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## The Suppression of Instabilities via Biphase Interfaces During Bulk Fabrication of Nanograined Zr

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Severe plastic deformation (SPD) is a common method to fabricate nano-grained metals. However for Zr, a structural metal for nuclear applications, obtaining a nanoscale grain structure via SPD has been problematic due to deformation twinning and phase transformations. Here, nanostructured hcp Zr is fabricated through a refinement process via the introduction of a biphase interface. Despite mechanical and thermal conditions known to chemically mix Zr and Nb, no intermixing is observed and the heterophase interfaces appear resistant to phase transformations and twinning. Increasing the density of chemically sharp Zr–Nb interfaces is a very different refinement mechanism than substructure development, stacking fault formation, or alloying.

**Keywords:** Severe Plastic Deformation, Zirconium, Nanomaterials, Interfaces

Nanomaterials have been found to exhibit remarkable strength and, in some cases, simultaneous ductility.[1–6] Nanolayered metallic composites often achieve these exceptional properties while retaining good thermal stability.[7–9] Realizing these outstanding material properties in structural applications requires methods of fabricating nanomaterials in the bulk form. Over the past few decades, severe plastic deformation (SPD) techniques, such as wire drawing,[10,11] high-pressure torsion (HPT),[12–15] equal channel angular pressing (ECAP),[16,17] and accumulative roll bonding (ARB),[18–23] have been employed to successfully make nanomaterials and nanolayered materials. The last two processes, ECAP and ARB, can potentially produce nanomaterials in sheets and rods in both sizes and quantities suitable for structural applications.[24–26]

Nanostructuring (< 100 nm characteristic length scales) has been achieved primarily for cubic metals, such as face-centered cubic (fcc) and body-centered cubic (bcc) as well as composite combinations of the two, i.e. fcc–fcc and fcc–bcc.[17,22,27–30] Many technically relevant metals have a hexagonal close packed (hcp) structure and, to date, making bulk nano-hcp metals has been a challenge with SPD techniques.[19,20,23,31,32] In this work, we develop a method using ARB to produce nanolayered Zr–Nb composites with layer thicknesses  $h$  of ~90 nm.

It has been notoriously difficult to make nanoscale hcp metals (not only Zr but also Mg and Ti) in engineering quantities. Prior attempts to make nanostructured Zr metals created materials in small quantities. For example, nanolayered Zr–Nb composites have been fabricated

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using bottom-up approaches such as magnetron sputtering or other deposition methods.[33] Nanoscale Zr grains can be fabricated utilizing HPT.[13–15] However, HPT does not lend itself to the production of engineering scale quantities.[34] In addition, HPT can cause an allotropic phase transformation from hcp  $\alpha$ -Zr to hexagonal  $\omega$ -Zr and bcc  $\beta$ -Zr.[13,15,35] Similarly, while ball-milling can create nanograin Zr ( $\sim 5$  nm), a fraction of Zr transforms into an fcc phase.[36] Earlier attempts to use ARB to make Zr–Nb layered composites ended with a layer thickness of  $h = 4$   $\mu$ m, since further rolling led to shear banding.[37,38] It was found in the ARB of monolithic Zr that an ultra-fine-grained microstructure developed with equiaxed grains of diameter  $\sim 300$  nm.[39,40] Cryo-rolling followed by annealing treatments has been utilized to fabricate bimodal distributions of ultra-fine-grained and nanocrystalline Zr in the past.[41,42] However, this nanocrystalline Zr was captured during the annealing treatment as opposed to during the refinement process and showed little thermal stability as evidenced by the sharp decrease (70%) in the volume fraction of nanograins with an increase of 50°C in the annealing temperature.[41]

