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Sun drying of residual annatto seed powder

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ABSTRACT. Residual annatto seeds are waste from bixin extraction in the food, pharmaceutical and cosmetic industries. Most of this by-product is currently discarded; however, the use of these seeds in human foods through the elaboration of powder added to other commercial powders is seen as a viable option. This study aimed at drying of residual annatto powder, with and without the oil layer derived from the industrial extraction of bixin, fitting different mathematical models to experimental data and calculating the effective moisture diffusivity of the samples. Powder containing oil exhibited the shortest drying time, highest drying rate ($\approx$ 5.0 kg kg^{-1} min^{-1}) and highest effective diffusivity (6.49 $\times$ 10^{-12} m^2 s^{-1}). All mathematical models assessed were a suitable representation of the drying kinetics of powders with and without oil, with R^2 above 0.99 and root mean square error values lower than 1.0.

Keywords: *Bixa orellana* L., agricultural waste, dehydration, mathematical modeling.

Secagem ao sol de farinhas de grãos residuais de urucum

RESUMO. Os grãos residuais de urucum são o resíduo proveniente da extração da bixina nas indústrias alimentícia, farmacêutica e cosmética. Atualmente, a maioria desse subproduto tem sido descartada, no entanto vê-se a possibilidade de utilização desses grãos na alimentação humana através da elaboração de farinhas, com a finalidade de adicioná-las a outras farinhas de grãos comerciais. O objetivo do estudo foi secar por exposição ao sol farinhas de grãos residuais de urucum, com e sem a camada de óleo proveniente do processo de extração industrial da bixina, ajustar diferentes modelos matemáticos aos dados experimentais e calcular a difusividade de umidade efetiva das amostras. Verificou-se que a farinha com óleo apresentou o menor tempo de secagem, as maiores taxas de secagem ($\approx$ 5,0 kg kg^{-1} min^{-1}) e difusividade efetiva (6,49 $\times$ 10^{-12} m^2 s^{-1}). Todos os modelos matemáticos avaliados representaram de forma satisfatória a cinética de secagem das farinhas com óleo e sem óleo, com valores de R^2 superiores a 0,99 e valores de desvio quadrático médio inferiores a 1,0.

Palavras-chave: *Bixa orellana* L., resíduos agrícolas, desidratação, modelagem matemática.

Introduction

Annatto (*Bixa orellana* L.) is a plant native to South America, and belongs to the family Bixaceae. Its seeds have high commercial value due to the thin serosal layer around, from which bixin, a reddish-orange pigment with applications in the food, pharmaceutical and cosmetic industries, is extracted (SANTOS et al., 2013). Chuyen et al. (2012) report that bixin from annatto can be extracted by immersing seeds in hot vegetable oil.

Bixin oil extraction generates a large amount of by-products in the form of annatto seeds that are impregnated with both the oil used in extraction, and residual pigment. According to Silva et al. (2005), the bixin extraction process produces 97 to 98% of the by-product, of which approximately 97% is not reused (RÊGO et al., 2010).

A number of researchers have investigated the application of annatto seeds in animal feed (SILVA et al., 2005; RÊGO et al., 2010). However, it was observed that this by-product might have potential use in human foods, since, like all seeds, they are rich in proteins, carbohydrates and fibers (SANTOS et al., 2012; DIÓGENES et al., 2013). Jittanit (2011) recently reported that the use of food processing waste is the focus of researchers worldwide, due to increasing environmental concerns, stimulating the search for better recycling alternatives.

A feasible option for annatto seeds is the powder form, which can be incorporated to other seed powders traditionally consumed in breads and cookies, among others. Since the hygroscopic behavior of powders differs from that of whole seeds in their smaller grain size and fragmented tegument, they exhibit unique characteristics during the drying process.

