

Dielectric Performance of a High Purity HTCC Alumina at High Temperatures - A Comparison Study with other Polycrystalline Alumina

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Abstract

A very high purity (99.99+%) high temperature co-fired ceramic (HTCC) alumina has recently become commercially available. The raw material of this HTCC alumina is very different from conventional HTCC alumina, and more importantly there is no glass additive in this alumina material for co-firing processing. Previously, selected HTCC and LTCC (low temperature co-fired ceramic) alumina materials were evaluated at high temperatures as dielectric and compared to a regularly sintered 96% polycrystalline alumina (96% Al_2O_3), where 96% alumina was used as the benchmark. A prototype packaging system based on regular 96% alumina with Au thick-film metallization successfully facilitated long term testing of high temperature silicon carbide (SiC) electronic devices for over 10,000 hours at 500°C. In order to evaluate this new high purity HTCC alumina for possible high temperature packaging applications, the dielectric properties of this HTCC alumina substrate were measured and compared with those of 96% alumina and a previously tested LTCC alumina from room temperature to 550°C at frequencies of 120 Hz, 1 KHz, 10 KHz, 100 KHz, and 1 MHz. A parallel-plate capacitive device with dielectric of the HTCC alumina and precious metal electrodes were used for measurements of the dielectric constant and dielectric loss of the co-fired alumina material in the temperature and frequency ranges. The capacitance and AC parallel conductance of the capacitive device were directly measured by an AC impedance meter, and the dielectric constant and parallel AC conductivity of the dielectric were calculated from the capacitance and conductance measurement results. The temperature and frequency dependent dielectric constant, AC conductivity, and dissipation factor of the HTCC alumina substrate are presented and compared to those of 96% alumina and a selected LTCC alumina. Other technical advantages of this new co-fired material for possible high packaging applications are also discussed.

Keywords: Packaging, high temperature, HTCC, alumina.

I. Introduction

High temperature co-fired ceramic (HTCC) alumina usually includes several percent of glass constituents to enable co-firing with conductors at a temperature below the regular sintering temperature of alumina. Recently, a very high purity (99.99+%) high temperature co-fired ceramic (HTCC) alumina has become commercially available [1]. The raw material of this HTCC alumina is based on nano alumina powders, so the raw material process is very different from conventional HTCC alumina, and more importantly there is no glass added in this alumina material designed for co-firing processing. Previously, selected HTCC and LTCC (low temperature co-fired ceramic) alumina materials were evaluated as dielectric for packaging applications at high temperatures, and compared to a regularly sintered 96% polycrystalline alumina (96% Al_2O_3), where 96% alumina was used as the benchmark [2], since a prototype packaging system [3] based on regular 96% alumina with Au thick-film

metallization [4] successfully facilitated long term testing of high temperature silicon carbide (SiC) electronic devices for over 10,000 hours at 500°C [5]. However, these prototype packages were not fabricated via an integrated commercial co-fire process, which is more suitable for large scale commercial production in comparison with those prototype packages integrated in a research laboratory. In order to evaluate the dielectric performance of this high purity high temperature co-fired ceramic alumina material for possible applications in high temperature packaging, both dielectric constant and AC conductivity of the material were measured from room temperature to 550°C at frequencies of 120 Hz, 1KHz, 10KHz, 100 KHz, and 1MHz. In addition to the dielectric properties of the material, other advantages of this material for high temperature packaging are also discussed.

