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RESEARCH ARTICLE

MODELING OF THE WATER ADSORPTION ISOTHERM OF MANGO SLICES (*Mangifera indica* L.) DRIED UNDER HOT AIR

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Abstract

The general objective of this work is to contribute to the reduction of post-harvest losses of mangoes by the transformation into stable dried products.This research aims to determine the best storage conditions for dried mangoes.Thus, the water adsorption isotherms of mango slices dried under hot air were experimentally determined by the static thermogravimetric method at 25°C.Eight mathematical models from the literature were used (GAB, BET, Hasley, Henderson, Harking and Dura, Oswin, Chung and Pfost, Smith) to describe the hygroscopic behavior of dried mango slices.Regression analysis was performed using MatLab R2016a software to determine the coefficients, model parameters and statistical parameters of the smoothing.The results revealed that the equilibrium is obtained from 35 days for the two samples and the adsorption isotherms determined are of type II characterized by a sigmoidal shape.In addition, the three-parameter (GAB) model showed good agreement with the experimental data (r= 0.999 and 0.997 respectively for slices with thicknesses of 1cm and 1.5cm) at 25°C.The water content of the mono-molecular layer is 2.295% at 25°C for the 1cm slices and 2.289% at 25°C for the 1.5cm slices.The net isosteric heat of adsorption tends towards zero for high water contents (0.052KJ/mol at 44.472% water).The linearity between enthalpy (net isosteric heat) and differential entropy of adsorption shows their compensation.

Introduction:-

Agriculture contributes to the growth of the Ivorian economy and continues to be its key (Nouar et al., 2013).Indeed, Côte d'Ivoire sees its economy being dominated by the export of so-called cash crops, in particular cocoa, coffee, oil palm, rubber, etc. (Abouo et al., 2015).It also exports fruits and vegetables, the most important of which (quantitatively) are pineapple, banana and mango respectively (Abouo et al., 2020).These three products contribute almost 4% to the formation of the gross domestic product (GDP).With a production of around 160,000 tonnes, including more than 33,000 tonnes exported to Europe, Ivorian production has risen to third place among mango suppliers to the European market after Brazil and Peru (CBIMFA, 2019).Despite this importance, producers face problems of post-harvest losses during storage.To reduce losses, extend the durability of the product and add value

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to the product, conventional drying, which is an operation widely used both in the food industry and in an artisanal way by farmers, is adopted (Bonazzi et al., 2003Abouo et al., 2021).This process stabilizes the product by lowering the water activity (Iglesias et al., 1982).Furthermore, to optimize the storage conditions of a product so as to ensure its physicochemical and microbiological stability, the determination of sorption isotherms is a necessity (Ahouannou et al., 2010; Koko et al., 2018).These isotherms are curves that provide valuable information on the hygroscopic balance of a product because they make it possible to know its domain of stability after drying by determining the final water content (Lamharrar et al., 2007; Abouoet al., 2020).Knowledge of water activity and sorption-desorption isotherms are of great importance in the food industry, storage and food preservation (Multon, 1982; Boudhrioua, 2004).During the last two decades, a large number of works have focused on the study of sorption isotherms of food products (Bolin, 1980), the influence of temperature on isotherms (Labuza et al., 1985; Ayranci et al., 1990) and the study of mathematical models describing sorption isotherms (Chirifie et al., 1983; Kim et al., 1985; Maroulis et al., 1988).However, very little work has been devoted to determining the water adsorption isotherms of conduction-dried mango slices.This study aims to contribute to improving the shelf life of dried mango during storage.Its objective is to evaluate the evolution of the hygroscopic behavior of dried mango slices using the static gravimetric method at a temperature of 25°C.Specifically, these are :

- ✓ Establish water adsorption isotherms of dried mango slices
- ✓ Characterize the isotherms (type, water content of the mono-molecular layer)
- ✓ Model the water adsorption isotherms of dried mango slices
- ✓ Quantify iso-steric heat and free enthalpy