Drying is a preservation method in which the moisture content and water activity of agricultural products are diminished by hot air in order to minimize chemical, biochemical and microbiological degradation (DOYMAZ, 2009). Drying can be in the sun or artificial, using solar driers or hot air (AKPINAR, 2011). In sun drying, the products are exposed to sunlight to produce heat. Part of this heat penetrates the product (provoking an increase in temperature and the formation of water vapor) and the rest is used to evaporate surface moisture. Moisture spreads from the inner portion to the surface to replace evaporated surface moisture. Part of the heat is lost to the atmosphere and the rest to the ground (PRASAD, 2009).

Sun drying is a simple, low-cost alternative for drying agricultural products, mainly for family farming. A large part of Brazil, especially the Northeast region, exhibits favorable environmental conditions for sun drying, since it lies in a tropical zone with high levels of sunlight, and little or no rainfall and low relative humidity at certain times of the year

In this respect, the aim of this study was to dry annatto seed powder in the sun, with and without the oil layer, resulting from bixin extraction, to fit different mathematical models to experimental data and to calculate effective moisture diffusivity of the samples.

Material and methods

Annatto seeds were donated by the Maratá food company, whose headquarters is located in Sergipe State, Brazil. Seeds were stored in plastic containers at -18°C until the onset of experiments. These seeds were impregnated with soybean oil and pigment residue, waste products from the bixin extraction.

For obtaining powders, annatto seeds were processed in two ways: in the first, seeds were preserved with the oil layer remaining from the bixin extraction, and in the second, the oil layer was removed by washings with water and neutral detergent. After washing, seeds were distributed in a thin layer on a laboratory bench until drying surface water.

Seeds with and without oil were ground in a knife mill, obtaining powders with (Treatment 1) and without (Treatment 2) oil. At the onset of drying, powder samples with and without oil had moisture content of about 20% w.b.

Drying experiments were performed with 4 repetitions, each one using around 15 g of sample, which were placed into aluminum containers with a diameter of 13.5 cm and 1.5 cm high.

Dryings were initiated at approximately 8 a.m and drying kinetics data were obtained by weighing samples at regular intervals of 15, 30 and 60 min. When samples reached final theoretical moisture content (5% w.b.), determined by the difference between initial mass and mass at a given weighing, moisture content was measured in an oven at 105°C for 24h, in order to calculate the final moisture content. Temperature and relative humidity were monitored during the sun drying process. Sunlight values were obtained from the National Meteorology Institute (INMET, 2011).

Equation 1 was used to determine the moisture ratio:

$$MR = \frac{X - X_e}{X_i - X_e} \qquad (1)$$

where:

MR = moisture ratio, dimensionless;

X = moisture content at any time t, % w.b.;

X_i = initial moisture content, % w.b.;

X_e = equilibrium moisture content, % w.b..

The mathematical models described in Table 1 were fitted to experimental data obtained in drying kinetics, using the Statistica 7.0 (STATSOFT, 2004) by a nonlinear regression method (Quasi-Newton).

Table 1. Mathematical models used to predict sun drying of annatto seed powders.

Model	Equation	
Diffusion Approach	$MR = a.\exp(-k.t) + (1 - a).\exp(-k.b.t)$	(2)
Two Term	$MR = a.\exp(-k.t) + b.\exp(-q.t)$	(3)
Midilli	$MR = a.\exp(-k.t^n) + b.t$	(4)
Page	$MR = \exp(-k.t^n)$	(5)
Thompson	$MR = \exp((-a - (a^2 + 4.b.t)^{0,5})/2.b)$	(6)

where:

t = drying time, min;

k = drying constant, min^{-1};

a, b, n, q = coefficients models.

The correlation coefficient (R^2) and root mean square error (RMSE) were used as criteria to assess if models fitted the experimental data well, in accordance with Equation 7:

$$RMSE = \left[\frac{1}{n}\sum_{i=1}^{n}(MR_{pred,i} - MR_{exp,i})^2\right]^{\frac{1}{2}} \qquad (7)$$

where:

RMSE = root mean square error, dimensionless;

$MR_{pred,i}$ = predicted moisture ratio, dimensionless;

$MR_{exp,i}$ = experimental moisture ratio, dimensionless;

n = number of observations.

