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ORIGINAL REPORT



Segregation-sandwiched stable interface suffocates nanoprecipitate coarsening to elevate creep resistance

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ABSTRACT

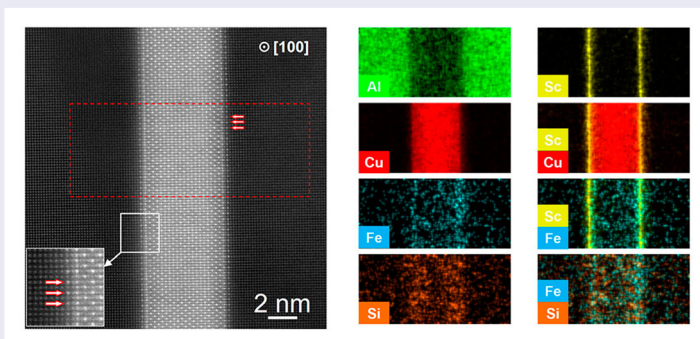
We demonstrate a strategy to stabilize nanoprecipitates in Al–Cu alloys, based on computational design that identifies synergistic solutes (Sc and Fe) that simultaneously segregate to the θ' -Al₂Cu/Al interface and strongly bond to one another. Furthermore, Sc and Fe are predicted to each segregate into a separate atomic plane, forming a sandwiched structure reinforcing the interface. This interfacial architecture was realized through a simple heat treatment in a Sc–Fe–Si triple-microalloyed Al–Cu model alloy. Such a back-to-back layered interface, thermodynamically stable and kinetically robust, is found to suffocate nanoprecipitate coarsening at 300°C, enabling a dramatic reduction in creep rate.

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KEYWORDS

Computational interface design; high-throughput calculations; nanoprecipitate stabilization; creep resistance



IMPACT STATEMENT

The segregant architecture of synergistic solute at θ' -Al₂Cu/Al interface was guided by computational calculations and artificially realized at atomic scale to achieve an ultra-high thermal stability of θ' -Al₂Cu, leading to a high creep resistance at 300°C.

1. Introduction

Precipitation hardening is widely used to strengthen structural materials used in aerospace, automotive, and construction settings. This strengthening route is particularly instrumental to aluminum (Al) alloys, the most

popular lightweight engineering material, in which a high number density of nanoprecipitates provides the resistance to dislocation motion to elevate strength [1]. However, the large volume fraction of precipitate/matrix interfaces is highly susceptible to thermal instability, i.e.

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precipitate coarsening at elevated temperatures [2]. The resultant loss of strength at temperatures above $\sim 200^\circ\text{C}$ hampers the load-bearing applications of commercial Al alloys in critical fields such as high-performance military aircrafts. It has been a challenge to engineer the precipitate/matrix interface in Al alloys to enable their structural uses at moderately elevated temperatures (such as 300°C).

According to the Gibbs absorption isotherm, the coarsening of precipitates is driven by the reduction of interfacial energy in the system. Therefore, a widely adopted strategy to stabilize nanoprecipitates is to minimize this driving force for coarsening [2]. This can be done by segregating a solute species to the precipitate/matrix interface [3,4]. Examples include Sc [5] (or Ag [6]) to the θ' -Al₂Cu/Al interfaces in Al–Cu-based alloys. The single-segregant decorated interface, although found to improve the stability of nanoprecipitates under hot conditions, often remains inadequate to survive long exposure to elevated temperatures [3,7].

In this paper, we report a novel interface design involving multiple solute elements segregated into multiple atomic layers. The model we demonstrate here is the case of an Al–Cu based alloy. Previously, Mn/Zr were used to co-segregate at the θ' -Al₂Cu/Al interface [4]; but the architecture we engineer is different: a stable interface structure is formed that has Sc and Fe each segregating in one atomic plane, forming back-to-back barriers reinforcing each other. Moreover, this Sc and Fe co-segregation ushers in a synergistic effect by forming tightly interlocked Sc–Fe bonding, further boosting the thermodynamic stability of the interface. The entire design, including the selection of the most effective solutes and the identification of their most preferable residence locations, is guided by our density-functional theory (DFT) prediction of the global minimum energy state. We have developed a heat treatment protocol that successfully fabricated the predicted stable interface, which almost eradicated the nanoprecipitate growth and elevated the creep resistance at 300°C by orders of magnitude when compared with the Al–Cu–Sc alloy that rely on single-solute segregation and the base Al–Cu alloy.

2. Experimental

An Al–Cu-based alloy with nominal compositions of Al–2.5wt.%Cu–0.22wt.%Sc–0.18wt.%Fe–0.04wt.%Si was used in this study. Detailed information of first principle calculations and experimental procedures were presented in SI.

