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Bifurcation in deformation behavior of Cu and Ta by accumulative roll-bonding at high temperature

Tarang Mungole [a,*], Bilal Mansoor [b], Georges Ayoub [b,c], David P. Field [a]

[a] School of Mechanical and Materials Engineering, Washington State University, PO Box 642920, Pullman 99164-2920, USA
[b] Mechanical Engineering Program, Texas A & M University, PO Box 23874, Doha, Qatar
[c] Department of Mechanical Engineering, University of Michigan, Dearborn 48128-2406, USA

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ABSTRACT

Cu and Ta are co-deformed at 673 K by the accumulative roll-bonding technique up to 8 passes. At an equivalent von Mises strain (ε_{vm}) of ~2 the deformation is bifurcated into shear accommodated by the 'soft' Cu, and plane-strain by the 'hard-but-ductile' Ta. This is attributed to transitions occurring collectively at ε_{vm} ~2 in crystallographic texture, partitioning of recovery and recrystallization, and the nature of interfaces as elucidated by orientation relations, Hall-Petch behavior and manifestation of instabilities at higher strain.

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Cu and Ta forms a class of an immiscible FCC/BCC material system, the study of whose co-deformation behavior is still in its infancy. Ta provides an effective barrier layer for diffusional transport of Cu into semiconductor substrates, due to its chemical inertness, so the Cu-Ta interface is of industrial importance [1]. The Ta facilitates adhesion between the Cu and dielectrics [2]. Furthermore, drawing of Cu/Ta and Cu/Nb multiphase systems has been proposed for the development of high strength and highly conductive composites for highly pulsed magnetic field applications [3]. In the current report, we investigate their co-deformation behavior by a well-established accumulative roll-bonding (ARB) [4] technique at 673 K due to some of the fundamental reasons mentioned here forth. Ta shows remarkably similar flow behavior (ductility of ~40%) at 773 K to Cu deformed at ambient temperature [5]. This similarity can even be extended to deformation of FCC metals [6]. Thus, Ta shows high ductility as opposed to other BCC metals in general. The reason is attributed to its relatively high stacking fault energy (SFE) (~57 mJ m^{-2} [7]) compared to other BCC metals and is similar to that of Cu (~70 mJ m^{-2} [8]). Ta also shows a negative or zero strain-rate sensitivity at 673 K (~0.2 T_m, where T_m is the absolute melting temperature) [6,9,10]. Compared to shear modulus ~30 GPa for Nb which is close to that of Cu, Ta displays a higher value of 69 GPa. Since Nb has a lower SFE (~40 mJ m^{-2} [7]) than Ta, it is hoped, that without sacrificing any ductility, Cu/Ta systems would possess higher strength, in general, than the relatively well-known Cu/Nb system, albeit being a similar FCC/BCC crystal system. The elastic modulus mismatch between components of a multi-layered material has been shown to promote instabilities at the interfaces [11] which have been observed in numerous reports in ARB of FCC/BCC metals [12–16]. Moreover, high temperature deformation of Cu is anticipated to alter the co-deformation characteristics of an FCC/BCC system by activation of non-octahedral slip (NOS) systems [17,18]. Thus a deviation from usual deformation textures in respective single phase materials as well as from usual FCC/BCC co-deformation texture is expected [19].

Plates of oxygen-free electronic Cu (99.99% pure) were purchased commercially and high purity Ta was received from Global Advanced Metals. Cu and Ta plates of dimensions 60 mm length, 10 mm width and 0.4 mm thickness were annealed for 1.5 h at 673 K (~0.5 T_m) and at 1373 K (~0.4 T_m) respectively and ARB was conducted in conventional manner [4] at 673 K up to 8 passes, leading to a total number of 2^N layers, where N is the number of ARB passes. Approximately 45% thickness reduction was achieved after each N. Samples were preserved after every pass, except for $N = 6$ and $N = 7$, for microtexture analysis by automated electron backscatter diffraction (EBSD). At each N, the analysis consisted of at least 5 EBSD measurements from individual layers on the normal direction (ND)-transverse direction (TD) plane, covering at least ~4000 grains. Microhardness measurements were recorded on individual Cu and Ta layers on the ND-TD plane up to $N = 5$ using the Vickers

* Corresponding author.
E-mail addresses: tarang.mungole@wsu.edu (T. Mungole), bilal.mansoor@qatar.tamu.edu (B. Mansoor), gayoub@umich.edu (G. Ayoub), dfield@wsu.edu (D.P. Field).

