Quantitative Comparison of Sink Efficiency of Cu-Nb, Cu-V, and Cu-Ni Interfaces for Point Defects

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Abstract

The efficacy of hetero-interfaces as sinks for point defects in Cu was characterized using local measurements of tracer-impurity, radiation-enhanced diffusion (RED). The measurements were performed as a function of irradiation temperature and Cu thickness in multilayer samples, with the results being compared to steady-state kinetic rate equations to determine sink strengths. Cu-Nb Kurdjumov-Sachs (K-S) interfaces are found to be nearly ideal sinks for point defects, whereas Cu-Ni (111) hetero-epitaxial interfaces are poor sinks; Cu-V K-S interfaces are intermediate. Quantitative analysis of the RED data also yields the defect production efficiency for freely migrating defects in Cu, which is on the order of 1% for MeV Kr irradiation.

Introduction

Large supersaturations of point defects are often created in driven systems, such as those exposed to severe plastic deformation or energetic particle irradiation. These non-equilibrium defects frequently lead to undesirable microstructural evolutions; swelling, solute redistribution, and creep, for example, are particularly troublesome in reactor materials subjected to neutron bombardment. [1-4] Controlling the production, transport, and annihilation of point defects consequently remains a primary focus in efforts to develop materials that are tolerant to irradiation.[5] General strategies for eliminating non-equilibrium Frenkel pairs seek to create high densities of preferential sites for promoting recombination and/or unbiased defect absorption. These efforts include controlling the volume fraction and size distribution of solutes, precipitates, and interfaces with an emphasis on optimizing both the Frenkel pair recombination rate and stability of the microstructure.[6-10] Nanostructured materials have recently gained much attention for this purpose as they contain significant concentrations of hetero-interfaces, and they can often be adequately stabilized against coarsening.[8, 10-12] At present, however, the efficacy on these structures for defect annihilation and whether they act as saturable or unsaturable sinks remain largely unknown.

Experimental and computational studies suggest the strength of interfacial sinks can depend strongly on interface crystallography and chemistry.[8, 13] For example, the width of bubble or void denuded zones around grain boundaries and Cu-Nb interfaces of different crystallographic character suggests that high-angle grain boundaries are good point defect sinks, while low-angle grain boundaries, coherent twin boundaries, and Kurdjumov-Sachs (K-S) interfaces produced by physical vapor deposition are poorer sinks.[8] Measurements of irradiation creep on nanocrystalline Cu samples also suggest that high-energy grain boundaries are more effective sink sites than the interfaces between Cu and W nano-precipitates (in this case W acquires a Bain-related misorientation relationship, which is close to K-S).[14, 15] Local diffusivity measurements performed as a function of distance from

Cu-Nb interfaces, on the other hand, result in calculated vacancy profiles and a temperature dependence relatively consistent with an ideal sink.[16] This latter study is in agreement with theoretical modeling, which suggests that Cu-Nb should indeed be a good sink.[17]

The current work seeks to clarify the role of hetero-interfaces as point defect sinks and to better understand how sink efficiency varies with interface structure and chemistry. We use here the definition of sink efficiency as proposed by Sutton and Balluffi [18] i.e., $\eta = J_{\text{measure}} / J_{\text{perfect}}$, where $J_{\text{measure}} / J_{\text{perfect}}$ is the ratio of point defect flux to the interface in question to that for a perfect sink. This definition of efficiency depends on the irradiation conditions and sink geometry, but this will be discussed below.

Characterizing the distribution of point defects adjacent to interfaces experimentally provides a direct approach for quantifying sink efficiencies, as the concentration of defects and its gradient at the interface determine the defect flux, and thus it can easily be compared to computational models.[5, 19, 20] Measurement of defect concentrations adjacent to the interface, however, presents experimental challenges. Defect concentrations, in general, are difficult to quantify and direct approaches such as positron annihilation[21] or Balluffi-Simmons type experiments [22] do not achieve appropriate spatial resolution. The formation of dislocation, void, or bubble denuded zones near interfaces offers some insight into the distribution of point defects near an interface.[23, 24], but these methods require detailed modeling to extract quantitative results. Radiation enhanced diffusivity measurements, on the other hand, provide direct information about the average point defect concentration, and when measured in idealized geometries with known sink densities, they provide a convenient measure of sink efficiencies. We have previously used local tracer-impurity diffusivity measurements performed as a function of distance from an interface to map vacancy concentrations with approximately 10 nm resolution.[16] Such data can be fit to predictions from the steady-state chemical rate equations. The exact concentration that determines the boundary condition at the interface, however, is difficult to accurately define, since the profile of the tracer ideally should not intersect the interface, otherwise modeling is needed to include solute-interface interactions.

