

Estimation of Thermodynamic Parameters for Better Conservation of Fresh Tomato (*Lycopersicum esculentum*)

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Abstract

Knowledge of the state of water balance of agro-food products is an essential step in drying or storage operations for preservation. Our study made it possible to determine the thermodynamic parameters which influence the storage conditions of fresh tomatoes grown in the south of Benin; and to predict its hygroscopic behavior during post-harvest storage. The desorption isotherms obtained at 40°C, 50°C and 60°C, by the static gravimetric method using saturated saline solutions, are compared with those of the theoretical models of Brunauer, Emmet and Teller (BET), SMITH, PELEG and Guggenheim-Anderson-Boer (GAB). An adjustment of the experimental points, on the theoretical models, was made thanks to the numerical method which exploits the fminsearch algorithm under the MATLAB software, version R2018a. The GAB model at 50°C faithfully reproduces the experimental desorption curves for water activities from 5.5% to 82.3%. The net isosteric heat of sorption was determined using the Clausius-Clapeyron equation, it increases when the degree of dehydration of the product increases. The applied isokinetic theory and enthalpy-entropy compensation are consistent.

Keywords

Tomato, Isotherms, Theoretical Trend Model, Thermodynamic Properties,

1. Introduction

Food is the first vital need of man and food self-sufficiency must be considered an imperative for both humanis and economics. Food production must therefore increase significantly in order to meet the needs of a constantly growing population [1] [2]. The food production must therefore increase significantly in view to meet the needs of a constantly growing population. Thus, the recourse available natural resources consisting essentially of agricultural, livestock and fishing products becomes imperative [3] [4]. Among agricultural products, tomato occupies a prominent place in the dietary habits of populations throughout the world [5] reported that tomato production occupies an important place among the gardening market speculations produced in Benin with an annual production of 253.150 tons, in 2018. Indeed, tomato is the second most consumed fruit vegetable in the world after potato [6], and the first most consumed fruit vegetable in Benin [7]. According to [8], vegetable farming represents the main activity for many families, which have it as an important source of financial revenue. It would contribute to the creation of 60.000 direct jobs and 25.000 indirect jobs. Tomato is an important food component, mainly rich in potassium, phosphorus, magnesium; vitamin C and B9; and lycopene and β -carotene and possesses many virtues; so it is a potential source of vegetable proteins and micronutrients; it is a very powerful antioxidant [9]. Nevertheless, despite its excellent nutritional potential and its high demand by the populations, tomato remains a highly perishable foodstuff with a relatively high speed of alteration, which causes its rarity and high cost at a given time of the year. In Benin, and like other countries in sub-Saharan Africa, tomato fails to meet the daily food requirements of the population. Thereby, the trade balance in market garden products of this species is in deficit and Benin is required to import the tomato which comes from Nigeria, Burkina Faso and Togo. In addition to this deficit balance, huge post-harvest losses are recorded and are evaluated at 45% for Africa and more than 50% for Benin [10]; which usually leads to a loss of income and a decrease in the quantity of the product available for food. The transformation of tomatoes for conservation therefore becomes a necessity in order to reduce post-harvest losses and ensure food security for populations. Thus, the access of smallholder farmers to effective and easy-to-use techniques for post-harvest conservation becomes a development priority [11]. Among these techniques, drying remains the most accessible. It is all the more so for a good number of African populations because of the permanent availability and intensity of the solar deposit [12]. For a drying operation, the determination of the desorption isotherms is an essential step because these sorption curves provide information on the limit values of the product's water content during the desorption and adsorption processes and there-

fore promote the optimization of operations. In the analysis and conception of various equipment for drying and storing agricultural products for preservation, the knowledge of thermodynamic parameters such as net isosteric heat of sorption and differential entropy is essential for the estimation of energy necessary for the dehydration or drying of the product. In this study, we determined the desorption isotherms of tomato for three temperatures: 40 °C, 50 °C and 60 °C, in order to understand the diffusional mechanisms that take place in the products and which influence the desorption process, and then to predict the hygroscopic behavior during post-harvest storage.

2. Materials and Methods

2.1. Experimental Material

Tomato fruit

Tomato fruits of the so-called “*akikon*” variety are used for the experimental study. They come from the local market “Dantokpa” in the city of Cotonou and are commercially ripe (neither too ripe nor too green). The selected tomato fruits are washed with drinking water, drained and weighed, then cut into 1 cm thick strips, **Figure 1**.

The static gravimetric experimental method is used to determine the desorption isotherms of fresh tomatoes. Through this method, salts such as KOH, LiCl, MgCl₂, KI, NaCl, KCl, are used to prepare saturated saline solutions in airtight jars which are kept at a constant temperature. Each saline solution is obtained by dissolving a salt mass M_i in the jar containing a volume V of distilled water. The mass of salt (M_i) and the volume of distilled water are mixed to obtain a saturated salt solution shown in **Table 1**. The masses obtained are distinct for the different types of salts used in saline solutions. This difference in the masses of the salts is due to the difference in solubilities. The different solubility values at 25 °C of the different salts are presented in **Table 1**.

2.2. Saturated Saltsolution Bottle and Dehydration Oven

In this atmospheric condition (**Figure 2**), these saturated saline solutions make



Figure 1. Selected tomato fresh variety “Akikon” sliced for testing.

Table 1. Variation of saturation from saline solutions.

Variables	KOH	LiCl	MgCl ₂	KI	NaCl	KCl
Masses of salt (g)	300	120	120	264	264	120
Volumes of water (ml)	200	150	200	200	200	200
Solubilities (g/l) at 25°C)	1100	769	542	1430	358	344

**Figure 2.** Ventilated desiccation oven for tomato slices/controlled temperature and humidity.**Table 2.** Standard value of water activity as a function of salts and temperature.

Salts	$\ln(a_w) = \frac{K_1}{T} - K_2$	a_w (%)		
		40°C	50°C	60°C
KOH	$\ln a_w = (2094.4890 \times 1/T) - 9.4977$	0.0630	0.0570	0.0550
LiCl	$\ln a_w = (10.8233 \times 1/T) - 2.2193$	0.1120	0.1110	0.1100
MgCl ₂	$\ln a_w = (151.0652 \times 1/T) - 1.6271$	0.3120	0.3050	0.2930
KI	$\ln a_w = (258.1545 \times 1/T) - 1.2388$	0.6610	0.6450	0.6310
NaCl	$\ln a_w = (23.1092 \times 1/T) - 0.3607$	0.7470	0.7440	0.7450
KCl	$\ln a_w = (157.0587 \times 1/T) - 0.6967$	0.8230	0.8120	0.8030

Source: [13].

it possible to obtain relative humidities varying in the range of values from 5.5% to 82.3%, for temperatures set at 40°C, 50°C or 60°C, **Table 2**.

