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Low-temperature thick-film dielectrics and resistors for metal substrates

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Abstract

In this work, a set of low-temperature thick-film dielectrics consisting of two high-lead low-temperature glasses, stabilised by various amounts of alumina filler, is characterised on alumina and aluminium metal, as a function of firing temperature. Corresponding resistors based on the same glasses as the dielectrics, but with RuO_2 as a conductive phase, were studied on the dielectrics. The purpose of these materials is to enable deposition of thick-film electronics onto substrates such as glass and metals (steel, aluminium, brass, titanium), which cannot be exposed to the standard high-temperature 850 °C thick-film firing cycle. Satisfactory insulating properties were obtained, and the properties of the resulting resistors are promising, but managing thermal mismatch was found to be a major issue regarding the mechanical integrity.

Keywords: Electrical properties; Thermal expansion; Glass; Al_2O_3; Sensors

1. Introduction

Thick-film technology applied to piezoresistive force or pressure sensing typically uses alumina as a substrate material because it is the standard for thick-film technology.[1] However, alumina is not optimal for piezoresistive sensing applications, as its elastic modulus is high and its strength rather low. Additionally, alumina is brittle and therefore ill suited to harsh environments and its heat dissipation capabilities are limited. Aluminium or aluminium alloys offer advantages in applications such as high power electronics or high-range load cells, due to their excellent thermal dissipation, mechanical sturdiness, easy packaging and adjustable thermal expansion coefficient α (Al–Si and Al–SiC systems).[2,3] However, the high temperatures associated with commercial thick-film processing (850 °C) are not compatible with aluminium owing to its low melting point: an appropriate low-temperature thick-film system (dielectrics, resistors and conductors) is therefore necessary.

In this work, we endeavour to investigate a novel low-temperature thick-film dielectric + resistor systems. In order to avoid interdiffusion problems between the dielectrics and resistors, both are based on the same glasses, differing only by the nature of the filler: Al_2O_3 for the dielectrics and RuO_2 for the resistors. We present the results of the properties of the dielectrics (adhesion, mechanical integrity, dielectric strength, dielectric constant and loss) and the resistors (value, temperature coefficient of resistance—TCR), on alumina and aluminium substrates.

2. Experimental

In this work, seven thick-film dielectric materials based on two lead borosilicate glasses (V6 and V8) were evaluated, with different glass compositions, filler loadings and processing temperatures. Glass V6 is 75%PbO (Aldrich, 99.9%) + 10% B_2O_3 (Aldrich. 99.99%) + 15% SiO_2 (Sihelco, Sikron), with 2% Al_2O_3 (Alfa Aesar, 99.99%) added. Glass V8 is 85% PbO + 10%B_2O_3 + 5% SiO_2, also with 2% Al_2O_3 (all compositions by mass). Both glasses were melted three times and the resulting frit was subsequently ball milled to ca. 1–5 μm. Dielectrics were prepared by adding various alumina powder concentrations (10–40 vol.%, 1 μm grain size) to the glass powders and an organic vehicle, and the resulting mix was homogenised with a three-roll mill. The alumina filler serves to improve the dimensional stability of the dielectrics. A commercial low-firing conductor composition (ESL 590G, silver with glass frit) was chosen as top con-

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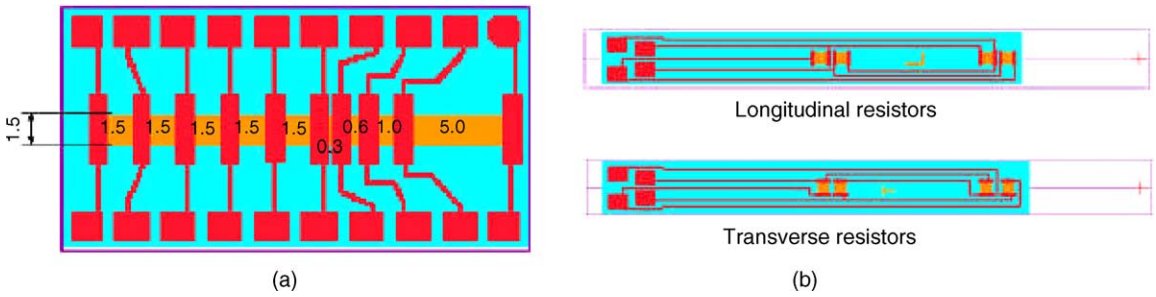

Fig. 1. Layout of the test sample for measurement of TCR (a) and GF (b).

ductor for characterisation of the dielectric, and as resistor termination material.

