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# Correlation between Morphology and Mechanical Properties of TPEs from PA (6, 66) / EPDM Blends

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**Abstract:** In the present work, a systematic study has been carried out to investigate the effect of incorporation of varying amounts of EPDM on mechanical properties of PA(6,66)/EPDM binary blends. First we have studied the effect of blend ratio on the mechanical properties of PA (6,66) /EPDM blends. From this we selected three blend ratios viz. 30/70 (EPDM rich blend), 50/50 (co-continuous blend) and 70/30 (nylon rich blend) for further studies. The phase morphology of the blends was investigated using scanning electron microscopy with special reference to blend ratio. Mechanical properties of the blends were correlated with the morphological parameters. Explanations and illustrations based on the results of above studies are presented in this communication.

**Keywords:** Blends, Mechanical properties, Morphology

## I. INTRODUCTION

Blending of an elastomer with a plastic provides thermoplastic elastomers (TPEs) of diverse nature. These materials have significant commercial interest [1-5]. The process of polymer blending is a versatile and widely used method for fabricating cost effective engineering products. Thermoplastic Elastomers (TPEs) are reprocessable materials [6]. The physical properties of the blends are controlled by the size of the dispersed phase, its dispersibility and its interfacial interactions [7,8]. The key factors affecting the mechanical properties of TPEs are the morphology and the compatibility between the blend components [9].

Nylon (PA) and EPDM could form blends with very interesting properties because nylon offers very good mechanical properties and EPDM offers good barrier to moisture. Nylon/rubber blends have been extensively studied by several researchers [10-19]. In contrast to PA 6 and PA 66 blends, less information is available on the properties of PA copolymer (PA6,66) blends. Therefore it is challenging and interesting to develop super tough thermoplastic elastomers with PA (6,66). It was observed that as the time of mixing increases the dispersed domain size decreases in the case of PA (6,66) / EPDM blends. Thomas and Groeninckx. [19] studied the effect of processing conditions on the morphology development of nylon6/EPM blends in 1999. Paul and co-workers [20] studied the mechanical properties of blends of nylon/EPM-g-MA. They observed strain hardening and cold drawing for the nylon rich blend systems.

## II. EXPERIMENTAL

### A. Materials and Methods

Nylon used in this work is a copolymer of PA6 and PA66, ( Tufnyl F-120 ) was procured from Sri Ram Fibers Ltd. Madras, India. The EPDM (Keltan 720) was obtained from DSM, Netherlands.

### B. Blend Preparation

PA(6,66) was preheated in vacuum oven at 80°C for 24 hrs and kept in vacuum desiccator. This preheated and cooled nylon was used for blend preparations. The blending was performed in Haake Rheocord 600 mixer. PA (6,66) was first charged into the melting chamber and melted at 180°C for 2 minutes at a rotor speed of 60 rpm. The masticated EPDM was then fed in to the mixer. Total mixing time was fixed at 8 minutes to ensure homogeneity. Time and torque were also noted while mixing. The formulation of the blends is given the Table 1.

Table 1: Formulations of the blends

Material	N <sub>0</sub>	N <sub>20</sub>	N <sub>30</sub>	N <sub>40</sub>	N <sub>50</sub>	N <sub>60</sub>	N <sub>70</sub>	N <sub>80</sub>	N <sub>100</sub>
PA (6,66)	0	12	18	24	30	36	42	48	60
EPDM	60	48	42	36	30	24	18	12	0

## III. RESULTS AND DISCUSSIONS

The processing characteristics of the blends have been studied from the Rheomix time-torque curves ( Fig. 1).

