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Characterization of PVC–PMMA Blend Thermoelectrets via TSDC Measurements

R Y Bakale [*], Y G Bakale , Y S Tamgadge , R P Ganorkar, S V Khangar

Department of Physics, Mahatma Fule Arts, Commerce and Sitaramji Chaudhari Science Mahavidyalaya, Warud, Maharashtra, India.
Department of ASH, Kavikulguru Institute of Technology and Science, Ramtek, Maharashtra, India
Department of Physics, Mahatma Fule Arts, Commerce and Sitaramji Chaudhari Science Mahavidyalaya, Warud, Maharashtra, India.
Department of Chemistry, Mahatma Fule Arts, Commerce, and Sitaramji Chaudhari Science Mahavidyalaya, Warud, Maharashtra, India.
Department of Physics, Shri Shivaji Education Society Amravati, Science College, Congress Nagar, Nagpur, India

ABSTRACT: Thermally stimulated discharge current (TSDC) measurements were performed on thermoelectrets prepared from blend films of polyvinyl chloride (PVC) and polymethyl methacrylate (PMMA) in varying compositions. The films were polarized under a DC electric field at elevated temperatures and cooled under constant bias to freeze-in dipolar and space charge orientations. The discharge current spectra, recorded upon linear heating, revealed multiple relaxation peaks corresponding to molecular dipole reorientation, interfacial polarization, and trap release mechanisms. The most intense relaxation appeared near 95 °C for a 70:30 PVC–PMMA blend, attributed to α-relaxation linked with cooperative segmental motion of the polymer chains. The activation energies of the relaxation processes were calculated using the initial rise method, showing values between 0.65 eV and 1.25 eV. The results indicate that PMMA incorporation increases the trap depth and stabilizes the electret state, suggesting potential applications in charge storage and sensor devices.

KEY WORDS: blend film, TSDC, Thermoelectric.

I. INTRODUCTION

Thermoelectrets are dielectric materials capable of retaining quasi-permanent electric polarization after being subjected to an electric field at elevated temperature. Among polymeric electrets, polyvinyl chloride (PVC) is well known for its high resistivity and charge storage capability, while polymethyl methacrylate (PMMA) offers excellent optical and mechanical stability. The blending of these polymers provides a means to tune their dielectric and relaxation properties through intermolecular interactions and morphological modifications.

Thermally stimulated discharge current (TSDC) analysis is a powerful technique to investigate charge trapping and relaxation phenomena in dielectrics. In this method, a polarized sample is heated at a controlled rate, and the resulting discharge current is recorded as a function of temperature. The obtained spectra yield valuable information about the nature, depth, and stability of traps, as well as molecular mobility within the polymer matrix.

Previous studies have shown that blending polar polymers can induce new relaxation processes due to interfacial polarization and altered free volume distributions. However, limited work has been reported on the TSDC behaviors of PVC–PMMA blends, particularly concerning the relationship between blend composition and relaxation characteristics. This work aims to fill that gap by systematically examining the TSDC spectra of thermoelectrets formed from PVC–PMMA blend films.

II. EXPERIMENTAL WORK

Materials and Film Preparation

Commercial-grade PVC (Mw ≈ 80,000) and PMMA (Mw ≈ 100,000) were used as received. Blend films with PVC: PMMA weight ratios of 100:0, 90:10, 70:30, 50:50, and 30:70 was prepared by dissolving appropriate amounts of the polymers in tetrahydrofuran (THF) and casting onto clean glass substrates. The solvent was evaporated slowly at room temperature, followed by vacuum drying at 60 °C for 12 hours to remove residual solvent. The average film thickness was about 50 μm, measured with a digital micrometre.

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III. ELECTRODE DEPOSITION AND POLARIZATION

For TSDC measurements, aluminium electrodes (diameter 10 mm) were vacuum-deposited on both sides of the films. The samples were polarized by applying a DC electric field of 1.5×10^6 V m^{-1} at a polarization temperature (Tp) of 100 °C for 30 minutes. The films were then cooled to room temperature under the same field to preserve the induced polarization.

IV. TSDC MEASUREMENT

The polarized electrets were short-circuited for 10 minutes at room temperature to eliminate stray charges, then heated at a linear heating rate of 3 °C min^{-1}. The discharge current was measured using a Keithley 6517B electrometer. The current density (J) was plotted against temperature (T) to obtain TSDC spectra.

V. EXPERIMENTAL RESULTS AND DISCUSSION

A. General Features of TSDC Spectra

Figure 1 shows the TSDC spectra for PVC–PMMA blend thermoelectrets of different compositions. Pure PVC exhibits a single broad discharge peak around 85 °C, associated with α-relaxation due to cooperative segmental motion in the amorphous phase. Incorporation of PMMA modifies the spectrum significantly, producing multiple peaks that reflect distinct relaxation modes. The 70:30 PVC–PMMA blend displays a well-resolved major peak at 95 °C and a secondary peak near 55 °C. The low-temperature peak (β-relaxation) is attributed to local dipolar reorientations of polar groups (C–Cl in PVC and C=O in PMMA), while the high-temperature α-relaxation corresponds to large-scale segmental motions coupled with space charge release.

