International Research Journal of Engineering and Technology (IRJET) e-ISSN: 2395-0056
Volume: 13 Issue: 07 | Jul 2026 www.irjet.net p-ISSN: 2395-0072

# Optimizing Carbon Black Dispersion through Sequential Mixing Strategies in Natural Rubber–Polybutadiene Rubber Compounds

**Mahaveer Joshi[1], Chandrakant Sinha[2], Bhupendra Singh Ken[3], Mukul Vijay[4], Kapil Nahar[5], Neetesh Kumar Dehariya[6]**

[123456] *Department of Chemical Engineering, Mewar University, NH - 48 Gangrar, Chittorgarh – 312901, Rajasthan, India*

---

**Abstract -** *Natural rubber (NR) and polybutadiene rubber (BR) blends are commonly used in tire and dynamic rubber products due to their mechanical and dynamic properties. The reinforcing efficiency of these blends largely depends on the uniform dispersion of carbon black, which is strongly influenced by the mixing process. In the present study, the effect of sequential carbon black addition on filler dispersion, rheological behavior, curing characteristics, and mechanical properties of NR/BR compounds was studied. Five formulations were prepared using a three-stage Banbury mixing process while maintaining an identical compound composition and varying only the carbon black split ratio and polymer addition sequence. In the proposed sequential mixing strategy, different proportions of carbon black were incorporated into the NR phase before the addition of BR to evaluate the effect of polymer–filler affinity on filler distribution. The results demonstrated that introducing a larger proportion of carbon black during the initial NR mixing stage significantly enhanced filler wetting and agglomerate breakdown, resulting in improved carbon black dispersion. Among the investigated formulations, the X-BR1 compound, containing 75% of the total carbon black in the first mixing stage, exhibited the highest dispersion value of 99.42%. Although rheological behavior showed slight improvements and mechanical properties remained largely comparable among all formulations, the optimized mixing sequence produced a more homogeneous filler network without altering the compound formulation or curing chemistry. The results demonstrate that sequential carbon black incorporation is a simple, cost-effective, and industrially applicable strategy for improving filler dispersion and process consistency in NR/BR rubber compounds, offering practical benefits for tire tread and other high-performance rubber applications.*

**Key Words: Natural rubber (NR), Polybutadiene rubber (BR), Carbon black dispersion, Sequential mixing, Polymer–filler interaction, Rubber compounding**

## 1. INTRODUCTION

Natural rubber (NR) and polybutadiene rubber (BR) are among the most widely used elastomers in the tire industry because of their characteristics. Natural rubber provides excellent tensile strength, tear resistance, and green strength because of its strain-induced crystallization behavior, whereas polybutadiene rubber has superior abrasion resistance, resilience, and low heat build-up due to its highly flexible molecular structure. The combination of these two elastomers has therefore become a preferred choice for manufacturing high-performance tire tread compounds, conveyor belts, and other dynamic rubber products where an optimum balance between mechanical strength, durability, and rolling resistance is required. However, achieving the desired performance from NR–BR blends [1] depends not only on the formulation but also on the efficiency of the mixing process, which controls the distribution of fillers and additives within the rubber matrix.

Carbon black remains the most important reinforcing filler used in rubber compounding because of its ability to significantly improve tensile strength, modulus, abrasion resistance, fatigue life, and processing characteristics [2]. The reinforcing efficiency of carbon black is strongly influenced by its state of dispersion within the elastomer matrix [3]–[5]. Poorly dispersed carbon black forms agglomerates that act as stress concentration sites, cause inferior mechanical performance, inconsistent curing behavior, increased heat generation, and reduced service life of the final product [6],[7]. Consequently, obtaining a uniform dispersion of carbon black has long been recognized as one of the most critical objectives [8] in rubber mixing. Numerous studies have demonstrated that factors such as rotor speed, fill factor, mixing temperature, mixing time, and filler loading influence the dispersion process [9]. Nevertheless, the sequence in which carbon black is incorporated into multi-polymer rubber blends has received comparatively less attention [10] despite its potential influence on filler distribution and polymer–filler interactions.

In NR–BR blend systems, the dispersion mechanism is considerably more complex than in single-polymer compounds because the two elastomers exhibit different viscosities, molecular structures, and affinities toward carbon black [11]. During mixing, carbon black particles tend [12] to migrate toward the polymer phase [13] that provides lower interfacial energy and better wetting characteristics [14],[15]. As a result, the sequence of filler addition can alter the initial wetting of carbon black [16], the

breakdown of filler agglomerates, and the final distribution of the reinforcing filler between the NR and BR phases [17]–[20]. This selective localization ultimately affecting compound viscosity, cure characteristics, and mechanical properties [21],[22]. Therefore, understanding the interaction between mixing sequence and polymer affinity is essential for designing rubber compounds with improved processing consistency and enhanced performance [23].

Although considerable research has been devoted to optimizing rotor speed, mixing temperature, fill factor, and carbon black loading [24], relatively few studies have systematically examined the effect of sequential carbon black incorporation in NR–BR blends [25],[26]. In industrial practice, carbon black is generally added according to established production procedures without fully considering [26] how different filler split ratios between the individual polymer phases [27] may influence dispersion efficiency [28]–[30]. Consequently, there remains limited scientific understanding of how varying the proportion of carbon black added during different stages of mixing affects filler distribution [31]–[33], polymer–filler interaction [34], and the resulting compound properties [35]. Addressing this knowledge gap is particularly important for modern tire manufacturing [36], where small improvements in filler dispersion can convert into significant gains in product quality, production efficiency, and energy consumption [37]–[40].

The present investigation focuses on optimizing carbon black dispersion through a sequential mixing strategy in natural rubber–polybutadiene rubber compounds [41],[42]. Different carbon black split ratios were employed during the masterbatch preparation to examine how the timing and proportion of filler addition influence dispersion quality [43]–[45]. By varying the amount of carbon black introduced into the NR and BR phases during sequential mixing, the study aims to evaluate the role of polymer affinity in controlling filler distribution and agglomerate breakdown. Unlike conventional approaches that primarily emphasize processing parameters, the proposed strategy explores the influence of filler addition sequence as an effective means of improving dispersion while maintaining the same overall compound formulation.

The significance of this work extends beyond laboratory-scale experimentation. In commercial rubber manufacturing, improved carbon black dispersion contributes to better process stability, reduced mixing energy, shorter mixing cycles, enhanced batch-to-batch consistency, and improved mechanical performance of finished products. The findings of this study are therefore expected to provide practical guidance for optimizing industrial mixing protocols used in tire tread compounds and other carbon black reinforced NR–BR applications.

