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Experimental Determination and Modeling of Sorption Isotherms of Tropical Fruits: Banana, Mango, and Pineapple

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Abstract: Sorption isotherms of banana, mango, and pineapple have been experimentally found at 40, 50, and 60°C by use of the salt method for a range of water activities from 0.056 to 0.85. The sorption capacity of these fruits increases with the temperature for a given water activity. The experimental curves have been simulated by the GAB and BET models. The BET model ensures a better representation of the experimental results for water activity lower than 0.35 with maximum deviation of 1.0, 1.5, and 2.0 kgw·kgdm⁻¹ for respectively banana, mango and pineapple. On the contrary, the GAB model enables the representation of the whole desorption isotherms and the estimation of the water content corresponding to monolayer saturation and of the isosteric heat of sorption. It also takes into account the temperature effect. The isosteric sorption heat of the three studied products are deduced from experimental results and empirical correlations are proposed leading to a satisfactorily representation.

Keywords: Banana; Mango; Ananas; Sorption isotherm; Isosteric heat; GAB; BET; Specific area

INTRODUCTION

Water activity a_w of a product essentially depends on its water content X and on its temperature θ . When the water in a product is in balance with the water vapor present in the surrounding air, the product temperature is equal to the air temperature (no heat transfer) and the water vapor

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pressure in the product is equal to the water vapor pressure of the air (no mass transfer): Water activity of the product is thus equal to air relative humidity. The curve representing, for a temperature θ , the water content X_{eq} of a product in equilibrium as a function of its water activity a_w (or of air relative humidity HR) is called the sorption isotherm. The desorption isotherms are obtained when the initial product is wet and reaches the equilibrium with air by losing water; the (ad)sorption isotherms are obtained when the initial product is dry and reaches the equilibrium with air by adsorbing water.

The sorption isotherm is particularly important when considering convective drying of a product by a hot air flow. It enables the calculation of the equilibrium water content X_{eq} , the limit of the product water content at the end of the drying. The equilibrium water content X_{eq} is a parameter that appears in models predicting the product water content evolution during its drying.

Furthermore, the analysis of the processes influencing physical, biochemical, and microbiological stability of a product (that determines its quality) is mostly dependent on the sorption isotherm. The sorption isotherm also gives information on the sorption mechanism and on the interaction between adsorbate and adsorbent as pointed out by Bizot and coworkers.^[1] The equilibrium water content of a product is one of the most important parameters to predict its behavior during storage.^[2,3] Nevertheless, it has been shown by Yang and Atallah^[4] that the drying method could have an effect on the characteristics of a dried product such as: sorption and desorption isotherms, porosity, specific area, and color.

Several authors^[5–18] have published results about sorption isotherms studies of various products, but very little information is available about sorption isotherms of tropical fruits, except that presented by Talla et al.^[19]

This article presents a study on the determination of sorption and desorption isotherms of three tropical fruits: banana, mango, and pineapple. The variety of mango used in our tests is *Mangifera indica*; there exist around 300 varieties of mango, which can be quite different from one to another.

MATHEMATICAL MODELS

Sorption isotherms of a material have been described by various mathematical models with two or more parameters as presented by Van der Berg and Bruin.^[20] Nevertheless, models with more than three parameters lead to uneasy use and physical interpretation. The model presented by Brunauer et al.^[5] known as BET model, and its modified version from Guggenheim et al. (GAB) as described by Bizot^[1] have been successfully used by many authors^[10–14,16,17] for modeling sorption isotherms. These

models include parameters that have physical meaning. Moreover, the GAB model takes into account the temperature effect.

BET Model

This model allows the study of the phenomenon of water molecules adsorption and desorption. Its mathematical expression is as follows:

$$X_{eq} = \left(\frac{X_m b a_w}{1 - a_w} \right) \left[\frac{1 - (n + 1)a_w^n + n a_w^{n+1}}{1 + (b - 1)a_w - b a_w^{n+1}} \right] \tag{1}$$

where

X_m = water content corresponding to monolayer $kgw \cdot kgdm^{-1}$;
 b = constant; and
 n = number of water molecules layers.

If the monolayer is not saturated, a simplified expression is:

$$X_{eq} = \left(\frac{X_m b a_w}{1 - a_w} \right) \left[\frac{1 - 2a_w + a_w^2}{1 + (b - 1)a_w - b a_w^2} \right] \tag{2}$$

GAB Model

The GAB model enables a representation of sorption isotherms for the whole values of water activity a_w . Its mathematical expression is as follows:

$$X_{eq} = \frac{X_m C K a_w}{(1 - K a_w)[1 + (C - 1)K a_w]} \tag{3}$$

where

X_{eq} = equilibrium water content ($kgw \cdot kgdm^{-1}$);
 X_m = water content corresponding to monolayer saturation ($kgw \cdot kgdm^{-1}$);
 a_w = product water activity;
 C = constant linked to monolayer sorption heat; and
 K = constant linked to monolayer sorption heat.

$$C = C_0 \exp\left(\frac{H_1 - H_m}{RT}\right) \text{ and } K = K_0 \exp\left(\frac{H_1 - H_q}{RT}\right) \tag{4}$$

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with H_1 , H_m , H_q , respectively, condensation heat of pure water, total sorption heat of the monolayer, and total sorption heat of the other layers
 where

T = absolute temperature (K); and
 R = perfect gas constant ($8,314 \text{ J} \cdot \text{mol}^{-1} \text{ K}^{-1}$).

With the hypothesis of an initial homogenous monolayer filling of the product surface, one can calculate its area. Supposing that the monolayer is recovered with aligned water molecules, it could easily be shown that its area can be calculated by a formula.^[5] The value of the specific area of a product may give information about its rehydration and its internal structure.

$$S_m = \frac{\sqrt{3}\sqrt{2}}{2} \left(\frac{N}{M\rho_w^2} \right)^{1/3} \rho_{dm} X_m \quad (5)$$

where

S_m = monolayer area or specific area ($\text{m}^2 \cdot \text{m}^{-3}$);
 N = Avogadro number = 6.023×10^{23} ;
 M = absorbing material molecular mass ($\text{kg} \cdot \text{mol}^{-1}$);
 ρ_w = water density ($\text{kg} \cdot \text{m}^{-3}$); and
 ρ_{dm} = solid matrix density ($\text{kgdm} \cdot \text{m}^{-3}$).