To fabricate nanolayered Zr/Nb, we started with  $h_0 = 2$  mm sheets of high-purity Zr [43] and reactor-grade, high-purity Nb (99.97% pure, ATI-Wah Chang) (see supplement for composition and initial microstructure). Both starting sheets showed measureable preferred texture and equiaxed grains with average sizes of 40  $\mu$ m for Zr and 300  $\mu$ m for Nb (see Figure S1 in supplement). The plastic deformation behavior of similar single-phase Zr has been well studied, and it has been found that it deforms primarily by slip with only a modest amount of

twinning at room temperature [44–46] and by an interplay of slip and twinning at 77 K.[37,46,47] Nb similar to that used here has been found to deform by slip, activating both  $\{110\}\langle 111 \rangle$  and  $\{112\}\langle 111 \rangle$  slip at room temperature in rolling and uniaxial testing.[38,48] The ARB process (see supplement for more detail concerning this process) consists of a repetitive sequence of cutting, cleaning, stacking, and roll bonding sheets with the initial step in our current process consisting of sandwiching a 2 mm Nb sheet between two 1 mm Zr sheets. Using this cladding technique, new Nb–Zr interfaces are only formed during the first rolling pass. As the number of bonding steps is repeated, the number of layers increases and the individual layer thickness  $h$  decreases. The total strain as a function of  $h$  is given by

$$\varepsilon = \ln \frac{h_0}{h} \quad (1)$$

Using the ARB process, we are able to obtain a final average layer thickness of 92 nm corresponding to an extreme strain level of 10.7. Figure 1(a) shows a representative transmission electron microscopic (TEM) image of the final nanostructure (see supplement for TEM methods). The interfaces are planar with modest morphological roughening. This is the first main achievement of this study—*fabrication of nanolayered Zr/Nb in quantities suitable for structural applications using deformation conditions amenable to mass production.*

While the ARB process is well known, a few key factors enabled success in this nanomaterial synthesis. Instabilities such as pinch-off (‘island formation’), debonding, and shear banding can prevent layer refinement. An earlier mathematical mechanics model [49]

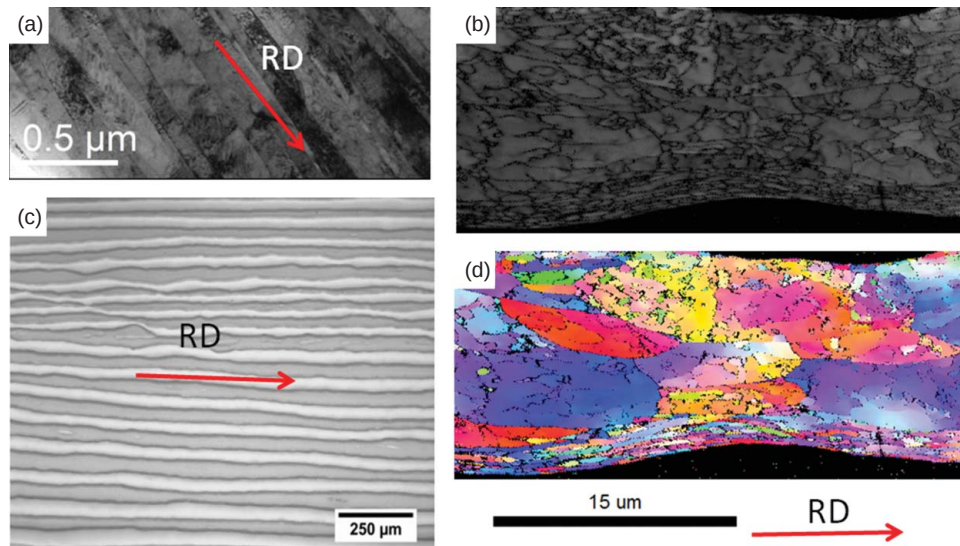


Figure 1. (a) Bright field TEM micrograph of Zr–Nb lamellar composite with  $h = 92$  nm. (b) Optical micrograph of Zr–Nb lamellar composite with  $h = 59$   $\mu$ m, illustrative of the bulk nature of the material fabricated. (c) EBSD image quality map showing sub-grain boundaries for  $h = 12$   $\mu$ m layer of Nb. (d) Orientation map from EBSD scan on material from (c). Note that dislocation cell structure is present at sub-grain scales for  $h = 12$   $\mu$ m.

