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Effects of Xanthan Gum, Carboxymethyl Cellulose, and Gum Arabic on the Properties of Bean Powder-Based Biofilms

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Natural biofilms are increasingly interested in replacing plastic films with serious long-term environmental consequences. This study aimed to investigate the role of Xanthan Gum (XG), Carboxymethyl Cellulose (CMC) and Gum Arabic (GA) on biofilm properties. Membrane preparations (by weight % of the mixture) with 2 % lablab or lima bean powder, 7 % rice flour, the ratio of XG, CMC, GA was 0.15 %, respectively; 0.3 %; 0.45 %. The viscosity of the film-forming solution was determined using a Brookfield DV III Ultra viscometer. The mechanical properties of the films were measured using a Brookfield CT3 instrument. The results showed that the polysaccharides had a significant effect on the properties of the formed film but did not significantly affect the deformation of the film. With the same ratio and type of polysaccharide, the viscosity of the film-forming solution from lima bean powder (LiBP) tended to be higher than that of lablab bean powder (LaBP). GA did not form an efficient film compared with XG and CMC. The biofilms were made from LiBP with 0.3 % XG and 0.3% CMC, and LaBP with 0.45 % CMC had mechanical properties higher than other mixing ratios. Research is promising when applied to producing edible biofilms used in the packaging of low-moisture foods, contributing to a sustainable and environmentally friendly food system.

1. Introduction

Biofilm is a thin film of food packaging and can be viewed as food or additive made from edible ingredients. In addition, it helps to control gas permeability, metabolic activity between fresh products and the atmosphere (Sanchez-Tamayo et al., 2020). In recent years, the issues related to environmental pollution and food safety have increased the interest in the search for new types of packaging materials that are biodegradable and environmentally friendly. Due to biodegradability, renewable ability, and abundant resources, biofilm can be a promising alternative solution to substitute plastic film. Besides, biofilm is also environmentally friendly and has broad applicability (Zhang and Li, 2021). Lipids, carbohydrates and proteins can be considered as the main building blocks of biofilms. Sources of animal proteins such as milk, collagen, gelatin and myofibrillar proteins and vegetable proteins such as corn zein, wheat gluten and soy proteins have been used in biofilm formation. So far, except for the well-published soybean protein film-forming studies, the studies on biofilm formation from other legume proteins are still limited. Lima beans (*Phaseolus lunatus*) belong to the Leguminosae family. The content of nutrients in lima beans includes 17 – 40 % of protein, 0.5 – 2 % of lipid, 4 – 7 % of fiber, 4 – 6 % of ash, and 54 – 60 % of carbohydrate (Yellavila et al., 2021). Lablab bean (*Lablab purpureus*) belongs to the family Fabaceae grown in tropical and subtropical India. Lablab beans are grown and popular in Southeast Asia, such as Vietnam, Thailand, Indonesia, Nepal, Philippines, etc. In fact, lablab beans are considered a multipurpose crop since it is used for food, forage, soil protection, and weed control. Recently, lablab beans have been an important source of therapeutic agents used in the modern and traditional systems of medicine, and it carries tremendous healing potential. The content of nutrients in lablab beans including 24.70 – 25.06 % of protein, 50.83 – 53.16 % of carbohydrate, 3.610 – 3.643 % of nitrogen, 3.020 – 3.124 % of potassium, 0.338 – 0.356 % of phosphorus, 1.764 – 1.804 % of calcium, 100 g of dry beans contains 25.6 g or 64 % of fiber (Naeem, 2009).

