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Interfacial interaction in PP/EPDM polymer blend studied by positron annihilation

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ABSTRACT

Positron lifetimes in polypropylene/ethylene-propylene-diene monomer blend (PP/EPDM, EPDM volume content = 40%), pure PP and EPDM have been measured in the temperature range of 95-370K. We observed the γ - and glass transitions in all samples and the transition temperatures T_γ and T_g are determined. The blend has two lower T_gs compared with pure EPDM and PP. At room temperature, the measured free-volume concentration in blend versus EPDM volume fraction ϕ_V is smaller than that calculated by the simple mixing. The different free-volume properties and T_gs between blend, PP and EPDM indicate the existence of interfacial interaction between PP and EPDM phases in blend.

1. INTRODUCTION

Polymer blends are of great importance in application because they have some different mechanical properties compared with that of components [1] e.g., PP/EPDM blend has a higher impact strength than pure PP, which depends on the content and size of EPDM particles [2]. The reason for this improvement is being investigated. A percolation model has been proposed to explain this improvement, where the enough interfacial interaction between two phases is assumed in the stress volume [1]. In order to understand the interfacial interaction in the blends, we performed the measurements of positron lifetime vs T and ϕ_V.

2. EXPERIMENT

Three kinds of samples, i.e. PP (melt index= 3 g/10min, density =0.905 g/cm^3), EPDM and PP/EPDM blends were prepared, where EPDM is a kind of uncrosslinked rubber with the Mooney viscosity of 45 and density of 0.870g/cm^3. Blends were prepared by melt extrusion after mechanical mixing of PP and EPDM at 500K. Positron annihilation lifetime (PAL) was measured using a fast-fast coincident system with a time resolution of 270ps. The raising temperature is controlled better than 0.5K. Half a million counts were collected for each spectrum.

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3. RESULTS AND DISCUSSION

Each spectrum was resolved into three components using the program PATFIT [3]. The longest-lived component τ_3, has strong temperature dependence, is due to o-Ps annihilation in free-volume holes in amorphous regions, and its intensity I_3 is sometimes considered to be proportional to hole concentration [4]. In the simple mixing of the filler EPDM and the matrix PP, i.e. there is no any interfacial interaction between two phases, the hole concentration and the fractional free-volume are conservable. If the transition rate of o-Ps into the holes is the same for PP, EPDM and PP/EPDM [6], we have

$$I_{3B}=I_{3F}\cdot\Phi_V+I_{3M}\cdot(1-\Phi_V) \tag{1}$$

$$R_B^3\cdot I_{3B}=R_F^3\cdot I_{3F}\cdot\Phi_V+R_M^3\cdot I_{3M}\cdot(1-\Phi_V) \tag{2}$$

where B, F and M denote as the blend, filler EPDM and matrix PP, respectively, and R is the average hole radius.

The temperature dependence of o-Ps lifetime τ_3 shows the thermal expansion of free-volume holes. From the variations of τ_3 and I_3 shown in Fig.1 and 2 the γ - and glass transition temperatures can be determined [5]. The detailed performance is presented in [6]. In Tab.1 the T_γ and T_g measured by PAL and Dynamic Mechanical Analysis (DMA) are presented. The existence of two T_g in blend shows the two phases in blend are immiscible.

Tab.1 The results of T_γ and T_g measured by PAL and DMA.

	T_γ(K)		T_g(K)	
Samples	PAL	DMA	PAL	DMA
EPDM	160	-	220	239
PP	165	-	280	286
PP/EPDM (ϕ_V=40%)	160	-	205, 270	223, 280

The variations of I_3 in pure PP and EPDM vs T are explained in [5,6]. Generally the increase in I_3 above T_g suggests the creation of new free-volume holes , due to the onset of micro-Brownian motion of segments [5]. However, for PP/EPDM blend, it is evident that I_3 is nearly constant in the region of $T_{g1}\leq T\leq T_{g2}$, which shows no new holes are created. It arises from the following reasons: 1). no new holes are

created in the glassy PP; 2). although EPDM phase is rubbery when $T_{g1} \leq T \leq T_{g2}$, the creation of new holes in EPDM is strongly restricted by the interaction between PP and EPDM phases.

There are other evidences for existence of interfacial interaction in PP/EPDM. First, The T_gs of the blend are shifted to a lower values from those of PP and EPDM, probably due to the formation of the emulsion interfaces with the excess free-volume [7]. In literature [8] the effects of several possible interfacial morphologies on the glass transitions are discussed. It is proposed that in an emulsified interface, there is small interfacial domains but no interfacial mixing, where the excess free-volume is developed. As result the mobility of macromolecules of both phases in the interface is increased, which may reduce the glass transitions temperature of the interface. A detailed calculation shows that the shift of T_g is dependent on the concentration and the T_g of interfacial layers [7].

