



## THE PHYSICAL TESTS OF DIFFERENT FIBERS FOR EMPTY CAPSULE FILM WITH DIFFERENT CONCENTRATION OF PLASTICIZER

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### ABSTRACT

Most of our capsules shells are made from derivative of animal products which mean they are not great option for those who are muslim, Kashmir religion and vegetarian people. In recent era, vegetable capsules are new approach that can replace gelatin capsule. The present research, study the physical properties of different Fibers for empty capsule film with different proportions plasticizer. The fibers used in this research were coconut husk, oil palm trunk and paddy husk, whereas the concentration of plasticizer that were used were 0.2% w/w, 1.0% w/w, 1.5% w/w and 2.0% w/w of glycerol. The film samples that were produced in this study were analyzed by using FT-IR, DSC and dissolving test. The FT-IR results indicate that the samples contained carbonyl compound and aromatic hydroxyl. The dissolving test of films in acid and alkali solution demonstrated that the films of all samples were not dissolved in 10 minutes. As a conclusion, oil palm trunk, coconut husk and paddy husk has great potential as a capsule materials.

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### 1. INTRODUCTION

In latin world, capsule word is 'capsula' means a small box or container [1]. Yongo et al., (2016) [2] stated that capsule is a solid dosage forms in which the medicinal is enclosed within shells that made from gelatin. The shells are usually formed from gelatin however; they also may be made from starch or other suitable substances. The proposed of capsule is to be swallowed whole by the patient [3]. It can be of liquid, solid or semi solid type and can be overseen orally, rectally or vaginally. Capsule also capable for providing protection to encapsulated drug from oxidization made by atmospheric oxygen, light, moisture etc. owing to the barrier effects of [4].

Gelatin is a biopolymer which is a translucent brittle solid, colorless, tasteless and odorless. The gelatin is derived from collagen extracted from various animal body parts such cattle bone, horse, fish, pork and bovine. The physical of gelatin is translucent,

colorless, brittle and flavorless foodstuff. From previous study, it state that the gelatin is an irreversibly hydrolyzen from a collagen. According to the [5], gelatin is a mixture of peptides and protein that can produces by a hydrolysis of extraction collagen from animal body part such as skin, bones and tissues. Basically the animal used in gelatin are fish, cattle bone, horse, chicken and the most used in gelatin is pigs and bovine. Based on the [6], structure of gelatin contains almost entirely of amino acids in L-configuration with traces in D-configuration linked by amide (peptide) bonds forming a linear polymer with a molecular weight ranging from 15,000 to 250,000 Daltons.

However, the consumption of gelatin capsule have several disadvantages, like in the previous researcher state that it give allergic effect for some individual people, potential toxicity, highly hydroscopic nature resulting in storage difficulty and limitation in hygroscopic preparation [2]. Meanwhile (Majee et al., 2017)[4] stated that the religious perspective may became an issues of gelatin from animal source where it may be concern for some religious who forbid the use of animal product and some vegan or vegetarian people. The cross linking occurs when reacted to the aldehydic end product of degradation resulting excipient interaction formation of insoluble skin or pellicle on the gelatin shell as well as cross linking of gelatin that due to formaldehyde produces as result of the decomposition of lactose. Gelatin also has limited compatibility with some liquid and gels, and gives allergic reaction to some consumer [5]. Hence, the green capsule is produced as an alternative these issues. The green capsule is synthesis from hydroxyl propyl methyl cellulose (HPMC), starch, pullulan and [1].

According to the [4] the alternative capsule shell material should be preferably of plant origin, should not undergo cross-linking reactions with various excipients, should be stable towards fluctuation of environmental temperature and humidity during production and storage, should not exhibit temperature-dependent dissolution and should be able to contain any fill material.

## **2. MATERIAL AND METHODS**

### **2.1. Materials**

Fibers obtained from different agriculture waste. Oil pump trunk, paddy husk and coconut fiber were selected for the study. These fibers were selected because they are

among common agriculture waste generate in Malaysia. Oil palm trunk were collected at Felda Jengka 3, Pahang, coconut husk at Cherating and paddy husk at Kedah.

## **2.2. Preparation of Extraction**

The extraction of polysaccharides from oil palm trunk, coconut fiber and paddy husk were involved by soaking them into high cooking pressure at 50 kPa about one hour to squeeze out the starch from dried fiber. Then the extractions were used in this research and the waste of fiber can be used several times by drying it in the room temperature.

