The effect of carbon black loading and structure on tensile property of natural rubber composite

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The effect of carbon black loading and structure on tensile property of natural rubber composite

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Abstract. Natural rubber composite has been continuously developed due to its advantages such as a good combination of strength and damping property. Most of carbon black (CB)/ Natural Rubber (NR) composite were used as material in tyre industry. The addition of CB in natural rubber is very important to enhance the strength of natural rubber. The particle loading and different structure of CB can affect the composite strength. The effects of CB particle loading of 20, 25 and 30 wt% and the effects of CB structures of N220, N330, N550 and N660 series on tensile property of composite were investigated. The result shows that the tensile strength and elastic modulus of natural rubber/CB composite was higher than pure natural rubber. From SEM observation the agglomeration of CB aggregate increases with particle loading. It leads to decrease of tensile strength of composite as more particle was added. High structure of CB particle i.e. N220 resulted in highest tensile stress. In fact, composite reinforced by N660 CB particle shown a comparable tensile strength and elastic modulus with N220 CB particle. SEM observation shows that agglomeration of CB aggregates of N330 and N550 results in lower stress of associate NR/CB composite.

1. Introduction

Natural rubber (NR) is widely used in today life. The important mechanical properties of NR are tensile property, tearing strength, wear resistance, etc. Carbon black can effectively reinforce NR because of the hydrophobic surface of CB filler matched to hydrophobic surface of NR [1,2]. In related to the particle size, the size of CB aggregate of 0.1-0.2 μm is also important in determining the utility of the CB reinforcement in NR. The aggregate then form agglomeration of 10-1000 μm in size. As the count of unit mass increases, aggregate interstitial spacing is reduced. Those properties of aggregates are called 'structure'. The aggregate that contained more particles were said has a higher structure [3]. The work on CB series of N990, N326, N351 and N358 reinforced NR shown that the frictional coefficient under abrasion test of SBR was increased as the structure of CB is higher [4]. The tensile strength and elastic modulus of CB reinforced EPDM was show the increase in tensile strength and elastic modulus as the surface area is increased, especially for N472 that has the largest surface area [5].

This work investigated the effect of CB loading on the tensile property of NR/CB composite. The effect of the CB structures i.e. CB N220, CB N330, CB N550 and CB N660 on tensile property also investigated. The tensile properties of NR without CB were investigated as comparison. SEM-

EDX images are used to explain the effect of the CB loading and structure on tensile property of NR/CB composite.

2. Materials and Methods

2.1. Materials

Materials use in this study was NR in form of latex. Latex was originated from NR cultivation in Lampung Province, Indonesia. Amoniac was use as anticoagulant of lateks during lateks transportation. Formic acid was used as coagulant after lateks was mixed with sulfur and carbon black. Carbon black N330 was used to investigate the effect of CB loading. Carbon black series of N220, N330, N550 and N660 were used to investigate the structure effect on the tensile property of NR/CB composite.

2.2. Composite Preparation

In preparation of composite, before mixing, all materials were weighted according to the percentage of its ingredients in Table 1. Table 1 show the composition of natural rubber with formic acid, refer as NRf1 and NRf2. NR/CB composite with different percentage of CB refer as NR/CB20, NR/CB25 and NR/CB30. Natural rubber with sulphur, refer as NRfS1, NRfS2, NRfS3 and natural rubber filled with carbon black structure, refer as NR/CB samples.

Table 1. Ingredients of NR and NR/CB composite

Sample	Natural Rubber	Formic acid	Sulphur	Carbon Black
NRf1	100	2	0	0
NRf2	100	3	0	0
NR/CB20	78	2.23	0	20
NR/CB25	73	2.23	0	25
NR/CB30	68	2.23	0	30
NRfS1	100	2	2	0
NRfS2	100	2	4	0
NRfS3	100	2	6	0
NR/CB	80	2	4	20

NRf : Natural rubber with formic acid.
NR/CB20: Natural rubber with addition of 20% carbon black. Carbon Black structure was N330.
NRfS1: Natural rubber with formic acid and sulphur.
NR/CB: Natural rubber with addition of different carbon black structure.

