

Effect of Shape of a Fruit Model-Product (Sodium Alginate Gel) Dried by Microwave and Modeling of Drying Kinetics

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ABSTRACT— *Model-products are products designed to imitate and understand the heat treatment behavior of real products. This is the case of a sodium alginate gel used to simulate the drying behavior of water-rich fruit products (e.g. tomatoes). The aim of this study was to investigate the effect of the shape of a sodium alginate gel as a model-product of microwave drying. Then, from this shape of the gel, find a semi-empirical model that simulated well its drying kinetics. Thus, a sodium alginate gel was developed in cylindrical and slab shapes, which had almost identical volumes and weights, to assess the influence of product shape on drying kinetics. Experiments were performed in a stereo-mode cavity at 2450 MHz with output microwave powers densities of 1, 1.5, and 2 W / g. A regular weighing of dried product mass was carried out to better appreciate drying behavior. In addition, the temperature at the product core was measured by an optical fiber to observe the evolution of gel temperature. The experiments showed that the cylindrical shape of the model-product dries faster than the slab shape. Moreover, the Midilli mathematical model best fit the drying kinetics of the cylindrical shape of the gel.*

Keywords— Microwave, model-product, alginate gel, modeling of drying

Nomenclature

a, b, c, n	empirical constants in drying models
D_{eff}	effective water diffusivity, (m ² /s)
k, k_0, k_1	empirical constants in drying models
L	slice half-thickness (mm)
X_0	initial moisture content (kg water/kg of dry matter)
X_e	equilibrium moisture content (kg water/kg of dry matter)
$Xr_{exp,i}$	i ith experimental moisture ratio (dimensionless)
$Xr_{pre,i}$	i ith predicted moisture ratio (dimensionless)
N	number of observations
n_p	number of constants
Ps	Specific power (W/g)
R^2	coefficient of determination
$RMSE$	root mean square error
t	drying time (h)
Xt	moisture content at any time (kg water/kg of dry matter)
Xr	moisture ratio (dimensionless)
χ^2	chi-square

1. INTRODUCTION

Throughout history, drying has been one of the processes most commonly used to improve the stability of foods by reducing the water activity of a product, reducing the microbiological activity, and minimizing the physical and chemical changes occurring during storage [1]. However, the behavior of agricultural products during microwave drying is very complex, very hard to accurately predict, and varies from one product to another [2], especially due to the heterogeneity of the products. The mathematical models that are needed for the analysis, design, simulation and control of the drying process should describe the coupled transfer of mass and heat. This is not simple, especially for real agricultural produce. To study the role of a new drying mode, using a more stable model-product is required. There are several kinds of model-products. In this study, a sodium alginate gel, which does not exhibit the variability of agricultural goods but is rich in water like them, is used as a model-product.

Alginates are polysaccharides (polymers constituted of -oses) that are obtained from a family of brown algae and have a molecular formula of $C_6H_8O_6$. Sodium alginate is a polyelectrolyte that consists of β (1–4) linked D-mannuronic acid units and some L-guluronic acid units [3]. Sodium alginate is a food additive (E401) used to increase viscosity and to stabilize emulsions. It is a white powder that is tasteless, odorless, and is very soluble in water. Sodium alginate overdose may have a laxative effect. As model-products, sodium alginate gels are porous materials that are mostly studied in the field of drying. Various techniques of model-products have advanced considerably in recent years [2]. Thus, in addition to traditional convective drying, other drying methods such as high frequencies or microwaves were added [4]. Microwave drying can be regarded as a rapid dehydration process. During the drying process, moisture content was reduced and the loss factor of the dried materials decreased. Also, the local pressure and temperature could be increased to speed up the drying process [5]. It was reported that increasing the microwave power increased the dehydration rate of carrots [6].

The objective of this study is firstly to characterize the combined microwave-air drying of a sodium alginate gel as a model-product, particularly with respect to the effect of its geometrical shape on drying kinetics, and secondly to model these kinetics.

2. MATERIAL AND METHODS

2.1. Preparation of samples

Alginate gel was obtained by combining the constituents listed in Table 1. Sodium alginate powder was slowly sprinkled into 70 mL of 6.9 pH phosphate buffer (K_2HPO_4 , KH_2PO_4). After shaking continuously for 30 min to achieve complete dissolution, the solution was cooled in ice water at $0^\circ C$. 30 mL of calcium pyrophosphate ($Na_4P_2O_7$, $10H_2O$ and $CaSO_4$, $2H_2O$) was added, and the mixture was blended until it became a homogeneous suspension, which was poured into cylindrical (radius 25mm, height 11 mm) and slab (43 x 42 x 11 mm) moulds. 7.3 g of dry reactants and 100 mL of distilled water were used for gel production.

