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High-Frequency Electrical Properties of Silver Thick Films Measured by Dielectric Resonator Method

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The electrical properties of silver films, prepared using a low-curing-temperature metallo-organic-decomposition (MOD) paste and a high-temperature silver paste screen-printed on polished and nonpolished alumina substrates, at microwave frequency were characterized in this study. Surface resistance and effective conductivity of the silver films at microwave frequency (approximately 4 GHz) were evaluated using the TE_{011} mode of the resonator cavities method. Devices of T-type resonator circuits were fabricated to determine the simulated and measured Q-values and to evaluate the effects of silver films and the surrounding substrate. The surface roughness of the fired films printed on nonpolished Al_2O_3 substrate is slightly less than those on polished substrate, because the surface energy of the nonpolished alumina (29.81 mN/m) is slightly less than that of the polished alumina (36.69 mN/m). The calculated effective conductivities at 4.3 GHz are slightly less than the DC conductivities of the films. Moreover, the films prepared using the high temperature silver paste have higher electrical conductivity ranging from 4.08×10^7 to 4.13×10^7 S/m, since the high-temperature firing process leads to an improved connectivity of the silver particles. The results indicate that the films screen-printed on the polished substrate have a higher Q and a lower ΔQ value than those of films that are screen-printed on the nonpolished substrate. For the silver films prepared using the high-temperature silver paste, both the Q and ΔQ values were the highest among the films studied, which is consistent with the observation of the dense microstructure of the silver film and the interfacial reaction between the glass in the film and the substrate as a result of high firing temperature. [DOI: 10.1143/JJAP.47.7289]

KEYWORDS: electrical properties, microwave frequency, silver film, metallo-organic decomposition compound

1. Introduction

Traditionally, commercial conductive inks designed for various printing processes have two types of formulation, one method is to use a conducting metal particle as the conducting phase and use resins as the binders and vehicles, and the other is to use a metal particle as the conducting phase while glass acts as the binding and adhesive agent. However, the glass, binder, and vehicles are generally nonconductive, which may hinder the contacts of metal powders and reduce the electron transmission.

For flexible applications, low-curing-temperature conductor technology with metallo-organic-decomposition (MOD) compound additions was investigated to form a three-dimensional metal network. There is no binder and glass addition in the MOD compounds. The MOD compounds are generally synthetic metallo-organic salts that decompose completely at low temperature to precipitate metal or oxide, depending on the metal and firing atmosphere.[1,2)] A previous study[1)] indicated that a paste containing 2-ethylhexanoate ($C_8H_{15}O_2Ag$) has a very low decomposition temperature (190.3 °C) among MOD agents, and it forms silver particles to promote the linking among silver flake particles and thus reduces the resistivity to $<13\ \Omega$ cm at temperatures as low as 200 °C. However, the electrical resistivity of the metal film was affected by the connectivity of metal particles in addition to the surface roughness and porosity of the film, which resulted from the evaporation of organic matter at high temperature.

Recently, there has been tremendous interest in the development of thick-film conductors for use in high-frequency applications, such as radio-frequency identification (RFID) tags. The behavior of an electromagnetic wave traveling through the metal film is dependent on the characteristics of the metal film and the surrounding substrate. Therefore, it is important to understand the electrical properties of metal films at high frequencies. To date, very little information regarding the electrical properties of silver thick-films at microwave frequency is found in the literature. In this study, a low-curing-temperature silver paste containing silver 2-ethylhexanoate[1)] and a conventional high-temperature silver paste containing a glass binder were screen-printed on Al_2O_3 substrates with polished and nonpolished surfaces and then fired at an optimum temperature. The surface resistance and effective conductivity of the silver films at microwave frequency (approximately 4 GHz) were evaluated using the TE_{011} mode of the resonator cavities method. The surface and cross-section microstructures of the films were characterized by scanning electron microscopy (SEM). The effects of the silver film microstructure and substrate characteristics on the electrical resistivity of the silver films are discussed.

