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Thermal conductivity measurements with galvanic metallization lines on porosified LTCC applying the 3-Omega technique

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Abstract

The reduction of the thermal conductivity of low temperature co-fired ceramics (LTCC) caused by a porosification process originally developed for the local modification of the permittivity is determined by the 3ω measurement technique. Therefore, metallization lines of small width down to 20 µm are applied by galvanic pulse plating onto the roughened LTCC surface. By this deposition technology it is possible, to cover the pores without any depth penetration. Compared to the 'as fired' state, a reduction in thermal conductivity of up to 70% is measured independent of the LTCC type.

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Keywords: LTCC; porosification; 3-Omega; electro deposition; silver

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1. Introduction

Low temperature co-fired ceramics (LTCC) feature a moderate dielectric dissipation factor compared to organic materials resulting in benefits, such as a decreased electric field expansion in combination with minimized radiation losses. For advanced radar sensors, the porosification of the glass-ceramic substrate is targeted to provide areas of low permittivity, so that radiating elements such as patch antennas can be directly applied onto the same surface. Therefore, air is surface-near embedded by a wet chemical etch process [1].

It is desirable for the LTCC to feature a high thermal conductivity in order to transfer heat away from the circuitry [2, 3]. Typically, LTCC feature thermal conductivities up to ten times higher than resin-based materials [2].

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However, the embedded air with its very poor thermal conductivity of 0.026 $Wm^{-1}K^{-1}$ reduces the total thermal conductivity of the LTCC substrate [4].

In this paper the reduction of thermal conductivity of different commercially available LTCC materials DuPont 951, Heraeus CT 702, CT 708 and CT 802 by the porosification process will be investigated using the 3ω method. For comparison, the organic polyimide DuPont Kapton MT MD is investigated.

2. The 3ω method

The 3ω method is a non-static or dynamic technique for measuring the thermal conductivity. It facilitates a metal strip-line as electrical heating and sensing element with a width in the micrometer range. There are various adaptations of this technique reported in standard literature [5]. The approach used in this study is based on that of Cahill which measures the thermal conductivity of bulk materials at low frequencies. This method works under the assumption of a semi-infinite substrate and with the adaptation for very small metal strip-lines. Basically an alternating current with the angular modulation frequency ω is applied to the strip-line which features a certain electrical resistance R. The Joule heating results in a static temperature gradient and a thermal diffusion wave at a frequency 2ω penetrating into the substrate [5, 6]. The exact solution for the resulting temperature oscillation within the material caused by an infinitely narrow line heat source is given by Carslaw and Jaeger [6]. The resulting very low voltage component 3ω is proportional to the amplitude of the temperature increase in the heated metallic strip line and can be detected by a lock-in amplifier [5].

3. Experimental details

On both, the original LTCC samples including the Kapton MT MD as well as on the porosified LTCC, two-pad 3ω structures were applied by different metallization techniques. The nominal strip-line widths varied from 3 µm to 50 µm with a constant length l of 10 mm.

The original LTCC samples were polished with 2000 SiC abrasive paper. The structures were fabricated by standard lithography using the lift-off technique with AZ 5214E (MicroChemicals) photoresist. The metal consisted of 300 nm of gold on 20 nm of titanium deposited by e-beam evaporation.

Further LTCC samples were prepared by a treatment in phosphoric acid with a concentration of 50 m.-% for 8 h at 100°C. After the porosification, a seed layer with 10 nm of titanium and a top layer consisting of 50 nm silver were deposited by DC magnetron sputtering. Subsequent to a lithographical step using the galvanic resist AZ 125nXT (MicroChemicals), silver coatings were produced from a weakly alkaline cyanide-free silver electrolyte applying the pulsed current technique. The depositions were performed at ambient temperature using a stainless steel sheet as an anode under moderate electrolyte agitation. Prior to the electro deposition, the substrates were degreased using a tartrate-based solution, rinsed with distilled water, activated (two steps activation) with 10% tartic acid and 10% amido sulphonic acid and rinsed again with distilled water. A computer controlled pulse reverse power supply system (Plating Electronic pe86) was used for the plating experiments. The applied average current density was varied in the range of 0.4 to 1.33 A/dm^2. The deposition time was calculated aiming for a 5 µm thin silver layer. After stripping the resist, the seed layer was removed by silver etch.

