Effect of Urethane Based Bonding Agent on the Cure Characteristics and Mechanical Properties of Styrene Butadiene Rubber – Whole Tyre Reclaim - Short Nylon Fiber Composite

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ABSTRACT

The cure characteristics and mechanical properties of short nylon fiber – styrene butadiene rubber/whole tyre reclaim (SBR/WTR) composites with and without an interfacial bonding agent based on 4,4 diphenyl methane diisocyanate and polyethylene glycol (MDI/PEG) have been studied. An 80:40 blend of SBR/WTR reinforced with 20 phr of short nylon fiber has been selected and the MDI/PEG ratio has been changed from 0.67:1 to 2:1. The minimum and maximum torques increased with isocyanate concentration. The scorch time and cure time showed an initial reduction. The cure rate showed an initial improvement. Tensile strength, tear strength and abrasion resistance increased with MDI/PEG ratio, these values were higher in longitudinal direction. Resilience and compression set increased with isocyanate concentration.

INTRODUCTION

Fiber–matrix interfacial bond is known to play an important role in composites since its interface has a decisive impact on the service property of the composite. Different techniques have been employed to achieve a strong interface or a good adhesion between fiber and matrix. Many researchers have evaluated the effect of conventional HRH bonding agent on short fiber elastomer composites and found an improvement in the properties of the composites[1-10]. Derringer evaluated the effect of HRH system with various fibers in nitrile rubber and NR and

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found that HRH system was not much effective with polyester fibers in many elastomeric matrices[11]. The effect of interfacial adhesion between nylon and vinylon short fiber reinforcements treated by different methods used in NBR and SBR matrices was studied by Zhou *et al*.[12]. Pukanszky studied the influence of the interfacial bonding on the ultimate tensile properties of the composite[13]. Kondo reviewed the selection of adhesives for bonding short fiber reinforcements in SBR and NR composites[14]. Bonding of polyester and aramid cord in EPDM and HNBR was developed by Janssen *et al*[15]. The adhesion of polyester fiber to different matrices was reported by Hirosuke *et al*[16]. Influence of adhesive system on the properties of composites of polyester fiber and SBR filled magnesium silicate has been studied by Ibarra[17]. Role of interface in fiber reinforced polymers with natural fiber was reported by Mukherjea[18]. The effect of urethane resin on polyester polyurethane composite was studied by Suhara *et al*.[19]. We have reported the effect of urethane resin on NR/WTR short nylon fiber composite and NBR/WTR short nylon fiber composite[20,21]. In this paper we are reporting the effect of urethane based bonding agent on the cure characteristics and mechanical properties of SBR/WTR/short nylon fiber composite.

The composite containing 20 phr of short fiber was selected for this study based on our earlier work[21].

EXPERIMENTAL

Materials

Styrene butadiene rubber (synaprene 1502) was obtained from Synthetics and Chemicals Ltd., Bareilly. Nylon fiber obtained from SRF Ltd., Madras was chopped to approximately 6 mm. Whole Tyre Reclaim (WTR) was supplied by Kerala Rubber and Reclaims, Mamala, India. The characteristics of reclaim used are given in Table 1. MDI (4,4' diphenyl methane diisocyanate) was obtained from BASF South Korea and PEG (Polyethylene

Table 1 Characteristics of WTR

Parameter	Value
Acetone extract (%)	13
Carbon content (%)	19

glycol) with a molecular weight 4000 was obtained from Manali Petrochemicals, Chennai.

All other ingredients are of commercial grade.

