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Modeling of Moisture Adsorption Isotherm of Selected Commercial flours of Bangladesh

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Abstract

The moisture adsorption behavior is a fundamental knowledge in the processing and storage of food materials. In this paper, the experimental adsorption behavior of wheat, rice and corn flours were compared with five widely recommended adsorption models (BET, GAB, Oswin, Smith and Halsey) in the literature. From the sorption data, monolayer moisture content of wheat, rice and corn flours were estimated as per BET and GAB models. For all flours, GAB model gave higher monolayer moisture content compare to the BET model. Linear regression analysis was done to determine the model constants where all models were compared using regression coefficient and standard error of the estimate. It was observed that there is no single model that could describe accurately the sorption isotherm over the whole range of water activity (a_w) and for all types of flours. BET model was found as the best model for the prediction of equilibrium moisture content (EMC) of the most stable region (i.e. EMC corresponding to a_w range below 0.52) but it was not suitable at a_w above 0.52. The GAB model was found to be the most suitable to predict the EMC for wide range of water activity (0.11 to 0.93).

Keywords: Wheat flour, rice flour, corn flour, sorption isotherm, water activity, equilibrium moisture content.

1. Introduction

An adsorption isotherm is an indispensable curve describing the relation of moisture mobility from an aquatic environment to solid phase environment at a fixed temperature and pressure. The mobility of moisture mainly depends on water activity (Fraction of equilibrium relative humidity), state and components of food (Doporto *et al.*, 2012). The research about water activity (a_w) came to the forefront as a major factor for controlling the spoilage of dried food and predicting the equilibrium moisture content (EMC) and shelf life of hygroscopic food products. EMC is the limiting moisture content of a food product after it has been exposed to a fixed a_w condition for an infinitely long time (Moreira *et al.*, 2010).

A number of theoretical, semi-theoretical and empirical models have been proposed for mathematical expression of the relation between water activity and equilibrium moisture content. In most cases, a specific model is suitable for certain food products, while, is not suitable for different ones. Specially, the model is only suitable to predict the equilibrium moisture content for a certain water activity range (Andrade *et al.,* 2011). Over the last few decades, a wide variety of moisture adsorption isotherm model has been formulated to predict the shelf life of foods. Among these models BET (Brunauer, Emmett and Teller), GAB (Guggenheim-Anderson-De Boer), Oswin, Smith and Halsey equations are generally used for most of the cereal and cereal products (Brett *et al.,* 2009; Al-Muhtaseb *et al.,* 2002 and Andrade *et al.,* 2011). Andrade *et al.* (2011) stated that the GAB model is the best method among all because it could be applied for a wide range of water activity (up to 0.93).

Rice, wheat and corn are the three major crops in Bangladesh (Leishman, 2013; Mahmud, 2013). Consumption of wheat flour, as well as rice and corn flour, is increasing day by day due to the change in food habit and lifestyle (Islam and Haque, 2011; Mahmud, 2013). As per time demand, Bangladesh needs enough knowledge, skills and technology for storage of hygroscopic food items like flours. Furthurmore, no information was found about the modeling of adsorption data of Bangladeshi commercial wheat, rice and corn flours.

Durakova and Menkov (2004) determined the adsorption and desorption behaviors of rice flour in a temperature range between 10 and 30 °C. For rice flour, they found modified Chung–Pfost and Oswin models for better description of the relationship between the equilibrium water content, the a_w, and the temperature. Stripatrawan and Jantawat (2006) studied the sorption characteristics of Jasmine rice crackers and concluded that the GAB model gave the best fit over a wide range of water activity. Moreira *et al.* (2010) found the GAB and Chung-Pfost as suitable models for predicting adsorption of chestnut and wheat flours. The GAB and modified Oswin models were found to be acceptable for prediction of moisture adsorption characteristics of maize flour (Oyelade *et al.,* 2008).

Based on above information the specified objectives of this study are:

i). Modeling of moisture adsorption data imployed by different model equations.
ii). Finding the best model based on the validity of the models by experimental data.

