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Reinforcement of Prevulcanised Natural Rubber Latex Films by Banana Stem Powder and Comparison with Silica and Calcium Carbonate

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This study investigates the effect of incorporating banana stem powder, surface modified with 2% of sodium hypochlorite solution, as a reinforcing filler on the properties of natural rubber latex (NRL) compound and its films. Increasing the banana stem powder loading did not affect much of the swelling index of the latex films but slightly increased the moduli [modulus at 100% (M_{100}) and 300% (M_{300}) elongations] of the latex films. Tensile strength and elongation at break of the latex films showed some reduction with the increase in the banana stem powder loading. The tear strength of the NRL films containing the banana stem powder is higher than the tear strength of the unfilled prevulcanised NRL films. In comparison with other commercial fillers, calcium carbonate and colloidal silica, banana stem powder produced prevulcanised NRL films of comparable mechanical properties.

Keywords: prevulcanised natural rubber latex films; banana stem powder; calcium carbonate; colloidal silica; mechanical properties

Excellent elasticity, flexibility, antivirus protection, good formability and biodegradability of natural rubber latex (NRL) made it a preferable material for the manufacture of medical products such as medical gloves, condoms, blood transfusion tubing and catheters[1–3]. Due to the worldwide spread of epidemic diseases such as AIDS, hepatitis B and influenza A (H1N1), it becomes increasingly urgent to develop high performance NRL protective products[4].

Poor tear resistance is a common problem encountered in medical gloves and condoms[4]. Fillers such as carbon black[5], ultra-fine calcium carbonate[6], silica[7] and starch[8] are normally added to the NRL products for reinforcement. These fillers, however, are not good enough. Besides strength, the NRL products should also be biodegradable. To produce totally biodegradable NRL products will require the complete change of the raw material (latex). However, to produce partial biodegradable

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NRL products is easier and it can be achieved by adding biodegradable material as one of the components in the NRL formulation. A suitable biodegradable material is natural fibres because natural fibres are abundant, cheap and due to their intrinsic properties could also reinforce the NRL products.

Natural fibres such as flax, hemp, banana and oil palm are known to reinforce polymers[9]. Researchers are investigating the exploitation of natural fibres as reinforcing filler in rubber composites[10–16]. The reinforcement of rubber with natural fibres is possible by combining the elastic behaviour of the rubber matrix with the strength and stiffness of the natural fibres.

The degree of reinforcement of rubber by natural fibres depends on the fibre concentration, fibre dispersion within the rubber matrix, void content and also the adhesion of the fibre to the rubber matrix[17]. Poor fibre dispersion is the biggest problem encountered when mixing rubber with natural fibres and this always results in the reduction of the overall strength of the composites[9]. Poor dispersion of the natural fibres with rubber could be improved by modification of fibre surface by the use of bonding agent and chemical modification of the fibre surfaces[18–20].

This study reports the effect of using banana stem powder, treated with 2% sodium hypochlorite, as filler in prevulcanised NRL compound and its films. The study also covers the effect of using commercial fillers, colloidal silica and calcium carbonate for comparison.

EXPERIMENTAL

The experiment was divided into two parts. The first part was to determine the amount of banana stem powder required to produce prevulcanised NRL films with optimum properties. The second part was to compare properties of the prevulcanised NRL films containing banana stem powder as filler with those prevulcanised NRL films containing the same amount of commercial fillers (calcium carbonate and colloidal silica).

Materials

High ammonia (HA) natural rubber latex (NRL), curative agents (sulphur, zinc oxide, potassium hydroxide, zinc diethyldithiocarbamate (ZDEC) and antioxidant), sodium hypochlorite solution, 33% of colloidal silica and calcium carbonate powder were purchased from ZARM Scientific and Supplies (Malaysia) Sdn. Bhd.

Preparation of Banana Stem Powder Dispersion

Banana stem was cut into small pieces, washed twice with water and shredded using a blender. Shredded banana stems were pressed by a hydraulic press to remove excess water followed by drying in the oven at 110ºC for 2 hours. The dried banana stem was then ground into powder form and sieved using a sieve of 53 µm aperture to remove any foreign matter and large banana stem powder particles.

About 30 g of the dried and sieved banana stem powder was bleached with 450 g of a 2% solution of sodium hypochlorite at 40ºC for 24 hours. After that, the banana stem powder was ball-milled at room temperature for 24 hours. About 5% of ammonia hydroxide was added to maintain the pH of the dispersion around 11 and also to prevent further reaction of sodium hypochlorite solution in the dispersion.

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Preparation of Calcium Carbonate Dispersion

Calcium carbonate dispersion was prepared using the formulation shown in *Table 1*. The preparation of initial coarse slurry was done by mixing the dispersing agent and colloid stabiliser first with water followed by calcium carbonate powder under high speed stirring. The resulting slurry was then ball-milled for 24 h before incorporation into the NRL compound.

Particle Size Measurements

The particle size distribution of the banana stem powder, calcium carbonate and colloidal silica was measured using the Malven Mastersizer 2000S. From the output of the machine, the mean particle diameter (d_{50}) was taken as the average particle size.