Materiel and Methods:-

a. Biological material

The biological material used for this study consists of Kent variety mango.They come from the local market in the town of Abobo (North Abidjan).The mangoes were purchased in the period of May 2022, round in shape and green-red in color.They were commercially ripe (ripe, whole fruit) and without injuries.These mangoes each had a weight of about 400g and a length of about 10cm.These mangoes (pulp) were sliced to varying thicknesses of 1 cm and 1.5 cm and dried at 50°C for 23 and 27 hours respectively on average under hot air in a BIOBASE brand oven (Abouo et al., 2020).These slices were then weighed on a digital scale and put in stainless cups and placed in jars (Figure 1).

b. Methods

- **Experimental procedure for carrying out adsorption isotherms**

The static gravimetric method was used to determine the adsorption isotherms of dried mango slices (Wolf et al., 1985 ; Kakou et al., 2015).In eight (8) airtight jars each containing 200 mL ofsulfuric acid solutions (Figure 1) at increasing concentrations, 5 g of dried mango slices were introduced, weighed in stainless cups.The samples are stabilized in temperature and hygrometry in an oven (Biobase, China) at 25°C.This temperature was chosen because the average temperature variation in Côte d'Ivoire is between 18 and 37°C.Temperature generally encountered in supermarkets (Akmel et al., 2009 ; Kakou et al., 2015).The weighing of the samples was carried out every 48 hours until the balance of the mass.Balance is considered to be reached when the variation in mass between three successive measurements is less than or equal to 0.001g. Knowing the wet masses, the dry masses are then obtained by placing the samples in an oven at 105°C ± 2°C for 24 hours.Thus, the equilibrium water content Xéq is deduced from formulas (1) and (2).The couples (Aw, Xéq) provide the points of the adsorption isotherm.

$$Xi = \frac{\mathrm{Mi} - \mathrm{MS}}{\mathrm{MS}} \times 100 \quad (1)$$

$$Xeq = \frac{\mathrm{M}}{\mathrm{Mi}}(\mathrm{Xi} + 100) - 100 \quad (2)$$

With: Mi: mass of the product at the initial time (g), M: mass of the product at time t (g), Ms: dry mass of the product (g), *Xi*: water content of the product atinitial instant (% ms), *Xeq*: equilibrium water content of the product.

Figure 1:- Experimental device for determining adsorption isotherms.

- **Modeling of adsorption isotherms**

Several mathematical models, knowledge or empirical relationships, describe the relationship between equilibrium water content, relative humidity at a given temperature.Eight different models found in the literature were used during the study (Table I).The goal is to determine the most appropriate model(s) for describing the water adsorption isothermsof biological material (Touati, 2008).

Table I:- Mathematical models used.

MODELS	MATHEMATICAL EQUATIONS	COEFFICIENTSAND PARAMETERS
G.A.B (Van der Berg and Bruin, 1981)	$X = \frac{X_m * C * K * aw}{((1 - K.aw) * (1 - K.aw + C.K.aw)}$	Xm, K, C
BET (Brumauer, Emmet et Teller, 1983)	$X = \frac{X_m * C * K * aw}{((1 - K.aw) * (1 - K.aw + C.aw)}$	Xm, C, aw
Hasley (Hasley, 1948)	$X = \left(\frac{A}{\ln\left(\frac{1}{aw}\right)}\right)^{1/B}$	A, B
Henderson (Henderson, 1952)	$X = \left(\frac{-\ln(1 - aw)}{A}\right)^{1/B}$	A, B
Iglesias-Cherif (Iglesias et al, 1978)	$X = A + B\left(\frac{aw}{1 - aw}\right)$	A, B
Oswin (Oswin, 1946)	$X = A\left(\frac{a_w}{1 - aw}\right)^c$	A, C

Caurie (Caurie, 1970)	$X = e^{(a+b.aw)}$	a, b
Smith (Smith, 1946)	$X = C_1 - C_2.\ln(1 - aw)$	C_1, C_2

- **Determination of parameters (Xm, A, B and C)**

The parameters Xm, A, B and C, of the various models were determined by identification with the experimental adsorption curves, by minimizing the sum of the Mean Square Deviations:

$$MSE=\frac{1}{N}\sum_{i=1}^{N}\left|\frac{Xeqi,exp - Xeqi,pre}{Xeqi,exp}\right|^2 \quad (3)$$

With: *Xeq*i, exp: equilibrium experimental water content (% dm: dry matter), *Xeq*i, pre: predicted equilibrium water content (% dm), N: Number of experimental points.

