Experimental Study on Mechanical Properties of EPDM/NBR Composite using Nanoclay

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Abstract:

Currently, significant research attention is focused on new rubbery materials obtained by blending two or more rubbers. The blending of Ethylene-Propylene-Diene Monomer (EPDM) and Acrylonitrile-Butadiene Rubber (NBR) were prepared to achieve the best properties from each component. EPDM has good ozone and heat aging resistance but it possesses poor solvent resistance. NBR has high resistance to swelling in the solvents and oils but suffers from poor heat aging and ozone resistance properties. The blend of such two rubbers attracts the attentions to tailor a rubber blend which withstands heat ageing, ozone, solvent and oils swelling with desirable mechanical properties. Thus, the product of this rubber blend will have excellent heat resistance, oil resistance, ozone resistance and mechanical properties. It could be used for the production of recognized rubber products such as automotive radiator hoses, automotive brake hoses, motor mounts, conveyor belts, transmission belts, sheets and rolls. Nanoclay was generally used to improve the physical, mechanical and thermal properties of the rubber and its composite compounds. This was due to the property of very high surface area of the nanoclay. The load applied in the composite rubber was transferred from the polymer (rubber) matrix to the reinforcement (nanoclay) particles. Nanoclay filler filled composites based on EPDM/NBR were prepared. The nanocomposites were prepared by mixing using a two-roll open mill. The effect of nanoclay filler on the mechanical properties, hardness, rebound resilience, abrasion resistance, swelling resistance and morphology of the EPDM//NBR nanocomposites were investigated. Tensile strength increases with increase of nanoclay upto 7.5 phr and then decreases. As the tensile strength and hardness increases, elongation at break and rebound resilience decreases. Percentage increase in tensile strength is 99% whereas percentage decrease in elongation at break is 18%. Abrasion and swelling resistance increases with increasing nanoclay loading.

Keywords: *EPDM/NBR blends; Nanoclay; Mechanical Properties, Swelling resistance.*

1. Introduction:

Composites, the wonder materials are becoming an essential part of today's materials due to the advantages such as low weight, corrosion resistance, high fatigue strength, and faster assembly. The basic difference between blends and composites is that the two main constituents in the composites remain recognizable while these may not be recognizable in blends. Composite material is a material composed of two or more distinct phases (matrix phase and dispersed phase) and having bulk properties significantly different from those of any of the constituents. Matrix phase is the primary phase having a continuous character. Matrix is usually more ductile and less hard phase. It holds the dispersed phase and shares a load with it. Dispersed (reinforcing) phase is embedded in the matrix in a discontinuous form. This secondary phase is called the dispersed phase. Dispersed phase is usually stronger than the matrix, therefore, it is sometimes called reinforcing phase.

SBR was the initial developed low price artificial rubber used in manufacturing industries. SBR in spite of having high-quality mechanical and abrasive properties, they are too insightful towards the environmental factors such as moisture, ozone, light, and heat. This was due to the occurrence of twice bond present in the polymeric chain which can be surmounting by combination it with well saturated elastomers like EPDM. EPDM was a special kind of rubber used in range of industrial purpose in day to day life. EPDM rubbers exhibit a superior oxidative and heat resistant characteristic due to the significant absence of unsaturated chain in polymeric backbone [1, 2]. Carbon black and amorphous silica was utilized to get better mechanical properties of rubber composites, and have been utilized as an vital reinforcing agent in the rubber industries in the current decades [3–5]. Later, other reinforcing materials such as nanoclay [6–12], nanosilica [13], calcium carbonate [14] and carbon nanotubes [15] have acknowledged a immense agreement of notice in rubber industries due to their unique structures and their easy accessibility in comparison with other reinforcing nano-particles. The use of nanoclay in the EPDM rubber [16, 17], SBR [18–20] and EPDM/SBR [21, 22] has been reported by many researchers in current decades.

