Synthesis and characterization of new poly(ethylene terephthalate)/poly(phenylene oxide) blends

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Abstract: Several compositions of poly(ethylene terephthalate) (PET) and poly(phenylene oxide) (PPO) were prepared by two different methods: *in situ* polymerization (IS-P) and direct mixing in the melt (DM). The existence of phase separation in the obtained PET/PPO systems has been confirmed by calculating the solubility parameter of compounds using Hoy's method. The combination of results obtained with DSC and DMTA points toward a complex morphology of PET/PPO systems.

Keywords: poly(ethylene terephthalate), poly(phenylene oxide) (PPO), *in situ* polymerization, melt blending, phase structure, water absorption.

Otrzymywanie i charakterystyka nowych mieszanin poli(tereftalan etylenu)/poli(tlenek fenylenu)

Streszczenie: Z zastosowaniem polimeryzacji *in situ* (IS-P) i mieszania w stanie stopionym (DM) wytworzono dwie serie mieszanin na bazie poli(tereftalanu etylenu) (PET) oraz poli(tlenku fenylenu) (PPO). Na podstawie obliczonych metodą Hoya współczynników rozpuszczalności poszczególnych składników potwierdzono występowanie separacji fazowej w otrzymanych układach PET/PPO. Technikami różnicowej kalorymetrii skaningowej (DSC) i dynamicznej analizy mechaniczno-termicznej (DMTA) potwierdzono złożoność struktury fazowej uzyskanych układów PET/PPO.

Słowa kluczowe: poli(tereftalan etylenu), poli(tlenek fenylenu), polimeryzacja *in situ*, bezpośrednie mieszanie w stanie stopionym, struktura fazowa, chłonność wody.

Poly(ethylene terephthalate) (PET), a plastic resin which is the most common type of polyester used commercially, is one of the most recycled plastics in the market today. It was first synthesized in 1946 [1] and shortly after, in 1953, when DuPont commenced its commercial production, PET became the most widely produced synthetic fibre in the world. Thereafter, in the 1970s, the development of stretch molding procedures enabled the production durable crystal-clear beverage bottles from bulk PET – an application that soon became second only to fibre production [2]. PET technology has been developed ever since and has advanced to such a level that its presence can be observed in almost every industrial application. Thanks to a unique chemical structure and composition the material provides many advantages. It is, among others, resistant to both heat and cold, transparent, resistant to chemicals, and abrasion proof. The presence of a large aromatic ring in the PET's repeating units gives the polymer notable stiffness and strength, especially when the polymer chains are aligned with one another to create an orderly arrangement by drawing or stretching. PET is often used as a blend with other fibres such as rayon, wool, and cotton, as fibre filling for insulated clothing, for furniture and pillows, as artificial silk when made in very fine filaments, and as carpets when in large-diameter filaments. Other industrial applications include automobile tire yarns, conveyor and drive belts, reinforcements for fire and garden hoses, seat belts, nonwoven fabrics for stabilizing drainage ditches, culverts, railroad beds, diaper top sheets, and disposable medical garments [2]. At a slightly higher molecular weight, PET can be combined with other thermoplastics and shaped, using all commonly known methods, into a high-strength plastic. Magnetic recording tapes and photographic films are produced by extrusion of PET film. However, the most commonly known PET application are

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the PET bottles which are one of the most commercially preferred food containers due to their nontoxicity, durability, transparency, light weight, resealability, shatter resistance, and recyclability.

