DTU Library

Determination of charge carrier mobility in doped low density polyethylene using DC transients

Khalil, M.Salah; Henk, Peter O; Henriksen, Mogens

[Link back to DTU Orbit]()

DETERMINATION OF CHARGE CARRIER MOBILITY IN DOPED LOW DENSITY POLYETHYLENE USING D.C. TRANSIENTS.

M. Salah Khalil, P.O. Henk, M. Henriksen

ABSTRACT

Charge carrier mobility has been determined for plain and doped low density polyethylene (LDPE) using d.c. transient currents. Barium titanate has been used as a strongly polar dopant and titanium dioxide as a semiconductive dopant. The values of the mobility obtained were within the range of 10^{-10} $cm^2v^{-1}s^{-1}$. Results indicate that the inclusion of 1% by weight of the $BaTiO_3$ and TiO_2 has a considerable effect on the conduction properties of the polymer. $BaTiO_3$ has increased the charge carrier mobility by a factor of 3 and also increased the conductivity of the polymer. TiO_2 has increased the charge carrier mobility by a factor of 5. Charge trapping and space charge formation are modified by the introduction of titanium dioxide.

INTRODUCTION

Space charge formation in polymer insulation under h.v.d.c. conditions is one of the main problems in the development of h.v.d.c. cables with polymeric insulation. The use of additives represents one of the methods to modify the space charge formation in polymeric materials [1]. Additives can also be used to provide an insight into the nature and origin of charge carriers as well as the electrical conduction mechanisms in polymers. It is well known that the nature of space charge is closely related to charge carriers and trapping effects. Recently, additives have been used to modify charge trapping and conductivity properties of linear low density polyethylene LLDPE [2].

Charge carrier mobility represents one of the essential parameters characterizing electrical conduction in polymers and is also closely related to charge carrier trapping in these materials. The experimental determination of the carrier mobilities in polymers is always beset with difficulties and the recorded results vary within large ranges. Mobility can be deduced from different techniques such as the time of flight measurements and surface charge dissipation method. Carrier mobilities in some polymers have also been evaluated from step voltage transient current measurements [3,4,5]. In the present work, the effect of both barium titanate as a strongly polar additive and titanium dioxide as a semiconductive additive on the charge carrier mobility in LDPE is investigated, using measurements of d.c. transients.

M. Salah Khalil, P.O. Henk and M. Henriksen are with the Technical University of Denmark, Dept. of Electric Power Engineering.

EXPERIMENTAL DETAILS

The polymer used was a commercial LDPE used for cable insulation with the following characteristics:density 0.925 g/cm³, melt index 0.25 g/10 min. The specimens were discs, [1,8-2 mm] thickness and 175 mm diameter. LDPE was mixed with 1% by weight of either $BaTiO_3$ or TiO_2 by means of a double screw extruder prior to pressing. Gold electrodes were applied to the specimens by evaporation under vacuum after washing with pure alcohol. The specimens were thermally conditioned, under vacuum, in an evacuated chamber ($2-3 \times 10^{-3}$ torr) at 40°C for 24 hours. Electrical conditioning was performed by applying 60 kV for 24 hours,after mounting the specimens in the test set-up. The specimens were then short-circuited for 24 hours at 40°C before starting the current measurements. D.C. transient currents were measured by applying 50 kV d.c. to the specimen for 6 days under 40°C. Details of the measuring technique are described elsewhere [6].

RESULTS AND DISCUSSION

Fig. 1 shows the current time dependence in plain LDPE. In this case the current passes through a well defined peak with a value of maximum current I_m = 102 nA after a time period t_m of 6300 s after which it decays towards to its steady state value, I_{st} of 14 nA. The current time dependence for 1% by weight $BaTiO_3$ doped LDPE is shown in fig. 2. In this case I_m = 189 nA and t_m = 1982 s. The steady state current, I_{st} is 40 nA. Fig. 3 shows the current time dependence in 1% by weight TiO_2 doped LDPE. In this case, the current reaches its maximum value, I_m = 48 nA after a time period, t_m of 1262 sec, then decays fast to its steady state value which is 35 nA. It is evident in this case that the steady state current is reached much more faster when compared with the previous two cases; more over, the steady state current is about 70% that of the maximum current while it is 14% and 21% in the case of plain LDPE and $BaTiO_3$ doped LDPE respectively.