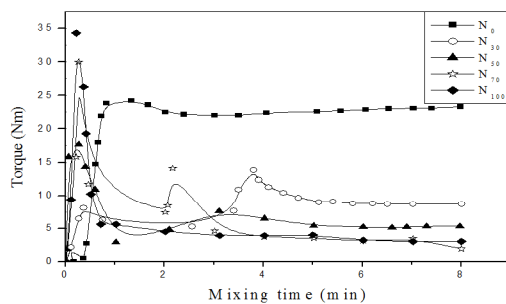


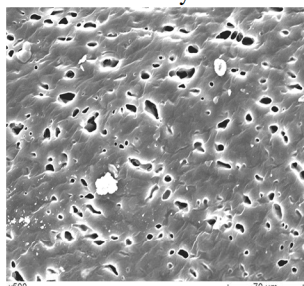
Figure 1: Rheographs showing torque-time relations

The time–torque curves of all blends have two peaks. The first peak is due to the increase in the viscosity by the introduction of the cold PA(6,66) granules into the mixer. The viscosity then decreases, showing the complete melting. Upon the addition of EPDM into the PA(6,66), the viscosity again increases which corresponds to the second peak. Thereafter the viscosity comes down showing the complete melting of the second phase and finally the curves level off to give uniform torque value. The levelling off of the torque may be related to the attainment of a good level of mixing. It is found that 180°C, 60 rpm and the mixing time of 8 minutes are the ideal conditions for mixing.

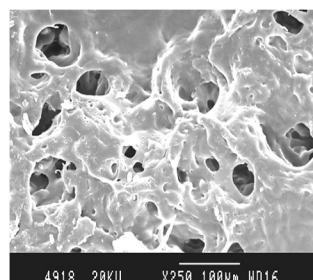
#### A. Phase Morphology of Blends

The main mechanism governing the morphology development in the blends is believed to be the result of both droplet breakup and coalescence. The scanning electron micrographs of PA(6,66)/EPDM blends ( $N_{20}$ ,  $N_{30}$ ,  $N_{40}$ ,  $N_{50}$ ,  $N_{60}$ ,  $N_{70}$  and  $N_{80}$ ) are shown in the Fig. 2(a) to (g). All the micrographs show a two-phase morphology due to the immiscible nature of the blends as a result of strong unfavourable interfacial interaction.

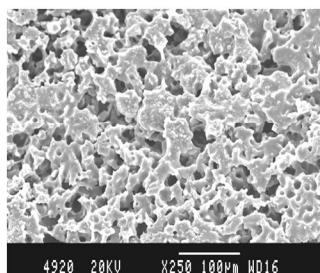
A careful evaluation of the micrographs suggests that, up to 30 wt % PA(6,66) concentrations, the PA(6,66) phase is preferentially dispersed in the high viscosity EPDM matrix with a notable difference in the size and its size distribution (Fig. 2 (a & b)). Spherical, elliptical and elongated elliptical domains of nylon can be observed in these blends.



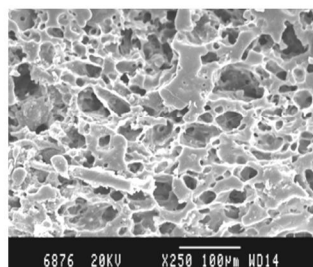
(a) Nylon20/80EPDM



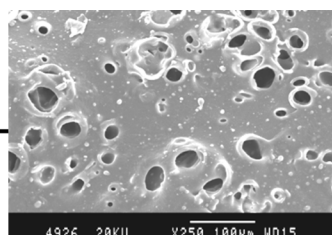
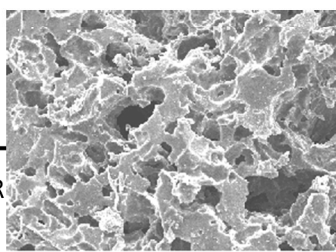
(b) Nylon30/70EPDM



(c) Nylon40/60EPDM



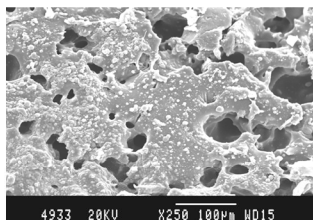
(d) Nylon50/50EPDM





(e) Nylon60/40EPDM

(f) Nylon70/30EPDM



(g) Nylon80/20EPDM

Figure 2: Scanning electron micrograph of nylon copolymer/EPDM blends at a magnification of 250 times (a) N<sub>20</sub>, (b) N<sub>30</sub>, (c) N<sub>40</sub>, (d) N<sub>50</sub> (e) N<sub>60</sub>