II. Experimental Details

An after-fire HTCC 99.99+% alumina substrate acquired from the material manufacturer was used for this work [1]. The substrate is light white color and slightly transparent. A capacitive device for measuring temperature dependent dielectric properties of HTCC alumina substrate was fabricated on a 5.8 cm x 5.9 cm x 0.33 mm (2.27 in. x 2.32 in. x 13 mil) HTCC (as the dielectric) with thin-film platinum (Pt) metallization (4000Å, used as the two electrodes) on both sides of the substrate. The substrate was prepared by the manufacturer. Multi layers of the green tape materials were laminated together with pressure of 21 MPa at 70°C, then were fired at 1550°C (in this case without any conductor) for two hours. The surface roughness of the substrates was ~ 0.38 μm (15 micro-inch) (rms). The electrode/metallization area was 5.9 cm x 5.9 cm (2.004 in. x 2.004 in.) and located at the center of the substrate. Au wires with 10 mil diameter were mounted on the Pt electrodes (metallization) areas to electrically connect the capacitive device with measuring instruments via 1 m long coaxial cables. The capacitive device was thermally soaked in a box oven in which the temperature was controlled to an accuracy of ±2°C during the experiment. Prior to data acquisition, the capacitive devices were first heated in 550°C air ambient for 24 hours to make measurements be more precisely repeatable. The AC impedance of the alumina capacitive devices was measured by an AC impedance meter (HP 4192A and Keithley 3322). Small signal (1 V rms) sinusoidal voltage at frequencies $f = 120$ Hz, 1 kHz, 10 kHz, 100 kHz, and 1 MHz was applied to these capacitive devices. The impedance meter was calibrated (short and open) with 1 m long coaxial cable to compensate the impedances of the cable. The AUTO mode of the impedance meter was used to select/verify that the RC parallel circuit model was suitable to the dielectric. The AC impedance of a capacitive device is usually composed of impedances of interconnectors (wires), metal/dielectric interfacial contact, and the dielectric material in series [6]. For a parallel plate capacitive device with high capacitance, at low frequencies, the interconnector (wires) impedance is much lower compared with that of the dielectric material. In case of devices with thin dielectric, such as capacitors based on thick-film and thin layer of co-fired-ceramic dielectric, the contact impedance can be comparable to that of dielectric [6,7]. However, since the dielectric material studied in this work is relatively thick and demonstrates reasonably high impedance in the temperature and frequency ranges, only the dielectric impedance is

considered for these capacitive devices. Electrically the alumina dielectric is modeled approximately as a parallel circuit composed of an ideal capacitor (without parasitic self-inductance and conductance) for polarization current and an ideal parallel resistor (without parasitic capacitance and self-inductance) for conduction and dielectric loss current. Both effective AC conductivity and effective dielectric constant (dielectric constant) are assumed as frequency and temperature dependent. Based on these discussions, the dielectric constant of the alumina substrate (the dielectric) is calculated from the capacitance measurement at frequency f , and effective AC conductivity of the dielectric is calculated from the admittance measurement. Using this linear parallel circuit model, the measured complex AC admittance, Y , at frequency f is a linear function of the relative dielectric constant, ϵ' , and AC effective volume conductivity, σ_{eff} [8]:

$$Y(T, f) = \sigma_{eff}(T, f)A/d + j2\pi f\epsilon_o\epsilon'(T, f)A/d \quad (1)$$

where ϵ_o is the vacuum dielectric constant, T is temperature, A is electrode area, and d is the parallel electrode separation distance (thickness of the Al_2O_3 dielectrics). Since $A^{1/2} \gg d$ (59 mm wide compared with 0.33 mm thick) the capacitive boundary effects at metallization edges can be approximately ignored. The tests were conducted in the temperature range $23^\circ C \leq T \leq 550^\circ C$, and frequencies at 120 Hz, 1 kHz, 10 kHz, 100 kHz, and 1 MHz in this work. $\sigma_{eff}(T, f=0)$ is the DC conductivity. Both the dielectric constant and the effective volume conductance of the dielectric (HTCC alumina or LTCC alumina in our case) can be directly calculated from the measured AC admittance through Equation 1. Typically, σ_{eff} decreases with temperature for good conductors such as metals, while it increases with temperature for semiconductors and good insulators such as ceramics. Ideally, for a packaging substrate material both dielectric constant and conductivity are ideally expected to be low and stable with respect to temperature and frequency so the packaging parasitic effects would be low and stable.

The dissipation factor D ($\tan \delta$) of an RC parallel circuit is defined as the ratio of the conductance to the reactance measuring the ratio of the in-phase current (with respect to the voltage source) to the current with a -90° phase shift (with respect to the voltage source):

$$D = \sigma_{eff} / 2\pi f \epsilon' \quad (2)$$

D also measures the ratio of the energy dissipated per cycle to the average energy stored in the capacitor and it is independent of the device (capacitor) dimensions. For an ideal dielectric material, the dissipation factor is expected to be low and stable with respect to temperature and frequency.