A single thick-film resistor compositions, based on the V8 glass, was prepared in the same way as the dielectrics, except that an usual conductive oxide filler for thick-film resistors,[4] RuO_2 (Johnson Matthey, 99.9%, 400 nm grain size, 8% volume), was used instead of Al_2O_3.

Samples were produced on two substrate materials: 96% pure alumina (Kyocera, Japan, A-476) as standard thick-film substrate and EN AW 6060 aluminium alloy (AlMgSi0.5) pieces. For each sample, two layers (40 μm) of dielectric were first screen-printed and fired, in order to guarantee good insulation, followed by the conductor and resistor. Firing was carried out in a lamp furnace. In all cases, the firing cycle started with a dwell at 370 °C for organic burn-out, followed by rapid (10 °C/min) heating to the peak temperature, remaining there for 20 min. Finally, the samples were cooled down at 20 °C/min. Firing temperatures were 525–600 °C for dielectrics based on V6 glass, 450–525 °C for dielectrics based on V8 and 450 °C for the resistor. The conductor was fired at 500 °C or at the same temperature as the dielectric, whichever was lower.

Samples were produced for three measurements: dielectric breakdown strength, sheet resistance and TCR, and piezoresistive gauge factor of the resistor.

Samples for breakdown strength measurement consisted of 2 mm diameter conductor dots screen-printed onto the final

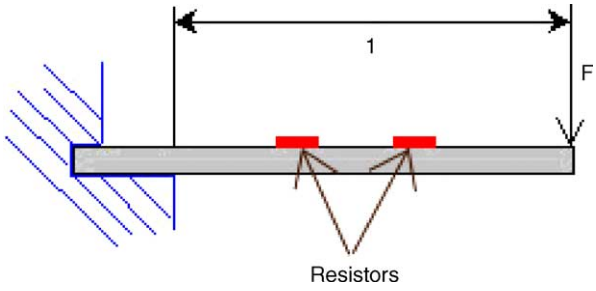

Fig. 2. Mounting of the beam for GF measurement.

dielectric layer itself screen-printed on aluminium substrates. Electric breakdown tests were carried out by applying dc and ac voltages with a ramp of 100 V/s to maximum voltage (5 kV dc or 4 kV ac) or breakdown. Breakdown was considered to have occurred when the current exceeded 0.1 mA dc or 1 mA ac. These samples allow measuring the dielectric constant and loss factor too.

For measuring sheet resistance and TCR (1.5 mm wide resistors of several lengths), and the piezoresistive gauge factor (GF, Wheatstone measurement bridge on cantilever beam, 1 mm × 1 mm resistors) two sample types were produced and depicted in Fig. 1.

Sheet resistance and TCR were measured between 30 and 100 °C using the samples depicted in Fig. 1a. GF, defined as the ratio between relative resistor change and mechani-

