

The Effect of Carbon Black and Types of Glass Fibre in Natural Rubber Latex Composite

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Abstract: *In this research, six formulations were prepared consisting of 0%, 0.5%, 1.0%, 1.5%, 2.0% and 2.5% of carbon black (CB) loading into natural rubber (NR) latex. The points corresponding to 0% CB reflect pure rubber latex. Here, the effect of CB with different loadings in NR latex composite was studied. The evaluation of the CB particle dispersion in NR latex was done using optical microscope. The tensile strength of pure NR latex gave the highest value followed by the 1.0%, 0.5%, 2.5%, 1.5% and 2.0% of CB loading. The tensile modulus exhibited maximum values at 2.5% CB loading. The density of these NR latexes increased almost linearly with the increasing percentage of CB loading. 2.5% CB loading was chosen as a standard formulation to produce the NR latex/glass fibre (GF) composite laminates. These laminates were prepared through dipping process. It was found that the mechanical properties of the laminates highly depended on the type of GF used. The presence of woven fibre in NR latex/GF composite laminates increased the tensile strength of the laminates. However, addition of tissue fibre increased the elongation at break of NR latex/GF composite laminates.*

Keywords: Natural rubber latex, glass fibre composite, carbon black loading, latex tensile strength, NR latex/GF composite laminates

1. INTRODUCTION

Natural rubber (NR) latex is one of the most important biosynthesised polymers and renewable natural resources. NR can be isolated from more than 200 different species of plants. However, only one tree source is commercially important, i.e., *Hevea brasiliensis*.¹ NR latex is a very versatile raw material because its behaviour is governed by the composite properties of the vulcanised and the base rubber. Thus, it is possible to alter the balance of strength, heat resistance, anti-viral properties, biodegradability, oil resistance, gas permeability, wet grip and elastic properties to suit the application.

NR latex has an outstanding combination of strength and resilience, as well as excellent overall performance in engineering applications like bearing springs and tires.²⁻⁶ The criteria in choosing the reinforcing filler are the homogeneity and fine dispersion into polymer matrix, strong interaction between filler-matrix, reduced tendency to flocculate and re-agglomerate during storage and at processing stage.⁷

Reinforcing carbon black (CB) into NR can improve the properties of the rubber since pure NR is soft and sticky.⁶ The incorporation of CB into NR latex gives rubber products its black colour and an ultraviolet (UV) absorber that protects rubber products from sunlight, especially for NR and styrene butadiene rubber (SBR).⁸ Studies show that the presence of both silica/carbon in NR vulcanisates produces better mechanical properties compared to those with only CB or silica.^{7,9}

On the other hand, glass fibre (GF) is commonly used as reinforcements to improve the impact damage tolerance and produce light weight composite materials.^{10,11} Several authors have been working with GF in their research.¹²⁻¹⁷ Glass is widely used due to its good properties in strength, stiffness, electrical, weathering, as well as its low cost and low modulus.^{11,13,14,18}

2. EXPERIMENTAL

2.1 Materials

The main materials used for this research were NR latex and GF. Latex concentrate with high ammonia was used in this research, supplied by Lembaga Getah Malaysia (LGM), Sungai Buluh, Selangor, Malaysia. CB (HAF) was used as reinforcement in NR latex. The compounding ingredients used were potassium hydroxide, vulcastab LW, zinc diethyl dithiocarbamate, zinc oxide and sulphur. Three types of E-glass fibre were used, namely the woven roving, chopped stand mat and tissue. GF woven roving 400 was supplied by Mycond Engineering Supply, Puchong, Selangor. GF chopped stand mat and tissue were supplied by Laupik Solution, Petaling Jaya, Selangor. The latex compounding formulation used in this research is presented in Table 1.

Table 1: Latex compounding formulation.

