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Application of Peleg's equation for modeling of water absorption kinetics of paddy (*Oryza sativa*)

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Abstract : Peleg's equation was applied to depict the nature of hydration characteristics of paddy grain during soaking temperature of 10°C, 20°C and 30°C for 30 minutes to 12 hr. This equation was adequately capable to predict the hydration characteristics of paddy under experimental conditions. The relationship between two Peleg's constant (k_1, k_2) and temperature showed that Peleg rate constant (k_1, h%$^{-1}$) reduces with the increase in hydration temperature while Peleg capacity constant (k_2, %$^{-1}$) increases with the rise in temperature. This indicated that water absorption rate increased and water absorption capacity decreased with increase in temperature. An Arrhenius-type equation was used to describe the temperature dependence of k_1 and k_2 (R^2>0.98) and activation energy was found to be 35.83kJ/mol. Using the value of activation energy different thermodynamic parameters were evaluated.

Keywords : Paddy; Water absorption; Peleg's equation; Hydration kinetics; Activation energy.

INTRODUCTION

Paddy (*Oryza sativa*) is one of the most widely grown food grain crop which serves as the staple food for about half of global population. This popular cereal crop is second only to maize in terms of worldwide annual production for food use.

Steeping of paddy grain has gained a lot of attention due to its usefulness in further processing of grain. After harvesting paddy is usually subjected to two types of moisture treatment namely drying for safe storage and water absorption for further processing[1]. The principal reason for soaking is to gelatinize the starch in the grain and it can be achieved either through conditioning below the gelatinization temperature and then cooking above the gelatinization temperature, or through direct cooking above the gelatinization temperature[2]. Palatability and digestibility are also improved due to soaking. Soaking is the first step in water penetration, which transforms the inactive tissue into living tissue[3]. Absorption of water activates grain metabolism which results in degradation of reserve grain nutrients into simpler form. Hydration is the important unit operation which facilitates further processing and quality of the

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final product[4]. Hence there is a need to study the hydration characteristics of paddy grain in order to control and predict process parameters[5].

Each cereal grains have its own optimum soaking time and temperature. Hydration of grain is influenced by time of soaking and temperature of water[6,7]. Therefore it is of practical significance to optimize soaking conditions and predict water absorption as a function of time and temperature. Thus equation modeling of hydration behavior of grain has attracted considerable attention.

Absorption of moisture by food materials have been analyzed earlier by Fick's laws of diffusion with appropriate equations[8]. Since the laws of diffusion are complex and involve numerous functions and parameters, they are not convenient for practical computations under most situations[9]. To simplify the modeling of water absorption by food components a two-parameter, non-exponential empirical equation (Eq. (1)) was proposed by Peleg and became known as Peleg's equation[10]. Peleg's equation can be written as

$$M_t = M_o \pm \frac{t}{(k_1 + k_2 t)} \qquad (1)$$

Where M_t= moisture content (% dry basis) at a known time (t), Mo= initial moisture content (% dry basis)-, t = soaking time (hr), k_1= Peleg's rate constant ($h\%^{-1}$) and k_2 = Peleg's capacity constant ($\%^{-1}$). In the above equation "±" becomes "+" when the process is absorption, and for drying process it becomes "–". The equilibrium moisture content is given by

$$\text{As } t \rightarrow \infty \; M_E = M_o \pm 1/k_2 \qquad (2)$$

Where M_E = equilibrium moisture content (% dry basis)

The momentary sorption rate is given by the first derivative of equation (1)

$$\frac{dM_t}{dt} = \pm \frac{k_1}{(k_1 + k_2 t)^2} \qquad (3)$$

At the beginning i.e. when t = 0 Peleg rate constant relates to the sorption rate and equation (3) becomes

$$(dM_t/dt)_{t=0} = \pm 1/k_1 \qquad (4)$$

Peleg equation (Eq.1) can be transformed to a linear relationship in the form

$$\frac{t}{(M_t - M_o)} = k_1 + k_2 t \qquad (5)$$

By plotting $t/(M_t-M_o)$ against t, a straight line will be obtained having k_1 as intercept and k_2 as the gradient or slope of the line. The plot allows determination of characteristics of Peleg's constant. Peleg's equation is applicable to the curvilinear segment of the sorption curve and the reciprocal of k_2 can be used to predict the equilibrium moisture content[11] (Eq.2). Due to its simplicity Peleg's empirical formula has been widely used to demonstrate water absorption kinetics of cereal grains and legumes[2,4,6,9,12,13]. The objective of this work was to study the suitability of Peleg's equation in modeling of hydration characteristics of paddy grain during soaking at temperatures below gelatinization temperature and determination of thermodynamic parameter for hydration of paddy.

