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Anelasticity study on electromigration effect in Cu thin films

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Abstract
Electromigration (EM) tests on Cu thin film circuits below or near 373 K with the current density between $1 \times 10^9$ A/m$^2$ and $8 \times 10^9$ A/m$^2$ were carried out by means of the composite vibrating reed method. The resonant frequency ($f$) and the internal friction ($Q^{-1}$) of the composite reed and the resistivity ($R$) of the thin film circuit were measured during isothermal EM tests for as-deposited Cu/Ta films, Cu/Ta films annealed at 450 K and as deposited Ta/Cu/Ta films. An increase in $f$, a decrease in $Q^{-1}$ and a decrease in $R$ were commonly observed. The activation energies found for the EM process ($E_{EM}$) range between 0.21 and 0.41 eV for the as-deposited Cu/Ta films, between 0.21 and 0.59 eV for the annealed Cu/Ta films and around 0.3 eV for as deposited Ta/Cu/Ta films. It is suggested that a previously unrecognized mass transport process with low $E_{EM}$ operates in these Cu thin films.

Key words: Electromigration, Cu interconnects, Young’s modulus, Internal friction
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1. Introduction
With increasing down-sizing in ultra-large scale integration (ULSI) devices, the current density in copper (Cu) leads goes up beyond $10^{10}$ A/m$^2$ and the electromigration (EM) tolerance comes to play a crucial role in the reliability of ULSI devices [1]. The data on the EM tolerance of Cu interconnects have been gathered above 573 K in accelerated tests, where the activation energy, $E_{EM}$, found ranges between 0.47 eV and 1.25 eV [2]. Most of these $E_{EM}$ values range between 0.75 eV and 1.2 eV which have been reported for the activation energy for the grain boundary diffusion, $E_{GB}$, in ultra-high-purity Cu specimens [3] and in conventional high-purity Cu specimens [4], respectively. On the other hand, it is also reported that the $E_{EM}$ values found depend on the Cu interconnect structure [5, 6], indicating that mass transport through the surface (interface) diffusion may play an important role. A recent review [7] emphasizes that although mass transport above 573 K in the accelerated EM tests may be largely affected by the GB diffusion, the EM failure at the operating temperature of 373 K in real devices is induced by surface diffusion. That is, EM tests near or below 373 K may be required to get insight into the EM process at the device operating temperature.

It is also reported that the stress induced by EM in Cu/low-dielectric-constant-material interconnects will be a serious issue because the low-dielectric-constant-materials tend to be mechanically weak [7]. For the mechanical properties of nanocrystalline specimens and/or
thin films of Au [8], Al [9] and Cu [10], the grain boundary anelastic process (GBAP) is thermally activated above 200 K and causes a considerable decrease in the Young’s modulus. On the other hand, an internal friction peak due to the GBAP in bulk Cu is observed near 500 K [11], indicating that the EM process associated with grain boundaries in nanostructured Cu may be not the same as that in bulk Cu. In other words, the EM process in nanostructured Cu interconnects may be affected not only by an increase in the fractional volume of the GB and surface (interface) regions but also by probable changes in the material properties of these regions. To gain insight into the EM process and the effect of EM on the mechanical property in nanostructured Cu interconnects near or below the device operating temperature of 373 K, measurements were carried out using a composite vibrating reed method.

2 Experimental

For a composite vibrating reed, a Si reed with a uniform thin film of metal, a change in the resonant frequency, $f$, of the composite vibrating reed, $\Delta f/f$, due to a change in the Young’s modulus of the film, $\Delta M_f/M_f'$, is given by,

$$\Delta f/f = (3d_f/2d_s)(M_f'/M_s)(\Delta M_f'/M_f'),$$

where $M_s$ and $M_f'$ are the Young’s modulus of the substrate and the film on the substrate, and $d_s$ and $d_f$ denote the thickness of the substrate-reed and the film, respectively [12]. The internal friction, $Q^{-1}$, of the composite reed is given by,

$$Q^{-1} = Q^{-1}_s + (3d_f/d_s)(M_f'/M_s) Q^{-1}_f,$$

and

$$\Delta Q^{-1} = (3d_f/d_s)(M_f'/M_s) \Delta Q^{-1}_f,$$

where $Q^{-1}_s$ and $Q^{-1}_f$ denote the internal friction of the substrate and the film, respectively [13].

