

Mechanical Behavior of Polyblended Polystyrene and Ethylene-Propylene-Diene Rubber by Cationic Reactions in the Melt

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Synopsis

Attempts have been made to obtain high impact polystyrene by Lewis-acid-initiated degradation process in the melt reactions of PS and EPDM. It is found that impact strength improves whereas tensile strength decreases. This can be interpreted by the increased solubility of the coupled rubber molecules in the continuous phase.

INTRODUCTION

In order to obtain high impact polystyrene (PS), polymeric carbocations have been generated via controlled electrophilic degradation in the melt under shear and these carbocations are trapped by alkylative grafting onto a preformed second polymer in the system.¹ The effect of various kinds of Lewis acids on the cationic melt reactions of PS and EPDM rubber has been investigated.² Mori et al. have used $\text{NaCl} \cdot \text{AlCl}_3$ double salt to minimize the degradation and at the same time to increase the number of crosslinks for the melt reaction of PS and EPDM. They have also studied the tensile properties of the resulting grafts.³ Due to presence of only Lewis acid (AlCl_3), the reactivity will be enhanced and the degradation of polystyrene is also possible, which may enhance the impact resistance of resultant polyblends. During the Lewis-acid-initiated reaction of PS and EPDM grafting, degradation and crosslinking, etc., take place at the same time. The rubber phase undergoes degradation and crosslinking. Similarly polystyrene phase undergoes degradation and then branching and crosslinking. The coupling reaction also occurs at the same time. In case the degradation of polystyrene is suppressed as in the work of Mori et al.,³ the increase in the impact strength may be limited. In the present investigation, only AlCl_3 has been used and melt reaction has been performed in the mixing chamber of Brabender plasticorder. The tensile and impact properties of the resultant polyblends have also been studied in the present investigation.

EXPERIMENTAL

Polystyrene (RAPRA, U.K.), anhydrous AlCl_3 (Analar B.D.H., India), and EPDM rubber (Exxon) have been used as received. Reactions are carried out in the mixing chamber of the Brabender Plasticorder (Brabender OHG Duisburg) fitted with cam type blades rotating at 60 rpm and 180°C. The total charge of PS and EPDM is 50 g and Lewis acid is added after 10 min of

TABLE I
Characteristics of EPDM and Polystyrene

Name of substance	Wt % composition based on IR analysis			Mol wt M_w	Intrinsic viscosity (dL/g)
	Ethylene	Propylene	Diene		
PS Rapra, U.K.	—	—	—	1.59×10^5	0.66
EPDM Vistalon 5600, EXXON	42.7	47.9	9.4	1.42×10^5	1.585

mixing i.e. after complete fusion and homogenization of the mixture. The torque is recorded from the very beginning of the mixing operation. After addition of $AlCl_3$ the torque is continuously recorded for another 20 min and after that resultant polyblend is taken out from the chamber.

The blend is dissolved in benzene, precipitated in excess of methanol, and then dried under vacuum for 48 h. Five blends are prepared containing various proportions of EPDM rubber. The amount of $AlCl_3$ in all the five blends is kept approximately the same. The dried blends are properly mixed with 0.2% zinc stearate and 0.3% di-*tert*-butyl paracresol (Ionol) and extruded in Brabender plasticorder and extrusiograph at 220°C. The extruded material is then compression-molded at 200°C to make a flat sheet. Tensile and impact specimens are cut for mechanical testing from this sheet. Similarly PS/EPDM blend is also prepared. The tensile and impact testing has been performed