Drying rates were calculated from the moisture content of samples and drying times, in accordance with Equation 8:

$$DR = \frac{X_{t+dt} - X_t}{dt} \tag{8}$$

where:

DR = drying rate, kg kg^{-1} min^{-1};

X_{t+dt} = moisture content at t +dt, kg water per kg dry matter;

X_t = moisture content at a specific time, kg water per kg dry matter;

t = drying time, min.

To determine the effective diffusivity, drying data of Treatments 1 and 2 were fitted to the mathematical model of liquid diffusion with approximation of three terms (Equation 9), considering the uniform initial water distribution and absence of thermal resistance. This equation is the analytical solution for Fick's second law, considering the geometric shape of the product as the approximate shape of a flat plate:

$$MR = \frac{X - X_e}{X_i - X_e} = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left[-(2n+1)^2 \pi^2 D \frac{t}{4L^2}\right] \tag{9}$$

where:

MR = moisture ratio, dimensionless;

D = effective moisture diffusivity, m^2 s^{-1};

n = number of terms;

L = characteristic dimension (half thickness of the plate);

t = time, s.

For statistical analysis of effective diffusivity values, a completely randomized experimental design was used, with two Treatments (samples with and without oil) and four repetitions. Data were subjected to analysis of variance (ANOVA) and Tukey's test ($p < 0.05$) was applied to compare means, using the 'Assistat' software.

Results and discussion

Figure 1 shows temperature, relative humidity and sunlight values observed during sun drying of annatto seed powder with (Treatment 1) and without (Treatment 2) oil. Mean temperatures of 27.55 and 26.82°C, mean relative humidity of 57.50 and 67.40% and mean solar radiation of 2832.80 and 2003.22 KJ m^{-2} were recorded for samples from Treatments 1 and 2, respectively. These data reveal better environmental conditions for drying samples from Treatment 1, given that higher drying temperatures result in a higher water removal rate due to the increased moisture gradient between the product and the air, as well as lower relative humidity values, which accelerate water transfer between the product and the air.

