Deformation mechanics of sputtered copper layers during nanoindentation tests

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ABSTRACT – The mechanical properties of the thin sputtered copper layer on the SiO₂-coated silicon substrate is needed as part of the requirements in quantifying the reliability of the Through-Silicon Via (TSV) interconnects. In this respect, two different Cu coating layers, each from the different sputtering process, are examined. A series of nanoindentation tests are performed on the Cu coating layer samples with indenter speeds ranging from 80 to 400 nm/s, and the indentation depths of 320 nm. The properties of elastic modulus, hardness and the hardening behavior of the Cu coating layers have been quantified. Results show that the coating with higher contamination of C at 8.41 wt. % displays a significant hardening and a peak load level, as reflected in the measured nanoindentation load-displacement curves. However, insignificant effect of the applied probe displacement speeds up to 400 nm/s on the resulting properties of the coating is registered. The Johnson-Cook constitutive equation adequately describes the strain rate-dependent hardening behavior of the Cu coating layer.

INTRODUCTION

Thin copper layers are commonly deposited as copper (Cu) traces on the printed wiring boards, and as a filler metal in Through-Silicon Vias (TSV)s. The conductive Cu traces and fillings function is to provide the current path for the microelectronic device. However, the temperature excursions during the reliability tests and operating conditions of the device induce stresses due to the mismatches of the coefficient of thermal expansions within the Cu/substrate bonded structure. Such a reliability concern over the failure of the copper layer has been addressed through experimental and computational studies e.g. [1,2]. Yielding and fracture of the copper layer has been identified as a cause of failure of the TSVs [3-6]. The failure processes of the TSV are dictated, to a large extent, by the deformation response of the conductive copper layer under the operating load conditions. Thus, the prediction of the device reliability and mechanical performance necessitates the deformation mechanics of the copper layer to be thoroughly understood and quantified.

Mechanical properties of Cu in the bulk material form are determined through tensile and fracture toughness tests on the samples of the metal. However, microtensile tests of freestanding Cu thin film specimen, and nanoscale cantilever beam tests for the Cu film have been performed to establish the properties of the material [6,7]. The Young’s modulus, hardness and fracture toughness of Cu in the form of a coating layer is very different from the bulk properties. The yield stress of the 20 nm-thick Cu film is ten times higher than its bulk counterpart [7]. The yield stress and elastic modulus are reported to be 765 MPa and 129 GPa, respectively, while the typical corresponding properties of bulk copper are much lower at 70 MPa and 117 Gpa, respectively. The observed difference in the mechanical properties is likely attributed to the different microstructures of the bulk and thin coating, as derived from the processing of the materials. Although the thin Cu coating layer produced by electroless plating and displayed similar grain size with that of sputtered coating layer, the latter exhibits diffused grain morphology [8].

Nanoindentation tests have also been performed on the coating layers to establish the load-displacement response, and extract the elastic modulus and hardness value of the materials [8-14]. During the test, the elastic-plastic response is measured during the loading of the hard indenter into the surface of the relatively softer coating layer. The peak load level is used to define the hardness value of the layer material. The Young’s modulus is extracted from the initial unloading part of the load-displacement curve, assuming a purely elastic response. The influence of nanoindentation test variables, including the depth of the indent, the degree of the orthogonality of the indenter to the coating surface, and the coefficient of friction between the indenter and the coating material, on the extracted property values has been examined [15]. The true hardening behavior of the layer could be obtained using the loading portion of the measured load-displacement curve, through the inverse analysis approach [16-19]. When the sample shows the sign of a crack, the fracture toughness of the material could also be established [20].

This paper examines the deformation response of the sputtered Cu coating layer during the nanoindentation test. The effect of different probe speeds on the resulting hardness and modulus properties of the layer is examined. The strain rate-dependent hardening behavior of the Cu coating layer is described.

