Open- and short-circuit thermally stimulated currents in ethyl cellulose (EC) : polymethyl methacrylate (PMMA) blend

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Abstract. Mechanisms of charge generation and its persistence in one and both-side vacuum-aluminized ethyl cellulose (EC) : polymethyl methacrylate (PMMA) blend thermoelectrets, prepared under different fields (10, 25, 50 and 100 kV/cm) and temperatures (40, 60, 80 and 100°C), have been analysed using short- and open-circuit thermally stimulated depolarization current (TSDC) technique. The TSDCs were recorded by reheating the samples at a linear heating rate of 4°C/min. The TSDC thermograms of polyblends containing EC:PMMA in different weight ratio are, in general, characterized with two peaks in lower and higher temperature regions. However, the polarity of the peaks was found to be just opposite in short- and open-circuit TSDC measurements. Moreover, results on 97:3, 93:7 and 90:10 EC:PMMA polyblends indicated that the current increases with concentration of PMMA. The results indicate the existence of heterocharge due to dipole orientation and ionic charge drift together with the injection of charge carriers from electrodes with their subsequent localization in surface and bulk traps. Further, the chances of charge trapping in polyblends, at the interfaces are greater than in the individual polymers.

Keywords. Thermally stimulated currents; heterocharge; homocharge; plasticization effect; induced dipoles.

1. Introduction

A rapid growth of interest, in recent years, in studying charge storage and transport phenomena in polymer electrets by means of techniques based on thermal stimulation has been indicated by a number of publications (Shrivastava et al 1979, 1981; Pillai et al 1981; Khare et al 1992, 1996). The thermally stimulated depolarization current (TSDC) is a powerful technique that has provided valuable information on the charge storage and transport behaviour of many polymers (Sinha et al 1981; Keller et al 1993; Khare 1994; Khare et al 1994a, b). The technique has shown that for improving charge storage properties of polymers and obtaining strong and stable electrets, a better understanding of the structural or morphological details and dynamical properties of the polymers is required on both molecular and super molecular levels (Frensch and Wendorff 1986). It has been reported that the electret state in a polymer can be produced not only by conventional methods, but also by bringing about some structural changes in the electret forming materials (Khare and Singh 1994). Investigations concerning the dependence of electrical properties on structure are, however, presently very sparse. Appropriate model systems are required to obtain definite conclusions.

Multicomponent polymers, specially blends of two chemically different polymers that are partially compatible, comprise such model systems.

Ethyl cellulose is a partially polar polymer having density 1·14, melting temperature 165-175°C and glass transition temperature, Tg, 60-75°C. Polymethyl methacrylate is strongly polar and mostly amorphous in character, having density 1·88, melting temperature 180°C and glass transition temperature, Tg, 114°C. The two polymers thus differ remarkably in their glass transition temperature, i.e. in their dynamical as well as structural properties. Blends of these two polymers are expected to yield strong and long-lived electrets useful for industrial applications.

Several reports on TSDC behaviour of EC and PMMA thermoelectrets and different relaxation processes contributing to the observed peaks in the corresponding thermograms are available (Shrivastava et al 1978; Vanderschueren and Linkens 1978; Jiska et al 1981; Khare and Srivastava 1993, 1994; Khare et al 1993, 1994c). However, the role of various polarization processes and their relative contribution to the electret state of the polymers is not yet fully understood. Particularly, the space charge relaxation mechanism and the details of trap structure (including the trap distribution in energy and also over the volume of the polymer) are still to be well understood. Such information can best be obtained by combined study of open- and short-circuit TSCs.
In this paper we report the results of open- and short-circuit TSDC measurements carried out on EC:PMMA blends.

2. Experimental

EC and PMMA used in the present investigation were obtained from M/s Redox Chemicals, Jabalpur. Isothermal immersion technique was utilized for preparing films. Three blend samples prepared were 97:3, 93:7, 90:10 by weight proportion of ethyl cellulose and polymethyl methacrylate, and designated as P₁, P₂ and P₃, respectively. The solution was prepared in a glass beaker by first dissolving 2.4 g EC in 30 ml of chemically pure benzene. Appropriate amount of PMMA was then mixed in it. The solution was continuously stirred for about 30 min by means of a teflon-coated magnetic stirrer. Thereafter it was stirred and heated up to 60°C to yield a homogeneous solution. The solution thus prepared was poured onto clean glass plate floating over the mercury pool and the solvent was allowed to evaporate inside an air oven at 40°C. The desired samples thus obtained were subjected to room temperature outgassing at a pressure of 10⁻⁵ torr for a period of 12 h to remove any residual solvent. Such preconditioned samples were then unilaterally and bilaterally vacuum-aluminized over a central circular area of 42 mm diameter.

The samples were thermally polarized with fields of 10, 25, 50 and 100 kV/cm at temperatures 40, 60, 80 and 100°C. After polarizing for 1 h at the desired temperature, the samples were cooled to room temperature under the application of the field in 1-1/2 h. Total time of polarization was thus adjusted to be 2.5 h in each case. Polarized samples were subsequently short-circuited for an arbitrary time of 5 min so as to remove the frictional and stray charges present, if any. The short-circuit TSDCs on bilaterally aluminized samples were then recorded by reheating the samples at a linear rate of 4°C/min. For registering TSDC in open-circuit, the unilaterally polarized sample was mounted in the electrode assembly with its non-metallized surface parallel to the sensing electrode at a distance of 3 mm, while the metallized surface of the sample rested on the other metal electrode.

A high voltage power supply EC 4800 D, provided stabilized DC voltages for polarization while TSDCs were measured with the help of a Keithley 610 C Electrometer.

3. Results and discussion

The characteristic open-circuit TSDC thermograms for P₁ and P₃ polyblends polarized at various temperatures are shown in figures 1 and 2, respectively. Initially, the

![Figure 1](image1.png)  
**Figure 1.** Open-circuit TSC thermograms for EC:PMMA (97:3) blends polarized at different temperatures (40, 80 and 100°C) with 50 kV/cm field.

![Figure 2](image2.png)  
**Figure 2.** Open-circuit TSC thermograms for EC:PMMA (90:10) blends polarized at different temperatures (60, 80 and 100°C) with 50 kV/cm field.