Bulgarian Chemical Communications, Volume 47, Special Issue B (pp. 121–126) 2015

Electret stability of gamma irradiated PP and PET films

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The influence of the gamma irradiation on electrets stability of the polymer films of polypropylene and poly(ethylene terephtalate) was studied by following the surface potential decay with time and with sample's storage temperature. The electret surface potential was measured by the method of the vibrating electrode with compensation. Polymer film samples were subjected to integral irradiation doses ($\langle E\gamma \rangle = 1.25$ MeV, ^{60}Co source) of 5 kGy and 25 kGy accumulated in air at a dose rate of 0.26 Mrad/h. After irradiation, the samples were charged in a corona discharge by means of a corona triode sistem for 1 minute under room conditions. Positive or negative 5 kV voltage was applied to the corona electrode and 1 kV voltage of the same polarity as that of the corona electrode was applied to the grid. Significant changes in the electret behaviour of the polimer films after gamma irradiation were established. The surface potential dacay depended on factors such as the corona polarity, the type of material and the irradiation dose. At 25 kGy the gamma-irradiation-induced enhancement of the electrets efficiency of the PP films achieved the highest value. The possible mechanisms of surface potential decay responsible for the observed irradiation dependent behaviour are discussed.

Key words: corona discharge, electrets, gamma irradiation, polypropylene, poly (ethylene terephthalate)

INTRODUCTION

Electrets are dielectric materials capable to retain electric charges over a long period of time and to create an external quasistatic electric field. The electret state is an important cross-scientific subject of dielectric physics, material science, sensor engineering, medical and bio-engineering [1–3]. Over the years, a considerable interest in the surface potential decay of corona-charged polymeric materials has been shown. Besides the electret's material and conditions of electret's manufacturing the surface potential decay depends on a number of factors under which the electrets have been stored or used, for example: temperature, humidity, pressure, gamma irradiation etc. [4–6]. The influence of these factors on the charge decay has been studied in order to obtain stable electrets for numerous applications. However, there are only a few publications about the influence of gamma irradiation on the charge decay of polymer electret films. In [7] the gamma irradiation effect (up to 100 kGy) on the electret behaviour of corona charged biobased polymer films of polylactic acid have been investigated. It has been shown by viscometric, DSC, and SEM studies that the degradation process is predominant. As compared to the non-irradiated samples the values of the surface potential of the irradiated samples, independently of the corona polarity and irradiation dose, were higher. In [8] the surface potential decay after dc corona charging of gamma irradiated low density polyethylene (LDPE) has been studied. The irradiation was carried out in air, nitrogen gas and vacuum to investigate the effect of the irradiation environment on the surface potential of LDPE. Significant changes occurred in the surface potential decay characteristics in dependence on irradiation dose. The dynamic behavior of the surface charge of gamma rays irradiated polybutylene naphthalate has been studied in [9] by applying a negative bias voltage between two aluminum plate electrodes. It was established that with the increase of the total dose of gamma irradiation, both the capacity of surface charge and the rate of charge decay decrease. The charge accumulation depends upon the density of localized surface states, which is varied by the radiation induced cross-linking and the degradation reactions of the molecule structure. The decay is caused by the recombination of surface charge with ions of the opposite sign in air. The gamma irradiation effect on the chain segment motion and charge detrapping in polyamide 610 has been investigated by means of thermally stimulated depolarization current in [10]. The gamma irradiation increases the stability of trapped charge in both amorphous phase and interphase, but does not affect the stability of trapped charge in crystalline phase. The effect of 1.25 MeV gamma radiation on the structural properties of pristine and gamma irradiated (0–2000 kGy) poly(ethylene terephthalate) polymer sam-

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ples have been analyzed in [11] and certain increase in the crystallinity was observed. Polymeric materials as polypropylene (PP) and poly(ethylene terephthalate) (PET) are widely used for the formation of electrets, because of their important commercial significance, structure and appropriate mechanical and electrical properties [12, 13]. In the literature however, there are no data available for the influence of gamma irradiation on the charge decay of PP and PET electret films. Therefore, we undertook such a study on some polymers. The present paper reports on the influence of low dose gamma irradiation on the stability of electret characteristics of corona charged PP and PET films.

