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Modeling water uptake in a cereal grain during soaking

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ABSTRACT

A diffusion equation to describe the isothermal absorption of liquid water in a spherical solid that undergoes uniform swelling was derived. The resulting partial differential equation was solved using a finite difference method, taking into consideration water content dependence of the diffusion coefficient. The developed model was applied to simulate the water uptake of brown rice for the soaking temperatures of 25, 45, 55 and 65 °C. The estimated "differential" diffusion coefficients were a strongly increasing function of moisture content for all temperatures tested, approaching to the self-diffusion coefficients of water for brown rice moisture contents near to the saturation values. The "integral" diffusion coefficient corresponding to range of moisture content resulting from soaking conditions were calculated and correlated according to Arrhenius equation with an activation energy of 32.5 kJ/mol.

1. Introduction

Considerable research was reported in the literature to develop an understanding of the mechanism of moisture movement in natural products, but findings are not yet conclusive. Fick's second law for diffusion was used by various investigators to describe water absorption in grains and legumes, accepting the hypothesis that the resistance to water flow is distributed throughout the material and that this does not swell during process (Becker, 1960; Fan et al., 1965; Engels et al., 1986; Patil, 1988; Thakur and Gupta, 2006). Others solutions have been formulated considering variable diffusion coefficient but neglecting shrinking (Chu and Hustrulid, 1968; Aguerre et al., 1985; Dutta et al., 1988; Tolaba et al., 1997; Landman and Please, 1999).

Swelling of biological products during soaking takes place simultaneously with water diffusion and thus may affect the water absorption rate. Hence, a study of the swelling phenomena is of importance for better understanding of the soaking process. Consideration of swelling in soaking process is generally difficult because of the lack of information about swelling velocity and its relationship with water diffusivity. A mathematical expression of Fick's second law for drying of an infinite slab that shrinks unidirectionally was formulated by Viollaz and Suarez (1984) assuming volume additivity for water and dry solid. Gekas and Lamberg (1991) derived relationships for the diffusion coefficient in systems where volume changes occur during drying. A mathematical model was developed by Hawlader et al. (1999) to describe heat and mass transfer within materials undergoing shrinking during drying.

The purpose of this study is to develop a mathematical model describing the water transfer in spherical solid that undergoes swelling, taking into consideration the dependence on water concentration of the diffusion coefficient. The governing equations and corresponding boundary conditions are solved using the finite difference method to simulate the soaking process of rice grain, and determine the liquid diffusion coefficient of the starchy endosperm and its dependency on water concentration.

1.1. Model developing

Moisture migration in a swelling solid during soaking was modeled as a diffusive process in a binary system, formed by a solid B whose mass remains constant and a diffusant A, in our case moisture migrating in the liquid phase. Fick's equation can be applied to describe the movement of moisture in a solid, assumed here of spherical shape. To facilitate the calculation procedure the following assumptions are made:

(a) Swelling are only due to moisture absorption.
(b) The absorption process is assumed to occur under isothermal conditions.
(c) The swelling is assumed uniform and its magnitude is equal to the volume of water gained.

The diffusion equation for shrinking and non-shrinking sphere is,

$$\frac{\partial \rho_A}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left\{ r^2 D(\rho_A)\frac{\partial \rho_A}{\partial r}\right\} \tag{1}$$

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In this equation ρ_A represents the local volumetric concentration of water and $D(\rho_A)$ is the diffusion coefficient, a function of moisture concentration, r, is the radius of sphere and t, time. Expanding the differentiation in Eq. (1) it results,

$$\frac{\partial \rho_A}{\partial t} = D(\rho_A)\frac{\partial^2 \rho_A}{\partial r^2} + 2D(\rho_A)\frac{1}{r}\frac{\partial \rho_A}{\partial r} \tag{2}$$

