# Carbon Black and Lignin Hybrid Filler in Designing NR Based Cushion Gum Composite Formulation

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**Highlights:**

- Lignin and pine tar oil are potential alternative bio-based rubber chemicals.
- The adhesiveness characteristic of cushion gum is affected by rubber chemicals.
- Good cushion gum improves the performance of retread tires.

**Abstract**. Lignin has high potential to be used as rubber cushion gum compound ingredient due to its unique properties. Cushion gum is an adhesive used to bind a new tread to the surface of used tires in a retread tire factory. An experiment was carried out to evaluate the performance of hybrid filler consisting of carbon black and lignin in affecting the physical and mechanical properties of natural rubber (NR) based cushion gum composite. The composition of carbon black/lignin was arranged at 40/10 phr and 35/15 phr. Carbon black grade N220, N330, and N550 were used in this research. Meanwhile, the lignin was specifically in the form of sodium lignosulfonate. The cushion gum compound was also formulated with the addition of pine tar oil as bio-based plasticizer. Commercial cushion gum was used as a comparison. The result showed that the hybrid filled NR cushion gum composite with N330/lignin at 40/10 (coded as SP2) was the most appropriate formula. Compared to commercial cushion gum, the SP2 hybrid filled cushion gum had comparable or even better processability and mechanical properties, attributed to high crosslink density. The excellent adhesive strength of the SP2 NR cushion gum composite was due to the presence of high content of the phenolic functional group in lignin and pine tar oil.

**Keywords:** *composite; cushion gum; hybrid filler; lignin; natural rubber; retread tire.*

## 1 Introduction

The tire retreading industry is the second largest domestic natural rubber consumer in Indonesia. The industry is growing rapidly since tire retreading offers economic benefit by extending the service life of tires. It also promotes environmental protection and energy conservation [1]. After frequent exploitation, a used tread tire can be further processed in a certain way, either by cold or hot retreading to allow reuse [2]. Retread tires reduce the amount of

hazardous waste ending up in the environment because of tire replacement due to their complex structure and varied composition [3-5].

Retreading is one the processes in the tire remanufacturing process. A retread tire is constructed from three layers of rubber composite, namely the used tire (casing layer), the cushion gum layer, and the new tread layer. Cushion gum functions as an adhesive layer. In the case of the cold retreading process, the new tread is already molded, cured, and applied to the outer casing surface with the help of the cushion gum compound layer [6,7]. In reference to its function, the cushion gum compound must have high adhesive strength in rubber-to-rubber bonding. Generally, the adhesion ability of a rubber compound is influenced by the chemical composition of the rubbers, their compatibility factor, their molecular weight and distribution, the additives used, their amorphous and crystalline contents, their surface nature and chemistry, and their crosslink density [8].

Some rubber chemicals used as rubber compounding ingredients that play an important role in adhesion strength (rubber to rubber or rubber to metal) are tackifier resin, plasticizer or rubber processing oil (RPO), and filler [9-12]. The addition of tackifier resin, mainly coumarone or phenolic resin types, into the cushion gum rubber compound formula results in an increase of green tack to facilitate build-up assembly and to achieve adequate adhesion. A special type of rubber plasticizer that can build tack or autohesion is pine tar oil. Pine tar oil is a sticky dark liquid and is a promising alternative bio-based rubber processing oil to substitute aromatic oil. Reinforcement of rubber by adding appropriate filler is primarily required to enhance tensile strength, tear strength, abrasion resistance, hardness, and modulus [13]. Commonly, carbon black and silica are used as the main reinforcing fillers in downstream rubber industries [14].

Recently, the hybrid filler system in rubber composite technology has attracted the attention of many researchers. According to Salim, *et al.* [15], the hybrid filler system is defined as two or more reinforcing and non-reinforcing fillers mixed in a single matrix. The combination of petro-based and bio-based reinforcing fillers has become the first choice in view of environmental and health concerns. Lignin is regarded as an interesting new material for bio-based reinforcing fillers in the rubber and plastic industry owing to its abundant availability, low cost, sustainability, and renewability [16,17]. Lignin, which can be categorized as a lignocellulosic class of biomass, has many functional groups on its surfaces, such as methoxyl, phenolic, hydroxyl, and carbonyl groups. The functional groups can adjust polarity to produce compatibility with appropriate matrix polymers [18-21]. The phenolic group in the lignin surface makes it a potential substitute for adhesives [22]. Jagadale, *et al.* [23] has explored the usage of lignin as plasticizer in nitrile rubber due to its aromatic content (methoxy and hydroxyl group).

