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Reinforcement of Flaxseed Mucilage-Based Edible Film with Nanocrystalline Cellulose

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Flaxseed mucilage-based edible film plasticized with glycerol was previously prepared, with relatively low strength and stiffness which limits its application as food packaging material. With the aim to reinforce the mechanical properties, nanocrystalline cellulose (NCC) was added into the film in the present study. 1 wt.% to 5 wt.% of nanocrystalline cellulose suspension (NCC1 to NCC5) were prepared and added into the optimized formulation of flaxseed mucilage with 1 wt.% of glycerol (FMG). The nanocomposite films were formed via solution casting. They were studied in terms of mechanical, morphological, physical and antioxidant properties. As the NCC suspension increased, the tensile strength and Young's modulus (or stiffness) significantly increased (to 2.3 MPa and 0.32 MPa) whereas the elongation at break decreased (to 40.3%) ($p<0.05$). From the morphology analyses of the FMG-NCC samples' tensile fracture surfaces, agglomeration of NCC within the matrix was observed. This resulted in the lower mechanical reinforcement than anticipated. The addition of various loading of the NCC suspension did not significantly change the samples' colour which remained within the yellowish hue and transparent ($p>0.05$). While FMG itself had low antioxidant (at 9.13% of free radical scavenging activity and 13.8 mg gallic acid equivalent (GAE)), the addition of NCC did not significantly affect these antioxidant activities ($p>0.05$).

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Introduction

For a long time, polymers have supplied most of the common packaging materials because they present several ideal characteristics such as softness, lightness and transparency. However, the increasing usage of synthetic polymers such as polyethylene, polypropylene, polystyrene and polyvinyl chloride has notably burdened the environment due to their non-biodegradability (Siracusa et al., 2008; Rhim et al., 2013). With the rising awareness of sustainability, packaging materials from renewable biological resources, namely biopolymers are continuous being sought after. In addition, biopolymer packaging materials are also outstanding vehicles for incorporating various additives such as antioxidants, antifungal agents, antimicrobials, colours, and nutrients (Rhim et al., 2013).

Flaxseed mucilage has been continuously investigated as a potential packaging or coating material (Tee et al., 2016; Tee et al., 2017). The mucilage percentage of flaxseed is considerably high, which contributes to its excellent film characteristic such as good water retaining ability, high viscosity in aqueous solution and good barrier properties (Salamanca et al., 2011). Tee et al., (2016; 2017) tested the formation of flaxseed mucilage-based film with the addition of glycerol as plasticizer. This resulted in the casted film that is transparent, stable and flexible. Despite of its advantages, the addition of glycerol has significantly decreased the strength of the developed film which could limit its potential to package a wider range of bioproducts.

With the aim to overcome the drawback, the present work added and studied the effect of adding various concentration of nanocrystalline cellulose (NCC) into the sample. As compared to cellulose fibres, NCC holds advantages such as nanoscale dimension, high strength and modulus and high surface area (Habibi et al., 2010; Peng et al., 2011). In addition, the percolation of the filler network in the polymer matrix and the filler's geometrical aspect ratio are also the major factors in affecting the mechanical performance of the composite (Peng et al., 2011). Several recent literatures have reported the reinforcement in tensile strength of various bio-based films upon the addition of NCC (Huq et al., 2012; Abdollahi et al., 2013; Reddie and Rhim 2014; Alves et al., 2015; Wang et al., 2017). By loading NCC into the present sample, it is envisaged that the strength of the film shall improve.

The viscous flaxseed mucilage is a mixture of rhamnogalacturonan I and arabinoxylan with novel side group substitution, where the two polysaccharide molecules form large aggregates in solution (Naran et al., 2008). Arabinoxylan is found to be able to scavenge free radicals (Barthet et al., 2014). Phenolic acids, which are also water-extractable, was hypothesized to contribute to the antioxidant activity in flaxseed mucilage (Barthet et al., 2014; Malunga and Beta 2015). In addition, Bouaziz et al., (2016) reported that flaxseed mucilage can be a potent antioxidant compound. Acknowledging the possible antioxidant activities in the flaxseed mucilage, in addition to the mechanical, physical, and morphological analyses, the present work also subjected the samples to antioxidant tests to investigate the samples' feasibility as an active packaging without the addition of other antioxidant compounds.

