

# Electret properties of ethylene–propylene random co-polymer

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## ARTICLE INFO

### Article history:

Received 9 September 2008

Received in revised form

1 January 2009

Accepted 11 January 2009

Available online 10 February 2009

### Keywords:

Ethylene–propylene co-polymer

Electret

TSC

Stable charge

## ABSTRACT

Thermally stimulated current (TSC) spectra were examined for ethylene–propylene (EP) random co-polymer at different charging voltages  $V_p$  with positive and negative polarities. Observed TSC spectra showed two well-separated TSC bands,  $B_L$  and  $B_H$ , which respectively appeared in the temperature regions below and above 100 °C. Observed  $V_p$  dependence of  $B_L$  was quite different from that of typical polypropylene homo-polymer: As  $V_p$  increased,  $B_L$  band grew keeping its peak position same at 65 °C, and the band shape unchanged, as if the traps responsible for the  $B_L$  band are a single set of traps with the same trap depth and capture cross section. The trap depth of  $B_L$  was about 1.9 eV and 1.7 eV for positively charged EP and talc-containing EP samples, respectively. EP samples also showed unique TSC bands above 100 °C: one is a narrow TSC band peaked at 120 °C and the other is an unusual TSC band which was non-vanishing even at 165 °C just before destruction of samples by their melting. Consequently, the utmost stable charge density in EP co-polymer above 100 °C was found to be  $3.5 \times 10^{-4}$  C/m<sup>2</sup> and  $6.0 \times 10^{-4}$  C/m<sup>2</sup> for positively and negatively charged samples, respectively. These equivalent surface charge densities are much larger than those of usual polypropylene homo-polymer.

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## 1. Introduction

Electret air filters in practical use are exclusively made of polypropylene because of its inexpensive cost and easy fabrication. Therefore, there have been many reports concerning material improvement by chemical modification due to acid-treatment [1], fluorinating of cellular PP [2], and by adding nucleating agents to change its crystalline state [3–6], by blending [7,8] and so on. By these improvements, unstable charges peculiar to PP, which begin to decay around 60 °C, are partly or fully eliminated from improved PP electrets. For application of electrets to sensing devices, charge stability is the most important. As for practical electrets for air filters, however, the most important factor is not stability in charge amounts but an absolute amount of stable charges. Therefore, the unstable charges peculiar to PP is not a serious disadvantage for practical electrets for air filters, provided that the electrets have sufficient amounts of stable charges.

Unlike monomer, e.g. ethylene introduced into PP main chains by co-polymerization, creates discontinuity in the main chains. This discontinuity affects on crystallization process and also has a possibility to produce crystalline defects, which act as deep traps.

For this reason, we examined the utmost amount of stable charges in deep traps in ethylene–propylene (EP) random co-polymer by the TSC method.

## 2. Experimental

Sample films used in this study were kindly supplied by Mitsui Chemical, Inc. They are 30 μm thick films fabricated by an air quenched blown method from 240 °C melt of EP random co-polymer resin (ethylene content: 0.4 mol%) with and without a magnesium silicate (talc) additive of 0.3 weight%. Talc (Hi-Filler 15DS, Matsumura Sangyo Co. Ltd.) was added with expectations that it creates crystalline defects, which act as deep traps. The weight-averaged molecular weight  $M_w$ , number averaged molecular weight  $M_n$ , melt flow rate (MFR), and crystalline melting temperature of this resin are  $1.9 \times 10^5$ ,  $2.7 \times 10^4$ , 2.4 g/10 min and 164 °C, respectively.

A circular thin Bi electrode of 20 mm diameter was vacuum deposited on one side of each sample film. The other side of the film was corona-charged in air for 3 min at different charging voltage  $V_p$  with a three electrode charging system. TSC spectrum of the charged sample was observed under open-circuit condition at a heating rate of 3.5 °C/min from room temperature to about 160 °C.

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### 3. Results and discussion

#### 3.1. Positively charged samples

TSC spectra of EP co-polymer samples, which were positively charged at different charging voltages  $V_p$  are shown in Figs. 1 and 2 for EP samples and talc-containing EP samples, respectively. As shown in these figures, observed TSC spectra have two well-separated TSC bands, a low temperature band  $B_L$  below  $100^\circ\text{C}$  and a high temperature band  $B_H$  above  $100^\circ\text{C}$ .

Comparing Fig. 1 with Fig. 2, it can be said that adding of talc did not have a large effect on the low temperature TSC band: With increasing  $V_p$ , the intensity of the  $B_L$  band increased in such a way that the band shape was almost unchanged and its peak position also kept the same temperature of  $65^\circ\text{C}$  with an exception of  $68^\circ\text{C}$  for the 1.8 kV charged sample without talc. For talc-containing samples, the band rather grows more regularly keeping its band shape and the peak position unchanged in comparison to the pure EP without talc. Observed  $V_p$  dependence of the  $B_L$  band is much more different from that of PP homo-polymer already reported [9,10]: For PP homo-polymer, intensities of low temperature bands below  $100^\circ\text{C}$  both for positively and negatively charged samples, increased in the way that the low temperature parts of these TSC bands grew with increasing  $V_p$ . Therefore, their peak position shifted toward a low temperature side and their band tails on their high temperature side formed an envelope as the charging voltage increased. This fact means that charge traps responsible for these low temperature TSC bands distribute in trap depth and their charge trapping probability decreases with decreasing trap depth because of decrease of the capture cross section in spite of increasing number of the traps. Consequently, the charge traps were filled with charges from deeper to shallower traps, that is, from high to low temperature side of the band.

