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Effect of drying methods on the sorption isotherms of plantain flour

^DGBEMISOLA JAMIU FADIMU^{1,2*}, LATEEF OLADIMEJI SANNI¹, ABDUL-RAZAQ ADESOLA ADEBOWALE¹, OLAJIDE PAUL SOBUKOLA¹, PETRAL BOLA ABDULSALAM-SAGHIR³, SEMIU ABOLORE RASAQ¹, MONILOLA KUDIRAT ADENEKAN⁴

¹Department of Food Science and Technology, Federal University of Agriculture, Abeokuta, Nigeria ²Department of Food Science and Nutrition, King Saud University, Riyadh, Saudi Arabia. ³Department of Agricultural Extension and Rural Development, Federal University of Agriculture, Abeokuta, Nigeria. ⁴Department of Food Technology, Moshood Abiola Polytechnic, Abeokuta, Nigeria

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ABSTRACT

The demand for plantain flour has increased in recent years due to its health benefits and industrial applications, however, there is dearth of information on the effect of drying methods on the sorption isotherm of plantain flour. In the present study, moisture sorption properties for unripe plantain flour using four drying methods at temperatures 27 °C, 37 °C and 42 °C were determined for water activity 0.10 to 0.80. The data generated were modelled using Peleg, GAB, Oswin, BET and Langmuir. The results revealed that the Equilibrium Moisture Contents decreased with an increase in temperature at all the water activities considered. Also, moisture isotherms of the plantain flour were not significantly (p>0.05) affected by temperature. The coefficient of determination (R^2) for the used models ranged from 0.640 and 0.986 and Peleg was the most appropriate for the adsorption isotherm of plantain flour out of all the models. The monolayer moisture content of the flour also showed that all the unripe plantain flours could be stored for longer periods at all the temperatures studied, as their Mo falls within acceptable range for storage.

Introduction

Plantains (Musa paradisiaca) is one of the important staple food crops consumed in the tropics right after rice, wheat and maize and is obtainable in about 120-130 tropical countries in the world (Kawongolo, 2013). It is an essential food crop in Sub-Saharan Africa, which serves as a source of nutrient and household income for many people around the world (Kawongolo, 2013). The aggregated world production is put at over 76 million metric tons (Olumba, 2014), out of which over 12 million metric tons are harvested yearly in Africa (Fakayode et al., 2011). Sizeable tons of plantains are annually harvested in Nigeria and Nigeria is the biggest producer of plantains in West Africa with an estimated production of about 2.7 million metric tons, majority of which are produced and harvested in the southern part of the country (FAO, 2009). In spite of large tons of plantains harvested yearly in Nigeria, Olorunda and Adelusola (1997) reported that over 50% of plantains harvested are lost due to unavailability of appropriate storage facilities to prevent postharvest losses. Usually harvested at matured, but unripe stage, plantains undergo rapid respiration after harvest, making it a short-lived agricultural product that requires urgent attention immediately after the harvest. Plantain may therefore be processed into flour when it is matured but not ripe. Traditionally, sun drying is the most common method used in processing plantains into flour. However, there are some problems associated with sun drying such as slowness of the process, uncertainty of the weather and uneven drying (Arinola et al., 2016). Falade (2009) reported that drying is one of the best methods in terms of cost efficiency to preserve plantain flour in order to have a plantain product with considerable shelf stability. In Nigeria, plantain flour is usually processed into a gruel called amala and it is often recommended for people with diabetes because of its low glycemic index (Falade,



^{*}Corresponding author E-mail: fadimugbemisola@gmail.com

2009). Drying is an ancient method used in preserving agricultural produce and different studies on the use of drying for different agricultural products have been conducted. Drying is basically done to reduce moisture to a level where enzymatic and microbial activities are minimized to confer preservation of a product (Kawongolo, 2013). According to Arinola et al. (2016), different drying methods have been used for drying plantain fruits with some success.

Understanding the thermodynamic relationship between equilibrium moisture content and water activity is very important in food systems, as it helps in the fabrication and design of appropriate drying equipment and the most suitable storage conditions (Ahmed et al., 2005). This understanding is needed for making appropriate predictions about changes that may likely occur during storage of the dehydrated products (Panjagari et al., Water sorption isotherm which provides 2015). information about the fundamental relationship between EMC and water activity of dehydrated food products at a given temperature is very important in the field of food science. Johnson and Brennan (2000) reported water sorption isotherms of plantain flour at different temperatures. Khawas and Deka (2017) also reported moisture sorption isotherms of a culinary banana at five different experimental temperatures. However, different studies on the moisture sorption isotherm of starch based agricultural products have been conducted, including plantain and banana. Nonetheless, information on the effect of different drying methods on water sorption isotherm of plantain flour are rare. The objectives of this paper are to evaluate the effect of drying methods on the moisture sorption isotherm of plantain flour and to model the experimental data with appropriate models.