EXPERIMENTAL

Formation of gamma irradiated polymer films

Initially, the 20 μm PP and 40 μm PET films were cleaned in an ultrasonic bath with alcohol for 4 minutes, followed by rinsing with distilled water and drying on filter paper in air at ambient temperature. Samples of 30 mm diameter were cut from the clean films and subjected to gamma irradiation treatment. The irradiations were performed in air by a ^{60}Co source with total doses of 5 kGy and 25 kGy accumulated in a single step at a dose rate of 0.25 kGy/h. It was checked by thermometric control that the sample temperatures during the irradiation did not appreciably increase above room temperature. This was expected because of the low dose rate of the irradiation.

Corona charging and surface potential measurement of the samples

The charging of the samples in a corona discharge was carried out by a conventional corona triode system (Fig. 1), consisting of a corona electrode (needle), a grounded plate electrode and a grid placed between them. The distance between the corona electrode and the grid was 10 mm and the distance between the grid and the grounded plate electrode was 3 mm.

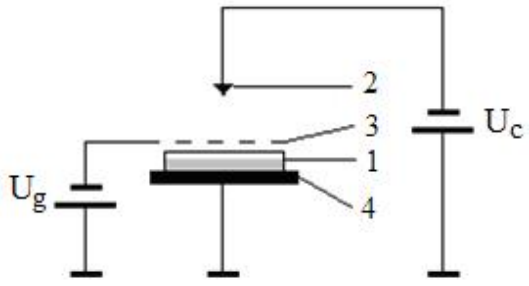

Fig. 1. Corona charging set-up: 1 – sample on a metal pad; 2 – corona electrode; 3 – grid; 4 – grounded plate electrode; U_g – grid voltage power supply; U_c – corona voltage power supply.

The samples of the non-irradiated (0 kGy) and gamma irradiated (5 kGy and 25 kGy) PP and PET films were charged at a room temperature for 1 minute. Positive or negative 5 kV voltage was applied to the corona electrode. Voltage of 1 kV of the same polarity as that of the corona electrode was applied to the grid. The electret's surface potential of the charged samples was measured by the vibrating electrode method with compensation and the estimated error was less than 5%.

RESULTS AND DISCUSSION

Influence of time storage on electrets surface potential decay

The dependences of normalized surface potential on the time of storage under room conditions for positively and negatively charged non-irradiated and gamma irradiated PP and PET films have been followed for 115 days. The surface potential was measured once a week except for the first 30 days because the charge was rapidly decaying. After this period (115 days) steady state values of the surface potential were established for all of the samples.

Time dependences of the normalized surface potential for PP electrets charged in a positive or in a negative corona are presented in Fig. 2 and Fig. 3, respectively. Similar dependences for PET electrets are presented in Fig. 4 and Fig. 5, respectively.

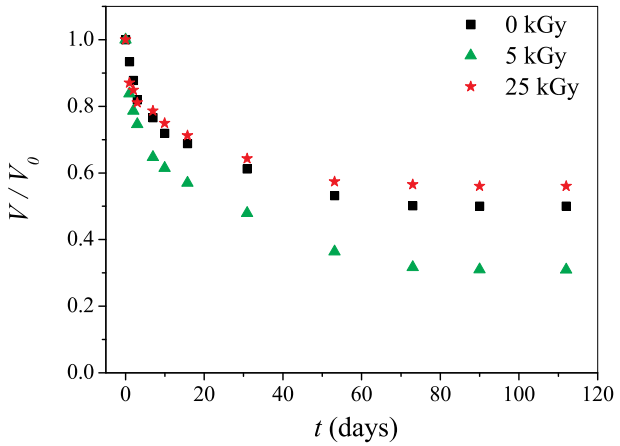

Fig. 2. Time dependences of the normalized surface potential for positively charged non-irradiated and gamma irradiated PP films.

