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ORIGINAL RESEARCH PAPER

INFLUENCE OF DRYING CONDITIONS ON THE EFFECTIVE DIFFUSIVITY AND ACTIVATION ENERGY DURING CONVECTIVE AIR AND VACUUM DRYING OF PUMPKIN

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The main purpose of the work is to investigate the efficiency of convective air and vacuum processing on pumpkin drying kinetics. The pumpkin samples were of two different geometrical shapes (cylinder and cube) and were dried in a laboratory scale hot air dryer using some specific parameters (constant air velocity of 1.0 m/s, three different temperatures 50, 60 and 70ºC suited to relative humidity (RH) values of 9.8, 6.5, and 5.4% respectively). The vacuum drying was led at constant pressures of 5 kPa and accordance temperatures of 50, 60 and 70ºC. Moisture transfer from pumpkin slices was described by applying Fick's diffusion model. Temperature dependence of the effective diffusivity was described by the Arrhenius-type equation.

Cylindrical samples have a slightly better behaviour compared to cubic samples, due to the disposition of the tissues, and the mass and thermic transfer possibilities. Analysing the results of both drying methods, it was deduced that the most efficient method is convective air drying at 70ºC.

Keywords: pumpkin, convective air drying, vacuum drying, moisture diffusivity, activation energy

Introduction

The pumpkin *(Cucurbita moschata)* belongs to the family Cucurbitaceae. It is a seasonal vegetable traditionally used in human food. The pumpkin flesh is an important source of carotenoids. It has numerous culinary uses either as fresh vegetable or as ingredient in pies, soups, stews and breads (Doymaz, 2007). About 24,679,859.41tons of pumpkins were produced in the world in 2013.The largest pumpkin producers are China – 7,155,250 tons, India – 4,900,000 tons, the Russian

Federation – 1,128,205 tons, Iran – 897,293 tons, U.S. – 796,872 tons (FAO, 2013). Fresh pumpkins are very sensitive to microbial spoilage; consequently, they are often used as a dried product available as snack food, dried premix, soup as a premixed dried powder, etc. (Perez and Schmalko, 2007). Dried pumpkin may be a finished product or a half-finished product, subject to further processing. A properly selected drying method of the raw material may increase the quality of the finished product (Sojak and Głowacki, 2010). Pumpkin processing may cause oxidation and/or isomerization of different compounds, consequently resulting in loss of biological activity and/or color. The intensity of isomerisation reactions depends directly on factors such as temperature and contact with oxygen and light (Provesi and Amante, 2015).

The major compound which can be isomerized is β-carotene. Isomerisation reactions could be the first step of oxidation. β-carotene can be easily attacked by oxygen on either side of the cis bond. This radical can degenerate in homolytic internal substitution, which forms epoxides, which in turn can result in final stable products such as apocarotenoids and apocarotenal.

Food drying is one of the most cost-effective ways of preserving foods of all varieties, relevant and challenging unit operations in food processing, although the art of food preservation through the partial removal of the water content is still used nowadays. One of the biggest advantages of dried foods is that they take much less storage space than canned or frozen foods (Jangam *et al.*, 2010).

Dehydration offers a means of preserving foods in stable and safe conditions, as it reduces water activity and extends shelf-life much longer than that of fresh fruits and vegetables. Many conventional thermal methods, including hot-air drying, vacuum drying, and freeze-drying, occur in low drying rates in the falling rate period of drying (Doymaz, 2007; Piotrowski *et al.*, 2007; Arévalo-Pinedo and Murr, 2006; Jiang *et al.*, 2011). Drying process efficiency depends on two intrinsic factors: time and temperature. The long drying times at relatively high temperatures during the falling rate periods often lead to undesirable thermal degradation of the finished products (Mousa and Farid, 2002).

Different methods of drying could have different effects on the effective moisture diffusivity, shrinkage and other physical properties of the drying material (Hashemi *et al.*, 2009).

The most common drying method employed for food materials is hot air drying, which is the simplest and most economical among the various methods.

As well, vacuum drying is a process in which moist material is dried under sub atmospheric pressures, allowed drying temperature to be reduced and higher quality to be obtained than with classical air conventional process at atmospheric pressure.

