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Moisture sorption isotherm of natural condiment cube made from juice water of *katsuobushi* plant, coated with edible coating and stored at ambient temperature

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Abstract. This research aimed to study the MSI of natural condiment cube coated with an edible coating, using the standard gravimetric method. Condiment pasta was performed into small cubes about 15 mm wide, dried, and coated with an edible coating made from carrageenan flour. Uncoated cube was used as a control sample. Samples of natural condiment cube were placed in a series of desiccators represented of a range of RH's, and allowed to equilibrate, indicated by constant weight of samples. Water content of equilibrated sample was then measured as equilibrium moisture content (EMC). Five models of the equation were used in this experiment to fix Moisture sorption isotherm (MSI) data. The results show that natural condiment cube coated with edible coating has lower water content than control. The equilibrium moisture contents of natural condiment coated with edible coating ranged between 7.0 and 76 % dry basis. Henderson models showed best-fit values of regression coefficients with mean relative determination (MRD) of 8.21, and moisture sorption isotherm curved were sigmoid. In conclusion, edible coated were effectively used as a coating of natural condiment cube, stored in ambient temperatures only in RHs lower than 75%.

Keywords: *katsuobushi*, natural condiment cube, moisture sorption isotherm

1. Introduction

One of the important factors influence properties of food such as physical, chemical and organoleptical, is water content. Water activity (a_w) can be described as a ratio between the water vapor pressure of water in food and the water vapor pressure of pure water, at the same temperature. Can be easily explaining as free water content in a food that can be used for biological activities, such as microbial or enzymatic activities [1]. Moisture sorption isotherm usually presented as a curve of relationship between equilibrium moisture content (EMC) as dependent variables and a_w as independent variables, for a certain temperature [2], and used for a number of purposes in food research especially for intermediate moisture food (IMF). It includes estimation for drying time, ingredient formulation, packing, and moisture changes during processing and storage, and the shelf life stability [3, 4].

Efforts to minimize environmental pressure have been developed using biodegradable materials for packaging. For example, application of edible films as coating material for some fresh fruits and vegetables, to retain quality and prolonged shelf life [5, 6]. The edible films function is also to minimize

the loss of water from food during storage [7]. However, during storage the changes of relative humidity can influenced the permeability of edible film [8]. Moisture sorption isotherms can be used also to analyze water binding capability of edible film [9]. Starch is example of one common promising biopolymer materials that are, available in abundant quantity, biodegradable, affordable, renewable, and more environmentally friendly [10]. Various studies of the starch-based film have been carried out. Information about moisture sorption characteristics of starch base film, such as cassava [11], pea [12], sago [13], rice starch [14] are widely available, there is lack of information on MSI of Carrageenan coated of natural condiment cube. Carrageenan starch is one of the potential materials for making edible films [15].

Condiments or seasonings can be provided from the mixture of ingredients and other, fermented or unfermented and used to alternate flavor or aroma to foods and beverages, and can improve the taste of monotonous diets [16]. Dried smoked skipjack or *Katsuobushi* is a popular type of smoked fish in Asia and widely used in traditional Japanese cuisine. There are five *Katsuobushi* industries established in North Sulawesi Indonesia and produce 50,000 to 75,000 liters of juice water every two days as by-product. This juice contains a lot of amino acids and omega-3 fatty acids as well and considered as a new source of ingredient and flavoring agents, instead of wasted and created pollution [17]. One of the food current issues is highly dependency of people especially China and Indonesia on flavoring products although some of the products are still controversial. Therefore, producing of natural condiment as an alternative is needed by utilizing high glutamic acid-containing juice. The objective of this study are to prepare and determine the moisture sorption isotherm of natural condiment cube coated with edible coating and stored at ambient temperature.

2. Materials and Methods

2.1. Materials
The juices used as a raw material of natural condiment were obtained from *Katsuobushi* plant of Cellebes Mina Pratama Company in Bitung. The ingredients were purchased from a local market, while the chemical reagents i.e. sodium hydroxide (NaOH), lithium chloride (LiCl), magnesium chloride (MgCl$_2$), potassium carbonate (K$_2$CO$_3$), magnesium nitrate (Mg(NO$_3$)$_2$), sodium nitrite (NaNO$_2$), sodium chloride (NaCl) and potassium sulfate (K$_2$SO$_4$), were purchased from Merck, Indonesia. All the reagents are analytical grade.