suggested that these can be avoided with low ratios ( $< 5$ ) in flow stress between the two constituents. On this basis, static annealing steps at 575°C for 60 min were performed in this work after every second roll bonding pass corresponding to incremental strain intervals of 1.4. The time and temperature combination was chosen in order to minimize intermixing of the Zr and Nb, which can occur above temperatures of around 500°C.[50]

To investigate the chemical sharpness of the Zr–Nb interfaces, energy-dispersive spectroscopy (EDS) was utilized within the TEM. EDS line scans on the  $h = 92$  nm sample showed no signs of intermixing and showed chemically distinct, pure phases in the layers to within 4.5 nm from the interfaces. Figure 2 provides a representative high-angle annular dark field scanning TEM image with accompanying EDS line scan showing the sharp transitions between Zr and Nb. Parameters utilized in obtaining the EDS line scans are specimen thickness  $\approx 50$  nm, electron probe size of  $\approx 1$  nm, and electron beam energy of 300 keV.

According to its equilibrium phase diagram, Zr–Nb shows miscibility at relatively low (500°C) temperatures [50] and thus the intermediate annealing temperatures applied here could have led to interdiffusion between Zr and Nb. Furthermore, mechanical deformation from ball-milling and wire drawing has been known to induce chemical mixing even between immiscible metals.[51–53] Preservation of two phases allows the layer thickness to limit the grain size, thus facilitating

grain refinement to the nanoscales. In addition, the thermal stability of the nanostructure is greatly enhanced by the presence of the Zr–Nb bimetal interfaces in comparison with the single-phase Zr nanograins in cryo-rolling studies.[41,42]

To detect possible changes in the primary deformation mechanisms and crystal structures of the phases, we measured the bulk textures for each phase at all length scales using neutron diffraction (NeD, see supplement). NeD was carried out at the High Pressure Preferred Orientation neutron time-of-flight diffractometer at the Los Alamos Neutron Science Center at Los Alamos National Laboratory.[54,55] Compared with other texture measurement techniques, NeD enables statistically relevant data without potential modifications due to surface preparation. Texture measurements also provide a robust indicator of the primary deformation mechanisms, such as when interface-driven plasticity,[28,56] grain boundary-mediated processes,[57] deformation twinning,[58–60] and partial-mediated slip [61] prevail. Figure 3 shows examples of textures at the micron scale and nanoscale in the form of pole figures, a compact way to present texture, as well as the standard texture presentation for Zr (albeit not for Nb). The analysis undeniably shows that the Zr retains an hcp crystal structure throughout the extreme deformation. Thus, unlike nanostructured Zr produced by HPT,[13,15,35] nanostructuring via ARB resulted in no measurable allotropic phase transformation.

We observe that a preferred texture develops at all length scales which is distinct from the initial texture (Figure S1), and indicates that the prevailing mechanism is a crystallographic slip. Zr's strong texture indicates that interface or grain boundary-mediated processes, which tend to randomize the texture, are not active.[57] More importantly, the textures are remarkably similar (apart from the usual modest variations) over the wide range of  $h$  for each phase. Abrupt transitions in texture would have significant changes in the predominant deformation mechanism when  $h$  crosses from the micron to nanoscale, such as twinning, partial dislocation-mediated slip, or interfacial sliding. In spite of the increased density of interfaces and nanoscale dimensions, such transitions are not present in the Zr–Nb composite when refining from the micron scale to the nanoscale.

Another significant finding is that the textures in Figure 3 are very similar to textures in coarse-grained Zr and Nb when rolled alone. In past studies of rolled single-phase Nb, a texture was observed consisting of  $\alpha$  (001) and  $\gamma$  (111) fibers with particularly strong intensity associated with the  $\{001\}\langle 110 \rangle$  and  $\{111\}\langle 110 \rangle$  orientations.[62,63] Monolithically rolled Zr develops texture with strong intensities associated with  $\{0001\}$  that are tilted 20–40° away from the normal direction (ND) about the rolling direction (RD).[40,64,65] The textures of our Zr–Nb nanolayered composites (Figure 2) clearly