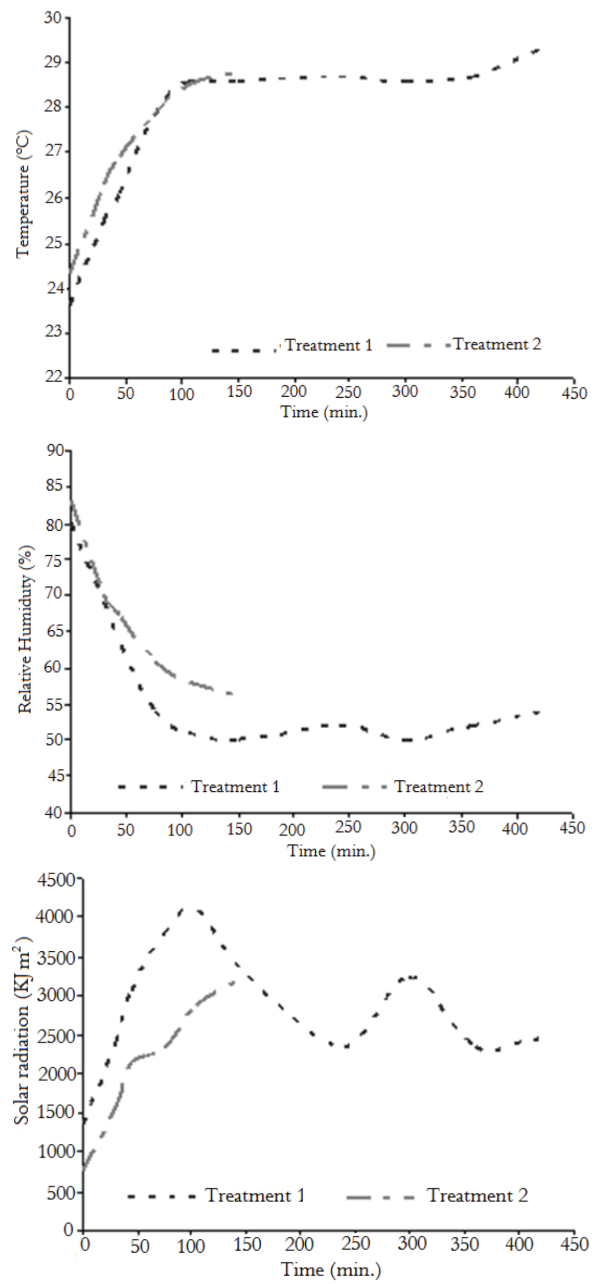

Figure 1. Variation of environmental conditions during sun drying of annatto seed powder samples.

Figure 2 shows moisture ratio (A) and moisture content (B) curves of samples from Treatments 1 and 2 subjected to sun drying. Treatment 2 samples required a shorter drying time to reach the desired moisture content (2.5h), while Treatment 1 samples required a nearly 3 times longer drying time (7.0h) to reach a moisture content of ≈5% w.b. Although better environmental conditions were observed for drying Treatment 1 samples, the oil layer likely formed a physical barrier between powder granules, hindering removal of moisture and justifying the prolonged drying time recorded for this treatment. Similar sun drying times were reported by Ronoh et al. (2009) in a study on drying amaranth seeds (6h), Akpinar (2006) on drying parsley, basil and mint (6.5h) and Al-Mahasneh et al. (2007), who dried sesame seeds in thin layers (≈ 3h).

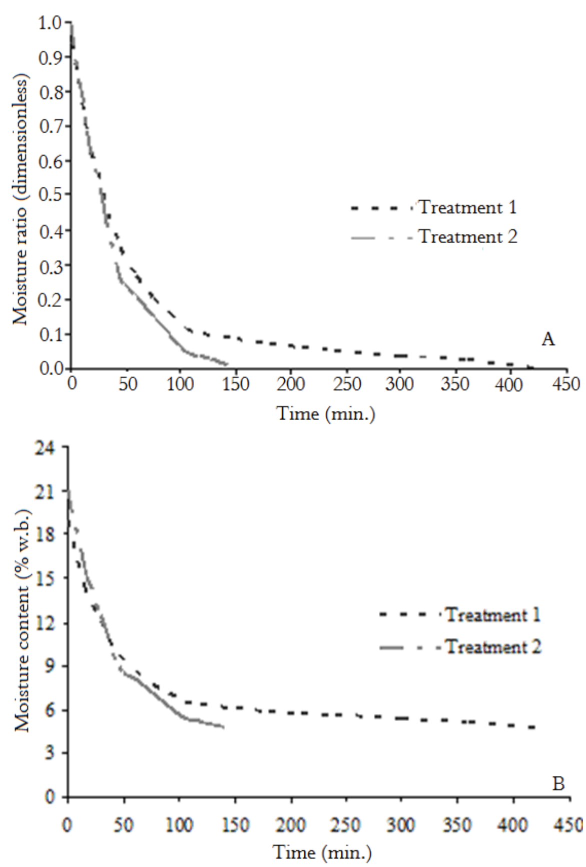

Figure 2. Sun drying curves of annatto seed powders with and without oil, expressed as moisture ratio (A) and moisture content (B).

Figure 3 illustrates the highest drying rates at the onset of the procedure, for both treatments, decreasing over time. There was no constant drying rate period on these curves, given that dehydration occurred in periods of falling drying speed. The absence of a constant drying rate period may be due to the thin layer of the product, which does not supply a constant moisture source during drying. Moreover, there may be resistance to moisture movement caused by surface shrinkage, which significantly reduces the drying speed (MEISAMI-ASL et al., 2010). Treatment 1 samples exhibited drying rates above 0.4 kg kg^{-1} min^{-1}, while Treatment 2 samples had rates of more than 0.5 kg kg^{-1} min^{-1}. The higher drying rate observed for Treatment 2 samples may be due to the absence of oil layer, since, according to Prasad (2009), drying rate depends on internal parameters, such as sample characteristics. Similar results were found by Nuthong et al. (2011), Ayim et al. (2012) and Hii et al. (2009) in studies on longan (*Dimocarpus longan* Lour.), banana and cocoa bean drying, respectively.