Literature review includes numerous papers on the mathematical modelling of drying kinetics of fruits and vegetables (Doymaz, 2005; Arévalo-Pinedo and Murr, 2006), but unfortunately there are few publications about the influence of geometric shape on the drying process.

The objectives of this study were to investigate the effect of different geometrical shapes of pumpkins during convective air and vacuum drying, and calculations of effective moisture diffusivity and activation energy.

Abbreviations

MR moisture ratio, dimensionless

DR drying rate, g water/g dry matter/min

M moisture content, g water/g dry matter

M_0 initial moisture content, g water/g dry matter

M_e equilibrium moisture content, g water/g dry matter

t drying time, min

D_{eff} effective moisture diffusivity, m^2/s

D_0 pre-exponential factor of the Arrhenius equation, $m^{2/}s$

E_a activation energy for moisture diffusion, kJ/mol

R^2 coefficient of determination, dimensionless

R gas constant, kJ/(mol·K)

T_a absolute drying air temperature, K

Materials and methods

Raw material

Pumpkins were purchased at a local market, in Galati, Romania, hand peeled and washed in running tap water. The pumpkins were cut into cylinders with 5 mm height and 25 mm base diameter, and cubes with equal sides of 25 mm. The moisture content of fresh pumpkin was 88.8 ± 1.2%. All the samples were prepared in triplicate. Only the flesh from the pumpkin mesocarp was considered for the drying experiment.

Drying experiments

Convective air drying. The hot-air drying experiments were carried in a pilot plant tray dryer (UOP 8 TRAY DRYER, Armfield, UK (capacity up to 2.1 kg of wet material, at a flow rate ranging from 0.4 to 3.0m/s and temperatures up to 80ºC) from the Department of Food Engineering, Gaziantep University, Turkey), at a constant air velocity of 1.0 ms^{-1}, temperature 50, 60 and 70ºC and the correspondent relative humidity (RH) 9.8, 6.5 and 5.4% respectively.

Vacuum drying. The vacuum drying experiments were performed also at three different temperatures, 50, 60 and 70ºC, and under constant pressure of 5 kPa, using a BINDER (23VD series, BINDER GmbH, Germany) vacuum drying oven (from the Department of Food Engineering, Gaziantep University, Turkey). In the vacuum oven chamber the drying temperature distribution is homogeneous and the effect on the product is gentle. All the drying experiments were replicated three times.

Mathematical modelling of drying curves

The moisture ratio (MR) of pumpkin samples during drying experiments was calculated using the following equation:

$$MR = \frac{M - M_e}{M_0 - M_e} \quad (1)$$

The drying rate (DR) of the pumpkin during the drying process can be determined using the following equation:

$$DR = \frac{M_{t+dt} - M_t}{dt} \quad (2)$$

There are many methods to determine drying parameters, of which the most common is Fick's second law of diffusion (Boulaoued and Mhimid, 2012; Chen *et al.*, 2012):

$$\frac{\partial M}{\partial t} = D_{eff} \frac{\partial^2 M}{\partial x^2} \quad (3)$$

where D_{eff} is the diffusion coefficient and M – the moisture content, g water/g dry matter.

In equation (4) are expressed the analytical solutions for various geometries and the solution for slab object with constant and unidirectional diffusion, described for the first time by Crank (1975. This equation has been widely used to describe the drying process (Arévalo-Pinedo and Murr, 2006; Chen *et al.*, 2012; Simal, 2005; Göğüs and Maskan, 2006; Darvishi *et al.*, 2013):

$$\frac{M - M_e}{M_0 - M_e} = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left(-\frac{(2n+1)^2 \pi^2 D_{eff} t}{L^2}\right) \quad (4)$$

where D_{eff} is the effective moisture diffusivity (m^2/s), L is the thickness of the samples (m), and *n* is a positive integer.