## **2.3. Preparation of Capsule Film**

3% (w/w) of methyl cellulose was added in 85% (w/w) of extraction polysaccharides beaker (oil palm trunk, coconut fiber and paddy husk) and blend together using magnetic stirring. The amounts of methyl cellulose 3% (w/w) were added in small amount to prevent the mixture became clumpy. Stir the methyl cellulose until it blends well. Different amount concentration (0.2%,1%, 1.5% and 2%) w/w of plasticizer (glycerol) was added into the solution and stirred for 15 minutes to get better homogeneity. After that, 2%w/w of tamarind seed polysaccharides was poured into mixture with continuously stirring. Finally, pour 10%w/w of cornstarch into the beaker, stirred it until became jelly liquid suspended. In these preparation, the bubble were appeared thus, discard the bubble using spatula and while it still hot, cast over the suspended onto the petri dish and dry it in room temperature.

### 3. RESULT AND DISCUSSION

#### 3.1. Effect of plasticizer on film

**Table 1:**The observation of plasticizer on film appearance

Fibers	Concentration of Glycerol (%)			
	0.20	1.0	1.5	2.0
Oil palm trunk				
Paddy husk				
Coconut Husk				

Around 5 films sample were observe by their appearance. Generally all the appearance of all plasticizers with different extraction was translucent yellowish brown color. All the films were smooth surface except coconut fiber with concentration of plasticizer 2.0% w/w the observation of 0.2% w/w concentration of glycerol was brittle and the at 2.0%w/w concentration it became flexible and soft surface. Thus, when more concentration of plasticizer was pour, the more flexible of the film appearance. According to the [7], at lower concentration, the film were brittle. It was due to the macromolecular properties that affected by the plasticizer. The color of the film was affected by the gelling agent, tamarind seeds polysaccharides extract, and which was a brown color powder and the concentration give effect to the film appearance.

The plasticizer effectively reduced internal hydrogen bonding while increasing intermolecular spacing thereby decreasing brittleness and increasing permeability of the film materials. Higher concentration bind with starch molecules is attached polymer chains and becomes weakened resulting the film more flexible [7].

There are some cracks were observed on surface of the sample film. The crack appeared when the film was dry at room temperature. It is because the rapid cooling, when the

jelly suspended was dry at the petri dish, the temperature of the jelly suspended was change from hot to room temperature. Thus, it gives the film crack result.

### 3.2. Difference Scanning Calorimetry (DSC)

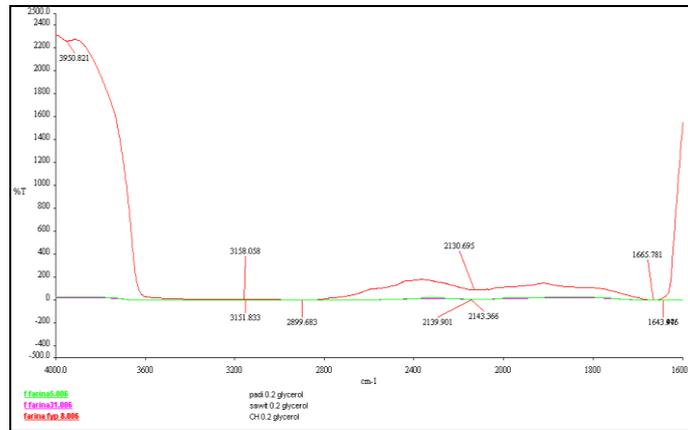
**Table 2:** The value of Tg containing different concentration

Concentration Of glycerol(%)	fibers	Tg(°C)	Tg (min)
0.2	Oil palm trunk	84.09	3.11
	Pady husk	97.55	3.61
	Coconut husk	82.65	2.97
1.0	Oil palm trunk	75.91	2.56
	Pady husk	95.82	5.42
	Coconut husk	90.84	3.84
1.5	Oil palm trunk	83.17	3.23
	Pady husk	87.28	3.06
	Coconut husk	73.57	2.55
2.0	Oil palm trunk	101.15	6.08
	Pady husk	90.10	3.33
	Coconut husk	95.39	3.16