The experiment consists of putting a portion of the tomato sample to be desorbed in the hermetically sealed isothermal jars, which respectively contain the saturated solutions of salts mentioned above, maintained at a constant relative humidity because of the nature of the salt and at a constant temperature imposed in the regulated oven (**Figure 2**). The desorption experiments are carried out at 40°C, 50°C and 60°C. Before putting the product to be desorbed in the jars, the latter are introduced and left in the regulated oven for 24 hours so that the saline solutions can adapt to the set temperature setpoint. The mass mea-

surement of the insulated jar + product is carried out every 24 hours until the variation in mass between two successive measurements becomes negligible. When the different equilibrium masses (M_{eq}) are obtained for the different climatic conditions, the samples tested are taken up and then immediately placed in the drying oven set at $103^{\circ}\text{C} \pm 2^{\circ}\text{C}$. This operation makes it possible to obtain the dry extract of the sample studied, which in this condition undergoes the total loss of the free and bound water it contains [11] [14]. The samples are regularly weighed and if after 24 hours their masses no longer vary, then the dry extract of the product is obtained. Thus, the mass of the dry extract measured for each sample is considered as the anhydrous mass (M_s). From this parameter obtained, we defined the different equilibrium water contents of the samples placed in the jars. The equilibrium water contents (X_{eq}) of the samples placed in the jars. The equilibrium water content of the fresh tomato on a dry basis is calculated by the following expression:

$$X_{eq} = \frac{M_{eq} - M_s}{M_s} \quad (1)$$

For a given temperature, the pairs of values obtained (H_{ri}, X_{eqi}) for $i = 1, \dots, n \in N$, made it possible to experimentally trace the desorption isotherms of the tomato. Therefore, the water content of the product at equilibrium can be expressed by the following relationship:

$$X_{eq} = f(a_w)_{T_s} \quad (2)$$

T_s : System equilibrium temperature in Kelvin; a_w : water activity of the product, as a decimal fraction given by the relationship:

$$a_w = \frac{P_v}{P_{vsat}} = \frac{H_r (\%)}{100} \quad (3)$$

The relationship between water activity, temperature and saturated salt solutions dissolved in distilled water is determined by the equation (Labuza *et al.*, 1985):

$$\ln(a_w) = \frac{K_1}{T} - K_2 \quad (4)$$

2.3. Theoretical Models Used to Determine Desorption Isotherms

The theoretical models of BET, SMITH, PELEG and GAB are used to reproduce the tomato desorption isotherm curves; they are compared with the experimental results. The usefulness of desorption isotherms is to know the optimal conditions of temperature and hygrometry favorable for conservation, in other words the conditions of hygrothermal equilibrium of the product with the environmental atmosphere. Thus, the various thermodynamic parameters calculated previously are deduced directly or indirectly from the equilibrium conditions offered by the isotherms. The desorption isotherms can be used to conduct an optimal drying operation. They make it possible to estimate the quantity of heat necessary and essential for a drying process to bring a product from its fresh

state to a state of favorable water content for its conservation. In this paragraph of our study, four (04) mathematical models taken from the literature are exploited for an optimal adjustment of the experimental data. They are:

Model of BET (1938)

The equation of Brunauer, Emmett and Teller "Equation of BET" is essential to appreciate the degree of the water content of the monolayer, an important parameter in the study of the deteriorations of food products [15]. This model is only valid up to $a_w = 0.5$, [11] [15]. It is represented by the following equation:

$$X_{eq} = \frac{X_m C a_w}{(1 - a_w)(1 + (C - 1)a_w)} \quad (5)$$

In this expression, X_{eq} is the equilibrium moisture content (g water/g, dry basis); X_m is the monolayer moisture content (g/g, dry basis); C , BET constant; a_w is the water activity. The transformation of expression (1) gives a linear equation $f(X) = b + a \cdot X$, such that:

$$\frac{a_w}{(1 - a_w)X_{eq}} = A a_w + B \quad (6)$$

$$\text{With } A = \frac{C - 1}{C X_m} \text{ and } B = \frac{1}{C X_m}$$

The resolution of Equation (2) by the method of least squares, makes it possible to determine the slope (A) and the ordinate at the origin (B). The parameters X_m and C being constants, they are obtained by the following relation:

$$X_m = \frac{1}{A + B} \text{ and } C = \frac{A + B}{B} \quad (7)$$

Model of SMITH (1947)

The SMITH equation has two constants [16]. It is of the form $f(X) = b + a \cdot X$ and is frequently used to simulate the isotherms of biological products. It is given by the following expression:

$$X_{eq} = K_1 - K_2 \cdot \ln(1 - a_w) \quad (8)$$

A linearization of the function $X_{eq} = f(\ln(1 - a_w))$ makes it possible to determine the constants K_1 and K_2 of Equation (4).

Model of PELEG (1993)

PELEG has developed an empirical equation in polynomial form [17]:

$$X_{eq} = C_1 a_w^a + C_2 a_w^b \quad (9)$$

GAB of model (1981)

The theoretical model of Guggenheim-Anderson-Boer (GAB model) established in 1985, represents a methodically elaborated extension of BET theories. The theoretical basis for the determination of the GAB sorption isotherm is the assumption of localized physical adsorption in the multilayers, with non-lateral interactions. This model is widely used nowadays and showed a good fit up to a water activity of the order of 0.9 [11] [18]. It allows a good evaluation of the quantity of strongly bound water at the level of the primary adsorption sites

(X_m). Beyond its reliable theoretical basis, this model includes parameters that physically reflect the sorption process. It also makes it possible to describe the effect of temperature on sorption isotherms using Arrhenius-type equations.

$$X_{eq} = \frac{X_m CKa_w}{(1 - Ka_w)(1 - Ka_w + CKa_w)} \quad (10)$$

Equation (10) rearranged in the form of a polynomial function gives expression (11):

$$\frac{a_w}{X_{eq}} = \frac{K}{X_m \left(\frac{1-C}{C} \right)} a_w^2 + \frac{C-2}{X_m C} a_w + \frac{1}{X_m KC} \quad (11)$$

which takes the general form:

$$\frac{a_w}{X_{eq}} = \alpha a_w^2 + \beta a_w + \gamma \quad (12)$$

With

$$\alpha = \frac{K}{X_m} \cdot \frac{C}{1-C}; \quad \beta = \frac{C-2}{X_m C}; \quad \gamma = \frac{1}{X_m KC}$$

A numerical resolution of Equation (7) using the quadratic regression method made it possible to approach the experimental values obtained. The parameters K , X_m and C of the G.A.B equation were calculated after solving the nonlinear Equation (11) by the least squares method as follows [19]:

$$K = \frac{\sqrt{f} - \beta}{2\gamma}, \quad X_m = \frac{1}{\beta + 2K\gamma}, \quad C = 2 + \frac{\beta}{K\gamma} \quad (13)$$

where $f \cong \beta^2 - 4\alpha\gamma$

We will also note that C and K are correlated with temperature by the Arrhenius equation [14]:

$$K = K_o \exp \frac{\Delta H_k}{RT}; \quad C = C_o \exp \frac{\Delta H_C}{RT} \quad (14)$$

With $\Delta H_C = H_m - H_n$ and $\Delta H_k = H_L - H_n$.

