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Phase behaviour in ternary polyamide 6/polyamide 66/elastomer blends

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Abstract

The phase behaviour of the ternary blend system polyamide 6/polyamide 66/elastomer has been interpreted by the binary interactions of the respective polymers. Three different elastomer components have been studied; random ethene–propene copolymer (EPM), ethene–propene–diene copolymer grafted with maleic anhydride (EPDM-*g*-MA) and ethene–1-octene copolymer also grafted with maleic anhydride (EO-*g*-MA), respectively. The miscibility of the polymers is predicted by the Flory–Orwoll–Vrij equation-of-state theory using the pVT data in connection with the solubility parameter concept. The data suggests that polyamide 6 (PA 6) and polyamide 66 (PA 66) are miscible within the practically accessible temperature range from 270 to 310°C and all types of elastomer are immiscible with both polyamides due to strong repulsive interactions mainly characterised by large differences of the characteristic pressure between polyamides and elastomers. Thus the interaction parameters χ_{ij} between the polyamides and the different elastomers exceed the critical χ parameter unambiguously. Therefore, all ternary systems are heterogeneous. This is confirmed by dynamic mechanical analysis experiments where a single glass transition was only observed for PA 6/PA 66 blends, whereas two glass transition temperatures are observed for all ternary blends. The heterogeneous systems show a glass transition temperature depression for the ethene–propene copolymer and ethene–octene copolymer grafted with maleic anhydride depending on the composition in the ternary blends. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Phase behaviour; Polyamides; Ternary blends

1. Introduction

Multicomponent polymer systems with more than two phases have properties, which can frequently not be achieved by binary polymer systems. Most ternary systems discussed in the literature are usually based on polymers and solvent or solvent and non-solvent [1,2] or two polymers and one copolymer or modified polymer as compatibiliser [3,4]. Other investigations have been made for ternary polymer blends. Only in exceptional cases, where all three respective binary blend pairs are miscible—as in the system poly(hydroxy ether of bisphenol A)/poly(vinyl methyl ether)/poly(ϵ-caprolactone) a complete miscibility over the whole composition range can be observed [5]. Most ternary blends have an immiscibility gap as studied for the system poly(methyl methacrylate)/poly(ethylene oxide)/phenoxy [6]. Ternary blends containing two miscible polymers and one polymer which is immiscible with both the other polymers are always systems with an immiscibility gap. In the case that the interaction parameters differ strongly from each other the ternary system is completely immiscible.

Theoretical and experimental results on the blend system polyamide 6/polyamide 66 (PA 6/PA 66) give strong interactions between both polymers [7,8]. Caused by the similar chain architecture of PA 6 and PA 66 a molecularly dispersed system can be expected in the melt. Ellis [7,8] considered polyamides as copolymers containing different numbers of methylene and amide groups occupying different volumes. The blend behaviour is then interpreted in terms of binary interaction parameters between different copolymer sequences. Since PA 6 and PA 66 have an identical volume fraction of methylene groups the miscibility is favoured.

In general, polyamides are immiscible with polyolefins or polyolefin based elastomers due to the structural differences (absence of specific interactions) and also caused by free volume effects resulting from different thermal expansion coefficients α. But such polyamide/elastomer blend systems have a relevant practical application. The elastomer component increases the toughness especially immediately after injection moulding and at low temperatures. This effect is mainly achieved by blending polyamides with elastomers with a T_g usually in the range of -30°C or lower. The

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Table 1
Materials

Polymer	M_n (g/mol)	M_w/M_n	ρ (g/cm^3)	γ_s[a] (mN/m)
PA 6	42 000	1.89[b]	1.13	36.38
PA 66	26 000	1.83[b]	1.13	35.23
EPDM-*g*-MA (8.7 wt% ENB, 55 wt% ethene)	117 000	2.6[c]	0.85	35.43
EPM (78 wt% ethene)	71 000	1.98[b]	0.88	27.87
EO-*g*-MA (11 wt% octene)	105 000	2.98[b]	0.87	29.02

[a] Surface tension determined by contact angle measurements.
[b] Determined by GPC.
[c] Specification given by Uniroyal Chemical.

increase of the toughness is only observed when the elastomer particles are chemically bound across the interface to the polyamides. Therefore, the elastomers, especially the polyolefin-based elastomers, are functionalised with carboxylic, ester or anhydride groups. Most widely used polyolefins are grafted with maleic anhydride. Recently, super tough materials were created by the preparation of ternary blends of two polyamides or a copolyamide with an elastomer [9–12]. Mainly the influence of the particle size, the interparticle distance and the interfacial adhesion on the toughness was studied [13–15].

This contribution deals with the phase behaviour of ternary blends based on PA 6, PA 66 and three different elastomers; ethene–propene copolymer (EPM), ethene–propene–diene copolymer grafted with maleic anhydride (EPDM-*g*-MA) and ethene–octene copolymer also grafted with maleic anhydride (EO-*g*-MA). The binary interaction parameters χ_{ij} were calculated based on the pVT data. The blend behaviour was determined experimentally using the dynamic mechanical analysis (DMA). From the results especially the T_g shift was taken into consideration for the evaluation of the phase interactions.