Preparations of the sample were as follow: firstly, latex was stirred for 2 min. For NRf sample, lateks was prepared for the second step. For NRfS sample, sulphur were added to latex and mixed. For NR/CB composite sample, the CB was added to latex and mixed. Secondly, each sample was mixed with formic acid for 20 min before being place in the mould. Thirdly, each sample mixtures was poured into 50 x 50 x 5 mm moulds then let them sit for 30 min. Fourthly, each sample was taken out from the plastic mould and pressed under the load of 8 tons for 30 min. After pressing, each sample was taken out from the mould and place outside to dry. Finally, after 24 h drying, each sample was placed in the oven for curing at 150°C for 30 min. Sample then were cut to form dumble specimen.

2.3. Tensile Test and SEM Observation

Tensile test was prepared according to ASTM D412 Standard Test Method for Vulcanised Rubber and Thermoplastic Elastomer-Tension. The tensile rate was 500 mm/min. Temperature and humidity was 23°C and 52%, respectively. The SEM observation was performed on the cross section of the specimen under tensile load. The equipment for SEM observation and EDX analysis was JEOL JSM-6510LA and SEM-EDX EVO MA10.

3. Result and Discussion

Latex was susceptible to host of strong micro bacterial activity during the maturation because of the richness of the organic compound. Maturation is the time period between tree tapping and coagula processing [6]. For preservation of the latex, the latex was mixed with ammoniac before coagulation process. Many researchers used commercial dry rubber in making the NR/CB composite. Rarely, CB was added directly into fresh latex. In this research, latex was used as composite matrix after coagulated with formic acid and catalysed with sulphur. Therefore, to find appropriate percentage of formic acid and sulphur to get sufficient tensile strength, the tensile tests were also carried out to unfilled latex that have coagulated with formic acid and vulcanised with sulphur. For coagulation process, the NR was mixed with formic acid as coagulant, refer as NRf sample. In the range of formic acid loading tested in this study, the optimum tensile strength and strain of NRf was 2% as listed in Table 2.

Vulcanization is a process of compounding NR with different chemical. The chemicals such as sulphur, organic peroxide, silane, amino, phenolic resin, etc. Sulphur was widely used by rubber industries. Vulcanisation increases the elasticity of the rubber. The isoprene polymer chain was locked and limited the amount of chain slipping [7]. Tensile strength of natural rubber with sulfur which the samples refered as NRfS sample is shown in Table 2. Tensile stress and elastic modulus of NRfs were increases when sulphur loading up to 6 %, however the strain was decreases. It is suggest that four percent as the best loading of sulphur in refer to balance tensile stress and strain.

Table 2. Tensile Property of NR coagulated with formic acid and vulcanised with sulphur

Sample	Tensile Strength (MPa)	Elongation (%)	E (ε=0.05%) (MPa)
NRf1	0.417	517.3	4.47
NRf2	0.356	309.0	7.77
NRfS1	0.494	383.7	7.57
NRfS2	0.618	397.2	7.44
NRfS3	0.637	377.4	7.82

3.1. The Effect of CB Filler Loading on Tensile Property

Stress-strain curve of NR and NR/CB composite were showed in Figures 1(a)-(d). The mean stress-strain curve of NR and NR/CB composite was shown in Figure 1(e). The stress-strain curve of NR in Figure 1(a) shows the sample break at very high strain as compare to the NR/CB composites in figure 1(b)-(d). Obvious different in stress-strain curve between NR and NR/CB composite was observed in figure 1(e) that shown low stress and high strain of NR compare to NR/CB composite. As the CB loading increases, the composite failure strain decreases. It indicated the brittle behaviour of NR after filled with CB particle. In reverse, elastic modulus increases with CB loading.