According to [20], C_o , K_o are entropic proportionality factors; T , absolute temperature ($^{\circ}\text{K}$); R , universal ideal gases constant (8.3145 J/mole-K); H_m , total heat sorption of monolayer; H_n , total heat sorption of multilayer; H_L , latent heat of condensation of liquid water, either 43.98 kJ/mole over the range (20°C - 80°C).

Statistical criteria for model validation

The numerical method of local optimization of Algorithm `fminsearch` was used under the software MATLAB version R2018a for the simulation of the experimental points. This localized optimization method aims to converge to the nearest local optimum. The highest correlation coefficient (r^2), the lowest mean squared (*RMSE*) and standard errors (*EST*) are the criteria for choosing the best model.

$$r^2 = \left| \frac{\sum_{i=1}^n (X_{eqi,pre} - \bar{X}_{eqi,exp})^2}{\sum_{i=1}^n (X_{eqi,exp} - \bar{X}_{eqi,exp})^2} \right|^{\frac{1}{2}} \quad (15)$$

$$RMSE = \frac{100}{N} \sum_{i=1}^N \left| \frac{X_{eqi,exp} - X_{eqi,pre}}{X_{eqi,exp}} \right| \quad (16)$$

$$EST = \left| \frac{\sum_{i=1}^n (X_{eqi,exp} - X_{eqi,pre})^2}{df} \right|^{\frac{1}{2}} \quad (17)$$

$$Ei = |X_{eqi,exp} - X_{eqi,pre}| \quad (18)$$

$X_{eqi,exp}$: ith experimental equilibrium water content; $X_{eqi,pre}$: ith predicted equilibrium water content on a dry basis (% dm). N : number of experimental points, {in our case, $N = 9$ } and $df = N - n$ is degree of freedom of the regression of the model, with n the number of constants of each model.

From the theoretical models presented above, it has been possible for us to determine distinct series of points (a_w, X_{eq}) in order to predict the desorption isotherm curves of tomato and to say which ones reproduce faithfully the experimental curves found.

2.4. Determination of the Thermodynamic Properties of the Desorption Process

Activation energy

From the Arrhenius equation, a correlation between the water content of the monolayer, X_m of GAB and the temperature, is established [1] [21].

$$X_m = X'_m \exp\left(\frac{-E_a}{RT}\right) = X'_m \exp\left(\frac{-E_a}{R} \frac{1}{T}\right) \quad (19)$$

With

E_a : activation energy (J/mol), X'_m : collision frequency (%MS). The representation of the graph $\ln(X_m) = f\left(\frac{1}{T}\right)$, makes it possible to determine the slope $\frac{-E_a}{R}$ and the ordinate at the origin $\ln(X'_m)$.

Net isosteric heat of desorption

The Clausius-Clapeyron equation, due to its theoretical basis, is often used in the determination of the total heat of sorption (Q_{st}) of agricultural products [22] [23] [24]. This equation is applied to estimate the quantity of heat useful for the evaporation of water (free + bound) in a product. It is given by the following expression:

$$\left. \frac{d(\ln P)}{dT} \right|_X = \frac{Q_{st}}{R_0 T^2} \quad (20)$$

For pure water in the free state, the equation (Eq.16) becomes:

$$\frac{d(\ln P_o)}{dT} = \frac{L_v}{R_o T^2} \quad (21)$$

With

P : water vapor pressure of the product at the specified temperature; P_o : saturation water vapor pressure at temperature T and L_v ; latent heat of vaporization of pure water in the free state.

Substituting Equation (20) into Equation (21) and arranging the result, we have:

$$\left. \frac{d\left(\ln \frac{P}{P_o}\right)}{dT} \right|_X = \left. \frac{d \ln a_w}{dT} \right|_X = \frac{q_{st}}{R_o T^2} \quad (22)$$

With

$q_{st} = Q_{st} - L_v$, net isosteric heat of sorption (kJ/mol) at equilibrium water content, X_{eq}

Integrating Equation (18) gives Equation (19):

$$\ln a_w = \left(\frac{-q_{st}}{R_o} \right) \frac{1}{T} + C_{st} \quad (23)$$

The graphical representation of equation (19), for a fixed water content and a known value of R , allows us to determine the slope $\left(\frac{-q_{st}}{R_o} \right)$ and the integration constant C_{st} .

From the value of the slope and for each water content X_{eq} we can deduce the corresponding net isosteric heat, and plot the curve $q_{st} = f(X_{eq})$. In the same vein, Tsami *et al.* [20] propose the following empirical correlation to describe a relationship between net isosteric heat of desorption and equilibrium water content.

$$q_{st} = q_o \exp\left(-\frac{X_{eq}}{X_o}\right) \quad (24)$$

With q_o , net isosteric heat of desorption of the first water molecules in the product (KJ/mol) and X_o , characteristic water content at which the net isosteric heat of desorption was deduced, *i.e.* 63% [22].

Enthalpy-Entropy compensation theory applied to the isosteric heat of desorption

The differential entropy is calculated from the Gibbs-Helmholtz equation, Equation (24) where the Gibbs free energy is obtained by the equation Equation (25), [11] [24].

$$\Delta S = -\frac{q_{st} - \Delta G}{T} \quad (25)$$

$$\Delta G = RT_o \ln\left(\frac{P}{P_o}\right) = RT \ln(a_w) \quad (26)$$

Substituting Equation (25) into Equation (26) and arranging the result, we get:

$$\ln(a_w) = -\frac{q_{st}}{RT} + \frac{\Delta S}{R} \quad (27)$$

From the ordinate at the origin of the lines of the isosteres and for each water content at equilibrium X_{eq} , we deduce the corresponding differential entropy of sorption and we draw the curve $\Delta S = f(X_{eq})$.

According to the compensation theory, the linear ratio between enthalpy and entropy for a specific reaction is given by:

$$q_{st} = T_\beta (\Delta S) + \Delta G_\beta \quad (28)$$

With T_β : isokinetic temperature for which all reactions in the series are at the same rate and ΔG_β : the free energy at temperature (T_β) used to assess whether the water sorption process is spontaneous: ($-\Delta G_\beta$) or not ($+\Delta G_\beta$). So, T_β and ΔG_β are obtained by drawing the curve $q_{st} = f(\Delta S)$.