Xanthan Gum (XG) is a high molecular weight exo-polysaccharide composed mainly of the bacterium Xanthomonas (a Gram-negative genus with several species). The food sector and many other industries have

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used XG as an additive, such as food, food packaging, water-based paints, petroleum, oil recovery, construction and building materials. XG is hydrophilic and can form gels when combined with proteins or polysaccharides with different mechanical properties (Kumar et al., 2018). Carboxymethyl cellulose (CMC) is a derivative of natural cellulose polymers abundant in nature, and it has an INS code of E466. CMC is an inert binder and thickener used to make structures soft and smooth (Nguyen and Nguyen, 2018). The biofilm made from CMC tends to have medium durability, transparent property, soluble in water, and average oxygen barrier (Morillon et al., 2002). Gum Arabic is an extract obtained from the trunk and branches of the Acacia Senegal tree. It can be soluble in hot or cold water and has the lowest viscosity of the types of hydrocolloid gum. The GA, Gum Guar, and XG mixture provide a uniform coating and good adhesion in the wet environment (Mei et al., 2002). Glycerol and sorbitol are commonly used as plasticizers to produce films and packaging applications in the food industry from raw materials rich in protein, polysaccharide, chitosan (Cuq et al., 1995).
There have been many studies on edible starch films from various agricultural products. However, research on using whole beans after cooking in combination with the above polysaccharides for film processing has not been published. The objective of this study is to investigate the role of XG, CMC and GA polymers on the film properties with the following research contents: 1) determine the viscosity of the film-forming fluid; 2) determine physical and mechanical properties of the films; 3) initial trial of using film to wrap and preserve the dried cakes.

2. Materials and methods

2.1 Materials and chemicals

Lima bean and lablab bean (Duc Trong District, Lam Dong Province, Vietnam), after being cooked, the bean cooking water (aquafaba, it's been used in a series of other studies) the bean residues were dried at 70 ºC for about 14-16 h. Next, the dead bean residues are ground with a Phillips blender, and then the bean powder is sifted through a sieve mould with a pore size of Ø 0.3 mm. The final powder has a moisture content of 7.5 -8.8 %, Lima bean powder (LiBP) and lablab bean powder (LaBP) contained 20.34 ± 1.33 and 24.22 ± 1.43 % of protein; 3.63 ± 0.16 and 3.52 ± 0.50 % of lipid; 2.22 ± 0.068 and 1.60 ± 0.04 % of ash.
AAA rice flour was procured from Sa Dec. Inc, Vietnam. The ingredients printed on the packaging are as follows: max 12 % of moisture, 0.8 – 0.9 % of protein, 86 – 87 % of carbohydrate, 0.6 – 0.7 % of lipid.
XG, CMC, GA, Glycerol and Sorbitol were products originating from China.

2.2 The formulation for mixing biofilm-forming solution

All powders involved in the film formulation were sieved through a Ø 0.3 mm sieve. XG, CMC, and GA were collectively referred to as polysaccharides. Each biofilm-forming solution survey weighed exactly 60 g. The film-forming solutions had the composition as in Table 1.

Table 1: The formulation of the film-forming solution

Ingredients	Types of polysaccharides ((% based on the weight)								
	XG			CMC			GA		
Content	0.15 % (A1)	0.3 % (A2)	0.45 % (A3)	0.15 % (B1)	0.3 % (B2)	0.45 % (B3)	0.15 % (C1)	0.3 % (C2)	0.45% (C3)
LiBP/LaBP	2 %								
Rice flour	7 %								
Glycerol	5 %								
Sorbitol	5 %								
Water	Add enough 100 %								

The polysaccharides and water were mixed with an electric mixer (Panasonic MK-GB1WRA) at 525 rpm for 2 min. The dry flour mixture with glycerol and sorbitol was added to the solution mixture and stirred for 5 min. After that, the combination was heated on an electric stove. The temperature of the film-forming solution was about 65 – 70 °C and kept for 20 min to collect the final film-forming solution. During the heating process, it was necessary to stir well. This process did not only help the mixture of the bean powder to be mixed with the additives and helped to remove the air in the mix, making the film after drying smooth, with fewer air bubbles, and does not affect the sensory properties of the mixture film. All the biofilms were obtained by the coasting method (Saberi et al., 2016). 10 mL of fibrogenic suspensions were poured onto Petri dishes size Ø 10 cm. Coated Petri dishes were at 50 - 60 °C until constant weight (about 8 - 10 h) for the film to form. The formed films were peeled off from Petri dishes and equilibrated in a desiccator at 25 °C for about 24 h. All films were preserved in zip bags before the further examination.