3. Results and discussions

3.1. Computational interfacial design

Our computational design of the novel interface consists of three key steps. First, we examined the single-solute segregation energy as a function of the substituted atomic positions. This evaluates the tendency for segregation of each solute species towards the θ' -Al₂Cu/Al interface, and its most favorable physical location (atomic plane) at the interface when segregation happens. Second, based on the spatial position of these preferable sites, we selected two co-segregating alloying elements that would form back-to-back layered interface. Third, we confirmed that the multi-segregation-sandwich structure would reduce interfacial energy more than that expected from single-solute cases, demonstrating the synergistic effect of the interacting solutes.

Our design begins with a supercell model for the base Al–Cu alloy. As shown in Figure 1(a) and Figure S1, the model starts out with a coherent interface between the Al matrix and the θ' -Al₂Cu precipitate, with the first atomic layer of θ' -Al₂Cu enriched in Cu, as found in recent HAADF-STEM experiments [8]. Such a Cu close-packed coherent interface is identical to a GP(I) zone, which implies a relatively high interfacial energy ($\sim 0.39\text{ J m}^{-2}$ [9]) when compared with the θ' -Al₂Cu

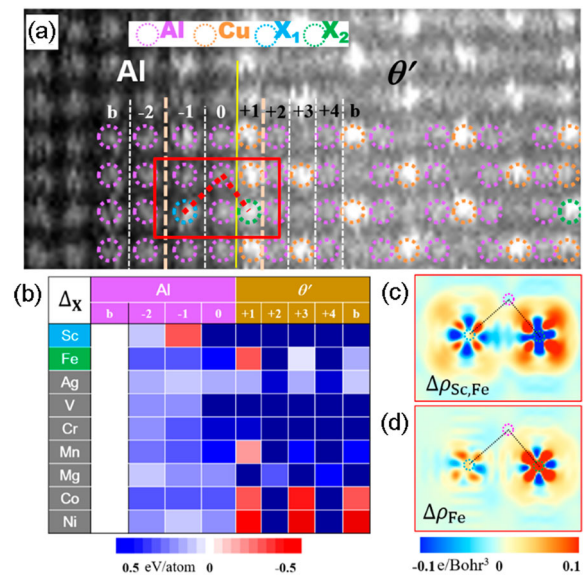


Figure 1. Targeted atomic locations at θ' -Al₂Cu/matrix interface from DFT calculations. (a) A representative HAADF-STEM image of the θ' -Al₂Cu/matrix interface, showing the supercell model used for computational design. (b) The DFT calculated segregation energy Δ_x corresponding to the crystallographically distinctive lattice planes (as denoted in (a)). (c) and (d) are the charge density distributions of the Sc–Al–Fe tri-layer, suggesting strong bonding and positive synergistic effect.

structure ($\sim 0.2 \text{ J m}^{-2}$ [4,10]). This kind of Cu-rich interface constitutes an intermediate state in the precipitate thickening process, as frequently found in the early growth stage of θ' -Al₂Cu. While in the later growth stage, the interface becomes less enriched in Cu and accompanies with severe θ' -Al₂Cu thickening. In our Al-Cu-Sc-Fe-Si alloy, the majority of θ' -Al₂Cu are characterized by extra Cu atoms at interface, which indicated the precipitate growth has been highly suppressed at early stage of aging, as a result of strong thermal stability of the precipitate. Solute X, one species at a time, was introduced into the supercell as the first step (one X atom into the 2×2 supercell depicted in Figure S1). The X atoms were progressively moved from the Al matrix (Al_b plane, which is sufficiently far away from the interface) towards the θ' -Al₂Cu precipitate, crossing the sharp interface (Al₀ plane) into the θ' platelet. To assess the propensity for segregation to each crystallographic plane at the interface (see Figure 1(a)), we evaluate the segregation energy $\Delta_X^i = E_X^i - E_X^b$, where E represents the total energy calculated from the state-of-the-art first-principles DFT supercell calculations, E_X^b is the total energy with X located at the Al_b plane, and i represents the plane (position) where solute X is located (see Methods for more details). The DFT calculated Δ_X^i corresponding to different planes is shown in Figure 1(b) and Table S1. We did systematic screening for a number of different X, one by one (see Figure 1(b) and Table S1). On the Al side, for segregation in the Al₋₁ plane, Sc was found to be the only solute that gives a negative Δ_X , i.e. it is the only choice to reside stably at this plane, among all single-solute possibilities. In the meantime, the Δ_X of Sc at this plane is also more negative than that at any other plane, indicating that this location represents the global minimum state for Sc. As such, Sc should be used when the objective is to build a solute-segregated Al₋₁ plane. The prediction of Sc segregation is consistent with earlier theoretical calculations [4], as well as with previous experiments showing that Sc lowers the interfacial energy and segregates towards the interface, and as a result reduces the coarsening of the precipitates at high temperatures [5,11]. On the θ' side, we aim to find the appropriate solute to segregate to the θ'_{+1} plane. This turns out to be Fe, which gives the lowest (the most negative) Δ_X among all solutes we have tried. In fact, this plane also corresponds to the global energy minimum for Fe, based on our trials of its location sampling all other planes. Taken together, we see that Sc and Fe prefer different planes, sandwiching the Al₀ plane (our computation showed that none of the solutes we screened would prefer to reside at the Al₀ plane). As such, Fe and Sc together could turn the originally sharp

