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Tensile Behaviour and Biodegradability of Natural Rubber Latex Incorporated with *Cucumis sativus L.*

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This research focused on the potential of cucumber (*Cucumis sativus L.*) powder (CP) as a reinforcing and biodegradable filler for natural rubber latex (NRL) films. The filler was incorporated into NRL at various loadings ranging from 0 to 10 phr. Unfilled NRL and NRL/CP films were subjected to swelling, tensile, and soil burial tests. The NRL/CP films exhibited a gradual decrease in swelling percentage, from 500% to 415%, and an increase in crosslink density (from 2.74 to 8.5 × 10^{-3} mol/cm^3) with higher filler loading compared to unfilled NRL films. Additionally, the NRL/CP films demonstrated improved results in tensile strength, elongation at break, and modulus at 100% elongation. A soil burial test was conducted for 4 weeks. The NRL films with 4 and 10 phr of CP filler underwent biodegradation as early as week 1, with further biodegradation occurring after 4 weeks of soil burial. The incorporation of CP filler at 4 phr in NRL resulted in a significant synergistic effect, enhancing both the mechanical and physical properties, as well as the biodegradability of NRL.

Keywords: *Cucumis sativus* L.; cucumber powder; natural rubber latex; tensile properties; biodegradability

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In the face of the COVID-19 pandemic, disposable latex gloves have been extensively used for personal protection against the virus. Increasing environmental pressures have led numerous studies to explore the production of biodegradable disposable latex gloves by incorporating natural fillers to improve their decomposition rate [1, 2]. According to Jayathilaka, the global demand for natural rubber latex (NRL) products has significantly increased across various sectors, including medical and personal care. The report indicated that in 2017, the global demand for rubber gloves reached 228 billion pieces, with an anticipated annual increase of 7.95% through 2023. Consequently, the waste generated from NRL-based products is expected to increase as a result of this rising demand [3]. Natural rubber latex is a biopolymer that undergoes vulcanisation to enhance its strength and elasticity. However, this process significantly reduces its degradability, resulting in waste that persists in the environment for extended periods [4]. Therefore, this study investigates the effect of incorporating biofillers derived from cucumber (*Cucumis sativus L.*) into NRL, with a focus on their mechanical properties and biodegradability.

Cucumbers have the potential to contribute to food waste if they exceed their shelf life before consumption. Once spoiled, cucumbers are commonly discarded, which adds to the accumulation of organic waste in landfills. In large-scale agriculture, cucumbers that do not meet market standards for size, shape, or appearance are often discarded as waste. In addition to being underutilised, cucumbers are rich in cellulose. Due to their high cellulose content, cucumbers may serve as an excellent substitute for conventional fillers, such as carbon black, clay, and silica in rubber production. This substitution can also aid in the development of biodegradable latex products with favourable mechanical properties. Cellulose provides structural strength, which can enhance the strength-related properties of NRL films. When used as a biofiller, it improves the stiffness and tensile strength of the rubber matrix, contributing to biodegradable latex products with higher durability [5].

Biodegradable latex products are made from natural latex materials that can break down over time through biological processes. These products are designed to decompose into environmentally harmless substances when exposed to microorganisms such as bacteria, fungi, and other living organisms in soil, water, or composting conditions [6]. Examples of biodegradable latex products include gloves, balloons, condoms, and other latex-based items engineered to minimise environmental impact through degradation. By incorporating cucumber powder (CP) into NRL, the degradation rate may increase compared to NRL incorporated with synthetic fillers, due to the high water content of cucumbers. This moisture content can enhance the overall moisture level within the rubber matrix, thereby accelerating the biodegradation rate of natural rubber. Additionally, cucumbers provide higher oxygen permeability compared to synthetic fillers, facilitating oxidative degradation processes that contribute to the faster bio-deterioration of the rubber matrix.

