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DYNAMIC MECHANICAL BEHAVIOUR OF VIRGIN PP/PET BLENDS

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ABSTRACT

In this paper, virgin Polyethylene tere phthalate has been blended in an elastic extruder with three different types of polypropylenes such as low viscosity Polypropylene (A 5012), high viscosity PP (K 7050) and modified PP (XKP 707W). Dynamic mechanical behaviour such as storage modulus (Er) and dissipation factor (tan δ) of these blends has been determined at different temperatures. Effects of blending ratio on storage modulus and dissipation factor have been determined, compared and analyzed. It has been found that addition of PET increased the storage modulus of PP due to the higher modulus of PET. An additional peak has been observed In tan δ plots obtained for PP/PET blends. This additional peak corresponds to a-relaxation of PET. Type of PP addition has also modified the dissipation factor (tan δ) and storage modulus Er behaviour of blends.

Key words : Polyethylene tere phthalate (PET), Polypropylene (PP), blends.

INTRODUCTION

Polypropylene (PP) has been modified by blending or addition of different types of fillers[1] to achieve the desired type of mechanical properties.Toughness of PP has been improved by incorporating different elastomers [2,3]. Development of PP/ elastomer/PE has provided high impact-properties of PP[4], Ternary blend of PP/ elastomer/mineral filler has good mechanical properties[5]. Rheological and morphological properties of short glass fibre reinforced PP / EPDM blends have been determined and reported by Gupta *et.al.*[6]. They found that a rubbery phase was indistinguishable in the fractured surfaces obtained in SEM.

Polypropylene has high crystallinity[7]. Crystallinity of the film depends on the modification of the structure of polypropylene. It has been reported in the literature that melting point of PP has been assigned to the monoclinic α form of the crystallites. Higher melting point of PP film has been attributed in the literature to an increased order and degree of molecular packing in the c/ystalline phase of polymer[8]. These changes occur during biaxial orientation of PP. Tenter type of orientation gives highest melting point than the tubular type orientation of PP. Tenter type, tubular type and unoriented PP had melting points 171,166 and 159°C respectively[8]. Crystallinity of the film depends on the crystallization temperature.

Recently, Ellis and Picot[9] have studied the effect of draw ratio on melting endotherm of PET. Different types of polypropylene are commercially available. Similarly, change of orientation in polyethylene terephthalate (PET) changes the structure of PET, which has been observed by dielectric relaxation

measurements and reported by Gupta er. a/.[10]. In order to improve the toughness of PP, PET has been added in the past[11]. However, no report is available in the literature on the effect of addition'of PET to different type of PPs on their storage modulus and on dissipation factor, which directly gives the idea about strength and structure of Polypropylene.

In this paper virgin PET has been melt blended with three different types of PPs in an elastic extruder. Dynamic mechanical behaviour of these blends have been determined, compared and analyzed at different temperatures.

Materials and Methods

The materials used in this study were blends of PP and PET. Polypropylenes were used as a matrix material and PET was present as second phase. Table-1 lists the source of polymers, physical properties and names of their suppliers.

Blend Composition Preparation

All the blend materials were extruded and then compressed in a hot compression moulding machine. PET concentration was 5, 10, 20, 30 and 50 wt% was supplied in to the extruder along with PP. The blends prepared by this machine were used as a base material for compression moulding. Table-2 lists the composition of ingredients along with their densities. The weighed amounts of the material for moulded samples were piled in the mould cavity. Then, the samples were compressed into 150 x 150 x 3 mm size sheet for 3 minutes in a hot press at 5 MPa pressure and then cooled in a cold water circulating press at the same pressure. The densities of blends were measured by using Shimadzu, Japan make Pycnometer model Accupyc 1330.

Dynamic Mechanical Sample Preparation

The 3 mm thick sheet was cut into 35 mm x 5 mm x 3 mm size samples for dynamic mechanical measurements.