The present work extends this idea and compares sink efficiencies of different interfaces during MeV Kr ion irradiation; Cu-Nb K-S interfaces, Cu-V K-S interfaces, and Cu-Ni hetero-epitaxial interfaces all prepared by physical vapor deposition are examined. In this work, vacancy concentrations at the center of Cu layers in multilayer samples are obtained from experimental measurements of tracer diffusivity using Au marker atoms. The Cu layer thickness is varied within the multilayer samples to vary the strength of the sink. Au serves as a convenient tracer impurity, as it diffuses in Cu by a vacancy mechanism [25, 26], with a thermal diffusivity close to that of self-diffusion in Cu, and it has a high solubility in Cu even at quite low temperatures. Vacancy mobilities in Ni, Nb, and V are anticipated to be negligible in the temperature regime investigated [27], and thus the sink efficacy for Cu Frenkel pairs should not be significantly influenced by any defect fluxes in the second phase. We anticipate, moreover, that these different interfaces will exhibit different sink efficiency based on molecular dynamics calculations reported in the

literature[17, 28-31] and preliminary results of our own molecular dynamics simulations for Cu-Ni. The current experimental results are interpreted in the context of kinetic rate equations, which we use to relate the excess vacancy concentrations in the center of the Cu layers to the interfacial sink efficiencies.

Experimental Procedure

Sample Preparation

We prepared **multilayer coarse-grained thin film samples with circular cross-sections**. Thin films were grown on **needle-shaped** Mo substrates, described below, via magnetron co-sputtering of Cu (99.99%), Nb (99.95%), Au (99.99%), Ni(99.99%), and V(99.5%). Growth was performed at a base pressure of 3×10^{-8} torr and an Ar pressure of 2×10^{-3} torr. The Au tracer atoms were deposited at the center of the Cu layers as part of Cu-10 at.%Au layer, 5 nm thick. Rutherford backscattering spectrometry measurements were employed to verify the thickness and composition of these layers. Coarse grain Cu films with (111) texture were obtained by initially growing a 400 nm Cu layer on the substrates at 350°C. **Large grain sizes are promoted to ensure that the hetero-interfaces, rather than grain boundaries, are the primary sinks for point defects.** Prior studies have shown that V and Nb deposit on (111) Cu as BCC (110) with the K-S orientation relationship.[29, 32] Ni (111) is expected to grow hetero-epitaxially on Cu (111).[33, 34] Due to these preferred orientation relationships the large grain size from the underlying Cu persists through the subsequent thin films, which are grown at room temperature in order limit oxygen contamination and possible interface roughening. As illustrated schematically in Fig. 1, the samples were all grown with the configuration 30 nm M, 120 nm Cu, 30 nm M, 60 nm Cu, 30 nm M, 30 nm Cu, and 30 nm M, where M=Nb, Ni, V. Note that each specimen contains Cu layers with three different thicknesses.

Needle-shaped Mo substrates were prepared by first electropolishing 100 μm Mo wires in 5% NaOH solution, by weight, at 2.5 V. The wire is supported in a 1 mm Cu tube in order to provide structural stability. The electropolishing step results in a fine tip, which is subsequently milled using a focused ion beam (FIB, FEI Helios 600i) to produce a flat surface, approximately 10 μm in diameter. The tip then serves as the substrate for subsequent thin film growth.

