Enhancements in cure rate index and mechanical properties of silica-filled natural rubber using octadecanol-fatty alcohol

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Abstract. The enhancements in cure rate index and mechanical/tensile properties of natural rubber (NR) filled with silica because of the addition of octadecanol (OCD)-fatty alcohol (FA) were investigated. The silica as a reinforcing filler was compounded into the NR at a fixed loading, 30 parts per hundred rubber (phr.). The OCD-FA was derived from the oil of palm kernel and compounded into the NR as an additive agent. The loading variations of OCD-FA were 0 to 4 phr. It was found that the OCD-FA functioned as a curative and plasticizing agent for the silica-filled NR compounds. As a co-curing agent, the OCD-FA enhanced the time of scorch and cure rate index-CRI of the compounds of NR. A higher the OCD-FA loading caused in a higher CRI degree. As a plasticizing agent, the OCD-FA decreased tensile modulus but increased elongation at break. It was also found that the OCD-FA increased both tensile strength and abrasion resistance of the compounds of NR up to a 2 phr. of loading. The addition of OCD-FA with a 2 phr. of was an optimum loading for the compounds of NR.

1. Introduction
Mixing of a type of raw rubber with some rubber additives is defined as rubber compounding operation. Each of raw rubbers and also the rubber additives has a certain function either in the cure, mixing process or rubber products [1]. Raw rubbers will deliver the rubbery behaviour; additives such as accelerator will enhance the cure state; the curing agent will connect chemically the chains of rubber; and plasticizer will reduce the viscosity, enhance deformability and change properties.

In rubber compounding, the additives are defined as curative and non curative. The curative will affect the curing process. The well-known ones of them are zinc oxide, stearic acid, accelerators and sulfur. Non curative such as fillers are utilized in order to enhance mechanical/tensile properties of Vulcanized products. Silica is one of the most exploited fillers in the enhancements in mechanical properties of rubber products [2]. Relatively, at higher loading of utilization, the micron-sized particles of the filler that tends to form some bigger aggregates during compounding and causing in a relatively poorer degree of filler dispersion. Besides, the silica filler also has numerous silanol groups on its surface and causes it becomes very polar filler. It can readily interact with zinc oxide during the curing process. The silica-zinc complexes unable to activate the accelerator and as a consequence, the activity of zinc is decreased and hence, the vulcanization or cure process is retarded [2].

As an attempt to solve such problems, a special rubber additive agent which is named as octadecanol (OCT) was added into the compounds of natural rubber (NR) filled with silica. It is fatty alcohol derived from the oil of palm kernel and it was utilized to solve both the cure retardation and filler dispersion problems. The material of OCT-fatty alcohol (FA) is oily with unique polar and non-
polar functional groups inside the material; oil can act as a plasticizing agent [3-5] that helps to disperse the filler homogeneously inside the rubber molecules.

This research investigated the effects of OCT-FA on cure rate index and also mechanical/tensile properties of the NR which was filled with silica.

2. Experimental

2.1. Materials of research
A SIR 3L-grade of NR was supplied by the PPTPN VII - Indonesia. IKD, PT locates in Medan of Indonesia prepared IPPD-N-isopropyl-N'-phenyl-p-phenylenediamine, precipitated silica, stearic acid, sulfur, MBTS-mercaptobenzothiazole disulphide, zinc oxide. Ecogreen, PT in Batam, Indonesia supplied OCT-FA. The molecule structure of OCT-FA is CH₃(CH₂)₁₆CH₂OH.

2.2. Compounding of NR
A semi EV cure system was applied in making the compounds of NR. Procedures of the making of compounds of NR were conducted according to ASTM D 3184-80. The compounds of NR were made on a laboratory two roll mill. The recipe of compounds of NR is presented in Table 1.

| Chemicals         | Content (phr.) |
|-------------------|----------------|
| SIR 3L            | 100            |
| Zinc oxide        | 5              |
| IPPD              | 2              |
| Stearic acid      | 2              |
| MBTS              | 1.5            |
| Sulfur            | 1.5            |
| Precipitated silica | 30.0        |
| OCT-FA            | 0; 1; 2; 3 and 4 |

2.3. Curing of compounds
Cure of compounds of NR was characterised based on an MDR 2000 – Rheometer. The Rheometer was operated to determine the times to scorch and cure according to ISO-3417. The respective compounds of NR were vulcanized at 150 °C.

2.4. The tensile properties
The vulcanized NR was cut from molded sheets based on ISO-37. The tensile properties were tested at a crosshead speed of 500 (mm.min⁻¹) and the testing was performed using the Instron 3366-universal tensometer to determine the stresses at 300% and 100% elongations-M300 and M300, tensile strength-TS, elongation at break-EB.

2.5. The abrasion test
The samples of NR compounds were moulded with a KAO – Tech moulding machine with a force 100 kg/cm² at 150 °C for optimum cure time plus 5 minutes. The samples were a disc with dimension 12.7 + 0.0 – 0.5 mm in thickness, 63.5 + 0.5 mm diameter with a central hole 12.7 mm diameter. Wallace Abrasion Tester was used to determine abrasion loss of the vulcanized rubber according to BS-903 part A9. The angle between the abrasion wheel and the rubber disc was 15°, the applied weight was 4.5 kg. The abrasion experiment was carried out at room temperature. The test specimen was driven at a constant speed and drives the abrasion wheel which was held against it by a constant force. The test specimen was weighed and mounted on the spindle of the machine by tightening the wing nut finger tight. After 500 revolutions of the trial run, the test specimen was weighted again. The weight of the
rubber loss was converted to volume loss by using the density of the vulcanized rubber. Based on this volume loss, suitable running in period and test run for the sample was determined. After the running in period, five test runs were carried out and the average was in term of volume loss (cm³) per-1000 revolutions.