Dynamic Mechanical Measurements

Dynamic visco elastic analyzer (DVA) model (DVA220) I.T.I. Co Ltd. Japan make was used to measure storage modulus (Er) and dissipation factor (tan δ) at different temperatures ranging from 20-270°C. Measurements were done in tension mode and at 1 Hz frequency. Storage modulus (Er) and dissipation factor (tan 5) graphs were plotted by using a plotter attached to DVA.

RESULTS AND DISCUSSION

Fig.-1 shows the variation of storage modulus (Er) and tan δ with temperature for virgin PP (A 5012) at 1 Hz frequency. In this case, it was found that storage modulus (Er) decreased from 1.06×10^9 to 1.0×10^4 Pa during the temperature change from 40.1 to

Table- 1: Source of Polymers

SN	Sample Name	State	Density (gm/cc)	Source
1	PP (A 5012)	Unmodified, High viscosity	0.9040	Chisso Polypro Co. Ltd., Japan
2	PP (K 7050)	Unmodified, Low viscosity	0.9060	Chisso Polypro Co. Ltd., Japan
3	PP (XKP 707 W)	Modified	0.9084	Chisso Polypro Co. Ltd., Japan
4	PET (MA 2101)	Virgin	1.3413	Unitica Co. Ltd., Japan

Table- 2: Wt % of ingredients of the Blends and their Densities

Table- 2a

Sample	PP (A 5012)	PET (MA 2101)	Density (gm/cc)
(A)	95	5	0.9324
(B)	90	10	0.9385
(C)	80	20	0.9723
(D)	70	30	1.0109
(E)	.50	50	1.0997

Table- 2b

Sample	PP (K 7050)	PET (MA 2101)	Density (gm/cc)
(F)	90	10	0.9322
(G)	80	20	0.9676
(H)	70	30	1.0038
(I)	50	50	1.0588

Table- 2c

Sample	PP (XKP 707W)	PET (MA 2101)	Density (gm/cc)
(J)	95	5	0.9275
(K)	90	10	0.9446
(L)	80	20	0.9810
(M)	70	30	1.0173
(N)	50	50	1.0801

176.2 °C.The tan δ value goes on increasing with temperature up to 6.09 x 10^{-1} at 176.2 °C. Maximum rate of decrease in Er was found at 165.1 °C. This sharp decrease is due to melting of Polypropylene.

Pure PP is highly crystalline. Er values for PP (K 7050) at 40.3 and at 173.1 °C were 1.18 x 10^9 and 1.04 x 10^4 Pa Fig.-2. Er goes on decreasing with increase of temperature. Er had maximum rate of decrease at 168.1°C and had the value 1.78 x 10^6 Pa. The tan δ at 173.1°C was 4.52 x 10^{-1}. This difference in Er behaviour is due to the higher modulus of this PP. Peak value ot tan δ was lowest in case of PP (K 7050), which was a low viscosity polymer and. gave lowest tan δ values at peak positions.

Fig.-3 shows that Er values for PP (XKP 707 W) at 40.2 and 175.1 °C were 1.53 x 10^9 and 1.39 x 10^4 Pa respectively. The tan δ at 175.1 °C was maximum 6.68 x 10 It was found that decrease in Er has two steps decrease for PP (XKP 707 W). These two temperatures were 88.2 and 143.1 °C respectively. Maximum decrease in Er and peak in tan δ for PET (MA 2101) appeared

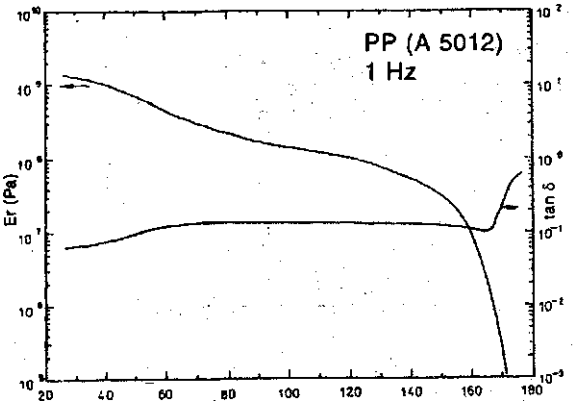

Fig. -1: Er and tan δ variation with temperature for PP (A 5012)

