Reactive Compatibilization of EPDM/NBR Blends via Glycidyl Methacrylate Grafting: Effect on Interfacial Adhesion and Mechanical Performance

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Abstract. This study focuses on improving the compatibility of EPDM/NBR (70/30) rubber blends through glycidyl methacrylate (GMA) grafting. Because EPDM is nonpolar and NBR is polar, their direct blending usually results in weak interfacial adhesion and poor mechanical performance. To solve this problem, GMA was grafted onto EPDM and NBR using peroxide-initiated melt and solution methods. FTIR analysis confirmed successful grafting by the appearance of new peaks at 1720 cm^{-1} (C=O ester group) and 910 cm^{-1} (epoxy group). The nitrile peak of NBR at 2235 cm^{-1} remained unchanged, showing that the main polymer structure was preserved. These results indicate chemical modification rather than simple physical mixing. Mechanical tests showed clear improvements after compatibilization. Tensile strength increased from 6 MPa to about 11 MPa, and elongation at break increased from 180% to over 300%. DMA results also showed higher storage modulus and lower tan δ peak intensity, indicating stronger interfacial bonding and reduced phase separation. Overall, GMA grafting is an effective and practical method to enhance interfacial adhesion and mechanical performance in EPDM/NBR blends, making these materials more suitable for engineering and sealing applications.

Keywords: EPDM, NBR, blends, materials, method

1. Introduction

Blending different elastomers is a pragmatic route to tailor material properties by combining complementary performance attributes within a single matrix. Ethylene–propylene–diene monomer (EPDM) is widely used for outdoor and heat-resistant applications because of its saturated backbone and outstanding weathering, ozone and thermal stability, while acrylonitrile–butadiene rubber (NBR) provides excellent oil and hydrocarbon resistance owing to its polar acrylonitrile segments. Direct physical blending of EPDM and NBR, however, typically yields macroscopically phase-separated morphologies with weak interfaces and poor stress transfer because of the strong

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disparity in polarity and solubility parameters between the two rubbers [1,2]. Such immiscibility manifests as reduced tensile strength, low tear resistance, and rapid crack propagation under cyclic load — attributes that limit the practical application of simple EPDM/NBR blends in demanding engineering contexts [3,4].

Reactive compatibilization — i.e., creating chemical functionality at one or both phase surfaces so that covalent bonds or strong secondary interactions form during mixing or curing — has emerged as the most effective strategy to reconcile these differences and to obtain fine, stable morphologies with enhanced mechanical integrity. Several reactive routes have been explored: (i) addition of pre-formed block or graft copolymers that localize at the interface and lower interfacial tension [5–7]; (ii) in-situ reactive extrusion/grafting where functional monomers are grafted onto a host polymer during melt processing (e.g., maleic anhydride, glycidyl methacrylate) [8–11]; (iii) use of multifunctional coupling agents or silanes that chemically link phases or fillers [12–14]; and (iv) peroxide-initiated co-crosslinking strategies that produce interpenetrating networks [15,16]. Each approach has trade-offs: pre-formed copolymers are effective but often costly and can suffer from poor dispersion if not properly designed [5]; silane coupling is powerful for filler–matrix adhesion but is less suited to direct polymer–polymer compatibilization in non-hydrolytic systems [12]; peroxide co-crosslinking can enhance cohesion but risks chain scission and loss of elasticity if process conditions are not carefully controlled [15].

Glycidyl methacrylate (GMA) grafting represents a particularly attractive in-situ compatibilization strategy for EPDM/NBR systems because the monomer provides two complementary functionalities in a single molecule: a polymerizable methacrylate double bond that can be grafted onto a hydrocarbon backbone via free-radical chemistry, and a glycidyl (epoxy) group that serves as a latent electrophile able to participate in ring-opening reactions or strong dipolar interactions with polar groups in the counterphase [9,17]. Prior studies have demonstrated the effectiveness of GMA grafting in polyolefin blends (e.g., PE/PA, PP/PA) and in rubber/filler systems, reporting improved dispersion, reduced phase domain sizes and enhanced mechanical properties compared with ungrafted controls [8,10,18]. In EPDM-based systems, peroxide-initiated GMA grafting onto EPDM has been shown to significantly increase adhesion to polar fillers and to polar elastomers—largely by lowering interfacial tension and by creating sites for covalent linkage or strong secondary bonding during subsequent blending or cure [19–21].