III. Results

The dielectric properties of regular 96% alumina substrate with yttrium oxide as firing agent are used as benchmarks to evaluate the high purity HTCC alumina material. The previously measured results of dielectric properties of 96% alumina at room temperature are in the range of those reported elsewhere [9,10]. The regular 96% alumina substrates were fired at $\sim 1700^\circ\text{C}$. The data of dielectric properties of 96% alumina were measured previously using the same method under the same conditions [11]. The dimensions of 96% alumina dielectric were 88.9 mm x 88.9 mm in. x 0.381 mm, and the Au electrode area was 63.5 mm x 63.5 mm at the center region of the substrate.

The dielectric constants of tested HTCC alumina substrate are shown in Figure 1a-e with those of 96% alumina for comparison as well as a previously tested LTCC alumina, between room temperature and 550°C at various frequencies (these figures are not in same scale). For HTCC alumina, the dielectric constant at room temperature is 10.5, at frequencies from 120 Hz to 100 kHz and it increases to 10.7 at 1 MHz, compared with 9.7 of 96% alumina at 120Hz and 9.9 at 1 MHz. The dielectric constant of the HTCC alumina increases slowly with temperature, and it increases more at lower frequencies. At 550°C , the dielectric constant of the HTCC alumina increases 40.6% to 14.7 at 120 Hz while that of previously tested 96% alumina substrate increases 173.6% to 26.54; at 1 kHz, the dielectric constant of HTCC alumina increases 18.2% to 12.4 while that of 96% alumina increases 80.4% to 17.5; at 10 kHz, the dielectric constant of HTCC alumina increases 10.1% to 11.51 while that of 96% alumina increases 30.9% to 12.7. At 100 kHz and 1 MHz, the dielectric constant of HTCC alumina increases 6.7% to 11.20 and 6.6% to 11.4, respectively, while that of 96% alumina increases 14.2% to 11.08 and 12.3% to 10.89, respectively. Overall, the dielectric constant of HTCC alumina is slightly higher than that of 96% alumina at temperatures below 300°C at all frequencies, but it relatively changes (increases) less with temperature at high temperatures compared with 96% alumina.

As previously reported, for LTCC alumina, the dielectric constant at room temperature is slightly lower compared with those of 96% alumina. It does not change significantly until 300°C , it increases rapidly with temperature above 300°C , especially at low frequencies. The dielectric constant of the LTCC alumina increases by a factor of 303 to 2181 at 120 Hz. But at 1 MHz, the dielectric constant only increases by a factor of 1.9 to 13.5 at 550°C .

The conductivity data of tested HTCC alumina substrates are shown in Figure 2a-e, with those of 96% alumina and selected LTCC alumina tested previously [2,11] for comparison, between room temperature and 550°C at various frequencies (these figures are not in same scale). For high purity HTCC alumina, the conductivity basically increases with temperature. At 120 Hz, the conductivity was not directly measurable at temperatures up to 150°C due to the limit of sensitivity ($0.0005 \mu\text{S}$) of the impedance meter, and it is assessed $< 6.4 \times 10^{-11} \text{ S/m}$ using the sensitivity of the conductance measurement of the impedance meter at 120Hz. The conductivity is $6.4 \times 10^{-11} \text{ S/m}$ at 200°C , and it increases with temperature and it is $1.90 \times 10^{-8} \text{ S/m}$ at 550°C while that of 96% alumina increases from $2.9 \times 10^{-11} \text{ S/m}$ at room temperature to $3.2 \times 10^{-7} \text{ S/m}$ at 550°C ; At 1 kHz, the conductivity was not directly measurable at temperatures up to 100°C , and it is assessed $< 6 \times 10^{-11} \text{ S/m}$ between room temperature and 100°C , and it is $6.4 \times 10^{-11} \text{ S/m}$ at 150°C , the conductivity increases with temperature and it is $6.4 \times 10^{-8} \text{ S/m}$ at 550°C while that of 96% alumina increases from $8.7 \times 10^{-10} \text{ S/m}$ at room temperature to $5.5 \times 10^{-7} \text{ S/m}$ at 550°C ; At 10kHz, the conductivity is assessed $< 6.4 \times 10^{-9} \text{ S/m}$ between room temperature and 150°C , the conductivity is $1.27 \times 10^{-9} \text{ S/m}$ at 200°C and it increases with temperature above 200°C and it is $2.22 \times 10^{-7} \text{ S/m}$ at 550°C while that of 96% alumina increases from $6.8 \times 10^{-9} \text{ S/m}$ at room temperature to $1.5 \times 10^{-6} \text{ S/m}$ at 550°C . At 100 kHz, the conductivity of the HTCC alumina is $9.6 \times 10^{-8} \text{ S/m}$ at room temperature and it increases with temperature very slowly with temperature to 1.1×10^{-7} at 200°C , and it is $1.15 \times 10^{-7} \text{ S/m}$ at 250°C and it increases with temperature to $8.4 \times 10^{-7} \text{ S/m}$ at 550°C while that of 96% alumina increases from $1.3 \times 10^{-7} \text{ S/m}$ at room temperature to $4.2 \times 10^{-6} \text{ S/m}$ at 550°C . At 1 MHz, the conductivity is $2.8 \times 10^{-6} \text{ S/m}$ at room temperature, it increases from $3.95 \times 10^{-6} \text{ S/m}$ at 200°C to $7.9 \times 10^{-6} \text{ S/m}$ at 550°C while that of 96% alumina increases from $2.0 \times 10^{-6} \text{ S/m}$ at 200°C to $1.6 \times 10^{-5} \text{ S/m}$ at 550°C . Overall, at frequencies up to 100 kHz the conductivity of HTCC alumina is lower than that of 96% alumina at all temperatures tested. At 1 MHz,