MATERIALS AND METHODS

Materials

Paddy grains were collected from local market. Then the grains were manually screened to remove foreign particles, chaff and broken kernels, immature and damaged seeds. Only paddy grains that were in good condition were selected for the experiment.

Moisture content determination

Moisture content of paddy grains was measured by standard air oven method.1 gm paddy grain was taken in aluminum cup and weighed, and then it was placed in hot air oven for moisture determination[14] at 130±3°C untill constant weight was obtained.

Measurement of physical properties

Linear dimensions of randomly selected 50 paddy grains were measured using micrometer (Mitutoyo Corporation, Japan.) having a resolution of 0.01mm. Geometric mean diameter (mm) was measured using the following formula[15].

$$D_{gm} = (LWT)^{1/3} \qquad (6)$$

where Dgm= geometric mean diameter, L,W,T are length, width and thickness in mm.

The sphericity (φ) is the ratio of the surface area of the sphere having the same volume as that of the grain to the surface area of the grain and it was determined using the relationship[15]

$$\varphi = (LWT)^{1/3}/L \qquad (7)$$

Surface area was measured using the following formula[16].

$$S = \frac{\pi B L^2}{2L - B} \qquad (8)$$

where $B = \sqrt{WT}$

Bulk density (kg/m^3) of grain was determined by filling the cylindrical container of 10 ml volume with the grains, then weighing the contents. The bulk density was calculated from the mass of the grain and the volume of the container. The grain density or true density (kg/m^3) was determined using toluene displacement method[17,18]. Toluene was used instead of water because it is absorbed by kernels to a lesser extent.

Porosity percentages were calculated from the difference between grain density and bulk density and divided by grain density and it was expressed by the following relationship[15].

$$\varepsilon = [(\rho_g - \rho_b)\,100]/\rho_g \qquad (9)$$

where ρ_g is grain or true density (kg/m^3) and ρ_b is bulk density (kg/m^3) of grain, ε is porosity (%).

Thousand grain weight was measured using electronics balance (Mettler Toledo) having a resolution of 0.02g.

Water absorption study

To study the hydration characteristics of paddy grains a definite amount of clean raw paddy were soaked into distilled water (1:10 w/v) at different temperature. The soaking temperature studied were 10°C, 20°C and 30°C. Paddy grains were taken in a glass beaker containing distilled water at the desired temperature. During hydration the samples were taken out at definite interval of time (initially 30 min, then 1 h and then 2 h). The soaked samples were placed on a filter paper to absorb the surface moisture and then weighed to an accuracy of ±0.02g. Moist grains were placed in an oven to determine moisture content. Gain in weight of paddy grain due to soaking was expressed in terms of moisture content (%dry basis). For equilibrium moisture content a separate set of sample was soaked for 24 hours.

The linearized form of Peleg's equation (Eq.5) as described before was used to model the hydration behavior of paddy grain under experimental conditions.

$$\frac{t}{(M_t - M_o)} = k_1 + k_2 t \qquad (5)$$

After evaluating two Peleg's constant in each temperature for paddy grain correlations between them and temperature were established.

Statistical analysis

All the experiments were conducted in triplicate and mean values were reported. Data obtained from linear regression analysis of Peleg's equation were subjected to Student "t" test (Origin 6.1) and means were considered significantly different when $P < 0.05$.

RESULTS AND DISCUSSION

Physical properties

Average values of physical properties like linear dimensions, geometric diameter, sphericity, surface area, porosity are given in TABLE 1. The average values of length; width and thickness are 9.5±0.5mm, 2.21±0.11mm and 1.75±0.25mm respectively at a moisture content of 14.3% (dry basis) which was the initial moisture content. These results are comparable with the earlier reports[1,19]. Porosity of paddy grains was found to be 57.89%. Sphericity of paddy grain was 0.349±0.039. True density and bulk density were 1228±2 kg/m^3 and 517±3 kg/m^3 respectively. These values are in agreement with the earlier report for paddy grain[1,18], and lupin seeds[4].