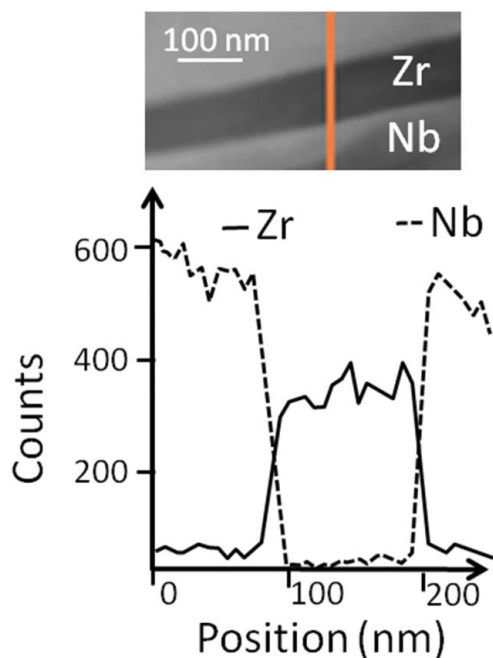


Figure 2. Zr–Nb composite micrograph where  $h = 92$  nm with accompanying EDS-based line scan. The sharp transition between Zr and Nb exhibited indicates chemically distinct phases to within 4.5 nm from the interfaces.

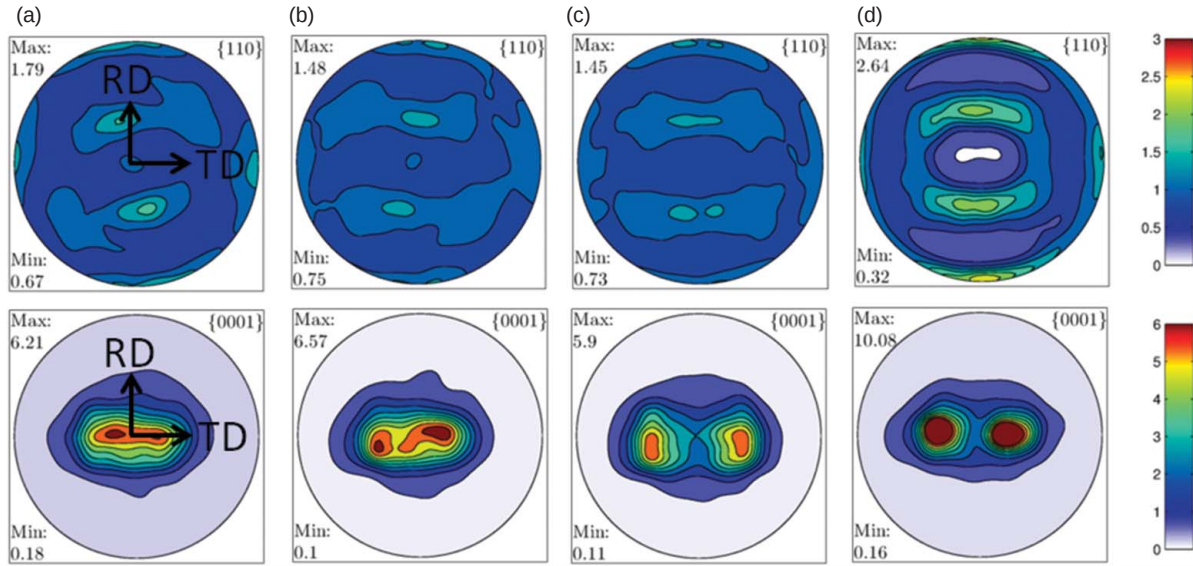


Figure 3. NeD-based pole figures for Nb  $\{110\}$  (top) and Zr  $\{0001\}$  (bottom) for length scales of  $h =$  (a)  $106\ \mu\text{m}$  as rolled, (b)  $26\ \mu\text{m}$  as rolled, (c)  $26\ \mu\text{m}$  annealed and (d)  $92\ \text{nm}$  as rolled. Zr pole figures are plotted on a scale of 0–6 MRD while Nb pole figures are plotted on a scale of 0–3 MRD.