2.3 Method for determining the viscosity of biofilm-forming solutions

Viscosity measurement was carried out using a Brookfield viscometer (model: HBDV-III U). 200 ml of each film suspension was gently moved to a 250 ml Becker. The probe was submerged into the suspension. The viscosity was determined at 100 rpm, 25 °C), using spindle No.2 for films from GA and spindle No.3 for films from CMC and XG. Viscosity value was recoded after 10 s of the stirrer rotation.

2.4 Method for determining the mechanical properties of films

The mechanical properties of the membranes were measured according to the procedure used by Farahnaky et al. (2013) with a slight modification: Film was fixed with tape on an acrylic mold of size (length x width = 100 x 89.5 mm). In the middle of the mold, there is a rectangle with dimensions (Length x width = 4 x 1.5 cm) Equipment (Brookfield CT3) with TA39 rod probe (2 mm D x 20 mm L) was equipped with a crosshead speed of 1 mm.s^{-1} and an initial grip distance of 75 mm. The films were placed on the rectangular mould measuring 100 mm x 89.5 mm with the starting force was on the films was 0.1 N.

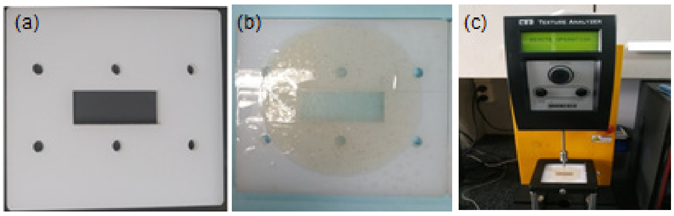

Figure 1: The images illustrate the preparation and measurement of film mechanical properties (a) Self-designed acrylic mold; (b) Fixed film on the mold; (c) Operation of the probe to film on Brookfield CT3 machine

2.5 Method for determining moisture content and film thickness

Film moisture content was determined by using a Stratorius moisture analyzer. Before analysis, films were equilibrated in a desiccator for 24 h at 25 °C. The films thickness was determined using a Mitutoyo micrometre (0 - 150 mm) with an accuracy of ± 0. 01 mm. Three measurements were randomly taken at different locations for each specimen, and the mean value was reported (Saberi et al., 2016).

2.6 Statistical Analysis

All experiments were performed in a randomized design and replicated three times. Data were presented at Mean ± SD. Analysis of variance was carried out, and the results were separated using Least Significant Difference's test ($P < 0.05$) with $\alpha = 0.05$ using Startgraphic Centurion XV. I software. The graphical construction for experiments by using Microsoft Excel 2016 software.

3. Results and Discussion

3.1 The viscosity of the film-forming solution

Figure 2a shows the difference in viscosity between the film-forming solutions. The viscosity of the CMC film-forming solutions is higher than that of the film-forming solutions from XG and GA. The reason is due to the colloidal properties of different additives that create the other mixtures. For example, CMC is easily dispersed in cold water, hot water, and alcohol. It is also a coagulant; it can form solid blocks with very high humidity (up to 98 %) (Quispe et al., 2013). The solution of XG dissolved at the right temperature will have a high viscosity. The increase of XG content made the gelatinization process was shortened and ageing earlier because the rapid aggregation of amylose led to the formation of a three-dimensional gel network (C.Kim and B.Yoo, 2006). The temperature of the solution has dramatically affected viscosity by controlling the molecules and their arrangement in the solution. The molecules of XG come in two forms: twisted and coiled depending on the dissolution temperature of the solution (Garcıa-Ochoa et al., 2000). GA has a lower viscosity than XG and CMC, and it is recommended that the concentration of GA be increased (> 12 %) to create a thick mixture (Williams and Phillips, 2009). The viscosity of LiBP tends to be higher than LaBP at the same polysaccharide ratio due to the amylose content of lima bean is more in starch than the amylose content of lablab bean amylose procedures a highly viscous solution when heated.