interface between the Al matrix and θ' -Al₂Cu precipitate into an energetically favorable solute-clad interface.

We proceeded to confirm this, by adding Sc and Fe at the same time into the interface model. When both solutes are present together in the tri-layer configuration above (Al₋₁, Al₀ and θ'_{+1}), the energy is found to be lower than any other combination (i.e. Al₋₂, Al₀ and θ'_{+1} , etc.). Therefore, the sandwich configuration in Figure 1(a) turns out to be the preferred stable interface, the most energetically preferable spatial distribution upon simultaneous segregation of Sc and Fe (denoted as X₁ and X₂, respectively, in Figure 1(a)). The bonding between Sc, Al and Fe atoms at the interface is depicted in the charge density plots in Figure 1(c,d), in which increased charge density is marked red and a depletion of charge is shown blue. Figure 1(c) maps out the change in bonding charge density, $\Delta\rho_{\text{Sc,Fe}}(r)$, defined as $\Delta\rho_{\text{Sc,Fe}}(r) = \rho_{\text{Sc,Fe}}(r) - \rho_0(r)$, where $\rho_{\text{Sc,Fe}}(r)$ and $\rho_0(r)$ is the self-consistent interacting charge density of the Sc/Fe co-segregated interface and the original interface without solute, respectively. The positive $\Delta\rho_{\text{Sc,Fe}}(r)$ between Sc(Al₋₁)/Fe(θ'_{+1}) and Al(Al₀) atoms indicates enhanced bonding strength at the interface due to the co-segregation of Sc and Fe atoms.

There is in fact a synergistic effect when the two solutes Sc and Fe reach their targeted positions in Figure 1(a). To see this, the change in bonding charge density, $\Delta\rho_{\text{Fe}}(r)$, defined as $\Delta\rho_{\text{Fe}}(r) = \rho_{\text{Sc,Fe}}(r) - \rho_{\text{Sc}}(r)$, where $\rho_{\text{Sc}}(r)$ is the self-consistent interacting charge density of the Sc-segregated interface, is shown in Figure 1(d). Our DFT calculations indicate a positive $\Delta\rho_{\text{Fe}}(r)$ between Sc(Al₋₁) and Al(Al₀) atoms, indicating an additional enhancement of the bonding between the segregated Sc atom and Al atom, when Fe is brought to the neighboring plane. This synergistic effect between Sc(Al₋₁) and Fe(θ'_{+1}) is also supported by the calculated interfacial adhesion energy (Table S2): the cohesive strength across the interface is increased significantly by the co-segregation of Sc(Al₋₁) and Fe(θ'_{+1}).

The challenging task, then, is how to use a simple experimental approach to realize the desired stable tri-layer interface. From the picture discussed above, Fe should be pre-stored in the θ' -Al₂Cu precipitate, rather than as dilute solute in the Al matrix; this way Fe can migrate to the θ'_{+1} plane without having to cross the Sc-walled interface from the Al side during heat treatment. However, the Δ_{Fe} inside the θ' -Al₂Cu precipitate (except the θ'_{+1} plane) is slightly positive, such that Fe would not preferentially reside inside the precipitate. To address this problem, we extended our computational design to incorporate a third microalloying element, Si in our case, based on the two criteria as mentioned in SI (Section 1.3).