Irradiation Conditions

Irradiations were performed at 300°C and 350°C using a 1.8 MeV Kr beam with an ion flux of $1.4 \times 10^{14} \text{ A-cm}^{-2}$. This leads to a defect production rate of $2.1 \times 10^{-2} \text{ dpa-s}^{-1}$ in Cu (dpa = displacements per atom), which is relatively constant (+/- $\approx 15\%$) throughout the Cu layers in the upper 330 nm of the samples utilized in this work [35]. Under these conditions freely migrating defects can reach interfaces before recombining. The total dose employed for the RED measurements was $8 \times 10^{14} \text{ cm}^{-2}$ ($\approx 1.9 \text{ dpa}$). Diffusion coefficients were also measured on reference samples that were held at the same temperatures and times as the irradiated samples, but not irradiated.

Characterization

After irradiation, the FIB was used to mill the samples to electron transparency using an annular milling routine. Characterization included transmission electron microscopy (TEM) and scanning transmission electron microscopy (STEM) in a JEOL 2010 LaB₆ and a JOEL 2010 FS, respectively. Selected area electron diffraction confirmed the orientation relationships between the thin films. Bright field and dark field TEM were used to confirm the absence of grain boundaries or significant lattice dislocations. High angle annular dark field (HAADF) STEM was used to distinguish the Au containing layer from the other constituents. The absence of grain boundaries in the thin TEM samples is consistent with grain size measurements based on scanning electron microscopy imaging in the FIB, which indicated grains at the surface were ≈ 450 nm. **The needle-shaped sample geometry facilitates energy dispersive spectroscopy (EDS) characterization by minimizing surrounding material that can contribute to absorption, fluorescence and other spurious signals, and it provides a well-defined measurable sample thickness that is useful for quantifying the local composition.**

Broadening of the Au tracer impurity was determined using EDS line scans. 5-10 separate line scans were performed at different locations, and the values were averaged to produce a single diffusion coefficient. This approach allowed us to collect more total counts with minimal effect of sample drift or contamination. Prior work using single scans resulted in larger overall error with poorer Gaussian fits, especially for dilute Au concentrations, due to the reduced statistics and/or greater drift [16]. Errors associated with the present diffusivity measurements derive principally from the uncertainty associated with fitting the Gaussian profiles. The uncertainties in our values for apparent activation energies are calculated from maximum and minimum values associated with a standard deviation at each temperature normalized by the square root of the number of independent measurements. **Prior experiments demonstrated that the chemical tracer diffusivity varies in Cu adjacent to Cu-Nb interfaces in a manner that correlates well with predictions from the steady-state rate equation [16] for radiation enhanced diffusivity. Therefore, the diffusivity in the current experiment is anticipated to vary as a function of position, which could affect the shape of the tracer profile. The diffusivity gradient is smallest at the center of the sample, where the current measurements were performed. Predictions from the steady-state rate equation indicate that in the worst case scenario of the 30 nm thick Cu-Nb sample over the full width at half maximum of the Gaussian the diffusivity varies by less than 10%. This is less than the magnitude of the experimental error bars.**

Results

Experimental Measurements of Radiation Enhanced Diffusion

Fig. 2 shows HAADF STEM, and SAED patterns for each of the samples. Cu-Nb and Cu-V both exhibit the K-S orientation relationship (note aligned fcc<111> bcc<110> in diffraction pattern), whereas data for Cu-Ni are consistent with a cube-on-cube hetero-epitaxial orientation (fcc<111>fcc<111>). The Cu-Nb and Cu-V interfaces are clearly distinguished in the images. The Cu-Ni interface is more difficult to observe due to the similar atomic masses of Cu and Ni. The dilute

Cu₉₀Au₁₀ tracer layer is also easily identified in each image.

Fig. 3(a) shows the composition profiles associated with samples thermally annealed at 350°C for times equivalent to those used for irradiation. The composition profiles do not broaden measurably relative to the initial 5 nm thickness, i.e., to within the ≈ 1 nm spatial resolution of the technique. Fig. 3(b) shows the measurements associated with compositional broadening during room-temperature irradiation. At this temperature, where thermally activated vacancy motion is negligible, compositional broadening is small, and it is independent of Cu-layer thickness, as expected for ballistic mixing, D_{mix} .