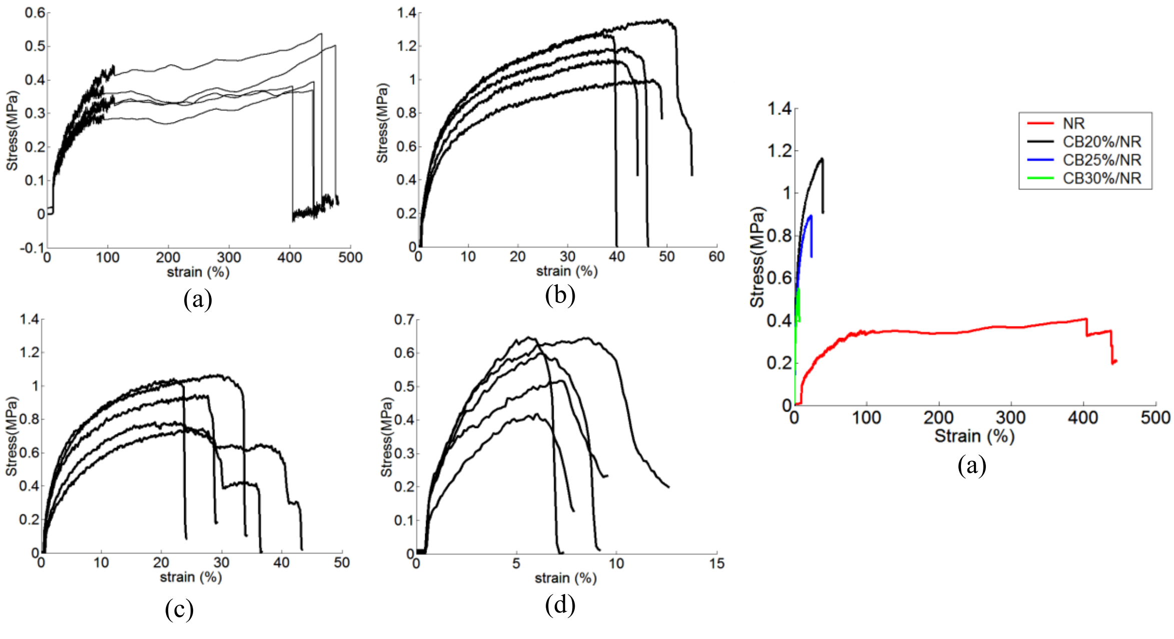

Figure 1. Stress Strain Curve of: (a) Natural Rubber (b) CB20%/NR Composite (c) CB25%/NR Composite and (d) CB30%/NR Composite (e) Mean value of NR and all composites

Composite of NR/CB20 show the highest tensile strength as listed in Table 3. This represents an effective reinforcement of CB in NR. The reinforcing potential is mainly come from formation of flexible filler network and strong polymer-filler coupling. The principal factors determined the capability of reinforcement were (1) Van der walls force between CB and polymer, (2) the chemical cross link of polymer into the filler surface due to free radical reaction between carbon atoms in filler and polymer and (3) the mechanical interlocking of the polymer on to the filler surface.

Table 3. Tensile property of NR and NR/CB composite for different CB loading

Sample	Tensile Strength (MPa)	Elongation (%)	E (ε=0.05%) (MPa)
NR	0.437	442.2	2.48
NR/CB20	1.189	42.5	47.32
NR/CB25	0.916	25.2	34.17
NR/CB30	0.565	6.7	95.75

3.2. The Effect of Structure on the Tensile Property

The NR/CB tensile property as a function of series number was listed in Table 4. High CB structure exhibited a high amount of primary particle per aggregate. In reverse, the aggregate of low CB structure contained a low amount of primary particle. The number system of CB structure means the structure level for example N220 series structure was higher than N330. The sequence of CB structure for highest structure to the lowest was N220>N330>N550>N660. As expected, the tensile strength of NR/CB N220 was the highest among other. In other side, it was interesting that the NR/CB (660) composite show high value of elastic modulus and strain compares with other NR/CB (330) and NR/CB (550) composites. The high strain of NR/CB (660) composite might arise from the shape of aggregate. The aggregate formed agglomerates by weak van der walls force [8]. Like in filler loading that has an optimum value of CB loading on NR, the structure also has the limit which higher structure were no longer contribute to increasing the tensile property of the NR/CB composite. It is probably because the balance between particle loading and aggregate structure was achieved. Carbon black of N472 and N330 has the smaller aggregate size compare with N550 and N770. N472 and N330 have the higher BET specific surface area than N550 and N770 [5].