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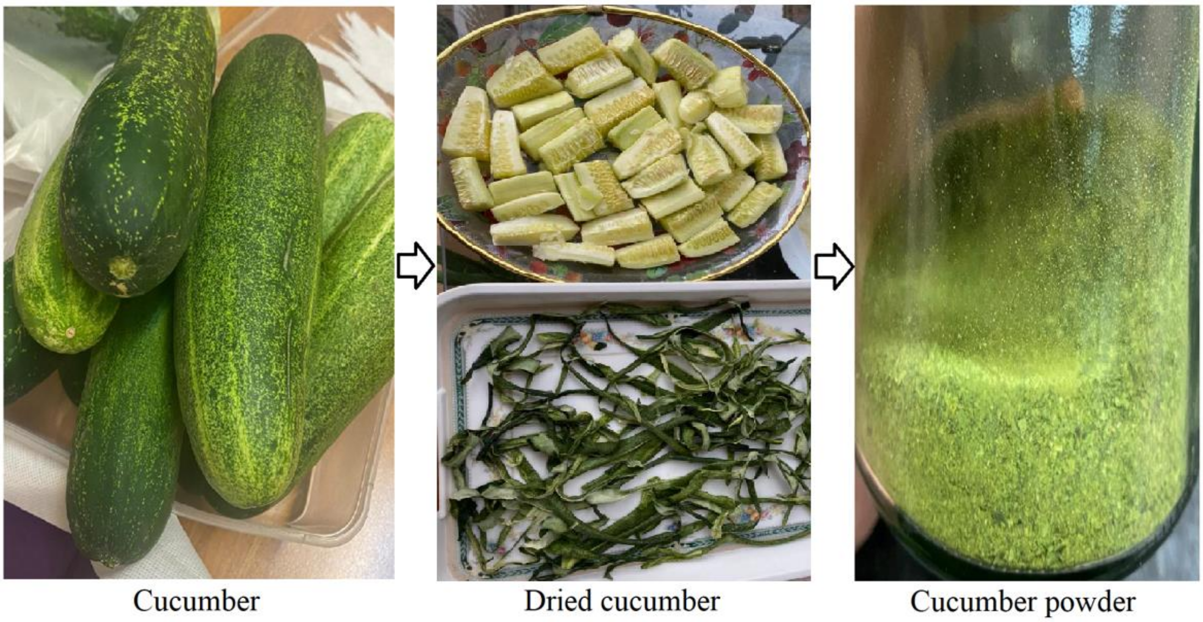

Figure 1. Preparation of CP.

EXPERIMENTAL

Materials

High ammonia NRL with 60 wt.% dry rubber content was purchased from Getahindus Sdn. Bhd. Chemicals and compounding ingredients such as potassium hydroxide (KOH), zinc diethyldithiocarbamate (ZDEC), zinc oxide (ZnO), Vulcastab LW, sulphur, and methyl cellulose were purchased from LGC Scientific Sdn. Bhd., Balakong, Selangor and Sigma-Aldrich (M), Subang Jaya, Selangor.

Cucumbers (*Cucumis sativus L.*) were obtained from a local market in Shah Alam, Selangor. The cleaned cucumbers were sliced and dried at 60 °C in a convection oven for 2 days, followed by grinding in a mixer-grinder to obtain a fine powder, as shown in Figure 1. Approximately 2–3 kg of cucumbers were required to produce 100 g of CP for this study.

Preparation of 50% Dispersion CP

The CP filler was prepared as a 50% dispersion, as shown in Table 1, prior to compounding with NRL.

The preparation began with the dissolution of 1.25 parts of methyl cellulose in a portion of water, with continuous stirring until a homogeneous solution was achieved. Separately, 4 parts of Vulcastab LW and 2 parts of KOH were dissolved in 42.75 parts of water, ensuring thorough mixing until both additives were fully incorporated. Once the additives were dissolved, 50 parts of CP were gradually added to the water phase while stirring continuously to prevent clumping. A high-shear mixer was used to ensure uniform dispersion of the filler. Subsequently, the methyl cellulose solution was slowly introduced into the CP mixture, maintaining constant stirring to achieve homogeneity. The mixture was then stirred for an additional 15–30 min to ensure complete and even distribution of all components. Finally, the prepared dispersion was stored in a sealed container to prevent contamination and sedimentation prior to use.

Table 1. Formulation of 50% dispersion CP.

Compounding Ingredients	Amount (phr)	Purpose
CP	50	Filler
Water	42.75	Solvent
Methyl cellulose (1.25%)	1.25	Dispersing agent
Vulcastab LW (20%)	4	Antioxidant/stabiliser
KOH (10%)	2	pH adjuster

Table 2. Formulation of NRL/CP films.

Components	Dry Weight (phr)
Latex concentrate (60%)	100
KOH (10%)	1.5
ZDEC (50%)	0.7
ZnO (50%)	1.3
Vulcastab LW (20%)	0.2
Sulphur dispersion (50%)	1.5
CP dispersion (50%)	0/2/4/6/8/10

Preparation of NRL/CP Films

Natural rubber latex was compounded according to the previous study conducted by Blanchard et al. [7] with some modifications, as shown in Table 2.

