Evaluation of effectiveness of three cellulose derivative-based edible coatings on changes of physico-chemical characteristics of 'Berangan' banana (*Musa sapientum* cv. Berangan) during storage at ambient conditions

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Abstract: Three edible coating formulations, namely methyl cellulose (MC)-glycerol, sodium carboxymethyl cellulose (Na-CMC) - glycerol and hydroxypropylmethyl cellulose (HPMC) - stearic acid-glycerol with three concentrations of main edible components, MC (2, 3.5 and 5% w/v), Na CMC (0.1, 0.8 and 1.5% w/v) and HPMC (2, 4 and 6% w/v) were applied on the peel surface of green mature Berangan banana and stored at 26±2°C and 40-50% relative humidity. Results showed that coated banana using high concentration (1.5% w/v) of Na-CMC and 1% (w/v) glycerol had the least weight loss, total colour difference and total soluble solids content (7.14%, 6.06 and 3.95° Brix, respectively) and had the highest firmness and pH (20.42 N and 5.36) at day 10 of storage while the control samples were already completely ripened. This was followed by banana coated with low concentrations of Na-CMC (0.1% w/v)-glycerol (1% w/v) and HPMC (2% w/v)-stearic acid (1% w/v)-glycerol (1% w/v), MC and control. However, no significant difference was observed between banana coated with low concentration of MC (2% w/v)-glycerol (1.25% w/v) and control.

Introduction

Edible coatings applied on fresh fruits to reduce the moisture transfer, oxidation, metabolic processes, respiration rate, physical/mechanical impacts and microbial growth and to maintain the sensory quality and safety are important to prolong their shelf-life (Nisperos-Carriedo *et al*., 1992). Edible coating acts as a physical barrier for the gas exchange between the fruit and their storage environment and create modified atmosphere. It slows down both the rates of generating energy and substrate degradation which in turn will slow down the biochemical reactions leading to fruit ripening (Maftoonazad and Ramaswamy, 2005).

The polysaccharides as coating materials for fruits have been used extensively in the past few years. These natural polymers, in addition to the above mentioned benefits, present advantages due to their availability, low cost, and biodegradability. The latter in particular is of great interest, as it leads to a reduction in the large quantities of non-biodegradable synthetic packaging materials (Zhou *et al*., 2008). In addition, their physico-chemical properties can also be improved by modifying them.

Cellulose is a naturally occurring polymer which is found abundantly and is usually present as a linear polymer of anhydroglucose (Kester and Fennema, 1986). At a molecular level, cellulose is a simple linear polymer comprising of β-[1,4] linked D-glucose molecules. Because of its regular structure and array of hydroxyl groups, it tends to form strong hydrogen-bonded crystalline microfibers, which are insoluble in several solvents (Zugenmaier, 2006).

Several cellulose derivatives such as methyl cellulose (MC), carboxymethyl cellulose (CMC), hydroxypropyl cellulose (HPC), and hydroxypropylmethyl cellulose (HPMC) are widely produced commercially (Olivas and Barbosa-Canovas, 2005). These edible coatings have been applied to a variety of fruits to provide moisture, oxygen and carbon dioxide barriers, and to improve adhesion of coating formulations (Kester and Fennema, 1986). Coatings and films that are based on these cellulose ethers are generally transparent, flexible, odourless, tasteless, water-soluble, and resistant to O_2 and CO_2 (Nisperos-Carriedo *et al*., 1992).

Edible coatings based on cellulose derivatives have also been used to delay ripening in some climacteric fruits like mango, papaya and avocado. Baldwin *et al*. (1999) used an edible coating based on HPC on mango and reported that it could delay ripening and increased concentrations of flavour

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volatiles of mango. The application of CMC sodium salt (Na CMC) coating was also found able to delay ripening of mango up to 32 days at 13°C and 85% RH (Carrillo-Lopez *et al*., 2000). Coating treatments based on CMC sodium salt extended storage life of pineapple fruit up to 5 weeks at 10°C and 90-95% RH, whereas the control fruit had a storage life of only 28 days (Nimitkeatkai *et al*., 2006). Maftoonazad and Ramaswamy (2005) applied MC coating on avocado. Their results indicated the strong effects of the coating in retarding the ripening behaviour of coated avocados as compared to control samples, at ambient conditions.

