Drying Characteristics of Starch-Gelatin Edible Films Plasticized with Glycerol

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***Abstract*— The aim of the experiment is to find out the drying characteristics of starch-gelatin films containing glycerol plasticizer by using drying characteristic analysis. Since the uncontrolled problem of waste management in Malaysia with many types of waste being produce, edible films is one of the alternative ways to reduce the waste because edible film is a one of biodegradable products. The potato starch-gelatin was prepared by casting the film-formation into petri dish and dried temperature 45, 50, 55 and 60ºC. Then the results obtain was plotted. The drying kinetics models was used to find out the best drying temperature and drying models for this experiment. As the results, the best drying kinetic model is Newton model at 45ºC.**

***Keywords*— Starch-gelatin film, Edible film, Modelling of Kinetic Drying, Drying Characteristics, Glycerol Plasticizer**

I. INTRODUCTION

The purpose of edible films is to reduce the waste being produce by community or even in industry [1]. The usage of plastics has been increasing day by day because of the increasing population of human in the world. Due to this problem, edible films is one of invented biodegradable product to reduce landfilling. Interest on biodegradable product arises since it can decrease the life span of products and decomposed by bacteria [20]. To improve the quality and economic starch-gelatin film product, a few of research and experiment was done.

These edible films and coating were used in foods product such as fruits to prevent moisture loss. On 1967, the commercial use of edible films is very little. 20 years later, there are increasing of company on business of edible films. Then another 10 years later, about 600 companies on business with edible films [2]. This shows that the films is important and useful in the industry as time continues.

Many effort was done to develop green product or environmental friendly product which is the most popular topic lately. Human nowadays are more alert on how climate changes happen and world are more pollute compare to few years ago. So then, they desired to produce more biodegradable product to save the Earth.

In this experiment is about starch-gelatin films. Starch in under types of polysaccharides films alongside with cellulose, chitin and chitosan film [3]. This types of films has low caloric content and oily surface. However, the permeability to water vapor is high so the films is poor barrier to water vapor [9].

Then, gelatin is derived from the partial degradation of collagen. Many industry choose gelatin film compare to the other types of film because of gelatin film easy to find which abundance, low cost needed, good forming ability with any types of plasticizer and biodegradability [4]. Based on the experiment, the gelatin film is in transparent form which is good oxygen and carbon dioxide barrier [5].

To improve the weakness properties of starch, gelatin was added to the starch film-forming solution. Hence, according to the Al-Hassan and Norziah (2012) when the protein predominates in the blend of polysaccharide and gelatin solution resulting good interaction between these two types of films [10].

To enhance the properties of edible film, the plasticizer is added to the film forming solution before drying the film. Plasticizer is a component where it is low molecular weight in purpose to reduce the brittleness of film. The plasticizer act as strengthen of the gelatin film by increase the polymer chain, flexibility of the film. Then it also decrease the intermolecular attraction force of film [6, 7].

There are many types of plasticizer. Currently researcher are more focus on non-toxic plasticizer to produce biodegradable film. The 'green' plasticizer has gain attention among the researcher because of the non-toxic properties even economic friendly to the environment. Moreover, based on previous study the plasticizer used for their film forming by using glycerol and plasticizer which is also natural based plasticizer [8].

The analysis of drying characteristics is important to characterize the drying materials [11]. There are several drying kinetic models that being used by previous study such as Page, Logarithmic, Newton, Henderson and Pabis models. The models were analyze to determine the best temperature for this study [12].

The target in this study is to prepare the starch-gelatin edible films at different temperature which is 45, 50, 55 and 60ºC and analyzed the drying characteristics to determine the best drying temperature. The drying kinetic used is Newton, Page, Pabis and Henderson models.

II. METHODOLOGY

A. *Materials*

Gelatin powder and glycerol from chemistry laboratory. Potatoes blend for starch source of films.

B. *Film Preparation*

The preparation of film forming solution by added 3 g of gelatin powder to the beaker. Then added 100 ml distilled water into the beaker containing gelatin powder. The solution then stirred using magnetic stirred at 90ºC. The solution continued stirred for 30 minutes. After 30 minutes, 1.5 mL of glycerol and 1.5 mL of potato juice is added. The solution then stirred for another 10 minutes.

