

The effect of carbon black composition in natural rubber compound

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ABSTRACT

This study aims to determine the effect of carbon black (CB) filler composition on natural rubber compounds on thermal properties, morphology, diffraction patterns, and functional groups. The method of making compound uses an open mill by mixing SIR 20 natural rubber as a binder and carbon black as a filler with a composition variation (0; 2; 4; 6 and 8)wt%. The diffraction pattern shows that there is a change from the shape of the amorphous structure to the semi-crystalline form with increasing CB composition. Morphological results showed that the mixture was homogeneous. FTIR graph shows that there is no significant difference between compound with filler and without filler. Differential scanning calorimetric analysis, the addition of carbon black composition to natural rubber increases the heat delta of the compound compared without CB.

1. Introduction

Rubber is a crop that can produce a variety of products that are useful in life. Rubber technology continues to develop over time, and more products will come from the rubber industry. There are two types of rubber commonly used in industry, which are natural rubber and synthetic rubber. The use of natural rubber (NR) in various industries due to its elasticity, low hysteresis, high durability, excellent toughness. In general, NR is an amorphous material. However, when stretched NR can crystallize. This crystallization contributes to the mechanical properties of NR such as tensile strength, tear resistance, and abrasion resistance, etc.

The rubber compound is the main derivative of rubber commodity. Almost all rubber compounds use carbon black (CB) as a filler. Carbon black filler functions to strengthen, increase the volume, improve the physical properties of rubber, and strengthen vulcanization. The results of the rubber compound can be useful in making shoe soles, gloves, and motorized vehicle tires. Making rubber finished goods is done through the process of mixing rubber with chemicals compositions. Furthermore, milling is carried out at a temperature and time according to the type of rubber and its purpose. Compounding is done to get a homogeneous mixture between rubber and chemicals [1].

Carbon black is a carbon material close to pure form combustion products derived from hydrocarbon or biomass products. The causes of aggregate size in carbon black are combustion temperature, combustion time, and material. The manufacture of tire compounds with carbon black as a filler aims to strengthen the bonds between compound-forming molecules. Carbon black as an active filler has a functional group that plays a role in strengthening the bonds between molecules forming rubber products. The structure of

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carbon black determines the optimal composition of the filler in a polymer matrix [2,3].

Carbon black is the type of filler most commonly used in making rubber compounds. Carbon black fillers have a strengthening effect on physical properties, especially those with small grain size [4–6]. The addition of carbon black will affect compound properties, viscosity, and strength of the compound. But the use of carbon black also has the disadvantage of reducing the stickiness. This makes carbon black not compact with other constituent materials at the time of mixing. Carbon black as an active filler can improve the performance of vulcanized rubber [7–9].

The natural rubber has limited properties. To increase the value of natural rubber modification is needed. One of the fillers is carbon black. Carbon black filler material is a filler material that can increase the hardness, tear resistance, abrasion resistance, and high breaking stress on the goods produced [10–12]. NR/CB compounding system had been popularly used for tire treads (areas where the tire comes in contact with the road surface) on passenger vehicles up until approximately a decade ago [13]. Carbon black grafted (GCB) is prepared to strengthen NR. The result is mechanical properties NR increases because of better compatibility between GCB and NR [14]. The addition of carbon black fillers in a natural rubber/polypropylene (NR/PP) can increase the tensile properties, but there is a decrease in the elongation at break. The decrease is a consequence of the contribution of carbon black properties that have relatively no elastic. Adding a filler is a way to modify natural rubber. Besides CB, the oil palm boiler ash (OPBA) can also be useful as a filler in NR, where is OPBA in the form of nanoparticles [15,16].

There have been many studies that make rubber compounds with different comparison ratios [17–19]. This study made a natural rubber compound with a ratio of natural rubber/CB (phr) 100/0, 98/2, 96/4, 94/6, 92/8. Furthermore, the diffraction patterns, morphology, thermal properties, and functional groups of the compounds will be analyzed.

2. Experimental

2.1. Material

Indonesia Standard Natural Rubber Ruber –20 (SIR-20), zinc oxide (ZnO), stearic acid, Wax, Tetra Methyl Thiura Disulfaric (TMTD), Sulfur, Marcapto Benzhoathizole Disulfide (MBTS) carbon black (CB) (N330), ISO Propylamino Diphenylamine (IPPD).