exhibit the same predominant features of monolithic rolling textures within normal variation. Moreover, prior modeling studies that calculated the relative contributions of the different deformation modes clearly showed that these Zr and Nb rolling textures are a consequence of dislocation slip.[37,38] The similarity of the nanoscale phase textures measured here to conventional rolling textures is another indication that interface-mediated processes are not the dominant mechanisms of plasticity, even at submicron layer sizes. It was not previously known that dislocation-mediated plasticity could persist in hcp Zr down to the nanoscale. *Persistent dislocation plasticity in Zr in the nanoscale regime is the second key finding of this work.*

As a highly influential factor facilitating nanostructuring, we investigated the effect of annealing on microstructure and texture development during ARB processing. Figure 3(b) and 3(c) presents the  $26\ \mu\text{m}$  textures before and after annealing. For these micron layers, Zr and Nb both respond to annealing in a similar fashion as severely rolled hcp and bcc materials in the monolithic form.[63] Specifically, Nb maintains its weak rolling texture prior to and after annealing and Zr shows a reorientation of the basal poles from  $10\text{--}20^\circ$  to  $30\text{--}40^\circ$  away from the ND about the RD after annealing. In order to assess accompanying changes in grain and layer morphology, multiple electron backscatter diffraction (EBSD) scans were conducted on samples before and after annealing. In Figure 4, we present the grain structure, where it is clearly seen that both Zr and Nb recrystallize during the annealing step. The low internal grain misorientations in the recrystallized grains in Figure 4 also provide evidence that the dislocation content was

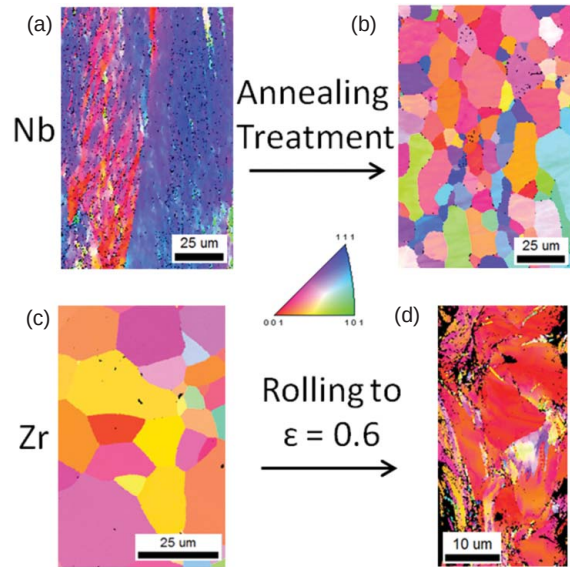


Figure 4. EBSD-based inverse pole figure maps for (a) Nb,  $h = 466\ \mu\text{m}$  as-rolled, (b) Nb,  $h = 466\ \mu\text{m}$  as-annealed, (c) Zr,  $h = 106\ \mu\text{m}$  as-annealed, and (d) Zr,  $h = 59\ \mu\text{m}$  as-rolled. Recrystallized microstructures are noted after annealing treatments.

sufficiently reduced during annealing. This reduction was likely instrumental in keeping the flow-stress ratio of Zr to Nb sufficiently low to suppress instabilities and enable nanoscale refinement.

The foregoing analysis warrants repeating for layer thicknesses approaching the submicron regime. Very recently in a study of ARB Cu–Nb composites, it was found that at finer layers  $h < 1.9\ \mu\text{m}$ , a typical recrystallized microstructure did not develop after

annealing.[66] A similar finding is revealed for the ARB Zr–Nb composites through an EBSD-based study. More than 20,000  $\mu\text{m}^2$  of Zr–Nb material ( $h = 1.5 \mu\text{m}$ ) was scanned in the as-rolled and annealed states with no difference in grain size noted. It should be noted that texture was consistent between NeD and the EBSD scans, implying that a representative area was scanned. This indicated that the annealing treatment had little microstructural and textural effects at this reduced length scale in comparison with Figure 4. Previously, it was proposed in [66] that the bimetal interfaces can act as sinks for dislocations, leading to a reduction in the dislocation content in the phases with strain and consequently a reduction in the driving force for recrystallization after deformation. Our analysis suggests that a similar phenomenon is operating in Zr–Nb. A reduction in dislocation content in the layers as they become single-grain thick is expected to be instrumental in continued layer refinement from the submicron to the nanoscales.

