



Vacuum-assisted void-free copper filling of high-aspect-ratio through-glass vias for advanced interposers

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ABSTRACT

Void-free copper filling in high-aspect-ratio (AR) through-glass vias (TGVs) remains a critical challenge owing to void formation within geometrically confined structures. In this study, the effects of different pretreatment processes, such as sputtering, electroless plating, and vacuum-assisted wetting, on via filling behavior were systematically investigated. Electrochemical analyses employing cyclic voltammetry stripping (CVS) and linear sweep voltammetry (LSV) revealed that electrolytes containing additives exhibit mass transport-dependent adsorption behavior, which governs spatially dependent deposition behavior in high-AR vias. Additionally, contact angle measurements indicated that metal-coated surfaces and high-AR via structures exhibit reduced wettability compared to bare glass, thereby further limiting electrolyte infiltration. To overcome these limitations, a vacuum-assisted wetting process was introduced. Under reduced pressure, the pressure difference at the gas–liquid interface increases significantly, promoting bubble expansion and facilitating the removal of trapped gas from the via interior. As a result, uniform and void-free copper filling was achieved across a wide range of applied current densities in structures with AR values up to 10. Moreover, no defects such as voids or seams were observed after thermal shock testing (−55 °C to 125 °C, 100 cycles), thereby confirming the structural reliability of the fabricated vias. These results show that vacuum-assisted wetting is an effective pretreatment strategy for improving electrolyte infiltration and enabling consistent and reliable void-free filling in high-AR TGVs, offering a practical approach for advanced glass interposer applications.

1. Introduction

With the rapidly increasing demand for high-density, large-area, and high-performance semiconductor packaging technologies, glass substrates have emerged as a highly promising next-generation platform capable of overcoming the limitations associated with conventional organic substrates and silicon interposers [1,2]. Glass substrates exhibit several advantageous properties, including low dielectric loss [3], superior insulation characteristics [4], excellent surface flatness [5], and a relatively low coefficient of thermal expansion (CTE) [6], while their compatibility with large-area panel-level manufacturing processes enables substantial cost benefits [7]. In addition, glass substrates have attracted attention as platforms for various functional materials with notable electrical and magnetic properties [8–12]. To facilitate the

practical deployment of glass substrates, through-glass vias (TGVs) are essential for achieving vertical electrical interconnection. Although TGVs are structurally similar to through-silicon vias (TSVs), they offer distinct advantages arising from the intrinsic insulating properties of glass, including reduced inter-signal crosstalk and enhanced high-frequency performance [13]. As packaging technologies continue to scale, TGV structures are progressively evolving toward high-aspect-ratio (AR) geometries, thereby enabling higher I/O and increased design flexibility. However, in comparison with low-AR structures, high-AR geometries introduce significant process-related challenges [14]. Restricted electrolyte diffusion within the vias increases the likelihood of defect formation and further constrains the process window. Therefore, achieving stable processing conditions and ensuring the reliability of high-AR TGVs have become critical

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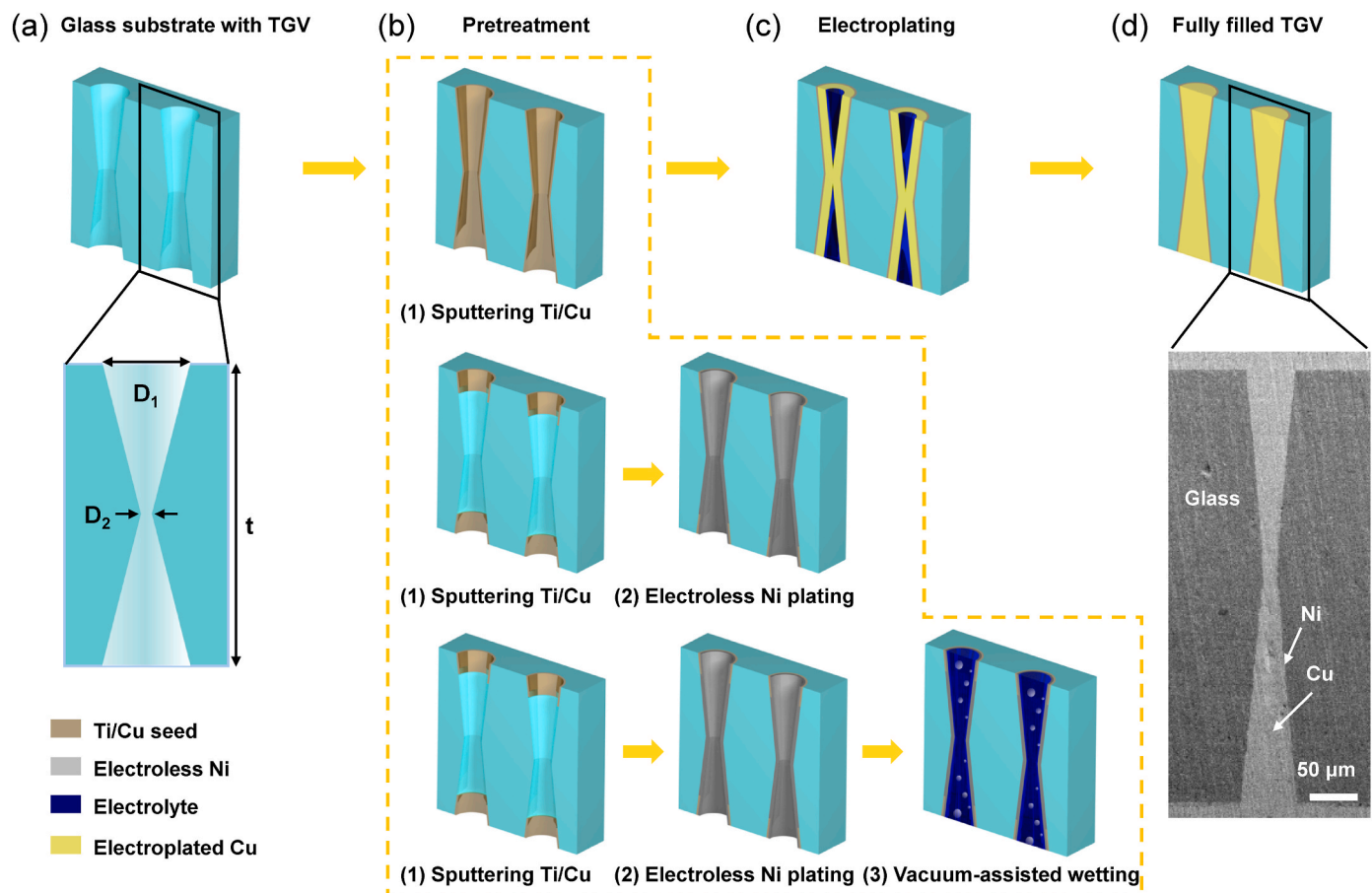


Fig. 1. Fabrication process and cross-sectional images of TGV: (a) substrate preparation with hourglass-shaped via structure; (b) pretreatment steps performed prior to electroplating, including (1) seed layer sputtering, (2) Ni electroless plating, and (3) vacuum-assisted electrolyte wetting; (c) Cu electroplating; and (d) fully filled TGV, as observed by SEM.

challenges.