Ingredients	Part by weight (g)
High ammonia latex	167
Vulcastab LW	0.25
Zinc oxide	6
Zinc diethyl dithiocarbamate	3
Sulphur	5
Potassium hydroxide	1
HAF black	0.5, 1.0, 1.5, 2.0, 2.5 (%)

2.2 Processing

Lamination of GF is an important process in the manufacturing of NR latex/GF composite laminates. It involves seven main steps: treatment of GF; determination of total solid contents; preparation of compounding ingredient and HAF carbon black dispersion; dipping; lamination; curing; and post-cure process. In the first steps of treatment, the GF was soaked into acetic acid container at pH 4 ± 1 for 30 min. Next, it was dried in an oven. Before adding CB into NR latex, the CB was dispersed using a ball milling system. Anchoind was used as a dispersing agent. The particle size of the HAF black was measured using light optical microscope.

The compounding ingredients (as in Table 1) were mixed with high ammonia (HA) latex concentrate. The mixture was poured into dipping containers for the dipping process. Each GF was left for 1 min to allow the compounded NR latex compound to penetrate into the GF. Three or two layers of GF were laid, one after another, on a glass plate for lamination process. The NR latex/GF composite laminates were dried in an oven. Subsequently, post-cured was carried out at a $50^\circ\text{C} \pm 5$ for 15 min.

2.3 Testing of NR Latex Composite

2.3.1 Tensile testing

The tensile properties were analysed using a universal tensile machine (Shimadzu Autograph Ag-X 20kN). The crosshead speed was set at 50 mm min^{-1} throughout the test. The samples used for the determination of tensile strength were dumb-bell shape (Type 2) and standard method referred to was ISO 37. The gauge length of each sample was 30 mm.

2.3.2 Density test

The density of NR latex/GF composite laminates can be measured by the standard density methods in accordance to ASTM D 2734 [53]. The density was examined by using Shimadzu (Uni-BLOC UX4205). 5 samples of NR latex/GF composite laminates with dimension of (2×2) cm² were tested and the results were averaged.

2.4 Morphological Studies

2.4.1 Optical microscopy

The CB distribution in NR latex compound was observed by using optical microscope. CB particles were applied on the glass slide and the optical micrograph can be visualised using magnification of between 10 to 50 lines at various intervals.

3. RESULTS AND DISCUSSIONS

3.1 The Effect of CB in NR Latex Composite

The influence of CB on tensile strength and Young's modulus of NR latex compound was investigated. The results of the tensile strength of pure NR latex and NR latex composite with different CB loadings are as illustrated in Table 2. It can be seen that the NR latex gave the highest tensile strength value. This value corresponds to typical pure NR latex, which has an excellent flexibility and low rigidity. When applying the stress to NR materials, e.g., stretching it, the polymer chain would be extended or elongated. However, when the stress is released, the polymer chain returned to the original shape because of elastomeric recoverability property. It is however observed that the incorporation of the CB into NR latex decreases the tensile strength value.

Referring to Table 2, the 1% CB loading gives the highest tensile strength followed by 0.5% and 2.5%. The lowest tensile strength corresponds to the 2.0% CB loading into NR latex. The difference between the highest and the lowest tensile strengths of NR latex/GF composite is about 73.78%. With the presence of CB, the Young's modulus increases with increasing CB-loading. NR latex shows the lowest Young's modulus because of high flexibility properties. However, the addition of CB shows the reduction in elastic properties of NR latex. Increasing the Young's modulus increases the ability of rubber elongating.

Table 2: Properties of NR latex composite.

CB loading (%)	Tensile strength (MPa)	Young's modulus (MPa)	Density (g cm^{-3})
0	25.52 ± 0.73	0.815 ± 0.04	0.938 ± 0.002
0.5	19.61 ± 0.55	1.103 ± 0.01	0.945 ± 0.001
1.0	21.88 ± 0.70	1.206 ± 0.03	0.949 ± 0.001
1.5	14.39 ± 0.77	0.964 ± 0.05	0.948 ± 0.002
2.0	12.60 ± 0.59	0.989 ± 0.06	0.953 ± 0.002
2.5	19.64 ± 0.69	1.294 ± 0.01	0.957 ± 0.002

For filled latex, the Young's modulus increased from 1.0% CB loading but dropped drastically at 1.5%. The Young's modulus seemed to increase slightly at 2.0% CB loading. It can be found that, the difference between 1.5% and 2.0% CB loading was only 2.7%. The increment was however considered insignificant due to the overlapping of the error bar. The CB particles were not completely dispersed into NR latex for 1.5% incorporation as shown in Figure 1. The presence of CB aggregates in NR latex is greater at 1.5% CB so it affect the Young's modulus of NR latex composite.

