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**TIME DOMAIN RESPONSE OF ELECTRICAL CERAMICS-
MICRO TO MEGASECONDS**

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TIME DOMAIN RESPONSE OF ELECTRICAL CERAMICS MICRO TO MEGASECONDS

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ABSTRACT

The electrical properties of ceramics can be measured in either the time domain or in the frequency domain. But for electrically nonlinear ceramics such as varistors, time-domain measurements provide insights that are different and more relevant to material performance as well as being more physically incisive. This article focuses specifically on the electrical properties of ZnO varistors, but much of it is of relevance for other materials, in particular those materials with grain-boundary barriers and disordered ceramics or glasses. The interpretation of electrical measurements in the time domain is profoundly influenced by such practical matters as source impedance and waveform characteristics. Experimental results are presented for both high and low source impedance relative to that of a test varistor, and the difference in experimental difficulty and ease of interpretation is described. Time-domain measurements of capacitance and of the inductive response of varistors to large, fast electrical pulses are presented and their implications for varistor theory are given. Experimental evidence is given of short- and long-term memory in varistors. These memory phenomena are ascribed respectively to the life time of holes that become trapped in barriers and to polarization currents originating from deep electron traps. Polarization current measurements are presented for a wide range of time and temperature. The power-law time dependence and "universal" behavior of these currents is discussed. The exponent that describes the power law behavior is seen to change with temperature, and the change is interpreted as a double transition from diffusive to dispersive transport that originates with current from two different electron traps.

INTRODUCTION

Electrically based characterizations of ceramic materials can be done in either the time domain or in the frequency domain. When a material's electrical response is linear, the same information is obtained, at least in principal. Frequency-domain measurements are convenient and most often employed. But for electrically nonlinear ceramics such as varistors, time-domain measurements provide insights that are different and more directly relevant to material performance as well as more physically incisive. This article describes the time-domain measurements done on ZnO varistors. Varistor measurements are profoundly influenced by the test apparatus that is used, and this linkage between test apparatus and device response is discussed. Some measurement problems of the time domain are described. The understanding of varistors obtained from time domain measurements is presented. Comparisons of time- and frequency-domain results are given to illustrate similarities and differences.

LUMPED PARAMETER CIRCUIT DESCRIPTION

Figure 1 is a simplified circuit description of a time-domain test apparatus that includes a lumped parameter portrayal of a varistor. An approximately square electrical pulse with a width varying from about 1 μ s to 1 Ms is applied to the varistor from a supply that has a resistive output impedance denoted as R_s . The voltage applied to the varistor and the resulting current flow are measured and interpreted to obtain an electrical characterization of the varistor. Though almost any wave form can be used, interpretation is easiest with square pulses. For fast pulses, the current and voltage measurements require fast digitizers and parasitic reactance becomes a big

Figure 1. A simplified description of a varistor in a test circuit.

problem. For slower pulses, simple electrical meters suffice, but stability is a concern. The output impedance of the pulser (i.e., R_s) is ideally much higher or much lower than that of the varistor that is measured. But this condition is difficult to achieve because the impedance of a varistor changes with voltage.

Varistors are ceramic grain-boundary-barrier devices, and they have an electrical response that can be only roughly described by lumped parameters. The nonlinear resistance that is the essence of varistors is contributed by Schottky-like grain boundary barriers. Since this resistance changes with voltage by 12 orders of magnitude or more, it is denoted in Fig 1 as the variable resistor R_b . However, the change in R_b with voltage is not instantaneous; R_b evolves in time and introduces phase shifts into the electrical response. As is discussed below, these phase shifts can cause R_b to appear to be either a capacitor or an inductor. The Schottky barriers also exhibit an easily measured capacitance C_b that is contributed by barrier depletion layers of ionized donors. Although the varistor grains are highly conductive compared to the grain boundaries they contribute a small resistance R_g (e.g., 1 ohm cm) that can profoundly influence the electrical response in some measurements. In addition, there are electronic traps in varistors that contribute a capacitance C_t which is much larger than C_b . One conduction path represents the contributions of all electron traps in Fig. 1. But, in actuality, there are several different traps, and each trap should be represented by a parallel conduction path. The response of deep electronic traps can be very slow; so in a lumped parameter model C_t is associated with a large resistance R_t . However, the traps do not exhibit the exponential time response suggested by the lumped parameter description. Instead, they have a distinctly nonlinear – often termed a non-Debye – response to the applied voltage. In the strict sense, neither the trapping phenomena nor the nonlinear resistance of varistors can be described in terms of lumped parameter circuits or even within linear response theory. The lumped parameter description of Fig. 1 is nevertheless the jargon of electrical measurements and it is useful in the discussions of varistor measurements that follow.