Among the various fabrication steps, the metal via-filling process is a key determinant governing the overall TGV performance and reliability [15]. The presence of internal voids or seams can lead to increased electrical resistance, accelerated electromigration, Joule heating, and long-term degradation in both the overall mechanical and electrical reliability performance [16]. In high-AR structures, ion diffusion and mass transport limitations further complicate the overall filling behavior [17]. To overcome these limitations, various electroplating strategies have been proposed, including current-density-based process parameter control [18], double-sided electroplating, and optimization of additive chemistry [19,20]. The microstructure of the resulting electrodeposited film is known to be sensitive to electrodeposition conditions and bath composition [21]. In particular, extensive research has been conducted on the use of functional additives, such as accelerators, suppressors, and levelers, to regulate deposition behavior and facilitate void-free filling [7]. Additionally, process optimization studies have investigated the influence of parameters including current density, additive concentration [22], and electrolyte agitation conditions. However, most prior studies have predominantly focused on the electroplating process itself [23], while systematic investigations of pretreatment processes, including seed-layer formation parameters, surface activation to enhance wettability, and pre-wetting procedures, remain relatively limited.

Pretreatment processes play a critical role in enabling uniform and consistent electrolyte infiltration into the via and stabilizing early-stage nucleation behavior. Non-uniformity introduced at this stage can result in localized growth non-uniformity during subsequent plating,

ultimately leading to the formation of defects such as seams and voids [24–26]. In high-AR TGVs, the open-ended through-via structure configuration is particularly susceptible to pinch-off at the via entrance and subsequent central void formation during conformal growth. Moreover, incomplete filling caused by insufficient electrolyte penetration has also been frequently reported [27,28]. According to previous studies, in TSVs, vacuum-assisted or multi-step pre-wetting has been employed as an effective pretreatment for removing trapped air and improving electrolyte penetration into the via interior, enabling void-free filling even in high-AR or nanoscale structures [24,29,30]. For glass substrates, related pretreatment efforts have mainly focused on forming continuous electroless seed layers in TGVs [31]. In contrast, the use of vacuum-assisted wetting in open-ended, high-AR hourglass-shaped TGVs has remained relatively underexplored, particularly in combination with electroless seed-layer formation. Therefore, to achieve reliable via filling in high-AR TGV structures, the influence of pretreatment processes on filling behavior must be systematically investigated, in addition to the optimization of plating conditions.

In this study, we examined the via-filling characteristics of high-AR TGVs as a function of pretreatment conditions to minimize defect formation. Void formation behavior was evaluated across multiple AR values, and the effects of process parameters at each stage were systematically examined. Based on these results, optimized processing conditions for seed-layer formation, vacuum-assisted processes, and electroplating were established. As a result, we demonstrate that void-free filling can be successfully achieved in high-AR TGVs with an AR of 10:1.

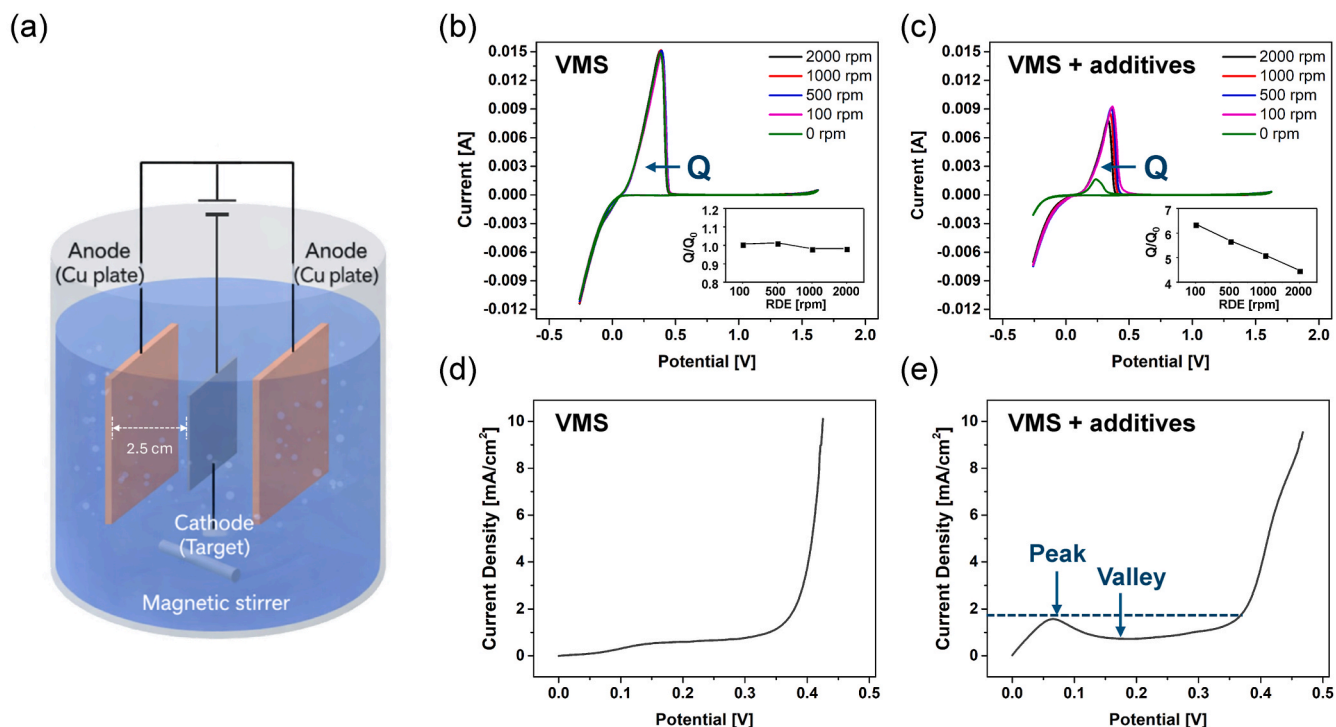


Fig. 2. Double-sided electroplating setup and electrochemical analysis using CVS and LSV. (a) Double-sided electroplating setup with two Cu plates as anodes and one target sample as the cathode, positioned centrally between the anodes. CVS results of (b) VMS and (c) electrolyte with additives. LSV results of (d) VMS and (e) the electrolyte containing additives.

2. Methods

2.1. Sample preparation

Fig. 1 presents the geometry of the hourglass-shaped via and the overall via-filling procedure. The TGV structure is defined in terms of the via entrance diameter (D_1), the via center diameter (D_2), and the substrate thickness (t) (Fig. 1(a)). TGV structures are generally designed as through-via geometries that remain open at both ends. In this study, glass substrates with hourglass-shaped vias, exhibiting ARs of 4, 6, 8, and 10, were prepared and fabricated, as shown in Fig. S1. The via holes were fabricated by laser modification of the glass, followed by an etching process [32,33]. During the etching step, a reduction in the overall substrate thickness may occur and vary [34], leading to variations in the resulting substrate thickness depending on the AR. The pitch of the vias was consistently maintained at approximately 200 μm for all samples. The entrance diameter (D_1) was varied within the range of 40–100 μm , and the ratio between D_1 and D_2 was controlled to be approximately 2:1. Detailed geometric information for the TGV structures is presented in Fig. S1 and Table S1.