- **Statistical analyzes**

Modeling adsorption isotherms requires statistical methods of regression and correlation analyzes (Abouo et al., 2021).The regression analysis was carried out under Excel 2013 software, using the nonlinear GRG algorithm of the solver.The correlation coefficient (r) was the first criterion for selecting the best model to describe the adsorption curves (Inci and Dursun, 2004).In addition to r, the calculations of the values of EMR (Relative Mean Error) and EQM (mean squared error) made it possible to justify the choice of the model (Sun and Woods, 1994).The best model is the one with the highest value of r (close to 1) and the lowest values of MRD and MSE (Benhamou et al., 2010).

$$r = \sqrt{\frac{\sum_{i=1}^{N} (Xeqi,pre - \bar{X}eqi,exp)^2}{\sum_{i=1}^{N} (Xeqi,pre - \bar{X}eqi,exp)^2}} \quad (4)$$

$$MSD = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{Xeqi,exp - Xeqi,pre}{Xeqi,exp}\right| \quad (5)$$

Resultsand Discussion:-

Results:-

- **Characterization of isotherms**

The hygroscopic equilibrium of the dried mango slices is reached from the 35th day of the study.The experimental adsorption curves obtained for each sample at 25°C are shown in Figure 2.They all show type II sigmoidal shapes.For the constant temperature of 25°C, the equilibrium water content of the product decreases with thewater activity of the product.Indeed, it goes from 44.472% to 0.102% for TMs of 1cm and from 32.93 to 0.105% forTMs of 1.5cm.

