Scientific Research Journal,
Vol 17, No 2 (2020) 221-245
https://doi.org/10.24191/srj.v17i2.10160

Moisture Adsorption Isotherm Model for Edible Food Film Packaging – A Review

Aqila Zulaikha Nazreen, Junaidah Jai*, Sherif Abdulbari Ali, Norasmah Mohamed Manshor

Faculty of Chemical Engineering, Universiti Teknologi MARA 40450 Shah Alam, Selangor, Malaysia

**Corresponding author's e-mail: junejai@uitm.edu.my*

Received: 27 March 2020
Accepted: 27 August 2020
Online First: 28 August 2020

ABSTRACT

Bioplastic has been extensively studied due to its ability to replace synthetic plastics. Its biodegradability is seen as a significant advantage to be used as edible food film packaging. As a food packaging, the film should be able to retain moisture from the food and the surrounding environment. The ability of the film to retain moisture varies with the type of material the film is made of. Moisture sorption isotherms are used to study the moisture content and water activity of food or film packaging at a given temperature. There are many models to describe moisture sorption isotherm. Some researchers modified the models to account for varying temperatures. The purpose of this paper is to review types of adsorption isotherms and the most common models used for film packaging. The general and modified models are described in this paper as well as its best-fit use on film packaging. The advantages of studying adsorption isotherm include shelf-life determination, physical and chemical stability, and others.

Keywords: *adsorption isotherm model, edible film, moisture sorption isotherm, moisture content, water activity*

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INTRODUCTION

In recent years, the focus of researchers is to develop bioplastic. This is due to the use of synthetic plastics that have been affecting our environment negatively. The demands for synthetic plastics have been increasing these past few years. One of the primary uses of synthetic plastics is food packaging. Therefore, bioplastic film is seen as an environmental-friendly alternative to synthetic plastics. Bioplastic film is defined as a thin layer made of natural polymers. The benefits of bioplastic films include environmentally friendly as it can degrade voluntarily, consumable with the food, non-toxic, and made of natural resources.

Generally, the function of bioplastic film as food packaging is to protect the food or produce from the surrounding environment [1]. From a more in-depth perspective, the role of a bioplastic film as food packaging is to act as a moisture barrier for the food to prevent moisture loss. Moisture loss from food could disrupt its texture, flavour, and colour. Other than that, it should be able to act as a gas barrier between the food and the surrounding environment to retard the deterioration of the food. Bioplastic film should also have the proper durability as food packaging to protect the food from mechanical impact during transportation and storage [2].

Since bioplastic films are made of natural resources, proper materials could be selected according to the properties we wanted. Mainly, bioplastic films are made using polysaccharides, lipids, protein, or a combination of these materials. Each material has its advantages and disadvantages. Starch is an example of polysaccharides which are made up of the hydroxyl group. Lipids are hydrophobic materials such as beeswax. Soy protein is an example of a protein which are made up of hydrophilic amino acid. A combination of all these materials would produce a composite edible film which is suitable to be tallied to the properties we desired for food packaging from the edible film [3].

The suitability of edible film as food packaging depends on its properties. Physical, chemical optical, mechanical, and barrier are among the properties studied by researchers. The water absorption property of an edible food film packaging is determined through moisture sorption isotherm [4]. It is an important property studied to evaluate the performance of the film when exposed to a variety of temperatures during packaging and storage. Through sorption isotherm, the equilibrium moisture content could be estimated; thus, the hydrophilic properties of the film, stability, and quality changes of the food product could be predicted [5].

CLASSIFICATION OF ADSORPTION ISOTHERM

Moisture sorption isotherm is the mathematical expression that describes the moisture content and water activity of food. Through these mathematical expressions, the graph of isotherm could be acquired in two ways, which are adsorption isotherm and desorption isotherm, as shown in Figure 1. The adsorption isotherm is acquired by measuring the addition of weight due to moisture uptake when a completely dry material was introduced to the environment with rising relative humidity. Meanwhile, desorption isotherm is obtained by measuring the decrease in weight when firstly wet material is introduced to the environment with constant relative humidity [6].