Preparation of compounds and moulding

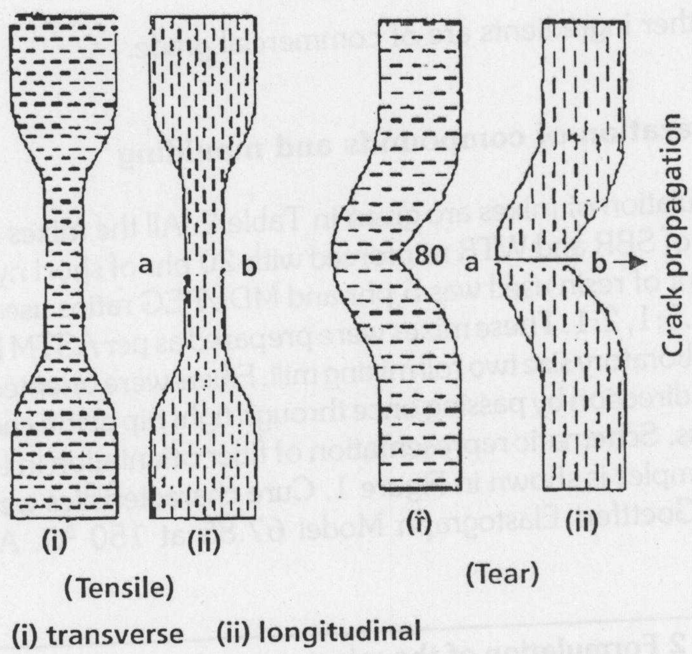
Formulation of mixes are given in Table 2. All the mixes contain 80/40 blend of SBR and WTR reinforced with 20 phr of short nylon fibers. The amount of resin used was 5 phr and MDI/PEG ratios used were 0.67:1, 1:1, 1.5:1, 2:1. These mixes were prepared as per ASTM D 3182 (1989) on a laboratory size two roll mixing mill. Fibers were oriented preferentially in one direction by passing once through tight nip at the end of the mixing process. Schematic representation of fiber orientation in tensile and tear test samples is shown in Figure 1. Cure characteristics were determined using Goettfert Elastograph Model 67.85 at 150 °C. All mixes were

Table 2 Formulation of the mixes

Ingredient (parts by weight)	Mix No.				
	A	B	C	D	E
SBR	80	80	80	80	80
WTR	40	40	40	40	40
Nylon short fiber	20	20	20	20	20
MDI	0	2	2.5	3	3.33
PEG	0	3	2.5	2	1.67
Zinc Oxide	4	4	4	4	4
Stearic Acid	2	2	2	2	2
HS	1	1	1	1	1
MBTS	0.5	0.5	0.5	0.5	0.5
TMTD	1.8	1.8	1.8	1.8	1.8
Sulfur	0.3	0.3	0.3	0.3	0.3

(MDI - (4,4` diphenyl methane diisocynate). PEG - (Polyethylene glycol) HS (1,2 - dihydro 2,2,4 trimethyl quinoline), MBTS (dibenzothiazyldisulfide), TMTD (tetramethylthiuramdisulfide))

Figure 1 Schematic representation of fiber orientation



vulcanized in an electrically heated hydraulic press at 150 ^{0}C and at a pressure of 180 kg/cm^2.

Testing of vulcanizates

The vulcanizates were tested for different mechanical properties according to the relevant ASTM standards. Tensile properties were measured using a Zwick UTM Model 1445 according to ASTM D 412 (die E). The test speed was 500 mm/min. Tear test was done on Zwick UTM as per ASTM D 624 (die C). The grip separation speed was 500 mm/min. Abrasion resistance of composites was measured using DIN abrader as per DIN 53516 and the values were expressed as volume loss per hour. Compression set at constant strain was measured according to ASTM D 395 – 86 method B. Resilience was measured according to ASTM D 2832 – 88. Properties such as tensile strength, tear strength and abrasion resistance were determined both in the longitudinal and transverse orientation of fibers. For ageing resistance, samples were aged in an air oven for 48 h at 70 ^{0}C (ASTM D 573 – 88). The properties were measured 24 h after the completion of ageing.

RESULTS AND DISCUSSION

Cure characteristics

The variation of minimum torque with MDI/PEG ratio is shown in Figure 2. The minimum torque shows an increase up to MDI/PEG ratio of 1.5:1. With further increase in isocyanate concentration the minimum torque value decreases. The initial improvement can be attributed to the low level of urethane formation in the processing stage. Due to the increase in stock viscosity as indicated by minimum torque values, the processability of the composite is reduced in presence of bonding agent. Similar results have been reported earlier[20,21].