2. Experimental

2.1. Materials

PA 6 (Ultramid B5) and PA 66 (Ultramid A4) from BASF and three types of elastomers were used: a random ethene–propene copolymer of Exxon (Exxelor PE805), an ethene–propene–diene random copolymer grafted with maleic anhydride of Uniroyal Chemical (Royaltuf 490) and an ethene–1-octene random copolymer grafted with maleic anhydride of DuPont (Fusabond MN493D). The characteristic data of the polymers are given in Table 1. Ternary blends of the system PA 6/PA 66/elastomer varied in the range (50:50):*x* (*x* ranges from 10 to 40 wt%) and binary blends of the system polyamide/elastomer were prepared by melt mixing in a laboratory single screw extruder with a mixing screw (Brabender) at temperatures between 275 and 300°C with a screw rate of 30 rpm.

2.2. pVT measurements

The thermodynamic data of PA 6 and PA 66 were taken from Zoller and Walsh [16]. The pVT data of EPM, EPDM-*g*-MA and EO-*g*-MA were measured with a Gnomix PVT apparatus (Boulder Co.). The change of the specific volume was measured in a temperature range between 25 and 300°C and for pressures between 10 and 200 MPa. For PA 6 the data were fitted to the Flory–Orwoll–Vrij (FOV) EOS in the temperature range from 250 to 300°C, for PA 66 from 265 to 300°C and for the elastomers from 180 to 300°C.

2.3. DMA measurements

DMA measurements were carried out in order to analyse the phase behaviour of the blends. The DMA measurements were done in the bending mode with a frequency of 1 Hz and a heating rate of 2 K min^{-1}. Specimens with the dimension of $8 \times 30 \times 1$ mm^3 were prepared from compression moulded plates. The temperature range of DMA measurements was between −120 and approximately 200°C. Prior to measurements the samples were dried in vacuum for 24 h at 110°C.

2.4. Phase-morphology study

SEM was used to evaluate the phase morphology of the ternary blends. The samples were fractured under liquid nitrogen and gold-coated before microscopy. A Cam Scan (CS 4) scanning electron microscope was used to observe the sample surface.

3. Results and discussion

3.1. Thermodynamics of the blends

3.1.1. Theoretical background

The simple models for polymer/polymer miscibility are based on the Flory–Huggins (FH) theory—often with various modifications, e.g. the consideration of the polydispersity of polymers, of occurrence specific interactions, the formation of hydrogen bonds between components [17], or

Table 2
Relevant physical parameters of the investigated polymers at 280°C

Polymer	$\alpha \times 10^4$ (K^{-1})	$\tilde{v}$	v^*_{sp} (cm^3 g^{-1})	$\tilde{T}$	T^* (K)	γ (MPa K^{-1})	p^* (MPa)
PA 6[a]	5.862	1.265	0.823	0.0596	9275	0.786	696
PA 66[a]	6.041	1.272	0.818	0.0606	9131	0.804	720
EPDM-*g*-MA[b]	8.948	1.369	1.001	0.0726	7618	0.452	468
EPM[b]	8.081	1.342	1.052	0.0696	7950	0.401	399
EO-*g*-MA[b]	8.254	1.343	1.042	0.0697	7855	0.440	438

[a] PVT Data acc. [16].

[b] PVT Data measured by Gnomix-PVT-apparatus (Boulder, CO).

free volume effects can be considered only by introducing of special terms [18–21].

The equation-of-state (EOS) theories consider the free volume effects originally neglected by the incompressible Flory–Huggins (FH) theory. However, the FH theory is simpler to implement than the EOS theory. So, the interaction parameter χ is often quantitatively from the EOS theory and used in the FH model [22].

The phase behaviour of ternary blends can be judged using the binary interactions expressed in terms of the Flory–Huggins interaction parameter (Eq. (1))

$$\chi_{ij} = \frac{V_{\mathrm{r}}}{\mathrm{R}T}(\delta_i - \delta_j)^2 \tag{1}$$

where δ_i and δ_j are the solubility parameters of the pure components, V_{r} the reference volume. In the case that one of the binary interaction parameters χ_{ij} is larger than the critical χ parameter (Eq. (2)), the whole system can be considered as immiscible.

$$\chi_{12\mathrm{crit.}} = \frac{V_{\mathrm{r}}}{2}\left(\frac{1}{V_1^*\phi_1} + \frac{1}{V_2^*\phi_2}\right) \tag{2}$$

The concept of solubility parameters is based on the theory of regular solutions. This leads to some restrictions as the strict absence of specific interactions, e.g. the occurrence of hydrogen bonds between the blend components. For the linear polyamides PA 6 and PA 66 it can be assumed that the hydrogen bonds are completely saturated at room temperature. The hydrogen bonds weaken with increasing temperature but they do not disappear completely [23]. Blending of PA 6 and PA 66 in the melt leads only in exceptional cases to hydrogen bonds between the different components, but, in the case that hydrogen bonds are formed they are not stable [24]. It can be assumed that there are no specific interactions between polyamide and the unmodified elastomer. In conclusion, it can be supposed that the solubility parameter concept is valid for blends of polyamides and blends of polyamides with elastomers. The concept can be used for the prediction of the blend miscibility entirely from the characteristic data of the pure components. The solubility parameter δ can be determined from the molecular cohesion energy ΔE_{v} and the molecular volume V_{m} [25]. ΔE_{v} cannot be determined directly but the term can be replaced by the negative configurational energy U [21].