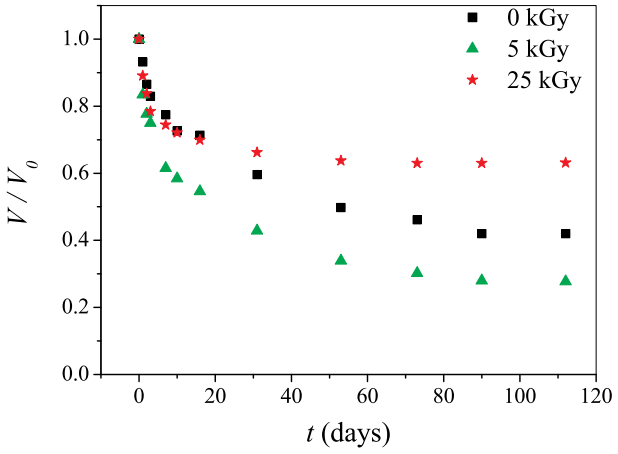

Fig. 3. Time dependences of the normalized surface potential for negatively charged non-irradiated and gamma irradiated PP films.

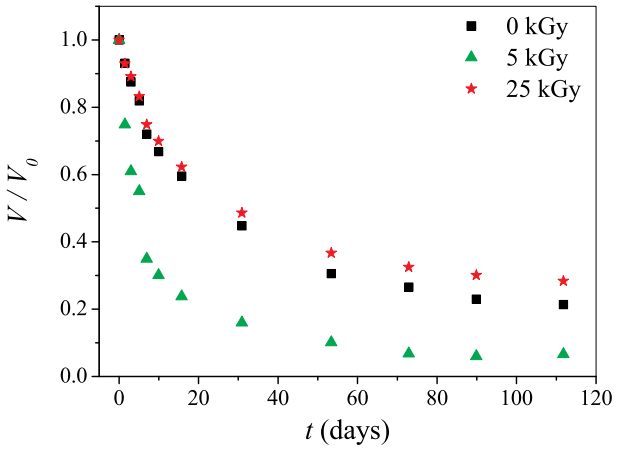

Fig. 4. Time dependences of the normalized surface potential for positively charged non-irradiated and gamma irradiated PET films.

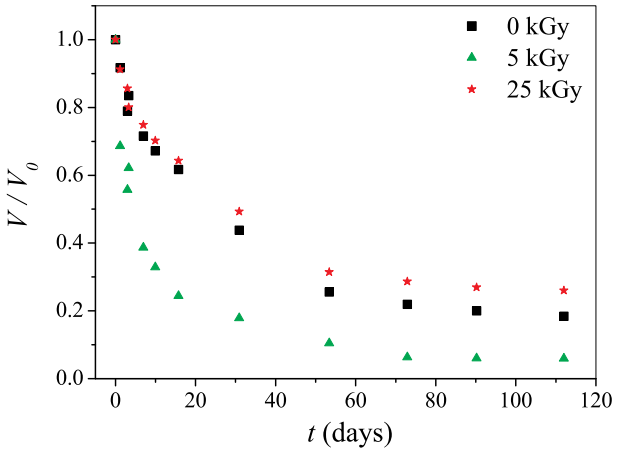

Fig. 5. Time dependences of the normalized surface potential for negatively charged non-irradiated and gamma irradiated PET films.

Each point in the figures is a mean value from 6 samples. The calculated standard deviation was better than 5% from the mean value with confidence level 95%.

The results, presented in Figs. 2-5 show the following peculiarities:

- For all investigated samples the normalized surface potential are initially decaying exponentially for the first 30 days and then are slowly decreasing and are practically stabilized to the 115th day.

The value of the surface potential of electrets depends on the amount of trapped charges in the different localized surface states of the samples. In the initial period of time after the charging in the corona, the surface potential rapidly decreases due to the release of the weakly captured charges from the shallow energy states. Then the surface potential stabilizes to a steady state value caused by the tightly captured charges in the deep energy traps.