Water absorption phenomena

Hydration behavior of paddy grain in terms of moisture content (% dry basis.) at different soaking time and temperatures is represented in Figure 1. The rate of water absorption was higher initially at all three soaking temperatures after that water absorption rate decreased

TABLE 1 : Physical characteristics of paddy grain

Parameters	Value±SD[1]
a) Length (L) (mm)	9.5±0.5
b) Width (W) (mm)	2.21±0.11
c) Thickness (T) (mm)	1.75± 0.25
d) Sphericity (φ) = [(L.W.T) ⅓]/ L	0.349±0.039
e) Surface area (mm^2)	32.80±0.2
f) 1000 grain weight (g)	30.75±0.25
g) Bulk Density (kg/m^3)	517±3.00
h) Density (kg/m^3)	1228±2.00
i) Porosity (ε) (%)	57.89±0.11

[1]Datas are mean of 3 replicate observations

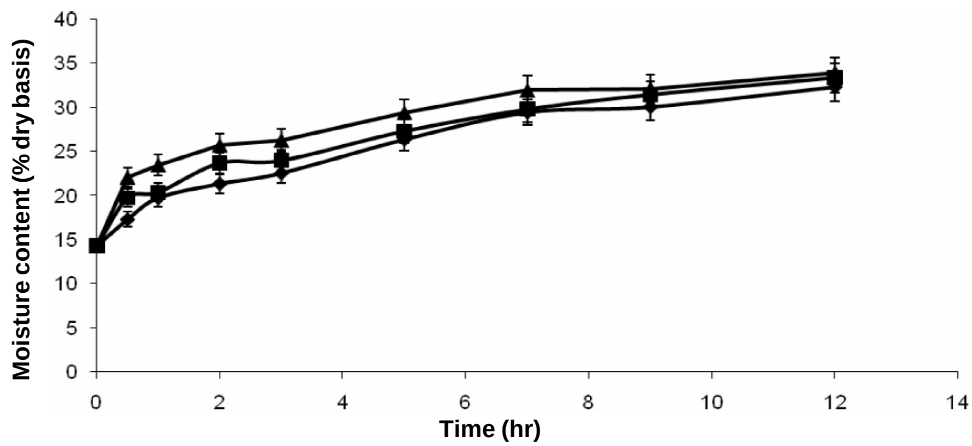

Figure 1 : Water absorption curve during soaking of paddy grain (▲30ºC, ▪20ºC, ♦10ºC) (5% error bar has been adjusted)

gradually as the moisture content approaches to saturation. The water uptake rate depends on the difference between moisture content at equilibrium and at a given time. With the progression of hydration process this difference decreases and as a consequence the water absorption rate also decreases. The soaking temperature influences the rate of absorption of moisture. The higher the soaking temperature (10-30°C) the higher was the rate of water absorption. This phenomenon can be linked to high rate of water diffusion at higher temperature[4]. Similar observations have been reported for cereal grains and legumes[1,4,6,12,13].

Modeling of hydration isotherms

The moisture absorption data under the specific experimental conditions was fitted to Peleg's equation (Eq.5) (Figure 2). Estimated Peleg's constants from the linear regression analysis are shown in TABLE 2. The correlation coefficients R^2 (TABLE 2) varied from 0.966 to 0.985. This established the sufficiency of the equation for describing the hydration kinetics of paddy grains within the studied temperature range.

Determination of Peleg rate constant k_1

Peleg rate constant (k_1) is linked with mass transfer rate. Sopade and Obekpa and Sopade *et al*. observed that k_1 was inversely related to temperature and its reciprocal defines the initial hydration rate[20,21] (Eq.4). In the present study the magnitude of k_1 showed a sta-

TABLE 2 : Values of Peleg's constants and correlation coefficient R^2 for hydrated paddy grains

Temperature (°C)	$k_1(h\%^{-1})$	$k_2(\%^{-1})$	R^2
10	0.180	0.0422	0.966
20	0.127	0.0445	0.965
30	0.076	0.0465	0.985

