The Hygroscopic Properties and Sorption Isosteric Heats of Different Chinese Wheat Types

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Abstract

The moisture sorption isotherm data of fourteen Chinese wheat varieties were determined using the static gravimetric method at five different temperatures (10, 20, 25, 30 and 35 °C) and relative humidity ranging from 11.3 to 96%. Eight models, namely Brunauer-Emmett-Teller, CAE, Chen-Clayton, Modified-Chung-Pfost (MCPE), Modified-Henderson, Modified-Guggenheim-Anderson-deBoer, and Modified-Oswin and Strohman-Yoerger, were used to fit the sorption data. MCPE shows the best fitting results. A significant hysteresis effect was found between wheat desorption and adsorption isotherm at lower ERH, but the similar hygroscopic properties remained for different wheat types like hard vs. soft, red vs. white, and winter vs. spring, respectively. The experimental results show that the isosteric heats for both wheat adsorption and desorption, and all the sorption heats for different wheat types decrease rapidly with increasing seed moisture initially, however, after the moisture is more than 15% w.b. they decrease tardily with increasing moisture content. The isosteric heats of wheat desorption were considerably higher than those of adsorption below 17.5% m.c., but the similar sorption isosteric heats were found for wheat types like hard vs. soft, red vs. white, or winter vs. spring, respectively. It is concluded that the wheat grains from different types have similar hygroscopic properties and sorption isosteric heats and can be synchronously dealt with during physical control in storage.

Keywords: Wheat (*Triticum aestivum*), Hygroscopicity, Sorption, Hysteresis effect, Isosteric heat of sorption, Thermodynamics

1. Introduction

Wheat (*Triticum aestivum*) is the major grain in China, with its annual production being around 100 million metric tons in recent years. In China, a portion of the wheat is stored for a longer period of time (3 to 5 years) than that in the developed countries with deterioration controlled largely through moisture content and temperature. In order to maintain the quality of the wheat during this storage time period, it is important and interesting to know the relationship between the storage condition (temperature and humidity) and the quality of the wheat. In other words, knowing the relationship between equilibrium moisture content (EMC) and equilibrium relative humidity (ERH) of the cereal grains is essential. This relationship has been widely studied and different models/equations have been introduced (Nellist & Dumont, 1979; Van den Berg & Bruin, 1981; Sun & Woods, 1994; Blahovec, 2004; de Carvalho Lopes et al., 2006). Among 77 isotherm equations compiled by Van den Berg & Bruin (1981), only ten equations are commonly used to fit EMC-ERH relationship for wheat data (Sun & Woods, 1993). Among these ten equation, Chen-Clayton (CCE), Day-Nelson, Henderson, Modified Chung-Pfost (MCPE), Modified Henderson (MHE), and Strohman-Yoerger (SYE) equations are used to describe the sorption behavior of wheat chaff and unthreshed kernels in wheat heads (Duggal, Muir & Brooker, 1982). Based on the results, it was known that among these equations, the smallest residual sums of squares of ERH were obtained using the SYE for wheat kernels and the CCE for chaff, and the minimum standard error of estimate of ERH by the MCPE, respectively. All these conclusions were made based on the study of one or two source sets of wheat data. Nellist & Dumont (1979) collected wheat data from thirteen sources and fitted the data using five common equations in order to obtain isotherm equations for general drying applications. This work indicated that the MCPE results in the best fit. As the authors mentioned, if more and better data became available, the coefficients of MCPE equation should be updated. Sun & Woods (1994) analyzed 29 source sets of wheat EMC/ERH data using five models (i.e. CCE, MCPE, MHE, Modified-Oswin (MOE), and SYE). It was concluded that the MCPE and the MOE are the preferred equations due to the fact that these two equations can describe each and all the individual data set and are three-coefficient invertible equations. Understandingly, the variety of the wheat has some influence on the results and all these studies did not include the Chinese wheat varieties. In this paper, the moisture sorption isotherm data of fourteen Chinese wheats are reported.

In respect of kernel hardness, wheat is divided into several types: durum, hard, soft, and mixed wheat residing between soft and hard wheat. Pfost et al. (1976) used MCPE analyzed the EMC/ERH data for hard, soft, and durum wheats. The results indicate that there is some variation in the resulting coefficients of the MCPE among these three types of wheat. Sun & Woods (1994) compared the MCPE curves for the three types of wheat (eight hard wheat varieties, six soft wheat varieties, and three durum wheat varieties) at three temperatures of 0, 30 and 60 °C. At the temperature of 0 °C, the hard-wheat curve lies well above the others and the durum wheat results in the lowest curve. At 30 °C the difference in the hygroscopic properties of the three types of wheat become very small due to the three curves coming closer. However at 60 °C, these three curves depart from each other again in the reverse manner, the curve for durum wheat lying above the curve for soft wheat, with the curve for hard wheat at the bottom. Recently, we studied the moisture sorption data of some Chinese wheat varieties (one mixed, two hard, two soft wheats). By using six equations, the Brunauer-Emmett-Teller (BET), CCE, MCPE, MHE, MOE and SYE, it is found that these hard and soft wheat varieties show similar hygroscopic properties and sorption isosteric heats (Li et al., 2011). In the literature, few reports are about the hygroscopic properties and sorption isosteric heats between winter and spring wheat, as well as between red and white wheat.

Thermodynamic, structural and dynamic approaches have been used to understand the properties of water and calculate the energy requirements of heat and mass transfer in biological systems (Fasina, Ajibola & Tyler, 1999). Thermodynamic functions/variables of the isotherm sorption deepen the understanding of experimental results. These thermodynamic functions/variables include isosteric heat of sorption, integral enthalpy, and integral entropy. The knowledge of sorption isotherms at different temperatures enables an evaluation of the heat of sorption, which determines the interaction between an adsorbent and adsorbate. All these provide a guideline for the drying process (Iglesias, Chirife & Viollaz, 1976). The level of material moisture content at which the net isosteric heat of sorption approaches the latent heat of vaporization of water is often taken as an indication of the amount of 'bound water' existing in the product (Kiranoudis et al., 1993). The heat of vaporization of sorbed water may increase to values well above that for the vaporization of pure water as food is dried to low moisture levels (Rizvi, 1986).