The throw away materials such as groundnut shell, rice husk (RH), sugarcane bagasse and bamboo leaves are well thought-out as a natural source of silica. These throw away materials are scalded and the ash obtained from it are rich in silica and carbon content, which has no exploitation yet. Rice husk ash was the material that consists of about 90–98% of silica after the complete combustion of it. Extraction of silica from rice husk was an emerging trend in the existing research field. Enormous amount of rice husk are treated as the waste materials and are disposed as the landfill. But burning of rice husk materials will lead to stern ecological pollutions. The suspended ash particles in the atmosphere lead to respiratory problems in human beings [23]. The commonly extracted silica (tetra ethyl orthosilicate) was more expensive, and hence rice husk ash (having adequate silica) can be used as an alternative source [24, 25, 26].

The present study to develop Ethylene Propylene Diene Monomer (EPDM) rubber / Acrylonitrile Butadiene Rubber (NBR) blends reinforced by nanoclay by the conventional method using a two-roll mill. The main objectives of the study are Preparation of Ethylene Propylene Diene Monomer (EPDM) rubber / Acrylonitrile Butadiene Rubber (NBR) blends with nanoclay by using a two roll mill and Studying the mechanical properties and characterization of EPDM/NBR composites.

2. Materials and Methods

2.1. Preparation of Ethylene-Propylene-Diene monomer (EPDM)/Acrylonitrile-Butadiene Rubber (NBR) with Nanoclay Composites

2.1.2. Ethylene Propylene Diene Monomer Rubber: In this work, EPDM (KEP-270) grade in the form of bulk polymerization material supplied by D.R polymer engineering work, Chennai is used.

2.2. Styrene Butadiene Rubber: In this work, Acrylonitrile-Butadiene Rubber in the form of 35 Kg packed material supplied by D.R polymer engineering work, Chennai is used.

2.3. Reinforcing Filler: Nanoclay supplied by Sigma Aldrich is used as reinforcing filler.

2.4. Compounding Ingredients: The following are the Compounding Ingredients is used to produce EPDM/NBR-Nanoclay composite.

- ✓ Ethylene-Propylene-Diene Monomer (EPDM)
- ✓ Acrylonitrile-Butadiene Rubber
- ✓ Zinc oxide(Activator)
- ✓ Stearic oxide(Co-activator)
- ✓ Nanoclay
- ✓ Sulphur (Cross linking agent)
- ✓ Dibenzothiazole Disulfide (MBDS) (Primary Accelerator)

✓ Tetramethyl Thiuram Monosulfide (TMTD) (Secondary accelerator)

2.5. Activator (Zinc oxide): Activator is added to rubber compounding to activate cross linking agents. The majority of organic accelerators are usually unable to exercise their maximum effect unless the rubber contains certain substance known as activators .These are two kinds-metallic oxides and organic acids. Both are usually required in order to achieve full effect with most accelerators. The most commonly used metallic oxide is zinc oxide. Zinc oxide are used as activator in our rubber compounds. The benefits are Reducing vulcanization time and To make the rubber durable

2.6. Reinforcing Fillers (Nanoclay): filler can be used very effectively to enhance the ultimate physical properties such as tensile strength, resistance to abrasion, tear and flexing. At the same time, however. Filler also affect the modulus and viscoelastic properties at small and intermediate strains. They generally increase the relative density of rubber compounds.

2.7. Cross linking agent or Vulcanizing agent (Sulphur): Vulcanizing agents most commonly used as sulfur creates cross-link between polymer chains changing the thermoplastic to the elastic condition. Vulcanizing agents is an ingredient to form cross-links between the rubber chains in the vulcanization process. Sulfur is used in our rubber compounding as cross linking agents for the benefits of Elastic strength will be increased

