Effective Moisture Diffusivity of Shrinking Particles Subjected to Infrared Drying

Camila F. Ferreira, Lidja D. M. Santos Borel, Luanda G. Marques, Manoel M. do Prado

Abstract— In order to contribute for a better understanding of the mass transfer in drying of shrinking particles, in this study shrinkage and drying characteristics of moringa seeds exposed to infrared (IR) radiation were experimentally determined. The IR source was located at a distance of 18 cm from the samples. Experiments were conducted with the seeds arranged in a single layer and exposed to three IR source temperature levels (51, 61 and 72°C). The surface temperature of the material was measured during drying through an optical pyrometer. The effects of the operating conditions on the drying kinetics and particle shrinkage were evaluated. Experimental results showed that increasing source temperature leads to an intensification of mass transfer rate, but restricts the volume contraction of the particles. Neglecting shrinkage of IR-dried moringa seeds led to an overestimation of the values of effective moisture diffusivity.

Index Terms— mass transfer, shrinkage, drying, infrared energy.

I. INTRODUCTION

Sustainable processes and products have been the focus of research and investments with growing interest in the current socioeconomic context. Within this context, seeds from *Moringa oleifera* L. have received special attention due to their various applications.

The moringa tree is a perennial plant, which has good development even when subjected to unfavorable weather conditions. It is fully usable. Its seeds are rich in proteins and lipids, having high nutritional value and pharmacological properties [1].

Moringa seed can be used as feedstock for biodiesel production due to high oxidative stability of its oil, which is related to the high content of oleic acid [2]. Its coagulant properties also make it an interesting alternative to replace the aluminum salts used in water treatment. The use of these salts has been related to various problems such as Alzheimer's disease and production of large volumes of sludge [3].

Moringa seeds are harvested with moisture content of approximately 70% wet basis (w.b.), which must be reduced by means a drying process to about 10% w.b. for a safe storage, in order to avoid the growth of microorganisms and the proliferation of fungus, to minimize enzymatic reactions of deterioration, losses of dry matter and germination as well as to inhibit the development of free fatty acids, which affects the quality of the extracted oil. Furthermore, drying allows reduction in seed weight and volume, minimizing the cost of packaging, storage and transportation.

Moringa seeds appear to be very attractive for infrared (IR) drying as these seeds usually contain high levels of protein and lipid, which have good absorption of infrared radiation [4]. However, there is still no information available in the literature regarding the use of IR drying for moringa seeds.

IR drying has several advantages over conventional drying, such as higher energy efficiency, higher drying rate and shorter drying time, thus reducing energy consumption without loss of the product quality. Other advantages are versatility, simplicity of equipment, easy incorporation to any existing dryer, and low capital cost.

In this drying technique heating elements like lamps or radiators produce rays/waves in the IR region that travel directly to the exposed material surface and penetrate it, being absorbed by some components of the product. The absorbed electromagnetic energy causes molecular vibration, thus generating internal heating [4], [5]. No energy is wasted directly heating the surrounding air. Moisture inside the material is rapidly heated and vaporized. Since heating is intense, the temperature gradient in the material reduces within a short period. Infrared radiation is also advantageous as the high drying rates suppress oxidation and prevents heat-sensitive substances being lost from the product during drying. In addition, energy saving in infrared drying process tends to be relatively higher.

Progress in the use of the IR drying technology for moringa seeds requires a deep knowledge of the mass transfer parameters like the effective moisture diffusivity, which is essential for proper design, simulation and optimization of the drying process by using mathematical modeling.

Effective moisture diffusivity describes all possible mechanisms of moisture movement within the solid, such as liquid diffusion, vapor diffusion, surface diffusion, capillary flow and hydrodynamic flow [6]. Several works have been carried out to determine the effective moisture diffusivity of different materials such as pomegranate arils [6], powdered biomass [7] and olive stone [8]. The heating mechanism and conditions within a material during IR drying are different from those during conventional drying. No information is available in literature on diffusion characteristics for IR drying of moringa seeds yet.

The IR drying of moringa seeds is a challenging assignment due to the complex relationship between the transport phenomena and the shrinkage characteristics. Due to their high moisture content, moringa seeds tend to undergo changes in their dimensions during drying, modifying the heat and mass exchange area, thus affecting the drying rate. The complexity increases as the extent of shrinkage is also process dependent. That is the result of the moisture gradient in the product, which, in turn, induces stresses and, thus, mechanical deformation [9].