ARTICLE HISTORY

Revised: 27th Oct 2019
Accepted: 20th Nov 2019

KEYWORDS

Hardness; nanoindentation test; sputtered copper coating; Young’s modulus.
MATERIAL AND EXPERIMENTAL PROCEDURES

Sputtered Copper Coating Layer

Copper is sputtered onto the $SiO_2$-coated silicon ($Si$) substrate. The $Si$ wafer with the orientation of [1 0 0] has a thickness of 525 µm, while the $SiO_2$ coating is 300 nm-thick. The wafer is diced into square-shaped substrates measured 10x10 mm$^2$ for the sputtering process. Two different sets of Cu-sputtered specimens are produced by different magnetron sputtering machines and processing parameters. The first set, labeled $SP1$ are sputter-coated using the Model TF450 PVD system. The other set of specimens, labeled $SP2$ are sputtered using the Evatec Clusterline CLC200 Metal Sputter. The substrate is maintained at room temperature during the sputtering process. The process parameters used are listed in Table 1.

| Equipment/Process Parameters | $SP1$ | $SP2$ |
|-----------------------------|-------|-------|
| Sputtering Machine          | TF450 Sputtering System, SG Control Engineering | Evatec Clusterline CLC200 Metal Sputter |
| Deposition Time (S)         | 3600  | 240   |
| Base Vacuum Pressure (Mbar) | $3.5 \times 10^{-5}$ | $1.25 \times 10^{-3}$ |
| Argon Flow Rate (Cm$^3$/Min) | 15    | 24    |
| Dc Voltage (V)              | 430   | 720   |
| Dc Power (Kw)               | 0.103 | 12    |
| Dc Current (A)              | 0.24  | 16.7  |

The thickness of the sputtered Cu coating samples is 8 µm. The samples are etched in a mixture of 25 ml HCl, 5 g ferric chloride and 100 ml distilled water to better reveal the microstructure. The resulting microstructures of the Cu coating layer for the $SP1$ and $SP2$ samples are shown in the micrographs Figure 1 and Figure 2, respectively. The microstructure of the sputtered copper sample $SP1$ shows a square-like shapes with sides measuring ranging from 13 to 20 µm. In contrast, the $SP2$ microstructure displays a smoother surface with randomly distributed fine sputtered copper particles. The relatively longer deposition time will likely cause the grains to grow and even recrystallize [21].

![Figure 1. Surface structure of the Cu coating layer, $SP1$.](image_url)
Figure 2. Surface structure of Cu coating layer, SP2.

The corresponding composition analysis of the SP1 and SP2 sample, as represented by the EDX elemental spectrums, are compared in Figure 3(a) and (b), respectively. The primary constituents of the sputtered Cu coating layer are compared in Table 2. A higher carbon content of 8.41 wt. % is detected in the Cu-coating for SP1 sample, possibly due to contamination induced from either Cu-target poisoning or through the incorporation of the residual gas [22]. The excess carbon would likely result in higher strength of the Cu coating material. However, due to increase in carbon percentage, the brittleness of the material is also increased. The carbon will act as a barrier to dislocation mobility, where the movement is related to the ductility of materials. Metals with less carbon content are relatively ductile because the dislocations can easily glide on crystallographic planes during the deformation.

Table 2. Chemical composition (wt. %) of the sputtered Cu coating layer.

| Element   | SP1   | SP2   |
|-----------|-------|-------|
| Carbon    | 8.41  | 2.08  |
| Oxygen    | 0.45  | 0.33  |
| Copper    | 91.14 | 97.58 |
The resulting surface morphology of the Cu coatings is compared in Figure 4 (a) and (b) for the sample SP1 and SP2, respectively. These micrographs are obtained using the scanning probe microscope. Based on the sampling area of 10x10 µm², sample SP1 exhibits a relatively flat surface with a low number of peaks and valleys. Although the highest peak, at 135 nm is found on sample SP1, a larger number of peaks at 93 nm describe the morphology of sample SP2, as shown in Figure 4 (b). The three deep valleys, as indicated in each micrograph, are the permanent indentation marks left by the nanoindentation tests to the specified depth of 320 nm, and at the probe speed of 80 nm/s. The measured peaks and valleys, however, are much less than the magnitude of the indentation depth (320 nm). The morphology of the sputtered Cu coating surface could significantly affect the measured properties through non-orthogonality of the axis of the indenter with the local surface during the test, as discussed in the next section.
Nanoindentation Test

Nanoindentation tests were performed on the sputtered copper coating samples using the Hysitron TI 750 Ubi Triboindenter. The indenter used is a Berkovich diamond indenter tip with a radius of 100 nm. The inclination of the three surfaces of the pyramid-shaped indenter to the horizontal plane would produce an indentation mark of an equilateral triangle. The load and probe displacement resolution of the system is 1 nN and 0.04 nm, respectively. The tests conformed to the ASTM E2546-07 standard [8]. A valid nanoindentation test requires the penetration depth not to exceed 10% of the thickness of the coating layer.