The solubility parameter can be expressed in terms of the configurational energy or as a function of the characteristic pressure and the reduced volume (Eq. (3))

$$\delta = \left(-\frac{U}{V_{\mathrm{M}}}\right)^{\frac{1}{2}} = \frac{p^{*1/2}}{\tilde{v}} \tag{3}$$

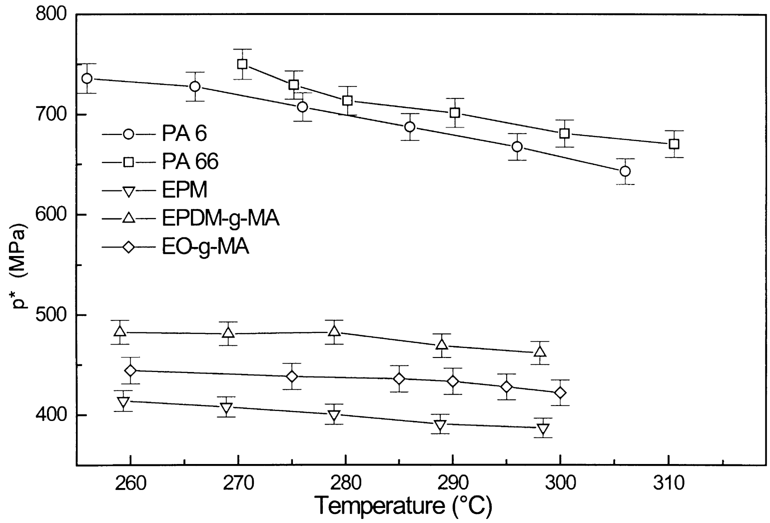

Fig. 1. Characteristic pressure of PA 6, PA 66, EPM, EPDM-*g*-MA and EO-*g*-MA as a function of temperature.

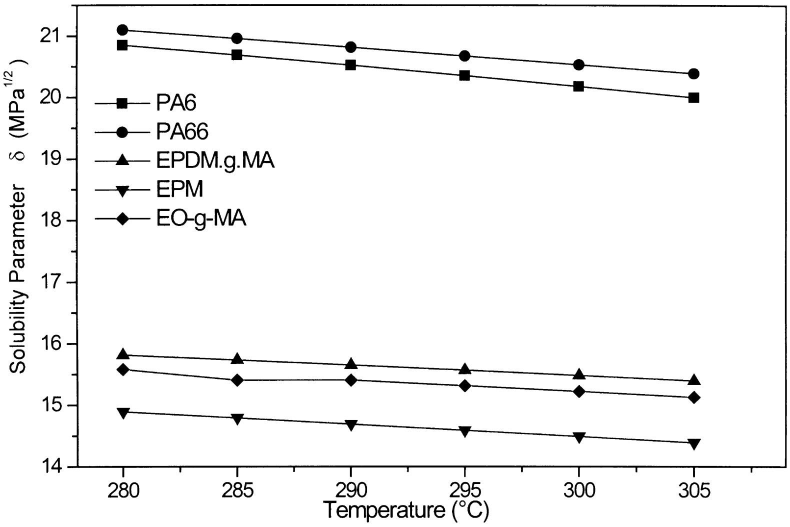

Fig. 2. Solubility parameters of PA 6, PA 66, EPM, EPDM-*g*-MA and EO-*g*-MA as a function of temperature.

By using the characteristic data p^*, V^* or v^*_{sp}, T^* of the pure components, the reduced quantities $\tilde{p}$, $\tilde{v}$ and $\tilde{T}$ can be obtained from Eq. (4).

$$\tilde{p} = \frac{p}{p^*}; \qquad \tilde{v} = \frac{v_{sp}}{v^*_{sp}}; \qquad \tilde{T} = \frac{T}{T^*} \tag{4}$$

The EOS of FOV (Eq. (5)) connects the reduced quantities.

$$\frac{\tilde{p}\tilde{v}}{\tilde{T}} = \frac{\tilde{v}^{1/3}}{\tilde{v}^{1/3} - 1} - \frac{1}{\tilde{v}\tilde{T}} \tag{5}$$

If the χ value exceeds the critical value of a binary blend $\chi_{ij\text{crit.}}$, it can be expected that the system separates into phases. The critical value is a function of composition, V_r and V^* expected, is given in Eq. (2).

3.1.2. pVT properties

All thermodynamic data of the polymers are summarised in Table 2.

The characteristic volume $v^*_{sp.}$ can be considered as the van der Waals volume and is additive from group increment contributions [25]. Since PA 6 has an identical ratio of CH_2 groups per CONH group as PA 66, it is clear that the values of $v^*_{sp.}$ are nearly identical.

The p^* values of all polymers are plotted in Fig. 1. The p^* values of all elastomers are significantly smaller compared to the values of the polyamides. Since the characteristic pressure p^* depends on the compressibility γ it correlates with the cohesion energy density. Therefore, a decrease of p^* with an increasing number of CH_2 groups can be expected. Generally, the rubbers have a smaller cohesion energy density compared to the polyamides. The p^* value of EPDM-*g*-MA is larger than that the value of EPM despite the fact that the propene content in EPM is smaller compared to EPDM (see Fig. 1). This is obviously a result of the presence of the diene groups in EPDM from the ethylidene norbornene.

Maier et al. [22] reported the solubility parameter of ethene–1-octene random copolymers from PVT data. The solubility parameter of the copolymer containing 64 wt% octene is about 14.6 $\text{MPa}^{1/2}$ at 280°C and increases to 15.5 $\text{MPa}^{1/2}$ when the comonomer content is only 2 wt%. Similar tendencies were observed from our calculation for ethene–propene copolymers: The solubility parameter decreases with increasing propene content.

The solubility parameter for EPDM-*g*-MA with 55 wt% ethene is higher than for EPM with 78 wt% ethene which is caused by the high compressibility as a result of cross-linking and increased polarity.

The solubility parameter as a function of temperature calculated from the pVT data are shown in Fig. 2. The solubility parameters of the melts of both polyamides are very similar; thus miscibility can be expected. Again a large difference of the solubility parameters exists between polyamides and the elastomers indicating complete immiscibility.