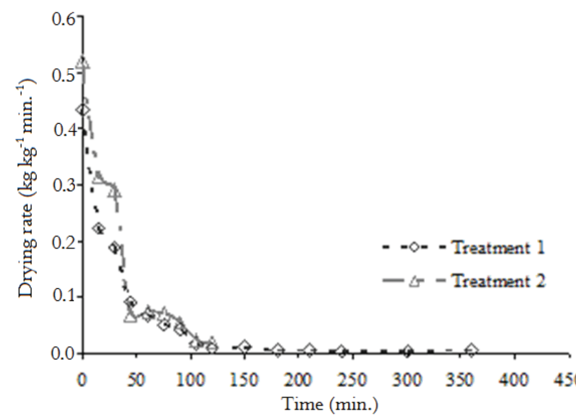

Figure 3. Sun drying rates of annatto seed powder samples.

Table 2 lists initial and final moisture content of annatto seed powder. Both treatments needed to lose more than 74% moisture to reach the final content of approximately 5% w.b. High water losses were also reported by Al-Mahasneh et al. (2007), Ronoh et al. (2009) and Doymaz (2011), in studies on sun drying of sesame seeds, amaranth seeds and green beans, reporting water loss of 94.12, 83.24 and 83.23%, respectively. Sun drying of Treatments 1 and 2 was generally enough to lower moisture content to a suitable level for storage and commercialization.

Table 2. Initial and final moisture content of samples of annatto seed powder.

Samples	Moisture content (% w.b.)		Water loss (%)
	Initial	Final	
Treatment 1	19.01	4.85	74.49
Treatment 2	21.03	4.60	78.13

Table 3 displays the parameters for Diffusion Approach, Two Terms, Midilli, Page and Thompson models fitted to the experimental points of sun drying kinetics of annatto seed powders, and their respective correlation coefficients (R^2) and root mean square error values (RMSE). The Two Terms model exhibited the best fit to the drying curves of Treatment 1 samples, while the Midilli model best fitted the drying data of Treatment 2 samples. Both models showed correlation coefficients (R^2) higher than 0.99 and the lowest RMSE values. The other mathematical models exhibited correlation coefficients (R^2) higher than 0.99 and low RMSE values, and can be also used to predict the drying kinetics of powders from Treatments 1 and 2.

Table 3. Parameters of mathematical models and respective correlation coefficients (R^2) and root mean square errors (RMSE) for the drying kinetics of annatto seed powders.

Model	Sample	Parameters						
		a	b	k	n	q	R^2	RMSE
Diffusion Approach	T1	0.8448	0.1791	0.0288	-	-	0.9991	0.0017
	T2	-0.0480	0.1674	0.1656	-	-	0.9971	0.0034
Two Terms	T1	0.1543	0.8445	0.0051	-	0.0287	0.9991	0.0014
	T2	0.5039	0.5039	0.0267	-	0.0268	0.9967	0.0076
Midilli	T1	1.0049	0.00007	0.0408	0.8464	-	0.9967	0.0053
	T2	1.0020	-0.00008	0.0245	1.0166	-	0.9972	0.0005
Page	T1	-	-	0.0465	0.8072	-	0.9947	0.0264
	T2	-	-	0.0226	1.0423	-	0.9970	0.0023
Thompson	T1	-7.6323	0.4722	-	-	-	0.9969	0.0150
	T2	-2632.1	8.3603	-	-	-	0.9967	0.0052

T1 - Treatment 1; T2 - Treatment 2.