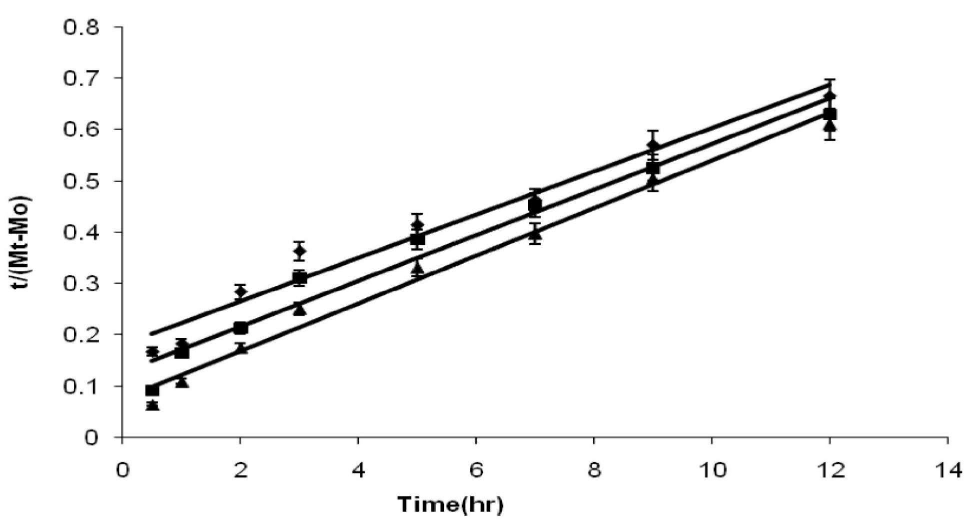

Figure 2 : Fitting of Peleg's model to water absorption data during soaking of paddy grain (▲30ºC, ▪20ºC, ♦10ºC) (5 % error bar has been adjusted)

tistically significant ($P<0.05$) decreasing trend (from 18×10^{-2} h%>$^{-1}$ to 7.6×10^{-2} h%>$^{-1}$) with the increase in soaking temperature from 10ºC-30ºC (TABLE 2). This indicates increase in water absorption rate with increase in soaking temperatures. This trend is in agreement with the previous study[6,12,13,22].

k_1 could be compared to a diffusion coefficient and the Arrhenius type equation (Eq.10) could be used to describe the temperature dependence of the reciprocal of Peleg's constant k_1[23]. This equation has been used previously to study the hydration behavior of different cereal grains and legumes[4,9,23].

$$\ln\left(\frac{1}{k_1}\right) = \ln k_o - \frac{Ea}{RT} \qquad (10)$$

where k_o is a constant (h%$^{-1}$), E_a is activation energy (Jmol^{-1}), R is universal gas constant (8.314 Jmol^{-1}K^{-1}) and T is the absolute temperature (K).

Arrhenius plot (Figure 3) with R^2 value 0.988, confirmed the temperature dependence of Peleg rate constant. Activation energy calculated from the slope of the plot (Figure 3) was found to be 35.83 kJ/mol. The reported value of activation energy of paddy grain was higher than rice[5], wheat[9] and sorghum[6], but lower than the value obtained for legume seed[4].

Determination of Peleg capacity constant k_2

Peleg showed that k_2 is linked with maximum water absorption capacity or equilibrium moisture content such that lower magnitude of k_2 indicate higher water absorption capacity and vice versa[10]. Peleg capacity constant, k_2 for paddy grain increased (from 4.22×10^{-2} %$^{-1}$ to 4.65×10^{-2} %$^{-1}$) significantly ($P<0.05$) with increase in temperature from 10ºC to 30ºC. The k_2 values (TABLE 2) showed that water absorption capacity declined with increase in temperature. This findings are in accordance with the earlier report for legume seeds[2,4,22]. The decreasing trend of k_2 with increasing temperature was reported for wheat[9], amaranth[13] and sorghum[6] etc. However other studies have shown that k_2 remains unaffected by temperature[20,23].

Like k_1 the temperature dependence of k_2 have also studied using Arrhenius type equation (Eq.11) and the Arrhenius plot (Figure 4) having R^2= 0.986 successfully characterized the Peleg capacity constant.

$$\ln\left(\frac{1}{k_2}\right) = \ln C + D\left(\frac{1}{T}\right) \qquad (11)$$

where C and D are the constants having values of 4.12 and 500 %K respectively.

Thermodynamic consideration

The value of activation energy (Ea) obtained from Eq.10 has been applied for determination of different thermodynamic parameters according to the expression[24].

$$\Delta H^* = E_a - RT \qquad (12)$$

$$\Delta S^* = R\left(\ln A - \ln\frac{k_b}{h_p} - \ln T\right) \qquad (13)$$

$$\Delta G^* = \Delta H^* - T\Delta S^* \qquad (14)$$

where ΔH^* is change in enthalpy, ΔG^* is change in free energy, ΔS^* is change in entropy R is universal gas constant, ln A is the ordinate intersection when regression analysis is applied to the plot obtained in calculation of E_a, k_b is Boltzman constant (1.38×10^{-23}JK^{-1}) h_p is Planck constant (6.626×10^{-34}Js) and T is absolute temperature.