2.8. Accelerator: During vulcanization the accelerator apparently converts the sulfur into a compound that reacts more rapidly with rubber than does sulfur itself. An alternative possibility is that the accelerator reacts first with the rubber, changing it into a form that combines rapidly with sulfur. Accelerating agents increase the rate of the cross linking reaction and lower the sulphur content necessary to achieve optimum vulcanizate properties. Dibenzothiazole Disulfide (MBDS) and Tetramethyl Thiuram Monosulfide (TMTM) are used as accelerator in our rubber compounding

2.9. Preparation of nanoclay filler-filled EPDM/NBR composites: Ethylene-Propylene-Diene Monomer (EPDM) and Acrylonitrile-Butadiene Rubber (NBR) compounds filled with nanoclay were made of Ethylene-propylene-Diene Monomer (EPDM), Acrylonitrile-Butadiene Rubber (NBR), Nanoclay, Cure activators (Stearic acid and ZnO_2) and curatives (Mercaptobenzothiazole Disulfide (MBDS), Tetramethyl Thiuram Monosulfide (TMTM), and sulphur). All compounds had the same composition, except for the amounts of nanoclay, which were varied to prepare compounds with different ratios of filler. However, the total amount of filler in each formulation was varies from 0 to 10 phr. The formulations are given in Table.1

Table. 1 NR/SBR compound formulation

Ingredients	Proportions (phr)				
	S_1	S_2	S_3	S_4	S_5
EPDM	70	70	70	70	70
NBR	30	30	30	30	30
Nanoclay	0	2.5	5	7.5	10
Stearic acid	2.5	2.5	2.5	2.5	2.5
Zinc Oxide	5	5	5	5	5
Sulphur	1.5	1.5	1.5	1.5	1.5
MBDS	1	1	1	1	1
TMTD	0.8	0.8	0.8	0.8	0.8

A two-step procedure is adopted to prepare Ethylene-Propylene-Diene Monomer (EPDM) and Acrylonitrile-Butadiene Rubber (NBR) with nanoclay. In the first step, Ethylene-Propylene-Diene Monomer and Acrylonitrile-Butadiene Rubber are thoroughly masticated on a two roll mill of Length 30 inches and roll diameter 13 inches. The Friction ratio is maintained at 1:1.25. In the Second step, masticated rubber is compounded with nanoclay and other ingredients at appropriate timings according to the formulation given in the Table. 2. Finally, 10 end-roll passes were made before sheeting off.

Table 2 Formulation for NBR-CB/Sio_2 composites

Constituent	Composition in phr	Processing time in min
EPDM	70	10
NBR	30	
Rubber vulcanization accelerator - stearic acid	2.5	2
Activator - zinc oxide	5	2
Reinforcing fillers – Nanoclay	0-10	0-10
Cross linking agent – sulphur	1.5	2
Primary Accelerator - Dibenzothiazole Disulfide (MBDS)	1	2
Secondary accelerator - Tetramethyl Thiuram Monosulfide (TMTD)	0.8	2

2.10 Curing of rubber: The compound of rubber thus obtained is dried at room temperature for 24 hours and then molded into sheets and button specimens using hydraulic press of 60 MPa load at 160^0C and at 10 minutes is shown in Fig.1 and die are shown in Fig.2, 3 & 4. To make approximately 2 mm thick rubber sheets is shown in the Fig.5.

Fig.1 Hydraulic press,

Fig.2 Die size 30*10,

Fig.3 Die size 15*15*2.5

Fig.4 Die size 16.7*21.5*12.8,

Fig.5 Image of vulcanized rubber sheets.

3. RESULTS AND DISCUSSIONS

3.1. Measurement of mechanical properties: The numerical values of mechanical properties are taken as the average of results obtained from 3 specimens.

3.1.1 Tensile properties: EPDM/NBR composites are tested for its mechanical properties such as tensile strength, elongation at break, hardness, rebound resilience and abrasion loss. Compression molded sheets having a thickness of about 2 mm are used for tensile testing. Standard dumbbell shaped specimens are used to determine the tensile strength according to ASTM D412. The test is carried out using an Instron Universal Tester (Model 4301) at a crosshead speed of 500 mm/min and with initial gauge length of 25 mm.

