

# Positron annihilation and differential scanning calorimetric study of poly(trimethylene terephthalate)/EPDM blends

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## Abstract

The blends of poly(trimethylene terephthalate) (PTT) and ethylene propylene diene monomer (EPDM) with different composition have been studied by positron lifetime technique (PLT) and differential scanning calorimetric (DSC) measurements. The DSC results for the blends of 50/50 and 40/60 show clear two glass transition temperatures indicating two-phase system. No melting point depression was observed for the blend system, which strongly supports the incompatibility. From the positron results an increase in free volume hole size and its concentration has been observed with the increase in EPDM content of the blend which indicates further that there is coalescence of free volumes of EPDM with the PTT to some extent and phase separation behavior continues. Another interesting aspect is that the relative fractional free volume exhibits neither negative nor positive deviation from the log additivity rule. It agrees well with the log additivity rule. The interchain interaction parameter evaluated from these results show some complex behavior. XRD results show the decrease in crystallinity of the blend with the increase in fractional free volume with the increase in concentration of EPDM.

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**Keywords:** *o*-Ps lifetime; Free volume; Differential scanning calorimetry

## 1. Introduction

Over the last few decades, it has been realized that many physical and mechanical properties of the polymers can be significantly improved by a process called blending [1]. Phase separation phenomena and miscibility of polymer blends have been pursued with great attention in polymer applications [2]. Understanding the nature and the underlying factors of blending at the molecular level is essential in order to predict and enhance the material properties of the blends. Rubber/plastic blends have been commercialized as rubber toughened plastics otherwise called thermoplastic elastomers (TPEs) [1]. Usually, if the blend contains relatively large composition of hard plastic, it can be used as an impact resistant plastic, whereas, if the blend contains more of rubber phase, the blend will be soft and will have at least some properties of thermoplastics [2]. It is, however,

known that most of the thermoplastic-rubber blends are immiscible and incompatible. In such blends, generally the physical, mechanical and viscoelastic properties depend not only on the constituent polymers but also on the morphology of the blends. Since the minor phase in an immiscible blend is of deformable nature [2], it is expected a wide range morphologies (size, shape and distribution of the dispersed phase) result during the blend processing. Many investigations have been focused on this aspect in the past [1]. In the present paper, we used a novel technique like positron lifetime spectroscopy to measure free volume size and its content in an immiscible system of PTT/EPDM blend in the light of theoretical models to understand the molecular level microstructure of the blend. Differential scanning calorimetry is used to characterize the glass transition temperature and the melting point of the blends.

There exist many physical probes for characterizing the structures and properties of polymer blends. However, only a limited number of probes are available for characterizing free volume properties. In recent years, positron

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annihilation spectroscopy has emerged as a unique and potent probe for characterizing the free volume properties of polymers [3]. Positron annihilation lifetime spectroscopy (PALS) is one of the sophisticated tools available for determining directly the angstrom sized free volume holes and their relative number density (concentration) in polymers [4–7]. To mention a few of these studies using this technique, diffusion kinetics in electron irradiated polycarbonate (PC) [8], water sorption in contact lens polymers [9], polymers containing silver nanoparticles [10], structural relaxation of polyethylene (PE) in the presence of silver oxide [11], etc. trust to show the versatility of the technique.

When positrons from a radioactive source (commonly Na-22) injected into a molecular medium, it reaches thermal energy in a very short time (around 1 ps), after interacting with surrounding molecules through elastic collision processes. During such a slowing down process and at nearly thermalized stage, a positron, the antiparticle of the electron, may pick up an electron from the medium and annihilate as a free positron, or get trapped in to defects present in crystalline, crystalline–amorphous interface regions and then annihilates, or form a bound state with an electron of the medium ( $e^+e^-$ ) called the positronium (Ps) atom. Ps exists in two allowed spin states: *para*-positronium (*p*-Ps), if the spins are aligned antiparallel, or *ortho*-positronium (*o*-Ps) if the spins are parallel. Due to the fact that *o*-Ps is preferentially localized in the free volume holes of polymer systems [4–13], the positron parameters  $\tau_3$  and ( $I_3$ ) have been widely used over the last few years in the study of microstructural behavior of this class of materials. It is usual to correlate to the relative concentration of the free volume holes to the viscoelastic properties of the system under investigation. Even though PLT has been successfully used in the study of polymers for more than two decades, only recently it has been used to study microstructural behavior of polymer blends [14,15].

## 2. Experimental

### 2.1. Blend preparation: melt blending

Samples of PTT and EPDM having density 1.33, 0.86 g/cc and the number average molecular weight of 38,000 and 60,000, respectively, were melt blended. The blends are designated as P100E0, P90E10... where P stands for PTT and E stands for EPDM, and the numbers indicate the weight percent of PTT and EPDM, respectively, in the blend. The blends were prepared in an internal mixer with a rotor speed of 60 rpm; the total mixing time was fixed as 4 min. PTT was melted first at a temperature of 230 °C and then EPDM was added after 2 min. Blending was continued for two more minutes. The blends having PTT/EPDM concentration 100/0–0/100 were prepared by melt blending.