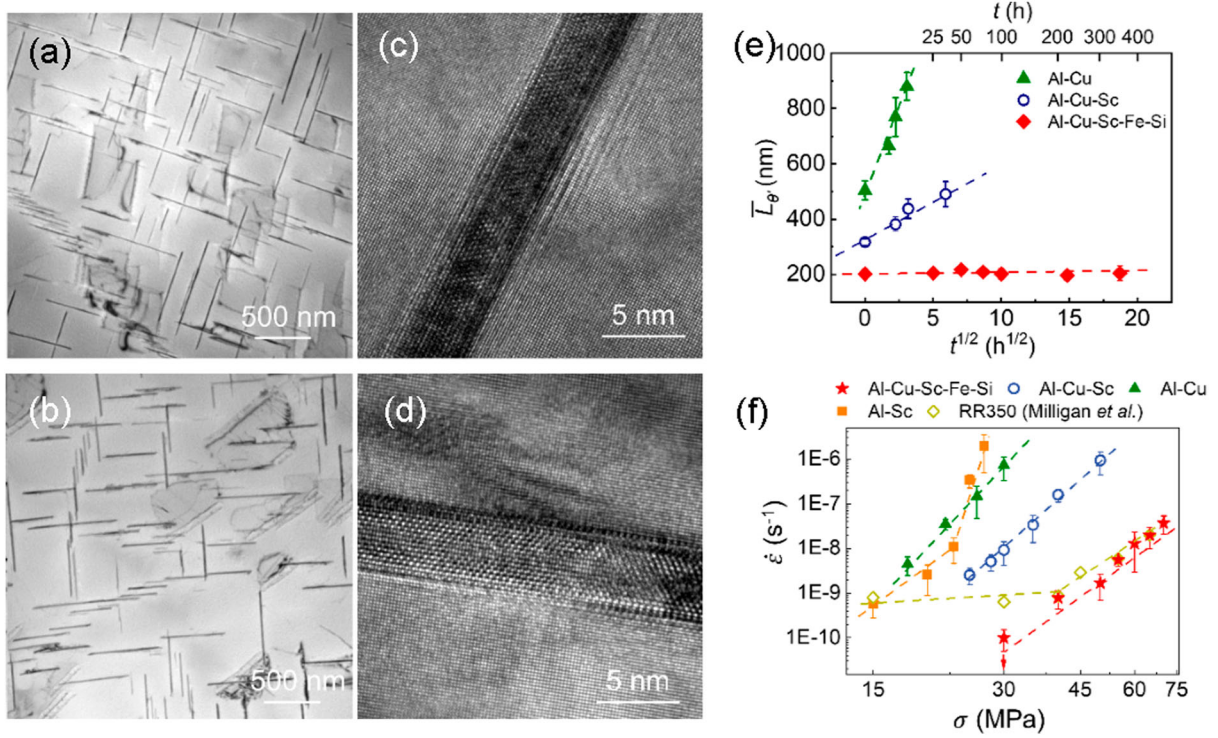


Figure 2. Unprecedented nanoprecipitate stability and creep resistance at 300°C. Representative (a–b) TEM and (c–d) HRTEM micrographs to compare the θ' -Al₂Cu precipitates before (a,c) and after (b,d) creep test at 300°C/30 MPa for 450 h. The θ' -Al₂Cu precipitates are highly stable. (e) Average half length $\bar{L}_{\theta'}$ as a function of square root of creep time $t^{1/2}$. (f) Double logarithmic plot of steady-state creep rate $\dot{\epsilon}$ vs applied tensile stress σ for the creep test at 300°C of Al–Cu–Sc–Fe–Si alloy, in comparison with the Al–Sc, Al–Cu and Al–Cu–Sc counterparts.

3.2. Experimental realization of multi-segregant interface

Following the guidance from the computational design, the experimental realization adopts triple (Sc–Fe–Si) solute elements in an Al–Cu alloy (see SI for details). Figure 2(a–d) compares the microstructure in the Al–Cu–Sc–Fe–Si alloy (see chemical composition in Table S3) after artificial aging (*as-aged* condition) and after creep test at 300°C under 30 MPa for 450 h. The coarsening kinetics of the average half length of the θ' -Al₂Cu plate, $\bar{L}$, is expected to follow [12]:

$$\bar{L}_{\theta'}^n - \bar{L}_0^n = k_{\theta'}(t - t_0) \quad (1)$$

where $\bar{L}_0$ is the average half length at time $t_0 = 0$, $k_{\theta'}$ is the coarsening rate constant, and n is the temporal exponent. The value of n depends on the coarsening mechanism [13] and was suggested to be $n = 2$ for plate-like θ' -Al₂Cu precipitates in interface-reaction dominated coarsening [14]. The evolution of $\bar{L}_{\theta'}$ is plotted against $t^{1/2}$ in Figure 2(e), comparing several Al–Cu based alloys [5,11]. The θ' -Al₂Cu precipitates in the Al–Cu–Sc–Fe–Si alloy are highly stable during 300°C creep test: it is remarkable that after the long test (450 h) $\bar{L}_{\theta'}$ remained practically unchanged from the ~ 220 nm before the test.