Banana fruit is climacteric and ripens rapidly after harvest. As reported by Marriott (1980), ripening of banana is associated with peel colour change from green to yellow, hydrolysis of starch to simple sugars, softening of the pulp and development of aroma. Banana has short storage life in ambient temperature (25 - 35°C) and it is usually harvested at mature green stage and stored (Stewart *et al*., 2005). Therefore, the purpose of the present study was to evaluate the effects of three cellulose derivative - based edible coatings (Na-CMC, MC and HPMC) on the changes in weight loss, firmness, total colour difference (ΔE) and total soluble solids content (TSS) of Berangan banana. Glycerol and stearic acid were used as plasticizers in the coating formulations to improve the water vapour and oxygen barrier and mechanical properties of the coatings.

Materials and Methods

Plant material

Mature green Berangan bananas (*Musa sapientum* cv. Berangan) from the second hand of a bunch that were free from any form of mechanical damage were used in this study. They were purchased from a local wholesale market (Pasar Borong Selangor, Malaysia). Fingers of banana were detached from the bunch and then the latex allowed to dry for one hour at ambient temperature (26 ± 2°C and 40-50% relative humidity). Banana fingers were surface disinfected by immersing in 0.1% (w/v) sodium hypochlorite solution for 2 min, washed and air-dried. The fingers were then separated into two lots; treated and control. Banana fingers from treated lot were coated with different edible coating solutions, while those from the control lot were dipped in distilled water. Both lots were then stored at ambient temperature. Each lot was replicated three times and each replicate had three representative samples.

Coating and storage procedures

All cellulose derivatives (Na-CMC, MC and HPMC) were purchased from Sigma-Aldrich (Sigma-Aldrich Co. Steinheim, Germany). Glycerol (87%) as plasticizer which was purchased from Fluka (Fluka Co. Steinheim, Germany) and stearic acid as an emulsifier and plasticizer purchased from Fisher Scientific (Fisher Scientific Co. Leicestershire, UK) were used in edible formulations as shown in Table 1.The three different edible solutions were prepared by dissolving Na-CMC (0.1, 0.8 and 1.5% w/v), MC (2, 3.5 and 5% w/v) and HPMC (2, 4 and 6% w/v) in distilled water with addition of additives (Table 1). They were then homogenised (Ultra-Turax T25, Janke and Kunkle, IKA-Labortechnik, Breisgau, Germany) at 20000 rpm for 4 min. Individual banana fingers were then immersed in the coating solutions for 1 min and then drained. The treated fruits were dried at ambient conditions (26±2°C and 40-50% relative humidity) to set a coat of edible film on their surface. Uncoated bananas as control samples were immersed in distilled water for the same duration of time. Treated and control samples were then stored at ambient conditions.

Table 1. Composition of edible coating formulations

Formulation	Components		Concentration	(% w/w)
		Low	Moderate	High
A	MC*	2	3.5	5
	glycerol	1.25	1.25	1.25
B	HPMC*	2	4	6
	glycerol	1	1	1
	Stearic acid	1	1	1
C	Na-CMC*	0.1	0.8	1.5
	glycerol	1	1	1

* Main edible component.

Quality evaluations

The weight loss, firmness, colour, TSS and pH of coated and control banana samples as internal and external indicators of banana quality were measured at day 0 and day 10 of storage. The monitoring of physico-chemical attributes of mature green Berangan banana revealed that at ambient conditions, control banana samples were fully ripened and edible at day 10 of storage.

The influence of the coatings on weight loss (%) was determined by weighing the coated and control banana samples on a digital balance (Presica 4000 C, Zurich, Switzerland) at days 0 and 10 of storage. Changes in peel colour of samples were evaluated at days 0 and 10 of storage using a Minolta Chroma meter (CR-300, Minolta Corp. Tokyo, Japan) coupled with a Minolta Chroma C (VO. 27) software. The total colour difference (ΔE) was calculated, for at least three different locations on each banana sample, as the root mean square of the differences in individual L^* (lightness or brightness), a^* (redness or greenness) and b^* (yellowness or blueness) values (equation 1)

(Maftoonazad and Ramaswamy, 2005).

$$[i.e., \Delta E = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{0.5}] \qquad (1)$$

Firmness was determined at the end of the storage period using a texture analyzer machine (TA-XT2i, Stable Micro System., Ltd, Surrey, UK) equipped with a 30N load cell. The maximum force (N) necessary to penetrate the fruit (5 mm) using a rounded 2 mm diameter probe, was measured for at least three different locations; apex, middle and peduncle end, and the results were averaged. Analysis of TSS and pH were done on sample homogenates using a refractometer (Palette PR-32, Atago Co Ltd., Tokyo, Japan) and pH meter (DELTA 320, Shanghai, China), respectively (Cano *et al*., 1997). TSS was expressed as degree Brix (°Brix).

