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Hygroscopicity of Castor Bean Seeds (*Ricinus communis* L.)

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Abstract

The hygroscopic balance of castor bean seeds (*Ricinus communis* L.) assists the proper handling of the product to preserve its water content at the levels recommended for safe storage. The objective of this work was to determine the water adsorption isotherms of castor bean seeds (EVF103 and EVF106) to obtain information on the amount of water that this product adsorb at temperatures of 10, 20, 30, 40oC and water activities between 0.20 and 0.89, as well as adjusting different mathematical models to the experimental data. To obtain the hygroscopic equilibrium, the indirect static method was used, using Hygropalm Model Aw 1 equipment, analyzing castor bean seeds of the genotypes EVF103 and EVF106. The Copace model (for the EVF103) and Modified Oswin model (for the EVF106), according to the statistical parameters and Akaike's information criterion (AIC) and Schwarz's bayesian information criterion (BIC) information criteria, represent the hygroscopicity of castor bean seeds in the temperature range of 10 to 40oC. In a same water activity, the water content of the castor bean seeds of the genotypes EVF103 and EVF106 reduces with increasing temperature.

Keywords: Equilibrium moisture content; Adsorption isotherms; Mathematical modeling

Introduction

The castor bean *(Ricinus communis L.)* is a plant that produces inedible oil seeds with 40 to 60% oil [1], which are widely used in the chemical and bioenergy industries [2]. Its seeds contain high levels of ricin, ricinin and certain allergens, which are highly toxic to humans and animals, but contain unique composition of ricinoleic fatty acid, which confers exclusive properties for industrial production [3,4]. Oilseeds are more prone to deterioration during storage because the intensity and speed of the deterioration process are linked to their chemical composition [5]. Seed longevity also depends on water content, environmental conditions, packaging, microorganism activity, among other factors [6].

Like all hygroscopic material, the seeds yield or absorb water from the surrounding air as a function of the vapor pressure of the water, since when equilibrium between the air and seed vapor pressures the equilibrium water content is reached [7]. The use of mathematical equations to estimate the water content of hygroscopic equilibrium has the advantage of predicting values of water activity of the product under conditions of difficult experimental determination. The relationship between the water content of a product and the equilibrium relative humidity at a given temperature can be expressed by means of equilibrium water content characteristic curves called isotherms. Numerous models can predict an isotherm [8]. However, according Corrêa et al. [9] for the establishment of isotherms that represents this equilibrium relation, empirical mathematical models are used, since no theoretical model developed has been able to accurately predict the equilibrium water content for a wide range of temperature and relative humidity. Currently, in the literature, there are more than 200 equations proposed to represent the phenomenon of hygroscopic balance of agricultural products. These models differ in their theoretical or empirical basis and in the amount of parameters involved [10]. According to Ayranci & Duman [11], the sorption isotherms are important to define dehydration limits of the product, as well as to estimate the changes of water content under a certain condition of temperature and relative humidity of the ambient air. In addition, they allow to define the adequate water contents so that the beginning of the activity of microorganisms does not occur that can cause the deterioration of the product during the storage.

Therefore, knowing the behavior of the seeds during storage is essential to make decisions about their handling. Thus, the objective was to evaluate the hygroscopicity of castor bean seeds of two genotypes (EVF103 and EVF106), which present distinct agronomic characteristics of the genetic materials used in Brazil. EVF 103 has larger races and more, with greater productive potential. EVF 106 is smaller in size, more compact and less sensitive to gray mold (the main disease of the crop). These materials originate from research in partnership with Israeli company and are the target of selections in producing regions in the country. The adsorption isotherms were determine using the indirect static method of determination of water activity, which describes the relationship between the amount of water absorbed by the product and the relative humidity at a temperature [12].

Material and Methods

The present work was developed in the Post-Harvest Laboratory of Vegetable Products and in the Seed Laboratory of the Federal Institute of Education, Science and Technology Goiano - Rio Verde Campus, Rio Verde, GO. Seeds of two castor bean genotypes (EVF 103 and EVF106) were obtained from cultivation in fields located at Fazenda 2P of the company Sementes Goiás LTDA. Seeds were harvested mechanically (PLM 08L Platform, specific for castor bean harvest), which were pre-cleaned and initially had a moiusture content of 6.64 ± 0.55% dry basis (EVF103) and 7.44 ± 0.35% b.s. (EVF106).