- The final values of the normalized surface potential are higher for the PP electrets than those for the PET electrets irrespective of the corona polarity. We assume that this is due to the different structures of the polymers, which leads to the formation of different localized surface states that capture electrical charges.
- The surface potential values of the samples irradiated by dose of 25 kGy, independently of the corona polarity and the type of material were higher in comparison with the non-irradiated samples and the irradiated ones by dose of 5 kGy. The final values of the normalized surface potential for the PP electrets charged in a negative corona and irradiated by dose of 25 kGy are the highest.

Therefore, the gamma irradiation by a greater dose of 25 kGy leads to an increase of the electrets surface potential value and to obtain the stable electrets. When the semicrystalline polymers as PP and PET are exposed to irradiation, the crystalline lamellas may be broken into two or more portions, whereas the long molecular chains of amorphous regions may be broken at different places, leaving a free smaller chains which immediately comes to a stable position by collecting its whole length into regularly arranged lamella form, of course, of smaller size [14]. Probably the low dose of 5 kGy causes changes that have blocked the formation of charge trapping surface state but the increase of dose to 25 kGy leads to a density

increase of the localized surface states and the captured surface charge increase.

Influence of the temperature storage on electrets surface potential decay

The dependences of the normalized surface potential on the temperature for positively and negatively charged non-irradiated and gamma irradiated PP and PET films were investigated. The surface potential measurements were performed by means of the surface potential measurement set-up at elevated temperatured. The surface potential was measured every two minutes for two hours at a constant temperature increase rate – 1.2°C/min. Temperature dependences for the normalized surface potential for PP electrets charged in a positive or in negative corona are presented in Fig. 6 and Fig. 7, respectively. Similar dependences for PET electrets are presented in Fig. 8 and Fig. 9, respectively.

The results presented in Figures 6-9 show that for all investigated samples two relaxation processes are observed.

The first process corresponds to the decay of the normalized surface potential in the range of 22°C to 40°C for positively and negatively charged PP samples and in the range of 22°C to 55°C to positively

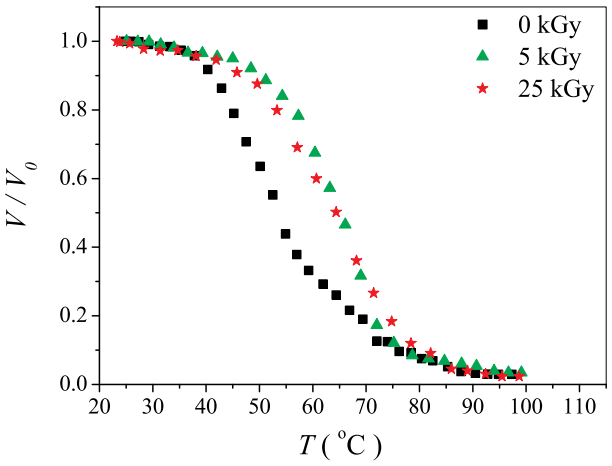

Fig. 6. Temperature surface potential decay curves for positively charged non-irradiated and gamma irradiated PP films.

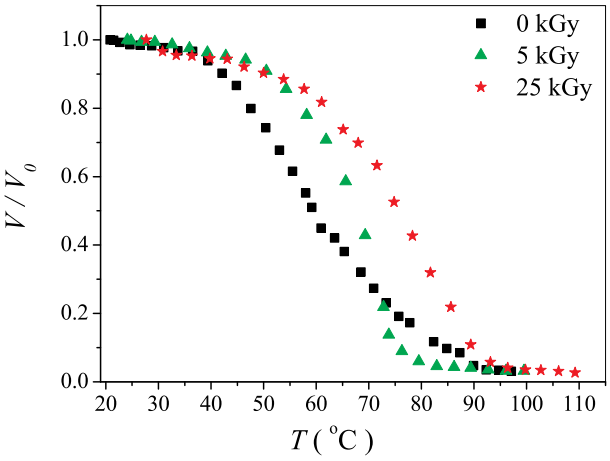

Fig. 7. Temperature surface potential decay curves for negatively charged non-irradiated and gamma irradiated PP films.