As for EP co-polymer studied in this paper, the band shape and its peak position unchanged. This unchangeable nature of the  $B_L$  band for EP co-polymer, irrespective of whether it contains talc or not, leads to a conclusion that the traps responsible for this band are a single set of traps with the same trap depth and capture cross section.

Next, we examine the high temperature band  $B_H$ : Samples without talc showed only one TSC band,  $B_{120}$ , which peaked at around  $120^\circ\text{C}$  with a narrow bandwidth of about  $16^\circ\text{C}$  and

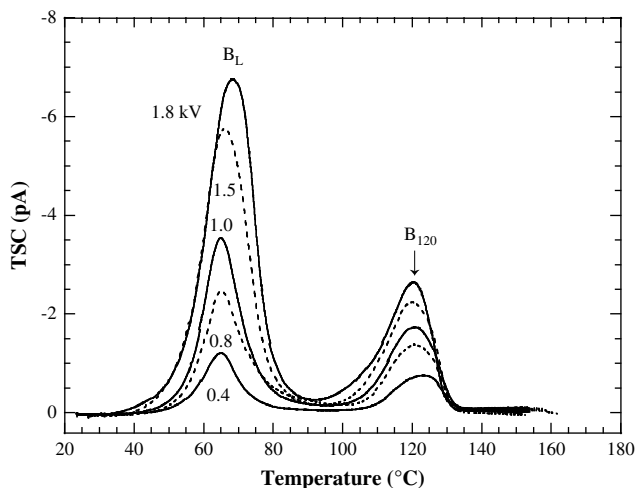


Fig. 1. TSC spectra of positively charged EP. The charging voltages are shown in the figure.

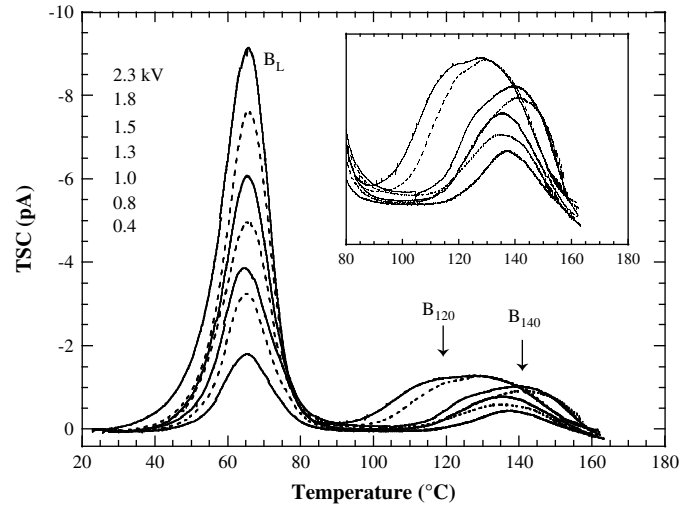


Fig. 2. TSC spectra of positively charged talc-containing EP. The charging voltages are shown in the figure in the same order of  $B_L$  band height.

disappeared in the temperature region above  $135^\circ\text{C}$ . On the other hand, talc-containing EP samples have two TSC bands in the high temperature region above  $100^\circ\text{C}$ : one is the  $B_{120}$  band and the other is a new broad band  $B_{140}$  with a peak around  $140^\circ\text{C}$  and the band extends to  $160^\circ\text{C}$ . The origin of this new band is thought to be some kind of crystalline defects introduced by talc. At low  $V_p$ , only the  $B_{140}$  band appeared and  $B_{120}$  was not observed. When the charging voltage increased to 1.5 kV, a small shoulder appeared on  $B_{140}$  as shown in the inset in Fig. 2. As the charging voltage further increased to 1.8 kV, this shoulder suddenly grew into  $B_{120}$  and the high temperature parts of  $B_{140}$  decreased. As a result, the TSC curves of 1.8 and 2.3 kV charged samples cross over those of 1.3 and 1.5 kV charged samples around  $140^\circ\text{C}$ . This fact means occurrence of some kind of charge redistribution from  $B_{140}$  to  $B_{120}$ .

Energy depth  $E_t$  of traps responsible for the  $B_L$  and  $B_{120}$  bands were estimated by the initial rise method [11]. The results are presented in Fig. 3. From Fig. 3, trap depths at low charging voltages were estimated to be 1.9 eV and 2.2 eV for  $B_L$  and  $B_{120}$  of the EP samples, and 1.7 eV for  $B_L$  of the talc-containing EP samples, respectively. Scattering of  $E_t$  values for  $B_{120}$  of the EP samples is due to fine differences in overlap of  $B_L$  and  $B_{120}$  for these samples. For

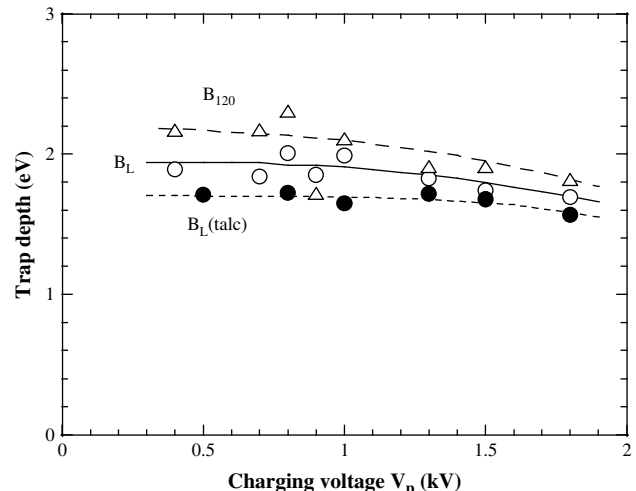


Fig. 3. Trap depths of  $B_L$  and  $B_{120}$  for positively charged EP and of  $B_L$  for talc-containing EP.

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