Dynamics of Nonlinear Charge Injection in Polymeric Films

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***Abstract*— Fundamental understanding of carrier mobility-related pre-breakdown phenomena in dielectrics provides insights into high field transport phenomena as well as the associated aging and onset of charge injection instability. A system for measuring resistive current through a planar dielectric film during a linear ramp voltage to breakdown has been developed to address the limits of conventional steady-state approaches in which the sample typically fails at ~60% of the breakdown field. With a dynamic range of 140dB and high linearity of the feedback loop, the system can facilitate cancelation of capacitive current during even substantial changes in sample capacitance as a function of field, which can occur in highly nonlinear materials. Based on this technique, the pre-breakdown conduction in polymeric films was characterized, which suggests highly nonlinear field dependence of charge injection and transport. Ability of such transient measurement of the dynamics of nonlinear charge injection in polymeric films provides a basis for conduction mechanism study and material engineering for improved high field performance.**

Keywords— ***high field conduction, dielectric breakdown, space charge limited current, polymer dielectrics***

I. INTRODUCTION

Dielectric films with high capacitive energy storage density are the most pervasive and least reliable devices, used in the DC-link of power conversion systems, filtering, power factor correction, and pulsed power systems [1]. In a broader sense, fundamental research on dielectric materials could be a critical enabler for payload efficiency and affordable high power density of various integrated electric propulsion and drive systems [2]. Through intensive research, promising candidate material families have been explored which include relaxor ferroelectric polymer based blends, copolymers, nanocomposites and nanolaminates, as well as modified glassy polymers through the addition of cyanoethyl, thiol, or sulfonyl polar side groups [3-5]. In addition to organic polymers, polymer chemical space searches have led to new promising organometallic polymer dielectric families [6-8]. These recent discoveries indicate the untapped opportunities within the vast polymer chemical space.

While rational design through hierarchical chemical space searches shall be continued [9-10], fundamental questions related to the degradation and breakdown of polymer dielectrics when subjected to large electric fields should be addressed. This is essential for the ultimate success of any capacitor and dielectric material development. Despite early seminal work [11-13], the behavior of a material experiencing ultra high fields has long defied a fundamental understanding due to the fact that dielectric degradation and breakdown process in engineering materials is extremely complex as a result of the interplay between the magnitude of the electric field, the time span of imposition of the field, the temperature, and the state and morphology of the material. Dielectric degradation which eventually culminates in breakdown is essentially the progressive creation and accumulation of atomic and nanoscale defects, assisted by electric field, time and temperature. The highest possible electric field that a defect-free material can tolerate has a special significance which corresponds to the intrinsic breakdown strength, determined solely by the material's chemistry and crystal structure [14]. The dielectric degradation and breakdown phenomena thus need much more understanding to guide ultimately the development of new electric field tolerant insulators with high breakdown strength. Conduction and defect accumulation provide positive feedback to each other, culminating in eventual catastrophic breakdown. As a result, pre-breakdown study is essential. This will involve studying the nature and onset of the injection and transport of charge carriers with high mobility, and the carrier mobility induced instabilities under extremely high electric fields.

II. TRANSIENT MEASUREMENT SYSTEM

Numerous past efforts on the characterization and study of the conduction and aging mechanism of dielectrics could only reach around two thirds of the breakdown field due to failure as a result of rapid aging [15]. Therefore, carrier mobility-related prebreakdown phenomena can only be studied within microscopic dimensions or under transient conditions to avoid thermal runaway as the result of power dissipation under extremely high fields [16]. Under transient condition, the current measured is a combination of the resistive current, the capacitive current, as well as, for the case of nonlinear polar dielectric, the polarization current,i.e , $j(t) = \varepsilon_0 \varepsilon \frac{\partial E(x,t)}{\partial t} + \frac{\partial P(x,t)}{\partial t} + j_c(x,t)$ The capacitive current is several orders of magnitude higher than the resistive current even near breakdown field. In order to reveal the resistive current as related to the charge injection, the capacitive current must be cancelled during the whole

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measurement. In this study, we utilize a newly developed active capacitive current cancellation system to study the dynamics of nonlinear charge injection in polymeric films under extremely high fields.