2. Theoretical Consideration of Surface Resisitivity Measurement

Metal films are widely used in many applications, such as copper cladded laminates, metallized low-temperature cofiring ceramic (LTCC) substrates, conductivity circuits, electric electrodes, and metallized antenna patterns. In the radio-frequency (RF) applications, the metal film conductivity is a critical factor for RF loss and impedance match. For metal films, the surface resistance (R_s) and effective conductivity at microwave frequency can also be measured using the resonator cavities method with a sapphire dielectric resonator operating on its TE_{011} mode, which is the most accurate measurement that can be performed. This method, which was originally used to determine the surface resistance of high-temperature superconductors,[3,4)] could also be

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used to measure the surface resistance of bulk conductors.[5] From the theoretical treatment of the TE_{011} resonant mode, the surface resistance and RF current density of the film can be calculated from the unloaded Q-value and the dissipated power. The advantage of using TE_{011} mode cavities for surface resistance measurements is that there are simple and exact formulas for the resonant frequency of such cavities as well as for the quality factor, and the unique surface current distribution allows one to avoid contact problems between bottom and lateral surfaces in such cavities. As presented by Krupka,[5] the total unloaded Q-factor of the resonant structure in the dielectric resonator measurements can be expressed as

$$Q^{-1} = Q_c^{-1} + Q_p^{-1} = \frac{R_{surface}}{G_b} + Q_p^{-1}, \quad (1)$$

where Q_c is the Q-factor associated with conductor losses in the measured metal layer, Q_p is the Q-factor due to parasitic losses from all other parts of the resonant structure, G_b is a geometric factor for bottoms of the resonant structure, and $R_{surface}$ is the surface resistance of the measured metal layer. If the Q-factor due to parasitic losses (Q_p) and the measured unloaded Q-factor value are known, the surface resistance of metal layers can be calculated using the formula as

$$R_{surface} = G_b(Q^{-1} - Q_p^{-1}). \quad (2)$$

In traditional bulk metals for RF applications, the electric field decays exponentially over a skin depth of $\delta = (2/\omega\sigma\mu_0)^{1/2}$.[6] If the thickness of the metal film is several times larger than the skin depth, the effective conductivity can be determined using the formula as

$$\sigma = \frac{\pi f \mu_0}{R_{surface}^2}, \quad (3)$$

here, $\mu_0 = 4\pi \times 10^{-7}$ H/m and f is the measurement frequency.

3. Experimental Procedure

3.1 Sample preparation

The low-curing-temperature silver paste used in this study was prepared using Ag flake (Ferro), a metallo-organic compound of silver 2-ethylhexanoate (STREM), and a solvent of α-terpineol (TCI). All the materials were mixed using a high-speed mixer (Thinky Mixer) for 3 min and de-bubbled for 1 min. Subsequently, a uniform paste was formed using a triple-roller grinder (EXERT), which causes the breakdown of pigment agglomerates. The weight ratio of the silver flake powder to solvent was fixed at 25 : 75. Silver 2-ethylhexanoate was added at a level equal to 5 wt % of the total silver flake powder and solvent. Also, a commercial silver paste (Ag paste 6142D, DuPont), which is a high-temperature silver paste for use in LTCC applications, was used for comparison in this study.

Large-area metal film samples were prepared for DC conductivity and high-frequency surface resistivity measurements. Two types of silver paste as described above were screen-printed on two types of alumina substrate, including polished and nonpolished substrates (Coors Ceramics). The films prepared using the low-curing-temperature silver paste were fired at 250 °C for 5, 10, 20, and 30 min. The films prepared using the high-temperature silver pastes were debindered at 350 °C for 30 min and then fired at 800 °C for 10 min.

3.2 Characterizations

The surface energies of polished and nonpolished alumina substrates were measured using a goiniometric system (A.S.C. Products VCA 2500XE), which is composed of a microliter syringe for dosing the liquids and an optical system.[7] The contact angles of the liquids, including deionized (DI) water, diiodomethane (Aldrich) and formamide (ACROS), on the substrates were measured. Liquids were dosed onto the substrate surface and the droplet was allowed to stabilize for a few seconds before the contact angle was measured. Five measurements were performed for each liquid to obtain the average value.

The long-term roughness of the silver films was measured using a contact-mode profile meter (Kosaka Surfcorder ET 4000A) for 8 mm line scanning. The short-term roughness of the silver films and bare alumina substrates was measured using a noncontact mode three-dimensional (3D) Optical Profilometer (Nano View NVE 10100) for 100 to 640 μm image area scanning. DC resistance measurements of the films were carried out with spiral silver metal lines with a length (l) of 216 cm, a line width (w) of 0.8 mm, and a line thickness (d) of 10–40 μm, which were screen-printed on a $12 \times 12\,cm^2$ area of the alumina substrate. A Keithely 2400 multimeter with a four-point probe was used to measure the resistance of the silver films. The electrical conductivity based on the equation, $\sigma = L/(R \times w \times t)$, where R is line resistivity, w is the line width, t is the line thickness and L is the line length, was calculated.