The TGV via-filling process involves three main steps: (1) substrate preparation, (2) pretreatment, and (3) electroplating. For substrate preparation, the substrates were thoroughly cleaned using acetone and isopropyl alcohol (IPA). To examine the effect of pretreatment conditions on via-filling behavior, three different processes were employed, as illustrated in Fig. 1(b). The first approach consists of performing electroplating onto a sputtered seed layer (Fig. 1(b-1)). In the sputtering process, titanium (Ti, 50 nm) and copper (Cu, 300 nm) layers were sequentially and uniformly deposited to form the conductive seed layer (Fig. 1(b-1)). The second approach involves the addition of an electroless plating step subsequent to sputtering (Fig. 1(b-2)). The third approach further includes the incorporation of a vacuum-assisted wetting step following sputtering and electroless plating to effectively eliminate trapped air bubbles inside the vias and significantly improve electrolyte penetration (Fig. 1(b-3)). Finally, via filling was performed

by electroplating the pretreated TGV structures (Fig. 1(c)). The completed TGV structures demonstrated fully filled vias, as confirmed by the scanning electron microscopy (SEM) image presented in Fig. 1(d).

2.2. Characterization

For cross-sectional analysis after electroplating, the samples were mechanically sectioned and subsequently polished using a polishing system (NA-P2001N, Nanotech, Korea). Polishing was performed in a stepwise manner using SiC abrasive papers (#2000 and #3000) at a controlled low rotational speed of about 100–200 rpm. The cross-sectional surfaces were examined using optical microscopy (OM; BX53M, Olympus, Japan) and SEM (Quattro ESEM, Thermo Scientific) to systematically evaluate the via-filling behavior. Additionally, energy-dispersive X-ray spectroscopy (EDS) analysis was conducted to examine the distribution of the seed layer inside the vias and verify its conformal coverage along the via sidewalls.

Contact angle (CA) measurements were carried out using a contact angle analyzer (Phoenix-MT(A), Surface Electro Optics, Korea) integrated with image analysis software (Image Pro 300). Measurements were performed on multiple surfaces, including glass, a sputtered Cu seed layer, and an electroless Ni surface. Deionized (DI) water, an additive-free electrolyte (VMS), and a commercial electrolyte containing additives (Cu520) were utilized as test solutions. Droplets with a volume of approximately 4 μL were consistently dispensed for all measurements. Images were immediately acquired after droplet deposition on the surface, and measurements were conducted for each liquid across three or more different locations. The obtained data were analyzed and reported as mean values with standard deviations to ensure the reliability of the measurements.

The electrochemical characteristics of the electrolyte were analyzed using a 894 Professional CVS (Metrohm) instrument operated with Viva software. A three-electrode system was utilized, consisting of a Pt rotating disk electrode (RDE) as the working electrode, an Ag/AgCl electrode as the reference electrode, and a Pt electrode serving as the

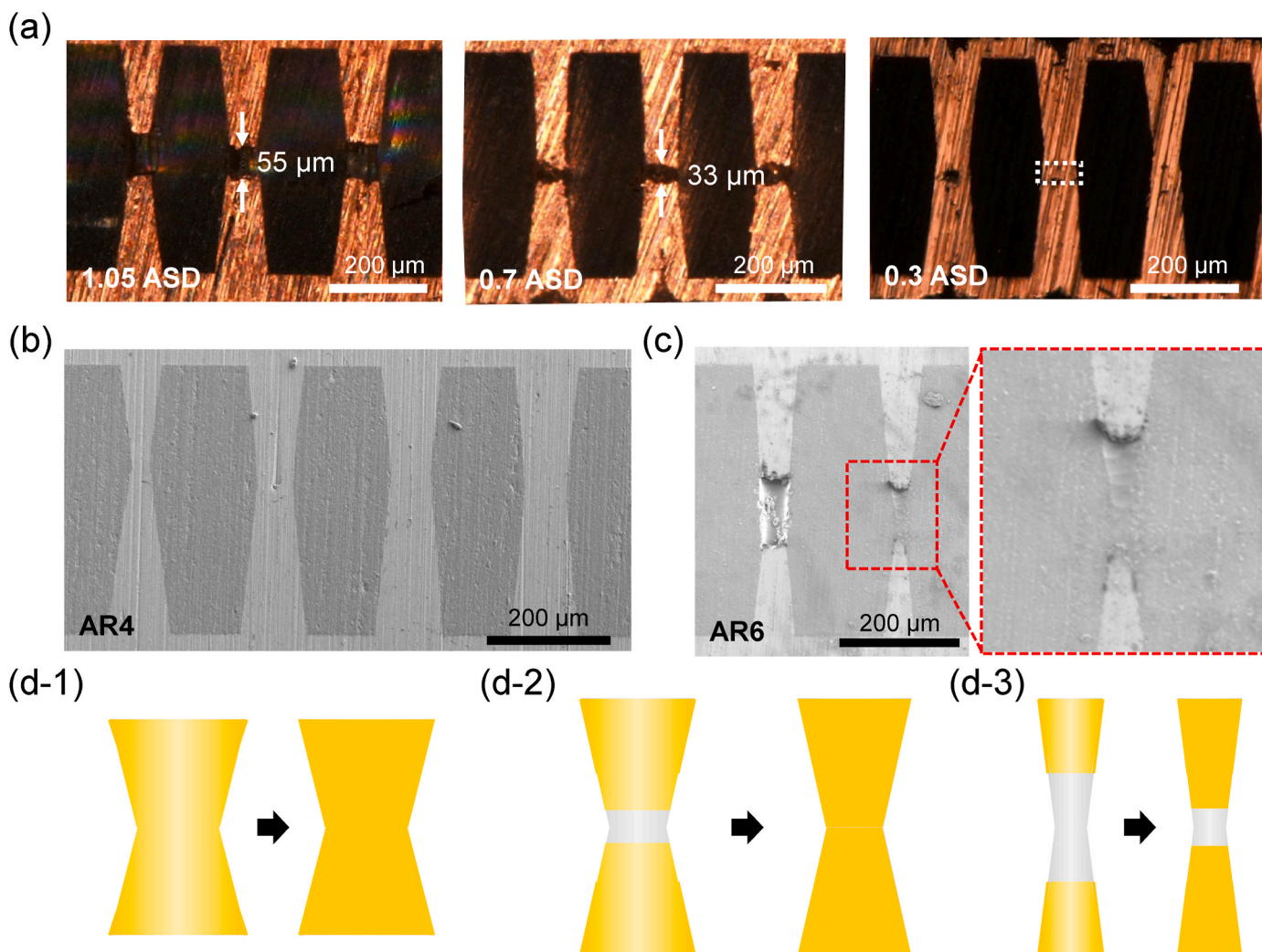


Fig. 3. Cross-sectional images of Cu-filling behavior using only a sputtered seed layer: (a) via-filling behavior of AR 4; (b) void-free filling under the conditions of Ti 50 nm/Cu 300 nm, 0.7 A/dm²; (c) void formation at a higher AR (AR 6) owing to insufficient seed layer coverage and schematic illustrations of via-filling behavior depending on via AR and sidewall coverage; (d-1) low-AR via with fully deposited seed layer coverage, (d-2) low-AR via with partially deposited seed layer, and (d-3) high-AR via with insufficient seed layer coverage resulting in void formation.

counter electrode. For cyclic voltammetry stripping (CVS) analysis, measurements were conducted at a controlled scan rate of 0.1 V/s over a potential range of 1.625 to −0.1749 V at rotation speeds varying between 0 and 2000 rpm. Linear sweep voltammetry (LSV) was performed under a slow scan rate of 0.001 V/s over a potential range from 0.5 to 0 V using an RDE rotating at 100 rpm. A commercial electrolyte (MICROFAB Cu520, Electroplating Engineers of Japan, Ltd.) was employed for the Cu electroplating process. The base electrolyte (VMS) contained CuSO₄ (50 g/L) and H₂SO₄ (25 g/L). Three types of additives, including accelerator, suppressor, and leveler, were selectively introduced into VMS.