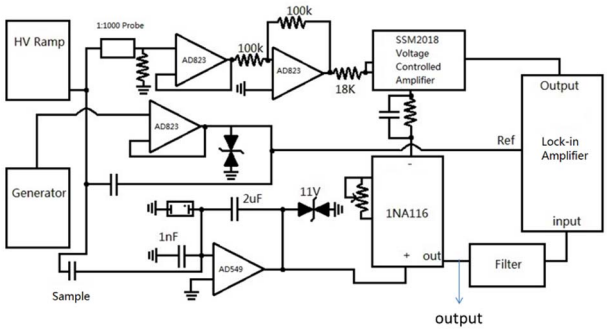

Fig.1 Block diagram of the prebreakdown measurement system with active capacitive current cancellation.

The principle of transient measurement system is based on active cancellation of the capacitive current with dynamic gain control. In details, a 1kHz small sinusoidal signal is superimposed on the high voltage ramp, causing a capacitive current through the sample. A high precision voltage probe measures the same high voltage modulated signal and provide a reference with a 1000:1 ratio. The amplitude of the reference signal is controlled by the VCA, and the gain of the VCA is controlled by a dual phase lock-in amplifier. The conditioned current signal from the sample and the reference are fed into a differential amplifier, the output of which is applied to the lock-in amplifier. The input voltage of the VCA is controlled by the feedback signal from the lock-in amplifier. The out-of-phase "Y" output is used as the feedback, which represents the remaining capacitive component. The reference phase is set to 128 where 90 is out of phase of the resistive component and 38 is due to the phase shift of the filter used to block out the DC component for the lock-in. The phase of the reference is set so that the lock-in can control the VCA to cancel all the capacitive current induced by the sinusoidal signal. As long as the system is linear, cancellation of the capacitive current caused by the sinusoidal signal results in cancellation of all capacitive current

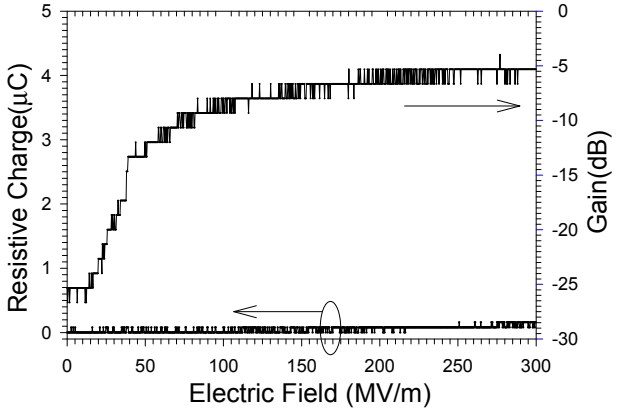

Fig.2 Demonstration of the principle of dynamic VCA gain control over a range of 30dB for effective active cancellation of the capacitive current.

through the sample. The remaining signal measured by the oscilloscope is the integration of the resistive current. The use of VCA results in an extremely broad dynamic range and fast response. The VCA has a 40dB – 100dB gain range following the 30mV/dB control principle, and the unity gain corresponds to a zero input voltage. Fig.2 demonstrates the capability of the VCA for an effective dynamic cancellation of the capacitive current over the range of 30dB. Typical working condition shall be centered around unity gain. Further development of this system shall provide complemental information to the traditional Seyer-Tower type D-E loop type of measurement regarding the polarization behavior.

III. EXPERIMENTAL

As shown in Fig.3, the polymeric film sample is sandwiched in between two metalized film electrodes with the top electrode facing down and the bottom electrode facing up. A 125μm Kapton film with a 1cm by 1cm window is used as a mask to set the active area of the sample. Two types of commercially available capacitor grade films are included in this study, i.e., biaxally-oriented polypropylene (BOPP) film of 12.5um formed by bubble process and polystyrene (PS) film of 10um in thickness.

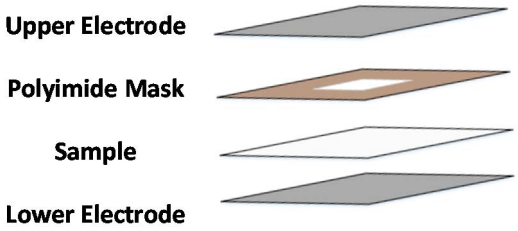

Fig.3 The metal-insulator-metal sample layout.