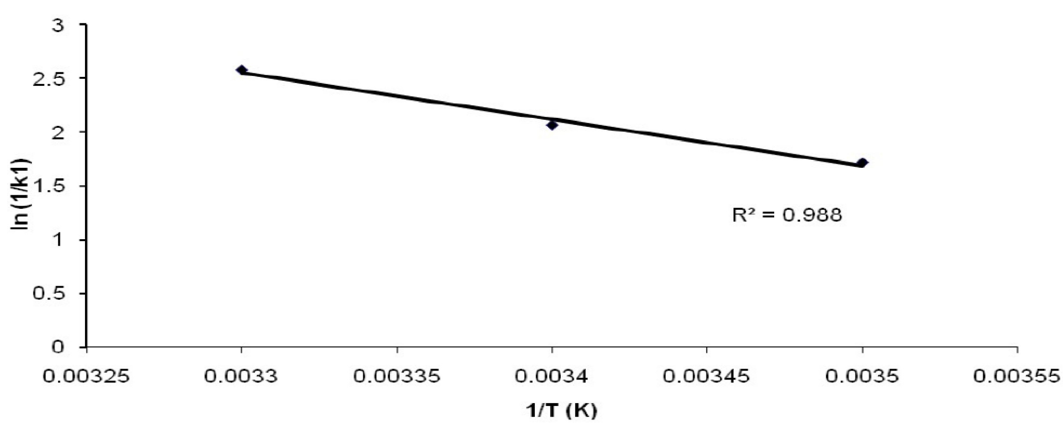

Figure 3 : Arrhenius plot of ln ($1/k_1$) vs. (1/T)

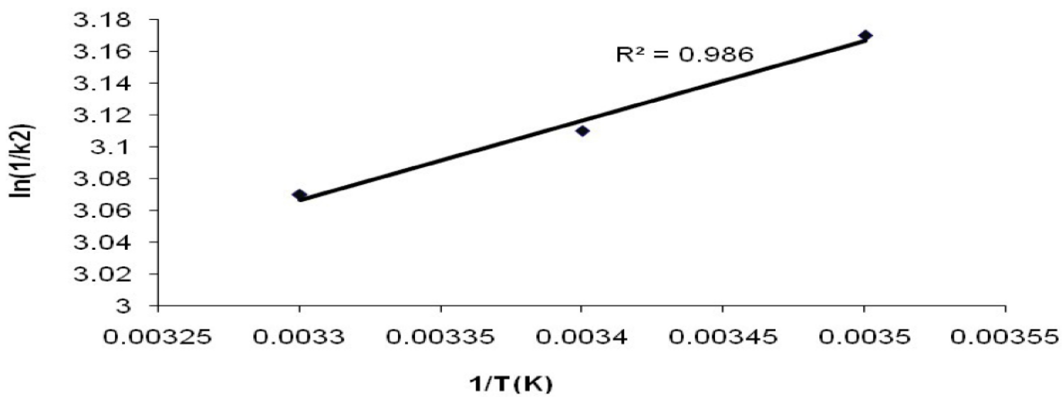

Figure 4 : Arrhenius plot of ln(1/k_2) vs.(1/T)

Values of these parameters are represented in TABLE 3. The negative values of enthalpy (ΔH*) indicate that changes during hydration of paddy were associated with exothermic and energetically favorable transformation[25]. The positive value of ΔG* indicates an enderogonic reaction that requires an input of energy from the surroundings[25]. Higher value of free energy of activation (ΔG*) (64.66kcal/mol at 30ºC) indicates that hydration was influenced by temperature. However the negative values of entropy (ΔS*) indicate an increase in system order and this is entropically unfavorable. Negative entropy of activation occurs when degrees of freedom of rotation or translation are lost during the formation of the activated complex[26].

TABLE 3 : Thermodynamic parameters for hydration of paddy grains

Temperature (°C)	ΔH* (cal/mol)	ΔS* (cal/kmol)	ΔG* (kcal/mol)
10	-2317.03	-221.04	60.32
20	-2400.17	-221.32	62.45
30	-2483.31	-221.60	64.66

CONCLUSIONS

The present study has shown the adequacy of Peleg's equation to analyze the hydration characteristics of paddy grains (*Oryza sativa*) during the hydration process at different soaking temperature, ranges from 10ºC to 30ºC. This model was also used to predict the moisture content at given soaking time and temperature within the experimental condition. Peleg's constant k_1, decreased with increasing temperature, indicated that water absorption rate increased with rise in temperature, where as reduction in water absorption capacity was observed as k_2 increased with increasing temperatures. The temperature dependence of Peleg's constants (k_1, k_2) was described by using Arrhenius type equation. The activation energy was used to study the thermodynamic parameters which in turn indicate that the changes during the hydration of paddy grain were associated with exothermic and energetically favorable transformation and the soaking procedure was a less random system. Therefore Peleg's empirical model could be used by the paddy grain processor to predict the soaking characteristics as well as hydration condition of paddy grain could be optimized.

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