3. Results and discussion

3.1. Electroplating process

Prior to analyzing the effect of pretreatment on the occurrence of void formation during via filling, an electroplating configuration suitable for high-AR TGV structures was carefully established. For the electroplating process, double-sided plating was implemented using the setup illustrated in Fig. 2(a). Copper plates (5 cm × 5 cm) were positioned on both sides of the sample as anodes at a distance of 2.5 cm, and the sample was centrally located between the two anodes to function as the cathode. The current was applied using a programmable linear DC

power supply (TL305P, Toyotech), and the current density was precisely controlled within the range of 0.3–1.05 A/dm² (ASD). The electroplating bath temperature was maintained at a constant value of 28 °C, and the electrolyte was continuously agitated using a magnetic stirrer at 300 rpm.

The applied current and plating time were determined based on a constant current density with respect to the plating open area. The deposition time was calculated based on Faraday's law:

$$m = \eta_f \frac{M}{nF} It, \quad (1)$$

where m denotes the total deposition mass; η_f represents the Faradaic efficiency; M and n represent the molar mass of Cu and the valence number of the Cu²⁺ ions involved in the reduction reaction ($n = 2$); and I and t denote the applied current and deposition time, respectively. The electrodeposition thickness was determined based on the calculated total deposition mass as follows:

$$d = \frac{m}{\rho A} = \frac{\eta_f M}{nF \rho A} It, \quad (2)$$

where d denotes the average thickness of the deposited Cu, ρ denotes the density of Cu, and A represents the deposition area. Under a constant

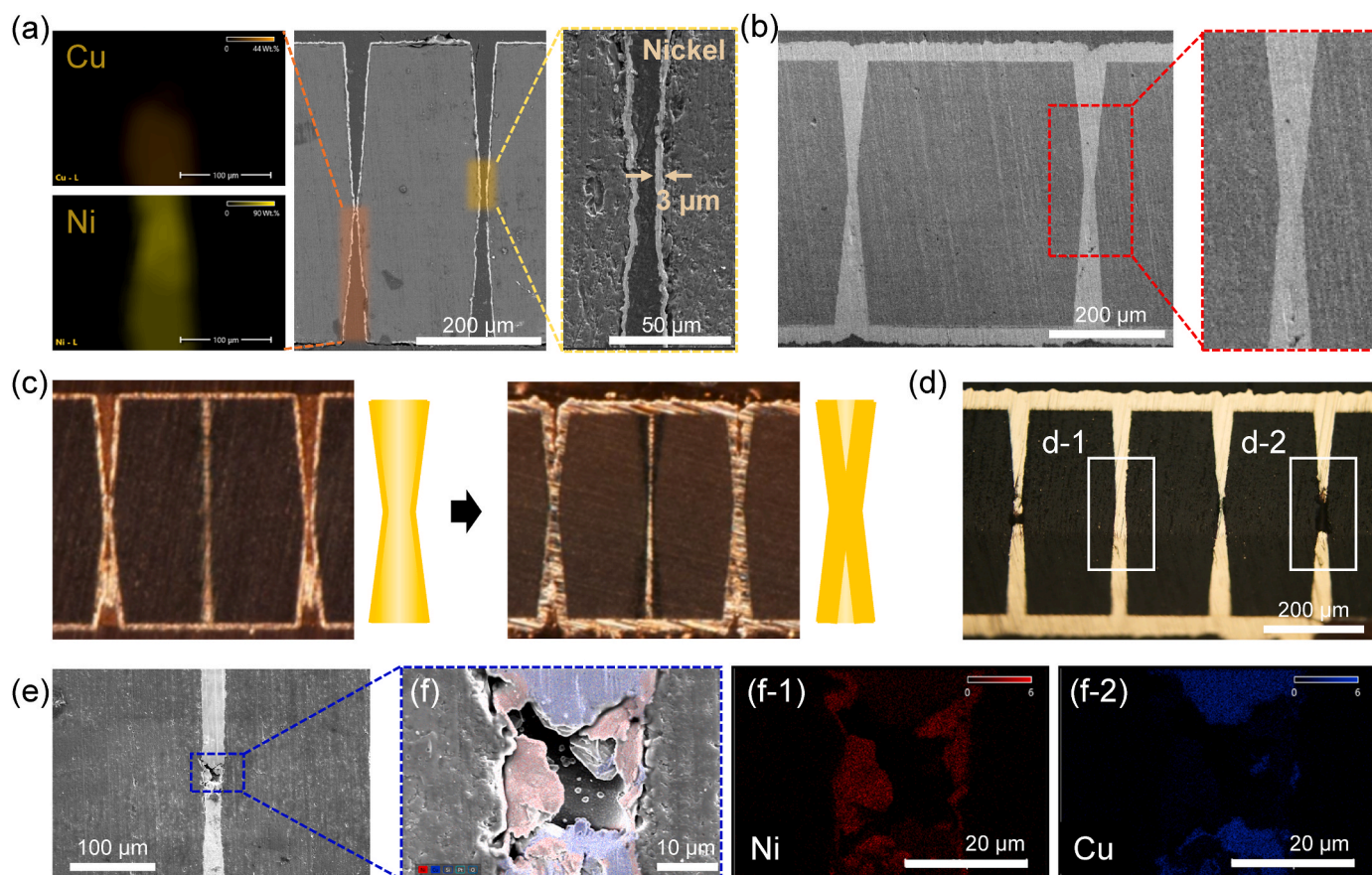


Fig. 4. Cross-sectional analysis with an electroless-assisted seed layer formation process: (a) EDS results showing insufficient Cu seed layer coverage and SEM images showing conformally deposited Ni seed layer, (b) void-free Cu filling under optimized electroplating conditions (0.5 A/dm^2), (c) OM images and schematic illustrations of via filling during electroplating, and (d) AR 8 vias showing both complete filling and void formation. (e) SEM image of the void interior, and (f) corresponding EDS elemental maps of (f-1) Ni and (f-2) Cu.

current density ($j = I/A$), the applied current (I) varies proportionally with respect to the open area (A).

The open area was calculated by considering the hourglass-shaped via geometry. The electrodeposition thickness inside the vias was estimated based on Eq. (2). However, the actual thickness may locally vary owing to non-uniformities in the current distribution. In particular, the presence of additives can substantially influence the deposition rate depending on electrolyte flow and via geometry, owing to their adsorption behavior.