For a solid that undergoes a volume change, Eq. (2) represents a Steffan problem because the domain of integration varies with time. To solve Eq. (2) uniform initial concentration and boundary conditions of the first time are assumed. The mathematical expressions of these conditions are,

$$\rho_A = \rho_{A_0} \quad \text{for } t = 0 \tag{3}$$

$$\rho_A = \rho_{Ai} \quad \text{for } t > 0 \quad \text{and} \quad r = R \tag{4}$$

$$\frac{\partial \rho_A}{\partial r} = 0 \quad \text{for } t > 0 \quad \text{and} \quad r = 0 \tag{5}$$

where ρ_{A_0} is the initial concentration of moisture and ρ_{Ai} the saturation moisture, assuming that the surface of the solid reaches instantaneously the moisture concentration corresponding to saturation, while R represents the instantaneous radius of the solid, a function of time owing to the swelling of the solid.

Eqs. (2)–(5) cannot be solved analytically owing to the movement of the solid boundary with respect to the fixed coordinate. So, the usual procedure of finite difference is quite difficult to apply. To overcome this difficulty, the following transformation introduced by Landau (1950) in his work of heat transfer with phase change is used here,

$$z = r/R \tag{6}$$

By means of this transformation it is possible to integrate over a fixed domain because the new variable, z, is zero at the center of the sphere and unity at the surface ($r = R$). In terms of the new variable, Eq. (2) can be transformed as follows:

$$\frac{\partial \rho_A}{\partial t} = \frac{1}{(zR)^2}\frac{\partial}{\partial z}\left\{z^2 D(\rho_A)\frac{\partial \rho_A}{\partial z}\right\} + \frac{z}{R}\left(\frac{\partial \rho_A}{\partial z}\right)\frac{dR}{dt} \tag{7}$$

Eq. (7) is undetermined at the center of the solid, $z = 0$, so the L'Hopital rule can be applied to obtain,

$$\frac{\partial \rho_A}{\partial t} = 3\frac{D(\rho_A)}{R^2}\frac{\partial^2 \rho_A}{\partial z^2} \tag{8}$$

To solve Eqs. (7) and (8) it is necessary to have an expression for the movement of the interfase, dR/dt. To generate an equation for the variation of R with time it is necessary to perform a mass balance for the water uptake in the solid, so

$$\frac{\rho_W}{A}\frac{dV}{dt} = \rho_W\frac{dR}{dt} \tag{9}$$

where ρ_W is the density of water and A and V are the surface and volume of the sphere. The flux of water at the surface of the sphere is,

$$\left|-D(\rho_A)\frac{\partial \rho_A}{\partial r}\right|_{r=R} = -\left|\frac{1}{R}D(\rho_A)\frac{\partial \rho_A}{\partial z}\right|_{z=1} \tag{10}$$

From Eqs. (9) and (10) we can obtained the following expression for surface movement:

$$\frac{dR}{d} = -\frac{D(\rho_A)}{\rho_W R}\left|\frac{\partial \rho_A}{\partial z}\right|_{z=1} \tag{11}$$

The instantaneous radius R can be estimated as a function of the solid average concentration, $\overline{\rho_B}$, taking into consideration that the mass of solid B is constant,

$$R = R_0\left(\frac{\rho_{B_0}}{\overline{\rho_B}}\right)^{1/3} \tag{12}$$

where R_0 is the initial radius of the solid and $\overline{\rho_{B_0}}$ is the initial concentration of solid mater.

1.2. Moisture dependence of diffusion coefficient

Even though it is generally recognized that the diffusion coefficient in starchy foods depends on moisture concentration (Saravacos and Raouzeos, 1984; Zhang et al., 1984; Leslie et al., 1991), until now few theoretical expressions of such dependence has been formulated. A rather simple expression can be obtained starting from the Arrhenius equation and using the relationship postulated by de Boer (1968) between the activation energy for the diffusive process, E_D, and the heat of sorption, E_S, as,

$$E_D = aE_S \tag{13}$$

Adopting an Arrhenius-type relation between $D(\rho_A)$ and the activation energy for diffusion, and taking into consideration that the value of $a = 0.5$ for a various food products (Aguerre et al., 1989), it results that,