The latex compound was stirred at 500 rpm for 2 h using a magnetic stirrer to ensure the maturation of the formulation, resulting in pre-vulcanisation that facilitated the development of films during the dipping process. A rectangular piece of tempered glass was first dipped in acetic acid, which acted as a coagulant agent, for 10 s and then dried at 65 °C for 20 min in an oven. After drying, the coagulant-coated glass was allowed to cool for 1 min at room temperature before the dipping process. The glass was then dipped in the latex formulation for 40 s to produce latex films. After dipping, the glass was cured at 100 °C for 1 h in the oven. This dipping procedure was repeated for all formulations containing different CP filler loadings (0–10 phr). The cured films were gently stripped from the tempered glass using China clay to prevent sticking. The films were then left to post-cure overnight to ensure they were fully cured before testing.

Particle Size Analysis of CP Fillers

A particle size analyser was used to determine the particle size distribution of CP fillers through laser diffraction. A Malvern Mastersizer 2000 (Malvern Instruments, Worcestershire, UK) was employed to obtain the volume distribution plots of particle size and diameter. The samples were dispersed in a 0.1% Tween solution in water using a Hydro 2000SM cell.

Morphology and Surface Characterisation of CP Fillers

The morphology and surface characterisation of CP fillers were examined using a scanning electron microscope (SUPRA 40VP; Carl Zeiss AG, Oberkochen, Germany) at a constant applied voltage of 10 kV. Prior to electron microscopy, the samples were gold-sputtered using a sputter coater (Type SCD005; Bal-Tec Inc., Balzers, Switzerland) to prevent electrostatic charging during analysis.

Measurement of Rubber-Filler Interaction (Swelling Test)

The interaction between rubber and filler was measured by swelling the cured rubber compounds in toluene in accordance with ASTM D471. Samples of unfilled NRL and NRL/CP films, each weighing about 0.2 g, were cut, and their initial weights were recorded prior to immersion. The samples were then immersed in toluene at room temperature for 48 h. After the immersion period, the samples were removed from the toluene bath, and the swollen samples were gently patted with a tissue to dry the samples before recording their final weights. The swelling index percentage and the rubber-filler interaction were calculated using the following equations:

$$Swelling\ index = \frac{Swollen\ rubber\ weight - Initial\ weight}{Initial\ weight} \times 100\% \qquad (1)$$

The rubber-filler interaction was calculated using the Flory-Rehner equilibrium swelling theory:

$$-[ln\ ln\,(1 - V_r)\ + V_r + \chi {V_r}^2 = V_0[X]\left({V_r}^{\frac{1}{3}} - \frac{V_r}{2}\right) \qquad (2)$$

Where V_r is the volume fraction of polymer in the swollen mass, V_o is the molar volume of the solvent, χ is the rubber-solvent interaction (0.847), and $[X]$ is the crosslink density.

Tensile Test of NRL/CP Films

The tensile test was conducted to measure the resistance to deformation of a material during stretching. The tensile properties of unfilled NRL and NRL/CP films were measured using a tensile tester (Instron 5569; Norwood, USA) with a crosshead speed of 500 mm/min. The samples were prepared by cutting strips of the films using a cutter to obtain dumbbellshaped samples. The thickness of each sample was measured prior to testing in accordance with ASTM D412. The important data obtained were recorded as the mean of five samples, which included the tensile properties of tensile strength, elongation at break, and modulus at 100% elongation.

Biodegradability of NRL/CP Films (Soil Burial Test)

The soil burial test was adapted from Yahya et al. [8] with some modifications. The decomposition of the films was assessed by measuring weight loss and observing changes in surface morphology over a four-week period. Films of unfilled NRL and NRL/CP were buried in a soil mixture consisting of fine sand, old humus, coco peat, and charcoal powder, all contained within a polybag to maintain stable aerobic conditions. The samples were watered every other day to maintain a moisture content of 80%–100%. The soil burial test was conducted in a laboratory under controlled temperatures ranging from 25 to 35 °C. The buried films were removed weekly, cleaned with distilled water, and dried at 40 °C for 2 h before calculating the percentage weight loss. The mechanical changes resulting from weight loss were measured using Equation 3, where W_i and W_f represent the initial weight (g) and final weight (g) of the soil-buried samples, respectively [8].

$$Biodegradation\ (\%) = \frac{W_i - W_f}{W_i} \times 100\% \qquad (3)$$

RESULTS AND DISCUSSION

Characterisation of CP Filler

The particle size distribution of CP filler consists of three peaks, as shown in Figure 2. This phenomenon may arise from the orientation of the filler during light scattering. The average particle size of the CP filler was approximately 40 µm, which corresponds to the dimensions observed in the scanning electron microscopy (SEM) image of the CP filler presented in Figure 3.