Comparative analyses in the literature show that GMA often outperforms other monomers for reactive compatibilization in mixed-polarity systems because the epoxide is relatively stable during grafting yet reactive enough during subsequent thermal processing to form interphase linkages; maleic anhydride (MAH), by contrast, can graft efficiently but may hydrolyze or form anhydride-derived crosslinks that alter cure chemistry and long-term stability [22–24]. Meanwhile, block copolymers engineered to contain polyolefin and polar blocks provide excellent interfacial coverage but require careful molecular weight and block ratio design to avoid micellization or poor processing [5,7]. Silane coupling agents are unparalleled for silica–rubber systems but are less direct when the target is two polymer phases rather than polymer–filler adhesion

[12]. Thus, the consensus emerging from multiple studies is that controlled GMA grafting offers a pragmatic balance of reactivity, processability and interphase chemistry for EPDM/NBR blends, provided homopolymerization and over-functionalization are avoided [9,11,21].

Mechanistically, the compatibilization afforded by GMA grafting has two sequential elements: (1) radical grafting of the methacrylate moiety onto a polymer backbone generates pendant glycidyl groups anchored to one phase; (2) during blending, heating, or curing, these glycidyl groups either undergo nucleophilic ring-opening reactions (with hydroxyls, amines, or carboxyls when present), react with functional coagents, or engage in strong dipole–dipole interactions (notably with nitrile groups in NBR), thereby forming an interphase region of increased polarity and, in favorable cases, covalent crosslinks between phases [9,17,25]. Quantitatively, prior work relates increases in grafting density to reductions in interfacial tension and domain size, but also points out a diminishing return and eventual embrittlement when graft density and homopolymer content rise beyond an optimal window [10].

Despite these advances, gaps remain in the literature. First, many studies focus on single-phase grafting (i.e., GMA onto the polyolefin phase alone) rather than systematic comparison of grafting on both phases (EPDM vs. NBR), which may yield different interphase chemistries and mechanical outcomes [21]. Second, kinetic and mechanistic understanding of the competitive pathways — grafting vs. GMA homopolymerization vs. backbone scission — under industrially relevant melt-extrusion conditions is incomplete; a predictive kinetic model would greatly aid scale-up and process design [11]. Third, the long-term stability of epoxide-mediated interfacial bonds under thermal, oxidative, and hydrocarbon exposure relevant to service conditions (e.g., sealing in oil environments) requires more systematic aging studies [20]. Addressing these knowledge gaps motivates the present work, which aims to (i) prepare EPDM-g-GMA and NBR-g-GMA under controlled conditions, (ii) compare their relative efficacy as compatibilizers in EPDM/NBR blends, and (iii) correlate spectroscopic evidence (FTIR), morphology (SEM), and mechanical performance (tensile, DMA) to grafting degree and processing variables.

2. Experimental Part

2.1 Materials

EPDM (ENB-type, ML_{1+4} = 60 at 125 °C) and NBR (34 wt% acrylonitrile) were used as base elastomers. Glycidyl methacrylate (GMA, ≥99%) was used as grafting monomer. Dicumyl peroxide (DCP) and benzoyl peroxide (BPO) were employed as radical initiators. Calcium carbonate ($CaCO_3$) was used as filler. Zinc oxide, calcium stearate, paraffinic oil, and Irganox 1010 were used as additives.