Table 1: Dry constituents used in the manufacture of the sodium alginate gel

Constituent	Weight (g)
Sodium alginate (SIGMA A2033-100G)	3
Dipotassium Hydrogenophosphate (K_2HPO_4)	0.1
Potassium phosphates (KH_2PO_4)	0.1
Tetra Sodium Pyrophosphate decahydrate ($Na_4P_2O_7$, $10H_2O$)	1.7
Calcium Sulphate dehydrate ($CaSO_4$, $2H_2O$)	2.4
Total dry product (DM)	7.3

2.2. Drying procedure

The study of gel drying kinetics was performed in a laboratory-pilot microwave dryer (Figure 1) (designed by the ex-Company MES Technologies), which has a stereo-mode cavity of 290 mm high, 215 mm wide, and 215 mm of deep (or long), at variable power. The maximum power emitted by the microwave generator is 1859 W at 2450 MHz frequency. In this cavity, a wave-guide emerges in front of which gels were placed with their smaller dimension, 11 mm thick, perpendicular to the wave-guide axis. A digital scale (Denver Instrument DI-12K) with ± 0.1 g of precision allowed for the measurement of the mass lost during the drying process. Two optic fibers (OPTOPRIM, France) allowed for the recording of the temperature in center of both shapes of the gel.

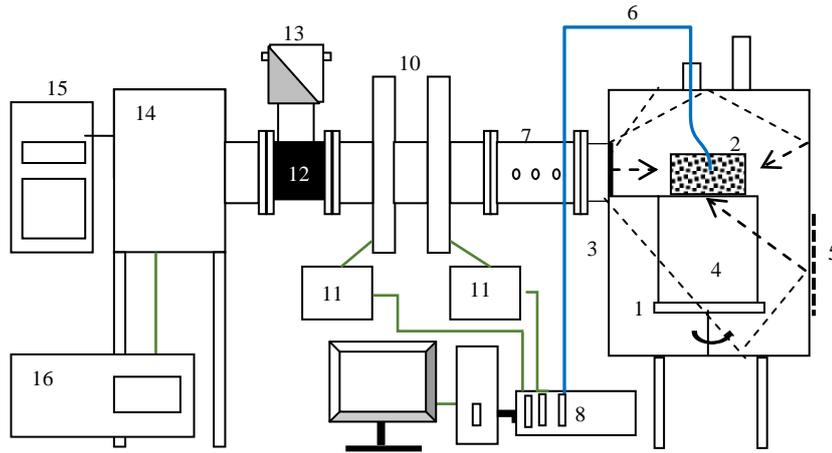


Figure 1: Schematic illustration of microwave drying with the stereo-mode cavity.

1- Rotatable plate, 2- Gel sample, 3- Waves inlet, 4- Aluminum Support, 5- Air outlet, 6- Optic fiber, 7- Waveguide, 8- Data acquisition center, 9- PC, 10- Directional couplers, 11- Watt-meters for measurement of applied and reflected powers, 12- Wave circulator, 13- Water trap, 14- Magnetron, 15- Electronic box, 16- electric generator.

Three of the four specific microwave power outputs (1, 1.5, 2 W/g) were used to study drying kinetics and the effective moisture diffusivity of both shapes of the gel (cylindrical and slab). Both shapes of the gel, each with a weight of 24.5 ± 1 g and a moisture content of 93.3% (wet basis), were dried at the same time by exchanging their position during each weighing. Each experiment was repeated three times. Recording the mass loss was done every three minutes at the beginning of the drying, then every six to eight minutes at the end of drying, until a final moisture of 7% was obtained (stable product) [7].

2.3. Modeling of drying kinetics and data analysis

The experimental moisture content data of the model-product (alginate gel) during microwave drying were converted to non-dimensionless moisture ratio (MR) using equation 1:

$$Xr = \frac{X_t - X_e}{X_0 - X_e} \quad (1)$$

where X_0 is the initial moisture content (g water per g dry matter), X_t is the moisture content (g water per g dry matter) at time t and X_e is the equilibrium moisture content (g water per g dry matter) [8-10]. However, assuming that at equilibrium (over a long period) X_e is negligible when compared to X_0 and X_t [11-12], the above equation can be further simplified to:

$$Xr = \frac{X_t}{X_0} \quad (2)$$

To describe the drying curves of both shapes of sodium alginate gel and to determine the best empirical equation, twelve thin layer drying models from literature (Table 2) were implemented. The following statistical parameters were used to evaluate the correlation between experimental results and the corresponding models:

- the determination coefficient (R^2) was one of the first criteria used to predict the best equation to reflect the drying curves.

