Sains Malaysiana 54(2)(2025): 535-545
http://doi.org/10.17576/jsm-2025-5402-18

Valorization of Chemically-Treated Recycled Carbon Black as a Filler in Biodegradable Cellulose-Based Mulching Films

(Valorisasi Karbon Hitam Kitar Semula Dirawat Secara Kimia sebagai Pengisi dalam Filem Sungkupan Berasaskan Selulosa Terbiodegradasi)

Nur Alia Sahira Azmi[1], Siew Xian Chin[2,3], Sarani Zakaria[1,3], Junfei Tian[4] & Chin Hua Chia[1,3,*]

[1]Materials Science Program, Department of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia
[2]ASASIpintar Program, Pusat GENIUS@Pintar Negara, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia
[3]Quantum Materials and Technology Research Group, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia
[4]State Key Laboratory of Pulp and Paper Engineering, School of Light Industry and Engineering, South China University of Technology, Guangzhou 510640, China

Received: 31 August 2024/Accepted: 12 November 2024

ABSTRACT

Researchers are exploring ways to incorporate carbon-based fillers like recycled carbon black (rCB) into cellulose films for use as biodegradable mulching films in agriculture. Adding dark fillers can increase opacity to control light exposure and moisture for optimizing crop environments. This study aimed to create an eco-friendly black mulching film by mixing treated rCB into a regenerated cellulose matrix derived from cotton linters. The rCB was chemically treated to modify its properties. Cellulose films were made with 10%, 25%, and 50% treated rCB mixed into a cotton linter cellulose solution. The films were characterized for properties like morphology, transparency, thermal stability, wettability, water vapor permeability, tensile strength, and soil biodegradation. Compared to untreated rCB, the treated filler improved the films' cross-section, surface area, and porosity. Higher rCB increased opacity but decreased tensile strength. Adding 10% treated rCB optimized the decomposition rate. Increasing to 50% progressively slowed decomposition. The rCB made the films more hydrophilic, and 10% treated rCB gave the best water vapor transmission performance. While rCB did not impact overall biodegradability, the 10% treated rCB film degraded fastest in soil once surface deterioration began. In summary, incorporating 10% chemically treated recycled carbon black into cellulose films derived from cotton linters produced an optimized eco-friendly black biodegradable mulching film material.

Keywords: Cellulose film; degradation; hydrophilicity; porosity; rCB; transparency

ABSTRAK

Para penyelidik sedang mengkaji cara untuk memasukkan pengisi berasaskan karbon seperti karbon hitam (rCB) kitar semula ke dalam filem selulosa untuk digunakan sebagai filem sungkupan biodegradasi dalam pertanian. Menambah pengisi gelap boleh meningkatkan kelegapan untuk mengawal pendedahan cahaya dan kelembapan untuk mengoptimumkan persekitaran tanaman. Penyelidikan ini bertujuan untuk mencipta filem sungkupan hitam mesra alam dengan mencampurkan rCB terawat ke dalam matriks selulosa yang dijana semula yang diperoleh daripada linter kapas. rCB telah dirawat secara kimia untuk mengubah suai sifatnya. Filem selulosa dihasilkan dengan 10%, 25% dan 50% rCB terawat yang dicampurkan ke dalam larutan selulosa linter kapas. Filem tersebut dicirikan untuk sifat seperti morfologi, ketelusan, kestabilan terma, kebolehbasahan, kebolehtelapan wap air, kekuatan tegangan dan biodegradasi tanah. Berbanding dengan rCB yang tidak dirawat, pengisi yang dirawat meningkatkan keratan rentas, luas permukaan dan keliangan filem. rCB yang lebih tinggi meningkatkan kelegapan tetapi mengurangkan kekuatan tegangan. Penambahan 10% rCB yang dirawat mengoptimumkan kadar penguraian. Peningkatan secara progresif sebanyak 50% dalam memperlahankan penguraian. rCB menjadikan filem lebih hidrofilik dan 10% rCB yang dirawat memberikan prestasi penghantaran wap air yang terbaik. Walaupun rCB tidak memberi kesan kepada kebolehbiodegradan keseluruhan, filem rCB terawat 10% terdegradasi paling cepat dalam tanah sebaik sahaja kemerosotan permukaan bermula. Ringkasnya, menggabungkan 10% karbon hitam kitar semula yang dirawat secara kimia ke dalam filem selulosa yang diperoleh daripada linter kapas menghasilkan bahan filem sungkupan biodegradasi hitam mesra alam yang dioptimumkan.

Kata kunci: Filem selulosa; hidrofilik; keliangan; kemerosotan; ketelusan; rCB

INTRODUCTION

Mulching involves distributing layers of materials to the planting site before, during, or immediately after sowing to support and spread the soil surface. Mulching is a water-efficient practice used in agriculture that enhances water infiltration into the soil, slows soil erosion, and minimizes surface runoff (Kader et al. 2017). Mulching is an effective strategy of modifying the crop-growing environment to boost crop production as well as enhance the quality of the goods by controlling soil temperature, preserving soil moisture, and lowering soil evaporation. Numerous mulching materials have recently been used in agriculture, including synthetic films made of petroleum-based materials (Kasirajan & Ngouajio 2012) and biodegradable polymer films consisting of PLA and cellulose (Yang et al. 2020). Recent research has concentrated on generating mulch from biodegradable polymer due to the material's renewability, biocompatibility, and biodegradability in order to avoid trash disposal and environmental problems (Yang et al. 2020). The use of synthetic mulching films, such as those made from polyethylene, in agricultural markets poses a disposal problem due to their limited biodegradability and slow degradation rates. Fortunately, biodegradable polymers may have limits in terms of processing range, brittleness, moisture absorption, gas barrier, and heat/mechanical resistance.

Cellulose is an unbranched homopolysaccharide, formed by two anhydro-glucose rings ($(C6H10O5)_n$) of β-D-glucopyranoside, linked by β-(1→4)-glycosidic bonds. Cellulose has a partially crystalline structure, formed by a strong network of intermolecular and intramolecular hydrogen bonds, resulting in an insoluble material in water or other common solvents (Geng et al. 2014), which are important properties that make cellulose a very interesting raw material to develop new biomaterials, and that differs from the rest of the polysaccharides. Cotton linters are some abundant resources that can serve as alternative materials in producing biodegradable mulching films.

