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# A fabrication strategy of nanotwinned metals with superior manufacturability and weldability for electronic packaging

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The emerging embodied intelligence and high-performance computing industries have prompted the demand for high-density integration, small feature sizes, and highly reliable interconnects in future advanced packaging technologies. Solid-state bonding exhibits excellent capacity for exceptional ultrafine-pitch interconnects, remarkable mechanical performance, and high reliability, being regarded as a promising technology for high-density electronic interconnects. Nanotwinned (NT) metals further enhance this potential due to their low-temperature weldability, excellent mechanical properties, and outstanding thermal and electromigration stability. They are applied as bonding, redistribution layer (RDL), and metallization materials, where their fabrication, bonding behavior, and joint reliability are key research focuses.

The controllable fabrication of NT-metals through electronics-manufacturing-compatible processes, such as electroplating and physical vapor deposition, is fundamental for their application in electronic interconnects. These methods enable the formation of NT-metals (e.g., NT-Cu and NT-Ag) characterized by high-density coherent twin boundaries (CTBs). Nanotwinned structures facilitate atomic diffusion at interfaces, thereby greatly improving weldability and allowing low-temperature bonding. Moreover, these CTBs remarkably enhance the microstructural stability and reliability of NT-metals under harsh electro-thermal service conditions. For instance, NT-Cu maintains structural integrity up to 250–300°C, resisting grain coarsening and detwinning that commonly occur in traditional nanocrystalline Cu. Additionally, NT-Cu demonstrates superior electromigration resistance; under a high current density ($3.6\times10^4$ A/cm$^2$), the presence of CTBs oriented perpendicular to the current direction significantly reduces void propagation and electrical resistance increase compared to conventional polycrystalline Cu.[1] Thus, the excel-