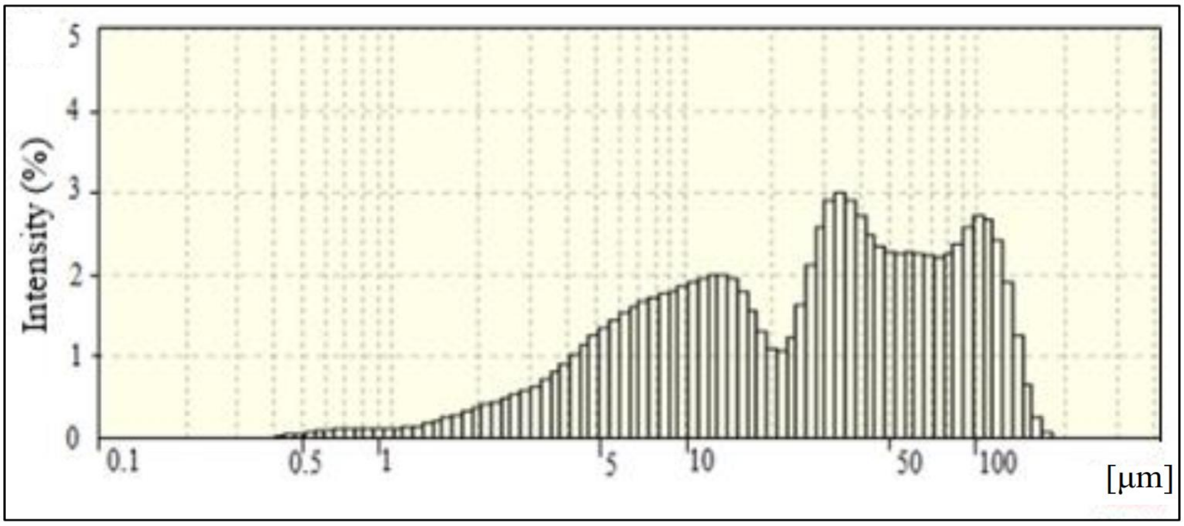

Figure 2. Particle size distribution of CP filler.

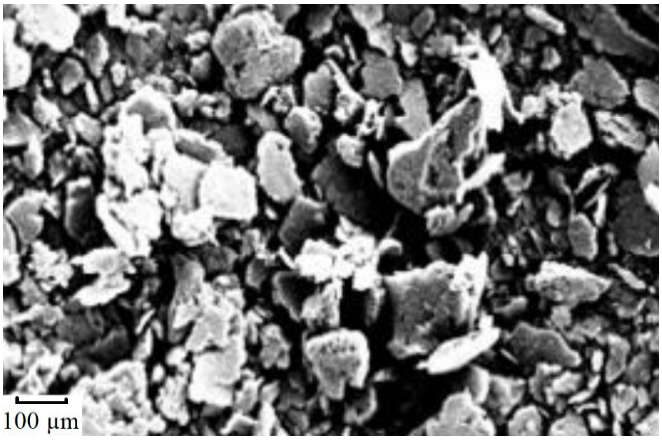

Figure 3. Scanning electron microscopy image of CP filler.

The SEM image of the CP filler in Figure 3 illustrates a morphology of irregular and complex particle shapes. The particles of CP fillers are composed of aggregates, and the irregularities in their shapes can be attributed to the mechanical grinding process, which results in a range of particle sizes. Small particles could be seen at 10 µm, while large particles could be observed at 100 µm, as indicated by the particle size analysis in Figure 2.

A smaller particle size of CP fillers enhances rubber-filler interaction due to the larger specific surface areas, which promote extensive contact area between the rubber and the filler, whereas larger particle size fillers, which consist of aggregates, have a higher tendency to form agglomerates [7, 8].

Swelling Behaviour of NRL/CP Films

The data from the swelling test were used to determine the crosslink density of the films. Figures 4 and 5 portray the swelling index and the crosslink density of unfilled NRL and NRL/CP films, respectively. As anticipated, the swelling index gradually decreased with increasing loading of fillers into the NRL.