Table 4. Tensile property of NR and NR/CB composite with diferent structure of CB loading

Sample	Tensile Strength (MPa)	Elongation (%)	E (ε=0.05%) (MPa)
NR/CB (N220)	1.216	163.4	5.75
NR/CB (N330)	0.836	137.0	5.62
NR/CB (N550)	0.610	183.4	4.10
NR/CB (N660)	1.210	199.8	6.88

3.3. The Scanning Electron Microscope (SEM) Observation

SEM micrograph of NR and NR/CB composite that vary in CB loading is shown in figure 2. The homogeneous surface of NR can be observed in Figure 2(a) hence the strain of NR was the highest among others. The homogeneous mixture of 20% CB particle in NR is shown in figure 2(b). Coarse mixture of NR at 25% and 30% CB is shown in Figure 2(c) and (d). This explained the highest tensile strength of NR/CB20 compare to NR/CB25 and NR/CB30.

Figure 3 shows the SEM micrograph of NR/CB composite for different CB structure. The morphology of NR/CB (N220) and NR/CB (N660) in Figure 3(a) and (d) show fewer agglomerates of 2 and 11 μm in size, respectively, compare with NR/CB (N330) and NR/CB (N550) of 8 and 18 μm in size, respectively in Figure 3(b) and (c) . Hence, it result in higher tensile strength of NR/CB (N220) and NR/CB (N660) compare to NR/CB (N330) and NR/CB (N550). Theoretically, the lower structure i.e. NR/CB (N660) has the lowest tensile property. In this research, the less agglomeration of CB N660 is seen in Figure 3(d). It result in quite high tensile strength of NR/CB (N660) composite. Table 5 shows the EDX result of NR/CB (220) and NR/CB (550) composites. Both composite surfaces contain C, N, S and O elements. Both composite surfaces also contained almost the same amount of Carbon and Nitrogen.