Fig. 4 shows the Au impurity-tracer composition profiles for different Cu film thicknesses adjacent to Cu-Nb interfaces, following irradiations at both 300°C and 350°C. Figs 5 and 6 plot similar Au impurity-tracer composition profiles in the Cu-Ni and Cu-V systems, respectively. Gaussian functions fit each profile well, and are used to calculate the impurity-tracer diffusivity. In each case, the diffusivities deduced from these profiles exceed those for thermal diffusivity by >4 orders of magnitude.[26] The large enhancement in diffusivity, D_{meas} , relative to thermally annealed samples is attributed to the non-equilibrium vacancy concentrations introduced into the lattice by irradiation and is indicative of radiation enhanced diffusivity (RED)[20]. We define RED here as, $D_{\text{RED}} = D_{\text{meas}} - D_{\text{thermal}} - D_{\text{mix}}$. The diffusivity at the center of Cu films adjacent to Cu-Nb interfaces exhibit a clear dependence on thickness, showing directly that the vacancy concentration varies as a function of Cu-layer thickness over the thickness range examined. The average apparent activation energy is $(0.04 \pm 0.11 \text{ eV})$; the negligible dependence on temperature indicates that under these irradiation conditions the system resides in the so-called sink-limited regime (more detail provided in discussion). Unlike the data for Cu-Nb samples, RED in Cu in the Cu-Ni specimens display no measurable dependence on thickness. The magnitude of the diffusivity, moreover, is significantly greater than in any of the Cu-Nb based samples. RED in these samples is temperature dependent, with an average apparent activation energy of $0.37 \pm 0.09 \text{ eV}$, which indicates that the system is in the recombination-limited regime (more detail is provided below). The RED measurements for Cu-V based samples irradiated at 300°C are similar to those in Cu-Nb samples; the radiation enhanced diffusivity depends strongly on Cu-layer thickness. These various sets of radiation-enhanced diffusion data are summarized in Fig. 7 in an Arrhenius plot.

Discussion

The experimental results are now interpreted in the context of chemical rate theory calculations. Within this framework, steady state is achieved when all defects can reach the most distant sinks. Since the characteristic time for vacancies to reach interfaces in even our thickest Cu layers, $\tau = L^2\pi^2/D_v \approx 0.012 \text{ s}$, is orders of magnitude shorter than the experimental irradiation time, $\approx 10^2 \text{ s}$, only steady state solutions are considered.

We first discuss the Cu films bounded by Ni layers. The lack of a variation in the radiation-enhanced diffusivity with Cu thickness in these samples indicates that Cu-Ni interfaces are, indeed, ineffective point defect sinks. We will express this more

quantitatively below. In this case, defects are eliminated primarily by recombination. Assuming D_{mix} and D_{thermal} are negligible in this temperature range, the measured diffusivity can be expressed as [20];

$$D_{\text{meas}} = D_{\text{RED}} = A \exp\left(\frac{-\Delta H_v^m}{2kT}\right) \quad (1)$$

where A is a prefactor which accounts for the irradiation flux. The measured activation energy in this regime is thus $\Delta H_v^m/2$ or half the vacancy migration enthalpy. The vacancy migration enthalpy in Cu is 0.72 eV [27], which is approximately double the activation energy measured here for the Cu-Ni samples, 0.37+/-0.09 eV. This agreement provides strong evidence that Frenkel pair recombination dominates in these samples, and that the Ni-Cu interface is a relatively ineffective sink.

As noted above, RED in Cu films bounded by Cu-Nb interfaces is nearly independent of temperature; the apparent activation energy is only 0.06+/-0.11 eV, i.e., to within the uncertainties, it is zero. This implies that in contrast to Cu-Ni interfaces, the Cu-Nb system is in the sink-limited regime, and that Cu-Nb interfaces provide an effective sink for point defects. While these results clearly demonstrate that Cu-Nb interfaces are far stronger sinks than Cu-Ni interfaces, a quantitative evaluation of the sink strength of these interfaces requires knowledge of the defect concentrations and defect fluxes in the vicinity of the interfaces. Since we have measured the defect concentrations only at the center of the Cu films, we use rate theory to relate these two concentrations.