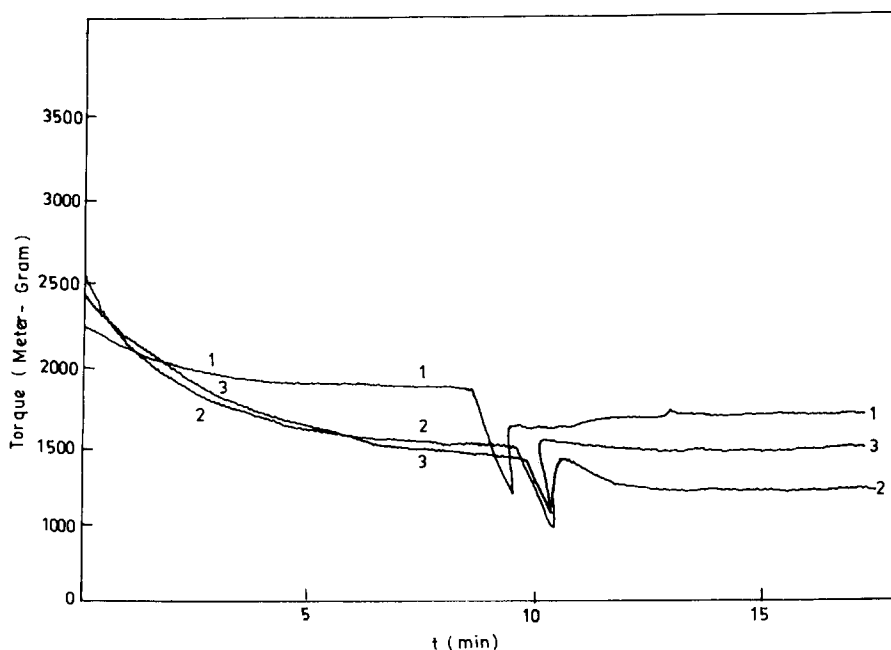


Fig. 1. Torque vs. time curves of the melt reaction of EPDM-PS mixture: (1) 0 wt %; (2) 5 wt %; (3) 10 wt % EPDM.

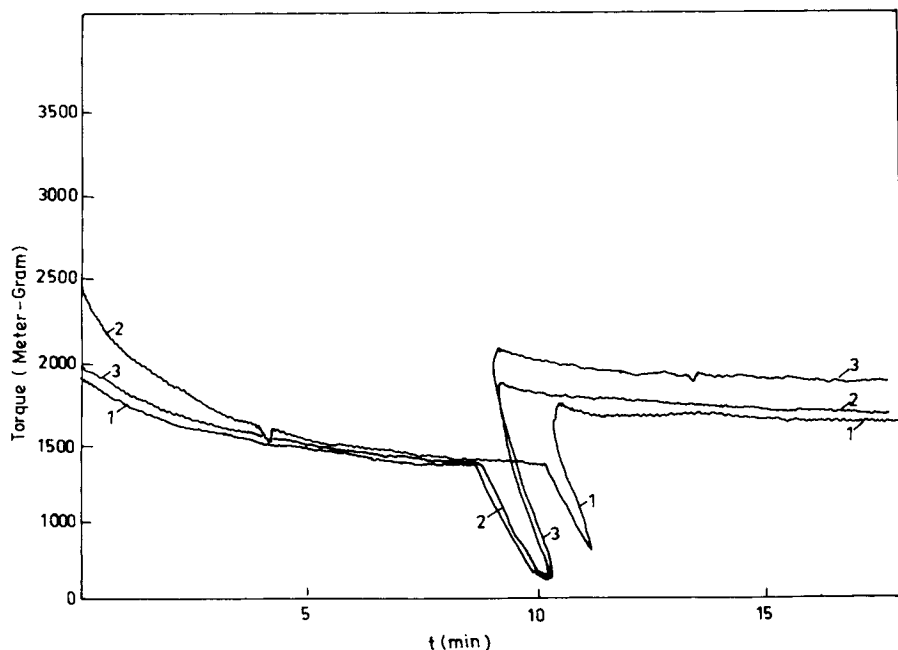


Fig. 2. Torque vs. time curves of the melt reaction of EPDM-PS mixture: (1) 15 wt %; (2) 20 wt %; (3) 25 wt % EPDM.

according to respective ASTM standards. SEM (Camscan, U.K.) was studied on the fractured surface of the blends.