Preparation of Prevulcanised NRL Films

The prevulcanised NRL compound was prepared by compounding the ingredients shown in *Table 2*, in a reaction flask, at 70 ± 1ºC. A chloroform number test was used to determine the degree of prevulcanisation of the compound. The prevulcanisation was stopped at chloroform number 3 after which the latex compound was allowed to cool and mature at room temperature for 1 day.

TABLE. 1. FORMULATION OF 50% CALCIUM CARBONATE DISPERSION

Ingredients	Parts by weight (p.p.h.r.)
Calcium carbonate powder	100
Anchoid dispersing agent	2
10% Potassium hydroxide	0.5
Water	97.5

TABLE. 2. FORMULATION FOR PREVULCANISED NRL COMPOUND

Ingredients	Parts by weight (p.p.h.r.)		
	Banana stem powder	Calcium carbonate	Colloidal silica
60% HA Natural rubber latex	100	100	100
10% Potassium hydroxide	0.5	0.5	0.5
50% Sulphur	1.5	1.5	1.5
50% Zinc oxide	1	1	1
50% Zinc diethyldithiocarbamate	1	1	1
50% Antioxidant 2246	1	1	1
10% Banana stem powder	5, 10, 15, 20	-	-
50% Calcium carbonate	-	5	-
33% Colloidal silica	-	-	5

Preparation of Prevulcanised NRL Dipped Films

Films were prepared by coagulant dipping using 10% calcium nitrate as the coagulant. Cleaned and dried aluminium plate formers were dipped in the coagulant for 10 s followed by drying in the oven at 100ºC for 15 minutes. The dried formers were then cooled at room temperature for 5 min and dipped into the latex tank with 10 s dwell time. After that, the formers were withdrawn and dried in the oven at 100ºC for 30 minutes. The dried NRL films were then stripped from the aluminium plates and stored in the desiccators before further tests were conducted.

Swelling Equilibrium Test

A small sample from each NRL film was taken and weighed (≥ 0.2 g). The sample was then immersed in toluene for 48 h at 40ºC with a replacement of toluene after the first 24 h of soaking. The swollen sample was then wiped with a tissue paper and weighed. Then the sample was dried in an oven at 60ºC until constant weight was achieved. Swelling index is calculated by the equation:

$$\text{Swelling index (\%)} = [(W_2 - W_1) / W_1] \times 100 \quad \ldots 1$$

where W_1 and W_2 are the initial and swollen weights of the latex films respectively.

Tensile Properties

The tensile properties of the films were determined by using an Instron Universal tensile testing machine according to *ASTM D412*. Dumbbell test pieces were cut from dipped films with a Wallace die cutter. A cross-head speed of 500 mm/min was used, and the tensile test was conducted at 25ºC. The tensile strength, modulus at 100% and 300% elongation (M_{100} and M_{300}) and elongation at break were evaluated from the tensile test data.

Tear Strength

The tear test was done by pulling the angled tear specimen using the Instron Universal tensile testing machine according to *ASTM D624*. The tear strength of the NRL films was measured by dividing the force required to completely tear the specimen with the thickness.

Scanning Electron Microscopy (SEM)

Fractured surfaces of the tensile test pieces were coated with chromium using an Emitech K575X sputter coater (Emitech, Houston, TX) for 30 seconds. The coated surfaces were next scanned using a ZEISS SUPRA 35 VP scanning electron microscopy (SEM).

Transmission Electron Microscopy (TEM)

The dispersions containing banana stem powder, calcium carbonate and colloidal silica were deposited onto glow discharged carbon coated grids and allowed to dry. These grids were analysed with a Philips CM 12 CRYO transmission electron microscope.

RESULTS AND DISCUSSION

Effect of Banana Stem Powder Loading on the Swelling Index of Prevulcanised NRL Films

The interaction between banana stem powder and the latex rubber matrix could be better assessed by doing the swelling test. The improved adhesion between banana stem powder particles and latex rubber matrix reduced the entry of solvent and resulted in lower swelling index of the films. However, factors such as the formation of voids/holes, types of filler and amount of filler also influence the solvent uptake. The results showed that banana stem powder loading has little effect on the swelling index and latex films with 10 p.h.r. powder loading showed the highest swelling index, 20% more compared to the control (0 p.h.r.) films. This slight increase was due to the formation of voids/holes that provided a pathway for the solvent to progress[21] as shown in SEM images. The formation of voids/holes was not observed in the control (0 p.h.r.) latex films thus the solvent uptake was lower.

TABLE 3. SWELLING INDEX OF PREVULCANISED NRL FILMS WITH VARIABLE BANANA STEM POWDER LOADING

BSP Loading (p.h.r.)	Swelling Index (%)
0	337.26
5	377.30
10	407.62
15	378.27
20	378.63

Effect of Banana Stem Powder Loading on the Mechanical Properties of Prevulcanised NRL Films

Initially, the moduli (M_{100} and M_{300}) reduced with the addition of 5 p.h.r. banana stem powder. However, upon further addition of banana stem powder, the moduli increased surpassing that of the control films. The initial drop in the moduli was due to the softening effect of the latex films by the addition of banana stem powder particles that have whiskers formation around them. This finding was the opposite of the reported observation by Mathew and co worker[22] where moduli increased with the addition of filler. This new finding is important especially in the latex glove industry where stronger and softer gloves can be produced without incorporating chemicals such as softeners. Normally, stronger gloves are stiffer and these gloves are uncomfortable to wear because it will reduce the blood circulation of the wearer's hand, especially on prolonged use.