In Fig. 8 we show results of rate theory for the steady state vacancy concentrations plotted as a function of position within a 120 nm Cu film, assuming different interfacial sink efficiencies. These calculations follow standard rate theory models (see e.g., Ref [36]), and similar to Ref [37], they assume the absence of internal sinks. The primary unknown parameter in this model is the rate for producing freely migrating Frenkel pairs, i.e., the defect production rate. For reasons discussed below, we assume it is $0.01K_{\text{NRT}}$ in these calculations, where K_{NRT} is the defect production rate obtained using the Norgett-Robinson-Torrens model [38], as calculated in SRIM [35]. Unit sink efficiency in this plot represents a perfect interfacial sink, i.e., $c(x=0, L) = c_{\text{eq}}$ or $c_{\text{excess}} = 0$. The interfacial flux under this condition, J_{perfect} , can be used to normalize the calculated fluxes, J_{measure} , in order to obtain the sink efficiency as defined above, $\eta = J_{\text{measure}}/J_{\text{perfect}}$. [18] The calculated vacancy concentrations at the center of the film for specific sink efficiencies can thus be compared with experimentally measured vacancy concentrations to determine η for different interfaces. Absolute vacancy concentrations are obtained from the RED data assuming that Au is a dilute tracer and that it diffuses by a vacancy mechanism. We then use literature values for the thermal diffusion of Au in Cu from ref. [26] and thermal vacancy concentrations in Cu from ref. [27] to relate diffusion coefficients to vacancy concentrations; see e.g. ref. [39] for details.

The concentrations of vacancies at the center of the Cu films calculated from rate theory are plotted in Fig. 8(b) as a function of film thickness for irradiations at 300°C, again assuming different sink efficiencies. For a perfect sink and absent mutual recombination in the film, the concentration should vary as the square of the

film thickness, since the process would depend only on diffusion to the interfacial sink. This is indeed found in Fig. 8(b) for the case of a perfect sink and thin Cu layers. Once the film thickness exceeds $\approx 100\text{nm}$, however, or the sink is not perfect, this simple scaling breaks down under the present irradiation conditions, as recombination begins to limit defect lifetimes. The slope on the log-log plot in Fig. 8(b) increasingly deviates from parabolic behavior with increasing film thickness until a maximum value of vacancy concentration is reached, where recombination dominates. The slope, moreover, also generally decreases with decreasing sink efficiency.

The experimental data for the vacancy concentrations **at the centers of the films** versus film thickness are plotted in Fig. 9 for comparison with the above calculations. We see that the slope in the curve for the Cu-Ni interfaces is to within experimental errors equal to zero. This provides convincing evidence that Cu-Ni interfaces are poor sinks ($\eta < 0.5$) and that the system is in the recombination regime for our irradiation conditions. As noted earlier, the temperature dependence of RED for Cu-Ni interfaces is also consistent with the system being in the recombination regime. We thus use these experimental observations to set the defect production rate to $0.01K_{\text{NRT}}$ in the rate theory calculations, as noted above. This normalization explains the perfect agreement between the absolute concentrations of vacancies calculated in the model and the experimental data. We note that the so-called defect production efficiency for freely migrating defect found here, i.e., the ratio the measured defect production rate to K_{NRT} , 0.01, is in good agreement with the less direct measurements of defect production efficiency in dilute Cu-Au alloys obtained using radiation-induced segregation behavior [25, 40, 41].

We next turn to the data for the Cu-Nb interfaces. These interfaces are clearly far more effective sinks than are the Cu-Ni interfaces. Comparison with rate theory calculations in Fig. 8 indicates, however, that while Cu-Nb interfaces are good sinks, they do not appear to be perfect sinks. Note that the slope of the experimental data in Fig. 9 does not show quadratic behavior. A sink efficiency between 0.95 and 0.99, on the other hand, fits the data quite well. Thus Cu-Nb interfaces present very effective sinks, if not perfect sinks. It should be mentioned that radiation-induced sinks for point defects, such as dislocation loops, could alter the behavior from that shown in Fig. 8(b). We discount that possibility here, since in that case the samples with Cu-Ni and Cu-Nb interfaces should behave more similarly, and the Cu-Ni samples should fall out of the recombination-limited regime. We thus expect that irradiation-induced sinks play at most a minor role in the current experiments. Lastly, we compare the experimental data for the Cu-V interfaces with Fig. 8(b). This comparison indicates that the sink efficiency for these interfaces lies between Cu-Nb and Cu-Ni, with $\eta \approx 0.9$ fitting the behavior quite reasonably.