When regarding the application of thermoplastics such as PET, the most important criteria which need to be taken into account are their thermal stability, stiffness, and strength. At a commercial level the aforesaid are achieved through an addition of suitable reinforcing agents such as fibres or fillers. Utilization of these fillers at the nanoscale in order to obtain optimal properties even at very low loading is the recent trend is this regard. There is a wide array of organic and inorganic nanofillers some of which have been studied more thoroughly than others. The most widely examined fillers include nanotubes [3, 4], graphene derivatives (*e.g.*, graphene [5], exfoliated graphite [6], expanded graphite [7], graphene oxide [8, 9], *etc.*), layered silicates [*e.g.*, montmorillonite (MMT), saponite], metal nanoparticles, metal oxides, semiconductors [10]. Additionally, the properties of poly(ethylene terephthalate) can be significantly improved by blending it with other polymers, both natural or synthetic. Over the course of the last several decades such blends have become an important line of research leading to new, high performance polymeric materials. However, in order to manufacture the same, some obstacles need to be overcome as most polymer blends are immiscible because of their high molecular weight and unfavorable interactions. They thus have a tendency to form multiphase structures which's properties are not only determined by the properties of the component polymers, but also by the formed morphology [11, 12]. In the case of melt-blended immiscible polymers two main kinds of morphologies can be distinguished: the matrix-dispersed structure and the co-continuous structure. Increasing attention is being paid to co-continuous blends as this morphology offers a far better combination of the component properties than the dispersed type structure [13–15].

PET blends can be prepared using five main techniques: graft copolymerization, melt solution, latex blending, partial block, and synthesis of interpenetrating networks. Melt blending is a simple mechanical process of creating a homogeneous mixture of two or more polymers. The major advantage of melt blending is the absence of any solvents which might be required in other methods of polymer blending [16]. PET contains hydroxyl and carboxyl functional groups both located at chain endings; *in situ* or reactive compatibilization is an effective approach for developing engineering thermoplastic blends from these materials. The combination of PET with polyolefins represents an important group of PET blends [17–19]. Murff *et al.* [20] prepared a melt blend of polycarbonate and PET by continuous extrusion and injection molded it into bars to study their thermal and mechanical behavior. Huang [21] studied and discussed the mechanism, morphology, and mechanical properties of PET/PA6 (polyamide 6) blends with low molecular weight bisphenol-A epoxy resin (E-44) as a reactive compatibilizer. Imashiro *et al.* [22] has described adding a carbodiimide compound to recycled PET such that the intrinsic viscosity and strength of the polyester resin during processing are retained. However, only a limited number of published papers on polymer blends of poly(2,6-dimethyl-1,4-phenylene ether) (PPO) and the polyesters like poly(ethylene terephthalate) [23] or poly(butylene terephthalate) (PBT) have been reported [24–27]. This is probably due to the incompatibility between these two classes of polymers. PPO demonstrated excellent mechanical properties at increased temperature, a good resistance to moisture and to wide range of chemicals [28, 29]. Due to its high glass transition temperature (T_g ~ 210 °C) [30], it is often blended with other thermoplastics, such as polystyrene [28, 31] or polyamide [32, 33], which reduces the T_g value and makes processing easier. PPO blends are widely used in many industrial applications, for instance in the automotive industry [34], computer and television casings and keyboard frames [35]. PPO was synthesized for the first time in 1959 by Hay *et al.* [36] by the oxidative polymerization of 2,6-dimethylphenol (2,6-DMP). Nowadays there are several methods of PPO preparation, such as microwave assisted polymerization [37], suspension polymerization [38], or reaction with immobilized catalysts [39]. Unfortunately those methods have very little technological importance. Therefore, the group from the Industrial Chemistry Research Institute investigated the effect of 2,4,6-TMP on the yield of PPO synthesis and polymer properties [40].

In this work, it was found that it is possible to obtain PET/PPO blends *via in situ* polymerization. Hence, the aim of this paper is to study the selected properties of the *in situ* prepared PET/PPO blends and compare their properties with those obtained using the classical method that is the direct mixing in the melt. Calculation of the theoretical Hildebrand solubility parameters for PET/PPO polymer blends confirm that PET and PPO are immiscible and exhibit phase separation. Moreover, the selected physicochemical properties (intrinsic viscosity, density, melt flow index) and water absorption in cold and boiling water of both systems were investigated. Additionally, the complex morphology of PET/PPO systems has been studied with differential scanning calorimetry (DSC) and dynamic mechanical thermal analysis (DMTA).

EXPERIMENTAL PART

Materials

For the poly(ethylene terephthalate) (PET) and PET blends synthesis the following chemicals were used: dimethyl terephthalate (DMT, Sigma Aldrich), 1,2-ethanediol (ED, Sigma Aldrich).