Several studies acknowledged that coloring mulch contributed to soil systems and processes, including nutrient uptake, water absorption, root growth, and the existence of soil-based microorganisms that depend on soil temperature (Amare & Desta 2021). Despite the accessibility and widespread use of different types of colored mulch, black plastic mulch is the most common and widely used. Black mulch increased soil heat by absorbing a large quantity of sunlight since it can absorb ultraviolet (UV), visible, and infrared wavelengths of solar radiation released by the sun (Haapala et al. 2015). The colour parameters of an agricultural mulch film together with the optical properties are directly related to its radiometric properties and will influence the temperature of the surface and the microclimate around the vegetable plants (Tarara 2000). This is because parameters, such as reflectivity, absorption capacity or transmittance, depend on incoming solar radiation (Lamont 2017).

Several studies have begun to use carbon in the manufacture of mulching due to its qualities in lowering bulk density, enhancing porosity, aggregation, water infiltration, and soil water holding capacity (Harada et al. 2019). Additionally, it darkens the color of the film, which has an impact on soil surface reflectance and helps to control soil temperature. Enhancing cation exchange capacity, raising soil pH, boosting nutrient supply and absorption, reducing nutrient leaching losses, capturing NH_3, and reducing nitrogen volatilization loss all contribute to nutrient availability in soils as a result of the addition of carbon (Fida Banu et al. 2023).

Carbon black is produced by incomplete combustion of hydrocarbons that are found in petroleum oil or natural gas (Galli 1982). Carbon black has gained attention for incorporating in film due to its ability to enhance polymer properties such as thermal, mechanical, and gas barrier also both safe material for animals and human and stability under harsh condition processes (Harada et al. 2016). Recent research sustainability strategies that focus on using renewable materials have started using carbon black from recycling sources from recovery pyrolysis activity of used waste tyres. This offers an effective method to mitigate pollution problems while also producing valuable alternative materials. However, rCB often suffers from quality issues like low surface area and active pore sites due to high sulphur and heavy metal. This has encouraged research interest in improving rCB properties technically through chemical treatment method before incorporating into films (Azmi et al. 2023; Dong et al. 2017). In this study, cellulose films with different composition percentages of chemically treated rCB that set at 10%, 25%, and 50% were prepared and the properties were determined in terms of morphology, thermogravimetric analysis, contact angle, water vapor permeability, tensile strength, and biodegradability.

MATERIALS AND METHODS

The pyrolytic recycled carbon black (rCB) was provided by Sun Rubber Industry Sdn. Bhd. The treatment rCB that chemically treated with toluene and acid as reported by previous study (Azmi et al. 2023). Cotton linter was provided by Hubei Chemical Fiber (Xiangfan, China). Sodium hydroxide (NaOH) (>99%, EMSURE® ACS), and urea (99%, ACS) were purchased from Merck Millipore (Burlington, MA, USA). Sulfuric acid (H_2SO_4) (95-98% ACS) was purchased from Thermo Fisher scientific (USA). Sodium Chloride (EMSURE® ACS) was purchased from Merck Millipore (Burlington, MA, USA).

PREPARATION OF CELLULOSE FILM LOADED WITH rCB TREATED

The preparation began with dissolving 7 g of NaOH, 12 g of urea, and 81 g of H_2O, followed by freezing the solution until it reached -20 °C. Next, 3.5 g of cotton linters were added to the solution when its temperature reached -13 °C, and the mixture was stirred at high speed. The solution was then centrifuged at 12,000 rpm for 10 min at 5 °C to produce a control cellulose solution (CT). Different composition percentages (10%, 25%, and 50%) of the treated rCB powder were subsequently incorporated into the cellulose solution and stirred to obtain a uniform mixture. The mixture was poured onto a thin glass plate and coagulated in a 5% H_2SO_4 solution to form regenerated cellulose films. Cellulose films with the different compositions treated rCB (rCB-T), (10%, 25%, and 50%), which have been denoted as CT-rCB-T (10%), CT-rCB-T (25%), and CT-rCB-T (50%). Cellulose film with commercial CB N330 was also produced and used as control sample (CT-N330 (25%)). The resulting films were thoroughly rinsed with distilled water to remove chemical residues until neutralization was attained. Finally, the rinsed film was dried in an oven on a PMMA sheet, preparing it for further characterization processes.

CHARACTERIZATION OF SAMPLES

The physical and chemical properties of films study were analyzed using various instruments. The morphology of sample was analyzed using a field emission scanning electron microscope (FESEM, FEI QUANTA 400F). The analysis considers the cross-sectional and surface image of the film measured at 500× magnification. The transparency analysis of film was analyzed using a UV-Vis spectrophotometer which measured the absorption and transmission of light in 300-800 nm. The thermogravimetric analysis (TGA) and derivative thermogravimetric analysis (DTG) were conducted using thermogravimetric analyzer (NETZSCH, STA 449 F3 Jupiter). Samples weighing 2 mg were analyzed under a nitrogen atmosphere with gas flow rate of 50 mL/min, a heating rate of 10 °C/min, and a temperature range from 25 to 800 °C. Contact angle analysis of the film was analyzed using Lauda Scientific, LSA-100 microscopy and lab-made software. Tensile tests were evaluated for each film using a universal testing machine (Gotech Al3000). Tensile strength (TS), tensile modulus, and elongation at break data were analyzed for each sample. Rectangular film specimens (10 mm × 60 mm) were randomly cut from the original film, following the ASTM D882 standard. The water vapor permeability analysis methodology used in this analysis followed the ASTM E96 standard 'desiccants cup' method to determine the rate of water vapor permeability of the celluloses films. The film was cut to an approximate size of 30 mm × 30 mm. Glass vials containing silica gel were covered with the film specimens and the vials were placed in a desiccator filled with a saturated NaCl solution to achieve relative humidity 75%. The bottles covered with containing silica gel were weighed every day and weight changes were recorded until the sample weight was stabilized. The water vapor transmission rate (WVTR) was calculated using Equation (1).

$$WVTR = ((w_f - w_i) / day \times A) \times L \qquad (1)$$

where w_i is the initial bottle weight and w_f is the bottle weight after testing each day. A is the effective film area in m^2 and L is the film thickness for normalization. Weight differences within each day were plotted against the number of days.