3.1.2 Hardness: The hardness of the EPDM/NBR composites is measured using Shore A durometer in accordance with ASTM D2240.

3.1.3 Abrasion resistance: The abrasion resistance of the samples is determined using a DIN abrader as per ASTM D5963. Samples having a diameter of 30 mm and a thickness of 10 mm are holding on rotating sample holder and 10 N load is applied. The difference in initial weight and final weight after the travel distance run of 42 m on a standard abrasive surface is taken as abrasion loss.

3.1.4 Rebound resilience: Rebound resilience is measured using a vertical rebound resilience tester as per ASTM D2632. A plunger weighing 28 ± 0.5g is dropped from a height of 40cm on the sample of thickness 10mm and the rebound height is measured.

3.1.5 Swelling resistance: The swelling test was done as per ASTM D-471 by the solvent immersion method. Swelling test was carried out at different penetrant such as aromatic (toluene), aliphatic (n-pentane) and chlorinated hydrocarbons (carbon tetrachloride). The mole percent uptake, Qt for solvent was determined using the formula.

$$Q_t(mol\%) = \frac{(M_t - M_0)/MW}{M_0} \times 100$$

where, M0 is the initial mass of the specimen, Mt is the mass of the specimen after time 72 h of immersion, and MW is the molecular weight of the solvent.

