Processing and Tensile Properties of Natural Rubber Filled with Calcium Carbonate or Silica in the Presences of Lauryl Alcohol

Indra Surya1*, Hanafi Ismail2, and Johan Anto3

1Department of Chemical Engineering, Universitas Sumatera Utara, Medan, Indonesia
2School of Materials and Mineral Resources Engineering, Engineering Campus, Universiti Sains Malaysia, Nibong Tebal, Malaysia
3P.T. Industri Karet Deli, Medan, Indonesia

Abstract. The processing and tensile properties of the compounds of natural rubber (NR) filled with calcium carbonate (CaCO3) or silica filler in the presence of lauryl alcohol have been investigated. The NR was filled with each of those fillers at a fixed concentration, i.e., 30 phr, and they were compounded with lauryl alcohol as a filler dispersant with varied concentrations on a laboratory mill by applying the semi-efficient vulcanization recipe. A typical oscillating disc rheometer (ODR) was utilized for the determination of processing properties. From the results, it was observed that CaCO3 or silica with lauryl alcohol had increased the rate coefficient of vulcanization (RV) of the filled NR compounds with a reduction in viscousness or minimal torsion. The results also showed that the lauryl alcohol additions have enhanced the tensile properties of obtained filled NR compounds. The greater the lauryl alcohol concentration, the greater the rate coefficient of vulcanization and tensile strength of both types of filled NR. The overall results have indicated that the lauryl alcohol performance in enhancements of processing and tensile properties was more pronounced on calcium carbonate than silica.

1 Introduction

Improving the mechanical properties, such as tear and tensile strengths, durability, and abrasion resistance is the practical meaning of rubber reinforcement with fillers [1-3]. Fillers are added materials to lift the performance and durability of rubber products. Several commonly utilized fillers for rubber reinforcement, i.e., carbon black (CB), precipitated silica, calcium carbonate (CaCO3), and clay with micron-sized particles. The choice of filler depends on the desired properties and specific application of the final rubber product. The quantity and type of filler, along with other rubber-compounding chemicals, are carefully selected to produce the desired balance of properties while considering factors including production cost, processing requirements, and environmental considerations.

* Corresponding author: isurya@usu.ac.id
CB is produced by the incomplete combustion of natural gas or oil. It is a popular type of reinforcing filler used in various industries, particularly in black-coloured rubber products, while silica and calcium carbonate are used to produce light-coloured rubber products. Silica is a white filler that is utilized for enhancing the abrasion resistance, tear resistance, and tensile strength of the final rubber products. This is particularly important in the production of vehicle tires, where these properties are crucial for safety and durability. In tire production, silica is often added to the tread compounds. This enhances the tire's grip and traction on wet surfaces, making it safer in wet conditions. Silica in combination with other additives, like silane coupling agents, can enhance the dispersion of silica particles in the rubber matrix, ensuring a more even dispersion and better performance. Silica in rubber compounds also can help reduce rolling resistance in tires. Lower rolling resistance results in cheap fuel consumption and reduced carbon emissions, making vehicles more environmentally friendly.

Like silica, calcium carbonate is also a white filler and is commonly utilized as a filler and reinforcing agent in the production of rubber and rubber products. It provides several benefits to rubber compounds, including cost reduction, improved processing, and enhanced physical properties. Calcium carbonate is used as a filler in rubber compounds to reduce the cost of the final product. It is a cost-effective alternative to more expensive fillers and extends the rubber material, making it more economical. Calcium carbonate can enhance the mechanical properties of rubber, such as tensile strength, modulus, and abrasion resistance. It contributes to the reinforcement of the rubber matrix and helps improve its performance.

In the production of various light-coloured types of rubber products, silica and calcium carbonate are normally utilized. However, at a relatively larger amount of their loadings, those filler particles like to form bigger filler agglomeration. This reality tends to poorer the filler distribution and degrade the mechanical properties of the rubber products. To solve those problems, a dispersant aid was used, namely lauryl alcohol, which can be produced from palm oil and it is structured molecule is visualized in Fig. 1. Therefore, this paper reports the effects of lauryl alcohol, \( \text{CH}_3(\text{CH}_2)_{10}\text{CH}_2\text{OH} \), on processing and tensile properties of calcium carbonate or silica-reinforced natural rubber (NR).

Fig. 1. Lauryl alcohol

2 Material and Methods

2.1 Chemical substances

NR is the raw or unprocessed rubber. Table 1 tabulates the chemical substances that were used for the NR mixing process. NR, with other chemicals, was used as bought from the local chemical sellers. P.T. Eco Green Oleo-chemical Factory, Medan, Indonesia, has supplied lauryl alcohol.