Using the solubility parameter obtained from pVT data, it is possible to extract the binary interaction parameter χ_{ij} of the polymer pairs, respectively, using Eq. (1). In the temperature range between 275 and 305°C the binary interaction parameter χ_{ij} of the pair PA 6 and PA 66 is smaller than the critical value indicating complete miscibility (see Fig. 3a). The calculations indicate a possible lower critical solution temperature (LCST) at temperatures higher than 320°C for 50/50 wt% blend ratio. The critical value $\chi_{ij\text{crit.}}$ is only a function of the degree of polymerisation and the specific molecular volume of the components. For calculation it was assumed that all elastomers have an identical molecular mass ($M_n = 100.000$ g/mol).

As expected the used polyamides are completely immiscible with elastomers investigated (see Fig. 3b). The critical

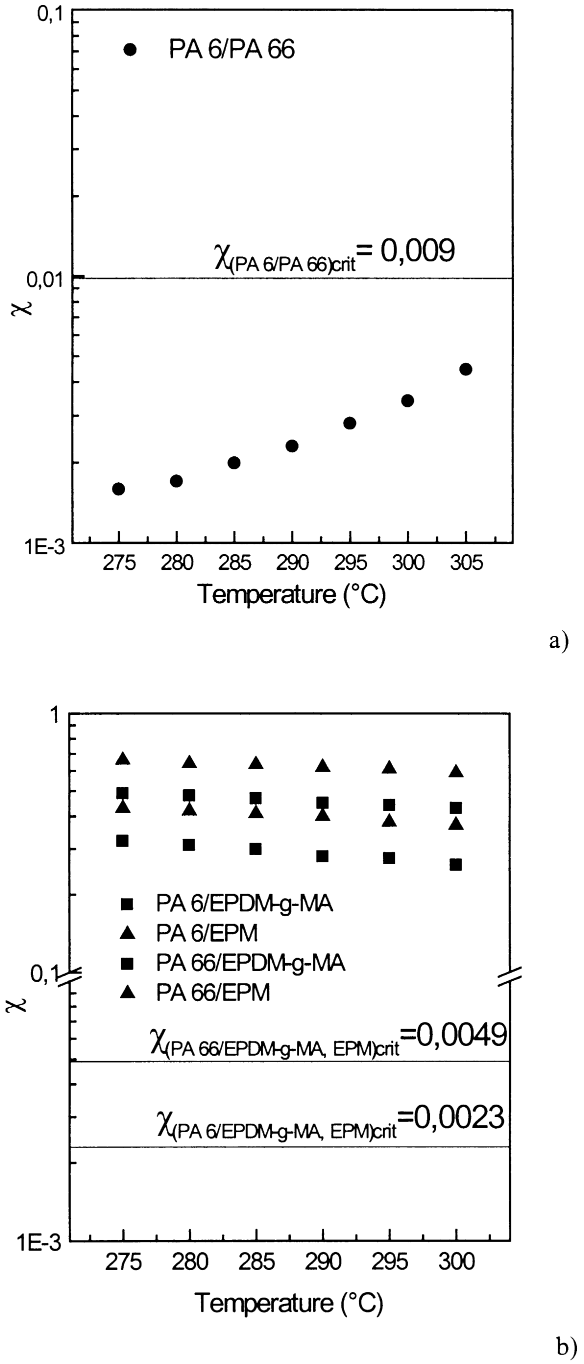

Fig. 3. χ parameters as a function of temperature for the polymer systems: (a) PA 6/PA 66; (b) PA 6/EPM, PA 6/EPDM-*g*-MA, PA 66/EPM, and PA 66/EPDM-*g*-MA.

value of $\chi_{ij\text{crit.}}$ between PA 6 and the elastomer is smaller compared to that between PA 66 and the elastomer due to the smaller molecular mass of PA 66.

Fig. 4 presents the phase morphology of blends of PA 6/PA 66 and of their blends with a non-reactive and reactive elastomer. The SEM micrograph in Fig.4a presents the homogeneous character of the morphology in the PA 6/PA 66 blend but strongly separated elastomer particles in the PA 6/PA 66/EPM blend with 10 wt% EPM (Fig. 4b). In compatibilised PA 6/PA 66/EPDM-*g*-MA blend at the same composition it is not possible to distinguish the elastomer phase. The fracture surface is characterised by a non-broken polyamide/elastomer interface (Fig. 4c). This is due to the chemical coupling across the interface. However, small elastomer particles with about 1 μm diameter embedded in the polyamide matrix can be observed.

3.2. Segment mobility and relaxation in polymer blends

3.2.1. Influence of the miscibility on the T_g in immiscible systems

The position of the glass transition temperature T_g of polymers depends on the structure and the co-operative mobility of the segments. The relaxation behaviour of blends seems to be an important indication for intermolecular interactions and thus for polymer/polymer miscibility of the amorphous regions. Strong changes of the segment motion can be induced by the thermodynamic miscibility in polymer blends. This behaviour is reflected by the occurrence of single composition dependent glass transition temperature T_g in miscible blends. In the case of partial miscibility, the glass transition temperatures of the blend components remain separated but the T_gs are shifted in comparison to the T_gs of the pure components toward each other. In completely immiscible polymer blends the T_gs remain largely unaltered but a change of the segment relaxation parameters has not been out of the question [26]. This can be caused by different mechanisms, e.g. by changes of the molecular mass distribution at the interface or so-called surface enrichment or the development of locale stresses as a result of different thermal expansion coefficients of the blend components. Depending on the ratio of the thermal expansion coefficients the T_g of the dispersed phases shift towards lower or higher temperatures. A negative shift toward lower temperatures can be observed in the immiscible polymer blend systems compatibilised PA 6/multifunctionalised polyethylene [27] and by isotactic polypropylene/polystyrene-*block*-poly(ethene-*co*-buthene)-block-polystyrene (SEBS) [28]. A change of the segment mobility of the system poly(butylene terephthalate)/polycarbonate prepared with a thermal history and blend composition which leads to immiscibility, was also observed by Kolesov et al. [26]. The interpretation of measurements of thermally stimulated depolarisation current comes to the result that the structure of the PC-rich phase is more relaxed because of the locally disordered interpenetration of the flexible PBT molecules through the PC volume.