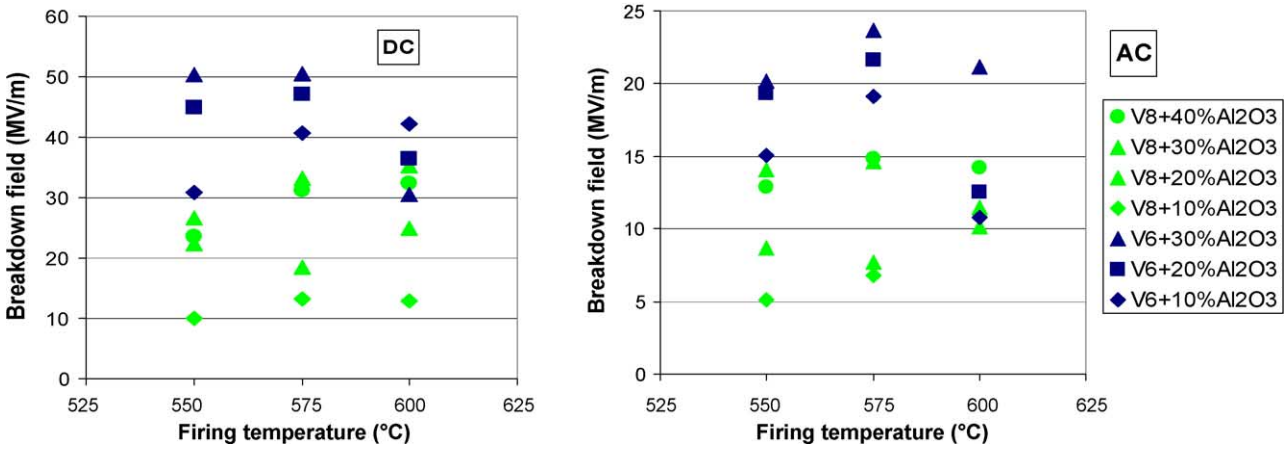

Fig. 3. Electric strength of dielectrics.

cal strain, was determined at RT (23–26 °C) by suspending weights at the end of the test cantilevers, depicted in Fig. 2.

3. Results and discussion

3.1. Dielectric strength

The results are represented in Fig. 3. We can notice that, for both ac and dc voltages, dielectrics based on V6 glass have a higher resistance to breakdown than those based on V8 glass. As the V8-based samples become more "liquid" during conductor firing than the V6-based ones due to the lower melting temperature of V8, we suppose this low electric breakdown resistance is due to diffusion of the conductor into the dielectric. This is confirmed by the fact that breakdown properties also improve with higher alumina powder content. In the case of V6 + 30% Al_2O_3 dielectric, we obtain very satisfactory dielectric insulation properties.

3.2. Dielectric constant and losses

The dielectric constant ε and the dissipation factor D of the dielectrics with 30% Al_2O_3 are given in Table 1. V8 tends to give a lower dielectric constant and higher losses. Further work is needed to more precisely evaluate the respective contributions of glass and filler.

3.3. Resistor value and TCR

TCR of both resistor sets measured as a function of underlying dielectric are summarised in Table 2. It must be mentioned that these resistive compositions are not optimised for a given TCR value.

On the alumina reference substrate, unreliable results were observed for the samples with dielectrics filled with low Al_2O_3 content (V6 + 10% Al_2O_3 and V8 + 10–30% Al_2O_3).

Table 1
Electrical characteristics at 10 kHz of dielectrics loaded with 30% Al_2O_3

Dielectrics	Dielectric constant, ε	Dissipation factor, D
V6 + 30% Al_2O_3	10.7	0.018
V8 + 30% Al_2O_3	4.3	0.035

Table 2
TCR (ppm/K) of resistors on various substrates and dielectrics

	Alumina	Aluminium
Alumina	−210 ± 30	–
V6 + 10%Al_2O_3	−400 ± 190[a]	−20 ± 14
V6 + 20%Al_2O_3	−240 ± 50	−25 ± 14
V6 + 30%Al_2O_3	−220 ± 30	–
V8 + 10%Al_2O_3	−1900 ± 1300[a]	+14 ± 9
V8 + 20%Al_2O_3	−2000 ± 1200[a]	−5 ± 9
V8 + 30%Al_2O_3	−2400 ± 2100[a]	−30 ± 14
V8 + 40%Al_2O_3	−200 ± 70	−110 ± 30

[a] Unreliable results ascribed to cracking.

This is ascribed to cracking due to tensile thermal stresses arising upon cooling after firing. Theses stresses are caused by the higher values of α of the glasses compared to alumina. The α values of V6 and V8 are 9 and 11 ppm/K, respectively,[5] compared to ca. 7 ppm/K for alumina, but this problem is mitigated by the alumina filler. This accounts for the fact that less problems are encountered at higher filler loadings and for V6 as opposed to V8.

When not affected by cracking, resistor values (10 kW) and TCR of resistors directly on alumina are very close to their values on the dielectrics independently of the dielectric composition, which is expected from the fact that the compositions of the resistor and dielectrics glass are identical (on V8 + Al_2O_3) or very similar (V6 + Al_2O_3), and that the dielectric filler material is identical to that of the substrate. One must also mention the low firing temperature (450 °C) of the resistor, which limits interdiffusion with dielectric. The firing temperature of the dielectric has no noticeable effect on the resistor properties, due to the absence of important chemical reactions within the dielectrics.