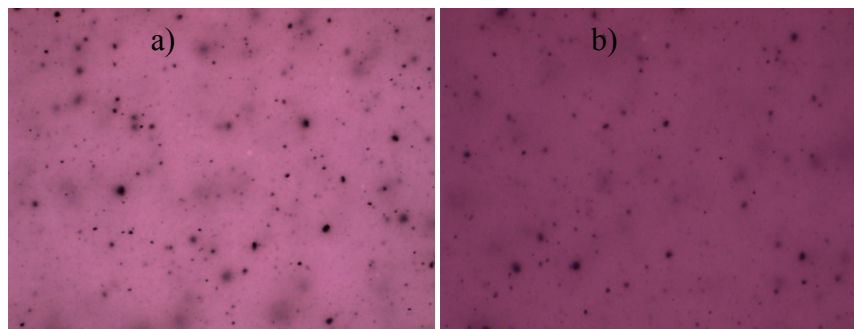


Figure 1: Comparison dispersion between a) 1.5% and b) 2.0 of carbon black in NR latex.

The aggregates would decrease the Young's modulus of NR latex because it reduced the interfacial surface between NR latex and CB particles. The larger particles of CB reduced the potential of CB to penetrate in between the rubber molecules. Increasing the incorporation of CB somehow increased the possibility of CB particle to penetrate in between the rubber particles and thus increased the Young's modulus of NR latex composite. The difference in the Young's modulus values between unfilled NR latex and NR latex with 2.5% CB loading is about 59% (from 0.81 to 1.30 MPa). The results may be due to the presence of CB in NR latex and strong interactions between CB particles and NR latex molecules.

The addition of CB reduced the flexibility and elasticity of NR latex by increasing the rigidity and stiffness of NR latex.

3.2 Effect of CB Loading on Density of NR Latex Composite

It can be seen that the density of these NR latexes increased almost linearly with increasing percentage of CB loading. Even incorporating a small amount of CB particles gave an enhancement in the density value as shown in Table 2.

The minimum density of the NR latex was at 0.9378 g cm^{-3} with no CB added into the NR latex compound. The density value increased to the maximum value of 0.957 g cm^{-3} at 2.5% CB loading. This increase of density values shows an improvement of CB dispersion in NR latex.

3.3 Mechanical Properties of NR Latex/GF Composite Laminate

The 2.5% of CB loading was chosen to produce the NR latex/GF composite laminates due to balanced properties of the composite. The mechanical properties of NR latex/GF composite laminates results are as shown in Table 3. The t-w latex composite showed the highest tensile strength and the t-c latex composite showed the lowest. This indicates that with the presence of woven fibre, the strength of the composite laminates increases drastically. The higher strength of t-w was due to the fibre-bundle arrangement of woven fibres itself. A fibre bundle consists of a lot of fibres and it affects the penetration of the rubber molecules in between of woven fibres.

Table 3: Mechanical properties of NR latex/GF composite laminates.

Samples	Tensile strength (MPa)	Young's modulus (MPa)	Elongation at break (%)
Tissue-tissue (t-t)	16.73 ± 0.63	158.6 ± 8.08	933.9 ± 17
Tissue-chopped strand mat (t-c)	5.263 ± 0.35	115.1 ± 5.02	645.4 ± 29
Tissue-woven (t-w)	65.59 ± 2.4	363.1 ± 6.99	28.4 ± 0.167
Tissue-tissue-tissue (t-t-t)	17.88 ± 1.73	212.3 ± 7.38	1085.9 ± 51
Tissue-chopped strand mat (t-c-t)	5.787 ± 0.28	87.30 ± 14.41	703.2 ± 20
Tissue-woven-tissue (t-w-t)	39.08 ± 1.36	205.9 ± 6.5	32.4 ± 1.14