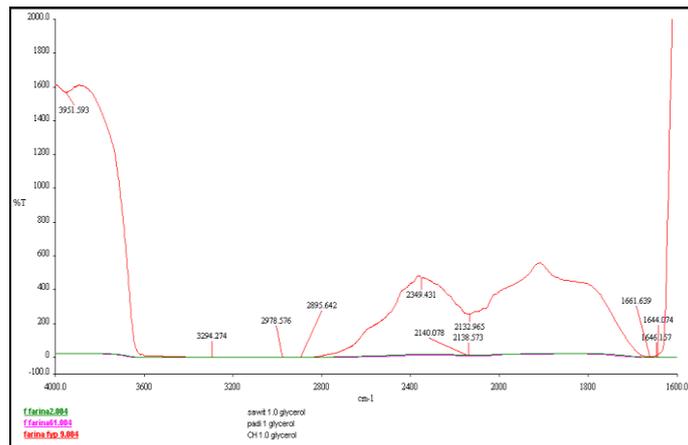
The degradation temperature of film and effect of glycerol concentration in different fiber were studied by DSC. A single endothermic transition occurs and the heating rate was 20K/min. The plasticizers of the peak were analyzed and the initial temperature of onset, delta cp ( $Jg^{-1}K^{-1}$ ) and also peak ( $^{\circ}C$ ) were measured. The Figure 4.3 shows result graph of different plasticizer in and Table 4.3 shows the value of Tg containing different concentration of plasticizer. The glass transition, detected at very low temperature than hard gelatin however, the 2.0% w/w of OPT was slightly higher than gelatin. This is may be due to the presence of highest crystallinity in the sample. The gelatin capsule have peak that almost straight line compare to others that have broad peak but different degradation temperature.

### 3.3. Fourier-transform infrared spectroscopy (FTIR)

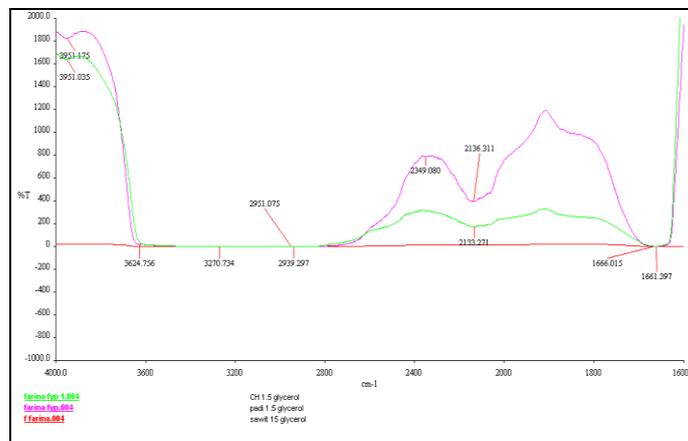
a)



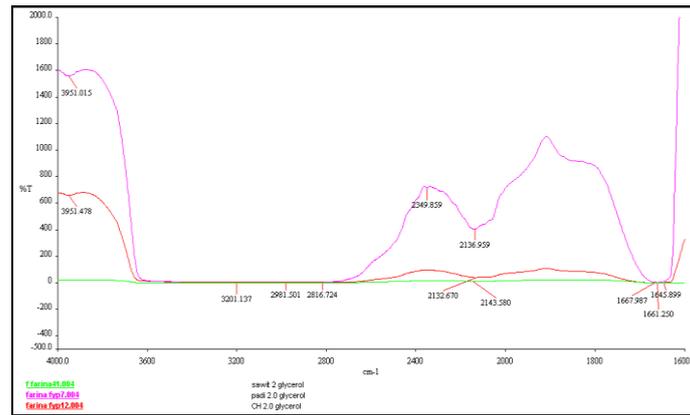
b)



c)



d)



**Figure 1** FTIR result of different fiber contains with different proportion of glycerol. a) 0.2% glycerol of different fiber b) 1.0% glycerol of different fiber c) 1.5% glycerol of different fiber d) 2.0% glycerol of different fiber.

Fiber films were run in FTIR spectroscopy to determine the functional groups that are present in the film capsule. Figure 3 shows the FTIR spectra of different concentration glycerol with different fiber film. The wavelength was observed at  $4000\text{cm}^{-1}$  until  $1600\text{cm}^{-1}$ . In figure 1 (a), many of the observed bands appeared in the fingerprint region which the wavelength number between  $4000\text{cm}^{-1}$  -  $1600\text{cm}^{-1}$ . The samples were produce similar spectra but different peak. The most intense peak in figure 3.3 (a) is coconut husk (CH). Where are broad peak at  $3950.821\text{cm}^{-1}$  has shown the possible involvement of substitute cyclopropane. The N-H stretch in amide was observed at the fingerprint  $3151.833\text{cm}^{-1}$  it also shown at  $1665.781\text{cm}^{-1}$ . This peak was involved of carbonyl compound. At the peak  $2898.17\text{cm}^{-1}$ , the peaks were slowly stretching due to the presence of strong aldehydes  $\text{CH}_3$ . The characteristic of alkynes  $\text{C}\equiv\text{C}$  at the broad peak is at  $2143.528\text{cm}^{-1}$  -  $2139.969\text{cm}^{-1}$  was observed. The broad peak at  $2143.366\text{cm}^{-1}$  -  $2130.695\text{cm}^{-1}$  which could arise the weak alkynes compound.