The resulting widths of the micro strips were measured with an optical microscope Axio Imager (Carl Zeiss Microscopy).

The used 3ω measurement setup facilitated a SR8030 – DSP lock-in amplifier (Stanford Research Sytem) with a frequency range between 1 mHz und 102.4 kHz. The used 3ω interface (microconsult, tribotec electronic) switches three shunts with 0.5, 5 and 50 Ω.

The measured 3ω voltage was interpreted according to [6] where the slope of the voltages real part versus ln(ω) of the thermal oscillation ΔV_3 is used to determine the thermal conductivity Λ. The power per length P_m coupled into the original samples was 0.5 W/m while for the porosified samples it varied up to 1.5 W/m. The measurements were conducted at 25°C. The resulting data points, as shown exemplarily for DuPont 951 in Fig. 1 (b), consist of one measurement each. The error bars visualize the samples covariance for a level of significance of 95% calculated by the Gaussian propagation of uncertainty. The measurement means were calculated using the Eq. 1. The slope of resistance over temperature is expressed by ΔR.

$$\Lambda\left(P_m,l,R,\Delta V_3,\Delta R\right)=\frac{\left(\sqrt{P_m\cdot l\cdot R}\right)^3}{4\cdot\pi\cdot l\cdot R^2\cdot\Delta V_3}\cdot\Delta R \qquad (1)$$

The thermal conductivity results were found to depend linearly on the strip line width, which is caused by the differences in electrical resistance between the lines [7]. This dependency was utilized to make the original and porosified samples comparable. The least squares method of Levenberg-Marquardt was used in order to approximate the data points of the original samples. Consequently, the thermal conductivity value at a theoretically infinite strip-line width was approached by the intersection of the fit line with the ordinate. Due to increased variations of the porosified results, the according intersection was determined by an averaged shift parallel to the original fit line.

4. Results and discussion

In Fig. 1 a it can be seen, that the pulsed plating technique is able to bridge the gaps, which result from the surface-near porosification openings. Thus, strip-lines down to 20 µm width could be manufactured on the rough surface.

The different thermal conductivities between the untreated and porosified DuPont 951 substrate at varying strip-line widths are shown in Fig. 1 b. The increase of the deviations with rising strip-line width is especially visible at the porosified samples of all investigated LTCC and rises with decreasing sample width. This behavior can be attributed to the limits of the utilized measurement method according to [6] as well as to the approximately 15 times larger strip height compared to the untreated samples. With increasing heater thickness and width, the frequency region where the vanishing heater-thickness approximation is in accordance with the finite-heater solution diminishes [8].

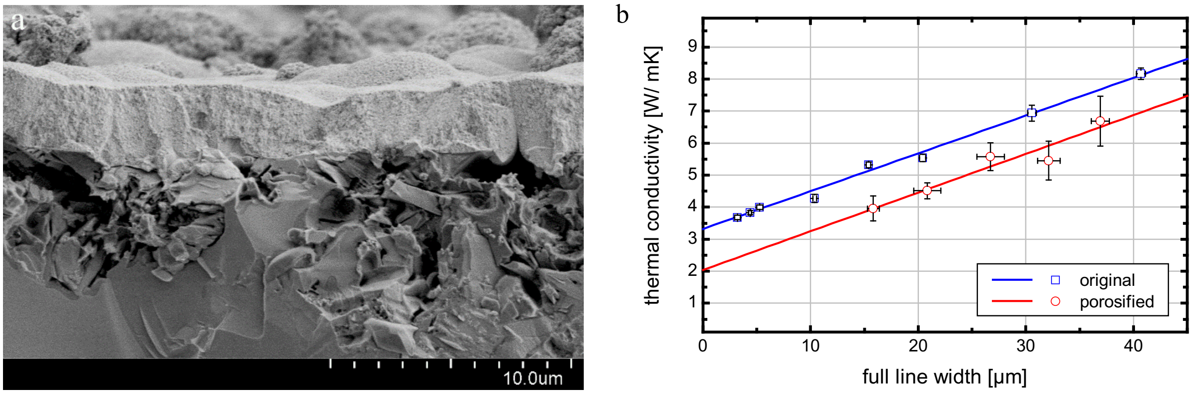

Fig. 1. (a) Cross-sectional view on a pulse-plated silver metallization on top of a porosified LTCC; (b) difference of the measured thermal conductivity between the original and porosified DuPont 951 sample as a function of the line width. The bars display the deviation on a 95% confidence interval.