Materials and methods

Materials

For this study, matured unripe plantain fruits of the same variety were procured from the plantain plantation of Federal University of Agriculture (FUNAAB), Abeokuta, Nigeria. Plantain (*Musa paradisiaca*). Maturity level was carefully selected, while the stage of ripeness was determined using colour chart. The used potassium metabisulphite was purchased from Libertas Laboratory in Abeokuta, Nigeria.

Plantain flour preparation

The matured unripe plantains were processed in the food processing laboratory of Food Science and Technology Department, FUNAAB, Abeokuta, Nigeria. The plantains were first washed, peeled and sliced using a plantain slicer. The plantain slices were then blanched using hot water at 50 °C for 7 min and sulphited using 1% potassium metabisulphite concentration. The pretreated samples were then dried using cabinet, solar, sun and oven drying methods. They were then milled and stored in ziplock bags (Fadimu et al., 2018). The plantain flours were coded as: CAPF = Cabinet dried plantain flour, SOPF = Solar dried plantain flour, SUPF = Sun dried plantain flour and OVPF = Oven dried plantain flour. *Cabinet drying*: The samples were dried using a cabinet dryer (LEEC Limited, Serial No 3114, Nottingham United Kingdom) for 24 h at 60 °C.

Oven drying: The samples were dried using a Gallenkamp oven (Model OV-160 size two BS Gallenkamp) for 24 h at $60 \,^{\circ}$ C.

Solar drying: Direct natural convection solar dryer designed and constructed in FUNAAB was used for this study. It is constructed with bricks on the sides and a transparent glass covering the top inclined at an angle of 10° to face the equator. The interior of the solar house is painted black to improve absorption of heat. The plantain samples were dried in the solar house at 50 °C for 48 h, the temperature of the solar house was constantly measured using thermometer.

Sun drying: The samples were kept in the sun between 10:30 am to 5:00 pm daily and were dried to constant weight for three days.

Equilibrium moisture content (EMC) determination

Equilibrium Moisture Content (EMC) of the plantain flours was determined using a static gravimetric method as described by Oyelade et al. (2001) and Famurewa et al. (2012). The samples were dehydrated in the hot air oven (Model OV-160 size two BS Gallenkamp) at 105±5 °C for 8 h (AOAC, 1990). Duplicate samples, 3.00±0.001 g each were weighed into moisture pans in the desiccators. Concentrated sulphuric acid quantities used to make up a 250 mL of desiccant with deionized water were prepared at 27, 37 and 42 °C, using water activity and temperature tables of Perry and Green (1984). The acid was dispensed into the dessicators according to their respective water activities (Table 1). The dessicators were held at water activity 0.1 and 0.8, and then placed in a Gallenkamp (England) incubator (Model M75CPD) to maintain the required temperature level (27, 37 and 42 °C). Each of the samples was weighed every day using a digital balance until constant weight was obtained in three consecutive recordings and the samples were presumed to be at equilibrium (± 0.001 g). The equilibration time ranged between 10 and 14 days, depending on the water activity in each of the desiccators; those at higher water activities attained equilibrium faster than those at lower water activities.

Water activities	Quantity of Conc. H ₂ SO ₄ /250mL water for 27	Quantity of Conc. H ₂ SO ₄ /250mL water for 37	Quantity of Conc. H ₂ SO ₄ /250mL water for 42		
0.00	<u>-C (IIIL)</u>	<u>- (IIIL)</u>	<u>-C (IIIL)</u>		
0.80	71.29	71.29	/1.13		
0.70	86.28	86.79	86.92		
0.60	99.36	100.71	101.07		
0.50	111.65	112.72	113.15		
0.40	123.30	124.55	125.11		
0.30	135.92	137.30	137.91		
0.20	149.08	150.35	150.94		
0.10	167.61	168.99	169.63		

Table 1. Desiccant preparation for 27 °C, 37 °C and 42 °C

Modelling of sorption isotherm

There are various models available for predicting sorption isotherms of foods. In this research, Brunauer-Emmitt-Teller (BET), Guggenhiem-Anderson-deBoer (GAB), Peleg, Oswin and Langmuir equations were used (Table 2). The selection of these models was based on the report of Ajisegiri et al. (2007) about their appropriateness to correctly predict moisture sorption isotherm of high carbohydrate foods. Data Fit (version 9.0.59) was used to calculate the various parameters of the models and appropriateness of each model was evaluated using coefficient of determination (R²) and mean relative percentage deviation (E%) expressed as:

$$RMSE = \sqrt{\frac{\sum (M_{exp} - M_{cal})^2}{n}}$$
(1)

$$\%E = \frac{100}{n} \sum \left| \frac{M_{exp} - M_{pred}}{M_{exp}} \right| \tag{2}$$

where M_{exp} and M_{cal} were determined and experimental values of EMC and n is the number of experimental values.