Meanwhile, lignin can also be used as antioxidant after being modified by NaOH extraction [24].

Puspitasari, *et al.* [25] have reported that the usage of 5 phr coumarone resin and 20 phr of pine tar oil, resulted from a cushion gum composite that was reinforced by the addition of 50 phr N330 carbon black, had level of hardness at 57 Shore A, tensile strength at 25.72 N/mm$^2$, and elongation at break at 620%. In accordance with this previous research report and the potential lignin usage described above, it is understood that the implementation of lignin in cushion gum production has potential to partially substitute carbon black as reinforcing filler and eventually improve cushion gum properties.

This study was aimed at evaluating the effect of a hybrid filler consisting of carbon black and lignin, specifically in the form of sodium lignosulfonate, on the performance of natural rubber-based green cushion gum composite that was also formulated by using pine tar oil as bio-RPO.

## 2      Material and Method

### 2.1      Material

The preparation of NR-based green cushion gum compound used Technically Standard Natural Rubber Grade Standard Indonesian Rubber (SIR 20) obtained from CV INIRO Indonesia. Three types of carbon black (CB) were tested in this work, grade N220, N330, and N550. All types of carbon black were manufactured by OCI Company Ltd, Republic of Korea. Sodium lignosulfonate, hereinafter called 'lignin', was purchased from LUG Chemical Indonesia. It was derived from fermented spruce wood sulfite liquor and used without further purification or chemical modification.

Pine tar oil as bio-based rubber processing oil was supplied by PT Organik Inti Indonesia. Zinc oxide (Lanxess), stearic acid (Rhein Chemie), 2,2,4-trimethyl-1,2-dihydroquinoline (TMQ, Lanxess), N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine (6PPD, Sinorgchem), paraffin wax (Rhein Chemie), coumarone resin (Lanxess), N-cyclohexyl-2-benzothiazole sulfenamide (CBS, Kemai), and sulfur (Miwon) were provided by PT Multi Citra Chemindonusa Indonesia. Commercial cushion gum (CCG) compound, kindly provided by a retread tire factory located in Bogor, was used as control.

The cushion gum-based natural rubber compound formulations are listed in Table 1. This formula was designed with the addition of 35 phr and 40 phr of three grades of CB (N220, N330, N550) to be combined with 10 phr and 15 phr of sodium lignosulfonate (lignin). The rubber composites were coded in accordance

with the hybrid filler composition (CB/lignin), i.e. SP1-SP3 for 40/10 phr and
SP4-SP6 for 35/15 phr.

**Table 1**    Green cushion gum compound formula.

| Materials | Composition (per hundred rubber, phr) | | | | | |
|---|---|---|---|---|---|---|
| | SP1 | SP2 | SP3 | SP4 | SP5 | SP6 |
| SIR 2 | 100 | 100 | 100 | 100 | 100 | 100 |
| ZnO | 5 | 5 | 5 | 5 | 5 | 5 |
| Stearic acid | 2 | 2 | 2 | 2 | 2 | 2 |
| TMQ | 2 | 2 | 2 | 2 | 2 | 2 |
| 6PPD | 2 | 2 | 2 | 2 | 2 | 2 |
| Paraffin wax | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 |
| Coumarone resin | 5 | 5 | 5 | 5 | 5 | 5 |
| Pine tar oil | 5 | 5 | 5 | 5 | 5 | 5 |
| Sodium lignosulfonate | 10 | 10 | 10 | 15 | 15 | 15 |
| CB N220 | 40 | | | 35 | | |
| CB N330 | | 40 | | | 35 | |
| CB N550 | | | 40 | | | 35 |
| CBS | 1.6 | 1.6 | 1.6 | 1.6 | 1.6 | 1.6 |
| Sulfur | 2.2 | 2.2 | 2.2 | 2.2 | 2.2 | 2.2 |