It was observed that increasing banana stem powder loading up to 20 p.h.r. contributed to fibre agglomeration, as shown in SEM images in *Figure 1*. These aggregates were responsible for the observed high modulus of

TABLE 4. PROPERTIES OF PREVULCANISED NRL FILMS WITH VARIABLE BANANA STEM POWDER LOADING

BSP Loading (p.h.r.)	M_{100} (MPa)	M_{300} (MPa)	Tensile strength (MPa)	Elongation at break (%)	Tear strength (N/mm)
0	0.790	1.672	23.25	1067	44.66
5	0.491	1.118	19.67	1103	65.55
10	0.571	1.260	16.20	1006	64.65
15	0.817	1.689	15.48	920	64.34
20	0.890	1.985	14.08	827	38.15

*BSP = Banana stem powder

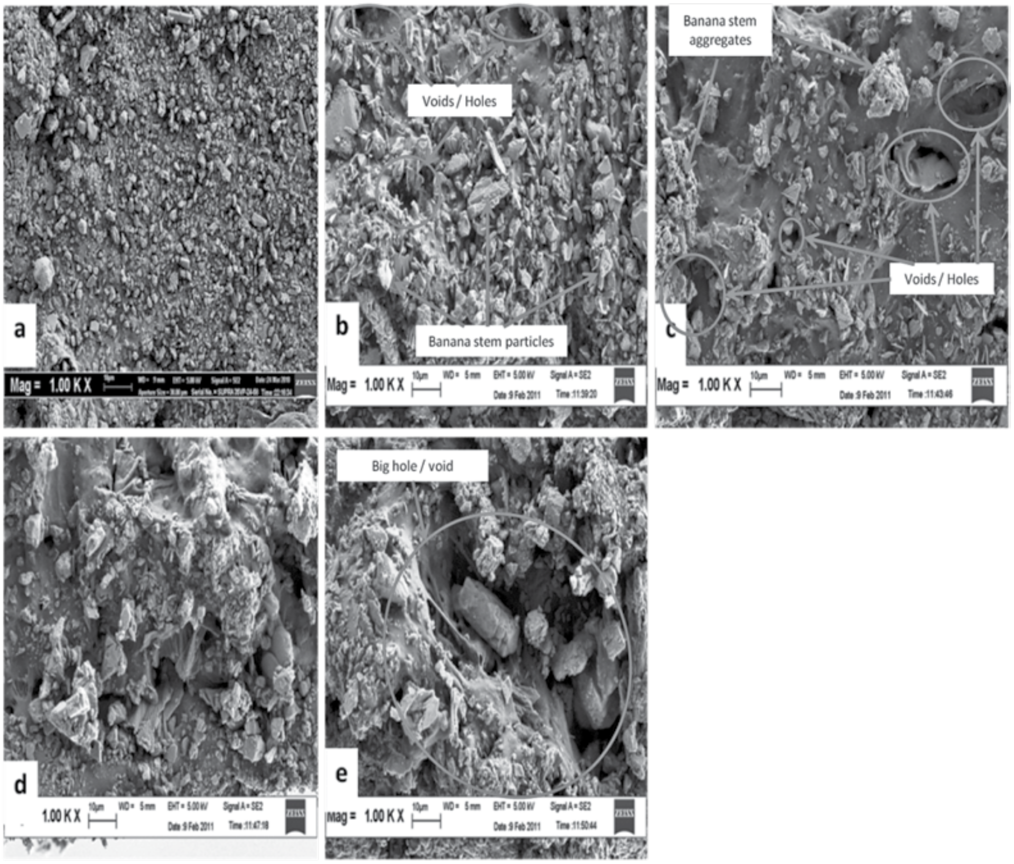


Figure 1. SEM images of tensile fractured surfaces of prevulcanised NRL film containing (a) no filler (control), (b) 5, (c) 10, (d) 15 and (e) 20 p.h.r. banana stem powder.

these films since the portion of latex rubber matrix surrounded by the agglomerates could not react to stress during deformation.

The films containing banana stem powder have lower tensile strength and the tensile strength of the prevulcanised NRL films reduced with increasing banana stem powder loading. This observation is similar to the results reported by Debasish and co workers[23]. The reduction of tensile strength with banana stem powder loading was due to the inefficient transfer of load from the latex rubber matrix[23] to the banana stem powder and the disruption of the formation of crystals by the powder in the prevulcanised NRL films during tensile test[24]. According to Debasish[23] incorporating porous natural fibre as filler in the rubber matrix provided weak sites for stress transmission to its surrounding which resulted in lower tensile strength. The formation of fibre aggregates was another factor that was responsible for the reduction of tensile strength with increasing fibre loading. The fibre aggregates acted as stress concentrators. When the prevulcanised NRL films were stretched, as during the tensile test, these aggregates were broken. This resulted in premature failure of the films.

From the SEM images, shown in *Figure 1*, the formation of aggregates increased the fibre-fibre interaction rather than rubber-fibre interaction. This contributed to the lowering of tensile strength as explained by Azura and co workers[25].

