**EFFECT OF GRAPHENE OXIDE ON THE MECHANICAL PROPERTIES OF THE EPDM/NBR COMPOSITES**

###### **A PROJECT REPORT**

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**ABSTRACT**

This study investigates the effect of graphene oxide (GO) on the curing kinetics, mechanical performance, abrasion resistance, and swelling behavior of ethylene propylene diene monomer/acrylonitrile butadiene rubber (EPDM/NBR) composites. The composites were prepared through melt blending using an open mill mixer, ensuring uniform dispersion of GO within the polymer matrix.

Rheological analysis revealed that GO incorporation enhanced the curing characteristics by increasing minimum torque, maximum torque, delta torque, and cure rate index while reducing scorch time and optimizing cure time. Mechanical testing demonstrated that tensile strength and stress at 100% elongation improved with GO addition up to 5 per hr, beyond which a decline was observed due to filler agglomeration. However, elongation at break and rebound resilience were negatively affected, indicating reduced elasticity.

Notably, the presence of GO significantly enhanced the swelling resistance and abrasion resistance of the composites. Field Emission Scanning Electron Microscopy (FESEM) analysis of fractured surfaces confirmed the homogeneous distribution of GO within the polymer network, contributing to its reinforcing effects. Performance evaluations revealed substantial enhancements in key mechanical properties: tensile strength increased by 61%, tear strength by 71%, stress at 100% elongation by 36%, and abrasion resistance by 17%, compared to the base vulcanizates. Conversely, elongation at break and rebound resilience decreased by 27% and 23%, respectively. These findings highlight the potential of GO as an effective reinforcing filler for improving the durability and performance of EPDM/NBR composites, making them suitable for applications requiring high mechanical strength and wear resistance.

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**CHAPTER 1**

**INTRODUCTION**

**1.1 COMPOSITE MATERIALS**

A composite is a type of structural material that is made by fusing two or more different components. The elements are not soluble in one another and are mixed at a macroscopic level. In general, a composite material is any substance made up of two or more parts, each of which has unique qualities and clear boundaries between them. In addition, man has employed the concept of combining various elements to create a material with properties that are not possible with the separate elements for thousands of years. Composites occur in nature. In accordance with this, the vast majority of natural materials that have developed as a result of a protracted evolutionary process can be thought of as composite materials. A piece of wood is a composite because it contains cellulose fibres to support the lignin matrix. Bones are also composites because they contain collagen fibres to strengthen the mineral matrix.

The reinforcing phase is one component of the composite, and the matrix is the component in which it is embedded. The material used in the reinforcing phase can take the shape of particles, fibres, or flakes. The aggregates of the discontinuous phase are kept in the bound form by the materials that make up the matrix phase, which are typically continuous. The matrix is important because without it, the aggregates are useless because they cannot be held in place. The matrix typically serves as a load transfer to the discontinuous phase despite having inferior mechanical qualities to the other phase. Reinforcement is the term for the discontinuous or scattered phase, which is typically chosen to have higher mechanical qualities. The composites' final qualities are significantly influenced by the reinforcement's characteristics.

**1.2 CLASSIFICATION OF COMPOSITES**

A composite material is one that comprises at least two distinct phases (the matrix phase and the dispersion phase) with bulk properties that are very different from those of any of the constituent parts. Composite materials are divided into two categories.

**1.2.1. Classification by matrix material**

1. Metal matrix composites

2. Ceramic matrix composites

3. Carbon matrix composites

4. Polymer matrix composites

1. Metal matrix composites

As their name suggests, Metal Matrix Composites (MMCs) are made of a metal matrix, such as Aluminium (Al), Magnesium (Mg), Zinc (Zn), Copper (Cu), or Titanium (Ti), and a reinforcement, such as Alumina (Al2O3), Silicon Carbide (SiC), Boron (B), graphite, or other particles. Metals are typically strengthened to change their characteristics in accordance with the design requirements. For instance, while metals' elastic stiffness and strength can be raised, their thermal expansion coefficients and electrical and thermal conductivities can be decreased by the inclusion of fillers like SiC. MMCs have better shear and compressive strength, higher specific strength, and higher modulus. These composites are mostly employed in turbine compressor parts, vehicles, maritime engineering, and aircraft. MMCs have less advantages than Polymer Matrix Composites (PMCs) due to greater processing temperatures and denser materials.

2. Ceramic matrix composites

Alumina reinforced by metals or a ceramic matrix, such as calcium aluminium silicate, makes up Ceramic Matrix Composites (CMCs). They are mostly utilised for cutting tool inserts, electronic industry, and high temperature applications. High strength, high service temperature, hardness, low density, chemical inertness are all characteristics of CMCs. Due of the high temperature processing required for CMCs, their development has lagged behind that of other composites. When CMCs are processed at high temperatures, the reinforcement and matrix have different coefficients of thermal expansion, which results in cooling-induced thermal stresses. Concrete is the ideal illustration of a ceramic matrix composite.

3. Carbon carbon composites

Carbon fibres are used in a carbon matrix to create carbon composites. They are 20 times stronger and 30% lighter than fibres, and they can withstand temperatures as high as 3315oC. Carbon is brittle and delicate to flaws on its own, much like porcelain. The composite can endure high temperatures, have minimal creep at high temperatures, low density, high fatigue resistance, good tensile and compressive strength, high coefficient of friction, high thermal conductivity thanks to the reinforcement of a carbon matrix. The disadvantages of these materials are their high cost, oxidation susceptibility, and poor shear strength. They are primarily utilised in fasteners for high temperature applications, aeroplane brakes, and space shuttle nose cones.

4. Polymer matrix composite

PMCs are made of a polymer as the matrix material and particles as the reinforcement material (particles reinforced polymer). Given their qualities at room temperature, low density, ease of production, ideal electrical and thermal properties, outstanding chemical and corrosion resistance, and low cost, these materials are used in the widest range of composite applications. Aerospace, automotive, civil construction, electrical, and marine industries all use polymer composites extensively.

A polymer is a substance made up of molecules with a significant molecular mass that are made up of monomers, which are repeating structural units, and covalent chemical bonds. The term comes from the Greek words polys, which means many, and meros, which means parts. Structure-wise, polymers are more complex than metals or ceramics. Although polymers are inexpensive and simple to produce, their strength, modulus, and low temperature resistance are lower than those of metals. Due to the predominance of covalent bonding, prolonged exposure to UV radiation and some solvents can result in the loss of polymer characteristics. They typically do not conduct heat or electricity well, but they are more chemically resistant than metals. Large chain-like molecules called polymers have the backbone of the chain formed by covalently bound carbon atoms. Polymerization is the process of creating large molecules from small ones. Hardening chemicals are incorporated into the liquid resin to accomplish this. The two primary types of polymerization are condensation polymerization and addition polymerization, often known as chain-growth and step-growth, respectively.

a. Addition polymerization

It entails joining together molecules with two or three chemical bonds. Due to their additional internal bonds, these unsaturated monomers can break and join with other monomers to form the repeating chain. Making polymers like polyethylene, polypropylene, etc. involves addition polymerization.

b. Condensation polymerization

It happens when condensation interactions between the monomers cause their bonding. These reactions are typically triggered by molecules reacting with those containing alcohol, carboxylic acid, amine, or other carboxyl derivative groups. Condensation polymerization occurs when an amine and carboxylic acid react to form an amide or peptide link while releasing water.

**1.3 Classification by reinforcement**

1) Particulate reinforced composites

CB, SiO2, clay, calcium carbonate (CaCO3), and conductive CB are among the materials used for reinforcing. Powdered metallic materials like Al and Cu have also been utilised as fillers. Mica flakes, carbon beads, glass beads, graphite powder, and amorphous materials including ceramics and amorphous SiO2 have also been employed as fillers in addition to metallic powders.

In the past, fillers were thought of as additives that, because of surface areas, poor geometrical characteristics, or chemical compositions on their surfaces, could only slightly raise the polymer's modulus while the strength (tensile, flexural) stayed the same or even declined. Their primary benefit was to reduce material costs by taking the place of the more expensive polymer. Faster moulding cycles because of improved thermal conductivity and fewer rejected parts because of warpage are further potential economic benefits. Other polymer qualities may be impacted by the type of filler; for instance, the addition of fibrous elements may considerably increase melt viscosity. On the other hand, thermal expansion and mould shrinkage would be lessened, which is a typical outcome of most inorganic fillers. Because inorganic reinforcing fillers are stiffer than the matrix and deform less, the overall matrix strain is reduced, especially where the particle and matrix interface is present. An automotive tyre, which contains CB particles in a matrix of polyisobutylene elastomer polymer, is an example of a particle reinforced composite. Concrete is another instance of a particle-reinforced composite, with cement serving as the matrix and aggregates (sand and gravel) as the particles. Higher tensile, shear, and compressive stresses are supported by particle reinforced composites. Particles like these are frequently utilised in polymer composites.

a. Carbon black

Carbon Black (CB) is a colloidal form of elemental carbon. Its colloidal morphology, the size and form of the final units, as well as its surface characteristics, are responsible for its reinforcing nature. CB is made up of fused clusters of individual particles rather than distinct particles. By thermally cracking hydrocarbons or by incomplete combustion, CBs are produced. One of the oldest and most popular fillers in the rubber business is CB. The tyre and rubber sectors use over 90% of the CB generated worldwide. Depending on how they are made, CBs are divided into furnace blacks, thermal blacks, channel blacks, acetylene blacks, and lamp blacks.

The furnace technique is used to produce the majority of CBs used in rubber reinforcing. Furnace type CBs are mostly used in elastomers for the reinforcing they provide to the vulcanisates. On the surface of CBs are reactive organic groups that give them an affinity for rubber. CB is added to rubber to improve its modulus, fatigue resistance, abrasion resistance, and other technological features.

b. Silica

Different SiO2 art forms are offered commercially for a range of industrial uses. Amorphous SiO2, crystalline SiO2, diatomaceous SiO2 (of fossil origin), and microcrystalline SiO2 are significant natural types. Precipitated, pyrogenic, aerogel, and hydrogel types of synthetic SiO2 are available. Precipitated SiO2 and pyrogenic (fume) SiO2 are two of these kinds that are utilised for elastomer reinforcing. Pyrogenic SiO2 is both too costly and active. Precipitated silicas are silicon dioxide particles having a water content of 10–14% and a size range of 1–40 nm. They serve as strengthening fillers in composite materials, resulting in composites with high tensile strength, tear resistance, hardness, and abrasion resistance. It is employed in the production of coloured and translucent items, as well as tyres and other mechanical rubber products. Silicon dioxide that has been pyrogenically altered contains less than 2% mixed water. These SiO2 are highly-reinforcing fillers with very small particle sizes that give silicone rubbers in particular strong tensile strength, tear, and abrasion resistance.

Additional agglomerates are created by the SiO2 filler's secondary particles. The so-called secondary structures they create resemble chains. The shear forces that occur during mixing more or less break the tertiary structures, despite the fact that they are also generally stable. The dispersion is better the higher the shear force.

c. Clay

The majority of non-black filler used in the rubber industry is comprised of clays. In this regard, they are second only to CB. Due to their cost-effectiveness in terms of delivering advantageous reinforcing and processing capabilities at a low cost, clay minerals are frequently employed in rubber compounding. The most significant clay mineral is kaolin, often known as china clay, and its derivative, which is created through chemical and thermal (calcining) processing. If clay strengthens rubber while also adding high tensile strength, high modulus, and resistance to abrasion, it is referred to as hard clay. If clay created a compound with weaker physical qualities, it is referred to as "soft clay." To enhance their performance in rubber, a number of commercial clays have been treated with silane coupling agents. Increased modulus and tensile strength are attained because the silane coupling agent offers a method of fusing the clay particles to the rubber. In most elastomers, clay can be formed to quite high loadings, with soft clays enabling slightly higher loadings than hard clays. Clay loading slightly increases viscosity, however formulations with 150–200 phr (part per hundred rubber) are said to be possible. The decrease in cure rate is the primary element to take into account when adding clay to the majority of formulations. An activator must be added, and/or the accelerator dosage must be increased, to achieve this reduction.

d. Organically Modified Nanoclay

Due to its superior qualities, montmorillonite (MMT), a form of nanoclay, is frequently utilised as a nanofiller in polymer and rubber matrix. Inorganic and hydrophilic in nature is MMT nanoclay. A quaternary ammonium salt is used to biologically modify MMT nanoclay, partially resolving the problem in the creation of the nanocomposite. Given that nanoclay has a high surface area, even a small amount of it can significantly improve the physico-mechanical characteristics of rubber. Based on filler parameters including aspect ratio, particle size, shape, and surface area, improvements in attributes are made. MMT has a surface area between 750 and 800 m2 g-1. By evenly dispersing nanoclay within the rubber matrix, technical qualities of nanocomposite are improved.

