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EXPERIMENTAL INVESTIGATION OF THE
EFFECT OF TITANIUM DIOXIDE AND BARIUM
TITANATE ADDITIVES ON DC TRANSIENT
CURRENTS IN LOW DENSITY POLYETHYLENE.

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ABSTRACT

The effect of titanium dioxide as a semi-conductive additive and barium titanate as a highly polar additive on the d.c. transient currents in low density polyethylene is investigated. Experiments were made using thick specimens under high electric field ($> 25 \times 10^6$ V/m) and constant temperature of 40°C. Results indicate that the incorporation of the foresaid additives has a remarkable effect on the d.c. transient currents in low density polyethylene.

INTRODUCTION

Transient electric currents flow from solid insulators, even though the insulators are under the influence of a constant potential difference. These currents are usually referred to as polarization, anomalous charging or discharging, absorption, after effect, residual or d.c. transient currents [1]. The measurement of these currents represents one of the methods in investigating the conductive properties of polymers. Despite the proliferation of literature on d.c. transient currents and the conductive properties of polymers, there is still little agreement about the mechanisms involved in the first [2,3], while no unified picture exists as yet in the latter [3,4]. Additives have been extensively used in polymers, not only as a means for improving certain properties such as thermal and electrical resi-

stivities, but also to provide an insight into the nature and origin of charge carriers as well as their transport mechanisms in polymers. Recently, it has been shown that additives can have a remarkable influence on space charge distribution in polymeric insulation [5,6]. The present paper describes an attempt to use titanium dioxide and barium titanate as additives to provide an insight into the mechanisms of d.c. transient current in PE and, generally, on the charge conduction and transfer in polymers.

EXPERIMENTAL DETAILS

The specimens were discs of about 2 mm thickness and 17.5 cm diameter. They were manufactured by pressing the material in a hot platten press. The materials were of plain LDPE and LDPE loaded with 1% by weight of either TiO_2 or BaTiO_3 . Mixing of the ingredients was accomplished by means of a double screw extruder prior to pressing. The discs were provided with thin insitu gold electrodes which were applied by evaporation in vacuum. The samples were thermally conditioned by placing them in an evacuated chamber ($2-3 \times 10^{-3}$ torr) at 40°C for 24 hours.

After thermal conditioning they were mounted in the test set up and electrically conditioned by applying 60 kV for 24 hours and then short circuited for 24 hours at 40°C before starting the measurements. All current measurements were conducted at 40°C and a voltage of 50 kV. Details of the measuring technique are described in [7].

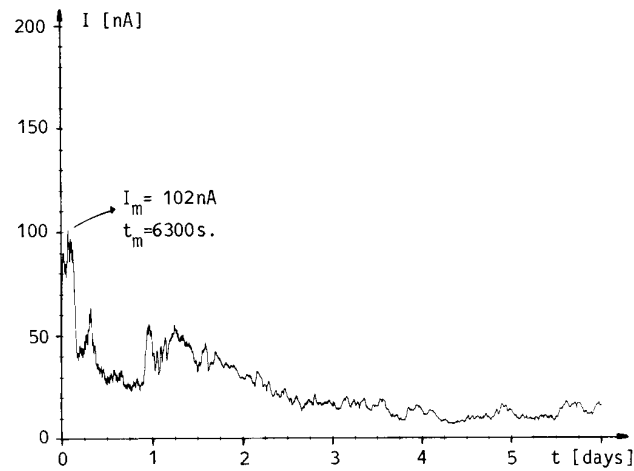


Fig. 1. Current-time dependence in plain LDPE.
D.C. voltage $V = 50$ kV, $T = 40^\circ\text{C}$.

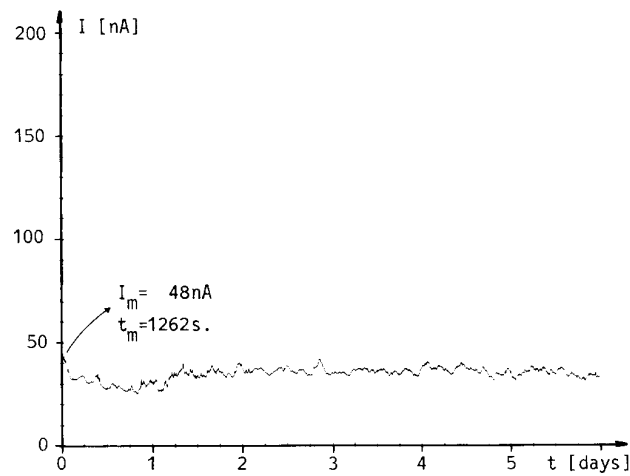


Fig. 2. Current-time dependence in TiO_2 -doped
LDPE. D.C. voltage $V = 50$ kV, $T = 40^\circ\text{C}$.