Determination of net isosteric heat of sorption and differential entropy

Heat of sorption (Q_{st}) of the plantain flours for specific moisture content was estimated from Clasius-Clapeyron equation given by Tsami (1991) as:

$$q_{st} = -R[d\frac{\ln a_w}{\left(\frac{1}{T}\right)}] \tag{3}$$

where a_w is a measure of the water activity, R is the gas constant and T the absolute temperature in K.

Net isosteric heat of sorption was determined from the slope of the plot of In a_w versus 1/T at constant moisture content. The differential entropy, also called sorption entropy (S_d) of moisture isotherm, was determined from Gibbs-Helmholtz equation given as follows:

$$G = RTIn(a_w) \tag{4}$$

$$S_d = \frac{(Q_{st} - G)}{T} \tag{5}$$

where G is the Gibbs free energy measured in (kJ/mol). Putting equation (4) into equation (5), the equation becomes:

$$In(a_w)|_x = \frac{Q_{st}}{RT} - \frac{S_d}{R}$$
(6)

plotting In (a_w) versus the inverse of temperature at constant moisture content S_d gives (S_d/R) , which can be calculated from the intercept.

Table 2. Linear forms of the sorption models used to fit the experimental values

Models	Equations	References			
GAB	$M = \frac{M_0 b a_w}{1 - c a_w 1 - c a_w + b a_w}$	Mc Kenna, 1984			
BET	$M_{W} = \frac{C a_{W}}{(1 - a_{W})(1 + (C - 1)a_{W})}$	Brunauer et al. 1938			
Oswin	$M = C(a_w/1 - a_w)^n$	Oswin, 1946			
Peleg	$M = C_1 a_w c^3 + C_2 a_w c^4$	Kaya et al. 2005			
Langmuir	$M = \frac{C m_0^2 a_w}{M_0 (1 + a_w)}$	Mc Kenna, 1984			

GAB = Guggenheim, Anderson and de Boer equation, M = equilibrium moisture content (%, dry basis); a, b, c = constant parameters; $T = temperature (^{\circ}C)$, $a_w = water activity$, $M_o=monolayer moisture content$.

Results and discussion

Moisture sorption isotherm as affected by temperature and drying methods

Understanding the concept of moisture sorption isotherm is required for predicting the shelf life and critical moisture content determination for acceptability and storage of food products (Alakali and Satimehin, 2007). Moisture sorption isotherm data for the plantain flour at different experimental temperatures 27, 37 and 42 °C are indicated in Table 3. The sorption analysis was replicated and the values displayed in Table 3 are the means of these replicates. As shown in Table 3, the equilibrium moisture content of plantain flour for different drying methods decreased with an increase in temperature for all the water activity range studied. In addition, the EMC of plantain flour from different drying methods differs for a particular temperature and water activity. To test whether there is a significant difference in the values of sorption isotherm data at different temperatures, one-way ANOVA was used and the results as presented in Table 4 revealed that the sorption data were not significantly affected by temperature at both 0.05 and 0.01 significant levels.

Effects of drying methods on the sorption isotherms of plantain flours at 27, 37 and 42 °C are shown in Figures 1, 2 and 3. The sorption curves showed that the equilibrium moisture contents of plantain flour decreased as water activity increased at a constant temperature. A similar trend has been reported by Moreira et al. (2008). It may be observed from the figures that drying methods, as well as storage temperature, had varying impact on the behaviour of the plantain flour to absorb and retain water during storage could decrease with an increase in the storage temperature. The reduction in EMC with the temperature rise could mean that when the temperature is high, the energy of the water molecules may change to higher energy levels and this leads to breaking of the

water binding sites of foods (Owo et al., 2016). Sanni et al. (1997) reported a comparable result for temperature effects on lafun and yam flour (elubo). In addition, the observed differences in the EMC of the unripe plantain flours may also be attributed to the differences in drying methods used during their processing.