The experimental setup of the composite reed with a Cu thin film circuit used here is similar to that reported previously [14]. For the present film on a Si-reed with $d_s = 200 \mu m$, $\Delta M_f'/M_f'$ $\sim$ 13x10$^3$($\Delta f/f$) and $\Delta Q^{-1}_f \sim$ 6.5x10$^3\Delta Q^{-1}$ for $d_f = 20$ nm and $\Delta M_f'/M_f'$ $\sim$ 10x10$^3$($\Delta f/f$) and $\Delta Q^{-1}_f \sim$ 5x10$^2\Delta Q^{-1}$ for $d_f = 200$ nm, where $M_f'$ is $\sim$100 GPa for the 20 nm-thick Cu film and $\sim$130 GPa for the 200 nm-thick Cu film [10] and $M_s$ is 130 GPa [14]. In order to minimize joule heating of a specimen film, thick Si-reeds with $d_f$ of about 200 $\mu m$ were used in the present work. As seen in Eqs. (1) and (3), the experimental errors in ($\Delta M_f'/M_f'$) and $\Delta Q^{-1}_f$ increase with decreasing $d_f$ for this method.

A Si reed-substrate was chemically etched and then subjected to thermal oxidation for 1 hour at 1120 K in a dry O$_2$ gas flow to form a SiO$_2$ surface layer about 10 nm thick. A Ta buffer layer 30 nm thick was deposited by means of DC magnetron sputtering at room temperature in 1.3 $\times$ 10$^{-1}$ Pa 6N-Ar gas atmosphere and then a Cu specimen film was deposited by sputtering. When capping was desired, a Ta layer 20 nm thick was subsequently deposited. Cu film specimens without and with a Ta capped-layer will be referred to as Cu/Ta and Ta/Cu/Ta, respectively. The Cu films were between 10 nm and 600 nm thick. EM tests were made isothermally in a vacuum of 10$^{-4}$ Pa for 2.5x10$^5$ s, where changes in $f$, $Q^{-1}$ and the electrical resistance, $R$, of a Cu thin film circuit were measured. X-ray diffraction (XRD) measurements were performed with Cu-K$\alpha$ radiation with the scattering vector normal to the
flat surface of the Cu film, and reflections from Si powders put on the film surface were used as reference. It is not shown here but the followings were observed for the XRD data. For Cu/Ta films, the XRD 111 reflection was predominant for $d_t$ less than about 50 nm and the fractional intensity of the XRD 200 reflection, $I_{200}/(I_{111}+I_{200})$ [hereafter], increased to about 0.2 with increasing $d_t$ to 600 nm. The mean grain size, $D_{111}$, determined from the peak width of the XRD 111 reflection was $\sim 15$ nm for $d_t$ near 20 nm and increased to $\sim 30$ nm with increasing $d_t$ to 600 nm. Outlines of the film thickness dependences of $I_{200}/(I_{111}+I_{200})$ and $d_t$ observed for Ta/Cu/Ta films were similar to those mentioned for Cu/Ta films. On the other hand, values of $I_{200}/(I_{111}+I_{200})$ and $d_t$ observed for Ta/Cu/Ta films were slightly larger than those for Cu/Ta films, indicating that capping by a Ta layer caused the spontaneous grain growth.