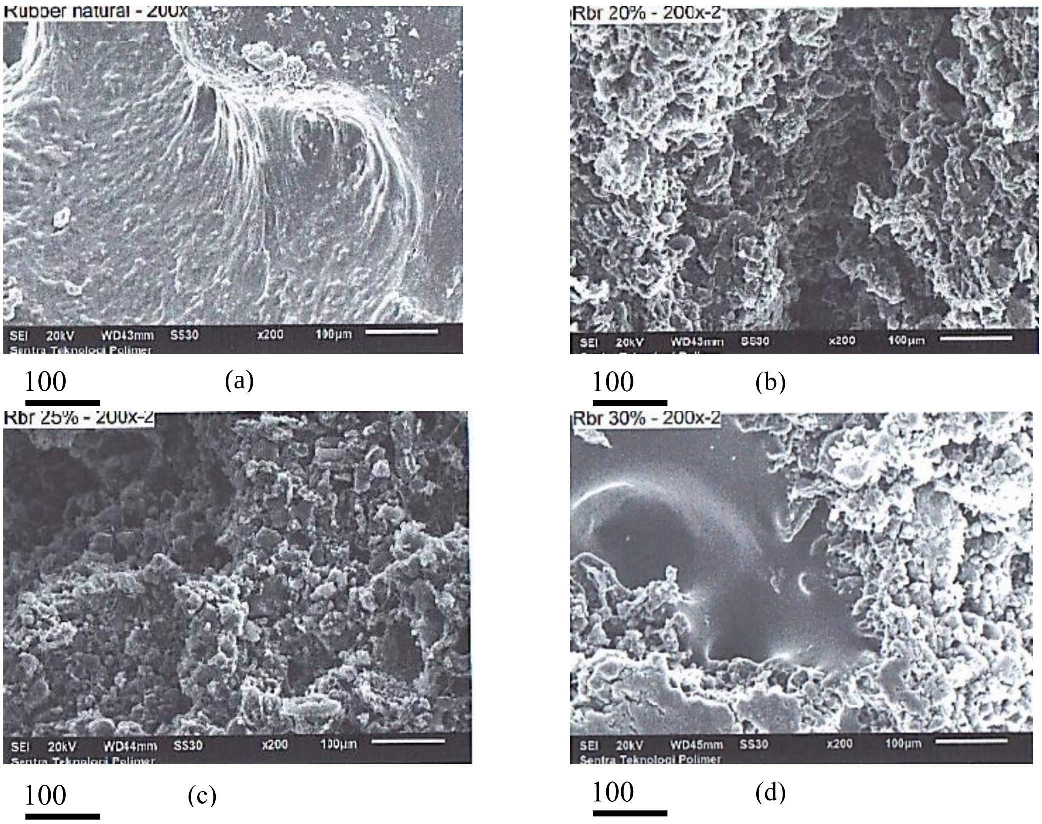

Figure 2. SEM Micrograph of NR/CB Composites: (a) NR (b) NR/CB20 (c) NR/CB25 (d) NR/CB30

The element in both composite was approximately the same. It indicated homogenity of both composite because the composition has the same composition. The different only in CB structure. The mapping of the element of C, N, S and O on the surface of each composite was depicted in Figure 4. It shown distribution of Sulphur and Carbon in composite. From Figure 4(a) and (b), it seems carbon evenly distributed in composite.

Table 5. EDX Result of NR/CB Composite with Different Structure of Carbon Black

Sample	Carbon (%)		Nitrogen (%)		Sulphur (%)		Oksigen (%)	
	wt.%	at.%	wt.%	at.%	wt.%	at.%	wt.%	at.%
NR/CB (N220)	81.40	84.61	14.72	13.12	1.98	0.77	1.91	1.49
NR/CB (N550)	81.59	84.65	13.95	12.41	3.07	2.39	1.39	0.54