Materials and Methods

Materials

Golden flaxseeds (*Linum usitatissimum* L.) (Organic Care2u, 150509R6) were purchased from NSK Sdn. Bhd., Kuala Lumpur, Malaysia. The seeds were packed in vacuum-sealed plastic bags and kept at room temperature. Nanocrystalline cellulose (CAS number: 9004-34-6) was purchased from Cellulose Lab, Canada. Reagent-grade glycerol and sodium hydroxide (NaOH) pellets were purchased from QReC (Asia) Sdn. Bhd., Selangor, Malaysia; 2,2-diphenyl-1-picryhydrazyl (DPPH), sodium carbonate (Na_2CO_3) and gallic acid ($C_7H_6O_5$) were purchased from Sigma-Aldrich (M) Sdn. Bhd., Selangor, Malaysia; and Folin-Ciocalteu reagent was purchased from Merck Sdn. Bhd., Selangor, Malaysia.

Extraction of flaxseed mucilage

A comprehensive methodology for the extraction of flaxseed mucilage was reported formerly (Tee et al., 2016). The flaxseeds were soaked with distilled water (1:30) and stirred at 1000 rpm at an elevated temperature of 80 °C to 100 °C for at least 3 h. Upon cooling to room temperature, the mixture was centrifuged and filtrated to separate the mucilage from the seeds.

Preparation of NCC suspension

The NCC suspension was prepared in reference to Khan et al., (2012). The NCC powder was added into distilled water at 0.1% w/w, followed by stirring at 700 rpm for 3 h using a hotplate magnetic stirrer (MS-H280-Pro, Dragon Laboratory, China) at room temperature. The NCC suspension was then subjected to ultrasonication (uSonic OS-6001, OSIM, China) for 30 minutes for the dispersion of NCC particles in the water.

Film Formation

Muñoz Hernández (2012) suggested the adjustment of mucilage's pH to 9 to enable the maximum hydration capacity of the mucilage. First, 0.1M of sodium hydroxide (NaOH) was added into the mucilage solution to increase the pH to 9. The solution was then stirred using a hotplate magnetic stirrer (MS-H280-Pro, Dragon Laboratory, China) at 120 rpm and 80 °C for 30 minutes. Then, the solution was weighed and added with 1% w/w of glycerol as plasticizer. The addition of NCC suspension was done in reference to Khan et al., (2012). The NCC suspension was added into the prepared solution according to the formulation as shown in Table 1, where the composite solution was then homogenized for 2 minutes at approximately 5400 rpm (HG-15A, Daihan Scientific, Korea). Then, 30 g of the solution was casted onto a 95 mm diameter Petri dish (also conveyed as 0.42 g cm^{-2}). The nanocomposite film was formed through evaporation of the cast mixture in a convective oven at 40 °C for 17 h. Finally, the film was removed from the Petri dish using a spatula and forceps. They were stored in a desiccator at 25 °C and 52% relative humidity for at least two days prior to the tests.

Table 1. Formulation of nanocomposite film with the addition of NCC

Sample	Composition of sample
FMG	Flaxseed mucilage + 1% wt glycerol
FMG-NCC1	Flaxseed mucilage + 1% wt glycerol + 1% wt NCC suspension
FMG-NCC2	Flaxseed mucilage + 1% wt glycerol + 2% wt NCC suspension
FMG-NCC3	Flaxseed mucilage + 1% wt glycerol + 3% wt NCC suspension
FMG-NCC4	Flaxseed mucilage + 1% wt glycerol + 4% wt NCC suspension
FMG-NCC5	Flaxseed mucilage + 1% wt glycerol + 5% wt NCC suspension

Tensile test

In reference to ASTM D882-12 (2012), tensile tests were done using a material testing machine (Lloyd Instruments, LF Plus, US). The samples were cut into the dimensions of 70 mm x 20mm. Each sample was clamped between grips, and the load and extension were recorded during the extension at 10 mm min^{-1}, with an initial distance between the grips of 60 mm. Tensile strength (TS), elongation at break (EB) and Young's modulus (YM) were measured in triplicates for each film formulation. The average values of three repetitions of each sample were reported with standard deviation.