2.5. Test on Compensation Theory

Some authors [25] [26] recommend a test for the compensation theory which consists in comparing the isokinetic temperature to the mean harmonic temperature (Th_m) which, is defined by [27] [28]:

$$Th_m = \frac{n}{\sum_{i=1}^n (1/T_n)} \quad (29)$$

Isokinetic temperature (T_β) is calculated by linear regression for all data sets with a 95% confidence interval.

$$T_\beta = T_\beta \pm t_{m-2, \alpha} / 2\sqrt{Var(T_\beta)} \quad (30)$$

$$T_\beta = \frac{\sum (q_{st} - \overline{q_{st}})(\Delta S - \overline{\Delta S})}{(\Delta S - \overline{\Delta S})^2} \quad (31)$$

$$Var(T_\beta) = \frac{\sum (q_{st} - \overline{q_{st}} - T_\beta \Delta S)^2}{(m-2) \sum (\Delta S - \overline{\Delta S})^2} \quad (32)$$

With n : total number of isothermal points; m : number of (q_{st} , ΔS) data pairs; ($\overline{q_{st}}$): the average enthalpy and $\overline{\Delta S}$: the average entropy.

3. Results and Discussion

The experimental determination of the desorption isotherms requires a fairly long time, because they are defined from several measurement points obtained under the conditions described above and for various stable atmospheric environments. Obtaining a first point of an isotherm curve lasts on average 7 to 10 days for all agricultural products with a high water content. For the other points, the time for the mass of the sample to stabilize was on average 96 hours. This observation confirms the results of [29] on the determination of the isotherms of

ginger, red pepper and cassava. Therefore, it will then take about 45 days to experimentally obtain a desorption isotherm curve of a product, with seven remarkable. This approach, although effective, is undoubtedly tedious and can be highly tainted with errors, when the manipulator is not very knowledgeable and well applied [18] [29]. We present in **Table 3**, the pairs of parameters (a_w , X_{eq}) representative of the points of the isotherms.

Table 3. Water content at equilibrium, X_{eq} (g water/100g ms) as a function of a_w (%) at 40°C, 50°C and 60°C.

Salts	$T=40^\circ\text{C}$		$T=50^\circ\text{C}$		$T=60^\circ\text{C}$	
	a_w	X_{eq}	a_w	X_{eq}	a_w	X_{eq}
KOH	0.0630	0.0945	0.0570	0.0683	0.0550	0.0500
LiCl	0.1120	0.1063	0.1110	0.0803	0.1100	0.0560
MgCl ₂	0.3160	0.1641	0.3050	0.1232	0.2930	0.0809
KI	0.6609	0.4120	0.6449	0.3400	0.6311	0.2208
NaCl	0.7470	0.5574	0.7440	0.4500	0.7420	0.2910
KCl	0.8230	0.8584	0.8130	0.6630	0.8030	0.5061

In this table, it can be seen that for the same saturated saline solution, an increase in the temperature in the regulated enclosure leads to a decrease in the ambient relative humidity and in the equilibrium water content of the product (**Figure 1**). Indeed, for high temperatures, the state of excitation of water molecules is greater leading to a decrease in the forces of attraction of water molecules between them [11] [29]. Our results on isotherms of food products are in perfect agreement with those of other authors such [30] [31] [32]. However, some authors [33] [34] have reported that for sugar-rich products such as fruits, there is an inversion effect where the curves cross for the values water activity (greater than 0.6), and this is due to the increased solubility of sugars where solubilization reduces water mobility. The dependence of temperature with the equilibrium water content has an important role on the concrete incidence of chemical and microbiological reactions associated with the deterioration of the product [33].

Validation of theoretical models

The experimental results obtained were compared with the theoretical models of BET, Smith, Peleg and GAB. The adjustments of these experimental data were made using the local optimization algorithm of Fminsearch under the software MATLAB version R2018a, and the parameters of the model are determined. These values are presented in **Table 4** along with the correlation coefficients (r) and the root mean square errors ($RMSE$) of the adjustment.

According to some authors such as [30] [35] [36], the models which present the high values of r ($r = 1$) and the low values of $RMSE$ ($RMSE = 0$) indicates a perfect fit of the experimental points. Among the models studied, that of G.A.B gives the greatest value of correlation coefficient and the lowest value of $RMSE$

Table 4. Estimated values of the adjustment parameters and statistical criteria for choosing the model.

$T(^{\circ}\text{C})$	Parameters	Theoretical models			
		BET ($n = 2$)	SMITH ($n = 2$)	PELEG ($n = 4$)	GAB ($n = 5$)
40	X_m	0.3378	-	-	0.1297
	C	2.4248	-	-	23.8905
	K	-	-	-	1.0329
	C_1	-	-	0.2722	-
	C_2	-	-	1.9166	-
	A	-	0.0207	0.4116	-
	B	-	0.4310	5.9572	-
	R^2	0.8888	0.9634	0.9782	0.9999
	$RMSE$	0.0680	0.0389	0.0085	0.00053
50	X_m	0.5765	-	-	0.1120
	C	1.1406	-	-	15.2213
	K	-	-	-	1.0244
	C_1	-	-	0.2168	-
	C_2	-	-	1.3189	-
	a	-	0.0153	0.4372	-
	b	-	0.3505	5.1493	-
	R^2	0.9118	0.9745	0.9796	1.0000
	$RMSE$	0.0479	0.0255	0.0106	0.00017
60	X_m	0.3343	-	-	0.0582
	C	1.3999	-	-	59.7818
	K	-	-	-	1.1001
	C_1	-	-	0.3041	-
	C_2	-	-	19.9214	-
	a	-	0.0069	0.8423	-
	b	-	0.2565	19.9214	-
	R^2	0.8491	0.9258	0.9624	1.0000
	$RMSE$	0.0460	0.0321	0.0905	0.00820

n = Number of model parameters.

in the range of temperatures studied. Thus, the GAB model remains the best model for smoothing experimental data. We also note that the Peleg model has high values of correlation coefficient r^2 and low $RMSE$. The theoretical curves

resulting from the four models studied are illustrated in **Figure 2**. According to [37], the values of the parameter C of the GAB equation make it possible to characterize the sorption isotherms. Indeed, when $C \leq 10$, the isotherm is of type III, while for $C \geq 10$, the isotherm is of type II. In our study, the values of the parameter C are higher than 10 for the model of G.A.B whatever the temperature. Thus, the desorption isotherms obtained are of type II. They are therefore of sigmoidal shape, characteristic of a multimolecular absorption, which reflects the progressive thickening of the absorbed layer. This type of isotherm is characteristic of the hygroscopic behavior of most food products [11] [18] [38]. The results of the isotherms obtained experimentally are in perfect agreement with the behavior of biological products, particularly products with a high-water content [27] [39] [40] [41]. We will retain the water activity domain for the BET model is between 0 and 35% according to several authors. The following curves present for each study temperature, the experimental points and the adjustment models, see **Figure 3**.