In this paper, a systematical study of 14 Chinese wheat varieties collected from different regions is reported. The EMC/ERH data is analyzed using eight equations in order to determine the most suitable EMC/ERH model for grain moisture sorption isotherms of fourteen Chinese wheat varieties, and compare the fitted sorption isotherms and isosteric heat of water sorption between different wheat types classified in respect of hardness, color and seedtime, providing theoretic basis for wheat treatments after harvest.

2. Materials and Methods

2.1 Wheat samples and experimental procedures

Fourteen varieties of wheat (*Triticum aestivum*) used in this work were collected from eight regions in China in 2007 and 2008. These include twelve winter wheat and two spring wheat as shown in Table 1. The hardness of wheat samples were measured with an SKCS 4100 (Perten Instruments AB, Sweden). Of these fourteen varieties, seven varieties are hard wheat, five varieties are soft wheat, and the remaining two varieties are mixed type wheat, or seven varieties are white wheat, the others are red wheat. The wheat seeds used for this study were intact, clean and plump. For adsorption experiment, the wheat seeds were dried to a wet bulb moisture content (m.c.) of 7-8% wet basis (w.b.) at 40.5 °C in an oven, and then dehydrated by P_2O_5 solid in a dessicator to below 5% w.b. For the samples of desorption experiment, the wheat seeds were re-moisturized to the m.c. of 23% w.b., and equilibrated at 4 °C for two weeks.

The static gravimetric method, with nine saturated salt solutions to maintain constant vapor pressure (Jayas & Mazza, 1991; Li et al., 2011), was used to obtain nine equilibrium moisture contents at each of five constant temperatures (10, 20, 25, 30, and 35 °C). The saturated salt solutions included lithium chloride, potassium acetate, magnesium chloride, potassium carbonate, magnesium nitrate, cupric chloride, sodium chloride, potassium chloride, and potassium nitrate. Twenty-seven wide mouth glass bottles (250-mL) each contained 65 mL salt solution, and were kept in one temperature controlled cabinet to maintain nine groups of different relative humidity (r.h.) levels ranging from 11.3 to 96% ERH. Every relative humidity at one temperature was triplicated and a total of 135 bottles was used in the experiment for five sorption isotherms of a wheat variety. The temperature of cabinets was monitored using a standard thermometer and controlled with an accuracy of ±0.5 °C. Each sample of wheat seeds (4-5 g) was placed into a small bucket (3 cm diameter × 4 cm length) made from copper wire gauze, and hung into the glass bottle on a copper wire pothook under a rubber plug, 2-3 cm above saturated salt solutions. The rubber plug was tightly pushed into the bottle mouth. From three weeks after exposing the samples in the saturated vapour at 35 °C, the copper wire buckets with samples were weighed every other day until the change in mass between two successive readings was less than 2 mg. When the sample was exposed to a lower temperature, the sample was left longer to equilibrate. However, the wheat seeds exposed over the saturated potassium nitrate solution for 3-4 days at higher temperatures were susceptible to molds growth, and removed immediately mould was observed on any seed. The moisture content of the sample at this constant stage was defined to be the EMC and was determined by the oven method (AOAC, 1980). The sample was dried to constant weight under 103.0±0.5 °C for 22-28 h.

2.2 Analysis of the adsorption and desorption data

Eight equations were used to fit the EMC data of wheat adsorption and desorption as given in Table 2. The fitting was conducted 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 determination coefficient (R^2), residue sum of squares (RSS), the standard error (SE), and mean relative percentage error (MRE) as defined below are used as the criteria to determine the best equation for the data analysis.

$$R^2 = \sqrt{1 - \frac{\sum_{i=1}^{n}(m_i - m_{pi})^2}{\sum_{i=1}^{n}(m_i - m_{mi})^2}} \tag{1}$$

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

$$SE = \sqrt{\frac{\sum_{i=1}^{n}(m_i - m_{pi})^2}{(n-1)}} \tag{3}$$

$$MRE = \frac{100\sum_{i=1}^{n}\left|\frac{m_i - m_{pi}}{m_i}\right|}{n} \tag{4}$$

Where m_i is the experimental value, m_{pi} the predicated value, m_{mi} the average of experimental values, and n the number of observations. The determination coefficient (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. The equations (1) - (4) were used for calculating R^2, RSS, SE, and MRE, respectively. The fit of an equation is good enough for practical purposes when MRE is less than 10% (Aguerre, Suarez & Viollaz, 1989).

2.3 Determination of the isosteric heat of sorption

The total energy required to remove a unit mass of water from wheat kernels, i.e. the differential heat of sorption (h_s), is conveniently partitioned into two components, namely the latent heat of vaporization of free water (h_v) and the differential heat of wetting (h_w). The h_s of adsorption and desorption of wheat grains were respectively calculated by the following six equations according to Thorpe (2001).

$$\frac{h_s}{h_v} = 1 + \frac{p_s}{r.h.} \times \frac{dT}{dP_s} \times \left. \frac{\partial r.h.}{\partial T} \right|_{m.c.} \tag{5}$$

$$h_v = 2501.33 - 2.363 \times t \tag{6}$$

$$P_s = \frac{6 \times 10^{25}}{(273.15 + t)^5} \times \exp\left(-\frac{6800}{t + 273.15}\right) \tag{7}$$

$$\frac{dP_s}{dT} = \frac{P_s}{(t + 273.15)} \times \left(\frac{6800}{t + 273.15} - 5\right) \tag{8}$$

$$\left. \frac{\partial r.h.}{\partial T} \right|_{m.c.} = \frac{C_1 \times r.h.}{(t + C_2)^2} \times \exp(-C_3 \times m.c.) \tag{9}$$

$$\left. \frac{\partial r.h.}{\partial T} \right|_{m.c.} = \frac{-1}{\left\{1 + \left(\frac{C_1 + C_2 \times t}{m.c.}\right)^{C_3}\right\}^2} \times \left\{ \frac{C_2 \times C_3}{m.c.} \times \left(\frac{C_1 + C_2 \times t}{m.c.}\right)^{(C_3 - 1)} \right\} \tag{10}$$