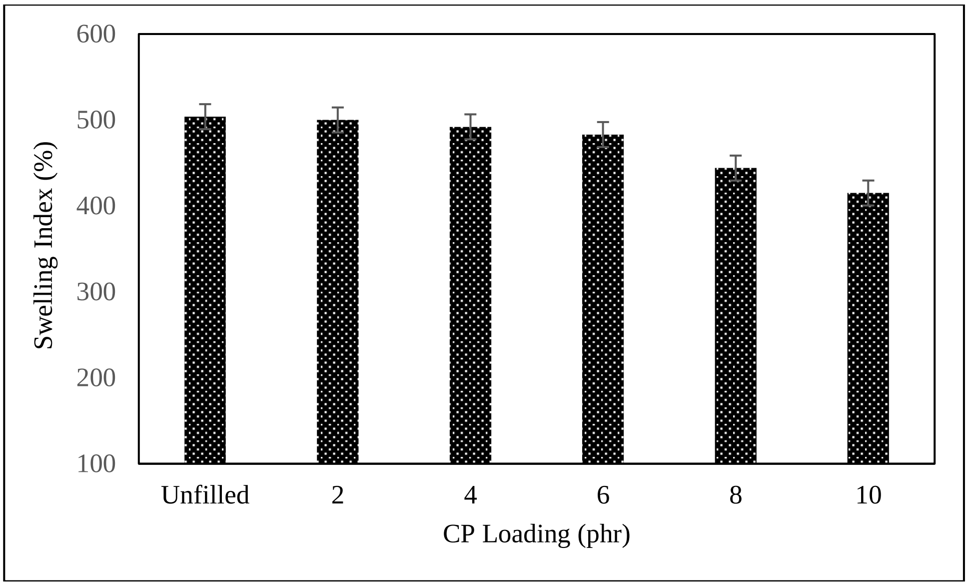

Figure 4. Swelling index of NRL/CP films.

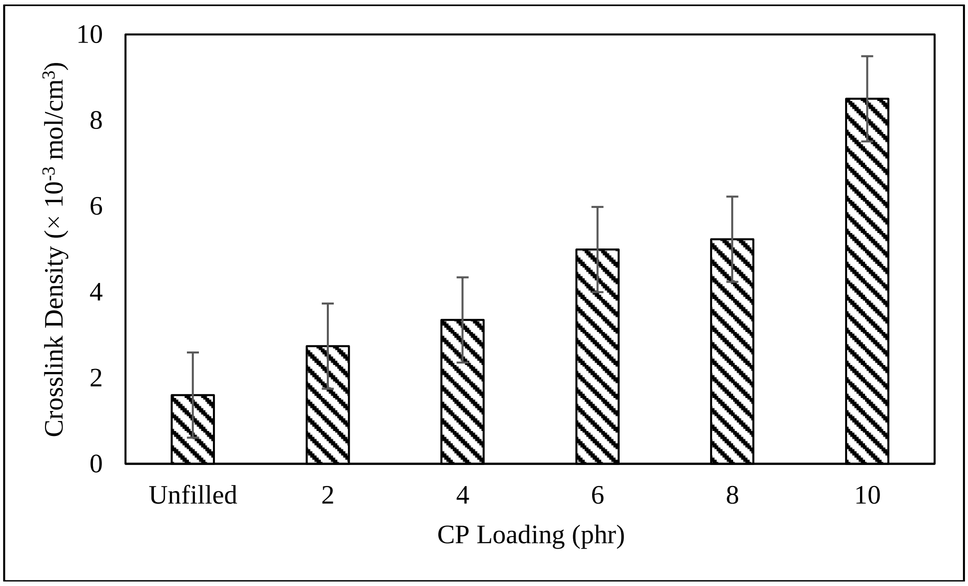

Figure 5. Crosslink density of NRL/CP films.