B. Effect of Blend Composition:

As PMMA content increases, the main TSDC peak shifts toward higher temperatures and decreases in magnitude, indicating enhanced trap depth and reduced mobility of charge carriers. This suggests that PMMA introduces deeper traps and increases dipole–dipole interactions, leading to more thermally stable electrets.

At high PMMA fractions (50:50 and 30:70), the α-relaxation broadens considerably, suggesting overlapping contributions from both polymer phases. The blending appears to promote interfacial polarization at PVC–PMMA boundaries, further broadening the relaxation spectrum.

C. Activation Energy Determination:

Activation energies (Ea) of the observed relaxation peaks were calculated using the initial rise method,

$$J\,(T)\;\alpha \exp\,[-\frac{E_a}{KT}]$$

where J is the current density, k is the Boltzmann constant, and T is the absolute temperature. The resulting Ea values ranged from 0.65 eV (β-relaxation) to 1.25 eV (α-relaxation), consistent with molecular dipolar reorientations and space charge entrapping mechanisms, respectively.

D. Mechanistic Interpretation:

The enhancement of TSDC intensity in moderate PMMA concentrations ($\leqslant$ 30 %) can be attributed to increased polarizability and better alignment of carbonyl dipoles within the PVC matrix. Excessive PMMA content, however, reduces free volume and charge mobility, resulting in diminished discharge currents. This balance indicates a synergistic effect between the two polymers, optimizing charge retention and relaxation behavior at intermediate compositions.

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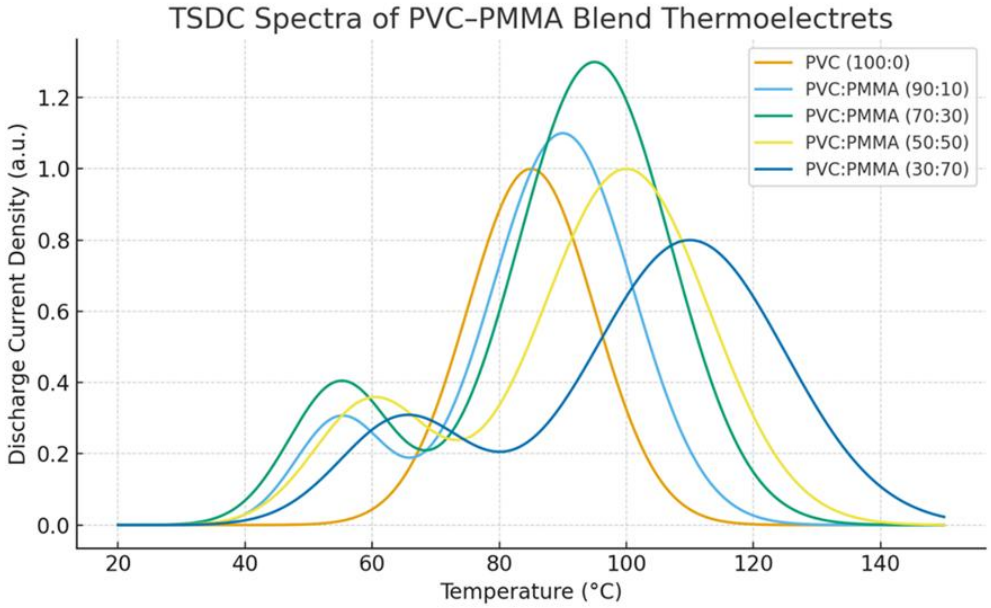

Figure 1: Simulated TSDC spectra of PVC–PMMA blend thermoelectrets at various compositions (Heating rate: 3 °C/min; Polarization field: 1.5 × 10⁶ V/m).

Here is the TSDC spectrum for PVC–PMMA blend thermoelectrets — showing the characteristic shift of discharge peaks toward higher temperatures and reduced magnitudes with increasing PMMA content, consistent with enhanced trap depth and electret stability.

V. CONCLUSION AND FUTURE WORK

1.PVC–PMMA blend thermoelectrets exhibit multiple TSDC peaks corresponding to β- and α-relaxations associated with dipolar and space charge mechanisms.
2.Increasing PMMA concentration shifts the relaxation peaks to higher temperatures, indicating higher trap depths and improved charge stability.
3.The 70:30 PVC–PMMA blend demonstrates optimal electret performance, with the most intense and thermally stable discharge current peak near 95 °C.
4.The calculated activation energies (0.65–1.25 eV) confirm that both molecular orientation and space charge relaxation contribute to the TSDC behavior.
5.The results highlight the potential of PVC–PMMA blends as tunable materials for charge storage and sensing applications.

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