2.2 Compounding the NR-silica/calcium carbonate-lauryl alcohol

Table 1 tabulates the NR and other chemicals for preparing calcium carbonate or silica-reinforced NR compound relating to a semi-efficient rubber compound [8].
Table 1. The calcium carbonate or silica-reinforced NR with and with no lauryl alcohol

<table>
<thead>
<tr>
<th>NR/Chemical substances</th>
<th>Lauryl alcohol (parts per hundred NR)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
</tr>
<tr>
<td>NR</td>
<td>100</td>
</tr>
<tr>
<td>ZnO</td>
<td>5.1</td>
</tr>
<tr>
<td>Stearic acid</td>
<td>2.2</td>
</tr>
<tr>
<td>Antioxidant</td>
<td>2.2</td>
</tr>
<tr>
<td>MBTS</td>
<td>1.7</td>
</tr>
<tr>
<td>Sulfur</td>
<td>1.7</td>
</tr>
<tr>
<td>CaCO₃ or silica</td>
<td>30</td>
</tr>
</tbody>
</table>

2.3 Processing properties

Regarding ISO 3417, a cure meter of MDR 2000 was used to determine the processing properties of the calcium carbonate or silica-reinforced NR with lauryl alcohol, including maximal torsion (Tmax), minimal torsion (Tmin), delta torsion (Tmax – Tmin), t₉₀ (minutes of cure time) and tₛ₂ (minutes of scorch time).

2.4 Tensile measurement

Tensile measurement was performed at a tensometer (ASTM D638). TS as tensile strength and EB as breaking elongation were determined.

2.5 Morphology properties

The fractured/broken surfaces of calcium carbonate or silica-reinforced NR with and without fatty alcohol specimens were analyzed for micro defects. The imaging was conducted using a Scanning Electron Microscope (SEM) to visualize the morphology properties of calcium carbonate or silica-reinforced NR in the existence of lauryl alcohol. The fractured specimens were sputter-covered with gold-palladium to minimize the electrostatic charging effect during the experiment.

3 Results and discussion

3.1 Lauryl alcohol affection on torsion properties

The torsion properties, such as maximal torsion (Tmax), minimal torsion (Tmin), and delta torsion (Tmax – Tmin), were determined through the rheography of the cure meter. Table 2 shows the torsion properties of the calcium carbonate or silica-reinforced NR with and without lauryl alcohol. Minimal torsion, as visualized in Fig. 2, adding 1 phr lauryl alcohol has decreased the value of Tmin. Increasing the lauryl alcohol concentration has caused a further lowering in the value of Tmin.

The property of minimal torsion indicates the viscosity of a rubber compound [9-10] and, in this investigation, the Tmin value and viscosity have been decreased; the viscosity reduction phenomenon was regarded as the softening of the NR-silica or calcium carbonate with lauryl alcohol ingredient. Hence, the lauryl alcohol acted as a softening chemical ingredient, which lowered the viscosity of the NR-silica or NR-calcium carbonate compound.

Minimal torsion value is also used to indicate the nature of filler-to-filler interactions [11-12]. A bigger value of minimal torsion means stronger filler-to-filler interactions and vice
versa. The reduction of minimal torsion trend was regarded as the function of lauryl alcohol as a softening agent, which lowered the viscosity but increased filler spreading level, respectively. Those chemicals that can make the viscosity of compounds are classified as softening agents [6, 13].

At a comparable filler concentration, the $T_{\text{min}}$ of calcium carbonate was lower than silica’s. It could be because of their molecule weight difference.

![Graph showing minimal torsion vs Lauryl alcohol concentration](image)

**Fig. 2.** Lauryl alcohol affection on minimal torsion

The property of delta torsion, $T_{\text{max}}-T_{\text{min}}$, is also tabulated in Table 2. As observed, the addition of 1 phr of lauryl alcohol has increased the values of delta torsion. An increase in the lauryl alcohol concentrations has caused a further lifting in the value of delta torsion.

**Table 2.** Processing properties of calcium carbonate or silica-reinforced NR with lauryl alcohol

<table>
<thead>
<tr>
<th>Filler</th>
<th>Lauryl alcohol (phr)</th>
<th>$T_{\text{max}}$, dN.m</th>
<th>$T_{\text{min}}$, dN.m</th>
<th>Delta, dN.m</th>
<th>$t_{90}$, min.</th>
<th>$t_{92}$, min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>CaCO$_3$</td>
<td>0</td>
<td>4.80</td>
<td>1.22</td>
<td>3.58</td>
<td>1.22</td>
<td>0.74</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>5.00</td>
<td>1.19</td>
<td>3.81</td>
<td>1.17</td>
<td>0.76</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>5.17</td>
<td>1.12</td>
<td>4.05</td>
<td>1.12</td>
<td>0.79</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>5.53</td>
<td>1.10</td>
<td>4.43</td>
<td>1.09</td>
<td>0.80</td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>5.32</td>
<td>0.86</td>
<td>4.46</td>
<td>1.05</td>
<td>0.82</td>
</tr>
<tr>
<td>Silica</td>
<td>0</td>
<td>11.38</td>
<td>2.74</td>
<td>8.64</td>
<td>1.65</td>
<td>0.72</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>10.43</td>
<td>2.33</td>
<td>8.10</td>
<td>1.64</td>
<td>0.73</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>10.61</td>
<td>2.22</td>
<td>8.39</td>
<td>1.62</td>
<td>0.74</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>11.30</td>
<td>2.15</td>
<td>9.15</td>
<td>1.57</td>
<td>0.79</td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>9.7</td>
<td>2.02</td>
<td>7.68</td>
<td>1.50</td>
<td>0.81</td>
</tr>
</tbody>
</table>