2.2 Preparation of GMA-Grafted Rubbers

EPDM-g-GMA (melt grafting): EPDM was grafted with GMA in an internal mixer at 160 °C and 60 rpm under nitrogen. After mastication, GMA (3 phr) and DCP (1 phr) were added. The product was extracted with acetone and vacuum-dried.
NBR-g-GMA (solution grafting): NBR was dissolved in MIBK at 70 °C. GMA (3 phr) and BPO (0.7 phr) were added, and the reaction was carried out for 3 h under nitrogen. The grafted polymer was precipitated in methanol and dried.

2.3 Blend Preparation

EPDM/NBR blends (70/30 phr) were prepared on a two-roll mill at 150 °C. Compatibilized blends contained 10 phr of EPDM-g-GMA or NBR-g-GMA. Compounds were peroxide-cured at 170 °C for 15 min.

Table 1. Formulation of EPDM/NBR Blends

Component	**Uncompatibilized (phr)**	**EPDM-g-GMA Blend (phr)**	**NBR-g-GMA Blend (phr)**
EPDM	70	60	70
NBR	30	30	20
EPDM-g-GMA	–	10	–
NBR-g-GMA	–	–	10
$CaCO_3$	50	50	50
ZnO	2	2	2
Ca-stearate	1	1	1
DCP	1.5	1.5	1.5
Paraffinic oil	5	5	5

2.4 Characterization Methods

FTIR spectroscopy (4000–600 cm^{-1}) was used to confirm grafting. Tensile properties were measured according to ASTM D412. Dynamic mechanical analysis (DMA) was performed from −80 to +100 °C at 1 Hz. Morphology was examined by SEM on tensile-fractured surfaces after gold sputtering [22-26].

3. Results and Discussion

3.1. FTIR Analysis

GMA-grafted EPDM and NBR showed new absorption bands at ~1720 cm^{-1} (ester C=O) and ~910 cm^{-1} (epoxide), confirming successful grafting.

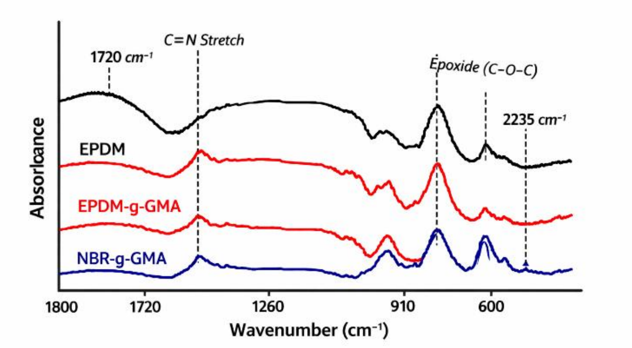

Figure 1. FTIR spectra of EPDM, EPDM-g-GMA, and NBR-g-GMA.

3.2. Mechanical Properties

Compatibilized blends showed significant improvement in tensile strength and elongation due to enhanced interfacial adhesion.

Table 2. Mechanical Properties of EPDM/NBR Blends

Sample	Tensile Strength (MPa)	Elongation at Break (%)	Modulus at 100% (MPa)
EPDM/NBR	6.0	180	2.5
+ EPDM-g-GMA	11.0	310	3.4
+ NBR-g-GMA	10.5	295	3.2

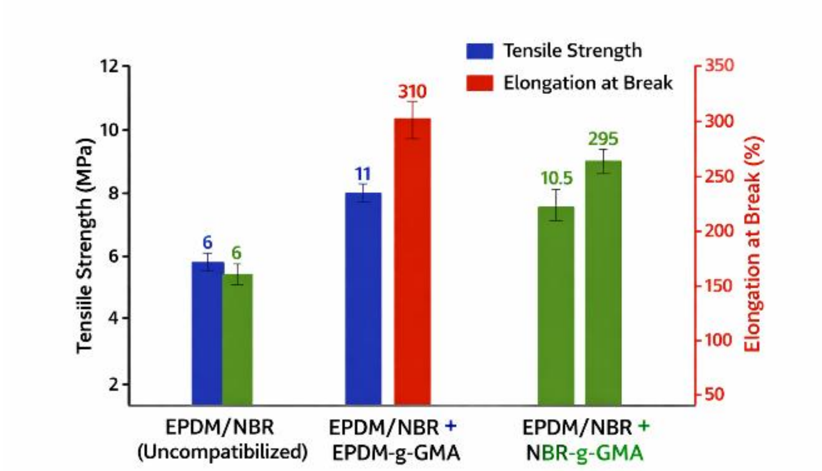

Figure 2. Tensile strength and elongation at break of EPDM/NBR blends.