The equation (5) enables one to calculate h_s/h_v, provided dP_s/dT and $\partial r.h./\partial T|_{m.c.}$ can be evaluated by equations (8) and (9), respectively. The h_v of free water in equation (6) is dependent on temperature. The saturated vapor pressure, P_s, can be calculated by equation (7). The derivative of *r.h.* with respect to t, $\partial r.h./\partial T|_{m.c.}$ depends on the sorption isotherm equation used, and the Modified Chung-Pfost (MCPE) in equation (9), or Modified Oswin (MOE) in equation (10) used in this study.

3. Results

3.1 Fitting of sorption equations to experimental sorption data

The results of fitting the sorption equations to the experimental data of adsorption and desorption isotherms by nonlinear regression analysis were respectively evaluated with the statistical indices such as RSS, SE, R^2 and MRE. Of eight equations, namely BET, CAE, CCE, MCPE, Modified Guggenheim-Anderson-deBoer (MGAB), MHE, MOE, and SYE (Table 2), seven equations such as CAE, CCE, MCPE, MGAB, MHE, MOE, and SYE gave the better fit to the experimental data of adsorption and desorption isotherms in a wide range of 11.3 to 96.0% ERH, but the BET equation gave the better fit in the range of 11.3 to 49.9% ERH. The further comparisons of the sorption equations in a form of $r.h. = f(M, t)$ or $M = f(r.h., t)$ for twenty-eight sets of isotherm data were given in Table 3. The average values of R^2 and error parameters (RSS, SE, and MRE) were calculated for the twenty-eight sets of isotherm data. In the form of $r.h. = f(M, t)$, the equations for desorption were ranked for accuracy in an order: CAE, SYE, MCPE, MOE, CCE, MHE and MGAB, but for adsorption the order was: CAE, SYE, MCPE, CCE, MHE, MOE and MGAB. The CAE model being used in Chinese stored grain aeration gave the least standard error of estimate, least mean relative percentage deviation, and explained variation on the ERH, thus it could be taken as the best model among the seven $r.h. = f(M, t)$ models because the residual plots showed a random deviation. In case of a form of $M = f(r.h., t)$, the equations for desorption were ranked in an order: MCPE, CCE, BET, MHE, MOE, and MGAB, the order for adsorption equations were MCPE, CCE, MHE, BET, MOE, and MGAB. However, CAE is five-coefficient, temperature dependent equation, and it can be not easily inverted to give EMC as a function of ERH. SYE is four-coefficient, temperature independent equation, and also cannot be explicitly invertible. CCE is a four-coefficient, temperature dependent and explicitly invertible equation. The other commonly used equations, such as MCPE, MHE, MOE and MGAB all are three-coefficient, temperature dependent and easily invertible equations (Table 2). MCPE fitted the data reasonably well. MHE, MOE and MGAB equations were again less effective in fitting the data. Thus, the MCPE in a form of $r.h. = f(M,t)$, or $M = f(r.h.,t)$ was considered to best describe the equilibrium moisture data of fourteen wheat varieties in a wide range of 11.3 to 96.0% ERH.

3.2 Comparison of hygroscopic properties of different wheat samples

The best fitted MCPE parameters for desorption and adsorption isotherms of different wheat samples were summarized in Table 4. For MCPE model in a form of $r.h. = f(M,t)$, the parameters C_1 and C_2 in adsorptive isotherm equation were significantly different from those corresponding to desorptive isotherm equation, respectively. In contrast, there was slight difference in corresponding parameters C_1, C_2 and C_3 between hard and soft wheat, as well as between red and white wheat, or between winter and spring wheat, respectively.

The experimental sorption data of wheat samples were fitted with MCPE and the predicted data were compared between wheat types. Figure 1 shows the fitted sorption isotherms of 14 data sets at 20 and 30 °C. The isotherms of desorption and adsorption for wheat samples were sigmoidal in shape. At a constant ERH, both types of EMC decreased with an increase in temperature. A substantial difference was observed between the adsorption and desorption data at the same temperature. The desorption data was higher than the adsorption data except at high *r.h.*, and the moisture sorption hysteresis effect was more significant at lower ERH. Both width and span of the hysteresis effect tended to decrease with an increase in temperature.

Figure 2 compared the predicted sorption isotherms between wheat types at 20 and 30 °C, respectively. The moisture sorption data of soft wheat were insignificantly higher than those of hard wheat at these two temperatures. The very similar moisture sorption data were also observed between red and white wheat, as well as between winter and spring wheat, respectively.

In a form of $M = f(r.h.,t)$, the deduced MCPE of each wheat variety was used to calculate the moisture content for grain safe storage with ERH equal to 70% (Table 5). At a borderline condition of 70% r. h., the average moisture contents of fourteen wheat varieties at different temperatures of 10, 15, 20, 25, 30, and 35 °C were 14.90%, 14.57%, 14.26%, 13.97%, 13.71%, and 13.46% w.b., respectively. At 20-25 °C the safe storage m.c. for desorption was 13.93-14.21%, these m.c. were 13.58-13.88%, 14.38-14.69%, 14.40-14.60% for hard, soft and mixed wheat, respectively. The safe storage m.c. (14.03-14.30%) of red wheat at 20-25 °C is similar to that (13.95-14.25%) of white wheat. Furthermore, the safe storage m.c. (14.08-14.35%) of winter wheat at 20-25 °C was slightly higher than that (13.48-13.84%) of spring wheat.