Thus, any attempt to accurately interpreting the mass transfer phenomenon during drying of shrinking particles like

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moringa seeds requires an experimental investigation to obtain reliable values of effective moisture diffusivity (D_{eff}), which should take into account the reduction in the distance required for the movement of water molecules. However, few works have been carried out on the effects of the shrinkage phenomenon on D_{eff} , even less on D_{eff} of moringa seeds.

The objectives of this work were therefore to study the thin-layer drying of *moringa oleifera* seeds using IR radiation, to determine their shrinkage characteristics, and to evaluate how the values of moisture diffusivity are affected by the volume contraction of the particle at different operating conditions.

II. MATERIALS AND METHODS

A. Materials

Moringa oleifera pods were collected from trees in open squares (-10.965078 to -10.944600 W; -37.075373 to -37.081825 S) in the city of Aracaju, Sergipe, Brazil.

Moringa seeds were removed from the green pods and a manual separation of damaged seeds was performed. Then the wings were removed from each remaining unshelled seed. The particle size distribution was determined, in order to identify the predominant seed size range to be used in the drying experiments, thus avoiding the effect of seed size variability on the studied processes. Medium size particles, with average diameter ranging from 11 mm to 12.5 mm, were found to represent the highest fraction in the seed samples.

The moisture content was determined by the oven method at 105°C for 24 hours [10]. The initial moisture content of moringa seeds used in the drying tests was (0.70 ± 0.03) w.b.

B. Infrared drying experiments

The experimental set-up used to investigate the single layer drying of moringa seeds using infrared radiation is shown schematically in Fig. 1. It consists of a drying chamber equipped with a 250 W incandescent lamp. The drying chamber of 200 x 200 x 300 mm was made from wood sheet of 10 mm thickness. The chamber walls were isolated and covered with an aluminum foil.

The temperature of the IR source was regulated using a temperature controller The distance between the heating source and the single layer of particles was set at 18 cm. Experiments were conducted with IR source temperatures of 51, 61 and 72°C, conditions under which the product has reached final temperatures of 40, 50 and 60°C, respectively.



Fig. 1: Schematic diagram of the IR dryer.

Two experiments groups were carried out. The aim of the first group was to determine the drying kinetics of the moringa seeds in terms of the moisture content and temperature evolutions at different IR radiation intensities. The second group had as objective to identify and quantify the shrinkage of the particles as a function of moisture content under the different IR drying conditions. Each experiment and subsequent analyses were performed in triplicate.

The drying operation consisted of initially regular the IR heating system in order to reach the desired operating temperature. After the equipment attaining steady state with respect to the pre-set operating condition, an aluminum tray (130 mm diameter) containing 50 seeds uniformly arranged in a single layer was placed in the dryer, starting the drying process.

To determine the drying kinetics curves the seed samples were withdrawn from the equipment at regular intervals along the total drying time and weighted on a digital balance (Sartorius, CP224S, Germany) with 10^{-4} g accuracy. All weighting processes were completed in 15 s during the drying process. Moisture content at a given time was calculated by the ratio between the mass of water retained in the material and the mass of dry solid, which was determined at the end of each experiment by the oven method at $(105 \pm 3)^{\circ}$ C for 24 hours [10]. The surface temperature of the particles was measured at different drying times by using an optical pyrometer (Impac, IP-850).

In the drying tests to quantify the particle shrinkage seeds samples were subjected to IR radiation for different periods to achieve the desired moisture content, with previously drying kinetics curves facilitating determination of the drying time. On attaining the predetermined drying time, three seeds were taken from the dryer and their dimensions were measured using a digital micrometer (Pantec, Model 13101-25, accuracy of 10^{-3} mm).

C. Analysis of shrinkage in infrared-dried seeds

The shrinkage of the moringa seeds exposed to IR radiation was quantified from the changes in volume of individual particles, which were calculated from the dimensions taken by the digital micrometer, assuming that moringa seeds are oblate spheroids. The particle volume (V_p) was then calculated using the following equation:

$$V_p = \frac{\pi}{6} D_1^2 D_2 \tag{1}$$

where D_1 is the largest major diameter, D_2 is the smallest diameter of the oblate ellipsoid.

Reduced dimensional changes in volume (V_p/V_{p0}) with respect to the moisture content constituted the so-called shrinkage curves. It must be pointed out that the shrinkage analysis, based on volume changes from the geometric measurements with the aid of a digital micrometer, was sufficiently accurate since moringa seeds keep its original shape throughout the whole drying process.