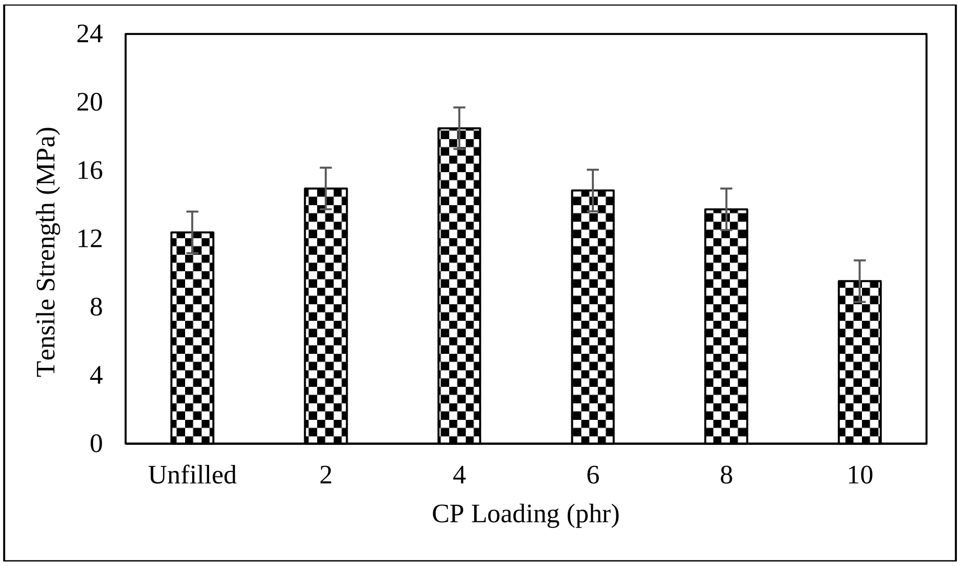

Figure 6. Tensile strength of NRL/CP films.

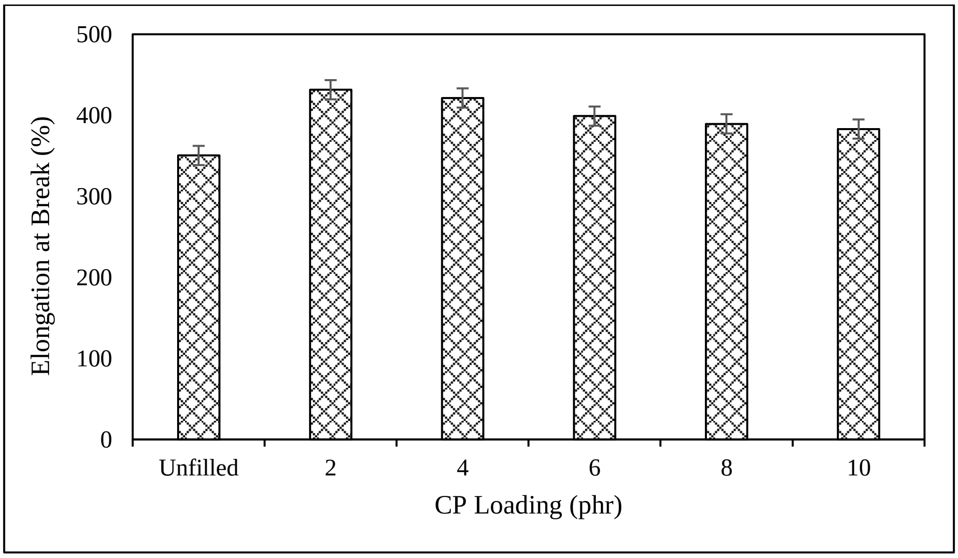

Figure 7. Elongation at break of NRL/CP films.

The declining trend of the swelling index for NRL/CP films is associated with the restriction of solvent to diffuse through the rubber matrix because the fillers occupy the free spaces between the rubber molecules, especially at higher filler loadings [9]. As stated by Sallehuddin et al. [10], fillers generally act as obstacles that inhibit the penetration of toluene. As filler loading increases, more crosslinking occurs between the filler and the rubber matrix, resulting in a decreased penetration of toluene into the rubber matrix. Therefore, the relationship between the swelling index and crosslink density is inversely proportional [10].

Mechanical Properties of NRL/CP Films

Figure 6 shows the effect of different CP filler loadings on the tensile strength of NRL/CP films. The tensile strength of the unfilled NRL film was found to be 12.38 MPa, and the highest tensile strength of 18.48 MPa was exhibited at a CP filler loading of 4 phr. This increase in tensile strength is attributed to the strong interfacial interaction between the filler and the NRL matrix [11]. However, the NRL/CP films exhibited a notable decrease in tensile strength after 4 phr of filler loading.

As the filler loading increases beyond 6 phr, the fillers tend to agglomerate or form clusters due to stronger filler-filler interactions, such as van der Waals forces, which complicate the achievement of uniform dispersion. At higher concentrations, the rubber matrix struggles to distribute the fillers evenly, resulting in poorly dispersed regions with densely packed filler particles. These clusters create weak points in the material where stress is concentrated. When the material is stretched, these agglomerates can lead to premature failure by acting as crack initiation sites, thereby reducing the overall tensile strength [12]. Rather than reinforcing the material, the fillers act as defects or voids, resulting in poor stress distribution and lower tensile strength [13].