Isosteric Sorption Heat

A heat that equals to the sum of phase change latent heat L_v and of sorption heat Q_{sorp} is necessary to evaporate one kilogram of water. The desorption heat may be calculated from the curves representing, for a constant water activity, product activity as a function of temperature, these curves are called the (water) sorption isosters. The equation of these curves is given by the Clausius-Clapeyron formula:

$$\ln(a_w) = - \left(\frac{Q_{\text{sorp}}}{R} \right) \left(\frac{1}{T} \right) + \text{Const} \quad (6)$$

In a two-dimensional diagram where $\ln(a_w)$ is represented as a function of $1/T$, the sorption isosters are straight lines according to the relation:

$$\left(\frac{\partial \ln(a_w)}{\partial \left(\frac{1}{T} \right)} \right)_{X=\text{Const}} = - \frac{Q_{\text{sorp}}}{R} \quad (7)$$

The isosteric sorption heat can be deduced from the slope of the sorption isosters representing $\ln(a_w)$ as a function of $1/T$ for each product

water content X , and then the curve $Q_{\text{sorp}} = f(X)$ can be plotted. It is an important parameter whose values show that at the end of the drying it cannot be neglected in energy balance since it has the same order of magnitude as the vaporization latent heat.

MATERIAL AND METHOD

Sorption Method

The static method of the saturated salts solutions has been used for sorption isotherms determination. It is a method where diffusion is the only way of mass transfer between the tested product and the surrounding air. The relative air humidity is fixed by contact with saturated salts solution whose water vapor pressure at a given temperature is perfectly known. This method is commonly used for product sorption isotherms determination.^[11,12,17,21,22]

The time necessary to reach equilibrium is quite long compared to the dynamic method since the diffusion rate is a limiting factor.

Experimental Process

The first step is the choice of the salts so that a large interval of water activities could be obtained. For our experiments, nine salts have been used that covered the range 0.056 to 0.85 for water activities; these include: KOH, LiCl, KCH_3CO_2 , MgCl_2 , K_2CO_3 , NaBr, CuCl_2 , NaCl, and KCl. The corresponding values of water activities for all these salts at the various experimental temperatures used are presented in Table 1 according to Bizot and Multon.^[23]

Three samples of the tested product (leading to an average value) are then set in each of the nine recipients containing the saturated salts solutions. Figure 1 shows the experimental facility. The recipients are set in a temperature regulated chamber. For adsorption isotherms determination, the samples were previously dehydrated at 60°C . For desorption isotherms, fresh products without any preparation were used.

Then, the samples were weighed at regular time intervals until the mass becomes constant (variation less than 1 mg by 48 h) and they could be considered as in equilibrium with air at (θ, HR) . The mass of a sample when equilibrium is reached with its bone dry mass measured after dehydration for 48 h at 102°C enables the calculation of the equilibrium water content by applying the following formula:

$$X_{\text{eq}} = 100 \left(\frac{m_{\text{eq}}}{m_{\text{d}}} - 1 \right) \quad (8)$$

Table 1. Air relative humidities imposed by saturated salt solutions for several temperatures^[23]

Salts	Relative humidity			
	25°C	40°C	50°C	60°C
KOH	8.2	6.3	5.7	5.6
LiCl	11.3	11.2	11.4	11.0
KCH ₃ CO ₂	22.5	21.6	21.6	21.6
MgCl ₂	32.8	31.6	30.5	29.3
K ₂ CO ₃	43.2	42.2	45.6	45.0
NaBr	57.6	53.2	50.9	49.7
CuCl ₂	68.5	68.0	65.5	63.3
NaCl	75.3	74.7	74.4	74.5
KCl	84.3	82.3	81.2	80.3

where

m_{eq} = sample equilibrium mass (kg); and

m_d = sample bone dry mass (kg · dm).

The sorption isotherm is given by the experimental couples (a_w , X_{eq}). The experimental procedure consists of introducing the recipients in the chamber regulated to the lowest temperature value of the series (40°C),

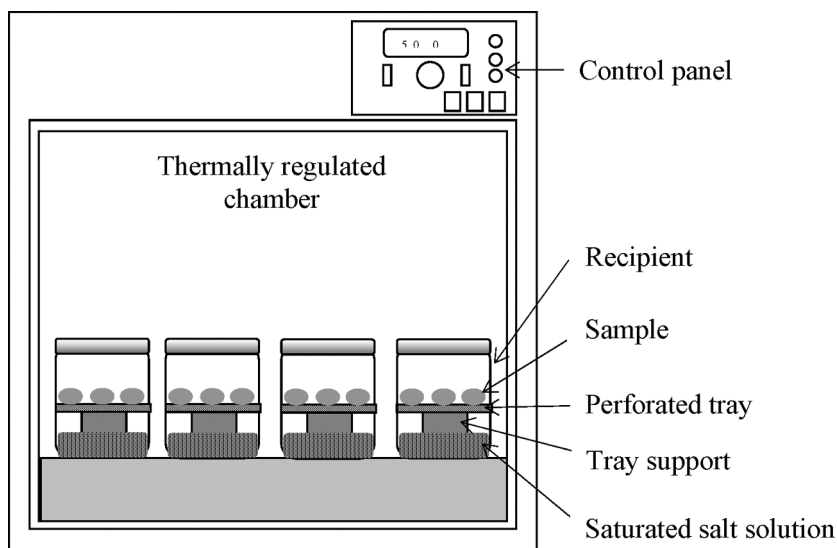


Figure 1. Experimental device for sorption isotherm determination.

weighing all the samples when the equilibrium was reached, and then raising the temperature to the next value (50°C). This procedure was repeated for 60°C. During the experiments, a partial vacuum was applied inside the recipients to increase the dehydration rate.

RESULTS AND DISCUSSIONS

Figures 2a to 4a represent the experimental desorption isotherms of banana, mango, and pineapple obtained at different temperatures. The isotherms are sigmoidal as usually presented for foodstuff as done by Iglesias and Chirife.^[7] Meanwhile, it can be observed for banana that these “static” equilibrium water contents are greater by a few percent than the “dynamic” equilibrium water contents reported by Talla et al.^[19] This result is in agreement with the conclusions of Nadeau and Puiggali^[24] related to the comparison between the static and dynamic methods for desorption isotherms determination. For a given value of the water activity, the equilibrium water content decreases with the temperature. Furthermore, it is not possible to obtain experimental couples (X_{eq} , a_w) for $a_w > 0.9$ with the static saturated salts method since the products (fruits) are decomposed after a few days before the thermodynamic equilibrium being reached.