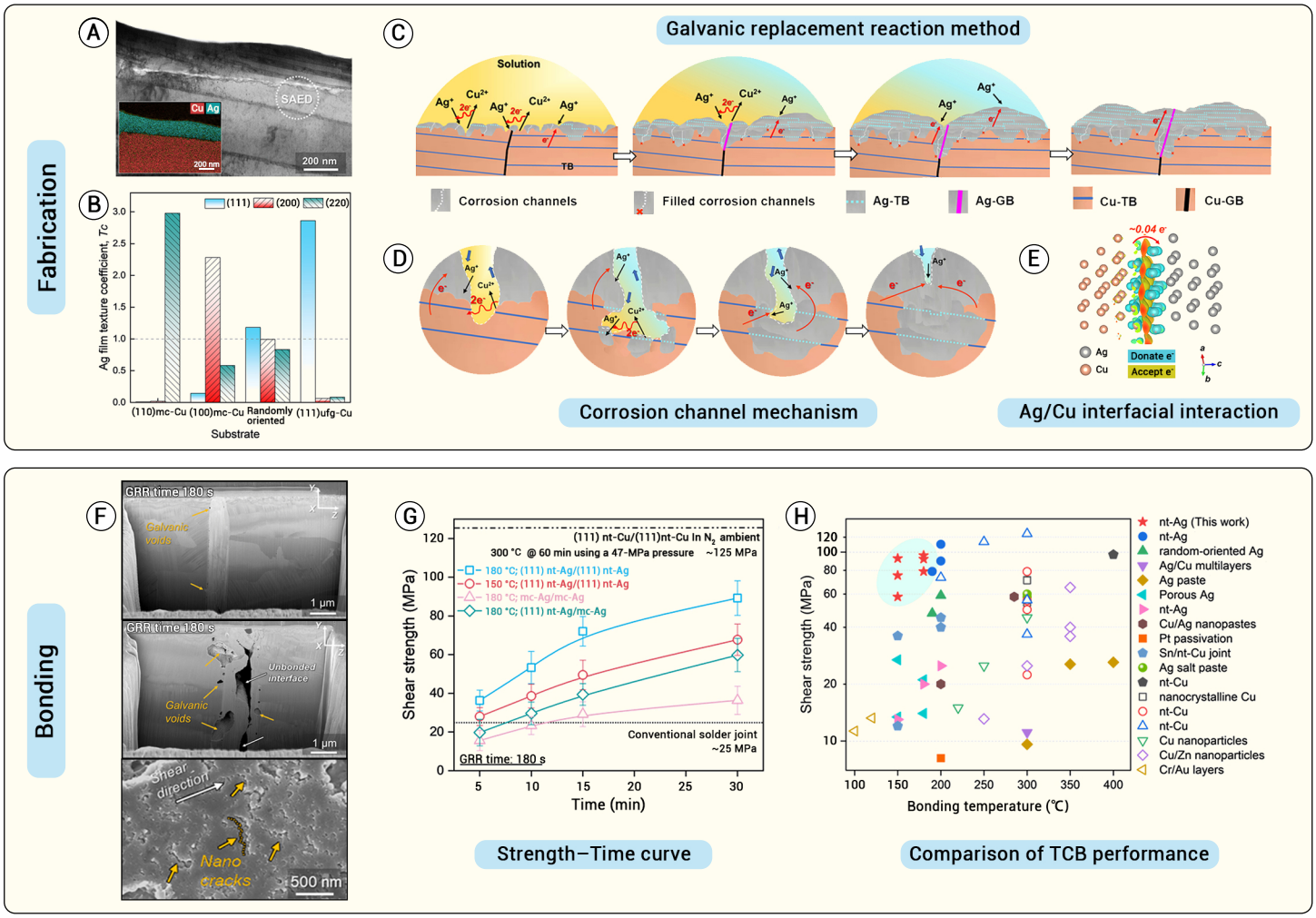


**Figure 1. Fabrication of NT-Ag via galvanic replacement reaction and its Bonding performance** (A) Nt-Ag/Cu interface (B) Crystallographic orientation relationship between the substrate and the Ag film (C) Schematic of in situ epitaxial thickening of (111) nt-Ag on polyerystalline (111) nt-Cu substrates (D) Schematic of a corrosion channel mechanism (E) Bader electron transfer at the Ag(111)/Cu(111) interface (F) The cross-sectional FIB-SEM images of the bonded and unbonded regions, showing zigzag nanocracks on the fracture surface (G) Shear strength-bonding time curves with a GRR time of 180 s (H) Comparison of thermo-compression bonding performance.

lent weldability and reliability of NT-metals are intrinsically derived from their unique nanotwinned microstructures, highlighting that precise and controllable fabrication is indispensable and foundational to practical applications.

Specifically, NT-Ag exhibits exceptional mechanical strength, electrical conductivity, and weldability as an interconnect material in electronic packaging. Ke et al. successfully fabricated NT-Ag films via magnetron sputtering, achieving high-density nanotwin lamellae oriented perpendicular to the growth direction.[2] These films demonstrated impressive electrical conductivity up to 55.6 MS/m (approximately 95.8% of the International Annealed Copper Standard, IACS) and a high hardness of approximately 1.95 GPa. In addition to the high surface diffusivity enabled by the characteristic {111} fiber texture, NT-Ag also displays enhanced oxidation resistance. However, the current fabrication of NT-Ag primarily depends on sputtering techniques, which are costly and relatively inefficient, thereby hindering its scalability for industrial applications.

NT-Cu can be fabricated through a more cost-effective and efficient electroplating method. However, although NT-Cu exhibits a lower oxidation rate compared to conventional Cu, it still retains the inherently high oxidation tendency of Cu, which negatively affects its weldability. The minimum required bonding temperature remains high at approximately 250°C, and the bonding process requires an extended duration of about 90 minutes under a high-vacuum environment of $1 \times 10^{-3}$ torr.[3] Moreover, the resulting bonding strength is limited to approximately 46.1 MPa, insufficient to meet the stringent mechanical strength requirements for solder joints in electronic devices. Overall, the inherent trade-off between manufacturability and weldability of NT-metals significantly constrains their broader application in electronic packaging.

## GRR-BASED FABRICATION OF NT-METALS: MECHANISM AND PERFORMANCE

To address the inherent trade-off between weldability and manufacturability, Zhan and Zhu et al. developed a novel galvanic replacement reaction (GRR) method enabling the in-situ epitaxial growth of wafer-scale, highly (111)-oriented NT-Ag films directly onto tailored polycrystalline (111) NT-Cu substrates (Figure 1A).[4] This approach synergistically integrates the advantages of NT-Cu and NT-Ag. Highly (111)-oriented NT-Cu was prepared by pulse reverse current electrodeposition from an electrolyte containing 200 g/L $CuSO_4 \cdot 5H_2O$, 70 g/L $H_2SO_4$ (98 wt%), and collagen as the surfactant, at 25°C. The current density was controlled between 1 and 12 A/dm$^2$. Subsequently, NT-Ag films were deposited onto the (111) NT-Cu by chemical immersion in a commercial silvering solution containing 0.9 g/L $Ag^+$. The innovative GRR method effectively overcomes conventional fabrication challenges associated with NT-Ag, such as high cost, process complexity, inefficiency, and dependence on single-crystal substrates. The resulting NT-Ag films accurately replicated the substrate's crystallographic orientation, exceptional thermal stability, and improved resistance to oxidation—critical attributes for achieving low-temperature direct metal bonding under ambient conditions.