The samples were compression molded at 230 °C with a pressure of 20 kg/cm<sup>2</sup> for 2 min into sheets (Fig. 1).

### 2.2. Differential scanning calorimetric measurements

The glass transition temperature ( $T_g$ ) of the homopolymers and their blends were measured using a Universal V3.0G TA instrument DSC 2010 connected to liquid nitrogen cooling accessory (LNCA) with a nitrogen purge. Each experiment began with cooling from room temperature to  $-65$  °C at a rate of 10 °C/min, then the samples were allowed to equilibrate for 10 min before starting the measurements. The glass transition temperature, and the melting point of the pure and blends of 80/20, 50/50, 40/60 (PTT/EPDM) samples weighing 10–20 mg were investigated with the heating rate of 10 °C/min from  $-65$  to 250 °C. The values of  $T_g$  and  $T_m$  so obtained from DSC scans are reported in the Table 1.

### 2.3. Positron annihilation lifetime measurements

Positron annihilation lifetime spectra were recorded for the homopolymers and the blend samples using positron lifetime spectrometer. The positron lifetime spectrometer consists of a fast–fast coincidence system with BaF<sub>2</sub> scintillators coupled to photo multiplier tubes type XP2020/Q with quartz window as detectors. The BaF<sub>2</sub> scintillators were shaped to conical to achieve better time resolution. The two identical pieces of the sample were placed on either side of a 17  $\mu$ Ci <sup>22</sup>Na positron source, deposited on a pure kapton foil of 0.0127 mm thickness. This sample–source sandwich was placed between the two detectors of PLT to acquire lifetime spectrum. The Co<sup>60</sup> prompt spectrum gave 180 ps as the resolution function. However, to have increased count rate, the spectrometer was operated at 220 ps. All lifetime measurements were performed at room temperature and two to three positron lifetime spectra with more than a million counts under each spectrum were recorded in a time of 1–2 h. Consistently reproducible spectra were analyzed into three lifetime

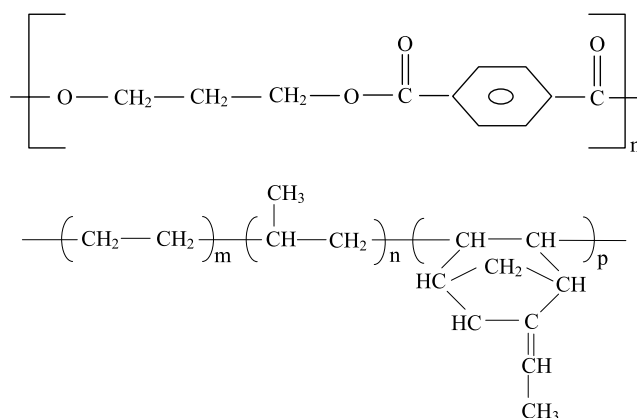


Fig. 1. Chemical structure of PTT and EPDM.

Table 1  
Glass transition temperature evaluated from DSC data

EPDM (wt%)	$T_{g1}$ (°C)	$T_{g1}$ (°C)	$T_{g1}$ (°C)	$T_{g1}$ (°C)	$\Delta H_m$ (J/g)
0	–	–	230	50.00	109.9
20	–	–	230	54.11	48.85
50	–52.00	–	229	54.29	33.20
60	–48.00	42.00	230	52.00	35.6
100	–55.00	42.60	–	–	–

components with the help of the computer program PATFIT-88 [16] with proper source and background corrections. Source correction term and resolution function were estimated from the lifetime of well-annealed aluminum using the program RESOLUTION [16]. Since the single Gaussian resolution function did not converge the lifetime spectrum, the resolution function was resolved further into three Gaussian components, which gave quick and good convergence. The net resolution function for this turned out to be 220 ps. The three Gaussian resolution functions so determined were used to estimate the lifetime parameters of some of the well-characterized polymers like polycarbonate, PTFE, etc. The lifetime parameters were found to be in good agreement with the values reported in the literature [8]. Therefore, the three Gaussian resolution functions were used in the present analysis of positron lifetime spectra in all the blend and homopolymer samples.

#### 2.4. X-ray diffraction measurements

X-ray diffraction scans were obtained on a Rigaku diffractometer (Japan) Model D/Max-IC in para focus geometry with the chromium target ( $\lambda = 2.2892 \text{ \AA}$ ) and a graphite monochromator in the diffracted beam. The measurements of the scattered intensity at different angles ( $2\theta$ ) starting from 5 to 60° were taken to study the state of the polymer samples in terms of the crystallinity of the samples.