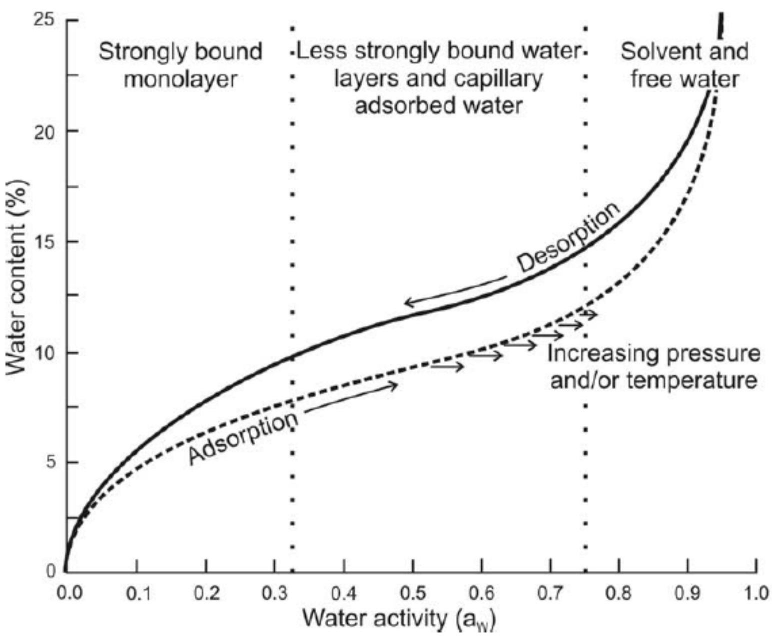

Figure 1: Graphical Representation of Moisture Sorption Isotherm for Food Products (Source: Airaksinen [7], 2005)

The adsorption isotherm can be further classified into five types according to the curve shape and process.

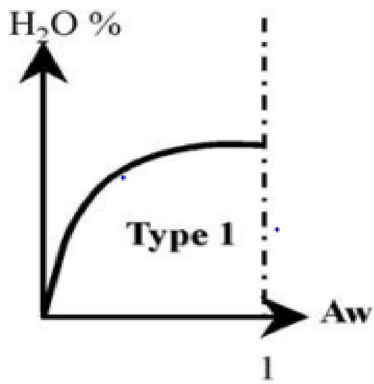

Figure 2: Adsorption Isotherm Type 1 (Source: Mathlouthi and Rogé [8],2003)

Figure 2 shows the Type 1 of the adsorption isotherm. Type 1 is called the Langmuir and could easily be described by the Langmuir Adsorption Isotherm [6, 9]. The graph is a convex upwards where the increase in water activity causes an increase the moisture content. Commonly, this graph could be found in processes with monolayer adsorption in which the surface of the adsorbent interacts with the one layer of molecules.

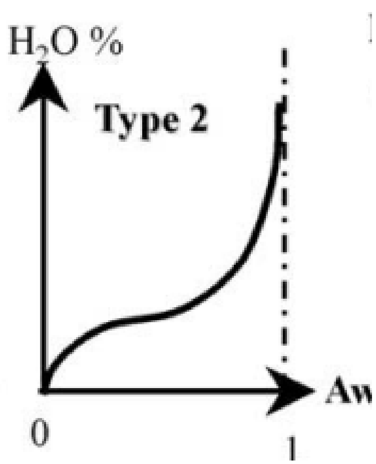

Figure 3: Adsorption Isotherm Type 2 (Source: Mathlouthi and Rogé [8],2003)

Figure 3 shows Type 2 of the adsorption isotherm. It is commonly known as the sigmoidal adsorption isotherm. From the shape of the graph that concave upwards, this type of sorption isotherm considers multilayer adsorption [6]. The knee of the curve is the point of completion for the first monolayer adsorption. It also marks the beginning of adsorption for the second and higher layers [10].