Lastly, soil degradation analysis was performed in this experiment to determine the biodegradability of film materials by microorganisms. Specimens with an estimated size of 40 mm × 40 mm were cut, weighed, and placed on the surface net before being buried in pot depth of 3 cm, with an additional 5 cm depth of soil added on top. Watering was done to regulate the moisture content and temperature of the area, simulating a natural environment. The samples were taken out within a 60-day period. The mass of samples was measured after 30 days, and the process was repeated for another 30 days. The state of biodegradability in terms of appearance was evaluated visually.

RESULTS AND DISCUSSION

MORPHOLOGICAL ANALYSIS

The cross-sectional and surface morphology of the cellulose films were analyzed and shown in Figures 1 and 2, respectively, at 500× magnification. These images compare the effects of adding different percentages of treated rCB to the cellulose films. As shown in Figure 1(a), the neat cellulose film exhibits a smooth and homogeneous cross-sectional structure. However, the addition of different percentages of rCB into the cellulose shown in Figure (1(b), 1(c) and 1(d)) results in varying degrees of cross-sectional compatibility. The composition of cellulose with treated-rCB displays relatively rougher surfaces with obvious porous carbon particles observed across the film. It can be observed that rCB particles start forming agglomeration at 25% as shown in Figure 1(c). The agglomeration of carbon present in the film has also been observed in previous study (Harada et al. 2016). This result can be described as in complete dispersion of a fraction of the carbon particulate. Furthermore, increasing the percentage composition of treated rCB introduces higher porous carbon particles content into the film. Based on Figure 2(a), neat cellulose film shows smooth surface despite there is some undissolved cellulose fibers observed. Cellulose films containing treated rCB show more visible rCB particles as increase of rCB treated content (Figure 2(b)-2(d)).

TRANSPARENCY

The transparency results of all samples are presented in Figure 3. Based on the figure, the neat cellulose film transmits light up to 80%. The addition of treated rCB significantly reduces light transmission, decreasing

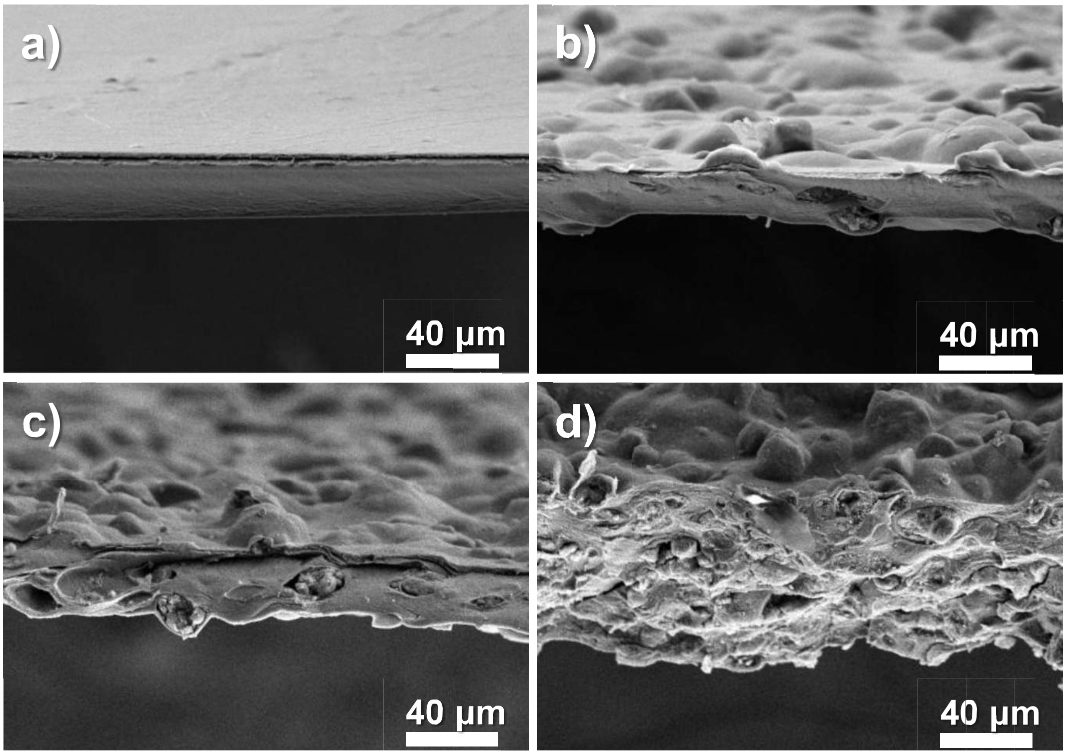

FIGURE 1. a) SEM cross-sectional images of the regenerated cellulose film samples (a) CT, (b) CT-T-rCB (10%), (c) CT-T-rCB (25%), and (d) CT-T-rCB (50%)

