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TECHNICAL REPORT

Leakage Currents in Degraded Multi-Layer Ceramic Capacitors

(Paper to be presented at IEEE Electronic Components Conference, May 14-16, 1984, New Orleans)

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Sponsor: Office of Naval Research (Contract No. N0014--83-k-0168)

Contractor: Virginia Polytechnic Institute and State University

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LEAKAGE CURRENTS IN DEGRADED MULTI-LAYER CERAMIC CAPACITORS

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Abstract

Leakage currents in new and degraded X7R type multi-layer ceramic capacitors show both ohmic and space charge limited behavior. The near-3/2 power voltage characteristic ($I \alpha V^{3/2}$) of new devices can be attributed to electron emission from electrode points. The quadratic behavior ($I \alpha V^2$) for moderately degraded devices represents space charge limited emission from planar electrodes. This emission may evolve from the point emission due to resistivity decreases that occur in the emission region as a result of ion movement. For these currents, electrons are believed to be the dominant charge carriers. Neither Schottky nor Poole-Frenkel currents were identified. Thermal activation energies were found to decrease from ~ 1.3 eV for new devices, to zero for degraded ones, corresponding to resistivity decreases from ~ 10^{13} Ω-cm (at 125°C) to 10^5 Ω-cm or less. Drift mobility increase (along with that of carrier concentration) may be an important factor in leakage current increase during degradation.

I. Introduction

It is known that degradation of insulation resistance, as contrasted to shifts in capacitance or dissipation factor, is the primary failure concern for high dielectric constant multi-layer ceramic (MLC) capacitors.[1,2] It is, however, not easy to measure the true DC leakage current for a good MLC device at room temperature due to the presence of additional currents that exist after a capacitor is charged, such as polarization and pyroelectric currents. The sum of these currents is usually larger than the DC leakage current over a period of many hours. It is not difficult, however, to measure steady state leakage at elevated temperatures, above roughly 100°C. This is because the DC resistivity is decreased, and the other current types decay faster.

DC leakage currents in new MLC devices of more or less standard composition (X7R for example) are probably due to the same mechanisms. When similar devices (X7Rs of different values) are degraded under the same conditions, similar trends are seen, both during degradation and in subsequently measured electrical characteristics.

The dominant leakage current charge carrier, and its mode of transport, have not been established. The current has been attributed, in MLC capacitors, to Poole-Frenkel emission[3], and indirectly (through the 1.19 eV activation energy) to oxygen vacancy diffusion.[1,4]

Numerous conductivity studies have been reported for $BaTiO_3$ ceramic[5-10] and single crystal[11-14] material. For the ceramic, electron (n-type) conduction seems predominant[5,6], with the conductivity strongly affected by impurities[7,8]. Space charge limited[6,9,11,12] and Schottky[10] currents have been reported. For the single crystal case, space charge limited currents[11,12] double injection[13], both ionic and electronic carriers in different temperature ranges[14] and small polaron hopping[15] have been reported. Some of those $BaTiO_3$ ceramic and single crystal studies may pertain to the various types of ceramic used in MLC capacitors.

Thus, there are several questions related to MLC capacitor leakage current that should be addressed. Some of these are:

1. What is the dominant charge carrier? Does it change (from say ionic to electronic) at some point during capacitor degradation?

2. What are the sources of the carriers (oxygen vacancies, electrodes, etc.)?

3. What is the mode of carrier transport (hopping, grain boundary tunneling, etc.) and does it also change with time?

4. What role does carrier transport play in degradation, as contrasted to carrier concentration?

These questions are addressed later in varying degrees. Before they are considered further, the types of current that might be expected to flow in MLC devices are reviewed.