2) Fiber-reinforced composites

The matrix is often made of a ductile material that is tough, whereas reinforcing components are typically strong and low density. If the composite is properly planned and constructed, it combines the reinforcement's strength and the matrix's toughness to generate a combination of desired attributes not found in any one conventional material. The amount, configuration, and kind of particle reinforcement in the polymer have a significant impact on the composite's strength. Generally speaking, the strength will increase as the reinforcement content does. A "hybrid" composite, which combines the qualities of many reinforcing materials, can occasionally be made by mixing fillers, such as Carbon Black (CB) or Silica (SiO2), with other fillers. In addition, fillers and additives that alter the composite's performance properties are frequently included in its formulation.

**1.4 TYPES OF POLYMERS**

Depending on their structural and chemical bonds, polymers can be categorised into three classes: thermoplastic polymers, thermoset polymers, and elastomers. Thermoset polymers include epoxies, urea-formaldehyde, vinyl esters, unsaturated polyesters, phenolics, polyurethanes, thermoplastic polymers include Poly-Ethylene (PE), Poly-Propylene (PP), Poly-Amide (PA), Poly-Carbonate (PC), Poly-Styrene (PS), Poly Ether Ether Ketone (PEEK), Poly Tetra Flouro Ethylene (PTFE), Poly Methyl Methacrylate (PMMA), etc and elastomers include Natural Rubber (NR), Styrene-Butadiene Rubber (SBR), Acrylonitrile-Butadiene Rubber (NBR), Ethylene-Propylene-Diene Monomer (EPDM), etc.

**1.4.1. Thermoplastics**

Thermoplastics make up around 85% of all polymers made worldwide. Depending on the kind of their distinctive transition temperature, they can be classified into two major classes: amorphous and crystalline. The Glass-Transition Temperature, Tg, of amorphous thermoplastics, above which the modulus rapidly declines and the polymer exhibits liquid-like qualities, serves as a defining characteristic. Amorphous thermoplastics are often processed at temperatures much above their Tg. Semi-crystalline thermoplastics, or crystalline thermoplastics as they are more properly known, can range in crystallinity from 20 to 90% and are often produced above the melting point. Due to their anisotropic character, thermoplastics have a propensity to melt. Some thermoplastics can be employed in applications like liners for pressure vessels or pipes due of their good chemical resistance. Most thermoset polymers cannot match the temperature stability of thermoplastics like Polyetherimide (PEI), PA, or PEEK. Because thermoplastic melts have a significantly higher viscosity and processing temperatures must be high, there are significant variations between thermoset and thermoplastic materials in terms of processing (up to 400oC for some polymers). Thermoplastics, on the other hand, may be processed far more quickly than thermosets since the thermoplastic polymer immediately hardens by cooling, but thermosets must be cured through a laborious heating and annealing process. In order to shape thermoplastic polymers using viscous flow, the material must first be heated until it softens or melts. The molecules are held together by weak bonds (Van der Waal forces) and have a linear structure with no intermolecular cross-linking. The Van der Waal forces between the molecules in thermoplastic materials are broken by heating, changing the shape from solid to liquid. Contrarily, covalent bonds in thermosetting materials prevent them from being easily destroyed by heating; as a result, they cannot be remelted and produced.

**1.4.2. Thermoset resins:**

The majority of the time, these are low viscosity liquids or low molecular weight monomers or oligomers with functional groups for cross-linking reactions. These resins can be polymerized (by the "curing reaction") either by addition reaction, which doesn't result in the generation of volatiles (like unsaturated polyester, epoxy resin, or vinyl esters), or by condensation reaction (e.g., phenolic resin). Thermoset resins produce a three-dimensional network structure that is highly cross-linked and insoluble in the majority of organic solvents. It is preferable to employ resins that don't release volatile compounds during curing in order to prepare moulded pieces without any voids. The usual methods for curing thermosets at room temperature include amines, acids, cyanates, anhydrides, etc. The post-curing process is done at one or more higher temperatures. These materials develop into a three-dimensional covalently bound network upon curing. Thermoset resins can be low or high viscosity liquids or solids, depending on the backbone structure. Without the use of high temperature or pressure, it is possible to accomplish a satisfactory wetting of the fibres by the resin in the low viscosity resin. High pressure and temperature are used to impregnate fibres with high viscosity resins. To lower viscosity, resins are typically diluted with reactive diluents. Many thermosets have been said to have been almost unusable without the addition of fillers and reinforcing fibres due to their severe brittleness.

**1.4.3. Elastomers**

When compared to other materials, the viscoelasticity of elastomers has a higher yield of strain. These polymers exhibit segmental mobility of polymer chains above their glass transition temperature and are amorphous. These are typically utilised as adhesives, seals, and moulded flexible parts since they are soft and deformable at the right temperature. Examples include silicone rubber, butyl rubber, thermoplastic elastomers, polybutadiene, NR, SBR, NBR, EPDM, fluoroelastomers, chloroprene, synthetic polyisoprene, and so forth.

a. Acrylonitrile-butadiene rubber

Nitrile Butadiene Rubber (NBR) is a synthetic elastomer known for its excellent resistance to oils, fuels, and chemicals due to its high acrylonitrile (ACN) content. The ACN content in NBR typically ranges from 18% to 50%, with higher ACN levels providing better oil resistance but reducing flexibility and low-temperature performance. In EPDM/NBR composites, the NBR content plays a crucial role in determining mechanical properties, swelling resistance, and overall durability.

The incorporation of NBR in EPDM/NBR blends enhances resistance to hydrocarbons and improves tensile strength while maintaining good flexibility. However, the proportion of NBR in the composite must be optimized to balance oil resistance with elasticity and thermal stability. The addition of fillers, such as graphene oxide, can further modify the properties of the blend by reinforcing the rubber matrix, enhancing mechanical strength, and improving swelling resistance.

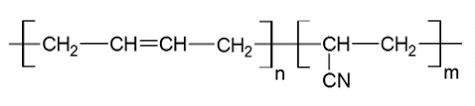


Figure 1.1 Structure of NBR

b. Ethylene-propylene-diene monomer

Using the catalyst technology for polymerizing a-olefins that Ziegler and Natta, who were given the Nobel Prize for Chemistry in 1963, discovered and perfected, this class of elastomers was first presented in Europe in the early 1950s. Initially, an amorphous-to-semicrystalline elastic material was created by polymerizing two straightforward ethylene, propylene, and olefins, in the occurrence of a Ziegler catalyst at roughly room temperature and atmospheric pressure.

Rubbers made of EPDM are amorphous terpolymers that contain a minor quantity of a nonconjugated diene. Unsaturation (double bonds) that are pendant or in a side chain are introduced by the diene. Dienes like Ethylidene Norbomene (ENB, Figure 1.2), 1,4-hexadiene (1,4-HD, Figure 1.3), and dicyclopentadiene (DCPD, Figure 1.4) are frequently utilised in the production of EPDM polymers. Vulcanization is made possible by the diene's provision of substituent unsaturation. Peroxide systems may also be used to crosslink EPDM polymers.



Figure 1.2 Structure of ENB

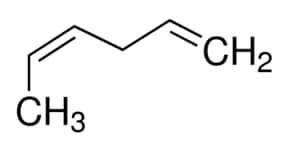


Figure 1.3 Structure of 1,4-HD

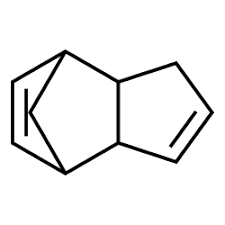


Figure 1.4 Structure of dicyclopentadiene

In EPDM, the monomer units are dispersed at random. The structure of EPDM is shown in Figure 1.5. In other words, the polymer has brief runs of both polyethylene and polypropylene interspersed between longer lengths of random copolymers rather than following a regular alternation sequence of ethylene and propylene units. Ethylidene norbomene is the termonomer that is used the most frequently because it is simple to include and has a higher vulcanization reactivity.

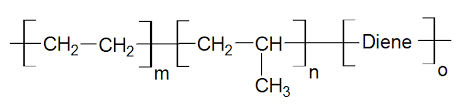


Figure 1.5 Structure of EPDM

The cure rate rises as the diene content does. A higher level of unsaturation allows for compounds with a quicker cure rate. These polymers work better when blended with diene rubbers. Some third monomers react more strongly with curing agents. In other words, at equal levels, ethylidene norbomene is faster than 1.4-hexadiene, which is faster than dicyclopentadiene.

The structure and makeup of EPDM polymers make them naturally resistant to ozone and the environment. Compounds don't require waxes, antiozonants, or any other specialised additives to be used in harsh weather or high ozone situations. EPDM materials offer good oxidation and heat resistance. For temperatures up to 100°C, no special compounding is necessary.

Other inherent characteristics of EPDM include resistance to water, aqueous solutions, resistance to polar fluids like ketones, specific esters, alcohols, and acetates, good low-temperature flexibility, high resilience over a fairly broad temperature range, and excellent electrical properties like low power factor, high dielectric strength, and corona discharge resistance and resistance to radiation.

Due to its low density (or specific gravity) and capacity to take significant loadings of filler and extender oils, it is possible to create competitive compounds for a number of applications.

The main applications of EPDM polymers are insulation, tubing, weatherstripping, automotive coolant system hoses, ignition cable, and isolator mounts. These materials are now widely utilised in a variety of moulded and extruded parts. Agricultural machinery, appliances, and the building and construction industries all have additional uses. The membrane sheeting for commercial building roofing is the most recent significant application.

**CHAPTER 2**

**LITERATURE SURVEY**

**2.1 Polymer blend**

Polymer blending allows for the creation of special features that cannot be acquired from individual components. In order to enhance physical and mechanical qualities, ease of processing, service life, and price, polymeric blends are frequently employed in rubber products (Vishvanathperumal et al. 2017; Vishvanathperumal et al. 2019). Arroyo et al. 2007 inquired about the impact of an Epoxidized Natural Rubber (ENR) and filler treatment on the shape and behaviour of NR nanocomposites. XRD was used to assess the kind and amount of clay dispersions in the filled nanocomposites. An exfoliated structure was formed in the presence of ENR, indicating that enough rubbery polymer has integrated into the interlayer gap. Rheological, mechanical, and swelling characteristics were used to examine the impact of clay in rubber composites. By using organoclay, a logical improvement in the nanocomposite's characteristics was seen. The extent of the silicate nanolayers' dispersion into the rubber matrix, the type of OC, and the compatibility with the elastomer have all been determined to have a significant impact on the characteristics of the compounds.

**2.2 Composites**

Das et al. (2008) have studied, Chloroprene Rubber (CR) vulcanizates' mechanical properties and crystallisation behaviour in relation to two chemically opposing types of nanofillers, namely Montmorillonite (MMT) and Layered Double Hydroxide (LDH), both in Pristine Layered Double Hydroxide (PLDH) and Organically Modified Montmorillonite (OMMT) forms. While a very small amount of pristine clays improves the physical characteristics of CR-based nanocomposites, MMT's reinforcing effectiveness is increased by organic modification more than LDH's. According to the results of the Dynamic Mechanical Analysis (DMA), MMT is discovered to increase the crystallisation tendency of CR chains, whilst LDH plays a function in restraining it. The DMA results also demonstrate that the presence of NC, even in tiny amounts, increases the modulus of vulcanizates based on CR, a phenomenon that is more prominent in the case of organically modified clays, particularly with OMMT.

Pal et al. 2009 studied the ENR and Cloisite 20A (C20A) nanocomposites were prepared by solution mixing. In blends of NR and High Styrene Rubber (HSR), the produced nanocomposites were included as reinforcing fillers together with CB of the Intermediate Super Abrasion Furnace (ISAF) and Semi-Reinforcing Furnace (SRF) types. The nanocomposites' morphology, curing traits, tensile, and thermal properties were examined. In comparison to ISAF N231 type of CB, the nanocomposites containing SRF N774 type of CB showed an increase in cross-link density, maximum torque (MH), and CRI. The nanocomposites containing SRF type CBs had better overall mechanical and thermal stability. The compounds in NR-HSR that contain ENR-Cloisite 20A (EC) have better barrier qualities than those that do not. Because more effective network chains are being formed as a result of the NC's greater reinforcing efficiency in the rubber matrix, EC with SRF N774 CB has demonstrated the lowest compression set value.

