#### Computers and Electronics in Agriculture 123 (2016) 40-48

Contents lists available at ScienceDirect

Computers and Electronics in Agriculture

journal homepage: www.elsevier.com/locate/compag

## Original papers Effective moisture diffusivity in wheat kernels during adsorption

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#### ARTICLE INFO

Article history Received 24 June 2015 Received in revised form 13 February 2016 Accepted 14 February 2016 Available online 26 February 2016

Keywords: Grain Moisture diffusion Aeration Modeling

#### ABSTRACT

Wheat samples with low (4.97-6.08%), normal (10.11-11.04%), and high (20.7-22.72%) initial moisture content (IMC, wet basis) were used to determine the rate of moisture adsorption by the gravimetric method at 10, 20, 25, 30, and 35 °C under 65%, 86%, and 100% relative humidity (RH), respectively. A moisture diffusion equation was modified to fit the relationship between the moisture ratio of samples and exposure time. From 65% to 100% RH, the IMC of wheat samples was inversely related to the moisture adsorption rate at temperatures from 10 to 35 °C. Moisture adsorption rates of samples increased with increasing temperatures. The moisture adsorption rate of samples with the same IMC increased with increasing RH at a given temperature. A single wheat kernel was considered geometrically to be a finite homogeneous slab shape, and the analytical solution of the partial differential equation for moisture diffusion was given. The effective moisture diffusivity was calculated using the slope method by plotting the experimental data in terms of ln (MR) versus rewetting time. In the range of 10–35 °C, the effective moisture adsorption diffusivity of wheat kernels with normal moisture was  $1.681\times 10^{-8}\text{--}1.516\times 10^{-7}$  $m^2 h^{-1}$ , and their activation energy was 23.651–28.434 kJ mol<sup>-1</sup>. For the same IMC, the effective moisture diffusivity of wheat kernels tended to increase with increasing temperature at a given RH, but decreased with an increase in RH at a given temperature. Activation energy and the pre-exponential factor of Arrhenius equation  $(D_0)$  tended to decrease with increasing RH. With similar initial moisture content, winter wheat and spring wheat exhibited similar effective moisture diffusivity.

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#### 1. Introduction

Agricultural products are typically at their highest quality immediately after being harvested and cleaned. Approximately one-half of the world's cereal grain production (i.e., one billion metric tons, USDA, 2007) goes into storage after harvest, where significant quality loss may occur (White, 1995). Post-harvest losses range from 9% in North America to 50% in developing countries (FAO, 2000). In recent years, computer simulations of grain storage have become a tool with which to improve grain storage procedures and reduce storage losses (Jayas, 2002; Lopes et al., 2006; Neethirajan and Jayas, 2007; Lawrence and Maier, 2011). However, simulating the storage of grain is hampered because of insufficient data to predict moisture adsorption rates and effective diffusion coefficients of grains during storage. Few studies in the literature provide data on moisture adsorption rates for important grains, such as corn (Muthukumarappan and Gunasekaran, 1990), soybeans (Osborn et al., 1991), and rice (Lan and Kunze, 1996). There is insufficient information to model moisture adsorption rates of

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wheat within the range of typical storage conditions. Babbitt (1949) provided some qualitative information regarding moisture adsorption by wheat kernels, but only determined constants for the spherical diffusion equation, with negligible external resistance for one set of storage conditions (25 °C, 75% relative humidity [RH]). Aldis and Foster (1980), Duggal et al. (1982), and Versavel and Muir (1988) presented some moisture adsorption information for wheat kernels and spikes, but did not include the needed adsorption rates for kernels. Casada (2002) adopted a spherical diffusion equation and cylindrical diffusion equation to describe wheat adsorption data under six test conditions (15 °C, 58% RH; 15 °C, 78% RH; 15 °C, 85% RH; 30 °C, 50% RH; 30 °C, 71% RH; 30 °C, 78% RH), but did not compare moisture diffusivity values of different wheat types. Moreover, no study has addressed moisture adsorption rates and diffusion coefficients of Chinese wheat under various storage conditions.

Considerable data are available on rates of grain moisture desorption during drying. Studies regarding the storage of grain and grain-type products mainly rely on moisture desorption data to describe moisture transfer, although moisture transfer during storage involves both moisture desorption and moisture adsorption in the kernel (Jayas, 2002; Casada, 2002). If moisture adsorption of grain kernels occurs at different rates than that of desorption, using









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desorption data alone for grain storage models will likely produce inaccurate predictions (Casada, 2002). Wheat is the major grain in China, with an annual production of approximately 100 million metric tons in recent years. A portion of wheat in China is stored longer (3–5 years) than in other developed countries, with deterioration curtailed largely through the control of moisture content and temperature. In order to maintain wheat quality during the storage period, it is important to understand the adsorption rate and effective diffusion coefficient of wheat under various storage conditions, with the goal of preventing moisture loss from wheat kernels during aeration in the autumn and winter seasons.

#### 2. Methodology

#### 2.1. Sample preparation

We used two spring wheat varieties, "Dan 2763" and "Jiangsu Chun", which are hard red grains, and two winter wheat varieties "Zhongmai" and "Zhoumai", which are mixed white grains. "Dan 2763" and "Jiangsu Chun" varieties were dehydrated to 4.97% and 6.4% wet basis (w.b.), respectively, using solid  $P_2O_5$  at 40 °C. "Zhongmai" and "Zhoumai" varieties with normal moisture were 10.1% and 11.0% w.b. and were dehydrated to 6.08% and 6.02% w. b., respectively, using solid  $P_2O_5$ . Samples of "Zhongmai" and "Zhoumai" were prepared with 22.7% and 20.7% w.b., respectively, by adding distilled water, equilibrating for two weeks in the refrigerator (4 °C), and shaking once every day.