Next 5mL of well-mixed solution was poured into the glass petri dish. The solution was dried at temperature 45, 50, 55 and 60ºC by using universal oven. The film solution was weighted first before drying started to record initial weight of film. Then the film was dried until reached constant weight.

C. Modelling of Drying Process

The weight was recorded to calculate moisture content at every hours. The equation of moisture content using as following equation 1 [13]:

$$\text{MC dry basis (g/g db)} = \frac{Wt - Wd}{Wd} \quad (1)$$

Where;
Wt = Weight at t (g)
Wd = Weight of dry (g)

The formula of drying rate shown as below [12]:

$$\text{Drying rate (g/g.hr)} = \frac{W_{t+1} - W_t}{W_d\,(\Delta t)} \quad (2)$$

Where;
W_{t+1}= Weight at t+1 (g)
W_t = Weight at t (g)
Wd = Weight of dry (g)
Δt = Interval of time (hr)

After calculated the drying rate, the graph of drying rate versus time was plotted to determine the trend of drying rate. The correlation coefficient (R2), chi-square and RSME formula is use to determine the result of the experiment. The best drying characteristics will show is when the highest value of R2 that approximately to 1. Then, the lowest number of chi-square and RSME were chosen as best drying temperature for the films.

$$R = 1 - \frac{\sum_{i=1}^{N}(MR_{pre,i} - MR_{\exp,i})^2}{\sum_{i=1}^{N}(MR_{pre} - MR_{\exp})^2} \quad (3)$$

$$RMSE = [\frac{1}{N}\sum_{i=1}(MR_{\exp,i} - MR_{pre,i})^2]^{1/2} \quad (4)$$

$$\chi^2 = \frac{\sum_{i=1}^{N}(MR_{\exp,i} - MR_{pre,i})^2}{N-n} \quad (5)$$

Where N is the number of the observations. $MR_{exp,i}$ and $MR_{pre,i}$ are the actual and predicted moisture ratios respectively. Then n is the number of constants in the model [14].

The models used in the experiment are shown in table 1 [13]:

Table 1: Summarize of mathematical models with their equation

Model	Equation	Slope	y-intercept
Newton	MR = exp(-kt)	-k	-
Page	MR = exp(-kt^n)	n	ln k
Henderson and Pabis	MR = a.exp(-kt)	-k	ln a

Based on table 1, the drying constant and coefficient resulted from the graph plotted. The values of x-axis and y-axis was plotted based on the arrangement for each model's equation as linear equation.

After that, the graph of predicted MR versus experimental MR was plotted to compare the accuracy of the experimental result. The values of RMSE and chi square determine using the equation 4 and 5. All the results was tabulated and compared to choose the best drying model based on values of correlation coefficient, RMSE and chi square.

III. RESULTS AND DISCUSSION

A. Drying Characteristics of films

The figure 1 shows that the graph of moisture content dry basis versus drying time. The dry basis moisture content is to describe the changes of moisture during drying. When the moisture changes, moisture content of dry basis changes and the weight also changes [15].

The drying temperature effect significantly toward moisture content to be constant. Figure 1 shows the graph of moisture content dry basis over time. The highest initial moisture content dry basis was started at similar values of moisture content which is around 25 to 20 g/g dry basis. Then from the figure 1 shows that the trend of moisture content at 60^{o}C steeper compare to other temperature. The fastest time taken for moisture content to constant is 2 hours at 60^{o}C and longest time taken is 10 hours at 45^{o}C. Similar behavior of the graph discovered by Carrion F.P. et.al (2009) found that the higher the temperature the faster moisture content to become constant [16]. The relationship shows that the drying time to reach constant moisture content significantly affect with drying temperature.