The tests on samples SP1 were initially conducted at 80 nm/s to determine the valid minimum indentation depth [23]. The indentation depths range from 80 to 320 nm and limited by the upper range of the load cell at 12 mN. Then, samples SP2 were tested at different specified probe speeds from 80 to 400 nm/s. The indenter is held for a period of 2 seconds at the maximum load followed by unloading, during each test. A minimum of three nanoindentation tests were repeated for each probe speed to establish the repeatability of the load-displacement (depth) response. The load-displacement data are recorded during the loading-unloading part of the nanoindentation cycle.

During the indentation process, the Berkovich indenter tip is driven into the surface of the Cu coating layer to the maximum specified depth, followed by unloading. A typical load-displacement curve is shown in Figure 5. The nonlinear loading part of the curve consists of the elastic-plastic deformation in forming the indent on the Cu coating surface, while the initial unloading part from the peak load level is assumed to be purely elastic. The residual displacement at zero load following the unloading process reflects the extent of the plastic deformation. The area enclosed by the curve represents the energy dissipated during the nanoindentation process.

Figure 5. Typical load-depth curve of nanoindentation test.
The mechanics of materials-based equations have been established to extract the hardness and elastic modulus of the material. The derivation of these equations is summarized here for convenience. The hardness measure, $H$ that quantifies the resistance of the material to plastic deformation is defined as [24]:

$$H = \frac{P_{\text{max}}}{A_p}$$  \hspace{1cm} (1)

where $P_{\text{max}}$ is the measured peak load and $A_p$ is the projected area between the Berkovich indenter and the $Cu$ layer, defined as [25]:

$$A_p = 24.5h_c^2$$  \hspace{1cm} (2)

In Equation. (2), the contact depth, $h_c = h_{\text{max}}$ corresponds to the peak load, $P_{\text{max}}$. The effect of the indenter shape on the calculated property values is represented by the coefficient of $24.5$ for the Berkovich type indenter in Equation. (2). The radius of the indenter tip need to be considered to ensure the correct determination of the projected contact area at the nanoscale [26].

The Young’s modulus of the copper layer, $E_c$ is determined as:

$$E_c = \frac{1 - \nu_c^2}{1 - \nu_i^2} \left( \frac{E_i}{E_r} \right)$$  \hspace{1cm} (3)

where $\nu_i = 0.07$ and $\nu_c = 0.30$ is the Poisson ratio of the diamond indenter, and $Cu$ coating layer, respectively. The Young’s modulus of the diamond indenter, $E_i = 1140$ GPa. The reduced modulus, $E_r$ is determined as:

$$E_r = \frac{\sqrt{\pi}}{2} \frac{\beta S}{\sqrt{A_p}}$$  \hspace{1cm} (4)

where $S = \frac{dP}{dh}$ is the initial slope of the unloading line at the peak load. The Berkovich constant $\beta = 1.034$.

RESULTS AND DISCUSSION

The deformation response of the $Cu$ coating layer on the $SiO_2$-coated $Si$ substrate is presented and discussed in terms of the load-displacement curve from the nanoindentation test, the resulting elastic modulus and hardness measure, and the hardening curves of the coating layer.

