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Moisture Sorbsi Isotherm (MSI) Model for Drying Bread Fruit (Artocarpus Altilis) Chips using a Tray Dryer

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Abstract:- The increase in breadfruit's water content and water activity causes the fruit to be susceptible to damage. Therefore, it is necessary to understand the absorption pattern of breadfruit's water content. The Water Sorption Isotherm (ISA) curve can be used to determine the pattern of water absorption. Therefore, it is necessary to know the knowledge of the sorption is other mic characteristics of water on breadfruit drying to predict the properties of these materials under conditions of relative humidity and air temperature in the storage area.

This research investigates the effect of storage temperature on the sorption isotherm characteristic of water of breadfruit chips. Dried breadfruit chips were conditioned at various water activity levels (aw) 0.1 - 0.8 using several types of saturated salt solution at 30°C and 40°C by gravimetric method. Furthermore, the obtained equilibrium moisture content and water activity data were plotted into a graph using several mathematical equation models, i.e., the BET, GAB, Oswin, Caurie, Halsey and Henderson models.

The curves were created using Microsoft Excel 2016 software. The exact model of the isothermic sorption equation of water for drying breadfruit was determined based on the equation model developed from the literature and adapted to the experimental data.

The results show that the isothermic sorption curve of water of breadfruit drying followed the sigmoid type 2 pattern. Four models were appropriate to describe the isothermic sorption pattern of the water of breadfruit drying at a storage temperature of 30°C, i.e., The GAB, Oswin, Caurie, and Halsey models.

The correct model suitability is indicated by the coefficient of determination constant (R^2) being high, the value of E is 5% < E < 10%, the value of the mean deviation (e_{ave}), and the value of the square root of the mean deviation (RMSE) is low.

The relative mean deviation value (%E) of 30°C storage temperature that can describe the isothermic sorption pattern is the CAB, Oswin,, Caurieand Halsey models (GAB= 6.713; Oswin=6.008; Caurie = 6.157; Halsey=6.189). The coefficient of determination(GAB=0.9544;Oswin= 0.7848; Caurie = 0.7705; Halsey = 0.8268), the average deviation value (e_{ave} GAB = 0.034; Oswin = 0.036; Caurie = 0.835; Halsey = 0.032) and the RMSE (GAB value. =0.183; Oswin=0.195;Caurie=0.416; Halsey=0.180).

Four models can describe the isothermic pattern of water absorption for a storage room temperature of 40°C, i.e., the GAB, Oswin, Caurie, and Halsey models.

The relative mean deviation (%E) of the model is (GAB=2.130; Oswin=5.509; Caurie = 5.928; Halsey=7.804). Coefficient of determination (GAB = 0.9928; Oswin = 0.8869; Caurie = 0.8654; Halsey=0.8336), mean deviation value (e_{ave} GAB = 0.001; Oswin=0.001; Caurie= 0.009; Halsey = 0.001) and RMSE values (GAB = 0.004; Oswin = 0.008; Caurie = 0.097; Halsey = 0.011).

I. INTRODUCTION

Breadfruit is gluten-free fruit and contains complete nutrients including carbohydrates, antioxidants, calcium, carotenoids, copper, iron, magnesium, niacin, omega 3, omega 6, phosphorus, potassium, protein, thiamine, vitamin A and vitamin C ((Biyumna *et al*., 2017; Teknologi *et al*., 2022). Breadfruit is one of the easily-damaged foodstuffs.

The protein and carbohydrate content of breadfruit starch consumed raw or cooked can compare to or even exceed other tropical plants such as sweet potato and cassava (Worrell *et al*., 1998; Arinola & Akingbala, 2018)

Harvested breadfruit can ripen within 1-3 days. It cannot be stored for more than five days and is stored in chilled conditions to delay the ripening process (Ragone, 2006; Arinola & Akingbala, 2018).

Breadfruit is one of the foodstuffs that, after being processed, has very hygroscopic properties, which can absorb water from the air in the surrounding environment and, on the other hand, can release some water into the air. This hygroscopic nature can affect the breadfruit's shelf life, quality, and stability.

Product stability can be determined by two main factors, i.e., the equilibrium relative humidity (RH) or water activity (aw) in the storage area and the equilibrium moisture content of food ingredients (Me) (Widowati *et al*. 2010).

The correlation between water activity (aw) and the water content of food products in a storage condition at a specific relative humidity (RH) value is also known as the water sorption isotherm (ISA).