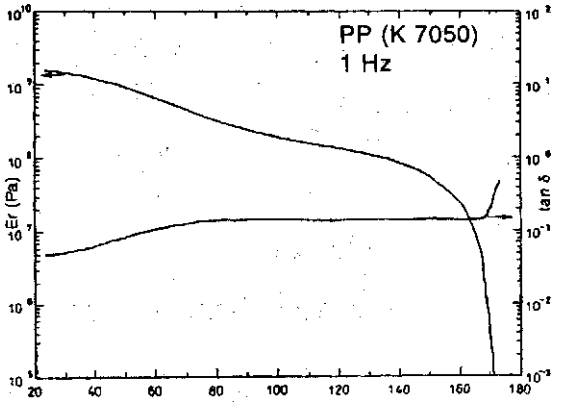

Fig. - 2: Er and tan δ variation with temperature for PP (K 7050)

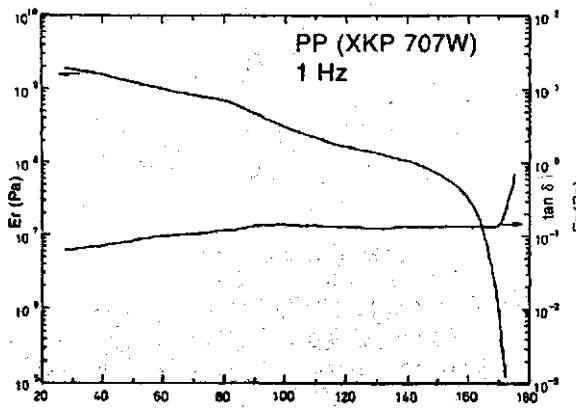

Fig. - 3 : Er and tan δ variation with temperature for PP (XKP 707 W)

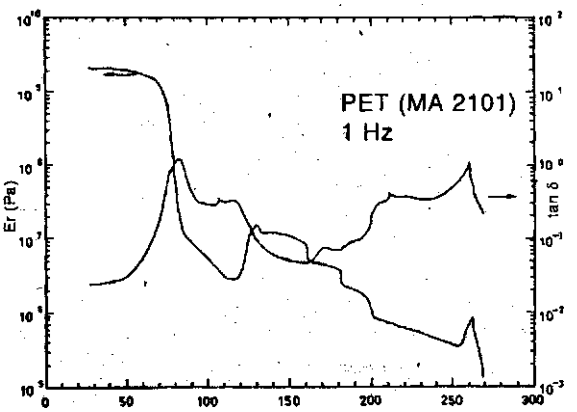

Fig. -4 :Er and tan δ variation with temperature for PET (MA 2101)

at 82.1 °C as shown in Fig.-4. Change occuring around 40°C in all the three types of PPs corresponds to β relaxation as is reported in the literature[12]. This is due to some impurities. This tan 8 peak corresponds to glass transition temperature of PET (MA 2101). Er value at 40°C was 2.08 x 10^9 Pa. Storage modulus of virgin PET (MA 2101) was highest as compared to all the three types of PP used in this study.

Umemura *et.al.*[8] reported that polypropylene crystallites obtained in unoriented polypropylene film have spherulitic structure. Spherulite size of this sample increased with higher crystallization temperature and or with longer crystallization time. Biaxially oriented films did not have spherulitic structure. Modulus is due to spherulitic structure. These PP sheets show that highest Er is for PP (XKP 707 W) and lowest is for PP(A5012). This is because PP (XKP 707 W) is a modified Polypropylene.