3.3. Comparison of isosteric heats of sorption between different wheat samples

3.3.1 The isosteric heats between wheat adsorption and desorption

The isosteric heat of sorption (h_s) was calculated from the equations (6) to (9). The coefficients C_1, C_2, and C_3 of MCPE equation with a form of $r.h. = f(M,t)$ in Table 4 were used as the coefficients in equation (9). Figure 3A shows the influence of grain moisture content ranging from 4 to 24% w.b. on the isosteric heats of wheat desorption and adsorption. The isosteric heats of both wheat desorption and adsorption decreased rapidly with an increase in seed moisture content until the m.c. of 15% w.b. was reached, but after the moisture is more than 15% w.b. they decreased slowly with increasing moisture content. At lower moisture contents below 15%, both isosteric heats of wheat desorption and adsorption at lower temperatures were higher than those at higher temperatures. The isosteric heats of wheat desorption were significantly higher than those of adsorption below 15% m.c., but above 15% m.c. there was no difference found between them.

The influence of another moisture sorption model such as MOE on the calculated isosteric heats of sorption was also compared in Figure 3B. The coefficients C_1, C_2, and C_3 of MOE equation (Table 6) with a form of $r.h. = f(M,t)$ were respectively used as the coefficients in equation (10). When the MOE model was employed to predict, the isosteric heats of both wheat desorption and adsorption decreased rapidly with an increase in seed moisture content from 7.5 to 17.5% w.b., but above 17.5% m.c. they decreased slowly with increasing moisture content (Figure 3B). The isosteric heats of wheat desorption were significantly higher than those of adsorption in the moisture range of 4 to 20% w.b. At lower moisture contents below 15%, both isosteric heats for wheat desorption and adsorption at lower temperatures tended to be similar to those at higher temperatures. However, above 15% m.c., both isosteric heats for wheat desorption and adsorption at lower temperatures were slightly higher than those at higher temperatures.

3.3.2 The sorption isosteric heats of sorption between hard and soft wheat

Figures 3C and 3D show both sorption isosteric heats of hard and soft wheat at different temperatures predicted by MCPE and MOE models, respectively. The isosteric heats for both sorption of hard and soft wheat were decreased rapidly with an increase in seed moisture content until the moisture content of 17.5% w.b. was reached, and thereafter they decreased slowly with increasing moisture content. The sorption isosteric heats of soft wheat were slightly higher than those of hard wheat under all moisture contents at a constant temperature. For the sorption isosteric heats predicted by MCPE (Figure 3C), at lower moisture contents below 17.5%, the sorption isosteric heats for both hard and soft wheat at lower temperatures were slightly higher than those at higher temperatures. However, the effect of temperature on the sorption isosteric heats of both hard and soft wheat was depleted when the MOE model was employed to calculate (Figure 3D).

3.3.3 The sorption isosteric heats between red and white wheat

Figures 4A and 4B show both sorption isosteric heats of red and white wheat estimated by the MCPE and MOE models, respectively. Similarly to both sorption isosteric heats of hard and soft wheat, it seems that both sorption isosteric heats of red and white wheat were decreased rapidly with increase in seed moisture content until a moisture content of 17.5% w.b. was reached, but above 17.5% they decreased slowly with increasing moisture content. Additionally, the sorption isosteric heats of red wheat were very similar to those of white wheat under all moisture contents at a constant temperature. For the sorption isosteric heats calculated by MCPE (Figure 4A), at lower moisture contents below 17.5%, the isosteric heats for the sorption of both red and white wheat at lower temperatures were slightly higher than those at higher temperatures, but the effect of temperature on the sorption isosteric heats of red and white wheat was depleted when MOE model was used (Figure 4B).

3.3.4 The sorption isosteric heats between winter and spring wheat

Figures 4C and 4D give both sorption isosteric heats of winter and spring wheat predicted by the MCPE and MOE models, respectively. In parallel to the changes in sorption isosteric heats for both hard and soft wheat, as well as for both red and white wheat, the sorption isosteric heats for both winter and spring wheat were decreased rapidly with increase in seed moisture content until the moisture content of 17.5% w. b. was reached, and thereafter they decreased slowly with increasing moisture content. The sorption isosteric heats of spring wheat were slightly higher than those of winter wheat under all moisture contents at a constant temperature. For the sorption isosteric heats calculated by MCPE (Figure 4C), at lower moisture contents below 17.5%, the isosteric heats for the sorption of both winter and spring wheat at lower temperatures were slightly higher than those at higher temperatures, but the influence of temperature on the sorption isosteric heats of winter and spring wheat was eliminated when MOE model was adopted (Figure 4D).

4. Discussion

The theoretical implications of moisture sorption hysteresis range from a depiction of the irreversibility of the sorption process to the question of validity of thermodynamic functions determined from such a system (Kapsalis, 1987). It has been accepted that there was significant hysteresis effect between wheat desorption and adsorption at lower ERH (Pfost et al., 1976; Sun & Woods, 1993; 1994). Sun & Woods (1994) analyzed thirty-three source sets of wheat EMC/ERH data with the preferred equations MCPE and MOE, and considered that the wheat hysteresis effect was not greatly influenced by temperature. However, in this study for the average fitted sorption data of fourteen wheat varieties, both width and span of the hysteresis effect tended to decrease with an increase in temperature. It was noted by Kapsalis (1987) that the span of hysteresis loop always decreases with increasing temperature, but the width of hysteresis loop could increase, remain unchanged, or reduce with increasing temperature. The hysteresis loops of wheat grains at 10-35 °C are of type three classified by Kapsalis (1987). In agreement with these results, the coefficients of MCPE and the isosteric heats for wheat desorption and adsorption were different. So far our knowledge on elucidation of hysteresis phenomenon of cereal moisture sorption is rather limited. The practical implications of moisture sorption hysteresis in wheat can deal with the effect on storage stability.