High-frequency surface resistances were determined using the TE_{011}-mode dielectric resonator, which was designed by Krupka[5] and manufactured by QWED, as indicated in Fig. 1. To evaluate the Q-factor due to parasitic losses, the dielectric loss tangents of the substrate and dielectric resonator have to be known. The dielectric loss tangent of the substrate was measured by the split-post dielectric resonator method.[8] A barium zirconate titanate (BZT) dielectric resonator of 17 mm diameter and of 8 mm height was used in this study. The measurement frequency ranged between 4 and 5 GHz. The electric energy filling factor and geometric factor were rigorously computed using the Rayleigh–Ritz method.[9] DC surface resistance measurements were performed on the large-area silver films with dimensions of $5 \times 5\,cm^2$. A Keithely 2400 multimeter with a four-point probe was used to measure the resistance of the fired silver films. The surface and cross-section microstructures of the silver films were investigated using a JEOL-6500F SEM.

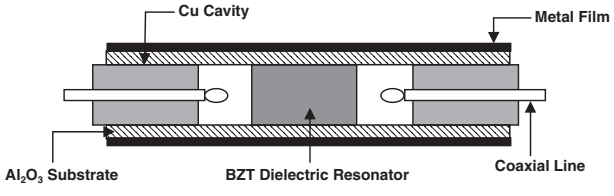

Fig. 1. Schematic of dielectric resonator.

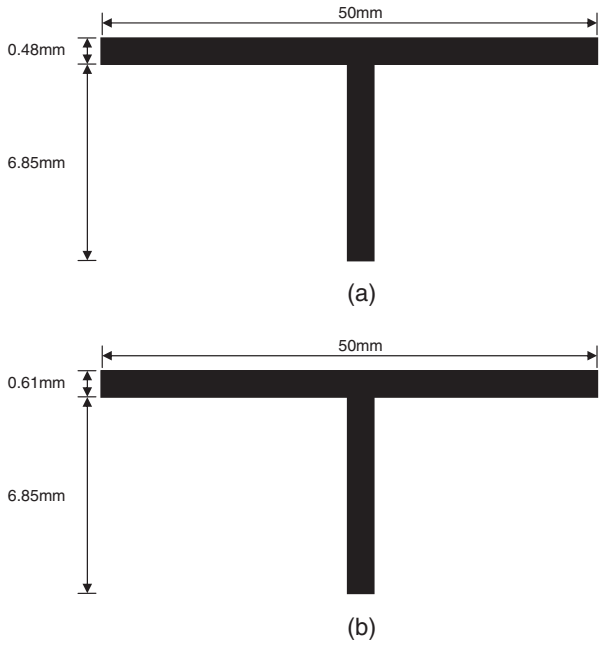

Fig. 2. Test patterns of "T-type" microstrip resonator for silver film printed on (a) polished alumina substrate and (b) nonpolished alumina substrate, resonated at 4.32 GHz.

To evaluate the effects of the silver films and substrates on the device performance, resonator patterns of high-frequency T-type circuits, which were designed to operate at 4.3–4.5 GHz, were fabricated and characterized. As shown in Fig. 2, the widths of the transmission line on the polished substrate (thickness $\approx$ 0.51 mm) and nonpolished substrate (thickness $\approx$ 0.64 mm) were calculated to be 0.483 and 0.610 mm, respectively, in order to meet the nominal resonating frequency at 4.32 GHz. The resonating frequency of S_{21} for the devices was measured using a vector network analyzer (VNA; Anritsu 37397C) with a TRL calibration fixture (Anritsu 3680V Universal Test Fixture) and a K-type calibration kit. Q_{unload} was calculated using the formula, of $Q_{\text{unload}} = f_0/\Delta f$ (f_0 = center frequency; $\Delta f = 3$ dB insertion loss bandwidth).