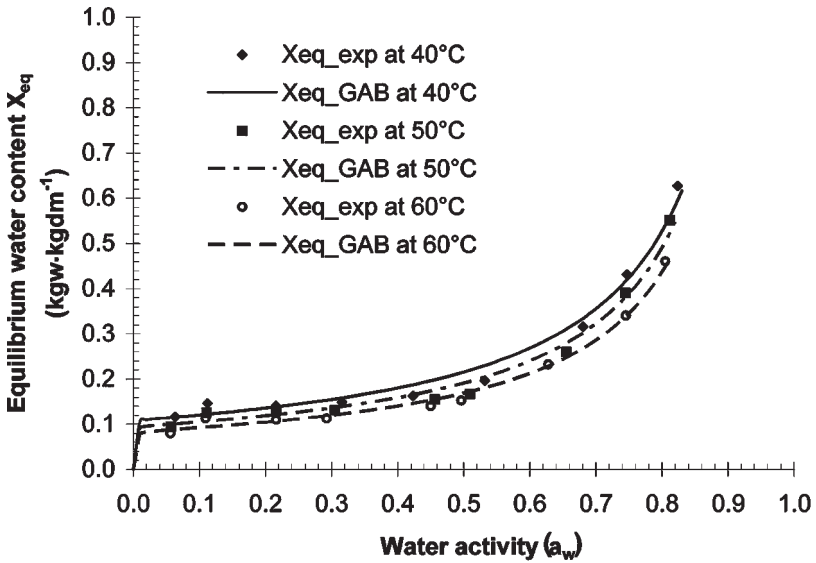
The constants X_m , C , and K of the GAB model depends on product characteristics and temperature, similar to the constants X_m and b of the BET model. All these constants are estimated from experimental results. For each temperature, these parameters are estimated by an optimization method minimizing the sum S of the quadratic errors calculated by formula^[9] between the experimental equilibrium water contents and the values calculated with relation.^[3]

$$S = \sum_{i=1}^n \left(\frac{\bar{X}_{exp} - \bar{X}_{mod}}{\bar{X}_{exp}} \right)_i^2 \quad (9)$$

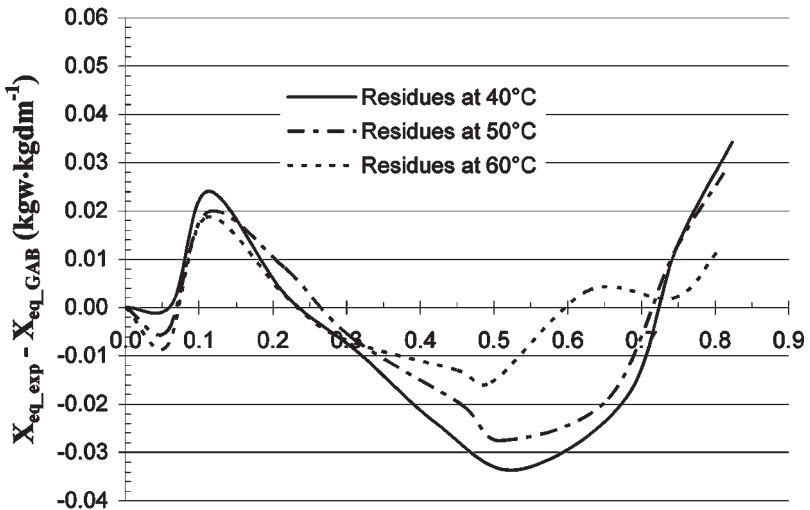
where n is the number of measurements for a temperature (nine in the present case).

Contrarily to the GAB model which represents the sorption isotherm on a wide range of a_w values, the BET model enables the representation of the sorption isotherms only in the range of activities from 0 to 0.35 according to Brunauer et al.^[5] Table 2 lists the whole results of the estimation. Furthermore, Figs. 2a to 4a and 5a to 7a present a comparison between the experimental curves and the curves simulated by the GAB model and by the BET model, respectively.

The representation of the residues on Figs. 2b to 4b shows higher deviation for high values of activities with the GAB model. The maximum deviation for the whole products is lower than 8% for a water



(a)



(b)

Figure 2. (a) Desorption isotherms of banana at several temperatures: simulation by the GAB model and experimental results; (b) Residues between experimental values of X_{eq} and values calculated by the GAB model for banana.

activity of 0.85. Nevertheless, the deviation is quite low for water activities lower than 0.35 and are thus interesting for drying modeling since the literature recommends a final water content between 0.15 and 0.20 for a