Morphological analysis

The morphology of the tensile fracture surface of sample was observed using a field emission scanning electron microscope (FE-SEM) (JSM-7600F, JEOL USA., US) at an accelerating voltage of 20 kV. Before scanning, the samples were sputter-coated with gold (Leica Microsystems Inc., Leica EM ACE200, US).

Film Colour

The sample colour was determined using a colorimeter (HunterLab, ColorFlex EZ, US). The degree of lightness (*L*) and chromaticity parameters, *a* (red-green) and *b* (yellow-blue) of the film were measured. The total colour difference (ΔE) was calculated using Equation 1, with the white standard coordinates of *L**= 93.82, *a**= -1.2 and *b**= 1.6. Values were expressed as the mean of five readings measured on different positions of the sample with standard deviation.

$$\sqrt{\{\quad\}} \qquad \text{(Eq. 1)}$$

Light Transmission and Transparency

The light transmittance and absorbance of the samples at the UV range from 200 to 800 nm was measured using a UV-VIS Spectrophotometer (UviLine 9400, Secomam, US). The samples were cut into rectangular shape with the approximate area of 8 mm x 35 mm prior to insertion into the sample cell. The absorbance was determined against air as a standard, where each obtained absorbance was used to calculate the transparency value of the samples using the equation of transparency value = *A*600/*x*, where *A*600 is the absorbance at 600 nm and *x* is the thickness (mm) of film (Han and Floros 1997). Greater transparency values represent lower film transparency.

Free Radical Scavenging Assay

The 2,2-diphenyl-1-picryhydrazyl (DPPH) free radical scavenging assay was done in reference to Jouki et al., (2014a,b). 25 mg of the film sample was dissolved into 5 mL of distilled water and was vortexed. 0.1mL of the film extract solution was then added with 3.9mL of DPPH solution (0.1mM methanol solution) followed by 60 minutes of incubation at room temperature. Incubation was done in dark ambient due to the sensitivity of DPPH to light. Following the incubation, absorbance of the sample solution was immediately recorded at 517nm using UV-visible spectrophotometer (UviLine 9400, Secomam, US). A reference sample was also prepared by replacing the film extract solution with distilled water. The percentage of the DPPH activity in scavenging free radical was calculated using Equation 2. All the tests were done in triplicate. The average values of three repetitions of each sample were reported with standard deviation.

% of free radical scavenging activity= $\frac{\quad}{\quad}$

(Eq. 2)

where $A_{reference}$ is the absorbance of the DPPH solution without the film sample, and A_{sample} is the absorbance of the DPPH solution with the addition of the film sample.

Total Phenolic Content Measurement

Colourimetrical measurement of total phenolic content through the Folin-Ciocalteu reaction was done in reference to Siripatrawan and Harte (2010), and Jouki et al., (2014a) with modification. 25mg of the film sample was dissolved in 5mL distilled water and was vortexed. 0.1mL of the extract solution, 7mL of distilled water and 0.5mL of Folin-Ciocalteu reagent were mixed and incubated at room temperature for 8 min. 1.5mL of sodium carbonate solution (2%, w/v) was added into the mixture. Subsequently, the mixture was topped up with distilled water until 10mL and was vortexed. The mixture was then kept in the dark at room temperature for 2 hours. The absorbance reading of the sample was performed at 765nm using a spectrophotometer (UviLine 9400, Secomam, US). The results of the present assay were expressed as mg gallic acid equivalent (GAE) per gram of dried sample according to Equation 3. The assay was done in triplicate. The average values of three repetitions of each sample were reported with standard deviation.

$$\frac{\quad}{\quad} \qquad \text{(Eq. 3)}$$

where *T* is total content of phenolic compound in the sample (mg GAE), *C* is the concentration of gallic acid obtained from the standard curve (mg/mL), *V* is the volume of film extract (mL) and *M* is the weight of dried film (mg).