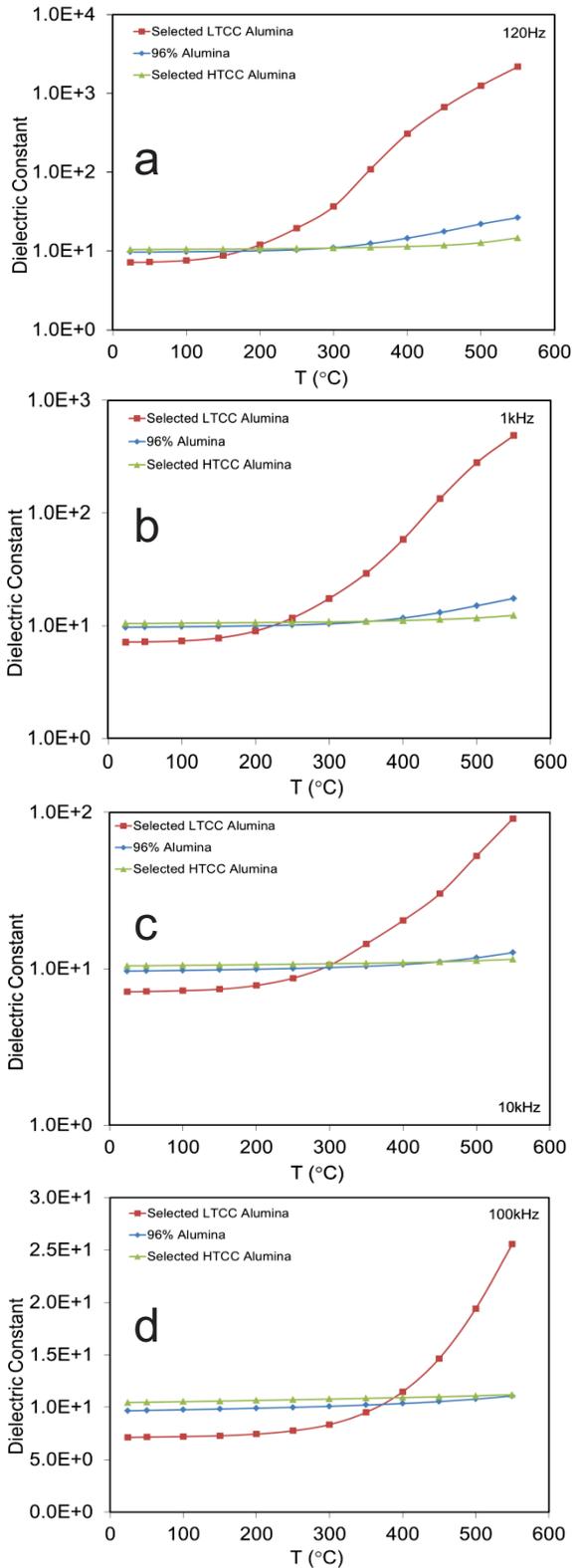
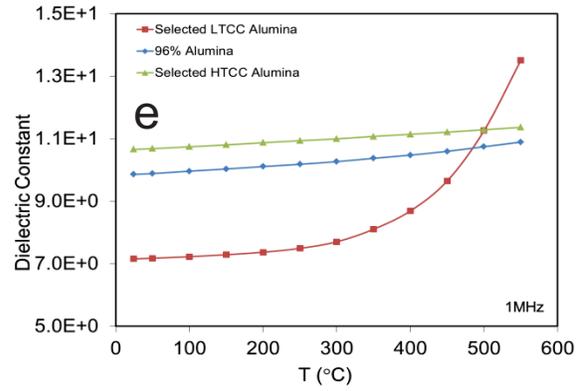