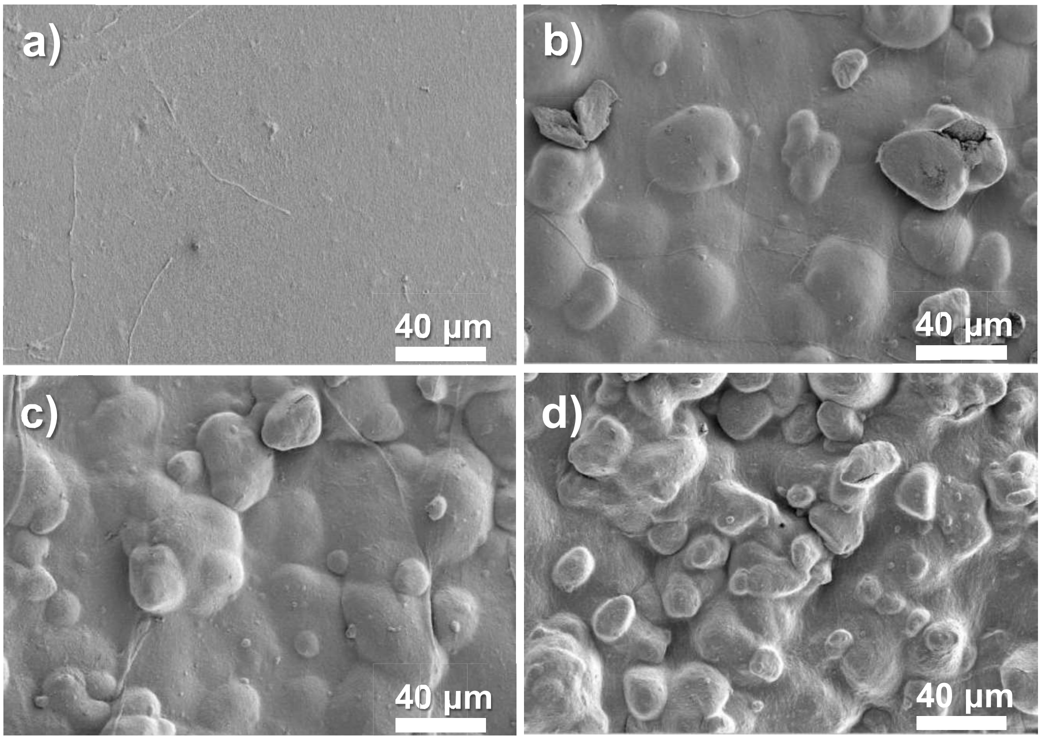

FIGURE 2. SEM surface image of the regenerated cellulose film samples: a) CT, b) CT-T-rCB (10%), c) CT-T-rCB (25%), and d) CT-T-rCB (50%)

from approximately 80% transmission to 15% with the incorporation of 10% treated rCB. The addition of 25% and 50% treated rCB results in a complete 100% opacity of the cellulose films. Higher loading percentages of treated rCB correspondingly decrease the optical transparency of the films. This phenomenon can be attributed to the dark color of the treated rCB and the decrease in crystallinity of the cellulose film (Liu & Zhang 2009). The presence of rCB has disturb the formation of hydrogen bonds between the cellulose chains to form crystalline areas during the regeneration process in the coagulation bath. The European standard EN 13655, which addresses mulch films used for weed control, specifies that black mulch films must have an emissivity value less than 1%. Additionally, as reported by Briassoulis and Giannoulis (2018), the measured coefficients should meet the standards set by EN 17033-18 (for all wavelengths), with the relative light transmittance required to be less than 3% for black and opaque mulch films. Therefore, CT-T-rCB (25%) and CT-T-rCB (50%) samples have met the standard requirement.

THERMOGRAVIMETRIC ANALYSIS

As can be seen from the thermogravimetric curves shown in Figure 4, the thermal decomposition investigated from 25 °C to 800 °C generally involved two distinct weight loss stages, corresponding to slow and fast decomposition processes. The initial slow weight loss at the early stage is associated with the evaporation of water. Although the mass of cellulose remained almost unchanged during this stage, a continuous weight loss was observed with increasing temperature due to changes in intermolecular forces and hydrogen bonding after the regeneration process (Yeng, Wahit & Othman 2015). The subsequent fast weight loss stage, occurring between 250 °C and 350 °C, corresponds to further dehydration and decomposition of the cellulose. During this fast pyrolysis stage, significant weight loss occurs due to the onset of oxidation processes, which begin at lower temperatures (270 °C and 305 °C). Regenerated cellulose almost burns during this stage due to oxidative thermal degradation and the destructive breakdown of its crystallite structure.

Table 1 shows that the original cellulose film exhibited the greatest decomposition compared to the films containing treated rCB. When comparing the addition of rCB, the cellulose film with 10% treated rCB decomposed to a higher extent than the film with untreated rCB, as evidenced by the higher char yield product during the pyrolysis stage. The incorporation of 10% rCB-treated into the cellulose film increased the stability temperature compared to the original cellulose film, as shown in Figure 4(b). This increased stability is expected to be due to the presence of leftover inorganic elements (carbonaceous material) and ash in the cellulose film, leading to a reduction in the rate of mass loss during the decomposition process due to the heat insulating characteristics of the residues (Puri, Hu & Naterer 2024). Furthermore, the percentage composition of rCB treated influenced the mass change and stability temperature of the cellulose films. As the percentage of rCB-treated in the cellulose film increased up to 50%, the decomposition process occurred at a slower rate, resulting in a slower mass loss. Higher percentages of rCB-treated may decompose more slowly under similar thermal conditions.

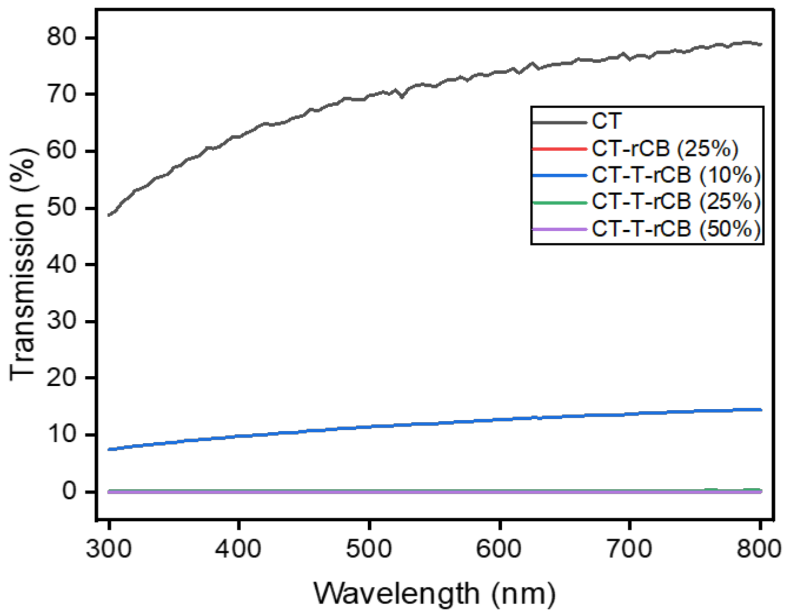

FIGURE 3. UV-vis spectra of the regenerated cellulose film samples CT, CT-rCB (25%), CT-T-rCB (10%), CT-T-rCB (25%), and CT-T-rCB (50%)