### 3. Results and discussion

#### 3.1. Differential scanning calorimetric results

Differential scanning calorimetric experiments were performed to characterize the  $T_g$ ,  $T_m$  of the blends. As suggested [1] these measurements help to distinguish whether the blending has resulted in phase separation or a single-phase system. It is known that  $T_g$  of the polymers and polymer blends is expected to depend on, the well-known factors such as the chain structure and conformation [2], degree of crystallinity and dispersion. Figs. 2 and 3 show the DSC curves for the PTT, EPDM and 80/20, 50/50, 40/60 blends respectively. In the present measurement  $T_g$  is taken as the onset of the transition. PTT is a crystalline polymer, which exhibits the  $T_g = 50 \text{ °C}$ , and a well-defined crystalline melting point of 230 °C with  $\Delta H_m = 109 \text{ J/g}$ , where  $\Delta H_m$  is

the change in enthalpy of the system. EPDM is an elastomer with  $-55$  and  $42 \text{ °C}$  as the  $T_g$  and  $T_m$ , respectively. In 50/50 and 40/60 blends, a low temperature  $T_{g1}$  and a high temperature  $T_{g2}$  are observed indicating micro phase separated structures consisting of micro domains of rubbery EPDM and glassy PTT segments [17]. Higher glass transition ( $T_{g2}$ ) is related to the rich PTT phase, whereas lower one corresponds to EPDM [18]. The melting point of the EPDM overlaps with the  $T_g$  of PTT in 40/60 blend, because of the melting component percentage is more, the heat flow signal produces a significant change in the base line, which is characteristic of heat capacity changes [18]. However, in the case of the blend with 80/20 composition, there appears to be a single  $T_g$  around 54 °C. Since the rubber concentration in this blend is only 20% the  $T_{g1}$  is not pronounced and can be considered as weak. However, it may be seen as an indication to the slight interaction between the two polymer components. It has been the general practice that blends can be judged miscible on the basis of a single  $T_g$  but there are some different argument against to this, i.e.  $T_g$  is surely a measure of the degree of dispersion but not of miscibility, and even if the blend is miscible at  $T_g$ , it does not necessarily mean that the thermodynamic miscibility exists under the rheological test conditions [1].

The melting point depression is normally observed for the homogeneous mixtures where the degree of interaction is high. The melting point depression is one way of measuring the interaction parameter ( $\chi_{12}$ ) of a crystallizable polymer in a homogeneous mixture in a solvent, plasticizer or second polymer. In a homogeneous mixture, the presence of second polymer decreases the melting point of the other by reducing crystallite size, and the interaction of two polymers is more favorable [2,14,19]. Fig. 3(b) shows the melting points of 80/20, 50/50 and 40/60 blends. It is important to note that all the blends exhibit a crystalline phase with a constant  $T_m$  of  $(230 \pm 2 \text{ °C})$ . The fact that no melting point depression is observed from the DSC results of 80/20, 50/50, 40/60 blends, strongly supports the formation of immiscible blends [19,20]. The endothermic melting of PTT completely dominates the features of the DSC curves. An overall analysis of the thermal data obtained in this work is tabulated in Table 1 and these results indicate that the blends studied were incompatible and the interaction between two components is very poor. As the EPDM concentration increases, we observe from Table 1 that  $\Delta H_m$  goes on decreasing. This suggest that the

