all 7 Zr and all 5 Nb orientations calculated based on the nano-grained Zr slip resistance ratio: prismatic : pyramidal : basal slip ratio of 1 : 1.8 : 1.12.

Fig. 17. DFT calculated GSFE curves for the prismatic, basal, and pyramidal slip systems for Zr. The displacement on the x-axis is normalized by the corresponding full dislocation Burgers vectors.

Nb {111}

Zr {0001}

Table 1. Selected Zr and Nb orientations studied here for their orientational stability in rolling.

#	Zr ($\varphi_1, \phi, \varphi_2$)	Nb ($\varphi_1, \phi, \varphi_2$)
1	(0°, 0°, 30°)	(0°, 0°, 45°)
2	(0°, 17.5°, 30°)	(0°, 25.24°, 45°)
3	(0°, 25°, 30°)	(0°, 35.26°, 45°)
4	(0°, 0°, 0°)	(0°, 46.69°, 45°)
5	(0°, 15°, 0°)	(0°, 54.74°, 45°)
6	(0°, 25°, 0°)	
7	(0°, 45°, 0°)	

Table 2. Average reorientation angle over the FE mesh for individual Zr components at a strain of 0.6: conventional and reduced anisotropy Zr CRSS ratio.

$\overline{\Delta\theta_{Zr}} [^\circ]$	Zr1	Zr2	Zr3	Zr4	Zr5	Zr6	Zr7
Nb1	8.8/ 4.5	20.8/ 1.5	17/ 0.6	0.8/ 0.3	23.3/ 9.6	19.7/ 7.8	22.6/ 1
Nb2	24/ 13.2	18.5/ 4.4	14.3/ 0.8	24.6/ 16	22.1/ 10.6	19.9/ 6.3	24/ 1.2
Nb3	26.6/ 12.2	17.8/ 3.4	13.5/ 0.7	28.5/ 17.7	21.8/ 9.8	20.1/ 5.5	24.1/ 4.1
Nb4	28.7/ 11.7	17.8/ 3.1	13.2/ 0.6	31.5/ 17.1	22/ 8.4	19.8/ 4.2	24.7/ 9
Nb5	28.7/ 11.2	18.2/ 2.7	13.5/ 0.5	30.6/ 16.8	22.4/ 7.7	19.7/ 4.4	24.9/ 9.8

Table A1. Twelve symmetry operations within the hexagonal close packed (HCP) rotational point symmetry group.

Symmetries 1–4	5–8	9–12
$(\varphi_1, \Phi, \varphi_2)$	$\left(\varphi_1, \Phi, \varphi_2 + \frac{\pi}{3}\right)$	$\left(\varphi_1, \Phi, \varphi_2 + \frac{2\pi}{3}\right)$
$(\varphi_1, \Phi, \varphi_2 + \pi)$	$\left(\varphi_1, \Phi, \varphi_2 + \frac{4\pi}{3}\right)$	$\left(\varphi_1, \Phi, \varphi_2 + \frac{5\pi}{3}\right)$
$(\varphi_1, \Phi + \pi, \varphi_2)$	$\left(\varphi_1, \Phi + \pi, \varphi_2 + \frac{\pi}{3}\right)$	$\left(\varphi_1, \Phi + \pi, \varphi_2 + \frac{2\pi}{3}\right)$
$(\varphi_1, \Phi + \pi, \varphi_2 + \pi)$	$\left(\varphi_1, \Phi + \pi, \varphi_2 + \frac{4\pi}{3}\right)$	$\left(\varphi_1, \Phi + \pi, \varphi_2 + \frac{5\pi}{3}\right)$