Fillers restrict the mobility of polymer chains. While moderate filler loading enhances reinforcement, excessive filler disrupts the polymer network, reducing the ability of the polymer chains to deform elastically. This explains the reduction in elongation at break values, as shown in Figure 7, when the loading of CP filler is increased. The material becomes stiffer and less elastic, leading to a loss of strength as it becomes less capable of distributing and absorbing mechanical stress efficiently [14].

The modulus at 100% elongation (M100) of unfilled NRL and NRL/CP composites at various filler loadings is presented in Figure 8. The highest M100 value of 0.92 MPa was observed for NRL/CP films at 2 phr. This peak in M100 suggests that at this filler concentration, the CP provides slight reinforcement, increasing the material's resistance to deformation at 100% elongation. However, beyond 2 phr, the M100 value decreased, indicating reduced reinforcement and ineffective load transfer within the matrix. At higher filler loadings, CP particles may agglomerate, forming clusters due to poor dispersion and limited compatibility with the rubber matrix. These agglomerates serve as stress concentration points, acting as weak zones during deformation [15, 16]. Consequently, these weak spots lead to premature material failure, resulting in a reduction in elongation at break, as shown in Figure 7.

The tensile properties of NRL filled with CP exhibited distinct trends at varying filler loadings, highlighting the influence of CP on the rubber matrix. The highest tensile strength was observed at 4 phr, indicating that this filler loading optimally reinforces the rubber matrix by enhancing stress transfer and crosslinking efficiency. However, at higher filler loadings, the tensile strength decreased, likely due to filler agglomeration, which creates stress concentration points and disrupts the uniform distribution of stress. Conversely, the highest elongation at break and modulus at 100% strain were observed at 2 phr. This suggests that at lower filler concentrations, CP enhances the elasticity and stiffness of the NRL matrix by maintaining polymer chain mobility and providing effective reinforcement at low strains [14]. As the filler content increases beyond 2 phr, the rubber matrix becomes more rigid, resulting in reduced flexibility and elongation at break. These findings highlight a trade-off between flexibility and strength, emphasising the importance of optimising filler loading to achieve the desired mechanical properties.

Biodegradation of NRL/CP Films

The biodegradability of a polymer can be qualitatively assessed by measuring the weight loss of the polymer after being buried for a specific duration [17]. Figure 9 shows the weight loss percentage (%) of unfilled NRL and NRL/CP films containing 4 and 10 phr of CP filler, recorded weekly over 4 weeks of soil exposure.

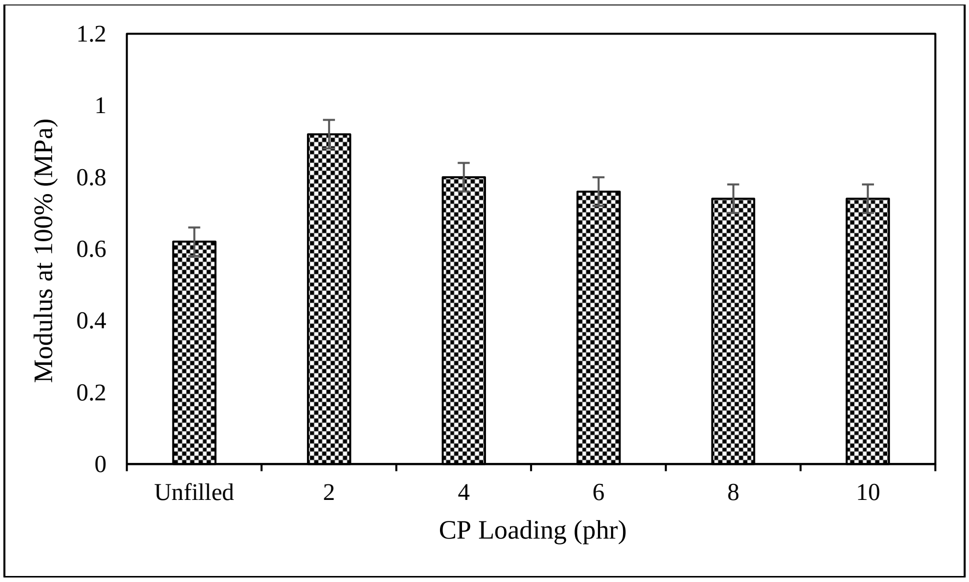

Figure 8. Modulus at 100% of NRL/CP films.