Three different relaxation processes α, β and γ for polyamides have been known: (i) the α relaxation belongs to the glass transition and is connected to the co-operative mobility of 25–50 chain atoms; (ii) the β relaxation is associated with the mobility of amide groups with weak hydrogen bonds to neighbour chains; (iii) the γ relaxation is a result of the motion of short sequences of methylene groups

[29,24]. The temperature of the α and β relaxation, respectively, is strongly influenced by moisture. The T_g shifts to lower temperatures with increasing water content, even to glass transition temperatures below room temperature. The shift of the β relaxation is not as strong as that of the glass transition but the intensity is additionally increased with the increased content of bounded water. This effect can be explained by the addition of water molecules to the amide groups, which is connected with a higher local degree of molecular motion. The β relaxation does not occur in that case if polyamide is slowly cooled from the melt and isothermally annealed so that the number of carbon amide groups with weak hydrogen bonds decreased. The β relaxation does also not appear in completely dry and slowly cooled samples [29].

3.2.2. Relaxation behaviour of ternary PA 6/PA 66/elastomer blends

The glass transition temperatures (α transition) determined by DMA for PA 6 and PA 66 are at 63 and 74°C, respectively (Fig. 5). This corresponds to the value of the dry polyamides [29]. At low temperatures a β peak with a small intensity can be observed probably due to the relatively fast cooling process of the test specimen. This results in the preservation of weak hydrogen bonds between the carbon amide groups. The binary PA 6/PA 66 blend (50/50) relaxes at the same temperatures as PA 6, however the peak comprises also the area of the PA 66 relaxation. The β peak is slightly broader compared with the β peaks of the pure components and the maximum is at −63°C (Fig. 5).

The temperature dependence of the loss factor tan δ and the storage modulus E' for EPM, EPDM-*g*-MA and EO-*g*-MA is shown in Figs. 6 and 7. The diene groups of EPDM-*g*-MA are cross-linked during the thermal treatment as observed by solvent-swelling measurement. Cross-links cannot be dissolved by the thermal motion. The rubbery state extends up to temperatures of the thermal degradation of polymers. The storage modulus decreases in the softening range and stays nearly constant after achieving the glass transition temperature. In contrast EPM and EO-*g*-MA used in this work are not cross-linked and remain only up to 50°C in the rubbery state.

The glass transition temperatures of EPDM-*g*-MA, EO-*g*-MA and EPM are at −53, −43 and −33°C, respectively, as shown in Fig. 6. The EO-*g*-MA and especially EPM relax in a broad temperature range.

In compatibilised ternary blends of PA 6/PA 66/elastomer a change of the relaxation behaviour in the range of the single signal of the glass transitions of PA 6/PA 66 cannot be observed. The peak maximums of EO-*g*-MA as well as of EPM are shifted with increasing polyamide content towards lower temperatures up to the β transition of polyamides, as shown in Figs. 8 and 9. This T_g depression is caused by an increase of the thermal stress across the elastomer domains attributed to differences in thermal expansion coefficients of the blend components resulting in a negative pressure of the