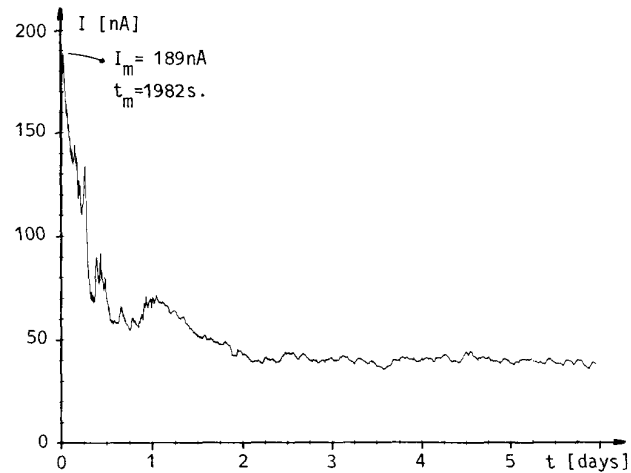


Fig. 3. Current-time dependence in BaTiO_3 -doped LDPE. D.C. voltage $V = 50 \text{ kV}$, $T = 40^\circ\text{C}$.

RESULTS AND DISCUSSION

Fig.1 shows the current time dependence in plain LDPE. It is evident that the current passes through a well defined maximum after 6300 sec. achieving a value of 102 n.A. The current then decays exponentially towards a constant final value of about 14 n.A. after about 1×10^5 sec.

Fig. 2 shows the current time dependence in TiO_2 doped LDPE. In this case the current passes through its maximum after 1262 sec. with a value of 48 n.A., and then decays towards a constant value of 35 n.A. after less than 5×10^3 sec.

Fig. 3 shows the current time dependence in BaTiO_3 doped LDPE. The current has the same pattern, arriving at a maximum value of 189 n.A. after 1982 sec. and decaying to a value 40 n.A. after about 1.7×10^5 sec.

D.C. transient currents in insulating solids can be attributed to one or more of the following mechanisms: (i) electrode polarization, (ii) dipole orientation, (iii) charge injection leading to trapped space charge effects (iv) tunneling of charges from electrodes to empty traps, (v) hopping of charge carriers through localized states. The assignment of the dominating mechanism(s), is by no means an easy task and will require experimental data covering an adequate range of field, temperature and other parameters. The transient electric current flowing in a solid insulator upon the voltage application is known, in most cases, to decay approximately by t^{-n} where t is the time after voltage application and $n \leq 1$. However, there are a few papers reporting the observation of d.c. transient current in PE passing through well defined maxima and decreasing monotonically with time of application of the voltage. [8,9]. Those maxima are also similar to the current transients maxima, according to the theory of transient space charge-limited currents in solids, earlier suggested by Many et.al. [10]. Those peaks are explained by the build up of space charge of injected carriers. The present results show a decrease in the time to maxima due to the presence of both TiO_2 and $BaTiO_3$. According to this theory, the time to maximum is in reverse proportionality with the charge carrier mobility. Thus the addition of those additives has increased the charge carrier mobility in the polymer.

As a consequence of weak molecular interactions in insulating polymers, charge transport through such materials may occur by tunneling between localized molecular electron states. The energies of these states are modified by local polarization, and this is particularly important at a metal electrode/molecular solid interface where charge transfer to the molecular states will occur. Polar additives, which will modify the energy states via their effect on

the polarization energy, will profoundly affect local charge transfer at an electrode boundary, where charge transfer will be facilitated [11].

The present results can be partly explained in terms of the previous theory. It is clear that both TiO_2 and BaTiO_3 have remarkably increased the conductivity in the polymer. The effect on TiO_2 is twofold. TiO_2 is considered to be n-type excess metal semiconductor with Ti^{+3} ions and anion vacancies occupying interstitial positions. In that case electronic relaxation polarization arises due to the thermally excited excess electrons. [12]. Thus the energy states of the polymer will be modified and the local charge transfer at the electrode/polymer interface will be encouraged. In addition to that, the existence of excess electrons in TiO_2 will increase the electronic conduction in the host material.

The presence of BaTiO_3 as a strong polar additive will again modify the energy states via its effect on the polarization energy and thus profoundly affect the local charge transfer at an electrode boundary.

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