II. Current Types

Ohmic

This can consist of both electronic (electron and/or hole) and ionic contributions. The voltage (V) dependence of current (I) is expressed as

$$I = qA\, n\mu\, V/L \qquad (1)$$

where q = electronic charge
A, L = cross sectional area and thickness
n = carrier density
μ = carrier drift mobility

The temperature dependence of I is contained in n and/or μ, and may be expressed in terms of an activation energy φ:

$$I = I_o e^{-\phi/kT} \qquad (2)$$

Space Charge Limited Current (SCLC): Planar Electrodes

In this case, electron (or hole) density injected from the electrodes exceeds the native bulk carrier concentration. This current applies to electronic carriers, since ions aren't expected to be injected from the electrodes in MLC devices. For planar electrodes, which might approximate MLC electrodes, a current-voltage relation is[16]

$$I = \frac{9}{8} A\, \theta\, \epsilon\mu\, V^2/L^3 \qquad (3)$$

where ε = permitivity and θ is a trapping parameter defined by

$$\theta = \frac{N_c}{2N_t}\, e^{-\Delta E/kT} \qquad (4)$$

where N_c = conduction band density of states and N_t = shallow trap density, located at energy ΔE below the conduction band edge.

These relations can be used to estimate μ, N_t and ΔE if the SCL characteristic defined by equation 3 is measured.

SCLS: Spherical (Point) Electrode

Current from a hemispherical point cathode emitting toward an anode of much larger radius can be described by the following I-V relation:[17]

$$I = \frac{2\pi\sqrt{2}}{3}\,\mu(\theta q \epsilon n)^{\frac{1}{2}}\,V^{3/2} \quad (5)$$

where all of the parameters were previously defined.

SEM pictures indicate the irregular nature of MLC electrodes. If SCLC is present, a non-planar electrode model such as that above may be necessary.

SCLC Variations

The two types of SCLC noted above are for specified electrode geometries and a discrete shallow trap. In reality, traps are unlikely to be discrete in a ceramic, and will probably be distributed across an energy range. Such extended trap distributions, in addition to the possible existence of diffusion currents (which were neglected above) modify the current-voltage dependence, usually towards a voltage exponent greater than two. Details are described in the literature.[16,17]

Schottky Current (Poole-Frenkel in bulk)

In this case, electron or hole currents increase due to lowering of a potential barrier by an external voltage, at the electrode (Schottky effect), or at a trap or grain boundary in the bulk (Poole-Frenkel effect).

The current-voltage relation for both of these currents has the form[18]

$$I = I_0 \exp\left[\frac{B}{T}\,(E/K)^{\frac{1}{2}}\right] \quad (6)$$

where B is constant, E is electric field, K is dielectric constant and T is temperature.

The Schottky effect only occurs at a rectifying contact, and probably doesn't apply to MLC capacitors. Bulk effects are more likely, due to electrons located in potential wells at traps and grain boundaries.

Field Dependent Factors

The dielectric constant appears in all of the above relations except the ohmic one. Voltage and temperature dependence of this factor must be included in these relations, and is more significant for Z5U material than for X7R.

Field dependent mobility could alter the I-V characteristics. This effect has been reported for a variety of single crystal semiconductors[19,20], and in these cases becomes predominant at low temperatures (< 300K). It is unlikely that it is a factor in polycrystalline ceramic above room temperature.

At high fields, the bulk carrier concentration can be increased due to avalanche effects. This is probably not a factor well below the breakdown strength of the dielectric, although it might be significant in localized regions of higher fields (caused by electrode non-uniformity, for example).

Other Factors

Additional current mechanisms, which are sometimes controlling factors in other structures, are probably not important for MLC devices. These include tunneling, surface leakage and field-induced exoemission.[21] Even though tunneling is important in other semiconductor devices,[22] and could potentially occur in MLC capacitors in high field locations (such as grain boundaries), we have seen no indication of it in I-V curves. Surface leakage and exoemission are probably not factors here because the electroded regions of these devices are buried in ceramic. Even though these external ceramic regions might serve as a local source of oxygen, they probably don't play an active part in current transport. External leakage paths on the package surface are not of concern to us here.

That a combination of the above currents may exist simultaneously, with particular ones dominant over specific voltage and temperature ranges, and that both electronic and ionic carriers may contribute, should be kept in mind during modelling of leakage currents for these devices. Also important is the high probability that leakage currents increase during lifetests in localized regions,[23] which may account for some of the variability seen in the I-V characteristics.