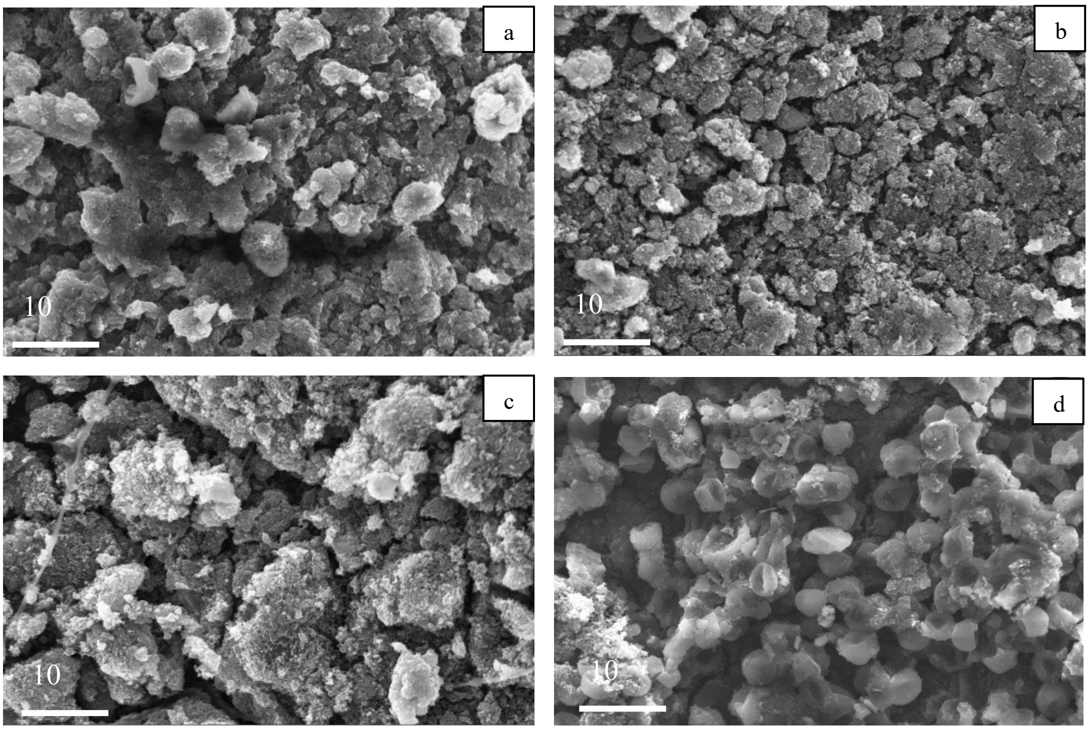

Figure 3. SEM Micrograph of NR/CB Composites contained the Different Structure of CB (a) NR (N220) (b) NR/CB (N330) (c) NR/CB (N550) (d) NR/CB (N660)

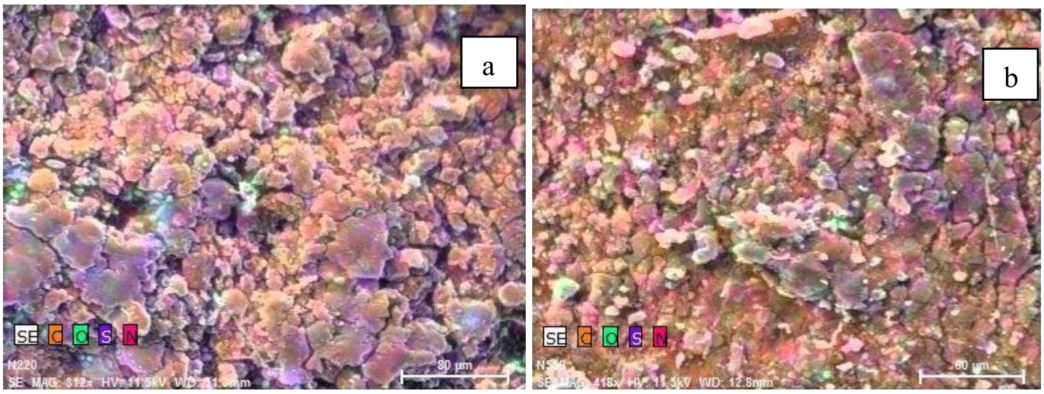

Figure 4. Mapping of Element from EDX Result: (a) NR/CB (N220) (b) NR/CB (N550)

4. Conclusion

Mechanical properties of composite were tested using tensile test. Natural rubber was coagulated with formic acid. It shows that 2% volume fraction of formic acid gives sufficient tensile strength. For the rubber vulcanize with sulphur, the optimal tensile property was shown by 4% sulphur addition. The loading of 20% CB into the NR results in highest tensile strength. Further addition of CB decreases the tensile property of NR/CB composite. The strain decreases with CB loading. In reverse, elastic modulus increases with CB loading. From SEM observation, the surface of NR/CB20 showed the homogeneous CB dispersion in NR matrix. As the loading of CB increases the agglomeration of CB, it decreased the reinforcement of CB that results in low tensile strength.

The structure of CB affected the tensile stress of NR/CB Composite. From SEM observation, less agglomeration of NR/CB (N220) composite results in highest tensile strength. Large agglomeration size of CB N550 and N440 aggregates were clearly seen. This resulted in lower tensile strength of these composites. Although, theoretically, NR/CB N660 composite has lower structure among others. However, high tensile property was achieved because of limited amount of agglomerations of CB N660 agrregates.

From this work, the loading and structure of CB affected the tensile property of NR/CB composite. However, the formation of aggregates and agglomerations of CB also contributed. From EDX result, the surface of NR/CB (N330) and NR/CB (N550) composite consisted of C, S, N and O which matched with the composition of NR/CB composite.

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