Accordingly, the objectives of the present study are to investigate the effect of sequential carbon black addition on filler dispersion in NR–BR compounds, to compare different carbon black split ratios and mixing strategies, to identify the most effective mixing sequence for achieving superior carbon black dispersion, and to establish the relationship between filler dispersion, rheological behavior, curing characteristics, and mechanical properties [46] of the developed rubber compounds.

## 2. MATERIALS AND METHODS

### 2.1 Materials

The rubber compounds in this study were prepared using commercially available elastomers, reinforcing fillers, processing additives, and vulcanization chemicals commonly using in tire tread formulations. Natural rubber (NR) was selected as the primary elastomer owing to its excellent green strength and fatigue resistance, while polybutadiene rubber (BR) was incorporated to improve abrasion resistance and reduce heat build-up during dynamic service. N326 grade carbon black was used as the reinforcing filler because of its balanced reinforcement and processing characteristics. Zinc oxide and stearic acid served as vulcanization activators, whereas paraffinic process oil was used to facilitate filler incorporation and improve compound processability. Microcrystalline wax and 6PPD were included as protective additives to enhance resistance against ozone and oxidative degradation. Vulcanization was carried out using a conventional sulfur curing system comprising sulfur, CBS accelerator, and PVI pre-vulcanization inhibitor.

**Table 1**: Materials used in the study

| Sr. No. | Compounding Ingredient | Grade | Purpose |
|---|---|---|---|
| 01 | Natural Rubber (NR) | TSR 20 | Base elastomer |
| 02 | Polybutadiene Rubber (BR) | High cis BR | Improve abrasion resistance and resilience |
| 03 | Carbon Black | N326 | Reinforcing filler |
| 04 | Zinc Oxide | Rubber Grade | Cure activator |
| 05 | Stearic Acid | Triple Pressed | Cure activator |
| 06 | Microcrystalline Wax | Rubber Grade | Antiozonant wax |

| | | | |
|---|---|---|---|
| 07 | 6PPD | Rubber Grade | Antioxidant / Antiozonant |
| 08 | CBS | Rubber Grade | Primary accelerator |
| 09 | Sulfur | Insoluble Sulfur | Vulcanizing agent |
| 10 | PVI | Rubber Grade | Scorch retarder |
| 11 | Process Oil | RAE Oil | Processing aid |

### 2.2 Compound Formulation

Five rubber formulations were developed to study carbon black distribution and polymer addition sequence on filler dispersion. The total rubber content (NR/BR ratio), curing package, and additive concentrations were maintained constant, while only the sequence of carbon black incorporation and BR addition was systematically varied. This approach enabled the effect of mixing strategy to be evaluated independently without introducing compositional differences.

**Table 2:** Experimental Formulations and their Composition

**Formulation of 1st Master Compound**

| Compounding Ingredient | X-A | X-HT1 | X-HT2 | X-BR1 | X-BR2 |
|---|---|---|---|---|---|
| NR | 60 | 60 | 60 | 60 | 60 |
| BR | 40 | 40 | 40 | 00 | 00 |
| N326 | 34 | 34 | 34 | 38 | 12 |
| | All other Compounding Ingredients are in same proportion in all the samples. ZnO 4; St. Acid 2 and RAE Oil 3 PHR | | | | |
| **Total PHR** | **143** | **143** | **143** | **107** | **81** |

**Formulation of 2nd Master Compound**

| Compounding Ingredient | X-A | X-HT1 | X-HT2 | X-BR1 | X-BR2 |
|---|---|---|---|---|---|
| Master - 1 | 143 | 143 | 143 | 107 | 81 |
| BR | 00 | 00 | 00 | 40 | 40 |
| N326 | 16 | 16 | 16 | 12 | 38 |
| | All other Compounding Ingredients are in same proportion in all the samples. M.C. Wax 1 and 6PPD 2 PHR | | | | |
| **Total PHR** | **162** | **162** | **162** | **162** | **162** |

**Formulation of Final Compound**

| Compounding Ingredient | X-A | X-HT1 | X-HT2 | X-BR1 | X-BR2 |
|---|---|---|---|---|---|
| Master - 2 | 162 | 162 | 162 | 162 | 162 |
| | All other Compounding Ingredients are in same proportion in all the samples. Sulfur 1.12; CBS 0.80 and PVI 0.20 PHR | | | | |
| **Total PHR** | **164.12** | **164.12** | **164.12** | **164.12** | **164.12** |

The experimental formulations used in this study are presented in Table 2. Five formulation, designated as X-A, X-HT1, X-HT2, X-BR1, and X-BR2, were prepared using an identical base formulation while systematically varying the mixing sequence of polybutadiene rubber (BR) and carbon black. The objective was to investigate the influence of sequential filler incorporation and polymer-filler affinity on carbon black dispersion without altering the overall compound composition.

The compounding process was carried out in three successive mixing stages. During the first masterbatch, natural rubber (NR) was introduced as the primary polymer along with zinc oxide, stearic acid, RAE oil, and a predetermined quantity of N326 carbon black. For the reference formulation (X-A) and the high-temperature formulations (X-HT1 and X-HT2), the complete rubber blend (60 phr NR and 40 phr BR) was incorporated in the first stage together with 34 phr of carbon black. In contrast, formulations X-BR1 and X-BR2 were designed to evaluate the effect of polymer affinity by delaying the addition of BR until the second mixing stage. Consequently, only NR was present in the first masterbatch, while the carbon black loading was varied to

38 phr for X-BR1 and 12 phr for X-BR2, corresponding to approximately 75% and 25% of the total carbon black content, respectively.

During the second masterbatch, the remaining quantity of carbon black was incorporated together with BR (where applicable), microcrystalline wax, and 6PPD. For X-A, X-HT1, and X-HT2, only the remaining 16 phr of carbon black was added because the complete NR/BR blend had already been introduced during the first stage. In X-BR1, BR was added together with the remaining 12 phr of carbon black, whereas X-BR2 received BR along with the remaining 38 phr of carbon black. This experimental approach maintained a constant total carbon black loading of 50 phr in all formulations while varying only its distribution between the two masterbatch stages.

The final mixing stage was identical for all compounds. The second masterbatch was blended with the curing system consisting of sulfur (1.12 phr), CBS (0.80 phr), and PVI (0.20 phr) to obtain the final compound with a total loading of 164.12 phr. By keeping the overall formulation constant and modifying only the sequence of carbon black and BR addition, any differences observed in filler dispersion, rheological behavior, or mechanical properties could be directly attributed to the effect of the mixing strategy rather than compositional variations.