III. Experimental Techniques

Most of the measurements were made on 10nF and 1μF X7R devices, both in the new and degraded states. Some were obtained from commercial vendors following accelerated lifetests (nominally at 125°C, twice rated voltage), and we degraded additional capacitors of the same types (typically at 180°, four or eight times rated voltage). It was not our objective to perform statistical lifetest studies, but to ascertain and model trends that were common in the leakage current behavior of devices aged under the above conditions. The types of behavior discussed below were similar for 10nF and 1μF devices, for both sets of accelerating conditions.

Current-voltage measurements were made using a Hewlett-Packard 6116A voltage supply and a Keithley 610B electrometer. Sample temperature could be maintained to within about ± 0.2°C, from room temperature to 200°C.

Current decay or instability was a problem in several instances. a) For new devices, very long time constants for current stabilization were seen near room temperature. For this reason, measurements were made on these devices in the 100-200°C range. b) Degraded devices exhibiting space charge behavior are inherently unstable in voltage regions of trap filling and emptying. In this region, average values were recorded, until the region was traversed and the currents became stable. c) Some degraded devices exhibited unstable behavior over the entire voltage range, and self-healing effects (suddent decreases in current by up to several orders of magnitude) were often seen in these cases. d) An unstable increase was seen as current exceeded the 10^{-4} to 10^{-3} amp range, probably caused by localized heating in the device. Since only DC voltages were used, currents were kept below about 10^{-4} A.

Time dependent characteristics such as those mentioned above are not further considered, unless it is felt that they were due to trap filling effects. (For example, as shallow traps fill, current increases rapidly with voltage due to the large mobility increase).

Capacitance and dissipation factor were measured for new and degraded devices from 10 Hz to 13 MHz on a Hewlett-Packard 4192A Impedance Analyzer, in order to ascertain if any significant changes are occurring in these parameters. Except in the extreme cases (insulation resistance less than a few KΩ), no major changes were seen. This seems to indicate that any changes occurring at grain boundaries, interfaces etc. are not significant enough to show up. These measurements, therefore, aren't discussed further below.

It should also be noted that early in the program, Z5U capacitors also showed SCL and ohmic behavior similar to that of the X7Rs. Tests on Z5Us were discontinued due to the large dependence of interpretation on their more variable dielectric constant, and to the fact that X7R devices are more relevant to higher reliability needs. Therefore, only X7R measurements are discussed below.

IV. Results and Discussion

Results

I-V curves for new 10nF and 1μF X7R devices are shown in Figure 1 (T = 170°C). The near 3/2 power dependence ($I \alpha V^{3/2}$), especially at higher voltages, is typical of new devices.