The observed current maxima show similarities to the current transients observed before by a number of authors [3,4,5]. They are explained by the build-up of space charge of injected carriers [7]. A rough estimate of the effective mobility "μ_{eff}" of charge carriers can be obtained from the time of the current maximum t_m using the equation:

$$\mu_{eff} = 0.786 \ a^2/U \ t_m \ \ldots \ldots \ cm^2 v^{-1} s^{-1} \tag{1}$$

where "U" is the applied voltage in volts, "a" the sample thickness in cm and t_m is the time of maximum current in seconds.

TABLE 1 - Time of maximum current and effective mobility.

Material	Time of maximum current t_m [sec.]	Effective mobility μ_{eff} [$cm^2 v^{-1} s^{-1}$]
Plain LDPE	6300	0.9×10^{-10}
LDPE + $BaTiO_3$	1982	2.6×10^{-10}
LDPE + TiO_2	1262	4.43×10^{-10}

Table 1 shows the effective mobility of charge carriers in the present specimens using equation 1. Barium titanate seems to increase the value of mobility and steady state current by an almost equal factor of about 3 times in comparison with plain LDPE.

Titanium dioxide has increased the mobility by a factor of 5 and the steady state current by a factor of 2.5. The pattern of the current time characteristics in TiO_2 doped LDPE (fig. 3),indicates possible modification of charge trapping and space charge formation due to the much less pronounced current peak and the fast arrival of the current to its steady state at a value very close to the peak value of the current, if compared with the case in plain and $BaTiO_3$ doped LDPE. The values of mobilities obtained in the present work compare favorably with the results obtained by other workers using different techniques as shown in table 2.

TABLE 2 - Mobility of charge carriers in PE previously determined using different methods.

Mobility	Experimental Method	Reference
$7 \times 10^{-11} - 2 \times 10^{-11}$ (T = 71°C)	Maxima of d.c. transients in oxidized LDPE	Fischer and Röhl [3]
$10^{-11} - 10^{-8}$ (T = 40°C - 80°C)	Maxima of d.c. transients in plain and oxidized LDPE and HDPE.	Ieda et al [4]
$10^{-11} - 10^{-9}$ (T = 60°C - 120°C)	Maxima of d.c. transients in graphite loaded LDPE	H.st. Onge [5]
$10^{-10} - 10^{-8}$ (T = 70°C)	Surface charge decay	Davies [8]
10^{-4} (T = 20°C)	Photo current investigation	Tanaka and Calderwood [9]

All experimental data on carrier mobility in polymers show that its value is very low, which is evidently due to trapping [10]. Trapping sites exert a strong influence on the concentration of free carriers and their mobility. Although much speculation has been made on the nature of these trapping sites, it is generally conceded that charge trapping is primarily due to the basic polymer structure. Traps due to polymeric structure can be of various types, including chemical and morphological heterogeneities and structural defects. Recent luminescence studies [11] have shown that metallic impurities such as titanium can modify the trapping effect by combining with the defect states and annihilating the trap center. The present results seem to coincide with this picture. TiO_2 is considered as an n-type excess metal semiconductor with Ti^{+3} ions and electrons occupying interstitial positions. The addition of TiO_2 will give rise to donor levels which will act as a reducing agent to annihitate some of the defect sites, thus annealing the defect states of the

system, and hence yielding the higher charge mobility observed in the present results. The presence of $BaTiO_3$, can be explained in terms of the molecular model suggested by Lewis [12] . The presence of $BaTiO_3$ as a strong polar ionic additive will modify the energy state of the polymer via its effect on the polarization energy and will profoundly affect local charge transfer at the electrodes boundaries, and hence, charge carrier mobility and conductivity. Further work is being continued to investigate the influence of additives on the conduction characteristics of polyethylene.