Few experiments have measured the efficiency of interfacial sinks, and these previous attempts have been rather indirect. Siegel et. al, for example, evaluated the sink efficiency of grain boundaries by measuring the density of stacking fault tetrahedral in the vicinity of grain boundaries in Au following quenching from high temperatures.[42, 43] Such experiments generally suggest that interface sink

efficiency depends on interface structure and is higher for higher energy grain boundaries.[44] More recent experiments have looked at formation of voids near different interfaces during energetic He irradiations. The strength of the interface sink in this case was assessed by the widths of void denuded zones near the boundary. [13] In this work the authors similarly concluded that the sink efficiency of high energy boundaries is high; they further concluded that the sink efficiency of Cu-Nb interfaces is intermediate, and that of low energy boundaries (e.g. $\Sigma 3$ or low angle) is low.[8, 13] We first note that the general results of this study, that Cu-Nb interfaces are good sinks, agrees with the present results, although a quantitative comparison of the two studies is difficult. For example, Fig. 8(a) illustrates that the vacancy concentration is very sensitive to the sink efficiency close to the interface, and so a small change in the sink efficiency, for example from 1.0 to 0.95 will measurably influence the widths of denuded zones. Calculating void densities, moreover, is difficult, and so there is no easy way of quantitatively estimating the vacancy concentrations near the interfaces, or sink efficiencies, in these experiments. This problem is made even more difficult in the case where He is present in the sample. Although the authors suggest the He implantation energy is high and that little He is implanted near the interfaces, SRIM suggests that the He concentration is approximately 0.25%. Since He is known to greatly facilitate void nucleation, these studies measure a convolution of sink efficiency for both He absorption and vacancy absorption. The current measurements yield only the sink efficiency for point defects.

Lastly we compare our results with calculations and experiments related to the sink efficiencies of Cu-Nb and Cu-V by Demkowicz et al.[29, 31] Cu-Nb and Cu-V interfaces were calculated to have 5% and 0.9% constitutional vacancy concentrations (i.e. equilibrium interfacial concentration). These calculations are consistent with experiments indicating that Cu-Nb stores He 4.5 times more effectively than Cu-V during He irradiation.[29] The constitutional vacancies can also trap interstitials and subsequently facilitate vacancy recombination. We use these findings to compare our results on Cu-Nb and Cu-V samples; we compare the results from the 30 nm films, since Frenkel pair recombination is negligible in this case. We first note that Kinetic Monte Carlo simulations (to be reported elsewhere [45]) show that if the probability of absorbing a vacancy increases from 1% to 5%, then the average concentration of vacancies in a 30 nm thick Cu sample decreases by a factor of 2. The concentration at the center of Cu-Nb in our experiments is, in fact, 1.9 times lower than that in Cu-V in such 30 nm samples, and thus our experimental results are quite consistent with the calculations cited above.

Conclusions

Significant variations in interfacial Cu vacancy sink efficacy were measured as a function of interface type varying from approximately ideal behavior for Cu-Nb K-S ($\eta > 0.9$) interfaces to a moderate sink strength associated with Cu-V K-S interfaces ($\eta \approx 0.9$) to ineffective hetero-epitaxial Cu-Ni sinks ($\eta < 0.5$). The trend is qualitatively consistent with calculated and published predictions from molecular

dynamics indicating that the average point defect absorption probability should be highest for Cu-Nb interfaces and lowest for Cu-Ni interfaces. Data from both highly effective and ineffective sinks indicate that the defect production efficiency for freely migrating defects is on the order of 1%.

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Fig. 1: Schematic of the thin film geometry utilized in this study. Note that ≈ 5 nm $\text{Cu}_{90}\text{Au}_{10}$ layers were grown in the center of each Cu layer.