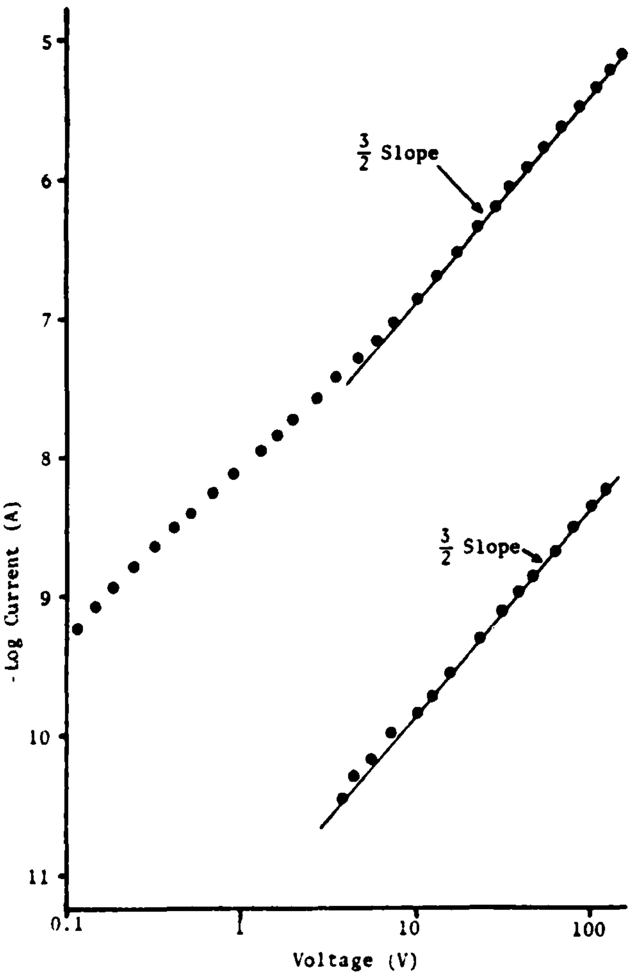

Figure 1. I-V curves for new 10nF (lower) and 1μF (upper) X7R devices.

Degraded devices show a variety of characteristics. The most commonly seen stable characteristics are: ohmic behavior at low voltages ($\lesssim$ 1 volt); SCLC square law or near square law behavior ($I \alpha V^2$) at higher voltages ($1 \lesssim V \lesssim 100$ v); a steep region on the I-V characteristic which is probably attributable to trap filling; ohmic behavior for severely degraded devices over the entire voltage range. Three device characteristics are shown in Figure 2, where the most degraded device is ohmic, the other two exhibiting SCLC characteristics.

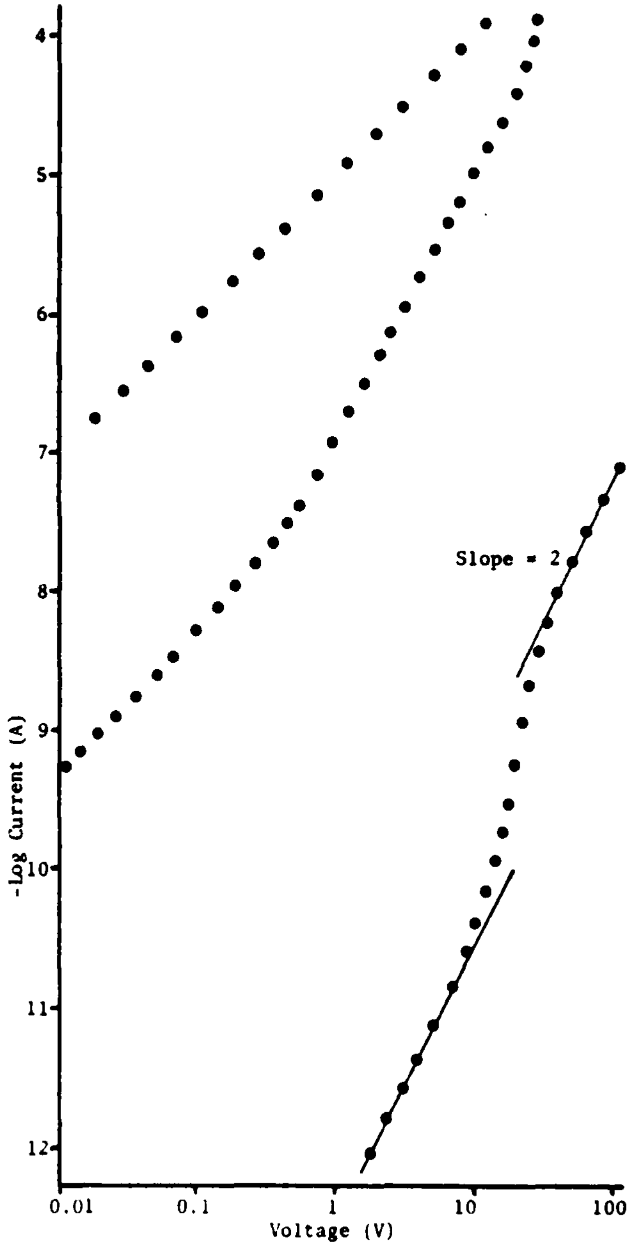

Figure 2. I-V curves for degraded 10nF devices.

A common characteristic seen in conjunction with SCLC is hysteresis, as shown in Figure 3, which is discussed further below.