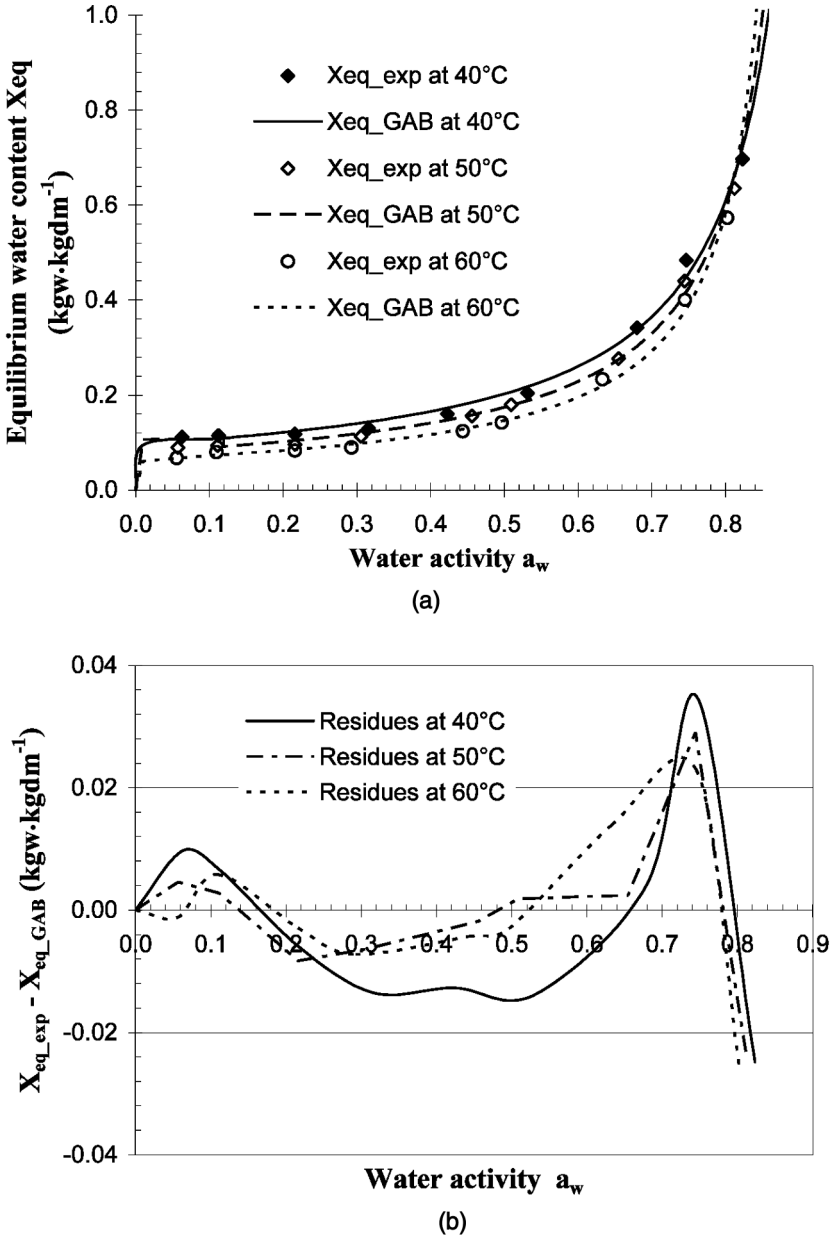


Figure 3. (a) Desorption isotherms of mango at several temperatures: simulation by the GAB model and experimental results; (b) Residuals between experimental values of X_{eq} and values calculated by the GAB model for mango.

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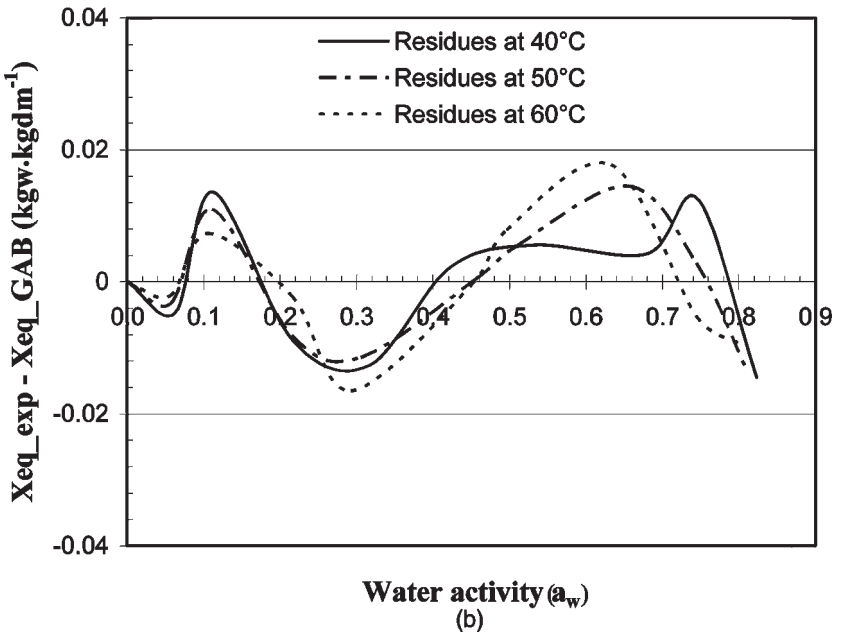
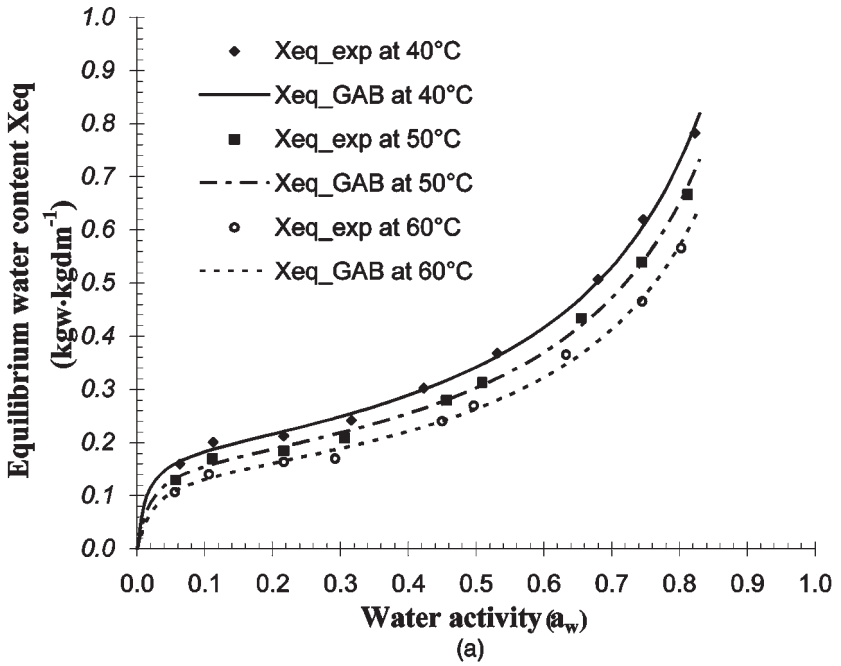


Figure 4. (a) Desorption isotherms of pineapple at several temperatures: simulation by the GAB model and experimental results; (b) Residues between experimental values of X_{eq} and values calculated by the GAB model for pineapple.

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Table 2. Estimated values of the parameters of the GAB and BET models