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indenter. Since the layer thickness was less than the indent size for $N = 8$, the hardness measurements of individual layers were not considered, owing to considerable effects due to interfaces.

Fig. 1a–b show the representative micrographs from the ND-TD plane of the rolled specimens after $N = 5$ and $N = 8$ leading to the formation of 32 and ~256 layers respectively. It is noticed that even in the ND-TD view where the inhomogeneities are least expected, the layers start to show waviness at $N = 5$ and through further straining, discontinuities at $N = 8$. These fluctuations of the interface morphology can be described in terms of a sinusoidal function $f(x) = e \sin(2\pi x/\lambda)$, where e and λ denote the amplitude and wavelength respectively. Under the action of a tensile stress σ_0, the energy of the interface perturbs and is balanced by elastic relaxation at the interface. The perturbed energy (ΔW) is described by [3,20]:

$$\Delta W = \frac{\pi \gamma e^2}{\lambda^2}\left(1 - \frac{\lambda}{\lambda_c}\right) \text{ with } \lambda_c = \frac{2\pi\mu\gamma}{(1-\nu){\sigma_0}^2} \tag{1}$$

where μ is the shear modulus, γ is the interface energy per unit area and ν is the Poisson's ratio. In our present investigation the effective tensile stress σ_0 is assumed to be of the order of 100 MPa with values of $\mu_{\mathrm{Ta}} =$ 69 GPa and $\nu_{\mathrm{Ta}} = 0.35$ [21]. Fig. 1b shows a zoomed-in RD-ND section at $N = 8$ from which the average wavelength can be calculated to be ~50 μm, hence λ_c ~ 25 μm. Cu/Ta interface energy at $N = 8$ can be estimated from (Eq. 1) as $\gamma_{\text{Cu-Ta}} = 0.38\ \mathrm{Jm^{-2}}$. This value is close to that reported ($\gamma_{\text{Cu-Ta}} = 0.48\ \mathrm{Jm^{-2}}$ [22]) by simulation of stable Cu/Ta interfaces. The waviness in the layers increases or equivalently the wavelength (λ), which is directly proportional to the interface energy ($\gamma_{\text{Cu-Ta}}$) from (Eq. 1), decreases with N. Thus the evolution of interfaces proceeds with decreasing interfacial energy making them relatively most stable at $N = 8$.

Equivalent von Mises strain (ε_{vm}) was estimated in each of the Cu and Ta layers by an embedded-pin technique. Cylindrical pins of Cu and Ta with 0.2 mm diameter were embedded in the respective plates and deformed to $N = 1$. Fig. 1c shows the central RD-ND plane of the embedded Cu and Ta pins after $N = 1$. The deflection curves $A'B'$ for the respective metals were used to measure the shear strain (γ_s) at $N = 1$ using [23]:

$$\gamma_s = \left[\frac{2(1-r)^2}{r(2-r)} \tan\theta\right] \ln\frac{1}{1-r} \tag{2}$$

where r is the thickness reduction during each pass and θ is the acute angle with respect to ND of the deflection curve $A'B'$. It can be noticed that a relatively high shear strain of ~3 is imposed in Cu after a single ARB pass. ε_{vm} for $N = 1$ was estimated by combining the shear strain with thickness reduction strain (ε_r) using the equations below:

$$\varepsilon_r = \frac{2}{\sqrt{3}} \ln\frac{1}{1-r} \text{ and } \varepsilon_{vm} = \sqrt{{\varepsilon_r}^2 + \frac{{\gamma_s}^2}{3}} \tag{3}$$

Evolution of ε_{vm} for $N > 1$ in individual layers during ARB was estimated by assuming that the additional or new strain accumulation after an ARB cycle is the same as that measured at $N = 1$, and distributed amongst Cu and Ta layers according to their spatial positions denoted by normalized thickness (t/t_0, where t is the distance from centre and t_0 the total thickness of the sample). Fig. 1d shows the complex nature of ε_{vm} as a function of t/t_0. Hence, this technique enables the ARB process to be viewed in terms of ε_{vm} which correlates with the spatial position across the thickness.

