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Studying and Modeling of Hydration Kinetics in Chickpea Seeds (*Cicer arietinum* L.).

SEYED MOJTABA SHAFAEI* AND AMIN ALLAH MASOUMI
Department of Agricultural Machinery Engineering, Faculty of Agriculture, Isfahan University of Technology, Isfahan, Iran.
*Corresponding Author: smshafaei@shirazu.ac.ir

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ABSTRACT

Standard models were used in several experiments in order to predict the moisture ratio of three major varieties of chickpea ('Desi', 'small Kabuli' and 'large Kabuli') cultivated in Iran. The experiments were carried out using distilled water at three temperatures (5, 25 and 45°C) in three replications. The amount of water absorption by seeds were calculated through measuring the increase in the mass of soaked seeds versus time. Fourteen standard models of water absorption were fitted to the experimental data. Coefficient of determination (R^2), chi-square (x^2) and root mean square error (*RMSE*) were used to evaluate the models. The appropriate model was chosen based on maximum value of coefficient of determination and minimum value of chi-square and root mean square error. The effective moisture diffusivity coefficient of three varieties in each temperature were determined according to Fick's equation. The result indicated that Binomial model is the most appropriate model for all studied varieties in three experimental temperatures in order to predict moisture ratio changes versus time in soaking. The plotted curves of water absorption of three varieties of chickpea indicated that moisture ratio decreases as temperature increases. Besides that, the effective moisture diffusivity coefficient of the three studied varieties showed an increasing trend by temperature increment from 5 to 45°C.

Keywords: binomial model, effective moisture diffusivity coefficient, immersion, moisture ratio, statistical index.

INTRODUCTION

Pulses are economical sources of protein, energy, vitamins and minerals. Food pulses diminished incidence of several diseases, for instance cancer, cardiovascular diseases, obesity and diabetes (Bhathena and Velasquez, 2002). Pulses contain relatively low quantities of the essential amino acid methionine, related to whole eggs, dairy products or meat. This means that a smaller proportion of the plant proteins, compared to proteins from eggs or meat, may be used for the synthesis of protein in humans.

Chickpeas (*Cicer arietinum* L.) are an important protein source in several developing countries. Chickpea is the third most commonly consumed legume in the world (Singh, 1990). There are two main varieties of chickpeas cultivated in Iran namely 'Desi' and 'Kabuli'. The 'Kabuli' type has thin, white seed coat while the 'Desi' type has a thick, colored seed coat and has smaller seed than 'Kabuli' type (Salunkhe *et al.,* 1985).

Since soaking the grains is usually used before dehulling and cooking, understanding water absorption of different seeds during soaking has been considered by researchers. Grains in different conditions of soaking have different water absorption rate and water absorption capacity (Sopade *et al.,* 1994). Understanding the water absorption in pulses during soaking is important since, it affects following processing operations and the quality of the final product (Turhan *et al.,* 2002). The water absorption of seed during soaking mainly depends on soaking time and water temperature. Applying warm water is a common method to reduce the soaking time, because higher temperature increases moisture diffusivity leading to higher hydration rate (Kashaninejad *et al.,* 2009).

Relationship between moisture content of seeds in soaking versus time has been expressed by different models (Shafaei and Masoumi, 2013b). Many theoretical and experimental methods have been employed and irregularly empirical models were desired because of their relative ease of use (Singh and Kulshrestha, 1987; Nussinovitch and Peleg, 1990). Also these models were used to predict dehydration of agricultural material. The most popular empirical and semi-empirical models, which has been used to model the water absorption process of agricultural products are 'Weibull' distribution function (Machado *et al.,* 1999; Marabi *et al.,* 2003; Garcia-Pascual *et al.,* 2006), and exponential model (Gowen *et al.,* 2007; Kashaninejad *et al.,* 2007). Empirical models are often preferred to the theoretical, due to their ease of computability and interpretation. Also, applying artificial neural network to predict water absorption of crop have been reported by many investigators (Kashaninejad *et al.,* 2009; Shafaei and Masoumi, 2013c).