A study by Chen & Morey (1989) indicated the necessity to choose the most appropriate moisture sorption equation for a specific crop. In this study, among the all eight acceptable moisture isotherm equations (BET, CAE, CCE, MCPE, MGAB, MHE, MOE and SYE) tested, the MCPE with a form of $r.h. = f(M,t)$, or a form of $M = f(r.h.,t)$ both best describe the EMC data of the fourteen wheat varieties in a wide range of 11.3 to 96.0% ERH. The obtained parameters of MCPE in a form of $r.h. = f(M,t)$ were similar to those of Sun & Woods (1994) though they considered MOE in a form of $M = f(r.h.,t)$ to best fit for the wheat sorption data. Thus, the MCPE in a form of $M = f(r.h.,t)$ was used to calculate the moisture contents of wheat safe storage at different temperatures. The safe storage m.c. for fourteen wheat varieties was predicted to be 13.46-14.90% w.b. at temperatures ranging from 10 to 35 °C. These values agree to the safe moisture level, usually taken as that in equilibrium with a maximum of 70% r.h. is about 14% w.b. for the starch cereal grains (Pixton, 1982). At six temperatures ranging from 10 to 35 °C, the standard deviation of m.c. for safe storage of fourteen wheat samples is around 0.7% w.b., close to standard deviation of 0.5% w.b. that the different methods for cereal grain moisture determination should not be beyond (AOAC, 1980).

To our knowledge, few studies have compared the hygroscopic properties between wheat types. We found that the very similar safe storage m.c. remained between red and white wheat. Considered the 0.5% standard deviation of moisture measurement, the similar safe storage m.c. were also found between hard and soft wheat, and between winter and spring wheat. Sun & Woods (1994) compared the fitted curves of MCPE for eight hard wheat varieties and six soft wheat varieties at three temperatures of 0, 30 and 60 °C. At the temperature of 0 °C, the curve for hard wheat lied well above that of soft wheat, but the difference in hygroscopic properties of these two types wheat become very small at 30 °C, then at 60 °C the curve of soft wheat lied above that of soft wheat. In this study, the fitted MCPE isotherms of soft wheat slightly lied above those of hard wheat at the temperatures of 20 and 30 °C. The similar hygroscopic properties between hard and soft wheat might be due to the overall effects of hygroscopic properties of their respective protein and starch (Li et al., 2011). The reason for the similar hygroscopic properties between red and white wheat, as well as between winter and spring wheat, needs further study on microstructural or morphologic characteristic of different wheat varieties.

The isosteric heats for both wheat desorption and adsorption, and for the sorption of both hard and soft wheat, decreased rapidly with an increase in seed moisture content till up to 15% w.b., but thereafter they decreased slowly with increasing moisture content. As mentioned above, similar trends of sorption isosteric heats were also observed for both red and white wheat, as well as for both winter and spring wheat. The isosteric heats of wheat desorption were dramatically higher than those of adsorption from 4.0 to 17.5%, but above 17.5% m.c. there was no difference found between them. These results were different from those reported by Öztekin & Soysal (2000) that the isosteric heats of wheat desorption were higher than those of adsorption from 9.1 to 13.0% w.b., but from 13.1 to 20% the isosteric heats of desorption were lower than those of adsorption. Their difference between desorption and adsorption isosteric heats below 13.0% is much smaller than ours. In their study, they also compared the sorption isosteric heats between hard and soft wheat in the moisture range from 9.1 to 16.7% w.b. The sorption isosteric heats of soft wheat were much higher than those of hard wheat from 9.1 to 12.3% w.b., but from 12.31 to 16.7% the sorption isosteric heats of soft wheat were lower than those of hard wheat. In this study, the sorption isosteric heats of soft wheat were slightly higher than those of hard wheat from 4 to 15% w.b., but above 15% no difference was found between the sorption isosteric heats of soft wheat and hard wheat. The same trends of the sorption isosteric heats were observed for winter and spring wheat, as well as for red and white wheat. In the study of Öztekin & Soysal (2000), the heat of sorption of wheat grains approached that of pure water at the moisture content of about 16.7% wet basis, but in this study the m.c. is around 15.0% w.b., close to those of melon seed, cassava, alfalfa pellets, gari, winged bean seed, and tea at moisture contents of about 11.5, 26.5, 13.8, 13.0, 13.0, and 13.0% w.b., respectively (Arslan & Toğrul, 2006). The rapid increase in the heat of sorption at low moisture content might be due to the existence of highly active polar sites on the surface of wheat grains, which were covered with water molecules forming a mono-molecular layer (Tsami, 1991). The decrease in the isosteric heats with higher amounts of sorbed water can be quantitatively explained by considering that sorption initially occurs on the most active available sites giving rise to high interaction energy. As these sites become occupied, sorption occurs on the less active ones, resulting in lower heats of sorption (Wang & Brennan, 1991). In low moisture contents, the values of the isosteric heats were higher than the latent heat of vaporization of water, indicating that the energy of binding between the water molecules and the sorption sites was higher than the energy which holds the molecules of pure water together in the liquid phase (Al-Muhtaseb, McMinn, & Magee, 2004). At high moisture contents, there was no significant difference between the sorption isosteric heat and the latent heat of vaporization of water over a broad range of moisture contents. Similar findings were reported for the isosteric heats of melon seeds and cassava (Aviara & Ajibola, 2002), starch powder (Al-Muhtaseb, McMinn, & Magee, 2004), and Brussels sprouts (Irzyniec & Klimczak, 2003). Comparison of the adsorption and desorption data shows that, at a specific moisture content, the isosteric heat of desorption was higher than the corresponding adsorption data. This indicates that there are more polar sites on the surface of the solid, and the energy of binding between the water molecules and the surface is higher (Tsami, 1991). Heat values for desorption give a measure of the energy that needs to be supplied to dehydrate the foodstuff.