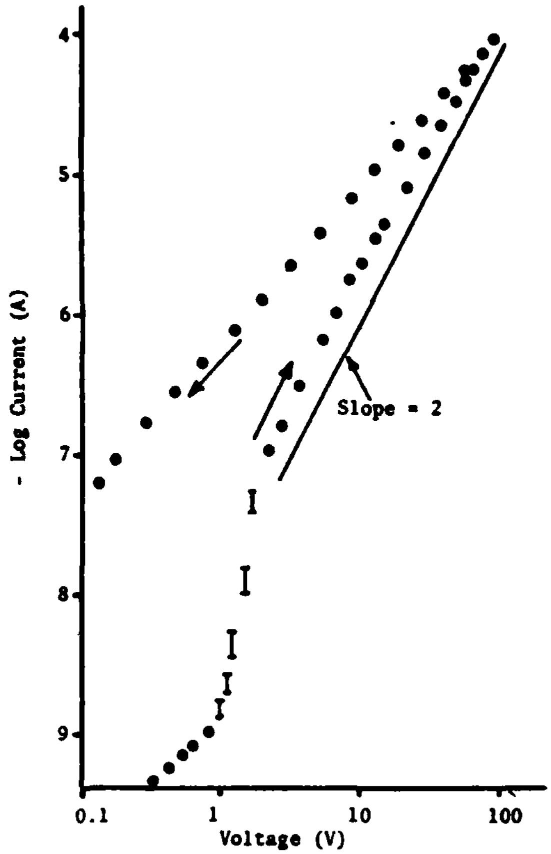

Figure 3. Hysteresis characteristic for degraded 10nF X7R device.

Activation energies for new X7R devices were measured to be 1.28 ± 0.03 eV. An example is shown in Figure 4.

It was found that activation energies were always less for degraded devices, i.e. as ceramic resistivity dropped so did activation energy. This dependence is shown in Figure 5 (using resistivity values at 125°C). Degraded devices with activation energies near zero have resistivities of about 10^5 Ωcm or less, and are ohmic. For 10nF devices, with area to thickness ratio of about 60, this corresponds to leakage resistance of roughly 2 KΩ or less.

The 3/2 or near -3/2 power dependence seen for new devices is not easy to fit to a planar electrode SCLC model, since due to trap filling, the voltage exponents are generally two or greater. (A point emission model that predicts 3/2 power behavior is discussed below).

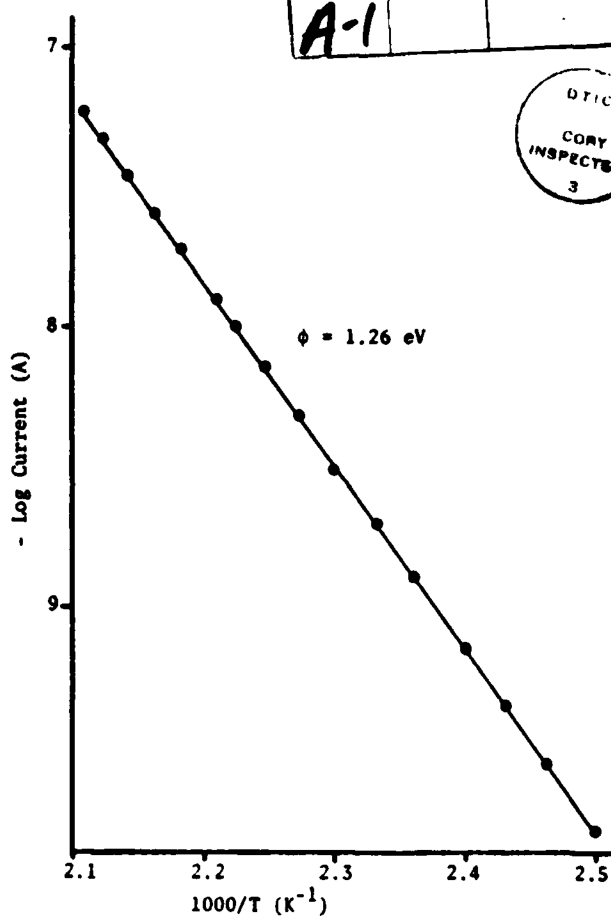

Figure 4. Arrhenius plot for new X7R device.