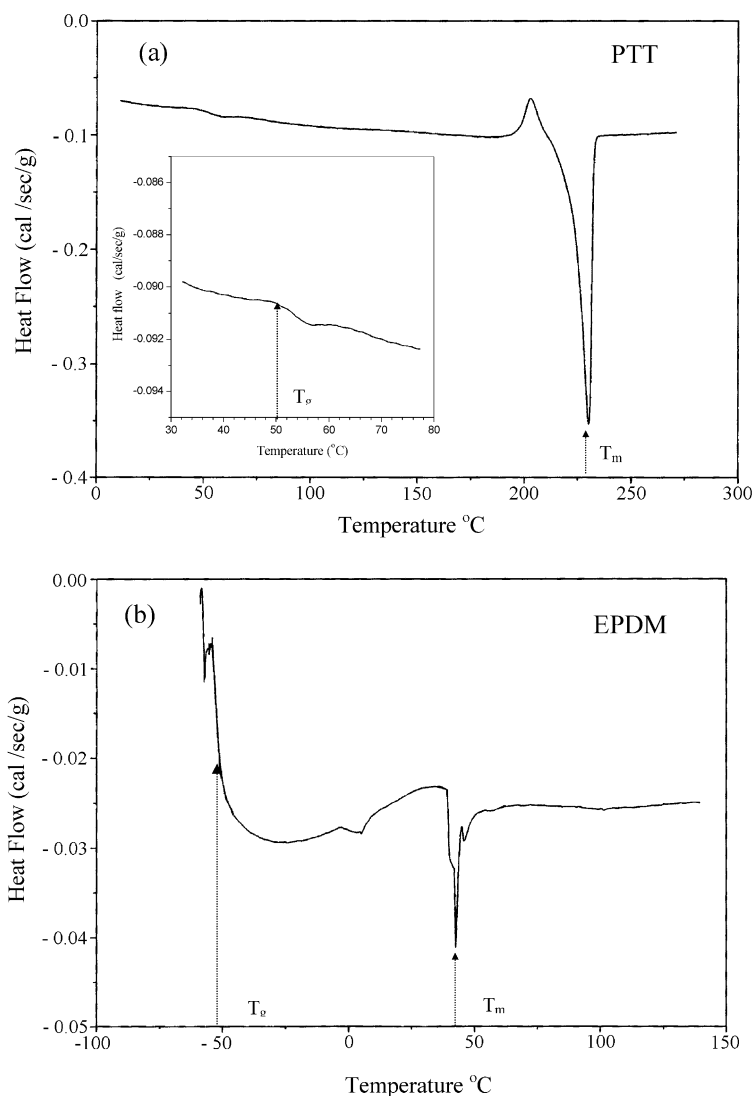


Fig. 2. (a) DSC scans of PTT. The insert shows higher magnification of  $T_g$  region. (b) DSC scans of EPDM.

crystallinity of the blend decreases with increase in EPDM component.

### 3.2. Positron annihilation lifetime results

The acquired lifetime spectra were resolved into three lifetime components  $\tau_1$ ,  $\tau_2$  and  $\tau_3$  with intensities  $I_1$ ,  $I_2$  and  $I_3$ , respectively. All the lifetime spectra were resolved into three lifetime components since it gave better chi square value and standard deviations than the two and four component analyses. The attribution of these lifetime components is as follows [5]. The shortest lifetime component  $\tau_1$  with intensity  $I_1$  is attributed to *para*-positronium (*p*-Ps) and free positron annihilations. The intermediate lifetime component  $\tau_2$  with intensity  $I_2$  is usually considered to be mainly due to annihilation of positrons trapped at the defects present in the crystalline regions or trapped at the crystalline–amorphous interface regions. Since  $\tau_2$  is not the main focus of the work, the second lifetime is not discussed in this

paper. The longest-lived component  $\tau_3$  with intensity  $I_3$  is due to pick-off annihilation of the *ortho*-positronium (*o*-Ps) in the free volume sites present mainly in the amorphous regions of the polymer matrix [21]. The *o*-Ps lifetime  $\tau_3$  is related to the free volume hole size by a simple relation given by Nakanishi et al. [22], which was developed on the basis of theoretical models originally proposed by Tao [23] for molecular liquids and later by Eldrup et al. [24]. In this model, positronium is assumed to be localized in a spherical potential well having an infinite potential barrier of radius  $R_0$  with an electron layer in the region  $R < r < R_0$ . The relation between  $\tau_3$  and the radius  $R$  of the free volume hole or cavity is

$$(\tau_3)^{-1} = 2 \left[ 1 - \frac{R}{R_0} + \frac{1}{2\pi} \sin \left( \frac{2\pi R}{R_0} \right) \right] \text{ns}^{-1} \quad (1)$$

where  $R_0 = R + \delta R$  and  $\delta R$  is an adjustable parameter. By fitting Eq. (1) with  $\tau_3$  values for known hole sizes in porous