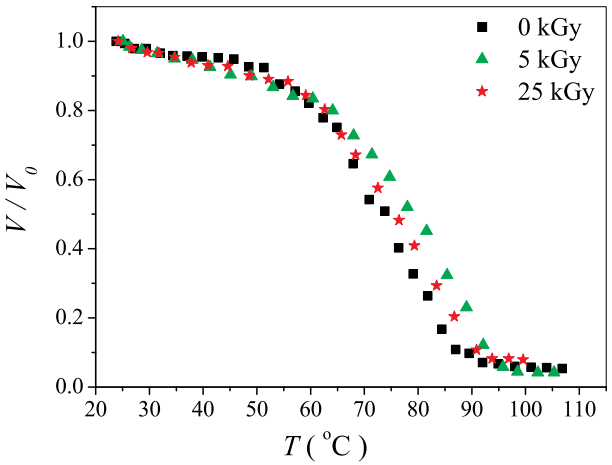

Fig. 8. Temperature surface potential decay curves for positively charged non-irradiated and gamma irradiated PET films.

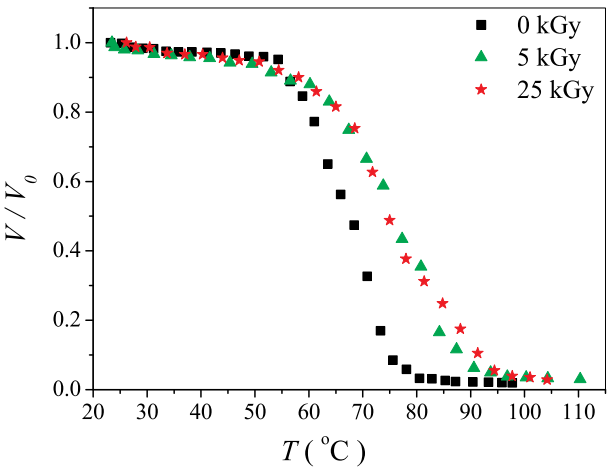

Fig. 9. Temperature surface potential decay curves for negatively charged non-irradiated and gamma irradiated PET films.

and negatively charged PET samples. These is the low energy process, i.e. it was observed at temperatures near room temperatur, and it is most likely due to the release of the weakly captured charges from the shallow energy states.

The second relaxation process depends on the type of material (PP or PET), the corona polarity (positive or negative) and the dose of irratiation (0 kGy, 5 kGy, 25 kGy). In Table 1 the temperature ranges for the second decay of the normalized surface potential are presented.

Table 1. Temperature ranges for the second decay of the normalized surface potential

Materials	Corona polarity	ΔT, °C 0 kGy	5 kGy	25 kGy
PP	positive	43–67	48–75	46–78
PP	negative	44–82	51–74	50–89
PET	positive	54–84	57–92	56–85
PET	negative	57–73	60–87	58–91

It can be seen from the data presented in Table 1 that the second process is observed at higher temperatures. According to our experimental data from DSC measurements with DSC 204 F1 Phoenix NETZSCH, Germany it has been established that there is a release of energy at these temperatures. Hence, we could assume that the charges are released from the deep energy traps and a sharp decrease of the surface potential is observed. The decrease of the normalized surface potential is observed. The decrease of the normalized surface potential is shifted to the higher temperatures for gamma irradiated samples compared to the non-irradiated samples, irrespective of the radiation dose. Consequently, the gamma irradiation of different polymer materials leads to creation of high energy traps. It has been established that the temperatures at which the decrease of the surface potential is shifted are higher for samples of PET, compared to those of PP, irrespectively of the corona polarity. Threfore, the generated charge trapping surface state in PET are more highly energetic than those in PP.

CONCLUSION

The observed significant changes in the electret behaviour of the polymer films induced by gamma irradiation are of complex origin. The surface potential decay depends on several factors. The corona polarity, polymer matrix type and gamma irradiation dose are factors that interplay. The PP electrets charged in a negative corona and irradiated by dose of 25 kGy achieved the highest surface potential. The possible surface potential decay mechanisms responsible for the above may be due to degradation, scissioning and crosslinking of the polymer chains with the increasing dose of gamma irradiation and formation of different charge trapping surface states.