Statistical Analysis

The statistical analysis was done using SPSS. Analysis of variance (ANOVA), Tukey Honest Significant Difference (HSD) multiple range test were performed to determine the significant difference among the mean value of the sample properties. Pearson correlation coefficient test was carried out for antioxidant assay to investigate the relationship between two assays. The significant difference test was conducted with significance level of 0.05 (p level of 0.05).

Results and Discussion

Tensile Properties

The tensile strength (TS), elongation at break (EB) and Young's modulus (YM) of the film samples were tabulated in Table 2.

The TS and YM of the samples increased significantly with the increase of NCC loading in the FMG ($p<0.05$). In contrary, the EB of the samples decreased significantly with the subsequent increase of NCC loading ($p<0.05$). FMG-NCC5 reported the highest TS and YM at 2.30 MPa and 0.32 MPa, with a decrease of EB to 40.33%. The reinforcing effect of the NCC might be due to the percolation of the filler network in the polymer matrix (Peng et al., 2011). The polarity and high surface area of NCC could have formed a strong interaction with the polymer matrix which enabled uniform envelopment by the matrix. This improves the dispersion of NCC (Oksman et al., 2006, Alves et al., 2015). From the present morphology analysis, it was observed that the NCC particles were indeed well covered with the mucilage matrix. Thus, it was hypothesized that a rigid network was formed between the fillers and the polymer matrix.

Table 2. Mechanical properties of flaxseed mucilage-based films with various concentration of NCC

Sample	Tensile strength (MPa)	Elongation at break (%)	Young's modulus (MPa)
FMG	0.55 ± 0.14^{a}	91.10 ± 6.73^{a}	0.007 ± 0.0006^{a}
FMG-NCC1	0.84 ± 0.15^{ab}	80.27 ± 5.66^{ab}	0.12 ± 0.09^{ab}
FMG-NCC2	1.13 ± 0.28^{bc}	76.23 ± 6.78^{bc}	0.19 ± 0.02^{bc}
FMG-NCC3	1.26 ± 0.29^{bc}	63.90 ± 7.47^{bcd}	0.21 ± 0.01^{bcd}
FMG-NCC4	1.68 ± 0.19^{c}	58.07 ± 10.37^{cd}	0.24 ± 0.03^{cd}
FMG-NCC5	2.30 ± 0.13^{d}	40.33 ± 7.03^{d}	0.32 ± 0.02^{d}

Mean ± standard deviation. Means followed by the different letters are signifying that they are significantly different ($p<0.05$), based on Tukey HSD test.

Morphology analysis

The micrographs of the NCC particles and the tensile fracture surfaces of some film samples were shown in Fig. 1. From Fig. 1(a), the NCC used in the present study was seemingly spherical. Spherical NCC was derived from the sulphuric acid hydrolysis of cellulose (Lu and Hsieh 2010; Reddy and Rhim 2014; Wang et al., 2017).

A good homogenization of NCC with polymer matrix is important. Without sufficient homogenization, the NCC can easily agglomerate and the reinforcing effect from their addition may not be optimized (Peng et al., 2011). In the present study, while good adherence or embedment of NCC particles to the matrix was observed, there were also several notable NCC agglomeration sites on the nanocomposites' fracture surfaces (Fig. 1(b) and 1(c)). The comparatively low homogenization rate (5400 rpm) used in the present study may have led to the uneven dispersion of NCC suspension as compared to other works with homogenization rates of 10000 to 20000 rpm (Khan et al., 2012; Abdollahi et al., 2013; Reddie and Rhim 2014).