### 2.3 Experimental Methodology

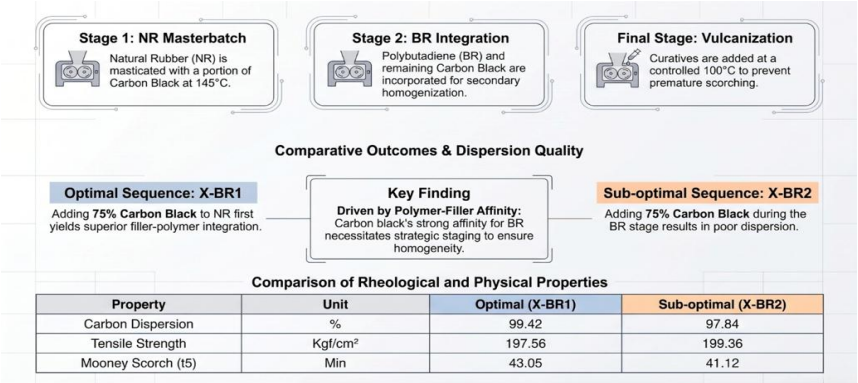


**Figure 1**: Visual Methodology

The experimental program was designed to evaluate the influence of sequential carbon black addition on filler dispersion in NR/BR blends. Five formulations (X-A, X-HT1, X-HT2, X-BR1, and X-BR2) were prepared using a three-stage mixing process. The reference compound (X-A) followed the conventional mixing sequence, whereas X-HT1 and X-HT2 were prepared by increasing the masterbatch dump temperature to promote improved filler wetting. Formulations X-BR1 and X-BR2 were specifically designed to investigate the effect of polymer affinity by altering the proportion of carbon black introduced before and after BR addition. In X-BR1, 75% of the carbon black was incorporated with NR during the first mixing stage and the remaining 25% after BR addition, whereas the reverse distribution was adopted in X-BR2. This systematic variation enabled the role of filler migration and polymer–filler interaction to be examined while maintaining an identical compound formulation.

### 2.4 Mixing Procedure

All rubber compounds were prepared using a 1.6 L laboratory Banbury internal mixer following a three-stage mixing procedure to ensure consistent filler dispersion and compound homogeneity. The processing parameters were maintained constant for all formulations so that the influence of the carbon black addition sequence could be evaluated without interference from changes in mixing conditions.

In the first masterbatch (Masterbatch-I), the base elastomer(s), reinforcing filler, processing oil, zinc oxide, and stearic acid were charged into the Banbury mixer according to the respective formulation (Table 2). Mixing was carried out at a rotor speed of 60 rpm with a fill factor of 0.72 for approximately 275 s, after which the compound was discharged at a dump temperature of 145°C. This stage was primarily intended to achieve effective mastication of the rubber and initial incorporation of the reinforcing filler.

The discharged masterbatch was subsequently processed in the second masterbatch (Masterbatch-II), during which the remaining carbon black (according to the designated formulation - Table 2), polybutadiene rubber, microcrystalline wax, and

6PPD were incorporated. Mixing was continued at the same rotor speed of 60 rpm for approximately 200 s, and the compound was again discharged at a dump temperature of 145°C to ensure uniform filler distribution and adequate dispersion within the rubber matrix.

After cooling the masterbatch, final mixing was performed by incorporating the curing ingredients, such as sulfur, CBS, and PVI. The curatives were blended for approximately 150 s, and the compound was discharged at a maximum temperature of 100°C to prevent premature vulcanization (scorch). The final compounds were then conditioned before further rheological, mechanical, and morphological characterization.

### 2.5 Testing Methods

The processing characteristics of the uncured compounds were evaluated by measuring the Mooney viscosity in accordance with ASTM D1646, while the curing behavior, including scorch time, optimum cure time, and maximum torque, was determined using a Moving Die Rheometer (MDR) following ASTM D5289. The vulcanized compounds were subsequently characterized for their physical and mechanical properties. Shore A hardness was measured according to ASTM D2240, whereas tensile strength, modulus, and elongation at break were determined using a universal testing machine (UTM) in accordance with ASTM D412. The dynamic resilience of the cured compounds was assessed by the rebound resilience test following ASTM D1054. In addition, the quality of carbon black distribution within the rubber matrix was evaluated using a carbon black dispersion analyzer in accordance with ASTM D7723. All tests were conducted under controlled laboratory conditions, and the results obtained were used to assess the influence of different sequential mixing strategies on processing behavior, curing characteristics, filler dispersion, and the overall performance of the NR/BR compounds.

## 3. RESULTS AND DISCUSSION

### 3.1 Mooney Viscosity

Mooney viscosity is one of the most important processing parameters because it reflects the flow behavior of an uncured rubber compound and indirectly indicates the extent of polymer–filler interaction developed during mixing. Figure compares the Mooney viscosity values obtained for the different sequential carbon black addition strategies. Although all compounds were prepared using identical ingredients and total filler loading, noticeable variations in viscosity were observed due to differences in the sequence of carbon black incorporation.

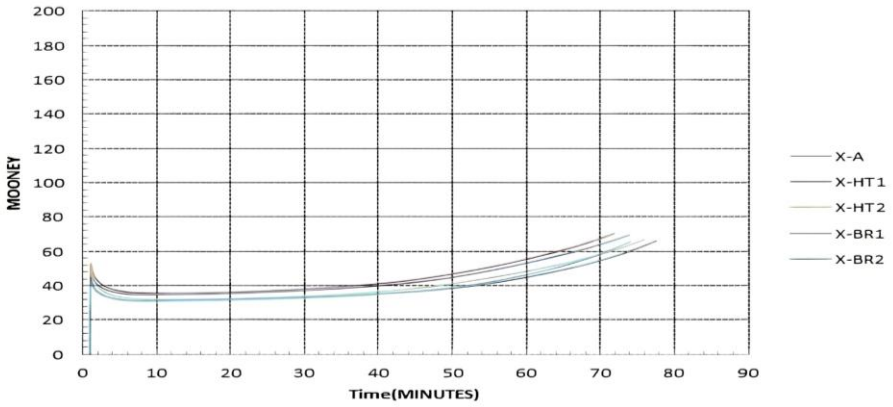


**Figure 2**: Mooney Viscosity of different Samples

The increase in viscosity observed for formulations containing a higher proportion of carbon black during the initial mixing stage can be attributed to the early development of a stronger filler network within the natural rubber matrix. Natural rubber possesses higher green strength and strain-induced crystallization characteristics than polybutadiene rubber, enabling carbon black aggregates to be wetted and dispersed more effectively during mastication. As a result, a larger fraction of filler becomes immobilized within the polymer chains, increasing resistance to flow and consequently increasing Mooney viscosity.

Conversely, when only a limited amount of carbon black was introduced during the first stage and the majority was added after the incorporation of BR, the viscosity tended to decrease. Polybutadiene rubber exhibits comparatively lower viscosity and reduced polarity toward carbon black, resulting in slower filler wetting and less efficient aggregate breakdown. Consequently,

weaker filler–polymer interactions and a less developed filler network were formed, leading to improved flow ability but comparatively inferior dispersion.