The seeds of the two genotypes were submitted to forced ventilation drying, regulated to 40°C, to standardize the initial water content to 3.63% dry basis (b.s.). This procedure was performed using 2 kg of seeds of each genotype, divided into 4 trays containing 500g. Drying was accompanied by loss of mass by sequential weighing to the desired final moisture content. Subsequently, seed re-watering was carried out for water contents 5.26; 6.38; 7.53 and 8.70% dry basis by monitoring the mass gains of the respective samples. For each water content, 400 g of seeds were used, placed in perforated baskets to allow the passage of water vapor through the seed mass. The samples remained in BOD (Biochemical Oxygen Demand) at 25°C and 85 ± 5% until reaching the related mass for each moisture content.

During drying and rewetting the moisture content of the seeds was monitored by gravimetry using the oven method at 105 ± 3°C for 24h in two replicates of approximately 12g [13]. The adsorption isotherms of castor bean seeds were determined using the indirect static method and the water activity (aw) was determined using the Hygropalm Model Aw1 equipment. For each moisture content, three samples of approximately 30g were used, which were placed individually in the equipment container and conditioned in BOD regulated at 10, 20, 30 and 40°C. To the experimental data of the equilibrium moisture content were adjusted mathematical models frequently used for hygroscopicity representation of agricultural products, whose expressions are presented in Table 1. For the adjustment of the mathematical models, a non-linear regression analysis was performed by the Gauss Newton method. In order to verify the degree of adjustment of each model, we considered the significance of the regression coefficient by the t test, using the 1% level of significance, the magnitude of the coefficient of determination (R^2), the estimated mean error (SE), the chi-square test (X^2) at the significance level of 1% and the confidence interval of 99% (P <0.01).

$$P = \frac{100}{N} \sum \frac{[\gamma - \hat{Y}]}{\gamma} \tag{13}$$

$$SE = \sqrt{\frac{\sum(\gamma - \hat{Y})^2}{GLR}} \tag{14}$$

$$X^2 = \frac{\sum(\gamma - \hat{Y})^2}{GLR} \tag{15}$$

Where, Y: experimental value; $\hat{Y}$: value estimated by the model; N: number of experimental observations; GLR: degrees of freedom of the model (number of experimental observations minus the number of coefficients of the model).

Table 1: Mathematical models used to predict the hygroscopicity of plant products.

Model Name	Model	
Xe* = a-b. In [-(T+c). In (aw)]	Chung-Pfost (Pfost et al., 1976)	(1)
Xe = exp [a-(b.T) + (c.aw)]	Copace [41]	(2)
Xe = exp [a-(b.T) - In(aw)]1/c	Modified Halsey (Iglesias & Chirife, 1976)	(3)
Xe = a.(aw.b) / Tc	Sabbah (Oliveira et al., 2017; [28])	(4)
Xe = exp {a-(b.T) + [c. exp(aw)]}	Sigma Copace (Oliveira et al., 2017; [28])	(5)
Xe = [log (1-aw) / (a.(T.b))]1/c	Cavalcanti Mata (Oliveira et al., 2017; [28])	(6)
Xe = {ln (1-UR) – a.(T+b)]}1/c	Modified Henderson (Thompson et al., 1968)	(7)
Xe = [In (1-aw) / (-a × T+273,16)]1/c	Henderson (Oliveira et al., 2017; [28])	(8)
Xe = (a+b.T) / (1-aw) / aw]1/c	Modified Oswin (Chen & Morey, 1989)	(9)
Xe = (a.b.aw). [(c/T)/(1-b.aw+(c/T).b.aw).(1-b.aw)]	Modified GAB (Jayas & Mazza, 1993)	(10)

Note: *Xe=equilibrium moiusture content, % d.b.; aw=water activity, decimal; T=temperature (°C), UR=relative humidity and a, b, c=model coefficients.

The Akaike information criterion (AIC) and the Schwarz Bayesian information criterion (BIC) were used as a secondary criterion for choosing the best mathematical model to predict the phenomenon. The AIC allows us to use the principle of parsimony in choosing the best model, that is, according to this criterion, the most parameterized model is not always the best [14]. The AIC is used to compare non-nested models or when three or more models are being compared, and lower AIC values reflect better fit [15]. Its expression is given by:

$$AIC = -2\log like + 2p \tag{16}$$

Where, p is the number of parameters and loglike the value of the logarithm as a function of the probability considering the estimates of the parameters.