3. Results

Figures 1(a), 1(b) and 1(c) show the changes in the resonant frequency, $f$, and the internal friction, $Q^{-1}$, of the composite reed as well as the resistance, $R$, of the Cu/Ta film circuit observed during EM tests at 300.9 K for an as deposited Cu/Ta film. $f$, $Q^{-1}$ and $R$ showed an increase, a decrease and a decrease showing saturation during the EM test for 2.4x10$^5$ s, respectively. The variations of these quantities, $\Delta f$, $\Delta Q^{-1}$ and $\Delta R$, with elapsed time were explained well by a simple exponential decay function, the time constant, $\tau$, showing good agreement with the experimental error, among these quantities. For Cu/Ta films after annealing at 450 K, changes in $\Delta f$, $\Delta Q^{-1}$ and $\Delta R$ observed during the EM tests at 300 K and 1x10$^9$ A/m$^2$ and the EM tests at 330 K and 4x10$^9$ A/m$^2$ were similar to those seen in Fig. 1 except that the magnitudes of $\Delta f$, $\Delta Q^{-1}$ and $\Delta R$ found for the EM tests at 300 K and 1x10$^9$ A/m$^2$ were one order smaller than those seen in Fig. 1 (not shown here). Figures 2(a) to 2(c) are similar to Figs. 1(a) to 1(c) but they were observed for a Ta/Cu/Ta film circuit with $d_t$ of 206 nm at $T_{EM}$ of 373. 0 K and $i_d$ of 8x10$^9$ A/m$^2$. The variations of $\Delta f$, $\Delta Q^{-1}$ and $\Delta R$ with elapsed time seen in Fig. 2 were very similar to those shown in Fig. 1 except that $\tau$ became shorter in Fig. 2. For the EM tests with increased $i_d$ shown in Fig. 2, the reverse changes in $\Delta f$, $\Delta Q^{-1}$ and $\Delta R$ were observed for $t > 2x10^5$ s, indicating that an outflow of Cu from the thin film circuit started.

As mentioned for Figs. 1 and 2, $\Delta f$, $\Delta Q^{-1}$ and $\Delta R$, were explained well by an exponential decay function for the present testing time of 2.5x10$^5$ s, where the magnitudes of these quantities, $\Delta f_0$, $\Delta Q^{-1}_0$ and $\Delta R_0$, were estimated from the observed data. Then, $\Delta M'^{-1}_{f,a}$ and $\Delta Q'^{-1}_{f,a}$ were determined by using Eqs. (1) and (3). Figures 3(a) to 3(c) show $\Delta M'^{-1}_{f,a}$, $\Delta Q'^{-1}_{f,a}$ and $\Delta R_{a}$, were explained well by an exponential decay function, the time constant, $\tau$, showing good agreement with the experimental error, among these quantities. For Cu/Ta films after annealed at 450 K, $M'^{f}$ showed a small increase for the EM tests at 300 K and 1x10$^9$ A/m$^2$ and a considerable increase for the EM tests at 395 K and 6x10$^9$ A/m$^2$. For as deposited Ta/Cu/Ta films, $M'^{f}$ showed a small increase for the EM tests at 300 K and 1x10$^9$ A/m$^2$ (not shown here) but a considerable increase for the EM tests at 330 K and $\sim 4x10^9$ A/m$^2$ and those at 370 K and $\sim 8x10^9$ A/m$^2$ as seen in Fig. 3(a). The $\Delta Q'^{-1}_{f,a}$ data seen in Fig. 3(b) were similar to the $\Delta M'^{-1}_{f,a}$ data mentioned above except that $Q'^{-1}_{f,a}$ showed a decrease. For the Cu film thickness dependences of $\Delta M'^{-1}_{f,a}$ and $\Delta Q'^{-1}_{f,a}$ observed for as deposited Cu/Ta films, the magnitudes of these quantities appear to show a minimum around 100 nm. The AFM observations on surface morphology showed that the root mean square surface roughness has a maximum around 100 nm for as deposited Cu/Ta films [15]. On the other hand, $M'^{f}$ and $Q'^{-1}_{f,a}$ for as deposited Ta/Cu/Ta films showed a monotonous increase and a monotonous decrease with increasing $d_t$.
above 50 nm.