Water absorption of 'Tarom' variety of rice was modeled by researchers. The results were demonstrated that the best equation for predicting the behavior of water uptake was 'Page' model. Also, the water uptake increased with increasing soaking temperature and soaking time. The effective moisture diffusivity coefficient through rice soaking in range 25 to 70 ° C was tested and described 5.58×10^{-11} to 3.57×10^{-10} respectively (Kashaninejad *et al*., 2007). Three mathematical models, 'Weibull', 'Peleg' and 'Exponential', for describing the water absorption kinetics of almond kernels were investigated by researchers. The studies on water immersion showed that 'Peleg' and 'Weibull' model were more accurate for recitation the water absorption characteristics of almond kernels (Khazaei, 2008). Water absorption process during wood soaking in water was studied on three varieties of wood. Two models were considered to describe the kinetics, the 'Peleg' and 'Khazaei' model, based on the viscoelastic properties of materials. The soaking data were fitted to the Fick's model to determine water diffusivity. The calculated diffusivity coefficients for 'Afra', 'Ojamlesh', and 'Roosi' wood varieties were reported 1.38×10^{-3}, 3.71×10^{-4}, and 4.88×10^{-4} (m^2s^{-1}) respectively (Khazaei, 2008). Other researchers reported that the effective moisture diffusivity coefficient varied from 8.376×10^{-12} to 2.22×10^{-12} (m^2s^{-1}) over the temperature range studied during sorghum soaking (Kashiri *et al*., 2010).

The objectives of the present study were to determine the best appropriate model for water absorption of three varieties of chickpea ('Desi', 'small Kabuli' ['Chico'] and 'large Kabuli' ['Kabuli']) to predict moisture ratio changes by passing the time during soaking and to determine the effective moisture diffusivity coefficient of chickpea during immersion.

MATERIALS AND METHODS

Sample Procurement:

Three varieties of chickpea were supplied from Legumes Seed Collection Center, Agricultural Organization, Khomein, Arak, Iran. The broken seeds and external materials were eliminated. Within each cultivar of chickpea, seeds were separated as three groups of size. Medium-size seeds were used, in order to abolish the effect of seed size on the soaking trials. The initial moisture content of samples was measured by following AACC 44-15A method (AACC, 1999).

Soaking Treatment:

Tests were conducted in distilled water at 5, 25 and 45°C for each variety at different durations. Containers and distilled water were kept in desired temperature for a few hours to reach the desire temperature, before each experiment.

For each duration that was included in the timetable, ten seeds of each cultivar were randomly selected and weighed, then placed in glass beakers containing 200 milliliters of distilled water. The amount of water absorption by various seeds were determined at 5, 10, 15, 30 minutes and one hour after immersion. The tests followed at intervals of one hour toward gelatinized seeds. After reaching at each fixed sampling time, the samples were drained on a paper and the excess water was eliminated with adsorbent paper, and the soaked sample were weighed.

A digital chronometer and an electronic weighing balance (AND, Model GF400, Japan) with a precision up to 0.001 (g) were used to control soaking duration and to measure weight of samples respectively, before and after soaking. Experiments were completed in three replications. The water absorption capacity was determined by follow equation (Mc Watters *et al.,* 2002):

$$W_a = \frac{W_f - W_i}{W_i} \times 100 \tag{1}$$

Where, W_a is water absorption (d. b. %), W_f is weight of seeds after immersion (g) and W_i is weight of seeds before immersion.

According to Peleg (1988), points were intentionally selected from recorded data, as that extremely small weight gains at the beginning of soaking were not included. Also, data with increasing losses of soluble solids of more than 1% of the initial samples mass were not included. Therefore, at each stage, the amount of solid material dissolved in water was controlled by measuring density of distilled water and drained water in each experiment.

Models Evaluation:

In most studies, water absorption and drying model are achieved based on the moisture ratio (*MR*), due to fewer data dispersion and optimize data (Akpinar *et al.*, 2003).

$$MR = \frac{M_c - M_e}{M_o - M_e} \tag{2}$$

Where *MR* is moisture ratio at time *t*, M_o is initial moisture content (d. b. %), M_e is saturated moisture

(d. b. %) and M_c is moisture content at time t (d. b. %).

The most common water absorption models for seeds, which were focused by researchers, are shown in Table (1) (Khazaei, 2008; Rafiee *et al*., 2009). The parameters of these models of each sample in water absorption during soaking were extracted using Matlab software. In order to evaluate the models, three parameters namely coefficient of determination (R^2), Chi-square (x^2) and root mean square error ($RMSE$) were determined based on equation (3), (4) and (5) respectively (Giner and Mascheroni, 2002; Garcia-Pascual *et al*., 2006).