$$D(\rho_A) = D_0 \exp\left(-\frac{E_S}{2R_g T}\right) \tag{14}$$

In Eq. (14) D_0 is a constant (equivalent to the diffusion coefficient at infinitely high temperature), T is the absolute temperature and R_g is the gas constant. To have an explicit relationship between the diffusion coefficient and moisture content it is possible to express E_S in terms of the isosteric heat of sorption,

$$E_S = Q_S(u) + \lambda(T) \tag{15}$$

where $\lambda(T)$ is the heat of water vaporization at the temperature of the process and $Q_S(u)$ is the isosteric heat of sorption, generally expressed in terms of the local moisture content in dry basis, u, related to local volumetric concentration, ρ_A, by means of the equation,

$$u = \frac{\rho_A}{\rho_B} \tag{16}$$

being ρ_B the local volumetric concentration of solid. A rather simple expression was obtained by Aguerre et al. (1986) to correlate the isosteric heat of sorption with moisture content,

$$Q_S(u) = R_g K_1 K_2^{u/u_i} \tag{17}$$

In this equation K_1 and K_2 are constants to be calculated from sorptional equilibrium data and u_i is the monolayer moisture content. Finally, if Eqs. (15) and (17) are replaced into Eq. (14) we obtain,

$$D(\rho_A) = D_0 \exp\left[-\frac{(R_g K_1 K_2^{u/u_i} + \lambda(T))}{2R_g T}\right] \tag{18}$$

2. Materials and methods

2.1. Materials

A local long-grain variety of rice obtained from a commercial mill was selected as the material for this study. The initial moisture content of grains, determined by AOAC (1996) vacuum oven procedure (5 h at 100 °C), was 0.108 kg water/kg dry solid. Grains were hand selected to remove foreign materials and damaged grains previous to experiments.

2.2. Dehulling of rice

The dehulling of rice grains was performed in a laboratory mill, Suzuki MT-95 (Japan), consisting of hull and bran removal sections. The dehulled or brown rice was graded and broken grains were separated.

2.3. Size characterization

As rice grain is irregularly shaped, the equivalent spherical radius was employed to represent rice grain for mathematical modeling. The total volume of 10 grains (dehulled) was measured with volume displacement of cyclohexane and replicated four times. The average grain volume was calculated by dividing the total sample volume by the total number of grains and the equivalent spherical radius was computed from the expression of the volume of the sphere. The equivalent spherical radius calculated by this procedure was 1.48 mm.

2.4. Dry solid density

Dry solid density of brown kernels, ρ_S, was determined following the experimental procedure used by Rovedo et al. (1997); the corresponding value is given in Table 1.

2.5. Soaking test

The dehulled rice was soaked in distilled water between 25 °C and 65 °C in 250 ml glass beakers. Approximately, 10 g of rice kernels were placed into each beaker containing 200 ml water. Several beakers were placed in a constant temperature shaker, controlled within ±0.5 °C of the testing temperature. The shaker was set at 30 cycles per minute, higher shaker speeds were tried and found to have no effect on the water uptake. For moisture content determination of the samples the beakers were removed at specified times from the shaker. The grains were then removed from the beakers and surface dried by using paper towel to eliminate the shiny layer of water from the rice. The kernels were then transferred onto a metal cup and weighed on precision balance to an accuracy of ±0.1 mg. The moisture content of rice was calculated based on the initial moisture content previously determined. All soaking tests were done in duplicate.

2.6. Numerical solution

To solve Eqs. (7) and (8) and the respective boundary conditions, together with Eq. (18) that gives the dependence of the diffusion coefficient with moisture concentration, the three-level implicit method developed by Lees (1959) was used. The input parameters of the numerical simulation were:

(a) the number of hypothetical shells into which grain kernel is divided,
(b) the initial moisture content of the kernel at all nodal points,
(c) kernel dimensions and water and grain properties listed in Table 1.

Table 1
Parameter values used to solve Eqs. (7) and (8) and Eq. (18).