2.2. Condiment Preparation
Juice water from the *Katsuobushi* factory was taken from Celebes Mina Pratama *Katsuobushi* Factory at Bitung. The juice water transported to Fisheries Products Processing Technology laboratory at Sam Ratulangi University for about 90 minutes by car. During transportation, water juice bottles was cooled using ice in a cool box. In the laboratory, the juice was filtered, and mixed with corn flour and salt, and then homogenate and evaporated using mixer vacuum evaporator at 60°C, for about two hours, until the texture became pasta. The formula of juice and corn flour was 5:1 (v/w). The raw condiments were prepared separately, consisted of onion, garlic, mushroom, tomato, and salt. Each raw condiment was blended, extracted, and then immediately mixed together with formula tomato: onion: garlic: mushroom: salt was 4:2:2:4:2. The juice water pasta and the mixed condiment were then mixed together, and evaporated in mixer vacuum evaporator for one hour until the desired texture of cubes was easily made. The final product is called natural condiment cube (figure 1).
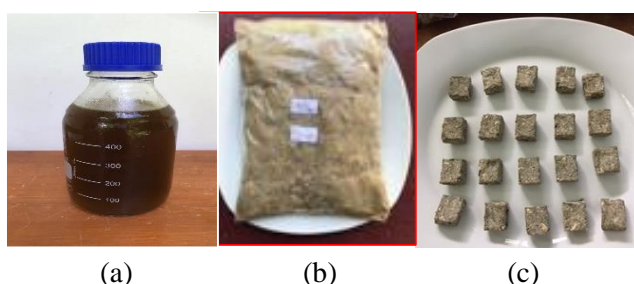
<center>(a)   (b)   (c)</center>

Figure 1. Juice water (a), natural condiment pasta (b) and natural condiment cube (c).

2.3. Coating preparation

Edible coating solution was prepared followed previous research with minor modification [18]. Carrageenan powder (0–2% w/v) was dissolved in distilled water, heated at 70°C while stirred using magnetic stirrer (Favorite HS0707V2 model) for 10 min. The solution was carefully adjusted to pH 5.6 by the addition of NaOH and Glycerol (0–2% v/v). The solution was then topped up to 500 ml. Furthermore, bee wax (0.2% v/v) as modification of the formula, was added and the solution was homogenized using a homogenizer (Wise Stir HS-100D) at 3,000 rpm for 5 min. For treatment samples, the fresh natural condiment cube samples were coated by dipped in the coating solution for 2 min. The coated samples were then dried for 2 h, at ambient temperature with RH of 60-70%. Samples were filled in polyethene bags and stored at a storing cabinet with ranged temperatures of 28±3°C, until analyzed time.

2.4. Moisture sorption isotherm

The EMC was determined using standard gravimetric method by placing the natural condiment cubes into constant relative humidity environment created by a saturated solution of a particular salt at ambient temperatures. Saturated salt solutions provided were: NaOH, LiCl, $MgCl_2$, K_2CO_3, $Mg(NO_3)2$, $NaNO_2$, NaCl, KCl and K_2SO_4, to maintain a respective relative humidity–(RH) inside separate vacuum desiccators in the range of 6.9; 11; 32; 44; 53; 64; 76; 87; and 97% [19]. The natural condiment cubes samples were allowed to reach EMC, indicated by a constant weight of the samples. The equilibrium moisture content of the samples with replication were determined by oven drying method at 105°C for 24 h [20]. Each treatment was done in two replication, and data were represented as mean value.

2.5. Water sorption isotherm curve modeling

Five mathematical MSI models were chosen to describe the MSI curves (table 1).

Table 1. Five models used in describing MSI curve of natural condiment.