Zinc acetate $Zn(CH_3COO)_2$ (Sigma Aldrich) and antimony trioxide Sb_2O_3 (Sigma Aldrich) were used as catalysts in transesterification and polycondensation, respectively.

Irganox 1010 (Ciba-Geigy, Switzerland) was used as antioxidant.

For the PET based blends, prepared *via* direct mixing, PET (ELPET, Bottle Grade) from Torlen Sp. z o.o. (Poland) with an intrinsic viscosity of 73–76 cm^3/g has been applied.

Poly(phenylene oxide) (PPO) has been provided by Industrial Chemistry Research Institute and prepared accordingly to the method published in [40]. The properties of PPO were as follows: $GLL = 49\ cm^3/g$, $MFR = 0.15$ g/10 min, $M_n = 18\,714$, $M_w = 70\,810$, $M_w/M_n = 3.784$.

Preparation of PET/PPO samples

The details of making PET/PPO blends *via in situ* polymerization (IS-P) can be found elsewhere [7–9, 41–45] and will not be repeated here. Samples with an addition of 2.5, 5.0, 7.5 and 10.0 wt % of PPO were prepared. Briefly, before polymerization, PPO (in the form of powder) was dispersed with 1,2-ethanediol using high-speed stirrer (Ultra-Turax T25) and sonicator (Homogenizer HD 2200, Sonoplus, with frequency of 20 kHz and 75 % of power 200 W) in both cases for 30 min. The polymerization process was conducted in two stages: transesterification in a temperature range of 160–180 °C, where PPO/ED mixture was charged along with DMT and TBT (tetrabutyl orthotitanate, Fluka) into 1 dm^3 steel reactor (Autoclave Engineers Inc., USA); the second stage – polycondensation, under vacuum conditions (~ 0.1 hPa) at the temp. of 275 °C. The obtained polymer/blend was extruded from the reactor under nitrogen flow in the form of polymer wire and subsequently granulated and injection molded.

In turn, the second series of PET/PPO blends, labeled in the text as DM, was prepared by melt mixing the PET (ELPET) and PPO in a laboratory scale twin screw extruder Leistritz Laborextruder LSM30 $L/D = 22.9$. The extruder temperature was 275 °C. This procedure allows to prepare polymer blends with higher concentration of PPO, *i.e.*, 5, 10, 15 and 20 wt %.

The dumbbell shape samples (type A3) for water absorption and DMTA measurements were obtained by injection molding using Boy 15 (Dr Boy GmbH & Co., Germany) injection molding machine with the following parameters: injection pressure 55 MPa, melt temperature 275 °C, mould temperature 30 °C, holding down pressure of 20 MPa for 15 s, and cool time 10 s. Additionally, the samples for DMTA measurements were annealed for 2 h at 100 °C, *i.e.*, the temperature above the T_g.

Methods of testing

– The intrinsic viscosity [η] of the samples was determined at 30 °C in the mixture of phenol/1,1,2,2-tetrachloroethane (60/40 w/w). The polymer solution had a concentration of 5 g/dm^3. The measurement was carried on a capillary Ubbelohde viscometer (type Ic, K = 0.03294).

– The melt flow index (*MFI*) was measured using a melt indexer (CEAST, Italy) as the weight of the melt flow in grams per 10 min, at temp. of 260 °C, and with orifice diameter of 2.095 mm and under 21.18 N load, accordingly to ISO 1133 specification.

– The density was measured at 23 °C on hydrostatic balance (Radwag WPE 600C, Poland), calibrated for standards with known density.

– Water absorption tests were conducted in cold and boiling water in accordance with the test procedures recommended in ASTM D570. The dumbbell shape samples were dried to constant mass at 50 °C within 24 h, cooled to the room temperature and weighed. To measure cold water absorption specimens were immersed in distilled water at 23 °C for 24 h. Boiling water immersion was conducted for 30 min, then specimens were allowed to cool down to room temperature in distilled water. All surface water was removed with filter paper and samples were weighed. Each reported value is an average of 5 test specimens.