3.1. Residual Error Curves between Experimental and Predicted

The residual error curves resulting from the confrontation of the experimental points and the theoretical models of GAB and Peleg at 40°C, 50°C and 60°C are presented in **Figure 4**. We note that these curves are almost merged with the abscissa axis in the range of water activities between 5.5% and 82.3%, especially

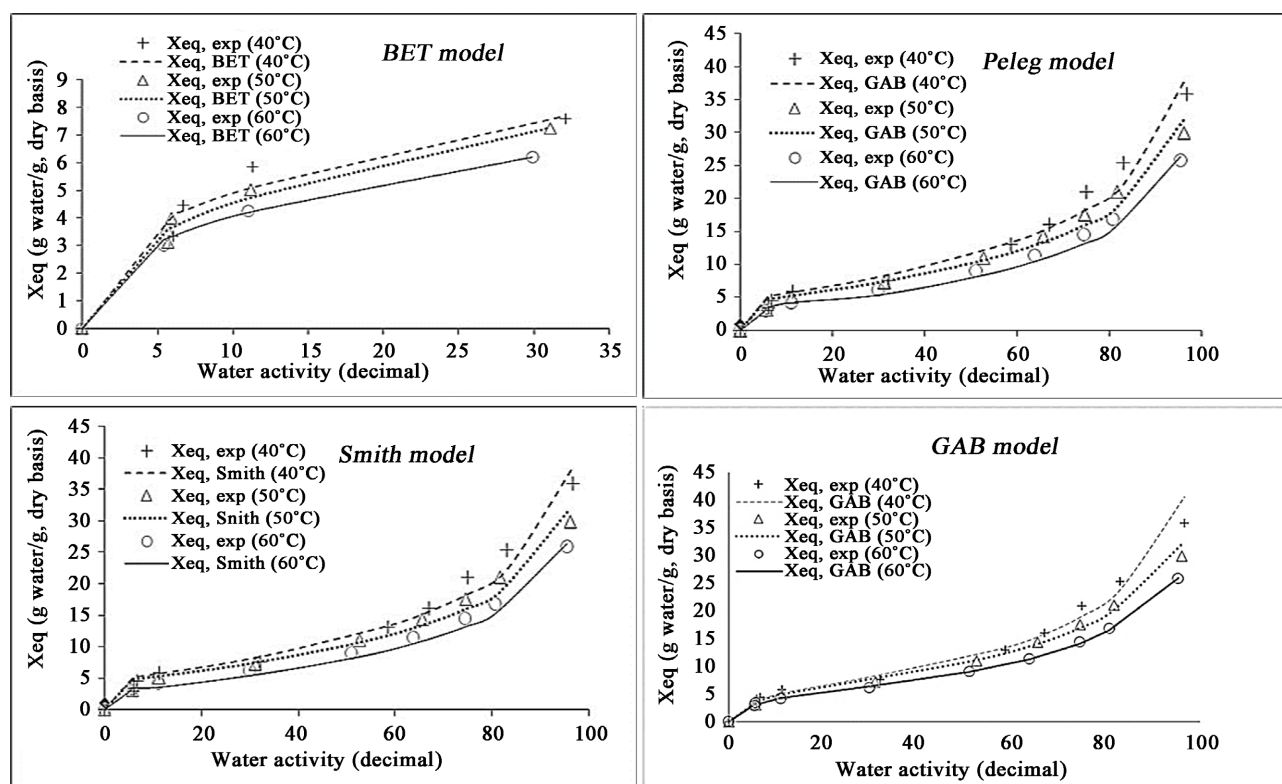
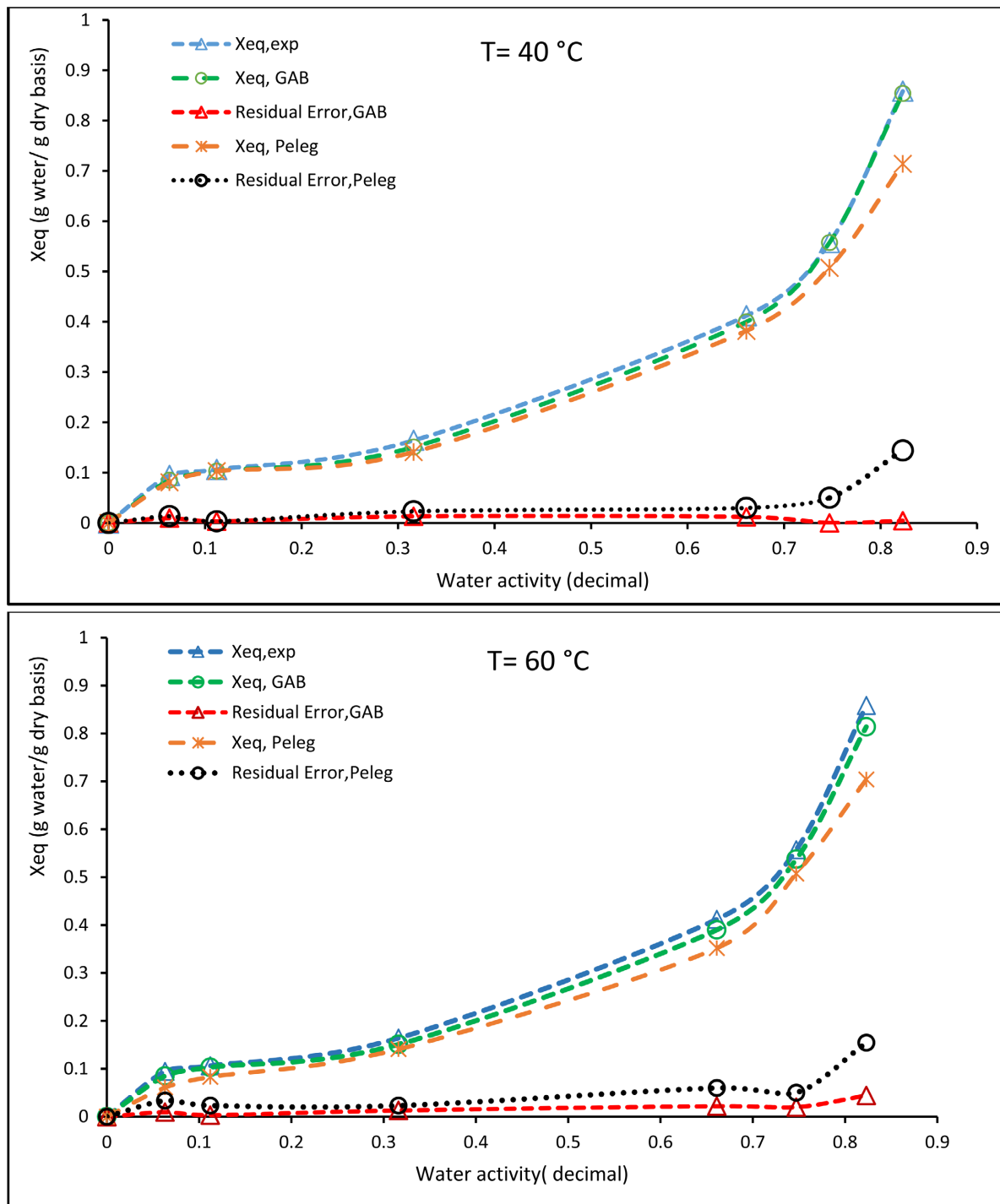


Figure 3. Representation of the experimental desorption isotherm curves compared to the theoretical models of BET, SMITH, PELEG, and GAB.

at 50 °C, which testifies to the effectiveness of these models at this temperature. This observation justifies the higher values of r^2 ($r = 1$) and the lower values of $RMSE$ ($R = 0.000106$) for GAB, compared to those at 40 °C and 60 °C.