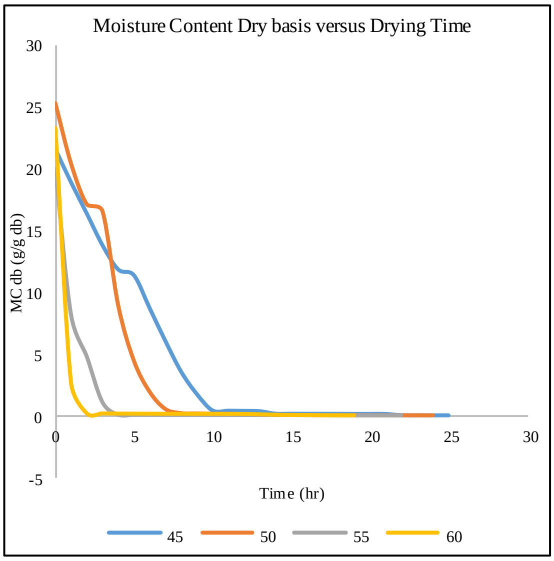

Fig. 1: Graph of moisture content versus time

Figure 2 shows the graph of drying rate for each temperature versus moisture content. The drying rate decrease throughout the drying period. Similar observation shown by Saxena D, et al, (2012) where the drying rate increase as time increase on sweet potato starch based films [17]. There are two different periods in drying processes which is constant rate period and falling rate period. Falling rate period took place for drying rate of 60^{o}C. The constant rate period is the period when moisture content are evaporating from the surface of the films by diffusion [18]. The moisture is evaporate to the surface at a rate sufficient to maintain saturation at constant rate period. The presence of constant rate period also because of presence of free surface water [19]. There are no surface evaporation took place at constant rate cause no constant rate period . It can conclude that during drying of films, the mass transfer of moisture took place predominantly by liquid diffusion [18]. The figure 2 shows the drying rate for each temperature versus moisture content graph.

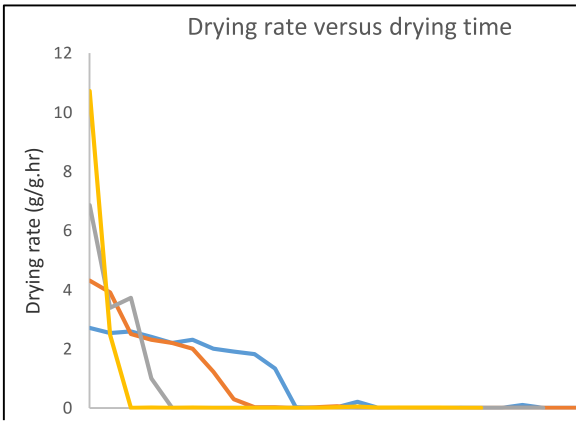

Fig. 2: Graph of drying rate over time

B. *Drying Kinetic Modelling*

The drying models that fitted into experimental data were Lewis, Page and Henderson and Pabis models. The graph of these three models was plotted as in figure 3 to figure 5 to find out the drying coefficient based on the linear and exponential equation at 45°C.