Product	θ ($^{\circ}\text{C}$)	GAB model				BET model			
		X_m ($\text{kgw} \cdot \text{kgdm}^{-1}$)	C	K	X_m ($\text{kgw} \cdot \text{kgdm}^{-1}$)	b	S_m ($\text{m}^2 \cdot \text{m}^{-3}$)	\bar{X}_0 ($\text{kgw} \cdot \text{kgdm}^{-1}$)	$\bar{X}_0 \rho_s / S_m$ ($\text{kgw} \cdot \text{m}^{-2}$)
Banana $\bar{X}_0 = 3.77$ ($\text{kgw} \cdot \text{kgdm}^{-1}$)	25	0.127	9295.7	0.979	0.181	74.49	8.91E+08	\bar{X}_0	5.92E-06
	40	0.108	6531.4	0.993	0.160	49.23	7.88E+08	\bar{X}_0	6.70E-06
	50	0.095	3420.9	1.009	0.147	35.66	7.24E+08	\bar{X}_0	7.29E-06
	60	0.083	1828.3	1.011	0.128	34.76	6.30E+08	\bar{X}_0	8.38E-06
Mango $\bar{X}_0 = 6.83$ ($\text{kgw} \cdot \text{kgdm}^{-1}$)	40	0.096	4798.6	1.054	0.129	94.30	6.35E+08	\bar{X}_0	1.51E-05
	50	0.080	2954.5	1.082	0.110	66.08	5.41E+08	\bar{X}_0	1.77E-05
	60	0.065	1104.0	1.111	0.094	45.87	4.63E+08	\bar{X}_0	2.07E-05
Pineapple $\bar{X}_0 = 6.15$ ($\text{kgw} \cdot \text{kgdm}^{-1}$)	40	0.185	83.292	0.934	0.266	24.45	1.31E+09	\bar{X}_0	6.58E-06
	50	0.164	57.285	0.937	0.234	21.94	1.15E+09	\bar{X}_0	7.47E-06
	60	0.143	45.769	0.938	0.199	20.64	9.80E+08	\bar{X}_0	8.79E-06

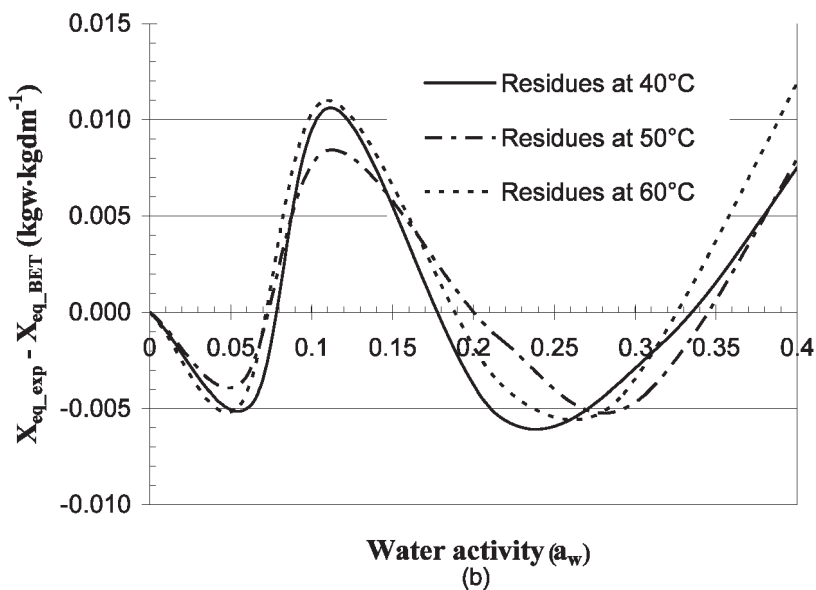
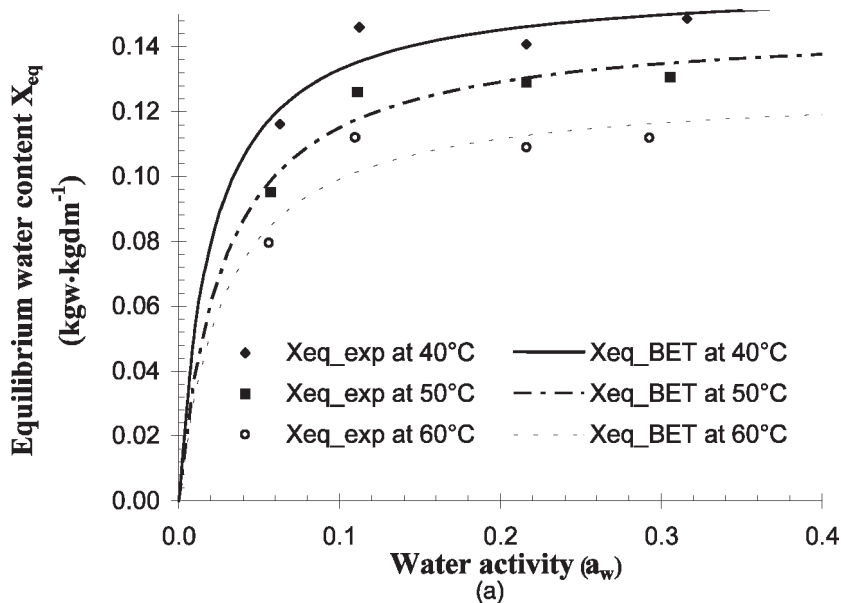


Figure 5. (a) Desorption isotherms of banana at several temperatures: simulation by the BET model and experimental results; (b) Residues between experimental values of X_{eq} and values calculated by the BET model for banana.

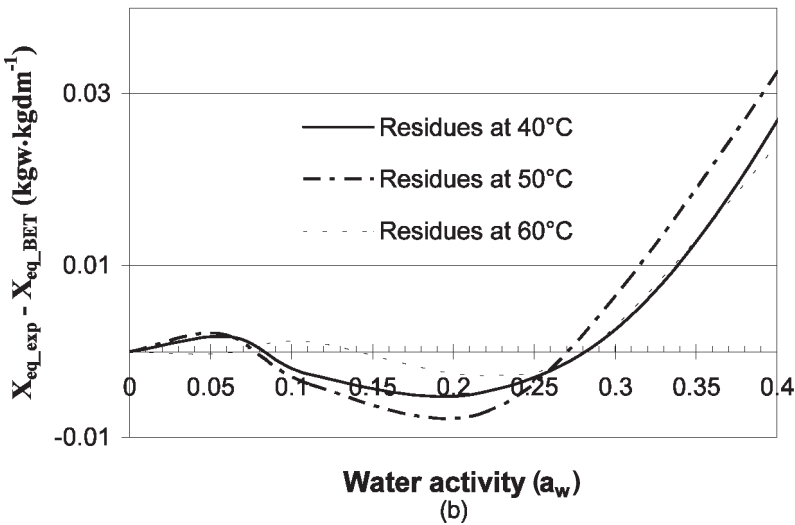
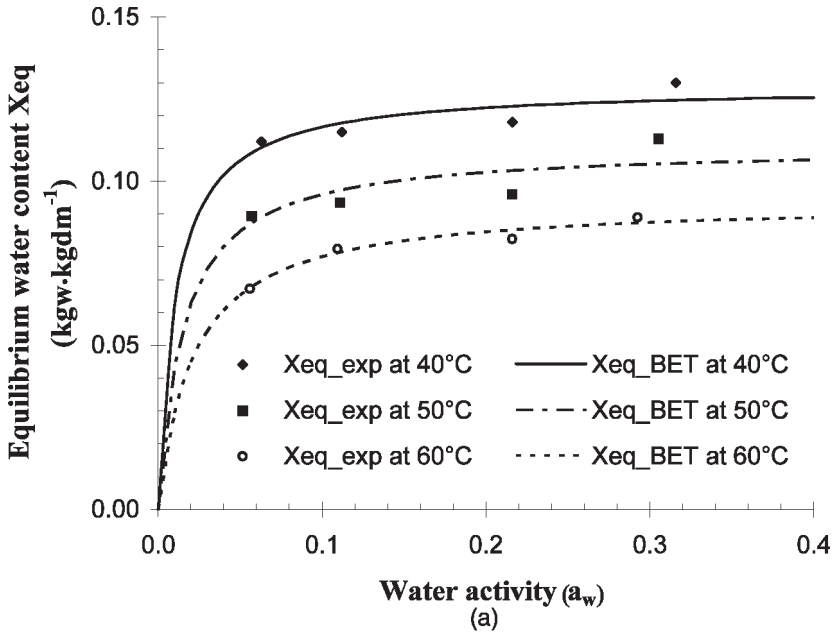