4. Results and Discussion

Long-term (contact mode, 8 mm) and short-term (noncontact mode) surface-roughness values of the silver films, prepared using the MOD silver paste as well as the high-temperature silver paste screen-printed on polished and nonpolished alumina, are shown in Table I. The short-term surface roughness values of polished and nonpolished bare alumina substrates were found to be 1.93 and 389.9 nm, respectively. The Al_2O_3 substrates used in this study were fabricated using tape casting technology. After sintering, pores or gain boundaries usually existed on the substrate surface, which determines the surface roughness. For the silver films studied, the short-term surface roughness values, shown in Table I, are slightly larger than the long-term surface roughness values. This is due to the fact that the short-term measurement employs area scanning, which traces the local structure of the surface, while long-term measurement operates on a line scanning, which may not map out all the defects or bumpy areas. Comparison of both types of silver film shows that the surface roughness values of the fired films printed on the nonpolished Al_2O_3 substrate are slightly less than those on the polished substrate. The fluidity and wettability of the pastes on the substrate surface may correspond to the differences in the surface roughness of the films. The surface free energies of the polished and nonpolished Al_2O_3 substrates, measured using a goiniometric system, were found to be 36.69 and 29.81 mN/m, respectively. The surface energy of nonpolished alumina is slightly lower than that of polished alumina, because the former has a higher surface roughness. An increase in the surface roughness and number of pores generally causes a decrease in the wettability of hydrophilic oxide surfaces.[10] It is reported that macroscopic pores are generally filled with low-surface-energy contaminants or absorb water. Silver pastes screen-printed on the nonpolished Al_2O_3 with lower surface free energy leads to a better wetting property and fluidity, which allows an improved paste leveling on the substrate and results in a reduced surface roughness of the film.

For films prepared using the low-curing-temperature silver paste heat-treated at 250 °C, the surface roughness slightly changes with soaking time. The surface roughness of the films slightly increases as the soaking time of the heat treatment is increased from 5 to 10 min, which is due to the shrinkage associated with the burnout of the organics. For the films prepared using the high-temperature silver paste, the high-temperature firing process causes the sintering of silver particles. Also, the softening of the glass binder at high temperature levels the silver film and enhances the

Table I. Long-term and short-term surface roughness values of silver films prepared using MOD silver paste as well as high-temperature silver paste screen-printed on polished and nonpolished alumina.

Paste	Substrate	Temperature (°C)	Soaking time (min)	R_a (long-term[a], 8 mm) (μm)	R_a (short-term[b], 640 μm) (μm)
MOD	Polished	250	5	3.46	3.51
			10	3.50	4.04
			30	3.51	3.99
	Nonpolished	250	5	2.74	3.14
			10	2.97	3.42
			30	2.91	3.31
HT	Polished	800	30	0.93	1.42
	Nonpolished	800	30	0.83	0.97

a) Contact mode line scanning roughness measurement.
b) Noncontact mode optical profile surface morphological measurement.

Table II. DC resistivities of silver films prepared using MOD silver paste as well as high-temperature silver paste screen-printed on polished and nonpolished alumina, measured using the four-point probe method.

Paste	Substrate	Temperature (°C)	Soaking time (min)	DC resistivity (Ω cm)	Electrical conductivity[a] (S/m)	Thickness (μm)
MOD[b]	Polished	250	5	0.0024	8.1×10^6	22.83
			10	0.0018	8.2×10^6	23.44
			30	0.0016	9.2×10^6	25.24
	Nonpolished	250	5	0.0025	8.2×10^6	20.93
			10	0.0016	8.2×10^6	25.20
			30	0.0015	9.1×10^6	21.72
HT[c]	Polished	800	30	0.0016	4.13×10^7	6.03
	Nonpolished	800	30	0.0016	4.08×10^7	8.56

a) The electrical conductivity was calculated using $\sigma(\mathrm{S/m}) = L/(R \times w \times t)$; R is the line resistivity, w is the line width, t is the line thickness, and L is the line length.
b) MOD: low-curing-temperature silver paste containing silver 2-ethylhexanoate.[6)]
c) HT: commercial LTCC internal electrode application (DuPont).

Table III. Surface resistance and effective conductivity of silver films prepared using MOD silver paste as well as high-temperature silver paste screen-printed on polished and nonpolished alumina, measured at microwave frequency range.