– The Hoy method has been applied in order to estimate the solubility parameter (δ). This method is based on three different contributions: a solubility parameter due to dispersion forces (δ_d) – all non-polar contributions, a polar contribution (δ_p) due to dipole forces, and hydrogen bond contribution (δ_h) – only present when the molecule can form hydrogen bonds or due to donor--acceptor interactions [46]. Hoy's system contains four additive molar functions, a number of auxiliary equations, and the final expressions for δ_{tot} and its components of δ.

Each of this can be regarded as a vector in three-dimensional space, so the total solubility parameter (δ_{tot}) is defined by [1]:

$$\delta_{tot}^2 = \delta_d^2 + \delta_p^2 + \delta_h^2 \tag{1}$$

The full equation that determines the solubility of a polymer (P) in an organic liquid (L) [2]:

$$\Delta\delta = [(\delta_{d,\mathrm{P}} - \delta_{d,\mathrm{L}})^2 + (\delta_{p,\mathrm{P}} - \delta_{p,\mathrm{L}})^2 + (\delta_{h,\mathrm{P}} - \delta_{h,\mathrm{L}})^2]^{1/2} \tag{2}$$

The smaller difference in solubility of block pairs $\Delta\delta \leq 5\ \mathrm{MPa}^{1/2}$ the more soluble they are [2]. It is possible to calculate the theoretical solubility parameters for polymers compositions.

– The melting and crystallization behavior of both series of PET/PPO blends was studied using a differential scanning calorimeter Q-100 (TA Instruments, USA, 2004) in the cycle: heating/cooling/heating in the temperature range of -50 – 300 °C. The heating and cooling rate of the samples was 10 °C/min. Moreover, the glass transition T_g temperature was determined as a midpoint of the heat capacity's change ($\Delta C_p/2$). The degree of crystallinity X_c was calculated using the following equation: $X_c = \frac{\Delta H_m}{\Delta H_m^0 \cdot x_{\mathrm{PET}}} \cdot 100\ \%$, where: ΔH_m – enthalpy of melting of material sample (J/g), ΔH_m^0 = 140.01 J/g – the theoretical value of enthalpy for 100 % crystalline PET [47], x_{PET} – mass fraction of PET content.

– The dynamic mechanical thermal analysis (DMTA) has been performed using a Polymer Laboratories MK II apparatus working in a bending mode in a temperature range from –100 °C to the polymer melt temperature, at a frequency of 1 Hz and the heating rate of 3 °C/min. The properties were determined on the basis of modulus changes and the ability of attenuation as a function of temperature and frequency of load changes.

RESULTS AND DISCUSSION

Physicochemical properties of PET/PPO blends

It is generally known that the morphologies of polymer blends depend on the shear history in the mixer (reactor, extruder, *etc*.), the viscosity ratio, and the interfacial tension between the matrix and components. Particularly, the interfacial tension between two polymers is very important for phase morphology and compatibilization plays a significant role in lowering interfacial tension or enlargement of interface adhesion, thereby forming a finer morphology [48]. The obtained PET/PPO blends were characterized by the intrinsic viscosity, density, melt flow index measurements. The addition of PPO affected the length of the PET sequences and thus the values of intrinsic viscosity. Moreover, with an increase of PPO content the values of melt flow index increased (Table 1). The highest values of melt flow index have been observed in the case of two IS-P blends with the highest concentration of PPO (increase of about 57 %). One can therefore conclude that some of the macromolecules of PPO reacted with the PET chains. In the case of DM blends almost no effect of the intrinsic viscosity and melt viscosity has been observed, even when the PPO content was 20 wt % in the PET/PPO system. This unambiguously confirms that in the case of PET/PPO blends prepared *via in situ* polymerization one can observed partially reactive blending. The transesterification reaction will probably occur between the hydroxyl groups of PPO and methylene groups of terephthalic ester. Thus, this method allows lowering the interfacial tension between PET and PPO and thus enlarging the interfacial adhesion between both phases.