Figure 6. (a) Desorption isotherms of mango at several temperatures: simulation by the BET model and experimental results; (b) Residues between experimental values of X_{eq} and values calculated by the BET model for mango.

good conservation.^[24] For water activities lower than 0.35, the BET model fits better of the experimental results as shown on Figs. 5b to 7b representing the residues. In this range of water activities, the maximum

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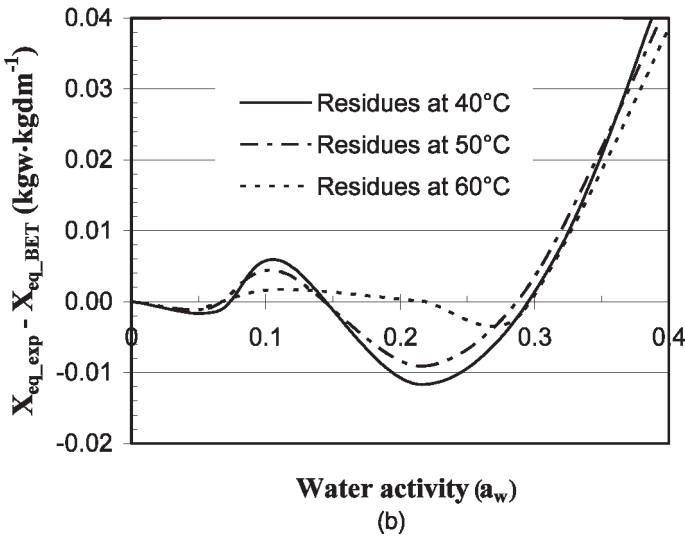
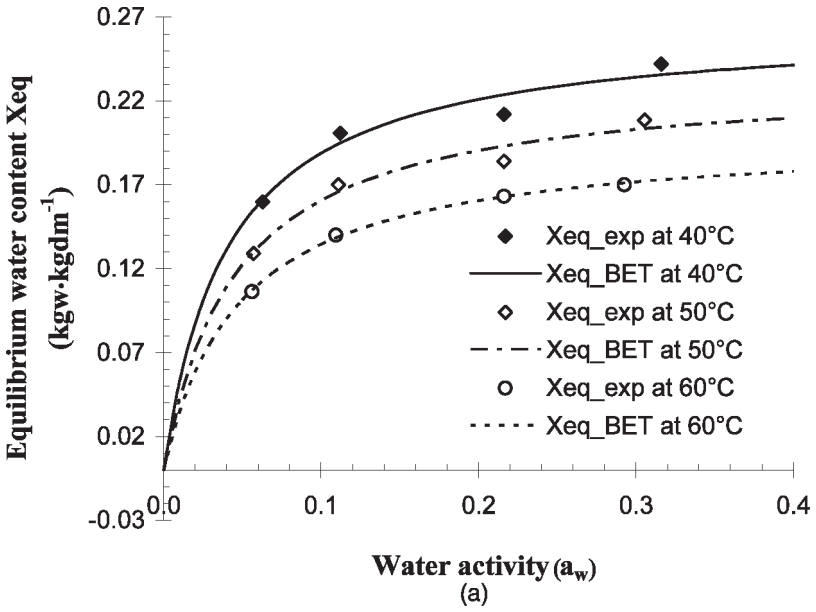


Figure 7. (a) Desorption isotherms of pineapple at several temperatures: simulation by the BET model and experimental results; (b) Residues between experimental values of X_{eq} and values calculated by the BET model for pineapple.

observed deviation is 1.0, 1.5, and 2.0%, respectively, for banana, mango, and pineapple. The relatively low deviation leads to the conclusion that these two models represent the desorption isotherm quite satisfactorily.

According to experimental results, the estimated parameters X_m and C decrease when the temperature rise since the parameter K increases with temperature (Table 2). This is in agreement with the reported behavior of other foodstuff.^[26–29] The sensitivity of the GAB model to the parameters X_m , C , and K is deduced from Eqs. (10) to (12) and represented in Fig. 8 for banana. This figure shows that the GAB model is quite insensitive to the parameter C : a relative variation of 10% on the parameter C has no effect on the simulated values of X_{eq} , which is also verified for mango and pineapple. This confirms, as pointed out by Rouquerol et al.,^[30] that the simplified theoretical expression of the parameter C given by formula (5) cannot lead to a reliable estimation of the total sorption heat H_m of the monolayer.

$$\frac{\epsilon}{100} X_m \frac{\partial X_{eq}}{\partial X_m} = \frac{\epsilon}{100} X_{eq} \tag{10}$$

$$\frac{\epsilon}{100} C \frac{\partial X_{eq}}{\partial C} = \frac{\epsilon}{100} X_{eq} \left[1 - \frac{CKa_w}{1 + (C - 1)Ka_w} \right] \tag{11}$$

$$\frac{\epsilon}{100} K \frac{\partial X_{eq}}{\partial K} = \frac{\epsilon}{100} X_{eq} \left[\frac{Ka_w}{1 - Ka_w} + \frac{1}{1 + (C - 1)Ka_w} \right] \tag{12}$$

