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Properties of SBR/NR Blend: The Efects of Carbon Black/Silica (CB/ SiO2) Hybrid Filler and Silane Coupling Agent

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Abstract

In the current research, investigation of natural rubber (NR)/styrene butadiene rubber (SBR) blend reinforced with carbon black (CB)/silica (SiO₂) and with and without silane coupling agent (SCA) was analyzed. The total hybrid filler (CB/SiO₂) concentration in the composite was fxed at 50 phr. Cure characteristics, mechanical properties and surface morphology were examined. The results revealed that as the $SiO₂$ content increase, the optimum cure time, scorch time, minimum torque, and maximum torque of $NR/SBR-CB/SiO₂$ composites with and without SCA increased. Except for maximum torque, the optimal cure time and minimum torque of $NR/SBR\text{-}CB/SiO₂$ composites with SCA were lower than those without SCA. Compared with NR/SBR composites with SCA, addition of $0/50$ CB/SiO₂ resulted in 53% decrease of tensile strength and 81% increase of elongation at break, superior to that of NR/SBR composites without SCA. The results revealed that as the $SiO₂$ content increase, the optimum cure time, scorch time, minimal torque, and maximum torque of $NR/SBR\text{-}CB/SiO₂$ composites with and without SCA increased. Except for maximum torque, the optimal cure time and minimum torque of NR/SBR -CB/SiO₂ composites with SCA were lower than those without SCA.

Keywords NR/SBR · Carbon black/silica · Silane coupling agent · Cure characteristics · Mechanical properties

1 Introduction

The development in industries and advancement in economic need results in never-ending demand for newer as well as low-cost materials. The polymer blending was a universal technology commonly used in the development of a newer product with superior properties. Because of its ability to adjust its shape crystallise under stretching, natural rubber (NR) has exceptional tensile properties [\[1,](#page-8-0) [2\]](#page-8-1), as well as better stress relief, electrical insulation, high abrasion, and fatigue resistance. NR, on the other hand, was vulnerable to ozone and had a lower resistance to petroleum-based fuels and oils [[3\]](#page-8-2). Because of its low cost, styrene butadiene rubber (SBR) was a commonly used rubber and was the frst synthetic rubber considered for manufacturing rubber components. Despite their strong mechanical properties,

 \boxtimes G. Anand anand.g@pec.edu SBRs have low abrasion resistance when exposed to conditions such as moisture, ozone, and sunlight. The presence of a double bond in the main polymeric chain was responsible for this [\[4](#page-8-3)[–6](#page-8-4)]. Due to their excellent mechanical and other engineering properties, NR and SBR blends are very appealing nowadays. Natural rubber (NR) offers stronger strength, reduced heat build-up, and better low-temperature performance than styrene butadiene rubber (SBR). SBR has superior crack resistance, wet grip, and weather resistance than NR. As a result, tyre manufacturers frequently use NR/ SBR mixtures. However, there are no defned principles for creating a compound for an NR/SBR blend in order to achieve a specifc set of mechanical properties [\[7\]](#page-8-5). SBR and NR are partially miscible. In SBR–NR systems, the largest amount of interface was observed at a 50:50 blend composition [[8](#page-8-6)]. Blends of natural rubber and butadiene rubber (NR/BR) or natural rubber and styrene-butadiene rubber (NR/SBR) are used in standard tyre formulations for cars and other vehicles. The blends of NR and SBR have been in use for a longer time period in many applications which also reduces the compounds cost [[9,](#page-8-7) [10\]](#page-8-8). Studies have been made by many researchers of such blends and the diferent properties are examined in detail [\[11](#page-8-9), [12](#page-8-10)].

