Temperature Dependent Dielectric Properties of Polycrystalline 96%Al_2O_3

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Abstract

Polycrystalline Al_2O_3 substrates have been proposed and tested for high temperature micro devices packaging intended for operation at temperatures up to 500°C. The dielectric properties of this material, including dielectric constant and effective volume conductivity, at elevated temperatures are of interest, especially for RF packaging applications. This article reports temperature dependent dielectric properties of polycrystalline 96% Al_2O_3 substrates from room temperature to 550°C measured by the AC impedance method at 120 Hz, 1 kHz, 10 kHz, 100 kHz, and 1 MHz. We observed negative temperature coefficients of volume electrical conductivity of 96% Al_2O_3 at 1 k, 10 k, and 100 kHz between room temperature and 50°C. The dielectric constant of the material increases significantly with temperature at frequencies below 10 kHz. The physical mechanisms of these dielectric behaviors of 96% Al_2O_3 at elevated temperatures are discussed.

Introduction

Polycrystalline aluminum oxide (Al_2O_3) is an important packaging substrate material which has low cost, high thermal stability, and high electrical resistivity. It has been extensively used in high reliability electronics packaging. Various thin-film and thick-film metallizations have been developed for Al_2O_3 substrates. In addition to conventional electronics packaging, Al_2O_3 has also been proposed for high temperature electronic devices and high temperature MEMS packaging [Salmon, 1998 and Chen 2002]. The packaging applications in elevated temperature environments, especially, those for high temperature radio frequency (RF) devices [Hunter, 2003, Spry and Neudeck, 2004] generated interests in the temperature dependence of dielectric constant and volume electrical conductivity of Al_2O_3 substrates. Limited data of temperature dependent dielectric properties of Al_2O_3 have been reported previously [Antula 1967]. In this article we report temperature and frequency dependent dielectric properties of polycrystalline 96% Al_2O_3 substrates in a temperature range from room temperature to 550°C measured by the AC impedance method at 120 Hz, 1 kHz, 10 kHz, 100 kHz, and 1 MHz.

Experimental Details

The room-temperature dielectric constant of polycrystalline 96% Al_2O_3 has been reported to be 9.5 between 1 k and 100 MHz, and room temperature dissipation factor has been reported to be 0.001 at 1 kHz, 0.0014 at 1 MHz, and 0.0004 at 100 MHz [MetWeb]. A 'capacitor' for measuring temperature dependent dielectric properties of 96%Al_2O_3 substrate was fabricated on a 3.5 in. x 3.5 in. x 0.15 mil 96%Al_2O_3 substrate (used as the dielectric) with gold (Au) thick-film metallization (used as the two electrodes) on both sides of the substrate. The surface roughness of the substrate was ~ 20 microinch (rms). The electrode/metallization area was 2.5 in. x 2.5 in. at the center region of the substrate. The Au thick-film material was screen-printed and cured at 850^{o}C in air. The rest of the Al_2O_3 surface areas (half inch wide surrounding the Au metallization areas) was passivated with thick-film glass in order to reduce surface electrical conductivity which can depend on environmental humidity, especially, at high temperatures. Au wires were bonded onto the Au electrodes (metallization) areas to electrically connect the 'capacitor' with measuring instruments via 1 m long coaxial cables. The 'capacitor' was thermally soaked in a box oven in which the temperature was controlled to an accuracy of 2^{o}C during the experiment. Prior to data acquisition, the 'capacitor' was first heated in 500^{o}C air ambient for 72 hours. The AC impedance of the Al_2O_3 'capacitor' was measured by an AC impedance meter. 1 V (rms) sinusoidal voltage at frequency *f* (or angular frequency $w = 2p \bullet f$, *f* = 120 Hz, 1 kHz, 10 kHz, 100 kHz, and 1 MHz) was applied to the 'capacitor'. The amplitude and the relative phase shift of the response current through the 'capacitor' were measured to construct a complex impedance (or admittance) at frequency f. The 'capacitor' was electrically modeled as a parallel circuit composed of an ideal capacitor (without parasitic self-inductance and conductance) for polarization current (with -90^{o} phase shift) and an ideal resistor (without parasitic capacitance and self-inductance) for conduction and dielectric loss current (with 0^{o} phase shift). The dielectric constant of Al_2O_3 is calculated from the capacitive imaginary part of the admittance at frequency *f*, and effective AC conductivity of Al_2O_3 is calculated from the real part of the admittance. With this linear parallel circuit model, the measured complex AC admittance, *Y*, at frequency *f* is a linear function of the relative dielectric constant, e', and AC effective volume conductivity, s_{eff}:

$$Y(T,w) = s_{eff}(T,w)A/d + jwe_o e'(T,w)A/d \qquad (1)$$

where e_o is vacuum dielectric constant, *T* is temperature, *A* is electrode area, and *d* is the parallel electrodes separation distance (thickness of the Al_2O_3 dielectrics). Since $A^{1/2} >> d$ the boundary effects at metallization edges can be approximately ignored. $23^{o}C \le T \le 550^{o}C$ in this experiment, and $f = w/2p$ = 120 Hz, 1 kHz, 10 kHz, 100 kHz, and 1 MHz depending on the setup of the impedance meter. Both DC leakage and the dielectric loss of the media in AC electric field are included in $s_{eff}(T,w)$. $s_{eff}(T,w=0)$ is the DC conductivity. Typically, s_{eff} decreases with temperature for good conductors such as metals, while it increases with temperature for good insulators such as ceramics. Both the dielectric constant and the effective volume conductance of the dielectric (96% Al_2O_3 in our case) can be directly calculated from the measured AC admittance through Equation 1.

One of the basic assumptions for measuring the dielectric constant and conductivity of a capacitor device using a parallel RC circuit model is that, in the range of the input voltage signal, the device is linear. Non-linearity may cause significant unexpected measurement error. The linearity of DC leakage of the capacitor device within ±1.5 V at various temperatures was examined by DC current – voltage (I-V) measurements.

Results and Discussion

Figure 1a shows the plots of the relative dielectric constant, $e'(T,w)$, of the polycrystalline 96%Al_2O_3 sample versus temperature at various frequencies. Below 200°C, e' changes very little with temperature at all five frequencies. Above 200°C, e' increases monotonically with the temperature, and the dielectric constant increases more rapidly with temperature at lower frequencies. At 120Hz, e' at 550°C is about 2.5 times of that at room temperature. In contrast, at 100 kHz and 1 MHz, with the temperature increase from room temperature to 500°C e' increases only 14.7% and 10.4%, respectively. Figure 1b shows frequency dependent e' at various temperatures. Temperature affects e' more at lower frequencies below 100 kHz. The inset of Figure 1b shows detailed variations of e' between 10 kHz and 1 MHz at various temperatures. At temperatures below 250°C, e' does not change much in the whole frequency range. This type of frequency dependence is typical. The higher measured dielectric constant at low frequencies is caused by displacement of quasi-free charge carriers which are thermally excited at elevated temperature [Bartnikas *et al.*, 1983]. These quasi-free charge carriers respond to external field but statistically do not get neutralized with charges from external sources, therefore, resulting in phenomenal polarization current in response to external fields. The quasi-free charge carriers excited/activated at high temperature are most likely from impurities/dopants used to control the grain sizes of Al_2O_3 (dielectric constant of 90% Al_2O_3 measured at elevated temperatures were found to be tremendously high [Chen et al., 2005]).

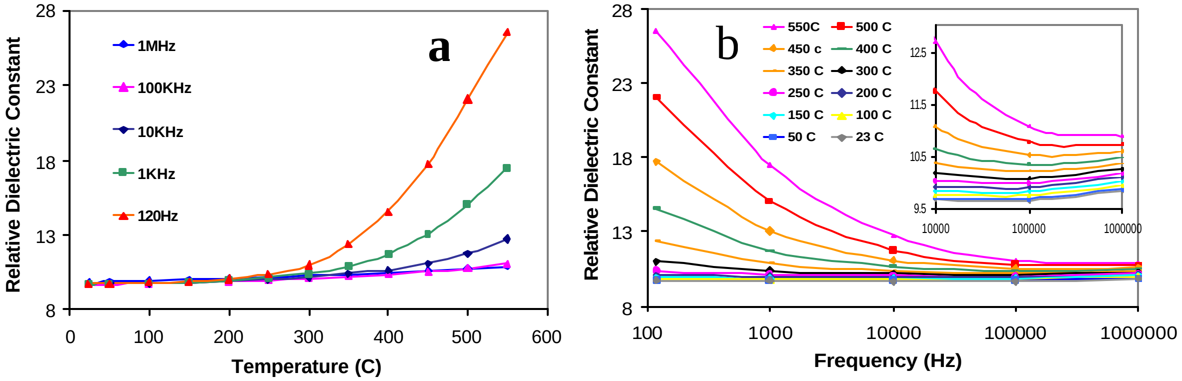

Figure 1: Temperature and frequency dependent dielectric constant of 96%Al_2O_3 substrate.