To investigate the influence of additives during the electroplating process, electrochemical analyses were systematically carried out using CVS and LSV by directly comparing electrolytes in the presence and absence of additives. The stripping charge area with and without convection was denoted as Q and Q_0 , respectively. As illustrated in Fig. 2(b) and (c), Q , which is directly proportional to the total amount of deposited Cu, was obtained by integrating the anodic current with respect to time at different rotation speeds of the RDE. In the additive-free electrolyte (VMS), the stripping charge exhibited relatively uniform and consistent behavior with increasing rotation speed (Fig. S2(a)). Conversely, when additives were present, the stripping charge varied significantly as a function of RDE rotation speed, suggesting that the deposition behavior is strongly governed by mass transport conditions (Fig. S2(b)). This behavior is primarily attributed to the surface adsorption characteristics of additives on the electrode surface [35]. Under high-flow conditions (i.e., high RDE speeds), the transport of suppressor species can be significantly enhanced, leading to increased surface coverage. Meanwhile, the relative effect of accelerator species can be correspondingly reduced due to competitive adsorption, leading

to a decrease in the overall net deposition rate.

Additionally, LSV analysis revealed distinct variations in the current-potential response behavior in the electrolyte containing additives compared to the VMS, as shown in Fig. 2(d) and (e). While the VMS exhibited relatively smooth and stable polarization curves, the additive-containing electrolyte showed a characteristic peak-valley feature, indicating the presence of complex electrochemical interactions occurring at the electrode surface [36,37]. This phenomenon is closely associated with the potential-dependent and competitive adsorption mechanisms of suppressor and accelerator species at the electrode interface. In confined geometries such as TGV structures, where electrolyte flow inside the via is inherently restricted, this flow-dependent adsorption behavior becomes more pronounced and influential. Consequently, the presence of additives induces spatial variations in deposition behavior, which can ultimately facilitate more uniform filling between the via entrance and interior.

3.2. Seed layer process condition effect

Prior to electroplating, a seed layer was formed as an essential pre-treatment step. As illustrated in Fig. 1(b-1), the via-filling behavior was initially investigated for samples prepared solely using the sputtering process. The seed layer was sequentially deposited with controlled thicknesses of Ti (50 nm) and Cu (300 nm). Electroplating was then conducted under current densities ranging from 0.3 to 1.05 A/dm^2 . As shown in Fig. 3(a), vias with an AR of 4 were filled with copper using only a sputtered seed layer, and the size of the void remaining at the via center varied depending on the applied current density. As seen in Fig. 3

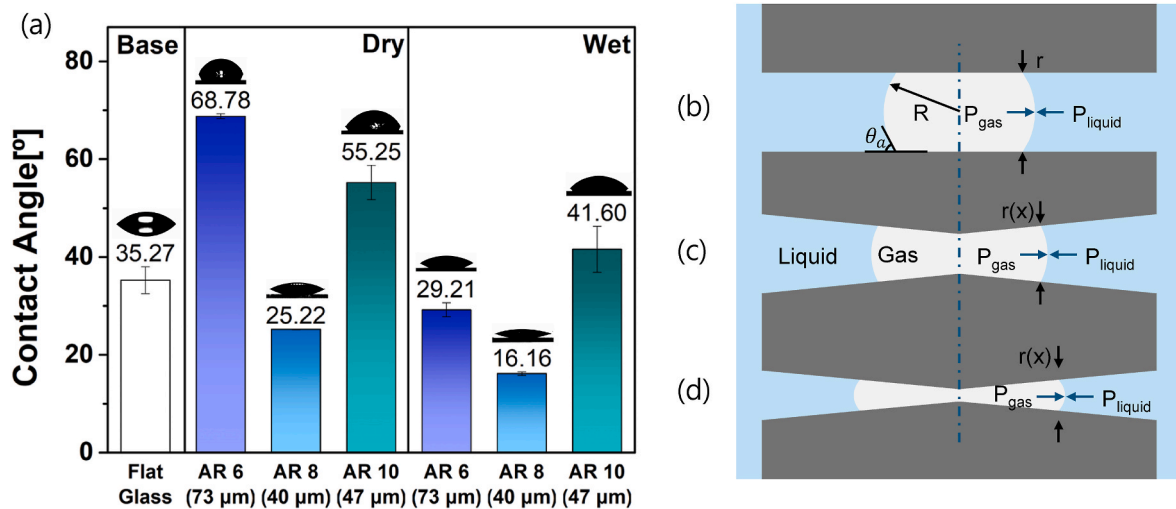


Fig. 5. CA measurements performed on glass substrates with and without via structures: (a) CA measured on a flat glass substrate and TGV substrates with ARs of 6, 8, and 10 under DRY and WET conditions; schematic illustration of TGV structures showing entrapment of bubbles at the center of the vias: (b) rod-shaped, (c) low-tapered structures, and (d) highly tapered structures.

(b), void-free complete filling of AR 4 vias was also obtained even when only sputtering was used for pretreatment. However, under identical current density conditions, the filling results varied considerably from sample to sample, ranging from complete filling to voids of several tens of micrometers, and this variation became increasingly pronounced with increasing AR. In the case of AR 6, as shown in Fig. 3(c), void formation was clearly observed at the central region of the via, and complete filling was difficult to achieve consistently even under lower current density conditions. This behavior is attributed to the intrinsic characteristics of the sputtering process. As sputtering deposits metal in a line-of-sight deposition manner, it is difficult to achieve sufficient conformal seed layer coverage on the via sidewalls. As the AR increases, the via entrance progressively becomes narrower, and the sidewall angle becomes increasingly steeper, further limiting effective sidewall deposition. Consequently, the seed layer coverage is non-uniformly formed from sample to sample, and this variation in coverage leads to the observed variation in filling results. As a result, in low-AR vias, the seed layer can be relatively uniformly deposited along the sidewalls, enabling successful and complete filling (Fig. 3(d-1)). In some cases, even with incomplete seed coverage at the via center, Cu growing from both ends can merge and form a continuous structure (Fig. 3(d-2)). However, in high-AR vias, insufficient seed layer coverage prevents the Cu growing from both ends from merging, resulting in void formation at the center (Fig. 3(d-3)). Therefore, the sputtering process alone cannot reproducibly control the filling results, and for high-AR TGVs, additional pretreatment steps are required to secure conformal seed layer coverage.

To address the limitation of seed layer deficiency at the via center region in high-AR structures, an electroless nickel plating step was applied to achieve a continuous conductive layer inside the vias, and the resulting filling behavior was subsequently evaluated. The detailed procedure of the electroless plating is presented in Fig. S3. Fig. 4(a) shows the SEM and EDS analysis results of the via cross-sectional region after electroless plating. The analysis revealed that the sputtered Cu seed layer was primarily distributed near the via entrance and was not sufficiently extended toward the central region of the via. Conversely, the electroless Ni layer was deposited along both the via entrance and the central region with a thickness of approximately 3 μm along the via. This indicates that the electroless plating provides continuous seed layer formation even in high-AR structures. As illustrated in Fig. 4(b), after forming a continuous seed layer along the via, electroplating at a current density of 0.5 A/dm² resulted in complete filling without void formation at the center. When the seed layer is uniformly distributed along the via

surface, the deposited Cu grows from the sidewalls and progressively closes at the narrowest region of the via center (Fig. 4(c)). However, as illustrated in Fig. 4(d), voids were still detected in certain vias, particularly at the center (Fig. 4(d-2)). To identify the origin of this residual defect, high-magnification SEM and EDS analyses were performed on the interior of a representative void (Fig. 4(e) and (f)). The EDS elemental maps clearly reveal that the electroless Ni layer remains along the wall of the void region (Fig. 4(f-1)), whereas Cu is absent inside the void volume (Fig. 4(f-2)). The absence of Cu in a region where a functional cathode surface is available indicates that the electrolyte physically failed to reach the via center during plating. In other words, even when a continuous seed layer exists, insufficient electrolyte supply due to locally trapped gas that excludes the electrolyte can limit uniform deposition inside the via. Consequently, the influence of process conditions becomes increasingly significant under such mass transport limitations.