Kotal et al. 2010 stated that one of the most affordable ways to create high-performance polymeric materials for a variety of applications has been generally acknowledged as polymer blending in combination with nanofillers. The production of Polyurethane (PU) blended with NBR (PU/NBR; 1:1 wt./wt.) nanocomposites, which were subsequently described, employed Dodecyl Sulfate Intercalated Mg-Al-based Layered Double Hydroxide (DS-LDH) as nanofiller. TS (156%) and Eb (21%) exhibit the greatest improvements for 1 wt.% filler loading as compared to the neat PU/NBR blend. Comparing the nanocomposites to the pure PU/NBR blend, the Storage Modulus (G´) and Loss Modulus (Gʺ), thermal stability, and Limiting Oxygen Index (LOI) of this blend are higher. DS-LDH loading in PU/NBR increases according to Glass Transition Temperature (Tg) and swelling measures by up to 3 wt% when compared to pristine PU/Acrylonitrile Butadiene Rubber (NBR) or its other equivalent nanocomposites. High-performance polymer nanocomposites are formed because the advantages in mechanical, thermal, and flame retardancy qualities are significantly greater than those of the neat blend.

Alipour et al. 2011 were created five distinct NR/EPDM blends [(0/100), (25/75), (50/50), (75/25) and (100/0)] using a two-roll mill and 1, 3, 5, and 7 phr Cloisite 15A (organo-modified NC). On the curing characteristics, mechanical, rheological and microstructure properties of the nanocomposites, the impact of organo-modified layered nanosilicates and composition was investigated. The d-spacing consequences of X-Ray Diffraction (XRD) patterns revealed that polymer chains had been intercalated into the silicate layers. XRD data were immediately verified by Transmission Electron Microscopy (TEM) micrographs as well. The tensile modulus of 75NR/25EPDM, which contains 7 wt% NC, increased by about 40% according to the mechanical characteristics of the nanocomposites.

Mahmood et al. 2012 deliberated about the preparation and characterisation of brand-new Thermoplastic Polyurethane–Urea (TPUU) and Carboxylated Acrylonitrile Butadiene Rubber (XNBR) blends. Following an ultrasonication, mixtures of various compositions were created in tetrahydrofuran using a solution approach. Both 1H-Nuclear Magnetic Resonance (1H-NMR) and Fourier Transform Infrared-Attenuated Total Reflectance (FTIR-ATR) spectroscopy were used to analyse the chemical interaction between the two intrinsically immiscible blend phases. The blend morphology can be seen in Scanning Electron Microscopy (SEM) and TEM as a homogeneous dispersion of the minor TPUU phase in the XNBR. Additionally, compared to the unfilled hybrid elastomeric material, the introduction of 5 wt% of organomodified MMT NC significantly enhanced the mechanical characteristics.

Bijarimi et al. 2013 reported a melt blend of Poly(Lactic Acid) (PLA)/liquid NR with Cloisite C30B (C30B). Investigations were conducted into the mechanical, thermal, and morphological characteristics of PLA/liquid NR and nanocomposites. Results showed that adding C30B to the PLA/liquid NR blend enhanced Young's modulus and flexural modulus. Comparing PLA/liquid NR to nanocomposite containing 1% C30B, the Elongation at Break (Eb) increased noticeably, rising from 37.3% to 62.4%. However, as the Nanoclay (NC) content rose above 3%, the Eb and impact strength gradually dropped, indicating that the addition of NC altered the blend systems' strain response. The Tg values in relation to PLA were reduced by adding NC to the PLA/liquid NR blends. This phenomenon might be explained by the fact that there is more free volume in nanocomposite blend systems than in pure PLA. According to this work, it is possible to create blend-toughened nanocomposites with a higher modulus and greater thermal stability.

In the studies conducted by Razavi-Nouri & Karami 2014, Thermoplastic Elastomer (TPE) nanocomposites based on NBR and Poly(Ethylene-co-Vinyl Acetate) (EVA) were created with various weight ratios (20, 40, and 60 wt% of NBR) and 5 wt% of Organoclay (OC) in an internal mixer. Investigating the phase morphology and particle size was done using SEM. The two-phase structure of the unfilled blends' SEM pictures showed that the NBR domains were scattered throughout the EVA phase. The mix with 60 weight percent NBR, however, showed a co-continuous morphology. The nanocomposites' Young's modulus and yield stress were higher than those of the unfilled materials, but their Eb and stress at break were lower.

Jolfaei et al. 2015 have successfully fabricated the dynamically vulcanised TPE/OC nanocomposites from PP and EPDM. To enhance the dispersion of commercial OC, two kinds of compatibilizers such as EPDM-g-MAH and PP-g-MAH, were added. It was demonstrated that the compatibilized TPE nanocomposites' superior OC dispersion contributed to the mechanical characteristics' improvement.

Khodabandelou & Razavi Aghjeh 2016 examined the impact strength and crystallite structure of un-vulcanized and dynamically vulcanised PP/EPDM blends, as well as PP/EPDM/Carbon Nanotube (PP/EPDM/CNT) mix nanocomposites with and without Polypropylene-Grafted-Maleic Anhydride (PP-g-MAH). The outcomes demonstrated that the addition of EPDM improved the PP matrix's impact toughness by encouraging the cavitation and debonding of rubber particles. Superior impact strength was greatly attributed to the improved interfacial adhesion caused by dynamic vulcanization. CNT addition altered the PP matrix's crystallisation behaviour and further improved impact resistance in PP/EPDM blends.

Vishvanathperumal & Gopalakannan 2017 were created the EPDM/SBR/NC nanocomposites varied NC loadings and crosslinking systems (sulphur, DCP, and the combined) using an open mill mixer. On the swelling, water uptake and compression set behaviour, abrasion resistance, hardness and rebound resilience of EPDM/SBR blends, the impacts of NC loading and crosslinking systems were examined. At 23°C, the nanocomposites' mole percent absorption of aromatic, aliphatic, and chlorinated hydrocarbons was examined. The lowest solvent uptake of any penetrant and the lowest uptake of aliphatic hydrocarbons were both displayed by the peroxide-cured nanocomposites. In all the crosslinking systems, an increase in NC loading led to decrease in rebound resilience and water uptake as well as increase in hardness, compression set and abrasion resistance.

Vishvanathperumal et al. 2018, examined EVA/SBR blends with CB and NC treated with and without Mercapto-Modified EVA (EVALSH) by using a two-roll mill. Investigation on the effects of NC and CB (N330)/NC hybrid fillers on the morphological, mechanical properties, and abrasion resistance of EVA/SBR blends were also conducted. A gradual increase in ML, MH, ∆M, and CRI has been observed in EVA/SBR blends filled with 20 phr CB and varying NC loading, whereas values for Scorch Time (ts2) and Optimum Cure Time (t90) have been found to decrease with increasing content of NC. Measurements have been made of mechanical characteristics like TS, Eb, M100, tear strength, abrasion resistance and hardness. Up to a level of 6 phr, the TS of composites increased with increasing NC content before decreasing. The Eb of composites reduced with increasing NC content while hardness, tear strength and abrasion resistance has improved. The EVA/SBR composites' overall characteristics were improved by compatibility with EVALSH.

Vishvanathperumal & Gopalakannan 2019 assessed the impact of both NC and crosslinking systems (sulphur, DCP, and a combination system of sulphur and DCP) on mechanical qualities, abrasion resistance, compression set, and swelling properties of EPDM/SBR blends. The two-roll mill was used to prepare nanocomposites. The EPDM/SBR nanocomposites' mechanical characteristics, like crosslink density, Tensile Strength (TS), Eb, M100, hardness, and tear strength, were investigated. Cure investigation showed that NC not only sped up the curing processes but also causes the torque values (ML, MH, and ∆M) to noticeably increase, indicating that the crosslinking density of the nanocomposites have risen in the presence of NC. After reaching an NC content of 7.5 phr, the TS and M100 of EPDM/SBR nanocomposites start to decline for all cross-linking systems. The NC content had a direct positive correlation with Eb, hardness, compression set and tear strength.

Vishvanathperumal et al. 2020, concluded that the mechanical characteristics of EPDM/SBR mixes were improved via NC. A tension test was used to determine the experimental data on the SS behaviour of EPDM/SBR blends under various NC loading. Strain energy functions were used to characterise the nonlinear mechanical behaviours of rubbers, ensuring that rigid body motions are not a factor in the constitutive law. The correct Cauchy-deformation Green's tensor or Green's strain tensor is necessary for the correct operation of mathematical models like the Mooney-Rivlin model, which is dependent on the presence of strain energy density functions. The rubber material constants are discovered by fitting the experimental data to the Mooney-Rivlin model. The crosslinking density is calculated using these constants. A comparison of the Finite Element Analysis (FEA) and experimental SS behaviour of a uniaxial tension test under various NC loads is shown.

Vishvanathperumal & Anand (2020) prepared and looked into hybrid EPDM/SBR composites reinforced with NC and Nanosilica (NS). On the mechanical characteristics of the EPDM/SBR hybrid composites, the interaction between NC and NS was investigated. The EPDM/SBR hybrid composites' swelling resistance, microstructure, mechanical properties (TS, Eb, M100, and tear strength), physical properties (hardness and rebound resilience), abrasion resistance, and compression set, were all assessed. The micro-structural, mechanical, and other properties of the nanocomposites were greatly influenced by the NS and NC concentrations. The nanocomposites with 7.5 phr of NC and 4 phr of NS clearly exhibit a maximum mechanical property in addition to their abrasion and swelling resistant properties. The EPDM/SBR hybrid composites with NC and NS content in particular exhibit the best mechanical and abrasion resistance qualities.

Vishvanathperumal & Anand 2021 prepared EPDM/SBR-NS nanocomposite. On a two-roll mill, the NS with EPDM/SBR (0–10 phr) was mixed. It looked into and studied the effects of NS (0–10 phr), crosslinking systems (sulphur, DCP, and mixed), and Silane Coupling Agents (SCAs) like Bis(triethoxysilylpropyl)–Tetrasulphide (TESPTS) on the curing and mechanical properties, rebound resilience and compression set, abrasion, swelling resistance property, and morphological characteristics of the EPDM/SBR–NC nanocomposites. It was obvious that as the amount of NS particles in it increased, so did the wear, mechanical, and swelling qualities. In general, the qualities of the sulphur-cured rubber composite are better than those of other crosslinking agents like peroxide and mixed systems. The characteristics of the nanocomposites were more significantly improved by the addition of SCA. It was evident that the rubber composite containing SCA had a greater influence of NS concentration than nanocomposites without it.

Vishvanathperumal & Anand 2022 explored the impact of NS particles on the EPDM/SBR-NS compound. Using an open mill mixer, the composite EPDM/SBR with and without SCA was treated. The NS particles were created in a lab and employed as the reinforcing ingredient in composites made of EPDM and SBR rubber. The composites were thoroughly researched and studied in terms of their curing and mechanical characteristics, hardness and rebound resilience, swelling and abrasion resistance, and compression set. The presence of NS particles increased the mechanical properties, swelling and abrasion resistance, hardness and compression set qualities of the EPDM/SBR rubber composites.

Ganeche et al. 2022, the mechanical characteristics, abrasion and swelling resistance of HNTs (ranging from 2 to 10 phr) filler filled EPDM/SBR were studied. Using electric universal tensile testing apparatus in accordance with ASTM D-412, the following parameters were assessed at room temperature: TS, M100, Eb, and tear strength. The findings demonstrated that increasing HNT content increases crosslink density, hardness, tensile strength, and tear strength. Field-Emission Scanning Electron Microscopy (FESEM) was used to examine the surface morphology of material that has undergone tensile fracture. According to FE-SEM data, the surfaces of rubber nano-composites loaded with HNTs were the roughest.

**2.3 EPDM/NBR blends**

For their many applications, elastomers based on more network predecessors have emerged as a technologically significant material. Their physical and chemical characteristics suggest them as suitable technical materials for electric insulators, the chemical sector, the manufacture of automobiles and aircraft, among many other applications. To get the best qualities out of each component, NBR and EPDM rubber were blended. The ozone resistance and heat ageing properties of NBR are weak, despite its great resistance to swelling in oils and solvents (El-Nashar & Turky 2003; Mohamed et al. 1998). Because its unsaturation sites in the side chain rather than the backbone, EPDM has good heat ageing and ozone resistance but low resistance to solvent (Azima & El-Sabbagh 2001, Ismail et al. 2003). Many researchers are interested in blending such two polymers to create a material that has good mechanical properties and is resistant to ozone, heat ageing, oil and solvent swelling. Thus, the end result of this combination will have exceptional mechanical strength, heat resistance, oil resistance, and ozone resistance. For example, automotive radiator hoses, automobile brake hoses, transmission belts, motor mounts, sheets, conveyor belts, and rolls all need such qualities and might be produced using this method.