Another intriguing question concerns the apparent decoupled texture evolution of Zr and Nb in spite of the fact that the phases were clearly coupled microstructurally and mechanically. Microstructural coupling is clear from our EBSD and TEM analyses, which indicate that one grain spans the layer thickness in the submicron and nanolayered composites ( $h < 1.5 \mu\text{m}$ ). Therefore, all Zr and Nb grains are joined by bimetal interfaces. Mechanical coupling can be proven via a simple calculation, which indicates that the applied rolling reduction should have reduced both layers to a nominal  $h = 92 \text{ nm}$ . Achievement of this average value of  $h$  by both phases in the actual material indicates that the Zr and Nb phases co-deformed. In spite of these strong links, each phase in the composite develops a classical rolling texture as if rolled alone. This texture remains stable into the nanoscale regime with no noticeable texture transitions. In contrast, microstructural and mechanical coupling in ARB Cu–Nb composites caused two abrupt transitions in texture evolution. The first occurred when the layers became fine enough that one grain spanned the layer thickness ( $h < 2 \mu\text{m}$ ). The second one occurred when the layers reduced below 100 nm, invoking deformation twinning in Cu. Based on this comparison and current texture analysis,

it appears that in ARB Zr–Nb, the bimetal interfaces and nanoscale dimensions amazingly do not promote coupling in texture or deformation mechanisms different from those in the single-phase coarse-grained material.

In order to highlight this difference in more detail, Figure 5 compares the textures of the Nb phase in the Zr–Nb and Cu–Nb ARB composites in the submicron regime ( $h = 676\text{--}770 \text{ nm}$ ) where significant contrast in texture is expected to occur first. The textures are strikingly different, with an unusually sharp, atypical rolling Nb texture for the Cu–Nb composite and a typical but weak rolling Nb texture for the Zr–Nb composite. To elucidate this distinctive behavior at the nanoscale, we repeat this comparison for  $h = 60\text{--}90 \text{ nm}$  in Figure 6. Again, we observe that the Nb phase within the present nanolayered Zr–Nb composite corresponds to a weak rolling texture, whereas the Nb phase within the nanolayered Cu–Nb composite is very sharp and strongly deviates from a typical rolling texture. Evidently, texture evolution was phase-driven in the Zr–Nb and interface-driven in the Cu–Nb.

This last point naturally brings us to the important issue of interface evolution. Interface character and properties in cubic systems have been found to play critical roles in dislocation and twin nucleation.[67–69] To date, the available data and numerical studies on hcp/bcc interfaces are insufficient for understanding their role in plasticity. Certainly, the sharpness and planarity of the Zr–Nb interfaces within the present layered Zr–Nb composites inspire further exploration of the interface character. The weak rolling texture in Nb (Figure 2) signifies that the characteristics of many grain orientations are dispersed about the ideal rolling components. Very strong, single-crystal like textures, however, are associated with development of a preferred interface character (as defined by five parameters describing the orientation relationship and interface plane), as seen in the Cu–Nb system.[70,71] It is, therefore, unlikely that a prevailing interface forms in Zr–Nb. We therefore conclude that two contrasting SPD behaviors are possible for nanoscale composites: one where texture is driven by grain orientation stabilization as for Zr–Nb composites and one driven by interface character stabilization as in the Cu–Nb composites.

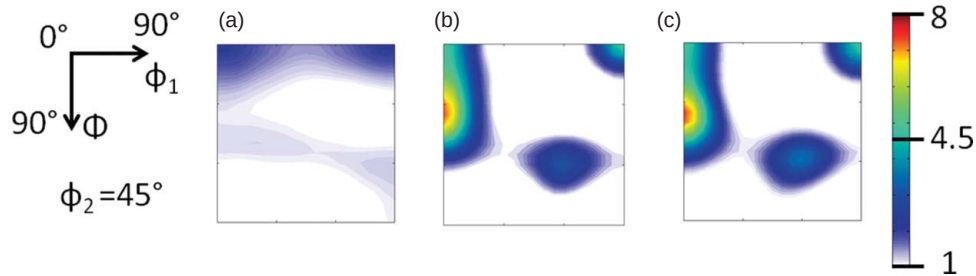


Figure 5. NeD-based orientation distribution function slices of  $\varphi_2 = 45^\circ$  for Nb in (a) Zr–Nb  $h = 676 \text{ nm}$  as rolled, last annealed at  $1.5 \mu\text{m}$ , (b) Cu–Nb  $h = 770 \text{ nm}$  as annealed,[21] and (c) Cu–Nb  $h = 714 \text{ nm}$  as rolled, with no prior annealing steps.[22]