$$R^2 = \frac{\sum_{i=1}^n (Xr_{exp i} - \bar{X}r_{exp i})^2 - (Xr_{pre,i} - Xr_{exp i})^2}{\sum_{i=1}^n (Xr_{exp i} - \bar{X}r_{exp i})^2} \quad (3)$$

Table 2: Drying models applied to the description of sodium alginate gel drying curves

N°	Model name	Model equation	Reference
1	Newton-Lewis	$Xr = \exp(-kt)$	[13]
2	Page	$Xr = \exp(-kt^n)$	[14]
3	Henderson and Pabis	$Xr = a.\exp(-kt)$	[15]
4	Logarithmic	$Xr = a.\exp(-kt) + c$	[16]
5	Wang and Singh	$Xr = 1 + at + bt^2$	[17]
6	Verma	$Xr = a.\exp(-kt) + (1-a).\exp(-gt)$	[18]
7	Two term exponential	$Xr = a.\exp(-kt) + (1-a).\exp(-kat)$	[19]
8	Midilli–Kucuk	$Xr = a.\exp(-kt^n) + bt$	[20]
9	Diffusion approach	$Xr = a.\exp(-kt) + (1-a).\exp(-kbt)$	[21]
10	Chavez-Mendez	$Xr = [1 - (1-b).at]^{1/(1-b)}$	[22]
11	Simplified Fick's diffusion	$Xr = a.\exp[-c(t/L^2)]$	[23]
12	Logistic	$Xr = b/(1 + a.\exp(kt))$	[24]

a, b, c, coefficients; n, specific exponent of each drying equation; k, g, specific coefficients of each drying equation; t is drying time; L, thickness of the gel.

- the statistic parameter khi-square (χ^2) was used to improve the smoothing precision. This parameter is computed as follows:

$$\chi^2 = \frac{\sum_{i=1}^N (X_{r_{exp,i}} - X_{r_{pre,i}})^2}{N - n_p} \quad (4)$$

- the root mean square error (RMSE) provided information on the short term performance. It was computed from the following equation:

$$RMSE = \sqrt{\frac{\sum_{i=1}^N (X_{exp,i} - X_{pre,i})^2}{N}} \quad (5)$$

where $X_{r_{exp,i}}$ stands for experimental moisture ratio found in the measurements, $X_{r_{pre,i}}$ is the predicted moisture ratio for this measurement, N is the number of observations, and n_p is the number of constants for a model. All of these statistical parameters as well as the drying constants and the coefficients for the semi-empirical drying models were computed by using a simple non-linear regression program carried out in MATLAB R2007b (Inc., Massachusetts, the USA). So, the best model to describe the gel kinetic drying curve was chosen according to the following criteria: R^2 near to unity, and then χ^2 and RMSE as low as possible.

2.4. Computation of effective diffusivity

In many studies, authors use water effective diffusivity (D_{eff}) to analyze and characterize the drying quantitatively [10]. Diffusivity is used in drying to describe the rate of flow of moisture out of material. Diffusivity is influenced by shrinkage, moisture content, case hardening, and the temperature of the product during drying [25]. It was assumed that our model-product is similar to an agricultural product rich in water (about 70% moisture). In the falling rate period of drying, moisture was transferred mainly by molecular diffusion according Fick's second law (Eq.6):

$$\frac{\partial X}{\partial t} = \nabla(D_{eff}(\nabla X)) \quad (6)$$

Fick's second law of unsteady state diffusion, given in Eq. (6), can be used to determine the moisture ratio in Eq. (7). The solution to the diffusion equation for an infinite slab was determined by Crank, (1975) [26]. Some assumptions were made in the estimation of moisture diffusivity of alginate gels during MWA drying [25]: (i) initial moisture content distribution is uniform throughout the product and tends towards zero with the time; (ii) water transfer is done exclusively by diffusion and there is no water transfer resistance on the surface; (iii) product shrinkage is negligible and diffusivity is constant and uniform.

$$MR = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left(- (2n+1)^2 \pi^2 \frac{D_{eff}}{L^2} t\right) \quad (7)$$

The simplified solution of Fick's model proposed by Crank (1975) [26] for infinite slab of thickness, L with a long drying time is as follows (Eq.8):

$$MR = \frac{8}{\pi^2} \exp\left(- \pi^2 \frac{D_{eff}}{L^2} t\right) \quad (8)$$

Eq. (8) could be further simplified to a straight-line equation as given below (Eq.9):

$$\ln(MR) = \ln\left(\frac{8}{\pi^2}\right) - \pi^2 \frac{D_{eff}}{L^2} t \quad (9)$$

Thus, effective moisture diffusivity was typically determined by plotting experimental drying data in terms of $\ln(MR)$ vs drying time and then calculating the slope ($\pi^2 D_{eff} / L^2$).