Botros and Tawfic (2005a) analysed that the mechanical qualities of EPDM/NBR vulcanizates produced by sulphur curing systems were superior to those of DCP vulcanizate. In comparison to sulphur vulcanizates, the DCP curing technique produced an EPDM/NBR vulcanizate with improved heat stability and less weight swell in braking fluid and motor oil. After combining EPDM and NBR, the rubber vulcanizates' fatigue resistance was significantly increased. The EPDM/NBR blend ratios of 50/50 and 75/25 produced the highest number of fatigue cycles before failure. After thermal ageing for seven days, all EPDM:NBR blend ratios studied deviated positively in terms of TS and Eb from the additive rule's straight line, demonstrating the effectiveness of EPDM and NBR's co-curing. The highest aged mechanical qualities were found in the 25/75 EPDM/NBR blend vulcanizate, which also had exceptional heat stability.

Botros and Tawfic (2005b) stated that the NBR content in the EPDM-NBR mix grew, as the MH, M100, and weight swell in motor oil decreased. The 50/50 EPDM-NBR blend, compatible with epoxidized EPDM, exhibited good thermal stability with the highest positive deviation of TS and Eb, after 7 days of ageing, from the additive rule. It also possessed the highest TS and the longest Eb throughout the thermal ageing or the UV exposure periods. The 25/75 EPDM/NBR mix had the maximum TS and the longest Eb during the thermal ageing or UV exposure period. It was compatible with aminated epoxidized EPDM. After 7 days of ageing, the 75/25 and 25/75 blends both deviated positively from the additive rule in terms of TS and Eb.

Botros and Tawfic (2006) made MAH-g-EPDM and evaluated it as a compatibilizer for EPDM/NBR blend and processed by a two-roll mill. The co-curing of EPDM rubber and NBR rubber was improved by MAH-g-EPDM. The EPDM/NBR blend with ratio of 25/75 wt./wt., compatible with MAH-g-EPDM, has the highest mechanical qualities of all the blend ratios studied. It also has the best heat resistance and UV stability, with a slightly weaker resistance to motor oil swelling.

Eid and El-Nashar (2006) unequivocally stated that the best mechanical properties can be attained by mixing EPDM and NBR in the ratio of 75:25. When loaded with various silica contents, the electrical and mechanical characteristics of such a sample (EPDM/NBR ratio is 75/25) were also investigated. This analysis has led us to the conclusion that silica concentrations of 50–60 phr are the most promising for both mechanical and electrical applications. EPDM/NBR multiphase blend's electrical and mechanical qualities are improved by the addition of Polyvinyl Chloride (PVC) as a compatibilizer. The inclusion of 10 phr PVC as a compatibilizer raises all mechanical properties (σR, σB, strain at rupture percent and strain at yield percent).

Manoj et al’s (2010) research studies examined the cure characteristics and mechanical features of filler reinforced EPDM/NBR blend systems in terms of filler loading and filler types. The ISAF filled sample was determined to have the highest MH levels. All of the filled systems showed increased TS as compared to the unfilled system. The ISAF filled system demonstrated the highest TS and tear strength among the various filler materials employed. With reference to the type of fillers, the barrier characteristics of CB filled EPDM/NBR vulcanizates utilising benzene, toluene, and xylene as penetrants have been investigated. When compared to the corresponding unfilled blends for a particular blend ratio, it has been discovered that the filled samples show better resistance to the uptake of the three organic solvents. The solvent uptake occurred in the following order for the three different forms of carbon black: SRF > HAF > ISAF filled samples. This is explained by the fact that as the size of the CB particles utilised decreases, there is a greater constraint on the total chain mobility and flexibility of polymeric chains. For a particular system, it can be seen that the swelling coefficient gradually increased as filler particle size grew.

Manoj et al. (2011) also studied in detail about the impact of filler loading and filler types on the cure characteristics and mechanical features of filler reinforced EPDM/NBR blend systems have been examined. The filler loading has been observed to boost the MH values. With a rise in the percentage of black particles in the mix system up to a threshold level, the t90 values declined while the CRI increased. The 15 phr loaded ISAF system has been determined to have the maximum scorch safety. For a specific blend ratio, it has been observed that filled samples exhibit less solvent uptake than the unfilled sample. Due to improved filler reinforcement, blends loaded with ISAF showed the lowest liquid uptake among black filled systems. All of the filled systems showed increased TS as compared to the unfilled system. The ISAF-filled system demonstrated the highest TS and tear strength among the various filler types employed. This has been explained by ISAF black's improved reinforcing effectiveness.

Jovanović et al. (2013) as the CB component in the NBR/EPDM rubber blend increases, the MH and ML, ∆M, and t90 values rise while the ts2 and CRI values fall. With a 70 phr CB, NBR/EPDM composite has a minimum Eac value of 5.5 kJ/mol and a maximum Activation Energies of Reversion (Ear)/Activation Energies of Cross-linking (Eac) ratio (66.27). The NBR/EPDM/CB composite has a maximum value for Ear (407.1 kJ/mol) and a CB content of 90 phr. For all NBR/EPDM rubber blend composites, the values of hardness and TS increase but Eb values drop as CB concentration increases. Mechanical property values fell with thermal age, and a degradation process could be seen. It is possible to notice two different Tg values for CB reinforced composites made of EPDM/NBR rubber mixtures.

**2.4 EPDM/NBR nanocomposites**

Ma et al. 2008, melt mixing was used to create rubber/OC nanocomposites. While nonpolar unsaturated rubber (EPDM) made it difficult to generate intercalation structure without the aid of other additives, polar or unsaturated matrices (such as NBR and SBR) could easily penetrate OC layers. When Stearic Acid (SA) was present, an intercalated structure for the EPDM system was seen in composites made of SA and OC. Clay dispersion in nonpolar saturated rubber matrix was significantly worse than in polar or unsaturated matrix, according to TEM observation. By contrasting the effects of Butyl Rubber (IIR)/OC and Bromobutyl Rubber (BIIR)/OC systems, the similar effect of polar matrix was established. Additionally, utilising OC pretreatment by SA (S-OC), the examined nanocomposites' clay dispersion was obviously improved because SA was intercalated into the OC interlayers. The intercalation structure was particularly visible in the nonpolar saturated EPDM system. TS and stresses at low strain of NBR and SBR based nanocomposites with S-OC were significantly improved relative to the corresponding nanocomposites using OC; however, with EPDM nanocomposite, using S-OC, only TS were improved but the stresses at low strain were nearly the same, which should be related to the different interfacial force between OC and different rubber matrices.

Ghassemieh (2009) looked at the qualities of EPDM and NBR as seal materials were affected by the addition of various concentrations of NC. Evaluate the impact of the inclusion of the NC, properties including TS, modulus at various extensions, Eb, compressive set, permeability and hardness as well as abrasion resistance are investigated. Results show that adding NC to some compositions may marginally diminish the rubber's strength. However, a more stable modulus at various strains is offered, the rubber's hardness is maintained and somewhat increased, and the permeability is decreased in both rubbers, with an especially notable decrease in EPDM, which is desired to lessen the effect of explosive decompression. The compression test also demonstrates that the NC enhances the rubbers' ability to operate well under compression, which is crucial for seal applications. The XRD analyses demonstrate the exceptional quality of the NC dispersion in the NBR samples. The dispersion in the EPDM samples has to be improved.

Gao et al. 2010, created and conducted a thorough investigation on a number of EPDM/NBR composites with short aramid fibre reinforcement and a certain Fiber Orientation Angle (β) between the short fibre orientation and the ablative surface. The impact of β on the composites' thermal stability and resistance to abrasion under an oxyacetylene flame is thoroughly examined. The char layer's porosity decreases as its hardness increases with an increase in β. Due to the increased heat conductivity of the char layer after the composites were carbonised, the Pyrolysis Rate (Rp) and Carbonization Rate (Rc) are accelerated. The rate of mass loss during the pyrolysis reaction increases as thermal degradation accelerates. However, as β increases because of the increased hardness of the char layer, the Erosion Rate (Rl), which is mostly brought on by the mechanical ablation of the high-speed heat flux, gradually decreases. The Mass Loss Rates (Rm), which are dominated by the two types of ablation techniques in opposition as β increases, first decline and then gradually increase.

Gomaa et al. 2010, using Positron Annihilation Spectroscopy (PAS), blends of EPDM and NBR with and without the Polyvinyl Chloride (PVC) compatibilizer were studied. The PAS results indicate that the size of the Free-Volume Hole (Vf), its Fraction Percentage (f%), and the S-parameter have a linear relationship with a negative slope as a function of the wt.% of NBR, indicating an increased interaction between the blend's constituents in the presence of a compatibilizer. Additionally, for blends without a compatibilizer, Vf, f%, and S-parameter demonstrate positive and negative divergence from the linear additive rule, indicating a multiple phase system. The EPDM/NBR blend is incompatible as seen by the widening in Vf size distribution caused by increasing the weight fraction of NBR in the blend without a compatibilizer. According to the filling effect of SiO2 on the free-volume characteristics of EPDM/NBR (75% /25%), SiO2 addition up to 50 phr is the concentration that offers the most promise for electrical applications. Additionally, correlations between the outcomes of the mechanical and electric systems and the size and percentage of Vf were determined.

Samaržija-Jovanović et al. 2011, studies focused on studying the various network precursors, such as NBR, EPDM, and their blend (NBR/EPDM) reinforced with NS, which were used to create nanocomposites. Mechanical properties before and after thermal ageing, rheometric parameters, curing kinetics, and morphology of these materials are discussed. Using a rheometer with an ODR, the network formation process was detected by the variable torque durations, and the curing kinetics and the rheometric properties of nanocomposites were determined. Using an air-circulating oven, the mechanical characteristics of the elastomeric composites were assessed both before and after thermal ageing. FTIR was used to identify the precise interactions between the rubber and filler. The surface morphology of fractured rubber was studied using SEM. The results showed a relationship between the mechanical properties of the sample EPDM/NBR = 20/80 and the calculated Activation Energies of Cross-linking (Eac) and Reversion (Ear). Maximum TS values and synergism were noted for this blend. Furthermore, SEM analysis revealed that this blend had a very consistent shape. The SiO2-reinforced EPDM/NBR rubber mixes were revealed to be immiscible by the DSC curves.

Ersali et al. 2012, in view of conducting an in-depth study, created three different kinds of EPDM/NBR blend-based nanocomposites: one kind contains C20A in the EPDM phase, the second includes C30B in the NBR phase, and the third category contains both C20A in the EPDM phase and C30B in the NBR phase at the same time (called EN-xAB). For all varieties of nanocomposites, XRD and TEM methods revealed intercalated or exfoliated structures. The EPDM/NBR/Cloisite 20A/Cloisite 30B (EN-xAB) nano-composites were found to have the roughest surfaces, per the SEM findings. The samples' curing properties revealed that EN-xAB samples had the shortest ts2 and t90 and the highest crosslink densities. The samples that contained both types of OCs instantaneously had the best mechanical properties, according to a comparison of the nanocomposites' mechanical characteristics.

Abd-El-Messieh et al. 2013, attempted to study the Permittivity (ε') and Dielectric Loss (ε'') of 50/50 NBR/EPDM mixes by taking measurements at various frequencies and room temperature (30°C). These blends included three different forms of nanoscaled CB that were examined by TEM. The electrical conductivity and dielectric properties both experienced a substantial rise at a specific CB concentration. This growth is due to the network creation, which was verified by SEM analysis and the use of several conductivity models. The composites under scrutiny also underwent a mechanical investigation.

Hoikkanen et al. 2015, Using a melt mixing technique, polar NBR and nonpolar EPDM mixes were created in the influence of MWCNTs. The masterbatch dilution and direct mixing method were the two mixing techniques employed to create MWCNTs loaded EPDM/NBR mixes. To understand how MWCNTs disperse, several physical and mechanical as well as morphological aspects are investigated. It was determined that the preparation procedure affects how evenly the MWCNTs disperse in the various rubber phases, and that the degree of dispersion and distribution of the MWCNTs in the dual phases determines the attributes of these blends.

In the research conducted by El-Gamal et al. 2017, different types [(N326-HAF) and (N774-SRF)] and proportions of CB fillers were added to blends of EPDM/NBR. Investigations were made into the mechanical characteristics of EPDM/NBR rubber mixes both loaded and unloaded with various CB ratios. When loaded and unloaded with CB, the mix of 75% EPDM and 25% NBR was found to have the maximum TS and Eb. The morphology as seen by SEM was connected to the observed changes in the mechanical characteristics of the mixes. Investigated were the variations in electrical resistivity of rubber blend composites during compression. The hypothesis used to explain the experimental results was that an external pressure causes an increase or decrease in the blend composites' resistivity depending on whether effective conductive routes are created or destroyed, respectively.