Load-Displacement Response

The deformation responses of the sputtered $Cu$ coating layer during the nanoindentation test at the probe speed of the 80 nm/s and to the indentation depth of 320 nm are compared in Figure 6. The loading stage to the peak load is nonlinear with increasing slope of the load-displacement curve, indicating the hardening of the $Cu$ layer due to the localized plastic deformation. The more gradual change in the slope of the loading curve for the coating layer sample $SP2$ suggests a lower hardening rate, while the lower peak load achieved reflects a relatively softer material than the coating sample $SP1$. The observed “blunt” peak of the curves at the loading-unloading transition could possibly be due to the inherent strain rate-dependent deformation process, particularly in the vicinity of the indenter tip region. However, the initial unloading response for both samples is linear, reflecting an elastic behavior. The displacement at zero-load denotes the residual depth of the indent. The relatively softer $Cu$ coating layer ($SP2$) experienced a deeper residual indent than that of the sample $SP1$. The larger magnitude of the total dissipated energy during the nanoindentation process is associated with the sample $SP1$, as shown by the greater area bounded by the load-displacement curve, than that for the sample $SP2$. The observed difference in the mechanical responses is closely related to the different microstructure of the sputtered $Cu$ coating layers.
A previous experimental study by the authors has indicated that a minimum indentation depth corresponding to 3% of the thickness of the coating is required to eliminate the effect of surface morphology on the measured properties of the coating layer [23]. The extracted properties of hardness and Young’s modulus of the Cu coating layer, at different applied probe displacement rates, are shown in Figure 7 and 8, respectively. Each symbol represents three repeats of the measurements. Repeatability of the extracted data is excellent, as indicated by the small uncertainty range of the data.

The hardness of the sample SP1 with an average value of 3.0 GPa is higher than that of sample SP2 at 1.3 GPa. This is likely due to the relatively higher contaminated C content in the sample, as deliberated earlier (see Table 2). The insignificant effect of the higher applied probe displacement rates of up to 400 nm/s on the hardness measure is observed. The Young’s modulus of the sample SP1 at an average value of 95 GPa is only slightly lower than that of the sample SP2 at 107 GPa. These measured values are comparable to the reported Young’s moduli of thin Cu layers that range from 125 to 170 GPa, and significantly higher than that of the bulk copper at 70 GPa [27, 28]. In addition, the modulus property values are independent of the probe displacement rates.

**Figure 6.** Load-displacement responses of the Cu coating samples during the nanoindentation test to the depth of 320 nm, at 80 nm/s.

**Figure 7.** Hardness measure of the Cu coating samples at different applied probe displacement rates, $h = 320$ nm.
Stress-Strain Response

The load-displacement curve for the loading stage of the nanoindentation test contains information on the hardening behavior of the Cu coating layer. However, the deformation is described by the 3-D stress and strain state. Nevertheless, the uniaxial true stress-plastic strain curve of the material could be extracted through the inverse analysis method. Details of the inverse analysis approach that employed the measured load-displacement curve from the nanoindentation test and the finite element simulation of the nanoindentation process is discussed elsewhere [19]. The resulting stress-plastic strain curves for the Cu coating layer, based on the sample SP2, for two different strain rates are compared in Figure 9. The quasi-static initial yield strength of the Cu coating layer is 279 MPa. The higher strain rate of 0.608/s results in higher level of the initial yield strength of 520 MPa. In addition, the strain rate-dependent hardening behavior of the sputtered Cu coating layer could be represented by the Johnson-Cook constitutive equation as [19, 29]:

$$
\sigma = \left[ 279 + (2633)\varepsilon^{(0.66)} \right] \left[ 1 + (0.275)ln\dot{\varepsilon} \right]
$$

The first term on the RHS of Equation (5) represents the hardening behavior while the second term represents the strain rate effects.

![Figure 9](image.png)

**Figure 9.** True stress-plastic strain curves of the sputtered Cu coating layer.
CONCLUSIONS

The deformation response of the sputtered Cu coating on SiO₂-coated Si substrate during the nanoindentation test has been established. The resulting properties of hardness, Young’s modulus and hardening behavior have been quantified for two different microstructures of the coatings. Results show that:

1. The Cu coating with higher contaminated C content of 8.41 wt. % displays a significant hardening and a peak load level, as reflected in the measured nanoindentation load-displacement curves.
2. The insignificant effect of the applied probe displacement speeds up to 400 nm/s on the resulting properties of the coating is registered.
3. The hardening behavior of the Cu coating layer is adequately described by a strain rate-dependent stress-plastic strain curves through the Johnson-Cook constitutive equation.

ACKNOWLEDGMENT

This work is funded by the Collaborative Research in Engineering, Science and Technology (CREST) Malaysia and Universiti Teknologi Malaysia under the Grant No. P18C1-13 (UTM Project No. 4B118).

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