The quality of most foods preserved by drying, such as breadfruit, is highly dependent on their physical, chemical, and microbiological stability. This stability is mainly a consequence of the correlation between the food's equilibrium moisture content (EMC) and the water activity (aw) at a given temperature.

The correlation between relative humidity, water activity, and water absorption isotherm during storage determines the reactivity in predicting foodstuffs' quality, stability, and shelf life (P *et al*., 2011). It is also essential to understand the connection of these parameters in designing and calculating the optimization of drying equipment and for calculating changes in humidity that may occur during storage.

Isothermal sorption describes the thermodynamic correlation between water activity and the equilibrium moisture content of food products at constant temperature and pressure. It also can predict the effect of the components of food ingredients on decreasing or increasing the aw value. This isothermal sorption is generally represented by an equation/mathematical model.

Many mathematical models are often used to describe the drying process, where the thin layer drying model is the most common. According to Al-Muhtaseb 2002; Ruth *et al*., 2020, mathematical models describing the drying mechanism of grains and foods can also provide the required temperature and moisture content information.

The equation models that are most commonly used to describe absorption in foodstuffs include the Langmuir equation or model, the BET (Brunauer-Emmett-Teller) model, the Oswin model, the Smith model, the Halsey model, the Henderson model, the GAB model (Guggenheim-Anderson- deBoer), and the Peleg model (Sahin *et al*., 2006; Ruth *et al*., 2020).

An experiment was conducted in this research to evaluate the ability and accuracy of the model in predicting the isothermic pattern of water absorption in the drying of breadfruit chips at 30°C and 40°C with various relative humidity values. The suitable model can determine the best storage stability conditions and minimize the water absorption rate to extend its shelf life.

II. MATERIALS AND METHOD

A. *Tools and materials*

The tools used in this research include a desiccator, analytical balance, a rectangular aluminum frame, oven, and measuring cup. The materials used include dry breadfruit chips with blanching and non-blanching treatments and five types of saturated salt, i.e.,NaOH,$MgCl_2$,K_2CO_3,NaCl dan KCL.

B. *Research Procedure*

a) Determination of Equilibrium Moisture Content
The equilibrium moisture content of breadfruit was conducted using the gravimetric method.

Moisture Absorption Study. Saturated salt solution for the required humidity was done by dissolving a certain amount of salt in distilled water until it was saturated or no longer dissolved. Before use, the saturated salt solution was first incubated in a desiccator for 24 hours at a temperature of 30±2°C. The water activity (aw) of several saturated salt solutions at 30°C is presented in table 1.

type of salt	aw	Amount	
		salt(g)	aquades(ml)
NaOH	0.18	150	85
$MgCl_2$	0.32	200	25
K_2CO_3	0.46	200	90
NaCl	0.75	200	60
KCL	0.82	200	80

Table 1: Preparation of Saturated Salt solution for determination of the Sorbsi Isotherm Curve

Source: (Spiess and Wolf,1987; *Umur Simpan Granula Ubi Kayu Dg ISA*, n.d.)

Samples of dried breadfruit chips with a size of 2x2x2 cm^3 with three replications were placed on top of the aluminum rectangular hole container. Breadfruit chips were put into a desiccator containing a saturated salt solution according to the desired water activity level at room temperatures of 30°C and 40°C. Samples were weighed weekly using a digital balance until they reached a balanced weight. The balanced weight is characterized by a difference of three consecutive weighing of ≤ 2 mg for RH ≤ 90 conditions and no higher than 10 mgg for RH 90 conditions (Liovonen and Ross, 2000; Adawiyah, 2006; Mona fitria, 2007). Samples that have reached a constant weight are then measured for their moisture content by the oven method (Sudarmadji *et al*., 1997; Ritonga & Masrukhi, 2020) at a temperature of 105°C for 72 hours. The water content obtained was the equilibrium water content.

b) Determination of Curves and Equation Models of Moisture Sorbsi Isothermic
Six equations models of water sorption isothermic of dry breadfruit developed from the literature were tested for accuracy on experimental data. The model that was tested for accuracy with experimental data is presented in table 2.