The maximum torque values show a reduction at lower MDI/PEG ratio, with further increase in isocyanate concentration the torque values increase (Figure 3). The initial reduction arise from less effective utilisation of monomer functionalities of the bonding agent. With further increase in isocyanate concentration the matrix becomes more restrained and the torque increases. At higher isocyanate concentration there is more effective formation of urethane resin which makes the fiber matrix bond stronger thus resulting in a more restrained matrix.

Figure 2 Variation of minimum torque with MDI/PEG ratio

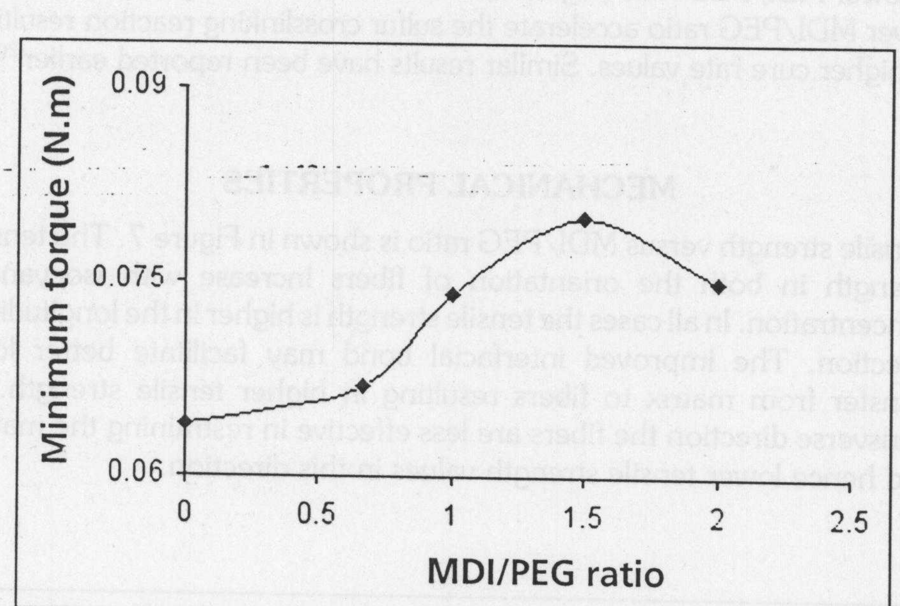
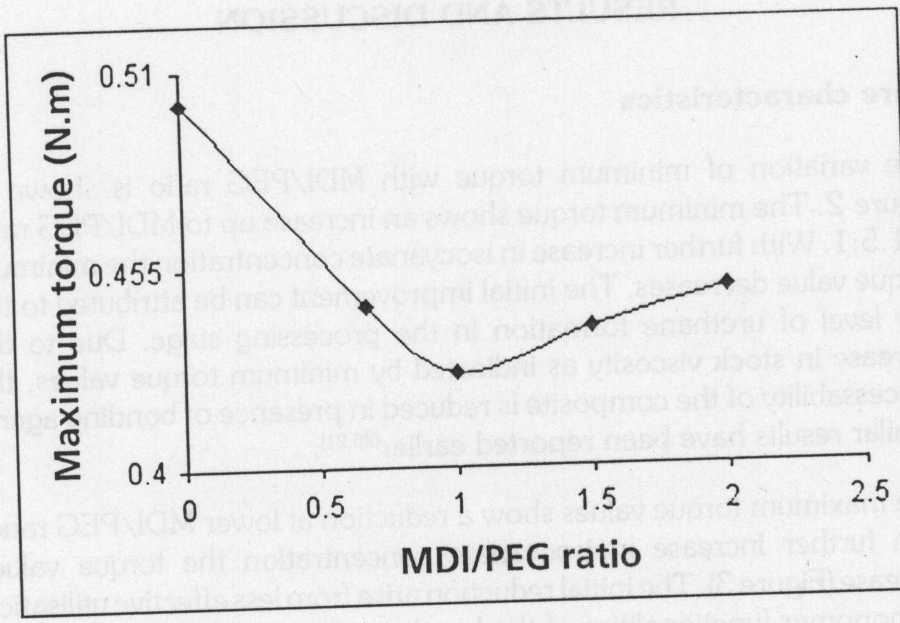