The sorption isosteric heats for both winter and spring wheat, as well as for both red and white wheat were also compared in this study. The minor difference between the sorption isosteric heats of winter and spring wheat at a particular temperature was very similar to that of soft wheat and hard wheat. No difference was found between the sorption isosteric heats of red and white wheat at all moisture contents from 4.0 to 24% w.b. at a constant temperature. These results suggest the similar hygroscopic properties and sorption isosteric heats occur for different wheat types, i.e. hard vs. soft wheat, red vs. white wheat, or winter vs. spring wheat, respectively.

When MCPE was used to calculate the wheat heat of sorption, at lower moisture contents below 17.5% w.b., the isosteric heats of both desorption and adsorption, of both sorption of hard and soft wheat, as well as of both sorption of winter and spring wheat, and those of both sorption of red and white wheat under lower temperatures all were higher than those under higher temperatures. However, when MOE was used for the calculation of the heat of sorption, regardless of either desorption or adsorption, either hard or soft wheat, as well as either winter or spring wheat, and either red or white wheat, there was no difference found in the isosteric heats of sorption at different temperatures at an EMC below 15%, but above 15% the isosteric heats of sorption at lower temperatures were slightly higher than those at higher temperatures. It has been noted that h_s/h_v was calculated to be dependent on temperature, but the dependence was small (Thorpe, 2001). In this study for two models MCPE and MOE employed respectively to calculate the wheat heat of sorption, MOE can eliminate the dependence of h_s on temperature. Thus, in contrast to a big difference in isosteric heats of wheat desorption and adsorption below the m.c. of 15% w.b., we consider that the similar sorption isosteric heats occur between hard and soft wheat, between red and white wheat, as well as between winter and spring wheat, and the grains from different wheat types could be concordantly treated after harvest.

5. Conclusion

This study determined the moisture sorption isotherms of wheat grains for fourteen Chinese varieties. It is found that MCPE model results in the best fitting to the sorption data. A significant hysteresis effect was found between wheat desorption and adsorption at lower ERH, but the similar hygroscopic properties remained between wheat types, i.e. hard and soft wheat, red and white wheat, or winter and spring wheat, respectively. The isosteric heats for wheat adsorption and desorption, and the sorption heats for hard and soft wheat, winter and spring wheat, as well as red and white wheat, all decreased rapidly with an increase in seed moisture up to 15% w.b., thereafter they decreased slowly with increasing moisture content. The isosteric heats of desorption were higher than those of adsorption below 15% m.c., but above 15 % m.c. there was no difference between the desorption and adsorption. The similar isosteric heats of grain sorption found between hard and soft wheat, as well as between red and white wheat, or between winter and spring wheat indicate that the wheat grains from different types have similar hygroscopic properties and sorption isosteric heats, and can be synchronously dealt with during drying, storage and aeration.

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Nomenclature

a_1, a_2, b_1, b_2, d	the coefficients of CAE equation	h_w	differential heat of wetting (KJ/Kg)
C_1, C_2, C_3, C_4	equation coefficients	R^2	coefficient of determination
d.b.	dry basis	P_s	saturate vapor pressure (Pa)
EMC	equilibrium moisture content	*r.h.*	relative humidity
ERH	equilibrium relative humidity	RSS	residue sum of squares
m.c.	moisture content	SE	standard error
m_i	experimental value	T	absolute temperature (K)
m_{mi}	average mean of experimental value	t	temperature (°C)
m_{pi}	predicated value	vs.	versus
MRE	mean relative percentage error (%)	w.b.	wet basis
n	number of observations		
h_s	isosteric heat of sorption (KJ/Kg)		
h_v	latent heat of vaporization of free water (KJ/Kg)		

Table 1. The characteristics of fourteen wheat samples adopted in this study

Sample No.	Wheat variety	SKCS Hardness	Wheat Classes	Producing region	Harvest time
1	Nanduan	68	Hard white winter	Shandong	June, 2008
2	Longyuan 2	74	Hard white winter	Shaanxi	June, 2007
3	Changwu 3297	73	Hard white winter	Shaanxi	June, 2007
4	Henan Bai	59	Hard white winter	Henan	June, 2008
5	Nongda Hong	50	Mixed red winter	Beijing	June, 2007
6	Sanyuan	51	Mixed red winter	Beijing	June, 2007
7	Shunyi 8433	40	Soft white winter	Beijing	June, 2008
8	Nongda 5177	40	Soft white winter	Beijing	June, 2007
9	Xuzhou Bai	36	Soft white winter	Jiangsu	May, 2008
10	Zhaozhuang 1	24	Soft red winter	Beijing	June, 2008
11	Lumai 1	18	Soft red winter	Shandong	June, 2007
12	Hebei Yongqing	74	Hard red winter	Hebei	June, 2007
13	Neimeng Chun	77	Hard red spring	Neimenggu	June, 2007
14	Longjiang Chun	69	Hard red spring	Heilongjiang	June, 2007