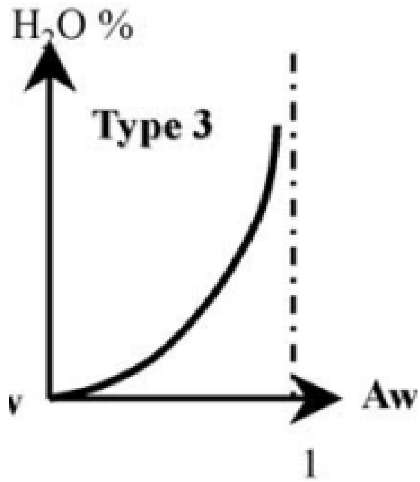

Figure 4: Adsorption Isotherm Type 3 (Source: Mathlouthi and Rogé [8],2003)

Figure 4 shows Type 3 of the adsorption isotherm, which is generally known as the Flory- Huggins isotherm [6]. Based on the graph, there is no knee point, which means there is no restricted multilayer formation. The graph is convex at all its points. This type of graph occurs due to the weak interaction between the adsorbent surface and adsorbate [10].

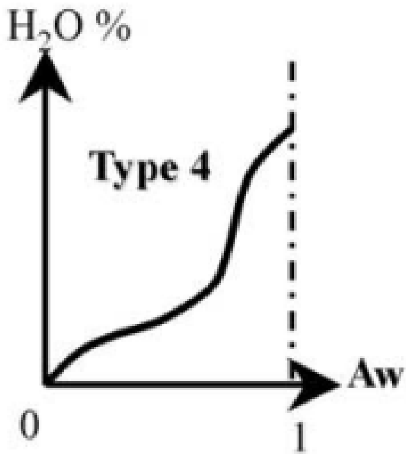

Figure 5: Adsorption Isotherm Type 4 (Source: Mathlouthi and Rogé [8],2003)

Figure 5 shows Type 4 of the adsorption isotherm. Type 4 is closely related with Type 2 as both curves contain a knee point. The curve shows the formation of monolayer and multilayer formation. It could also be used to describe the swelling of a hydrophilic solid through adsorption until it reaches maximum site hydration [11].

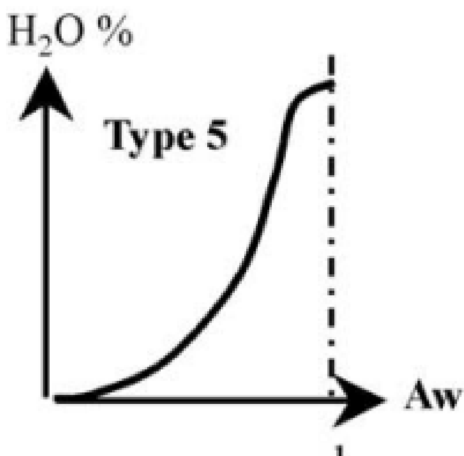

Figure 6: Adsorption Isotherm Type 5 (source: Mathlouthi and Rogé [8],2003)

Figure 6 shows Type 5 of the adsorption isotherm. Type 5 is the same as Type 3, where there is an absence of knee point. Both curves describe the formation of multilayer adsorption and weak interaction between adsorbent-adsorbate interactions [11].

Based on the types of adsorption isotherms, Type 2 is the most popular for food products [9]. This shows that most types of food have multilayer adsorption and that the water contained in the food exhibits vapour pressure relatively near to pure water. The vapour pressure remains unchanged until the moisture content of the food is at 22%, and after that, the vapour pressure decreased. The sigmoid shape is the result of the difference in the atmospheric moisture [6]. An example of a food with Type 2 adsorption isotherm includes cassava starch [12], indica rice starch [13], banana [14], and others. For films, Type 2 and Type 3 are frequently found. Sago starch film [15] is an example for Type 2, while chitosan film [16] is an example for Type 3 adsorption isotherm.

The classification of moisture sorption isotherm is essential to understand the interactions between the adsorbates and the adsorbent surfaces, which leads to the understanding of the stability of the edible food film packaging. The sensitivity of the film to moisture and the type of surroundings it is exposed to needs to be determined to maintain the shelf life of the packaging as well as the quality of the food product.

MODELS OF ADSORPTION ISOTHERM

There are many models developed to describe the sorption isotherm of edible food film packaging. Some models are developed with the theoretical background to describe the sorption mechanism, while other models are a simplification of the theoretical models or entirely empirical [9]. Inedible food film packaging, the study of sorption isotherm is vital to determine the shelf life of packaging, quality, and stability of food during transport and storage. Some well-known models used for edible food film packaging are described below. The models depicted include its general and modified forms. The main difference between the general model and the modified model is the inclusion of the temperature parameter. Some researchers find it necessary to include the temperature parameter in the models. Temperature is one of the factors that need to be considered because the water uptake of an edible food film packaging also depends on temperature other than relative humidity and chemical composition.