Moreover, for PET/PPO blends, the experimental density is lower than the density of neat poly(ethylene terephthalate). This means that the addition of PPO doesn't occupy the free volume of the PET network. The variation in the densities can be associated with the differences in the values of degrees of crystallinity. It can be assumed the PPO doesn't affect the nucleation and thus disturbs the crystallization of PET matrix.

The water absorption test showed that PET/PPO blends are almost impermeable. PET/PPO blends can be immersed in cold and hot water, with water absorption of 0.25 % and 0.40 %, respectively. Small addition, up to 10 wt % of PPO, in melt blending process results in a slight decrease of cold and boiling water absorption. Hindered absorption of water indicates the hydrophobic nature of poly(phenylene oxide). In melt blended samples (DM) containing 15 wt % and more of PPO one can deal with non-homogeneous distribution of PPO in the whole volume of the PET matrix, resulting from phase separation (Fig. 1, see below the solubility parameters). PPO agglomerates seem to be wrapped within PET matrix, thus, the water absorption values for PET/PPO blends (DM) are comparable to neat PET. Similar phenomena occur in PET/PPO samples obtained by *in situ* synthesis. Especially, since Bae *et al*. [49] observed in poly(ethylene terephthalate)/polypropylene (PET/PP) blends that the water absorption of pure PET was reduced by the addition of PP. It was explained, that the reduction in water absorption was proportional to the PP volume content in the blends. Moreover, the compatibilized blends of PET and PP grafted with 2-hydroxyethyl methacrylate isophorone diisocyanate (PP-*g*-HI) was proved to exhibit significantly improved water absorption resistance. This was due to the finer dispersed phase size, which created the increase of the total surface area for the PP phase to the PET phase.

Table 1. Physicochemical properties and water absorption of PET/PPO blends

Sample	Content of PPO, wt %	[η], cm^3/g	*MFI*, g/10 min	*d*, g/cm^3	Cold water absorption, wt %	Boiling water absorption, wt %
PET IS-P	0	55	45.67 ± 0.76	1.3358 ± 0.0011	0.23 ± 0.01	0.39 ± 0.03
PET/2.5 PPO IS-P	2.5	54	47.93 ± 0.79	1.3293 ± 0.0024	0.21 ± 0.01	0.39 ± 0.01
PET/5.0 PPO IS-P	5.0	52	53.75 ± 0.90	1.3236 ± 0.0031	0.22 ± 0.01	0.38 ± 0.02
PET/7.5 PPO IS-P	7.5	49	72.59 ± 1.21	1.3230 ± 0.0024	0.23 ± 0.01	0.44 ± 0.03
PET/10.0 PPO IS-P	10	46	71.81 ± 1.20	1.3231 ± 0.0016	0.23 ± 0.01	0.39 ± 0.02
PET DM	0	59	45.19 ± 0.75	1.3355 ± 0.0014	0.24 ± 0.01	0.40 ± 0.02
PET/5.0 PPO DM	5.0	53	43.67 ± 0.73	1.3183 ± 0.0016	0.23 ± 0.01	0.37 ± 0.00
PET/10.0 PPO DM	10	65	47.51 ± 0.79	1.3073 ± 0.0032	0.22 ± 0.01	0.36 ± 0.01
PET/15.0 PPO DM	15	66	49.67 ± 0.83	1.2916 ± 0.0024	0.25 ± 0.01	0.41 ± 0.01
PET/20.0 PPO DM	20	65	45.80 ± 0.76	1.2928 ± 0.0030	0.24 ± 0.01	0.42 ± 0.01

[η] – intrinsic viscosity; *MFI* – melt flow index at 260 °C; *d* – density measured at 23 °C.

