Investigation of Thermally Stimulated Depolarization Current in Poly-Methyl-Methacrylate Magneto-Electrets

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Abstract

Thermally stimulated depolarization currents (TSDC) have been investigated in poly-methyl-methacrylate Magneto-Electrets (PMMA-MEs) in the temperature region of 60-160°C under various polarizing fields, ranging from (1 to 9) Kilo Gauss of uniform magnetic field with different heating rates. The TSDC spectra show a peak (P1) around 110-120°C and pronounced peak (P2) around 150-160°C attributed to the joint reorientation of the polar side-groups, together with the adjustment of main chain segments for peak (P1). The peak (P2) is considered to be due to decay processes related to conduction.

Keyword: Magneto-Electrets, Thermally stimulated discharge currents, PMMA.

Introduction

Polymethylmethacrylate (PMMA), CH$_2$=C(CH$_3$)CO$_2$CH$_3$ is one of the best polymeric materials broadly used for insulation devices manufacture. Its electric properties are highly influenced by many environmental parameters such as temperature, UV radiations, oxidation, therefore, investigations on the effects of these parameters have been made by numerous research teams[1-4]. In our case, we have chosen to submit PMMA samples to magnetic field to identify nature and origin of polymer chain dynamics, dipolar rotations and space charge in magneto electrets of PMMA[5-6]. This paper focuses on, the dielectric relaxation behavior of PMMA by investigating the TSDC currents obtained under various test conditions, such as different magnetic fields, heating rates, different electrode combinations and thickness of the sample.
Experimental Details
Measurements were carried out on samples of commercial PMMA (Altuglasw), which were cut from sheets of 20 mm thickness. The usual method of preparation of Magneto-Electrets was to place a pristine sample of PMMA in the form of a disc (diameter =1.23 cms, thickness = 20mm) in a cavity made out of insulating material. Raw sample was repeatedly washed with benzene so that any frictional charge may be get rid off after that it was covered from both sides with a 99.9% pure aluminum foil. Now the sample was put inside magneto-Electret container which comprises of an intermediate plate of brass, having a circular cavity for the sample. The sample was covered from both sides with aluminum foil to create good thermal contact, and then two outer brass plates were screwed together. This M.E container was held vertically in between the pole pieces of the electromagnet along with a thermocouple (K-type), probe inserted in cavity at the top of the container for recording temperature. To keep the temperature uniform over the entire area of the M.E. sample, non-magnetic heating elements are embedded in the outer plates of the container.

Results and Discussion
The TSDC current spectra of PMMA sample, for various polarizing fields (H_p), ranging from 1 to 9 KGauss at the poling temperature of 140°C. In all the cases current peak (P_1) is observed around 110° – 120°C known as α peak. The peak is more pronounced at higher values of (H_p). In most of the cases there is a presence of current maximum around 155°-160°C known as ρ peak. A steady fall in the current is observed in the high temperature region approximately beyond 160°C. I_{max} shows increasing trend of 0.55- 0.65 pA in the field strength of 1600 Gauss to 5600 Gauss although this increase is not phenomenal whereas I_{max} increases a bit more upto 0.9pA during application of 8600 Gauss as shown in Fig[1,2]

![Figure 1](image.png)
These observations are in conformity with the non-isothermal charging mechanism of a polymer where the polarizing current is supposed to comprise of two components behaving differently as a function of temperature. The first one is the orientation of dipoles as a transient process resulting in the formation of $\alpha$-peak, whereas the second component $\rho$-peak is the conduction current derived due to decay processes related to conduction, the motion of excess charges by space charge limited drift and diffusion. In PMMA the presence of carbonyl groups ($>\text{C}=\text{O}$) constitute the bulk of permanent dipoles. In presence of polarizing magnetic field as the heating of sample is started, the relaxation time ($\tau_o$) of the dipoles decreases and they are progressively oriented. This orientation becomes more strong and rapid with increasing polarizing field. Consequently, the current maximum appears with greater magnitude. The linear relationship between the magnetic field strength ($H_p$) and the peak magnitude $I_m$ shows the occurrence of a uniform induced bulk polarization confirming the dipole origin of the $\alpha$- peak.

The location of $\alpha$-peak is such that we cannot attribute the dipole nature of $>\text{C}=\text{O}$ groups as the cause for this current maximum. It has been shown earlier that the dipolar relaxation due to $>\text{C}=\text{O}$ groups TSD current maximum occurs at around 120°C. The dipolar nature of peak is also independent of the sample thickness Fig[3] as well as by the kind of electrode material Fig[4].
Turnhout has shown that usually thin samples release more charge than very thick ones and the peak intensity is maximum when \( r_0/s = 0.5 \), where \( r_0 \) represents the thickness of the charged layer and \( s \) that of the sample. The activation energy estimated from Initial – rise method for peak P1 comes around 1.54 – 2.18 eV. The distinction between the actual polarization current and the conduction current can be made by adopting different heating rates during the TSDC process as it affects the characteristic of the former components, the faster heating rate shifts the TSDC maximum towards higher temperature and with enhanced magnitude Fig[5].
In addition to dipoles, the electrets usually contain immobilized space charges. These are nonuniformly stored, often residing near the electrodes. During heating, they will be mobilized, and neutralized either at the electrodes or in the sample by recombination with charges of opposite sign. The forces driving the charges are their drift in the local magnetic field, and diffusion, which tends to remove concentration gradients. In general, the field-controlled self-drift prevails. At high temperatures, the self-motion of space charges becomes accompanied by a second neutralization mechanism, namely, recombination with thermally generated carriers. These carriers are generated uniformly in the entire specimen by dissociation of neutral entities. They are responsible for the conductivity of the material. The conductivity can be either electronic or ionic; in polymer it seems to be (impurity) ions which contribute most to the ohmic conduction, because polymers show an appreciable conduction only above $T_g[7]$. In our case also $\rho$ peaks appear at $160^\circ C$ and activation energy comes around $1.45 - 1.61 \text{ eV}$.

**Conclusion**

TSDC depolarization process found to be dependent on the experimental conditions. These observations confirm the charging mechanism of PMMA in magnetic field. Combined effect of polarizing magnetic field and temperature shows increase in conductivity of PMMA MEs with increasing polarizing magnetic field. The dielectric relaxation measurements of nonlinear optical polymer are very useful for obtaining detailed information of the underlying molecular origins as well as for process control in manufacturing.
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Reference