Non-linear Dielectric Testing at High AC Voltages using Waveforms and Harmonics

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Abstract: Complex impedance and non-linear response were measured for high-dielectric composite films at AC voltages using a waveform technique. With decreasing dielectric thickness, these materials operate at higher electric fields, which cause non-linear dielectric effects and lead to a time dependent dielectric-breakdown. The waveforms were Fourier transformed from time domain to frequency domain and analyzed at fundamental and higher order harmonic frequencies. It was found that the conventional epoxy resin laminates exhibit an impedance characteristic that is nearly independent on applied electric field, up to the material dielectric breakdown. When the electric field approaches near dielectric breakdown conditions, the second-order non-linear dielectric susceptibility decreases abruptly while the third harmonic response dominates non-linear dielectric behavior. In comparison, the impedance of dielectric materials made of organic resins and ferroelectric ceramics decreases continuously with increasing voltage. The non-linear dielectric effect is dominated by the third harmonic response that originates from a polarization reversal of the ferroelectric component. When the electric field approaches near dielectric breakdown conditions, again, the second-order non-linear dielectric susceptibility decreases abruptly.

Introduction

Dielectric composite materials with enhanced dielectric properties are being increasingly used in electronics as building blocks for functional circuits and as insulation materials for power distribution. These materials are complex chemical formulations consisting of a polymer resin, a dielectric modifier, and mechanical and aging stabilizers that are bound by interfacial coupling agents. They are typically more conductive and show a time dependent dielectric deterioration. The mechanism responsible for the dielectric-breakdown is not well understood. With decreasing dielectric thickness, these materials are operating at higher electric fields that can induce non-linear behavior.

Under a static higher electric field, E, the dielectric displacement, D, and the dielectric permittivity, ε, become field dependent: $D = P_0+\varepsilon_1E+\varepsilon_2E^2+\varepsilon_3E^3+\ldots$, where P_0 is the zero-field static polarization, ε_1 is the linear, field-independent part of ε and the ε_2, ε_3,.. ε_n coefficients represent the non-linear dielectric constant. For dielectrics consisting of freely rotating dipoles, P_0 and ε_2 are equal to zero, and the effect is measured by $\varepsilon_3 = \Delta\varepsilon/E^2$, where $\Delta\varepsilon = \varepsilon(E) - \varepsilon_1$ is the non-linear dielectric increment [1]. Measurements of the non-linear dielectric effect have been effectively used to study Langevin saturation, intermolecular interactions and cooperative molecular dynamics in dipolar fluids [2-4].

A convenient method of probing the field induced non-linear characteristics is to measure the harmonic components of the non-linear polarization under a sinusoidal (ac) electric field. The amplitude of harmonics reflects the magnitude of the non-linear polarization. The methodology was pioneered by Furukawa [5] to measure non-linear dielectricity in ferroelectric polymers. Non-linear alternating current response in composite materials in which the non-linear component is not inversion symmetric ($\varepsilon_2 \neq 0$) has been analyzed by Levy [6]. Several authors extended this technique recently and applied it to study fundamental molecular dynamics in dipolar fluids and molecular dielectrics [7, 8].

In this paper, we describe measurements of complex impedance and non-linear dielectric response by recording and analyzing AC waveforms at their fundamental frequency and at higher order harmonic frequencies. In particular, we are seeking to discover corelation between the dielectric breakdown conditions and non-linear dielectric response. We have examined and will demonstrate this effect for thin epoxy resin laminates and for novel composites with enhanced dielectric properties.

Experiment

Materials: We performed impedance measurements on several fiber-glass reinforced epoxy resin laminates, and on newly developed hybrid materials with enhanced dielectric properties. Here we present results obtained for a 50 μm thick fiber-glass epoxy resin laminate (FR4) and for a 40 μm thick high dielectric constant (high-k) composite made of an organic resin filled with ferroelectric ceramic sub-micron size particles. Both materials are representative of high capacitance layers that are being used in advanced electronic circuits for power-ground de-coupling [9]. The test pattern was made in accordance to the ASTM standard [10], with a diameter of the top electrode being 14.1 mm.