The viscosity trend therefore provides indirect evidence that sequential mixing significantly influences filler distribution within the blend. A moderate increase in viscosity indicates enhanced polymer–filler interaction rather than excessive filler agglomeration, suggesting that appropriate carbon black loading during the first mixing stage promotes efficient filler incorporation without adversely affecting process ability.

### 3.2 Cure Characteristics

The curing behavior of the rubber compounds was evaluated using a Moving Die Rheometer, and the obtained parameters are presented in Figure 5. These parameters provide valuable information regarding process safety, crosslink development and curing efficiency.

The minimum torque (ML), representing the viscosity of the uncured compound, followed a trend similar to that observed in the Mooney viscosity measurements. Formulations exhibiting improved carbon black dispersion generally showed slightly higher ML values due to increased polymer–filler interaction before vulcanization.

The maximum torque (MH), which reflects the stiffness and crosslink density of the cured compound, increased for formulations exhibiting superior filler dispersion. Well-dispersed carbon black contributes to a larger effective reinforcing surface area and improves stress transfer within the rubber matrix, resulting in higher final torque values.

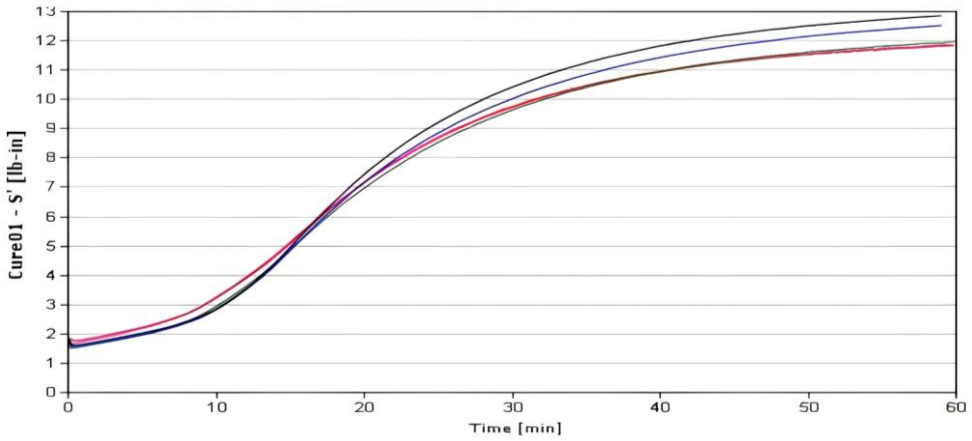


**Figure 3**: Rheographs of different Samples

Scorch time ($Ts_2$) remained relatively unchanged among the compounds, indicating that the sequential filler addition strategy did not significantly alter the onset of vulcanization. This observation suggests that the accelerator and sulfur distribution remained sufficiently uniform despite differences in filler incorporation sequence.

**Table 3**: Rheological Properties

| Physical Properties | Unit | X-A | X-HT1 | X-HT2 | X-BR1 | X-BR2 |
|---|---|---|---|---|---|---|
| ML | lb-in | 1.65 | 1.77 | 1.57 | 1.52 | 1.59 |
| MH | lb-in | 11.88 | 11.84 | 11.98 | 12.55 | 12.88 |
| Ts1 | Minutes | 7.20 | 7.84 | 8.20 | 8.21 | 8.35 |
| $Ts_2$ | Minutes | 10.54 | 10.99 | 11.24 | 11.43 | 11.54 |
| Tc10 | Minutes | 7.95 | 8.42 | 8.85 | 9.27 | 9.50 |

| | | | | | | |
|---|---|---|---|---|---|---|
| Tc15 | Minutes | 9.80 | 10.15 | 10.60 | 11.11 | 11.32 |
| Tc25 | Minutes | 12.78 | 13.10 | 13.43 | 13.85 | 13.99 |
| Tc50 | Minutes | 18.92 | 19.04 | 19.45 | 19.67 | 19.52 |
| Tc90 | Minutes | 38.70 | 38.31 | 39.72 | 40.17 | 39.30 |
| Torque @60' | lb-in | 11.85 | 11.82 | 11.91 | 12.52 | 12.86 |
| MH-ML | lb-in | 10.23 | 10.07 | 10.41 | 11.03 | 11.29 |
| Reversion | % | 0.29 | 0.20 | 0.67 | 0.27 | 0.18 |
| CRI | $Minute^{-1}$ | 3.55 | 3.66 | 3.51 | 3.48 | 3.60 |

Similarly, the optimum cure time ($Tc_{90}$) exhibited only marginal variations. However, compounds with improved filler dispersion generally reached optimum cure slightly earlier due to better heat transfer and more homogeneous distribution of curing ingredients throughout the compound.

The Cure Rate Index (CRI), calculated from $Ts_2$ and $Tc_{90}$, showed a slight improvement for the formulations prepared with higher first-stage carbon black loading. Enhanced filler dispersion provides a more uniform reaction environment, facilitating efficient crosslink formation during vulcanization.

Overall, the rheometric results indicate that sequential carbon black addition primarily influences the extent of reinforcement rather than fundamentally altering the vulcanization chemistry.

### 3.3 Mechanical Properties

The mechanical performance of filled rubber compounds is strongly governed by filler dispersion, polymer–filler adhesion and the efficiency of stress transfer across the rubber matrix. The results presented here clearly demonstrate that sequential carbon black incorporation significantly influences these characteristics.

The tensile strength increased progressively as a larger proportion of carbon black was incorporated during the initial mixing stage, reaching its maximum for the BR1 formulation. This improvement can be attributed to enhanced dispersion of carbon black within the natural rubber phase, leading to a more uniform reinforcing network. Better dispersion reduces stress concentration sites and allows applied loads to be distributed more effectively throughout the matrix during deformation.

**Table 4**: Physical Properties

| Physical Properties | Unit | X-A | X-HT1 | X-HT2 | X-BR1 | X-BR2 |
|---|---|---|---|---|---|---|
| Elongation @ Break | % | 708 | 709 | 702 | 686 | 689 |
| 100% Modulus | $Kgf/cm^2$ | 15.75 | 15.49 | 16.23 | 15.77 | 15.64 |
| 200% Modulus | $Kgf/cm^2$ | 33.27 | 31.51 | 33.86 | 33.28 | 33.25 |
| 300% Modulus | $Kgf/cm^2$ | 60.74 | 57.43 | 61.93 | 62.06 | 61.55 |
| Tensile Strength | $Kgf/cm^2$ | 199.12 | 199.68 | 203.58 | 197.56 | 199.36 |
| Hardness | Shore A | 59 | 58 | 59 | 57 | 58 |
| Rebound Resilience | % (@100 °C) | 56 | 54 | 56 | 56 | 56 |

The elongation at break exhibited only moderate variations among the compounds. Improved filler dispersion generally maintains higher extensibility because uniformly distributed filler particles delay crack initiation and propagation. In contrast, poorly dispersed agglomerates behave as localized defects that promote premature failure under tensile loading.