The BIC also considers the degree of parameterization of the model and, likewise, the lower the BIC value [16], the better the fit of the model. It is an asymptotic criterion whose adequacy is strongly related to the magnitude of the sample size. In relation to the sanction applied in the quantity of parameters, it will be more rigorous than the AIC for small samples. Its expression is given by:

$$BIC = -2\log like + p.\ln(n) \tag{17}$$

Where, n: number of observations used to adjust the curve, p is the number of parameters and loglike the value of the logarithm as a function of the probability considering the estimates of the parameters.

Results and Discussion

The coefficients of the models adjusted to the observed data of hygroscopic equilibrium of the castor bean seeds (EVF103 and EVF106) obtained by adsorption, for the different temperature and relative humidity conditions of the air, with their respective values of the coefficient of (R^2) of the standard deviation of the estimate (P) and the relative mean error (SE), together with the AIC and BIC information criteria (Tables 2 and 3). For the seeds of the EVF103 genotype, it was verified that, among the adjusted models, the Cavalcanti Mata, Henderson and Modified GAB showed a relative mean error (P) above 10%, which, according to Rosa, Moraes & Pinto [17] is an indication that this model is not adequate to represent the phenomenon studied. The models showed high values of the coefficient of determination (above 80%), except for Chung-Pfost, Cavalcanti Mata and Modified GAB. The Sigma Copace model presented the highest values of 80.84 and 86.67% for the genotypes EVF103 and EVF106, respectively. However, the coefficient of determination (R^2) and the relative mean error (P) are not sufficient to gauge the fit of a hygroscopic equilibrium model [18]. Thus, the estimated mean error (SE) is evaluated, verifying that the Sigma Copace model presented the lowest value. The ability of a model to adequately represent a given physical process is inversely proportional to the estimated mean error value [19].

Table 2: Parameters of the hygroscopic equilibrium models for castor bean seeds EVF103, with their respective mean (SE) and relative (P), chi-square (χ^2), determination coefficients (R^2) and AIC and BIC information criteria.

Models	Parameters	SE***	P (%)	χ^2***	AIC	BIC	R^2 (%)
Chung-Pfost	a = 39.1726**	0.786	9.99	0.6182	42.4622	45.5525	79.94
	b = 9.2031**						
	c = 32.4697**						
Copace	a = 0.0959ns	0.71	8.82	0.5039	42.3475	39.2571	83.65
	b = 0.02926**						
	c = 4.6576**						
Modified Halsey	a = 1.2470**	0.768	9.44	0.5896	41.7606	44.851	80.87
	b = 0.0192**						
	c = 0.6746**						
Sabbah	a = 171.1117ns	0.77	9.56	0.5936	47.6657	50.756	80.74
	b = 2.3664**						
	c = 0.5753**						
Sigma Copace	a = -1.9732ns	0.693	7.99	0.4799	41.7984	44.8798	80.84
	b = 0.0286**						
	c = 2.6495**						
Cavalcanti Mata	a = -0.0731**	0.829	10.33	0.6868	44.1577	47.248	77.72
	b = 0.3660**						
	c = 0.6539**						
Modified Henderson	a = 0.0043**	0.7	8.845	0.489	38.778	41.8684	84.13
	b = 33.5289**						
	c = 0.6096**						

Henderson	a = 0.0002ns	1.301	17.39	1.6938	41.3643	44.4547	40.83
	b = 1.4596**						
Modified Oswin	a = 9.3943**	0.758	9.17	0.5751	41.3643	44.4547	81.34
	b = -0.1448**						
	c = 0.9590**						
Modified GAB	a = 6.7227**	0.924	11.743	0.853	44.2759	47.3662	72.31
	b = 0.9449**						
	c = 18.7556ns						

Note: ** Significant to 0.01 probability by the t test; *** in decimal; ns not significant. Akaike Information Criterion (AIC) and the Bayesian Information Criterion (BIC).

Table 3: Parameters of hygroscopic equilibrium models for castor bean seeds EVF106, with their respective mean (SE) and relative (P), chi-square (χ^2), determination coefficients (R^2) and AIC and BIC information criteria.