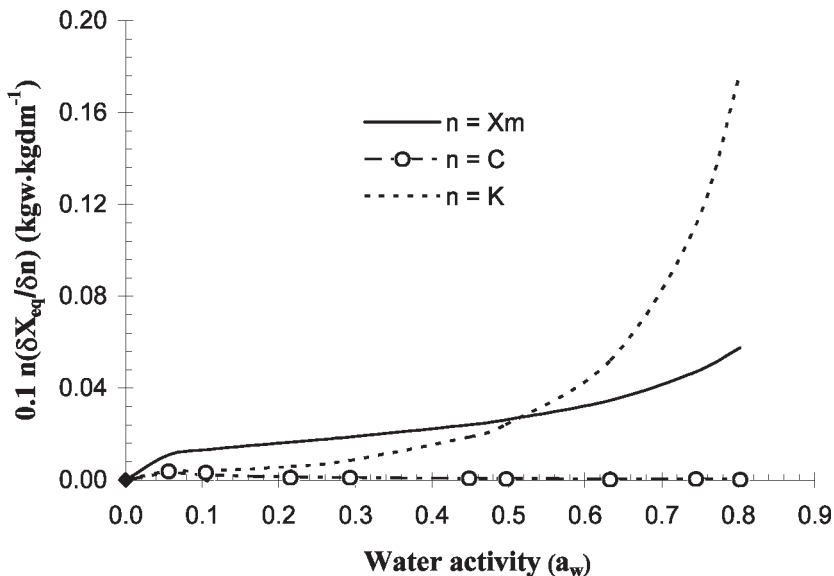


Figure 8. Variation of X_{eq} induced by a relative variation of 10% of each parameter.

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These three formulas represent the variation of the estimated value of X_{eq} indices by a relative variation of $\varepsilon\%$ of respectively the parameters X_m , C , and K from their nominal values.

The parameter X_m is the water content corresponding to the monolayer saturation. Table 2 shows that the values of X_m estimated from the BET model are always greater than the values obtained from the GAB model. This difference is linked to the fitting of the model with the experimental curve specially in the area corresponding to the monolayer saturation (water activity lower than 0.2). The values obtained from the BET model which better fits the experimental values in this area must be more reliable than those obtained with the GAB model.

Furthermore, it can be observed that the specific area (calculated from the BET model) is not linked to the initial product water content, which is rather linked to the porosity (Table 2). The specific area is directly proportional to the water content at the saturation of the monolayer. Table 2 also indicates the values of $X_0\rho_s/S_m$, which represents, at the initial state, the water content filling the pores by unit of specific area and so has the dimension of a water thickness. The density of the solid phase is deduced from the results presented by Lozano,^[31] who has measured the density of several fruits and vegetables as a function of their water content. For a zero water content, the density varies from 1300 to 1550 $\text{kg} \cdot \text{m}^{-3}$; a mean value of 1400 $\text{kg} \cdot \text{m}^{-3}$ has been used in our calculation. Table 2 shows a lower value for mango than for banana and pineapple, which may indicate that mango will be easier to dry; this prediction is in agreement with the results of Talla et al.^[19] On the other hand, according to the values in Table 2 for banana and pineapple, these two products might have a rather similar behavior during drying.

The parameters C_0 and $H_1 - H_m$ have not been estimated since it will not be reliable as previously pointed out. Table 3 presents the estimated values of K_0 and $H_q - H_1$. The values of $H_q - H_1$ represent the mean value of sorption heat of the studied fruits, which is greater than the vaporization heat of water.

Figures 9 to 11 represent the sorption isosters $\ln(a_w)$ vs. $1/T$ that must be straightlines according to formula (6), for, respectively banana, mango, and pineapple. For each water content X , a sorption isoster has been plotted and the isosteric sorption heat was calculated from its

Table 3. Estimated parameters for K calculation

Product	K_0	$H_1 - H_q$ ($\text{kJ} \cdot \text{mol}^{-1}$)
Banana	1.359	-97.74
Mango	2.533	-274.63
Pineapple	1.003	-22.04

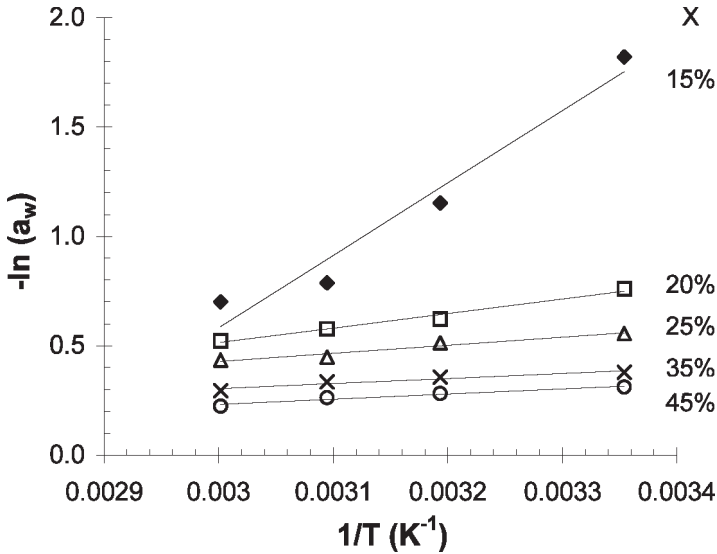


Figure 9. Sorption isosters for banana.

slope. For each product, the representation of Q_{sorp} vs. X has then been plotted on Fig. 12. It can be observed that the isosteric sorption heat is high for the low water content, indicating the strong link between the adsorbate (water) and the adsorbent, but it is quite negligible compared to vaporization latent heat for high water content. The same

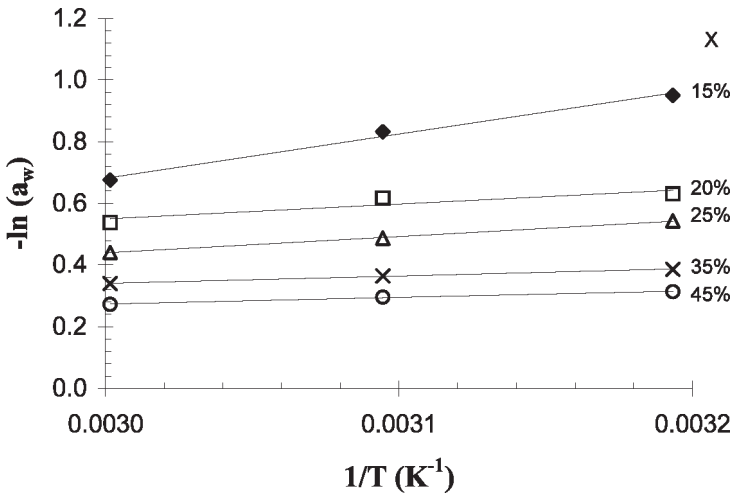


Figure 10. Sorption isosters for mango.

