Tensile and rheometric properties of calcium carbonate-filled natural rubber compounds without/with lauryl alcohol

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Abstract. The tensile and rheometric properties of CaCO3, calcium carbonate-filled natural rubber (NR) compounds without and with lauryle alcohol (LA) as a new rubber additive were examined. The NR was filled by CaCO3 with a certain addition, thirty phr. The LA was synthesized from palm kernel oil and incorporated into CaCO3-filled NR compounds at 1, 3, 5 and 7 phr. From the overall results, the LA was a curative and also plasticizing material. As a curative material, LA enhanced both tensile strength and difference in torque up to a five phr of addition. As a plasticizing material, LA reduced tensile moduli but enhanced elongation at break of CaCO3-filled NR vulcanizates. A five phr of LA was an optimum addition.

1. Introduction
Reinforcing fillers are non black and black ones. Black one is carbon black (CB) which is used to produce rubber products with black-coloured, and non black ones are clay, precipitated silicas and CaCO3 (calcium carbonate). The non black fillers are used to produce rubber products with black-coloured is not wanted. Practically, CaCO3 and other non black are having relatively lower reinforcing efficiency compared to CB [1]. It is due to chemistry of their surfaces that are more hydrated and also polar and hence, causing them to become more difficult to interact with natural rubber (NR) which is a non-polar rubber [2]. Some attempts have been performed to improve their reactivity to NR. One effort was the utilization of a silane coupling agent. This rubber additive changes the polar fillers surfaces and serves as a more active one which will contribute positively during curing, serving some coupling-bonds upon the additive and both NR - polar fillers [3]. Those modified rubber vulcanizates exhibit an excellent performance.

The CaCO3 is one kind of polar fillers. It is ready to be dispersed in polar rubbers compared to NR [4] and hence, the recipes of CaCO3-NR compounds should have a compatibilizer that can promote a stronger rubber to filler interactions. In the case to handle the weakness of CaCO3, this work has incorporated lauryle alcohol (LA) to CaCO3-filled NR compounds. The LA comes from the palm tree and due to its oily property, it is potentially to be utilised as a compatibilizer. This work reports the tensile and rheometric properties of CaCO3-filled NR compounds without/with LA.

2. The experimental
2.1. The materials for research
An SIR 3L (NR), sulfur, stearic acid, zinc oxide, antioxidant-IPPD, accelerator-MBTS and CaCO3 was supplied by P.T. IKD, Medan, Indonesia. The LA (CH3(CH2)10CH2OH) was supplied by P.T. Ecogreen, Batam, Indonesia.
2.2. The rubber compounding
The semi efficient vulcanisation was performed for compounding and procedures of compounding were based on ASTM (D3184 – 80). The rubber compound preparation was done on the two roll mill with XK-160 Model. Table 1 presents recipe of NR compound with different LA concentrations.

| Materials       | Composition |
|-----------------|-------------|
| SIR-3           | 100         |
| ZnO             | 5           |
| IPPD            | 2           |
| Stearic acid    | 2           |
| S               | 1.5         |
| MBTS            | 1.5         |
| Calcium carbonate| 30         |
| LA              | 0; 1; 3; 5; 7 |

2.3. The curing
Scorch and optimum cure times, maximum and minimum torques and also torque difference of filled NR compounds with and with no LA were determined utilising an MDR2000 (Monsanto Moving Die Rheometer) ASTM, D2084-11. The temperature of the testings was at 150 °C.

3. Results and Discussion

3.1. Rheometric properties
Scorch and optimum cure times of CaCO₃-filled NR compounds without/with LA are presented in Fig. 1. Scorch times of CaCO₃-filled NR compounds with LA were higher compared to with no LA. A higher LA addition gave a longer scorch times and hence, the additive LA was considered as a scorch delay material.

![Figure 1](http://example.com/figure1.png)

**Figure 1.** The scorch and optimum cure times of CaCO₃-filled NR compounds without/with LA

Figure 1 also shows that optimum cure times of CaCO₃-filled NR compounds plus LA were shorter compared to CaCO₃-filled NR compounds without LA. A shorter optimum cure time is a faster cure rate. In this case, LA caused improvement in curing. The higher the addition of LA, the more...
significant was cure improvement. The LA acted as a curative material in the vulcanisation of NR vulcanizates.