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Model	Equation	Source
BET	$M = \frac{M_0 . C . a_W}{(1-a_W)(1+C.a_W-a_W)}$	Sahin and Sumnu, 2006;Aini *et al.*, 2014
GAB	$m_e = \frac{m_o k C a_W}{[1-ka_W][1-ka_W+1-Cka_W]}$	Labuza dan Altunakar, 2007; Sahin and Sumnu, 2006; Aini *et al.*, 2014
Oswin	$\ln m_e = \ln k + c.\ln[a_W/(1-a_W)]$	Oswin, 1946; Sahin and Sumnu, 2006; Cahyanti, 2016
Caurie	$m_e = A + B.\ln(-\ln a_W) + C\ln(-\ln a_W)^2 + D\ln(-\ln a_W)^3$	Caurie, 1970; Mariem and Mabrouk, 2015 (Sugiyono *et al.*, 2011)
Halsey	Log [ln(1/1-aw)] = log K + n log Me	Bonner dan Kenney 2013; Crude, 2016
Henderson	Log [ln(1/1-aw)] = log K + n log Me	Chirife dan Iglesias, 1978; Van den Berg dan Bruin, 1981; Molenaar *et al.*, 2014

Table 2: Model test accuracy

Six equation models, i.e., the BET, GAB, Oswin, Caurie, Halsey and the Henderson model, were tested for accuracy using the experimental model. The model being tested was modified into a linear equation with logarithmic transformations and normal logarithmic to facilitate the calculation process.

These six models were tested based on previous studies, where these equations can describe the isothermic sorption curve over a wide range of water activity values (a_w) (Chirife and Iglesias, 1978; Mona Fitria, 2007). In addition, the equation model above has parameters less or equal to three; therefore, it is simpler and easier to solve.

c) Model Accuracy Test

The accuracy test of the isothermic sorption equation was conducted using the calculation of Mean Relative Determination (MRD) (Walpole, 1990; Ritonga & Masrukhi, 2020). The analysis of the accuracy between the mathematical model and the experimental data refers to the percentage value of the relative mean deviation (MRD or E%), the mean deviation value (e_{ave}), and the square root value of the low mean deviation (RMSE) (Lomauro et al., 1985; Crude, 2016). If 5<E%<10, then the model rather accurately describes the actual situation. If MRD>10, then the model is not appropriate to describe the actual condition (Juliana et al., 2020) with the following equation:

$$E(\%) = \frac{100}{n}\sum_{i=1}^{n}\left|\frac{M_i - M_{pi}}{M_i}\right| \ldots\ldots\ldots\ldots\ldots\ldots(1)$$

$$e_{ave} = \left(\frac{M_i - M_{pi}}{n}\right) \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(2)$$

$$RMSE = \sqrt{\frac{1}{n}\left[\sum_{i=1}^{n}\left(M_i - M_{pi}\right)^2\right]} \ldots\ldots\ldots\ldots (3)$$

Where n = the number of experimental data, mi is the experimental data for the measurement of isothermic absorption, and Mpi is the prediction data using the model. The model is considered suitable/proper if the E value < 10% (Mc.Laughlin and Magee, 1998; Adawiyah *et al*., 2010). Another opinion expresses that the modeling has good accuracy if the MRD value is less than 10% (McLaughlin and Magee, 1998). ; Azizah, 2017. The RMSE (Root Mean Square Error) value indicates the matching ability of a model concerning the number of data points.

III. RESULT AND DISCUSSION

Determination of the equilibrium moisture content uses the static gravimetric method by conditioning five types of saturated salt solution at different RH at storage temperatures of 30°C and 40°C. The MSI curve obtained from measuring the equilibrium moisture content of the sample plotted on each aw can be observed in Figure 1,2,3 and Figure 4. The MSI curve is closely related to the stability of food ingredients under various storage conditions and the product packaging process to maintain the stability of its shelf life (Budijanto *et al.*, 2010) and can determine changes that may occur to food during the storage process (Dyah Purnomosari, 2008).

The value of the equilibrium moisture content will increase with the value of relative humidity or water activity. This process can occur because of the transfer of water vapor from the environment to the sample, which is hygroscopic. There is an adsorption process on breadfruit chips at high water activity; thus, the equilibrium water content increases. However, a desorption process occurs in the sample when water activity is low. Therefore, the equilibrium water content decreases (Banoet, 2006; Azizah, 2017).

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The MSI curve of breadfruit chips follows the shape of the MSI type 2 curve in the form of a sigmoid. This sigmoid curve is the most common in foodstuffs and is specific for each food ingredient. Foodstuffs in the form of carbohydrates, tapioca, and casein as their main constituents show an isothermic sorption pattern in the form of a sigmoid commonly found in amorphous food systems (Adawiyah *et al*., 2010).

This is also reinforced by the opinion of Labuza *et al*., 1985, that foodstuffs with low water content tend to have a sigmoid isotherm curve. However, the slope of the sigmoid sorption isotherm curve can vary due to the nature of the food ingredients, temperature, and speed factors of adsorption and desorption that occur during storage (Fennema, 1985; Azizah, 2017).