Statistical analysis

Experiments were designed according to a completely randomized design for three concentrations of each edible coating formulation with three replications. Analysis of variance (ANOVA) was performed to evaluate significant differences between independent variables and Tukey's comparison test was used to compare the mean values of variables. All comparisons were made at the 5% level of significance (P< 0.05). Minitab v. 14 statistical software (Minitab Inc., PA, USA) was used for data treatment and statistical analysis.

Results and Discussion

Weight loss

Weight loss which occurs in banana fruit during ripening is due to both two transpiration and respiration. Transpiration is associated with water vapour pressure of the surrounding atmosphere and the banana surface. Weight loss contributed by respiration is a result of carbon atoms, in the form of carbon dioxide molecules, leaving the fruits (Park, 2000). As clearly observed in Figure 1, by increasing the concentrations of cellulose derivatives; MC, HPMC and Na-CMC in edible formulations A, B and C, respectively, the weight loss of coated banana samples decreased. Although coated banana samples had lower weight loss as compared to control samples, the difference between weight loss of coated and control banana samples was not drastic. While the weight loss of control banana samples were 8.67 % at day 10 of storage, this value for coated bananas with high concentrations of MC, HPMC and Na-CMC were 7.44, 7.56 and 7.14% at same storage time. The result was most likely due to the fact that

edible coatings based on hydrocolloids, because of their hydrophilic natures have poor barrier properties towards water vapour transition (Olivas and Barbosa-Canovas, 2005). Because of the regular structure and array of hydroxyl groups of cellulose derivatives, they tend to absorb and form strong hydrogen- bonds with water molecules of fruits and environment (Togrul and Arsalan, 2004). Therefore, addition of stearic acid as a lipid component in order to increase the water resistance of edible formulation based on HPMC did not show significant (P< 0.05) effect to decrease the weight loss as compared to other two edible formulations. Banks (1984), Maftoonazad and Ramaswamy (2005) and Navarro-Tarazaga *et al*., (2008) found that edible coatings based on Na-CMC, HPMC and MC effectively decreased weight loss of banana, mandarin and avocado, respectively.
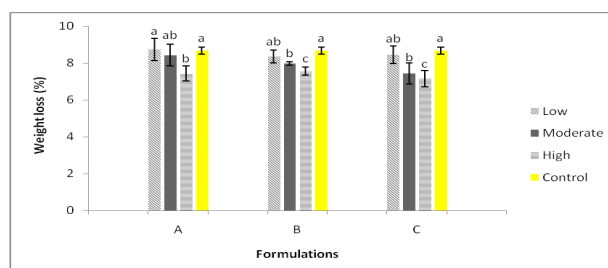

Figure 1. Weight loss of coated and control banana samples at day 10 of storage. Values are means of three replicates (3 representative samples per replicate). Mean values labeled with different letters for each formulation are significantly different (P < 0.05)

Total colour difference

There are normally two types of pigment in the peel of banana: chlorophylls and carotenoids (Marriott, 1980). During ripening of banana, chlorophyll in the peel, enzymatically degrades and banana peel colour changes (Thompson and Burden, 1995). Colour evaluation of banana peel as affected by types and concentrations of edible materials is shown in Figure 2. As clearly observed, total colour difference (ΔE) of coated banana samples are lower than that of control samples. However, the changes in ΔE of coated banana samples with different edible based coatings did not follow the same trends, where edible coatings based on moderate concentration of MC, low concentration of HPMC and high concentration of Na-CMC indicated minimum ΔE, 13.28, 9.62 and 6.06, respectively, among their other concentrations. The result may be related to degree of adhesion between coatings and fruits and cohesion of the molecules within the coatings, where higher cohesion improves barrier and flexibility properties of edible coatings. The degree of adhesion depends on the chemical and electrostatic affinity of the coating material with the surface of the fruit (Olivas and
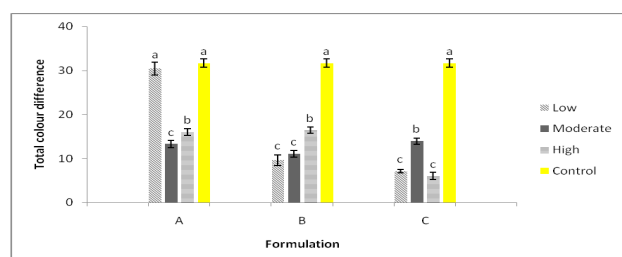
Figure 2. Total colour difference of coated and control banana samples at day 10 of storage. Values are means of three replicates (3 representative samples per replicate). Mean values labeled with different letters for each formulation are significantly different (P < 0.05)