Table-3a shows the changes occurred in storage modulus (Er) and tan δ behaviour along with the melting peak position obtained for PP (A 5012), PP (XKP 707 W) and in case of PET (MA 2101). This shows that PP do not show any peak around 180 °C, while Fig.-4 shows a peak at 82.1 °C, which corresponds to Tg of PET. Which is in agreement with the reported Tg peak of PET, which is 69°C[13]. This peak is a weak endothermic peak. Melting endotherm is obtained at 260.1 °C in Fig.-4 is due to PET only. The peak obtained in tan δ around 117.1 °C corresponds to crystallization exotherm of PET, which is similar to the DSC curves obtained by Qujnn[11]. He found a weak endotherm at 80°C in unstressed PET yarn, which he proposed the Tg of PET. He also found another crystallization exotherm at 115 °C along with a melting endotherm at 255 °C.

Fig. - 5a and b show Er and tan δ plot for PP (A 5012) / (MA 2101) / (90 / 10) and (50 / 50) composition respectively. This shows that Tg of PET in blend appeared around 79.1°C, while melting appeared in both the cases around 173 °C. In this graph melting is dominated by PP melting. In case, when PET content is 50 % a clear Tg peak of PET is obtained in Fig.-5b.This is not very clear in case of lower PET content blends.

Table-3b lists the temperature at which storagemodulus (Er) changes.Table-3b also shows the Tg peak positions and melting peaks. Peak around 80 is clearly visible in blends on changing the PET content from 10 % to 50 %. Fig.-6a and b are the typical plots for 10 wt % PET blend 50 wt % PET blend with PP (K 7050).These show that Tg peak shited to higher temperature to around 82.2 °C due to addition of more PET. Melting peak observed around 175 °C.

An important observation is that type of PP decides the Tg and Er change temperature. Melting peak observed in tan 5 plots is not influenced much on adding PET

Table- 3a: Er/tan δ Changes

Sample Name	Ist change in tan 5 (°C)	IInd change in Er (°C)	Melting Peak (°C)
1. PP(A 5012)	Increased at 41.1	132.1	176.2
2. PP(K7050)	Increased at 40.3	141.1	173.1
3. PP(XKP 707W)	Increased at 40.2	88.2/143.1	175.1
4. PET(MA2101)	Peak 82.1 (Tg)	117.1	260.1

Table- 3b: tan δ peaks

Sample	Peak I[st] (°C)	Temp, of Er change (°C)	Melting Peak (°C)
PP (A 5012)/ PET (MA 2101)			
A (95/5)	66.1	133.1	170.1
B (90/10)	79.1	140.1	172.1
C (80/20)	80.1	141.1	176.1
D 70/30)	81.1	140.1	172.1
E (50/50)	79.1/115	142.0	173.1
PP(K 7050) / PET(MA 2101)			
F (90/10)	78.1	136.1	178.1
G (80/20)	83.2	133.1	172.1
H (70/30)	79.1	134.2	170.1
I(50/50)	82.2	141.1	175
PP(XKP 707W)/ PET(MA 2101)			
J (95/5)	93.1	145.1	175.1
K (90/10)	94.2	150.1	174.1
L (80/20)	95.1	147.1	176.1
M (70/30)	82.1	147.2	174.1
N (50/50)	82.2	151.2	173.1

because, it corresponds to PP Sample E has an additional peak at 115°C. This corresponds to α_c peak of PP as reported in literature for pure PP Fig.-7a and7b show the variation of Er and tan δ with temperature at 1 Hz frequency for PP (XKP 707 W) and PET (MA 2101) blend having 10 and 50 wt. % of PET. Table- 3b shows first peak around 93.1 °C in case of 5 wt % of PET. The tan 8 peak in PP (XKP 707 WJ/PET (MA 2101) appeared at 82.2 °C on adding 50% wt. of PET. Which corresponds toTg of PET. There is an increase in temperature for change in Er from 150.1 to 151.2 °C on increasing the PET content. The melting endotherm appeared around 173.1 °C, which is higher temperature as compared to PP (A 5012) blend.