This is a dramatic suppression of the obvious precipitate coarsening measured for the Al–Cu or Al–Cu–Sc counterparts, as contrasted in Figure 2(e). In terms of $k_{\theta'}$, we see from the slopes in Figure 2(e) the sizable $k_{\theta'}$ of these reference alloys is reduced drastically, to almost zero, after the triple Sc–Fe–Si microalloying.

To examine the chemistry at the θ' -Al₂Cu/matrix interfaces, the atom probe tomography (APT) mapping and corresponding proxigrams compare the distribution and elemental concentrations at the coherent θ' -Al₂Cu/matrix interface in crept Al–Cu–Sc alloys with versus without Fe and Si microalloying, see Figure 3(a–e) and Figure S4(a–e). For the Al–Cu–Sc alloy, relatively weak Sc segregation (peak concentration ~ 3.0 at.%, see Figure S4(e)) is observed at the θ' -Al₂Cu/matrix interface, accompanied by extremely low Fe and Si aggregation originating from trace impurities. In comparison, for the Al–Cu–Sc–Fe–Si alloy, both Sc and Fe are significantly depleted in the matrix and strongly segregate to the θ' -Al₂Cu/matrix interface, with peak concentration of ~ 7.5 at.% and ~ 1.0 at.%, respectively, see Figure 3(c–e) and Table S4.

In particular, securing the coherent interfaces (CIs) together with semi-coherent interfaces (SCIs) is quite important in preventing the θ' -Al₂Cu from coarsening

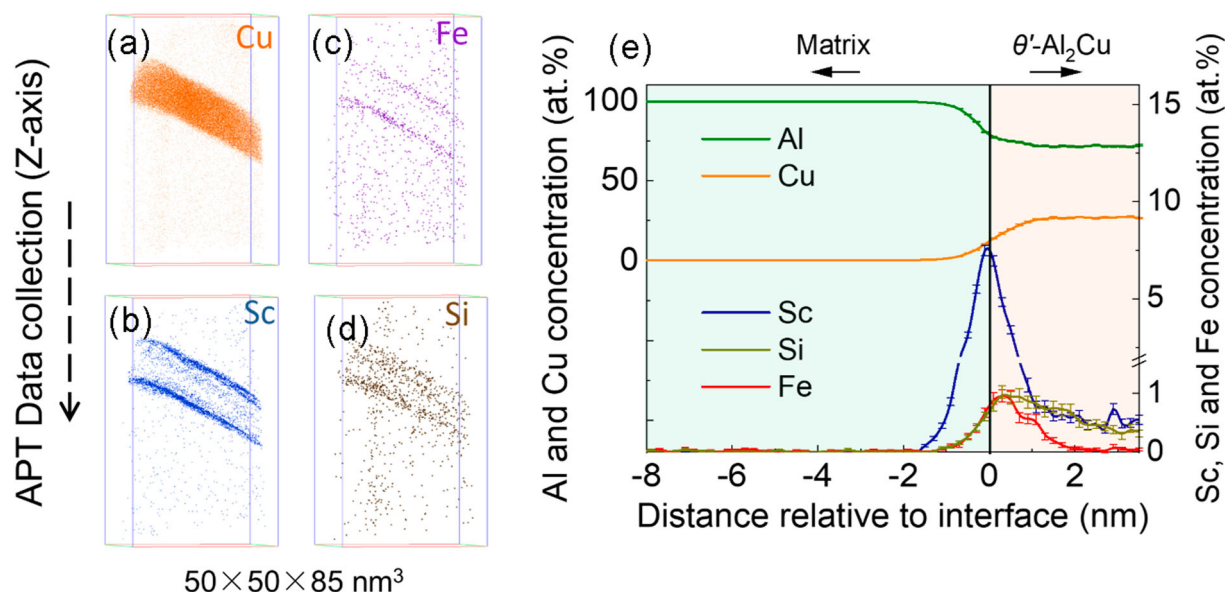


Figure 3. APT results showing a representative θ' -Al₂Cu/matrix interface with multiple segregation and the distributions of (a) Cu, (b) Sc, (c) Fe and (d) Si atoms in Al–Cu–Sc–Fe–Si alloy crept at 300°C for 450 h. The corresponding concentration profiles in (e) clearly reveal the strong Sc and Fe segregation at the interface and the more spread-out Si distribution.