Models	Form Equation	
	Form Linear	Linerization (Y = a + bx)
Hasley[ac)]	$Log(\ln(1/a_w) = \log P(1) - P(2) \log Me$	$Y = Log(\ln(1/a_w); a = \log P(1); b = -P(2)$ $x = Log Me$
Chen Clayton[bc)]	$Ln(\ln(1-a_w) = \text{lon} P(1) - P(2) Me$	$Y = Ln(\ln(1-a_w); a = \text{lon} P(1); b = -P(2)$ $x = Me$
Henderson[cd)]	$Log(\ln(1/(1-a_w) = \log K + n \log Me$	$Y = Log(\ln(1/(1-a_w); a = \log K; b = n$ $x = \log Me$
Caurie[bc)]	$Ln Me = Ln P(1) - P(2) a_w$	$Y = Ln Me; a = Ln P(1); b = -P(2)$ $x = a_w$
Oswin[ac)]	$Ln Me = \ln P(1) + P(2)(a_w/(1-a_w))$	$Y = Ln Me; a = \ln P(1); b = P(2)$ $x = (a_w/(1-a_w))$

Source: [a)] [21] [b)] [22] [c)] [23]

The most appropriate model was indicated by mean relative determinant (MRD) values. Equation (1) was used to determine MRD:

$$MRD = 100/n \sum_{i=1}^{n} \left(\frac{Mi - MPi}{Mi} \right)$$

<center>(1)</center>

Notes: Mi = Levels of experimental results, MPi = water content calculation and n = number of data. MRD lower than 5 indicates that the MSI model is good describe the actual data, the value of 5<MRD< 10, indicates the MSI model describes the state of approaching the truth, and MRD >10, the model cannot describe the actual data [22].

3. Results and Discussion

The moisture content of natural condiment was 40-50% for uncoated condiment and 54-60% for a coated condiment. Five models MSI curve of natural condiment cube uncoated and coated samples are presented in figure 2 and 3. The equilibrium moisture content (EMC) of natural condiment cube achieved during 7 days of storage in RHs ranged 6.9 – 97%. Samples placed in higher RHs showed that EMC were reached in more longer time.
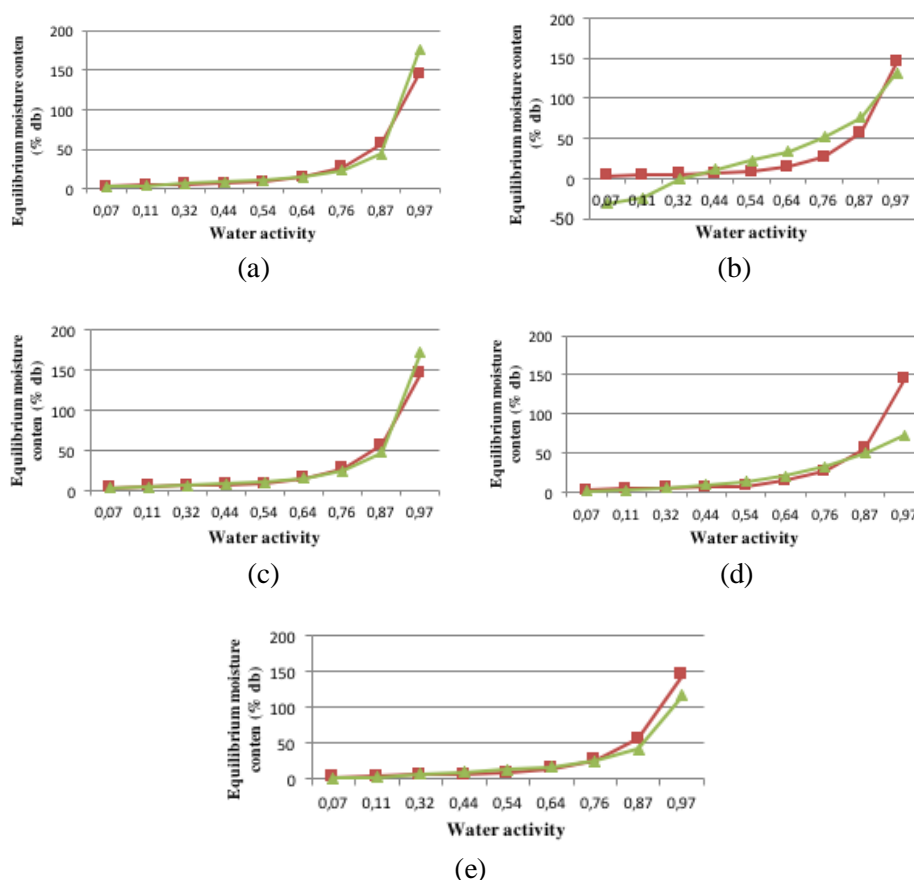