2. Materials and Methods

2.1 Raw materials

Locally available commercial wheat flour (Bosundhora atta), corn flour (Noor-Nobi) and milled rice flour (BRRI-28 variety) purchased from the local market of Bangladesh Agricultural University campus, were used in this study. Moisture contents (As per AOAC, 1990) of raw samples were 0.1383 $kg.kg^{-1}$(d.b) for wheat flour, 0.138 $kg.kg^{-1}$(d.b) for rice flour and 0.1286 $kg.kg^{-1}$(d.b) for corn flour.

2.2 Determination of EMC

The EMC of selected flours at different humid condition was determined by the static gravimetric method at temperature 30 °C. Each flour, with a triplicate sample, were put into the ceramic crucible and placed inside the glass desiccator. The saturated salt solutions were used to maintain a specific water activity inside the desiccator. The saturated salt solution of LiCl, $KC_2H_3O_3$, $MgCl_2 6H_2O$, K_2CO_3, $Mg(NO_3)2.6\ H_2O$, $CaCl_2$, NaCl, KCl and KNO_3 were used to maintain a specific a_w of 0.11, 0.2, 0.33, 0.44, 0.52, 0.68, 0.75, 0.85 and 0.93 respectively. The Genlab Incubator (Model M75CPD) was used to maintain desiccator temperature 30°C with deviation ±1°C. When the constant weight of the sample was reached, within maximum 4 weeks, the equilibrium moisture content (EMC) of the samples was determined by AOAC (1990) method.

2.3 Modeling of sorption data

The experimental adsorption data obtained corresponding to the water activity was fitted to the following models.

2.3.1 Brunauer-Emmett-Teller (BET) Model

The BET equation (equation 1), proposed by Brunauer, Emmett, and Teller (1938), is most widely used to characterize food isotherms. This equation has only one constant, C (related to the net heat of sorption). The BET model usually fits the experimental sorption data at water up to 0.52 (Islam, 1980).

$$m = \frac{m_0 C a_w}{(1-a_w)(1+(C-1)a_w)} \quad \text{........ (1.a)}$$

This equation can be rearranged as follows

$$\frac{a_w}{(1-a_w)m} - \frac{1}{m_0 C} + \frac{(C-1)}{m_0 C} a_w \quad \text{.......................... (1.b)}$$

Where, m_0 and m are the monolayer moisture content and EMC, respectively.

2.3.2 Guggenheim, Anderson and de Boer (GAB) Model

The GAB model (Guggenheim, 1966) takes into account the different properties of water in the multilayer region. The model is shown in equation 2.

$$m = \frac{m_o CKa_w}{\{(1-Ka_w)(1-Ka_w+CKa_w\}} \quad \text{.............................. (2)}$$

Where, m_0 is the monolayer moisture content (d.b), C is constant and K is the correcting factor (where $0 < K \leq 1$). Mathematically, BET equation is a special case of GAB with K equal to 1.0. (Andrade *et al.,* 2011).

Equation (2) can be converted into polynomial form as follows (Schär and Rüegg, 1985)

$$\frac{a_w}{m} = \alpha a_w^2 + \beta a_w + \gamma \quad \text{..................... (3)}$$

Where,

$$\alpha = \frac{K}{m_0}\left(\frac{1}{c} - 1\right)$$

$$\beta = \frac{1}{m_o}\left(1 - \frac{2}{c}\right)$$

$$\gamma = \frac{1}{m_0 C.K}$$

The constants α, β and γ are obtained from a plot of $\frac{a_w}{m}$ *Vs* a_w as 2nd order polynomial line.