Basha et al.’s 2018, study provides a thorough characterization of the cure properties, degradation kinetics and cure kinetics of nanocomposites made from an EPDM-NBR blend and filled with NC. Because EPDM rubber has the best level of radiation protection among elastomers, it is frequently utilised in nuclear power plants. However, EPDM components that are subjected to settings with the combined effects of radiation and hydrocarbons malfunction because of its weak resistance to aliphatic hydrocarbons. Combining EPDM with a hydrocarbon-resistant polymer, such as NBR, can increase the lifespan of such components. Qualities like hydrocarbon and radiation resistance, and mechanical properties can be increased by adding nanoparticles like NC to the rubber blend. As a function of the NC content, the degradation and cure kinetics of the rubber blend nanocomposites were examined. For the degradation and cure reactions, the order of the reaction was discovered to be one. Additionally, the rate constant of the degradation and cure reactions increased with NC concentration before decreasing as NC content increased. The observed results were connected to the nanocomposites' morphology.

Ashok et al. 2018, by utilising maleic anhydride as a compatibilizer during melt compounding in an internal mixer, created nanocomposites of blends based on EPDM and NBR. OMMT was added in varied amounts to the blends (0, 2, 4, 6 and 8 phr NC). In a hydraulic compression press, the nanocomposites were vulcanised at 170°C for the t90 to produce sheets that were about 2mm thick. NC intercalation and exfoliation at lower concentrations, as well as a small amount of agglomeration at higher concentrations, were confirmed by morphological investigation using XRD and AFM. With increasing NC content, the barrier properties—characterized by diffusion, sorption, and penetration coefficients—also improved. However, due to the production of agglomerates at greater NC concentrations, both barrier and mechanical properties showed a minor reduction.

Paran et al. 2020, Using a two-roll mill mixer, hybrid rubber nanocomposites based on EPDM, NBR, various concentrations of GNPs, and Graphene Oxide Nanoplatelets (GONPs) were created. SEM and TEM micrographs were used to carefully examine the impact of 1.5 wt% GNPs and GONPs on the micro-structure of the vulcanised EPDM/NBR/GNPs/GONPs compounds. Furthermore, on the basis of tensile tests, DMTA, and the Rubber Processing Analyzer (RPA), their mechanical and rheological properties as well as stiffness were discussed. When compared to the fracture surface of the EPDM/NBR compound blend, SEM micrographs from rubber nanocomposites containing 1.5 wt% of either GNPs or GONPs showed a rough fracture surface. The tensile test analysis for the nanocomposites comprising both kinds of platelet-like nanoparticles showed a 100% improvement in the TS. The results of experimental testing were in good agreement with theoretical analysis. With the addition of nanoplatelets to the EPDM/NBR combination, DMTA measurements showed a greater storage modulus of up to 1.59 MPa. Dielectric spectroscopy revealed that adding both GNPs and GONPs to the EPDM/NBR matrix increased conductivity by almost 7 times.

Nihmath & Ramesan 2020, Chlorinated NBR (Cl-NBR) and Chlorinated EPDM (Cl-EPDM) elastomeric mix nanocomposites were created with varying amounts of Hydroxyapatite (HA) nanofillers. FTIR, XRD, TEM, SEM, and TGA were used to evaluate the blend nanocomposite's production. As a function of nanoparticle loading, the fire resistance, dielectric behaviour, and petroleum fuels diffusion through the mix membrane had been examined. The rubber mix's HA absorption band was visible in the FTIR, indicating a successful interaction between the rubber mix and the HA nanofiller. The use of nanoparticles reduced the amount of amorphous rubber mix, according to XRD plots. The homogeneous dispersion of nanoparticles in the rubber mix was visible in TEM and SEM photos. According to TGA data, Cl-NBR/Cl-EPDM blend had a worse thermal stability than rubber mix nanocomposite. The increased fire resistance of mix composites produced by lowering the oxygen index proved that HA nanoparticles were a great fire-retardant additive for a mix of Cl-NBR and Cl-EPDM. The strengthening of the mix matrix by the HA nanoparticles was evidenced by an increase in dielectric constant with nanoparticle loading. The mix nanocomposite's solvent absorption decreased in the HA-loaded sample by up to 7 phr. With the penetrant molecules' growing molecular size, a decrease in solvent uptake was also noted. The diffusion mechanism was abnormal in some way. Overall, the sample containing 7 phr of the rubber mix loaded with HA demonstrated a notable improvement in flame resistance, dielectric characteristics, and minimal solvent uptake.

Azizli et al. 2020, made use of a two-roll mill to create hybrid rubber nanocomposite blends based on (80/20) XNBR/EPDM, which contained 5 phr Cloisite 10A (C10A), Cloisite 15A (C15A), Cloisite 20A (C20A), Cloisite 25A (C25A), Cloisite 93A (C93A), Cloisite 30B (C30B), CB, and SiO2 nanofiller, as well as two varieties of EPDM-g-MA and ENR-50, all of which contained 5 phr. The curing of the blends was studied using a rheometer test, the mechanical strength of the nanocomposite was studied using a tensile test, and the morphology of the created nanocomposite blends was assessed and observed using XRD, FESEM, and TEM tests. The rheometer test findings demonstrate that the C30B nanofiller decreased in comparison to other nanofillers while also boosting torque and t90. According to the XRD test results, the ENR-50 compatibilizer considerably enhanced the openness of the layers by 2.21 nm when used with the C30B nanofiller compared to other nanofillers and when used with the EPDM-g-MA compatibilizer. The improved distribution of C30B nanoparticles in the XNBR/EPDM sample with ENR-50 compatibilizer was validated by the results of FESEM and TEM. Additionally, XRD results demonstrated that C30B's nanofiller layers opened up more than those of the other nanofillers. Additionally, the XNBR phase is the location of all nanoparticles. Tensile test results revealed that when compared to other nanocomposites, the modulus and TS of the nanocomposite containing the C30B nanofiller and ENR-50 compatibilizer increased.

**2.5 Summary**

The primary derivative of rubber is rubber compound. Rubber mixes are widely used in a variety of applications; this statement is especially true for the production of tyres. Because of its excellent mechanical qualities and remarkable resistance to ageing, ozone, UV rays, and weathering, EPDM rubber is one of the most often used synthetic elastomers. As a result, it is used to make gaskets, belts, O-rings, window profiles, and waterproofing membranes. Insufficient resistance to hydrocarbons, including oils, kerosene, aromatic chemicals, gasoline, and halogenated solvents, is the fundamental drawback of EPDM rubber. NBR, on the other hand, has a low resistance to ozone and heat ageing but is quite resistant to swelling in oils and solvents. In the automotive sector, NBR rubber is used to make gasoline hoses, seals, and O-rings. There is no research on these systems in the open literature, despite the potential benefits of employing EPDM/NBR blends for O-rings and waterproofing membranes and the recent Indian standards for these uses in the automotive industry and outdoor strip. Depending on service requirements and tyre components, customised elastomer mixes are also used in addition to blends of the conventional rubbers.

CB is almost always used as a filler in rubber formulations. Rubber is strengthened, given more volume, has its physical characteristics improved, and the vulcanization process is made stronger due to CB filler. The rubber compound's output can be used to create glove material, motorised vehicle tyres, and shoe bottoms. By combining rubber with chemical compounds, completed rubber products are created. Additionally, milling is done at a temperature and for a duration that corresponds to the type of rubber and its intended use. To create a uniform mixture of rubber and chemicals, compounding is used. The history of tyres is connected with the use of CB. Despite losing some ground to other reinforcing fillers like silica and nanoclay, it is still the most well-liked and often utilised reinforcing filler because to its unmatched efficacy. But the main characteristics of CB are often governed by surface activity, surface area, particle size, and structure, and they are frequently connected. Enhancing strength and strength-related qualities, such as hardness, abrasion resistance, and Young’s modulus, as well as lowering material prices and streamlining the manufacturing process, is the main goal of reinforcement.

Rubber-GO nanocomposites have been developed and examined in this study. Highly distributed GO nanocomposites are achieved by combining two primary aspects. To achieve more GO dispersion in the polymer matrix, the first aspect includes the compatibility between the polymer and GO. Polar polymers make it simpler for GO to distribute than non-polar polymers do. Typically, a polymeric compatibilizer is present in the formulations of GO and polar polymeric systems. The second aspect is the technique used to create nanocomposites. In situ polymerization intercalation, solution intercalation, melt intercalation, and eventually co-coagulation of rubber latex and clay aqueous suspension have all been used to prepare rubber/GO nanocomposites. The compatibilizer should be more polar than the matrix polymer while yet being compatible with it. In this investigation, melt mixing was used to incorporate GO into an EPDM/NBR blend in order to achieve uniform GO dispersion. Investigations have been done on the modifications that the curing study, morphological, mechanical, swelling, and abrasion properties produced.

**CHAPTER 3**

**MATERIALS AND EXPERIMENTAL**

**3.1. Materials**

The materials utilized in this study, along with their specifications, suppliers, and locations, are detailed as follows: EPDM (KEP-270) was procured from Swastik Group of Companies, Gujarat, India, featuring a maximum volatile content of 0.75%, a specific gravity of 0.86, an ENB content of 4.5 wt.%, an ethylene content of 56.5 wt.%, and a Mooney viscosity of 71 ± 6 (ML1+4, 125°C). NBR (JSR N230SL) was sourced from Hind Elastomers Pvt. Ltd., Mumbai, India, with an acrylonitrile content of 35% and a Mooney viscosity of 42 ± 1 (ML1+4, 100°C). Graphite flake, characterized by a carbon content of 99%, a particle size of approximately 325 mesh (≥99%), and a molecular weight of 12.01 g/mol, was obtained from Sigma-Aldrich, Puducherry, India. Graphene oxide (GO) was synthesized using a modified Hummers' method. Industrial-grade curing agents, used without further purification, were supplied by Vignesh Chemicals, Chennai, India. Additional chemicals, including NaNO₃, H₂SO₄, KMnO₄, H₂O₂, HCl, 3-Aminopropyltriethoxysilane, N,N-Dimethylacetamide (DMAc), Dibutyltin dilaurate (DBTDL), and 4,4-Diphenylmethane diisocyanate (MDI), were also acquired from Sigma-Aldrich, Puducherry, India.

**3.2. Preparation of nanocomposites**

The GO-EPDM/NBR composites were prepared following the formulation outlined in Table 1. Initially, GO fillers were incorporated into the EPDM/NBR gum using an open mill mixer to ensure uniform dispersion. The compounding process involved blending EPDM at 70 parts per hundred rubber (phr) with NBR at 30 phr. Modified GO was added at varying loadings of 0, 1, 2, 3, 4, 5, 6, 7, and 8 phr. Essential curing agents were also included in the formulation, comprising sulfur (2.5 phr), zinc oxide (ZnO, 5 phr), stearic acid (2 phr), benzothiazyl disulfide (MBTS, 1 phr), and tetramethylthiuram disulfide (TMTD, 1 phr). The thoroughly compounded mixtures were subsequently molded using hydraulic pressing at a temperature of 160°C and a pressure of 30 MPa for the optimum curing time (t90), ensuring complete vulcanization. The final cured composites, with a sheet thickness of approximately 2 mm, were then cut into ASTM-standard specimens for mechanical property evaluations, including tensile, hardness, and elongation tests [73].