Measurement*: Figure 1 shows the block diagram of the measurement system that we described previously [11]. An IEEE 488.2 controlled function generator (Agilent 33250A) is used to source a sinusoidal voltage wave at a frequency f. The source voltage wave is amplified by an operational voltage amplifier (Trek Model 610C), and a high AC voltage is applied to the specimen. The specimen current is monitored using a standard reference resistor R as shown in Figure 1.

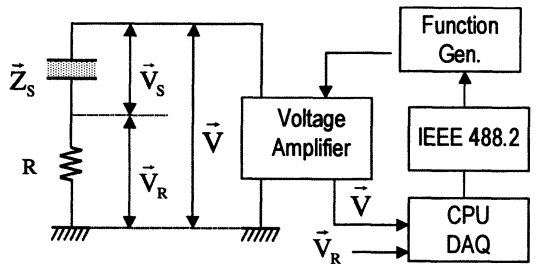

Figure 1: Measurement diagram.

Separate channels of the data acquisition (DAQ) card record the waveform of the monitoring signal (which is proportional by a gain factor to the output voltage, $\vec{V}$, from the voltage amplifier), and the voltage wave at the reference resistor, $\vec{V}_R$ (which is proportional to the specimen current). We used a two channel 12 bit analog-to-digital converter (NI PCI-6111) clocked at a frequency of 20 MHz, which has the capability to simultaneously sample both channels at frequencies of up to 100 kHz. The sampling was assumed coherent, i.e., containing an integer number of the sine-wave periods and an integer number, N, of the data points. In our experiments $N = 1000$ and $f = 50$ Hz. During measurements the specimen voltage was increased at rate of 5 V/s. The discrete quantization time and amplitude errors [12] were assumed to be within the manufacturer's specification for the NI-6111. The experimental uncertainty depends primarily on the tolerance of the reference resistor and its phase characteristic. The combined relative uncertainty was 0.5 % for the measured voltage wave amplitude and 1.5% for the phase. The complex impedance of the specimen, $\vec{Z}_S$, can be obtained from (1):

$$\vec{Z}_S = R\left(\frac{\vec{V}}{\vec{V}_R} - 1\right) \qquad (1)$$

In (1) $\vec{V}$ and $\vec{V}_R$ are phasor transforms from the time domain to the frequency domain representing complex quantities, each having an amplitude and phase [11]. The amplitude of the n-th order harmonic response, i_n, can be calculated from (2):

$$i_n = \frac{2}{N}\left[\left\{\sum_{l=1}^{N}\frac{v_R}{R}(l)\cos(2\pi nl/N)\right\}^2 + \left\{\sum_{l=1}^{N}\frac{v_R}{R}(l)\sin(2\pi nl/N)\right\}^2\right]^{1/2} \qquad (2)$$

where $\frac{v_R}{R}(l)$ represents an individual data point of the specimen current, $i(l)$, recorded at the time $t_l = (2\pi l)/(\omega N)$ and n (1, 2, 3,..) is the harmonic order. The specimen impedance and the higher order harmonics of the current wave response are presented as a function of the specimen electric field $E_S = |\vec{V}_S|/d$, where $\vec{V}_S = \vec{V} - \vec{V}_R$ and d is the specimen thickness. Thus for a sinusoidal electric field oscillating at an angular frequency $\omega = 2\pi f$, the non-linear dielectric response contains harmonic components at frequencies of $n\omega$.

Results and Discussion

The impedance and phase plots measured for FR4 as a function of the applied field are shown in Figure 2. The impedance magnitude remains at a level of about 22.3 MΩ up to 3.5×10^5 V/cm indicating a linear relation

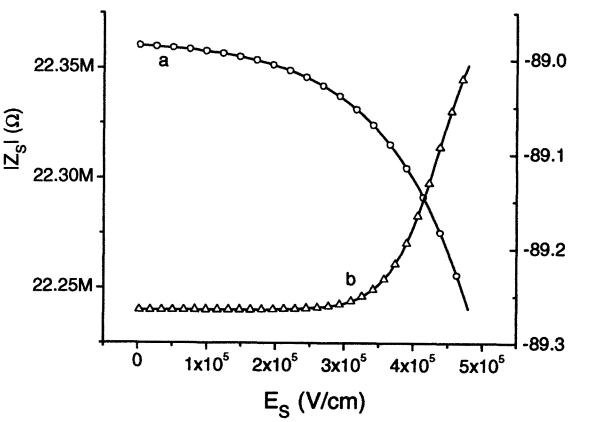

Figure 2: Impedance magnitude (a) and phase angle (b) of epoxy resin laminate ($\varepsilon_1 = 4$).