Table 2. The isotherm equations used in this study

Models	Equation[a]	Reference
Brunauer-Emmett-Teller (BET)	$M=\frac{(C_1+C_2\times t)\times C_3\times r.h.}{(1-r.h.)\times(1-r.h.+C_3\times r.h.)}$ ($r.h.$ <50%)	Brunauer et al., 1938
CAE	$r.h.=\exp\{\{\frac{d}{222}[\exp(\frac{b_1-M}{a_1})-\exp(\frac{b_2-M}{a_2})](1737.1-\frac{474242}{273+t})$ $+d[1-\exp(\frac{b_1-M}{a_1})]+202\}/87.72\}$	Wu et al., 2011
Chen-Clayton (CCE)	$M=\frac{1}{-C_3\times(t+273.15)^{C_4}}\ln[\frac{(t+273.15)^{C_2}\ln(r.h.)}{-C_1}]$ or $r.h.=\exp\{\frac{-C_1}{(t+273.15)^{C_2}}\exp[-C_3\times(t+273.15)^{C_4}\times M]\}$	Chen & Clayton, 1971
Modified Chung-Pfost (MCPE)	$r.h.=\exp[-\frac{C_1}{t+C_2}\exp(-C_3\times M)]$ or $M=-\frac{1}{C_3}\times\ln[-\frac{(t+C_2)\times\ln(r.h.)}{C_1}]$	Pfost et al., 1976
Modified Guggenheim-Anderson-deBoer (MGAB)	$r.h.=\frac{2+\frac{C_3}{t}\times(\frac{C_1}{M}-1)-\{[2+\frac{C_3}{t}\times(\frac{C_1}{M}-1)]^2-4\times(1-\frac{C_3}{t})\}^{\frac{1}{2}}}{2\times C_2\times(1-\frac{C_3}{t})}$ or $M=\frac{C_1\times C_2\times(\frac{C_3}{t})\times r.h.}{(1-C_2\times r.h.)\times(1-C_2\times r.h.+\frac{C_3}{t}\times C_2\times r.h.)}$	Jayas &Mazza, 1993
Modified Henderson (MHE)	$r.h.=1-\exp[-C_1\times(t+C_2)\times M^{C_3}]$ or $M=[-\frac{\ln(1-r.h.)}{C_1\times(C_2+t)}]^{\frac{1}{C_3}}$	Thompson et al., 1986
Modified Oswin (MOE)	$r.h.=\frac{1}{1+(\frac{C_1+C_2\times t}{M})^{C_3}}$ or $M=\frac{C_1+C_2\times t}{(\frac{1}{r.h.}-1)^{\frac{1}{C_3}}}$	Chen & Morey, 1989
STYE	$r.h.=\exp[C_1\times\exp(-C_2\times M)\times\ln(P_s)-C_3\times\exp(-C_4\times M)]$	Strohman & Yoerger, 1967

[a] $r.h.$, relative humidity; M, equilibrium content, percentage wet basis; t,temperature (°C); P_s, saturated vapor pressure; C_1, C_2, C_3, and C_4; a_1, a_2, b_1, b_2 and d are coefficients in the equations.

Table 3. Summary of the results of fitting equations to the 28 data sets

Sorption type	Model function	Models	Statistical parameters				Order
			RSS[a]	SE	R^2	MRE%	
Desorption	$r.h. = f(M,t)$	CCE	0.02884	0.00070	0.98714	4.93836	4
		MCPE	0.02166	0.00052	0.99331	4.59279	2
		MGAB	0.03934	0.00094	0.98785	5.51814	6
		MHE	0.03870	0.00077	0.98805	6.43200	5
		MOE	0.03528	0.00060	0.98911	7.02714	3
		STYE	0.01912	0.00047	0.99339	4.07907	1
	$M = f(r.h.,t)$	BET	42.12103	1.03414	0.98417	4.82737	3
		CCE	12.86221	0.31371	0.99078	3.76914	2
		MCPE	11.89029	0.28314	0.98875	3.23164	1
		MGAB	40.27414	0.95891	0.96213	7.10179	6
		MHE	24.61518	0.58607	0.97685	4.89171	4
		MOE	24.72369	0.58866	0.97661	5.47250	5
Adsorption	$r.h. = f(M,t)$	CCE	0.01369	0.00033	0.99218	3.56929	3
		MCPE	0.01349	0.00032	0.99584	3.64493	2
		MGAB	0.03183	0.00076	0.99017	6.49014	6
		MHE	0.01819	0.00043	0.99442	4.27700	4
		MOE	0.02627	0.00047	0.99190	5.78950	5
		STYE	0.01291	0.00031	0.99600	3.58414	1
	$M = f(r.h.,t)$	BET	29.78256	0.72409	0.98333	4.87981	4
		CCE	7.99557	0.19501	0.99151	3.03871	2
		MCPE	8.45751	0.20136	0.99281	2.97600	1
		MGAB	32.94837	0.78454	0.97197	9.32607	6
		MHE	11.96531	0.28596	0.98966	4.03821	3
		MOE	25.92843	0.61734	0.97829	6.80193	5

[a] RSS, residue sum of squares, SE, the standard error, R^2, correlation coefficient, and MRE, mean relative percentage error.

Table 4. The best fitted MCPE parameters for the moisture sorption of wheat samples

Data set	Total wheat varieties	Coefficients of MCPE in a form of $r.h. = f(M,t)$			Statistical parameters			
		C_1	C_2	C_3	RSS	SE	R^2	MRE%
Desorption	14	529.932	41.687	0.223	2.31E-02	2.40E-04	0.9969	3.1117
Adsorption	14	920.105	150.246	0.206	1.98E-02	1.79E-04	0.9977	2.4708
Average[a]	14	622.365	72.117	0.214	1.91E-02	1.78E-04	0.9977	2.0693
Hard wheat	7	601.664	72.665	0.214	2.12E-02	1.89E-04	0.9975	2.323
Soft wheat	5	580.889	61.186	0.213	1.76E-02	1.69E-04	0.9978	2.052
Mixed wheat	2	930.351	118.819	0.217	1.96E-02	2.58E-04	0.9967	2.748
Red wheat	7	644.263	74.867	0.215	1.99E-02	1.95E-04	0.9975	2.412
White wheat	7	602.627	69.642	0.214	1.88E-02	1.76E-04	0.9977	2.017
Winter wheat	12	634.661	73.938	0.215	1.94E-02	1.87E-04	0.9976	2.164
Spring wheat	2	557.89	62.448	0.213	2.11E-02	2.27E-04	0.9971	3.141

[a] Average is the mean of desorption values and adsorption values of 14 wheat varieties.