RESULTS AND DISCUSSION

Figures 1 and 2 show the torque vs. time curves of the melt reaction of EPDM and PS mixtures. The mechanism of Lewis-acid-initiated reaction is given elsewhere.⁴ The sudden decrease of the torque after addition of AlCl_3 is due to the formation of low molecular weight polystyrene and the subsequent homogenization. The increment of torque above the initial value after homog-

TABLE II
Composition of Melt Reaction Product

Composition of EPDM by weight (%)	AlCl_3 (wt %)	MEK ^a soluble (wt %)	Petroleum ether ^b soluble (wt %)	Benzene ^c soluble (wt %)	Cross-linking of rubber (%)
5	1.03	99.0	—	—	—
10	1.03	92.5	3.55	—	39.2
15	1.03	91.5	4.62	1.24	30.8
20	1.03	82.8	8.72	2.24	32.4
25	1.03	76.0	11.96	4.425	30.2

^aPolystyrene and low molecular weight graft.

^bUnreacted EPDM rubber.

^cGraft copolymer after extraction with MEK and petroleum ether.

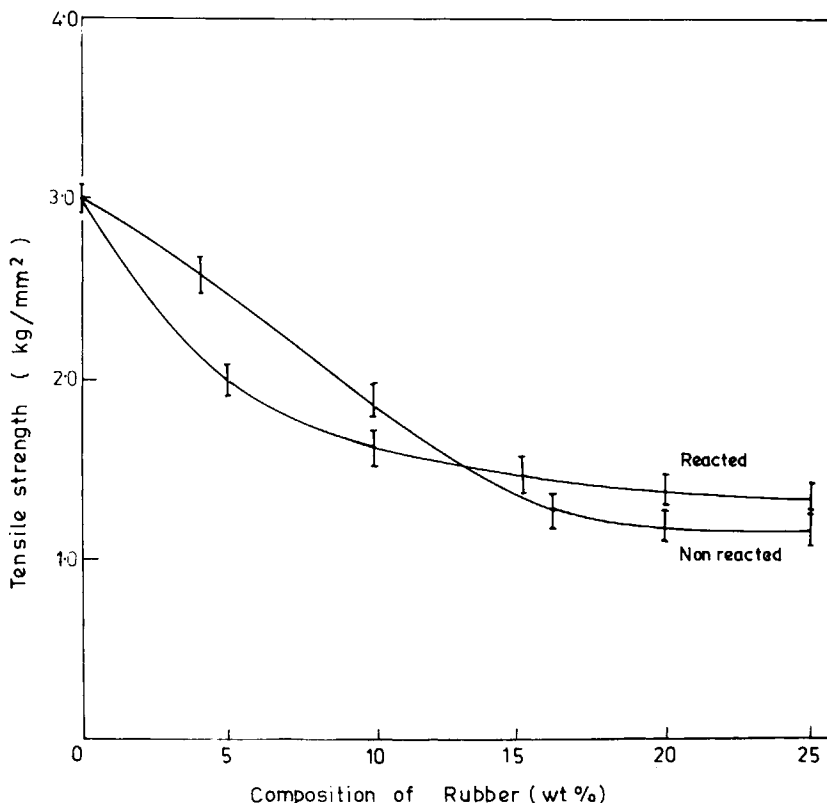


Fig. 3. Tensile strength of nonreacted and reacted mixtures as a function of rubber composition by weight percent.

enization indicates the effect of crosslinking in the rubber phase. Crosslinks in the rubbery phase of the graft are advantageous for impact modification; slightly crosslinked rubbers exhibit higher impact strength than uncured rubbers.⁵

From Table II, it can be seen that, with the increase of rubber content in the melt, the grafting reaction increases. This is due to the fact that, with increasing rubber content, the number of double bonds also increases, which results in a higher extent of grafting. But the crosslinking reaction is predominant. The higher solubility in MEK is due to the polystyrene and grafted rubber. It can also be seen that the amount of polystyrene content in the insoluble rubber is negligible.