For longer drying periods it is necessary to make some simplifying assumptions. It is assumed that the moisture transfer is unidirectional, the initial moisture is uniformly distributed and the diffusion coefficient of moisture is constant and negligible shrinkage:

$$\frac{M - M_e}{M_0 - M_e} = \frac{8}{\pi^2} \exp(-\pi^2 D_{eff} t / L^2) \quad (5)$$

Effective moisture diffusivity is actually calculated by using the slope of Eq. (5), namely, when the natural logarithm of MR versus time was plotted, straight line with a slope k_0 and Eq.(6) was obtained:

$$k_0 = \frac{\pi^2 D_{eff}}{L^2} \quad (6)$$

The effect of temperature on effective moisture diffusivity is generally expressed using an Arrhenius-type relationship, since temperature has a significant effect over the drying process rather than the initial moisture content of the product. Using Arrhenius Equation, as a relationship between temperature and effective moisture diffusivity, the activation energy can be calculated using Equation (7):

$$D_{eff} = D_0 \exp\left(-\frac{E_a}{T_a R}\right) \quad (7)$$

The activation energy expresses the effect of drying temperature on effective moisture diffusivity.

Results and discussion

Drying kinetics

To establish drying curves, the samples were weighed every 30 minutes, until they reached an equilibrium moisture content of 10.5±0.5%. Figure 1 shows how the moisture ratio depending on temperature varies in time, at constant air velocity – 1.0 m/s (in convective tray drier) and constant pressure – 5 kPa (vacuum drier oven) for different geometry of samples. Increasing the air temperature from 50˚C to 70˚C, the drying rate loss increased and the drying time decreased (from 420 min at 50ºC up to 270 min at 70ºC during hot-air drying and from 420 min at 50ºC up to 300 min at 70ºC during vacuum drying).

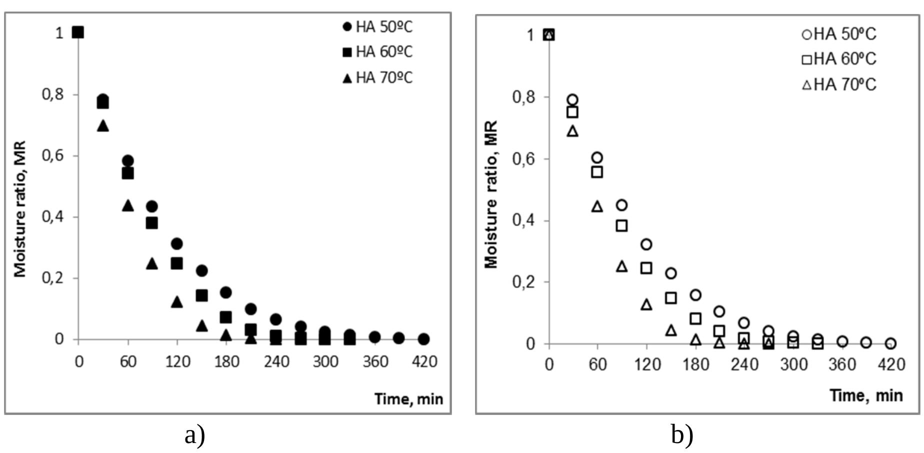


Figure 1. Effect of drying parameters (temperature and time) on MR of pumpkin: (a) cylinder shape, (b) cube shape

In the initial phase of the drying process (Figure 2), the moisture content (88.8 ± 1.2%) and the rate of moisture loss from pumpkin samples are high, in the range 0.7902 – 0.6905. During the drying processes, the moisture content of the pumpkin samples decreases up to 9.0±0.5%.

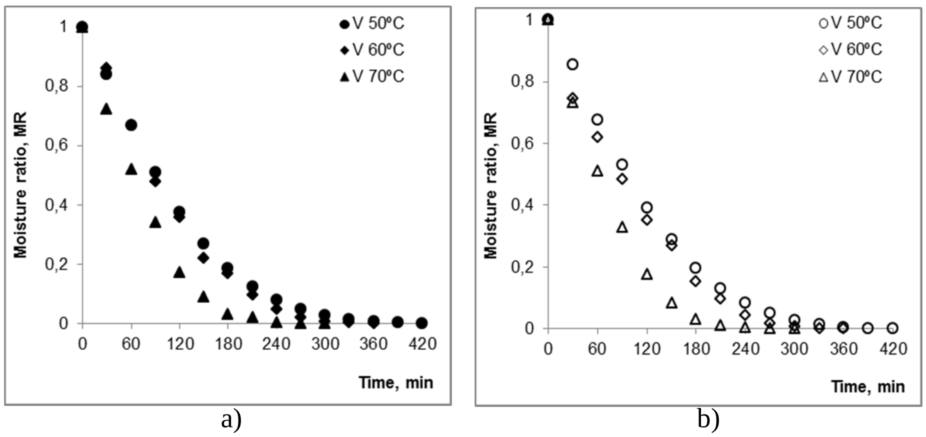


Figure 2. Influence of temperature and time on the MR of pumpkin during vacuum drying: (a) cylinder shape, (b) cube shape