$$R^2 = \frac{\sum_{i=1}^{N}(M_{exp,i}-M_{exp\,ave})^2-\sum_{i=1}^{N}(M_{exp,i}-M_{pre,i})^2}{\sum_{i=1}^{N}(M_{exp,i}-M_{exp\,ave})^2} \quad (3)$$

$$x^2 = \frac{\sum_{i=1}^{N}(M_{exp,i}-M_{pre,i})^2}{N-n} \quad (4)$$

$$RMSE = \left[\frac{1}{N}\sum_{I=1}^{N}(M_{pre,i}-M_{exp,i})^2\right]^{1/2} \quad (5)$$

Where, $M_{exp,i}$ is the ith experimentally observed moisture content (d. b. %), $M_{pre,i}$ the ith predicted moisture content (d. b. %), $M_{exp\ ave}$ is average moisture content observed (d. b. %), N is the number of data and n is the number of the constant coefficient of model.

Table 1. Regression models used for modeling moisture ratio in chickpeas.

Model	Equations
Nyton	$MR=exp(-kt)$
Page	$MR=exp(-kt^n)$
Modified Page	$MR=exp[-(kt)^n]$
Henderson and Pabis	$MR=aexp(-kt)$
Modified Henderson and Pabis	$MR=aexp(-kt)+bexp(-gt)+cexp(-ht)$
Logarithmic	$MR=aexp(-kt)+c$
Binomial	$MR=aexp(-k_0t)+bexp(-k_1t)$
Modified Binomial	$MR=aexp(-kt)+bexp(-gt)+c$
Binomial exponential	$MR=aexp(-kt)+exp(-mt)$
Wang and Sang	$MR=1+at+bt^2$
Diffusion	$MR=aexp(-kt)+(1-a)exp(-kbt)$
Midili *et al.*	$MR=aexp(-kt^n)+bt$
Werma *et al.*	$MR=aexp(-kt)+(1-a)exp(-gt)$
Weibull	$MR=exp(-(t/\beta)^\alpha)$

Regression index in each temperature were calculated and compared together. The best model was chosen based on maximum value of coefficient of determination and minimum value of chi-square and root mean square error.

Effective Moisture Diffusivity Coefficient:

Previous studies have shown that moisture transfer occurs mainly through the distribution process, during water absorption of food. Fick's second law can be expressed this distribution by following equation (6) (Doymaz and Pala, 2003):

$$\frac{\partial M}{\partial t} = \nabla^2(D_{eff}M) \quad (6)$$

Where M is moisture content at time t (d. b. %), D_{eef} is the effective diffusivity coefficient (m^2s^{-1}). Equation (6) can be written as spherical coordinates as equation (7):

$$\frac{\partial M}{\partial t} = \left(D_{eff}\left(\frac{\partial^2 M}{\partial r^2}+\frac{2}{r}\frac{\partial M}{\partial r}\right)\right) \quad (7)$$

Note that, (r) is grains spherical radius. The algebraic solution of equation (7) can be written as form of equation (8):

$$MR = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}exp\left(\frac{-D_{eff}n^2\pi^2}{r^2}t\right) \quad (8)$$

Higher-order terms that there is no significant change in the results were ignored. Thus, the first term of equation (8) can be rewritten as equation (9):

$$MR = \frac{6}{\pi^2}exp\left(\frac{-D_{eff}\pi^2}{r^2}t\right) \quad (9)$$

Equation (9) can be presented as equation (10) in logarithm form:

$$Ln(MR) = Ln\left(\frac{6}{\pi^2}\right)-\frac{D_{eff}\pi^2}{r^2}t \quad (10)$$

A linear line with a slope (S) is obtained, by plotting the natural logarithm of the data collected during the soaking test versus time; which is equal to coefficient (t) in relation (10). the effective moisture diffusion coefficient can be assisted as equation (11). This method has been used by many researchers (Ozbek and Dadali, 2007; Wang *et al.*, 2007).

$$S= \frac{D_{eff}\pi^2}{r^2} \quad (11)$$

Seed Spherical Radius:

The seeds spherical radius (r) was determined by measuring the average volume of seeds of each variety and using the volume of the equivalent sphere of seeds.