D. Analysis of mass transfer in infrared drying

The experimental data sets from the different drying tests were initially expressed as moisture content (X) versus time (t). The moisture contents values were also converted into the dimensionless moisture ratios (XR), defined as the ratio of the free water still to be removed at any time to the total free water initially available, and expressed as:

$$XR = \frac{\overline{X} - X_e}{X_0 - X_e} \tag{2}$$

where \overline{X} is the average moisture content at a given time, X_0 and X_e are the initial and equilibrium moisture content of the seeds, respectively. The value of X_e in (2) was set to zero since prolonged exposure of material to infrared radiation will eventually cause a complete moisture removal [11].

The drying rate per mass unit of dry solid (-dX/dt) was also determined from the drying curves by numerical derivation.

The mass transfer in IR drying of moringa seeds was analyzed in terms of effective diffusivity, which was determined at each IR source temperature by applying diffusion model to describe the drying kinetics under the consideration of both negligible and significant shrinkage.

Non-steady mass transfer equation based on Fick's second law for the diffusion of water during drying is expressed as:

$$\frac{\partial \left(\rho_{d} X\right)}{\partial t} = \nabla \cdot \left[D_{eff} \nabla \left(\rho_{d} X\right)\right] \tag{3}$$

where D_{eff} is the effective moisture diffusivity, ρ_d is the local concentration of dry solid (kg dry solid per volume of the moist material) that varies with moisture content, because of shrinkage and X is the local moisture content (dry basis).

Assuming one-dimensional moisture transfer with negligible particle shrinkage and considering that the process is isothermal, initial moisture distribution is uniform, effective water diffusivity into material is constant, and equilibrium conditions at the particle surface prevail, the analytical solution for spherical geometry is given as [12]:

$$XR = \frac{\overline{X} - X_e}{X_0 - X_e} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(-\frac{n^2 \pi^2 D_{eff}}{R_0^2} t\right)$$
(4)

where R_0 is the particle radius and D_{eff} is the effective moisture diffusivity without considering shrinkage.

In order to include the shrinkage effects, substituting the density of dry solid ($\rho_d = m_d/V_p$), Equation (3) for constant mass of dry solid could be expressed as [13], [14]:

$$\frac{\partial \left(X/V_{p}\right)}{\partial t} = \nabla \cdot \left[D_{eff} \nabla \left(X/V_{p}\right)\right]$$
(5)

where V_p is the particle volume. Substituting $Y = X/V_p$, the following equation is obtained:

$$\frac{\partial Y}{\partial t} = \nabla \cdot \left(D_{eff}^* \nabla Y \right) \tag{6}$$

For one-dimension moisture transfer in conjunction with the following initial and boundary conditions:

$$t = 0, \ Y = X_0 / V_{p0} \tag{7}$$

$$t > 0, \ z = 0, \ \frac{\partial Y}{\partial z} = 0$$
 (8)

$$t > 0, \ z = L, \ Y = X_e / V_{p,e}$$
 (9)

a solution similar to (4) is obtained:

$$YR = \frac{\overline{Y} - Y_{e}}{Y_{0} - Y_{e}} = \frac{\frac{X}{V_{p}} - \frac{X_{e}}{V_{p,e}}}{\frac{X_{0}}{V_{p,0}} - \frac{X_{e}}{V_{p,e}}} = \frac{6}{\pi^{2}} \sum_{n=1}^{\infty} \frac{1}{n^{2}} \exp\left(-\frac{n^{2}\pi^{2}D_{eff}^{*}}{R^{2}}t\right)$$
(10)

where V_p is the sample volume at a given time, V_{p0} is the initial volume and V_{pe} is the sample volume on attaining the equilibrium moisture, D_{eff}^{*} is the effective moisture diffusivity of shrinking particle and R is the time averaged radius during drying.

Equations (4) and (10) are valid only for describing the falling rate period when the drying process is controlled by internal moisture diffusion and moisture content is below the critical value. Therefore, the water diffusivity into moringa seeds undergoing IR drying was estimated from these equations by setting the initial moisture content to the critical value X_{cr} and by setting the drying time to zero when the mean moisture content of the sample reaches that critical moisture content. Equations (4) and (10) were then fitted to the experimental data of XR and YR, respectively. Deff and D_{eff}^{*}-values were estimated by applying a non-linear regression procedure using STATISTICA® software. The estimation method was based on the well-established Levenberg-Marquardt algorithm. The statistical criterion used to evaluate the goodness-of-fit of the aforementioned equations was the medium relative deviation (MRD).

III. RESULTS AND DISCUSSION

Typical results on the IR drying kinetics in terms of the time evolution of the average moisture content and surface particle temperature are presented in Fig. 2 and 3, respectively.