The modulus values increased with improved filler dispersion due to stronger polymer–filler interactions and greater restriction of molecular chain mobility. Carbon black particles effectively anchor polymer chains at their surfaces, thereby increasing resistance to deformation. The sequential mixing strategy enhanced this reinforcing effect by maximizing the contact between natural rubber and carbon black during the initial stage of mixing.

Similarly, Shore A hardness increased with increasing filler reinforcement. Better carbon black distribution produces a denser reinforcing network that restricts elastic deformation under indentation loading. However, the increase in hardness remained moderate, indicating that improved reinforcement was achieved without excessive filler agglomeration.

Overall, the mechanical property results confirm that the sequential mixing strategy enhances reinforcement primarily through improved carbon black dispersion rather than changes in compound formulation or curing chemistry.

### 3.4 Carbon Black Dispersion

Carbon black dispersion represents the most critical parameter in evaluating the effectiveness of the proposed sequential mixing strategy because it directly determines the reinforcing efficiency of the filler. The dispersion analysis clearly demonstrated that the order of carbon black addition substantially influenced the final microstructure of the NR/BR blend. Natural rubber possesses comparatively higher affinity toward carbon black due to its higher viscosity, superior green strength and greater capability to generate shear stresses during mixing. When a major fraction of carbon black was introduced together with NR in the first mixing stage, the filler aggregates experienced intense shear deformation and efficient wetting by the polymer chains. This promoted progressive breakdown of agglomerates into finer particles, resulting in a more homogeneous filler distribution.

**Table 5**: Carbon Black Dispersion

| Physical Properties | Unit | X-A | X-HT1 | X-HT2 | X-BR1 | X-BR2 |
|---|---|---|---|---|---|---|
| X | - | 7.25 | 7.83 | 8.15 | 8.10 | 6.68 |
| Y | - | 9.84 | 9.82 | 9.86 | 9.92 | 9.74 |
| Dispersion | % | 98.80 | 98.54 | 98.93 | 99.42 | 97.84 |

After incorporation of BR during the second stage, the already dispersed carbon black particles were redistributed throughout the blend without significant re-agglomeration. Since the filler had already been effectively wetted by NR, the lower-viscosity BR phase could flow around the dispersed particles, producing a continuous and uniform filler network across both polymer phases.

In contrast, formulations receiving most of the carbon black after BR addition exhibited noticeably poorer dispersion. At this stage, the blend viscosity had already decreased because of BR incorporation, reducing the shear stresses required to fracture carbon black agglomerates. Consequently, larger filler clusters persisted throughout mixing and acted as defects within the vulcanized compound.

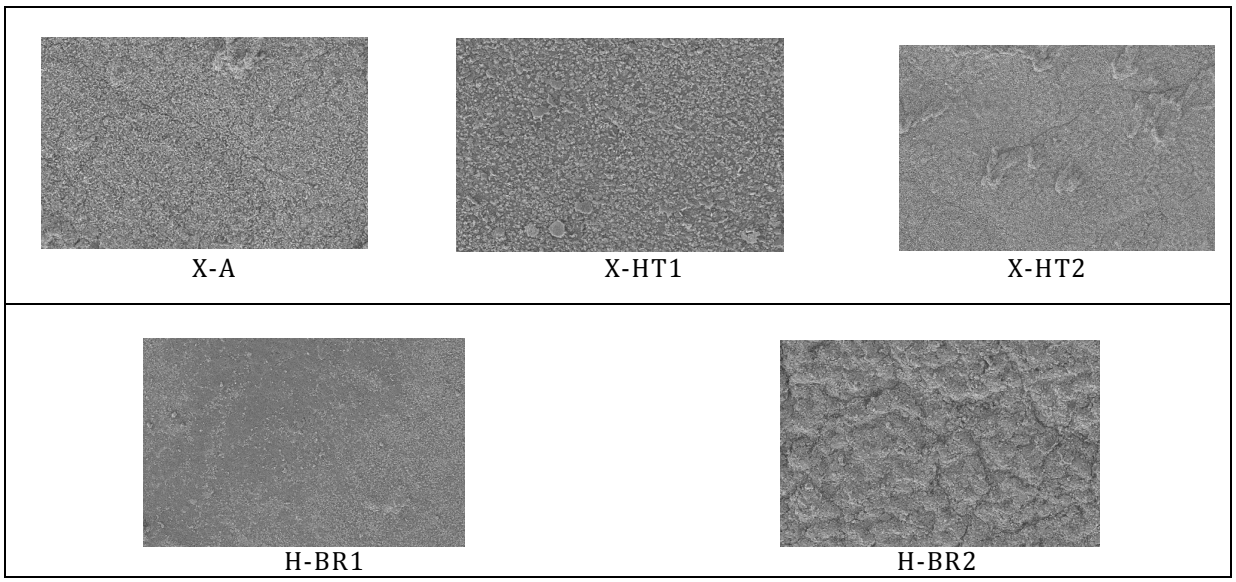


**Figure 4**: Microscopic images of different Samples

Microscopic observations supported these findings by revealing fewer and smaller carbon black aggregates in BR1 compared with others. The improved microstructure explains the higher tensile strength, modulus and cure torque observed for BR1. A clear correlation was observed among carbon black dispersion, rheological behavior and mechanical performance. Improved dispersion increased the effective interfacial area between the filler and polymer chains, strengthening polymer–filler interactions. This enhanced interaction resulted in higher Mooney viscosity due to greater restriction of chain mobility in the uncured compound.

The same improvement in filler distribution also contributed to increased maximum cure torque, indicating a stiffer and more effectively reinforced vulcanized network. Enhanced stress transfer across the well-dispersed carbon black network ultimately produced higher tensile strength and modulus while maintaining satisfactory elongation at break.

These relationships demonstrate that the sequential mixing strategy influences the entire structure–property chain, beginning with filler dispersion during mixing and extending to rheological characteristics, vulcanization behavior and final mechanical performance. Such comprehensive correlation confirms that optimizing the sequence of carbon black addition is an effective approach for improving the overall performance of NR/BR compounds without altering the formulation composition.