at high temperature, as well-documented in [15]. In our Al–Cu–Sc–Fe–Si alloy, such strategy was also considered, see Figure 4(a–f). It is clear that Sc and Fe co-segregation is also evident at the SCI. Meanwhile, the concentration profiles of Sc and Fe near the SCI are close to those of the CI (Figure 3(a–e)). It is thus concluded that both CIs and the SCIs of θ' -Al₂Cu are also well-protected in our Al–Cu–Sc–Fe–Si alloy during 300°C-creeping. The Si concentration is found to be close to zero in the matrix side within experimental error, as a result of Si enrichment in the θ' -Al₂Cu precipitates. The Si segregation towards the interface is known to play only a minor role in improving θ' -Al₂Cu stability, as the Si distribution tends to spread out spatially around the interface, with low interfacial excess [16]. Its main function is to serve as a facilitating element to attract Fe into the precipitate, as discussed above. In fact, kinetically Si also helps to sweep Fe into θ' -Al₂Cu before creep. The volume-diffusion of Fe alone is sluggish at 300°C [17], insufficient to carry Fe into the precipitate before Sc segregates to the θ' /Al interface. The fast Si diffusivity [17], together with the favorable Si–Fe binding [18], as well as the multiple favorable Si sites in the θ' -Al₂Cu precipitates [16], allows the Si solutes (albeit at a low concentration of $\sim 0.04 \text{ wt.}\%$ in the matrix) as a vehicle to pull Fe along with them into the precipitate interior (Figure S5). This Si strategy was also employed previously to accelerate Sc and Zr aggregation in Al–Sc-based alloys [19]. The Fe collection in θ' -Al₂Cu then sets the stage for its subsequent segregation to the interface during high-temperature creep (see below).

APT shows a clear Sc peak at the interface. But to directly observe the atomic locations in the crystal lattice, we performed HAADF-STEM imaging and EDS mapping. The results are presented in Figure 5(a–i). Sc atoms, with a medium intensity in Z contrast ($Z = 21$) between Al ($Z = 13$) and Cu ($Z = 29$), are greatly enriched in a single layer on the Al-side of the interface. When viewed along both the $\langle 100 \rangle_{\text{Al}}$ (Figure 5(a–i) and Figure S6) and $\langle 110 \rangle_{\text{Al}}$ axes (Figure 5(j–r)), these Sc atoms are seen to locate in a periodic fashion (brighter column with Sc, alternating with a dimmer one without) within the single Al₁ layer. This pattern arises because of the assembly of V-shaped Sc–Al–Fe trimers, which are in fact also observed in our DFT prediction (Figure 1). Unfortunately, the Fe atoms are not distinguishable from Cu in the Z-contrast HAADF-STEM, as they are not sufficiently different in atomic number. However, atomic EDS mapping obtained from two crystal directions indicates that Fe atoms are most likely localized in a single layer next to Sc, due to the strong Sc–Fe binding. These observed interfacial Sc and Fe layers are in full agreement with the DFT prediction based on minimized interfacial energy. Besides, we note that EDS mapping in Figure 5(b–i) does not have the resolution of HAADF-STEM to observe the Al₀ plane seen in Figure 5(a), or Figure 1(a). The Sc content in the Al₁ plane can be estimated by assuming that the alternating brighter atomic columns observed in Figure 5(a) are composed mostly of Sc; its concentration is then approaching 50 at. %. A more reliable estimate can be obtained by integrating the APT-measured concentration profile of Sc. Translating these Sc solutes into