3.2 Physical and mechanical properties of the films

As shown in Figure 2b, the moisture content of the three types of polysaccharides added to the mixture does not have a statistically significant difference. The moisture content of the biofilms changes when there is a change in the concentration of polysaccharides such as XG, Gelatin, CMC in the film formulations, leading to a difference in the film moisture content after casting on a Petri dish (Nur Hazirah et al., 2016). Moisture content is critical in preserving food, and it is partly related to the solubility of the film when used as a packaging material. The moisture factor helps prolong the shelf life of food and protects the integrity of the product in the packaging. According to the study of Coupland (2000), the effect of glycerol on the water absorption of starch films shows that the film's moisture content would increase gradually until the water activity reached close to 0.75. When the moisture content increased, the weight of the film will increase but not significantly. Besides, the increase of plasticizer increases the moisture absorption capacity, demonstrating the hydrophilic nature of glycerol.

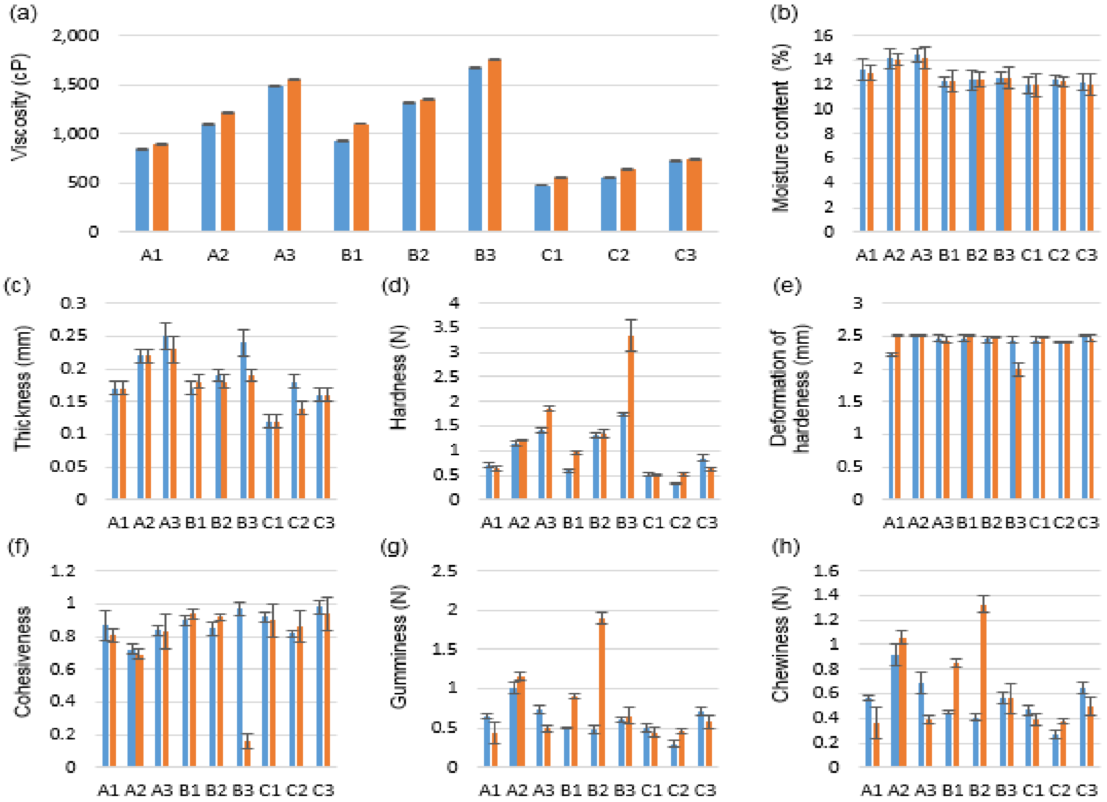

Figure 2: The charts of films properties with blue chart: LaBP; orange chart: LiBP. (a) Viscosity of film-forming solution; (b) Moisture content; (c) Thickness; (d) Hardness; (e) Deformation of hardness; (f) Cohesiveness; (g) Gumminess; (h) Chewiness