3.3. Vacuum-assisted wetting

To evaluate the liquid penetration behavior on glass substrates, the inner-surface wettability as a function of TGV geometry was analyzed. Fig. 5(a) shows the CA measurement results as a function of AR. Considering the structural characteristics of TGVs with a through-via structure, the wettability inside the via, particularly at the bottom surface of the via, can influence the overall liquid infiltration behavior. Therefore, CA measurements were performed under two conditions: a dry condition (DRY), where the via bottom was unfilled, and a wet condition (WET), where liquid was present beneath the substrate. For a flat glass substrate, the CA was measured as 35.27°. Under DRY conditions, the CA values of TGV substrates were 68.78°, 25.22°, and 55.25° for AR 6, AR 8, and AR 10, respectively. These results demonstrate that the CA does not vary monotonically with AR but is influenced by a combined effect of via diameter and structural geometry. Under the DRY condition, the via bottom is unfilled, so the air trapped inside the via remains beneath the droplet, and the measured CA reflects an apparent wetting state in which the droplet rests partly on this trapped air, analogous to a Cassie-Baxter-type state. The AR 6 vias, having the largest entrance diameter ($D_1 = 73 \mu\text{m}$), present the largest opening area associated with the trapped air and thus exhibit the highest CA. Under WET conditions, the CA values decreased for all samples, indicating enhanced wettability. Because liquid is present beneath the substrate, the trapped air can escape downward and the liquid infiltrates the via

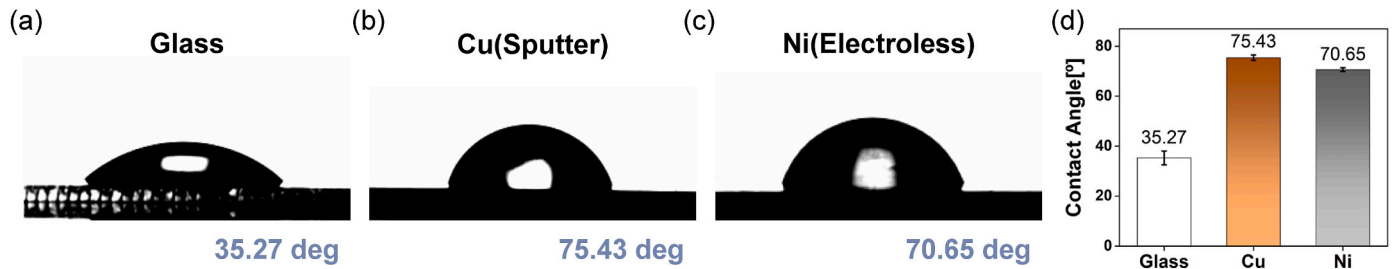


Fig. 6. CA measurements on various flat surfaces: (a) bare glass substrate, (b) Cu-coated glass substrate fabricated by sputtering, (c) Ni-coated glass substrate fabricated via electroless plating, and (d) comparison of average CA values, including standard deviation values. The substrate appearance differs among (a)–(c) owing to the differing surface reflectivity; the CAs were determined identically.

interior, shifting the system toward a Wenzel-type state in which the liquid contacts the via walls. In particular, for AR 6 and AR 8, the CA values were lower than those measured for the flat glass substrate, suggesting enhanced wetting induced by capillary effects. The extent of this reduction depends on the amount of liquid drawn into the via, which is governed primarily by the entrance diameter. The AR 6 vias, having the largest entrance diameter, allow more liquid to penetrate and thus show the most pronounced decrease in CA, becoming lower than that of AR 10 and reversing the trend observed under the DRY condition. Accordingly, the magnitude of the CA reduction from the DRY to the WET condition follows the order $AR\ 6 > AR\ 10 > AR\ 8$, consistent with their entrance diameters ($D_1 = 73, 47, \text{ and } 40\ \mu\text{m}$, respectively). In addition, a gradual decrease in CA over time was observed, suggesting time-dependent wetting behavior (Fig. S4). However, for the high-AR case (AR 10), the change in CA was relatively limited compared to the DRY condition, and the final CA value remained at 41.60° , which is higher than that of the flat glass substrate. This is because the narrow entrance of the AR 10 vias restricts liquid infiltration, limiting the transition toward the Wenzel state. Moreover, no significant time-dependent variation in wetting behavior was observed, suggesting that liquid infiltration is restricted in high-AR structures.

Based on the schematic illustrations in Fig. 5(b)–(d), the mechanism of bubble formation in hourglass-shaped high-AR vias was analyzed. The behavior of air pockets inside the vias is governed by the capillary pressure formed at the liquid–gas interface, which can be described by the Young–Laplace equation [38] as follows:

$$\Delta P = P_{\text{gas}} - P_{\text{liquid}} = \sigma k = \sigma \left(\frac{1}{R_1} + \frac{1}{R_2} \right), \quad (3)$$

where ΔP denotes the pressure difference between the gas and liquid phases; σ represents the interfacial tension; and R_1 and R_2 represent the principal radii of curvature of the interface. Assuming equal curvature ($R_1 = R_2 = R$), the equation can be simplified as follows:

$$\Delta P = \frac{2\sigma \cos \theta}{r}, \quad (4)$$

where θ represents the apparent contact angle, and r is the effective radius of the via (Fig. 5(b)).

$$\frac{d}{dx} (\Delta P) = \frac{-2\sigma \cos \theta}{r^2} \frac{dr}{dx} \quad (5)$$

$$\frac{d}{dx} (P_{\text{gas}} - P_{\text{liquid}}) = -\frac{dP_{\text{liquid}}}{dx} \quad (6)$$

$$\frac{dP_{\text{liquid}}}{dx} = \frac{2\sigma \cos \theta}{r^2} \frac{dr}{dx}. \quad (7)$$

In hourglass-shaped structures, the via radius changes continuously along the axial direction, thereby creating a capillary pressure gradient. Assuming that the gas pressure (P_{gas}) remains approximately constant, the pressure difference is governed primarily by variations in the tube radius (r) as shown in Eq. (7). In regions where the radius decreases ($dr/dx < 0$), the capillary pressure increases, forming a driving force that promotes liquid infiltration toward the narrower region of the via. In hourglass-shaped vias, liquid can enter simultaneously from both ends, which leads to the entrapment of gas at the center of the via (Fig. 5(c)). In such cases, the trapped gas cannot be easily removed owing to the balance between capillary pressure and viscous resistance, leading to the persistence of bubbles [39]. Consequently, in high-AR tapered structures, although capillary pressure increases, the combined effect of limited fluid supply and increased viscous resistance leads to the formation of air pockets at the via center. This phenomenon serves as a major limiting factor for uniform electrolyte infiltration in high-AR vias (Fig. 5(d)).