Figure 3 shows the tensile strength (TS) variation with the rubber composition. The tensile strength decreases continuously up to 15 wt % of rubber and then becomes horizontal. This is due to the increasing amount of rubber content and poor adhesion between the rubber and polystyrene phases. The steeper decrease of TS in the case of the graft system is due to the presence of low molecular weight degradation products. After 15 wt % rubber, there is an improvement of TS in the graft system. The presence of crosslinks as well as the improved adhesion due to the presence of graft accounts for this improvement.

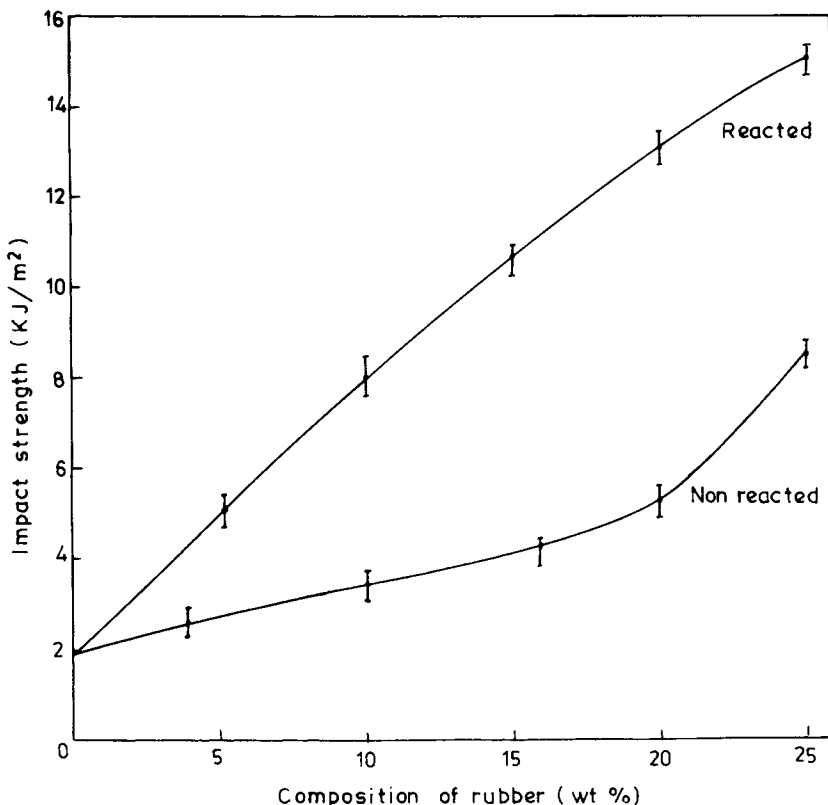


Fig. 4. Impact strength of nonreacted and reacted mixtures as a function of rubber composition by weight percent.

Figure 4 shows the impact behavior of the graft system with respect to the mechanical blend of PS-EPDM. The sufficient improvement of impact strength of graft system is largely due to the presence of crosslinks. The graft copolymer component serves to increase the compatibility⁶ between the two phases, thus increasing mutual adhesion since the grafted molecules tend to migrate to the interface and bond the two principal grafted phases together. The increase of impact strength can also be visualized by SEM microphotographs (Fig. 5). The grafted blend system contains smaller domains than the ungrafted blend system. The decrease of particle size increases the interfacial surface between rubber particles and polystyrene matrix. This can favor the formation of larger numbers of crazes (as craze starts at the said interface) increasing the impact strength. The irregular feature of the grafted system may be due to higher deformability of rubber particles during fracture propagation ahead of a traveling crack, which results in greater energy absorption and consequently higher impact strength.⁷ Hence, it may be concluded that melt reactions in the presence of Lewis acid in PS-EPDM blends increase the impact strength of the blend.