Acknowledgements. *This paper was performed with the financial support of the projects: MU-15-FFIT-003, Department of Scientific Research at the Plovdiv University "Paisii Hilendarski" and BG161PO003-1.2.04-0012-C0001 "Enhancing the capacity of the Plovdiv University for applied research by equipping laboratories for biocompatible materials and molecular biosensors".*

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ЕЛЕКТРЕТНА СТАБИЛНОСТ НА γ-ОБЛЪЧЕНИ ПОЛИМЕРНИ ФИЛМИ ОТ ПОЛИПРОПИЛЕН И ПОЛИЕТИЛЕНТЕРЕФТАЛАТ

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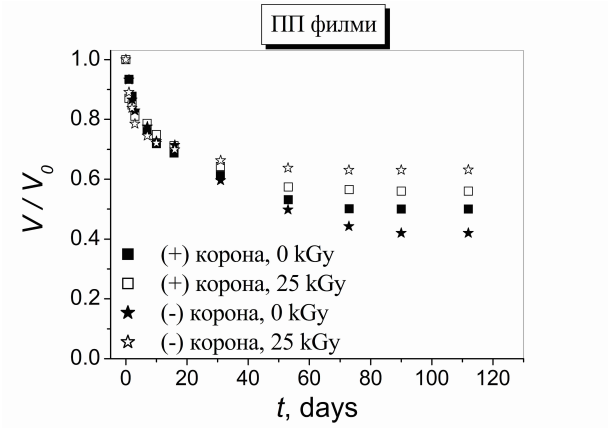
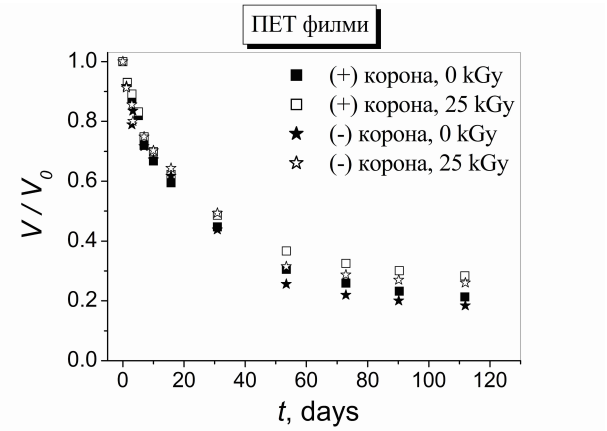
(Резюме)

В настоящата работа е изследвано влиянието на γ-облъчването върху електретната стабилност на полимерни филми от полипропилен (ПП) и полиетилентерефталат (ПЕТ). Първоначално ПП и ПЕТ филмите са облъчвани с кобалтов източник ^{60}Co с различни дози: 0 kGy, 5 kGy и 25 kGy. След това облъчените образци са зареждани в коронен разряд с помощта на триелектродна система в продължение на 1 минута при стайна температура. На корониращия електрод е подавано напрежение ±5 kV, а на решетката напрежение ±1 kV със същата полярност както на корониращия електрод. Повърхностният потенциал на получените електрети е измерван по метода на вибриращия електрод с компенсация.

За определяне на влиянието на γ-облъчването върху стабилността на електретните филми е изследвано спадането на повърхностния потенциал с времето и с температурата на съхранение на образците. Времевите зависимости на нормирания повърхностен потенциал са показани на Фиг. 1 и Фиг. 2.



Фиг. 1. Времеви зависимости на нормирания повърхностен потенциал за необлъчени и γ-облъчени ПП образци.



Фиг. 2. Времеви зависимости на нормирания повърхностен потенциал за необлъчени и γ-облъчени ПЕТ образци.

Получените резултати показаха значително изменение в електретното поведение на полимерните филми след облъчването им с различни дози γ-лъчи. Беше установено, че спадането на повърхностния потенциал зависи комплексно от няколко фактора: полярност на короната, вид на материала и доза на облъчване. В статията са дискутирани механизмите, отговорни за спадането на повърхностния потенциал. Експериментално беше доказано, че γ-облъчването с доза 25 kGy води до увеличаване на електретната ефективност на изследваните филми.