Models	Parameters	SE***	P (%)	χ^2***	AIC	BIC	R^2 (%)
Chung-Pfost	a = 19.4561**	0.833	11.52	0.6947	35.4268	37.6866	85.37
	b = 2.9197**						
	c = 170.4701**						
Copace	a =0.8370**	0.681	9.36	0.4631	30.2334	32.4932	90.25
	b = 0.0023**						
	c = 1.6850**						
Halsey Modificado	a = 2.9327**	0.874	11.8	0.7644	37.0507	39.3105	83.9
	b = 0.0055**						
	c = 1.9687**						
Sabbah	a = 11.0889**	0.783	10.75	0.6128	38.105	40.3648	87.09
	b = 0.8767**						
	c = 0.0427**						
Sigma Copace	a = 0.0926ns	0.597	6.91	0.3561	34.4837	36.7435	86.67
	b = 0.0025**						
	c = 0.9546**						
Cavalcanti Mata	a = -0.0360**	0.847	11.96	0.7169	37.1146	39.3744	83.32
	b = 0.3968**						
	c = 1.0292**						
Henderson Modificado	a = 0.0002**	0.744	10.034	0.553	32.5166	34.7764	88.36
	b = 217.7906**						
	c = 1.7425**						
Henderson	a = 0.00013**	1.175	18.51	1.3805	43.6729	45.3678	67.7
	b =1.7426**						
Oswin Modificado	a = 5.3983**	0.657	9.2	0.4312	29.3866	31.6464	90.92
	b =-0.0134**						
	c =2.5823**						

Note: ** Significant to 0.01 probability by the t test; ns not significant; ***decimal; Akaike Information Criterion (AIC) and the Bayesian Information Criterion (BIC)

In relation to the chi-square test (χ^2), all models analyzed were within the 95% confidence interval and the Sigma Copace and Modified Henderson (EVF103), Sigma Copace, Copace and Modified Oswin (EVF106) models presented the lowest values. The smaller the chi-square, the better the fit of the model to the experimental data [20]. The Sigma Copace model presented the lowest values of SE, χ^2 and P and could be selected to represent the adsorption isotherms of the castor bean seeds of the genotypes EVF103 and EVF106 for the temperature range of 10 to 40°C. However, some of these parameters have caveats, requiring the adoption of additional criteria in the selection of models to reinforce and support decision making. In this context, the Akaike Information Criterion (AIC) and the Bayesian Information Criterion (BIC) consist of evaluating the models according to the parsimony principle, since the number of parameters in the model's changes [21]. The indication of the best model may be more concise, since these criteria consider other factors, such as the analysis of the degree of parameterization of the models compared [22].

The Modified Henderson model presented lower AIC and Copace lower BIC value, considering the seeds of the EVF103 genotype. However, BIC is an asymptotic criterion whose adequacy is strongly related to the magnitude of the sample size. Regarding the sanction applied in the quantity of parameters, BIC will be more rigorous than AIC for small samples [16]. Thus, the Copace model was chosen to represent the adsorption isotherms of the castor bean seeds of EVF103, according to the BIC criterion (Figure 1a). For the seeds of the EVF106 genotype, the Modified Oswin model showed the lowest AIC and BIC criteria, indicating the most appropriate model (Figure 1b). According to the results of Ferreira Júnior, Resende, Oliveira & Costa [23], the Modified Oswin model fitted better to the experimental values obtained by the static method to determine the moisture content of *Hymenaea stigonocarpa* Mart seeds. The modified Oswin model was also more suitable to describe the isotherms of yellow pepper *(Capsicum chinense L.)* (Ferreira, et al. 2011), among other species [24, 25].