Brunauer – Emmett – Teller (BET) model.

General BET model

The BET equation was first introduced by Brunauer, Emmett, and Teller in 1938. Type 2 and Type 3 of the adsorption isotherm; it gives an approximation of the monolayer value. The BET model is widely used in the application of a gas-solid equilibrium system. The BET equation was modeled based on the following assumptions:

1. The rate of condensation of the first layer is equal to the rate of evapouration from the second layer
2. The binding energy of all the adsorbate on the first layer is equal
3. The binding energy of the other layers is equal to those of pure adsorbate

In most cases, the BET model only provides a linear graph for water activity between 0.05 and 0.045 [9]. Therefore, due to its limited range of water activity, the BET model is mainly used to determine the surface area of an adsorbent [17]. Besides, it could also be used as a basis to determine an ideal moisture content to provide stable storage of foods, especially for dehydrated food products [9].

The BET equation is given as:

$$M_w = \frac{M_0 C a_w}{(1-a_w)(1+(C-1)a_w)} \qquad (1)$$

Where;
Mw = Equilibrium moisture content (kg / kg dry solid)
Mo = Monolayer moisture content (kg/ kg dry solid)
C = energy constant related to the net heat of sorption
a_w = water activity

A study Bertuzzi, Castro Vidaurre [18], where corn starch and glycerol were used to form a film, BET equation sufficiently describes the sorption isotherm for the film where the coefficient of determination (r^2) > 0.99. Another study by Monte, Moreno [19], where a film was formed using chitosan and glycerol. The BET model was one of the models used to determine the equilibrium moisture content of the film. The BET model showed a reasonably good fit over the experimental data with r^2 > 0.98. In general, the BET model is used in the food system to estimate the value of monolayer moisture content for a small range of water activity. It is essential to estimate the value of monolayer moisture content as it will give a general idea of the moisture content for drying process and storage of the food to inhibit chemical and biological growth.

Modified BET Model

In a study by Aguirre-Loredo, Rodriguez-Hernandez [16], the sorption data for chitosan films were fitted to the modified BET. The model is suitable for water activity range from 0 to 0.75 with a coefficient of regression, r^2 = 0.9244. The BET model in terms of temperature is given by (2) below:

$$\boldsymbol{X = \frac{(A+BT)C\,a_w}{(1-a_w)(1-a_w+Ca_w)}} \qquad (2)$$

Where;
X = moisture content on a dry base (kg/kg dry solid)
T = Temperature
a_w = Water activity
A, B and C = constants

Guggenheim – Anderson – de Boer (GAB) Model

General GAB Model

The equation came from Guggenheim, Anderson, and de Boer, who each derived the equation independently. The GAB equation is an extension of the BET physical adsorption isotherms. This model proposed that the state of sorbate in the second layer is equivalent to the following layers, except for those in liquid form. This model introduces a new parameter K, which is the difference of the potential chemical standard between molecules in the second and following layers as well as molecules in the form of liquid. The GAB equation is given by (3):

$$M_W = \frac{M_0 c k a_w}{(1-ka_w)(1-ka_w+cka_w)} \quad (3)$$

Where;
M_w = moisture content (kg/kg dry solid)
M_0 = Monolayer moisture content (kg/kg dry solid)

c and k = constants where the energies of interaction between first and further molecules at individual sorption site. They are given as (4) and (5), respectively.

$$C = C_0\, exp(\frac{H_0 - H_n}{RT}) \quad (4)$$

$$K = k_0\, exp\,(\frac{H_n - H_l}{RT}) \quad (5)$$

Where;
c_o and k_o = entropic accommodation factors
H_0, H_n, and H_l = molar sorption enthalpies for monolayer, multilayers on top of the monolayer, and bulk liquid, respectively (kJ mol^{-1}).