Current Types

Neither Schottky nor Poole-Frenkel types of currents were identified. Even though a near-linear ℓn I vs. $V^{1/2}$ characteristic was occasionally obtained over a certain range of voltages, the slope of the curve was much too large to be attributed to Schottky or Poole-Frenkel emission. As determined from equation 6, assuming a dielectric constant of 2000 and electrode spacing of 6×10^{-5} m, a Poole-Frenkel slope of about

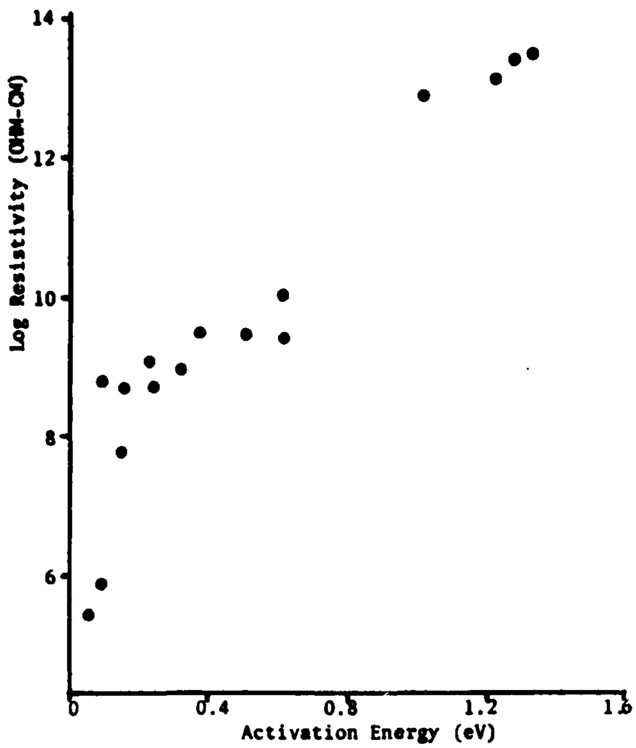

Figure 5. Dependence of resistivity on activation energy for X7R devices.

10^{-5} $(m/V)^{1/2}$ results, whereas experimental values are closer to 10^{-2} $(m/V)^{1/2}$. If one assumes a dielectric constant of only 10, a slope of about 10^{-3} $(m/V)^{1/2}$ results, which is still too small by an order of magnitude. In addition, since capacitance values remain essentially unchanged even for quite severely degraded devices, there is no indication that the dielectric constant is altered over a substantial volume of the capacitor.

What is the charge carrier? For all degraded devices (where a power law higher than 3/2 is followed) the SCL and ohmic currents are best described by electron flow. Thermoelectric coefficients for X7R chips with no internal electrodes are negative, indicating n-type behavior. Since the charge carriers for SCLC originate at the electrodes, ionic carriers can be ruled out.

For the undegraded capacitor also, a case can be made that the electron is the main carrier, under steady state conditions. Firstly, the 3/2 power model proposed below is based on electron emission, since it pertains to SCLC. Secondly, consider the leakage current exhibited by a 10nF X7R capacitor under lifetest at 180°, 800V. Its current remained steady at ∿ 0.5 µA for 5 days. If this current were due solely to doubly charged ion movement, it would result in over 10^{20} oxygen vacancies per cm^3. It is unlikely that the current could remain constant as such a large oxygen vacancy concentration developed. A more likely state is that electron flow predominates, accompanied by a smaller, ultimately degrading ion current in the same direction.