The torques properties of CaCO₃-filled NR vulcanizates without/with LA are presented in Figure 2. As presented, minimum torques (Mₐ) of NR vulcanizates were lower. The Mₐ value reflects the vulcanizate viscosity [1, 6-9]. A lower Mₐ is a lower viscosity and plasticizing effect LA reduced the NR vulcanizates viscosity. A higher LA addition gave a lower viscosity.

![Figure 2. The torques properties of CaCO₃-filled NR compounds without/with LA](image)

Figure 2 also shows that the torque differences (Mₕ - Mₐ) of CaCO₃-filled NR compounds with LA were higher. The difference in torque was further enhanced by the incorporation of LA up to five phr and reduced after the addition. A greater difference in torque gave a higher crosslinking density degree [9-13]. Improvement in crosslinking density up to optimum LA incorporations (five phr) was because of both physical and chemical properties of LA. Active groups of LA reacted not only to NR but also to sulfur in vulcanisation and as a consequent, causing a better degree of cure.

The oily properties of LA caused it acted as a plasticizing material. Since it reduced the filled NR vulcanizates viscosity, the degree of dispersion and also rubber to filler interactions were enhanced. Rubber to filler interaction is considered as additional physical crosslinking [1, 7] and it enhanced total crosslinking density of the NR vulcanizates [2, 14].

The reducing in torque difference, after optimum addition, was because of excessive amounts of LA which absorbed parts of CaCO₃ filler and also the others curatives and hence, total crosslinking density was reduced.

3.2. The tensile properties

Tensile properties of CaCO₃-filled NR compounds without/with LA are presented in Table 2. LA reduced the tensile modulus. A higher LA addition gave a lower M100/M300. The LA has made CaCO₃-filled NR compounds with lower tensile moduli and it was because of its usage as a plasticizing material.

The TS was enhanced up to an optimum LA addition (five phr) and then reduced by further increases the addition. TS improvement up to five phr of LA addition was because of the enhancements in CaCO₃ dispersion and crosslinking density. The deterioration in TS after a five phr addition was because of curative absorbing effect of a more amount of LA, which reduced density of crosslinking.
Table 2. Tensile properties of CaCO$_3$-filled NR compounds without/with LA

| CaCO$_3$-filled NR compounds | LA additions (phr) | 0.0 | 1.0 | 3.0 | 5.0 | 7.0 |
|-----------------------------|-------------------|-----|-----|-----|-----|-----|
| M100, MPa                   |                   | 0.85| 0.84| 0.79| 0.75| 0.72|
| M300, MPa                   |                   | 1.99| 1.88| 1.60| 1.58| 1.57|
| TS, MPa                     |                   | 15.70| 17.75| 19.70| 20.30| 18.00|
| EB, %                       |                   | 740 | 748 | 750 | 770 | 790 |

The EBs of CaCO$_3$-filled NR compounds were increased by LA incorporations. A higher LA addition gave a higher EB. Again, it was because of the usage of LA a plasticizing material. The LA was based on natural oil/fat and oil is one of the sources of a plasticizer [4]. Increases LA addition enhanced the amount of plasticizer in NR compounds which improved flexibility of filled NR vulcanizates.

4. Conclusions
The lauryl alcohol was a curative and plasticizing material in calcium carbonate-filled natural rubber compounds. It enhanced scorch time but reduced optimum cure time and also minimum torque.

Lauryl alcohol reduced tensile moduli but enhanced torque difference and also tensile strength up to an optimum addition. Five parts per hundred was an optimum addition of lauryl alcohol. Lauryl alcohol enhanced the elongation at break or extensibility.

Acknowledgements
The authors thank P.T. IKD, Medan, Indonesia and DRPM - Kemristek Dikti for a research grant of Penelitian-PDUPT 2018 (contract number 62/UN5.2.3.1/PPM/KP-DRPM/2018).

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