Seed Volume:

Fifty seeds of each variety was selected randomly in order to determine seed volume. To measure volume of each type of seed equation (12) was used (Mohsenin, 1978):

$$V_s=\frac{(W_{pf}-W_p)-(W_{pfs}-W_{ps})}{\rho_f} \quad (12)$$

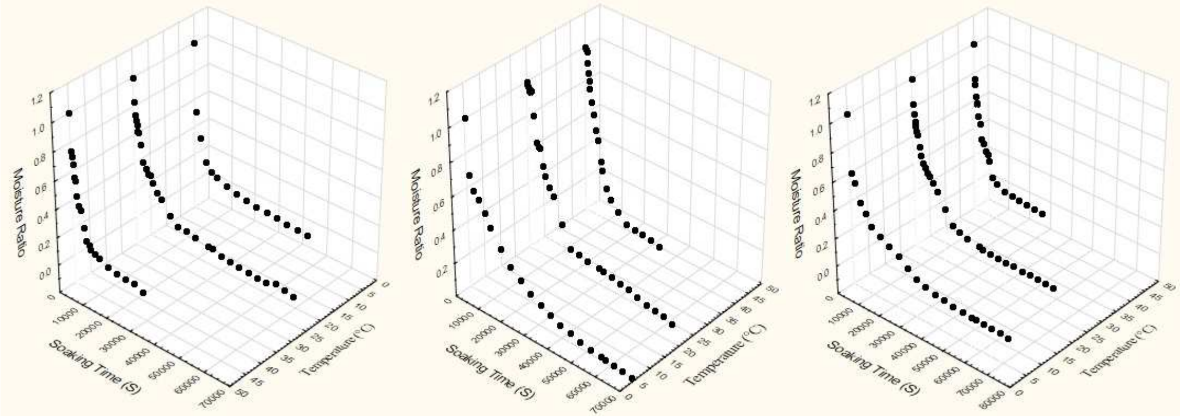
Where V_s volume of solid particles or seeds (m^3), W_p is empty weight of Pycnometer (g), W_{pf} is weight of filled Pycnometer with fluid (g), W_{ps} is weight of Pycnometer containing seeds (g), W_{pfs} is weight of Pycnometer containing seeds and fluid (g), ρ_f is density of fluid (g m^{-3}).

Ethanol was used as Pycnometer fluid because water can penetrate in seeds and it is not suitable as Pycnometer fluid.

RESULTS AND DISCUSSION

Moisture Ratio Curves:

Values of initial moisture content of chickpea were 8.74, 7.79 and 8.86% dry basis for 'Desi', 'Chico' and 'Kabuli', respectively and they were not significantly different at $P>0.05$. The decreasing moisture ratio of samples during soaking time is shown in Fig. 1. Moisture ratio curves show the rate of water absorption increased with increasing temperature, thus with increasing temperature moisture ratio is decreasing and tending to zero faster. In higher water temperature, the time needed to reach saturated moisture was shorter for samples. The reason of this phenomena is increasing of propagation velocity of water in seeds. Higher temperatures result to the grain gelatinization and will lead to the expansion and softening of grain. Therefore, more pores and cracks opened and finally transmission of water through the seed increases (Ranjbari *et al.*, 2011). Thus, high temperatures can cause the seeds to soften and expand. The water absorption rate will be higher, if the soaking temperature is closer to gelatinization temperature of seed. Therefore, use of higher temperatures on short time has resulted to reach equilibrium moisture in shorter time during soaking. Similar results have been reported for various legumes such as chickpea, cow chickpea, soybean, chick peanuts and bean (Sopade and Obekpa, 1990; Sopade and Kaimur, 1999; Turhan *et al.*, 2002; Pan and Tangratanavalee, 2003, Shafaei and Masoumi, 2013a).



(a): 'Chico' (b): 'Desi' (c): 'Kabuli'

Fig. 1. Moisture ratio curves of chickpea varieties during soaking.

By comparing each variety, result indicate that water absorption value are not significantly difference ($P>0.05$). It seems that this is due to the same cultivation condition and partial difference on morphology and physiologic properties of these varieties of chickpea in Iran (Shafaei and Masoumi, 2014).

Choosing Appropriate Model:

Fourteen standard models of water absorption were fitted to the experimental data in present study, as mentioned in Table (1). According to the value of R^2, x^2 and *RMSE* (which described moisture ratio changes during soaking) the corresponding models are listed for each type of chickpea (Table 2, 3 and 4). The appropriate model was chosen based on maximum value of coefficient of determination (R^2) and minimum value of chi-square (x^2) and root mean square error (*RMSE*). Results demonstrated that, Binomial model is the most appropriate for three varieties in three experimental temperatures to predict moisture ratio changes versus time of soaking. The coefficients of Binomial model of each variety at different temperatures are shown in Table (5). The moisture ratio versus time was plotted for varieties, using Binomial model (Fig. 2). Similar to our findings, Rafiee *et al.* (2009) reported modified Binomial model as an appreciate model for drying thin layers of orange slices.