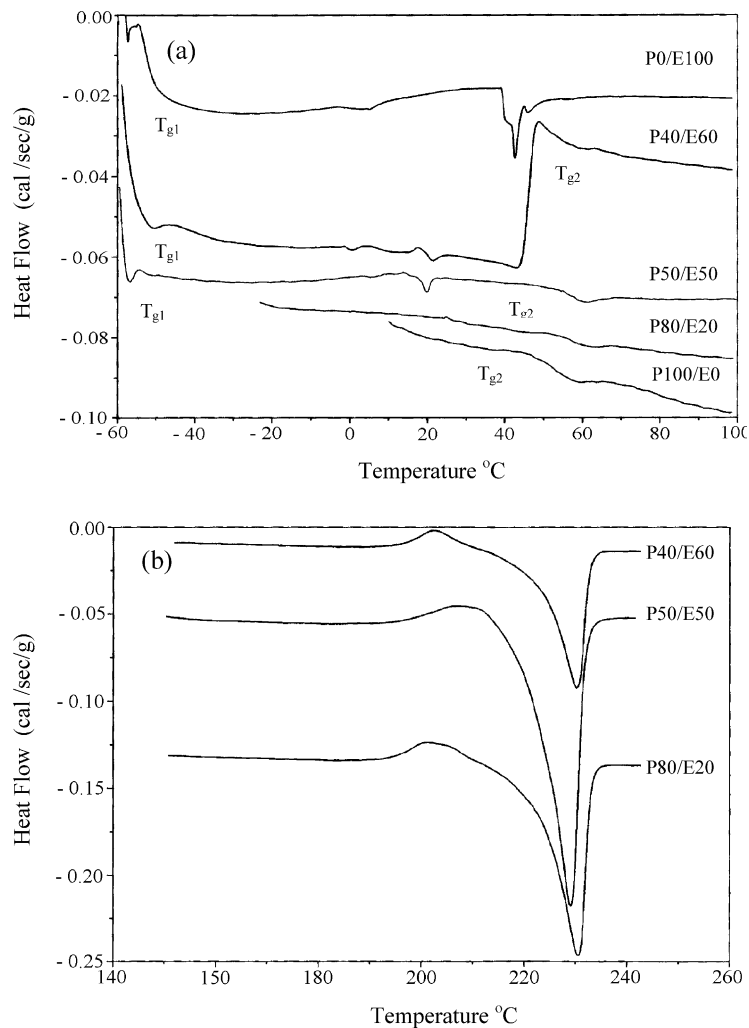


Fig. 3. (a) DSC scans for the PTT/EPDM blends at 0, 20, 50, 60, and 100 wt% of EPDM. (b) DSC scans for the PTT/EPDM blends at 20, 50, and 60 wt% of EPDM representing the melting points.

materials like zeolites, a value of  $\delta R = 0.1657$  nm was obtained. With this value of  $\delta R$ , the free volume radius  $R$  has been calculated from Eq. (1) and the average size of the free volume holes  $V_f$  is evaluated as  $V_f = (4/3)\pi R^3$ . The fractional free volume or the free volume content ( $F_v$ ), can then be estimated as

$$F_v = CV_f I_3 \quad (2)$$

where  $C$  is structural constant,  $V_f$  and  $I_3$  are the parameters described above. The parameter  $C$  has to be estimated from a different experiment. In the present case, we use the relative fractional free volume  $F_v = V_f I_3$  in the absence of  $C$  to understand the changes in fractional free volume.

Therefore, it seems appropriate that we consider only *o*-Ps lifetime  $\tau_3$  and its intensity  $I_3$  in understanding the changes in free volume properties with the concentration of EPDM. Since the Ps atom probes the local molecular environment and the free volume is the result of it, then the PLT results should be more appropriate for understanding

the interactions at molecular level [3]. The positron results for different blend compositions are summarized in Table 2.

From the positron data, it is clear that the average size of the free volume ( $V_{f3}$ ) and its number density ( $I_3$ ) increase with increase in concentration of EPDM. According to Eq. (1),  $\tau_3$  should increase with an increase in hole size and  $I_3$  should increase with an increasing number of free volume sites. Therefore, the changes in the amorphous structure, is actually probed by the *ortho*-positronium species [25].

The behavior of positron parameters as a function of the polymer blend composition has been explored as possible criterion in understanding the interaction between the blend components [18,20,26,27]. The literature reports that the study of polymer blends by positron method is very scanty and only few blends have been studied so far. Out of these, few blends have showed negative deviation from the log additivity rule and hence concluded as miscible [26]. Only one or two of them showed positive deviation and hence concluded as immiscible and only one agreed with the log

Table 2  
Free volume size evaluated from positron data

EPDM (wt%)	$\tau_3$ (ns)	$I_3$ (%)	$R$ (nm)	$V_f$ ( $\text{\AA}^3$ )	$F_v = V_f I_3$ (%)
0	$1.57 \pm 0.015$	$15.26 \pm 0.29$	0.242	59.58	9.1
10	1.82	13.89	0.268	81.07	11.3
20	2.04	14.97	0.290	101.94	15.3
40	2.07	18.37	0.292	104.71	19.2
50	2.16	19.54	0.304	113.65	22.3
60	2.21	23.56	0.310	117.79	27.8
80	2.28	25.76	0.317	124.77	32.1
100	2.37	28.57	0.317	133.90	38.3

additivity rule, which is also interpreted as immiscible [18, 27].

Fig. 4(a) and (b) shows the variation of  $V_f$  and  $I_3$  as a function of EPDM (wt%). The free volume hole sizes of EPDM and PTT are  $133.9$  and  $59.58$  ( $\text{\AA}^3$ ), respectively, while the free volume hole concentration of EPDM ( $I_3$ ) is  $28.57\%$  and that of PTT is  $15.26\%$ . The plot of  $I_3$ ,  $\tau_3$ ,  $V_f$

against EPDM wt% shows continuous increase of free volume parameters ( $V_f$  and  $I_3$ ) with the increase in EPDM concentration in the blend. Slight decrease of  $I_3$  is observed at 10 and 20-wt% of EPDM, and increase in glass transition temperature of about  $4$  °C is also observed from the DSC results. This might be due to the fact that the minor amorphous EPDM content is dispersed in the major