The GAB model parameters (m_0, C and K) are then obtained by solving the following equations (Blahovec and Yanniotis, 2010)

$$\gamma.K^2 + \beta K + \alpha = 0$$

$$C = \frac{\beta}{\gamma.K} + 2$$

$$m_0 = \frac{1}{\beta + 2K.\gamma}$$

2.3.3 Oswin model

Owsin mode (Oswin, 1946) consists a series expansion for the sigmoid shaped curve. The model is expressed as follows:

$$m = C\left[\frac{a_w}{1-a_w}\right]^n \quad \text{......................... (4)}$$

Equation (4) can be rearranged as

$$\ln m = \ln C + n.\ln\left(\frac{a_w}{1-a_w}\right) \quad \text{... .... (5)}$$

The value of C and n are determined by the linear regression of $\ln m$ versus $\ln\left(\frac{a_w}{1-a_w}\right)$

2.3.4 Smith model

Smith (1947) suggested that sorption isotherms of biopolymers could be represented as

$$m = k_2 - k_1 \ln(1 - a_w) \quad \text{........... (6)}$$

Where, m is the EMC, k_1 and k_2 are the smith constants.

From a linear regression of m versus $\ln(1-a_w)$ the Smith constants were computed. The smith equation has been found to fit the experimental isotherms of biopolymers and food products with a_w of 0.3 to 0.5 and higher (Heldman *et al.*, 2006).

2.3.5 Halsey model

Halsey equation (1948) is a good representation of adsorption data regarding isotherms type I, II, or III. The model is expressed as

$$a_w = \exp\left(-\frac{A}{RT\theta^r}\right) \quad \text{.................. (7)}$$

Where, A and R constant and $\theta = m/m_o$

The Halsey equation was modified by Chirife and Iglesias (1978) as the following form

$$\ln m = a + b\ln(-\ln a_w) \quad \text{............ (8)}$$

Where, a and b constants can be estimated from a linear plot of ln(m) versus ln (-lna_w)

The accuracy of each moisture adsorption model was justified by using different indicators namely standard error of estimate (SEE), and regression coefficient (R^2). These indicators are defined as follows

$$SEE = \sqrt[2]{\frac{(EMC_{mea} - EMC_{Pre})^2}{n}} \quad \text{.......... (9)}$$

$$R^2 = 1 - \frac{RES}{TES} \quad \text{......................... (11)}$$

Where, EMCmea., EMCpre. and TES are the measured EMC, the model predicted EMC and total error sum of squares respectively, where n is the number of the data point.

3. Results and Discussion

The mean value of measured EMC and standard deviation for different water activity is shown in Table 1. Adsorption isotherm according to the GAB model shown in Figure 1. From Table 1 and Figure 1, for all flour, it can be seen that the EMC values increase with the increase of water activity. The moisture isotherms of commercial wheat, rice and corn flours at wide range of temperatures are well described by Ahmed and Islam (2018). The published adsorption trend of wheat flour (Moreira *et al.*, 2010), rice flour (Brett *et al.*, 2009) and corn flour (Oyelade *et al.*, 2008) were found similar to our result with little higher moisture adsorption trend of corn flour specially at water activity more than 0.75.

Table 1. Equilibrium moisture content, m (% d.b) of wheat, rice and corn flour obtained by adsorption at different water activities (a_w) at 30°C temperature

a_w	Wheat flour		Rice flour		Corn flour	
	m*	s.d	m*	s.d	m*	s.d
0.11	8.45	0.11	10.5	0.09	9.35	0.04
0.2	9.87	0.12	11.45	0.06	10.23	0.08
0.33	9.89	0.09	12.32	0.10	11.87	0.18
0.44	10.56	0.12	12.89	0.12	13.08	0.15
0.52	10.67	0.05	12.9	0.08	14.03	0.16
0.68	13.68	0.04	14.76	0.17	16.97	0.12
0.75	15.78	0.14	17.6	0.15	21.76	0.16
0.85	23.04	0.17	26.78	0.08	31.86	0.18
0.93	32.67	0.32	36.98	0.17	42	0.45