2.2. Rubber compound preparation

SIR-20 natural rubber, zinc oxide (ZnO), stearic acid, Wax, IPPD, TMTD, MBTS are fed into the open mill machine as shown in Fig. 1a, then ground until the rubber is solid. As the rubber milling process progresses, the materials are added one by one in stages, with a rubber compound formulation as shown in Table 1. The compound sheet vulcanization process was carried out at 160 °C for 15 min and a pressure of 135 MPa. Fig. 1b shows the compound results (see Table 2).

3. Result and discussion

3.1. X-ray diffraction (XRD) analysis

XRD testing was carried out to obtain diffraction patterns and crystalline structures. The XRD used was Shimadzu 6100 (40 kV, 30 mA) with a wavelength of $\text{Cu-K}\alpha_1 = 1.5405 \text{ \AA} = 0.15406 \text{ nm}$, at a rate of $2^\circ/\text{min}$ in the angular range of $2\theta = 5^\circ - 70^\circ$. Fig. 2 shows the XRD diffraction pattern for natural rubber compounds with the addition of carbon black into the matrix polymer. The decrease in intensity was at $d_{\text{hkl}} 22^0$ at CB composition 2% wt and 4% wt. In the CB composition, 6% wt and 8% wt weight intensity disappears. This shows the change in structure from amorphous to crystal. This change shows that there is intercalation between natural rubber polymers and CB. With the increase of CB in natural rubber compounds, the Fe_3O_4 content was seen to increase in $d_{\text{hkl}} 440$. Likewise, the addition of CB with carbon intensity at $d_{\text{hkl}} 101$ angles $2\theta = 45^\circ$. It can change the amorphous phase to the crystal phase as a consequence of molecular diffusion during treatment [20].

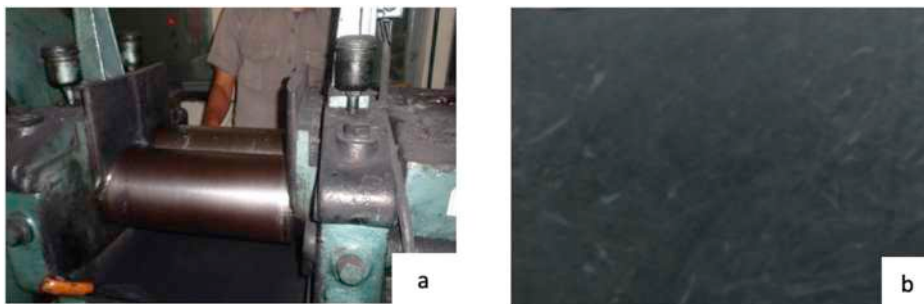


Fig. 1. (a) The grinding process in the roll mill, (b) Natural rubber compound.

Table 1
Composition of compounds with CB fillers.

No	Materials	S ₀ (without filler)	S ₁ (2 wt%)	S ₂ (4 wt%)	S ₃ (6 wt%)	S ₄ (8 wt%)	Function
1	NR SIR-20	100	98	96	94	92	Binder
2	Wax	1.5	1.5	1.5	1.5	1.5	Antilux
3	Filler (CB)	0	2	4	6	8	Filler
4	ZnO	5	5	5	5	5	Activator
5	Stearic Acid	2	2	2	2	2	Activator
6	Sulfur	3	3	3	3	3	Curing agent
7	IPPD	2	2	2	2	2	Antioxidant
8	TMTD	1.5	1.5	1.5	1.5	1.5	Accelerator
9	MBTS	2.5	2.5	2.5	2.5	2.5	Accelerator

Table 2
Melting point of natural rubber compound with CB filler.