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Despite NR and SBR are proven to have a wide range of desirable qualities, reinforcing fllers are often required to provide the desired properties for industrial applications. In the rubber industries, particle fllers are widely used for diferent of functions, the most important of which being reinforcements, material cost reduction, and processing improvements [[13\]](#page-8-11). More than 200 diferent types of organic and inorganic fllers have been examined and reported over the years [\[14](#page-8-12), [15](#page-8-13)]. Unfortunately, not all of them are reinforcing material, but some have limited use. Diferent fller materials, such as carbon black (CB) and silica $(SiO₂)$, are commonly used to enhance the mechanical properties of rubber composites [[16,](#page-8-14) [17](#page-8-15)], but talc and calcium carbonates can only be used for reducing the cost of compound [[18](#page-8-16)]. CB and $SiO₂$ have been commonly used as reinforcement materials in most engineering applications to improve the degree of rubber compounds. Tensile strength, tear strength, abrasion resistance, and modulus are all improved when CB is reinforced with either natural or synthetic rubber [[19](#page-8-17)]. As compared to silica reinforcement, CB reinforced rubber composites have a higher modulus. The interaggregate interactions of CB, the nature of the polymer, hydrodynamic efects due to CB, and extra cross-linkage due to fller-rubber linkage all infuence the elastic modulus of a rubber composite flled with CB. Size and surface area, surface activity and chemical properties, and form and structure are the key characteristics of carbon black fller. When it comes to interacting materials with rubber compounds, particle size and surface area are crucial. Carbon black particles range in size from 8 nm for furnace blacks to 300 nm for thermal blacks. Finer particles improve tensile strength, reinforcing, and abrasion resistance $[20]$. Whereas, silica offers properties such as improved tear strength, adhesion behavior, abrasion and aging resistance [\[21\]](#page-8-19). In comparison to CB reinforcement, silica reinforcement reduces rolling resistance, wet grip, and abrasion resistance in tyre treads [[22\]](#page-8-20). In general, tyre tread compounds have a high fller percentage (above 40 phr), and silica has been included in numerous tread compounds in the previous two decades [[23](#page-8-21)].

Since carbon black and silica both have their own set of benefts, using hybrid fllers (a mix of CB and silica) in rubber compounds can give you the best of both worlds. Wires and cloth coatings, tyre tread, rubber-covered belts, conveyor belts, hoses, engine mounts, cable jackets, and bumper strips can all be made with CB-reinforced rubber composites that have silica added to them $[24, 25]$ $[24, 25]$ $[24, 25]$ $[24, 25]$. The reinforcing efficiency of these fllers is known to be dependent on the kind of rubber matrix, fller parameters such as size, structure, and surface functioning, and mixing conditions [[26](#page-8-24)]. The surface chemistry of carbon black and silica is vastly diferent. On the carbon black surface, there are many functional groups such as hydroxyl, carboxyl, lactone, pyrone, ketone, quinone, and phenol, but the number is modest [\[27\]](#page-8-25). Silica is a fller for rubbers with particle sizes ranging from 10 to 40 nm. Chemically bound water makes up 25% of the silica, with adsorbed water accounting for the remaining 4–6%. In the polymeric industry, silica served as non-black reinforcing fller. In general, silica has a lot of polar groups on its surface, which helps the fller-fller interaction by hydrogen bonds to be stronger [\[28](#page-8-26)]. As compared to CB, adding silica to the rubber composite causes a slew of problems, most notably poor dispersion. Because the intermolecular hydrogen interactions between hydroxyl groups on silica's surface are especially strong, it can aggregate tightly $[29, 10]$ $[29, 10]$ [30\]](#page-9-1). Its property can result in a silica-flled rubber compound's scorch time being prolonged and its delta torque being reduced due to poor dispersion of curatives by silica. The introduction of a particular coupling agent [[31](#page-9-2)] will overcome the problems associated with silica incorporation. Silane coupling agents (SCAs) like bis(3-(triethoxysilyl) propyl)-tetrasulfde (TESPT) are commonly employed to increase fller dispersion and minimize curvative adsorption on the silica surface [[32](#page-9-3)[–35](#page-9-4)]. A siloxane bond is formed when the silane coupling agent combines with silanol on the surface of silica. A crosslink between the silica and the polymer is formed when a silane molecule bound to the silica surface reacts with a rubber molecule.