FAST PULSE RESPONSE

The response of varistors to fast electrical pulses is a test of their performance as surge arresters that also reveals a lot about the fundamental mechanisms that control the electrical nonlinearity. (The focus here is on fundamental understanding rather than performance testing.) The pulse response of varistors gives clear insights into varistor capacitance, and it illustrates more directly than frequency domain measurements an inductive response of varistors that reveals the influence of electron holes on varistor barriers.

The response of a varistor to a fast electrical pulses from high- and low-impedance circuits is shown in Fig 2. The pulses are classified as current or voltage pulses when they are supplied by circuits with output impedances that are higher or lower, respectively, than the varistor under test. The pulses drive the varistor above its breakdown voltage at about 300 V, and because the applied pulses are essentially square (i.e., 50 ns rise time), it is easier to obtain qualitative and quantitative information.¹

Qualitative examination of the turn-on and turn-off responses as seen in the leading and trailing edges

of the pulses show voltage lagging current below breakdown, but leading current above

Figure 2. Varistor response to fast pulses from high- and low-impedance circuits.

breakdown. Hence, the electrical behavior is described as capacitive below breakdown and inductive above. A quantitative interpretation follows from linear circuit theory, and the values of capacitance obtained for voltages below breakdown are given in Table I.

TABLE I. Varistor capacitance from pulse response and ac admittance.

	Voltage Rise	Voltage Fall	Current Spike	1 MHz Admittance
C (nF)	1.5	1.5	1.8	1.8

The capacitance from voltage rise in Fig. 2(a) is obtained from $dV/dt = I/C$. The same value can be obtained from the RC time constant of the decay since it is known that the output impedance of the pulse generator $1.7 \text{ k } \Omega$ with the voltage off. The capacitance obtained from the current spike of Fig. 2(b) is found by integrating over the initial spike to obtain the stored charge and using $C = Q/V$. The value may be a little high because the integration includes a little over barrier current. However, the value is the same as that found from 1 MHz admittance measurements, which also might be a little high because traps may contribute even at high frequencies. The varistor capacitance varies greatly because the contributions of traps are frequency dependent, but the capacitance found from admittance measurements is a minimum value at about 1 MHz.

It is possible to obtain values for the varistor inductance that appears when the varistor is above its breakdown voltage (i.e., $\sim 300\text{V}$) by assuming linear circuit theory and analyzing the rate of approach to steady state of either the voltage overshoot or the current undershoot in Fig. 2. But such different values as $1 \text{ } \mu\text{H}$ and $50 \text{ } \mu\text{H}$ are found from Figs. 2(a) and 2(b), respectively. There are two reasons for this difference: First, these values do not represent real inductance. They only reveal voltage lead relative to current that is interpreted as inductance in the linear-circuit lumped-parameter model of reality. Second, linear response theory is not applicable because the electrical conduction in the barrier breakdown region is well described by the distinctly nonlinear empirical equation:

$$I = (V/V_0)^\alpha, \quad (1)$$

where α is a nonlinearity coefficient that is typically 20 to 60 and V_0 is a constant. A small change in voltage corresponds to a large change in current, and this difference explains the small voltage overshoot relative to current undershoot that is seen in Fig. 2. To a first approximation, Eq. 1 implies that current undershoot is greater than voltage overshoot by the factor α :

$$(I_s - I_i)/I_s = -\alpha (V_s - V_i)/V_s, \quad (2)$$

where the subscripts s and i refer to steady state and initial values.¹ Inductance values found from current undershoot and voltage overshoot measurements differ by this same factor of α .