Fig. 2: HAADF STEM images of (a) the Cu-Nb, (b) Cu-Ni, and (c) Cu-V samples. Note that the left side of each image shows a Pt layer deposited to protect the sample, the right side of each image has the Mo wire and 400 nm Cu layer, and the multilayer investigated here is in the center. In HAADF STEM images the Nb, Ni, and V are brighter, comparable, and darker than Cu. Selected area diffraction patterns (inset), taken from the entire multilayer region, are consistent with K-S orientations for Cu-Nb and Cu-V, where the $\text{fcc}\langle 111 \rangle$ align with $\text{bcc}\langle 110 \rangle$ (note that these diffraction spots overlap in Cu-V), and cube-on-cube hetero-epitaxial $\text{fcc}\langle 111 \rangle \text{bcc}\langle 111 \rangle$ for Cu-Ni.

Fig. 3: EDS line scans of the Au composition profiles in (a) a 350 °C thermally annealed sample and (b) a room temperature irradiated sample.

Figure 4: EDS line scans of the Au composition profiles from Cu-Nb samples irradiated to $8 \times 10^{14} \text{ cm}^{-2}$ at (a) 300°C and (b) 350°C. The average values of diffusivity are $2.6 \times 10^{-20} \text{ m}^2 \text{ s}^{-1}$, $5.2 \times 10^{-20} \text{ m}^2 \text{ s}^{-1}$, and $8.6 \times 10^{-20} \text{ m}^2 \text{ s}^{-1}$ at 300°C, and $3.0 \times 10^{-20} \text{ m}^2 \text{ s}^{-1}$, $5.0 \times 10^{-20} \text{ m}^2 \text{ s}^{-1}$, and $1.0 \times 10^{-21} \text{ m}^2 \text{ s}^{-1}$ at 350°C for the 30 nm, 60 nm, and 120 nm samples, respectively.

Figure 5: EDS line scans of the Au composition profiles from Cu-Ni samples irradiated to $8 \times 10^{14} \text{ cm}^{-2}$ at (a) 300°C and (b) 350°C. The average values of diffusivity are $1.9 \times 10^{-19} \text{ m}^2 \text{ s}^{-1}$, $2.6 \times 10^{-19} \text{ m}^2 \text{ s}^{-1}$, and $2.4 \times 10^{-19} \text{ m}^2 \text{ s}^{-1}$ at 300°C, and 4.4×10^{-19} , 4.3×10^{-19} , and $4.3 \times 10^{-19} \text{ m}^2 \text{ s}^{-1}$ at 350°C for the 30 nm, 60 nm, and 120 nm samples, respectively.

Figure 6: EDS line scans of the Au composition profiles from Cu-V samples irradiated to $8 \times 10^{14} \text{ cm}^{-2}$ at 300°C. The average values of diffusivity are $5.1 \times 10^{-20} \text{ m}^2 \text{ s}^{-1}$, $8.0 \times 10^{-20} \text{ m}^2 \text{ s}^{-1}$, and $1.1 \times 10^{-19} \text{ m}^2 \text{ s}^{-1}$ at 300°C for the 30 nm, 60 nm, and 120 nm samples, respectively.

Figure 7: Summary of the radiation enhanced diffusivity data measured in this work plotted along with a line for thermal diffusivity.

Figure 8: (a) Vacancy concentration profile in 120 nm Cu thin films with interfaces of different sink efficiency calculated using kinetic rate equations. Note that changes in the vacancy concentration are more pronounced at the interfaces than in the center of the film. (b) Calculated vacancy concentration in the center of the film as a function of film thickness for interfaces of different sink efficiency.

Figure 9: Vacancy concentration **at the center of the film plotted** as a function of film thickness measured at (a) 300°C and (b) 350°C. Calculated trends for the bounding sink efficiencies of $\eta=0$ and $\eta=1$, assuming a defect production efficiency of 1%, are plotted as a guide.

Fig. 1

Fig. 2

Fig. 3a

(a)

Fig. 3b

(b)

(a)

(b)

(a)

(b)

Fig. 6

Fig. 7a

Fig. 7b

(b)

Fig. 8a

(a)

Fig. 8b

(b)

Fig. 9a

(a)

Fig. 9b

(b)