## 2.2. Determination of changes in wheat kernel moisture with time during adsorption

Changes in sample moisture over time were determined by the static gravimetric method (Li et al., 2011). At each of five constant temperatures (10, 20, 25, 30, and 35 °C), two saturated salt (NaNO<sub>2</sub> and K<sub>2</sub>CrO<sub>4</sub>) solutions and distilled water were used to maintain constant vapor pressure for a RH of 65%, 86%, and 100%, respectively. Three wide-mouth glass bottles (250 mL) contained 65 mL salt solution, and were kept in a temperature-controlled cabinet to maintain the three relative humidity (RH) levels. Each relative humidity value for each temperature was used in triplicate and a total of forty-five bottles were used in an experiment for one wheat sample with the same initial moisture content (IMC). The temperature of the cabinets was monitored using a standard thermometer and controlled with an accuracy of ±0.5 °C. Each sample of wheat seeds (ca. 5.0000 g) was placed into a small bucket (3 cm diameter  $\times$  4 cm length) made from copper wire gauze, and hung in the glass bottle on a copper wire hook under the rubber plug, approximately 2-3 cm above the saturated salt solutions. The rubber plug sealed the mouth of the bottle. After exposure to the saturated vapor began, the copper wire buckets with samples were weighed every 2 h for 24 h, and thereafter every 4 h until the change in mass between two successive readings was less than 2 mg. When the sample was exposed to a lower temperature, it took longer to equilibrate. However, wheat seeds exposed to saturated K<sub>2</sub>CrO<sub>4</sub> and distilled water solution for 10–12 days at higher temperatures were susceptible to mold growth, and samples were removed immediately if mold was observed on any seed. The moisture content of the samples the equilibration stage was defined as the equilibrium moisture content (EMC) and two samples of the triplicate were determined by the oven method. The sample was dried to constant weight at  $103.0 \pm 0.5$  °C for 24 h.

#### 2.3. Determination of length and width of wheat kernel

The length and width of wheat kernels was determined by a quality detection instrument for rice (JMWT12, Satake). Approximately 100 kernels were used for length and width determination at 65% and 86% RH for each of five temperatures. For samples at 100% RH, whole kernels were used for length and width determination after moldy kernels were discarded.

#### 2.4. Determination of moisture sorption rate of wheat kernels

It is generally agreed that moisture flow within a grain kernel occurs by diffusion (liquid and/or vapor). Partially coupled heat and mass transfer equations have been solved for an isotropic sphere with constant material properties, and coupled equations with varying material properties were used to study the drying of barley, soybeans, and corn kernels (Haghighi et al., 1990; Irudayaraj et al., 1993). Coupling effects of moisture and temperature, although important for accurately modeling desorption (Parti, 1993), is not important for adsorption because the adsorption process takes much longer (48–50 h) than the desorption process (6–10 h). The diffusion equations have been largely based on the simplifying assumption that grain kernels are homogenous. Page (1949) altered the exponential equation by adding an exponent to the time variable to improve the fit of corn drying data, which yielded

$$MR = \exp(-kt^n) \tag{1}$$

$$MR = (M_t - M_e) / (M_0 - M_e)$$
(2)

where MR is the average moisture ratio of the material at any given time (decimal);  $M_t$  is the average moisture of the material at any given time (t) (decimal wet basis);  $M_e$  is the equilibrium moisture content (decimal wet basis);  $M_0$  is the initial moisture content (decimal wet basis); t is the time from beginning of the process (h); and k and n are product specific constants.

We provide the modified form of Page's equation as

$$MR = a \exp\left[-kt^n \times \exp(-b/(\theta + 273))\right]$$
(3)

where  $\theta$  is temperature (°C), and *a*, *b*, *k*, and *n* are equation constants.

From Eq. (3) we obtain

$$\frac{d(M_t)}{dt} = (M_0 - M_e) \times a \times \exp(-kt^n \exp(-b/\theta)) \times (-k \times n)$$
$$\times t^{n-1} \exp(-b/\theta)$$
(4)

where  $d(M_t)/dt$  is the moisture sorption rate of grain kernels  $(10^{-5} h^{-1})$ .

Changes in the average moisture ratio of wheat kernels with time, given the combination of temperature  $(10-35 \,^{\circ}\text{C})$  and RH (65–100%), were fitted to Eq. (3) for samples with different initial moisture content using the non-linear regression procedure in SPSS 13.0 for Windows (SPSS Inc., 2006), which minimizes the sum of squares of deviations between experimental and predicted data in a series of iterative steps. The goodness-of-fit of Eq. (3) was evaluated using the determination coefficient ( $R^2$ ), residue sum of squares (RSS), the standard error (SE), and mean relative percentage error (MRE). The  $R^2$  was one of the primary criteria for selecting the best equation to fit the experimental data. In addition to  $R^2$ , the other statistical parameters, MRE (as a percentage), RSS, and SE were used to determine the quality of the fit. Eqs. (5)–(8) were used for calculating  $R^2$ , RSS, SE, and MRE, respectively.

$$RSS = \sum_{i=1}^{n} (m_i - m_{pi})^2$$
(5)

$$SE = \sqrt{\sum_{i=1}^{n} (m_i - m_{pi})^2 / (n-1)}$$
(6)

$$R^{2} = 1 - \sum_{i=1}^{n} (m_{i} - m_{pi})^{2} / \sum_{i=1}^{n} (m_{i} - m_{mi})^{2}$$
<sup>(7)</sup>

$$MRE\% = 100|m_i - m_{pi}|/n$$
(8)

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