Table 2. Average statistical index of the fitted models of soaking at three different water temperatures for 'Desi' variety.

Model	R^2	$x^2 \times 10^{-3}$	*RMSE*
Nyton	0.776	0.874	0.0376
Page	0.982	0.263	0.0313
Modified Page	0.992	0.265	0.0263
Henderson and Pabis	0.995	0.211	0.0220
Modified Henderson and Pabis	0.996	0.179	0.0223
Logarithmic	0.996	0.159	0.0200
Binomial	0.996	0.152	0.0201
Modified Binomial	0.996	0.163	0.0210
Binomial exponential	0.996	0.154	0.0198
Wang and Sang	0.976	0.945	0.0957
Diffusion	0.994	0.293	0.0254
Midili *et al.*	0.996	0.160	0.0196
Werma *et al.*	0.978	0.428	0.0373
Weibull	0.992	0.263	0.0263

Effect of Temperature on Moisture Diffusivity Coefficient:

Fig. 3 shows the natural logarithm of the data versus time plotted during soaking tests for ‘Chico’ type. The slop of lines express the effective diffusivity moisture coefficient based on Fick’s equation. Analogizing line slope indicate that increasing water temperature result in increasing effective moisture diffusivity coefficient which in its turn does not have a significant difference at $P>0.05$.

Table 3. Average statistical index of the fitted models of soaking at three different water temperatures for ‘Chico’ variety.

Model	R^2	$x^2 \times 10^{-3}$	*RMSE*
Nyton	0.853	0.332	0.0497
Page	0.993	0.124	0.0191
Modified Page	0.981	0.209	0.0253
Henderson and Pabis	0.990	0.402	0.0234
Modified Henderson and Pabis	0.990	0.139	0.0221
Logarithmic	0.995	0.118	0.0177
Binomial	0.996	0.088	0.0157
Modified Binomial	0.996	0.087	0.0160
Binomial exponential	0.949	0.170	0.0543
Wang and Sang	0.962	0.145	0.0957
Diffusion	0.988	0.203	0.0256
Midili *et al.*	0.995	0.149	0.0172
Werma *et al.*	0.988	0.203	0.0256
Weibull	0.993	0.124	0.0191

Table 4. Average statistical index of the fitted models of soaking at three different water temperatures for ‘Kabuli’ variety.

Model	R^2	$x^2 \times 10^{-3}$	*RMSE*
Nyton	0.886	0.794	0.0696
Page	0.985	0.205	0.0254
Modified Page	0.985	0.201	0.0255
Henderson and Pabis	0.992	0.343	0.0207
Modified Henderson and Pabis	0.992	0.129	0.0214
Logarithmic	0.992	0.139	0.0205
Binomial	0.994	0.086	0.0168
Modified Binomial	0.994	0.085	0.0171
Binomial exponential	0.945	0.290	0.0567
Wang and Sang	0.964	0.443	0.0769
Diffusion	0.994	0.093	0.0171
Midili *et al.*	0.994	0.105	0.0180
Werma *et al.*	0.993	0.094	0.0172
Weibull	0.985	0.205	0.0255

Table 5. The coefficient of Binomial model fitted to chickpea varieties.

Variety	**Temperature (°C)**	α	β	K_0(h^{-1})	K_1(h^{-1})
‘Desi’	5	0.7540	0.0004	0.2126	0.2561
	25	0.1164	0.9525	0.0750	0.4223
	45	0.4878	0.5686	0.5897	0.5971
‘Chico’	5	0.5410	0.1762	1.6020	0.2088
	25	0.3098	0.5331	0.1803	0.8882
	45	0.0890	0.7493	0.1738	1.1922
‘Kabuli’	5	0.7540	0.5621	4.4080	0.2141
	25	0.2093	0.7002	4.3430	0.3483
	45	0.7194	0.1459	0.6527	5.3531

This phenomena is due to dense texture of chickpea seeds and their resistance to water penetration in deep layers. By increasing the temperature of water used for soaking, seeds tend to increase their moisture content in a short period. Thus, volume and surface of seeds increase faster which results in increasing effective moisture diffusivity coefficient. Same results was obtained for other studied varieties in the present study. The effective moisture diffusivity coefficient of seeds in three experiment temperatures are reported in Table (6). Similar reports on corn seeds during soaking process has been published by Kashiri *et al.,* (2010).