CB–silica dual phase fller is a popular commercial hybrid fller (CSDPF). Cabot Corporation [\[36\]](#page-9-5) produces it by pyrolyzing petroleum- and silicon-based feedstocks in a novel co-fuming process. CSDPF has a strong carbon domain surface activity and a high silica surface [[37](#page-9-6)]. In compared to traditional CB and silica fllers, it has been demonstrated to provide improved overall mechanical qualities. A typical truck-tread compound has some features that are superior to the vulcanizate flled with CB N110, such as abrasion resistance and low rolling resistance. Because of the high polymer–fller interaction and low fller–fller interaction, this is the case. The CSDPF is reported to have strong abrasion resistance and tear strength advantages over silica [\[38\]](#page-9-7). Despite this, the CB and silica ratio in CSDPF has remained unknown. The types and contents of fllers have an impact on the curing characteristics of flled rubber compounds [\[39–](#page-9-8)[42](#page-9-9)]. Silica-flled rubber compounds cure diferently than carbon black-flled rubber compounds [\[35,](#page-9-4) [43–](#page-9-10)[45\]](#page-9-11). The structure and surface chemistry of the reinforcing fller have an impact on crosslink density. A flled rubber compound cures faster than an unflled rubber compound. Carbon black improves reversal resistance by increasing crosslink density [[46](#page-9-12)].

The area between carbon black agglomerates decreases as carbon black loading increases, resulting in signifcant fller-fller interactions. This could be because gum vulcanizates have a lower cross-linking density than flled vulcanizates, which have a higher crosslinking density that increases as carbon black loading increases. This demonstrates carbon black's potential to provide flled vulcanizates more rigidity, which lowers the rubber chains' mobility [\[47](#page-9-13)].

On the other side, Bandyopadhyay et al. [[48\]](#page-9-14) found that adding silica to the rubber matrix reduces tensile characteristics due to less efficient interaction at the rubber/silica interfaces due to the aggregated silica particles' smaller surface area. The silane coupling agent is prevented from interacting with the silica by the adsorption of acrylonitrile-butadiene rubber on the silica surface. As a result, there will be less interaction between the fller and the rubber [\[49\]](#page-9-15).

To improve the mechanical properties of silica-flled rubber composites, silane coupling agents are used. It is required to neutralize silanol groups on silica's surface while also increasing silica distribution in rubber blends. Many studies have been done on the impact of silane coupling agents on the mechanical and physical properties of rubber composites [[50](#page-9-16)[–55\]](#page-9-17). From the literature, it was clear that not much systematic research work have been reported with $CB/SiO₂$ hybrid filler on the properties of NR/SBR composites. Replacement of CB with $SiO₂$ in passenger car tire tread compounds, after the patent of Michelin in 1992 [[56\]](#page-9-18), offers tire compounds with lower rolling resistance and higher wet grip [[57\]](#page-9-19), so less fuel consumption and better driving safety. The present study employs high specifc surface area and structure CB N330 in synergy with $SiO₂$ in NR/SBR tire tread composites. In the present work, the hybrid filler of $SiO₂/CB$ -filled NR/SBR with various ratios is investigated keeping the total hybrid fller content constant at 50 phr. The effect of CB N330/SiO₂ on the properties of NR/SBR composites with and without SCA is investigated with respect to cure characteristics, tensile properties and morphology, all of importance for tire performance. CB/ $SiO₂$ is not added beyond 50 phr due to the formation of cluster in rubber matrix and cost factor.

2 Experimental

2.1 Materials and Methods

The natural rubber (NR-RSS1) was purchased from Asian Rubber, Ambattur, Chennai, India and SBR(SBR-1502) was procured from Arihant Reclamation Pvt. Ltd., Delhi, India. The reinforcement (silica (SUPERSIL 140), carbon black (N330)), silane coupling agent (SCA), and curatives (zinc oxide, stearic acid, sulphur, mercaptobenzothiazyldisulphide (MBTS), tetramethylthiuramdisulphide (TMTD) andprocessing oils) were of commercial grade and purchased from Vignesh Chemicals, Chennai, India.