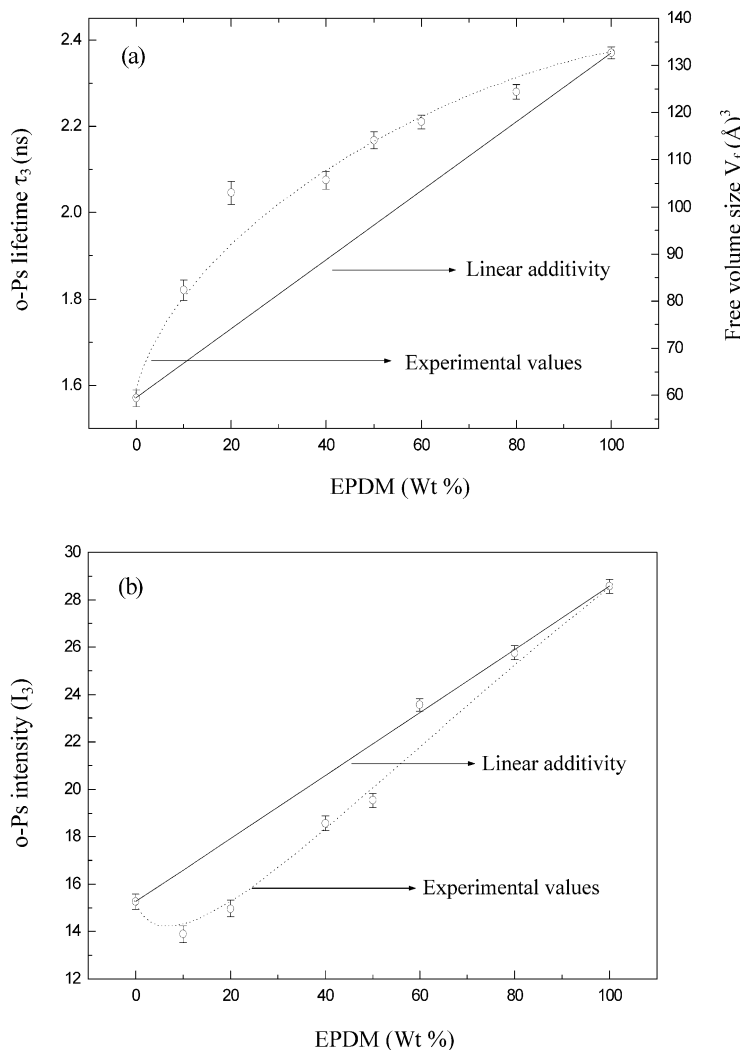


Fig. 4. (a) Variation of  $o$ -Ps lifetime as a function of EPDM weight percentage. The corresponding hole size ( $V_f$ ) is plotted on the right side y-axis. The solid line represents the linear additivity rule, symbols are the experimental points. A dotted line is drawn to show how the experimental data varies. (b) Variation of  $o$ -Ps intensity  $I_3$  as a function of EPDM weight percentage.

continuous PTT phase at lower weight fractions. Continuous increase in the free volume hole size of the blend above 30-wt% of EPDM is observed. This increase is possibly due to coalesce of the smaller free volumes of PTT with the large size free volumes of EPDM. Alternatively, it can be interpreted as follows: since EPDM and PTT are very dissimilar polymers the physical and chemical interactions across the phase boundaries are very poor. This leads to very weak interphase. As a result there is possibility of void formation. As such there is an increase in free volume with increase in EPDM content.

Of the few existing free volume theoretical models applied to polymer blends, we consider the one due to Kelley and Bueche [28], which is derived on the basis of additivity of free volumes of the blend constituents at all temperatures. According to this, the relative fractional free volume of a polymer blend will be the sum of the free volumes of the two components of the blend and is given by

$$h_b = w_1 h_{b1} + w_2 h_{b2} \quad (3)$$

where  $h_b$  is the relative fractional free volume of the blend,  $h_{b1}$ ,  $h_{b2}$  and  $w_1$  and  $w_2$  are the relative fractional free volumes and the weight fractions of the constituents 1 and 2, respectively.

Fig. 5(a) shows the variation of the relative fractional free volume as a function of EPDM wt%. For a blend to be miscible the relative fractional free volumes of the blend should show negative deviation from Eq. (3). The negative deviation of fractional free volume in miscible blends can be interpreted as the result of favorable interactions of segmental conformation and packing between dissimilar molecules [3]. In the present investigation, blends exhibit neither positive nor negative deviation from the linear additivity relation, instead there is linear increase of relative fractional free volume ( $F_v$ ) with increase in EPDM concentration and agree with Eq. (3). This is also an indication of the immiscible nature of the polymer blends [18]. It has been suggested that for the blends, the free volume can be correlated to interaction between dissimilar chains and the segmental conformation and packing of the component polymers. For a simple binary interchain interaction parameter, one can express the mean free volume hole fraction in the blend as [3]

$$F_v = F_{v1}w_1 + F_{v2}w_2 + \beta F_{v1}w_1 F_{v2}w_2 \quad (4)$$

Where  $F_{v1}$ ,  $F_{v2}$ ,  $w_1$  and  $w_2$  are the free volume hole fractions and the weight fraction of component 1 and 2, respectively.  $\beta$  is a parameter that could be related to the interaction between dissimilar chains. Liu et al. have reported negative values of  $\beta$  for PS/TMPC system [3]. It was reported that for a miscible blend the inter chain interaction parameter  $\beta$  should show negative deviation. In terms of interaction between dissimilar chains, a contraction of free volume may result in a decrease in Gibb's free energy. This can also be considered as a general criterion for miscible blend. In the