As tabulated in Table 2, lauryl alcohol has reduced the cure times but increased the scorch times of calcium carbonate or silica-reinforced NR. The bigger the lauryl alcohol concentration, the lower the cure time, but the longer the scorch time. It was observed that the lauryl alcohol was a scorch delay chemical for both filled-NR.

The lauryl alcohol affection on the rate coefficient of vulcanization, $R_v$, of calcium carbonate or silica-reinforced NR is visualized in Fig. 3. The lauryl alcohol has increased the $R_v$ value for both fillers. All prepared calcium carbonate or silica-reinforced NR with lauryl alcohol have a higher $R_v$ value compared with the filled NR with no lauryl alcohol. Since the rate coefficient of vulcanization indicates the activity of the components of rubber compounds [14], the increased $R_v$ was due to the great interactions between the active
function of both fillers and lauryl alcohol, hence causing a more pronounced enhancement of the vulcanization process.

**Fig. 3. Lauryl alcohol affection on the coefficient of vulcanization**

### 3.2 Lauryl alcohol affection on the swelling properties

The lauryl alcohol affection on the swelling properties of calcium carbonate or silica-reinforced NR is visualized in Fig. 4. The swelling properties are the degree of compound crosslink density. The lower swelling percentage is a higher degree of compound crosslink density. The lauryl alcohol has decreased the swelling percentages of both calcium carbonate and silica-reinforced NR. Those crosslink density enhancements were due to the role of lauryl alcohol as a plasticizing chemical, which improved the degree of filler dispersion and rubber-to-filler interactions, respectively. Since the delta torsion property reflects the degree of crosslink [15], the swelling properties agree with the properties of delta torsion in Table 2.

**Fig. 4. Lauryl alcohol affection on swelling percentage**

### 3.3 Lauryl alcohol affection on the tensile properties

Tensile properties of calcium carbonate or silica-reinforced NR with and without lauryl alcohol are visualized in Figs. 5 and 6. As observed in Fig. 5, elongations at break (EB) of calcium carbonate or silica-reinforced NR with 1 phr of lauryl alcohol were longer than those without lauryl alcohol. The bigger the lauryl alcohol concentration, the longer the EB. It could be attributed to the role of lauryl alcohol as a plasticizing chemical, which improved the extensibility of filled NR vulcanizates.
Fig. 5. Lauryl alcohol affection on breaking elongation

From Fig. 6, the tensile strength, TS, was lifted as the lauryl alcohol was added up to optimum lauryl alcohol concentrations at 5 phr for both filled NR. These TS enhancements were due to crosslink enhancement as the consequence of better filler dispersion of both fillers.

Fig. 6. Lauryl alcohol affection on tensile strength

3.4 SEM and morphology study

Fig. 7 shows the images of the surfaces of vulcanizates NR-CaCO₃ or silica-lauryl alcohol (magnification: 740X). The images of (B and E) have indicated an improvement in calcium carbonate or silica distribution as the lauryl alcohol was incorporated. The poorest filler dispersion is at the (A and D) image, whereas relatively big calcium carbonate or silica agglomerates have appeared. The (B and E) images show the biggest matrix breaking line than the images of (C and F). It has altered the cracking path, which provided increasing crack initiation resistance and strength properties. Enhanced rupture energy was because of a greater NR-filler action [16-17].
4 Conclusion

The lauryl alcohol has improved the processing and tensile properties of calcium carbonate or silica-reinforced natural rubber compounds. The minimal torsion or viscosity of the filled compounds has been decreased but the rate coefficient of vulcanization has been increased by the lauryl alcohol additions. Lauryl alcohol has increased the crosslink density through the plasticizing or reducing viscosity process and has increased the tensile properties through the increasing crosslink density. The morphology analysis has proved that the highest tensile strength for calcium carbonate or silica-reinforced natural rubber whenever the highest crosslink degree was achieved. Lauryl alcohol can be utilized as a filler degree enhancer for calcium carbonate or silica-reinforced natural rubber to improve the processing properties and tensile properties. The overall results tell that the performance of lauryl alcohol in enhancements of processing and tensile properties was more significant on calcium carbonate than silica.

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References