CONCLUSION

1). All types of PP show a Er change around 40 °C and a melting peak in tan 5 plot.

2). Addition of PET shows clearTg peaks around 82 °C in all the blends. 3). In case of all 50 / 50 blends two sharp peaks appeared due to addition of PET. These two peaks at 82.1 and 117.1°C correspond to weak endothermic peak of Tg of PET and second peak is exothermic crystallization peak.

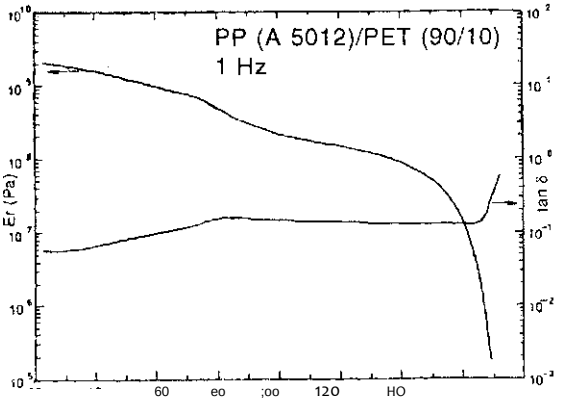

Fig. - 5a: Er and tan δ variation with temperature for PP (A 5012) / PET (MA 2101)/(90/10)

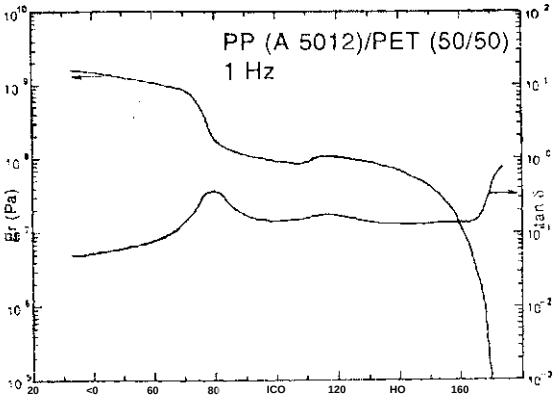

Fig. - 5b: Er and tan δ variation with temperature for PP (A 5012)/ PET / (MA 2101)/(SO/SO)

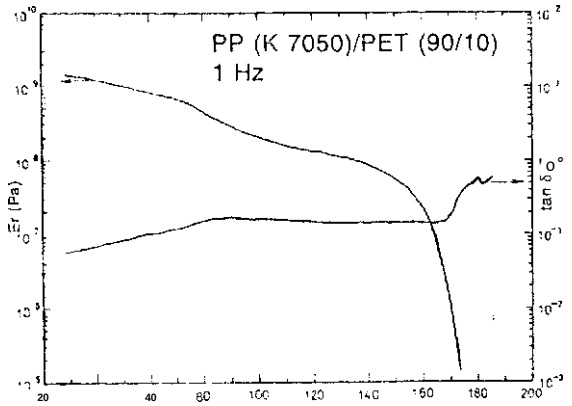

Fig. - 6a : Er and tan δ variation with temperature for PP (K 7050) / PET (MA 2101) / (90/10)

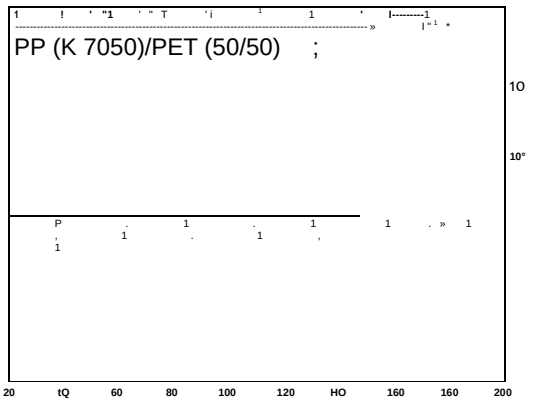

Fig. - 6b: Er and tan δ variation with temperature for PP (K 7050) / PET (MA 2101)/(SO/SO)