As the concentration of the nylon increases there is an onset of co-continuous morphology at N<sub>40</sub>. The blend systems N<sub>40</sub>, N<sub>50</sub>, and N<sub>60</sub> have got a co-continuous phase structure and all other blend systems have got typical matrix/droplet morphology. The nylon phase and EPDM phase are completely continuous at the co-continuous region. One can see a channel-like co-continuous phases of both components running through one another in Fig.(2d).

In the PA(6,66) rich blends (N<sub>70</sub> and N<sub>80</sub>) EPDM phase has been extracted (Fig. 2 (f and g)), a phase inversion occurs where nylon forms the continuous phase and EPDM exists as dispersed domains. The blend N<sub>70</sub> has got a clear and sharp interfacial boundary, which may be attributed to high interfacial tension indicating poor adhesion at the phase boundaries and is a manifestation of the incompatibility of the polymer components in these blends. It is well known that blends based on immiscible polymer components are characterized by high interfacial tension, which makes the dispersion during the blending

Table 2 presents the morphological parameters from SEM analysis of cryogenically fractured etched surfaces of the blends.

Table 2: Morphological Parameters of PA(6,66) /EPDM Blends from SEM analysis

Sample code	Composition of nylon/EPDM	$\overline{D}_n$ (μm)	$\overline{D}_w$ (μm)	$\overline{D}_w / \overline{D}_n$	$A_i$ (μm) <sup>2</sup> /(μm) <sup>3</sup>	IPD(μm)
N <sub>20</sub>	20/80	8.6	13.9	1.61	0.23	4.6
N <sub>30</sub>	30/70	13.8	20.3	1.47	0.14	3.4
N <sub>70</sub>	70/30	15.8	21.9	1.39	0.30	3.5
N <sub>80</sub>	80/20	14.5	19.1	1.32	0.28	2.8

It can be seen from the table that the dispersed phase domain size increased as the concentration of the dispersed phase increased. The extent of increase in the particle size ( $\overline{D}_n$ ) suggests that the phenomenon of coalescence is more predominant at high concentrations of the dispersed EPDM phase. However, on the other hand, when PA(6,66) is the dispersed phase, the influence of increasing nylon concentration on the coalescence is less predominant than the situation where EPDM is dispersed phase. This is associated with the high viscosity of the rubber phase (matrix) which resists the agglomeration of the nylon domains. In fact when the matrix phase is more viscous, the higher shear forces and, hence, the decreasing collision times along with a more difficult matrix interlayer film drainage between the colliding droplets reduce the coalescence probability.

This is, in general, related to the droplet agglomeration during melt mixing, which is well known to be a random process. As the result of mixing, drops of dispersed phase may tend to collide and coalesce eventually. The distribution of dispersed particles in continuous matrix can be evaluated from the polydispersity,  $\overline{D_w} / \overline{D_n}$ . It is obvious from the table that N<sub>30</sub> has got lower value of interfacial area per unit volume than N<sub>20</sub>. Interfacial area is a measure of interfacial thickness, which in turn is a measure of compatibility of blends. N<sub>30</sub> blend is highly incompatible. So it has got a very narrow interface compared to other blends, which may fail to transfer stress between the phases. The low values of the interparticle distance (IPD) indicate that the blends are not very brittle.

### B. Mechanical Properties

Tensile stress–strain behaviour of the simple blends at a crosshead speed of 50mm/min is shown in Fig. 4. The difference in the deformation characteristics of the blends under an applied load is evident from the stress-strain curves. Addition of non-crystalline elastomer in small amounts to semicrystalline PA(6,66) changes the nature of the curve considerably. At the crosshead speed of 50mm/min, neat nylon has got a well defined stress-strain curve typical that of a flexible plastic. Blends of varying component ratio show different failure characteristics. Stress-strain curves of PA(6,66) and PA(6,66) rich blends (>50%) show a linear elastic region followed by yielding in the inelastic region. The curve up to the yield point shows clear elastic deformation, thereafter the plastic deformation predominates. In the case of neat PA(6,66), the sharp increase in stress with strain beyond the yield point is associated with the orientation of the crystalline hard segments of the PA(6,66). As the rubber content increases, the initial modulus as well as the yielding tendency decrease. The phase change morphology can be understood from the stress-strain curves. In the case of N<sub>30</sub> the stress initially increases slightly and then decreases till the failure occurs.