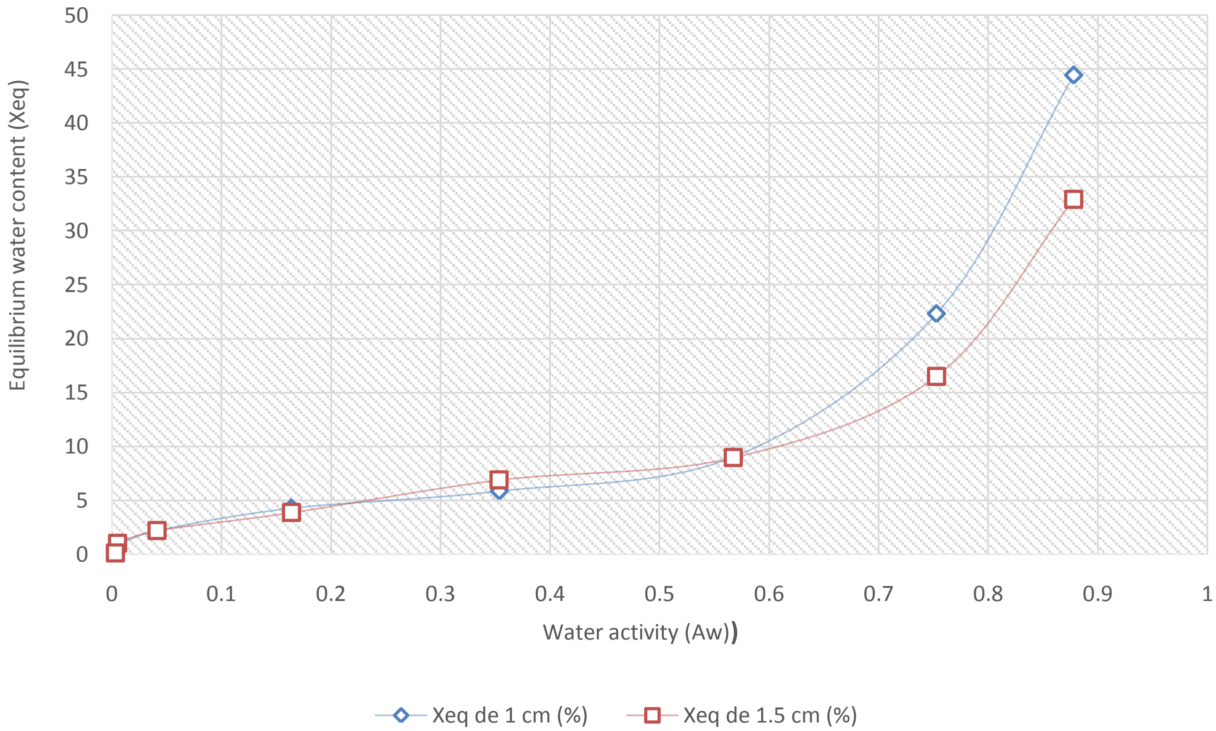


Figure 2:- Water adsorption isotherm at different thicknesses.

- **Modeling of isotherms**

Table II presents the parameters of the models used and the statistical selection criteria.The correlation coefficients r are all high for the same temperature.They vary from 0.941 to 0.999 for 1cm mango slices and from 0.948 to 0.997 for 1.5cm mango slices.A decrease in the value of the parameters (Xm, r, MSE, etc.) of the models used is observed when the diameter of the slices increases for the same drying temperature, with the exception of the parameters of the Chung-Pfost and Smith models.where the value of the parameters increases when the diameter of the TM increases.Thus, the water contents of the monolayer of the BET and GAB models are dependent on the thickness of the TMs at 25°C with relatively very close values (respectively 2.30 and 2.29% for the 1cm TMs.2.29 and 2.28 for 1.5cm TMs for BET and GAB).Statistical analysis of the eight models shows that the 3-parameter GAB model has the highest correlation coefficient r (0.999 for 1cm TMs) except for 1.5cm where the GAB model and Halsey have the 'r' highest (0.997).In terms of estimation errors for the two samples of mangoes, it is the GAB model which presents the lowest estimation errors of MSE (mean squared error), MSD (mean squared deviation) for the TM of 1cmand the highest MSE, MSD for 1.5cm TMs.Moreover, the values of the MSE and MSD estimation errors of the GAB model are relatively close to those of Halsey.Thus, the GAB model seems to be the most appropriate to describe the adsorption isotherms of TMs dried under hot air.

Table II:- Values of estimated parameters and statistical selection criteria (TM).

Models	**Parameters**	**1cm**	**1.5cm**
	Xm	2.304	2.297
	C	93.446	95.996
BET	r	0.968	0.965
	MSE	0.007	0.030
	MSD	13.785	12.623
	Xm	2.295	2.289
	C	16175.841	16208.718
	K	0.944	0.944
GAB	r	0.999	0.997
	MSE	0.040	0.021