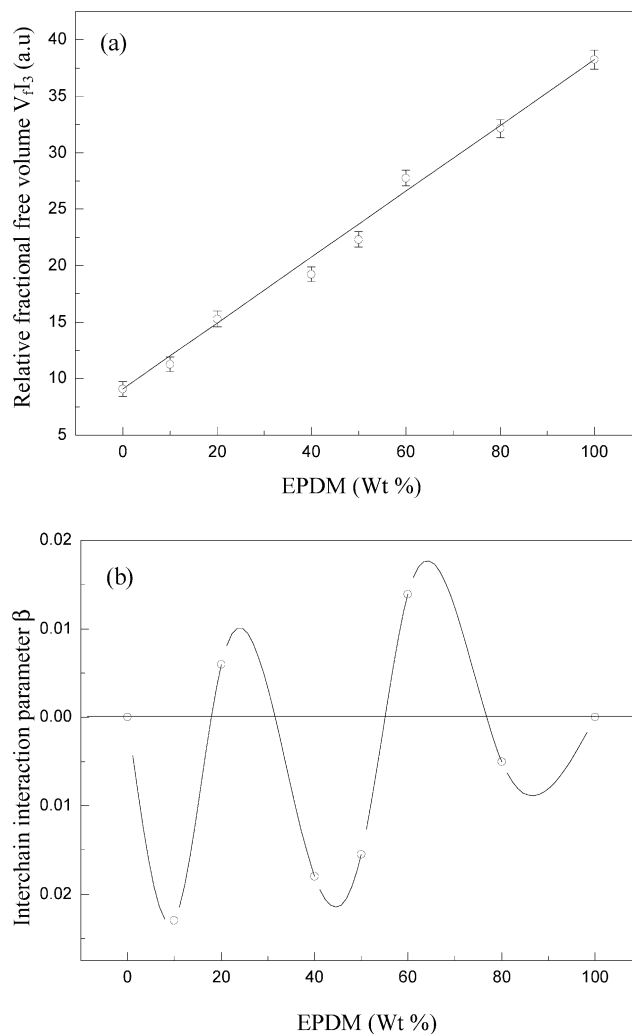


Fig. 5. (a) Variation of relative fractional free volume ( $V_f3$ ) as a function of EPDM weight percentage. Solid line represents the linear additivity rule. Symbols represent experimental points. (b) Variation of interchain interaction parameter  $\beta$  as a function of EPDM weight percentage. Solid curve is drawn to guide the eye.

present case,  $\beta$  calculated as per Eq. (4) shows fluctuating positive and negative values which is a complex behavior (Fig. 5(b)). Liu et al. obtained similar results for the PS/PC and for PS/PMMA immiscible blends [3]. They interpret this complex variation of free volume hole fractions due to immiscible nature of the blends [3]. In the light of these results, we conclude that the complex behavior of the  $\beta$  parameter is due to immiscible or incompatible nature of the blends, which agree with earlier conclusions.

### 3.3. X-ray diffraction results

Fig. 6(a)–(d) shows XRD scans of the PTT, 90/10, 80/20, 50/50 blends of PTT/EPDM, respectively. The ratio of the area under the crystalline peaks to that of the total area is taken as the measure of crystallinity [29–31], and the results so obtained are tabulated in the Table 3. From the table it is