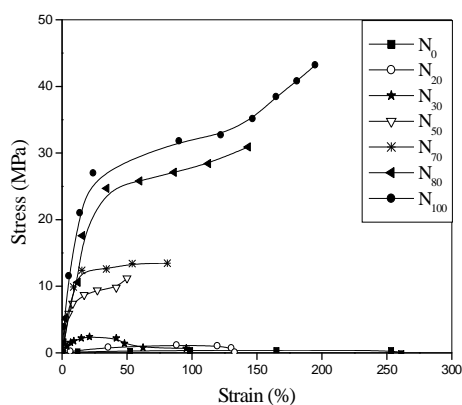


Figure 4: Tensile stress-strain curves of nylon/EPDM blends

The blend N<sub>50</sub> which is having a co-continuous morphology exhibits a stress-strain behaviour, which is intermediate to those of the other blend compositions. It is also observed that upon the addition of EPDM the strain increases and the stress decreases. Various tensile properties such as tensile strength ( $\sigma_m$ ), elongation at break ( $E_b$ ) and Young’s modulus (E) were determined from the stress-strain curves. The variation of tensile strength with wt% of PA(6,66) is shown in the Fig. 5.

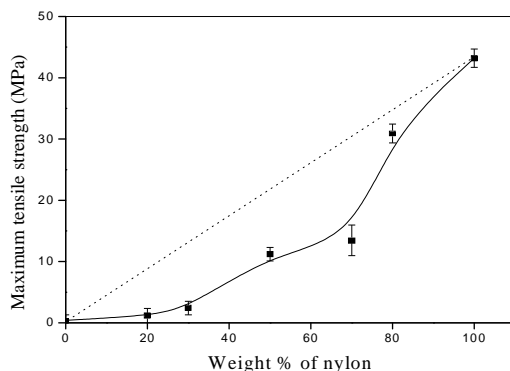


Figure 5: Variation of tensile strength with weight % of nylon (PA(6,66))

The tensile strength of the PA(6,66) /EPDM blends depends on the strength of the PA(6,66) matrix which in turn depends on the crystallinity of the PA(6,66) phase. As evident from the Fig. 5, PA(6,66) is a semi crystalline material having very good tensile strength, while EPDM is an amorphous material having very poor tensile strength. The curve shows a negative deviation. The blends show much lower tensile strength than projected from the additivity line. The negative deviation is due to the poor interfacial adhesion between the crystalline polar PA(6,66) and amorphous non-polar EPDM rubber, which prevents the stress transfer between the matrix and the dispersed phase. The failure stress depends on the interfacial interaction between the two polymer phases. The lowering of the tensile strength in the PA(6,66) /EPDM blends may be attributed to the presence of rubbery EPDM particles acting as stress concentrators. It is clear from the Fig. 5 that tensile strength increases as the PA(6,66) content increases. A sudden increase in the tensile strength is seen in blends where the PA(6,66) concentration is greater than 50%. This sharp increase in the tensile strength is associated with the phase inversion of PA(6,66) from dispersed to continuous phase. A clear change in the slope of the tensile strength-composition curve is seen between the composition ranges  $N_{30}$ - $N_{50}$ .