3.2 Characterization of Composites:

3.2.1 Surface morphology: Surface morphology is studied under Hitachi (model S-2500) scanning electron microscope (SEM).

3.3. Tensile Strength:

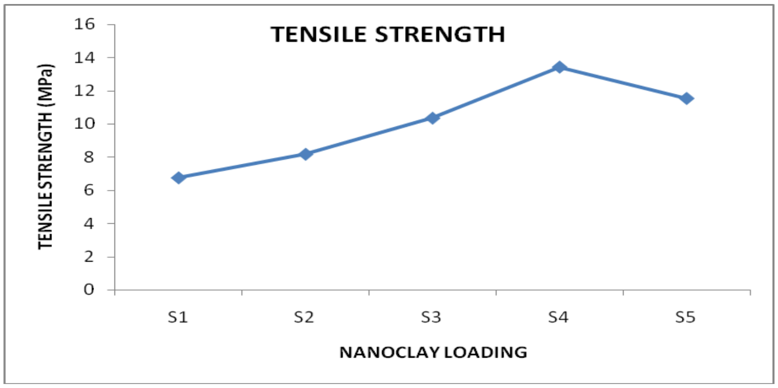

Figure 8.1 Tensile strength of the EPDM/NBR composites

3.4. Elongation at break:

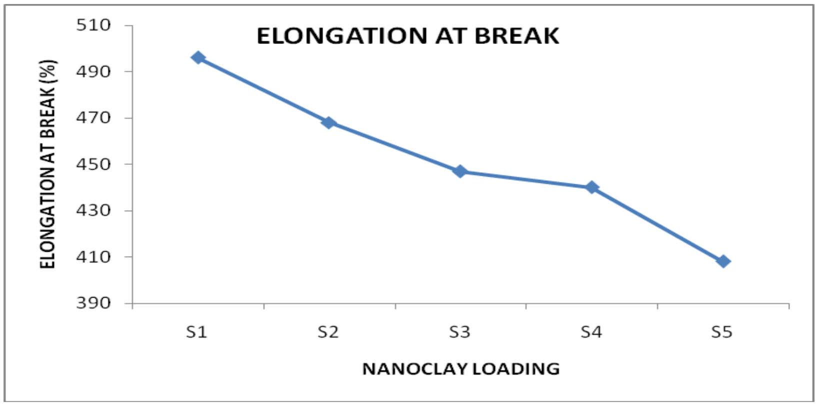

Figure 8.2 Elongation at break of the EPDM/NBR composites

3.5. Hardness:

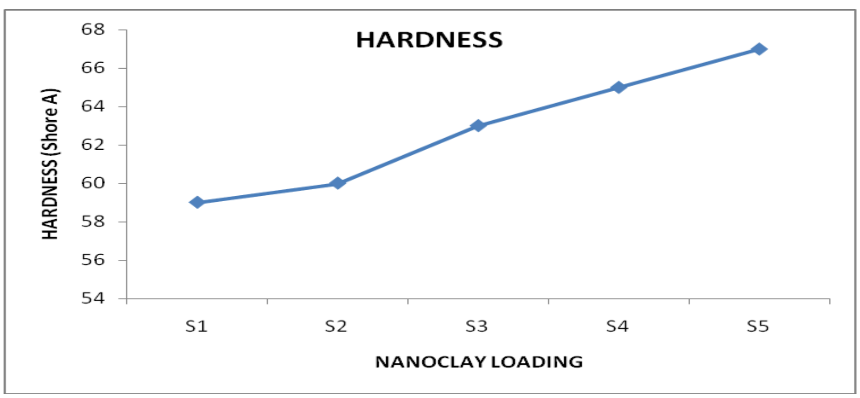

Figure 8.3 Hardness of the EPDM/NBR composites