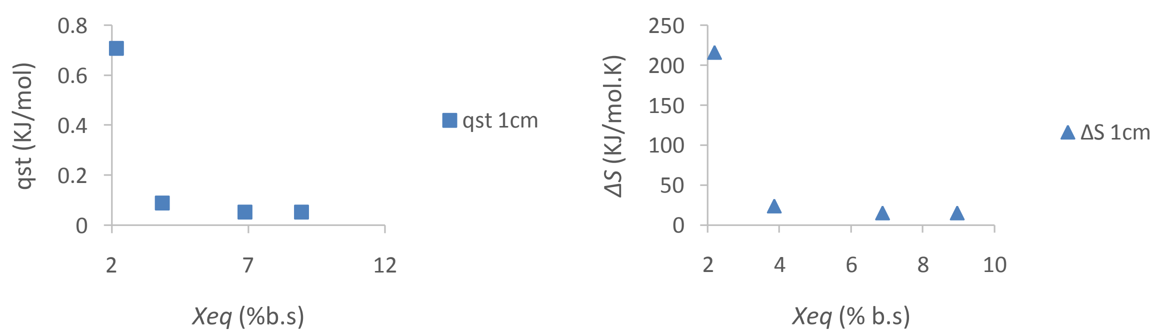
	MSD	12.490	11.462
	A	3.184	3.177
	B	0.326	0.327
Chungand Pfost	r	0.987	0.937
	MSE	0.005	0.0315
	MSD	13.394	12.025
	A	11.4309	11.034
	B	16.063	15.972
	r	0.979	0.971
Harking andJura	MSE	0.006	0.020
	MSD	12.869	10.638
	A	2.954	2.932
	B	0.292	0.298
Henderson	r	0.973	0.969
	MSE	0.005	0.004
	MSD	11.768	15.166
	A	-0.437	-0.436
	B	-6.323	-6.319
Smith	r	0.986	0.981
	MSE	0.011	0.068
	MSD	13.108	12.668
	A	4047.605	3932.376
	B	2.387	2.400
	X0	2.808	2.868
Hasley	r	0.958	0.997
	MSE	0.052	0.060
	MSD	13.798	11.113
	A	4.060	4.051
	B	0.506	0.484
Modified Oswin	r	0.941	0.948
	MSE	0.039	0.004
	MSD	12.178	12.647

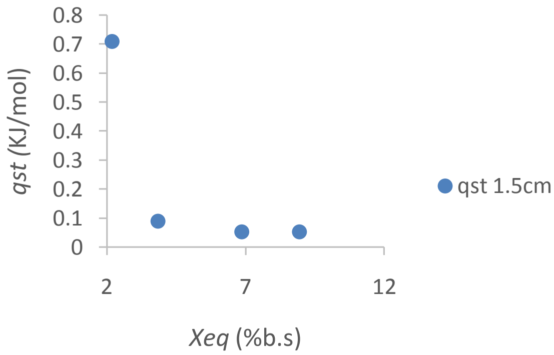
- **Thermodynamic properties**

The net isosteric heat (qst) and differential entropy (ΔS) of adsorption of conduction-dried TMs were calculated from the adsorption isosteres.Figure 3 shows respectively the net isosteric heat (qst) and the differential entropy (ΔS) of adsorption of the different TMs dried at the same temperature of 25°C.(should not reason with sulfuric acid concentrations) These curves show that for high water contents, the net isosteric heat and the differential entropy (ΔS) of adsorption tend towards zero.The experimental data of net isosteric heat (qst) and differential entropy (ΔS) were correlated with satisfaction (r = 0.999) according to the following relations:

$$qst = 0.052 + 29.147 E\exp(-1.733 Xeq) \text{(KJ/mol)} \quad (6)$$

$$\Delta S = 15.256 + 11782.956 E\exp(-1.860 Xeq) \text{ (J/mol)} \quad (7)$$





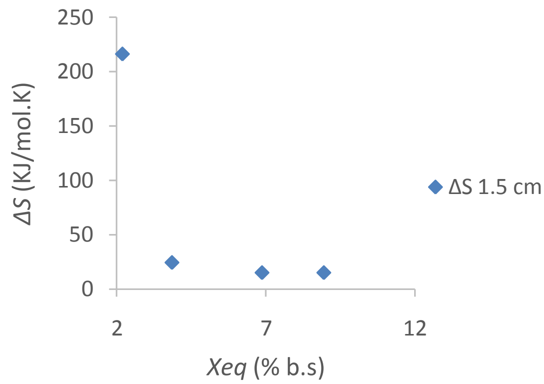


Figure 3:- Isosteric Heat (qst) and Differential Entropy (ΔS) at 1 and 1.5cm.

- **Compensation theory**

Figure 4 presents the enthalpy/entropy compensation theory of mango slices. This curve shows a linearity between enthalpy (net isosteric heat) and differential adsorption entropy.The isokinetic temperature $T\beta$, is 2.75°K and the free energy $\Delta G\beta$ is 0.773 J/mol.