*Mean of three replications, s.d.= Standard deviation

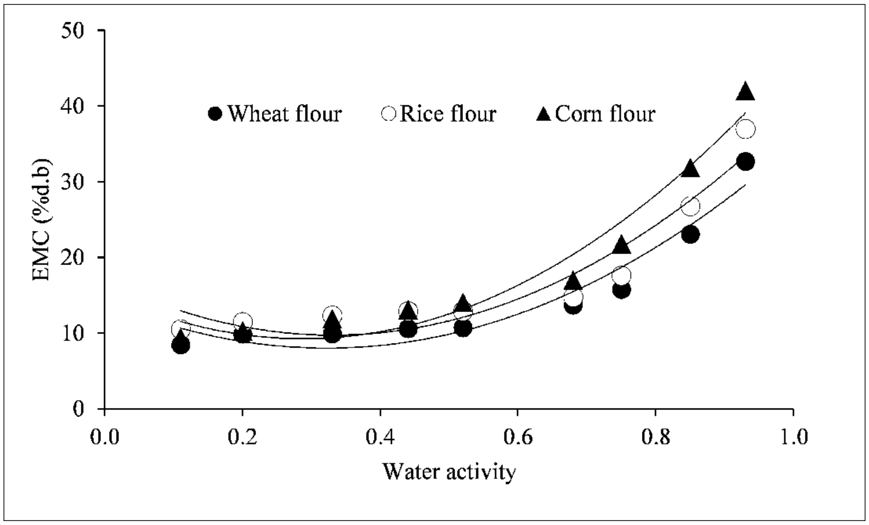

Figure 1. Adsorption isotherm of wheat, rice and corn flour according to GAB model

The monolayer moisture content (m_0) of different flours, at room temperature (30°C), was estimated using the BET and GAB models, shown in Table 2. Estimation of monolayer moisture content is very important because it is preferable to have moisture content at or slightly above monolayer value for extending the shelf life of cereal grains in storage (Blahovec and Yanniotis, 2010). From the BET model, monolayer moisture content of wheat, rice and corn flour were found to be 0.0512, 0.0623 and 0.0677 g water/g dry solid respectively. It can be seen from Table 2 that, GAB model gave the higher amount of monolayer moisture content which is 0.0567, 0.0667 and 0.0738 g water/g dry solid for the flour from wheat, rice and corn respectively. Similar observation for BET and GAB monolayer moisture content was found by several researchers (Ajisegiri *et al.,* 1994; Andrade *et al.,* 2011 and Brett *et al.,* 2009). The monolayer moisture contents of rice flour, for temperature range 5 to 50°C are 0.080–0.064 g water/g dry solids (BET model) and 0.111–0.065 g water/g dry solids (GAB model) reported by Brett *et al.* (2009). The calculated monolayer moisture content of wheat and corn flours are quite similar found by Moreira *et al.,* (2010) and Oyelade *et al.,* (2008). From the comparative study, it was found that GAB energy constant is higher than the BET energy constant for all the flours. From Table 2, it is seen that the wheat flour gave the lowest BET energy constant (14.20) where rice flour gave lowest GAB energy constant (18.52). The higher GAB energy constants (also called Guggenheim constant, C) determined for different flour at different temperature compared to those determined from BET are good agreement with Islam (1980) and Moreira *et al.,* (2010).

Without BET and GAB, other models do not have monolayer incorporation. Along with BET and GAB, the calculated model parameters of Oswin, Smith, and Halsey Model are given in Table 2. The calculated fitting parameters of the mentioned models of this study are in good agreement with several authors (Al-Muhtaseb *et al.,* 2002; Andrade et *al.,* 2011). For example, Gálvez *et al.,*(2006) reported fitting parameters of Oswin model (C= 0.106, n= 0.299), Smith model ($k_1 = 0.84$ and k_2=0.83) and Halsey model (a= 2.516 and b=0.002) which are supporting our findings.