As shown in Fig. 6, the effect of surface conditions on wettability was systematically analyzed. DI water droplets were placed on flat substrates, including glass, Cu-coated, and Ni-coated surfaces, for comparison. While the CA of the flat glass substrate was 35.27° , the values increased to 75.43° and 70.65° for the Cu- and Ni-coated surfaces, respectively, suggesting reduced wettability on metal-coated substrates. Although clean metal surfaces are intrinsically high-energy and hydrophilic, they readily adsorb airborne hydrocarbons upon exposure to ambient air, which lowers their surface energy and shifts the wetting

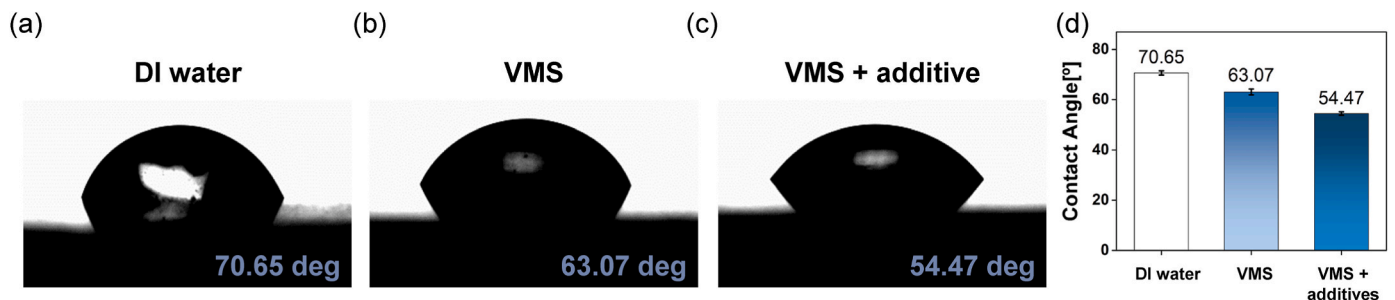


Fig. 7. CA measurements on Ni-coated surfaces: (a) deionized water, (b) additive-free electrolyte (VMS), (c) electrolyte containing additives, and (d) comparison of average CA values with corresponding standard deviations.

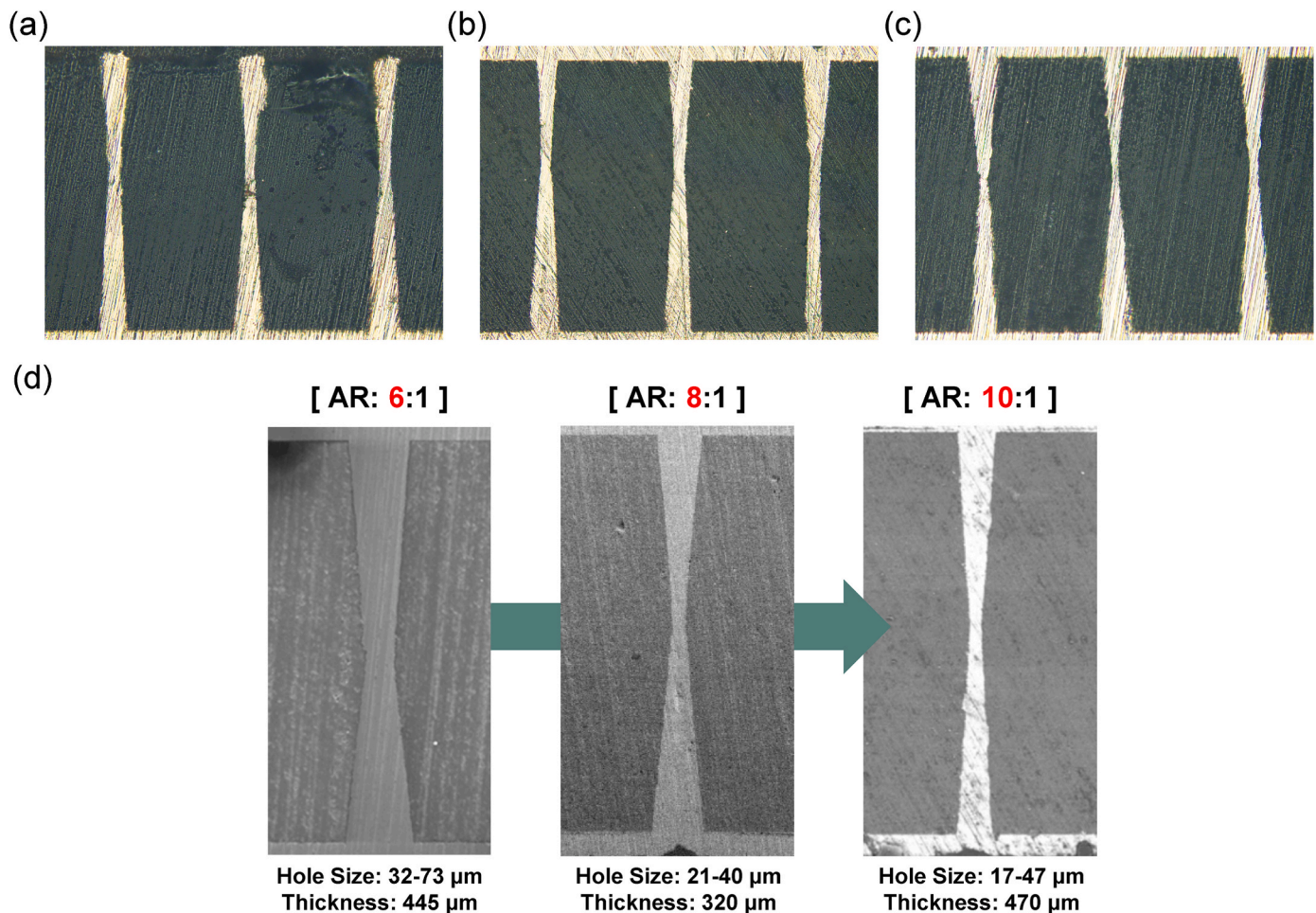


Fig. 8. Cross-sectional images of Cu filling after vacuum-assisted wetting: (a–c) void-free filling at various current densities: (a) 0.3 A/dm², (b) 0.5 A/dm², and (c) 0.7 A/dm²; and (d) void-free filling across a range of aspect ratios (AR 6–10).

behavior toward a more hydrophobic state [40,41]. This adsorption-driven transition accounts for the higher CAs measured on the Cu- and Ni-coated surfaces compared with the bare glass, whose hydrophilic character is largely retained. This reduction in wettability indicates that electrolyte penetration into the vias may be restricted, thereby increasing the probability of bubble entrapment and subsequent void formation. For the analysis of CA on the substrate with vias, Ni-coated TGVs with an AR of 6.6 were evaluated using DI water, as presented in Fig. S5(a). Consistent with the results for flat surfaces (Fig. 6(a) and (c)), the CA on the Ni-coated vias was higher than that measured for the bare glass via substrate (Fig. S5(b)). This indicates that complete wetting of the via sidewalls becomes more difficult following Ni electroless plating.

To address the limitation of liquid penetration into high-AR vias, a vacuum-assisted wetting process was additionally implemented. To select a suitable solution for the vacuum wetting process, the surface wettability of different solutions was evaluated and compared. Fig. 7(a)–(c) present the CA measurements for DI water, VMS, and electrolyte with additives, respectively. Fig. 7(d) presents the average CA values and standard deviations calculated from measurements obtained at three or more distinct locations for each solution, which were 70.65°, 63.07°, and 54.47°, respectively. Based on these results, the electrolyte containing additives exhibited the lowest CA value, indicating the highest level of wettability on the surface.