Sample(wt%)	Onset(C)	Tm Peak (C)	Endset(C)	Heat Area(mJ)	Heat Delta (J/g)
0	370.44	374.44	384.14	65.29	8.37
2	338.82	366.25	384.03	403.28	51.70
4	342.15	372.05	381.34	482.11	63.44
6	356.81	367.67	381.70	372.05	45.93
8	340.97	370.78	380.48	360.68	48.09

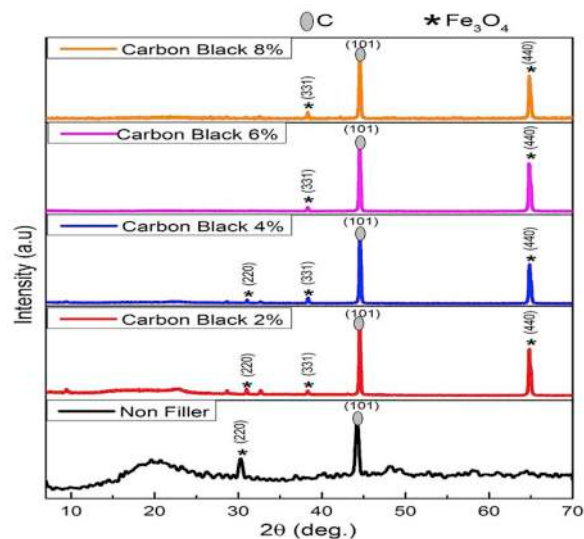


Fig. 2. Diffraction Patterns of natural rubber compound without filter and with CB filler.

3.2. Scanning Electron Microscopy (SEM) analysis of natural rubber compound with CB filler

Scanning Electron Microscopy (SEM) characterization on rubber was carried out to determine the distribution of carbon black fillers that affect the nature of natural rubber. SEM characterization results in Fig. 3 show the distribution of CB fillers in the composition 0, 2, 4, 6, and 8 wt%. The spread of filler occurs evenly. This is because of the interaction between the filler and rubber. Thus, increasing the mechanical properties of the rubber. Empty cavities arise due to the tendency of CB to form agglomeration because silica in CB has hydroxyl groups, which will help hydrogen bond with silica molecules or other polar chemical materials. This is consistent with the diffraction pattern of the XRD results in Fig. 2. Fig. 3a to d shows no de-adhesion phenomena occurring at the interface of the natural rubber matrix and fillers as found by Ref. [21] in natural-chitin rubber composites as the number of filler increases.

SEM analysis is a supporter of XRD analysis. The results of SEM analysis indicate the presence of SiO₂ morphology, which acts as an amplifier. The characterization of XRD and SEM shows that the majority of the amplifier used is amorphous. The presence of hump informs of the amorphous phase content of the sample [22]. The crystal will have a significant effect on the chemical elements of the sample. Generally, the higher the crystallinity, the lower the solubility of the elements [23]. From SEM observations, the surface of NR/CB20 shows homogeneous CB dispersion in the NR matrix. The presence of CB increases agglomeration to produce low tensile