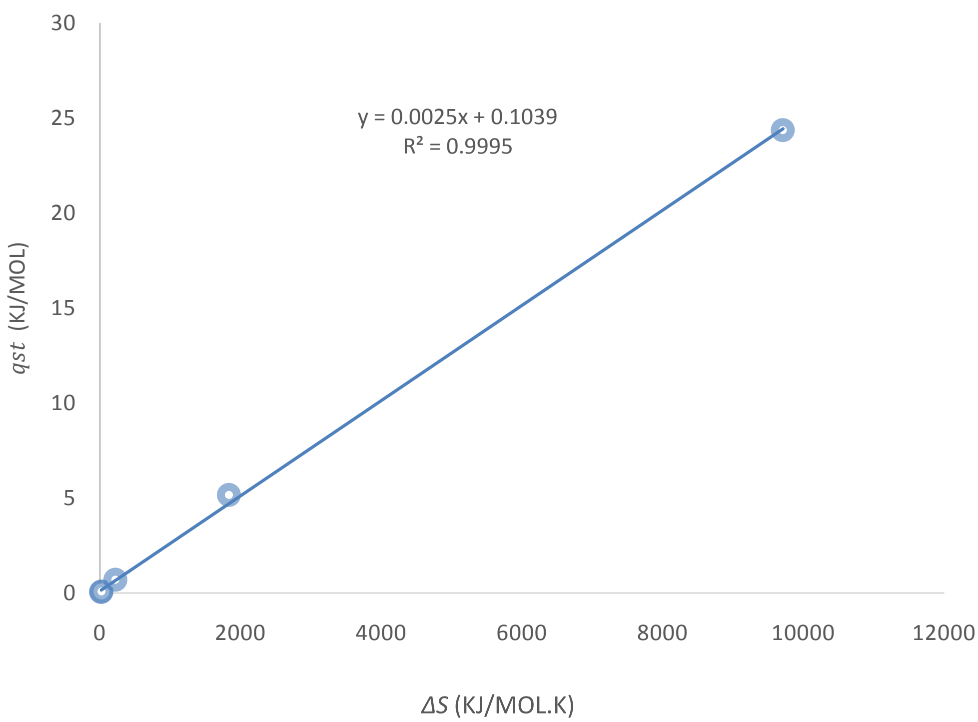


Figure 4:- Enthalpy/entropy compensation theory of dried mango slices.