3.6. Rebound resilience:

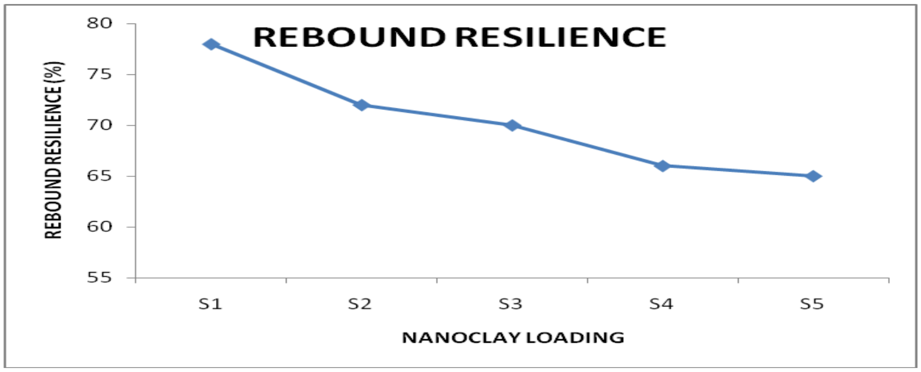

Figure 8.4 Rebound resilience of the EPDM/NBR composites

3.7. Abrasion loss:

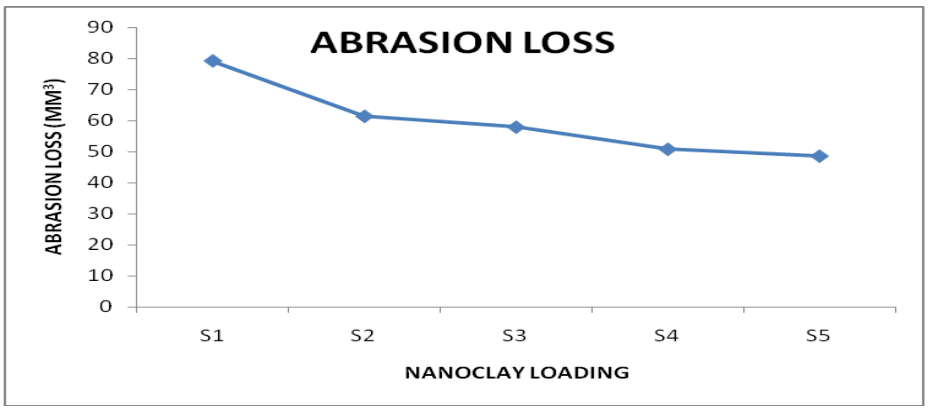

Figure 8.5 Abrasion loss of the EPDM/NBR composites

3.8. Swelling resistance

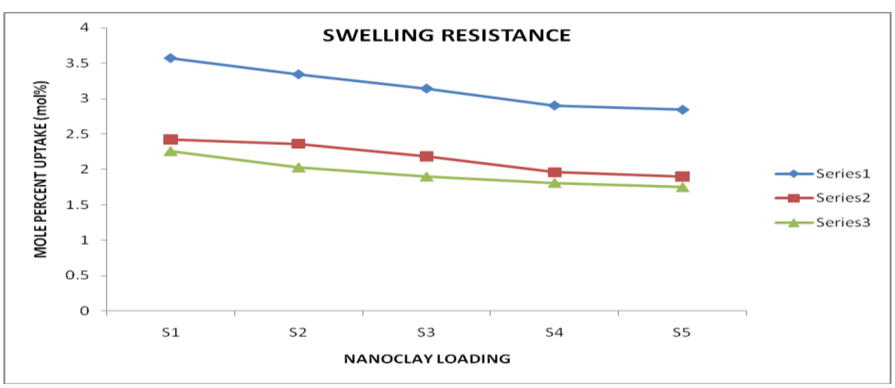

Figure 8.6 Swelling resistance (in terms of mole percent uptake) of the EPDM/NBR composites: Series 1 (Toluene solvent), Series 2 (pentane solvent) and Series 3 (carbon tetrachloride)