The alteration of silica with TESPT was carried out in solution to see how SCA affects silica reactivity and to eliminate any confounding efects of the NR/SBR matrix. In a three-necked fask, 10 g of silica were dispersed in 300 mL of toluene, followed by 1.0 g of SCA. The silanization reaction was carried out at 80 °C for 12 h with constant stirring. After that, the result was centrifuged and washed with ethanol and toluene multiple times to remove the ungrafted TESPT before being dried at 50 °C.

The NR/SBR composite was formulated with a blend ratio of 50/50 as shown in Table [1](#page-2-0). A two-roll mill at 50 °C was used to make the rubber compounds. NR was masticated for 4 min before being blended with SBR for another 4 min in the frst stage. Before incorporating constituent or reinforcing materials, it was important to soften the compound blend. The CB/ SiO₂ was incorporated in the NR/SBR blend with two major portions. Half of the constituents (zinc oxide and stearic acid) will be induced together after half of the $CB/SiO₂$ is added. The other half of $CB/SiO₂$ and its constituents were treated in the

Sl. No	Constituents (phr)											
		SBR	NR	CB	SiO ₂	SCA	Zinc oxide	Stearic acid	sulphur	MBTS	TMTD	Pro- cess- ing oil
	Without SCA	50	50	50	$\mathbf{0}$	٠	5	$\overline{2}$	2			5
2		50	50	40	10	٠	5	2	\mathfrak{D}			
3		50	50	30	20	٠	5	2	\overline{c}			ጎ
4		50	50	20	30	٠	5	\overline{c}	2			
5		50	50	10	40	۰	5	2	\overline{c}			
6		50	50	$\mathbf{0}$	50	$\overline{}$	5	\overline{c}	\mathfrak{D}			
7	With SCA	50	50	50	$\boldsymbol{0}$	3	5	2	\mathfrak{D}			
8		50	50	40	10	3	5	\overline{c}	$\overline{2}$			
9		50	50	30	20	3	5	\mathfrak{D}	$\overline{2}$			
10		50	50	20	30	3	5	\overline{c}	\overline{c}			
11		50	50	10	40	3	5	\overline{c}	\mathfrak{D}			
12		50	50	$\overline{0}$	50	3	5	\overline{c}	2		L	5

Table 1 Formulation for SBR/NR blend with and without SCA

same way. In the fnal stage, the sulphur was gradually added followed by MBTS and TMTD into the blend respectively. The total compounding time was done approximately from 25 to 35 min and compound dumping temperature were range from 50 to 80 °C.The prepared samples cure characteristics were investigated using a Monsanto Oscillating Disc Rheometer (ODR 2000) at 160 °C. The rubber compound was then compressed and moulded at 160 °C in an electrically heated hydraulic press in accordance with the cure times [\[58](#page-9-20)[–60\]](#page-9-21). A die cutter was used to make the dumbbell and nicked tab end- specimens.

2.2 Cure behaviors

The cure behaviour of the unvulcanized rubber samples was carried out as per ASTM D-2084 standard by oscillating disc rheometer at 160 °C, oscillation arc of 0.5° and 100 cycles per minute (1.66 Hz) conditions.

2.3 Mechanical properties

According to the requirements, dumbbell and nicked tab end shaped samples are cut from the moulded sheets. Tensile and tear checking was carried out in accordance with ASTM D 412 and ASTM D 624. Dak System Inc is used to perform tensile and tear tests at a crosshead speed of 500 mm/min (model: T-72102). The tensile strength, elongation at break and modulus were determined by Universal testing machine at 23 °C.The average values of six replicate measurements were used to arrive at the fnal outcome.

The samples hardness was determined using an ASTM D 2240 Shore-A Durometer. The hardness of composites was evaluated by the penetration of the Durometer tester into the rubber material, kept under formerly set conditions.The sheets with an effective thickness of 6 mm were used to test hardness.