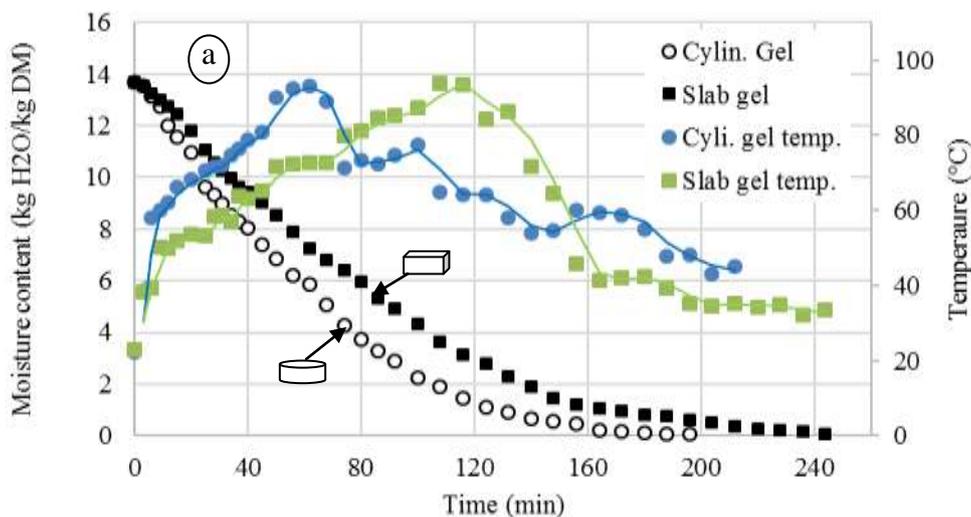
3. RESULTS AND DISCUSSION

3.1. Effect of the gel shape and microwave power on drying kinetics

Significant ($p < 0.05$) variation is observed during the evolution of gel moisture content of both of the shapes (cylindrical and slab) with the three specific powers (1 W/g, 1.5 W/g, 2 W/g) applied (Fig. 2a, 2b, 2c).

An analysis of these three figures shows that, whatever the output power used (1, 1.5, 2 W / g), the kinetics of microwave drying the cylindrical gel are faster than those of the slab gel. This means that the cylindrical gel dries quicker than the slab one. Indeed, at 1 W/g of microwave specific power, the cylindrical gel dried in 196 min whereas the slab gel dried in 244 min. It is the same for the application of 1.5 and 2 W/g, where the cylindrical gel was dried respectively in 172 and 108 min while the slab gel dried respectively in 220 and 132 min.

This difference was confirmed by the evolution of the gel temperatures. It was observed that temperature in center of the cylindrical shape sample increased more quickly than that of slab one, whatever the specific power used (fig. 2a, 2b and 2c). When the temperature of the product was higher, internal pressure increased, resulting in a greater water removal of the product by exudation [27]. This is characteristic of microwave volumetric heating [28]. Moreover, research showed that water removal from the product (drying rate) increases with temperature [29]. The periods of warming-up and evaporation of the water were reached more rapidly for the cylindrical shape than for the slab one. The same applies to cooling period. In this period, except for the convective heat losses, drying is isenthalpic. The absorbed power is used locally to evaporate water, which moves towards the surface under the influence of a favorable temperature and gas pressure gradient. As absorbed power and resistance to vapor transport decrease simultaneously, the temperature and gas pressure also decrease [30].