Figure 1: Dielectric constant of HTCC alumina substrates, compared with 96% polycrystalline Al_2O_3 , and LTCC alumina between room temperature and 550°C, a) at 120 Hz, b) 1 kHz, c) 10 kHz, d) 100 kHz, and e) 1 MHz. The data of 96% alumina and LTCC alumina are from Reference [11, 2].



the conductivity of HTCC is higher than that of 96% alumina at temperatures up to 350°C, but the conductivity of HTCC alumina is lower than that of 96% alumina between 400°C and 550°C where the conductivity is highest and most concerned for packaging parasitic effects for 500°C packaging applications. The conductivity of high purity HTCC alumina increases from 2.8×10^{-6} S/m at room temperature to 7.9×10^{-6} S/m at 550°C while that of 96% alumina increases from 7.7×10^{-7} S/m at room temperature to 1.6×10^{-5} S/m at 550°C.

As previously reported and discussed, for LTCC alumina, the conductivity at room temperature is close to or higher than that of 96% alumina, and it increases with frequency at all test temperatures. It increases from 3.9×10^{-10} S/m at 120 Hz to 5.5×10^{-7} S/m at 1 MHz at room temperature. The conductivity increases rapidly with temperature at all frequencies, especially, above 300°C. At 550°C, the conductivity of LTCC alumina ends in the order of $\sim 10^{-4}$ S/m at all frequencies.

The dissipation factor data of HTCC alumina are shown in Figure 3a and compared with those of 96% alumina and LTCC alumina in Figures 3b and 3c, respectively. Basically, dissipation factors of all three materials increase with temperature above 50°C (between room temperature and 50°C the conductivity of 96% alumina, at 1 kHz and 10 kHz, decreases with temperature so the dissipation factor of 96% alumina at these frequencies actually decreases slightly with temperature). Compared with 96% alumina, the dissipation factor of high purity HTCC alumina is always lower at frequencies up to 100 kHz. The highest point of dissipation factor is at 550°C and 120 Hz where it is about an order lower than that of 96% alumina. However, the dissipation factor of the HTCC alumina is higher than that of 96% alumina at 1 MHz at temperatures up to 400°C. At temperatures of and above 400°C the dissipation factor of HTCC alumina becomes lower compared with that of 96% alumina. In other words, the dissipation factor of HTCC alumina is always lower compared with that of 96% alumina at temperatures at and above 400°C.