Paste	Substrate	Temperature (°C)	Soaking time (min)	Frequency (GHz)	Q_{unload}	R_{surface} (Ω)	Conductivity (10^6 S/m)	Skin depth (μm)
MOD	Polished	250	5	4.3231	2555.74	0.05434	5.78×10^6	3.18
			10	4.3242	3073.98	0.04452	8.61×10^6	2.61
			30	4.3241	2802.82	0.04921	7.05×10^6	2.88
	Nonpolished	250	5	4.2536	2637.73	0.05560	5.43×10^6	3.31
			10	4.2486	3244.17	0.04466	8.41×10^6	2.66
			30	4.2461	3197.08	0.04506	8.25×10^6	2.69
HT	Polished	800	30	4.3232	5852.61	0.02149	3.69×10^7	1.26
	Nonpolished	800	30	4.2493	5976.20	0.02213	3.42×10^7	1.32

adhesion with the Al_2O_3 substrate. Sintering and leveling reduces the thickness and surface roughness of the silver film.

Table II shows the results of DC resistivity of the silver films prepared using the MOD silver paste as well as the high-temperature silver paste screen-printed on polished and nonpolished alumina, measured using the four-point probe method. The films prepared using the low-curing-temperature MOD silver paste have electrical conductivities ranging from 8.1×10^6 to 10.5×10^6 S/m after the thermal treatment, which is relatively close to the bulk resistivity of Ag. Although the differences are insignificant, it appears that the electrical conductivity slightly increases with soaking time, owing to the organic burnout and coalescence of the silver particles. The films prepared using the high-temperature silver paste have higher electrical conductivity ranging from 4.08×10^7 to 4.13×10^7 S/m, because the high-temperature firing process leads to a better connectivity of the silver particles. Apparently, the slight difference in the surface-roughness of substrate has no effect on the DC conductivity of the screen-printed silver films.

Table III shows the results of the surface resistance and effective conductivity of the silver films prepared using the MOD silver paste as well as the high-temperature silver paste screen-printed on polished and nonpolished alumina, measured at the microwave frequency range. The surface resistance (R_s) was determined at a frequency of ≈4.3 GHz using the TE_{011} mode of the resonator cavities method. The skin depths of the electric field at microwave frequency for the silver films were calculated, as shown in Table III. The results confirm that the thicknesses of the silver films, shown in Table II, are at least 5 times larger than the skin depth values. The films prepared using the high-temperature silver pastes have surface resistance values smaller than those prepared using the low-curing-temperature MOD silver paste. Comparison of the films prepared using the low-curing-temperature MOD paste after heat treatment at 250 °C for various times shows that the surface roughness increases slightly and the surface resistance slightly decreases with soaking time. This implies that the decrease in the surface resistance of the films is mainly due to the improved connectivity between the silver particles. The surface roughness of the substrate and the film's free surface has no effect on the high-frequency surface resistance of the screen-printed silver films. This may be due to the fact that the films are sufficient to eliminate the surface scattering (size effect),[11,12)] or the differences in the magnitude of the surface roughness are too small to cause a discrepancy in the high-frequency surface resistance. The effective conductivities of the films were determined using eq. (3) and the results are shown in Table III. The calculated effective conductivities at 4.3 GHz are slightly lower than the DC conductivity of the films. Moreover, similar to the DC conductivity, there is no significant difference in the high-frequency effective conductivity among the silver films prepared using the low-curing-temperature MOD silver paste.

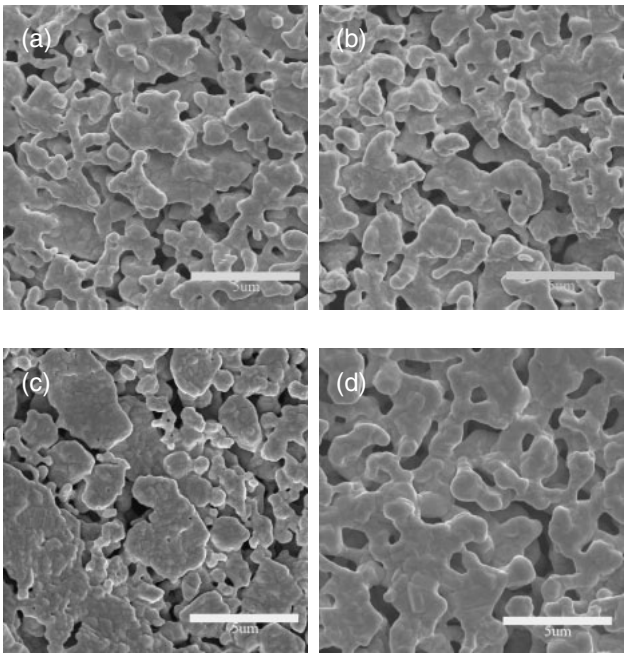

Fig. 3. SEM surface images of films prepared using low-curing-temperature MOD silver paste screen-printed on polished substrate and fired at 250 °C for (a) 10 and (c) 30 min, as well as on nonpolished substrates and fired at 250 °C for (b) 10 and (d) 30 min.