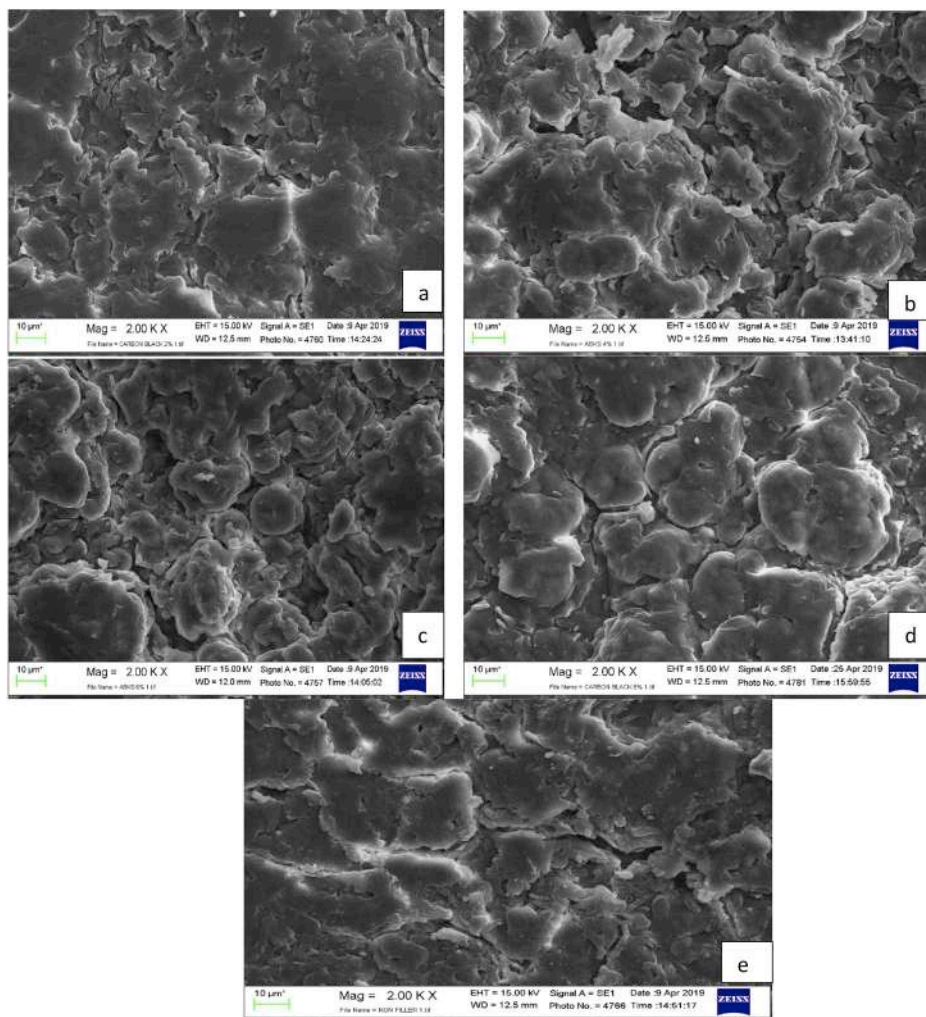


Fig. 3. Morphology of natural rubber compound with Filler CB (a) 2 wt%, (b) 4 wt%, (c) 6 wt%, (d) 8 wt%, (e) non filler.

strength [24].

3.3. Analysis of differential scanning calorimetric (DSC)

From Fig. 4, we can see the melting point from the onset temperature of 338 °C to the endset temperature of 384 °C. The change in peak temperature is not significant from the composition of the CB fillers 2 wt% to 8 wt% from temperatures 366.25C to temperatures 370.78C. However, a decrease in Heat occurs with increasing CB content from 403.38 to 360.68 mJ. The results from the DSC analysis are useful for determining enthalpy by measuring the differential heat flow needed to keep the sample and the inert reference at the same temperature. The information obtained in the semi-crystalline polymer is the material's crystallinity content [25]. The Melting temperature is an important parameter for thermoplastics because it represents the minimum temperature required for polymer processing. The melting behavior is highly dependent on the chemical structure of the material along with the size and regularity of crystallization found in the crystalline phase [26] (see Fig. 5).

3.4. Analysis of fourier transform infrared (FTIR)

The results of FTIR characterization on nanocomposite samples with Carbon Black fillers indicate the existence of several vibrational bonds. C–H bonds found in carbon absorbed in the area of 2853–2962 cm^{-1} . Peak 1475–1300 also shows C–H bending. Whereas the peak 1000–650 shows C = C–H bending [22]. For natural rubber, the stretching characteristics of C–H attached to the double bond C = C are at the absorption peak of 835 cm^{-1} , and the stretching characteristics of the alkene (C = C) are at the absorption peak of 1659 cm^{-1} .

FTIR spectra between nanocomposites with the addition of CB fillers and without fillers showed no significant difference. This was