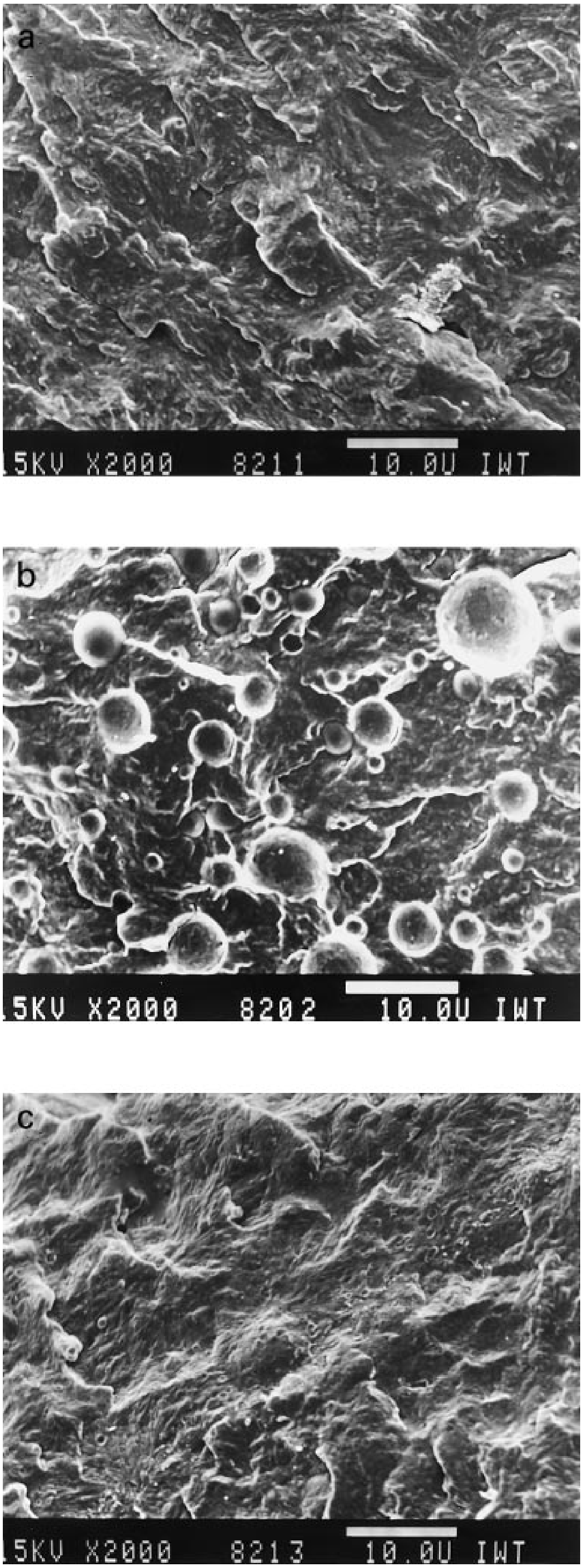

Fig. 4. SEM micrographs of PA 6/PA 66/elastomer blends by weight ratio: (a) PA 6/PA 66 50/50; (b) PA 6/PA 66/EPM 45/45/10; (c) PA 6/PA 66/EPDM 45/45/10.

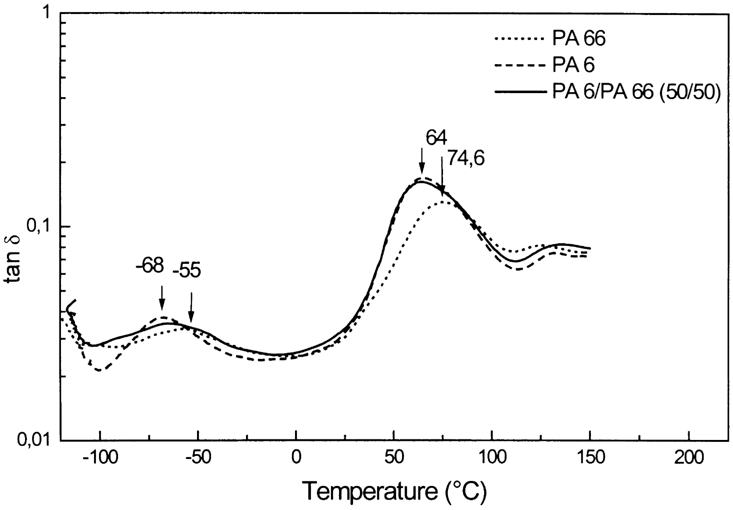

Fig. 5. Loss factor tan δ of PA 6, PA 66 and their blend by weight ratio 50:50.

elastomer domains. This is associated with an increase of the free volume of the rubber component and therefore with increasing the motion of rubber molecules. In the presence of a coupling agent at the interface, the rubber phase is finely dispersed as e.g. in the case of EO-*g*-MA blends. A direct result of the interfacial coupling is an increase of the interfacial thickness [30]. The extended interfaces (interfacial thickness, interfacial area) contribute to a damping of the mechanical stresses and therefore the glass transition temperature depression is small (Fig. 8). This damping in the blends with EPDM-*g*-MA is obviously stronger than in the case of EO-*g*-MA containing blends. A possible reason is the higher surface tension of EPDM-*g*-MA compared to the value of EO-*g*-MA, as shown in Table 1. Thus the interfacial tension between EPDM-*g*-MA with polyamide is smaller compared to that of EO-*g*-MA with polyamide. Thus, there does not exist any shift of the relaxation of EPDM-*g*-MA in blends with polyamides (Fig. 10).

The storage modulus E' of the ternary blends, as displayed in Figs. 11–13, at the temperature range between the T_g of the components decreases with increasing rubber content as expected. In the blend PA6/PA 66/EPDM-*g*-MA (45/45/10) it can be seen that small rubber contents do not cause an immediate decrease of the storage modulus E' (see Fig. 12).

4. Conclusions

The miscibility behaviour of ternary PA 6, PA 66 and elastomer blends has been interpreted by binary interactions employing the FOV EOS theory using the concept of solubility parameters. There is a clear indication from experimental results and from theoretical calculations that PA 6 and PA 66 are miscible in the amorphous phase at all temperatures and blend compositions. In contrast, all blends

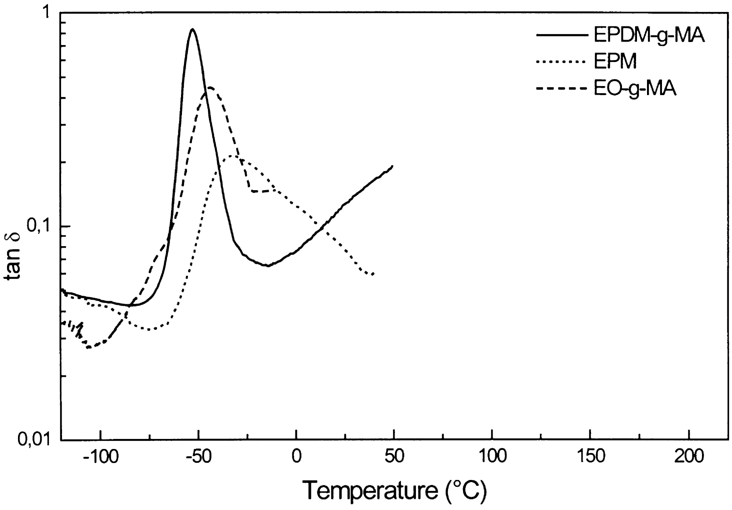

Fig. 6. Loss factor tan δ of EPDM-*g*-MA, EPM and EO-*g*-MA.