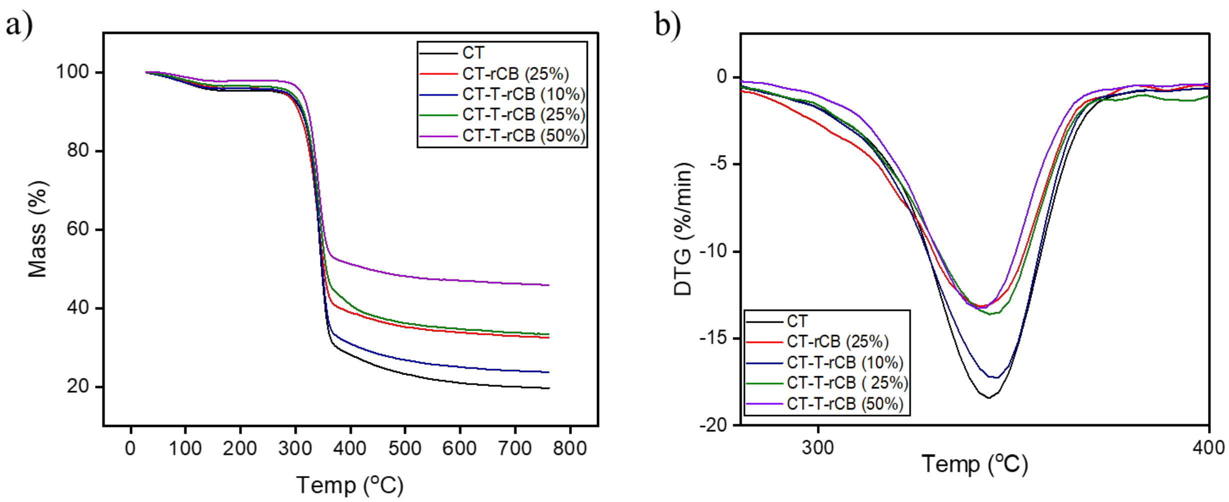

FIGURE 4. a) TGA analysis of the regenerated cellulose film samples and 4 b) DTG analysis of the regenerated cellulose film samples of CT, CT-rCB (25%), CT-T-rCB (10%), CT-T-rCB (25%), and CT-T-rCB (50%)

TABLE 1. Mass change and temperature change observed after decomposition process of the regenerated cellulose films of CT, CT-rCB (25%), CT-T-rCB (10%), CT-T-rCB (25%) and CT-T-rCB (50%)

Sample	Thermogravimetric analysis	Derivative thermogravimetric analysis
	Mass change (%)	Temperature (°C)
CT	75.6%	343.6 °C
CT-rCB 25%	63.2%	341.8 °C
CT-T-rCB 10%	72.1%	345.0 °C
CT-T-rCB 25%	63.1%	344.4 °C
CT-T-rCB 50%	52.0%	341.3 °C

TENSILE ANALYSIS

The tensile strength, elongation at break, and modulus of the cellulose film with added rCB are illustrated in Figure 5(a), 5(b), and 5(c), respectively. Generally, the original cellulose film exhibited the highest tensile strength of up to 45.50 MPa. However, the addition of rCB has significantly reduced the tensile strength of the films. This can be explained by the disruption of the formation of hydrogen bond between the cellulose chains by the rCB particles during the regeneration process of the cellulose solution (Qiu & Netravali 2012). In addition, the agglomeration of the rCB particles at higher loading (25% and 50%) has seriously impacted the strength of the films.

WATER VAPOR PERMEABILITY

Water vapor transmission rate (WVTR) of mulching films play important role in maintaining moisture content of soil (Ning et al. 2021). As shown in Figure 6, the neat cellulose film exhibited the lowest WVTR, indicating its superior moisture retention ability. Incorporating untreated rCB into the film increased the WTVR, reducing its moisture retention capacity. When varying the composition percentage of treated rCB added to the film, increasing the treated rCB up to 50% resulted in an increase in the WVTR, indicating lower moisture retention capacity. However, the addition of 10% treated-rCB to the film shows the optimum WVTR, being the sufficient composition to achieve the desired moisture retention capability as compared to other compositions. This can be attributed to the increasing of porosity of the cellulose film which influences the water vapor permeability performance of the cellulose film (Bumrungnok et al. 2023). These results are consistent with morphological analysis presented in Figure 1. As shown in Table 2, the water vapor transmission rates (WVTR) of different types of polysaccharide films, including regenerated cellulose and starch, were compared. The results suggest that the regenerated films produced in this study had WVTR values comparable to some of the films reported in other studies.