Figure 2. Five models of the equilibrium moisture content of uncoated natural condiment cube, ▬■▬Me; ▬▲▬ Me Hasley (a); Me Chen Clayton (b); Me Hederson (c); Me Caurie (d); Me Oswin (e).

According to water content, water of foods can be classified into three groups i.e. monolayer, multilayers, and free water layer [24]. This category shows that monolayer is dominant in the food with Aw of <32%, multilayer in Aw of 32%-76%, and free water layer in Aw >76%. It indicates that the EMC in the uncoated natural condiment cube, in general, are slightly higher than the coated one. The different interval of EMCs is due to product water absorption of RHs >76%. In this condition, the experimental environment has a high EMC, probably because of the free water is remained inside the coating samples [25]. Moisture sorption isotherm of natural condiment are presented in figure 2 and 3, show a sigmoid MSI curve which generally found in food and its product. The sigmoid shape is influenced by the combined effects of colligative, capillary and solid surface interaction with the temperature and the composition of the material. It could also affect the changes in the interaction of water molecules in the food [21].
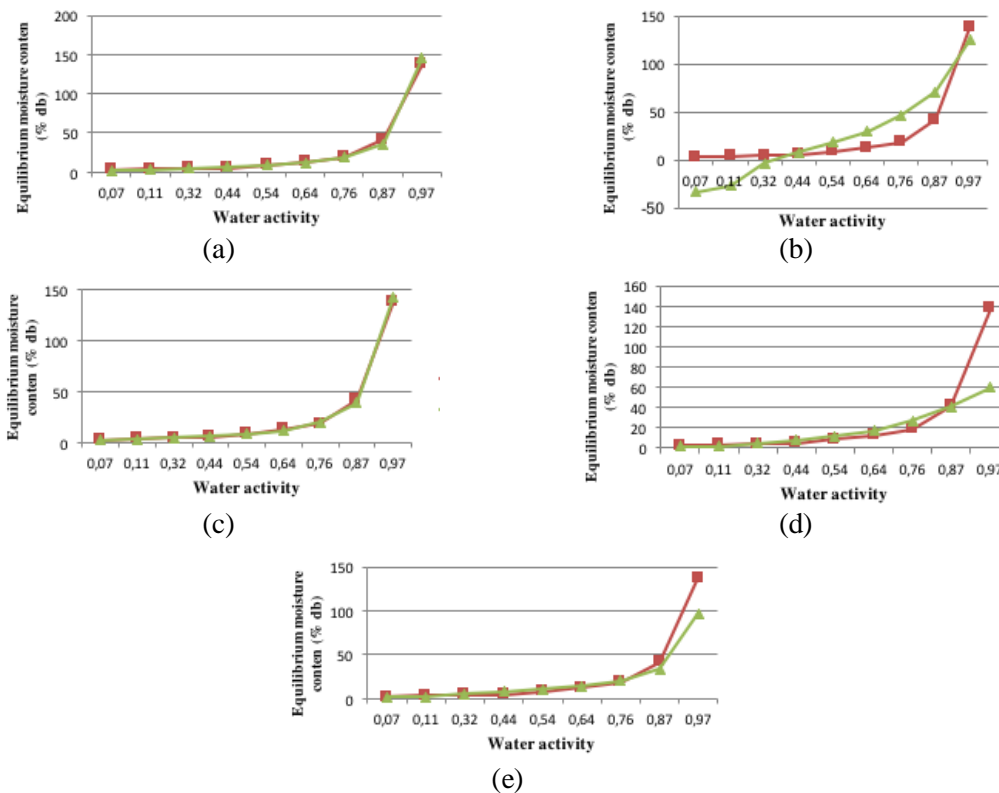
(a)

(b)

(c)

(d)

(e)

Figure 3. Five models of the equilibrium moisture content of coated natural condiment cube, ━■━Me; ━▲━ Me Hasley (a); Me Chen Clayton (b); Me Hederson (c); Me Caurie (d); Me Oswin (e).