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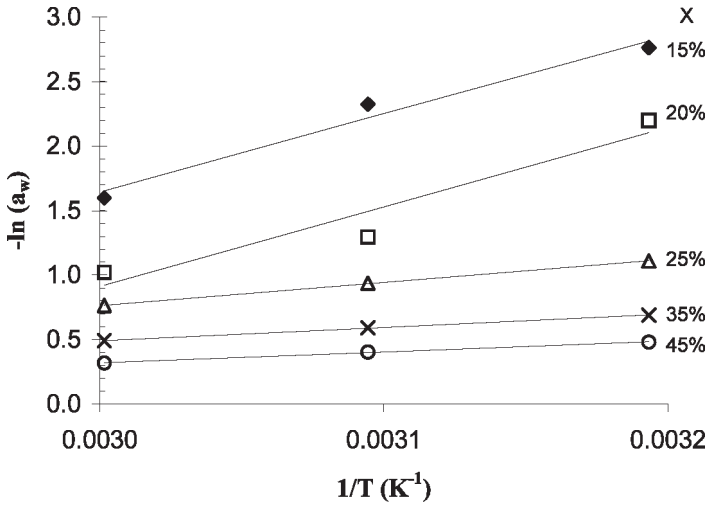


Figure 11. Sorption isosters for pineapple.

procedure has been repeated for mango and pineapple and all the experimental results have been correlated satisfactorily by relations (13) to (15).

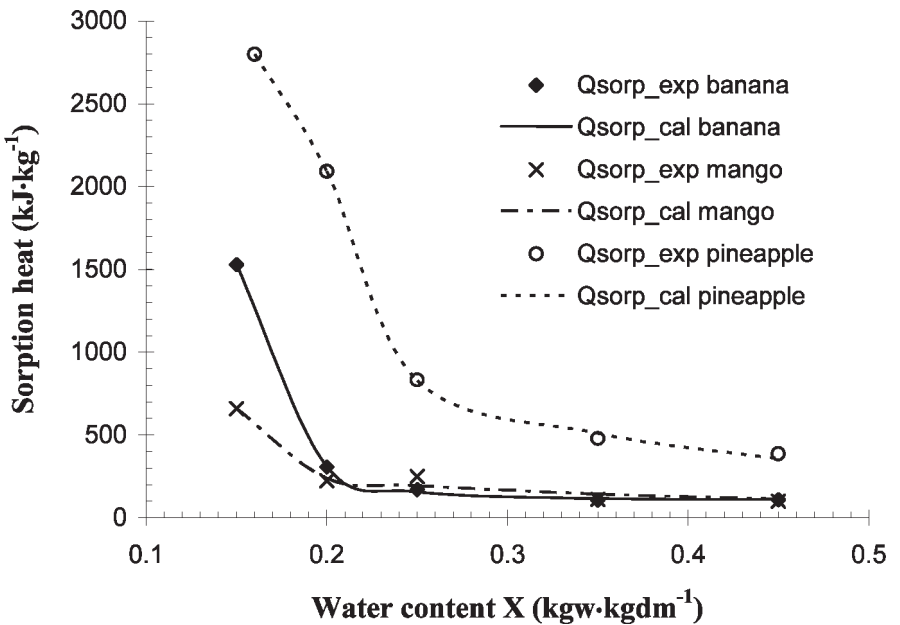


Figure 12. Sorption heat of banana, mango, and pineapple vs. water content.

$$\text{Banana: } Q_{\text{sorp}} = \frac{722.3 + 0.021\bar{X}^{-6.822}}{6.14 + \bar{X}^{1.028}} \text{ (kJ} \cdot \text{kgw}^{-1}) \quad (13)$$

$$\text{Mango: } Q_{\text{sorp}} = \frac{-44.6 + 90.8\bar{X}^{0.370}}{-0.03 + \bar{X}^{1.807}} \text{ (kJ} \cdot \text{kgw}^{-1}) \quad (14)$$

$$\text{Pineapple: } Q_{\text{sorp}} = \frac{82.49 - 0.320\bar{X}^{-3.653}}{-0.433 + \bar{X}^{0.542}} \text{ (kJ} \cdot \text{kgw}^{-1}) \quad (15)$$

CONCLUSION

In this study, the desorption isotherms of banana, mango and pineapple have been experimentally established and then modeled by BET and GAB models. The BET model enables the calculation of the monolayer water content leading to the value of the specific area for different temperatures. A comparison between BET and GAB models shows that the latter one gives a better fitting with experimental results for water activities lower than 0.35. Nevertheless, the maximal deviation between experimental results and calculated values of the water content X by the GAB model is around $0.08 \text{ kgw} \cdot \text{kgdm}^{-1}$ for $X = 0.85$, which remains acceptable. This model also leads to the calculation of the isosteric desorption heat of each fruit.

Furthermore, the curves giving the isosteric desorption Q_{sorp} heat vs. water content X have been deduced from the experimental desorption isotherms. The high values observed for low water content indicates the strong link between the adsorbate (water) and the adsorbent. Empirical relations $Q_{\text{sorp}} = f(X)$ have been established for each product.

NOMENCLATURE

a_w	Product water activity
C	Parameter linked to monolayer heat of sorption
K	Parameter linked to multilayers heat of sorption
m	Product mass (kg)
Q	Sorption heat ($\text{kJ} \cdot \text{mol}^{-1}$)
S	Specific area ($\text{m}^2 \cdot \text{m}^{-3}$)
X	Dry basis water content ($\text{kgw} \cdot \text{kgdm}^{-1}$)
\bar{X}	Mean dry basis water content ($\text{kgw} \cdot \text{kgdm}^{-1}$)

Greek Symbols

ρ	Density ($\text{kg} \cdot \text{m}^{-3}$)
θ	Air temperature ($^{\circ}\text{C}$)

Subscripts

0	Initial
w	Water
eq	Equilibrium
i	Sample number
m	Monolayer
n	Number of layers of water molecules
p	Number of measurements for a sorption isotherm
dm	Dry matter
sorp	Sorption
exp	Experimental
mod	Model

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