## 2.2    Experiment

Hybrid-filled rubber cushion gum composite was prepared by using a laboratory-
scale two-roll open mill (Berstorf) with the compounding procedure described in
ASTM D 3182. Firstly, SIR 20 was masticated into a softening mass to facilitate
the mixing of the rubber chemicals, mainly high content of hybrid filler. The
rubber ingredients were added continuously into the softening rubber, in the
following order activator: (ZnO, stearic acid), resin (coumarone), antidegradant
(TMQ, 6PPD, paraffin wax), RPO (pine tar oil), filler (CB/lignin), accelerator
(CBS), and finally vulcanizing agent (sulfur). The rubber mixture was then
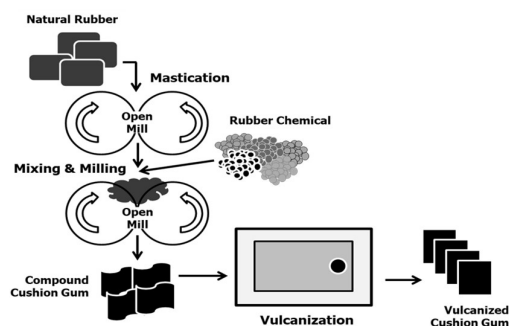blended and milled before being kept at room temperature for at least 20 hours.



**Figure 1**   Research stages.

## 2.3　Analysis and Calculation

For the curing characteristics analysis (ASTM D 2084-17), 50 g of SIR 20/CB/lignin composite was sampled by using an Alfa 2000 Moving Die Rheometer (Alfa Technologies, Akron, USA) at 150 °C for 30 minutes. Furthermore, the residual composite was press-cured into a vulcanized rubber plate at a temperature of 150 °C for the optimum cure time (tc90) obtained from the curing characteristic analysis by using a hydraulic press machine.

Rubber vulcanizates were used as the samples for the evaluation of mechanical properties, including the following parameters: hardness (ASTM D 2240-15, Frank Durometer Shore A), tensile strength and elongation at break (ASTM D 412-16, UTM Llyod 2000R), modulus 300% (ISO 37, UTM Llyod 2000R), tear strength (ISO 34, UTM MTS), specific gravity (ASTM D 297-2015), and adhesion strength (ASTM D 413, UTM Instron). The reinforcement index of the hybrid filler was evaluated in accordance with the analysis developed by Aini, *et al.* [21].

Crosslink density was determined referred to the Mooney-Rivlin theory, applying the stress ($\sigma$)-strain ($\lambda$)correlation approach. Stress-strain data were obtained from one cycle of tensile strength analysis by using UTM MTS in accordance with ASTM D 412. The data was then plotted in an XY graph with X as $\lambda^{-1}$ and Y as $\sigma/2(\lambda-\lambda^{-2})$. The intercept of the graph ($C_1$) correlated with crosslink density ($V_e$) is shown in Eqs. (1) and (2).

$$\sigma = 2\left(C_1 + \frac{C_2}{\lambda}\right)\left(\lambda - \lambda^{-2}\right) \tag{1}$$

$$V_e = \frac{2C_1}{\rho\,R\,T} \tag{2}$$

## 3　Result and Discussion

## 3.1　Curing Characteristics

The curing characteristics analysis evaluates the rheological property of rubber compounds. This analysis includes parameters such as maximum torque (MH), minimum torque (ML), scorch time (ts2), 90% of optimum curing time (tc90), and curing rate index (CRI). Torque differences (MH-ML) reflect the crosslink density. Scorch time indicates the processability of the rubber compound, while optimum curing time is the time required by the rubber compound to reach 90% state of cure [26].

Table 2 summarizes the curing characteristics of the NR-based cushion gum as a result of the addition of various hybrid filler types and content.