The summarized results of all substrates are shown in Fig. 2 a. First and foremost, the deviation of the values for the LTCC Heraeus CT 702, CT 708 and CT 802 with respect to those given in the datasheet is apparent. The differences can be most probably attributed to a different measurement technique, which is not reported. In contrast, the results gained from DuPont 951 and Kapton MT MD show an excellent agreement [9-13].

Depending on the degree of porosification, the thermal conductivity decreases substantially. The results presented here, show a maximum reduction of 70% at a mass reduction of 2.6% in the surface-near region (Figs. 2 a and b).

The degree of porosification resulting from acid concentration, treatment time and temperature strongly depends on the chemical composition of the LTCC. Thus, Fig. 2 b displays the relative thermal conductivity as a function of the corresponding relative mass reduction. The data show a nonlinear dependency which indicates a typical logarithmic mixing rule, which is often applied to ceramics [2].

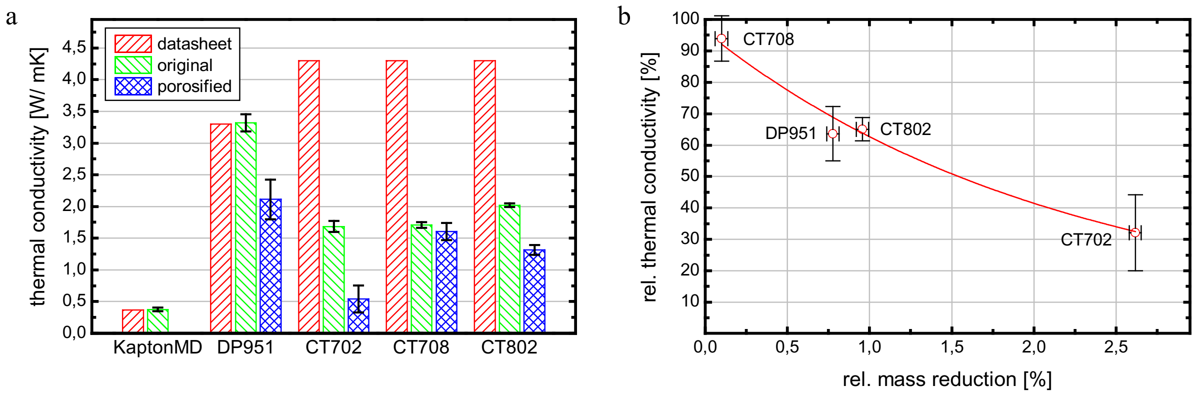

Fig. 2. (a) Thermal conductivity of Kapton and different LTCC substrates in comparison to the corresponding data sheet values; (b) relative thermal conductivity as a function of relative mass reduction. The inserted graph serves as guide to the eye. The bars display the deviation on a 95% confidence interval.

5. Conclusions

The thermal conductivity of low temperature co-fired ceramics (LTCC) is a key parameter for their implementation as advanced printed circuit board material. The reduction of the thermal conductivity caused by the relative permittivity decreasing porosification was determined by a 3ω measurement technique.

Metallization strips of small width down to 20 µm were applied by galvanic pulse plating onto the rough surface of porosified LTCC substrates. The maximum reduction, compared to their 'as fired' state, was determined to be up to 70%. Furthermore, a discrepancy between the original thermal conductivity values of the datasheets on three of five investigated samples was found. The conductivity reduction depends on the mass reduction and showed a nonlinear characteristics, which indicates a logarithmic mixing rule being typical for ceramics.

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