The thickness of the film depends greatly on the type and content of the polymer used. According to the study of Valenzuela et al. (2013), the thickness of the film is affected by the substances added to the film-forming solution. So that the properties of the film will increase with the addition of polysaccharides to the film-forming solution. Figure 2c shows that XG significantly increases the thickness of the film with the same polymer ratio. That can be explained based on Nur Hazirah et al. (2016) study shows the higher concentration of XG makes the denser binding of molecules in the film, the higher concentration of XG developed the higher crosslinking effect within the film matrix and help to form the compact film networkl. In the sol-gel, glycerol acts as a plasticizer that reduces the interaction of polymers to help thin films (Da Matta et al., 2011). This work fixed the glycerol and bean meal content; changed only the content of XG, CMC and GA polymers. As reported by Da Matta et al. (2011) increasing the content of XG in solution does not affect the physical and mechanical properties of the film.

Figure 2d shows the difference in the hardness between the films. Films manufactured of GA have weak strength, with C1 having the lowest hardness. Films made of CMC have the best hardness, with B3 being preferred. The hardness of B3 films created from LiBP and LaBP are 3.36 N and 1.75 N, respectively, demonstrating that films made from LiBP provide better hardness parameters than films made from LaBP. This reason can be explained the protein content of lima bean is higher than lablab bean, and the crosslinking protein chains increase resistance to film puncture (Harnkarnsujarit, 2017). The polysaccharide content affects the hardness of the film, and the higher concentration makes the hardness of the film tends to increase (Sahin et

al., 2005). The films using CMC are the best because of the inherent plasticity of CMC; this plasticity has made the film more durable and tough than the films using XG or GA.
The deformation at hardness is the study's standout feature. When the type and amount of polysaccharides used in the film compositions are changed, there is no discernible difference. When utilizing the same amount of glycerol (5 %), it is vital to evaluate the role of glycerol in causing the same deformation at the same hardness of the films. The study results by Jouki Mohammad et al. (2013) with a film made from watercress seeds mixed with glycerol came to the same conclusion.
In terms of cohesiveness, there is a substantial difference between the films. The film made from lima beans has the lowest cohesion (B3) (0.16 mm). The films from GA have a higher level of cohesion than the other films (the films from CMC and XG). The cohesiveness of the film tends to be inversely related to its hardness. There is a distinct difference in the cohesiveness of the investigated experiments. At the same proportion, the cohesion of CMC films is better than that of XG and CMC. CMC films also have a higher Gumminess and Chewiness index than the other films (from XG and GA). The films from GA have the lowest Gumminess and Chewiness index. Besides, the results in Figure 2g and Figure 2h show that the change in Gumminess and Chewiness do not follow any rules at the same rate. The films from XG give better Gumminess and Chewiness parameters at the rate of 0.15 % and 0.45 %, whereas the films from GA and CMC at the rate of 0.45 % give it's better than 0.15 % and 0.30 %. Based on the results obtained, we choose the three best films to apply in preserving cake are 0.30 % of XG with LiBP, 0.45 % CMC with LaBP, and 0.30 % CMC witLiBP.

3.3 The Initial trial of using film to wrap and preserve the dried cake

The Vietnamese snow-flaked cake (moisture of 2.35 - 2.50 %) was used for preservation application and stored for three days with a temperature of 32.0 ± 1.0 °C and relative humidity of about 67 ± 1 %. Three biofilms that could be initially considered for packaging application are presented in Table 2.