Second, between 280K-340K, PP/EPDM has a smaller thermal expansion coefficient of fractional free-volume than PP and EPDM, which may result from the residual thermal stresses in the matrix and the filler due to the differences in coefficient of thermal expansion, Young's modules and Poisson's ratio between EPDM and PP [9]. From the temperature dependence of relative fractional free-volume $F_r=I_3 \cdot V_f$ (V_f is the measured free-volume by PAL), the thermal expansion coefficient of relative fractional free-volume, i.e., $\alpha_r=(\partial F_r/\partial T)_p$ can be evaluated [6]. For PP, EPDM and PP/EPDM, the values of α_r in the range of 280-340K obtained by the linear fitting are 0.299, 0.349 and 0.290, respectively. It is evident that PP/EPDM has the smallest value of α_r in the three samples.

Third, at room temperature, the measured I_3 and R is smaller than the expected values obtained by eq.(1-2), as shown in Fig.3-4. This observation proves that the average packing density of segments in the bulk of PP/EPDM increases and the interaction between two phases is enhanced with the increase in the EPDM content.

We have measured the curve of impact strength with the rubber EPDM volume fraction ϕ_r for PP/EPDM [2] and observed an onset increase of the impact strength at ϕ_r=25%, corresponding to a brittle-ductile transition [1]. Experimental studies show that the impact strength improvement of PP/EPDM blends is directly connected with the shear yield of PP matrix around EPDM particles [1,2]. The interfacial interaction is beneficial for the stress transfer between two phases and can restrict the glide of polymer chains at the interface, so the shear yield of matrix molecules will become more difficult under the impact force and the blends will have a higher impact strength. In this work, the measured free-volume properties suggest the existence of the interfacial interaction and the thermal tensile stress at the interface, which should be one of essential conditions for the improvement of mechanical properties of PP/EPDM [2]. The quantitative relation of free-volume and the mechanical properties in polymer blend is necessary for future study.

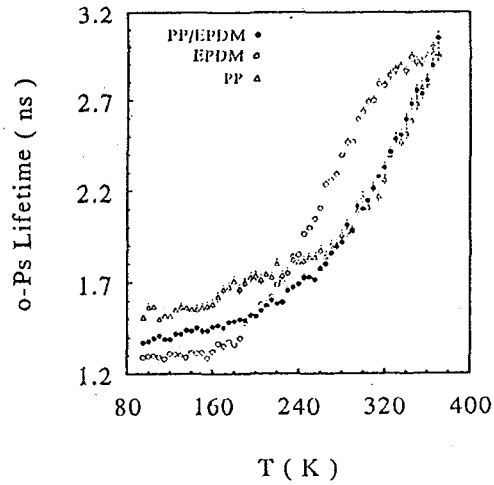

Fig.1 The variation of o-Ps lifetime τ_3 with T

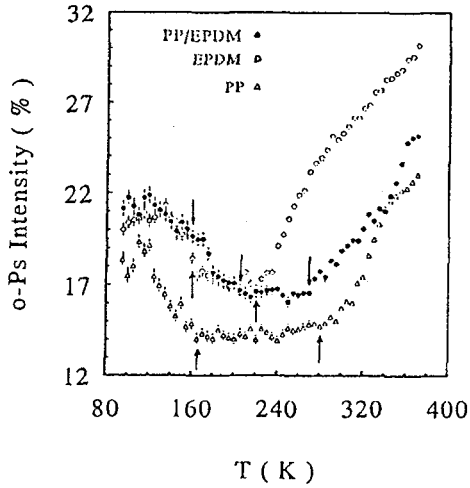

Fig.2 The variation of o-Ps intensity I_3 with T

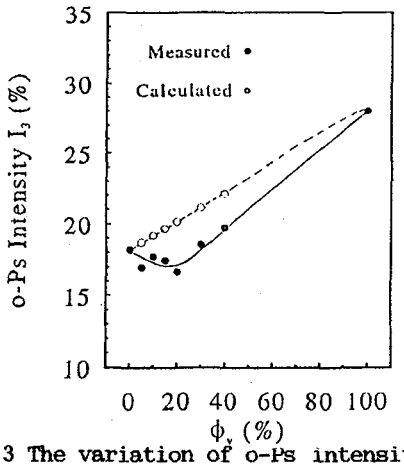

Fig.3 The variation of o-Ps intensity I_3 with EPDM content ϕ_V

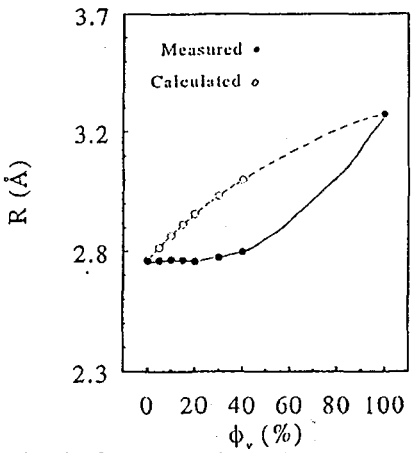

Fig.4 The variation of average radius of free-volume hole with ϕ_V

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