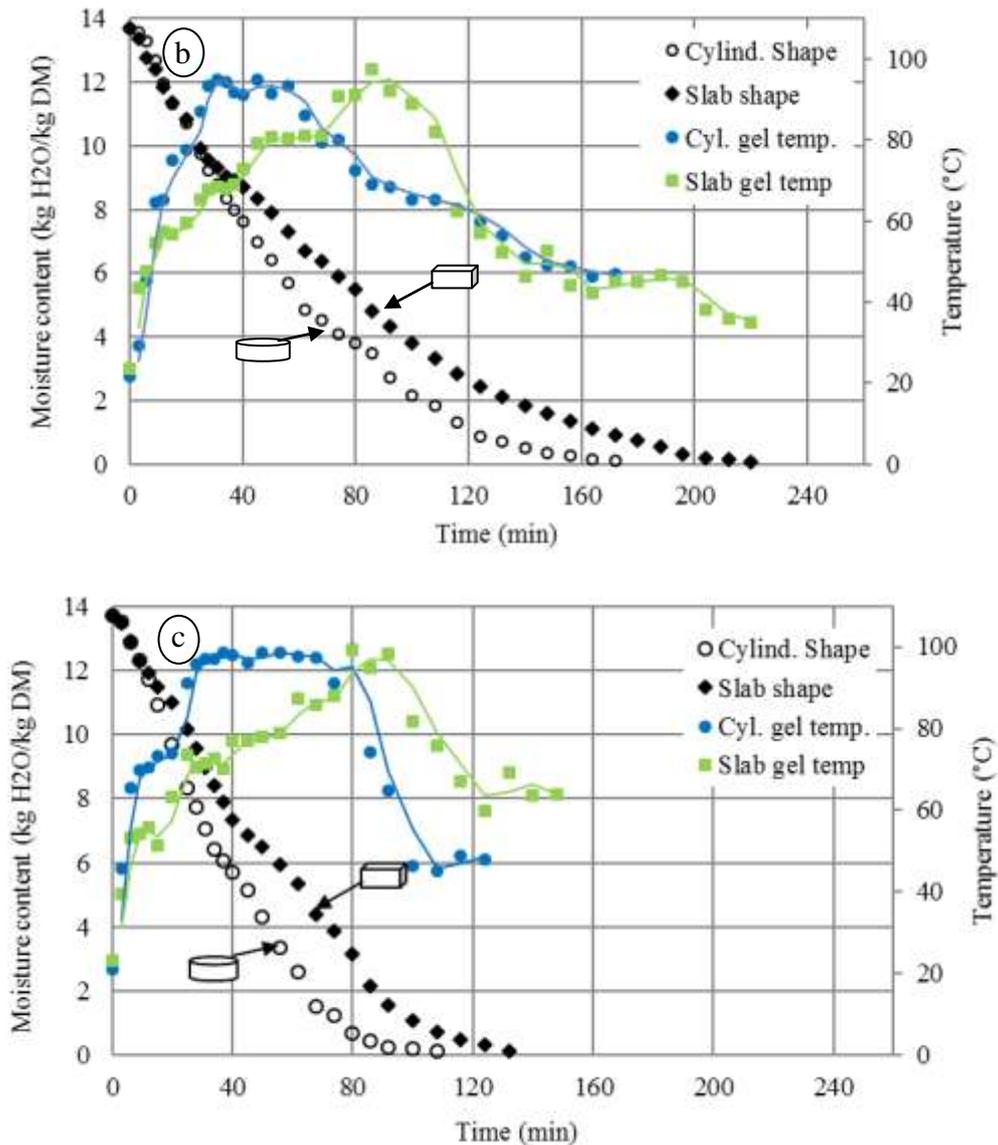


Figure 2: Effect of gel shape on microwave drying kinetics at different power densities: a) $P_s=1$ W/g; b) $P_s=1.5$ W/g; c) $P_s=2$ W/g.

In the final period, as the moisture content decreases, a large part of the energy is lost to convective heat or heating the medium. In Figures 2a to 2c, due to the very low absorbed power at the end of the drying cycle, an equilibrium is reached between the microwave heating and the convective losses by decreasing the gels temperatures [30]. McMinn et al., (2003) [21] has shown that during modelling the mass transfer of microwave drying of solid slab-shaped and cylindrical potatoes, their effective moisture diffusivities were respectively 1.91×10^{-8} and 19.99×10^{-8} m²/s for 90 W of microwave power (MP) applied, and respectively 3.73×10^{-8} and 24.22×10^{-8} m²/s for 650 W of MP. These results are consistent with our results and show that the cylindrical shape of the gel would favor more rapid drying of the food products relative to the slab shape. Therefore, this cylindrical shape will be used for the modelling of the drying kinetics.

3.2. Effect of the gel shape and microwave power on moisture diffusivity

Effective moisture diffusivity D_{eff} is usually computed by a graphic method, where drying experimental data is plotted in terms of the logarithm of moisture ratio X_r versus drying time (Figure 3a and 3b).