The effect of increasing banana stem powder loading on the elongation at break is in tandem with the trend shown by the moduli (M_{100} and M_{300}) of the films. The elongation at break seemed to increase slightly when the powder loading was increased to 5 p.h.r. This increase was due to the softening effect as reported earlier for the modulus. As more powder was added, the prevulcanised NRL films became stiffer, as evident from the increase in the modulus (M_{100} and M_{300}), and this reduced the elongation at break of the films. This behaviour was also reported by Jacob and co workers[26].

The tear strength of prevulcanised NRL films reached a maximum value at 5 p.h.r. and reduced on further banana stem powder loading. The tear strength of the filled prevulcanised NRL films except for those containing 20 p.h.r., was still higher than that of the unfilled prevulcanised NRL films. This result showed that the presence of banana stem powder from 5 p.h.r. to 15 p.h.r. improved the tear strength of the prevulcanised NRL films. When 20 p.h.r. of highly porous banana stem powder was incorporated into the latex compound, the porous areas were not filled with the latex rubber and formed voids. These voids could act as initial flaws, which lead to localised stress concentration during deformation. For this reason, both tensile strength and tear strengths reduced with increasing powder loading[27].

The observed increase in tear strength with powder loading was due to the ability of the powder particles to deviate cracks, as illustrated in *Figure 2*.

For the unfilled film, during the tear test, the crack propagated straight across the tear specimen because there was nothing to divert the crack. Therefore, the unfilled films showed smooth tear fractured surfaces. However, for the filled films, as the crack propagated across the tear specimen it encountered the powder particles. The hard powder particles deviated the cracks and thus the filled films exhibited "knotty' tearing behaviour which was responsible for the observed increase in the tear strength and also the rough tear fractured surfaces of the tear specimens as illustrated in *Figure 3*.

The tearing mode of the prevulcanised NRL films is shown in *Figures 3* and *4*. The films containing 5, 10 and 15 p.h.r. of banana stem powder showed knotty torn surfaces with multiple tearing load peaks whereas those films containing 0 and 20 p.h.r. banana stem powder showed smooth torn surfaces with single load peak. The knotty tearing mode showed by the films containing 5, 10 and 15 p.h.r. banana stem powder was regarded as being symptomatic of reinforced rubber films[28–31]. The banana stem powder particles or aggregates of the banana stem powder that were formed in the films, were able to deviate the propagation of crack and transformed the mode of tearing from smooth to knotty tearing as shown in *Figure 2*. However, the ability of the aggregates to deviate the propagation of tear reduced as the size or the amount of the aggregates in the films increased as in the case of films containing 20 p.h.r. banana stem powder. This is because the fibre-fibre interaction is more dominant than the fibre-rubber interaction. As a result, the tear strength dropped.