As expected, an increase in infrared heat flux produced a steeper drying curve resulting in a fast moisture removal. A similar trend has been reported in the literature for the IR drying of fruits, vegetables and grains [15], [16]. This can be attributed to a significant increase in particle temperature, as shown in Fig. 3, caused by a higher intensity of radiant energy applied, resulting into an increase in the water vapor pressure inside the product and thus in higher drying rates [15].



Fig. 2: Dimensionless moisture content as a function of time during IR drying at different temperatures.



Fig. 3: Surface seed temperature as a function of time during IR drying at different temperatures.

The surface temperature of moringa seeds was found to increase rapidly during IR drying, reaching steady state values at only few minutes. The process can therefore be considered as isothermal since heat transfer was faster than mass transfer. This means that all or most supplied energy is used as latent heat for evaporation of water contained in the seed.

Figure 4 shows typical results of drying rate versus dimensionless moisture content at different operating conditions. It can be noted that IR drying of moringa seeds took place predominantly in the falling rate period. The process was therefore controlled by mass transfer internal resistances. This means that moisture diffusion inside the solid was the main physical mechanism governing mass transfer in IR drying of moringa seeds.



Fig. 4: Drying rate as a function of dimensionless moisture content during IR drying at different temperatures.

Experimental shrinkage data of moringa seeds, expressed by V_p/V_{p0} ratios, were plotted against the dimensionless moisture content at different IR radiation source temperatures and are presented in Fig. 5.

Particle shrinkage was found to increase with decrease in temperature of IR source. For example, after removal of 50% of free moisture, that is, at a XR-value of 0.50, the reductions in particle volume were nearly 49%, 23% and 11%, at 51, 61

and 72°C, respectively.

The significant influence of IR source temperature on the volume contraction of the seeds can be explained by the relationship between the drying conditions and the mass transfer resistances. Exposing the particles under low drying temperatures results in low drying rates, so that external resistance controls the mass transfer, moisture profiles into the particles are uniform and stresses are minimum. As result, particle shrinks uniformly and magnitude of shrinkage is higher [17].

On the other hand, at severe drying conditions, internal resistances control the moisture transfer, which is not uniform inside the particle and does not compensates the evaporation rate from the particle surface and dried regions appear on it. As soon as the surface dries up a rubber–glass transition may occur, producing a porous outer rigid crust or shell [18], thus hindering the shrinkage phenomenon.

These results stress the need to take into account the shrinkage in the drying models, in order to obtain a more realistic description of the drying process of moringa seeds.

In what follows, the analysis of the decreasing rate period based on the application of the diffusion model is presented, aiming to evaluate the influence of particle shrinkage on the estimation of effective moisture diffusivity. Diffusion models without and with shrinkage, Equations 4 and 10, were then used to estimate the effective diffusion coefficient of moringa seeds subjected to IR-drying. Table 1 shows the results obtained for the three IR source temperatures investigated in this study.

It can be verified that the values of diffusivity calculated without consideration of the shrinkage were higher than those obtained taking into account the phenomenon. For example, at 61°C the effective diffusivity of moringa seeds without considering the shrinkage ($D_{eff} = 2.18 \times 10^{-12} \text{ m}^2/\text{s}$) was found to be 202% higher than that estimated taking into account the phenomenon ($D_{eff}^* = 0.72 \times 10^{-12} \text{ m}^2/\text{s}$).



Fig. 5: Dimensionless volume change (V_p/V_{p0}) of the particles as a function of dimensionless moisture content (XR) during IR drying at different temperatures.

Table 1: Values of effective moisture diffusivity estimated without and with consideration of shrinkage during IR drying of moringa seeds.

T _{IR} (°C) –	Without shrinkage		With shrinkage	
	$D_{eff} \ x \ 10^{12} (m^2\!/s)$	MRD (%)	${D_{eff}}^* \ge 10^{12} (m^2/s)$	MRD (%)
51	1.48	4.79	0.28	3.95
61	2.18	6.91	0.72	8.52
72	2.68	6.78	1.77	7.90

Neglecting shrinkage of the particles during IR drying leads, therefore, to an overestimation of the mass transfer by diffusion. The explanation is on the fact that particle shrinkage produces a variation in the distance required for the movement of water molecules, therefore making the effective diffusivity be overestimated when obtained from analytical solution of diffusion model without considering volume contraction. This finding agrees with results of previous reports on other products [13], [14].

Since the effective moisture diffusivity measures the ease with which the moisture inside the particle diffuses and reaches the surface, the increase in the vapor pressure caused by an increase of the drying temperature led to larger values of effective diffusivity.

IV. CONCLUSION

Moringa seeds subjected to drying using infrared radiation had significant volume changes during moisture removal.