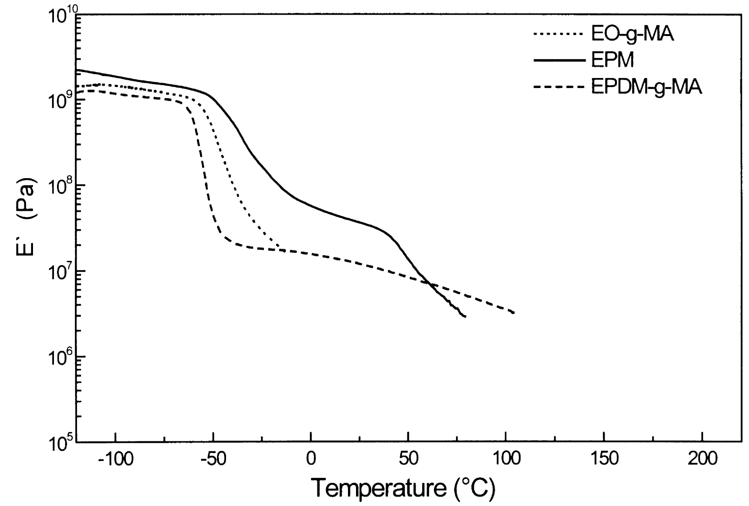

Fig. 7. Dynamic modulus of elasticity E' of EPM, EPDM-*g*-MA and EO-*g*-MA.

containing polyamide and a rubber phase show complete immiscibility.

Furthermore, it was observed that the solubility parameter of ethene–propene random copolymers increases with the decrease in propene content. But this tendency is not sufficient to approach these to the higher solubility parameters of the polyamides. Thus the immiscibility remains. The EOS theories can be used only for additional volume effects, but they are not able to take into consideration a chemical reaction. The EPDM and EO copolymers used were grafted with maleic anhydride. This leads in the melt to a chemical coupling across the interface during the blending with polyamides. The DMA and SEM results indicated a clear phase separation in ternary blend systems of PA 6/PA 66/elastomer. The development of local mechanical stresses at the interface caused by differences in the thermal expansion coefficients leads to a decrease of the T_g of the elastomer. In chemically compatibilised systems it is possible that the stresses can relax more due to the relatively large extension of the interfacial thickness and has an increase of the interfacial area as a result of smaller rubber domains. Due to these mechanisms, which are partly contrary, the T_g of the rubber in ternary EO-*g*-MA blends is only slightly decreased and it remains constant in blends with EPDM-*g*-MA.

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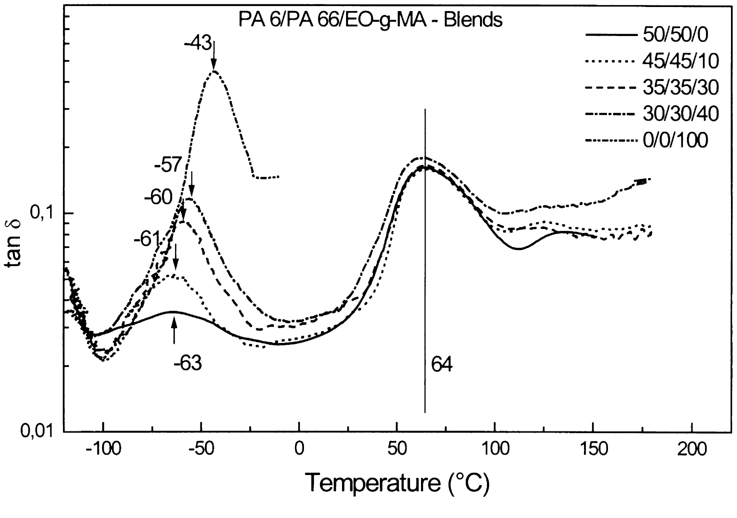

Fig. 8. Loss factor tan δ of ternary PA 6/PA 66/EO-*g*-MA blends.

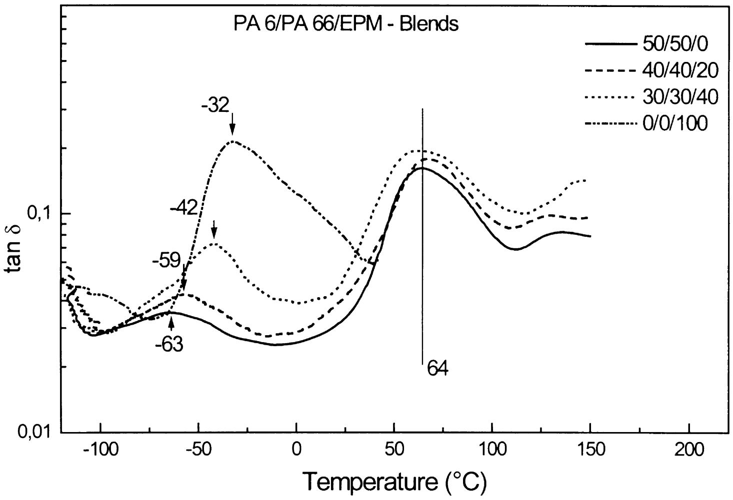

Fig. 9. Loss factor tan δ of ternary PA 6/PA 66/EPM blends.