As seen in Fig. 3(c), \((\Delta R_a/R_0)_{d_t}\) showed always a decrease, where the magnitude of the decrease tended to increase with increasing \(d_t\). It is known that conduction electrons in a thin metal film are scattered by surfaces (interfaces), GBs, lattice defects and phonons, and surface scattering becomes to play the major role with decreasing \(d_f\) \cite{16}. Since a change in \(d_t\) is not expected in the present EM test conditions, the decrease in \(R\) observed here is associated with a decrease in electron scattering by GBs and lattice defects. Such a decrease in \(R\) becomes more sensitively detected with increasing \(d_f\) because of the diminishing role of surface scattering. As seen in Fig. 3(d), \(\Delta I_2/(I_1+I_2)\) showed a decrease except for as deposited Cu/Ta films for \(d_f > \sim 150\) nm. For the as deposited Cu/Ta films, where both \(M'_t\) (Fig. 3(a)) and \(I_2/(I_1+I_2)\) (Fig. 3(d)) showed an increase.

Figures 4(a) to 4(c) show the \(R\) vs. \(t\) data observed for an annealed Cu/Ta film during the EM tests sequentially made at 330 K, 360 K and 395 K with \(6 \times 10^9\) A/m\(^2\), respectively. For the diffusion oriented EM process, the relaxation time, \(\tau\), and the activation energy, \(E_{EM}\), may be related as,

\[
\ln(T/\tau) = \ln[(D_0/k)z^*e\rho i_d] - E_{EM}/kT, \tag{4}
\]

where \(D_0\) is the pre-exponential factor of the diffusion coefficient, \(z^*\) is the effective charge number for the EM effect, \(e\) is the elementary electronic charge, \(\rho\) is the specific resistivity and \(k\) is Boltzmann’s constant. Figure 4(d) shows the Arrhenius plots for the \(\tau\) data found in Figs. (4(a) to 4(c), where \(E_{EM}\) of 0.27 eV is found after application of Eq. (4). Figure 5 shows the present \(E_{EM}\) data together with the \(E_{EM}\) and \(E_{GB}\) data reported (see \cite{1, 2, 5-7, 17} and references therein.

4. Discussion and Conclusion

The present work reports the EM-induced changes in the physical properties of Cu thin films prior to an outflow of Cu from the circuit region. As mentioned, for as deposited Cu/Ta films for \(d_t > \sim 150\) nm, both \(M'_t\) (Fig. 3(a)) and \(I_2/(I_1+I_2)\) (Fig. 3(d)) showed an increase. An increase in \(I_2/(I_1+I_2)\) is expected to cause a decrease in \(M'_t\) because of the strong directional anisotropy of the Young’s modulus of Cu. Thus, the increase in \(M'_t\) found for as deposited Cu/Ta films indicates that an increase in \(M'_t\) due to a decrease in the GBAP should be larger than the counter change due to an increase in \(I_2/(I_1+I_2)\). The \(\Delta Q_{la}^{-1}\) data observed for as deposited Cu/Ta films (Fig. 3(b)) suggest a decrease in the GBAP due to the EM tests too. For most cases, \(I_2/(I_1+I_2)\) showed a decrease after the EM tests, indicating that the increase in \(M'_t\) was composed of a constitutional increase due to a decrease in \(I_2/(I_1+I_2)\) and that associated with a decrease in the GBAP.