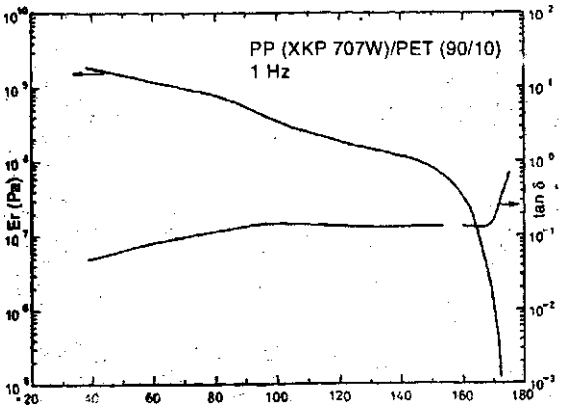

Fig. - 7a: Er and tan 6 variation with temperature for PP (XKP 707 W) / PET (MA2101)/(90/10)

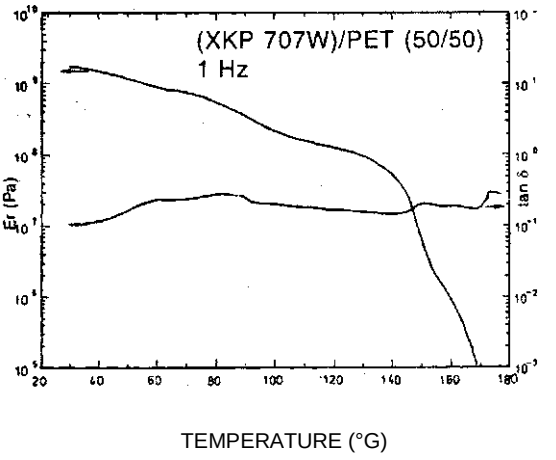

Fig. - 7b: Er and tan 8 variation with temperature for PP (XKP 707 w) / PET (MA 2101)/ (SO/SO).

REFERENCES

1. Wilen C. E., Falden M. Nylund and Nasman J., *J. Appl. Polym. Sci.,* **50** 1541(1993)
2. Michler G. and Strake J., *Chap. 17 in Toughned Plastics II, K, Riew and A.Kinloch, eds.,* ACS (1996)
3. Utracki L A., ed., Two-Phase Polymer Systems, Hanser Pub. (1991)
4. Fortelny, Navratilova E. and Kovar J., *Die Ange. Makrom. Chem.,186* 195(1991)
5. Striker F. and Mulhaupt R., *Polym. Eng. Sci.,* **38** 1463(1998)
6. Gupta A. K., Kumar P. K. and Ratnam B., *J. Appl. Polym. Sci.,* **42,** 2295(1991).
7. Umemura T. and Condese. A. **I,** *Ann. Rep.CEIDP* 586(1982)
8. Umemura T. Akiyama K. and Couderc D., *IEEE Trans. Elect. Ins.* E/-21.2 137(1986)
9. Ellis J. W. "and Picot J. J. C, *Polym. Engineering and Science.* **40,**7 (2000) 1622
10. Gupta A. K., Chand Navin and Man Singh A., *Anlsotropy of dielectric relaxation in PET fibers, Polymer,* **20** 875(1979)
11 Qujnn R. G., *J. Appl. Polym Sci,* **16** 3393(1972)
12. Umemura Kashiwazaki T. and Suzuki T., Japan *J. Applied Phys,,* **19,** 767(1980).
13. Di Lorenzo M.L., Errico M. E., Avella M., *Journal of Material Science* **27** 2351-2358(2002)