Figure 2a shows effective volume conductivity $s_{eff}(T,w)$ as a function of temperature at various frequencies. Between 23°C and 50°C the effective AC conductivity at 1 kHz, 10 kHz, and 100 kHz decreases slightly with temperature showing negative temperature coefficients. A negative temperature coefficient of the conductivity is not usual for insulator materials suggesting interesting thermal effect(s) on AC conductivity of 96% Al_2O_3 between 23 and 50°C. Above 100°C, volume conductivity increases monotonically and rapidly with temperature. The conductivity increases with frequency over the entire temperature range reflecting higher dielectric loss at higher frequency. Figure 2b shows the frequency dependence of $s_{eff}(T,w)$ at various temperatures. The negative temperature coefficients of $s_{eff}(T,w)$ at 1 k and 10 kHz cause the intersection of room temperature data with those of 50 and 100°C.

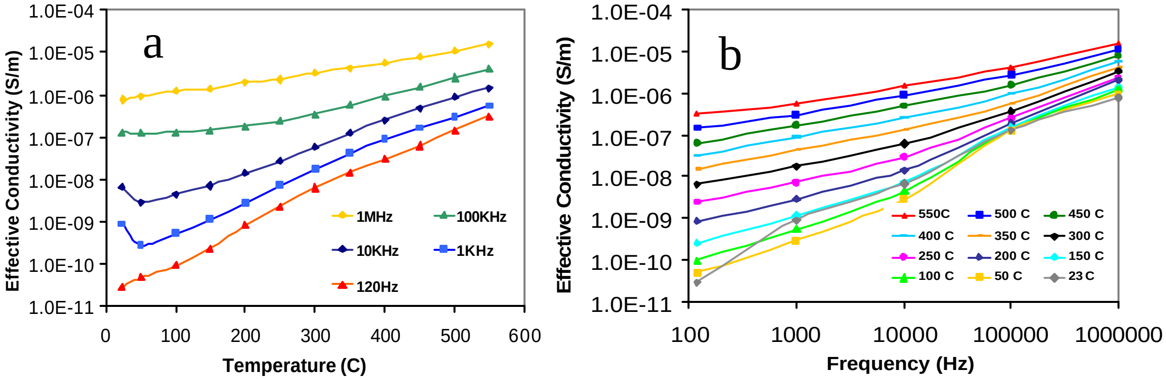

Figure 2: Effective volume conductivity of 96% polycrystalline Al_2O_3. a) Temperature dependences at various frequencies. b) Frequency dependences at various temperatures.

The Quality Factor Q of an RC parallel circuit is defined as the ratio of the reactance to the conductance (for parallel RC circuits) measuring the ratio of the current with a -90° phase shift (with respect to the voltage source) to the in-phase current:

$$Q = we' / s_{eff} \qquad (2)$$

Q also measures the ratio of average energy stored in the capacitor to the energy dissipated per cycle. It is independent of the device (capacitor) dimensions (assuming the boundary effects are negligible). Figure 3a shows measured Q as a function of temperature at all five frequencies. Between 23 and 50°C, the dielectric constant does not change much with temperature while the conductivity $s_{eff}(T,w)$ decreases so Q increases rapidly with temperature. Above 50°C, both e' and $s_{eff}(T,w)$ increase with temperature, but $s_{eff}(T,w)$ increases more rapidly than e' does so Q decreases with temperature. At 120 Hz, 1k Hz, and 10 kHz, Q peaks at ~ 50°C.

The quality factor Q at 500°C is ~1 at 120 Hz, 2.8 at 1 kHz, 7.4 at 10 kHz, 23.2 at 100 kHz, and 55.1 at 1 MHz, as shown in the inset of Figure 3a. Frequency dependent dissipation factor, which is defined as $1/Q$, at various temperatures is plotted in Figure 3b. The dissipation factor decreases rapidly with frequency in the entire temperature range.

The measured capacitances (or dielectric constant) are quite stable and reproducible after the initial 72 hours annealing at 500°C. The relative deviations of measurements of effective conductivity, before and after a thermal cycle, are 2% or less at room temperature, and 5% or less at 550°C for most data points (10% for a few data points).

The measured dielectric constant and conductivity at room temperature are in the range of previously published data for 96% polycrystalline Al_2O_3 [MetWeb].