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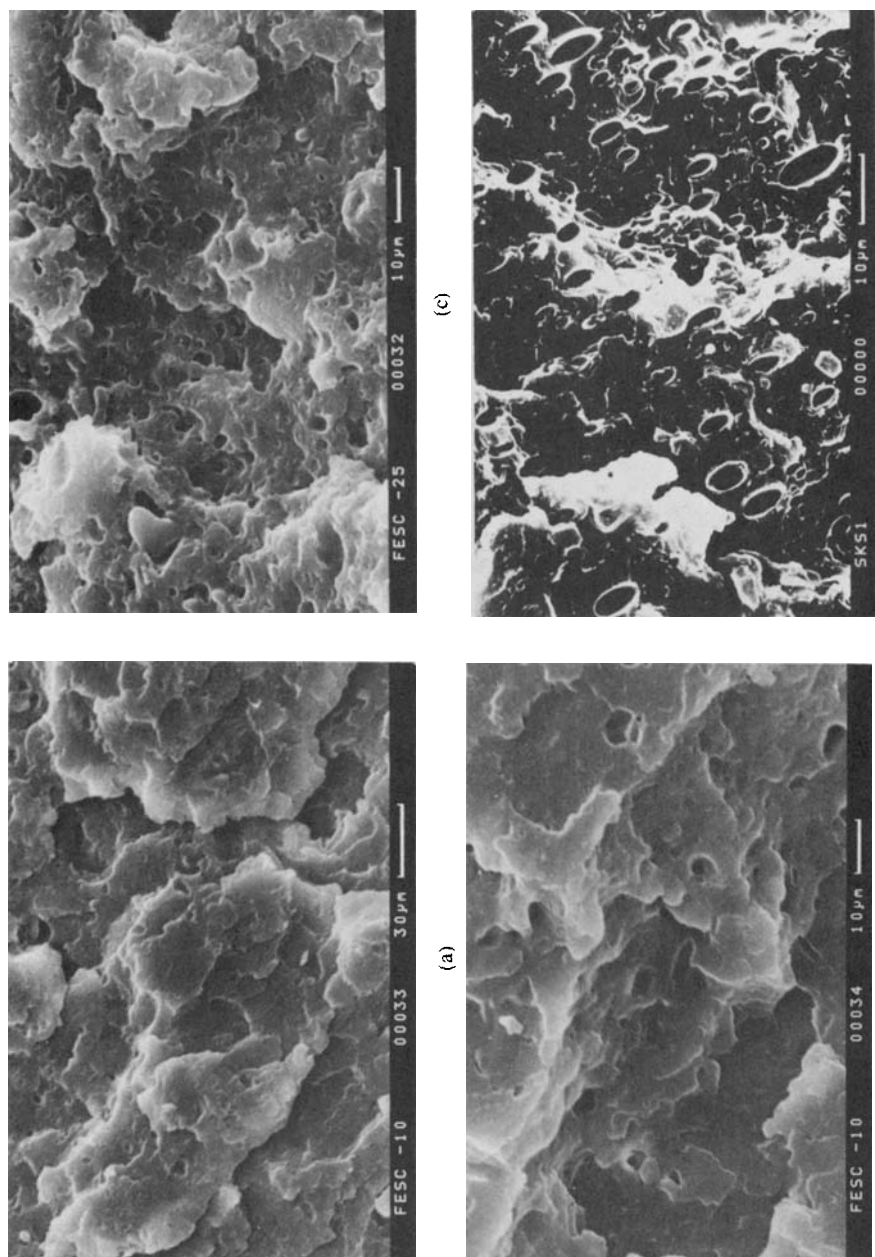


Fig. 5. Scanning electron micrographs: (a) and (b) 10 wt % rubber in PS/EPDM nonreacted blend; (c) 25 wt % rubber in PS/EPDM reacted blend; (d) 10 wt % rubber in PS/EPDM reacted blend.

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