Before the actual measurement, the sample was conditioned to approximately 100 MV/m to ensure good contact and elimination of the air bubbles trapped in between the electrodes and the sample. A half rectified triangular high voltage waveform with a ramping rate of 300V/s is applied to the film sample. The high voltage waveform is generated by a 100kV Spellman high voltage power supply driven by a function generator. The ramping rate is kept the same for all the different applied voltages.

Two types of measurements were performed for each material. In the first measurement, the sample is subjected to repetitive high voltage waveforms with progressively increased magnitude. After the first run, most of the shallow trap shall be filled and the resistive charge measured over the subsequent runs will be due mainly to conduction current. In the other type of measurement, every run involves a fresh sample. In this measurement, there is no prior history for each sample and the measured charge represents a combination of mostly the conduction current with the absorption current.

IV. RESULTS AND DISCUSSION

The charge injection in two polymer films under the transient triangular high voltage waveform is shown in Fig.4. The applied voltage ramped up to ~4 kV before ramping down to zero. The last portion of the voltage waveform lost its linearity due to the relaxation time constant.

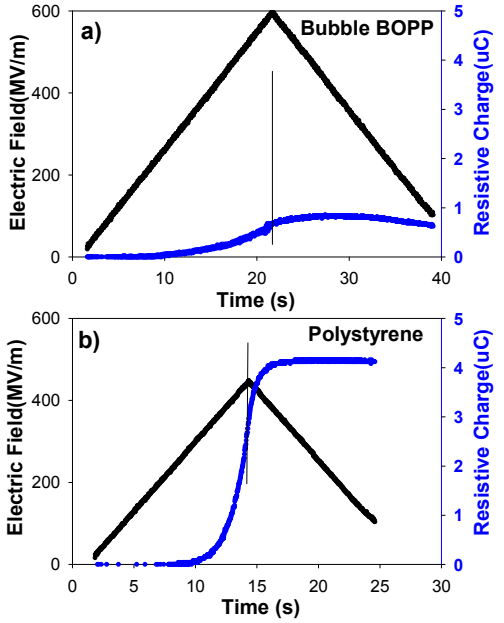

Fig.4 Nonlinear charge injection in (a) bubble BOPP and (b) polystyrene.

The first half of this measurement is similar to the prebreakdown conduction study reported in a previous paper [17]. Once above a threshold field of ~150kV/mm for polystyrene, the Ohmic behavior for low field conduction turns rapidly into a highly nonlinear charge injection. The shape of this experimental determined Space Charge Limited Current curve reflects the increment of the space charge with respect to the shift of Fermi energy. Further analysis of the first and higher order derivatives of the temperature modulated current density vs field curves yields the density of states for bulk traps inside polymeric films, measured in reference to the mobility edge of conduction band [18].

It shall be noted that such supralinear charge injection keeps increasing even after the maximum voltage is reached. The charge injection stops only when the inception voltage is reached again during the voltage ramping down. Such quantitative measurement of transient charge injection provides a unique opportunity for the observation of the charge dynamics as a result of space charge modified potential distribution inside the sample. When comparing the results for different materials, amorphous polystyrene definitely shows significantly more charge injection than semi-crystalline BOPP. One interesting observation is that the transient effect during the voltage ramping down cycle of the measurement. The resistive charge through polystyrene film stops increasing once the applied voltage is lower than a certain threshold and comes to a steady state. However, for BOPP, the charge starts decreasing when the applied voltage is lower than the charge injection threshold, which suggests some of the charge is actually "flowing back" against the applied voltage. The resistive charge is a direct integration of the resistive current. The charge through BOPP decreases once the applied field reaches once again the threshold field for charge injection, indicating a negative current flow under positive applied voltage. For the case of polystyrene, the observed behavior may be explained by the transient effect proposed by Rose and Smith [19,20]. When the applied voltage is reduced, it requires certain time for the trapped charges forced into the material at a higher voltage to be thermally released. The long time required for the transient current to subside is a direct measure of the capture cross section of traps in polystyrene (deep trapping) [19]. As for the case of BOPP, the "flow-back" of the charge during the voltage ramping down cycle indicates