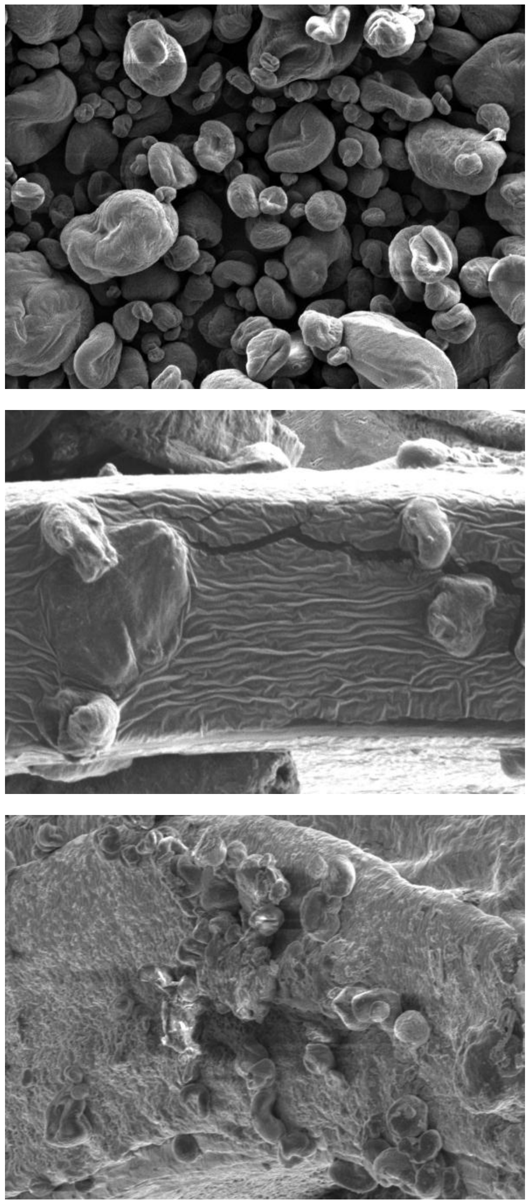
Film colour

The colour parameters of all the film samples were tabulated in Table 3, where *L* is the degree of lightness, *a* represents the green-red region, *b* represents the blue-yellow region and ΔE is the total colour difference of the sample from the white standard (ΔE).

The colour of the plasticized flaxseed mucilage-based film (FMG) was slightly greenish (-*a)* and yellowish (+*b*) with dark appearance (low *L* value). The addition of various loading of NCC into the film sample did not significantly affect the colour. This could be due to the low concentration of NCC suspension used and its nanoscale size which may not be visualized under the visible wavelength (Bandyopadhyay-Ghosh et al., 2010).

Light Transmission and Transparency

Table 4 showed the light transmittance at wavelength of 200nm to 800nm and the transparency values of the film samples. A lower transparency value indicates a more transparent film.



(a)
(b)
(c)

Fig. **1.** FE-SEM micrographs of (a) NCC; and the tensile fracture surfaces of (b) FMG-NCC1 and FMG-NCC5 at x1000 magnification.

As shown in Table 4, light was unable to transmit through all of the films between the 200 to 280nm wavelengths, namely the UV region. This indicated high UV barrier capability of the flaxseed mucilage-based films, which can be essential to to halt the photo-oxidation of the foodstuff and hence, prolonging their shelf life (Dick et al., 2015). Film sample without NCC was the most transparent, indicated by its low transparency value. There was no significant effect of adding up to 4 % of NCC suspension. In contrary, adding 5% of NCC suspension into the film significantly affected the films' transparency. The addition of the NCC should not significantly affect the transparency of the films due to its nanoscale dimension (Bandyopadhyay-Ghosh et al., 2010). The present occurrence may be due to the increased NCC aggregation within the polymer matrix from the inadequate homogenization procedure. Wang et al., (2017) reported a reduction in alginate-based films' transparency with the increase of NCC loading and concluded that the decrease was due to the hindrance of the light passage caused by the fillers in the polymer matrix.

Table 3. Colour measurements of the flaxseed mucilage-based films with various concentration of NCC

Sample	Colour			ΔE
	L	*a*	*b*	
FMG	22.28 ± 0.68[a]	-0.89 ± 0.04[a]	3.20 ± 0.12[a]	71.55 ± 0.68[a]
FMG-NCC1	22.44 ± 0.99[a]	-0.87 ± 0.04[a]	3.51 ± 0.24[a]	71.41 ± 0.98[a]
FMG-NCC2	23.08 ± 0.62[a]	-0.89 ± 0.06[a]	3.46 ± 0.31[a]	70.76 ± 0.61[a]
FMG-NCC3	21.31 ± 0.48[a]	-0.90 ± 0.02[a]	3.28 ± 0.31[a]	72.53 ± 0.49[a]
FMG-NCC4	22.05 ± 1.26[a]	-0.88 ± 0.03[a]	3.50 ± 0.23[a]	71.80 ± 1.26[a]
FMG-NCC5	22.47 ± 1.41[a]	-0.88 ± 0.04[a]	3.33 ± 0.18[a]	71.37 ± 1.41[a]

Mean ± standard deviation. Means followed by the different letters are signifying that they are significantly different ($p<0.05$), based on Tukey HSD test.