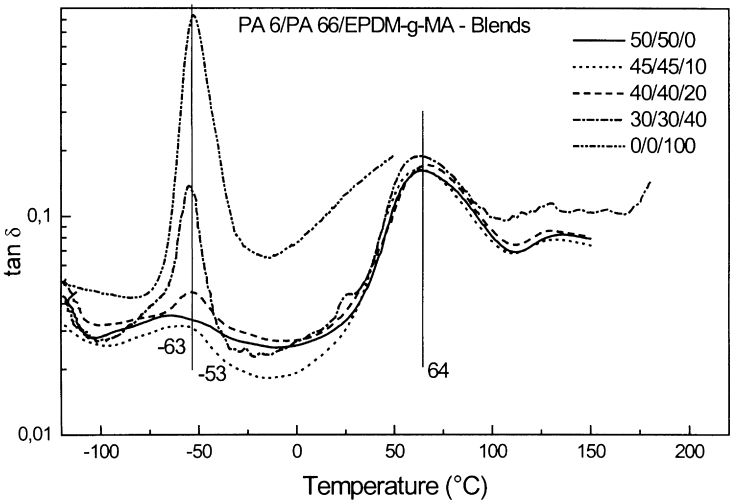

Fig. 10. Loss factor tan δ of ternary PA 6/PA 66/EPDM-*g*-MA blends.

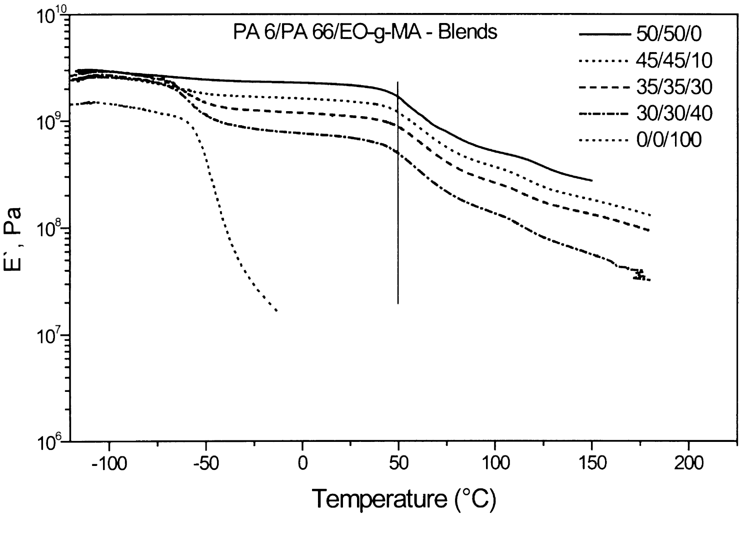

Fig. 11. Dynamic modulus of elasticity E' of PA 6/PA 66/EO-*g*-MA blends.

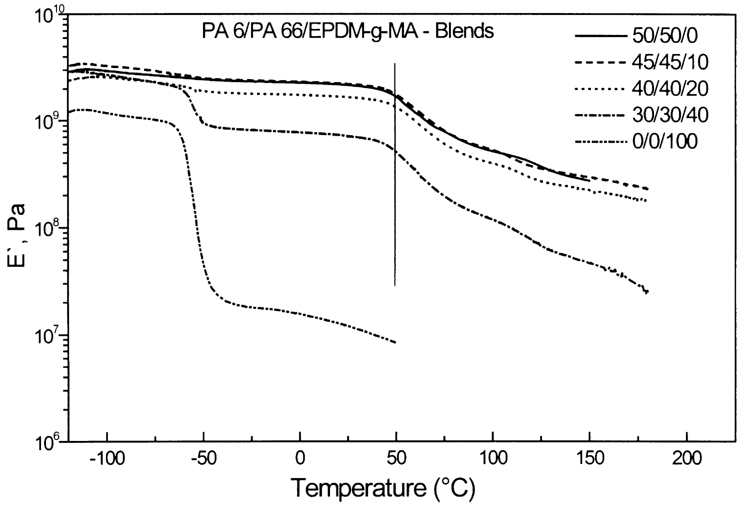

Fig. 12. Dynamic modulus of elasticity E' of PA 6/PA 66/EPDM-*g*-MA blends.

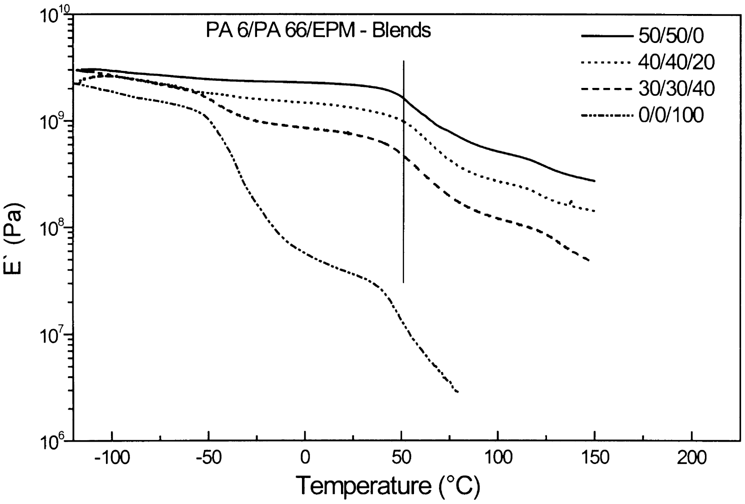

Fig. 13. Dynamic modulus of elasticity E' of PA 6/PA 66/EPM blends.

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