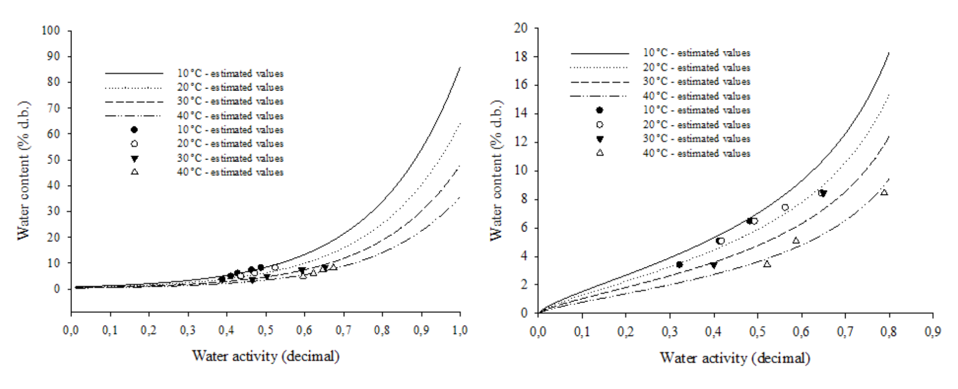

Figure 1: Water adsorption isotherms for castor bean seeds of the genotype EVF103 Estimated by the Copace model, and EVF106.
Estimated by the modified Oswin model, for different temperature and relative humidity conditions.

Experimental values of the equilibrium moisture of the castor bean seeds of the genotype EVF103 (A) and EVF106 (B) obtained by adsorption, and the values estimated by the Copace and Modified Oswin models, respectively, are presented in Figure 1. It is observed that the adsorption isotherms of castor bean seeds are classified as type III [26], as observed for the seeds of crambe [27], jatropha [28], forage turnip [29] and fruits of *Coffea canephora* [30]. With increasing temperature for the same water content, there was an increase in water activity and, for a constant water activity, the equilibrium moisture values decreased as the temperature increased, following the same trend of most agricultural products [8, 30-35].

The hygroscopic behavior represented by the isothermal curves is effective for the management of the environmental conditions of warehouses so that the water content of the product is kept at adequate levels that reduce the possibility of development of fungi and other pathogens and preserve the physical characteristics, chemical and physiological properties of the product during storage [12]. Santos et al. [36] studying the isotherms of hygroscopic equilibrium of jatropha seeds found that the Henderson model was the one that best represented the desorption isotherms of the product. However, Chaves et al. [28] observed that the Sigma Copace model presented higher coefficient of determination and lower values of χ^2, P and SE, being recommended for prediction of the phenomenon.

On the other hand, for the castor beans, Goneli [37] recommended the modified Halsey model to represent the equilibrium water content by the adsorption and desorption processes. In other oilseeds, such as colza, linseed and sunflower, Lazouk et al. [38] recommended the GAB model, considering the values of R^2 and P, which described the hygroscopic behavior of these species and presented a maximum equilibrium water content of 8.3% dry basis for water activity 0.7. Therefore, due to the isotherms obtained, castor bean seeds can be properly handled, as Chaves et al.

[28] point out that maintaining the water content at safe levels for storage, between 7.0 and 8.0% dry basis, controls the water activity and thus minimizes the processes that trigger the development of micro-organisms, insects as well as metabolic activities in the product. Also, according to Brooker et al. [39] the optimum range for storage of oil seeds is between 6.0 and 10.0% dry basis.

In this sense, the water contents recommended for safe storage (aw = 0.7) of castor bean seeds of the genotype EVF106 are at most 11.8; 9.6; 7.9; 6.4% dry basis, and for seeds of the EVF103 genotype are 19.5; 14.7; 11.0; 8.3% dry basis for the respective temperatures of 10, 20, 30 and 40°C. The water activity described by mathematical models contributes to predict phenomena involving the exchange of water vapor, and to maintain the quality of castor bean seeds. In addition, studies on isotherms help to understand the relationships between water content and relative humidity of post-harvest air at a given temperature, such as drying and storage [40].

Furthermore, the Akaike information criterion (AIC) and the Schwarz Bayesian information criterion (BIC) collaborated in the choice of the best mathematical model to estimate the adsorption isotherms of *Ricinus communis L.*, as well as Ferreira Júnior et al. [23] verified that this methodology can be included in the criterion of selection of models to estimate the sorption isotherms of vegetal products [41,42].

Conclusion

The Copace model (EVF103) and Modified Oswin model (EVF106), according to the statistical parameters and AIC and BIC information criteria, represent the hygroscopicity of castor bean seeds in the temperature range of 10 to 40°C. For a constant water activity, the moisture content of the castor bean seeds of the genotypes EVF103 and EVF106 reduces with increasing temperature. The adsorption isotherms of castor bean seeds (EVF103 and EVF106) exhibit curve type III.

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