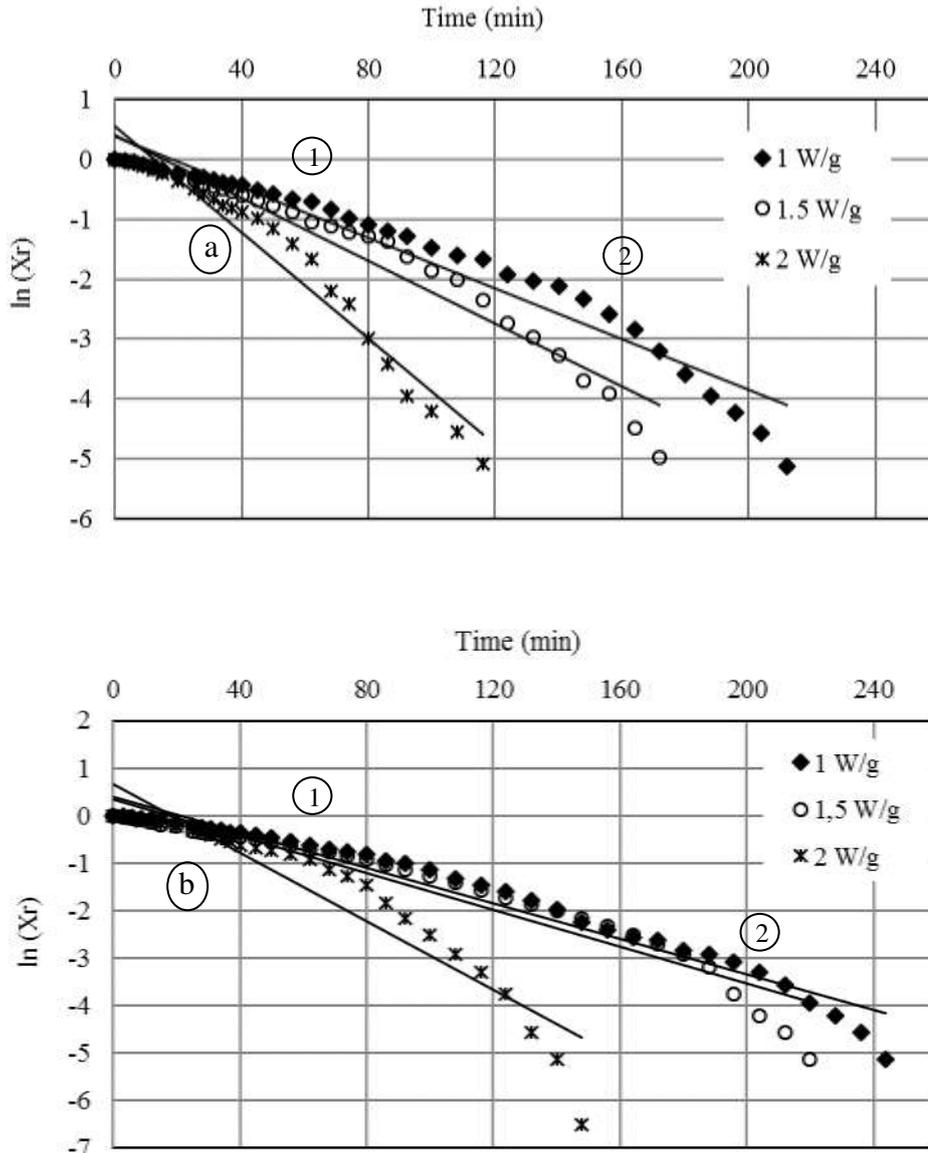


Figure 3: Variation of $\ln(X_r)$ versus time at different microwave powers of a) cylindrical and b) slab shaped gels

Figures 3a and 3b show a non-linear $\ln(X_r)$ function plot for the three powers levels. We observed two distinct and visible stages on the two almost linear parts of the curves. According to Maskan, (2001) [31], this indicates that drying takes place in the first and second slowing down periods. Indeed, according to Nadeau and Puiggali, (1995) [32], for hygroscopic raw material (case of alginate gel), during the slowing down period of drying, two stages exist.

The first one starts at critical point 1, where the evaporation zone "drying face" moves from the surface of material towards the interior. The abrupt reduction of the effective surface of transfer due to an insufficient interstitial water supply is the cause of this falling. The second stage or the "finale pace period" is the passage at critical point 2 (single with hygroscopic materials). On the other hand, it was noticed that, contrary to what is usually observed, effective diffusivity is higher in the second stage than the first one, the slowing down period. The same phenomenon was also observed by Giovanelli et al., (2002) [33] and Hawlader et al., (1991) [34] in the case of tomato hot air drying. This phenomenon was due to the shrinkage of samples during drying and, hence, to a reduction of thickness, resulting in faster water removal. The slope that was found allows for the determination of the various values of effective moisture diffusivity presented in Table 3.

Table 3: Effective diffusivity values of sodium alginate gel obtained at various microwave output powers

Ps (W/g)	D_{eff} ($\times 10^{-7}$ m ² /s)			
	Cylindrical Gel	R ²	Slab Gel	R ²
1	2.60 ± 0.03	0.94	2.29 ± 0.09	0.96
1.5	3.21 ± 0.02	0.95	2.38 ± 0.07	0.93
2	5.21 ± 0.20	0.96	4.43 ± 0.31	0.89

Effective diffusivity values increase with the microwave power applied. For the 3 levels of power, the effective diffusivity of the cylindrical gel is higher than that of the slab, which is expected. The found values of about 10⁻⁷ m²/s are markedly higher than the diffusivity matter values in food products, which ranged from 10⁻⁹ m²/s to 10⁻¹¹ m²/s [35], and are a little lower than the values fitted by Al-Harahsheh et al., (2009) [10], which are about $\times 10^{-6}$ m²/s for powers ranging between 3,2 W/g and 16 W/g.