Based on the CA results, the vacuum-assisted wetting process was carried out using the additive-containing electrolyte. When a vacuum is applied, the liquid pressure (P_{liquid}) decreases, resulting in an increase in

the pressure difference ($\Delta P = P_{\text{gas}} - P_{\text{liquid}}$). Under this condition, the pressure imbalance satisfies the relation expressed as follows:

$$\Delta P = P_{\text{gas}} - P_{\text{liquid}} > \frac{2\sigma}{R}. \quad (8)$$

As a result, the gas bubble expands to restore pressure equilibrium, leading to an increase in the bubble radius (R) [42]. This expansion promotes the detachment and removal of trapped gas bubbles from the via interior.

The vacuum-assisted wetting process was performed as follows. First, a beaker was filled with the additive-containing electrolyte, and the pretreated (sputtered and electroless-Ni-plated) TGV sample was fully immersed in this electrolyte. The beaker containing the immersed sample was then placed inside the vacuum chamber, and the chamber was evacuated to a gauge pressure of -95 ± 5 kPa using a vacuum pump (WK-2S4CSV, Dasheng Zhongheng, China) and a vacuum chamber with a volume of 7.85 L. To promote complete removal of the trapped air, the evacuation was repeated for three cycles, with each cycle consisting of approximately 1 min of reduced-pressure holding followed by a brief return toward atmospheric pressure (total duration ≈ 3 min). After the final cycle, the wetted sample was transferred, while still wet, into a beaker containing the Cu anodes, where the via-filling electroplating was carried out under the conditions described in Section 3.1. The via-filling behavior at various current densities was systematically analyzed following the application of the vacuum-assisted wetting process. Fig. 8(a)–(c) show cross-sectional images of the vias after electroplating at current densities of 0.3, 0.5, and 0.7 A/dm²,

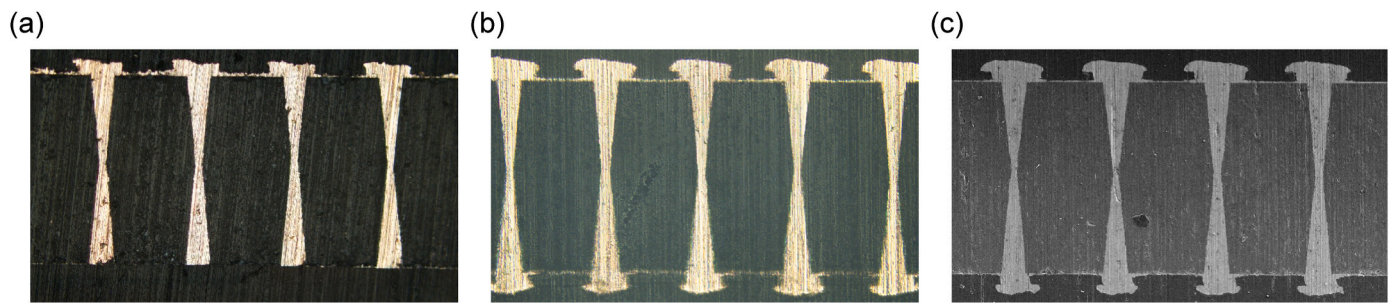


Fig. 9. Reliability evaluation after thermal shock test: (a) cross-sectional image before the thermal shock test, (b) OM image after 100 cycles, and (c) SEM image after completion of the test (AR 8, $-55\text{ }^{\circ}\text{C}$ to $125\text{ }^{\circ}\text{C}$).

respectively. In the case where only electroless plating was applied, the occurrence of voids at the via center was observed to vary as a function of the applied process conditions (Figs. 4(d-2)). Conversely, when the vacuum-assisted wetting process was applied, uniform, void-free filling was consistently achieved under all current density conditions. Fig. 8(d) shows the filling results for high-AR structures, demonstrating that stable and void-free filling behavior was obtained for vias with aspect ratios ranging from AR 6 to AR 10. This improvement is attributed to enhanced electrolyte infiltration into the via interior as a result of the vacuum-assisted wetting process. Consequently, the vacuum-assisted wetting process effectively reduces the limitations associated with electrolyte infiltration in high-AR structures, decreases sensitivity to variations in process conditions, and enables reliable void-free filling.

3.4. Reliability (TS test)

To evaluate the influence of the vacuum-assisted wetting process on the reliability after electroplating, a thermal shock test (TS test) was conducted. The test was performed over a temperature range from $-55\text{ }^{\circ}\text{C}$ to $125\text{ }^{\circ}\text{C}$, and the detailed temperature profile and cycle conditions are explicitly provided in Fig. S6. Reliability evaluation was performed using samples with different AR values, and representative AR 8 samples were selected for analysis. The test was performed for a total of 100 cycles. Fig. 9(a) presents the cross-sectional image before the TS test, while Fig. 9(b) and (c) present the OM and SEM analysis results obtained after the test, respectively. After 100 cycles, no noticeable structural degradation or damage was observed in the evaluated sample structures. The internal filling structure was also maintained without the presence or formation of defects such as seams or voids. These results indicate that the filling structure formed via the vacuum-assisted wetting process exhibits structural stability under thermal shock conditions and demonstrates its applicability and suitability for high-AR structures.

4. Conclusion

In this study, the pretreatment stage of Cu electroplating was comprehensively investigated with the objective of achieving reliable void-free via filling in high-AR TGVs. The main findings drawn from each subsection are summarized as follows:

- (1) Electroplating and additive behavior. CVS and LSV analysis demonstrated that additive adsorption is mass-transport dependent, producing spatially non-uniform Cu deposition in the flow-restricted interior of high-AR TGVs.
- (2) Seed-layer formation. A sputter-only seed layer failed to cover high-AR sidewalls conformally, causing central voids for $\text{AR} \geq 6$. Electroless Ni plating provided a continuous seed layer and enabled void-free filling at 0.5 A/dm^2 , but complete void suppression was not guaranteed owing to restricted electrolyte penetration.

- (3) Vacuum-assisted wetting. Contact-angle analysis revealed that metal-coated surfaces and highly tapered geometries reduce wettability and promote gas entrapment at the via center. Applying a vacuum ($-95 \pm 5\text{ kPa}$ for 3 min) enlarged the pressure difference across the gas-liquid interface, causing trapped bubbles to expand and detach. Consequently, uniform void-free Cu filling was consistently achieved for aspect ratios up to 10 across current densities of $0.3\text{--}0.7\text{ A/dm}^2$, substantially broadening the process window.
- (4) Reliability. Cu-filled AR 8 TGVs subjected to 100 cycles of thermal shock ($-55\text{ }^{\circ}\text{C}$ to $125\text{ }^{\circ}\text{C}$) exhibited no seams, voids, or interfacial delamination, confirming the structural robustness of the vias fabricated with the proposed vacuum-assisted wetting process.

Overall, vacuum-assisted wetting is established as an effective pretreatment strategy for reliable void-free filling in high-AR TGVs, providing a practical basis for next-generation glass-interposer packaging.

Data availability statement

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jmrt.2026.08.005>.

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