Varistor performance testing relies on time-domain measurements, while varistor theory has relied largely upon ac immittance measurements. However, the time-domain results have many clear theoretical implications. The most accepted of the varistor theories attribute electrical breakdown to barrier collapse that occurs when electron holes are trapped in the grain boundaries.²⁻⁸ And, strong evidence of holes is given by the inductive response of varistors, which is clearly seen in the time response of Fig. 2. (That minority carriers in semiconductor junctions can contribute an inductive response has been recognized for a long time,⁹ and a clear explanation of the inductive response in ZnO varistors is given by Pike.³) Thus, while much elegant experimental and theoretical work has been done to verify the existence and explain the role of holes in varistor breakdown,^{3-5,8} strong evidence is easily found in time response measurements. Moreover, the time response measurements distinguish between different interpretations of the observed change in varistor capacitance with voltage. On one hand, a huge increase in capacitance just below breakdown has been interpreted as real capacitance (i.e., real energy storage) stemming from hole

accumulation in the barriers.² On the other hand, ac impedance data showing a capacitance that increases below breakdown before decreasing to negative values above barrier breakdown has been attributed to phase shifts that result from a modulation of the barrier height by electron and hole trapping.^{3,6,8} Below the barrier breakdown voltage, Figs. 2(a) and 2(b) show only a well behaved capacitance without the charge storage that a large increase in capacitance would produce, but an inductive response – some times called negative capacitance – is seen above breakdown. Thus, the time domain measurements confirm the explanation³ of the voltage dependence of varistor capacitance in terms of phase shifts produced by electrons and holes that respectively raise and lower the barriers when they are trapped.

INFLUENCE OF TEMPERATURE AND RESISTANCE

The interpretation of electrical measurements in the time domain is profoundly influenced by such practical matters as waveform characteristics and source impedance. The measurements are easiest to interpret when the source impedance is either high or low relative to the sample being tested. But such conditions can be difficult to achieve in measurements of electrically nonlinear materials because the sample impedance can change dramatically. Consequently, pulse response data has often been presented that is a mixture of such results as are shown in Fig. 2. Neither current nor voltage remain constant, and interpretations are less straight forward.

The source impedance has a strong influence on the decay time of voltage overshoot and on the rise time associated with undershoot. The recognition of this influence proved the key to understanding the inductive response of varistors and its temperature dependence. The current rise time is found to have a value of a few microseconds at room temperature and to change with temperature in a manner suggesting a curious negative activation energy.^{10,11} As seen in Fig. 3, the voltage overshoot exhibits a similar decay time (i.e., 2.5 μ s) at room temperature, but it has no significant temperature dependence. This difference is surprising and difficult to explain if the voltage overshoot and current undershoot are two aspects of the same hole trapping process. The explanation lies in the source resistance of the pulser, which is greatly different in voltage overshoot and current undershoot measurements. The influence of even small amounts of source resistance is shown in Fig. 4(a). And Fig. 4(b) is a mathematical simulation of experimental results that confirms source impedance as the origin of the difference.

Figure 3. The temperature dependence of of the time constants describing voltage overshoot (denoted x) and current undershoot (denoted o). Thermal activation energies of -7.2 and +0.4 meV are deduced.

The simulation assumes the empirical description of the nonlinear current-voltage characteristic,

$$I(V) = I_c(V/V_c)^\alpha, \quad (3)$$

where I_c and V_c are scaling constants and α is the nonlinearity coefficient. The varistor voltage V is reduced from the source voltage V_s by the drop across the series resistance R_s ,

$$V = V_s - IR_s. \quad (4)$$

For simplicity, it is assumed that the varistor has an exponential time response described by

$$dI(t)/dt = [I(V) - I(t)]/\tau, \quad (5)$$

where τ is a time constant associated with the generation and trapping of holes. (It has been shown that a time lag in the response to voltage change of either the barrier resistance or the barrier capacitance can explain the inductive characteristics of varistors.¹²⁾ For a reasonable choice of parameters (e.g., $I_c = 5$ A, $V_c = 200$ V, $\alpha = 25$, and $\tau = 7$ μ s), the equations can be numerically integrated for values of R_s that correspond to different values of pulser output impedance in the case in which a rectangular voltage pulse of 8 μ s duration is applied, and the results are shown in Fig. 4(b). The experimental and simulated results are in excellent agreement. Of course, the current spikes associated with the charging and discharging of the varistor capacitance were not simulated. Thus, even small amounts of source impedance have a large effect, and measurements of rise time are only accurate in the limit of zero source impedance.¹¹