Another characteristic of interest is the hysteresis seen in Figure 3, which may be indicative of trap filling. It was noticed that upon heating to ∿ 100°C following such a run, the characteristic had reverted back to the lower curve, perhaps due to the thermal emptying of traps. If the thermally stimulated current (TSC) is measured during such heating, immediately after the device has seen 100 volts, a substantial current peak (about -3 nA for a 10nF capacitor) is seen at ∿ 50°C, which does not occur if another heating cycle is followed immediately thereafter. Such a TSC behavior might indicate the emptying of traps that were filled on the previous I-V run. However, it was found that this TSC peak was essentially the same for new and degraded devices, indicating that it is probably largely a depolarization current.[24] More extensive and refined TSC measurements on degraded devices such as these might yield trap parameters such as energy, lifetime etc.

Modelling

A model has been devised related to the 3/2 or near -3/2 power behavior seen for new X7R devices. It is known that 3/2 power behavior can be used to describe electron emission into a solid from a hemispherical electrode, an example for shallow traps being given in equation (5). Electron emission from electrode protuberances could result in a $V^{3/2}$ current dependence for MLC capacitors. Then one must ask why the 3/2 dependence transforms into a higher power (quadratic being common) as the device degrades.

Consider an electrode model where conductor protuberances occur due to the existence of ceramic pores, grain boundaries etc. in the electrode region (Figure 6).

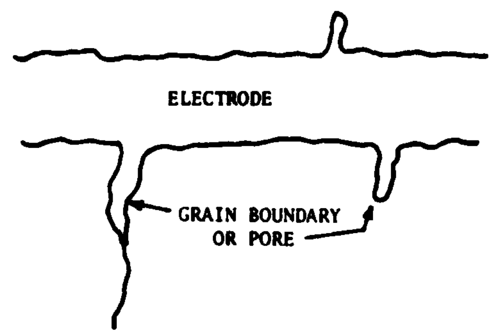

Figure 6. Electrode protuberances which can preferentially emit electrons.

Processes near the electrodes that could change the current from a 3/2 power SCLC behavior to quadratic (αV^2) as degradation occurs might be as follows: Initially, 3/2 power current from the cathode protuberance dominates. The field adjacent to the tip will be much larger than at the more planar cathode region. Oxygen ions will diffuse away from the tip preferentially, and a lower resistivity region will thus spread outward from the tip. The effective tip radius is thus increased due to the reduction of the field in the reduced resistivity region, and 3/2 power emission becomes less predominant. For more severe degradation, such effects could be so pronounced that tip emission would be more or less nullified, and the electrodes would appear (as far as electron emission is concerned) nearly planar. A simultaneous increase in oxygen vacancy and conduction electron densities in the ceramic body results in overall resistivity decrease and current increase. Hence the transformation from a 3/2 power to square low voltage dependence, with simultaneous increase in leakage current.

Even an order of magnitude estimate of the current emitted from such a point or network of points is at present difficult, because most of the key parameters are unknown, including density of points, carrier mobility and concentration, trap parameters, etc., in the ceramic adjacent to the emitter.

This model is therefore somewhat speculative, but does predict the observed voltage behavior. Other models are of course available, such as field dependent ion-flow[18], and field dependent electron mobility[19,22], but these are even more speculative since the voltage dependence is arrived at indirectly by means of field dependent charge transport.

For the degraded MLC X7Rs that we have measured, the quadratic or near quadratic voltage behavior is the most common type seen. What information can be obtained from such a characteristic? Consider the generic SCLC characteristic shown in Figure 7, for the case of shallow traps.

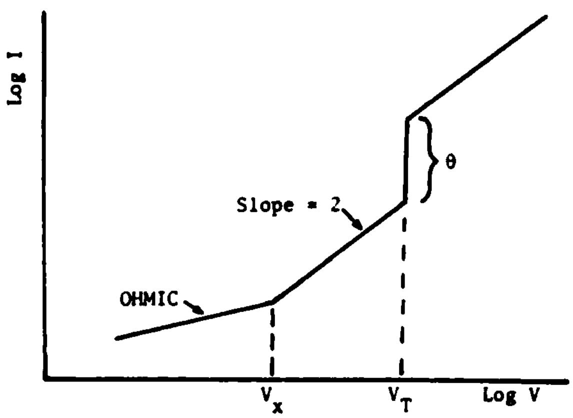

Figure 7. Generic ohmic-SCLC characteristic for degraded device.