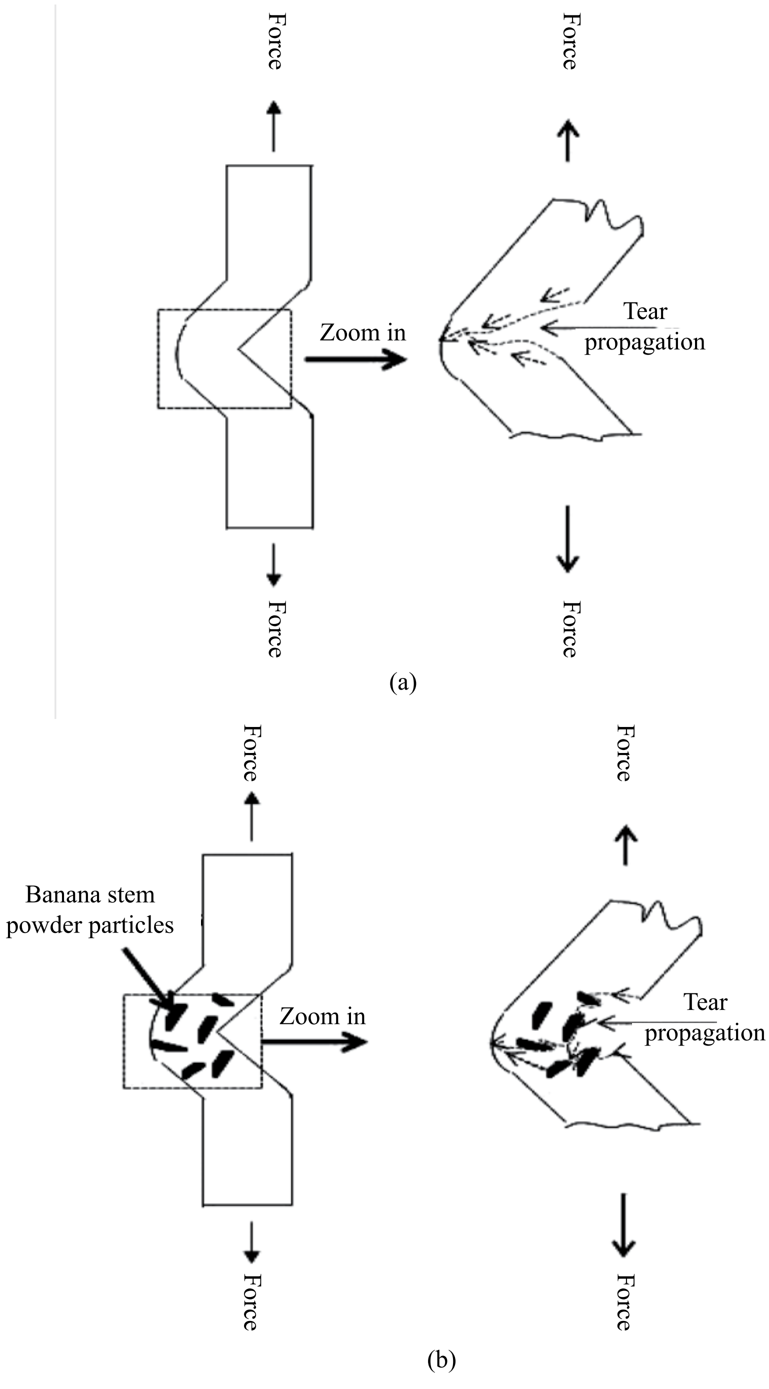


Figure 2. Schematic representation of crack propagation of the prevulcanised NRL films (a) without banana stem powder (0 p.h.r.), and (b) containing banana stem powder as filler.

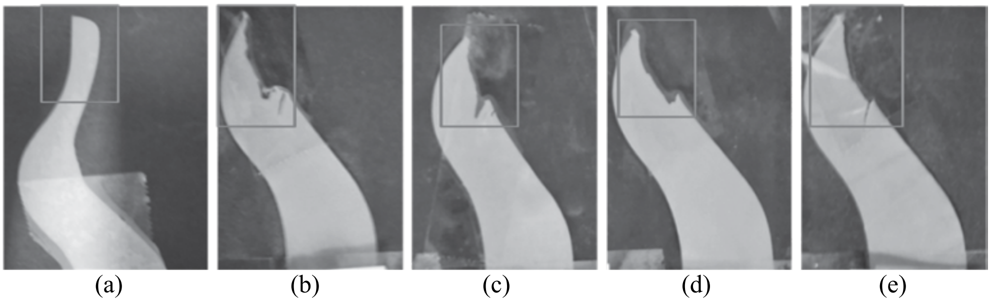


Figure 3.Tear test pieces after rupture of prevulcanised NRL films containing (a) no filler (0), (b) 5, (c) 10, (d) 15 and (e) 20 p.h.r. banana stem powder.

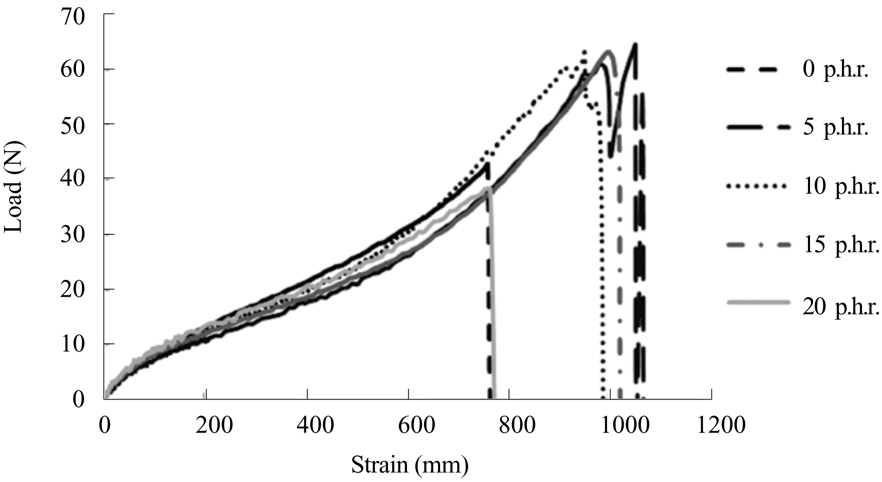


Figure 4. Graph of load vs. strain in prevulcanised NRL films containing variable banana stem powder loading.

It was also observed in *Figure 4* that the films containing 5 to 15 p.h.r. powder showed multiple tearing peaks. The formation of multiple tearing peaks was due to the cyclic increase and decrease of tearing force during the propagation of the tear across the tear specimen. Each increase in the tear forces produced a rapid tear that released the concentrated stress and resulted in increased torn length[32].

Comparison of Banana Stem Powder and Filled Films with Commercial Fillers

The results obtained from the first part of this study showed that the prevulcanised NRL films containing 5 p.h.r. banana stem powder have better mechanical properties compared to the other films. Therefore, in the second part of this study, these films were used to compare with those films containing the same amount of commercial fillers (silica and calcium carbonate).

Figure 5 showed the particle size distribution and the mean particle size (d_{50}) of banana stem powder, calcium carbonate and colloidal silica, measured by particle size analyzer. Bimodal particle size distribution was observed for banana stem powder. The bimodal distribution was due to the semi-crystalline regions that were present in the fibrous materials; fibre whiskers (responsible for the small particle size) and crystalline area (responsible for the large particle size). It also was noted that the number fraction of particles in the second peak was very large compared to the fraction of particles in the first peak. The first peak showed the presence of small particles in the range of 0.2 – 1.52 µm. Meanwhile, the particle size distribution for calcium carbonate is the polymodal distribution, the range of particle size wherein 0.2 µm to 1.52 µm, 1.52 µm – 4.79 µm, 8.48 µm to 18.21 µm, and the highest particle size was in the range of 1.52 µm – 4.79 µm. The colloidal silica, on the other hand, exhibited a unimodal distribution (between 0.10 – 0.18 µm) with mean particle size of 0.14 µm, as shown in *Figure 5 (b)*.