Table 3.1 Formulation for EPDM/NBR/NC rubber blends

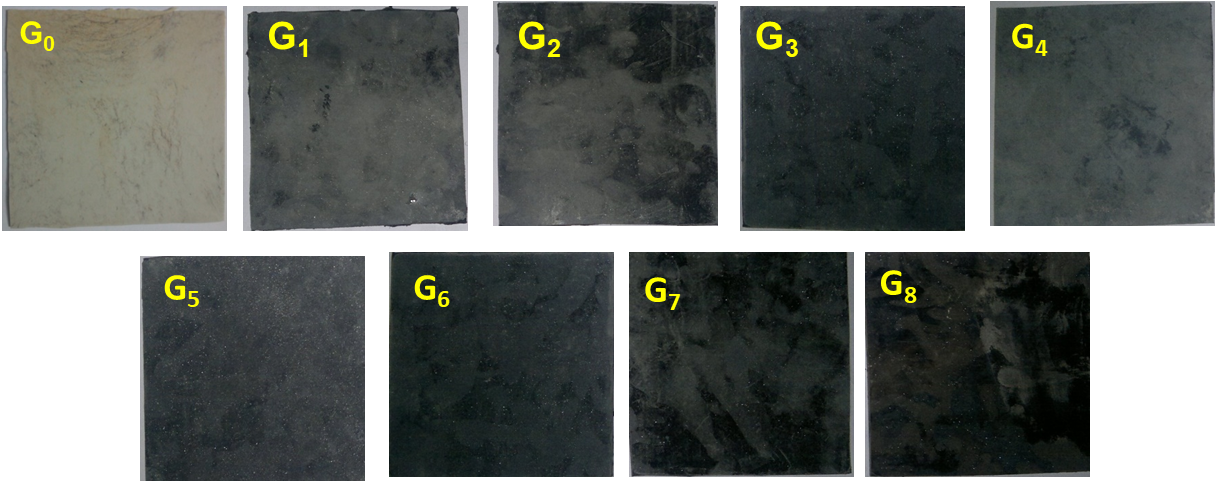
|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Sample code | EPDM | NBR | GO | Zinc oxide | Stearic acid | MBTS | TMTD | Sulphur |
| GO0 | 70 | 30 | 0 | 4 | 1.5 | 1.2 | 1 | 2.5 |
| GO1 | 70 | 30 | 1 | 4 | 1.5 | 1.2 | 1 | 2.5 |
| GO2 | 70 | 30 | 2 | 4 | 1.5 | 1.2 | 1 | 2.5 |
| GO3 | 70 | 30 | 3 | 4 | 1.5 | 1.2 | 1 | 2.5 |
| GO4 | 70 | 30 | 4 | 4 | 1.5 | 1.2 | 1 | 2.5 |
| GO5 | 70 | 30 | 5 | 4 | 1.5 | 1.2 | 1 | 2.5 |
| GO6 | 70 | 30 | 6 | 4 | 1.5 | 1.2 | 1 | 2.5 |
| GO7 | 70 | 30 | 7 | 4 | 1.5 | 1.2 | 1 | 2.5 |
| GO8 | 70 | 30 | 8 | 4 | 1.5 | 1.2 | 1 | 2.5 |



**Figure 3.1 Open mill mixer**

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**Figure 3.2 Hydraulic press**

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**Figure 3.3 EPDM/SBR nanocomposite samples**

**3.3. Characterization**

**3.3.1. Rheometric Characteristic**

The rheological test of unvulcanized rubber samples was carried out according to ASTM D-2084 by the oscillating disc rheometer under 160oC, oscillation arc 0.5o and 100 cycles per minutes (1.66 Hz) testing conditions. Minimum torque (Ml), maximum torque (Mh), scorch time (tS2 – the period in the curve in which the minimum torque increase by two Mooney units [38]), optimum curing time (t90 – the period in the curing curve where 90% increase in torque [38]) and cure rate index (CRI) are measured. CRI was calculated using the equation.

CRI =

**3.3.2. Mechanical Properties**

Tensile and tear properties were performed at 23oC respectively, on dumbbell- and un-nicked 90o angle test-shaped specimens according to ASTM standards D 412-C and D 624-B on a universal testing machine - series 7200 by Dak System Inc (model: T-72102) at a crosshead speed of 500 mm/min. Hardness is measured with a Shore-A Durometer as per ASTM D-2240. The average of five readings is reported for each sample. Tensile (tensile strength, elongation at break and 100% modulus) test samples, tear and hardness test samples were heated at 100oC in an air circulating oven for 96 hours. The sample is then cooled down for 30 minutes and conditioned at room temperature. The dimensions of each sample were measured and tested.

**3.3.3. Crosslinking Density**

The degree of crosslinking density has been calculated by the following equation [39, 66-67].

(2)

where Mc is the molar mass of the polymer between crosslinks. Molar mass between the crosslinks of the nanocomposites has been calculated from the Flory-Rehner equation [39, 65-67].

(3)

where, ρp is the density of the polymer, Vs is the molar volume of the solvent (106.3 mL/gmol), Vr is the volume fraction of polymer in the solvent-swollen filled compound, χ is the interaction parameter of the polymer (0.3) [68], and Vr is given by following equation [69]

(4)

where, Qm is the weight swell of the EPDM/SBR nanocomposites in toluene.

**3.3.4. Rebound Resilience**

The rebound resilience of the nanocomposites was carried out according to the ASTM D 2632. In this method, a plunger suspended from a given height above the specimen was released and the rebound height was determined. The ratio of the rebound height and the original height is stated to as the rebound resilience and expressed as a percentage. The test was performed on the vertical rebound tester as resiliometer. The average of five readings is reported for each sample.

**3.3.5.Abrasion Resistance**

Abrasion resistance of the rubber nanocomposites in terms of volume loss was determined on DIN abrader (Zwick Abrasion tester, model 6102) according to DIN 53516, ASTM D5963 and ISO 4649. The cylindrical shape specimen (10 ± 0.2 mm in diameter and 12 ± 0.2 mm in thickness) was placed in the starting position of the DIN abrader testing machine. After, the specimen was abraded across the emery paper of grade 60 at a constant force of 10 N and at a constant speed of 0.32 m/s. Then an abraded distance of 40 m was reached, the test specimen was automatically elevated from the DIN abrader emery paper. The DIN volume loss or abrasion loss of the rubber nanocomposites was calculated using Equation (3). Minimum of 5 specimens per sample was tested and recorded and then the average value was taken as the abrasion loss for each sample.

(3)

where Δm is a mass loss (mg); ρ is a density (mg/mm3); S0 is a value of nominal abrasive power (200 mg); S is an average abrasive power (mg).

**3.3.6. Compression set measurements**

Compression set test was carried out according to ASTM D395 [40]. The test sample is placed in the middle of the rectangular plates of the compression device by the spacers arranged on each side of it, allowing adequate clearance for bulging of the rubber when a compressive load is applied. The bolts tightened; therefore they are drawn together uniformly until in contact with the spacers. The percentage of compression working is 25% of its original thickness. Then the assembled device was placed at 23oC, 70oC, and 100oC for 1 day and 23oC for 2 and 3 days in an air circulating oven. After completion, compression device is taken from the air circulating oven and then the test specimen removed instantaneously and allowable to cool for 30 min. The final thickness is measured by an electronic digital Vernier caliper with 0.01 mm accuracy. The compression set was determined using formula

Percentage of compression set, C% = X 100 (5)

where t0 is the original thickness of the specimen, t1 is the specimen thickness after removed from the compression device and ts is the spacer bar thickness which is used.

**CHAPTER 4**

**RESULTS AND DISCUSSIONS**

The mechanical properties of the composites, including tensile strength (TS), elongation at break (EB), stress at 100% elongation (M100), and tear strength (TAS), were evaluated. Figure 4.1 shows the TS of EPDM/NBR composites with different GO concentrations. TS increases with GO loading up to 5 phr, after which a decrease is observed. The initial improvement in TS up to 5 phr can be attributed to GO's reinforcing effect, as its high mechanical strength and stiffness enhance the load-bearing capacity. Well-dispersed GO sheets within the polymer matrix act as bridges between polymer chains, promoting efficient load transfer and strengthening the composite structure. At lower GO loadings (up to 5 phr), the filler disperses effectively within the polymer matrix, enabling efficient stress transfer and strengthening mechanisms. This optimal dispersion enables effective reinforcement, which contributes to the observed increase in TS. Conversely, beyond the threshold of 5 phr, further incorporation of GO may lead to agglomeration of filler particles within the matrix. Agglomerated GO sheets can act as stress concentration sites, disrupting the uniform stress distribution and weakening the mechanical integrity of the composites. Additionally, increased filler-filler interactions may restrict polymer chain mobility, resulting in a decrease in TS. At higher GO loadings, a saturation point in polymer-filler interactions may be reached, beyond which the excess GO fails to effectively interact with the EPDM/NBR matrix. This reduces the efficiency of stress transfer, ultimately leading to a decline in TS. The presence of excessive GO can also influence the microstructure of the composites. The presence of large aggregates or agglomerates can compromise the integrity of the EPDM/NBR matrix, creating structural defects that diminish mechanical properties, including TS. Notably, the TS of the EPDM/NBR composites exhibited a 61% improvement compared to the EPDM/NBR blends. Composites typically exhibit a higher CLD compared to their blend counterparts due to the incorporation of nanofillers. The enhanced CLD in composites promotes improved dispersion and alignment of nanofillers within the EPDM/NBR matrix. Well-dispersed nanofillers provide additional reinforcement by acting as reinforcing agents, enhancing the load-bearing capability of the composites. This enhanced reinforcement leads to a significant increase in TS observed in the EPDM/NBR composites compared to the blends. The elevated CLD in the composites restricts polymer chain mobility, thereby minimizing chain slippage and improving the material's resistance to deformation. This reduced chain mobility results in a more robust and cohesive structure, leading to higher TS in composites compared to blends where CLD is lower.



Figure 4.1. Tensile strength of GO-filled EPDM/NBR composites

Figure 4.2 illustrates the EB of EPDM/NBR composites reinforced with GO. With increasing GO content, EB decreases due to restricted polymer chain mobility within the EPDM/NBR matrix. The higher filler loading limits the chains' ability to deform under stress, leading to reduced elongation. Additionally, the inherent rigidity of GO further constrains polymer chain movement, diminishing the material's capacity to stretch before failure and resulting in lower EB. The presence of GO in the composites can introduce stress concentration points, particularly when GO particles are not uniformly dispersed or when they form agglomerates. These stress concentration points can initiate crack propagation and accelerate failure, thereby reducing EB. In certain cases, insufficient interfacial bonding between the EPDM/NBR matrix and GO filler may arise, particularly at higher filler concentrations. This weak adhesion can lead to premature failure, contributing to a decline in EB. The EPDM/NBR composites exhibited a 27% reduction in EB compared to the EPDM/NBR blends. Additionally, the presence of nanofillers results in a higher CLD in composites, which further restricts polymer chain mobility, making it more challenging for the material to undergo significant elongation. With fewer free chains available for deformation, the composites exhibit reduced EB compared to blends, where the CLD is lower and polymer chain mobility is higher. The increased CLD in composites enhances the reinforcement effects of the nanofillers. Well-dispersed fillers within the polymer matrix effectively bridge polymer chains and strengthen the material. However, this improved reinforcement can also lead to reduced EB as the nanofillers restrict chain mobility and prevent extensive deformation of the material.



Figure 4.2. Elongation at break of GO-filled EPDM/NBR composites

Figure 3 presents the M100 values for the EPDM/NBR composites incorporating GO. The M100 values increase with GO content up to 5 phr. However, when the GO content exceeds this threshold, a decrease in M100 is observed. Initially, the incorporation of GO up to 5 phr improves the M100 of the composites. This is primarily due to the reinforcement effect of GO, which has high mechanical strength and stiffness. The uniformly dispersed GO sheets within the polymer matrix create effective linkages between polymer chains, enhancing load transfer and increasing stress-bearing capacity. At lower GO concentrations (up to 5 phr), the filler is evenly distributed, facilitating efficient stress transfer and reinforcing the material. This optimal dispersion allows for effective reinforcement, resulting in increased M100. However, beyond the threshold of 5 phr, further incorporation of GO may lead to agglomeration of filler particles within the matrix. Clusters of GO sheets can act as stress concentration sites, disrupting the uniform stress distribution and compromising the mechanical integrity of the composites. Furthermore, stronger filler-filler interactions may limit polymer chain mobility, leading to a decline in M100. At higher GO loadings, there may be a saturation point in terms of the polymer-filler interaction. Beyond this threshold, the surplus GO may fail to interact effectively with the EPDM/NBR matrix, reducing stress transfer efficiency and ultimately causing a decline in M100. The presence of excessive GO can also influence the microstructure of the composites. The formation of large aggregates or agglomerates may disrupt the continuity of the polymer matrix, leading to structural defects and decreased mechanical properties, including M100. The EPDM/NBR composites demonstrated a 36% increase in M100 compared to the EPDM/NBR blends. Composites typically exhibit a higher CLD compared to their blend counterparts due to the incorporation of nanofillers. The higher CLD in composites enhances the dispersion and alignment of nanofillers within the EPDM/NBR matrix. Well-distributed nanofillers serve as reinforcing agents, enhancing the material's load-bearing capacity. This enhanced reinforcement leads to a significant increase in M100 in the EPDM/NBR composites compared to the blends. Additionally, the higher CLD restricts polymer chain mobility, minimizing chain slippage and enhancing the material's resistance to deformation. This reduced chain mobility results in a more robust and cohesive structure, leading to higher M100 in composites compared to blends where CLD is lower.