For the figure (b), the peaks have some similar peaks however at 1.0% glycerol of Coconut Husk (CH) was slightly different with other peaks where it has broad stretching peak from  $4000\text{cm}^{-1}$  to  $3600\text{cm}^{-1}$ . It may because the structure of the aromatic hydroxyl was strong than the others starting at  $3616.033\text{cm}^{-1}$ . Strong O-H was broadly stretching at the fingerprint  $3294.274\text{cm}^{-1}$ . This was due to the O-H stretching of hydroxyl groups in

polysaccharides and water [8] which was present in aliphatic of aromatic and present in their main component [9]. The peaks were broadly stretching until  $2895.642\text{cm}^{-1}$ . At this region, strong C-H alkenes were observed. This band shifted from  $2349.420\text{cm}^{-1}$  in CH indicating an increase in the intermolecular hydrogen bonding. The FTIR spectra of PH and OPT were similar, indicating that asymmetric stretching. The peaks at  $2349.420\text{cm}^{-1}$  were observed of C-H stretch from aliphatic saturated compound. The present of aliphatic compound shows that the content of glycerol in gelatin films at the highest glycerol concentration is high enough to exhibit a signal in an infrared spectrum. Two broad bands in the region  $2394.08\text{cm}^{-1}$  to  $2136.311\text{cm}^{-1}$  were shown the strong C-O peak.

In the figure 1 (c) and (d) is shows the functional group of 1.5% and 2.0% glycerol with different fiber. At the peak around of  $3634.065\text{cm}^{-1}$  have observe strong aromatic hydroxyl. Band was shifting around the peak at  $3624.886$  involving hydroxyl groups. The asymmetric stretching peaks were involving C-H stretching at  $327.810$ . The C-H stretching causes the vibration of CH, CH<sub>2</sub> and CH<sub>3</sub> [10] observed band at  $330\text{cm}^{-1}$  and  $2900\text{cm}^{-1}$  and stretched the O-H and C-H link. The major changes can be seen are at the increment O=C≡O of carbon dioxide at  $2348.839\text{cm}^{-1}$ . The peak around  $1640.030\text{cm}^{-1}$  corresponded to the C=O stretching that may be attributed to the hemicellulose and lignin aromatic groups, [11]. The stretching C=C indicate alkene groups of aromatic groups.  $\alpha$ - $\beta$  unsaturated were present at the region  $1667.987\text{cm}^{-1}$  which was composition of oil palm trunk.

### 3.4. Dissolving Test

**Table 3:** Dissolving test in hydrochloric acid (Hcl) and distilled water

Fiber	Concentration of glycerol	In distilled water
Gelatine	Fully dissolve	Fully dissolve
Oil palm trunk	Not dissolve	Not dissolve
Paddy husk	Not dissolve	Not dissolve
Coconut husk	Not dissolve	Not dissolve

In table 3.4 shows the result of dissolving test film in Hcl and water. The sample film were cut at size 1cm x 1cm and soaked in 50 ml of 0.1M Hcl and water at constant

temperature,  $37\pm 2^{\circ}\text{C}$  for 10 minutes. The solution of HCl is represented acid in gastric fluid. Meanwhile, the constant temperature,  $37\pm 2^{\circ}\text{C}$  is represented the human body temperature. From the result, the sample films were not completely dissolved in 10 minutes in 50 ml water or in 50 ml HCl (pH 1.2) solution. This result was similar with previous researcher [12]. This result show that the film from different fiber cannot dissolve in gastric fluid.

However, hard gelatin capsule was dissolved rapidly and completely dissolved within 10 minutes of testing time. In a meantime, the color of solution was change into pink solution due to the colorant of gelatin capsule. Gelatin capsule contained a high percentage of moisture content than film fiber, thus the moisture can be released and cause easily dissolving [13].

#### **4. CONCLUSIONS**

In this research that have been conducted, the objective to analysis the best percentage of plasticizer use in fiber film were achieved. It can conclude that, the glycerol molecules interact with the fiber film and the more concentration can create the strong bonding fiber also a decrease in rigidity of the film. Furthermore, the film became more flexible as the plasticizer concentration increase. Thus, the best percentage of glycerol can used was 2.0% (w/w).

According to the FTIR result obtained, the functional group that appeared in fiber film mostly was carbonyl compound and aromatic hydroxyl. This is due to the mixing of others natural excipient such as tamarind seed polysaccharides. However, the mixing with tamarind seed polysaccharides gives an effect of the fiber film whereas after 3 month of producing film, microorganisms were appeared. In the term of dissolving test, this work proved that samples were not dissolved in both acid and slightly alkaline in 10 minute.

From the observed results, this work shows that all the fibers have a potential result for an alternative of gelatin capsule based on their performance and physiochemical.

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