3.2. Isotheric Heat of Sorption

The net isotheric heat of sorption is obtained from the slope of the curve deduced from Equation (33), for a given equilibrium water content (Figure 5).



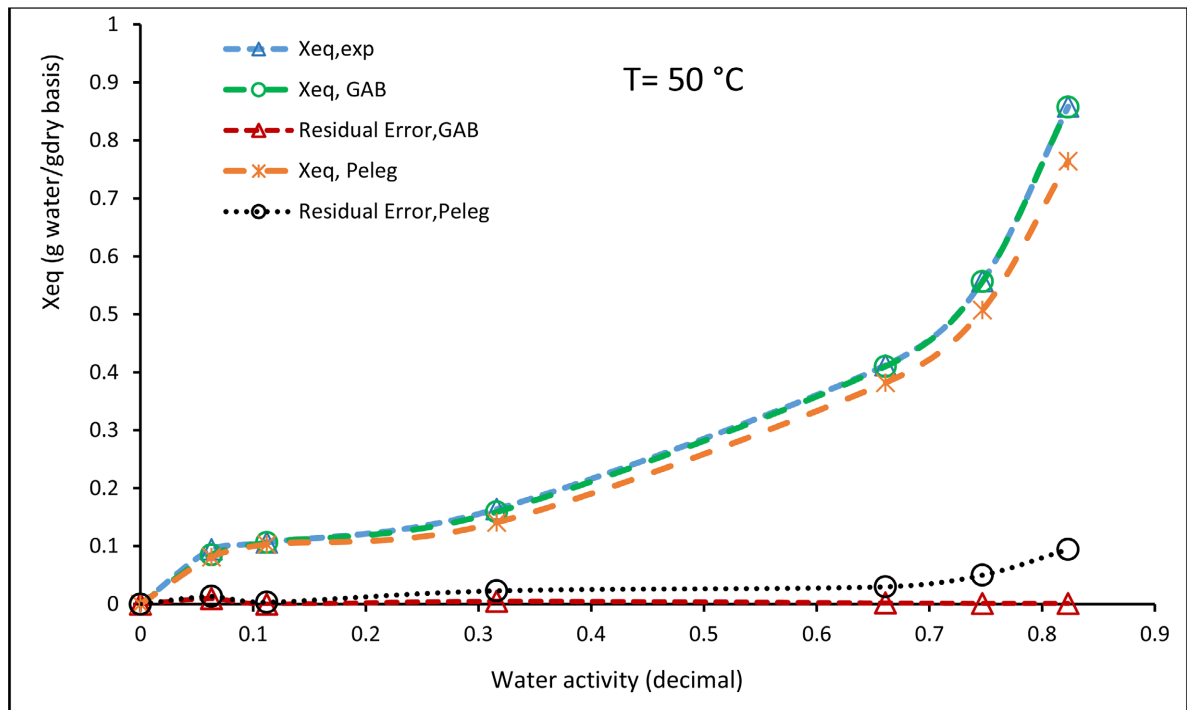


Figure 4. Residual error curves between experimental and predicted isotherms by Peleg and GAB at 40°C, 50°C and 60°C.

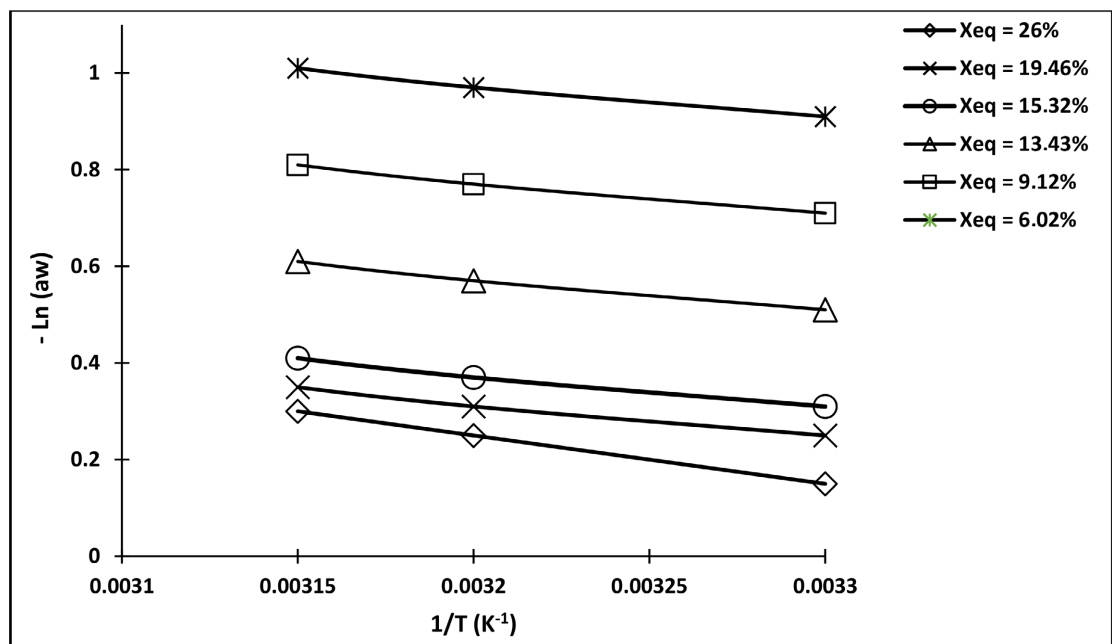


Figure 5. Desorption isosteres at different of equilibrium humidity values.