Parameter	Reference
R_0 = 1.48 mm	This work
u_0 = 0.108 kg water/kg dry solid	This work
ρ_S = 1530 kg/m^3	This work
K_1 = 12,920 K; K_2 = 0.2740	Tolaba et al. (1997)
$\lambda(T)$ = 2495.46 + 1881(T − 273) − 4180(T − 273)	Treybal (1980)
ρ_W = 1000 kg/m^3	

Once performed the numerical integration for each time interval, moisture profile in terms of volumetric local concentration, ρ_A, was calculated. After that, the mean moisture concentration, $\overline{\rho_A}$, was obtained by integration of moisture profile by means of Simpson's rule. Assuming volume additivity it is possible to calculate the new mean value of solid concentration, $\overline{\rho_B}$, as follows:

$$\frac{\overline{\rho_A}}{\rho_W} + \frac{\overline{\rho_B}}{\rho_S} = 1 \tag{19}$$

where ρ_S is the density of dried solid and ρ_W the water density. Finally, the instantaneous radius of the sphere can be estimated substituting the value of $\overline{\rho_B}$ obtained from Eq. (19) into Eq. (12).

3. Results and discussion

The relationship between moisture gain by brown rice and time of soaking for different temperatures are shown in Fig. 1. Most of the curves follow a similar pattern, the grains gain moisture rapidly during the initial stages of hydration until absorption ceases when they attained the saturation value. The rate of water uptake increased with increasing temperature as suggested by the slopes of the absorption curves, getting steeper with increasing temperature. The saturation moisture contents were obtained from soaking data by extrapolation the rate of change of moisture, $d\bar{u}/dt$ against the moisture content, $\bar{u}$, to the point where $d\bar{u}/dt = 0$; the saturation values are given in Table 2. The marked increase of the saturation moisture content observed for the soaking temperature of 65 °C can be attributed to some degree of gelatinization of rice kernels, which accelerate the absorption rate.

3.1. Modeling of water absorption of rice

In order to fit experimental data of brown rice to the present model a non-linear regression analysis was employed; the fitting parameter was D_o. The mean relative deviation modulus (P) between the experimental and predicted mean moisture contents ($\bar{u}$) has been used as a measured of model adequacy,

$$P = \frac{100}{n}\sum_{1}^{n}\left|\overline{u_{actual}} - \overline{u_{predicted}}\right|/\overline{u_{actual}} \tag{20}$$

Modeled absorption curves (Fig. 2) followed the observed data closely showing a reasonable agreement. The values of the parameter D_0 and of the deviation modulus P are given in Table 2. Certain discrepancies between experimental and predicted values were observed for the soaking temperature of 65 °C, which are likely due to the fact that some of the starch is becoming gelatinized increasing the water holding capacity of grains.

Typical moisture profiles in terms of local moisture content, u, in dry basis are shown in Fig. 3a for different soaking times and for the soaking temperature of 45 °C. Moisture profiles were also calculated in terms of local moisture concentration per unit of volume, ρ_A, making use of the following relationship:

$$\rho_A = \frac{\rho_W \rho_S u}{\rho_W + \rho_S u} \tag{21}$$

These, represented in Fig. 3b, show certain differences with that shown in Fig. 3a attributed not only to the non-linear relationship between both magnitudes but also to the fact that moisture and solid concentrations are not uniform distributed within the solid, i.e., solid concentration is much higher in the center of the solid than in the surface. Moisture profiles illustrated in Fig. 3a and b show certain convexity near the surface of the solid (unlike the concave pro-