Barbosa-Canovas, 2005). Higher adhesion ensures longer durability of the film on the surface of the fruit. Type and concentration of edible components have strong effect on cohesion (Nisperos-Carriedo *et al.*, 1992). By increasing the cohesion, gas permeability of coatings towards O_2 and CO_2 decrease and this leads to changes in the internal atmosphere of fruit, which in turn will decrease the rate of enzymatic reactions causing degradation and synthesis of banana peel pigments. Momen *et al.* (1997) and Baldwin *et al.* (1999) found that suitable concentration of main edible components and plasticizers in edible coating formulations could cover the stomatal apertures, suppress initial respiration and decrease the rate of ethylene production which ultimately retard degreening and significantly delay ripening of banana and mango, respectively.

Firmness

In general, the firmness of banana fruit during ripening decreases. Softening in banana fruit may be largely due to the breakdown of starch to form sugars since starch granules could have a structural function in the cells (Cano *et al.*, 1997). This could possibility be due to the breakdown of celluloses and other non-pectic polysaccharides under the catalysis of cell wall hydrolysis enzymes (Wills *et al.*, 1998). Figure 3 shows the firmness of coated and control banana samples. Coated bananas with high and moderate concentrations of Na-CMC and low concentration of HPMC in edible coating formulations C and B, respectively, had maximum firmness (20.42, 20.24 and 20.29N) as compared to other coated (having value in the range of 13.36N – 19.91N) and control (11.15N) banana samples. As clearly shown in Figure 3, all edible coatings significantly (P< 0.05) retard the changes in banana firmness as compared to control samples. However, increasing the concentration of MC and Na-CMC had direct effect on the retention of banana firmness while increasing the concentration of HPMC showed the reverse effect. In edible formulation B, both stearic acid and HPMC have a combination of hydrophobic

and hydrophilic groups. The result obtained in this study may be explained by the fact that by increasing the concentration of HPMC, the cohesion of edible coating decreased and permeability of edible coating based on HPMC-stearic acid-glycerol towards O_2 and CO_2 increased. Therefore, increasing the concentration of HPMC could not change the internal atmosphere of fruits and reduce the activities of cell wall hydrolysis enzymes. Navarro-Tarazagav *et al.* (2008) also found that by increasing the concentration of fatty acids in edible coating based on HPMC, the weight loss of coated mandarins decreased but permeability of coating towards O_2 and CO_2 increased.
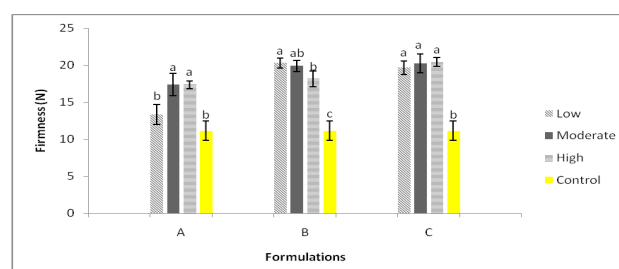

Figure 3. Firmness of coated and control banana samples at day 10 of storage. Values are means of three replicates (3 representative samples per replicate). Mean values labeled with different letters for each formulation are significantly different (P < 0.05)