The explanation of the temperature dependence of the current rise time follows from the understanding of the influence of the source resistance. It originates in the temperature dependence of the grain resistivity. The grain resistivity is about 0.5 Ω cm at room temperature, but it increases by an order of magnitude at liquid-nitrogen temperature due to the decrease in the carrier density. In the varistor used for Fig. 4(a), the grain resistivity contributes about 0.1 Ω at room temperature and about 2.4 Ω at 77 K. The grain resistivity reduces the barrier voltage in proportion to the current just as an external resistance does. (See Fig. 1.) Thus, a small change in the grain resistivity with temperature completely explains the temperature dependence of the current rise time. On the other hand, the grain resistivity has little effect on the voltage overshoot because it is negligible compared to the high source resistance.

PULSE MEMORY PHENOMENA

The inductive response of a varistor is either reduced or enhanced, depending upon the competing influences of a long- and a short-term varistor memory of preceding pulses. These phenomena are complex, transitory by nature, and most easily studied in the time domain. Figure 5 illustrates the complexity of these memory effects for voltage overshoot. The first pulse of Fig. 5 exhibits a voltage overshoot of 10%, which is less than the 13% overshoot seen in the second pulse. [Overshoot is roughly calculated as 100% $(V_{\max} - V_s)/V_s$ with V_s a steady-state voltage measured at 1.2 μ s.] Although the second pulse is delayed by only a few microseconds, it is an

Figure 4. The influence of circuit resistance on varistor current response to rectangular voltage pulses. Parts (a) and (b) respectively show actual and simulated results. The solid, long-, and short-dashed curves are for resistances of 3, 1, and 0.1 Ω , respectively.

Figure 5. The voltage overshoot of a varistor measured for current pulses with a 1.2- μ s duration and a 4- μ s separation. The solid curve is the initial response. The dotted curve was measured after a number of other pulses had been applied, and it is arbitrarily placed on the same time scale.

example of long-term memory because this enhancement of the second pulse can be seen even when it is delayed for several minutes. However, after several large current pulses have been applied, less overshoot is exhibited by the second pulse of such a pair, but only if it is applied within some tens of microseconds. Hence, this is a short-term memory phenomenon. The latter pair of pulses in Fig. 5 exhibit 17% and 12% overshoot for the first and second pulse, respectively.

The pulse memory phenomena are even more clearly disclosed by the current undershoot seen in a varistor's response to repeated pulses from a low-impedance (e.g., $\sim 1 \Omega$) source. Figure 6 shows the response of a varistor to double pulses of $6 \mu\text{s}$ duration and $50 \mu\text{s}$ separation. After long-term memory is suppressed by repeating the pulses several times at a rate of about once per second, the second pulse of a pair clearly exhibits much less current undershoot (i.e., 30% compared to 66%, gauging undershoot as $100\% (I_s - I_i)/I_s$ where I_s and I_i refer to steady-state and initial values).

Long-term memory is most clearly seen when the first pulse response is compared to the response of a subsequent pulse delayed by more than 10^{-4} s. (Short-term memory decays in about 10^{-4} s and long-term memory decays in about 10^3 s.) In Fig. 7, the first and tenth pulse of a sequence of pulses with $8 \mu\text{s}$ duration and 0.5 s separation are superimposed. In contrast to Fig. 6, the later pulse of Fig. 7 exhibits a greater undershoot than the first when the undershoot is measured as a percentage of the pulse amplitude. Note, however, that the initial value of the current is less for the later pulse (neglect the current spike) and the subsequent current rise is nearly identical for the two pulses. This result reveals that there are two components of the current with only one of these components behaving inductively. (In addition, the current spike reveals a capacitive component.) The later pulse exhibits a reduced noninductive component rather than an increased amount of inductive current. The undershoot is increased as a percentage of the pulse amplitude only because the inductive current represents an increased fraction of a diminished total.