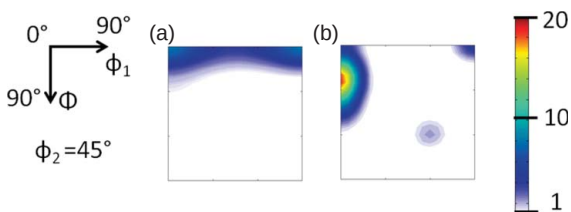


Figure 6. NeD-based orientation distribution function slices of  $\varphi_2 = 45^\circ$  for Nb in (a) Zr–Nb  $h = 92$  nm as rolled, (b) Cu–Nb  $h = 60$  nm as rolled, with no prior annealing steps.[22]

A key finding of this work is that nano-refinement arises as a result of the reduction in spacing between the bimetal interfaces. TEM results in Figure 1(a) provide a clear evidence that Zr and Nb are co-deforming as indicated by the similarity in layer thicknesses. Furthermore, our accompanying chemical analysis shows a lack of intermixing at the interface. Figure 1(c) and 1(d) both shows that thick Nb layers ( $h = 12 \mu\text{m}$ ) contain grain boundaries and a sub-grain dislocation cell structure, while Figure 1(a) shows that nanoscale layers exhibit little dislocation accumulation. Taken together, the results suggest that new grain boundary formation and intra-granular dislocation wall formation are not driving the refinement process at the nanoscale. Instead the bimetal interface is driving the refinement process through deformation carried by dislocation slip within the grains. An additional outcome of the texture analysis is that twinning and phase transformations are suppressed through the use of a bimetal interface as a refinement mechanism.

Despite thermal and mechanical conditions that could lead to chemical mixing of Zr and Nb, these two elements are found to maintain separation at the nanoscale under ARB with intermittent annealing treatments to form sharp Zr–Nb interfaces. The results imply that the Zr–Nb interface is in a lower energy state compared with a chemically varying interface and hence limits instabilities such as twinning and grain boundary sliding which have been observed to prevent nanostructuring of hcp Zr via SPD in the past. The lack of deformation-induced instabilities may indicate that the associated changes such as twinning and grain boundary sliding to the interface would induce an energy penalty. Phase transformations observed under HPT [13–15,35] arose as a result of a combination of stress state and pressure making it unclear if the interface, itself, is responsible for limiting the phase transformations.

In summary, we demonstrate a way to exploit the ARB SPD process to produce a 50/50 volume fraction hcp/bcc nanostructured composite material. The material fabricated is a two-phase hcp Zr–bcc Nb composite possessing a lamellar architecture with single nanocrystals spanning layer thicknesses. NeD measurements of texture indicate that dislocation-mediated plasticity persists even at the nanoscale with no evidence of interface-driven processes. This is in remarkable contrast to

cubic-based nanolayered systems produced by the same technique. Thus, critical length scales below which size-dependent or interface-driven mechanisms emerge in the hcp–bcc system, should they exist, must lie below 90 nm. Furthermore, based on the exceptional properties discovered for ARB Cu–Nb systems,[9,56,71–74] we envision that the Zr–Nb nanolayered material will exhibit many unusual and interesting properties, potentially far exceeding those of current coarse-grained Zr. This study opens up an innovative route to manufacturing hcp and bcc nanostructured metals as well as a novel material for pioneering hcp/bcc interface and nanoscale hcp research.

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**Supplementary Online Material.** A more detailed information on experiments is available at <http://dx.doi.org/10.1080/21663831.2014.941954>.

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