3.3. Dynamic Mechanical Analysis

DMA results showed higher storage modulus and reduced tan δ peak intensity for compatibilized blends, indicating stronger interfacial bonding.

Table 3. DMA Results

Sample	Tg (°C)	E′ at 25 °C (MPa)	tan δ (peak)
EPDM/NBR	−34.5	6.5	0.156
+ EPDM-g-GMA	−38.0	8.2	0.102
+ NBR-g-GMA	−37.2	7.9	0.110

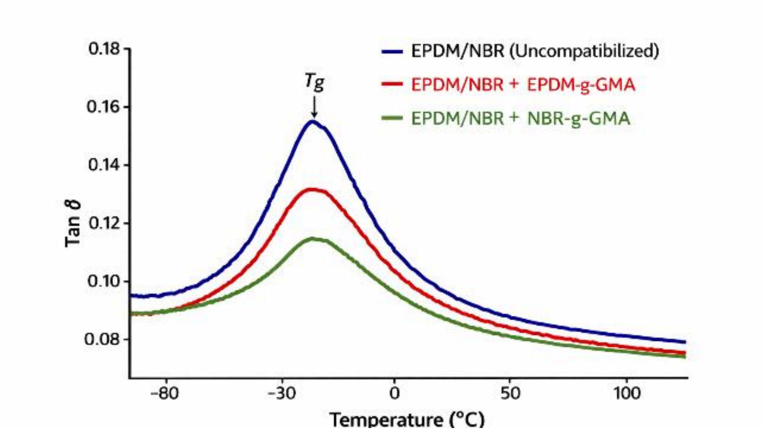

Figure 3. Tan δ versus temperature curves.

3.4. Morphological Analysis (SEM)

SEM images of the uncompatibilized blend revealed large, poorly bonded NBR domains within the EPDM matrix, indicating weak interfacial adhesion. In contrast, compatibilized blends exhibited finer and more uniform phase dispersion with blurred phase boundaries. The EPDM-g-GMA system showed the most homogeneous morphology, confirming efficient interphase bonding.

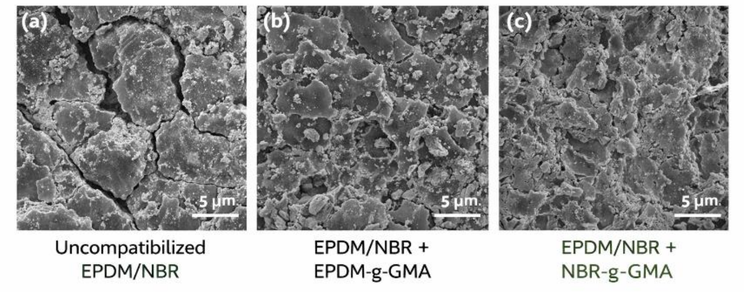

Figure 4. SEM micrographs of (a) uncompatibilized EPDM/NBR, (b) EPDM/NBR + EPDM-g-GMA, and (c) EPDM/NBR + NBR-g-GMA.

4. Conclusion

Reactive compatibilization of EPDM/NBR blends via GMA grafting significantly improved interfacial adhesion, morphology, and mechanical performance. FTIR confirmed successful grafting, while mechanical and DMA results demonstrated enhanced stress transfer and viscoelastic coupling. SEM observations supported the formation of finer morphologies in compatibilized systems. GMA grafting is therefore an effective strategy for improving polar–nonpolar rubber blends and can be extended to other elastomeric systems.

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