Discussion:-

A high water content of TM could promote chemical and enzymatic reactions and the development of microorganisms leading to product deterioration (Amrouche, 2016).Indeed, this water content of food products plays a key role in preservation (Cheftel et al., 1984).It therefore appears essential to determine the minimum water content that could promote the conservation of TM.This is how the TM adsorption isotherms were determined and characterized.In addition, the hygroscopic equilibrium of these samples was obtained from 35 days.This time obtained is in agreement with those reported by kakou et al.(2015) who had to work on the cocoa bean isotherm in which the hygroscopic equilibrium was reached after 35 days for the last points of the curve.This modeling work has identified the GAB model with 3 parameters (Xm, K, C) as the most accurate model for determining information on adsorption isotherms.Indeed, according to GAB, the type of adsorption isotherm is determined by considering the values of the parameter C. When the parameter C ≥ 10, the isotherm is of type II and when C ≤ 10, the isotherm is of type III (Medeiros et al., 2006).The results of this study revealed values of the parameter C greater than 10 at 25°C.Therefore, the isotherms obtained with the two TM samples are type II with a characteristic sigmoidal shape.This type of adsorption isotherm was also observed in kernels of Irvingia gabonensis (Koko et al., 2018).This implies a formation of a monolayer, then a multilayer (Danion, 2004).The general shape of the adsorption isotherm curves of the dried mango slices is increasing with three (03) zones emerging.A first zone where the water content is relatively low due to the existence of a quantity of water strongly bound to the molecules contained in the product. It is characteristic of the action of Vander Waals forces between hydrophilic groups and water molecules.The adsorption of the water molecules of the TM is done gradually until a monolayer is formed covering the entire external surface of the pores of the product.Water is in a rigid state due to the strong bonding forces between water molecules and the surface.The transition to the next zone occurs when the entire surface is saturated.Zone (2) where the water content is moderately high corresponds to the adsorption of molecules on the initial monolayer.The isotherm is linear in this area and the water is in an intermediate state between solid and liquid in the product.Finally, zone 3 where the water content is very high corresponds to the water present in the liquid state in the pores of the product.The thickness of the film is sufficient for the water to be present in the liquid state in the pores of the product.In addition, the water content of 1cm TMs being higher than that of 1.5cm TMs due to the difference in thickness between these TMs.Indeed, the volume of the 1 cm TM being less voluminous will be in contact with the air which surrounds it, this will favor the rapid rehydration of the product and therefore its deterioration.Unlike that of 1.5 cm TM where the thickness is voluminous, the air which surrounds it will not quickly promote its rehydration and therefore its deterioration.The net isosteric heat and the differential entropy of the 1cm and 1.5cm slices are greater than 0 which means that the rehydration of the dried mango is not a spontaneous phenomenon but it takes place over a given time.These results are consistent with the behavior of biological products (Kouhila et al., 2002 and Lahsasni et al., 2003).A second determined characteristic is the water content of the mono-molecular layer.In this regard, the evaluation of the Xm parameters of the G.A.B model made it possible to reveal the water contents of the monolayer.Thus, at the storage temperature of 25°C, the water content is 2.295 – 2.289 % d.m.These values are similar to those obtained with the B.E.T model (2.30–2.29% d.m.) at the same temperature.They make it possible to ensure ideal conservation of the TMs.Indeed, the loss of quality of dehydrated products, due to chemical reactions, microorganisms and insects is unlikely below the value of the water content of the monolayer (Kakou et al., 2015).In addition, Karel et al. (1975) observed that at or below this value, the chemical alteration reactions are weak and the stability of the products is satisfactory during storage.In addition, many food products have an optimum water content at which stability is maximized.This is the case for dates with 6.52% (Ferradji et al., 2008) and cocoa beans (2.12%) (KaKou et al., 2015).The different absorption isotherms determined experimentally at a temperature of 25°C were adjusted by eight empirical models.Taking into account the largest correlation coefficient r and the smallest error estimates (MSD and MSE), the models best describing the experimental points were chosen.The analysis of these selection criteria revealed that the G.A.B model provides the best fit at 25°C.Indeed, he gave at this temperature, the largest r and the smallest error estimates.These results are in agreement with numerous works carried out on sorption isotherms where the G.A.B model simulated better, over a wide range of water activity, the experimental data (Ahouannou et al., 2010; Sandoval et al., 2002).The determination of the sorption isotherms is an essential step and a preferred means of determining the final water content to be achieved in order to optimize the storage and drying conditions of these products.They also provide valuable information on the hygroscopic balance of the product to be stored.

Conclusion:-

This study was carried out with the aim of determining the adsorption isotherms of different dried mango slices (under hot air) at 25°C in order to assess and identify the mango slice that could be kept for a long time in supermarketswhich requires a temperature of 25°C.The adsorption isotherms were determined by the static gravimetric method at 25°C.The analysis of the results obtained indicate that the hygroscopic equilibrium is obtained after 35 days.The adsorption isotherms determined are of type II with a characteristic sigmoidal shape for the two samples.This proves high hygroscopicity of dried mango slices.The theoretical curves obtained coincide with the experimental results.The GAB model is the one that correctly predicts the hygroscopic behavior of dried mango slices.No difference is observed between the equilibrium water contents of slices 1 to 1.5 cm thick.The compensation curve between enthalpy and entropy shows that the linear part correlates well for water adsorption of dried mango.The net isosteric heat and the differential adsorption entropy tend to zero when the water content of the different mango samples increases.Thus the positive value of the free energy (+ΔG) shows that the adsorption does not occur automatically.

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