Figure 3 Variation of (maximum) with MDI/PEG ratio



The scorch time is plotted against MDI/PEG ratio is given Figure 4. The scorch time shows a reduction at lower MDI/PEG ratio and with further increase the scorch time increases. Similar trend is seen in the case of cure time also (Figure 5). There is a corresponding increase in cure rate values at lower MDI/PEG ratio (Figure 6). The excess free diol groups present at lower MDI/PEG ratio accelerate the sulfur crosslinking reaction resulting in higher cure rate values. Similar results have been reported earlier[20].

MECHANICAL PROPERTIES

Tensile strength versus MDI/PEG ratio is shown in Figure 7. The tensile strength in both the orientation of fibers increase with isocyanate concentration. In all cases the tensile strength is higher in the longitudinal direction. The improved interfacial bond may facilitate better load transfer from matrix to fibers resulting in higher tensile strength. In transverse direction the fibers are less effective in restraining the matrix and hence lower tensile strength values in this direction.

Figure 4 Variation of scorch time with MDI/PEG ratio

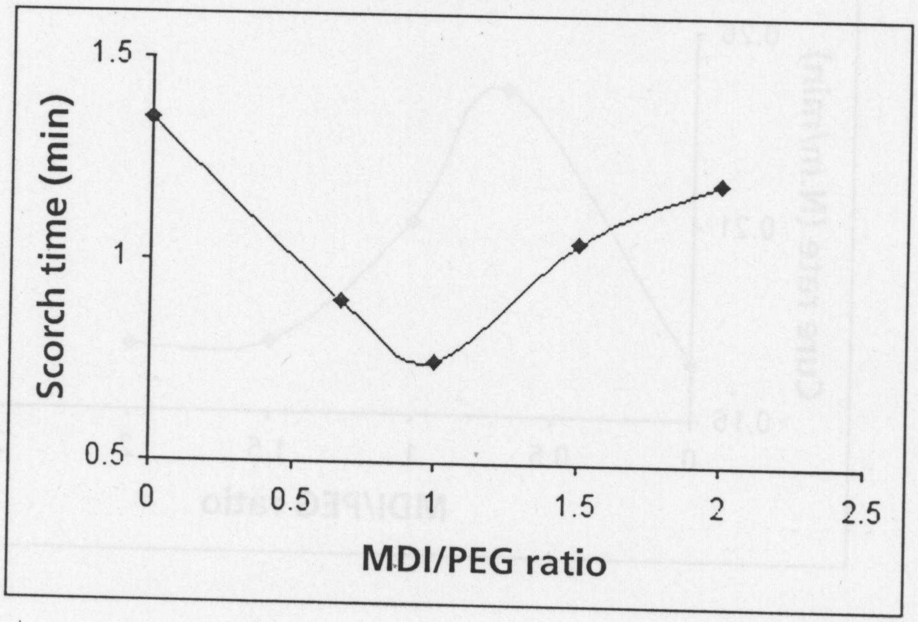


Figure 5 Variation of cure time with fiber MDI/PEG ratio

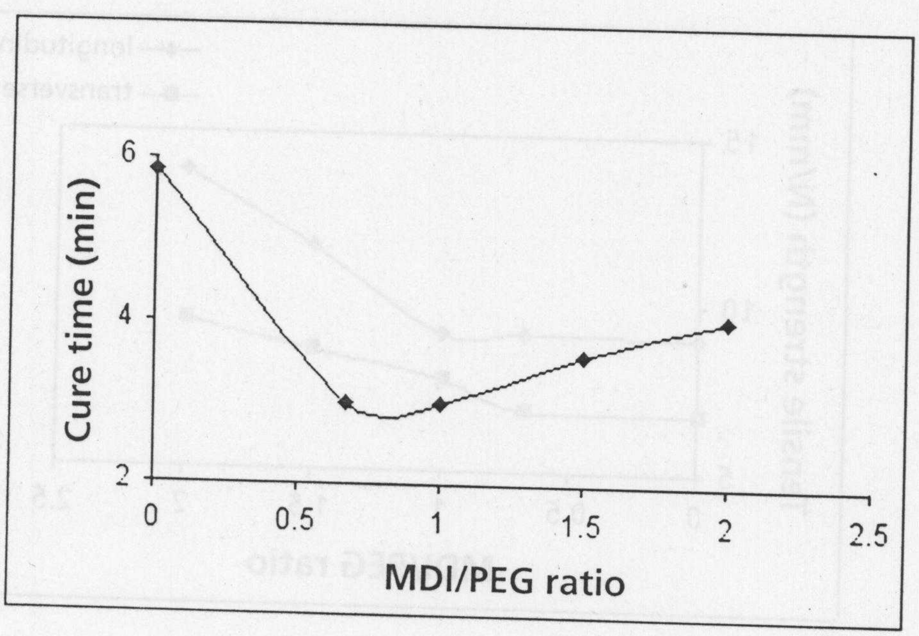


Figure 6 Variation of cure rate with MDI/PEG ratio

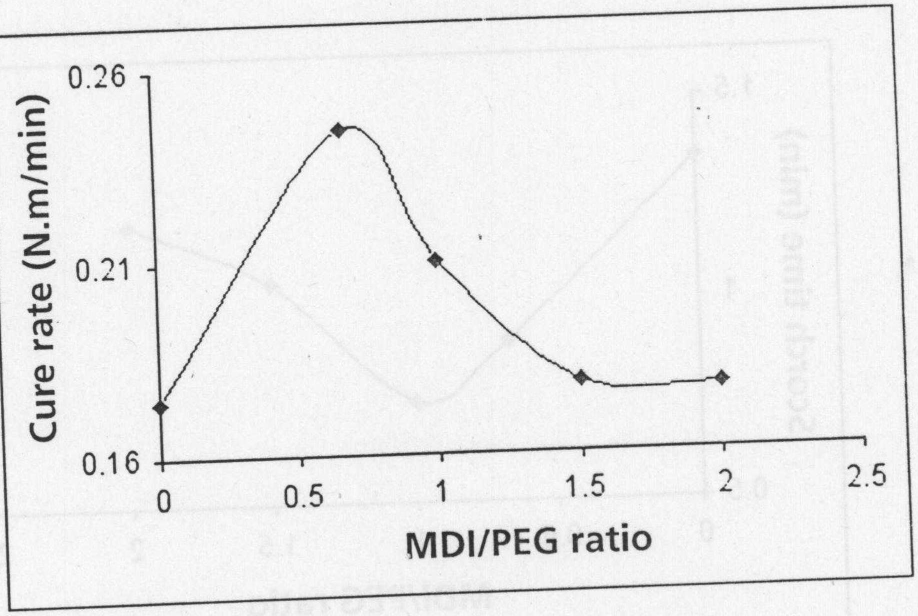
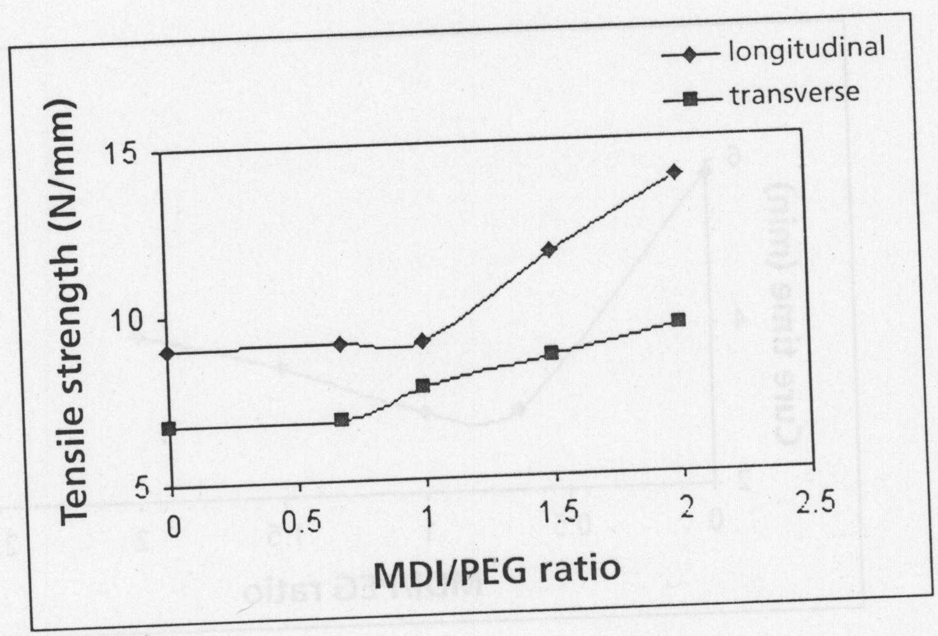