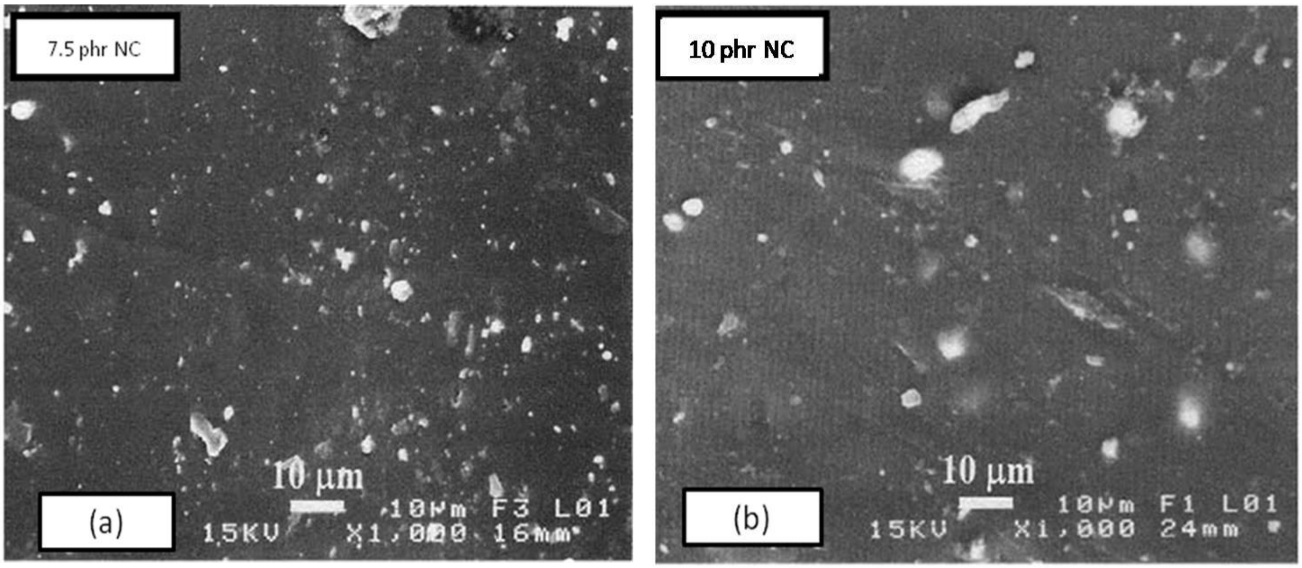

Figure 8.7 SEM micrographs of EPDM/NBR composites

Strong rubber-filler interaction is important to achieve maximum tensile strength by maximizing the nano dispersion of fillers. From the Figure 8.1, it is clear that the tensile strength of the EPDM/NBR composites increases with increasing content of nanoclay upto 7.5 phr and then decreases. Figure 8.2 represent the elongation at break. Tensile strength of the composites increases with increase in nanoclay content due to the fact that the crosslink density increases which in turn decreases mobility of rubber chains resulting in decreases in elongation at break. Hardness and rebound resilience of EPDM/NBR composites are presented in Figure 8.3 & Figure 8.4, respectively. From this result, hardness of the EPDM/NBR composites increases with increasing content of nanoclay. As expected when hardness increases rebound resilience decreases. The abrasion resistance of EPDM/NBR composites is shown in Figure 8.5. From this result, it can be observed that the abrasion loss decreases with increase in nanoclay filler. The higher weight loss infers the lower abrasion resistance of the composites. This increase in abrasion resistance of the composites depends upon hardness, and tensile strength.

The effect of nanoclay content on the swelling properties of penetrant (toluene, pentane and carbon tetrachloride) through EPDM/NBR composites was investigated. Figure 8.6 shows the swelling resistance in terms of mole percent uptake of the EPDM/SBR composites reinforced with nanoclay content. The uptake of toluene is reduced for the nanoclay filled EPDM/NBR composites compared to unfilled compounds. The graph clearly shows that the EPDM/NBR blend has the highest uptake, and incorporation of nanoclay in the composites, the mole percent uptake regularly decrease. The swelling of the penetrant solvent depends on the free space available in the polymer matrix to accommodate the penetrant molecule. The incorporation of nanoclay reduced the availability of these free spaces in the EPDM/NBR matrix and the restricted segmental mobility of EPDM/NBR matrix. The same trend is observed with pentane, and carbon tetrachloride as shown in Figure 8.6.

The surface morphology of the composites is shown in Figure 8.7. The surface morphology characterizes the arrangement of nanoclay molecules in EPDM/NBR composites.

Conclusions:

From the above experimental investigation about the influence nanoclay content in the EPDM/NBR composites the following conclusions are derived:

1. Tensile strength increases with increase of nanoclay upto 7.5 phr and then decreases.
2. As the tensile strength and hardness increases elongation at break and rebound resilience decreases.
3. Percentage increase in tensile strength is 99%.
4. Percentage decrease in elongation at break is 18%.
5. Abrasion and swelling resistance increases with increasing nanoclay loading.

References

1. Franta I (1989) Elastomers and rubber compounding materials: manufacture, properties and applications. Elsevier, Amsterdam

2. Morton M (1999) Rubber technology. Springer Science BusinessMedia, Dordrecht

3. Heinrich G, Klüppel M, Vilgis TA (2002) Reinforcement of elastomers. Curr Opin Solid State Mater Sci 6(3):195–203

4. Mark JE, Erman B, Roland M (eds) (2013) The science and Technology of Rubber. Academic Press, UnitedStates

5. Ray SS, Okamoto M(2003) Polymer/layered silicate nanocomposites: are views from preparation to processing. Prog Polym Sci 28(11):1539–1641

6. Gopi JA, Patel SK, Chandra AK, Tripathy DK (2011) SBR-clay carbon black hybrid nanocomposites for tire tread application. J Polym Res 18(6):1625–1634

7. Karger Kocsis J,Wu CM (2004) Thermoset rubber/layered silicate nanocomposites. Status and future trends. Polym Eng Sci 44(6): 1083–1093

8. Ma J, Xiang P, Mai YW, Zhang LQ (2004) A novel approach to high performance elastomer by using clay. Macromol Rapid Commun 25(19):1692–1696