The mechanism of NT-Ag formation via GRR is elucidated in two distinct stages: A heteroepitaxial growth stage followed by a homoepitaxial growth stage (Figure 1C). By employing an electrochemical Ag-deplating technique, the heteroepitaxial growth stage was experimentally demonstrated. Initially, driven by the potential difference, silver ions substituted copper atoms, forming a serrated Ag/Cu interface predominantly characterized by cube-to-cube orientation relationships. Analysis of the interface energy of this serrated Ag/Cu interface revealed that the 8 × 9 coincident site lattice (CSL) boundary possessed superior thermodynamic stability. Consequently, despite a substantial lattice mismatch of 11.6%, the Ag film preferentially replicated the twin structures of the underlying Cu substrate, with the fraction of (111)-oriented grains exceeding 91.9%. This theory was further validated through comparative experiments using Cu (100) and Cu (110) oriented substrates (Figure 1B). Additionally, a corrosion channel mechanism was identified during this stage (Figure 1D). Silver ions continued to deposit through these corrosion channels while copper ions departed the surface through the same pathways, resulting in prolonged reaction times and thicker Ag deposits. When the corrosion channels became sealed, homoepitaxial growth became the sole mechanism; however, the experimentally observed corrosion depths were significantly smaller than theoretical predictions. Employing density functional theory (DFT), the previously overlooked interfacial interaction within Ag/Cu was revealed (Figure 1E). Specifically, the redox interaction between the negatively charged interface and incoming Ag ions facilitated the continued incorporation of Ag ions into the crystal lattice. During the subsequent homoepitaxial growth stage, both thermodynamic and kinetic factors played critical roles in promoting nanotwin formation. Thermodynamically, the relatively low stacking fault energy (SFE) of Ag favored nanotwin formation, while kinetically, rapid deposition rates contributed further to this phenomenon. The detailed elucidation of the GRR mechanism provided in this study offers valuable theoretical guidance and practical reference for future preparation of NT-metals.

The weldability of (111)-oriented NT-Ag films fabricated via GRR was demonstrated through thermo-compression bonding (TCB) at significantly reduced temperatures (150-180°C). Notably, successful bonding was achieved in ambient air at a temperature as low as 150°C, which represents the lowest reported TCB temperature for NT-metal in air to date (Figures 1G-H). Under bonding conditions of 180°C for 30 min, an optimal shear strength of 86.2 MPa was obtained, significantly exceeding that of conventional solder joints. Remarkably, even when the GRR time was shortened to 45 s—resulting in an NT-Ag film thickness of only ~65 nm—the bonded joints still exhibited excellent shear strength. However, no ductile dimples were observed on the fracture surface; instead, many zigzag nanocracks were formed (Figure 1F). A thinner Ag film is more likely to generate micro-damage, which facilitates crack deflection during propagation and leads to the formation of zigzag nanocracks. This increases the crack surface area and promotes stress relief, thereby improving the fracture toughness of Ag-Ag joints.

## CHALLENGES, LIMITATIONS, AND FUTURE PROSPECTS

It is also important to recognize the limitations of the GRR process and explore possibilities for further improvement. On one hand, the deposition rate of chemical Ag plating is relatively low, and the driving force for nanotwin formation is limited, which affects the structural uniformity of NT-Ag films. Introducing electrochemical deposition with appropriate overpotential could further enhance the deposition rate and film quality. On the other hand, the two-step approach involving electroplated NT-Cu followed by GRR NT-Ag increases process complexity, cost, time, and uncertainty in industrial applications. Meanwhile, the presence of the Ag/Cu interface can lead to severe localized galvanic corrosion, negatively impacting joint reliability. Surface roughness or contamination on the Cu substrate can hinder the timely deposition of Ag in these regions, leading to the formation of voids and the entrapment of solution. During the GRR process, these areas are continuously corroded, and galvanic coupling between Ag and Cu can further sustain corrosion even after the GRR process is completed, ultimately resulting in the formation of galvanic voids. The microalloying strategy for NT-Ag-Cu alloys provides a promising solution: it combines the complementary advantages of Ag and Cu, eliminates the Ag/Cu interface, and enables one-step fabrication. If such Ag-Cu alloys can be prepared by electrodeposition, this would further reduce costs and enhance process feasibility.

In summary, the GRR process demonstrates the possibility of harnessing the complementary advantages of NT-Ag and NT-Cu, providing new ideas and directions for the development of advanced bonding materials, and thus warrants further investigation and optimization in future research.

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## DECLARATION OF INTERESTS

The authors declare no competing interests.