Volume contraction of the particles was found to be affected by IR source temperature, with an increase in temperature reducing particle shrinkage.

The IR drying of moringa seeds took place predominantly in the falling rate period and the mass transfer was limited by diffusion inside the material.

Neglecting the shrinkage of the particles led to a significant overestimation of the effective moisture diffusivity and thus of the energy required for drying the material.

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REFERENCES

- [1] A. Pandey, R. D. Pandey, P. Tripathi, P. P. Gupta, J. Haider, S. BHATT, and A. V. Singh "Moringa Oleifera Lam. (Sahijan) A Plant with a Plethora of Diverse Therapeutic Benefits: An Updated Retrospection", Medicinal Aromatic Plants. vol. 1. 2012.
- [2] R. Ayerza, "Seed yield components, oil content, and fatty acid composition of two cultivars of moringa (*Moringa oleifera* Lam.) growing in the Arid Chaco of Argentina", Industrial Crops and Products. vol. 33, pp. 389–394, 2011.
- [3] J. Beltrán-Heredia, J. Sánchez-Martína, A. Muñoz-Serrano, and J. A. Peres, "Towards overcoming TOC increase in wastewater treated with Moringa oleifera seed extract", Chemical Engineering Journal. vol. 188, pp. 40–46, 2012.
- [4] C. Sandu, "Infrared radiative drying in food engineering: A process analysis", Biotechnology Progress, vol. 2, pp. 109-119, 1986.
- [5] R. Khir, Z. Pan, A. Salim, B. R. Hartsough, and S. Mohamed, "Moisture diffusivity of rough rice under infrared radiation drying", LWT - Food Science and Technology, vol. 44, pp. 1126-1132, 2011.
- [6] Manish Dak, and N.K. Pareek, "Effective moisture diffusivity of pomegranate arils under going microwave-vacuum drying", Journal of Food Engineering, vol. 122, pp. 117-121, 2014.
- [7] Dengyu Chen, Xu Liu, and Xifeng Zhu, "A one-step non-isothermal method for the determination of effective moisture diffusivity in powdered biomass", Biomass and Bioenergy, vol. 50, pp.81-86, 2013.
- [8] Francisco J. Gómez-de la Cruz, José M. Palomar-Carnicero, Pedro J. Casanova-Peláez, and Fernando Cruz-Peragón, "Experimental determination of effective moisture diffusivity during the drying of clean olive stone: Dependence of temperature, moisture content and sample thickness", Fuel Processing and Technology, vol. 1375, pp. 320-326, 2015.
- [9] S. Eichler, O. Ramon, and I. Ladyzhinski, "Collapse processes in shrinkage of hydrophilic gels during dehydration", Food Research International, vol. 30, No. 9, pp. 719 – 726, 1997.
- [10] Brazil (2009). Rules for Seed analysis. (1st ed.). Brasília: Ministry of Agriculture, Livestock andSupply. (in Portuguese)

- [11] O. Fasina, B. Tyler, M. Pickard, G. Zheng, and N. Wang, "Effect of infrared heating on the properties of legume seeds", International Journal of Food Science and Technology, Vol. 36, pp. 79-90, 2001.
- [12] J. Crank, "The mathematics of diffusion", 2nd Edition, Clarendon Press, Oxford, London, 1975.
- [13] G. Hashemi, D. Mowla, and M. Kazemeini, "Moisture diffusivity and shrinkage of broad beans during bulk drying in an inert medium fluidized bed dryer assisted by dielectric heating", Journal of Food Engineering, vol. 92, No. 3, pp. 331-338, 2009.
- [14] A. M. Barbosa Neto, L. G. Marques, M. M. Prado; and D. J. M. Sartori, "Mass Transfer in Infrared Drying of Gel-Coated Seeds", Advances in Chemical Engineering and Science, vol. 4, pp. 39-48, 2014.
- [15] P. B. Pathare, and G. P. Sharma, "Effective moisture diffusivity of onion slices undergoing infrared convective drying", Biosystems Engineering, vol. 93(3), pp. 285–291, 2006
- [16] R. J. Brandão, L. D. M. Santos, L. G. Marques, and M. M. Prado, "Heat and mass transfer and energy aspects in combined infrared-convective drying of bee-pollen", Defect and Diffusion Forum, vol. 364, pp. 9-17.
- [17] C. Ratti, "Shrinkage during drying of foodstuffs", Journal of Food Engineering, vol. 23, pp. 91-105, 1994.
- [18] L. Mayor, and A. M. Sereno, "Modeling shrinkage during convective drying of food materials: a review", Journal of Food Engineering, vol. 61, pp.373-386, 2004.

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