Figure 6 showed the TEM images of dispersions of banana stem powder, calcium carbonate and colloidal silica. It seemed that the fillers have different particle size, shape, and distribution. Banana stem powder has irregular shape particles and there were fibre whiskers formed around the particles. According to Zuluaga and co workers[33] the fibre whiskers contributed to the formation of amorphous regions around the banana stem powder particles. These fibre whiskers were responsible for the low stiffness (low moduli) and high toughness (high elongation at break) of the latex films.

Calcium carbonate, a mineral filler that has regular shaped particles[34], usually decreases the impact energy of the polymers. It is a highly crystalline material in contrast to the colloidal silica, which is a suspension of fine amorphous, non-porous and spherical silica particles in water. From *Figure 6 (c)*, it can be seen that colloidal silica particles seemed to aggregate together and form a highly porous cluster. In this study, the total solid content for colloidal silica used was 32%. This is because the colloidal silica contained a large amount of water.

The effect of different types of filler on swelling index, M_{100}, M_{300}, tensile strength, elongation at break and tear strength of the prevulcanised NRL films is summarised in *Table 4*. It was observed that the latex films containing banana stem powder and colloidal silica swelled more than the unfilled films and the films containing calcium carbonate. However, a slight increase in the swelling index of the filled prevulcanised NRL films over the unfilled films suggested that incorporation of small amount (5 p.h.r.) of fillers did not significantly affect the interaction between filler particle and latex rubber matrix.

The ability of the filled latex films to swell in toluene was mainly due to the behaviour of the filler itself. The increase in swelling of the films containing banana stem powder and colloidal silica was due to absorption of the solvent by amorphous regions (the whiskers) of the banana stem powder particles and the porous colloidal silica aggregates, respectively.

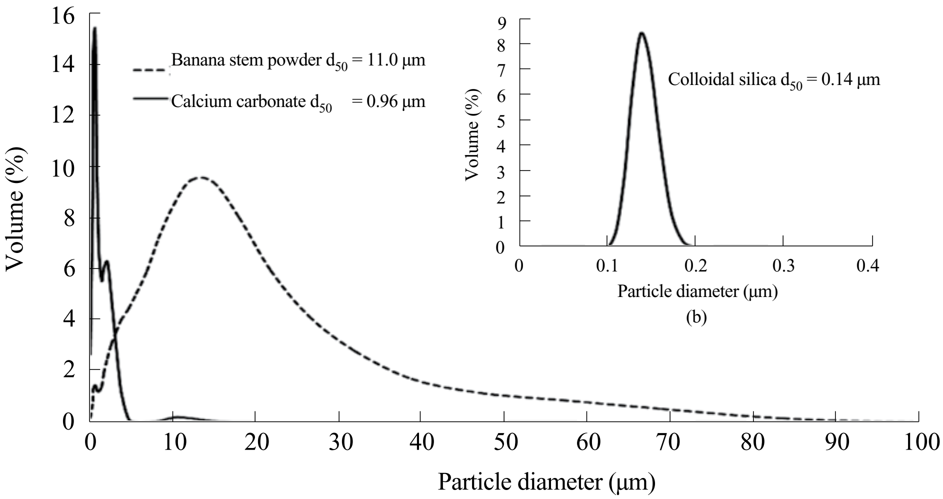


Figure 5. Particle size distribution and mean particle size (d_{50}) of different types of filler, (a) banana stem powder and calcium carbonate and (b) colloidal silica respectively.

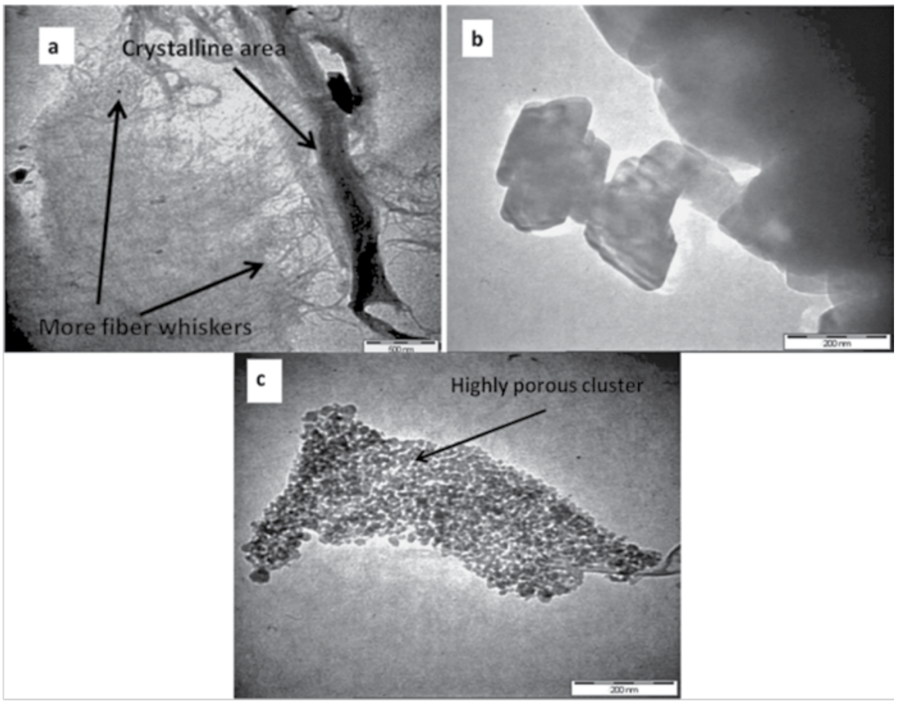


Figure 6. TEM images of (a) banana stem powder, (b) calcium carbonate and (c) colloidal silica.