The shorter-term memory of preceding pulses is attributed to the life time of the electron holes that become trapped in varistor barriers. Varistor breakdown occurs when trapped holes reduce the barrier height. But the drop in the barrier lags the applied voltage by the time required to generate and trap these holes. This time lag introduces a phase shift that is inductive in character. But once the hole traps are filled, the inductive character of any subsequent pulse response is diminished. (Actually, holes might immediately recombine with trapped electrons, and the extents to which they remain in trapped states is still unclear.) Thus, the shorter-term memory of preceding pulses is related to the lifetime of the holes trapped in the barriers. The longer-term memory of preceding

Figure 6. Current undershoot and short-term memory seen in the response of a varistor to double voltage pulses of $6\text{-}\mu\text{s}$ duration and $50\text{-}\mu\text{s}$ separation.

Figure 7. The response of a varistor to a sequence of voltage pulses with an $8\text{-}\mu\text{s}$ duration and a 0.5-s repetition rate. The first response (solid curve) and the tenth response (dashed curve) are superimposed.

pulses is attributed to a slow polarization-current discharge. As will be discussed below, these polarization currents can be large at short times, despite a slow-discharge classification.

POLARIZATION CURRENTS AND NON-DEBYE RESPONSE

Varistors display substantial polarization currents that are transient in nature and governed by distinctly non-Debye processes that decay slowly. Time domain measurements have provided the most definitive information on the slow decay of varistor polarization currents, and they are preferable to frequency domain measurements because of the higher information band pass they provide. As discussed above, polarization currents are significant at short times, and under breakdown conditions. But the focus here is on varistor response at low voltages, where the over-barrier currents can be neglected, and at times between 10^{-1} and 10^5 s, where depletion layer capacitance gives a negligible response. Under these conditions the polarization currents are predominant and time-domain measurements excel in revealing the nature of slow transients.^{13,14}

Figures 8 and 9 show charging and discharging currents for varistors at different voltages and temperatures. Figure 8 clearly shows that at room temperature about 10^3 s must elapse before polarization currents become negligible in comparison to steady-state conduction. At lower temperatures much longer times are required. Figure 9 shows the discharge currents that were measured after voltage was removed. Because there is no steady-current in this case, a current that decreases almost inversely with time is readily discerned. (A similar time dependence is seen if the steady-state current is subtracted in the results of Fig. 8.)

Because the polarization currents induced in varistors are large and decay slowly, it is tedious if not difficult to measure the steady-state current-voltage characteristics of varistors. As can be seen in Fig. 8, the approach to equilibrium is extremely slow at low voltages, and waiting a thousand seconds per data point is none too long at room temperature. At low temperatures, it can be virtually impossible to reach steady-state conditions. It also is worth noting that this slow approach to electrical equilibrium influences electrical properties measurements even at relatively high frequencies. Figure 10 shows hysteresis loops which illustrate the slow changes in capacitance and dissipation that are induced by the application of a voltage. As with measurements of I-V characteristics many hours are required to measure a steady-state capacitance as a function of voltage. This alone makes a Mott-Schottky analysis of the varistor capacitance difficult. But, there are other difficulties as well.¹⁵

The polarization currents are described reasonably well by the power law

Figure 8. Transient varistor currents induced by applying voltages that differ by factors of 2. Solid line: 295 K; dashed line 77 K.