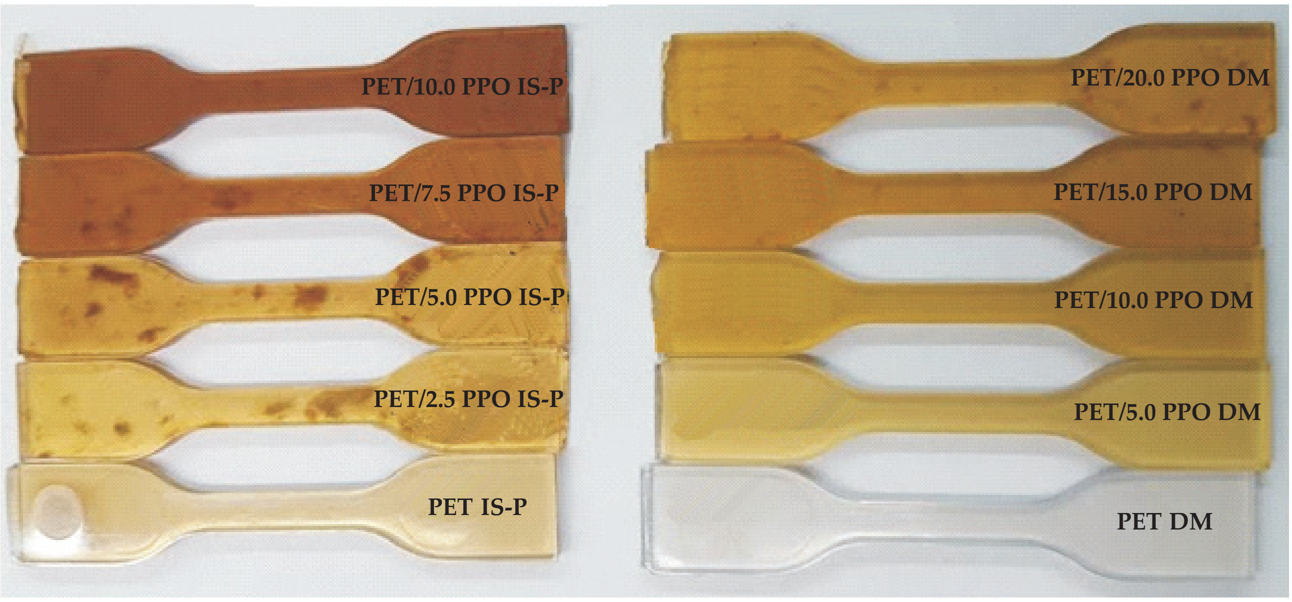

Fig. 1. Dumbbell shape samples for physicochemical properties' and water absorption measurements

Multiphase structure of PET/PPO blends

It can be assumed that the multiphase polymer blends have good functional properties, only when they are compatible with one another. These materials, depending on the temperature, may exhibit the phenomenon of phase solubility and the phase separation. This is due to the large differences between the phase transitions temperatures of each phase. The characteristic feature of copolymers is that in the cooled state they form a multiphase structure resulting from micro- and nanophase separation, characterized by thermodynamic immiscibility [50, 51]. The Hoy solubility parameters' method is the one of the simplest way to evaluate whether one material is miscible or soluble with/in another organic material (solvent, polymer, *etc*.). The contributions are added to the total for each chemical group in the compound. This method is mostly used for structural features like: cis, trans (around double bonds), ortho-, meta-, para- substitution (aromatics), branching (conjugation of double bonds, and rings) [52]. The total solubility parameters and its components were used to select miscible blend system for the model PET/PPO and were calculated by using Hoy's method (Table 2).

Table 2. Solubility parameters of the PET/PPO polymer blend calculated using Hoy's method

Solubility parameters	PET δ MPa$^{1/2}$	PPO δ MPa$^{1/2}$	PET/PPO $\Delta\delta$ MPa$^{1/2}$
δ_{tot}	23.75	15.43	14.04
δ_p	13.39	2.36	
δ_h	15.21	6.64	
δ_d	12.36	13.78	

$\Delta\delta$ – difference in the solubility parameters of polymer pairs δ_d, δ_p, δ_h – resulting from dispersion forces, polar interaction, and hydrogen bonding.

Accordingly to the Hoy's calculations method, the obtained blends exhibit significantly higher values of $\Delta\delta$ parameter (14.04 MPa$^{1/2}$) than parameter appointed for completely miscible materials. This proves that PET and PPO polymers are immiscible and exhibit phase separation. Based on the calculations one can deduce that in the cooled state the blends form a heterogeneous structure with two separate phases.