## 4. CONCLUSIONS

This study systematically investigated the influence of sequential carbon black addition on the dispersion behavior and mechanical performance of Natural Rubber (NR)/Polybutadiene Rubber (BR) compounds. The results clearly demonstrate that the sequence of filler incorporation plays a crucial role in determining the final dispersion quality of carbon black within the rubber matrix. Introducing a larger proportion of carbon black into the NR phase during the initial mixing stage promoted more efficient filler wetting, reduced agglomeration, and facilitated the formation of a well-developed reinforcing network before the addition of BR. Among all formulations, the X-BR1 mixing sequence exhibited the highest carbon black dispersion value of 99.42%, confirming the effectiveness of the proposed sequential mixing strategy.

Although significant improvements in carbon black dispersion were achieved through different split ratios, the overall mechanical properties of the vulcanized compounds, including tensile strength, elongation at break, modulus, hardness, rebound resilience, and abrasion resistance, remained comparable across all formulations. This indicates that optimizing filler distribution can substantially improve compound homogeneity without adversely affecting the final performance characteristics. The enhanced dispersion observed in the NR-rich initial mixing route is attributed to stronger polymer–filler affinity, improved shear-assisted deagglomeration, and more uniform filler migration during subsequent blending with BR. Overall, the findings demonstrate that sequential mixing is an effective and practical approach for controlling filler morphology in NR/BR compounds. Since the proposed method requires only modification of the mixing sequence without additional chemicals or processing equipment, it offers a simple, cost-effective, and industrially applicable strategy for improving carbon black dispersion in rubber compounding operations.

## 5. FUTURE WORK

The present investigation establishes the significance of sequential mixing in improving carbon black dispersion; however, further studies can provide deeper insight into the underlying reinforcement mechanisms. Advanced morphological characterization using Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM) would enable direct visualization of filler distribution, aggregate size, and polymer–filler interfaces at micro- and nanoscale levels. Dynamic characterization through Dynamic Mechanical Analysis (DMA) and Payne effect measurements could further elucidate filler network formation, viscoelastic behavior, and breakdown of carbon black structures under cyclic deformation.

Future work may also include bound rubber measurements to quantify polymer–filler interactions developed during different mixing sequences. Investigating other reinforcing carbon black grades such as N220, N330, and N550 would help determine whether the observed mixing effects are independent of filler particle size and structure. In addition, extending the study to hybrid reinforcement systems containing carbon black and silica could provide valuable information for the development of high-performance and low rolling resistance rubber compounds. Finally, validation of the proposed sequential mixing strategy under industrial-scale Banbury mixer conditions would establish its practical feasibility for commercial tire and general rubber product manufacturing, thereby facilitating its adoption in large-scale rubber compounding processes.

## DECLARATION

**Funding:** This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

**Ethical approval:** Not Applicable.

**Informed consent:** Written informed consent was obtained from all participants prior to their participation in the study.

**Author Contributions:** Mr. Mahaveer Joshi conducted the literature review, compiled and analyzed relevant research findings, performed field trials, and prepared the original draft of the manuscript. Mr. Chandrakant Sinha performed the data analysis and provided guidance in drafting and structuring the review paper to ensure its scientific clarity. Prof. Mukul Nijay supervised the overall study, provided continuous guidance throughout the research, and directed the development of the manuscript. Dr. Neetesh Kumar critically reviewed the entire manuscript to ensure its scientific accuracy, technical coherence, and overall quality, and provided valuable support in addressing the chemistry-related aspects of the research. All authors actively participated in scientific discussions, critically reviewed the manuscript, contributed to its refinement, and approved the final version for submission.

**Data Availability Statement**: The datasets generated and analyzed during the current study are available from the corresponding author upon reasonable request.

**Conflict of Interest:** The authors declare that they have no competing interests.

**Clinical Trial Number:** Not Applicable.

## REFERENCES

1) Shi, Y.; Li, Y.; Xiang, F.; Huang, T.; Chen, C.; Peng, Y.; Wang, Y. Carbon Nanotubes Induced Microstructure and Mechanical Properties Changes in Cocontinuous Poly( L-Lactide)/Ethylene-Co-Vinyl Acetate Blends. Polym. Adv. Technol. 2012, 23 (4), 783–790. https://doi.org/10.1002/pat.1959.

2) Xiao, J.; Zhang, Z.; Luan, Y.; Wu, S.; Wu, Y. Influence of Carbon Black Surface Characteristics on CB-NR Interfacial Interaction: Molecular Simulation and Experimental Study. Compos. Part A Appl. Sci. Manuf. 2024, 182. https://doi.org/10.1016/j.compositesa.2024.108198.

3) Feng, J.; Chan, C. M.; Li, J. X. A Method to Control the Dispersion of Carbon Black in an Immiscible Polymer Blend. Polym. Eng. Sci. 2003, 43 (5), 1058–1063. https://doi.org/10.1002/pen.10089.

4) Georgousis, G.; Pandis, C.; Chatzimanolis-Moustakas, C.; Kyritsis, A.; Kontou, E.; Pissis, P.; Krajči, J.; Chodák, I.; Tabačiarová, J.; Mičušík, M.; Omastová, M. Study of the Reinforcing Mechanism and Strain Sensing in a Carbon Black Filled Elastomer. Compos. B Eng. 2015, 80, 20–26. https://doi.org/10.1016/j.compositesb.2015.05.021.

5) Sivaselvi, K.; Varma, V. S.; Harikumar, A.; Jayaprakash, A.; Sankar, S.; Krishna, C. Y.; Gopal, K. Improving the Mechanical Properties of Natural Rubber Composite with Carbon Black (N220) as Filler. In Materials Today: Proceedings; Elsevier Ltd, 2020; Vol. 42, pp 921–925. https://doi.org/10.1016/j.matpr.2020.11.836.

6) Chen, J.; Shi, Y. Y.; Yang, J. H.; Zhang, N.; Huang, T.; Wang, Y. Improving Interfacial Adhesion between Immiscible Polymers by Carbon Nanotubes. Polymer (Guildf). 2013, 54 (1), 464–471. https://doi.org/10.1016/j.polymer.2012.11.042.

7) Omnès, B.; Thuillier, S.; Pilvin, P.; Grohens, Y.; Gillet, S. Effective Properties of Carbon Black Filled Natural Rubber: Experiments and Modeling. Compos. Part A Appl. Sci. Manuf. 2008, 39 (7), 1141–1149. https://doi.org/10.1016/j.compositesa.2008.04.003.

8) Shukri, N. M.; Azura, A. R. Effect of Different Types of Rubbers and Carbon Black Grades on Curing Characteristics and Dynamic Properties for Anti-Vibration Application. In IOP Conference Series: Materials Science and Engineering; Institute of Physics Publishing, 2019; Vol. 548. https://doi.org/10.1088/1757-899X/548/1/012002.

9) Burgaz, E.; Gencoglu, O.; Goksuzoglu, M. Carbon Black Reinforced Natural Rubber/Butadiene Rubber and Natural Rubber/Butadiene Rubber/Styrene-Butadiene Rubber Composites: Part I: Rheological, Mechanical and Thermomechanical Properties. Research on Engineering Structures and Materials 2019, 5 (3), 233–247. https://doi.org/10.17515/resm2019.74ma1123.