TSS

During the ripening of banana, starch hydrolyses and break downs and the amount of sugar in fruits usually increases (Cano *et al.*, 1996). In most ripe fruits, including banana, sugar and organic acids form the main component of soluble solids (Wills *et al.*, 1998). TSS of control and coated banana samples are shown in Figure 4. TSS of all coated banana were less than that of control samples (17.55 °Brix) after 10 day of storage. Edible coating formulation C with high and low concentrations of Na-CMC and edible coating formulation B with low concentration of HPMC had minimum TSS: 3.95, 4.15 and 4.55 °Brix, respectively. This indicated significant (P< 0.05) effect on changes of TSS of coated banana samples. These edible coatings seemed to strongly decrease the metabolic processes of coated samples and finally reduced their respiration rates. Sugars and organic acids of fruits are substrates that are consumed by respiration during storage (Zhou *et al.*, 2008). Among all edible coating formulations, edible coating formulation A with low concentration of MC had no strong effect in reducing the metabolic processes of coated banana samples. The results obtained in this study could be related to the fact that low concentration of main edible components creates a weak matrix in combination with plasticizers such as glycerol (Olivas and Barbosa-Canovas, 2005). It seems that glycerol as a plasticizer reduced

intermolecular forces along the polymer chain in MC and increased free volume among MC matrix.
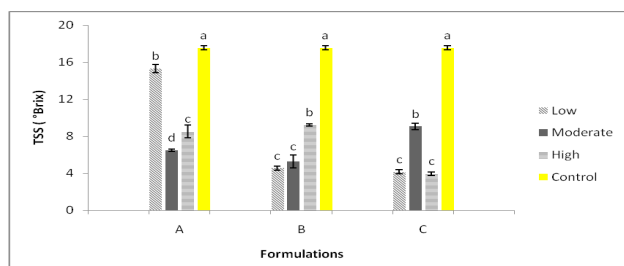

Figure 4. TSS of coated and control banana samples at day 10 of storage. Values are means of three replicates (3 representative samples per replicate). Mean values labeled with different letters for each formulation are significantly different (P < 0.05)

pH

Generally, when banana fruits are harvested at mature green stage, the pulp pH is high but as ripening progresses pH drops (Marriott, 1980). The effect of different edible coatings on pH of coated bananas as compared to control samples is shown in Figure 5. As clearly observed, at day 10 of storage the control banana samples were fully ripened and had minimum pH (4.70). High pH of coated banana with high and low concentrations of Na-CMC in edible coating formulation C (5.36 and 5.35) and, low and moderate concentrations of HPMC in edible coating formulation B (5.35 and 5.26) indicated that these edible coating formulations had strong effect in delaying of banana ripening process. Ethylene is a plant hormone which probably acts in concert with other plant hormones (auxins, gibberellins, kinins and abscisic acid) to exercise control over the fruit ripening process (Wills *et al*., 1998). High concentration of O_2 increases the ethylene production rate. The results obtained in this study were most likely due to the fact that edible coatings decrease the rate of anabolic and catabolic reactions in the fruit (Nisperos-Carriedo *et al*., 1992). It seems that by this reduction, the production of predominant organic acids in banana such as ascorbic and malic acids (Smith *et al*., 1989), as substrates that are consumed by respiration during storage decreased. This reduction in rates of respiration and ethylene production, respectively, in turn delayed the banana ripening process.
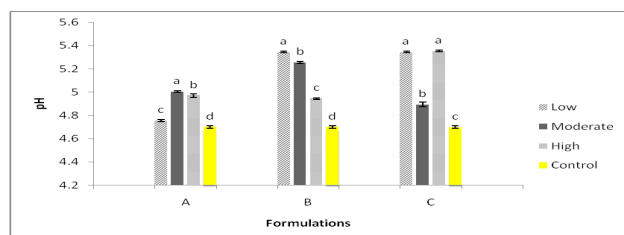

Figure 5. pH of coated and control banana samples at day 10 of storage. Values are means of three replicates (3 representative samples per replicate). Mean values labeled with different letters for each formulation are significantly different (P < 0.05)

Conclusion

Application of edible coatings based on cellulose derivatives (MC, HPMC and Na-CMC) on banana fruits were shown to be beneficial in retarding the ripening behaviour. Banana ripening is the result of a series of complex internal and external changes that are related to internal atmosphere and metabolism variation of banana during ripening. Types and concentrations of components in edible coating formulations are two important factors that strongly affect the delaying of banana ripening process. Among the three edible coating formulations based on cellulose derivatives, results obtained indicate that high concentration of Na-CMC in combination with glycerol had stronger (P< 0.05) effect in retarding the ripening of banana at ambient conditions.

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