R = Ideal gas constant
T = absolute temperature

The introduction of parameter K for the GAB equation considers the interactions of energy between multilayer molecules and the sorbent. Using the GAB equation, when the value of K is less than 1, the value of the sorption isotherm is much lower than calculated by using the BET equation. This gives an advantage to the GAB equation since it can be used at a wider range of water activity between 0 and 0.9. When K is equal to 1, the GAB model becomes the BET model. Other advantages of the GAB model include reliable theoretical background and considers temperature using the Arrhenius equation.

According to Leceta, Arana [20], the GAB equation decently described the chitosan film water sorption data. The equation is suitable to use for all range of water activity and could also be used to determine the monolayer moisture content. It is essential to determine the monolayer moisture content because it shows us the physical and chemical stability of the film, which could help determine the capability of the chitosan film to act as a water barrier.

Modified GAB Model

In a study of chitosan films by Aguirre-Loredo, Rodriguez-Hernandez [16], equation 6 was used and was found to be the best fit for moisture data obtained with $r^2 = 0.9847$. The modified equation for GAB, which includes temperature, is given as equations below:

$$X = \frac{X_m + \left(\frac{C}{T}\right) k a_w}{(1 - k a_w)(1 - k a_w + \frac{C}{T} k a_w)} \qquad (6)$$

Where
X = moisture content on a dry base (kg/kg dry solid)
X_m = Monolayer moisture content (kg/kg dry solid)
T = Temperature
a_w = water activity
A, B, C, and k = constants

Halsey Model

General Halsey Model

This model was developed by Halsey. Halsey equation is applicable for use when evaluating the condensation of multilayers at a relatively large distance from the surface. This model inferred that the potential energy of a molecule changes as the inverse nth power of its distance from the surface. Halsey model is represented by (7)

$$M_W = M_O\left(-\frac{A}{RT\ln a_w}\right)^{1/n} \quad (7)$$

Where;
M_w = Equilibrium moisture content (kg/kg dry solid)
M_0 = Monolayer moisture content (kg/kg dry solid)
A and n = constants
R = Universal gas constant
T = Absolute Temperature

The Halsey model has been seen as applicable to many types of food that have a water activity range between 0.4 and 0.9 [4]. The model could easily give a good description regarding moisture sorption data of Type 1, 2, or 3. Other than that, the Halsey equation is used to depict the sorption behaviour of starch-containing products. In a study regarding chitosan – polyvinyl alcohol film, the Halsey model proved to be more suitable than the BET model and could be used for a broader range of water activity [21]. Another study by Monte, Moreno [19] regarding chitosan – glycerol film, the Halsey equation was proven to have a good fit over the experimental data with $r^2 > 0.99$.

Modified Halsey Model

The modified Halsey was used to study the sorption isotherm of chitosan films. It is applicable for water activity of range between 0 and 0.9 with $r^2 = 0.9703$ [22]. The modified Halsey equation for temperature is given by (8):

$$\boldsymbol{X} = [-\frac{exp(A+BT)}{ln\, a_w}]^{\frac{1}{C}} \qquad (8)$$

Where;
X= Moisture content on a dry basis (kg/kg dry solid)
T = Temperature
a_w = Water activity
A, B and C = constants

Henderson Model

General Henderson Model

The Henderson equation is extensively used to describe the relationship between water activity and the amount of water adsorbed. When ln (-ln ($1-a_w$) is plotted against ln M, a straight line is obtained. However, it was found that the model could not provide exact information on the physical state of water for three localised isotherms. A study of polyethylene oxide – blended starch films found that the Henderson model is suitable for water activity ranged from 0.33 to 0.86 [4]. Another study conducted by Raj, Eugene Raj [23] for polyvinyl alcohol – corn starch film, the Henderson model showed a good fit for water activity ranged between 0.1 and 0.6.