10) Hoshikawa, Y.; An, B.; Kashihara, S.; Ishii, T.; Ando, M.; Fujisawa, S.; Hayakawa, K.; Hamatani, S.; Yamada, H.; Kyotani, T. Analysis of the Interaction between Rubber Polymer and Carbon Black Surfaces by Efficient Removal of Physisorbed Polymer from Carbon-Rubber Composites. Carbon N. Y. 2016, 99, 148–156. https://doi.org/10.1016/j.carbon.2015.12.003.

11) Zhao, Q.; Niu, F.; Liu, J.; Yin, H. Research Progress of Natural Rubber Wet Mixing Technology. Polymers. Multidisciplinary Digital Publishing Institute (MDPI) July 1, 2024. https://doi.org/10.3390/polym16131899.

12) Madera, R. C.; Lejeunes, S.; Euchler, E. Modeling the Payne Effect in Filled Rubbers: Accounting for Time Effect and Partial Reversibility. Polymer Bulletin 2025, 82 (9), 3527–3552. https://doi.org/10.1007/s00289-025-05680-z.

13) Osman, F. K.; Hou, D.; Han, L.; Zhou, Q.; Gao, J.; Zhang, C.; Miao, L.; Iranzo, A. Power Consumption and Rubber Phase Evolution in an Intermeshing Mixer: A Three-Dimensional Non-Newtonian Volume-of-Fluid Computational Fluid Dynamics Analysis. Polymers (Basel). 2026, 18 (10). https://doi.org/10.3390/polym18101163.

14) Robertson, C. G.; Hardman, N. J. Nature of Carbon Black Reinforcement of Rubber: Perspective on the Original Polymer Nanocomposite. Polymers. MDPI AG February 2, 2021, pp 1–28. https://doi.org/10.3390/polym13040538.

15) Chauhan, M.; Patel, B.; Desai, R. Enhancement of Soil Fertility Using Waste Natural Rubber Latex Gloves. Discover Soil 2026, 3 (1), 110. https://doi.org/10.1007/s44378-026-00272-3.

16) Won, Y. Y.; Meeker, S. P.; Trappe, V.; Weitz, D. A.; Diggs, N. Z.; Emert, J. I. Effect of Temperature on Carbon-Black Agglomeration in Hydrocarbon Liquid with Adsorbed Dispersant. Langmuir 2005, 21 (3), 924–932. https://doi.org/10.1021/la047906t.

17) Zhu, L.; Tian, X.; Pan, Y.; Chang, T.; Wang, K.; Niu, G.; Zhang, L.; Wang, C.; Han, W. Optimization of Serial Modular Continuous Mixing Process Parameters for Natural Rubber Composites Reinforced by Silica/Carbon Black. Polymers (Basel). 2020, 12 (2). https://doi.org/10.3390/polym12020416.

18) Fröhlich, J.; Niedermeier, W.; Luginsland, H. D. The Effect of Filler-Filler and Filler-Elastomer Interaction on Rubber Reinforcement. Compos. Part A Appl. Sci. Manuf. 2005, 36 (4), 449–460. https://doi.org/10.1016/j.compositesa.2004.10.004.

19) Liang, J.; Wu, J.; Wang, C.; Sang, J.; Ren, K.; Ngai, T.; Lin, W. Ethylene Vinyl Acetate as a Multifunctional Compatibilizer for Natural Rubber/Cis-1,4-Polybutadiene Rubber Blends with Enhanced Compatibility, Filler Dispersion and Mechanical Properties. Collagen and Leather 2025, 7 (1). https://doi.org/10.1186/s42825-024-00181-7.

20) Vishvanathperumal, S. Effect of Octavinyl-Functionalized Polyhedral Oligomeric Silsesquioxane/Carboxylated Carbon Nanotube (OV-POSS/COOH-CNT) Hybrid Filler on the Properties of Chloroprene Rubber/Natural Rubber (CR/NR) Nanocomposites. Emergent Mater. 2026, 9 (1–3). https://doi.org/10.1007/s42247-026-01383-x.

21) Mofrad, F. J.; Ahmadpour, A.; Ostad Movahed, S. The Effect of Modified Carbon Black and Nano-Silica, Individually and in Combination, on the Curing, Mechanical, and Dynamical Properties of Butyl Rubber (IIR). Polymers and Polymer Composites 2024, 32. https://doi.org/10.1177/09673911241256553.

22) McGlasson, A.; Rishi, K.; Beaucage, G.; Narayanan, V.; Chauby, M.; Mulderig, A.; Kuppa, V. K.; Ilavsky, J.; Rackaitis, M. The Effects of Staged Mixing on the Dispersion of Reinforcing Fillers in Elastomer Compounds. Polymer (Guildf). 2019, 181. https://doi.org/10.1016/j.polymer.2019.121765.

23) Thuong, N. T.; Long, N. T.; Nghia, P. T.; Linh, N. P. D.; Lam, N. B.; Kawahara, S. Graft Copolymerization of 2-(Dimethylamino)Ethyl Methacrylate onto Natural Rubber and Its Effect on Vulcanization of Natural Rubber. Journal of Polymer Research 2025, 32 (11). https://doi.org/10.1007/s10965-025-04619-2.

24) Chauhan, M.; Padhiyar, S.; Desai, R.; Patel, B.; Shukla, P. Sustainable Reinforcement of Rubber Compound Using Recycled PET (Polyethylene Terephthalate): A Review. Nature Environment and Pollution Technology. Technoscience Publications March 1, 2026. https://doi.org/10.46488/NEPT.2026.v25i01.B4342.

25) Włoch, M.; Ostaszewska, U.; Datta, J. The Effect of Polyurethane Glycolysate on the Structure and Properties of Natural Rubber/Carbon Black Composites. J. Polym. Environ. 2019, 27 (6), 1367–1378. https://doi.org/10.1007/s10924-019-01437-8.

26) Huang, Y.; Jiang, S.; Li, G.; Chen, D. Effect of Fillers on the Phase Stability of Binary Polymer Blends: A Dynamic Shear Rheology Study. Acta Mater. 2005, 53 (19), 5117–5124. https://doi.org/10.1016/j.actamat.2005.07.021.

27) Aggarwal, A.; Hackel, N.; Grunert, F.; Ilisch, S.; Beiner, M.; Blume, A. Investigation of Rheological, Mechanical, and Viscoelastic Properties of Silica-Filled SSBR and BR Model Compounds. Polymers (Basel). 2024, 16 (22). https://doi.org/10.3390/polym16223212.