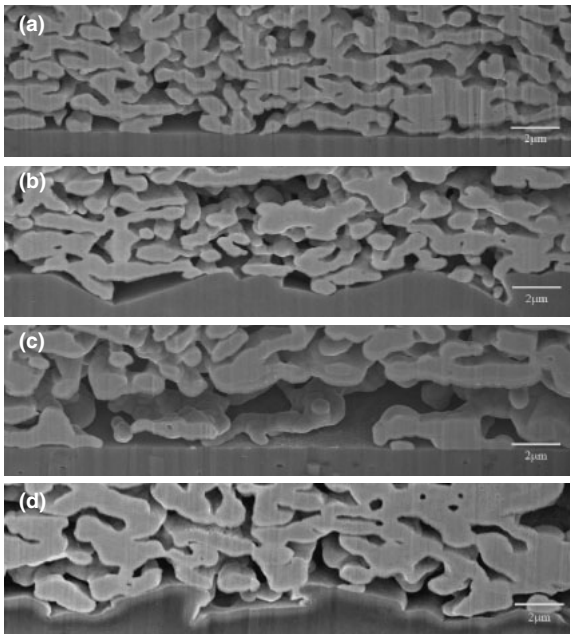

Fig. 4. SEM images of crosssections of the films shown in Fig. 3.

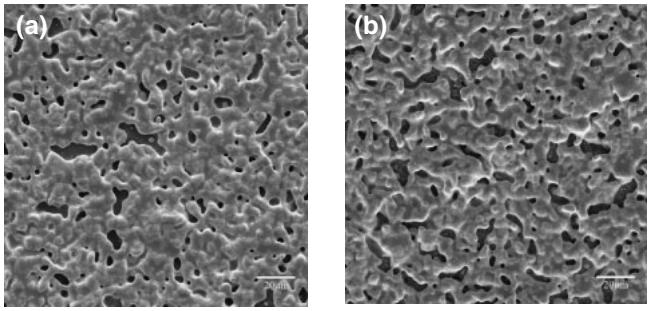

Fig. 5. SEM surface images of films prepared using high-temperature silver paste screen-printed on (a) polished and (b) nonpolished substrate and fired at 800 °C for 10 min.

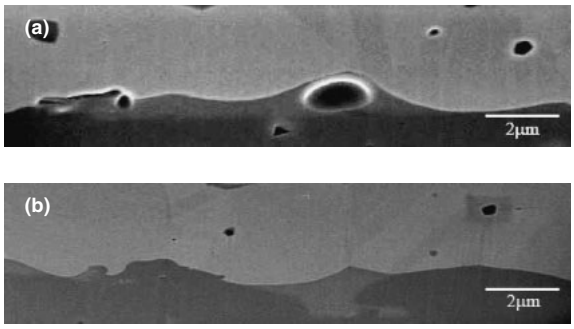

Fig. 6. SEM images of the crosssections of the films shown in Fig. 5.

Figure 3 shows the SEM surface images of the films prepared using the low-curing-temperature MOD silver paste screen-printed on polished as well as nonpolished substrates after heat treatment at 250 °C for 10 and 30 min. Silver grains with neck growth and interconnected porosities are found in the microstructures, which confirm the low resistivities of the silver films. It seems that all the samples have very similar microstructures, although the silver grains are slightly bigger in size for the films being heat treated for 30 min, compared with those films heat treated for 10 min. It is apparent that the nature of the substrate has no effect on the microstructural evolution of the films. Figure 4 shows the SEM images of the cross-sectional view of the films shown in Fig. 3. It can be seen that the microstructures of the films are porous and the coalescent grains and porosities become larger as the soaking time increases. The surface morphologies of the polished and nonpolished substrates do not change after firing because there is no chemical reaction or interdiffusion at the interface at the firing temperature of 250 °C. Figures 5 and 6 show the SEM images of the surfaces and the crosssections of the films, respectively, prepared using the high-temperature silver paste screen-printed on polished and nonpolished substrates. The sizes of the silver grains ranged between 5 to 10 μm. Both the films printed on the polished and nonpolished substrates have an irregular interface. In the high-temperature firing process, the glass added to the film not only densifies the silver film but also wets and reacts with the substrate. This results in a perfect adherence as well as an irregular interface between the films and substrate.