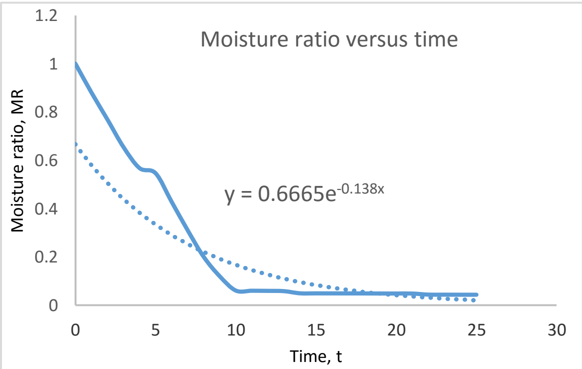

Fig. 3: Graph of MR versus t based on Newton equation for 45°C

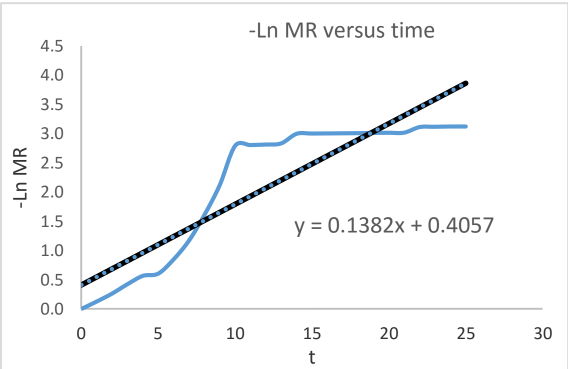

Fig. 4: Graph of –Ln MR versus time based on Page model at 45°C

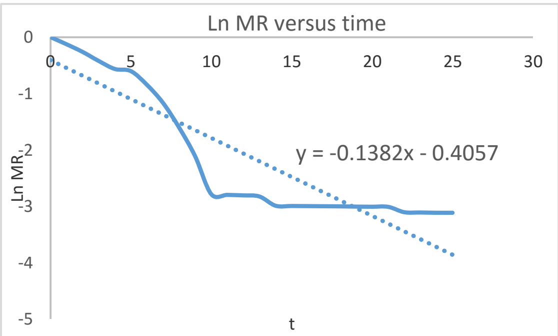

Fig. 5: Graph of Ln MR versus time based on Henderson and Pabis model at 45°C

The similar graph was plotted for 50, 55 and 60°C. Table 2 shows the constant and coefficient of drying at different temperature.

Table 2: Values of drying coefficient at different temperature and models

Model	Temperature	Drying constant	Drying coefficient
		k	n/a
Newton	45	0.1380	-
	50	0.1290	-
	55	0.1993	-
	60	0.1042	-
Page	45	1.5004	n =0.1382
	50	2.7030	n =0.1286
	55	5.0168	n =0.0704
	60	9.5927	n =0.0665
Henderson and Pabis	45	0.1382	a =0.6665
	50	0.1280	a =0.3699
	55	0.0704	a =0.1993
	60	0.0665	a =0.1042

The drying coefficient then be used to find out predicted moisture ratio. After predicted moisture ratio was calculated, the RMSE, SSE and $\chi 2$ was calculated. Table 3 shows the statistical results for three selected models. The models were chosen based on the closeness of value correlation coefficient to 1. Then $\chi 2$ and RMSE is better close to 0 shows good short term performance.

Table 3: Statistical results for drying kinetic models at varies temperature

Model	Temp e-rature	R^2	RMSE	SSE	X^2
Newton	45	0.9666	0.2257	0.0506	0.0509
	50	0.8766	0.2629	0.0691	0.0691
	55	0.6455	0.1259	0.0158	0.0173
	60	0.5990	0.1998	0.0399	0.0399
Page	45	0.9473	0.1668	1.8684	0.8684
	50	0.8309	0.0669	1.1383	0.1444
	55	0.7450	0.0374	1.0764	0.1781
	60	0.5328	0.5324	2.3484	0.0336
Henderson and Pabis	45	0.9343	0.2255	0.0508	0.0522
	50	0.9123	0.2623	0.0688	0.0691
	55	0.6777	0.1248	0.0155	0.0171
	60	0.4290	0.2003	0.0401	0.0412

Based on table 3, the most suitable drying kinetic model is Newton model. The results shows that the values of R2 is closer to 1 compared to Page and Henderson and Pabis model. The highest value for R2 is 0.9666 at 45°C. The lowest values of RMSE and $\chi 2$ is Newton model compare other two models which is 0.1259 and 0.0173 respectively. Hence, Newton model is chosen as most suitable drying model for process of drying gelatin film.

The results shows best drying kinetiv model is Newton model with equation $MR = e^{-0.1380t}$. The highest values of R2 is 0.9666 at 45°C. Compare to the values of R^2 at other temperature, the R^2 at 45°C is the highest and has value more than 95%. It can be conclude that the 45°C is the most suitable temperature for drying gelatin film

IV. Conclusion

As conclusion, the fastest time taken for films to dry is shorter at higher temperature. The moisture content decrease as the drying temperature increase. For drying rate, the region of falling rate period followed by constant rate period was observed. The drying temperature of 45°C is the best temperature as the drying rate at that temperature has best manner and curve compare to other three temperature. Then, the best drying model for the films is Newton models as the values for R^2 is highest and RMSE is the lowest.

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