A significant part of the product moisture is lost during the first drying period, and much time is needed to evaporate the remaining moisture to reach the equilibrium. This is explained as the first phase which represents the initial period, where sensible heat is transferred to the product directly influencing the moisture content. This marks the heating up point of the product from the inlet condition to the process one. The rate of evaporation increases dramatically during this period with most free moisture being removed. Thus, the drying curves of the pumpkin samples exhibit a fast drying rate at the beginning, followed by a long period of slower drying rate.

Usually, the drying process consists of two phases: a constant rate and a falling rate phase. Furthermore, the major part of the drying process runs at the falling rate phase. During the second phase, or constant rate period, free moisture persists on the surfaces and the rate of evaporation slightly decreased in direct relation to moisture content reduction.

In this study a drying constant rate period was not detected in the drying curves (Figure 3). In fact, drying only occurred in the falling rate period for all the temperatures investigated. In this period, the migration of moisture from the inner interstices of each particle to the outer surface becomes the limiting factor that reduces the drying rate. It can be seen that air temperature strongly affects the drying rate.

The results indicate in both cases that increased temperature leads to decreasing of moisture content and as a consequence the moisture ratio decreases proportionally with the drying rate. Several authors reported that drying rates increased with the increase in temperature and decreased in time for drying various vegetables such as pumpkin (Doymaz, 2007), mango (Corzo *et al*., 2008), red beetroot (Figiel, 2010).

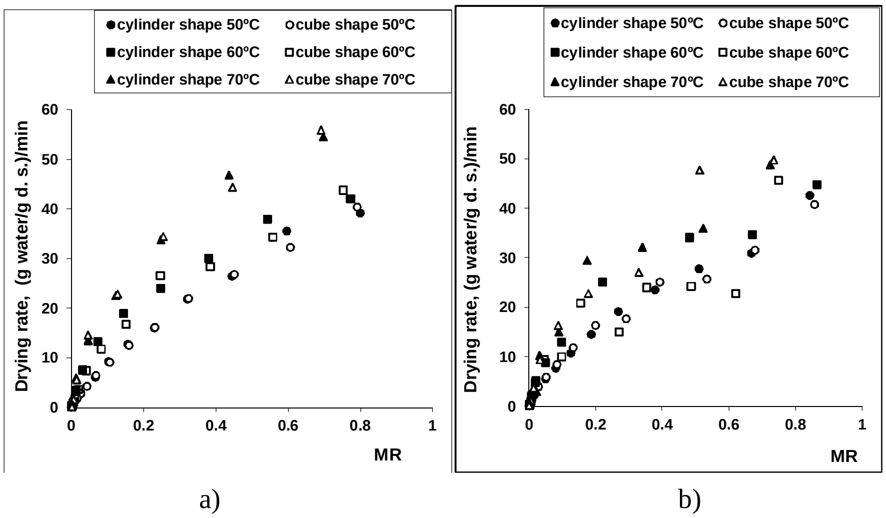


Figure 3. Effect of drying air temperature and moisture ratio on the drying rate of pumpkin samples: (a) convective air drying, (b) vacuum drying

Calculation of effective diffusivity and activation energy

Effective diffusivity values for different shapes at convective air and vacuum drying are presented in Table 1. Effective diffusivity was calculated using equations (Eq.5.) described in the Materials and Methods section.

Calculated effective moisture diffusivity (D_{eff}) increased directly proportional with drying temperature for both shapes of pumpkin samples and both drying methods. The minimum value of moisture diffusivity was 3.70–3.83 x10^{-8} (m^2/s) at 50ºC while the maximum value of moisture diffusivity was 6.19-7.10 x10^{-8} (m^2/s) at 70ºC. The experimental points were obtained for each temperature separately, by applying linear regression to the data after obtaining the slopes ln (MR) versus time plots.