The results of dynamic mechanical thermal analysis of PET/PPO blends (Fig. 2) confirmed the observations described above. For neat PET (IS-P) one relaxation peak is observed in the temperature range of 79–113 °C which corresponds to both, glass transition (beta relaxation) and cold crystallization temperatures. The same observations were made for the whole series of *in situ* prepared blends (Fig. 2a). Moreover, the formation of phases with a reduced content of PET (beta temperature of about 125 °C and ~ 170 °C) were demonstrated (Fig. 2a). In the case of samples prepared *via* direct mixing no such effect was visible (Fig. 2b). It is therefore possible and very interesting to obtain a reactively modified PET using PPO with enhanced thermal parameters such as T_g above 100 °C. From the DSC thermograms one can observe only one glass transition temperature at about 80 °C and the melting endotherm in the temperature range of 205–275 °C (Fig. 3). The PPO exhibits the T_g at about 210 °C, which herein is covered by the melting effect of PET crystals. Therefore on the DSC thermograms no T_g of PPO can be observed. The presence of PPO in the PET/PPO systems caused the decrease of the values of PET crystallization temperature (Table 3). Probably, firstly the transition of PPO occurs and subsequently the crystallization of PET. One can say that PPO disturbs the crystallization of PET. The PPO domains, which were set up following the glass

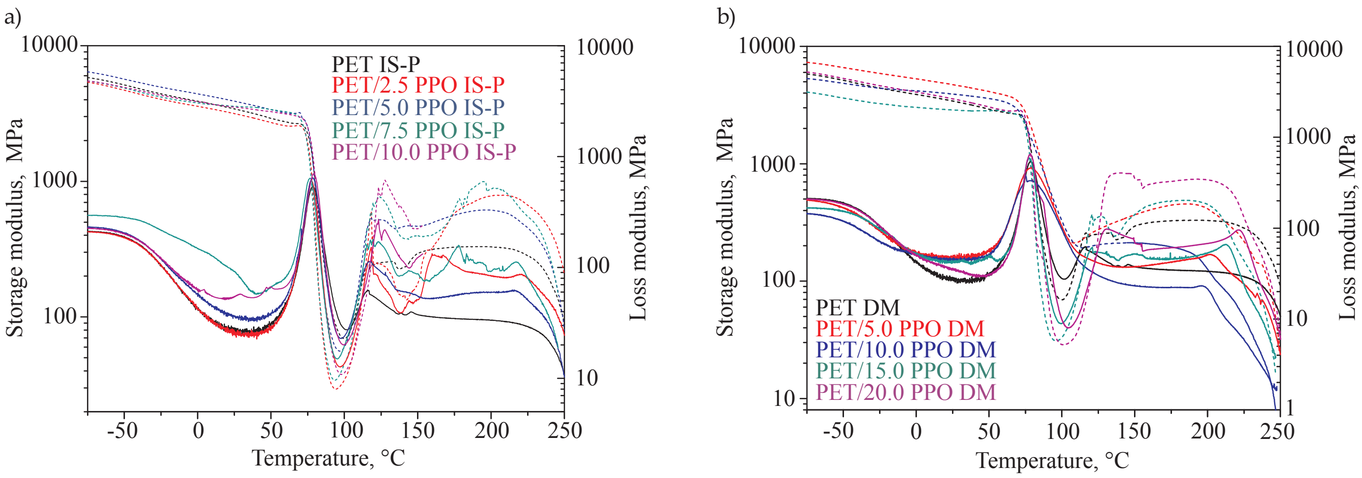

Fig. 2. The dependence of storage and loss moduli: a) PET/PPO IS-P blends, b) PET/PPO DM blends