between the specimen voltage and the resulting current. This linear voltage range corresponds to the dielectric withstanding voltage, which is a functional performance indicator for the dielectric material. Above 3.5×10^5 V/cm, the impedance starts to decrease, which is accompanied by an increase of the phase factor. Such a change of phase indicates that above 3.5×10^5 V/cm the leakage current increases non-linearly with voltage and

*Certain equipment, instruments or materials are identified in this paper in order to adequately specify the experimental details. Such identification does not imply recommendation by the National Institute of Standards and Technology nor does it imply the materials are necessarily the best available for the purpose.

the character of the specimen changes from dielectric to more resistive. At higher fields impedance also become becomes time dependent.

In comparison to FR-4, the impedance of the high-k specimens decreases considerably with increasing voltage, which leads to an increased specimen current. The drop in impedance is accompanied by a significant change in phase (Figure 3). Under the applied electric field, the material undergoes an apparent transformation from dielectric to resistive. A comparison of the specimen equivalent impedance indicates that the linear voltage range is much narrower for the high-k organic

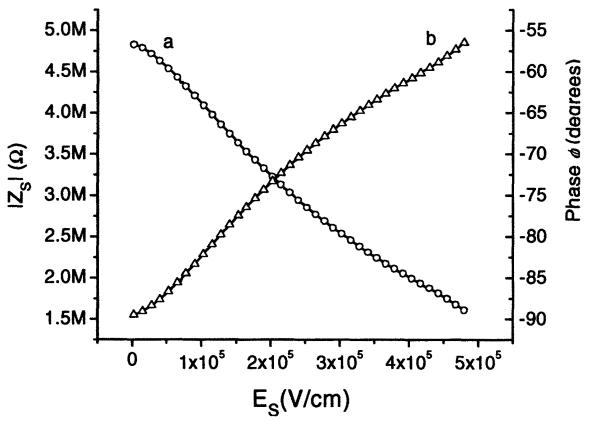

Figure 3: Impedance magnitude (a) and phase angle (b) of high dielectric constant composites ($\varepsilon_1 = 12$).

composites than that for the conventional dielectrics, suggesting that these materials would withstand only a small fraction of the conventionally measured dielectric withstanding voltage. At a sufficiently high power level such behavior may lead to thermal run-away due to an excess of dissipated current. This mechanism is fundamentally different than the dielectric breakdown that occurs in typical dielectric materials, where the dielectric failure is due primarily to voltage excited avalanche ionization.

To analyze the non-linear dielectric effects in more detail, we calculated the relative amplitude of the higher order harmonic currents, (i_n / i_1), using (2). Figures 4 and 5 illustrate these results for both types of materials, where the relative amplitude of the second harmonic, (i_2 / i_1), and the square root of the third harmonic, $(i_3 / i_1)^{1/2}$, are plotted as a function of the specimen voltage. Since the fundamental, i_1, is linearly dependent on voltage, the slope of the (i_2/i_1) plot is proportional to the relative magnitude of the second-order dielectric susceptibility while the slope of the $(i_3/i_1)^{1/2}$ plot reflects the relative magnitude of the third-order susceptibility. In the case of epoxy-resin specimens, the normalized

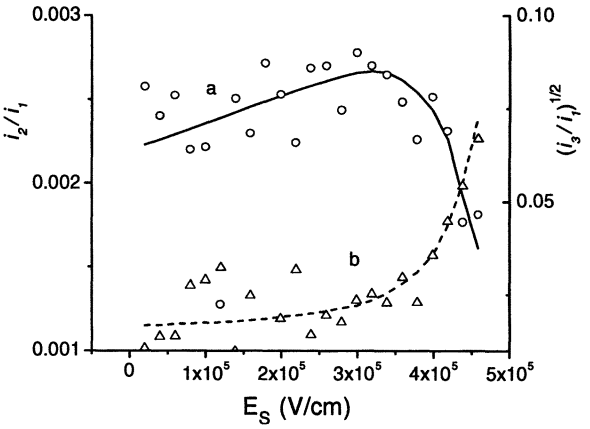

Figure 4: Normalized amplitude of the second harmonic (a) and third harmonic (b) current response for the epoxy resin dielectric material.