Thus, water activity values at different temperatures are calculated for each equilibrium water content, by applying the model parameters of GAB in its transformed form, according to [42] [43]; confers Equation (34). From the slope of the isosteres and for each water content X_{eq} one can deduce the corresponding net isosteric heat and plot the curve $q_{st} = f(X_{eq})$. The experimental data of net

isosteric heat and differential entropy were correlated with satisfaction; $R^2 = 1$.

$$a_w = \left[2 + \left(\frac{X_m}{X_{eq}} - 1 \right) C - \left\{ \left(2 + \left(\frac{X_m}{X_{eq}} - 1 \right) C \right)^2 - 4(1-C) \right\}^{\frac{1}{2}} \right] / [2K(1-C)] \quad (33)$$

From **Figure 6**, we notice that for high water contents, the net isosteric heat and the differential entropy of desorption tend towards zero. This means that the isosteric heat of sorption is equal to the heat of condensation of water. In this zone of high water content, the net isosteric heat is positive and this indicates that the interaction between water and the substrate of the product is endothermic, which corresponds to the dissolution of sugars. The very high initial heat of sorption at low water contents indicates that the interaction between agricultural product components and water is important. When the water content increases, the available sorption sites decrease and thus lead to a decrease in the values of the heats of sorption. The isosteric heat of desorption (q_{sd}) becomes negligible compared to the latent heat for high humidity. The existence of highly active polar sites in tomato, when covered with water molecules forming the monolayer could explain this fact [20]. According to other authors [44] [45], this phenomenon is due to the fact that in a very restricted range of humidity, when the water content increases; certain products swell, favoring the opening of new adsorption sites for strong bonds; which increases the isosteric heat.

The values of the characteristic water content of the product at which the net isosteric heat of desorption was deducted, *i.e.* 63% (X_o) and that of the net isosteric heat of desorption of the first molecules of water in the product (q_o) determined from the linear empirical Equation (22), by the method of least squares are transcribed in **Table 5**. From the analysis of this table, it is observed that the

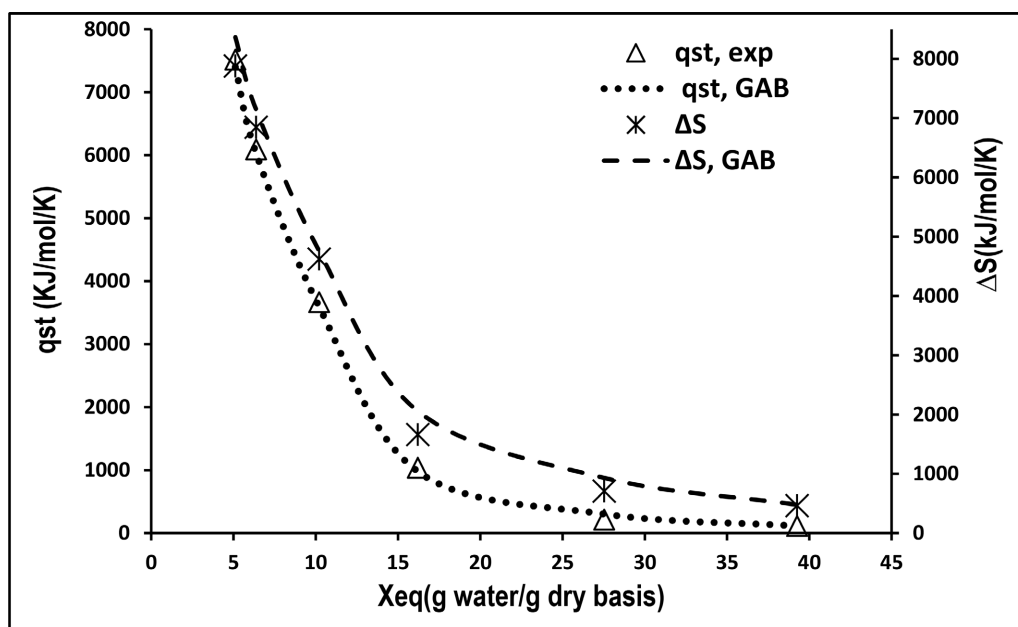


Figure 6. Experimental and predicted isosteric heat and differential entropy by GAB.

Table 5. Values of X_o and q_o of tomato fruit.

Parameters	$T = 40^\circ\text{C}$	$T = 50^\circ\text{C}$	$T = 60^\circ\text{C}$
X_o (g d'eau/gms)	0.070	0.060	0.050
q_o (KJ/mol)	16.300	16.390	16.370
r^2	0.937	0.988	0.999

increase in temperature leads to a decrease in the characteristic water content ($X_o = 0.0808$ g water/gms). The net heat of isosteric desorption of the first layers of water molecules in the product (q_o) is almost constant at 16.37 ± 0.98 kJ/mol in the range of study temperatures. These results are similar to 16.99 kJ/mol obtained by [32] for the desorption of fig fruit. Indeed, the q_o parameter provides important information on the physico-chemical interactions of water with the major constituents of the product and the state of the water in it, an essential parameter for estimating energy requirements during drying [22].

Activation energy

The activation energy is the amount of energy required to initiate a reaction, *i.e.* the free enthalpy that separates the energy of the transition state and that of the initial state. **Figure 7** presents the logarithmic variation of the water contents of the monolayer (X_m) of GAB and BET as the function of the inverse of the temperature.

A linearization of the curve $\ln(X_m) = f(1/T)$, by the theory of least squares made it possible to determine the slope $-E_a/R$ and the ordinate at the origin $\ln X^o$. Thus, the values of the activation energy (E_a) determined from the monolayer of GAB and BET are respectively -13.1009539 K/mol and -13.674442 K/mol, with a respective collision frequency X^o of 0.45646 and 0.30546 (%MS). Indeed, the negative values (E_a) obtained explain that the tomato desorption process is exothermic. The comparison of the logarithmic variation of the monolayer (X_m) of GAB and that of BET gives an affine straight line reflecting the existence of a proportionality between the monolayer (X_m) of GAB and BET with a coefficient of proportionality equal to 1.037 and a correlation coefficient $R^2 = 0.992$.

3.3. Enthalpy-Entropy Compensation

The enthalpy-entropy compensation theory is widely applied to evaluate the physical and chemical phenomena that accompany sorption reactions [11]. This theory confirms the natural changes in the interaction between the solvent and the substrate; and provides information on the perfect linearity of the relationship between these two state functions [37]. The curves obtained (**Figure 8**) show perfect agreement ($R^2 = 0.999$) between these two parameters. The values of the isokinetic temperature T_β and the harmonic mean temperature Th_m are 314.171 K and 314.459 K respectively, and the free energy ΔG_β is 308.18 J/mol. The value of T_β obtained for tomato is lower than that found for garlic (348 K) [30], mango (463 ± 4 K) [30], spaste (330.7 K) [31]. On the other hand, [42] obtained very low T_β value (0.0034 K) for cocoa beans. The mean harmonic temperature

