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Morphological stability of Cu-Nb nanocomposites under high-energy collision cascades

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We use molecular dynamics and phase field simulations to demonstrate that Cu-Nb multilayered nanocomposites with individual layer thicknesses above 2–4 nm remain morphologically stable when subjected to 100 keV collision cascades, characteristic of neutron or heavy ion irradiation. The probability of morphological instability rapidly increases as the layer thickness decreases to 1 nm, which we attribute to overlap of zones of liquid-like interdiffusion inside radiation-induced thermal spikes at neighboring interfaces in the multilayer. © 2013 AIP Publishing LLC [http://dx.doi.org/10.1063/1.4817785]

Several types of nanocomposites have exhibited remarkable resistance to radiation,1,2 motivating research into potential uses of nanocomposite structural materials in future fission and fusion reactors.3 Studies on model systems have revealed that the radiation resistance of nanocomposites is due to the fact that interfaces between adjacent components are often excellent sinks for radiation-induced vacancies and interstitials,4–6 as well as for implanted impurities, such as helium.7–11 Reducing microstructural dimensions increases radiation resistance because it shortens the average diffusion distance of point defects to interfaces. On the other hand, energy deposited by incoming neutrons or ions may lead to extensive intermixing across interfaces, promoting morphological instabilities in these materials.

We present molecular dynamics (MD) and phase field simulations that assess the limits of morphological stability in one type of model nanocomposite material: a layered composite of copper (Cu) and niobium (Nb) with individual layer thicknesses in the nanometer range.1,2,13 We show that the survival probability of Cu-Nb layered structures approaches unity for individual layer thicknesses above 2–4 nm. This finding suggests that it is possible to synthesize Cu-Nb multilayered nanocomposites that are both radiation resistant and morphologically stable under neutron or heavy ion bombardment.

We carry out our investigation in atomic models of Cu-Nb multilayers, such as the one shown in Fig. 1(a). Composites with four different individual layer thicknesses are investigated: 10, 18, 32, and 37 Å. Our models reproduce the crystallography of magnetron sputtered Cu-Nb multilayers, where Cu and Nb are in the Kurdjumov-Sachs orientation relation and the interfaces between them lie along closest packed planes ([111] in Cu and [110] in Nb).14 The simulation cells are cubic with ~300 Å edge lengths and contain approximately 1.9 million atoms, each. We use a newly constructed Cu-Nb embedded atom method (EAM) potential that has been fitted to the experimental phase diagram of Cu-Nb (including the liquid phase regions)15 and models short-range interactions using the Ziegler-Biersack-Littmark (ZBL) form.16

Collisions of neutrons or heavy ions with atoms in the multilayer are modeled by imparting a large kinetic energy—several tens of keV or more17–19—to randomly selected Cu or Nb “primary knock-on atoms” (PKAs). The initial structure is first equilibrated at 300 K and zero pressure for 5 ps, followed by initiation of a 100 keV PKA with a randomly selected velocity direction. The system is then allowed to evolve under constant energy and volume for up to 200 ps with dynamically varying time steps to maintain both high numerical accuracy and simulation efficiency. Ten such simulations are conducted for each layer thickness. For 100 keV PKAs, electronic stopping in Cu and Nb accounts for only about 10% of the stopping power16 and is therefore ignored.

As a PKA moves through the multilayer, it initiates a “collision cascade,” dissipating its kinetic energy through numerous collisions with surrounding atoms until it comes to rest. These collisions may lead to breakdown of the layered morphology of the composite by displacing atoms ballistically over distances larger than a layer thickness, or by creating transient pockets of molten material where rapid interdiffusion takes place.20 To identify collision cascade-affected regions, we subdivide the system into cubic voxels

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**Fig. 1.** (a) An atomic model of a Cu-Nb multilayer nanocomposite with 18 Å-thick layers. (b) A collision cascade initiated by a 100 keV PKA spans multiple layers.
with ∼6 Å edge length and calculate the local temperature by averaging the kinetic energy of all atoms in each voxel. In our simulations, regions where dissipation of PKA energy leads to local temperatures exceeding 1000 K (i.e., more than three times the ambient temperature) are connected and have characteristic dimensions of 100–200 Å, spanning multiple layers of Cu and Nb, as shown in Fig. 1(b).

After a collision cascade has come to completion, we search for regions of incipient morphological instability. Cu and Nb do not form compounds and have extremely low mutual solid solubility. Therefore, equilibrium Cu-Nb interfaces are atomically sharp and isolated Cu or Nb impurities created by irradiation de-mix over time scales governed by their diffusivities and the layer thickness. When the dose rate is low, the probability of multiple collision cascades occurring within a few seconds at the same position is negligible, so a single collision cascade can provide a reasonable estimate of multilayer stability under continued irradiation.

In our analysis, we adopt layer pinchoff as the criterion for onset of morphological instability. Pinchoff is known to initiate spherodization of Cu-Nb multilayers under high temperature annealing. It occurs when neighboring interfaces come into contact, forming a bridge between neighboring Cu or Nb layers. Besides irradiation, the likelihood of pinchoff may be affected by factors such as interface roughening due to plastic deformation, misfit strain, and the properties of triple junctions. Furthermore, at high dose rates, interfaces might not fully relax after each recoil event and defects created by multiple collision cascades may accumulate. Then, impurity precipitation and growth may initiate instabilities. In this work, we focus on the low dose rate case.