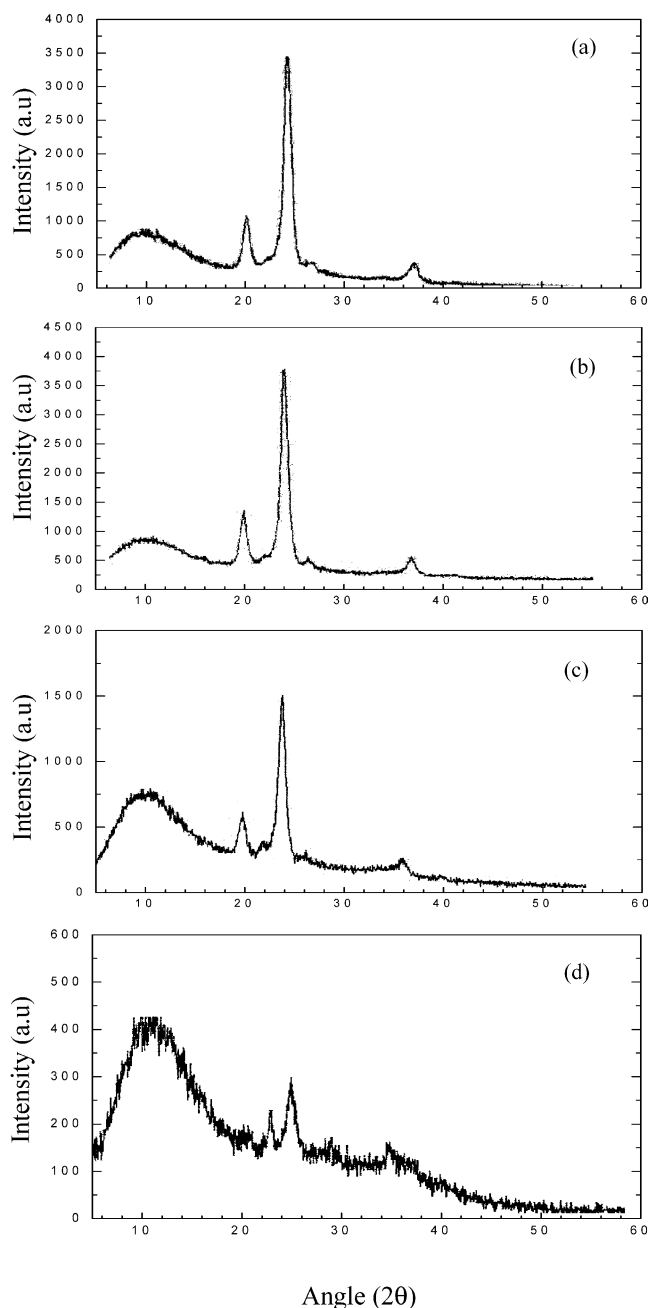


Fig. 6. (a) XRD scans of PTT. (b) XRD scans of 90/10 PTT/EPDM blend. (c) XRD scans of 80/20 PTT/EPDM blend. (d) XRD scans of 50/50 PTT/EPDM blend.

Table 3  
Crystallinity of the blend evaluated by XRD data

EPDM (wt%)	Crystallinity $\chi$ (%)
0	55.6 ± 1
10	54.5
20	44.0
50	23.0

clear that PTT is a crystalline polymer, which exhibits 55.6% of the crystallinity, and this value decreases as the EPDM wt% increases, which is in consonance with positron parameters.

Crystalline phases have been shown to directly influence the regions of semi crystalline homopolymers and the amorphous miscible regions in polymer blends. The mobility in the amorphous regions of a semi crystalline homopolymers will be reduced by the crystalline structures imposing restrictions on the segmental mobility. It has been shown that the  $I_3$  value is a measure of the crystallinity in the sample and that in a 100% crystalline polymer, the  $o$ -Ps formation probability approaches to zero [21,32]. In literature there are some interesting evidences regarding the decrease of free volume hole concentration ( $I_3\%$ ) in polymers and polymer blends with the increase in crystallinity. Recently, Dlubek et al. [29] has been done an interesting work on poly(ethylene) of different crystallinity, they found that the exponential decrease of  $o$ -Ps intensity with the crystallinity and approaching zero for 100% crystalline polyethylene. Ribeiro et al. [20] reported an increase in  $\tau_3$  and decrease in  $I_3$  with the wt% of PEG in PDMAM/PEG, PAM/PEG blends. According to them the change in the amorphous structure and crystalline phase content and their interphases, played a role in the overall material nanostructure.

In the present case, at the higher concentration of PTT in the blend, the free volume parameters ( $\tau_3$ ,  $I_3\%$ ) are minimum. As the PTT concentration decreases, the increase in the values of  $\tau_3$ ,  $I_3$  and  $F_v$  are observed. Correspondingly the reduction in the value of  $\Delta H_m$  in DSC measurements and the decrease in crystallinity by XRD are observed. This is on expected line that decrease of crystallinity results in an increase of free volume.

#### 4. Conclusions

- (1) From the DSC data, there is no observation of single  $T_g$  with the composition of EPDM and there is no shifting of  $T_g$ s towards each other. This indicates poor interaction between the two components, and high degree of incompatibility.
- (2) The melting point data shows that, there is no decrease in melting point of the blend with the EPDM concentration, and this further supports that the interaction between PTT and EPDM is very poor and blends obtained are immiscible.
- (3) The positron lifetime results show continuous increase in free volume size  $V_f$  and free volume concentration ( $I_3$ ) with increase in concentration of EPDM. The relative fractional free volume ( $h_b$ ) in the present case obeys Kelly and Bueche relation. Since no negative deviation from this relation is observed, this is a clear sign of incompatibility between the blend components.
- (4) The interchain interaction parameter  $\beta$  with the EPDM



concentration shows fluctuating positive and negative values further suggesting that the interaction between dissimilar chains is very poor.

- (5) XRD and DSC results shows the decrease in crystallinity of the blend with the increase in EPDM concentration and this is in agreement with positron results.

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