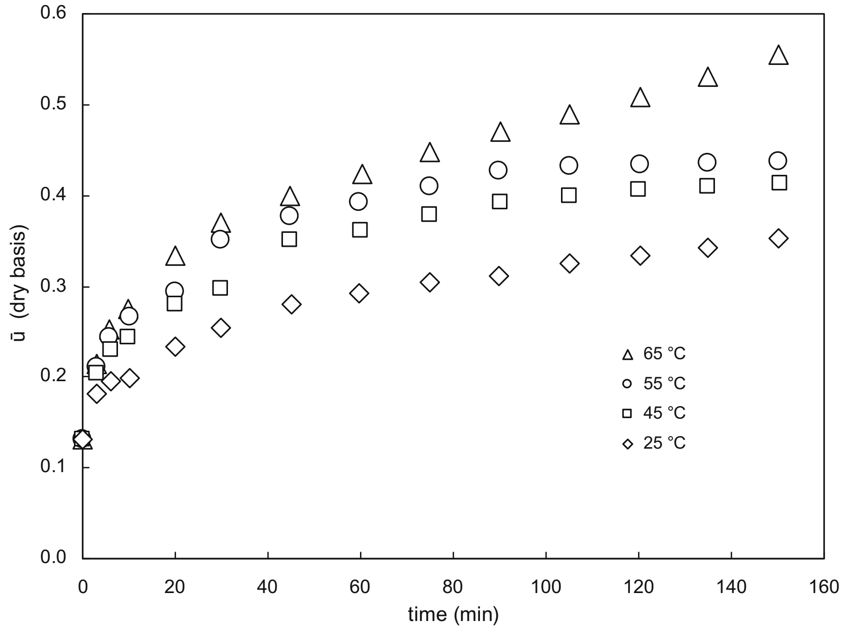

Fig. 1. Effect of temperature and soaking time on mean moisture content ($\bar{u}$) of brown rice.

Table 2
Saturation moisture content (u_i), pre-exponential of Eq. (18), D_0, time average diffusion coefficient ($\overline{\overline{D}}$) and mean relative deviation modulus (P) for brown rice soaked at different temperatures.

Temperature (°C)	u_i (dry basis)	D_0 (m^2/s)	$\overline{\overline{D}}$ (m^2/s)	P (%)
25	0.369	4.71×10^{-6}	7.80×10^{-11}	3.93
45	0.423	4.73×10^{-6}	1.79×10^{-10}	4.93
55	0.439	4.63×10^{-6}	2.60×10^{-10}	4.85
65	0.477	4.81×10^{-6}	3.38×10^{-10}	5.02

files corresponding to the classical solution of Fick's second law). It is also observed that during the first times of soaking moisture migration conducts to a kind of frontier that movies within the solid at an almost constant rate (this can be seen by observing the position where moisture profiles intercept the horizontal axis). As time of soaking increases moisture content is almost uniform across the solid. Such rapid increase of moisture concentration near the solid surface may be due to the variation of the diffusion coefficient with moisture concentration. The increase of the diffusion coefficient in the outer zones of the solid tends to accelerate the absorption rate with the corresponding sudden increase of moisture concentration.

Moisture dependence of the diffusion coefficient was estimated from Eq. (18) making use of the respective moisture profiles predicted from the model. With such purpose it was defined the "instantaneous" mean diffusion coefficient, $\overline{D}$, as,

$$\overline{D} = \frac{\int_0^R D(u) u r^2 dr}{\int_0^R u r^2 dr} \tag{22}$$

The variation of $\overline{D}$ with moisture content is shown in Fig. 4 for the four soaking temperatures investigated. So, at 65 °C the value of $\overline{D}$ changed from an initial value of 0.48×10^{-10} m^2/s to 3.75×10^{-10} m^2/s for the interval of maximum water uptake.

Such increase of the diffusion coefficient with moisture content can be attributed to the decrease in the heat of adsorption as kernel hydration proceeds (at high moisture content molecules are less firmly bound to the sorption sites). It can also be seen that as grain moisture content approaches the saturation value, the value of $\overline{D}$ is almost constant and coincident with that corresponding to self-diffusion coefficient of water (broken lines in Fig. 4).