Figure 7 Variation of tensile strength with MDI/PEG ratio



With increase in MDI concentration the elongation at break values tend to decrease (Figure 8). The elongation at break values are higher in the transverse direction. The improved interfacial bond formed inhibit the molecular orientations resulting in lower ultimate elongation.

The tear strength shows an improvement with increase in the MDI/PEG ratio in both the orientation of fibers (Figure 9). Due to the improved interfacial bond the crack propagation is hindered more effectively and the tear strength increases.

Resilience shows an increase with isocyanate concentration (Figure 10). In presence of urethane resin the elasticity of the composite is improved.

The compression set increases with isocyanate concentration (Figure 11). The increased compression set indicates an increased viscous value of the matrix. This may be due to the elevated test temperature which renders the matrix more soft and increase the flow under stress.

Abrasion loss shows a reduction at lower isocyanate concentration in both the orientation of fibers (Figure 12). The improved interfacial bond makes the matrix more stiff resulting in lower abrasion loss.

Figure 8 Variation of elongation at break with MDI/PEG

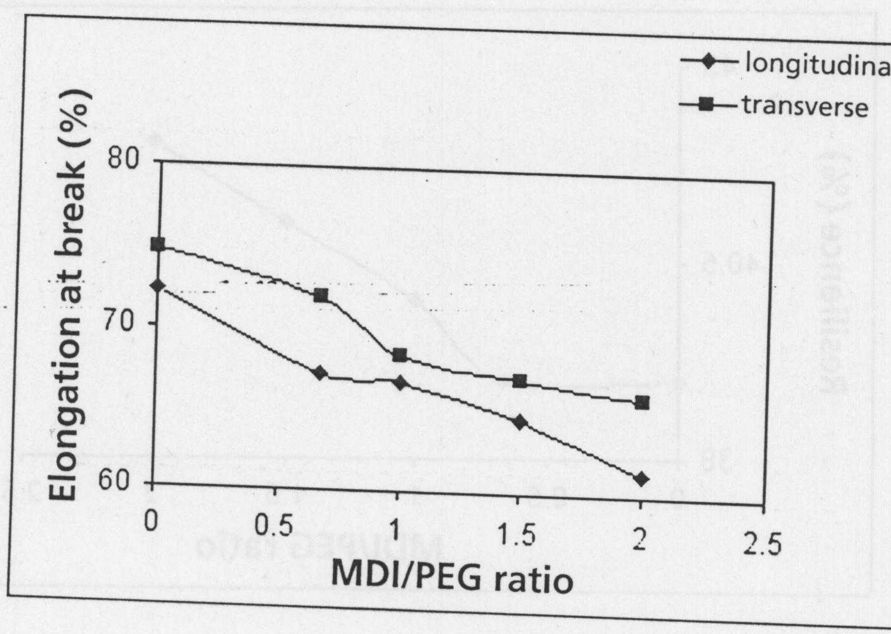


Figure 9 Variation of tear strength with MDI/PEG

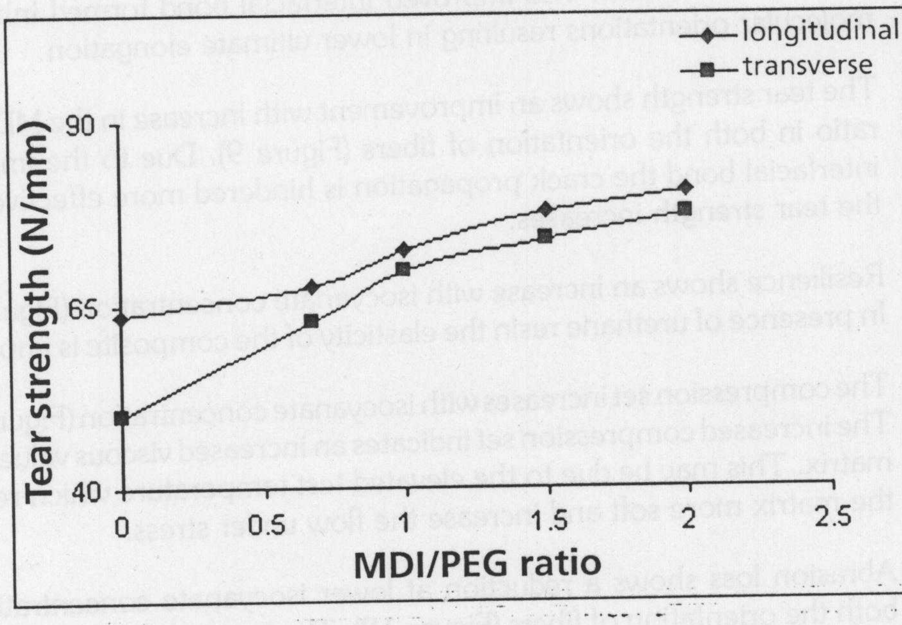


Figure 10 Variation of resilience with MDI/PEG

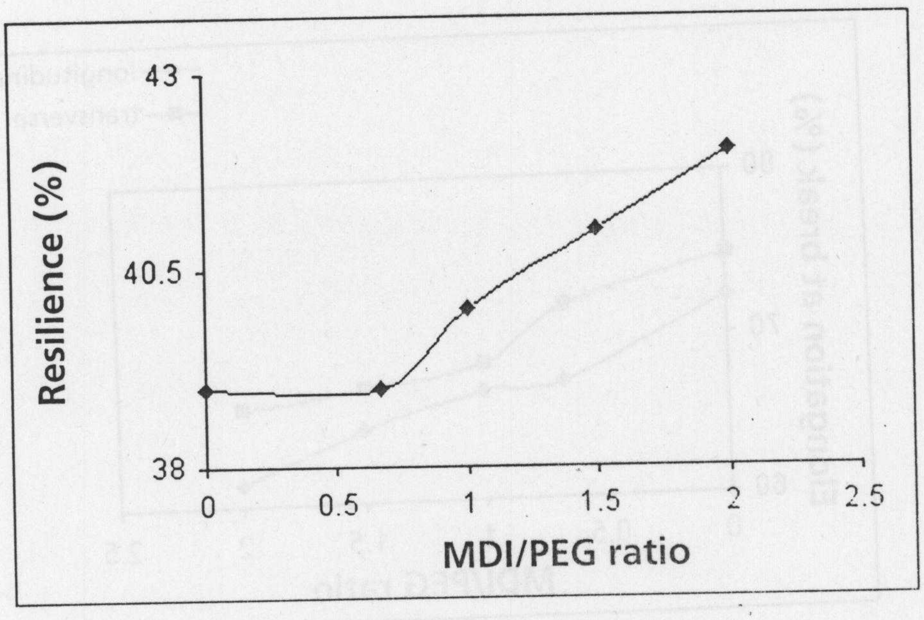


Figure 11 Variation of compression set with MDI/PEG

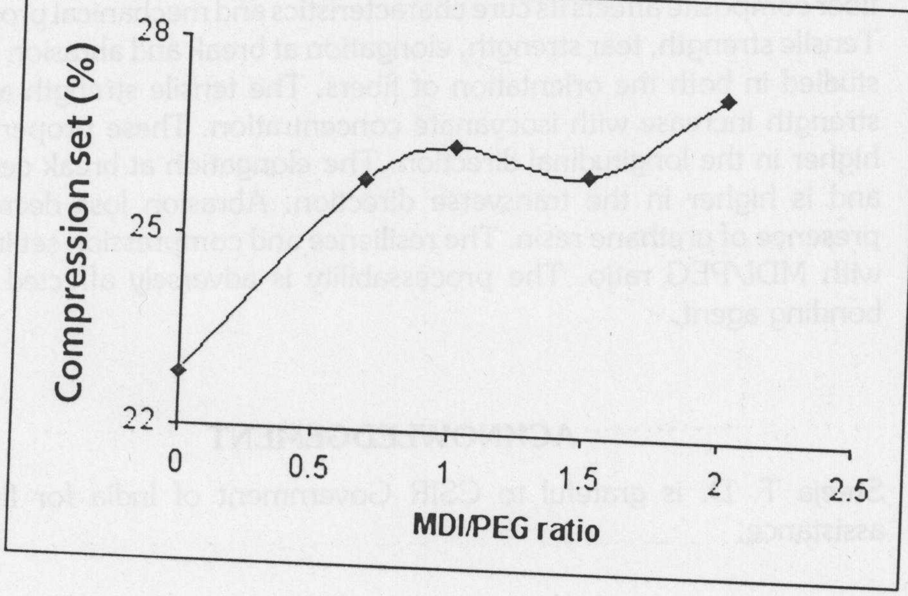
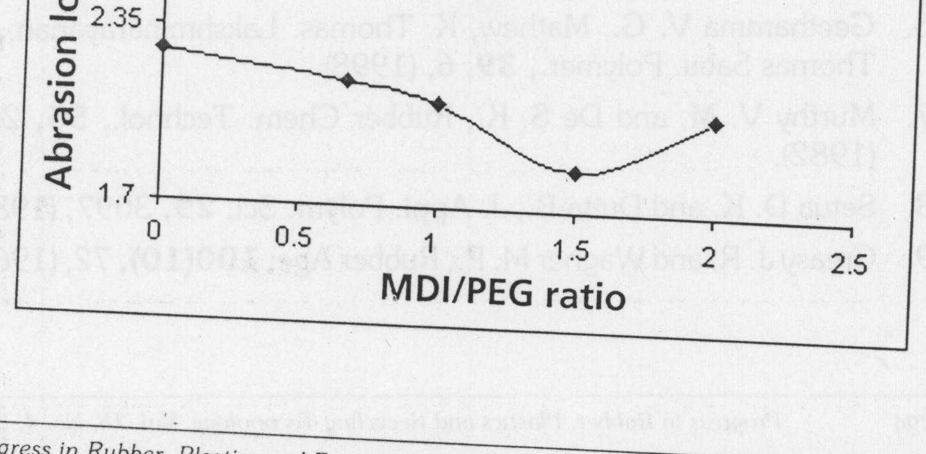


Figure 12 Variation of abrasion loss with MDI/PEG ratio



CONCLUSIONS

The conclusions from the above study are:

The addition of urethane resin (MDI/PEG) to SBR/WTR/short nylon fiber composite affects its cure characteristics and mechanical properties. Tensile strength, tear strength, elongation at break and abrasion loss are studied in both the orientation of fibers. The tensile strength and tear strength increase with isocyanate concentration. These properties are higher in the longitudinal direction. The elongation at break decreases and is higher in the transverse direction. Abrasion loss decrease in presence of urethane resin. The resilience and compression set increase with MDI/PEG ratio. The processability is adversely affected by the bonding agent.

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