As shown in Fig. 5, \(E_{GB}\) is 0.75 eV in ultra-high-purity Cu specimens \cite{3} and 1.2 eV in conventional high-purity Cu specimens \cite{4}. Most of the \(E_{EM}\) data reported range between 0.75 eV and 1.2 eV, for which the mass transfer through GB diffusion has been suggested (see \cite{1, 2, 5-7, 17} and references therein). Previously reported values of \(E_{EM}\) were as low as 0.47 eV claimed to be the activation energy of the surface diffusion, \(E_{SF}\), \cite{17} and as high as 1.4 eV claimed to be due to a Ta capping \cite{18}. These reported \(E_{EM}\) data have come from Blech tests, the EM failure of Cu interconnect or voiding in the accelerated EM tests, where in addition to the mass transfer through the GB diffusion and the surface diffusion, EM-induced damage occurring away from the high current region has been reported \cite{7}. In the present work, values of \(E_{EM}\) range from 0.21 eV to 0.41 eV for the as deposited Cu/Ta films, 0.21 eV to 0.59 eV for the annealed Cu/Ta films and around 0.3 eV for the as deposited Ta/Cu/Ta film. Thus,
the values of $E_{EM}$ found for Cu/Ta and Ta/Cu/Ta films in the resent work are similar to each other but lower than values reported in the accelerated tests. For Cu/Ta films, the magnitudes of $\Delta f$, $\Delta Q^{-1}$ and $\Delta R$ observed during the EM tests at 300 K and $1\times10^9$ A/m$^2$ were much lower in the annealed state than in the as deposited state. On the other hand, the magnitudes of $\Delta f$, $\Delta Q^{-1}$ and $\Delta R$ observed during the EM tests at 330 K and $6\times10^9$ A/m$^2$ for the annealed Cu/Ta specimens were similar to those in the as deposited Cu/Ta films for the EM tests at 300 K and $1\times10^9$ A/m$^2$. It is indicated that the volume fraction of the GB regions and the surface regions responsible for the EM process were decreased after annealing at 450 K, but $E_{EM}$ remained almost the same. On the other hand, the recoverable lattice defects with activation energies in the range between 0.21 eV and 0.59 eV are expected to be annealed out at 450 K. For Ta/Cu/Ta films, the recoverable lattice defects with the activation energy of 0.3 eV are expected to be eliminated after the grain growth due to a Ta capping. From those considerations, the present work suggests that a previously unrecognized mass transport process with low $E_{EM}$ operates in the nanostructured Cu thin films. To get insight into this process, further work is in progress.

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References

Fig. 1.
An example of the EM tests for an as deposited Cu/Ta film, where $d_f$, $T_{EM}$ and $i_d$ were 48 nm, 300.9 K and $1 \times 10^9$ A/m$^2$, respectively. (a) $f$, (b) $Q^{-1}$ and (c) $R$. 

$f_0=799.49$ Hz
48nm Cu/Ta 300.9 K, $1 \times 10^9$ A/m$^2$

$Q^{-1}_0=2.74 \times 10^{-5}$

$R_0=18.51$ Ω
Fig. 2.
An example of the EM tests for an as deposited Ta/Cu/Ta film, where \( d_f, T_{EM} \) and \( i_d \) were 206 nm, 373.0 K and \( 8 \times 10^9 \) A/m\(^2\), respectively. (a) \( f \), (b) \( Q^{-1} \) and (c) \( R \).
Fig. 3.
The Cu film thickness dependence of the EM-induced changes in (a) $\Delta M'_{f,a}$, (b) $10^3 \Delta Q^{-1}_{f,a}$, (c) $(\Delta R_a/R_0)_f$, and (d) $\Delta I_2/(I_1+I_2)$. ○; for as-deposited Cu/Ta (300 K, 1x10^9 A/m^2), △; for annealed Cu/Ta (300 K, 1x10^9 A/m^2), ▲; for annealed Cu/Ta (395 K, 6x10^9 A/m^2), ■; for as-deposited Ta/Cu/Ta (330 K, 4x10^9 A/m^2) and ◆; for as-deposited Ta/Cu/Ta (370 K, 8x10^9 A/m^2). See text for details.
Fig. 4.
(a) to (c): An example of the temperature dependence of the EM-induced changes in $R$ observed for a Cu/Ta film, where $d_f$ and $i_d$ were 30 nm and $6 \times 10^9$ A/m$^2$, respectively. (d) The Arrhenius plots to find $E_{EM}$.

$E_{EM}=0.27\text{eV}$
Fig. 5.
$E_{EM}$ and $E_{GB}$ data are plotted against $d_f$. Present $E_{EM}$ data, ○: as deposited Cu/Ta, ▲: annealed Cu/Ta, ■: as deposited Ta/Cu/Ta. Reported $E_{EM}$ data, +: Cu without a capped layer, ●: Cu with a capped layer. Reported $E_{GB}$ data, ◇: bulk Cu [3,4]. For the reported EM data, see [1, 2, 5-7, 17] and references therein.