The sigmoid pattern of the MSI curve is characteristic of dry foods rich in carbohydrates (Wolf *et al.*, 1972; Yusa, 2014). In addition, Adawiyah, 2006; Yusa, 2014) state that this sigmoid shape is due to the effect of capillarity and the interaction between the surface of the material and water molecules, as well as the accumulation effect of hydrogen bonds, Raoult's law and the interaction between water and the surface of food (Sahin and Sumnu 2006; Juliana *et al.*, 2020).

Another factor that affects the shape of the MSI curve is the physical interaction and components of food ingredients consisting of carbohydrates, proteins, fats, and minerals (*Shelf Life of Cassava Granules with ISA*).

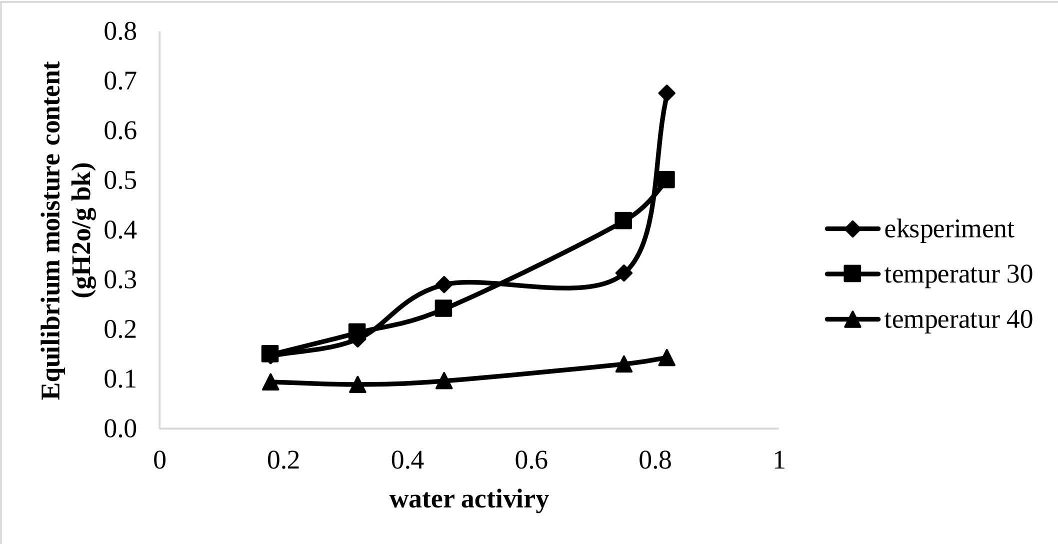

Fig. 1: Graph of GAB Model

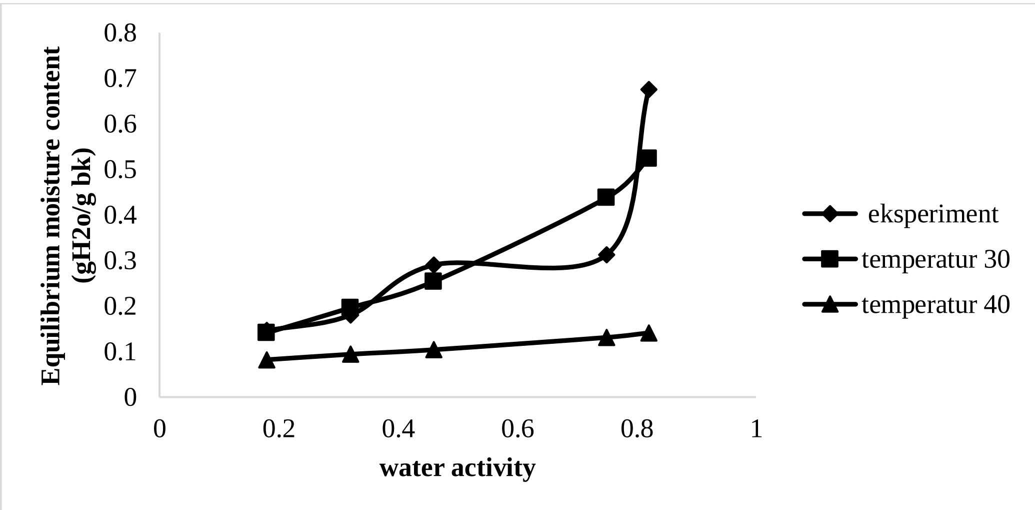

Fig. 2: Graph of Oswin Model

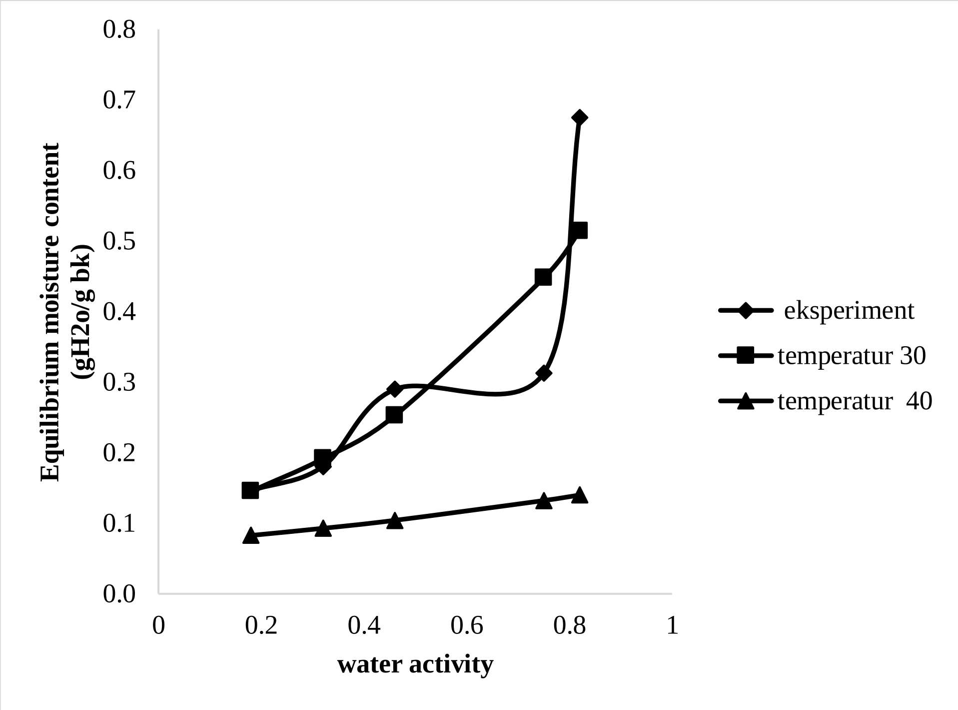

Fig. 3: Graph of Caurie Model

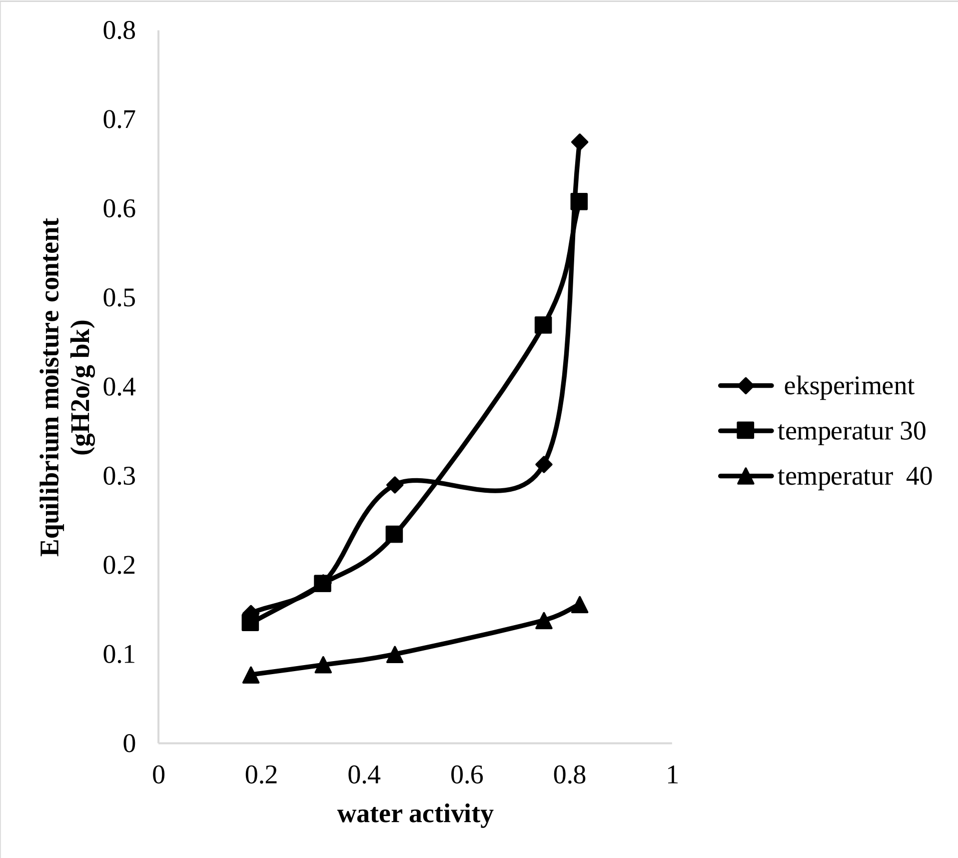

Fig. 4: Graph of Halsey Model