3.3. Empirical models of microwave drying kinetics

The drying results used to obtain the reduced moisture content (Xr) were used to fit the models presented in Table 2. The values of the statistical parameters that were computed are listed in Table 4 below. The best model describing the microwave drying process of cylindrical gel is that with the highest R² and the lowest χ^2 and RMSE.

Table 4: Non-linear regression analyzes resulting from gel microwave drying at different specific powers

N°	1 W/g			1.5 W/g			2 W/g		
	R ²	χ^2	RMSE	R ²	χ^2	RMSE	R ²	χ^2	RMSE
1	0.98162	0.00191	0.04551	0.98222	0.00198	0.04444	0.96271	0.00368	0.06753
2	0.99785	0.00025	0.01578	0.99760	0.00028	0.01659	0.99721	0.00028	0.01884
3	0.98810	0.00138	0.03715	0.99025	0.00112	0.03346	0.97756	0.00229	0.05347
4	0.99724	0.00033	0.01814	0.99818	0.00022	0.01471	0.99158	0.00089	0.03346
5	0.99837	0.00019	0.01374	0.99754	0.00029	0.01681	0.99431	0.00072	0.02692
6	0.99777	0.00027	0.01607	0.99727	0.00032	0.01801	0.99578	0.00056	0.02318
7	0.99740	0.00030	0.01737	0.99700	0.00034	0.01856	0.99514	0.00050	0.02488
8^a	0.99882	0.00015	0.01206	0.99885	0.00014	0.01189	0.99774	0.00025	0.01773
9	0.98162	0.00219	0.04616	0.98222	0.00211	0.04519	0.96271	0.00393	0.06892
10	0.98090	0.00221	0.04639	0.98153	0.00212	0.04530	0.96177	0.00390	0.06837
11	0.98810	0.00138	0.03715	0.99025	0.00112	0.03346	0.97756	0.00229	0.05347
12	0.99865	0.00016	0.01269	0.99758	0.00029	0.01697	0.99698	0.00032	0.02003

^a This model of Midilli gives best results for sodium alginate gel by microwave drying.

In Table 4, it was observed that all of the studied models give a good drying curve adjustment with R² values greater than 0,95 and low values of χ^2 and RMSE. However, for each microwave power, the model best adjusted (best R², χ^2 and RMSE) is that of Midilli-Kucuk. It best describes the drying of gel behaviour (Figure 4) for the three powers levels. Similar results were reported for the microwaves drying of tomato pulp by Al-Harahsheh et al., (2009) [10]. It was the same for Simha et al., (2016) [36], who showed that, from ten empirical models, Midilli's model best describes the drying kinetics and microwave assisted extraction of bioactive compounds from *Adathoda vasica* and *Cymbopogon citratus*. The computation of Midilli's model parameters allowed us to obtain the constants (a, b, k and n) for each of the three power levels (Table 5).

Table 5: Constants of Midilli’s model according to the various microwave output powers

Specific power	Constants				R ²	χ ²
1 W/g	a = 0.9913	b = -0.0002	k = 0.0046	n = 1.229	0.9988	0.0001
1.5 W/g	a = 1.0174	b = -0.0003	k = 0.0087	n = 1.1409	0.9989	0.0001
2 W/g	a = 1.0021	b = -0.0002	k = 0.005	n = 1.409	0.9977	0.0002

This allowed for the creation of model curves for gel drying kinetics (Figure 4), which fit well with experimental data at 99% (R² value) with values of RMSE and khi² minimal.

3.4. Experimental and predicted moisture ratios

Curves of the moisture ratios of gel samples versus time of microwave drying at different specific outputs powers are shown in Figure 4.