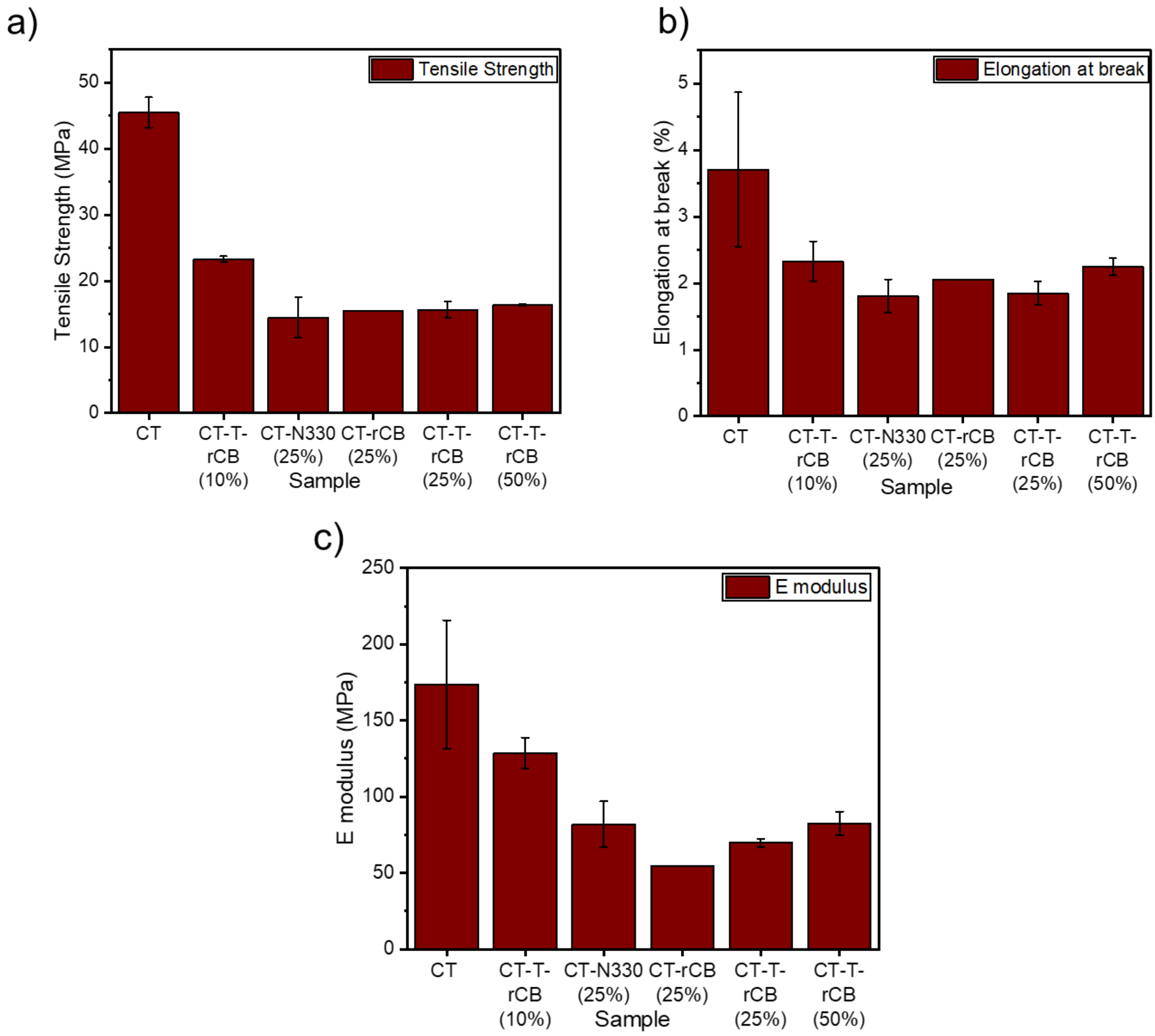

FIGURE 5. a) Tensile strength, b) elongation at break and c) elongation modulus of the regenerated cellulose films of CT, CT-rCB (25%), CT-T-rCB (10%), CT-T-rCB T (25%), and CT-T-rCB (50%)

TABLE 2. Water vapour transmission rate (WVTR) of polysaccharide-based films reported in previous studies

Material	Testing condition	WVTR (g.mm/m^2.d)	Reference
Regenerated cellulose	23 °C	3.995	Wang et al. (2018)
Cellulose/chitosan film	38 °C, 90% RH	0.00752	Ning et al. (2021)
Pulp/NMMO	25 °C, 85% RH	0.00165	Zhang et al. (2023)
Regenerated cellulose	25 °C	0.0112	Amalini et al. (2019)
Pineapple stem starch film	75% RH	45	Bumrungnok et al. (2023)
Potato starch film	25 °C, 90% RH	878	Zhang, Wang & Cheng (2018)

CONTACT ANGLE

Figure 7 illustrates the result of contact angle analysis for cellulose films with incorporation untreated-rCB, and rCB-treated with their respective composition percentages. The observed values range from 62.0° to 30.5°. The neat cellulose film exhibits the highest contact angle value, which can be attributed to smoother and lower roughness surface of the film than that of rCB. A significant reduction of contact angle value is observed as incorporating untreated rCB into the cellulose film, this difference might be due to variations in impurities of untreated rCB. Detailed analysis of varying higher percentage compositions of treated rCB incorporated into the film shows a decreasing trend until the addition of rCB treated up to 25% shows lower contact angle value as compared with untreated rCB. However, additional rCB treated up to 50% show insignificant increase while still within the range of other treated rCB. The decreasing contact angle value of the films with treated rCB can be attributed to the hydrophilic nature of the treated rCB due to the oxidation of treated rCB with (Azmi et al. 2023).

SOIL DEGRADATION ANALYSIS

Figure 8 shows the activity progress of regenerated cellulose films before the soil degradation period, after 30 days, and after 60 days. Based on the figure, the surface of the original cellulose film turns darker and develops small defects on the edges after 60 days. When comparing the cellulose films with treated rCB and untreated rCB, the physical matrix begins to appear broken and defective on the film surfaces. The degradation of the films can be attributed to microorganism activity in the soil (Zhang et al. 2023). In terms of different percentages of treated rCB in the films, the higher addition of rCB-treated up to 50% does not influence the degradation rate based on the observation of defects and the broken physical matrix film character. However, weight measurements could not be significantly evaluated due to lumps of soil sticking to the film surfaces, preventing accurate weight measurements.

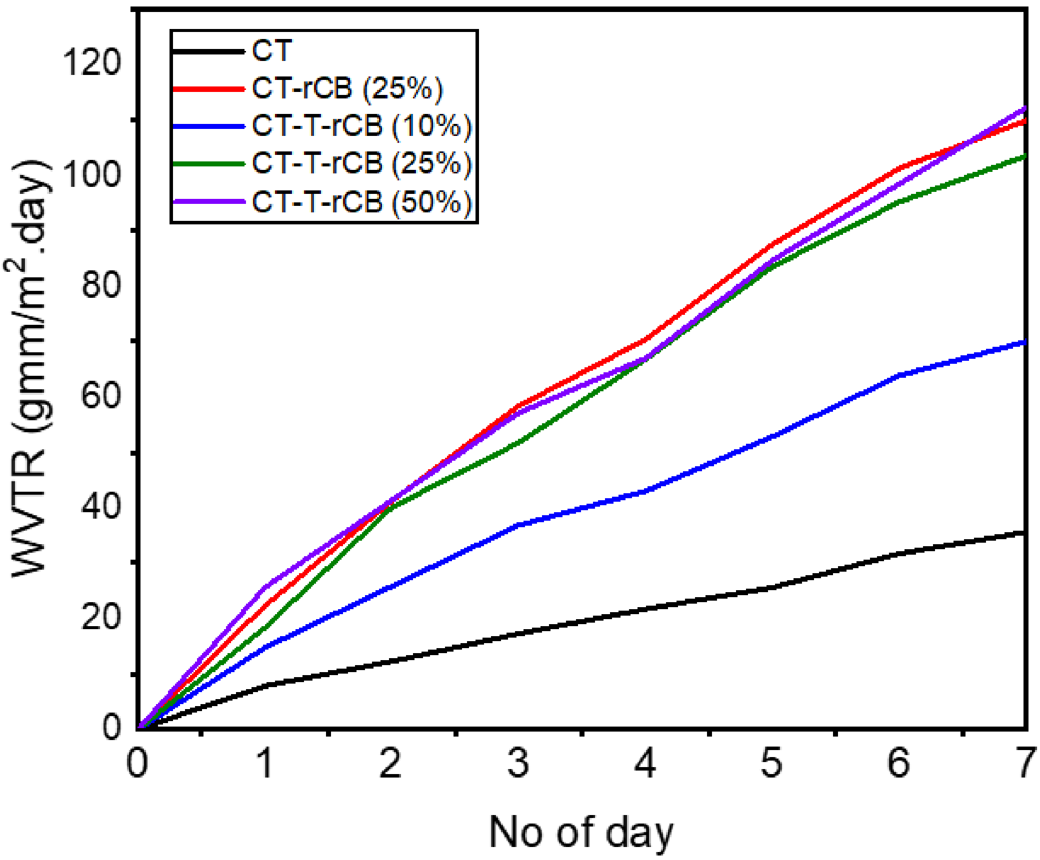

FIGURE 6. Water vapor transmission rate (WVTR) of the regenerated cellulose films of CT, CT-rCB (25%), CT-T-rCB (10%), CT-T-rCB (25%), and CT-T-rCB (50%) after 1 week