The rebound resilience tests were carried out using a vertical rebound resilience tester in accordance with ASTM D 2632 guidelines. The rolling slide test was used to assess the abrasion resistance, which was determined as the relative volume loss from the wear test using a Zwick DIN Abrader (model: 6102), as per the ASTM D 5963 standard testing technique. A 0.1 kN force was given to the rolling sliding cylinder on SiC paper in this experiment. A minimum of fve measurements were taken, with the average values reported.

2.4 Crosslink density

Cure test pieces of 20 X 20 X 2 mm were weighed and immersed in a glass vessel containing toluene (50 mL) at 23 °C. To avoid oxidation, the vessel was kept in a dark place. The samples were removed from the glass difusion vessel and tissue paper was used to remove the excess toluene from the sample. To prevent toluene evaporation, the samples were stored in a closed vessel and the weights of the swollen samples were calculated. The sample was then re-immersed in toluene and the process was repeated until the sample reached a constant swollen weight. For the volume fraction of swelling rubber, the sample was de-dried in a vacuum at room temperature to a constant weight. The Flory-Rehner equation [\[61](#page-9-22)] was used to compute the molecular weight between two crosslinks (Mc) using the swelling data.

$$
M_c\left(\frac{g}{mol}\right) = \frac{-\rho_p V_s V_r^{1/3}}{\ln(1 - V_r) + V_r + \chi V_r^2}
$$
(1)

$$
V_r = \frac{1}{1 + Q_m} \tag{2}
$$

where, $\rho =$ density of the rubbers, $V_s =$ molar volume of the toluene, V_r =volume fraction of the swollen rubber, χ =interaction parameter of the NR/SBR rubber, and Q_m = weight swell of the SBR/NR rubber blends in toluene. The degree of crosslinking density (v) is given by;

$$
v\left(\frac{mol}{cm^3}\right) = \frac{1}{2M_c} \tag{3}
$$

The degree of crosslinking density of NR/SBR composites was determined using the following constant values.

- ρ (SBR)=0.921 g/cm3,
- $ρ$ (NR) = 0.912 g/cm3,
- *χ* (NR/SBR)=0.3557 [\[62\]](#page-9-23), and
- V_s 106.29 cm³/mol.

2.5 Scanning electron microscopy (SEM)

With an acceleration voltage of 3 kV and a film of gold over it, a scanning electron microscope (Hitachi S-2500, Japan) was used to research the surface morphology of the tensile fractured surface.

3 Results and discussion

3.1 Cure behavior

Figures [1](#page-4-0), [2](#page-4-1), [3](#page-4-2) and [4](#page-4-3) depicts the cure behaviours (optimum cure time, scorch time, minimum torque, and maximum torque) of $CB/SiO₂$ filler filled NR/SBR composites with and without silane coupling agent. The graph for the cure time is shown in Fig. [1.](#page-4-0) The optimal cure time for both composites increases as the silica content increases. This was due to the presence of the silanol group in it [\[63\]](#page-9-24). Silica's surfaces are normally hydrophilic and polar. It also

Fig. 1 The optimum cure time of $CB/SiO₂$ hybrid filler filled NR/ SBR composites with and without SCA

Fig. 2 The scorch time of CB/SiO₂ hybrid filler filled NR/SBR composites with and without SCA

has a high tendency for absorbing moisture, which has a negative impact on the rubber composites' cure properties [\[64\]](#page-9-25). Figure [1](#page-4-0) shows that when a silane coupling agent was added to an NR/SBR composite with a similar composite ratio, the optimum cure time (t_{c90}) was shorter than when the silane coupling agent was not added. The composites with the presence of silanol groups which results in causing less hydrophilic characteristic, thus ensures a faster optimum cure time [[65\]](#page-9-26). The silanol groups in sulfur-cured rubbers can cause accelerator adsorption, resulting in a delay in the vulcanization reaction and, as a result, a loss in crosslinked density [[66](#page-9-27)].