The addition of one layer of tissue fibre increased the tensile strength of composite laminates of the t-t-t and t-c-t fibre. However, the tensile strength values do not show obvious differences between the two layers and the three layers between (t-t and t-t-t) and (t-t and t-c-t) fibre, which were 6.4% and 2.8%

respectively. However, the addition of more than one layer of tissue fibre reduced the tensile strength of t-w fibre. This happened because of the difficulty of the rubber particles to impregnate between the tissue fibres, which have close arrangement to each other. However, for woven fibre, rubber can easily penetrate between the woven fibres. Woven fibre is a combination of fibre bundles, and the gaps between bundle to bundle fibres give higher possibility to rubber penetrate and adhere into fibre bundles as shown in Figure 2.

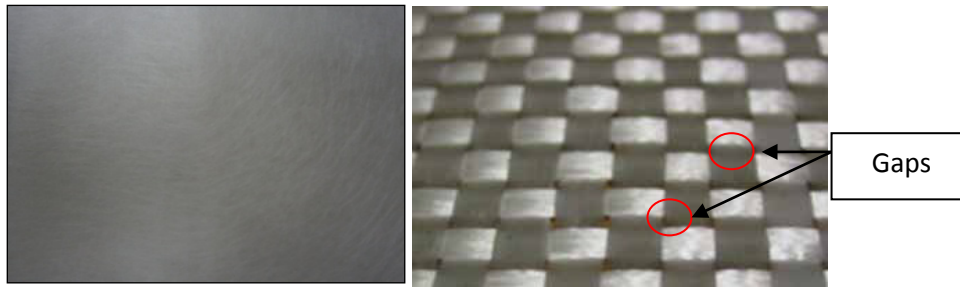


Figure 2: Different arrangement of glass fibre: a) tissue fibre and b) woven fibre.

For the two layers laminating system, the maximum mechanical properties of Young's modulus are 363.10 MPa shows by t-w fibre. Referring to Table 3, the maximum elongation at break is at 933.9% for the t-t fibre. Elongation at break for composite laminates, which decreased linearly in order of $t-t > t-c-t > t-w$ was shown in Table 3. The increasing value of elongation at break is most probably due to presence of woven fibre in NR latex/GF composite laminates.

For the three-layer laminating system, the maximum values of elongation at break and tensile modulus are 1085.87% and 212.26 MPa respectively. These mechanical properties are for t-t-t fibre as shown in Table 3. The addition of tissue fibre increased the elongation at break of NR latex/GF composite laminates. It is because this addition increased the ability of t-t-t fibre pulled until it reached the elastic limit. The arrangement of tissue fibre is close to each other causing incomplete wetting. NR latex compound faced difficulties in impregnation and wetting the fibres, as more rubber seems to adhere at a surface of the tissue fibre. Referring to Table 3, adding one layer of tissue fibre into t-t-t increases the elasticity of the composite laminates due to increase in the Young's modulus. However, addition of one layer of tissue fibre in laminating system for t-c-t and t-w-t reduces the Young's modulus.

4. CONCLUSION

In this study, the influence of CB loading on the types of GF was examined. The tensile strength of NR latex composite is much higher with the addition of 1% CB loading. The maximum tensile modulus and density were found to be 1.294 MPa and 0.9566 g cm³ respectively. The density of these NR latexes increased linearly with the increasing percentage of CB loading. For the NR latex/GF composite laminates, the properties of the laminates depend on the type of glass fibre used.

For the two-layer fibre, the presence of woven glass fibre increased the tensile strength of laminates. However, the addition of tissue fibre in laminate composite reduced the tensile strength of the three layer of fibres. It happens because of the difficulty of the rubber particles to impregnate between the fibre tissue. For elongation at break of the two-layer and three-layer fibres, addition of woven fibre reduced the ability of laminates elongating, but adding one more tissue fibre increases the elongation at break and tensile modulus of NR latex/GF composite laminates.

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