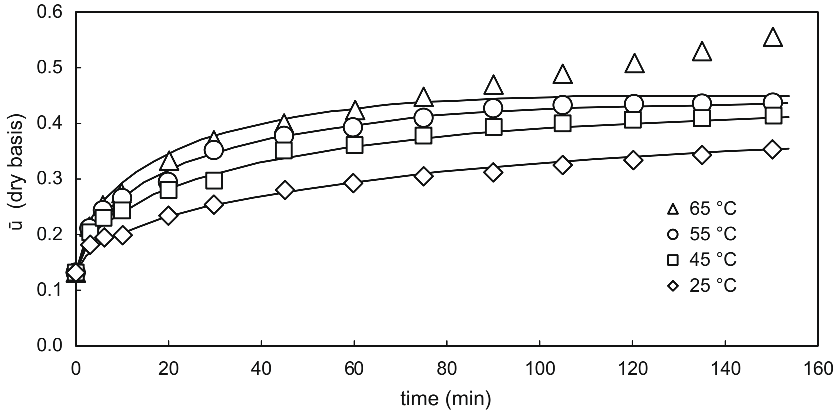

Fig. 2. Comparison of observed and predicted mean moisture contents ($\bar{u}$) of brown rice at 25, 45, 55 and 65 °C.

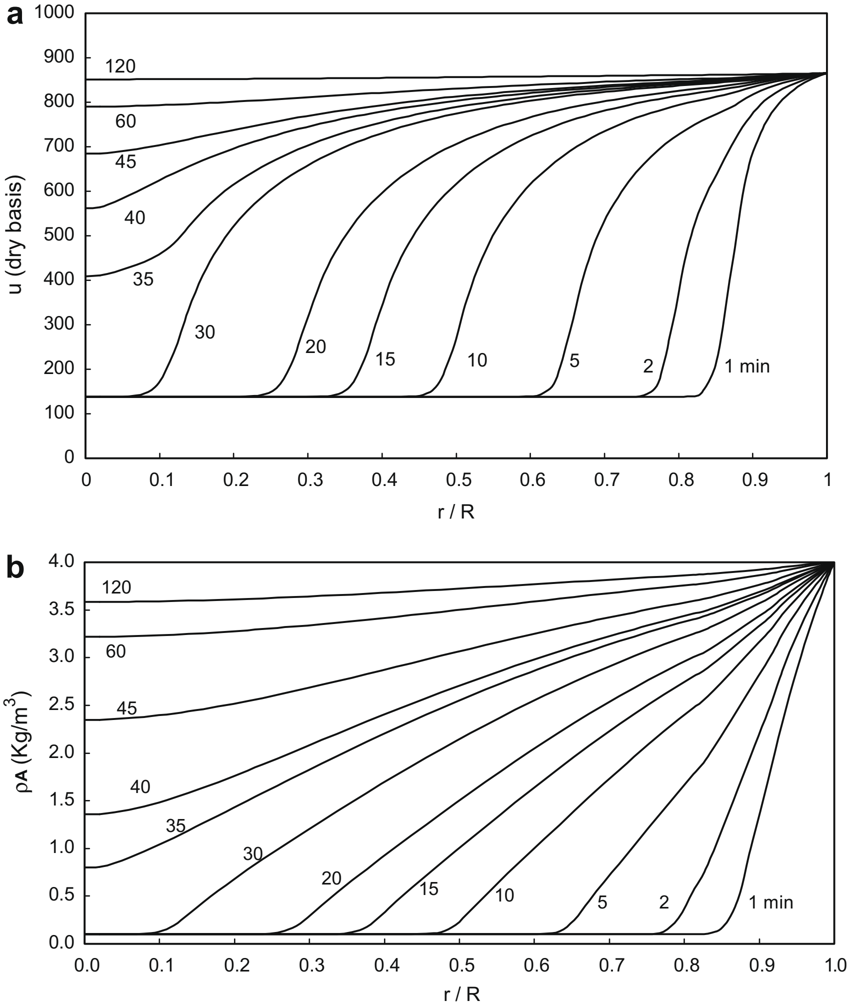

Fig. 3. (a, b) Local moisture variation with time as predicted by the model for soaking temperature of 45 °C: (a) moisture content, u (dry basis) and (b) moisture volumetric concentration, ρ_A.