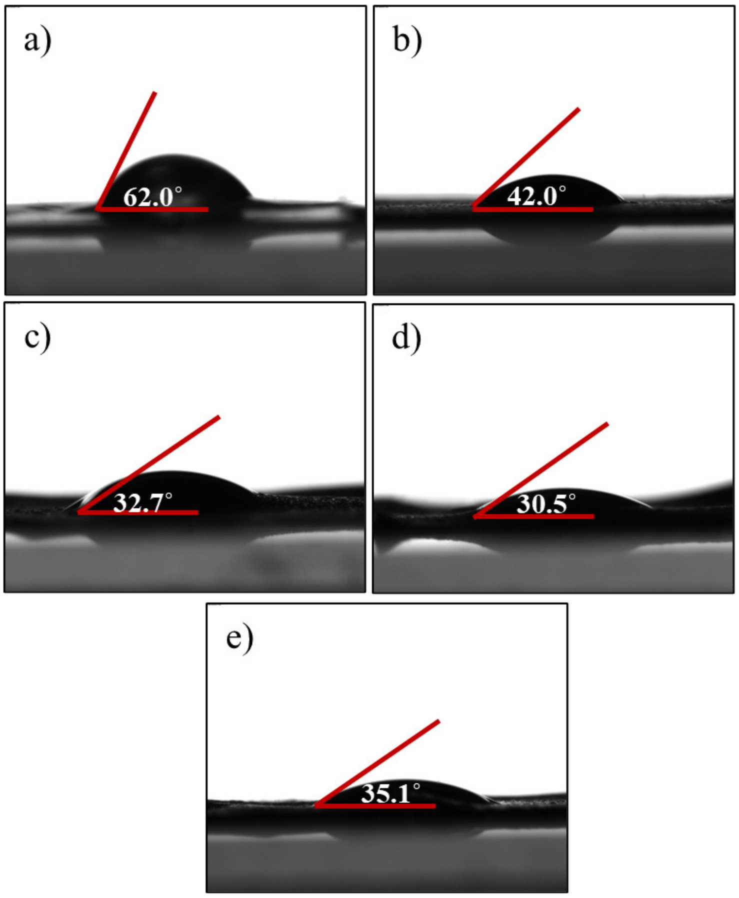

FIGURE 7. Water contact angle on surface films of the regenerated cellulose of a) CT, b) CT-rCB (25%), c) CT-T-rCB (10%), d) CT-T-rCB (25%), and e) CT-T-rCB (50%)

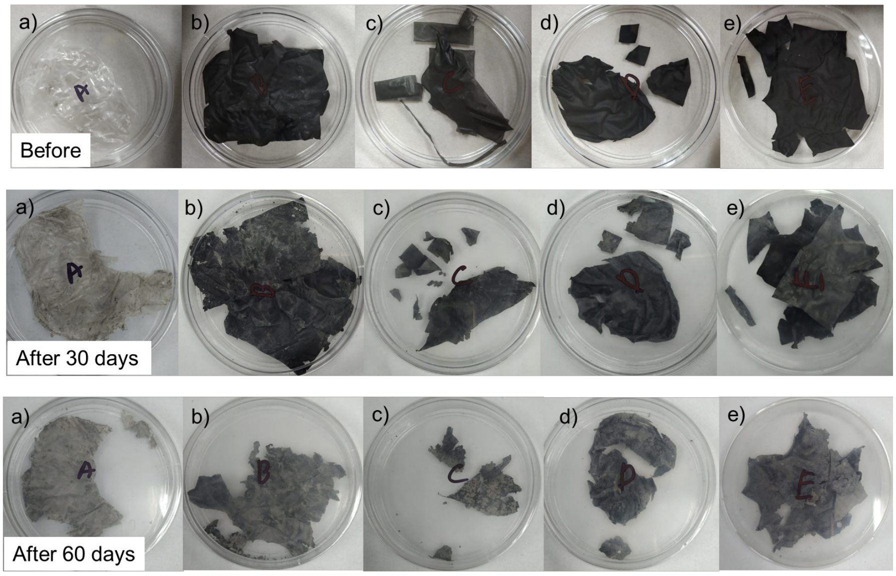

FIGURE 8. Visual of the regenerated cellulose film samples before, after 30 days and after 60 days soil degradation analysis of a) CT, b) CT-rCB (25%), c) CT-T-rCB (10%), d) CT-T-rCB (25%) and e) CT-T-rCB (50%)

CONCLUSION

In summary, incorporating treated rCB into the cellulose film improved its physical properties, including porosity, content, and compatibility. Films with untreated rCB appeared rougher due to impurities. Increasing the treated rCB percentage improved the film's opacity and provided optimum thermal stability at 10%. However, tensile strength decreased with treated rCB addition. Water vapor permeability tests showed that 10% treated rCB gave the best moisture retention, indicated by a lower water vapor transmission rate (WVTR). Treated rCB increased the film's hydrophilicity and influenced its degradation in soil over time. Using 10% treated rCB in the cellulose film offered optimal performance in thermal stability, moisture retention, and hydrophilicity, making it suitable for biodegradable black mulching. However, the reduction in tensile strength must be considered for applications needing high mechanical strength.

ACKNOWLEDGEMENTS

This research was funded by the Ministry of Higher Education, Malaysia through research project grant LRGS/1/2019/UKM-UKM/5/1 and Skim Zamalah Universiti Penyelidikan by Universiti Kebangsaan Malaysia (UKM). The authors acknowledge Sun Rubber Industry Sdn. Bhd. for providing the rCB. The authors are also thankful to the Centre for Research and Instrumentation Management (CRIM) as well as the Faculty of Science and Technology, UKM for the instrument facilities provided.