Table 2: Change of cake and film moisture content during storage time

	The values of moisture contents					
	Day 1		Day 2		Day 3	
	Cake	Film	Cake	Film	Cake	Film
A2 (LiBP)	3.01^{c}±0.02	2.32^{a}±0.25	6.55^{b}±0.60	5.25^{a}±0.22	12.06^{b}±0.88	10.03^{a}±0.81
B3 (LaBP)	2.92^{c}±0.12	2.21^{a}±0.17	3.31^{d}±0.03	2.68^{b}±0.39	3.80^{d}±0.07	4.60^{b}±0.15
B2 (LiBP)	3.56^{b}±0.12	2.08^{a}±0.37	4.02^{c}±0.04	2.82^{b}±0.39	5.12^{c}±0.13	5.38^{b}±0.34
Control 1	2.48^{d}±0.14		3.04^{e}±0.06		3.65^{e}±0.06	
Control 2	4.39^{a}±0.35		9.43^{a}±0.65		22.39^{a}±0.72	

Different lowercase letters in the same column denote significant differences ($p<0.05$). Caption: Control 1: The PE film; Control 2: The sample cake with no film.

Day 1: The data in Table 2 shows that the difference in cake moisture content is not too significant between the films, except for the PE film with the best value of 0.15 % among the four types of films used to preserve cakes. Cakes were kept by biofilm after one day, shown there is no difference significantly moisture content.
Day 2: There was a significant difference in cake moisture content between the films. The film A2 LiBP has a relatively higher moisture value than the others during storage cake. The PE film still retains its good role because it almost does not affect the moisture content of the cake. The film moisture content has no difference between B2 LiBP and B3 LaBP.
Day 3: There was still a difference in cake moisture value between the films. Specifically, the film A2 LiBP increased highly with 12.06 % shows poor moisture stability for cakes after three days of storage. Cakes were wrapped with the PE film had increased in moisture content but not significantly. Because the cake has initial moisture of about 2.2 %, the ability to absorb water back from the storage environment is speedy. The unwrapped cake sample reached the maximum value at 22.39 %.
The films with B2 LiBP and B3 LaBP give lower moisture content to the film and cake. The reason may be that CMC's structure is a linear chain with β(1→4) linkage – glucopyranose radical. The polysaccharide chain contains carboxyl groups that are hydrophilic and hydrophobic, exhibiting amphoteric properties (Su et al., 2010). The difference in film moisture is not significant between the B3 LaBP and B2 LiBP, which can be explained by the starch of lima bean than lablab bean so that the amylose content of LiBP is higher than LaBP and so lower water absorption (Zhang and Li, 2021). Because of the limited survey time, the cake was stored for three days, it was not possible to thoroughly observe the change of moisture in more prolonged conditions. However, the data showed the difference in cake moisture between the films used to preserved cakes. The film B2 LiBP reached the moisture value closest to the PE film. So that this film demonstrates the lowest back moisture absorption, it is suitable for preserving foods with low moisture content.

4. Conclusion

The roles of the three polysaccharides on the film's mechanical properties can be arranged in order from highest to lowest as follows CMC > XG > GA. With the same content of polysaccharides, the film using CMC has the best hardness, gumminess, and chewiness. XG has the effect of significantly increasing the thickness of the film, so when used with too high a concentration, it will reduce the cohesiveness and strength of the film. The appropriate XG content was recorded as 0.3 %. The strain of the films did not differ significantly with the change in the type and amount of polysaccharides. The cohesiveness of the film tends to be inversely proportional to the hardness of the film. When varying the content of polysaccharides produced a film with a significant difference in moisture content. LiBP films with the content of 0.3 % XG and 0.3 % CMC, or LaBP films with the content of 0.45 % CMC will be carefully considered in the future when applied to the preservation of specific food products.

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