TABLE 4. PROPERTIES OF PREVULCANISED NRL FILMS WITH DIFFERENT TYPES OF FILLER

Samples	Swelling Index (%)	M_{100} (MPa)	M_{300} (MPa)	Tensile strength (MPa)	Elongation at Break (%)	Tear Strength (N/mm)
NRL (Unfilled)	337.26	0.790	1.672	23.25	1067	44.66
NRL / BSP	377.30	0.491	1.118	19.67	1103	65.55
NRL / $CaCO_3$	333.10	0.698	1.478	27.48	1126	50.58
NRL / CS	401.03	0.661	1.353	22.11	1063	85.29

*BSP = Banana stem powder
*$CaCO_3$ = Calcium carbonate
*CS = Colloidal silica

The non-porous calcium carbonate particles cannot absorb the solvent and therefore, the swelling of the films was lower.

The films containing calcium carbonate exhibited the highest tensile properties followed by the unfilled films, colloidal silica filled films and finally by the banana stem powder filled films. The increase in the tensile properties of prevulcanised NRL films by incorporation of a small amount of calcium carbonate (below 15 p.h.r.) was expected since it has been reported before[6,27,35]. The trend observed for the tensile properties of the films was mainly due the particle size distribution of the fillers and the capability of the particles to rearrange when incorporated into the latex rubber matrix. Calcium carbonate with a polymodal particle size distribution has a well packed and good inter particles interaction with latex rubber matrix. Smaller particles fill the gaps between larger ones, thus improving the particle packing which resulted in improved interaction between the calcium carbonate particles and the latex rubber matrix. This enhanced the tensile properties of the prevulcanised NRL films. Meanwhile, the observed low tensile strength of the latex films containing banana stem powder and silica particles was due to the formation of voids or small holes as shown in *Figure 7*, which could act as initial flaws, leading to the localised stress concentration during deformation.

The effect of the fillers on the tear strength of the prevulcanised NRL films showed that filled NRL films have higher tear strength compared to the unfilled NRL films. This was due to the reinforcing effect of the fillers in the latex rubber matrix. Prevulcanised NRL films containing banana stem powder have higher tear strength than those containing calcium carbonate but lower than that containing colloidal silica. The presence of different types of fillers with different mean particle sizes provides a potential to restrict the crack growth during deformation as discussed earlier as in *Figure 2*. The latex films containing colloidal silica showed the highest tear strength. This was mainly due to the formation of small aggregates as shown in *Figure 7*. These aggregates were able to deviate the propagation of crack and produced knotty tearing thus increasing the tear strength of the latex films.

Figures 8 and *9* showed the mode of tearing and tearing curves of the prevulcanised NRL films containing different types of fillers. It can be seen that latex films containing banana stem powder and colloidal silica showed knotty tear mode with high tear strength. This behaviour has been reported by many researchers

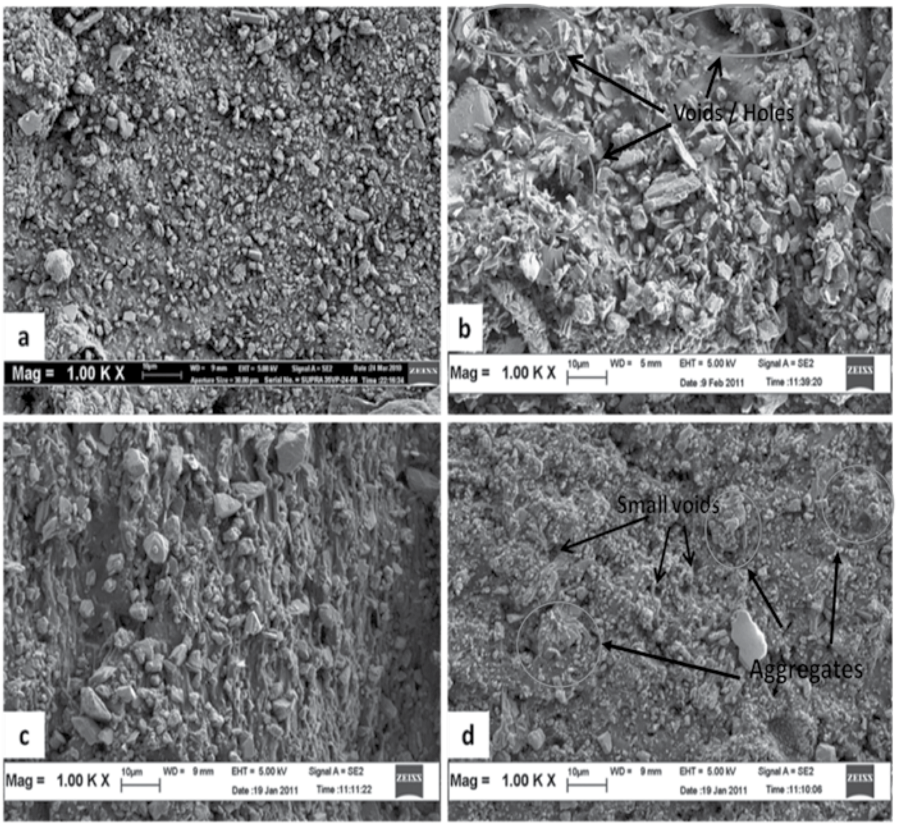
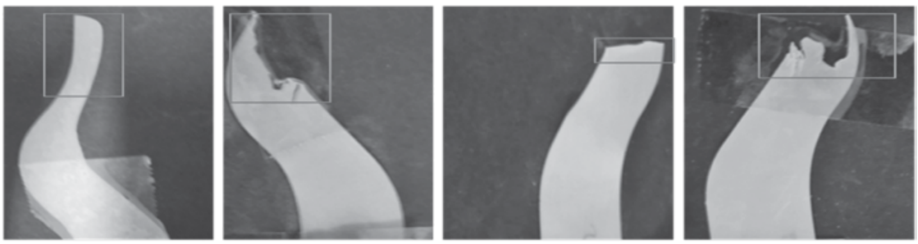