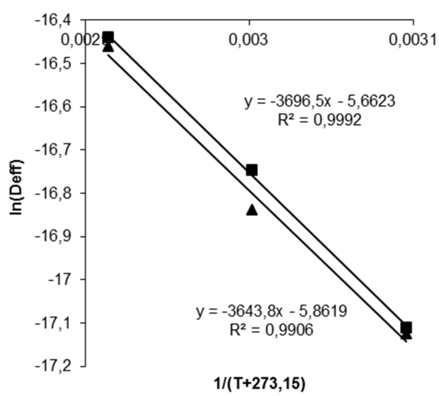
From Table 1 the increasing of the effective moisture diffusivity proportional with the temperature increasing can be observed. For the range of studied temperatures, the diffusivity for convective air drying is higher than for vacuum drying. This can be explained as a consequence of forced hot air drying velocity of 1 m/s, which transmits to the air a better contact with the sample surface.

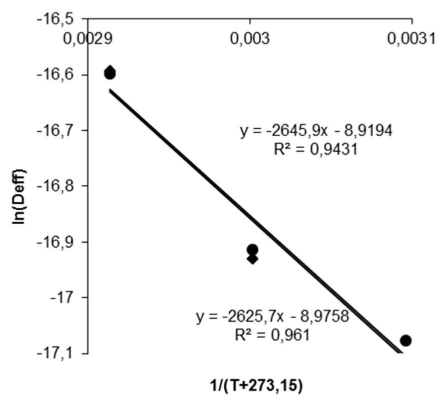
These values are higher than those found by Doymaz (2007) for convective air-drying (3.88×10^{-10} to 9.38×10^{-10} m^2/s) and by Arévalo-Pinedo and Murr (2007) for the vacuum drying of pumpkin samples in the range of temperatures from 50ºC to70ºC, which vary from 1.65×10^{-9} m^2/s to 2.63×10^{-9} m^2/s.

Table 1. Values of effective diffusivity of pumpkin samples at different temperatures

Drying method	Temperature	Geometrical shape	R^2	D_{eff} x10^{-8} (m^2/s)
Convective air	50ºC	cylinder	0.968	3.70
		cube	0.970	3.65
	60ºC	cylinder	0.953	5.32
		cube	0.962	4.87
	70ºC	cylinder	0.961	6.21
		cube	0.960	7.10
Vacuum	50ºC	cylinder	0.951	3.83
		cube	0.944	3.83
	60ºC	cylinder	0.926	4.44
		cube	0.918	4.51
	70ºC	cylinder	0.949	6.21
		cube	0.963	6.19

The activation energy (E_a) was calculated from the slope of the plot ln(D_{eff}) versus 1/(T + 273.15) in Fig.4. The value obtained for the diffusion coefficient at an infinite temperature, D_0 was 0.0034 m^2/s for convective air drying, and 0.0035 m^2/s for vacuum drying.





a) b)

Figure 4. Relationship between diffusivity and absolute temperature: (a) convective air drying, (b) vacuum drying

The results for the activation energy reported values from 12.7 to 110 kJ/mol for most food materials (Mirzaee *et al.*, 2009). The activation energy for moisture diffusion, E_a, was found to be 30.74 – 30.30 kJ/mol during convective air drying and 21.99 – 21.83 kJ/mol for vacuum drying. These values are lower when compared to the value reported by Doymaz (2007), 78.93 kJ/mol, for forced convective air drying, and similarly, the values of the Arrhenius constant and the activation energy for the convective mass transfer reported by Guiné *et al*. (2012).

Conclusions

Both convective air drying and vacuum drying seem to have similar influence on the pumpkin samples. Moisture diffusivity increased with drying air temperature. So, the highest moisture diffusivity 6.19-7.10 x10^{-8} (m^2/s) was found at 70ºC, while the lowest value 3.70–3.83 x10^{-8} (m^2/s) was reached at 50ºC.

The activation energy had higher values at convective air drying compared to vacuum drying. The energy of activation obtained for convective air drying is 30.74 – 30.30 kJ/mol and for vacuum drying is 21.99 – 21.83 kJ/mol. Even if it is known that the dimensions and shape of the samples influence the drying process performances, the present study demonstrated that the surfaces of heat transfer are almost similar for both cylinder and cube pumpkin samples.

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