To determine whether a layer has pinched off, we simulate microstructure evolution after collision cascades using the phase field method. We solved for the local volumetric concentration of Cu, $\phi$, modeled as a conserved order parameter using Cahn-Hilliard equations implemented in 3-D in the COMSOL finite element method (FEM) code. The local free energy density function we used has a double well form

$$ f(\phi) = 4\Delta f \phi^3 (1 - \phi)^2, $$

where $\Delta f$ is the average of the free energies of mixing of Cu and Nb in FCC and BCC crystal structures at 600 K. Interface thickness is set to be ∼2 Å: comparable to the spacing between Cu {111} and Nb {110} planes and small enough to allow for convergence with respect to mesh element size. We ignore the elastic energy from the lattice mismatch between Cu and Nb, as well as local distortions due to defects and defect clusters created during collision cascades. Since we are interested only in the final state of the multilayers (pinched off or not), we performed all simulations in reduced time units.

An example of our phase field simulations is shown in Fig. 2. The initial concentration field, illustrated in Fig. 2(b), is obtained from atomic structures created by MD collision cascade simulations, such as that in Fig. 2(a). Pinchoff channels connecting some cascade-affected layers (mostly Nb) are formed during the phase field simulation while other layers return to their initial, flat morphology, as shown in Fig. 2(c). The width of pinchoff channels continues to increase with time, consistent with onset of the breakdown of layered morphology and eventual spherodization.

By analyzing all layers in all the simulations, we obtained the layer pinchoff frequency and multilayer survival probability as a function of layer thickness, shown in Fig. 3. A multilayer is considered to have survived if no layers in it have
pinched off. Only \( \sim 30\% \) of 10 Å multilayers survived after one collision cascade, while all other thicker layers above 18 Å are morphologically stable.

To better understand the initial conditions that give rise to layer pinchoff, we further determined the concentration of impurities in cylindrical regions with a basal diameter of 10 Å and spanning the entire thickness of individual layers. We then counted the number of occurrences when the concentration of impurities is greater than threshold values of 10%, 20%, or 30% across a single cylinder. For the thinnest (10 Å) layers, all the layers in the cascade-affected region contain cylinders with impurity concentrations that exceed the threshold. For larger layer thicknesses, the frequency of occurrence of such cylinder decreases and the “survival probability”—now understood to mean complete absence of cylinders with impurity concentrations above the threshold—increases.

For a threshold impurity concentration of 30%, the occurrence frequency and survival probability obtained this way are close to those predicted by the phase field method. Thresholds of 20% and 10% may be viewed as providing more conservative estimates of layer thicknesses that are stable against pinchoff. Therefore, referring to Fig. 3, we conclude that nanolayered composites of low solubility elements with layer thicknesses above 2–4 nm are likely to remain morphologically stable even when subjected to high-energy collisions with incoming neutrons or heavy ions.

We interpret our findings in light of current theories of radiation-induced mixing. The dissipation of energy by a PKA may be roughly divided into two stages: the “ballistic phase” and the “thermal spike.” During the ballistic phase, PKAs, like billiard balls, undergo high-energy, two-body collisions with surrounding atoms, which consequently become displaced. The ballistic phase lasts for a few tenths of a picosecond and—in sufficiently high-energy collisions—is followed by a thermal spike, in which the kinetic energy of the PKA is Boltzmann-distributed among all atoms participating in a collision cascade as a function of their kinetic energy to much heavier target atoms, such as Cu or Nb, creating isolated vacancy-interstitial pairs rather than intermixing. The ballistic phase lasts for a few nanoseconds.

These findings indicate that mixing in Cu-Nb multilayers does not take place by ballistic displacements, but rather by interdiffusion within the liquid cores of thermal spikes.

If mixing across each interface were independent of mixing at neighboring interfaces, the number of impurities per interface would be a constant independent of layer thickness. This is indeed the case for layer thicknesses of 32 and 37 Å. Fig. 4, however, demonstrates that for layer thicknesses below 32 Å, the number of impurities per interface after a thermal spike does depend on layer thickness: nanolayered composites with thinner layers exhibit more intermixing per interface than those with thicker layers. Thus, mixing across an interface in a nanocomposite with sufficiently thin layers is not independent of mixing across neighboring interfaces, but rather is enhanced by it. This enhancement likely arises from the overlap of regions of interdiffusion formed at neighboring interfaces during a thermal spike.

Experiments have shown that Cu-Nb multilayer nanocomposites with 2.5 nm layer thickness are morphologically stable under 150 keV He ion irradiation. However, such experiments are not a good measure of multilayer morphological stability under neutron or heavy ion irradiation because light ions such as He only transfer a small fraction of their kinetic energy to much heavier target atoms, such as Cu or Nb, creating isolated vacancy-interstitial pairs rather than the dense cascades and thermal spikes investigated here.

Previous experiments on Cu-Nb multilayers under heavy ion bombardment have focused on materials with layer thicknesses of 10 Å and occasionaly, 18 Å.

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Previous experiments on Cu-Nb multilayers under heavy ion bombardment have focused on materials with layer thicknesses of 10 nm and above.
on Cu-Nb multilayers with layers thicknesses in the range of 2–4 nm to confirm their morphological stability.

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