**Table 2** Curing characteristics of cushion gum compounds.

| Conditions | MH (dNm) | ML (dNm) | MH-ML (dNm) | tc90 (Minute) | ts2 (Minute) | CRI |
|---|---|---|---|---|---|---|
| CCG (control) | 12.20 | 0.82 | 11.38 | 9.18 | 1.59 | 13.18 |
| SP1 | 8.19 | 0.31 | 7.88 | 7.47 | 3.07 | 22.73 |
| SP2 | 10.19 | 0.70 | 9.49 | 7.39 | 2.46 | 20.28 |
| SP3 | 8.65 | 0.46 | 8.19 | 7.30 | 3.23 | 24.57 |
| SP4 | 7.84 | 0.44 | 7.40 | 7.57 | 3.33 | 23.58 |
| SP5 | 8.71 | 0.70 | 8.01 | 7.26 | 3.04 | 23.70 |
| SP6 | 9.90 | 0.93 | 8.97 | 7.15 | 2.45 | 21.28 |

From Table 2, it can be understood that at the same composition of hybrid filler, N220 carbon black resulted in relatively the lowest maximum and minimum torque values. This indicates that incorporation of hybrid filler consisting of N220/lignin (SP1) into the natural rubber matrix led to the lowest viscosity and modulus of the rubber cushion gum composite. Accordingly, the cushion gum compound designed with SP1 formula had better processability. Considering the hybrid filler composition, it was seen that the CB/lignin composition at 35/15 phr had a greater effect on decreasing the maximum torque value but increasing the minimum torque occurred primarily with the N220 and N330 formulas. Meanwhile, N550 showed the opposite effect. The mixture of N550/lignin at higher lignin loading increased the torque values. This shows that the hybrid filled cushion gum compound containing N550 carbon black created more interaction between hybrid filler particles and rubber molecules, which restricted the mobility of the rubber chain. This may be because N550 has a larger average particle diameter than N220 and N330.

Compared to the commercial cushion gum, which only contains carbon black, the minimum and maximum torque values of the hybrid filled cushion gum compounds decreased with the presence of lignin as bio-filler. This is due to the weaker interfacial adhesion between the lignin and the rubber matrix segment. This finding is in line with previous research carried out by Wang, *et al.* [27]. In the previous study, lignin was also found to act as a plasticizer agent by observing the reduction trend in minimum torque value [21]. The commercial cushion gum compound showed the longest optimum curing time, however, in contrast, it had the shortest scorch time. Nevertheless, it cannot be directly compared to hybrid filled cushion gum compound since an unidentified curing system was selected by the manufacturer during the formulation of the commercial cushion gum.

Gradual substitution of carbon black with lignin tends to delay the initial stage of the vulcanization reaction of the rubber molecules, shown by the increasing scorch time trend. The hindered phenolic group contained in the lignin molecular chain plays the role of radical scavenger in the vulcanization reaction [28]. The surface of the lignin molecule also potentially adsorbs the rubber chemicals as

activator (zinc complex), which contributes to the disruption of sulfur vulcanization efficiency [21]. Thus, the incorporation of lignin could lower the crosslink density of the rubber composites. Moreover, after reaching the curing stage, the curing rate increased significantly. This could be attributed to the presence of free carboxylic acid in lignin and pine tar oil, which led to an increased optimum cure time due to their acidity [29]. A long scorch time and a short optimum curing time have been determined as an appropriate condition, since they can provide safety and consume less energy during the manufacture of rubber compounds.
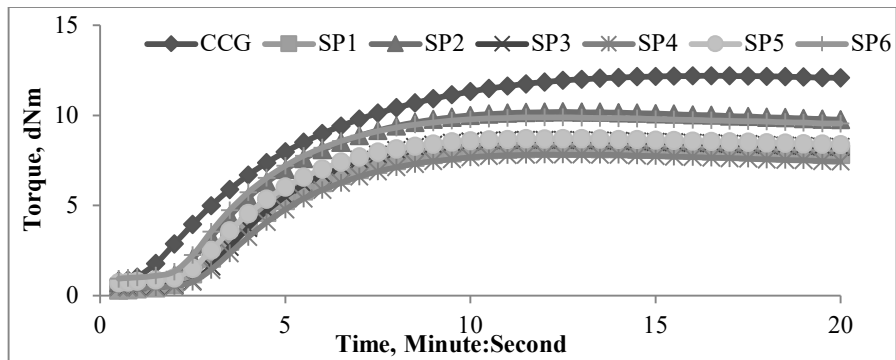


**Figure 2** Rheograph of the cushion gum compound.

The rheograph of the hybrid filled cushion gum compound in Figure 2 shows an identical pattern as the commercial cushion gum (CCG). The curing behavior of rubber compounds is strongly affected by the curing system. In this research, the cushion gum formulation was designed with a conventional curing system. Conventional curing systems also play role in delaying the curing reaction, aided by the inclusion of lignin in the formulation of the cushion gum. This mechanism was confirmed by the induction phase of the rheograph. The induction phase is measured in accordance with tc10 (10% of cure state). The tc10 value of all hybrid filled cushion gum compounds was higher compared to that of the commercial cushion gum.