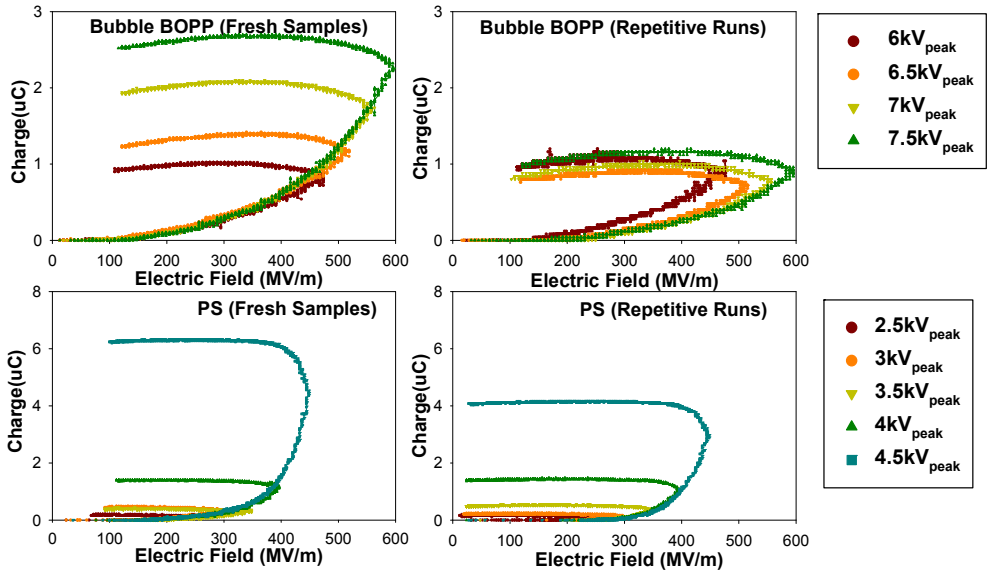

Fig.5 Resistive charge in BOPP and polystyrene films subjected to triangular voltage waveforms with different peak voltages. For BOPP, the peak voltages varied successively from 6kV to 7.5kV and for PS the peak voltages increased over five runs from 2.5kV to 4.5kV. Each material was tested under two different condictions, with one involved fresh sample for every run and the other involved only one single sample subjected to repetitive testing runs.

the presences of carriers of high mobility under extremely high field [21].

Fig.5 shows all the experimental data from these two materials. The applied voltage is converted to electric field and used as x-axis. Four rounds of measurement were performed for BOPP from 6kV to 7.5kV and five round measurement for PS from 2.5kV to 4.5kV. The voltage step between each round was 500V and the ramping rate was kept at 300V/s.

Comparing the results for test runs involving one single sample repetitively with the other using a fresh sample for every run, the one with fresh samples shows significantly higher charge injection. This is due to the fact that the resistive charge measured includes also the absorption current in addition to the conduction current. Furthermore, the nonlinear resistive charge curves overlap well with each other during the voltage ramping up cycle. For the repetitive runs involving only one single sample, the absorption current associated with the shallow traps will present mostly in the first run. Majority of these charges will be trapped and charges measured during the consecutive runs represent nearly the pure conduction. For BOPP, the second and third runs show actually lower resistive charge than the first run even though the applied voltages are higher.

Additional transient conduction study will be conducted under different ramping rates to probe the dynamics of space charges as well as the field dependent dielectric time constant. Such transient conduction study will be assisted with nonlinear finite element numerical computation.

V. Conclusion

With a dynamic range of 140dB and high linearity of the feedback loop, a special system was developed to facilitate the cancelation of capacitive current during even substantial changes in sample capacitance as a function of field so that prebreakdown conduction in polymeric films can be characterized to provide a quantitative basis for the study of charge injection and transport. Transient space-charge-limited-current measurement and analysis suggest distinctive behavior between BOPP and polystyrene. Furthermore, such transient resistive current measurement provides insight into the dynamics of nonlinear charge injection in polymeric films under extreme electric field. With further development, such pre-breakdown measurement is expected to advance the fundamental understanding of conduction mechanisms in polymeric dielectrics.

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