CONCLUSION

The Summary of the results that were obtained in the present experiment indicate that all recommended models by researchers that were fitted to chickpea soaking data are appropriate. The Binomial model was the most suitable model in order to predict moisture content of different types of chickpea during soaking and could be applied to assessment the moisture content at given soaking time and temperature within the considered experimental conditions. The corresponding plotted curves of each variety of chickpeas in three experimental temperatures demonstrated that moisture ratio decrease by temperature increment; and the effective moisture diffusivity coefficient increases with water temperature increment according to the Fick's equation.

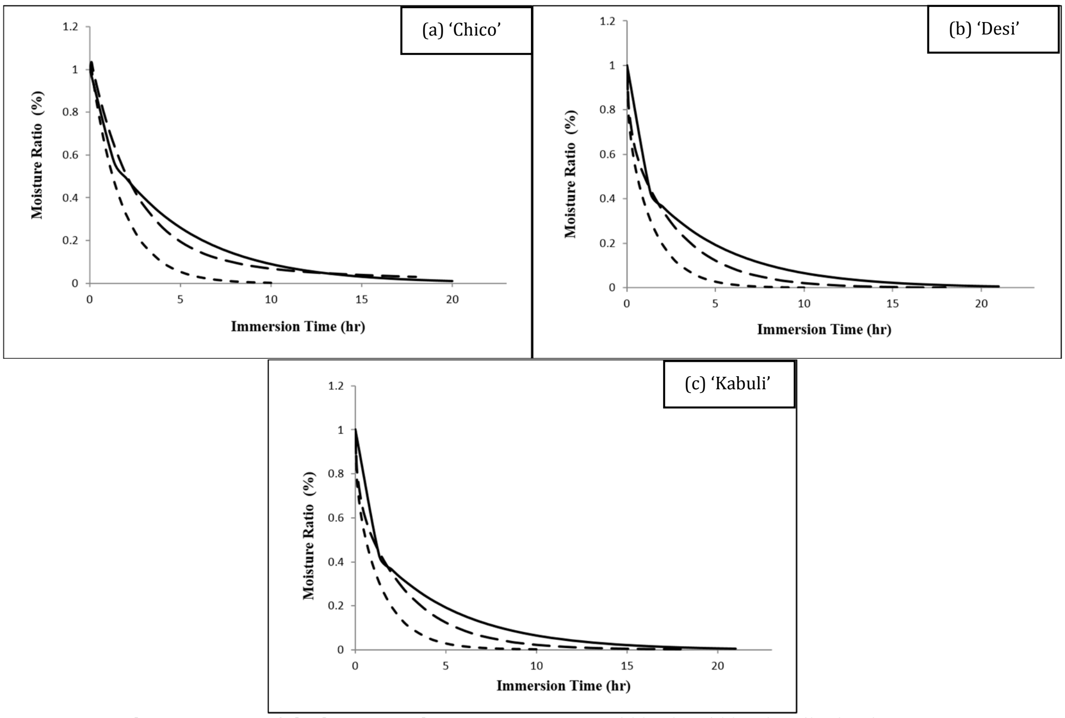


Fig. 2. Moisture ratio characteristics of chickpea type during immersion at: 5 ˚(____), 25 ˚(_ _ _), 45˚C (……).

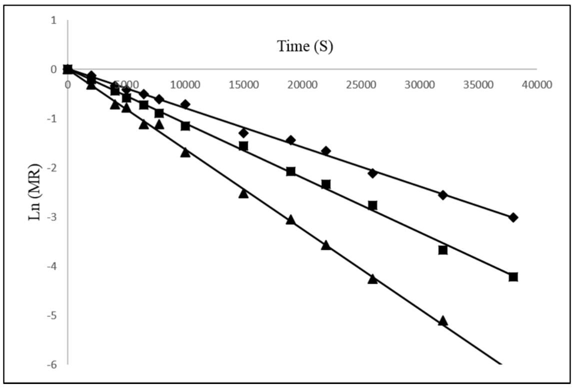


Fig. 3. The effective moisture diffusivity coefficient of 'Chico' variety during soaking (♦5˚, ■25˚, ▲45˚C).

Table 6. The effective moisture diffusivity coefficient Value of chickpea varieties during soaking.

Variety	Temperature (˚C)	The effective moisture diffusivity coefficient (m^2s^{-1}) $\times 10^{-6}$
'Chico'	5	1.023
	25	1.348
	45	1.751
'Desi'	5	1.591
	25	1.901
	45	2.024
'Kabuli'	5	1.503
	25	1.671
	45	1.943

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