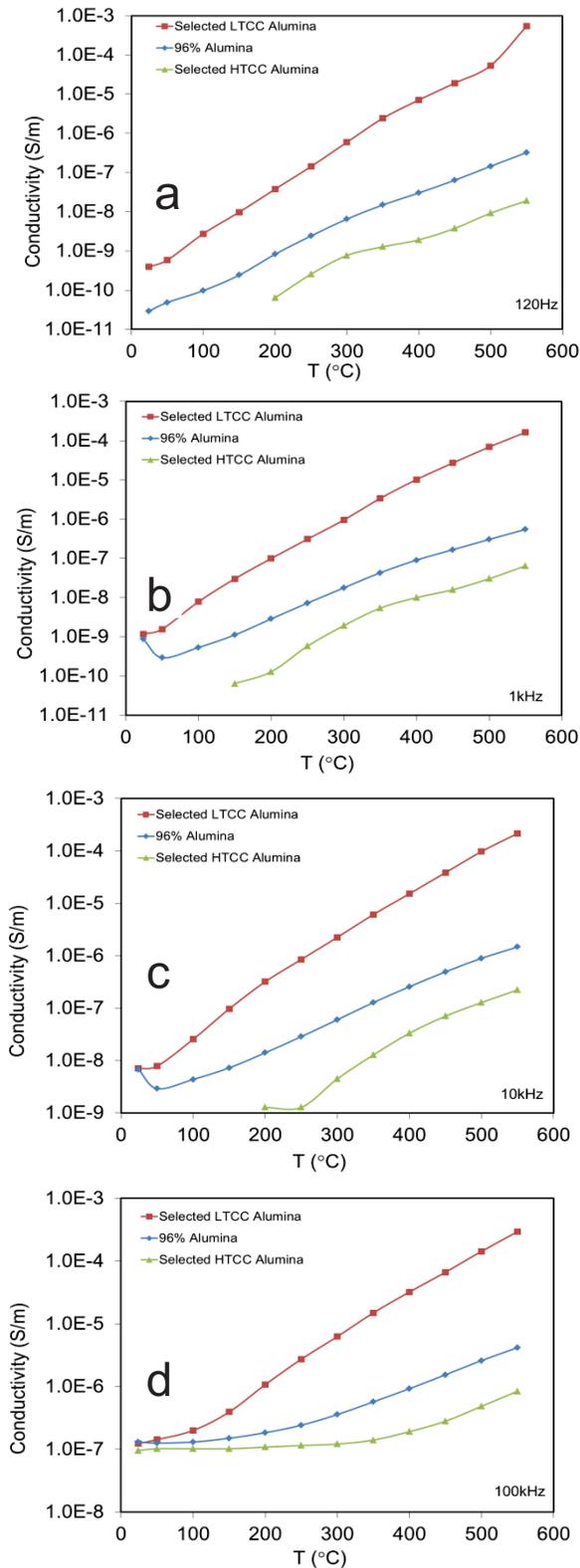
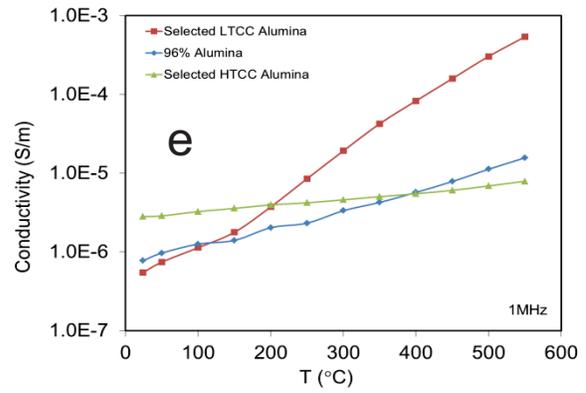


Figure 2: AC conductivity of HTCC alumina substrates, compared with 96% polycrystalline Al_2O_3 , and a selected LTCC alumina between room temperature and 550°C, a) at 120 Hz, b) 1 kHz, c) 10 kHz, d) 100 kHz, and e) 1 MHz. The data of 96% alumina and LTCC are from Reference [11, 2].



For LTCC alumina, at temperatures above 100°C the dissipation factor decreases monotonically with frequency while increases monotonically with temperature.

Parallel plate capacitive devices based on many polycrystalline ceramic alumina dielectrics, such as 96% alumina, for dielectric property test demonstrated slow DC (or low frequencies < 40 Hz) charging and discharging (with large time constants) at high temperatures. The slow charging/discharging indicated high DC (or low frequency < 40 Hz) capacitance which could not be quantitatively measured at such low frequencies by the experiment setup for this work. However, the capacitive device based on high purity HTCC alumina didn't show this type of unusual slow charging or discharging phenomena at temperatures up to 550°C.