In the curing phase of the rheograph, it can be seen that commercial cushion gum has the highest curve, followed by hybrid filled cushion gum designed with the SP2 and SP6 formulas. The mechanism of the curing phase of the rubber cushion gum compound was proportional to the different torque values (MH-ML), which indicate the degree of crosslink density. A slightly lower crosslink density of the hybrid filled cushion gum compound was predicted due to poor dispersion ability and interaction of the lignin with the rubber matrix. The interaction between the

filler and the rubber is affected by their compatibility. Lignin filler is known to have low compatibility with non-polar rubber matrices such as natural rubber. Consequently, it exhibits less crosslinked networks in the composite and the reinforcement performance of lignin is not as good that of carbon black alone [20].

The difference in curing behavior was also seen in the post-cure phase. From Figure 2, the rheograph of the commercial cushion gum showed a plateau curve, which indicates better thermal resistance. Otherwise, the reverse tendency was observed in the post-cure phase of the hybrid filled cushion gum compound. The formulation of the hybrid filled cushion gum compounds was designed with a conventional curing system. The system produced more poly-sulphidic bridges, which have lower thermal resistance due to the low S-S bonding energy. Poly-sulphidic bridges are easily changed into di-sulphidic and mono-sulphidic bonds by thermal scission during the vulcanization reaction [30].

## 3.2    Crosslink Density

Similar to the MH-ML value resulted from the curing analysis, the crosslink density measurement of the commercial cushion gum by the Mooney-Rivlin approach showed a relatively higher degree than the hybrid filled cushion gum compound, as shown in Figure 3. Figure 3 also confirms that the addition of a higher dosage of lignin to the hybrid filler that was composed of N220 and N330, reduced the crosslink density. Furthermore, with the ratio of N550/lignin at 35/15, the crosslink density was also slightly increased compared to the ratio of 40/10. The SP2 formula designed with CB N330/lignin at 40/10 had the closest degree of crosslink density to the commercial cushion gum.
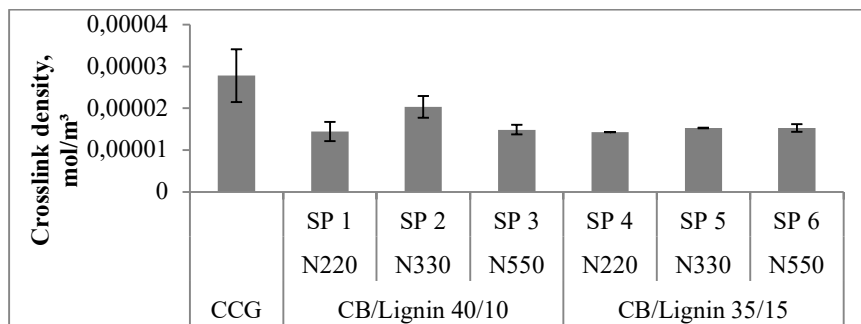