Table 4. Light transmittance and transparency value of flaxseed mucilage-based films with various concentration of NCC.

Sample	Light transmittance (%) at different wavelength (nm)								Transparency value
	200	280	350	400	500	600	700	800	
FMG	0.06	0.01	2.97	23.37	48.77	58.97	61.07	66.5	3.35 ± 0.21[a]
FMG-NCC1	0.06	0.02	2.14	16.1	38.87	37.9	51.27	53.37	4.61 ± 0.20[a]
FMG-NCC2	0.05	0.01	1.43	12.73	29.17	40.4	48.9	47.37	3.91 ± 0.27[a]
FMG-NCC3	0.05	0.02	2.41	12.35	28.23	35.3	34.97	43.27	4.39 ± 0.90[a]
FMG-NCC4	0.05	0.02	2.25	16.46	32.3	39.4	42.57	51.27	4.69 ± 0.58[a]
FMG-NCC5	0.04	0.01	0.46	6.36	21.13	27.37	33.2	37.97	6.60 ± 0.56[b]

Mean ± standard deviation. Means followed by the different letters are signifying that they are significantly different ($p<0.05$), based on Tukey HSD test.

Antioxidant activity analyses

The flaxseed mucilage-based film samples were tested for their potential to be classified as active food packaging material. The two antioxidant assays, DPPH and total phenolic content were performed and the average results were tabulated in Table 5.

From Table 5, all of the films have the potential to scavenge free radicals. However, the films' activity on scavenging free radicals was considerably weak. FMG had the highest free radical scavenging activity and decreased with the increase loading of NCC. Nonetheless, the decrease were insignificant ($p>0.05$). The total phenolic content of the samples also reported similar trend, where FMG had the highest phenolic content while FMG-NCC5 had the lowest. From the Pearson correlation coefficient test, the result of both antioxidant assays showed statistically insignificant or weak correlation ($p>0.05$).

Both results from the antioxidant assays showed that the films had an innate ability to scavenge free radicals. Nonetheless, they were not substantial enough to be considered as active packaging material at the present stage.

Table 5. Antioxidant assay results of flaxseed mucilage-based films with various concentration of NCC

Sample	Antioxidant assay	
	% of free radical scavenging activity	Total phenolic content (mg GAE)
FMG	9.13 ± 0.47[a]	13.08 ± 0.74[a]
FMG-NCC1	8.88 ± 0.16[a]	12.63 ± 0.95[a]
FMG-NCC2	8.81 ± 0.17[a]	12.44 ± 0.33[a]
FMG-NCC3	8.72 ± 0.47[a]	12.25 ± 0.72[a]
FMG-NCC4	8.49 ± 0.54[a]	11.77 ± 0.84[a]
FMG-NCC5	8.15 ± 0.71[a]	11.45 ± 0.59[a]

Mean ± standard deviation. Means followed by the different letters are signifying that they are significantly different ($p<0.05$), based on Tukey HSD test.

Conclusions

The development of nanocomposite films from flaxseed mucilage reinforced with NCC suspension up to 5% wt was successfully performed. As aimed in the present study, the addition of NCC into the flaxseed mucilage-based film did notably reinforced its strength and stiffness, with the acceptable compromise on the elasticity. The presence of NCC within the polymer matrix did not significantly changes the colour, transparency, free radical scavenging activity and phenolic content of the FMG film. The films produced were transparent, able to absorb light at the UV region (200-280 nm), and with its yellowish colour, may retard photo-oxidation of the food if used as coating or packaging material. As the antioxidant activities of the developed film was fairly weak, it will need to be added with other antioxidant compounds for it to be considered as active packaging.

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Conflict of Interest

All the authors declare that they have no conflict of interest.

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