Figure 9. Transient discharge currents induced in a varistor at 295 K by removing voltages that differ by factors of 2. The inset is an Arrhenius plot of the current measured at 0.1, 1, and 10 s after voltage removal.

$$I = I_0 e^{-E/kT/t^m}, \quad (6)$$

where E is a thermal activation energy and the exponent m is near unity. The inset of Fig. 9 suggests two traps with activation energies of 10 meV and 150 meV, and measurements at higher temperatures than are shown suggest an additional trap with a 660 meV activation energy.¹⁴ These activation energies are taken directly from the slopes of Arrhenius plots and are not highly accurate because differentiation of Eq. (6) gives

$$\partial \ln(I)/\partial(1/T) = -E/k - \ln(t) \partial m(T)/\partial(1/T), \quad (7)$$

and as is discussed below m is the slope of a plot of $\ln(I)$ versus $\ln(t)$, and it is not temperature independent. However, errors in the activation energies contributed by the temperature dependence of m are no more than 30%. The 660 meV energy can be attributed to the deep interface traps that give rise to the barrier because it is essentially identical to the measured activation energy of 680 meV energy for over-barrier conduction. The 150 meV activation energy is close to that of a bulk trap seen in DLTS measurements. The 10 meV activation energy is too low to be a trap depth, but it may be evidence of tunneling.^{14,16}

As can be seen in Fig. 11, the slope of $\ln(I)$ vs. $\ln(t)$ at eighteen different temperatures varies between a value that is equal to, but not in excess of, unity as an upper bound and a value of one half as a lower bound. At two temperatures (i.e., 110 and 330 K) the slope is close to one, and at two other temperatures (i.e., 230 and 450 K) m is equal or close to one half. As the temperature varies the value of m cycles back and forth between these bounds.

Jonscher¹⁷ has pointed out that the power law dependence of Eq. (6) is nearly universal in dielectrics and its validity for ZnO varistors has been confirmed for times varying over 12 orders of magnitude.¹³ Temperature variations in m are known in other materials, and changes in m between 0.5 and 1 have been reported, though not the cycling back and forth seen in varistors. Scher et al.¹⁸ have explained the transition from $m = 0.5$ to 1 in terms of the convergence of moments of the distribution of transit times. In their explanation, such behavior results as the limit of a statistical distribution. Hence, no particular process is assumed, and Jonscher's universality is implicitly contained. The transition from $m = 0.5$ to 1.0 as temperature drops is a change from diffusive to dispersive electrical transport. In varistors the nonmonotonic behavior of $m(T)$ can be interpreted as due to the resolution of polarization currents from two different traps (i.e., 660 and

Figure 10. Capacitance (solid curves) and dissipation (dashed curves) measured at 10 kHz versus applied voltage. (a) The voltage changed at 10^{-2} V/s. (b) The voltage changed at 10^{-4} V/s.

Figure 11. The slope of $\ln(I)$ plotted against $\ln(t)$. The curves are only guides to the eye.

150 meV), each of which make a current contribution that evolves in turn from diffusive to dispersive transport as the temperature is lowered.¹⁴

CONCLUSIONS

The time-domain characterization of electronic properties of materials offers unique and physically incisive insights, particularly for electrically nonlinear ceramics. Time-domain measurements on ZnO varistors disclose electronic transport of an exceptionally complex nature. Electrical currents in varistors are distinctly nonlinear in both their resistive and reactive components. And different types of nonlinearity appear at low and high applied voltages. At low voltages, polarization currents originating from deep electronic traps contribute to transient conduction that persists for long times, even though it is large at short times. A power-law rather than an exponential time dependence is observed. The power-law behavior is interpreted as "universal" behavior in the sense of Jonscher. The power-law exponent changes with temperature as seen in some other materials. But a unique double transition from diffusive to dispersive transport that originates in two different electron traps is observed in varistors. At high applied voltages, the electrical nonlinearity is contributed by Schottky barriers that form at grain boundaries. The time response measurements reveal an inductive behavior that is strong evidence of electron holes; they thereby support theoretical models that attribute the collapse of the varistor barriers at high applied voltage to hole trapping in the barriers. Also, the time response measurements distinguish between different theoretical interpretations of the observed change in varistor capacitance with voltage. Evidence is found of both a short- and a long-term memory of previously applied electrical pulses. These memory phenomena are ascribed respectively to the life time of the holes that get trapped in barriers and to the persistence of the polarization currents that originate from deep electron traps.

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