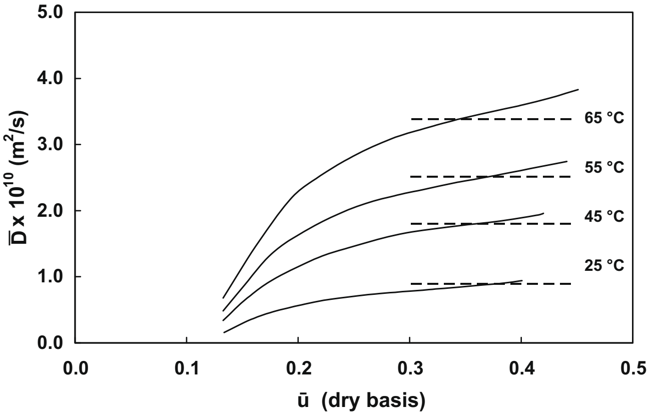

Fig. 4. Variation of the "instantaneous" mean diffusion coefficient ($\overline{D}$) with mean moisture content ($\bar{u}$) as a function of soaking temperature (broken line, self-diffusion coefficient of water).

It was also calculated a "time" average diffusion coefficient, $\overline{\overline{D}}$, by means of the following expression:

$$\overline{\overline{D}} = \frac{1}{t}\int_0^t \overline{D}dt \tag{23}$$

where t is the soaking time varying between initial and final moisture contents; the respective values of $\overline{\overline{D}}$ are given in Table 2.

This diffusion coefficient is representative of the entire soaking process and may be used for the purposes of comparison with moisture diffusivity data, which are usually obtained from the analytical solution of Fick's diffusion equation for constant diffusion coefficient. Values of $\overline{\overline{D}}$ varied between 7.80×10^{-11} m^2/s to 3.38×10^{-10} m^2/s for the respective temperatures of 25 and 65 °C. For water soaking of brown rice between 30 °C and 50 °C, Engels et al. (1986) report diffusion coefficients between 0.2×10^{-10} m^2/s and 0.72×10^{-10} m^2/s, while Lu et al. (1994) found values much larger (2.17×10^{-9} and 3.39×10^{-7} m^2/s) for soaking of rough rice between 20 °C and 50 °C. More recently, Thakur and Gupta (2006) found for water absorption of brown rice values of diffusion coefficient ranging from 3.89×10^{-11} to 1.46×10^{-10} m^2/s between 30 and 60 °C.

Finally, the effect of temperature on mean diffusion coefficient was described by an Arrhenius-type equation and the activation energy calculated from the slope of plot of log $\overline{\overline{D}}$ versus inverse temperature; the estimated activation energy was 32.5 ± 5.2 kJ/mol. The resulting activation energy is in reasonable agreement with the data reported by other investigators. So, for example, Engels et al. (1986) found activation energies varying from 22.5 to 64.5 kJ/mol, while Thakur and Gupta (2006) reported activation energy for brown rice of 37.32 kJ/mol.

4. Conclusions

The diffusion equation for sphere that swells uniformly during hydration process was derived by means of Landau transformation that transform the problem of diffusion with moving boundary, into one with fixed domain of integration. The resulting equation, that includes an explicit term for the rate of swelling of solid boundary, was numerically solved to simulate hydration of brown rice assuming volume additivity for water and solid and taking into consideration the variation of the diffusion coefficient with moisture concentration.

Predicted moisture profiles and their variation with time show marked differences depending on the unit used to represent local moisture concentration, i.e., volumetric concentration of moisture, ρ_A, or moisture content on dry basis, u. Such differences are the result of factors such as the non-linear relationship between ρ_A and u, the increase of the length of diffusion path as solid swells and to the marked decrease of the diffusion coefficient as moisture penetrates into the solid sphere.

The present model predicts that the 'differential' diffusion coefficient ($\overline{D}$) increases as the mean moisture content of rice grain increases with time, approaching to the self-diffusion coefficient of water as the moisture content reaches the saturation value. According to the present model, such increase is consequence of the decrease of the heat of adsorption as rice grain hydrates.

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