These results corroborate those obtained by Doymaz (2011) for sun drying of green beans. The author attained R^2 values above 0.99 for the Two Terms and Diffusion Approach models; however, they were greater than 0.98 for the Page and Midilli models. Sobukola et al. (2007) found R^2 values above 0.99 for the Page and Midilli models, in a study of sun drying of *Vernonia anyadalina* and *Ocimum viride* leaves. Fits to Diffusion Approach models were greater than 0.97 for *V. anyadalina* leaves and higher than 0.95 for *O. viride* leaves. Akpinar (2006) also reported R^2 values above 0.99 when fitting Page, Two Terms and Diffusion Approach models to kinetic data related to sun drying of basil and parsley.

Figure 4 shows the comparison between experimental moisture ratio data and those estimated by the Two Terms model, for drying Treatment 1 samples, and those estimated by the Midilli model for drying Treatment 2 samples. Predictions using the Two Terms and Midilli models demonstrated experimental data close to the straight line that runs through the origin, which theoretically represents equality between experimental and estimated values, evidencing the suitability of models in representing sample drying.

Table 4 shows mean values of effective moisture diffusivity determined for annatto seed powders. There was a significant effect at 1% level of probability (F-test), with mean diffusivity values differing to each other according to Tukey's test ($p < 0.05$). Effective diffusivity values showed magnitudes of 10^{-12} m^2 s^{-1}, corroborating the results obtained by Gorjian et al. (2011) and Akbulut and Durmuş (2009), who also reported effective diffusivity of 10^{-11} m^2 s^{-1} for drying barberries (*Berberis vulgaris* L.) and mulberries, respectively. Treatment 2 samples exhibited the highest value of effective moisture diffusivity, possibly owing the highest drying rate observed during the experiments.

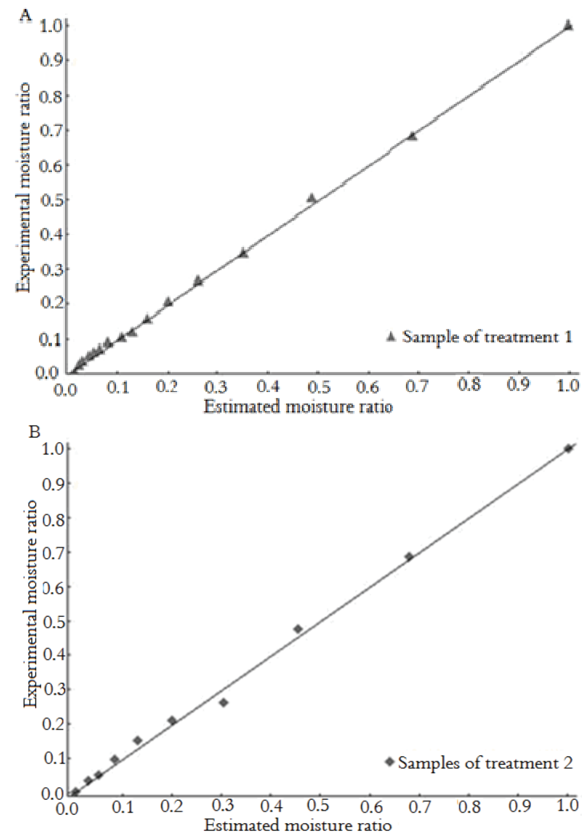

Figure 4. Relationship between experimental and estimated moisture ratio values according to the Two Terms (A) and Midilli (B) models for the drying kinetics of annatto seed powders with and without oil, respectively.

Table 4. Mean values of effective diffusivity obtained in sun drying of annatto seed powders.

Sample	Effective diffusivity (m^2 s^{-1})
Treatment 1	4.72×10^{-12} b
Treatment 2	6.49×10^{-12} a
F-test	415.87**

Means followed by the same letter are not significantly different by Tukey's test at 5% probability; **Significant at 1% probability (F-test).

Conclusion

Sun drying of annatto seed powder with and without oil was effective in lowering initial moisture content from approximately 20% w.b. to around 5% w.b., in addition to promoting short drying times for both samples. The highest drying rate and effective moisture diffusivity was found for the Treatment 2 (powder without oil). All the evaluated models have satisfactorily represented the drying kinetics of the powders, with R^2 values above 0.99 and RMSE values below 1.0.

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