Figure 3. Stress at 100% elongation of GO-filled EPDM/NBR composites

Figure 4 illustrates the hardness measurements of EPDM/NBR composites with GO incorporation. The hardness increases with rising GO content, as GO exhibits excellent mechanical properties, including high tensile strength and stiffness. Within the EPDM/NBR matrix, GO serves as a reinforcing filler, enhancing material strength and subsequently improving hardness. The rigid, planar structure of GO particles boosts the composites' load-bearing capacity, contributing to higher hardness. Achieving uniform GO dispersion within the matrix is essential for maximizing its reinforcing effectiveness. Well-dispersed GO particles ensure uniform reinforcement throughout the material, leading to enhanced hardness. Homogeneous dispersion of GO prevents the formation of localized weak points and promotes uniform mechanical properties across the composites. The incorporation of GO enhances the CLD of composites by acting as crosslinking sites during vulcanization, leading to a more tightly interconnected network within the EPDM/NBR matrix. This increase in CLD improves material cohesion and stiffness, thereby elevating hardness. Strong interfacial interactions between GO and the EPDM/NBR rubber matrix further enhance mechanical properties, including hardness, by enabling efficient stress transfer and load distribution. Additionally, GO may induce microstructural modifications, resulting in a denser and more rigid structure. These alterations in polymer chain and filler arrangement contribute to the overall hardness increase observed with higher GO content.



Figure 4. Hardness of GO-filled EPDM/NBR composites

Figure 5 illustrates the TAS measurements of EPDM/NBR composites with varying GO contents. The TAS increases as the GO content rises, primarily due to GO’s outstanding mechanical properties, including high tensile strength and stiffness. Acting as a reinforcing filler, GO enhances polymer chain interactions and facilitates effective load transfer, thereby improving tear resistance. Achieving uniform dispersion of GO within the EPDM/NBR matrix is essential for maximizing its reinforcing effect. Well-distributed GO particles provide consistent reinforcement, reducing localized weak points and enhancing tear resistance. Additionally, strong interfacial adhesion between GO and the polymer matrix promotes efficient stress transfer, minimizing crack initiation and propagation. Strong interfacial interactions help enhance tear resistance by preventing the separation of polymer chains during deformation. Additionally, the presence of GO can influence the CLD of the composites, further contributing to their improved TAS. Higher CLD, facilitated by the incorporation of GO, enhances the cohesion of the material and its resistance to tearing. The addition of GO may alter the microstructure of the composites, leading to improved tear resistance. GO sheets act as physical barriers that impede crack and tear propagation within the material, enhancing its overall TAS. The EPDM/NBR composites demonstrated a 71% increase in TAS compared to the EPDM/NBR blends. The enhanced CLD in the composites restricts polymer chain mobility, making them less prone to deformation and tearing. As a result, the material exhibits enhanced tear resistance, becoming more resistant to failure under applied stress. In contrast, blends with lower CLD may exhibit lower TAS due to increased chain mobility and susceptibility to tear propagation. The presence of a higher density of crosslinks in composites facilitates more effective stress distribution within the material. Crosslinks act as structural barriers that hinder crack and tear propagation, thereby enhancing tear resistance. In blends, where CLD is lower, stress concentrations may occur more readily, leading to localized failure and reduced TAS. Nanofillers like GO or HNTs also contribute to reinforcement, which further enhances TAS in composites. Evenly distributed nanofillers establish strong connections between polymer chains, promoting efficient load transfer and improving tear resistance compared to blends without nanofillers.



Figure 5. Tear strength of GO-filled EPDM/NBR composites

Figure 6 illustrates the rebound resilience (RR) of EPDM/NBR composites with varying GO content. As the GO content increases, RR decreases. The higher filler loading within the EPDM/NBR matrix limits polymer chain mobility, reducing their ability to return to their original shape after deformation, leading to lower RR. Due to its rigidity and immobility, GO further restricts polymer chain movement, diminishing the material’s capacity for elastic deformation and recovery. Additionally, the presence of GO increases interfacial friction between the filler and the polymer matrix, causing energy dissipation during deformation and further reducing RR. At higher GO loadings, the filler particles may agglomerate, creating stress concentration points and impeding uniform deformation, which also decreases RR. Moreover, the incorporation of GO enhances the CLD of the composites, forming a more rigid network structure that restricts the material’s ability to recover its original shape after deformation, leading to a decrease in RR. The EPDM/NBR composites showed a 23% reduction in RR compared to the EPDM/NBR blends. Composites typically exhibit a higher CLD than blends due to the presence of nanofillers like GO, which act as crosslinking sites during vulcanization, forming a more interconnected network within the polymer matrix. The enhanced CLD limits polymer chain mobility, reducing the material’s capacity for elastic deformation and recovery after compression, thereby leading to a lower RR compared to blends. Blends, with their lower CLD, allow for more flexible polymer chains, resulting in greater elasticity and better RR. Stronger interfacial bonding in composites, associated with higher CLD, may also cause more energy dissipation during deformation, further reducing energy recovery efficiency and contributing to the decreased RR in composites.



Figure 7 illustrates the abrasion loss values of EPDM/NBR composites with varying GO content. As the GO content increases, abrasion loss decreases, signifying enhanced abrasion resistance (AR). GO exhibits outstanding mechanical properties, such as high TS and stiffness. When incorporated into the EPDM/NBR matrix, GO acts as a reinforcing filler, significantly enhancing the material's strength and abrasion resistance. The rigid, planar structure of GO particles helps the material withstand abrasive forces, minimizing wear. Achieving uniform dispersion of GO within the EPDM/NBR matrix is crucial for maximizing its reinforcing benefits. Well-dispersed GO particles ensure uniform reinforcement throughout the material, leading to enhanced AR. Homogeneous dispersion of GO prevents the formation of localized weak points and promotes uniform wear resistance across the composites. Robust interfacial interactions between GO and the EPDM/NBR matrix are crucial for enhancing mechanical properties, including AR. Strong bonding between GO and the EPDM/NBR matrix enhances the material's resistance to abrasive wear by preventing filler detachment and reducing material loss. The inclusion of GO may also contribute to an increase in the CLD of the composites, further enhancing their durability. Higher CLD results in a more cohesive and durable material that is better able to resist abrasive wear. The denser network structure formed by crosslinks provides additional support and reinforcement against abrasive forces. The presence of GO can reduce the coefficient of friction between the contacting surfaces, thereby decreasing abrasive wear. The lubricating effect of GO particles helps to minimize frictional forces and prevent excessive material loss due to abrasion. Composites typically exhibit a higher CLD compared to blends due to the presence of nanofillers like GO. During vulcanization, GO particles act as crosslinking sites, promoting the formation of a denser network structure within the EPDM/NBR matrix. This increased CLD strengthens the material and improves its resistance to abrasive wear. The higher CLD in composites results in a more cohesive and durable material. The densely crosslinked network effectively holds the polymer chains together, preventing them from disintegrating or wearing away under abrasive conditions. As a result, composites with higher CLD exhibit enhanced AR compared to blends with lower CLD. The presence of a dense network of crosslinks in composites helps to minimize material degradation during abrasive wear. The tightly interconnected polymer chains exhibit greater resistance to breakage or detachment from the surface, resulting in minimized material loss and enhanced AR. In contrast, blends with lower CLD may experience more extensive material degradation under abrasive conditions, resulting in lower AR. The presence of crosslinks in composites facilitates more efficient load distribution across the material during abrasive wear. The interconnected network structure helps to distribute the applied forces more evenly, reducing localized stress concentrations and minimizing wear at specific points. The increased CLD facilitates better load distribution, which significantly enhances the AR of the composites.



Figure 7. Abrasion loss of GO-filled EPDM/NBR composites

Figure 8 depicts the compression set (CS) data for EPDM/NBR composites with different GO contents. The CS increases with rising GO concentration due to enhanced filler-filler interactions and possible agglomeration, which hinder the material’s ability to regain its original shape after compression. As a rigid and immobile filler, GO restricts polymer chain mobility within the composite matrix, thereby reducing its capacity for elastic deformation and shape recovery. Additionally, the presence of GO particles increases interfacial friction, which dissipates energy during compression and further impairs the material’s resilience, leading to a higher CS. The incorporation of GO can also influence the CLD of the composites. A higher CLD, promoted by GO, forms a more rigid network structure that is less adaptable to compression deformation, further contributing to increased CS values. At higher GO concentrations, filler agglomeration can generate stress concentration points, disrupting uniform compression and recovery, thereby increasing CS. Additionally, the presence of GO in the composites may influence shape memory retention, reducing the material's ability to return to its original form after deformation. Increased GO content can alter the material's viscoelastic properties, impacting its memory retention capacity. Consequently, the composites may show reduced memory retention, leading to greater permanent deformation and higher CS values.



Figure 8. Compression set of GO-filled EPDM/NBR composites

Figure 9 illustrates the CLD measurements of EPDM/NBR composites with varying GO content, demonstrating a rise in CLD with increasing GO concentration. GO nanoparticles possess functional groups that interact with the EPDM/NBR matrix during vulcanization, serving as crosslinking sites. As GO content increases, more crosslinking sites are introduced, resulting in a higher CLD within the composite matrix. Additionally, GO facilitates effective crosslink formation during vulcanization. Its rigid, planar structure promotes polymer chain alignment and enhances crosslinking reactions, leading to a denser network structure with elevated CLD. Strong interfacial interactions between GO and the polymer matrix improve filler-polymer bonding, enhancing the transfer of crosslinking agents to GO particles. This further supports crosslink formation, contributing to an overall increase in CLD. Moreover, GO incorporation restricts polymer chain mobility, preventing rearrangement during vulcanization and fostering a more interconnected network. Proper dispersion of GO ensures a uniform distribution of crosslinking sites throughout the matrix, leading to a homogeneous network with consistent CLD. CLD plays a crucial role in determining the mechanical properties of rubber-based composites, including TS, M100, TAS, hardness, and AR. A higher CLD results in a more tightly interconnected network, improving the material’s ability to withstand tensile forces without permanent deformation, thereby enhancing TS. The restriction of polymer chain mobility with increased CLD results in greater resistance to elongation, leading to higher M100. A denser crosslinked network also improves tear resistance by preventing crack formation and propagation, enhancing TAS. Furthermore, increased CLD contributes to material rigidity and stiffness, leading to greater hardness. The well-structured crosslinked network enhances resistance to wear and abrasion by providing mechanical support and minimizing material loss during abrasive contact, thereby improving AR.



Figure 9. Crosslinking density of GO-filled EPDM/NBR composites

Table 4.1 displays the mole percent uptake (MPU) of toluene in EPDM/NBR composites with varying GO loadings, showing a decrease in MPU as GO content increases. This decline is attributed to GO’s excellent barrier properties, which arise from its high aspect ratio and impermeable structure. When integrated into the EPDM/NBR matrix, GO creates a tortuous diffusion pathway, effectively restricting molecular transport within the composite. This impediment lowers the overall absorption of substances by the composite. Furthermore, GO enhances the CLD of the polymer matrix by interacting with polymer chains through its oxygen-containing functional groups, including hydroxyl, epoxy, and carboxyl groups. This enhanced cross-linking reduces the polymer's free volume, thereby lowering MPU. Overall, GO significantly contributes to improving the mechanical properties of polymer composites. The rigid and strong GO sheets swelling or uptake of molecules. The enhanced mechanical strength contributes to a lower MPU. GO has both hydrophilic and hydrophobic regions due to its functional groups. Depending on the nature of the substances interacting with the composite, GO can alter the surface properties of the composites. For instance, if the uptake involves polar molecules, the hydrophilic nature of GO can lead to reduced affinity and, consequently, lower uptake. Incorporating GO into the EPDM/NBR matrix reduces the free volume within the EPDM/NBR. The GO sheets occupy space in the matrix, minimizing the voids and gaps that would otherwise facilitate higher molecular uptake. As the GO content increases, its dispersion within the EPDM/NBR matrix becomes more uniform, resulting in a well-structured composite. This improved distribution creates a denser and less permeable material, thereby lowering the uptake.