IV. Discussion

Compared with 96% alumina, the dielectric performance of high purity HTCC alumina is better at elevated temperatures. Overall, the dielectric constant of the HTCC alumina is slightly higher at low frequencies compared with 96% alumina but it increases less with temperature. The AC conductivity (dielectric loss) of this material is lower than that of 96% alumina at frequencies up to 100 kHz in the entire test temperature range, and at 1 MHz the conductivity of high purity HTCC alumina is lower than that of 96% alumina at temperatures at and above 400°C where the parasitic parameters are most concerned for 500°C packaging systems. So the high purity HTCC alumina is suitable for high temperature packaging applications.

This high purity HTCC alumina material can be co-fired with compatible platinum conductor inks at temperatures ~1550°C in noble environment. Traditionally, metals with low CTE (thermal expansion coefficient) such as tungsten and molybdenum are conventionally used for co-firing with HTCC alumina. However, these conventional metal/alloys have relatively high electrical resistivity,

and may have oxidation issues for long term operation in high temperature air ambient. Platinum (Pt) thick-film inks designed for HTCC alumina substrates are more suitable for 500°C packaging applications. For these Pt inks, aluminum oxide is usually used as the binder for the inks designed for HTCC alumina substrates. As discussed previously [2], alumina-alumina bonds at metal/substrate interface are expected to be more thermodynamically stable in a wide temperature range compared with glass binder.

The temperature dependence of dielectric properties of the tested LTCC alumina is very similar to those of low purity alumina [12]. Compared with HTCC alumina, the major advantages of LTCC alumina include the much lower co-firing temperature and more choices of conductors for co-fire.

The observed slow charging / discharging process in many polycrystalline alumina materials at high temperatures may generate unwanted parasitic effects from an alumina packaging substrate. The high purity alumina, however, did not demonstrate this type of unusual slow charging / discharging at high temperatures, this may indicate that the DC dielectric constant (or very low frequencies) of high purity HTCC alumina may not rise dramatically at high temperatures. This provides a very important advantage as packaging substrate material for applications at high temperatures compared with other ceramic alumina.

The difference in dielectric performance of these alumina materials basically results from the different chemical compositions and electronic structures of these materials. The detailed physical mechanisms of the temperature dependence of dielectric properties of polycrystalline metal oxides with impurities can be complicated, and needs to be further explored.

There are many types of co-fired alumina materials with different chemical compositions and firing processes available on the market; these material systems, including HTCC alumina material systems, were all originally developed for packaging applications in conventional temperature environments. The test of these selected material systems at elevated temperature range is aimed to explore the possibility to expand the temperature range for application of these materials. Because of different compositions of these products, the dielectric properties of the high purity HTCC (and LTCC) alumina substrates reported and discussed in this article do not represent all of these types of co-fired alumina materials because of different chemical compositions and firing process. Further testing using more LTCC and HTCC alumina are planned