Figs. 2a–d show the representative orientation and grain boundary (GB) maps for Cu and Ta after $N = 3$ and 8. GB maps show boundaries categorized as high-angle (HAGB), low-angle (LAB) and very-low angle (VLAB) having the misorientations 15°–65°, 5°–15° and 2°–5° respectively. At ε_{vm} ~2.25, it can be observed from Fig. 2a that Cu is partially recrystallized whereas in Fig. 2b, Ta shows a deformed microstructure with grains elongated in TD. Fig. 2c shows that Cu is fully recrystallized with equiaxed grains at ε_{vm} ~5.8. However, Ta shows a fully recovered structure consisting of high-angle boundaries parallel to TD (also known as lamellar boundaries) in Fig. 2d. Low-angle interconnecting boundaries or incidental dislocation boundaries are shown by arrows. This is similar to observations of grains in rolling of pure Al [24], in Cu and Nb layers during ARB of Cu/Nb [25,26], and high-purity Al layers during ARB of Al of dissimilar purities [27]. The grain elongations were reported to be in RD with a similar or lower aspect ratio than in the present case. Occurrence of vast grain elongations in Ta towards the TD in the present report is not obvious from the usual plane-strain deformation during any rolling process. This can be attributed to a combined effect of Ta showing increasing ductility with increasing strain-rate at 673 K [9] and inhomogeneity of strain along t/t_0 depicted in Fig. 1d. The strain inhomogeneities directly correlate with inhomogeneous strain-rates in the specimen. Furthermore, it can be inferred from the evolution of the fraction of boundaries in Fig. 2e that the microstructure evolves until ε_{vm} ~3 in both Cu and Ta. The HAGB, LAB and VLAB fractions show identical values as the deformation progresses afterwards indicating the emergence of a steady microstructure for $\varepsilon_{vm} > 3$.

Generally, texture components evolving during rolling and shear of FCC metals coincide with those evolving in shear and rolling of BCC

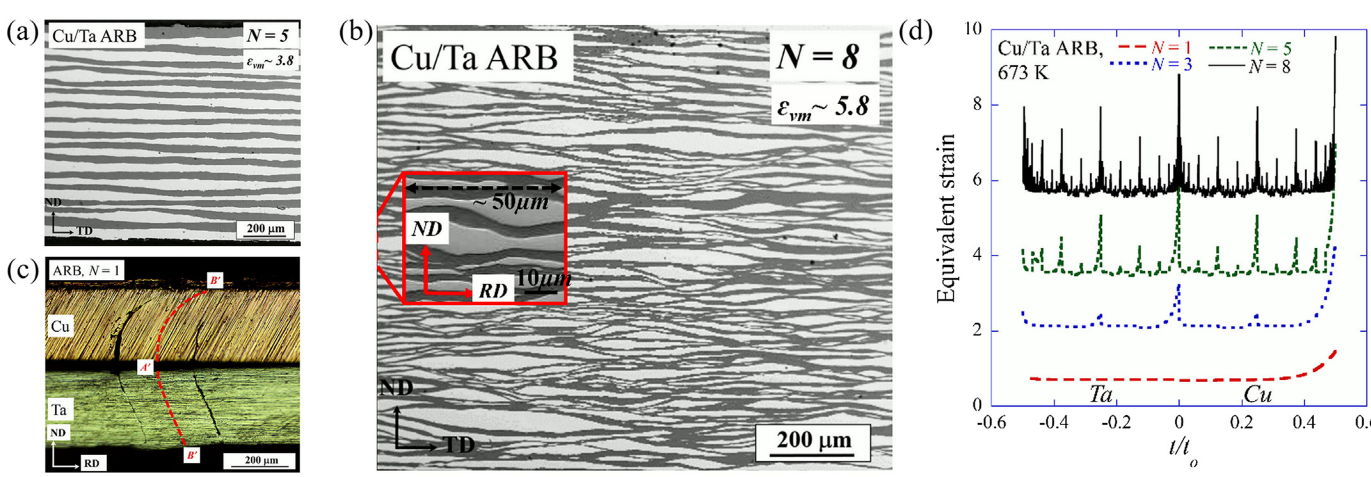

Fig. 1. (a) Representative image of the ND-TD plane of the Cu/Ta sample co-deformed via ARB at $N = 5$ and (b) at $N = 8$ with zoomed-in image of the RD-ND plane. Interfaces show waviness at $N = 5$ and additional discontinuities at $N = 8$. (c) shows the RD-ND plane with embedded Cu and Ta pins after $N = 1$ with average deflections ($A'B'$) and (d) shows the calculated variation of equivalent von Mises strain with the normalized thickness t/t_0 with rightmost layer always consisting of Cu. A complex strain variation is observed.