**Figure 3**  The crosslink density of the cushion gum compounds.

### 3.3 Hardness and Specific Gravity

The hardness of rubber composites has a strong correlation with the specific gravity due to the selection of filler type and dosage, attributed to the natural rubber compound formulation. High hardness is commonly achieved by the incorporation of high loading filler as hard material. Thus, high specific gravity is produced, which increases the rubber composite's weight. Comparing all hybrid filled cushion gum composites, the composition of N330/lignin at 40/10 (SP2) showed a desirable hardness value, even though it had the highest specific gravity value, as can be seen in Figure 4. Nevertheless, the SP2 cushion gum composite exhibited a lower specific gravity value than the commercial cushion gum. The harder rubber cushion gum composite originating from the SP2 formula may also be due to the influence of the high degree of crosslink density.
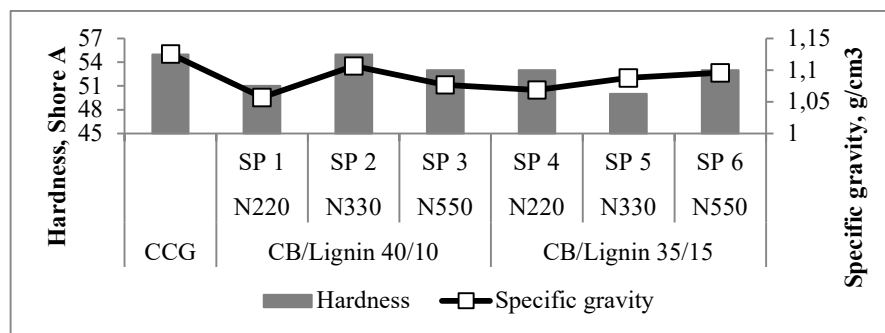


**Figure 4** Correlation of hardness and specific gravity of the cushion gum compounds.

### 3.4 Tensile Properties

The failure properties of rubber composite are reflected by tensile strength and elongation at break. Both parameters truly indicate the reinforcement behavior of the hybrid filler, while the tensile modulus 300% in this experiment captures only the effect of the stiffness of the hybrid filler particles [19]. The relation between tensile properties such as tensile strength and tensile modulus 300% and elongation at break of the rubber cushion gum composite are depicted in Figures 5 and 6, respectively. As can be seen in Figure 5, the tensile modulus 300% of the hybrid filled cushion gum compounds exhibited decreases with an increase of the lignin content, except the hybrid filled cushion gum compound using N220. For SP4 (N220/lignin 35/15), the tensile modulus 300% showed a higher value than the other hybrid filled cushion gum composites. The inclusion of lignin into the SP4 formula plasticized the rubber composite, which facilitated rubber chain

mobility and the dispersion of the carbon black resulted in higher elasticity. It is well known that the tensile modulus is correlated with composite elasticity [31].

It was found that an increasing amount of lignin combined with carbon black type N330 and N550 led to a drop in tensile strength, as can be seen in Figure 5. The reverse trend was shown in the case of the elongation at break parameter (Figure 6). A similar finding has been reported by Aini, *et al.* [21]. Datta & Parcheta [31] state that a decrease in tensile strength can result from filler-dependent factors. Partially dispersed or irregularly shaped filler particles cause stress concentrations, which result in faster rubber composite breaking. Furthermore, an increased amount of lignin in the composite with a lower particle size of carbon led to a more rigid composite due to the molecular chain mobility being hindered by the occurrence of filler agglomeration in the composite microstructure. The filler agglomeration may be attributed to low compatibility between lignin with natural rubber, which further causes low rubber-filler interaction.
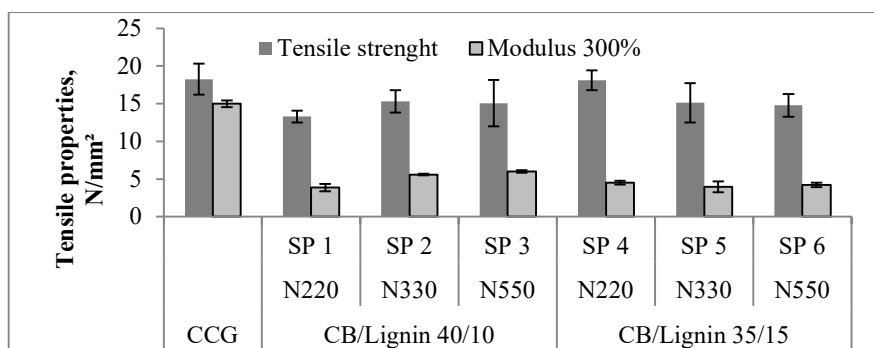