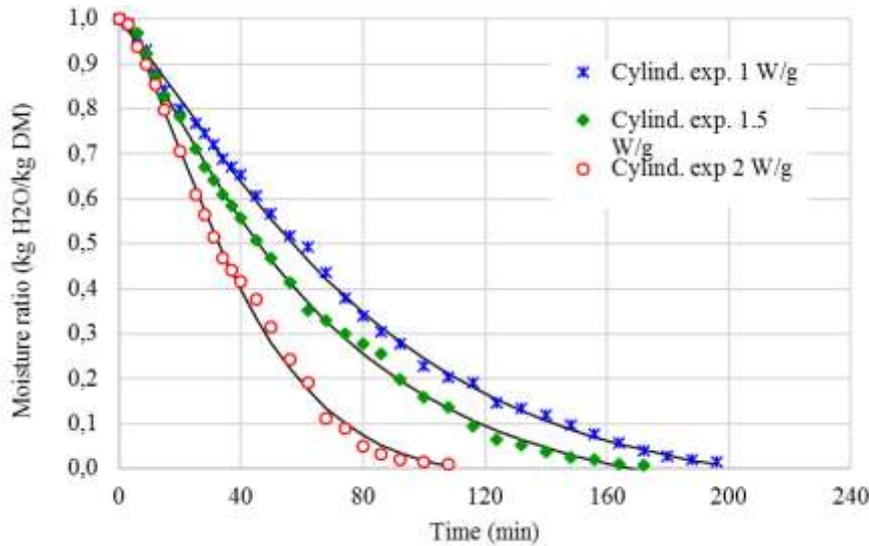


Figure 4: Impact of output power on drying kinetics and variation of experimental and predicted moisture ratio by Midilli model at different microwave powers (1 W/g, 1.5 W/g, 2 W/g).

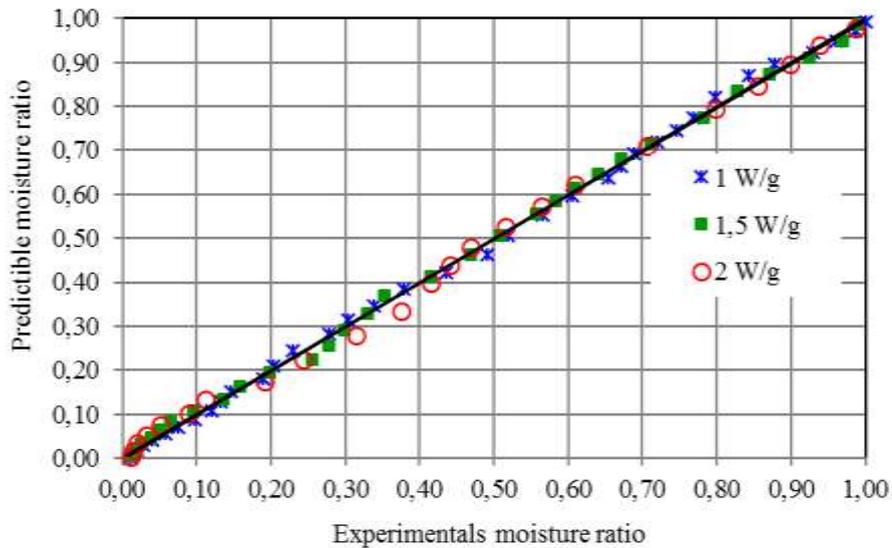


Figure 5: Evolution of the drying rate versus the moisture content at different microwave output powers (1 W/g, 1.5 W/g, 2 W/g).

With an increase in the microwave output power to the sample, the time necessary to achieve the moisture content of 8% ($\text{kg [H}_2\text{O].kg}^{-1}$ db) decreased. Thus, for the specific powers of 1, 1.5 and 2 W/g, respectively 212, 172 and 124 minutes of drying time were needed to reach the 8%. So, the increase in the drying microwave power resulted in a drying time decrease (Figure.4). The same effects of microwave power were observed by Soysal, (2004) [37] for parsley, Wang and Xi, (2005) [6] for carrot slices, and Gögüs and Maskan, (2001) [38] for olive pulp.

The drying curves of the moisture ratios and the processing times for alginate gels undergoing microwave drying were plotted. The simulations of the moisture ratios as drying time progressed were plotted using various model equations. The drying curves using the Middili et al model showed the best fit. Figure 5 shows the graph of the experimental moisture ratio and the predicted moisture ratios using Middili et model for microwave gel drying. Correlation coefficient are near 1 (0.99), which confirms that the Midilli model fits the experimental data set well.

4. CONCLUSION

The effect of the shape of a model-product (sodium alginate gel) on microwave drying was investigated and its drying kinetics were modeled. The results show that cylindrically shaped gels dry more quickly because its temperature increases faster of in its center. Thus, the cylindrical shape rather than the slab shape would be the most appropriate geometrical shape of a product for microwave drying. The analysis of gel moisture ratio during microwave drying seems to show two distinct aspects of drying, which is characteristic of hygroscopic materials. The experimental results of the cylindrical shaped gels were used to evaluate various semi-empirical models of drying that have been described in the literature. Among these models, for whichever power, the Midilli-Kucuk model gave the most satisfactory results. As for the microwave power, it significantly influenced the drying behavior of the model-product. In fact, a higher microwave power allowed for the reduction of the drying time of real products.

5. ACKNOWLEDGEMENT

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