Table 2. Fitting parameters of sorption models

Model	Parameters	Wheat flour	Rice flour	Corn flour
BET	m_0	0.0512	0.0623	0.0677
	C	14.14	14.20	25.03
GAB	m_0	0.0567	0.0667	0.0738
	C	20.82	18.52	40.31
	K	0.87	0.86	0.89
Oswin	C	0.0936	0.1071	0.1185
	n	0.1628	0.1617	0.1789
Smith	k_1	0.1179	0.1330	0.1537
	k_2	0.0074	0.0102	0.0129
Halsey	a	2.4702	2.3255	2.1989
	b	0.5225	0.512	0.5267

Table 3. Statistical parameters showing the degree of fitness of the models with experimental data

Model	Applied a_w range	Constants	Sample Wheat flour	 Rice flour	 Corn flour
BET	0.11-0.52	R^2	0.9755	0.9724	0.9892
		SEE	0.0064	0.0056	0.0031
GAB	0.11-0.93	R^2	0.9585	0.9196	0.9606
		SEE	0.0092	0.0132	0.0101
Oswin	0.52-0.85	R^2	0.9845	0.9974	0.9821
		SEE	0.0490	0.0197	0.0579
Smith	0.52-0.93	R^2	0.9874	0.9737	0.9845
		SEE	0.0114	0.0187	0.0165
Halsey	0.52-0.93	R^2	0.9945	0.90	0.9771
		SEE	0.0378	0.038	0.0784

R^2 = Regression co-efficient, SEE = Standard Error of Estimate

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Comparison of sorption models based on statistical parameters is shown in Table 3. From the data it is seen that R^2 values of BET model are almost same for wheat flour (0.9755), rice flour (0.9724) and corn flour (0.9892) while the corresponding SEE values are 0.0064, 0.0056 and 0.0031. Thus the BET, which represents a_w up to 0.52 gives the best fit for corn flour followed by rice flour and wheat flour. Similarly, GAB model which predicts EMC up to a_w of 0.93 gives the best fit for wheat flour (least SEE Value) and is followed by corn flour and rice flour. The range of R^2 values being 0.9196 to 0.9606 and SEE value being 0.0092 to 0.0132 for GAB model, as the predictor of EMC in the

a_w range of 0.11 to 0.93, would be highly acceptable. The Smith and Halsey models were tested in the a_w range of 0.52 to 0.93 and Oswin model in the a_w range of 0.52 to 0.85 as these were not applicable in the lower (below 0.52) a_w range. For corn flour, however, Smith equation undoubtedly gives the best fit and successively followed by Oswin and Halsey model. On the other hand, for rice flour, Oswin model gives best fit and is closely followed by Smith while Halsey gives the lowest fitness as considered with respect to R^2 and SEE value (R^2= 0.90 and SEE= 0.038). The GAB model was reported as the best fit at wide range of water activity by many researchers (Andrade *et al.*, 2011; Brett *et al.*, 2009 and Oyelade *et al.*, 2008) while BET for a_w below 0.50 (Andrade *et al.*, 2011; Gálvez *et al.*, 2006).

Thus, no single equation describes accurately the sorption isotherm over the whole range of relative humidity and for all types of food materials. Labuza (1975) noted that no sorption model could fit data over the entire range of relative humidity because water is associated with the food matrix through different mechanisms in different water activity regions. BET model is undoubtedly the best model for the prediction of EMC of the most stable region (a_w below 0.52) but is not enable at higher a_w range (above 0.52). The GAB model is quite acceptable for the prediction of EMC at higher water activity range (from 0.11 to 0.93). So, GAB is the best model for predicting EMC for all selected flours in water activity range from 0.60 to 0.65, as the range considered as the stable zone for most dried products.

4. Conclusions

The five sorption models (BET, GAB, Oswin, Smith and Halsey) were used to predict the equilibrium moisture content at different water activities. From the moisture sorption isotherm data, the monolayer moisture content was estimated by BET and GAB model. It was found that the GAB equation gave much higher value of monolayer moisture content compared to those for all flour types obtained from BET equation. No single equation describes accurately the sorption isotherms over the whole range of relative humidity for all types of flours. However, based on the study the GAB model was found to be more appropriate to predict the EMC according to statistical parameters for the wide range of water activity.

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