Estimates can be made of certain parameters, under the assumption that the steep characteristic is due to shallow trap filling as more electrons are injected from the contacts. If trap filling occurs at voltage V_T, then equation (3) can be solved for mobility. One obtains

$$\mu \approx \frac{I L^3}{A\theta\epsilon V_T^2} \qquad (7)$$

The trap density can be estimated from the relation[16]

$$N_t \approx \frac{10^6 \ K \ V_T}{L^2} \qquad (8)$$

where K is the dielectric constant and other parameters were defined previously. The trap distance, in energy, from the conduction band edge can be expressed (using equation 4)

$$\Delta E = kT \ \ell n \frac{N_c}{2\theta N_t} \qquad (9)$$

We have used the following parameters related to 10nF X7R devices that show SCLC trap filling such as that seen in Figure 2: $A = 0.36$ cm^2, $L = 0.006$ cm, $\epsilon = 1.8\times10^{-10}$ F/cm.

The following ranges were obtained:

mobility: $10^{-7} < \mu < 10^{-2}$ cm^2/Vsec

trap concentration: $10^{14} < N_t < 10^{15}$ cm^{-3}

trap location: $0.4 < E_t < 0.6$ eV

The large mobility range results from equation (7) due to the large increase in I seen as devices degrade, with V_T remaining relatively constant, in the 1-10 V range. This is consistent with the fact that the ohmic and SCL quadratic currents both increase as a device degrades, each being proportional to mobility. In this regard, it should be noted that if only the carrier concentration increased during degradation (and if mobility remained constant), then only the ohmic part of the curve would increase in current; V_x in Figure 7 would increase, and the SCL current would be unchanged for $V > V_x$. This is because the SCL current does not depend on the background carrier concentration in the ceramic, but on emission from the electrodes, and, for transport, mobility. This implies for these devices, that a mobility increase may be just as significant a contributor to leakage current as the carrier concentration increase, during degradation. Mechanisms for such a mobility increase are at present not known, but may include grain boundary barrier height reduction (due to charge accumulation), hopping potential decrease, or other factors, depending on what actually controls the mobility. One indicator of a reduction in transport potential barriers (two examples mentioned above) could be the decrease in current activation energy ϕ, as shown in Figure 5. In equation (2), ϕ can be represented as $\phi = \phi_c + \phi_m$, where ϕ_c and ϕ_m are carrier concentration and mobility activation energies respectively. Since equation (2) is followed experimentally, we anticipate that carrier concentration and mobility might follow similar relations, firstly because of the large changes in μ noted above, and secondly because this is what was conclusively demonstrated by Nagels for $LiNbO_3$[25]. He reported electron conduction by means of small polarons, with both carrier concentration and mobility varying exponentially with temperature. Whether or not this is the case for X7R ceramic is being investigated by us by means of thermal emf and conductivity, and will be reported in a separate communication.

V. Conclusions

1. Steady state DC leakage currents for X7R MLC capacitors are ohmic at low voltages; at higher voltages follow a near-$V^{3/2}$ dependence when new, evolve into a V^2 behavior during degradation, and subsequently, with further degradation, become ohmic over the entire voltage range of the device;

2. The 3/2 and quadratic voltage behavior can be modelled after space charge limited currents; the 3/2 case attributed to electron emission from cathode points, the quadratic case to emission from planar electrode regions;

3. Electrons appear to be the dominant charge carrier, with thermoelectric polarity on X7R chips indicating n-type behavior;

4. Schottky or Poole-Frenkel currents were not identified;

5. Thermal activation energies decrease from ~ 1.3 eV to zero as resistivity (at 125°C) decreases from $\sim 10^{13}$ Ω-cm to $\sim 10^{5}$ Ω-cm or less;

6. Estimates of drift mobility, trap concentration and trap energy ranges were made from quadratic SCLC characteristics;

7. It is possible that mobility increase, (in addition to carrier concentration increase), plays a dominant role in leakage current increase during degradation.

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