Table 3 shows the sorption data at different water activities and the results obtained are presented in Table 5. The parameters of various sorption models used for the sorption data revealed that the experimental adsorption isotherms of plantain flour were satisfactorily depicted by Peleg, GAB, BET and Oswin models owing to their higher coefficient of determination (R^2) values and low root mean square error (RMSE). However, the use of R^2 and RMSE does not suggest that the models precisely fit the experimental data. Therefore, the use of monolayer moisture content (M_o) estimate was necessary to make conclusive judgement. The GAB, BET and Langmuir monolayer moisture content (M_o) of all the unripe plantain flours varied between 0.77 - 7.50 kg water/kg dry solids and were within the range for storage stability (<10% dry basis) according to Labuza et al. (1985). Monolayer moisture content is the lowest amount of water covering the hydrophilic sites on the surface of food materials and the parameter is required to accomplish storage with little quality loss for a considerable period of time. Hence, the most appropriate temperature for storing food at a particular water activity is the temperature that is equivalent to that of monolayer moisture.

Moisture sorption isotherms of plantain flour indicated that all the samples absorb more moisture at 27 °C than at 42 °C, which implied that the samples could be stored longer at temperature above room temperature (25±2 °C). It was also observed that the Moisture Adsorption Isotherm of all the unripe plantain flours had a sigmoid shape, which is typical of type II isotherm curve classified by Brunauer et al. (1938). Comparable reports for sorption curves have been described in literatures for various starchy foods (Samapundo, 2007).

Table 3. Experimental sorption isotherm data for plantain flour from different drying methods

Sorption data for plantain flour												
	CAPF			SOPF		OVPF						
	27 °C	37 °C	42 °C	27 °C	37 °C	42 °C	27 °C	37 °C	42 °C	27 °C	37 °C	42 °C
0.1	0.9	0.62	0.53	4.3	2.49	0.42	3.01	0.63	0.1	6.13	3.26	0.19
0.2	3.7	2.59	0.69	5.36	3.16	0.63	3.53	2.63	0.62	7.5	4.09	0.71
0.3	6.48	4.68	1.1	5.87	4.02	0.89	5.92	3.82	1.56	7.86	5.89	0.83
0.4	9.15	5.86	1.46	7.74	5.63	1.8	7.87	5.89	2.3	9.4	7.82	1.56
0.5	10.16	7	2.44	8.55	6.84	3.01	8.31	7.02	2.97	9.71	8.84	2.68
0.6	10.98	8.9	3.49	10.81	8.23	3.86	10.2	8.54	4.12	10.74	11.23	3.89
0.7	11.76	10.92	4.91	12.85	9.99	5.86	11.73	10.14	5.76	11.53	13.56	5.39
0.8	17.76	13.9	7.71	13.6	13.17	7.1	14.49	11.5	7.13	13.84	14.56	7.11

*Readings are average of 3 replicates, CAPF: Cabinet dried plantain flour; SOPF: Solar dried plantain flour; OVPF: Oven dried plantain flour; SUPF; Sun dried plantain flour



Table 4. Analysis of variance for the sorption isotherm data

Fig 1. Effect of drying methods on the sorption isotherm of plantain flour at 27 °C



Fig 2. Effect of drying methods on the sorption isotherm of plantain flour at 37 °C



Fig 3. Effect of drying methods on the sorption isotherm of plantain flour at 42 °C

Thermodynamics properties

Effect of drying methods on net isosteric heat of sorption and differential entropy

Figure 4 shows the net isosteric heat of sorption of plantain flour as affected by drying methods. The slope of the plot between the values of ln (a_w) and 1/T at specific moisture content was used to calculate the net isosteric heat of sorption. The net isosteric heat of sorption decreased exponentially when the moisture content increased for cabinet, solar, sun and oven dried plantain flours. The reduction in the heat of sorption with an increment in absorbed water in plantain flours showed that water-solid exchange is more cohesive at the reduced moisture content, which justifies the increase in the exchange of energy between the water molecules and the plantain flour monomolecular layer (Owo et al., 2016). Consequently, the energy needed to separate the water molecules from the plantain

flour is large, compared to when the force of the attraction between the water and the plantain flour is low. The differential entropy was computed from the intercept of In a_w against 1/T at constant moisture content (Owo et al., 2016). A forceful connection existed between differential entropy, which was similar with what was observed in the net isosteric heat of sorption. The differential entropy decreased as the moisture content increased in all the plantain flour samples. Differential entropy is a measure of how much a material is disordered. Sorption entropy of a material is related to the amount of available sorption sites at any particular energy level (Moreira et al. 2008). Reduction in the moisture content prevents the movement of water in the sample as a rearrangement of the internal molecules grows. Fasina (2006) reported a comparable result for the sweet potato flour (Owo et al., 2016).