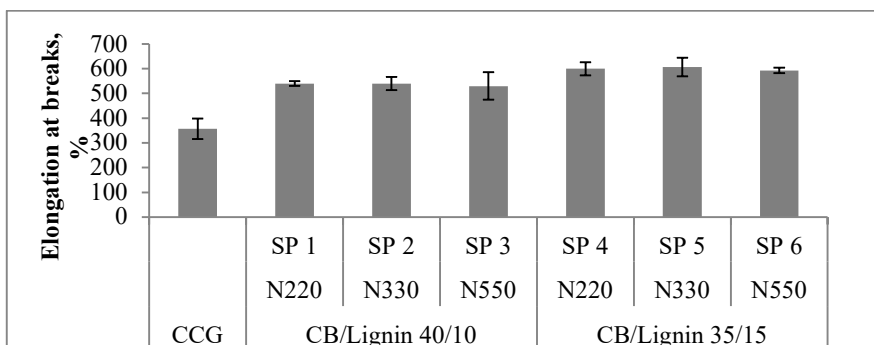**Figure 5** Tensile strength and modulus 300% of cushion gum compound.



**Figure 6** Elongation at break of the cushion gum compounds.

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It should be mentioned that in terms of the elongation at break parameter, the addition of lignin had no negative impact on the elongation character of the rubber cushion gum composites. In every type of hybrid filler, the elongation at break of the rubber cushion gum composite composed of 35/15 hybrid filler was always higher than that of the rubber cushion gum composite filled with 40/10 hybrid filler. It is believed that the type of carbon black dominated the elongation at break value of the rubber cushion gum composites. The elongation at break increased gradually with the reduction of the carbon black particle size. The commercial cushion gum solely containing carbon black showed the highest tensile strength and tensile modulus 300%, but the lowest elongation at break, since carbon black has greater dispersibility into rubber composites than lignin.

## 3.5 Tear Strength

The tear strength, which represents crack propagation resistance, is considered to be an important property of cushion gum in tire tread application. Excellent tear strength provides good quality of the retread tire, since defects due to tearing, which are often found in retread tires, can be minimized. Eventually, it can avoid tearing of the new tread on the surface of a used tire. The obtained tear strength value of both the commercial cushion gum and the hybrid filled cushion gum composites are presented in Figure 7. Lignin inclusion in the hybrid filled rubber cushion gum composite resulted in relatively lower tear strength. Unlike the other composites, the hybrid filled rubber cushion gum composites with N330/lignin at 40/10 (SP2) and N550/lignin at 35/15 (SP6) resulted in higher tear strength values compared to the commercial cushion gum. The improvement of tear resistance for the SP2 and SP6 formulas was presumably due to their higher degree of crosslink density.
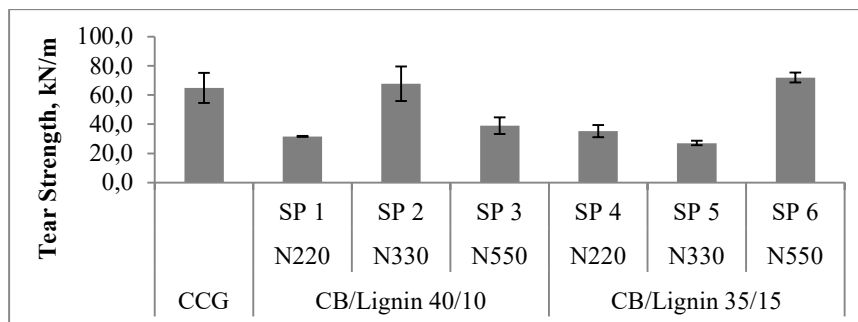