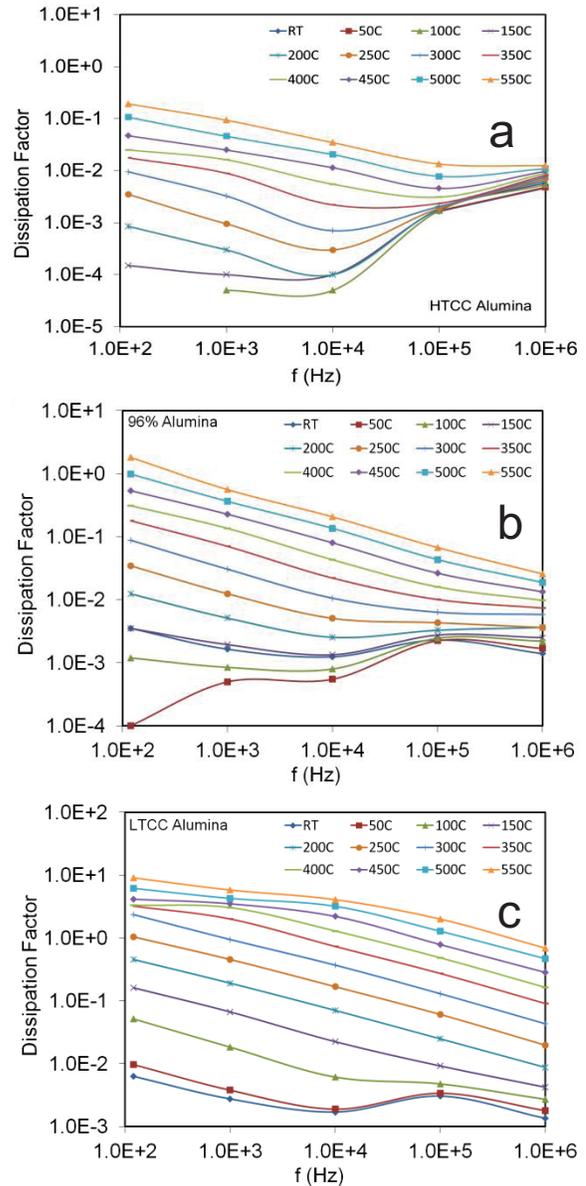


Figure 3: Dissipation factors of (a) HTCC, (b) 96% polycrystalline Al_2O_3 , and (c) LTCC alumina substrates compared, between room temperature and 550°C, at 120 Hz, 1 kHz, 10 kHz, 100 kHz, and 1 MHz. The data of 96% alumina is from Reference [11]. The data of LTCC alumina is from Reference [2]. These figures are not in same scale.

for future work in this direction to further down select the optimal substrate/metallization system for packaging technologies for 500°C SiC electronics.

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Reference

[1] http://www.electrosience.com/ceramic_tapes.html

[2] L.-Y. Chen, "Electrical Performance of Co-Fired Alumina Substrates at High Temperatures," in Proceedings of 2012 iMAPS International Conference on High Temperature Electronics, (HiTEC), May 8-10, Albuquerque, NM.

[3] L.-Y. Chen, D.J. Spry, and P.G. Neudeck, "Demonstration of 500°C AC Amplifier Based on SiC MESFET and Ceramic Packaging" in: Proc. 2006 iMAPS (International Microelectronics and Packaging Society, Washington, DC) International High Temperature Electronics Conference (2006 HiTEC), Santa Fe, NM, 2006, p. 240.

[4] J.S. Salmon, R.W. Johnson, and M. Palmer, (1998) "Thick Film Hybrid Packaging Techniques for 500°C Operation," in Trans. Fourth Int. High Temperature Electronics Conf. (HiTEC), June 15-19, Albuquerque, NM.

[5] P.G. Neudeck, S.L. Garverick, D.J. Spry, L.-Y. Chen, G.M. Beheim, M.J. Krasowski, and M. Mehregany, "Extreme Temperature 6H-SiC JFET Integrated Circuit Technology," Phys. Status Solidi A, 1-17, 2009.

[6] E. Miś, A. Dziejczak, and K. Nitsch, "Electrical Properties and Electrical Equivalent Models of Thick-film and LTCC Microcapacitors," Microelectronics International, vol.26 no 2 (2009), p.45-50.

[7] M. Unger, J. Fleig, M. Ahrens, W. Smetana, G. Radosavljevic; Permittivity and Conductivity of Various LTCC-Tapes at High Temperatures, Proc. 33rd Int. Spring Seminar on Electronics Technology, 2010, p.93-98.

[8] R. Bartnikas and R. M. Eichhorn (Edited), Engineering Dielectrics, Vol. IIA, Electrical Properties of Solid Insulating Materials: Molecular Structure and Electrical Behavior, ASTM Special Technical Publication 783, Philadelphia, PA, 1983.

[9] <http://www.coorstek.com/materials/ceramics/alumina.php>

[10] R.C. Buchanan, *Ceramic Materials for Electronics; Processing, Properties, and Applications* (2nd ed.), Marcel Dekker Inc., New York, 1991.

[11] L.-Y. Chen and G.W. Hunter, "Temperature Dependent Dielectric Properties of Polycrystalline 96% Al₂O₃ Substrate," Proceedings of Symposium G (Materials, Integration, and Packaging Issues for High-Frequency Devices II), 2004 MRS Fall Meeting, Boston, MA, November 2004.

[12] L.-Y. Chen, "Temperature Dependent Dielectric Properties of Polycrystalline Aluminum Oxide Substrates with Various Impurities," in Proceedings of 2007 8th International Conference on Electronics Packaging Technology, Shanghai, China, Aug. 14-17, 2007.