The model is given by (9):

$$M_W = [\frac{ln(1-a_w)}{-A}]^{1/n} \quad (9)$$

Where;
M_w = Equilibrium moisture content (kg/kg dry solid)
A and n = constant

Modified Henderson Model

The Modified Henderson that includes the temperature parameter was used to study the sorption isotherm of chitosan films. The sorption data is suitable for water activity range between 0 and 0.9 with $r^2 = 0.9729$ [22]. The model is given by (10):

$$X = [-\frac{ln(1-a_w)}{A(B+T)}]^{\frac{1}{C}} \quad (10)$$

Where;
X= moisture content on dry basis (kg/kg dry solid)
a_w= water activity
T = Temperature
A, B and C = constants

Oswin Model

General Oswin Model

Oswin model is a three-parameter, semi-empirical model developed by Oswin in 1946 that dwells on an array of expansion for sigmoid shaped curves. It was observed that the Oswin Equation is commonly used to express the isotherms of starchy foods. Other than that, it gives an acceptable description of the isotherms of vegetables and meats. In a study of edible food film packaging made from xanthan gum, tapioca starch, and potassium sorbate, the Oswin model describes the sorption data adequately [24]. Another study by Enyinnaya Chinma, Chukwuma Ariahu [25], it was found that the GAB model and Oswin model perfectly describe the sorption data for the edible film made from cassava starch – soy protein concentrate blend with root mean square of error (%RMS) of less than 10 for the entire water activity range. The BET model was also used in this study to describe the sorption data. Even though the RMS (%) is also less than 10, it is only applicable for water activity less than 0.5 [6].

The model is given by (11):

$$\boldsymbol{M_w = A[\frac{a_w}{1-a_w}]^B} \quad (11)$$

Where;
M_w = Equilibrium moisture content (kg/kg dry solid)
A and B = constants

Modified Oswin Model

The modified Oswin was used to study cornstarch edible film incorporated with an antimicrobial agent. The modified Oswin has the best fit for the sorption isotherm data for temperatures of 25°C to 45°C. The values of r^2 for all data are near 1, and the values for mean relative percentage deviation modulus (MSE%) is narrow between 1.524 and 2.103 [26]. Thus, it is considered the best model when compared with the GAB and Peleg model. Modified Oswin is suitable to describe the sorption data for products containing high starch [26]. The Modified Oswin is given by (12)

$$\boldsymbol{X = (A + BT)[\frac{a_w}{1-a_w}]^{\frac{1}{C}}} \quad (12)$$

Where;
X = moisture content on dry basis (kg/kg dry solid)
T = Temperature
a_w = water activity
A, B and C = constants

For the chitosan films by Aguirre-Loredo, Rodriguez-Hernandez [16], the modified Oswin is suitable for water activity ranging from 0 to 0.9. The r^2 value is 0.9781, and the %E value is 9.6822. However, when compared the modified Oswin to the general model, the general model has lower r^2 and mean percentage error (%E) values for the same water activity range.

Peleg Model

Peleg developed an entirely empirical model that could describe the moisture sorption isotherms of food as good as the GAB model due to its four parameters model. Monolayer moisture content is not considered in this equation. An edible film made from rice starch – carboxymethyl cellulose (from Durian rind) blend was ideally described using the Peleg model. Compared to other models used in the study, the Peleg model has the highest r^2 value, and the lowest %RMS value for water activity range ranged between 0.11 and 0.85 [27]. Another study by Suriyatem, Rachtanapun [28] regarding edible film made from rice starch – carboxymethyl chitosan blend, the Peleg model proved to be the best model among others (GAB, BET, Lewicki, and Oswin) for the sorption data obtained with r2 ≈ 0.997 and the lowest %RMS value.

Peleg model is given by (13).

$$M_W = C_1 a_W^{C_3} + C_2 a_W^{C_4} \qquad (13)$$

Where;
M_w = Equilibrium moisture content (kg/kg dry solid)
a_w = water activity
C_1, C_2, C_3, and C_4 are constants; $C_3 < 1$ and $C_4 > 1$

Langmuir Model

Langmuir developed a simple empirical model that is used in physical and chemical adsorption. The model was developed with the following assumptions [17]:

1. There is only monolayer adsorption whereby there is only one layer of molecules that are absorbed on the absorbent surface
2. The surface of the adsorbent is homogenous, and there is orderly adsorption energy for all sites
3. Once all the site is occupied, adsorption will no longer occur
   With increasing distance, there will be loss of attractive intermolecular forces
4. There will not be any interaction between molecules adsorbed on the adjacent sites (ideal adsorbate behaviour)
5. There is localised adsorption on the surface

The equation of the Langmuir model is given by (14):

$$\boldsymbol{a_w} = \left(\frac{1}{M_w} - \frac{1}{M_0}\right) = \frac{1}{CM_0} \qquad (14)$$

Where;
M_w = Equilibrium moisture content (kg/kg dry solid)
M_0 = Monolayer moisture content (kg/kg dry solid)
a_w = Water activity
C = Constant

No study could be found on this model concerning edible food film packaging. However, there is a study of pear and garden mint leaves using this model for desorption study [9].