Kusnandar *et al*., 2010; Azizah, 2017 stated that the type of isothermic curve of each food ingredient varies depending on how dominant the hygroscopic character of the food material; highly hygroscopic, moderate hygroscopic, and low hygroscopic. Isothermal sorption describes a complex hygroscopic ability and is an interaction, physically or chemically, between the components of the food ingredients and is also influenced by the heating process or pretreatment.

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The sigmoid shape on the MSI curve occurs because of differences in water binding in foodstuffs. Water bound to foodstuffs can be categorized into three; primary bound water, secondary bound water, and tertiary bound water (Labuza, 1968, Suyitno, 1995, Candra, 1998, Sukmono, 1998; Dyah Purnomosari, 2008). Primary bound water or single layer bound water is located at aw below 0.25, secondary bound water is located between a_w 0.25-0.75, and tertiary bound water is located above 0.75 a_w (Suyitno, 1995; Sukmono, 1998; Dyah Purnomosari, 2008).

The sigmoid shape of the MSI curve on breadfruit chips illustrates that the hygroscopic ability of the material is Type 2 or moderate hygroscopic.

Testing for each model of the isothermic sorption equation is conducted by testing the accuracy of the model using the criteria for the percentage value of the relative average deviation (MRD or E%), the average deviation value (e_{ave}), and the low square root value of the average deviation (RMSE) as shown in equations (1), (2) and (3). The value of the accuracy test of the breadfruit chips model can be seen in the following table 3:

Perlakuan	uji ketepatan				
suhu	model	E%	e_{ave}	RMSE	R^2
30 ^{o}C	BET	65,930	2,605	1,614	0,9089
	GAB	6.713	0,034	0,183	0,9544
	Oswin	6.008	0,036	0.195	0,7848
	Caurie	6.157	0.835	0.416	0,7705
	Halsey	6.189	0.032	0,180	0,8268
	Henderson	174,220	0,086	0,293	0,8514
40 ^{o}C	BET	65,479	0,014	0,117	0,9331
	GAB	2.130	0,001	0,004	0,9928
	Oswin	5.509	0,001	0,008	0,8869
	Caurie	5.928	0,009	0,097	0,8654
	Halsey	7.804	0,001	0,011	0,8336
	Henderson	174,220	0,086	0,293	0,7878

Table 3: The value of the accuracy test of the Moisture Sorbsi Isothermic (MSI) model of breadfruit chips

The graph of the correlation between equilibrium moisture content and water activity based on various mathematical models tested can be seen in the image below:

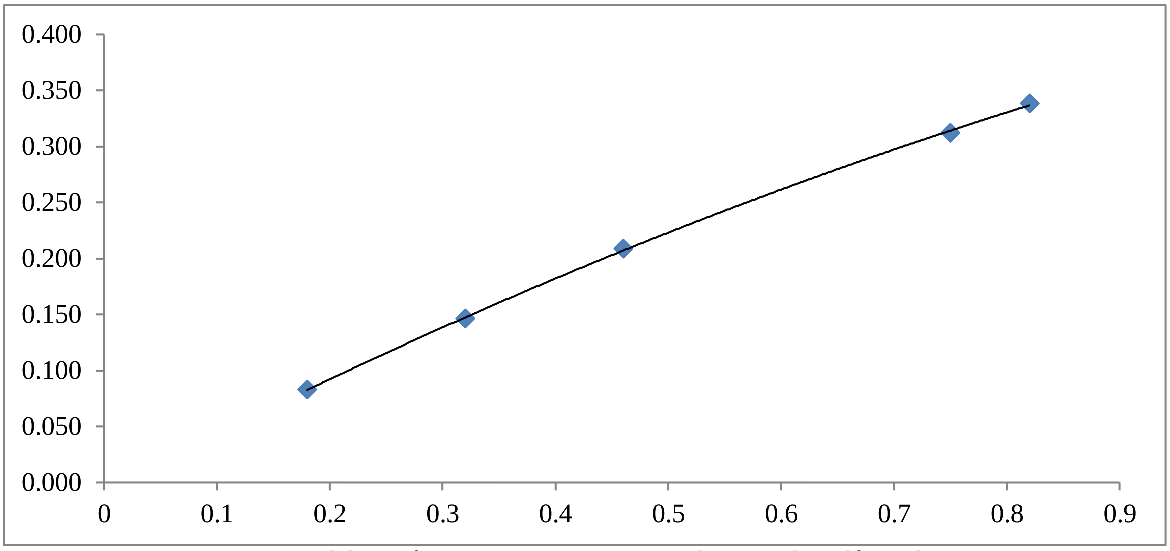

Fig. 5: Modeling of GAB Water Sorption Isotherm on breadfruit chips

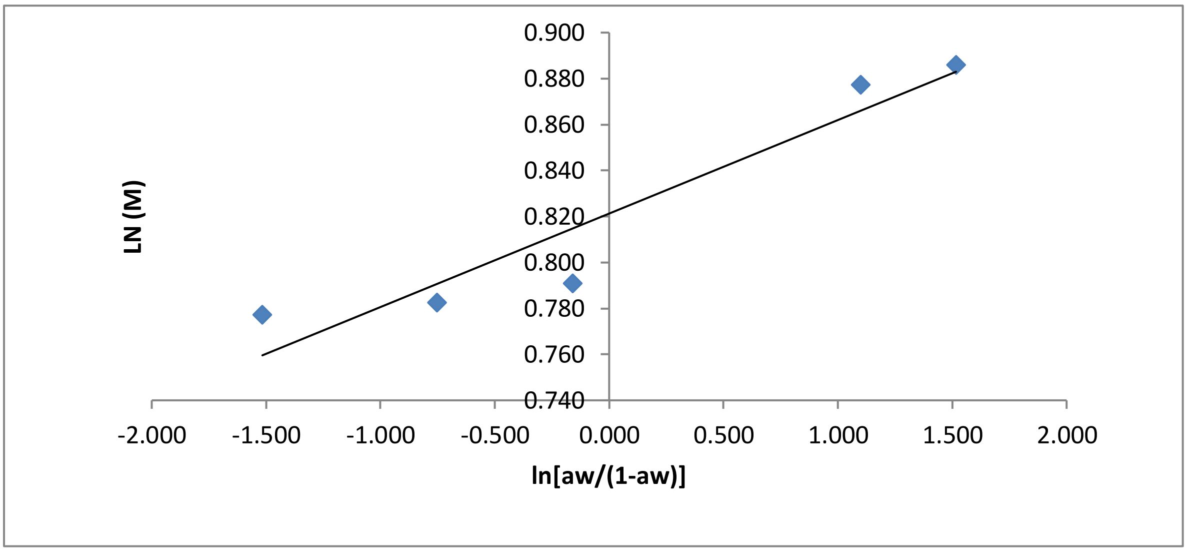

Fig. 6: Modeling of Oswin Water Sorption Isotherm on breadfruit chips

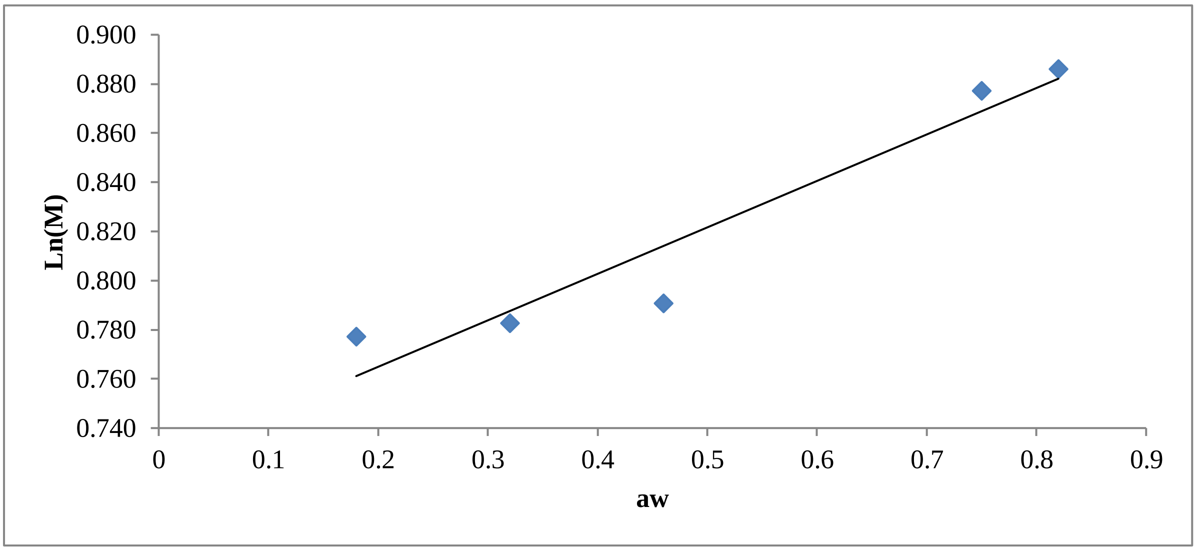

Fig. 7: Modeling of Caurie Water Sorption Isotherm on Breadfruit Chips

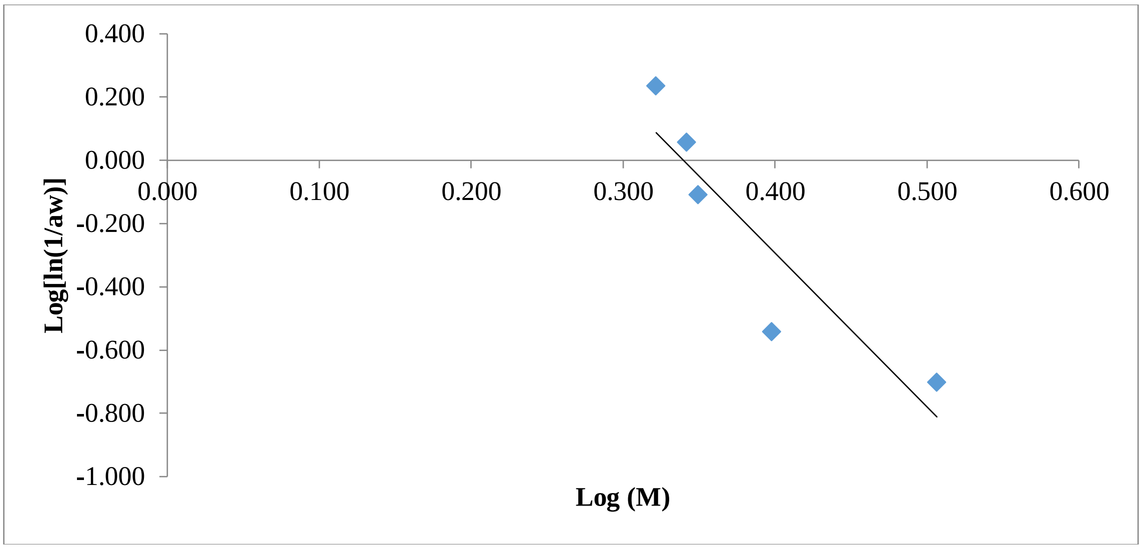

Fig. 8: Modeling of Halsey Water Sorption Isotherm on Breadfruit Chips

The table above shows that at a storage temperature of 30°C, the E% values of the GAB, Oswin, Caurie, and Halsey models are < 10%, while the e_{ave} and RMSE values are low, and the R^2 value is high. Therefore, the model can accurately describe the actual state of the sorption isotherm. The E% test value above 10% is considered unable to describe the actual situation. The lower the E% value, the water sorption isotherm model can accurately describe the actual situation (Kusnandar *et al*., 2010; Hartanto, 2019).

Meanwhile, atastorage temperature of 40°C, the appropriate models describing the actual isothermic sorption are the GAB, Oswin, Halsey and Caurie models with E% <10%, e_{ave}, and RMSE values are low, while R^2 is high.

IV. CONCLUSION

The water sorption isotherm curve on breadfruit chips is in the form of sigmoid type 2. The appropriate modelings to predict the phenomenon of water sorption isotherm on breadfruit chips with a storage room temperature of 30°C are GAB, Oswin, Caurie and Halsey. Meanwhile, the appropriate models at a storage temperature of 40°C are GAB, Oswin, Caurie and Halsey

At the same temperature, the increase in water activity (a_w) causes the equilibrium moisture content of the breadfruit chips to be higher. This is because the humid air contains more water content. Therefore, the equilibrium moisture content of the breadfruit chips will be higher.

At the same a_w, an increase in temperature causes the value of the equilibrium moisture content to be lower. This is because the high air temperature causes the storage air to be drier and allows the sample to undergo a desorption process. The desorption process is the activity of releasing water from the sample into the storage air; thus, the water content becomes lower.

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