Figure 7. SEM images of tensile fractured surfaces of (a) NRL film containing no filler (control), filled NRL films with different types of filler (b), (c), and (d) for banana stem powder, calcium carbonate and colloidal silica respectively.



(a) (b) (c) (d)

Figure 8. Tear test pieces after rupture of prevulcanized NRL films containing different types of filler (a) no filler, (b) banana stem powder, (c) calcium carbonate and (d) colloidal silica respectively.

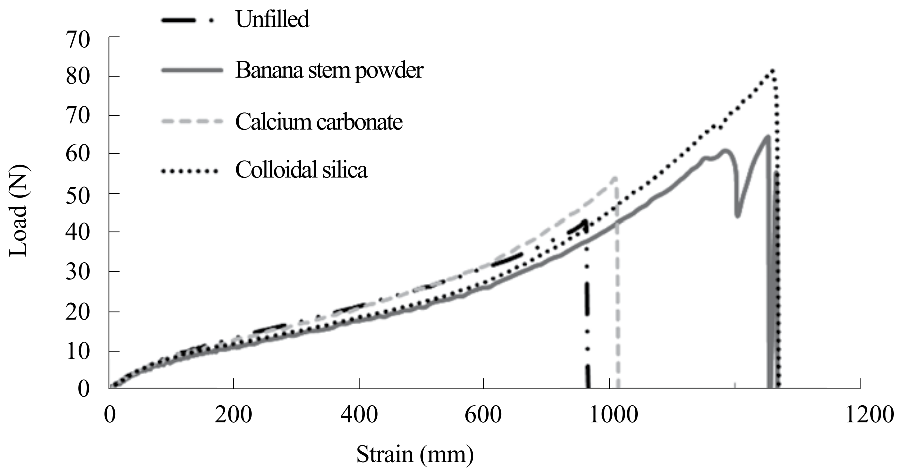


Figure 9. Graph of load vs. strain in prevulcanised NRL films containing different types of filler loading.

studying the reinforcement of rubber by fillers[29–31]. Mullin[29] reported that the role of reinforcing fillers was to increase the strength of rubbers. It appeared that a process giving rise to rapid relaxation of stress ahead of the tip of a growing tear can in principle result in broadening of the tip of the tear and this leads to increased strength. It is considered that this type of process played an important and sometimes dominant role in the reinforcement of rubbers by fillers or by crystallisation. It provided a consistent and coherent mechanism which indicated a source of the roughening of rupture surfaces of reinforced rubbers and of their possible branching and knotty tearing.

CONCLUSIONS

Increasing banana stem powder loading up to 20 p.h.r. did not significantly affect the swelling index of the prevulcanised NRL films. The moduli (M_{100} and M_{300}) of the prevulcanised NRL films decreased initially on addition of banana stem powder but increased again when banana stem powder loading was above 5 p.h.r. Increasing banana stem powder loading reduced the tensile strength of the prevulcanised NRL films. The elongation at break of the films increased initially with banana stem powder loading but reduced below that of the unfilled films after more than 5 p.h.r. of banana stem powder was added. The tear strength of the prevulcanised NRL films showed the same trend as the elongation at break with banana stem powder loading but the tear strength value reduced below that of the unfilled films only when 20 p.h.r. banana stem powder was loaded. Banana stem powder showed bimodal particle size distribution with an average particle size of 11.00 µm where as calcium carbonate has a polymodal particle size distribution with an average size of 0.96 µm. Colloidal silica has unimodal particle size distribution with an average particle size of 0.14 µm, which was smaller than that of the banana stem powder and the calcium carbonate. The prevulcanised NRL films containing banana stem powder have properties comparable to those films containing the same amount of calcium carbonate and colloidal silica.

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