Freundlich Model

Freundlich model is an empirical equation that is suitable to use in multilayer adsorption. The model follows a few assumptions [17, 29]:

1. The adsorbent surface is heterogeneous, and there is exponential energy distribution for the active sites
2. Towards the end of the adsorption process, there is an exponential reduction in the adsorption energy as the more powerful binding sites are occupied first

The equation of the Freundlich model is given by (15):

$$q_e = K_f C_e^{\frac{1}{n}} \tag{15}$$

Where;
q_e = Amount of adsorbate per unit mass (mol/kg)
K_f = Adsorption coefficient
C_e = Equilibrium concentration of the adsorptive
n = correction factor

The value of slope 1/n is between 0 to 1, where it indicates heterogeneity. As the value approaches 0, the surface is more heterogeneous. When the value of the slope falls between 0 to 1, it shows good adsorption isotherm [17].

The application of the Freundlich model is mainly on organic compounds and to understand soil chemistry [29, 30]. There is no study of edible food film packaging using the Freundlich model. The major disadvantage of this model is the inability to estimate maximum adsorption [30].

SUMMARY OF MOISTURE SORPTION ISOTHERM STUDIES

Table 1: Summary of Moisture Sorption Isotherm Studies of Various Type of Food Packaging

No.	Polymer	Plasticiser/ Surfactant	Antimicrobial	Moisture Sorption Model	Reference
1	Pullulan	Alginate	-	GAB	[31]
2	Yuba (soybeans)	Water	-	Peleg, GAB	[32]
3	Balangu seeds	Glycerol	-	GAB, BET	[33]

4	Whey protein	Glycerol	-	Peleg, GAB, BET	[34]
5	Fish gelatin and sugars	Glycerol, Sorbitol -	-	GAB	[35]
6	Wheat gluten and lipid	Glycerol,	-	GAB	[36]
7	Pea starch and guar gum	Glycols, Sugars, Polyols	-	GAB, Peleg, Ferro-Fontan	[37]
8	Chitosan- Caseinate	Glycerol	-	GAB, Peleg	[38]
9	Peanut protein	Glycerin, Sorbitol, Propylene Glycol, Polyethylene Glycol	-	GAB, BET, Smith, Henderson	[39]
10	Cassava flour	Glycerol, Sorbitol, Polyethylene Glycol	-	GAB, Lewicki, Peleg, Oswin	[40]
11	Chitosan - PVA	-	-	BET, GAB, Halsey, Oswin, Smith	[21]
12	Corn starch	Glycerol	-	BET	[18]
13	Hydroxypropylmethyl cellulose	Tween85 (Surfactant)	Tea Tree Oil	GAB	[41]
14	Chitosan	Glycerol	-	Oswin, Smith, Halsey, Henderson, BET, GAB	[42]
15	Cassava starch	Glycerol, Sorbitol, Glycerol-Sorbitol	-	GAB	[43]
16	Gum tragacanth, Whey protein	Glycerol	-	GAB	[44]

Table 1 shows a summary of previous MSI studies for various types of edible food film packaging. The most used model was found to be GAB. The use of the GAB model to determine moisture sorption isotherm of food is very popular as it is applicable for a wide water activity range and variety of edible food film packaging.

CONCLUSION

In conclusion, BET, GAB, Halsey, Oswin, Henderson, and Peleg are among the known models used to determine the moisture sorption of edible films. Among these models, the GAB is widely used due to parameter have physical meaning. The moisture sorption is essential to determine the stability and shelf life of the film as food packaging. Modified models are not widely used in determining moisture sorption studies. Other than temperature, the concentration of polymer should be considered in moisture sorption isotherm studies. It is better to understand how the concentration of polymer relates to MSI.

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