REFERENCES

1. Khalil, M.S., Zaky, A.A. and Hansen, B.S., 1985 Ann.Rep., Conf. on Electr. Insul. & Dielectr. Phenom, 143-149.

2. Perlman, M.M., and Haridoss, S., 1987, IEEE Trans on Electr. Insul., EI-22, 9-12.

3. Fischer, P. and Röhl,P., 1977, Prog. Colloid & Polym.Sci, 62, 149-153.

4. Ieda, M., Mizutani, T., Tsukahara, T., and Suzuoki, Y., 1979 Ann.Rep., Conf. on Electr. Insul. & Dielectr. Phenom, 106-113.

5. St. Onge, H., 1975 Ann. Rep., Conf. on Electr.Insul. & Dielectr. Phenom, 632-639.

6. Henriksen, M., Larsen, E., and Johannesson, H., 1987, Ann. Rep., Conf. on Electr. Insul. & Dielectr. Phenom, 106-113.

7. Many, A. and Rakavy, G., 1962, Phys. Rev, 126, 1980-1988.

8. Davies, D.K., 1972, J.Phys. D: Appl. Phys., 5, 162-188.

9. Tanaka, T., and Calderwood, J.H., 1974, J.Phys. D: Appl.Phys., 7, 1295-1302.

10. Kryszewski, M., 1975, J. Polymer Sci., Symposium, 50, 359-404.

11. Cheng, Y.C., Loutfy, R.O., 1980, J. Chem.Phys. 73, 2911-2918.

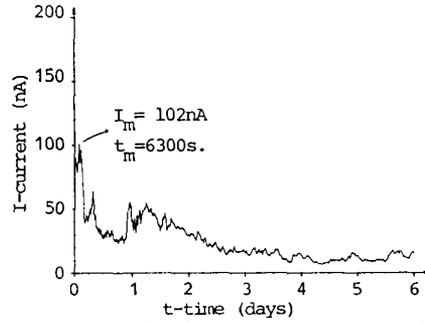12. Lewis, T.J., 1984, IEEE Trans. on Electr.Insul., EI-19, 210-216.

196

Figure 1. Current-time dependence in plain LDPE
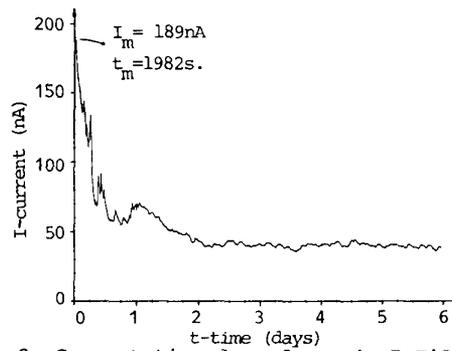D.C. voltage = 50 kV, T = 40°C.

Figure 2. Current-time dependence in BaTiO$_3$ doped
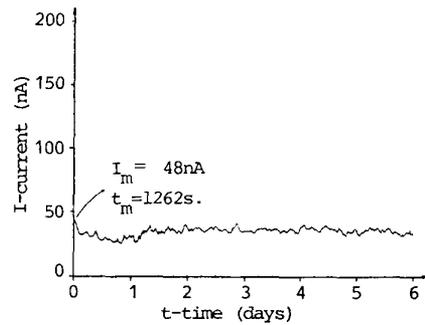LDPE. D.C. voltage = 50 kV, T = 40°C.

Figure 3. Current-time dependence in TiO$_2$ doped
LDPE. D.C. voltage = 50 kV, T $\leq$ 40°C.