On aluminium, the TCR values are positively shifted by ca. 200 ppm/K compared to on alumina. On dielectrics based on V6, no noticeable dependence of TCR on filler loading is observed. This is not the case for V8-based ones, where higher filler loadings give somewhat lower TCR values. In contrast to alumina, dielectrics on aluminium are under strong compressive stresses, which is higher for V6 than for V8 and for higher filler contents. This stress caused cracking for V6 + Al_2O_3.

3.4. Gauge factor measurement

The results are depicted on the Fig. 4. Not all combinations could be measured, due to the abovementioned cracking problems, which are much more acute for this measurement, where the sample is subjected to mechanical stress. This is why no samples could be measured on alumina, except directly on the substrate.

On Al, the resistor gauge factors are quite low (GF_L = 3–4), which lies in the range of previous results on similar resistors

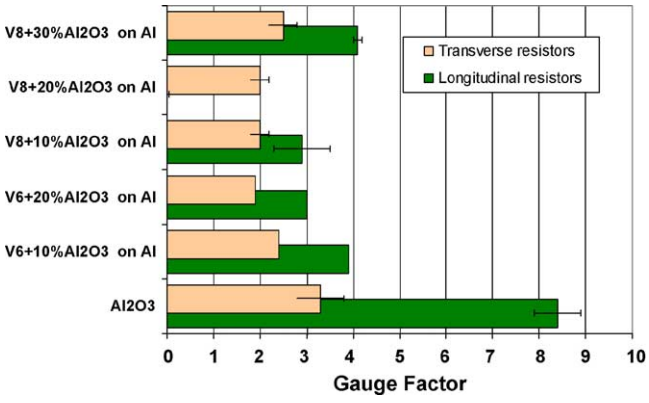

Fig. 4. Transverse and longitudinal GF according to combinations dielectrics/substrate.

based on V6.[6] The results directly on alumina are suspect, due to the abovementioned potential cracking problems. In fact, this sample was found to be damaged (probably cracked) after the gauge factor measurements.

3.5. Comparison of TCR and GF on aluminium and alumina

For a given resistor, the underlying substrate influences TCR in two ways: through chemical interaction and through thermal expansion. Chemical interaction may modify the resistor material and hence TCR, and cannot be ruled out, even with a separating dielectric.

On two substrates with different values of α, a thermomechanical shift of TCR is expected through the gauge factors[7,8]:

$$\Delta\mathrm{TCR} = (\mathrm{GF_L} + \mathrm{GF_T}) \times \Delta\alpha$$

ΔTCR : expected difference of TCR between alumina and Al
$\mathrm{GF_{L,T}}$: longitudinal and transverse gauge factor
$\Delta\alpha$: difference of substrate thermal expansion coefficients

A positive shift of TCR on aluminium versus alumina is therefore expected, as aluminium has a much higher value of α: 23 ppm/K versus 7 ppm/K. From our gauge factor measurements, we get: $\mathrm{GF_L} + \mathrm{GF_T} \cong 6$, which gives us an expected shift of ca. +100 ppm/K. This value agrees well with the shift observed on V8 + 40% Al_2O_3. On the other dielectrics, a further shift of TCR is observed, which is probably chemical in nature. This issue will be clarified in further experiments.

4. Conclusion

The studied experimental thick-film compositions constitute the first steps towards a low-temperature thick-film materials system suitable for deposition on heat-sensitive substrates such as steel, glass, titanium and aluminium alloys. Good electrical insulation and first resistive and piezoresistive properties have been demonstrated.

From a processing point of view, V6-based dielectrics and dielectrics with higher alumina loading are more difficult to obtain crack-free on aluminium, as they have a lower α and a higher stress relaxation temperature, which gives rise to considerably higher thermal stresses. The V8-based dielectrics are better suited for application onto aluminium.

Future work will therefore concentrate on achieving a good thermal match between dielectric and substrate. This can be accomplished by altering the substrate (for instance using Al–Si or Al–SiC composite instead of Al) or incorporating different fillers into the dielectrics. For substrates with very low (alumina, Si) or high α (Cu, Al), other fillers such as SiO_2 or CaF_2 will be explored.

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