second harmonic current, (i_2/i_1), increases linearly with the specimen voltage, reaching a maximum at about 3×10^5 V/cm (Figure 4a). This second-order non-linear dielectric susceptibility, which originates from a residual static polarization, starts to decrease above 3×10^5 V/cm. Such decrease in (i_2/i_1) can be attributed to an electric field driven molecular rearrangement process where the material approaches near dielectric breakdown. In comparison, the $(i_3/i_1)^{1/2}$ plot (Figure 4b) changes slope at about 3.5×10^5 V/cm, again indicating a field induced molecular mobility, which results in an increased third-order dielectric susceptibility above 3×10^5 V/cm.

Figure 5 shows the corresponding amplitude of the second harmonic and the third harmonic currents obtained for the high-k dielectric material. The third-order susceptibility dominates the overall non-linear dielectric response. Such behavior can be attributed to the polarization reversal in the ferroelectric ceramics component, dispersed in the organic phase. Polarization reversal in ferroelectric materials usually generates odd order harmonics where the magnitude of the third harmonic is typically the largest. The change in slope of the $(i_3 / i_1)^{1/2}$ plot (Figure 5b) at about 1.5×10^5 V/cm indicates a change in the dielectric polarization mechanism of the ceramic filler rather than a dielectric softening of the organic phase. Initially large, the third-order susceptibility tensor saturates above 2.5×10^5 V/cm, due to ferroelectric saturation in the ceramic filler. Consequently, the overall character of the third-harmonic response is fundamentally different than that shown in Figure 4b for epoxy-resin specimens. Both the impedance characteristic (Figure 3) and the third-order non-linear dielectric response (Figure 5b) primarily reflect the dielectric properties of the ceramics rather than organic phase. However, in the background

of the third harmonic response the second harmonic is present (Figure 5a). The normalized second harmonic current, (i_2/i_1), decreases somewhat with the increasing electric field. The (i_n/i_1) drops rapidly at about $3.2x10^5$ V/cm, indicating an electric field driven

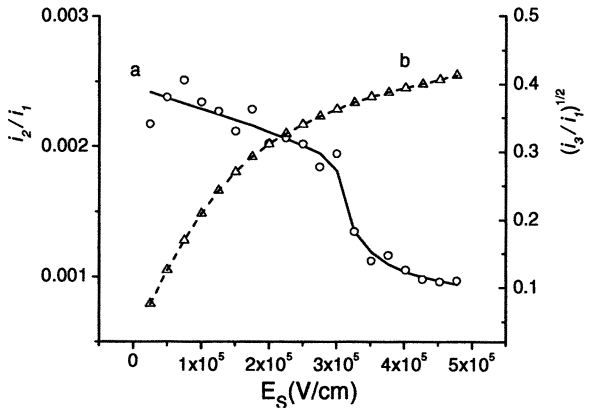

Figure 5: Normalized amplitude of the second harmonic (a) and third harmonic (b) current response for the high-k composites.

molecular rearrangement process where the material approaches near dielectric breakdown.

Non-linear dielectric effects can result from polarization reversal, rectifying barriers and material degradation near dielectric breakdown conditions. Such effects may appear at relatively low voltages in nano-sized composites and sub-micron thin dielectric films that are of interest to new electronic technologies. The presented procedure of recording and analyzing waveforms allows the evaluation of specific characteristics of dielectric materials that cannot be readily evaluated using conventional techniques.

Conclusion

Conventional dielectrics, such as epoxy-resin laminates exhibit a linear impedance characteristic nearly independent of electric field, up to the materials' breakdown conditions. In contrast, the impedance of dielectric hybrid materials with high dielectric constant is highly non-linear and decreases considerably with increasing electric field, making it difficult to determine mechanism of their dielectric breakdown. The results demonstrate, however, that the electric field induced dielectric deterioration mechanism can be inferred from the second-order and third-order susceptibility characteristics. The presented testing procedure represents a compatible extension of the existing standard test methods for evaluating dielectric breakdown, but is better suited for low impedance thin film materials and materials with a high-dielectric constant.

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