REFERENCES

Amalini, A.N., Haida, M.K.N., Imran, K. & Haafiz, M.K.M. 2019. Relationship between dissolution temperature and properties of oil palm biomass based-regenerated cellulose films prepared via ionic liquid. *Materials Chemistry and Physics* 221: 382-389.

Amare, G. & Desta, B. 2021. Coloured plastic mulches: Impact on soil properties and crop productivity. *Chemical and Biological Technologies in Agriculture* 8: 4.

Azmi, N.A.S., Lau, K.S., Chin, S.X., Zakaria, S., Chowdhury, S. & Chia, C.H. 2023. Study on effect of toluene-acid treatments of recycled carbon black from waste tyres : Physico-chemical analyses and adsorption performance. *Sains Malaysiana* 52(9): 2689-2696.

Briassoulis, D. & Giannoulis, A. 2018. Evaluation of the functionality of bio-based plastic mulching films. *Polymer Testing* 67: 99-109.

Bumrungnok, K., Threepopnatkul, P., Amornsakchai, T., Chia, C.H., Wongsagonsup, R. & Smith, S.M. 2023. Toward a circular bioeconomy: Exploring pineapple stem starch film as protective coating for fruits and vegetables. *Polymers* 15(11): 2493.

Dong, P., Maneerung, T., Cheng, W., Zhen, X., Dai, Y., Tong, W.Y., Ting, Y., Nuo, S., Wang, C. & Gee Neoh, K. 2017. Chemically treated carbon black waste and its potential applications. *Journal of Hazardous Materials* 321: 62-72.

Fida Banu, M.R., Rani, B., Kavya, S.R. & Nihala Jabin, P.P. 2023. Biochar: A black carbon for sustainable agriculture. *International Journal of Environment and Climate Change* 13(6): 418-432.

Galli, E. 1982. Carbon blacks. *Plastics Compounding* 5(2): 1-5.

Geng, H., Yuan, Z., Fan, Q., Dai, X., Zhao, Y., Wang, Z. & Qin, M. 2014. Characterisation of cellulose films regenerated from acetone/water coagulants. *Carbohydrate Polymers* 102(1): 438-444.

Haapala, T., Palonen, P., Tamminen, A. & Ahokas, J. 2015. Effects of different paper mulches on soil temperature and yield of cucumber (*Cucumis sativus* L.) in the temperate zone. *Agricultural and Food Science* 24(1): 52-58.

Harada, J., Macedo, J.R.N., Machado, G.A.F., Valenzuela-Díaz, F., Moura, E.A.B. & Rosa, D.S. 2016. Effects of carbon black incorporation on morphological, mechanical and thermal properties of biodegradable films. In *Characterization of Minerals, Metals, and Materials,* edited by Ikhmayies, S.J., Li, B., Carpenter, J.S., Hwang, J-Y., Monteiro, S.N., Li, J., Firrao, D., Zhang, M., Peng, Z., Escobedo-Diaz, J.P. & Bai, C. Springer, Cham. pp. 697-704.

Harada, J., de Souza, A.G., de Macedo, J.R.N. & Rosa, D.S. 2019. Soil culture: Influence of different natural fillers incorporated in biodegradable mulching film. *Journal of Molecular Liquids* 273: 33-36.

Kader, M.A., Senge, M., Mojid, M.A. & Ito, K. 2017. Recent advances in mulching materials and methods for modifying soil environment. *Soil and Tillage Research* 168: 155-166.

Kasirajan, S. & Ngouajio, M. 2012. Polyethylene and biodegradable mulches for agricultural applications: A review. *Agronomy for Sustainable Development* 32(2): 501-529.

Lamont, W.J. 2017. Plastic mulches for the production of vegetable crops. In *A Guide to the Manufacture, Performance, and Potential of Plastics in Agriculture*, edited by Orzolek, M.D. Elsevier Ltd. pp. 45-60.

Liu, S. & Zhang, L. 2009. Effects of polymer concentration and coagulation temperature on the properties of regenerated cellulose films prepared from LiOH/urea solution. *Cellulose* 16(2): 189-198.

Ning, R., Liang, J., Sun, Z., Liu, X. & Sun, W. 2021. Preparation and characterization of black biodegradable mulch films from multiple biomass materials. *Polymer Degradation and Stability* 183: 109411.

Puri, L., Hu, Y. & Naterer, G. 2024. Critical review of the role of ash content and composition in biomass pyrolysis. *Frontiers in Fuels* https://doi.org/10.3389/ffuel.2024.1378361

Qiu, K. & Netravali, A.N. 2012. Fabrication and characterization of biodegradable composites based on microfibrillated cellulose and polyvinyl alcohol. *Composites Science and Technology* 72(13): 1588-1594.

Tarara, J.M. 2000. Microclimate modification with plastic mulch. *HortScience* 35(2): 169-180.

Wang, J., Gardner, D.J., Stark, N.M., Bousfield, D.W., Tajvidi, M. & Cai, Z. 2018. Moisture and oxygen barrier properties of cellulose nanomaterial-based films. *ACS Sustainable Chemistry and Engineering* 6(1): 49-70.

Yang, Y., Li, P., Jiao, J., Yang, Z., Lv, M., Li, Y., Zhou, C., Wang, C., He, Z., Liu, Y. & Song, S. 2020. Renewable sourced biodegradable mulches and their environment impact. *Scientia Horticulturae* 268: 109375.

Yeng, L.C., Wahit, M.U. & Othman, N. 2015. Thermal and flexural properties of regenerated cellulose(RC)/poly(3- hydroxybutyrate)(PHB)biocomposites. *Jurnal Teknologi* 75(11): 107-112.

Zhang, R., Wang, X. & Cheng, M. 2018. Preparation and characterization of potato starch film with various size of Nano-SiO_2. *Polymers* 10(10): 9-12.

Zhang, S., Zhang, X., Wan, X., Zhang, H. & Tian, J. 2023. Fabrication of biodegradable films with UV-blocking and high-strength properties from spent coffee grounds. *Carbohydrate Polymers* 321: 121290.

*Corresponding author; email: chia@ukm.edu.my