28) Taguet, A.; Cassagnau, P.; Lopez-Cuesta, J. M. Structuration, Selective Dispersion and Compatibilizing Effect of (Nano)Fillers in Polymer Blends. Progress in Polymer Science. Elsevier Ltd 2014, pp 1526–1563. https://doi.org/10.1016/j.progpolymsci.2014.04.002.

29) Du, M.; Wu, Q.; Zuo, M.; Zheng, Q. Filler Effects on the Phase Separation Behavior of Poly (Methyl Methacrylate)/Poly (Styrene-Co-Acrylonitrile) Binary Polymer Blends. Eur. Polym. J. 2013, 49 (9), 2721–2729. https://doi.org/10.1016/j.eurpolymj.2013.06.006.

30) Ranaei, O.; Aghakouchak, A. A. Experimental and Numerical Study on Developed Elastomeric Layers Based on Natural and Butyl Matrix Rubbers for Viscoelastic Dampers. Mech. Time. Depend. Mater. 2022, 26 (1), 211–233. https://doi.org/10.1007/s11043-021-09484-2.

31) Ulfah, I. M.; Fidyaningsih, R.; Rahayu, S.; Fitriani, D. A.; Saputra, D. A.; Winarto, D. A.; Wisojodharmo, L. A. Influence of Carbon Black and Silica Filler on the Rheological and Mechanical Properties of Natural Rubber Compound. Procedia Chem. 2015, 16, 258–264. https://doi.org/10.1016/j.proche.2015.12.053.

32) Burgaz, E.; Gencoglu, O.; Goksuzoglu, M. Carbon Black Reinforced Natural Rubber/Butadiene Rubber and Natural Rubber/Butadiene Rubber/Styrene-Butadiene Rubber Composites: Part II. Dynamic Mechanical Properties and Fatigue Behavior. Research on Engineering Structures and Materials 2019, 5 (3), 1–13. https://doi.org/10.17515/resm2109.75ma1124.

33) Tran, D.-L.; Tran, M.-T.; Phan, T.-N. Superhydrophobization of Hydrophilic Silica via Solid Phase Reaction Enhances Dispersion in Deproteinized Natural Rubber. Discover Chemistry 2026, 3 (1). https://doi.org/10.1007/s44371-026-00565-0.

34) Fang, S.; Wu, S.; Huang, J.; Wang, D.; Tang, Z.; Guo, B.; Zhang, L. Notably Improved Dispersion of Carbon Black for High-Performance Natural Rubber Composites via Triazolinedione Click Chemistry. Ind. Eng. Chem. Res. 2020, 59 (48), 21047–21057. https://doi.org/10.1021/acs.iecr.0c04242.

35) Al-Saleh, M. H.; Sundararaj, U. An Innovative Method to Reduce Percolation Threshold of Carbon Black Filled Immiscible Polymer Blends. Compos. Part A Appl. Sci. Manuf. 2008, 39 (2), 284–293. https://doi.org/10.1016/j.compositesa.2007.10.010.

36) Lee, S.; Ahn, K.; Hong, S. J.; Kim, Y. T. Effect of Sequence-Based Incorporation of Fillers, Kenaf Fiber and Graphene Nanoplate, on Polypropylene Composites via a Physicochemical Compounding Method. Polymers (Basel). 2025, 17 (14). https://doi.org/10.3390/polym17141955.

37) Tian, C.; Cui, J.; Ning, N.; Zhang, L.; Tian, M. Quantitative Characterization of Interfacial Properties of Carbon Black/Elastomer Nanocomposites and Mechanism Exploration on Their Interfacial Interaction. Compos. Sci. Technol. 2022, 222. https://doi.org/10.1016/j.compscitech.2022.109367.

38) Abbas-Abadi, M. S. The Effect of Process and Structural Parameters on the Stability, Thermo-Mechanical and Thermal Degradation of Polymers with Hydrocarbon Skeleton Containing PE, PP, PS, PVC, NR, PBR and SBR. Journal of Thermal Analysis and Calorimetry. Springer Science and Business Media B.V. February 1, 2021, pp 2867–2882. https://doi.org/10.1007/s10973-020-09344-0.

39) V, B.; K, A.; John, H. Effect of Thermally Exfoliated Graphite on the Performance Characteristics of Epoxidized Natural Rubber Compatibilized Natural Rubber/Polybutadiene Tyre Tread Formulations. Journal of Polymer Research 2024, 31 (12). https://doi.org/10.1007/s10965-024-04201-2.

40) Hamdani, M. H.; Harmain, G. A.; Goswami, M.; Wani, S. S.; Chattopadhyay, S. Fracture Assessment of Single Edge Notched Elastomeric Composite under Mixed-Mode Loading. Mech. Soft Mater. 2025, 7 (1). https://doi.org/10.1007/s42558-025-00066-7.

41) Vayyaprontavida Kaliyathan, A.; Huskic, M.; Thomas, S. Partitioning of Carbon Black within Natural Rubber and Butadiene Rubber Blends Depending on Its Concentration. Next Research 2025, 2 (2), 100353. https://doi.org/10.1016/j.nexres.2025.100353.

42) Vayyaprontavida Kaliyathan, A.; Rane, A. V.; Thomas, S. Nanoscale Partitioning of Carbon Black in Styrene Butadiene Rubber and Butadiene Rubber Miscible Blends and Its Effect on Physico-Mechanical Properties. Journal of Rubber Research 2023, 26 (2), 81–98. https://doi.org/10.1007/s42464-023-00197-4.

43) Plattier, J.; Benyahia, L.; Dorget, M.; Niepceron, F.; Tassin, J. F. Viscosity-Induced Filler Localisation in Immiscible Polymer Blends. Polymer (Guildf). 2015, 59, 260–269. https://doi.org/10.1016/j.polymer.2014.12.044.

44) Zhang, M.; Sheng, Y.; Li, H.; Tang, L.; Li, Z.; Zou, X. Evaluation of Rubber Powder Distribution on the Ductility of Rubber Asphalt: Characterization and Mesoscopic Simulation. J. Mater. Eng. Perform. 2026, 35 (10), 9508–9522. https://doi.org/10.1007/s11665-025-12350-8.

45) El Mogy, S. A.; Chawraba, K. Rubber Nanocomposites: A Review of Processing, Thermo-Mechanical Properties, and Industrial Frontiers. Journal of Polymer Research. Springer Science and Business Media B.V. December 1, 2025. https://doi.org/10.1007/s10965-025-04686-5.

46) Knauert, S. T.; Douglas, J. F.; Starr, F. W. The Effect of Nanoparticle Shape on Polymer-Nanocomposite Rheology and Tensile Strength. J. Polym. Sci. B Polym. Phys. 2007, 45 (14), 1882–1897. https://doi.org/10.1002/polb.21176.