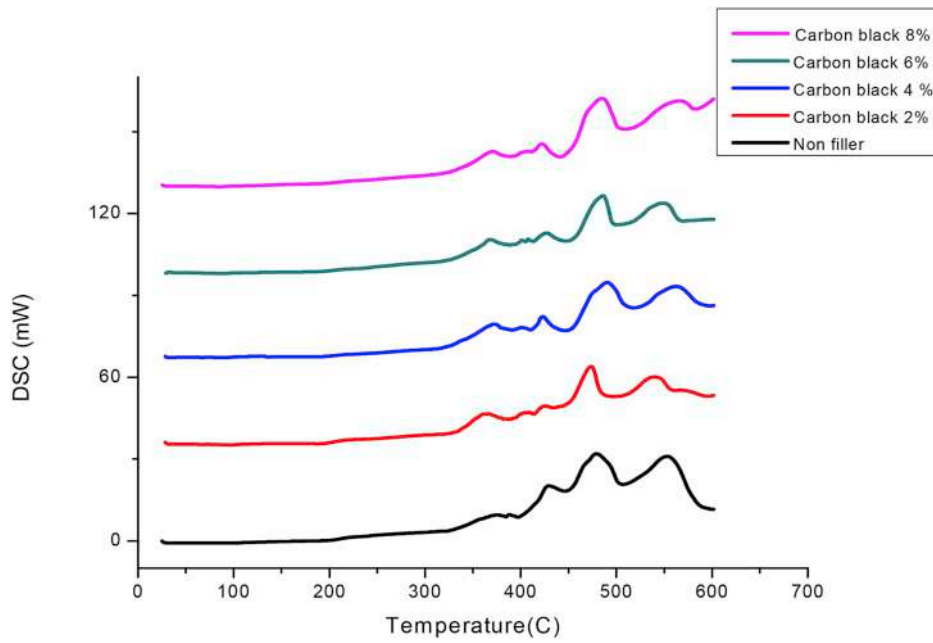


Fig. 4. DSC Thermogram of natural rubber compound with CB filler (0–8) wt%.

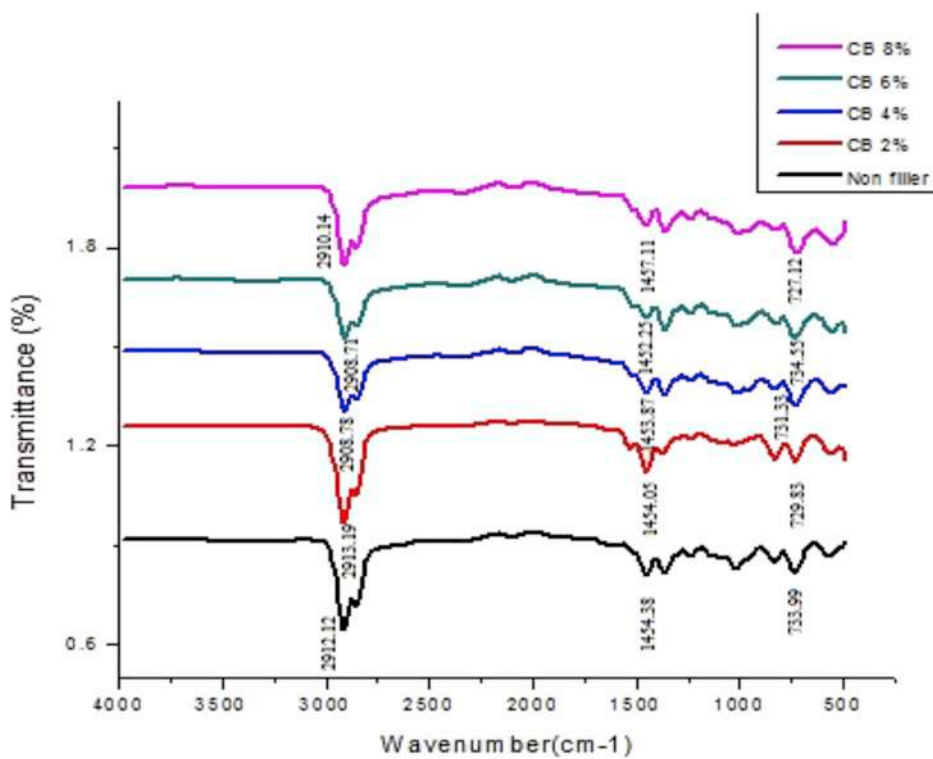


Fig. 5. FTIR Spectra of natural rubber compound with CB filler combined (0–8)wt %.

probably because CB fillers were spread evenly on the rubber compound and also possibly due to the number of fillers that were not large enough to affect the bond on natural rubber.

4. Conclusions

The XRD analysis results showed a change in the structure of the sample from amorphous to the crystal structure. SEM characterization shows an even CB filler. The change in peak temperature is not significant from the composition of the CB fillers 2 wt% to 8 wt% from temperatures 366,25C to temperatures 370,78C. However, a decrease in Heat occurs with increasing CB content from 403.38 to 360.68 mJ. FTIR spectra between nanocomposites with the addition of CB fillers and without fillers showed no significant difference. This was probably because CB fillers were spread evenly on the rubber compound and also possibly due to the number of fillers that were not large enough to affect the bond on natural rubber.

Declaration of competing interest

Authors confirmed that have not conflict of interest in our article.

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