Table 3. Thermal properties of PET/PPO blends

Sample	T_g °C	T_m °C	ΔH_m J/g	T_c °C	ΔH_c J/g	X_c %
PET IS-P	78	258	51.5	207	51.7	36.7
PET/2.5 PPO IS-P	82	256	41.9	198	42.4	30.6
PET/5.0 PPO IS-P	82	255	42.5	199	47.6	31.5
PET/7.5 PPO IS-P	82	252	36.6	188	36.5	28.2
PET/10.0 PPO IS-P	79	249	38.7	191	37.1	30.4
PET DM	76	251	39.2	194	38.8	28.0
PET/5.0 PPO DM	77	248	32.1	185	33.0	23.8
PET/10.0 PPO DM	79	247	32.2	188	32.6	25.5
PET/15.0 PPO DM	79	249	29.5	181	31.7	25.2
PET/20.0 PPO DM	80	249	31.4	183	35.8	28.0

T_g – glass transition temperature; T_m – melting temperature; T_c – crystallization temperature; ΔH_m, ΔH_c – enthalpy of melting and crystallization; ΔH_m^0 = 140 J/g; X_c – mass fraction of crystallinity determined from DSC.

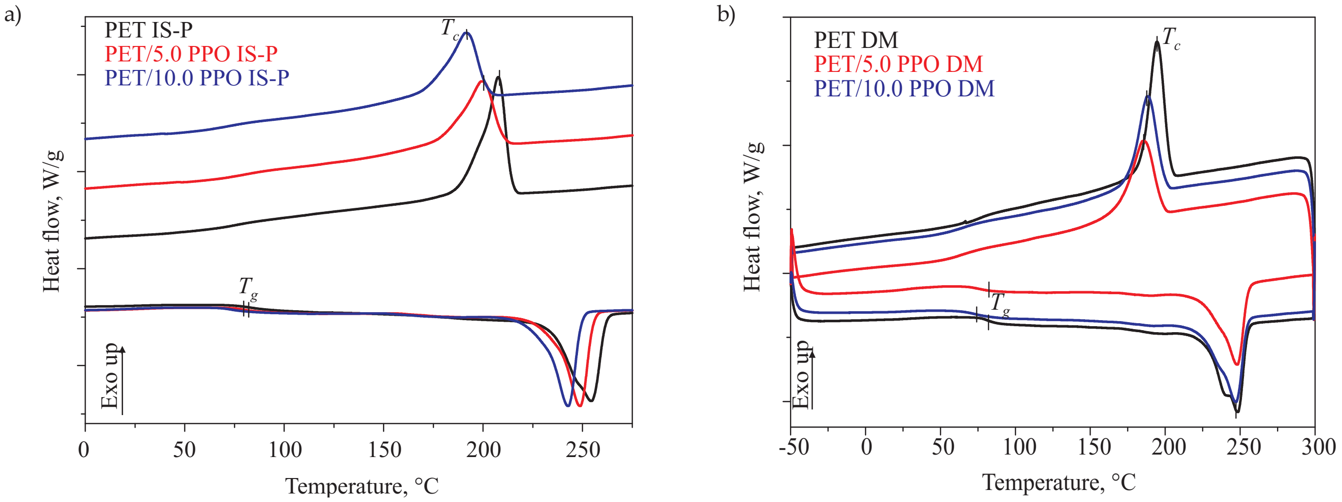

Fig. 3. DSC thermograms during second heating and cooling for: a) PET/PPO IS-P blends with 5 and 10 wt % of PPO, b) PET/PPO DM blends with 5 and 10 wt % of PPO

transition, might restrict the mobility of PET chains. As a result, the crystallization temperature is lowered.

CONCLUSIONS

It has been presented that the addition of PPO into the PET matrix during both *in situ* polymerization (IS-P) and direct mixing (DM) only marginally affected the properties of the investigated PET/PPO systems. Basically, this is due to the lack of miscibility of the two polymers, which was confirmed by determining the solubility parameters using Hoy method. However *in situ* polymerization method allows obtaining and studying new polymer materials from PET/PPO blends. This method is also more effective than the typical mixing in the molten state during extrusion. The possibility of modifying PET with small amount of PPO gives great prospect for new application of these materials in packaging, where the antinucleating effect of PPO on the PET is more than desirable. This study will be continued in order to improve the compatibility, and thus to improve the properties of PET/PPO systems using appropriate compatibilizers.

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