The graph for scorch time was shown in Fig. [2.](#page-4-1) The scorch time of hybrid fller-flled NR/SBR composites with

Fig. 3 The minimum torque of $CB/SiO₂$ hybrid filler filled NR/SBR composites with and without SCA

Fig. 4 The maximum torque of CB/SiO₂ hybrid filler filled NR/SBR composites with and without SCA

and without silane coupling agent increases as the concentration of silica in the composite increases. When comparing composites with and without SCA, it was discovered that composites with Si69 have a longer scorch time (t_2) , indicating that there is good reaction between the SCA-silica-NR/ SBR compounds in the composite. When the silica content is high, the amount of SCA utilised in this study is thought to be sufficient (as the SBR, NR, and SCA are constant except for silica content). As a result, the reaction between silica-SCA-NR/SBR was slightly increased. The NR/SBR tends to adsorbed on the silica surface in mixes without SCA, preventing the silica from adsorbing the accelerator and thereby reducing scorch time [\[67](#page-9-28)]. When silica concentration is high, especially greater than 30 phr, the scorch time increases steadily. The steady increase in scorch time when silica concentration is larger than 30 phr could be due to the fact that these compounds have had reduced heat history during mixing.

The incorporation of high specifc surface area and high structure CB N330 that has better compatibility with NR/ SBR compared to polar silica, causes an increased interfacial contact and consequently higher viscosity of the composites. The graph for the minimum torque was shown in Fig. [3.](#page-4-2) With an increase in silica concentration, the minimum torque of NR/SBR composites flled with hybrid fller with and without silane coupling agent increases. However, when comparing the composites with and without silane coupling agent, the composites with silane coupling agent exhibit a lower torque at all ratios, indicating that they are more processable [[68–](#page-9-29)[70\]](#page-9-30).

The graph for the maximum torque was clearly shown in Fig. [4.](#page-4-3) From the fgure it was clear that the maximum torque for NR/SBR hybrid composites with and without SCA was enhanced with an increase in silica concentration. Incorporation of hybrid fllers reduces the movement of the macromolecular chains in the composites, henceforth increases the maximum torque. However, when compared to NR/SBR composites without silane coupling agent, hybrid fller reinforced NR/SBR composites with silane coupling agent have a higher maximum torque value. The increased interaction between the hybrid fller and the NR/SBR matrix, especially in the presence of a silane coupling agent, may be responsible for the increased maximum torque [[71](#page-9-31)[–73](#page-9-32)].

3.2 Mechanical properties

From Fig. [5,](#page-5-0) [6](#page-5-1), [7](#page-6-0), [8](#page-6-1), [9](#page-6-2) and [10](#page-6-3), the mechanical properties of $CB/SiO₂$ hybrid fillers reinforced NR/SBR composites with and without silane coupling agent are shown. Figure [5](#page-5-0) shows the tensile strength of NR/SBR composites flled with hybrid fillers with and without a silane coupling agent. The figure clearly shows that both composites follow the same pattern, with the tensile strength of the composite decreasing as the silica content rises. The poor interaction of hybrid fller with NR/SBR rubber, as well as the rise in silica-silica interaction, resulted in a decrease in tensile strength at higher silica concentrations. Furthermore, as compared to the dispersion of carbon black, the dispersion of silica in rubber blends was very poor. This was due to the fact that silica forms a tight hydrogen bond, causing reinforcing material to clump together. The tensile strength of the hybrid composites was improved to a greater extent with the addition of a silane coupling agent. It was also apparent from the results that the composite with silane coupling agent had higher tensile strength than the composite without silane coupling agent at a comparable ratio of hybrid fllers (especially when the concentration of silica was more). The reinforcing material

Fig. 5 The tensile strength of $CB/SiO₂$ filler filled NR/SBR composites with and without SCA

Fig. 6 The 100% modulus of CB/SiO₂ filler filled NR/SBR composites with and without SCA

and the silane coupling agent formed a stable bond (Si–O-Si) when the silane coupling agent was incorporated into the composite [[74](#page-9-33)]. The presence of a silane binding agent improved the interaction of hybrid fllers in the NR/SBR matrix, resulting in better dispersion. By lowering the interfacial energy between the stages, the hybrid fller's interfacial adhesion to the NR/SBR matrix improves. The tensile strength of carbon black and NR/SBR matrix will be higher than silica due to good interaction and dispersion. In comparison to silica fller, carbon black fller has a stronger rein-forcing effect [[75\]](#page-9-34).