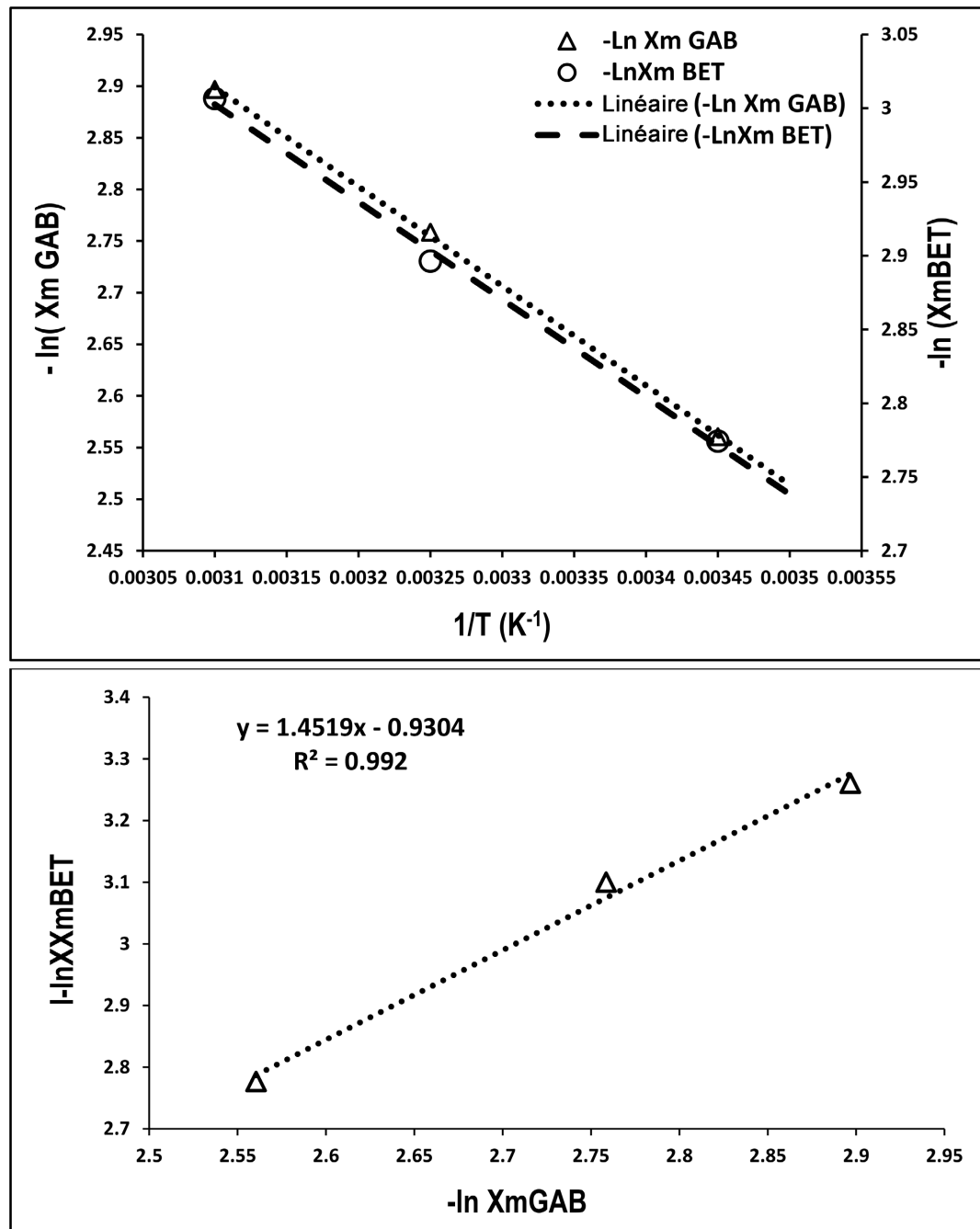


Figure 7. Logarithmic variation of X_m GAB and X_m BET as a function of the inverse of temperature.

$T_{hm} = 314.459$ K clearly confirms the relevance of the isokinetic theory for water desorption from tomatoes. The values of (T_β) and (Th_m) obtained show that the enthalpy process is driven; when $T_\beta < Th_m$, the enthalpy process is said to be driven, on the other hand if $Th_m < T_\beta$ the process is considered as controlled entropy. The positive value of the free energy ΔG_β (308.18 J/mol) found shows that the desorption process is non-spontaneous [46]. In fact, the enthalpy-entropy compensation process thermodynamically manifests the structuring-destructuring of water. The variation of the enthalpy corresponds to the variations of energy

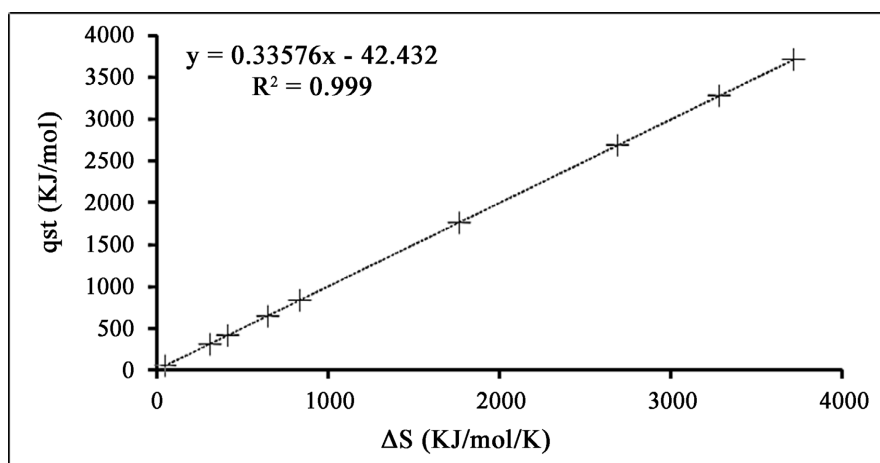


Figure 8. Enthalpy-Entropy compensation of tomato for temperatures 40°C, 50°C, 60°C.

which occur between the molecules of water and that of the matrix during the operations of sorption. While entropy reflects the disorder and quality of conversions [11] [18].

4. Conclusion

The determination of the desorption isotherms constitutes an essential step for the conservation of tomato. These isotherms determined by the static gravimetric method were successfully correlated by the GAB model at 40°C, 50°C and 60°C. These results showed that the equilibrium water content decreases with increasing temperature in the whole range of water activity (5.5% to 82.3%). These isotherms obtained have a sigmoidal shape (type II) characteristic of a multimolecular absorption. The curves of residues between the experimental isotherms and those of GAB theory show low values at 50°C. The net isosteric heat of sorption of tomato was determined using the Clausius-Clapeyron equation. It increases as the degree of dehydration increases, it varies exponentially with the equilibrium water content. An empirical exponential correlation has been established to describe the dependence of the heat of sorption on the equilibrium water content. Enthalpy-entropy compensation and isokinetic theory have been successfully applied to explain the influence of temperature on the desorption process of tomato. The study of these thermodynamic parameters will make it possible to simulate the optimal conditions for tomato drying.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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