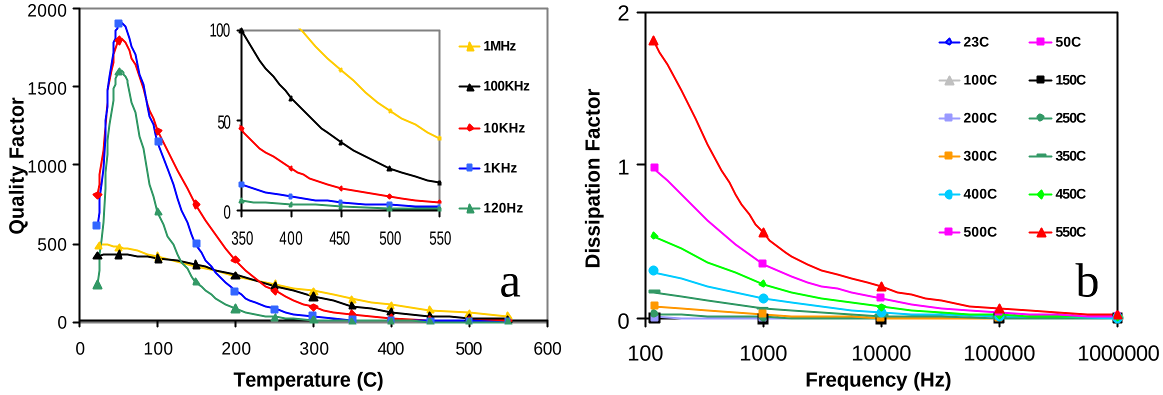

Figure 3: Quality Factor and Dissipation Factor of 96% Al_2O_3 substrate at various temperature and frequencies.

The measurements were conducted in a typical lab air ambient without strict gas ambient control. However, because a) the measured dielectric constant data are very repeatable within a few percent, and b) the room temperature data are consistent with previously published results, the contribution of gas ambient, including moisture, to the measured dielectric constant is believed to be negligible after the initial annealing.

Conclusions

The dielectric constant of 96% Al_2O_3 is ~9.5 at room temperature at all five frequencies. At 500°C, the dielectric constant is 2.27 times of that at room temperature at 120 Hz, 1.11 times of that at room temperature at 100 kHz, and 1.13 times of that at room temperature at 1 MHz. At 120 Hz, the effective volume conductivity of 96% Al_2O_3 increased by four orders of magnitude to $3.21 \bullet 10^{-7}$ S/m from 23°C to 550°C, but at 1 MHz, it increased by about one order to $1.57 \bullet 10^{-5}$ S/m from room temperature to 550°C. Negative temperature coefficients of AC volume conductivity at 1 kHz, 10 kHz, and 100 kHz were observed between room temperature and 50°C. The higher measured dielectric constant, at elevated temperatures and frequencies below 100 kHz, is attributed to the response of quasi-free charge carriers excited at elevated temperatures. 96%Al_2O_3 substrates show acceptable dielectric loss at high temperatures and high frequency, and the dielectric constant at 100 kHz and 1 MHz is stable between room temperature and 550°C. Therefore, 96%Al_2O_3 qualifies as a substrate material for high temperature RF packaging and passive applications.

Acknowledgements

The authors want to thank Drs. Lawrence G. Matus, Philip G. Neudeck and Carl W. Chang for proofreading the manuscript. This work is supported by the NASA Intelligent Propulsion System Fundamental Technologies (IPSFT) project at NASA Glenn Research Center and the NASA Electronic Parts and Packaging (NEPP) program.

References

Antula, J. (1967) Temperature Dependence of Dielectric Constant of Al2O3, Phys. Lett. A A 25(4): p308.

Bartnikas, R. and Eichhorn, R.M. (1983) Engineering Dielectrics, Vol.IIA, Electrical Properties of Solid Insulating Materials: Molecular Structure and Electrical Behavior, American Society for Testing and Materials (ASTM) Special Technical Publication (STP) 783, Philadelphia, PA.

Chen, L.-Y. *et al*. (2005) Dielectric Properties of Polycrystalline AlN and 90%Al_2O_3 Substrates at Elevated Temperatures, to be published.

Chen, L.-Y. and Lei, J.-F. (2002) Packaging of Harsh-Environment MEMS Devices, Chapter 23, CRC MEMS Handbook, CRC Press, Boca Raton, LA.

Hunter, G.W. (2003) Morphing, Self-Repairing Engines: A Vision for the Intelligent Engine of the Future, AIAA 2003-3045, AIAA/ICAS International Air & Space Symposium, July 14-17, Dayton, OH.

Http://www.MatWeb.com

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

Spry, D., Neudeck, P.G., Okojie, R., Chen, L.-Y., Beheim, G., Meredith, R., Ferrier, T., and Mueller, W. (2004) Electrical Operation of 6H-SiC MESFET at 500°C for 500 Hours in Air Ambient, Proceedings of 2004 IMAPS International Conference and Exhibition on High Temperature Electronics, May 17-20, Santa Fe, NM.

This paper was presented on 2004 MRS Fall Meeting, Nov. 20-Dec.3, 2004, Boston, MA. Copy Right MRS.