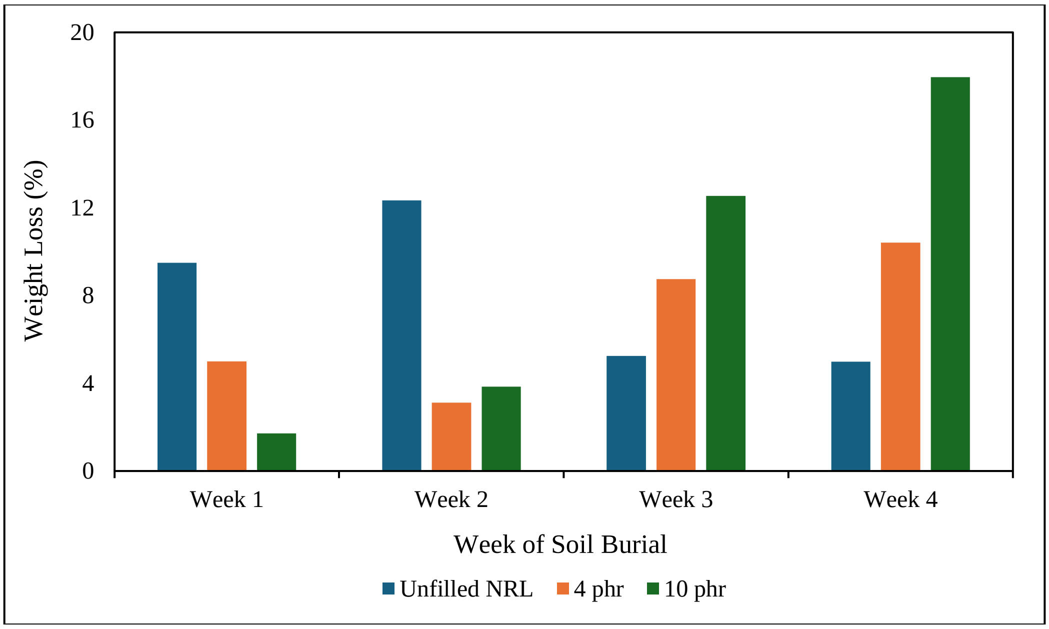

Figure 9. Weight loss percentage of NRL/CP.

In weeks 1 and 2, the unfilled NRL film exhibited a higher initial weight loss compared to the NRL/CP films, as it is more susceptible to microbial degradation and hydrolytic processes. The natural rubber matrix is more exposed to soil microorganisms and moisture, without any protective barriers or additional materials. Cucumber powder, being an organic filler, may initially slow the degradation process. The filler can absorb some moisture, thereby limiting the immediate exposure of the rubber matrix to soil microorganisms [17]. Additionally, the filler may alter the microenvironment in the soil, providing initial protection to the NRL matrix against microbial activity in weeks 1 and 2.

As the test progresses into the 3rd and 4th weeks, the dynamics of degradation change. The CP filler may begin to degrade or interact with soil microorganisms in a manner that accelerates the overall degradation of the rubber matrix. As shown in Figure 9, the NRL/CP films with 4 and 10 phr of CP filler show higher weight loss percentages compared to unfilled NRL films. The CP filler could decompose and contribute to the breakdown of the rubber or create conditions that promote microbial activity. Over time, soil microorganisms may adapt to break down the filler material more efficiently, thereby enhancing their ability to attack the NRL matrix [18]. Additionally, the filler may provide additional nutrients or create a more favourable environment for microbial growth, resulting in accelerated degradation. This phenomenon explains the highest weight loss percentage observed in NRL/CP films with the highest CP filler loading.

CONCLUSION

The incorporation of CP into NRL has enhanced the strength-related properties of the films compared to unfilled NRL films. The highest tensile strength was recorded at a CP loading of 4 phr, which correlates with an increase in crosslink density. As the CP loading increases, the crosslink density also increases, contributing to the improved mechanical properties. The highest elongation at break and modulus at 100% strain were recorded at 2 phr, indicating that lower CP levels improve elasticity and stiffness without making the rubber too rigid. As the filler content increases, the material becomes stiffer and less flexible, reducing its ability to stretch. These findings suggest that CP effectively reinforces NRL, with 2 phr enhancing flexibility and 4 phr maximizing strength, depending on the application needs. In the soil burial test, unfilled NRL initially experienced a higher weight loss during the 1st and 2nd weeks due to more direct exposure to degradation, whereas NRL/CP films showed a lower initial weight loss. This is attributed to the protective or limiting effect of the CP filler. However, as time progressed, the degradation of CP itself, along with changes in microbial activity, led to increased weight loss percentages for NRL films containing 4 and 10 phr of CP filler. The higher CP content accelerated degradation by interacting with both the soil and the rubber matrix. In conclusion, CP has the potential to be developed as a biofiller in NRL films for the production of biodegradable latex products.

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