**Figure 7** Tear strength of cushion gum compounds.

### 3.6 Adhesive Strength

Rubber adhesion to other materials is mainly affected by the surface tension or surface energy of solids [32]. In addition, the adhesive strength is influenced by the chemical composition of its ingredients. The phenolic nature of lignin makes it prone to substitution of phenol-formaldehyde petro-based resin in the formulation of adhesives [33]. Pine tar oil, which was used as bio-based rubber processing oil in this research, also contains a phenolic functional group, which is a viscous sticky black liquid. The mixture of lignin and pine tar oil increases the adhesive strength of the hybrid filled rubber cushion gum composites, as shown in Figure 8. The hybrid filled cushion gum composite containing 40 phr N330 and 10 phr lignin (SP2) was selected as the best formulation, since it combines good processability with better physical-mechanical properties, although its adhesive strength is weaker than that of the hybrid filled cushion gum composed of N550. Lignin dosage at 15 phr revealed a reduction in the adhesive strength of the hybrid filled cushion gum composites. It is presumed this is because of the agglomeration of lignin particles among the rubber molecular chain.
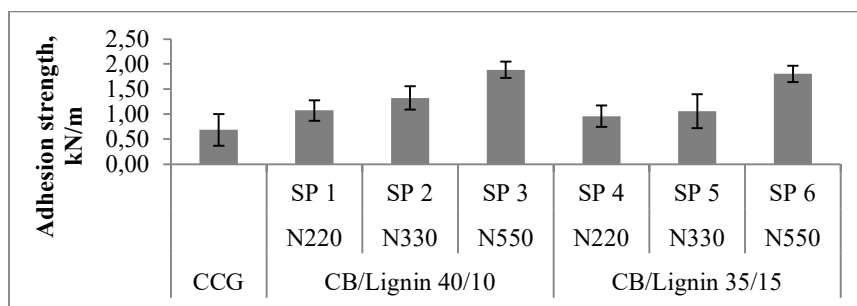


**Figure 8** The adhesive strength of the cushion gum compounds.

### 3.7 Reinforcement Index (RI)

The reinforcement index (RI) of the hybrid filler was measured by comparing the tensile modulus at 100% and 300% (M300/M100). Although RI was not involved in the evaluation of cushion gum specification, it is important to identify the influence of filler on the stress-strain curves of the rubber compounds [34]. The result of the RI measurement is illustrated in Figure 9. From Figure 9 it can be seen that the RI values of all cushion gum compounds were not significantly different with increasing lignin content. Additionally, the RI of the cushion gum compound composed of CB N330/lignin, both at 40/10 phr or 35/15 phr, was relatively higher than that of CB N220/lignin and CB N550/lignin. This was due

to better dispersion of the lignin throughout the rubber matrix filled by CB N330, as confirmed by the crosslink density analysis in Figure 3.
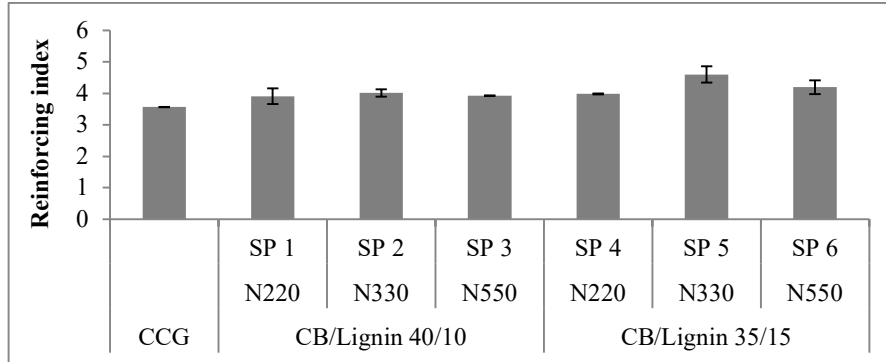


**Figure 9** Reinforcing index (RI) of the cushion gum compounds.

## 4 Conclusion

The present experiment result showed that the usage of hybrid filler consisting of carbon black (N330) and lignin (sodium lignosulfonate) at a ratio of 40 phr and 10 phr (the formula coded as SP2) has potential to be developed as rubber cushion gum composite for the tire retreading industry. The performed study indicated comparable or even better processability, physical and mechanical properties of the hybrid filled rubber cushion gum composite (SP2) in comparison to commercial cushion gum. The hybrid filled rubber cushion gum composite obtained by applying formulation SP2 had low minimum torque (0.7 dNm), a short optimum cure time (7.39 minutes), and a long scorch time (2.46 minute) along with a high torque difference, which represents crosslink density at 9.49 dNm (equal to $2.03 \times 10^{-5}$ mol/m$^3$). In addition, it revealed higher hardness (55 Shore A), better tensile strength (16 N/mm$^2$), elongation at break (550%), RI (M300/M100 at 4.01), and tear strength (65.3 kN/m) but slightly lower tensile modulus 300% (5.6 N/mm$^2$), and higher specific gravity (1.107 g/cm$^3$). The excellent adhesive strength of the SP2 cushion gum composite (1.33 kN/m) was due to the presence of high content of the phenolic functional group in lignin and pine tar oil.

## Acknowledgements

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