The graphs for 100 percent modulus and elongation at break of hybrid composites with and without silane coupling agent are shown in Figs. [6](#page-5-1) and [7](#page-6-0). With increasing silica

Fig. 7 The elongation at break of CB/SiO₂ filler filled NR/SBR composites with and without SCA

Fig. 8 The hardness of $CB/SiO₂$ filler filled NR/SBR composites with and without SCA

54 **Without SCA** 52 With SCA 50 Rebound resilience (%) 48 46 44 42 40 38 36 $40/10$ $30/20$ $20/30$ $10/40$ $50/0$ $0/50$ CB/SiO₂ (phr/phr)

Fig. 9 The rebound resilience of CB/SiO₂ filler filled NR/SBR composites with and without SCA

Fig. 10 The abrasion loss of $CB/SiO₂$ filler filled NR/SBR composites with and without SCA

concentration, the 100 percent modulus tends to decrease, while the elongation at break tends to increase. The matrixfller interaction decreases as the concentration of silica increases. In contrast, the modulus of hybrid composites with silane coupling agent is higher than that of composites without silane coupling agent at equivalent composite ratios. This was due to the presence of a silane binding agent, which increased cross-link density and improved fller dispersion. The elongation at break values increase with increasing silica concentrations when a silane coupling agent is added. This is because the silane coupling agent aided chemical bonding between the hybrid fller and the NR/SBR matrix, increasing elongation at break values [[76](#page-9-35)]. Elongation at break increment due to the interfacial adhesion force and movement of rubber molecular chains. Because of the poor dispersion of fller in NR/SBR composites flled with CB/ silica, the interfacial adhesion force between rubber and particles is weakened, then lots of rubber molecular chains can move easily and get sufficient extension when composite is stretched, and that induces a high elongation at break.

Figures [8](#page-6-1) and [9,](#page-6-2) clearly indicates the hardness and rebound resilience of hybrid fllers reinforced NR/SBR composites with and without silane coupling agent. The hardness of the composite tends to decreases whereas the rebound resilience increases with the increase in the concentration of silica in it. Lower crosslink density (as illustrated later in Fig. [11\)](#page-7-0) of the composite was responsible for the minimum hardness. This was because, as the crosslink density of the

composite decreases, the harder matrix turns into a softer one [[77,](#page-9-36) [78](#page-9-37)]. The increase in the concentration of silica leads in decrease in the rubber-fller interaction. Nevertheless, at a similar composite ratio, the hardness and rebound resilience of hybrid composite with silane coupling agent shows a greater value as compared to the composite without silane coupling agent [\[79\]](#page-9-38).

The Fig. [10,](#page-6-3) show the graph for the abrasion loss of hybrid NR/SBR composites with and without SCA. The graph shows that as the concentration of silica increases, the abrasion resistance decreases. The abrasion resistance of the composites was measured by the friction coefficient and by the modulus of the composite [[80\]](#page-9-39). The lower frictional coefficient and higher modulus indicates that the composite have a better abrasion resistance. The diferences among the composites that are filled with $CB/SiO₂$ and $CB/silical$ SCA could match the theory of modulus and dispersion of fller well according to what we have discussed in Fig. [5.](#page-5-0) As compared to composites with mediocre fller dispersion, the composites with excellent fller dispersion were said to have improved abrasion properties [[81–](#page-9-40)[84\]](#page-9-41). As a result,

Fig. 11 The crosslinking density of $CB/SiO₂$ filler filled NR/SBR composites with and without SCA

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the histrionic reduction in abrasion resistance at high silica content was expected to be due to weak silica dispersion or a heavy filler–filler relationship [[85\]](#page-9-42). As to the CB/SiO₂ composites with this experimental conditions, the friction coefficient might have a great influence on the wear resistance of composites [\[86](#page-9-43)].