The Young's modulus of PA(6,66) /EPDM blends as a function of blend ratio is given in the Fig. 6. Young's modulus values followed a trend opposite to the strain at break. Modulus is a measure of the strength of the material at low strains. So PA(6,66) rich blends give comparatively good Young's modulus values. Pure PA(6,66) has got a Young's modulus of  $\approx 205$  MPa. Addition of EPDM decreases the Young's modulus. The curve has got a negative deviation. This is due to the high interfacial tension between the two phases and the low modulus value of EPDM phase. From 60wt% of PA(6,66) onwards the modulus increases remarkably due to the presence of high modulus of PA(6,66) as continuous phase. The yield stress also got the same trend as that of young's modulus.

The decrease in the tensile modulus in the blends may be due to the softening effect of the EPDM copolymer, since the tensile modulus of EPDM is considerably lower than that of pure PA(6,66). The introduction of EPDM, a low modulus material, in to the PA(6,66) matrix causes an overall lowering in the tensile modulus of the blends, and this in fact is contributed by the low interfacial adhesion between the two mixtures.

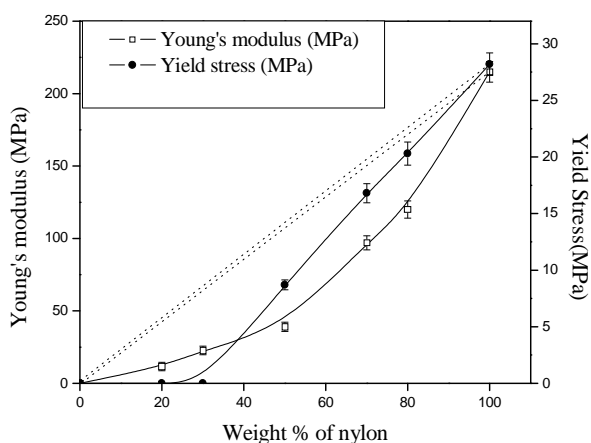


Figure 6: Effect of blend composition on the Young's modulus and yield stress of PA(6,66) /EPDM blends

As seen from the Fig. 7 the elongation at break also shows a negative deviation. EPDM has got higher elongation at break value than PA(6,66). The value decreases as the PA(6,66) content increases and is found to have more or less same values for  $N_{40}$ ,  $N_{50}$  and  $N_{60}$  composition. Thereafter the elongation at break is found to be increased. The blends have intermediate values which are much lower than projected from additive level. The low value of the elongation at break for the blends are due to the incompatibility and the poor adhesion between the phases.

The tension set after failure also increases as the PA(6,66) (nylon) content increases (Fig. 3.8). The considerable increase in the tension set values for the blends of high PA(6,66) content greater than 40% is attributed to the poor elastic recovery of the PA(6,66) phase after deformation.

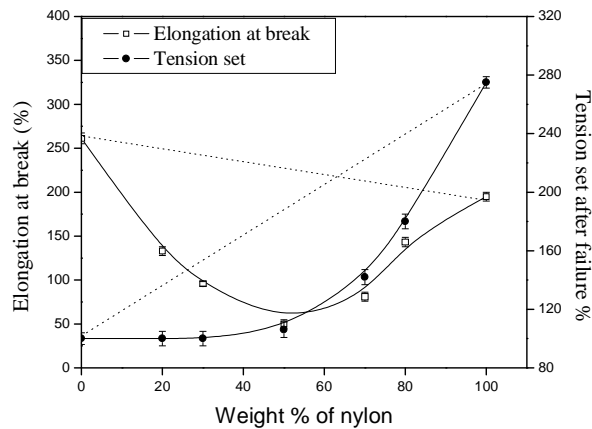


Figure 7: Effect of blend ratio on elongation at break and tension set of PA(6,66) (nylon) and EPDM blends

The variation of tear strength with weight % of PA(6,66) (nylon) is shown in the Fig. 8. The tear strength values of the blends also exhibit same trend as tensile strength. Tear strength decreases as the rubber content increases. This is due to the decreases in the crystallinity caused by the incorporation of the rubber phase. Nylon copolymer (PA(6,66)) is a semi crystalline plastic with much better strength and EPDM is an amorphous elastomeric material with poor strength. From the Fig 8, it is clear that blends with higher wt % of PA(6,66) have got higher tear strength. In these blends PA(6,66) behaves as a continuous phase.