In both MSI curves of coated and uncoated natural condiments show that in the range of RHs >76%, the equilibrium moisture contents increased sharply, precisely at 87% RH. The MSI of salted dried yellowtail has also the same trend [19]. Figure 2 and 3 explain that the product will absorb water when kept at room temperature with relative humidity >76%, and suggested that the product should be packaged in those condition. Storing in RHs <76 % makes the product be more stable. It indicates that the natural condiment cube at RHs >87% has a high ability to absorb water from the environment, and it occurred faster in uncoated natural condiment than the coated one (figure 3). It reflects that edible film (coating) could prevent organic compound interactions with water.

3.1. The accuracy of the models
Five equations model have been used to fix the MSI curves, and each model has been analyzed for MRD using equation 1. The MRD values can be seen in table 2.

Table 2. MRD of natural condiment.

Natural condiment	Hasley	Henderson	Chen Clayton	Caure	Oswin
A (uncoating)	15.18	6.58	262.11	32.78	28.27
B (coating)	12.19	8.21	335.89	33.74	28.61

Table 2 shows that one model has the MRD value <10, i.e. Henderson model, with MRD of 6.58 for uncoating natural condiment and 8.21 for coated condiment. Henderson model has a most suitability in predicting of MSI on foods that have RHs of 0-85%, and most precisely for coating condiment compare to uncoating one. This data supported that edible coating could prevent the water interaction between condiment and environment, as explained before.
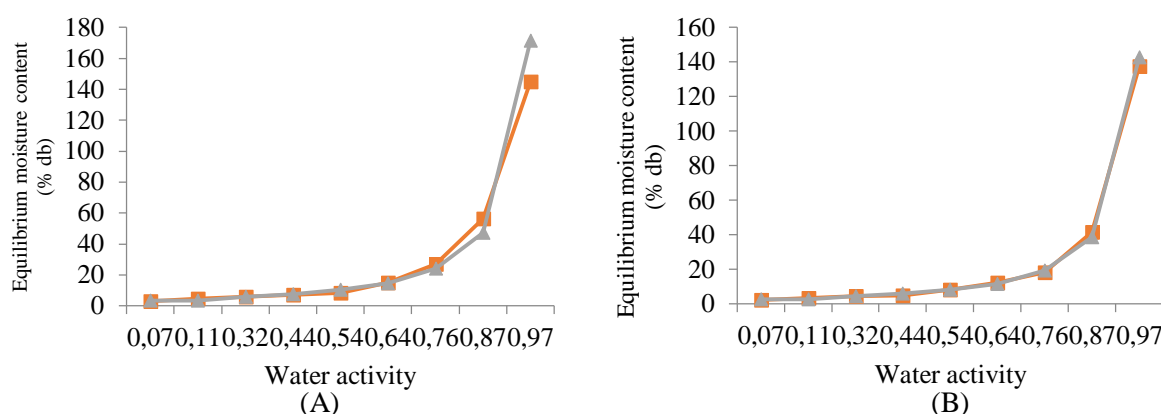
Figure 4. Moisture sorption isotherm of uncoated condiment (A) and coated condiment (B), ▪ Me;
— Me Hederson.

Based on the values of MRD, Henderson model was comsidered to fix the MSI curve for all treatments (figure 4). The accuracy of the predicted MSI curve is determined by form of function and parameters of the equation. Henderson models was the best model to fit the experiment and gave reliable results for the dried product.

4. Conclusion

Equilibrium moisture contents (EMCs) were obtained in different storage days, and can be classified in three groups i.e. RHs less than 32%, 32% to 76%, and higher than 76%. The MSI curve was a sigmoid shaped, and Henderson equation has given more precise predictions, with MRD range of 6.58 to 8.21. Equilibrium water content of natural condiment cube increased faster at RHs 76% to 97%, and therefore, in those RHs packing of the natural condiment is considered to be important.

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