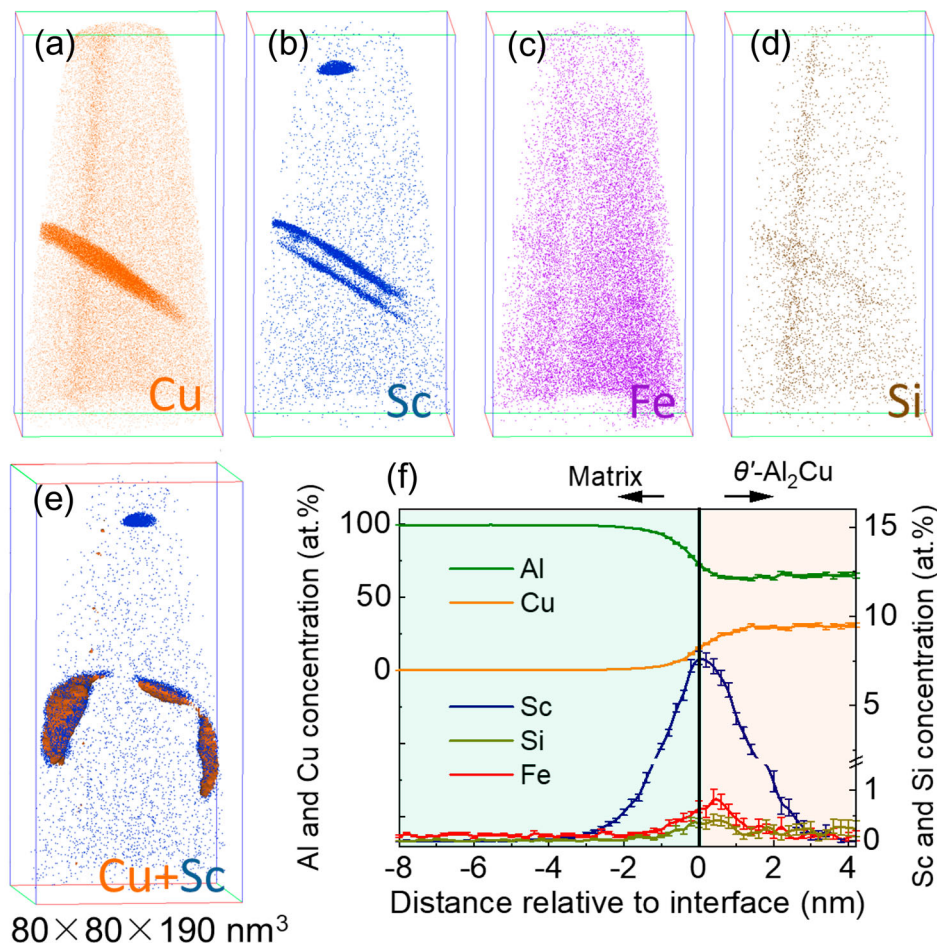


Figure 4. Multiple solute segregation at semi-coherent interface (SCI) of θ' -Al₂Cu precipitate. The representative APT maps were viewed in (a–d) *face-on* and (e) *edge-on* directions showing the elemental distribution of (a) Cu, (b) Sc, (c) Fe and (d) Si at the SCI of a θ' -Al₂Cu plate in Al–Cu–Sc–Fe–Si alloy after creep at 300°C for 450 h. The θ' -Al₂Cu/matrix interface was highlighted by 10 at.% Cu iso-concentration surface in (e). The proxigram in (f) display the Sc and Fe segregation peaks well as Si enrichment in concentration profiles.

single-plane concentration as they actually all reside in the AL₁ plane, the result is ~ 47 at.%, in line with the high Sc percentage estimated from TEM. The same integration method from the APT result in Figure 3(e) gives a Fe concentration of ~ 7 at.% in the θ'_{+1} plane. These experimentally measured planar concentrations of Sc and Fe are actually within the range we covered in modeling, see Table S2.

Based our computational design, this segregation-sandwiched interface is expected to diminish the driving force for coarsening. In terms of kinetics, Sc and Fe diffusion is known to be sluggish compared with Cu [17,19]. The back-to-back Sc–Fe segregation layers at the θ' -Al₂Cu/matrix interface are expected to significantly reduce the interfacial mobility. From both the thermodynamic and kinetic perspectives, we expect highly stabilized θ' -Al₂Cu nanoprecipitates against coarsening. This explains the remarkable performance in Figure 2(f): during the creep tensile test at 300°C, the coarsening is suffocated, and the creep strain rate is reduced by

about two orders of magnitude relative to the single-solute stabilized alloys, not to mention the Al–Cu base alloy.

4. Conclusion

In this paper, we demonstrate a novel approach to engineer the θ' -Al₂Cu/matrix interface to suffocate the θ' -Al₂Cu coarsening at a high homologous temperature of $0.61T_m \approx 300^\circ\text{C}$ (where T_m is the melting temperature of the Al matrix). This strategy is derived from a synergy of computational and experimental design, aiming at assembling a novel interfacial architecture with the appropriate atomic configurations contributed by multiple solutes (Sc, Fe and Si). Specifically, Sc and Fe were identified by the DFT calculations to simultaneously segregate to the θ' -Al₂Cu/Al interface, and their strong binding forms a sandwiched structure reinforcing the interface. This interfacial architecture was experimentally realized through an assembly of V-shaped