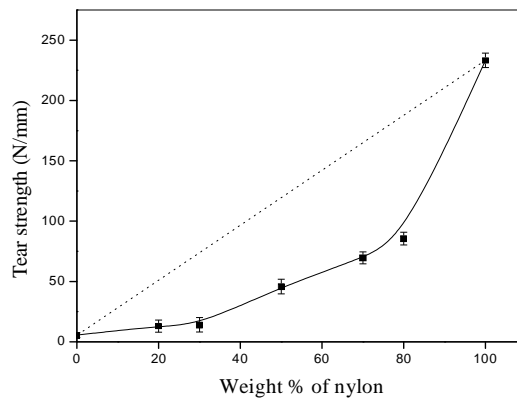


Figure 8: Effect of blend composition on the tear strength of PA(6,66) /EPDM blends

One of the important advantages of TPEs is that, they exhibit wide range of hardness. In Fig. 9 the Shore A hardness as a function of blend composition is given. The hardness values ranges from 31 to 99 Shore A.

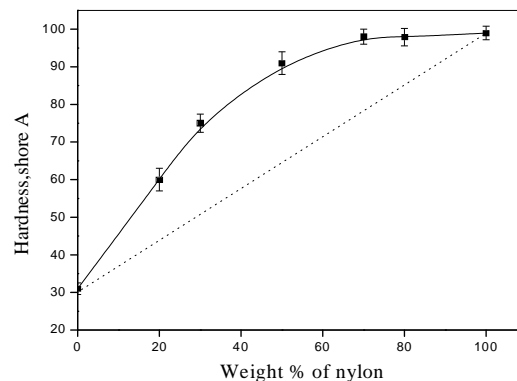


Figure 9: Variation of Shore A hardness with wt% of nylon (PA(6,66))

The neat nylon (PA(6,66)) shows the highest value of Shore A hardness, while EPDM shows the lowest. The curve shows a slope change beyond 50wt% of EPDM. The reduction in the hardness and the slope change in the curve at higher concentration of EPDM can be explained by the phase inversion of EPDM from dispersed to continuous phase. The useful working range [21] of the Shore hardness measurements is in between 10 and 90 for Shore A. Therefore reliable results were obtained for blends. The hardness values show a positive deviation. The values lie above the additive line because it is a surface property and is much less related to the interfacial bonding.

It is interesting to note that as the wt % of the minor phase increased the properties decreased. This is in good agreement with the morphological parameters, which showed that as the weight % of the minor phase increased, the morphological stability decreased. In short, all the properties except hardness show a negative deviation from additivity line. The inferior mechanical properties of the uncompatibilised nylon/EPDM blends are due to the lack of interfacial interactions between the phases.

#### IV. CONCLUSIONS

Morphology of PA(6,66) /EPDM blend system indicated a two-phase structure in which low viscosity nylon phase was dispersed as domain in the continuous high viscosity EPDM matrix up to 40 wt% of PA(6,66) concentration. A co-continuous morphology was obtained for 40/60, 50/50 and 60/40 PA(6,66)/EPDM compositions. At high PA(6,66) concentrations (70wt%), the EPDM phase was dispersed as domains in the continuous nylon matrix. All the results confirmed that the blends of PA(6,66) with EPDM show poor mechanical properties because of their immiscibility and owing to their poor interfacial adhesion due to the coarse morphology and lack of favourable interactions at the interface between PA(6,66) and EPDM. We observed a definite correlation between the phase morphology and mechanical properties. The mechanical properties of the blends were found to be strongly influenced by the blend ratio and morphology. Mechanical properties such as tensile strength, Young's modulus, tear strength, and hardness increased with the increase in PA(6,66) content. The increase was sharper when the PA(6,66) content was more than 60% where it formed a continuous phase. It is found that when the elastomer (EPDM) content increased, Young's modulus decreased and elongation at break increased. All the mechanical properties except hardness were found to have a negative deviation due to the high level of incompatibility.

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