Table 4.1. MPU values of EPDM/NBR-GO composites in different solvents

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Sample code | Mole percent uptake (mol%) at 30oC | | | | | | | | | | |
| Aromatic | | | | Aliphatic | | | | Chlorinated | | |
| Benzene | Toluene | Xylene | Mesitylene | n-pentane | n-hexane | n-heptane | n-octane | Dichloromethane | Chloroform | Carbon tetrachloride |
| GO0 | 4.12 | 3.85 | 3.65 | 3.1 | 2.08 | 2.04 | 2.02 | 1.88 | 2.75 | 2.65 | 2.38 |
| GO1 | 4.11 | 3.84 | 3.64 | 3.08 | 2.07 | 2.02 | 2 | 1.87 | 2.73 | 2.63 | 2.36 |
| GO2 | 4.1 | 3.83 | 3.62 | 3.06 | 2.06 | 2 | 1.98 | 1.85 | 2.71 | 2.61 | 2.35 |
| GO3 | 4.09 | 3.81 | 3.61 | 3.04 | 2.04 | 1.98 | 1.96 | 1.84 | 2.7 | 2.59 | 2.33 |
| GO4 | 4.07 | 3.8 | 3.59 | 3.02 | 2.02 | 1.97 | 1.95 | 1.82 | 2.68 | 2.57 | 2.31 |
| GO5 | 4.05 | 3.77 | 3.57 | 2.99 | 2 | 1.95 | 1.93 | 1.8 | 2.65 | 2.55 | 2.29 |
| GO6 | 4.06 | 3.78 | 3.59 | 3 | 2.02 | 1.97 | 1.95 | 1.82 | 2.67 | 2.56 | 2.3 |
| GO7 | 4.07 | 3.79 | 3.6 | 3.02 | 2.03 | 1.98 | 1.96 | 1.83 | 2.68 | 2.58 | 2.32 |
| GO8 | 4.09 | 3.81 | 3.62 | 3.04 | 2.05 | 1.99 | 1.98 | 1.84 | 2.7 | 2.59 | 2.34 |

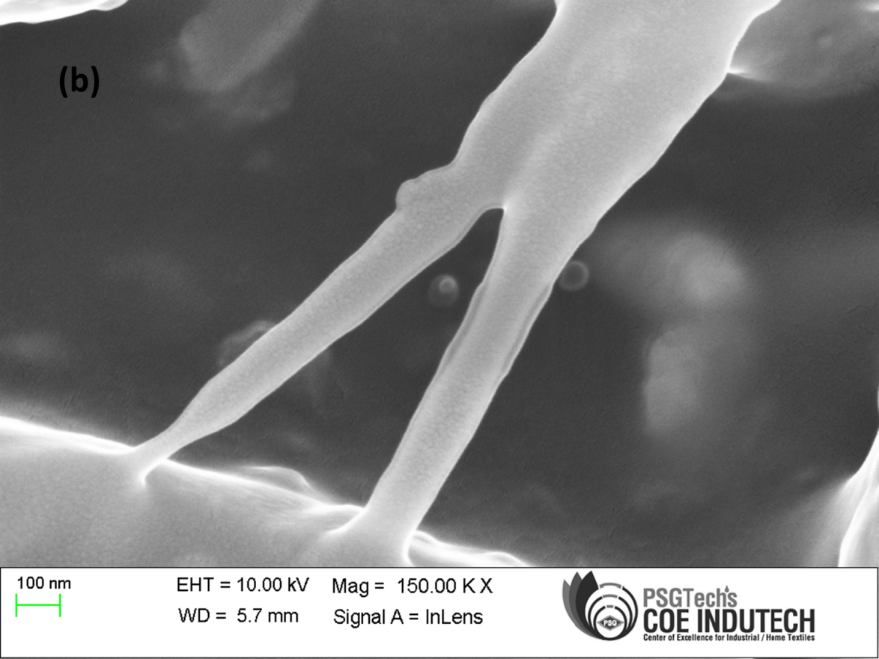
The aromatic solvent uptake follows the order of benzene, toluene, xylene, and mesitylene, with benzene exhibiting the highest uptake and mesitylene the lowest. Benzene is the smallest and simplest aromatic hydrocarbon, with a single six-carbon ring structure. Its smaller molecular size allows it to penetrate and be absorbed more easily into the EPDM/NBR matrix. Toluene is slightly larger than benzene, with a methyl group attached to the benzene ring. The additional methyl group increases its molecular size, which slightly hinders its uptake compared to benzene. Xylene has two methyl groups attached to the benzene ring. The presence of two additional methyl groups increases the molecular size relative to toluene, reducing its ability to diffuse into the EPDM/NBR matrix. Mesitylene has three methyl groups attached to the benzene ring, making it the largest among the four solvents. The increased molecular size and steric hindrance significantly reduce its ability to be absorbed into the EPDM/NBR matrix. Benzene has the highest symmetry and relatively low polarity among the solvents listed, which allows it to interact easily with the polymer chains. The addition of a methyl group to benzene increases its polarity slightly, but it still remains relatively low, allowing moderate interaction with the EPDM/NBR matrix. With two methyl groups, xylene's polarity is slightly higher than toluene, but the steric hindrance from the two methyl groups reduces its overall uptake. Mesitylene, with three methyl groups, has the highest polarity and steric hindrance, making it the least absorbed by the EPDM/NBR matrix due to its larger size and more complex structure. Smaller molecular size allows for more efficient van der Waals interactions with the polymer chains, enhancing its uptake. The single methyl group slightly hinders the van der Waals interactions compared to benzene, reducing its uptake. The presence of two methyl groups further weakens van der Waals interactions with the EPDM/NBR matrix. The presence of three methyl groups significantly enhances steric hindrance, further weakening van der Waals interactions and resulting in the lowest uptake.

The uptake of aliphatic solvents follows the order of n-pentane being the highest, followed by n-hexane, n-heptane, and n-octane. Among them, n-pentane has the shortest carbon chain, consisting of five atoms. Its smaller molecular size allows for easier diffusion into the EPDM/NBR matrix, resulting in the highest uptake. In contrast, n-hexane, with a slightly longer carbon chain (6 carbons), has a marginally larger molecular size, which reduces its ability to penetrate the EPDM/NBR matrix and leads to a slightly lower uptake compared to n-pentane. n-heptane has a seven-carbon chain. As the carbon chain length increases, the molecular size expands, leading to reduced diffusivity and lower uptake by the EPDM/NBR matrix. Among the four solvents, n-octane, with its eight-carbon chain, has the largest molecular size. Its longer chain length restricts penetration into the polymer matrix, leading to the lowest uptake. In contrast, smaller aliphatic molecules such as n-pentane experience weaker van der Waals interactions, enabling easier diffusion into and within the EPDM/NBR matrix. Slightly stronger van der Waals interactions compared to n-pentane due to its longer chain, but still relatively weak, allowing moderate uptake. Further increased van der Waals interactions due to its longer chain reduce its ability to diffuse as effectively as n-pentane and n-hexane. Strongest van der Waals interactions among the four, combined with its larger size, make it the least diffusive and thus the least absorbed by the EPDM/NBR matrix. The solubility parameter of n-pentane may align more closely with that of the EPDM/NBR matrix, promoting greater uptake. n-Hexane has a slightly different solubility parameter, which may result in a moderately lower compatibility and uptake compared to n-pentane. With further differences in the solubility parameter and increased molecular size, n-heptane's uptake is lower. The least compatible due to the longest chain and greatest difference in solubility parameter, resulting in the lowest uptake.

The solvent uptake follows the order of dichloromethane being the highest, followed by chloroform, and then carbon tetrachloride. Dichloromethane, with its smallest molecular size and simplest structure, diffuses most easily into the EPDM/NBR matrix, leading to the highest uptake. Chloroform, being larger than dichloromethane but smaller than carbon tetrachloride, exhibits moderate diffusion and uptake. In contrast, carbon tetrachloride, the largest molecule in this series, has a more complex structure, making it the least effective at penetrating the EPDM/NBR matrix, resulting in the lowest uptake. Additionally, dichloromethane's higher polarity enhances its interactions with the polymer matrix, further increasing uptake. Chloroform, with an intermediate polarity, shows moderate interaction and uptake, while carbon tetrachloride, with the lowest polarity, exhibits the weakest interaction and lowest uptake. Carbon tetrachloride is non-polar, which reduces its interaction with many polymer matrices, leading to the lowest uptake. Dichloromethane has the lowest density and highest diffusion coefficient among the three solvents, allowing it to move more freely and be absorbed more efficiently into the EPDM/NBR matrix. Chloroform has a higher density and a lower diffusion coefficient than dichloromethane but remains lower than that of carbon tetrachloride, leading to a moderate uptake. In contrast, carbon tetrachloride, possessing the highest density and the lowest diffusion coefficient, has the least ability to penetrate the EPDM/NBR matrix, resulting in the lowest uptake.

Figure 4.10 presents FESEM micrographs of the tensile-fractured surfaces of EPDM/NBR composites, including: (a) the blend; (b) 5 phr GO; and (c) 7 phr GO. EPDM is well blended with NBR, forming a homogeneous blend between the two due to their compatible polymer structures and similar solubility parameters. This compatibility facilitates thorough mixing, resulting in no gaps or separation, as seen in Figure 16(a). The uniform dispersion of polymers improves the MPs and stability of the blend. As shown in Figure 16(b), the 5 phr GO filler exhibits excellent dispersion within the EPDM/NBR composites. This even distribution enhances the composite's mechanical performance, leading to increased tensile strength by facilitating more efficient stress transfer and minimizing stress concentration points. By contrast, Figure 16(c) depicts less effective dispersion of the 10 phr GO filler, which can be attributed to agglomeration at higher filler loadings. This agglomeration leads to the formation of weak points within the composite structure, causing lower TS and more pronounced tear lines. The insufficient distribution of GO at higher concentrations prevents the optimal reinforcement of the EPDM/NBR matrix, highlighting the importance of achieving proper filler dispersion to maximize composite performance.





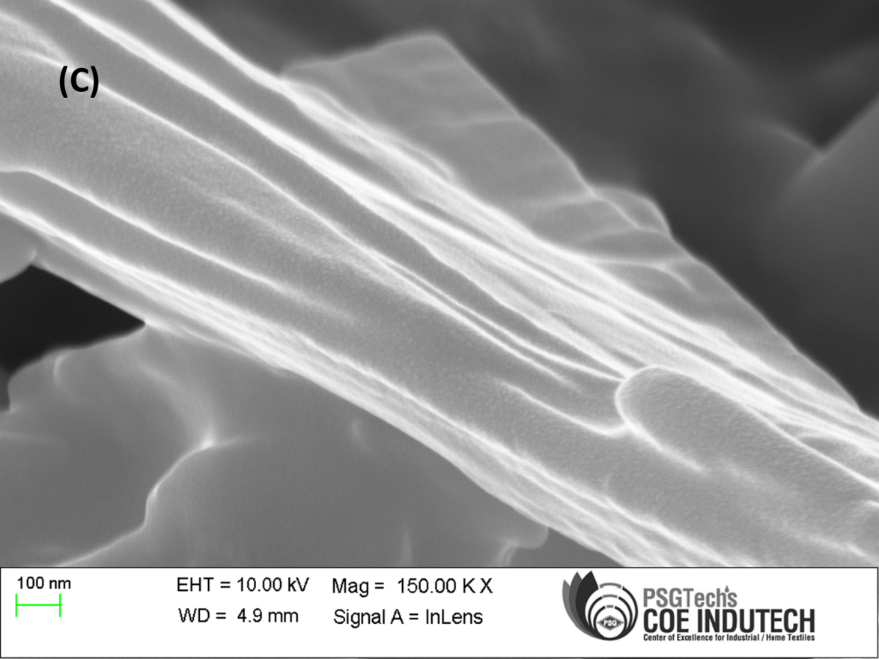


Figure 4.10. FESEM images of (a) GO0, (b) GO5, and (c) GO7.

**CHAPTER 5**

**CONCLUSIONS**

EPDM/NBR composites incorporating GO were systematically prepared using an open mill mixer. The findings from this study provide valuable insights, leading to the following conclusions:

1. TS and M100 increase with GO content up to 5 phr due to improved reinforcement and filler dispersion. However, beyond 5 phr, agglomeration of GO particles leads to localized stress concentrations, resulting in diminished mechanical properties.
2. EB decreases with increasing GO content as the rigid nature of GO restricts polymer chain mobility and elongation. Similarly, RR declines since higher GO content impairs the polymer matrix's ability to recover its original shape after deformation, reducing resilience.
3. TAS improves with increasing GO content due to GO’s reinforcing effect, which enhances interfacial adhesion and dispersion, thereby increasing resistance to crack propagation.
4. Hardness rises with higher GO content as GO reinforcement strengthens the EPDM/NBR matrix. The increased CLD and rigidity contribute to greater resistance to indentation.
5. AR improves with higher GO content due to the reinforcement effect of GO particles, which strengthen the matrix and minimize wear. Enhanced dispersion of GO further improves interfacial adhesion, further boosting AR.
6. CS increases with GO content as reduced flexibility and increased stiffness limit the material’s ability to recover after deformation.
7. Swelling resistance improves with higher GO content due to GO’s impermeable nature, which acts as a barrier against the penetration of swelling agents, reducing overall swelling.
8. CLD increases with GO content as the functional groups on GO facilitate crosslinking reactions with the polymer matrix, resulting in a denser network structure.
9. FESEM revealed homogeneous blending of EPDM/NBR blends due to their compatible structures. The effective dispersion of 5 phr GO in the EPDM/NBR matrix suggests proper mixing, while the agglomeration of 10 phr GO indicates excessive filler loading, leading to clustering within the matrix.
10. EPDM/NBR composites filled with GO show superior mechanical properties and swelling resistance, making them ideal for robust sealing solutions like automotive gaskets. Their improved performance, driven by enhanced interfacial adhesion and dispersion, highlights their potential to enhance sealing efficiency in demanding environments

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