The Fig. [11,](#page-7-0) show the graph for the crosslinking density of hybrid NR/SBR composites with and without SCA. Increases in silica concentration result in a decrease in fllerrubber interaction (increases in silica-silica interaction), afecting the crosslinked density value [\[87](#page-9-44)[–90](#page-9-45)]. The tensile strength, tensile modulus, elongation at break, and hardness of hybrid fllers flled NR/SBR blends with SCA are higher than those of NR/SBR blends without SCA at similar blend ratios.In the presence of SCA, this is simply owing to enhanced fller dispersion and greater crosslinked density. The SCA signifcantly reduced fller-fller interaction, resulting in enhanced rubber-fller interaction and improved fller dispersion.

3.3 Scanning electron microscopy

The Fig. [12a](#page-7-1)-[b](#page-7-1) shows the SEM micrographs of tensile fractured specimen reinforced with hybrid fllers with and without silane coupling agent at 20/30 composite ratio. Figure [12a](#page-7-1) shows a rougher surface with numerous tear lines, indicating that more energy was needed to break the composite than Fig. [12b](#page-7-1), which was nearly smooth. This is because the presence of a silane coupling agent improves the interaction of the CB and silica with the NR and SBR rubber matrix.In SEM micrographs of SBR/NR composites, the black region represents the rubber matrix and CB/ $SiO₂$ hybrid filler, while the white line represents the silane coupling agent. The cross-section of silane coupling agent is represented by the white lines or areas. The uniformly distributed hybrid fllers in NR/SBR matrix have an ability to resist the crack initiated and propagates which results in better tensile property of the composite [\[91\]](#page-9-46). The addition of the silane binding agent to the composite improves fller dispersion and decreases the silica-silica interface, resulting

Fig. 12 The tensile fracture surfaces of hybrid fllers flled NR/SBR composites with $\times 300$ magnifcation, (**a**) with silane coupling agent at 20/30 composite ratio, (**b**) without silane coupling agent at 20/30 blend ratio

in improved matrix-reinforcing material interaction. Further, due to the development of hydrogen bonds between the silanol groups on the silica surface, the composites without SCA are non-uniformly dispersed in SBR/NR matrix with agglomerates [\[92\]](#page-9-47). In contrast, in composites containing SCA, practically all of the silanol groups on the surface of the silica particles were reacted, causing the silica fller to be compactable with the SBR/NR rubber matrix and welldispersed in SBR/NR composites, as illustrated in Fig. [12a.](#page-7-1)

4 Conclusions

The optimum cure time of hybrid fllers reinforced NR/ SBR composites with silane coupling agent was less than the composite without silane coupling agent, according to the report. This was attributed to the decrease in curative adsorption phenomena observed on the silica surface.The scorch time of the hybrid fller reinforced NR/SBR composites with silane coupling agent is higher. With increasing silica concentration, the minimum and maximum torque for the composite with and without silane coupling agent increases. Hybrid fller reinforced NR/SBR composites with silane coupling agent had higher tensile strength, modulus, elongation at break, hardness, rebound durability, and abrasion resistance than composites without silane coupling agent.This was due to the composite's improved matrixfller interaction due to the presence of the silane coupling agent. The presence of silane coupling agent in hybrid fllers reinforced NR/SBR composites is revealed by SEM micrographs, which reveal better dispersion of the hybrid fllers in the NR/SBR matrix.

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Author contributions 1. G. Anand – Corresponding Author – First Author – Fabricated the composite samples and conducted the testing as per the ASTM standard and have written the manuscript, both authors read and approved the manuscript.

2. S. Vishvanathperumal – Second Author - Helped in Fabrication the samples and conducted the testing as per the ASTM standard and helped in writing the manuscript, both authors read and approved the manuscript.

Data Availability All data generated or analyzed during this investigation are included in this published article.

Declarations

Competing Interests None of the authors have any relevant fnancial or non-fnancial competing interests and any confict of interest to publish the article.

Conflict of Interest The authors declare no confict of interest.

Compliance with ethical standards No, all testing are conducted in this study as per ASTM standards.

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