9. Monfared A, Jalali-Arani A (2015) Morphology and rheology of (styrene butadiene rubber/ acrylonitrile–butadiene rubber) blends filled with organo clay: the effect of nanoparticles localization. Appl Clay Sci 108:1–11

10. Nematollahi M, Jalali-Arani A, Golzar K (2014) Organo clay maleated natural rubber nanocomposite. Prediction of abrasion and mechanical properties by artificial neural network and adaptive neuro-fuzzy inference. Appl Clay Sci 97:187–199

11. Thomas S, Stephen R (2010) Rubber nanocomposites: preparation, properties and applications. John Wiley & Sons, Asia, pp 169–173

12. Zhang L, Wang Y, Wang Y, Sui Y, Yu D (2000) Morphology and mechanical properties of clay/styrene–butadiene rubber nanocomposites. J Appl Polym Sci 78(11):1873–1878

13. Pal K, Rajasekar R, Kang DJ, Zhang ZX, Pal SK, Das CK (2010) Effect of fillers on natural rubber/high styrene rubber blends with nano silica: morphology and wear. Mater Des 31:677–686

14. Mishra S, Shimpi NG, Patil UD (2007) Effect of nano CaCO3 on thermal properties of styrene butadiene rubber (SBR). J Polym Res 14:449–459

15. LorenzH, Fritzsche J, Das A, Stockelhuber KW, Jurk R, Heinrich G (2009) Advanced elastomer nano-composites based on CNT-hybrid filler systems. Compos Sci Technol 69:2135–2143

16. Gatos KG, Karger-Kocsis J (2005) Effects of primary and quaternary amine intercalants on the organo clay dispersion in a sulfur –cured EPDM rubber. Polymer 46(9):3069–3076

17. Zheng H, Zhang Y, Peng Z, Zhang Y (2004) Influence of clay modification on the structure and mechanical properties of EPDM/ montmorillonitenano composites. Polym Test 23(2):217–223

18. Ahmed S, Basfar AA, Aziz MA (2000) Comparison of thermal stability of sulfur, peroxide and radiation cured NBR and SBR vulcanizates. Polym Degrad Stab 67(2):319–323

19. Meneghetti P, Shaikh S, Qutubuddin S, Nazarenko S (2008) Synthesis and characterization of styrene–butadiene rubber – clay nanocomposites with enhanced mechanical and gas barrier properties. Rubber Chem Technol 81(5):821–841

20. Sadhu S, Bhowmick AK (2004) Preparation and properties of styrene–butadiene rubber based nanocomposites: the influence of the structural and processing parameters. J Appl Polym Sci 92(2): 698–709

21. Durandish M, Alipour A (2013) Investigation into morphology, m i c r o s t r u c t u r e a n d p r o p e r t i e s o f SBR/EPDM/ organomontmorillonite nanocomposites. Chin J Polym Sci 31(4): 660–669

22. Naseri ASZ, Jalali-Arani A (2015) A comparison between the effects of gamma radiation and sulfur cure system on the micro structure and cross link network of (styrene butadiene rubber/ ethylene propylene diene monomer) blends in presence of nanoclay. Radiat Phys Chem 115:68–74

23. Bock J, Su SJ (1970) Interpretation of the Infrared Spectra of Fused Silica. J Am Ceram Soc 53:69–73

24. De Caillerie JB, De la Kermarec M, Clause O (1995) Si NMR observation of an amorphous magnesium silicate formed during impregnation of silica with mg (II) in aqueous solution. J Phys Chem 99:17273–17281

25. Hunt LP, Dismukes JP, Amick JA, Schei A, Larsen K (1984) Rice hulls as a raw material for producing silica. J Electrochem Soc 131: 1683–1686.

26. Sabanayagam S, Chockalingam S, (2019) Analysis of high temperature oxidation behaviour of SS316 by Al_2O_3 and Cr_2O_3 coating, Materials Today: Proceedings, https://doi.org/10.1016/j.matpr.2020.01.218