Devices of T-type resonator circuits were designed according to the effective conductivities of the silver films at frequencies of 4.24–4.30 GHz. The simulated and measured Q-values and resonance frequency values of T-type resonators prepared from the films using both low-curing-temperature MOD silver paste and high-temperature silver paste are shown in Table IV. The resonator circuit was designed to have a resonance frequency at 4.3–4.5 GHz. The measured Q-values of T patterns were used to calculate the ΔQ value [$\Delta Q = (Q_{HF} - Q_M)/Q_{HF}$; Q_{HF}: simulated value; Q_M: measured value]. ΔQ indicates the difference between the simulated and measured values of Q. The results indicate

Table IV. Simulated and measured Q-values and resonance frequency values of T-type resonators prepared from the films using both low-curing-temperature MOD silver paste and high-temperature silver paste.

Paste	Substrate	Firing temperature (°C)	Soaking time (min)	Simulation using effective conductivity[a)]		Measured		ΔQ (%)
				Q_{HF}	f_0 (GHz)	Q_M	F_0 (GHz)	
MOD	Polished	250	10	122.6	4.24	93.1	4.26	24.1
			30	114.2	4.26	89.9	4.25	21.3
	Nonpolished	250	10	151.5	4.24	86.7	4.28	42.8
			30	141.3	4.20	89.0	4.30	37.0
HT	Polished	800	30	251.3	4.28	140.0	4.30	44.3
	Nonpolished	800	30	273.9	4.27	134.6	4.30	50.9

a) $\Delta Q = (Q_{HF} - Q_M)/Q_M$

that films screen-printed on the polished substrate have a higher Q and a lower ΔQ value than those of the films screen-printed on the nonpolished substrate. It is known that the device loss at high frequency is mainly due to the conductor loss, dielectric loss, and radiation loss. In this study, the higher roughness of the substrate surface results in a higher ΔQ value (Fig. 4), while the free-surface roughness of the silver film does not have a significant effect on the ΔQ value. For the silver films prepared using the high-temperature silver paste, the Q values are higher than those of the films prepared using the low-curing-temperature MOD paste, because the former has a dense microstructure (Fig. 5), which leads to a higher electrical conductivity and lower conductor loss. However, the difference between the simulated and measured Q values for the films printed on the polished substrate is similar to that printed on the nonpolished substrate, which is the largest among the films studied. This is due to the fact that the glass in the film reacts with the substrate during the high-temperature firing process, which results in a perfect adherence as well as an irregular interface between the film and substrate, as shown in Fig. 4, and a higher dielectric loss.

5. Conclusions

The electrical properties of silver films, prepared using a low-curing-temperature MOD paste and a high-temperature silver paste screen-printed on polished and nonpolished alumina substrates, at microwave frequency were characterized in this study. On the basis of the results obtained, several conclusions could be made as follows:

a. The surface roughnesses of the fired films printed on nonpolished Al_2O_3 substrate are less than those printed on polished substrate owing to the lower surface energy of the former.
b. The calculated effective conductivities at 4.3 GHz are slightly less than the DC conductivities of the films.
c. The films prepared using the high-temperature silver paste have a higher effective conductivity ranging from 4.08×10^7 to 4.13×10^7 S/m, because the high-temperature firing leads to a better connectivity of the silver particles.
d. The results for the device with T-type resonator circuits indicate that the films screen-printed on the polished substrate have a higher Q and a lower ΔQ value than those of the films screen-printed on the nonpolished substrate.
e. For the silver films prepared using the high-temperature silver paste, the Q values are higher than those of the films prepared using the low-curing-temperature MOD paste, because the former has a dense microstructure that leads to a higher electrical conductivity and a lower conductor loss. However, the ΔQ values of the films prepared using the high-temperature silver paste are the largest among the films studied, owing to the interfacial reaction between the glass contained in the film and the substrate during the high-temperature firing process, which results in an irregular interface between the film and substrate and leads to a higher dielectric loss.

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