3. Results and discussion

3.1. The cure rate index of compounds of NR

The effects of OCT-FA on scorch time, optimum cure time and cure rate index (CRI) of the NR compounds are shown in Figs. 1-2. As shown in Fig. 1, the scorch times of silica-filled NR compounds with OCT-FA were higher compared to the compounds without OCT-FA. A higher OCT-FA loading caused a higher in scorch time. In this investigation, the OCT-FA acted as a scorch delay agent. Any material that enhances the scorch time can be defined as a scorch delay material [6-8].

![Figure 1. Effects of OCT-FA addition on times to scorch and cure of compounds of NR](image1)

![Figure 2. Effects of OCT-FA addition on cure rate index of compounds of NR](image2)
From Fig. 1, the addition of one phr. of OCT-FA decreased the time of optimum cure. Shorter cure time is defined as a faster cure rate. Therefore, OCT-FA caused in cure improvement. A higher OCT-FA caused in a more pronounced in cure improvement. In this study, the OCT-FA functioned as a cocuring agent for the compounds of NR. Any material that affects the curing properties of a rubber compound is defined as a cocuring or curative agent [9-11].

As shown in Fig. 2, the CRI of compounds of NR with OCT-FA were higher compared to the filled NR compounds without OCT-FA. A higher OCT-FA loading caused in a higher in CRI. Again, it was due to the function of OCT-FA as a cocuring agent.

3.2. Tensile properties of the compounds of NR
Effects of OCT-FA on tensile properties of the compounds of NR are visualized in Figs. 3-5. As visualized in Fig. 3, the OCT-FA decreased the tensile moduli. The higher the OCT-FA, the lower the stresses at 300% and 100% elongations was. The OCT-FA caused in the tensile moduli of compounds of NR with lower values. It was due to the function of OCT-FA as a plasticizing agent which softened filled NR vulcanizates.

![Figure 3. Effects of OCT-FA addition on tensile modulus of compounds of NR](image)

From Fig. 4, the TS was increased slightly up to a two phr. of OCT-FA loading and then, was decreased with further increases the loading of OCT-FA. TS enhancement up to a two phr. of OCT-FA might be due to the effect of plasticizing of OCT-FA which increased the degree of silica dispersion and rubber to filler interaction, respectively. Consequently, the crosslink density of the filled NR vulcanizates was enhanced. The deterioration in TS after a two phr. of OCT-FA loading was due to active ingredients absorbing the effect of the excessive OCT-FA that decreased the crosslink density.

As shown in Fig. 5, the addition of a one phr. of OCT-FA into NR control compound increased elongation at break (EB). The addition with a more amount of OCT-FA caused further increases in the EB. Again, it was due to the plasticizing effect of OCT-FA to the compounds of NR [12-15]. The additive enhanced the deformability of a polymeric composite. Increasing the OCT-FA loading caused further increases the deformability or extensibility of the compounds of NR.
3.3. Abrasion loss
The effect of OCT-FA on abrasion loss of the compounds of SIR 3L-NR is visualized in Fig. 6. As shown, the abrasion loss of the SIR 3L-NR samples (in grams) was decreased up to an optimum OCT-FA loading (at a two phr.) and then it was increased with further increasing the OCT-FA loading. The abrasion resistance of the SIR 3L-NR was relatively poorer; the OCT-FA improved the resistance. It was due to a higher degree of wetting and silica dispersion inside the OCT-FA plasticized SIR 3L-NR molecules. A higher degree of silica dispersion caused in both a greater degrees of rubber to filler interaction and abrasion resistance [16]. The rubber to filler interaction is a type of physical crosslink which affects the enhancement in overall crosslink density of vulcanized rubber, significantly [17-19]. The enhancement in abrasion resistance was due to the enhancement in crosslink density of the silica-filled with OCT-FA. The deterioration in abrasion resistance after the two phr. of OCT-FA was due to the excessive effect of the additive which decreased the crosslink density of the NR filled vulcanizates.
4. Conclusions
The octadecanol-fatty alcohol acted as a co-curing agent for SIR 3L-natural rubber compounds that were filled with silica-filler. It enhanced both times of scorch and cure rate index of the silica-filled SIR 3 L-natural rubber.

The octadecanol-fatty alcohol was also acted as a plasticizing agent. It softened the compounds of SIR 3L-natural rubber and hence, enhanced both the rubber to filler interaction and silica dispersion degree. The octadecanol-fatty alcohol lowered the tensile moduli but increased tensile strength, abrasion resistance and extensibility of the vulcanized SIR 3L-natural rubber.

The enhancements in mechanical properties were due to the crosslink density enhancement as well as cure state enhancement of the compounds of SIR 3 L-natural rubber.

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Figure 6. Effect of OCT-FA addition on abrasion loss of compounds of NR
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