Fig 4. Effect of moisture content on the net isosteric heat of sorption (Qst) for plantain flour



Fig 5. Effect of moisture content on differential entropy (Sd) of plantain flour

		CAPF		SOPF			OVPF						
		27 °C	37 °C	42 °C	27 °C	37 °C	42 °C	27 °C	37 °C	42 °C	27 °C	37 °C	42 °C
PELEG	А	4.71	6.75	8.25	7.46	3.49	5.77	6.29	0	4.1	5.44	0.04	3.35
	В	4.71	2.06	2.28	0.65	8.74	0	6.5	6.96	2.93	0.52	8.6	4.85
	С	1.12	2.07	6.92	1.17	0.53	2.27	0.67	4.46	5.83	0.28	1.33	1.49
	D	1.12	0.12	1.02	0.36	4.76	0.98	6.86	1.04	1.22	6.42	1.13	6.74
	\mathbb{R}^2	0.946	0.984	0.989	0.973	0.988	0.982	0.989	0.988	0.962	0.995	0.968	0.988
	RMSE	0	0	0	0	0	0	0	0	0	0	0	0
GAB	А	2.57	6.31	3.35	15.68	13.63	0.11	8.59	3.24	1.48	10.47	5.43	0.76
	В	0.57	0.83	0.99	0.71	0.93	0.98	0.74	0.78	0.82	0.59	0.69	0.8
	С	5.83	2.21	0.77	2.94	1.59	1.98	2.91	4.97	1.51	3.28	3.51	2.14
	\mathbb{R}^2	0.947	0.979	0.989	0.955	0.988	0.974	0.989	0.975	0.96	0.989	0.956	0.987
	M_0	5.83	2.21	0.77	2.94	1.59	1.98	2.91	4.97	1.51	3.28	3.51	2.14
	RMSE	0	0	0	0	0	0	0	0	0	0	0	0
BET	А	0.73	0.97	0.17	1.47	0.5	0.19	1.06	0.88	0.25	5.52	0.87	0.19
	В	7.5	5.97	4.4	6.1	6.08	4.36	6.37	5.54	3.85	5.42	6.73	4.27
	\mathbb{R}^2	0.932	0.881	0.959	0.871	0.894	0.985	0.927	0.963	0.948	0.777	0.926	0.981
	M_0	7.5	5.97	4.4	6.1	6.08	4.36	6.37	5.54	3.85	5.42	6.73	4.29
	RMSE	0	0	0	0	0	0	0	0	0	0	0	0
OSWIN	А	3.9	3.53	1.2	4.05	2.86	1.21	3.82	3.14	1.28	4.57	3.82	1.19
	В	0.51	0.42	0.78	0.35	0.53	0.78	0.42	0.44	0.7	0.22	0.44	0.78
	\mathbb{R}^2	0.928	0.98	988	0.951	0.985	0.954	0.985	0.956	0.954	0.99	0.949	0.982
	RMSE	0	0	0	0	0	0	0	0	0	0	0	0
LANG.	А	3.75	3.47	2.26	3.66	3.22	2.27	3.61	3.3	2.27	3.78	3.63	2.25
	В	3.75	3.47	2.26	3.66	3.22	2.27	3.61	3.3	2.27	3.78	3.63	2.25
	\mathbb{R}^2	0.858	0.88	0.655	0.907	0.796	0.645	0.923	0.914	0.711	0.525	0.888	672
	M_0	3.75	3.47	2.26	3.66	3.22	2.27	3.61	3.3	2.27	3.78	3.63	2.25
	RMSE	0	0	0	0	0	0	0	0	0	0	0	0

Table 5. Statistical parameters and model of sorption isotherms for plantain flour at 27, 37 and 42 °C

*R²: Coefficient of determination; RMSE: Root Mean Square Error; M₀: Monolayer moisture content; CAPF: Cabinet dried plantain flour; SOPF: Solar dried plantain flour; OVPF: Oven dried plantain flour; SUPF; Sun dried plantain flour; A,B,C,D: Constant parameters

Conclusion

The moisture sorption isotherms of plantain flours at 27, 37 and 42 °C was determined over water activity range of 0.10 to 0.80. The EMC of the plantain flour decreased as the storage temperature increased and represented a type II sigmoidal shape, which is typical for most high carbohydrate foods. The Peleg model adequately described the sorption isotherm of the plantain flour in terms of coefficient of variation and root mean square error. The monolayer moisture content (M_0) of the samples was higher in CAPF and lower in SOPF. However, all the plantain flours regardless of the drying method could be stored for longer periods at all the temperatures studied, as their M_0 falls within an acceptable range for storage.

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