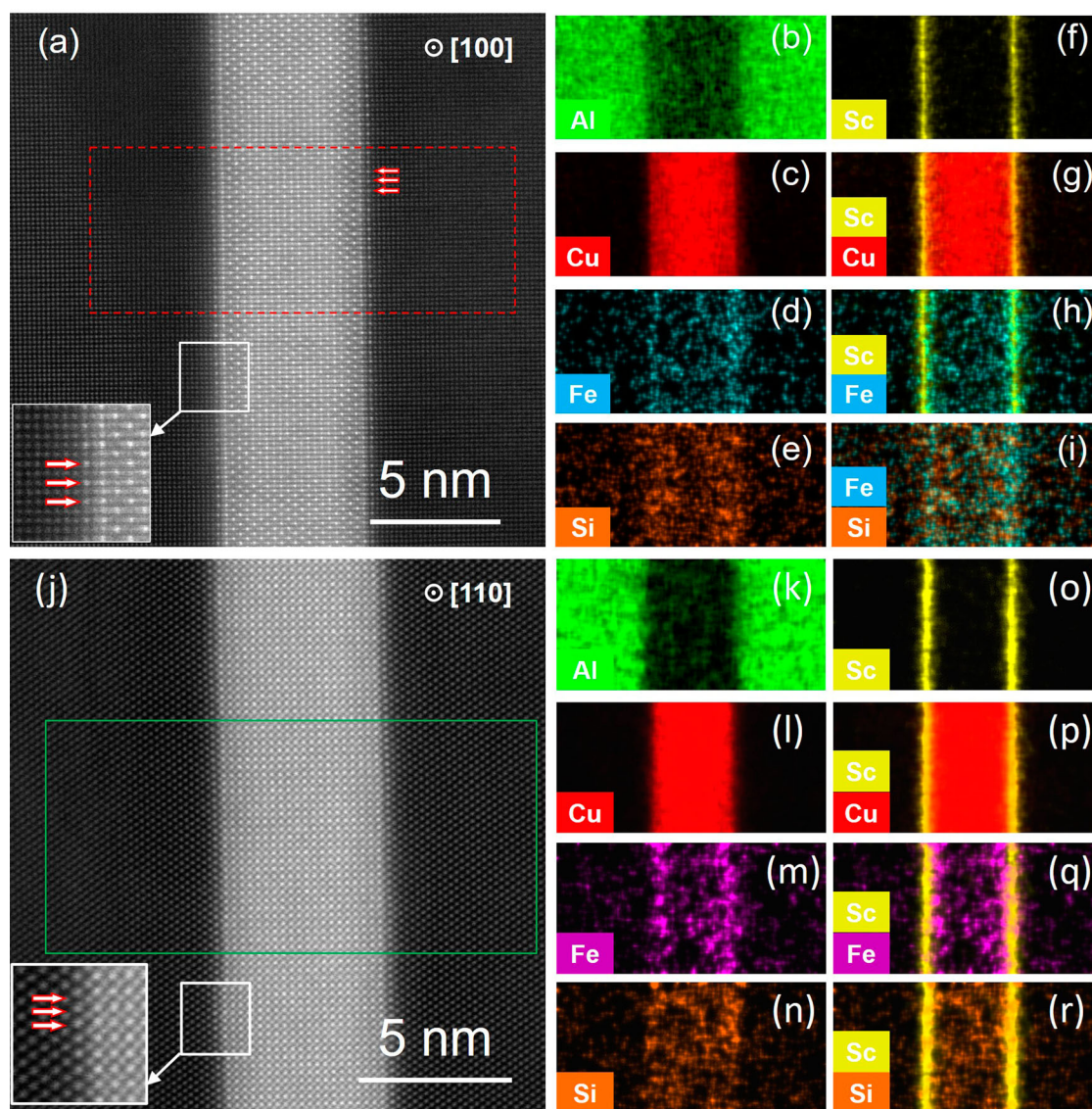


Figure 5. Atomic-scaled details at θ' -Al₂Cu/matrix interface after Sc-Fe-Si microalloying viewed along (a–i) $\langle 100 \rangle_\alpha$ and (j–r) $\langle 110 \rangle_\alpha$ axes, respectively. (a and j) HAADF-STEM images of a θ' -Al₂Cu precipitate in the Al–Cu–Sc–Fe–Si alloy after creep test at 300°C/30 MPa for 450 h. The Sc segregation in a single atomic layer Al₋₁ can be clearly observed at the interface, as highlighted by red arrows (also see the enlarged view in the left bottom inset). (b–i) and (k–r) are EDS maps corresponding to the red dash squared region in (a) and (j), showing atomic columns rich in (b and k) Al, (c and l) Cu, (d and m) Fe, (e and n) Si, as well as (f and o) Sc, respectively. The maps superimposing pairs of elements, for (g and p) Sc and Cu, (h and q) Sc and Fe, as well as (i and r) Fe and Si, respectively, clearly reveal the relative locations of these segregates.

Sc–Al–Fe trimer at the θ' -Al₂Cu/matrix interface in a model Al–Cu alloy with minor Sc, Fe and Si additions. Owing to the back-to-back layered, thermodynamically stable and kinetically robust interface, the coarsening of θ' -Al₂Cu nanoprecipitate is nearly suffocated at 300°C, enabling a two orders-of-magnitude reduction in creep rate in comparison with the counterparts with single-solute segregation. These results are expected to open new opportunities for remarkable property advances in high-temperature resistant engineering alloys.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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