Table 5. Estimation of the moisture content for wheat safe storage with the MCPE isotherm

Sample no.	Parameters of MCPE in a form of $M = f(r.h.,t)$			Moisture contents (% w.b.) of wheat grains at 70% ERH					
	C_1	C_2	C_3	10°C	15°C	20°C	25°C	30°C	35°C
1	689.488	53.566	0.224	15.24	14.91	14.59	14.30	14.02	13.76
2	1006.036	109.277	0.224	14.12	13.94	13.76	13.59	13.43	13.27
3	438.700	51.009	0.220	13.65	13.30	12.96	12.65	12.36	12.09
4	585.597	52.761	0.216	15.11	14.76	14.43	14.12	13.83	13.56
5	2394.201	296.777	0.219	14.09	14.02	13.94	13.87	13.80	13.73
6	744.465	45.296	0.228	15.92	15.55	15.20	14.87	14.57	14.29
7	521.849	48.853	0.220	14.61	14.24	13.89	13.57	13.28	13.00
8	475.356	36.445	0.204	16.45	15.95	15.50	15.08	14.70	14.34
9	665.515	55.459	0.221	15.16	14.83	14.52	14.23	13.95	13.69
10	987.527	75.072	0.231	15.08	14.83	14.60	14.37	14.16	13.96
11	722.930	55.033	0.220	15.63	15.30	14.98	14.69	14.41	14.15
12	688.704	63.555	0.231	14.15	13.86	13.59	13.34	13.10	12.88
13	385.071	39.355	0.214	14.42	13.97	13.56	13.18	12.83	12.50
14	478.517	45.377	0.214	14.89	14.49	14.12	13.77	13.45	13.15
Average value[b]				14.90± 0.78	14.57± 0.73	14.26± 0.70	13.97± 0.68	13.71± 0.67	13.46± 0.67
Desorption[a]	635.479	57.093	0.221	14.84	14.51	14.21	13.93	13.66	13.41
Hard wheat	564.602	54.757	0.22	14.53	14.19	13.88	13.58	13.31	13.05
Soft wheat	638.063	51.729	0.219	15.37	15.02	14.69	14.38	14.09	13.82
Mixed wheat	1015.397	89.640	0.223	15.03	14.81	14.60	14.40	14.21	14.03
Red wheat	689.899	60.829	0.222	14.90	14.59	14.30	14.03	13.78	13.54
White wheat	588.872	53.871	0.218	14.92	14.57	14.25	13.95	13.67	13.40
Winter wheat	684.633	60.503	0.221	14.95	14.64	14.35	14.08	13.82	13.58
Spring wheat	428.858	42.234	0.214	14.66	14.23	13.84	13.48	13.14	12.83

[a] Desorption is the average of desorption data of fourteen wheat varieties. [b] Average value is the means of samples from no. 1 to 14 plus standard deviation.

Table 6. The fitted MOE parameters for the moisture sorption of wheat samples

Data set	Total wheat varieties	Coefficients of MOE in a form of $r.h. = f(M,t)$			Statistical parameters			
		C_1	C_2	C_3	RSS	SE	R^2	MRE%
Desorption	14	12.638	-0.071	3.529	0.02311	5.50E-04	0.9929	6.1188
Adsorption	14	10.333	-0.029	2.879	0.01979	4.71E-04	0.9939	5.2387
Average[a]	14	11.492	-0.049	3.191	0.01912	4.55E-04	0.9941	5.078
Hard wheat	7	11.288	-0.048	3.134	0.0212	5.05E-04	0.9934	5.371
Soft wheat	5	11.978	-0.057	3.267	0.01758	4.19E-04	0.9946	4.901
Mixed wheat	2	10.961	-0.032	3.193	0.01956	4.66E-04	0.994	4.919
Red wheat	7	11.474	-0.048	3.202	0.01993	4.75E-04	0.9938	5.177
White wheat	7	11.507	-0.051	3.181	0.01883	4.48E-04	0.9942	5.177
Winter wheat	12	11.456	-0.048	3.191	0.01943	4.63E-04	0.994	5.122
Spring wheat	2	11.706	-0.056	3.194	0.02108	5.02E-04	0.9935	5.814

[a] Average is the mean of desorption and adsorption data of 14 wheat varieties.

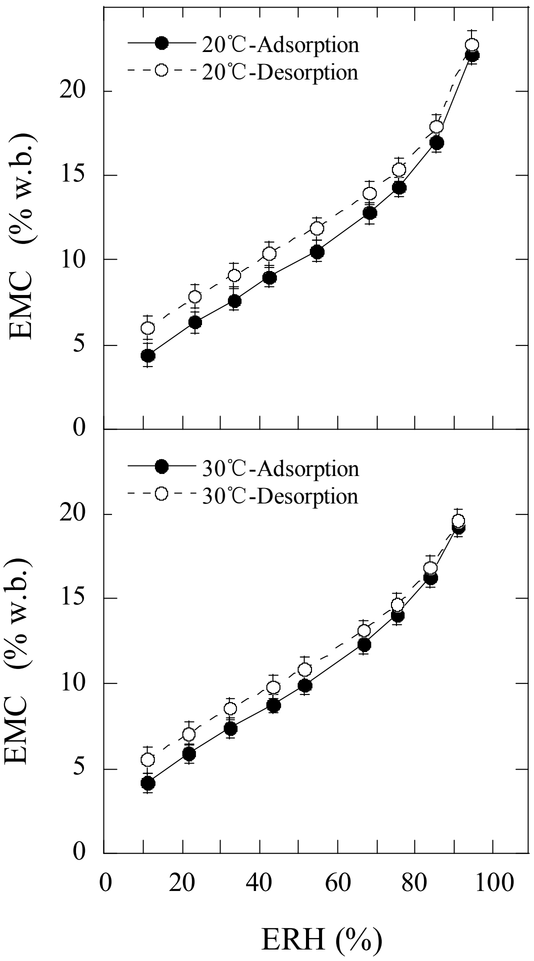

Figure 1. Comparison of the wheat desorption and adsorption isotherm at 20 and 30 °C predicted by the Modified Chung-Pfost equation (MCPE)

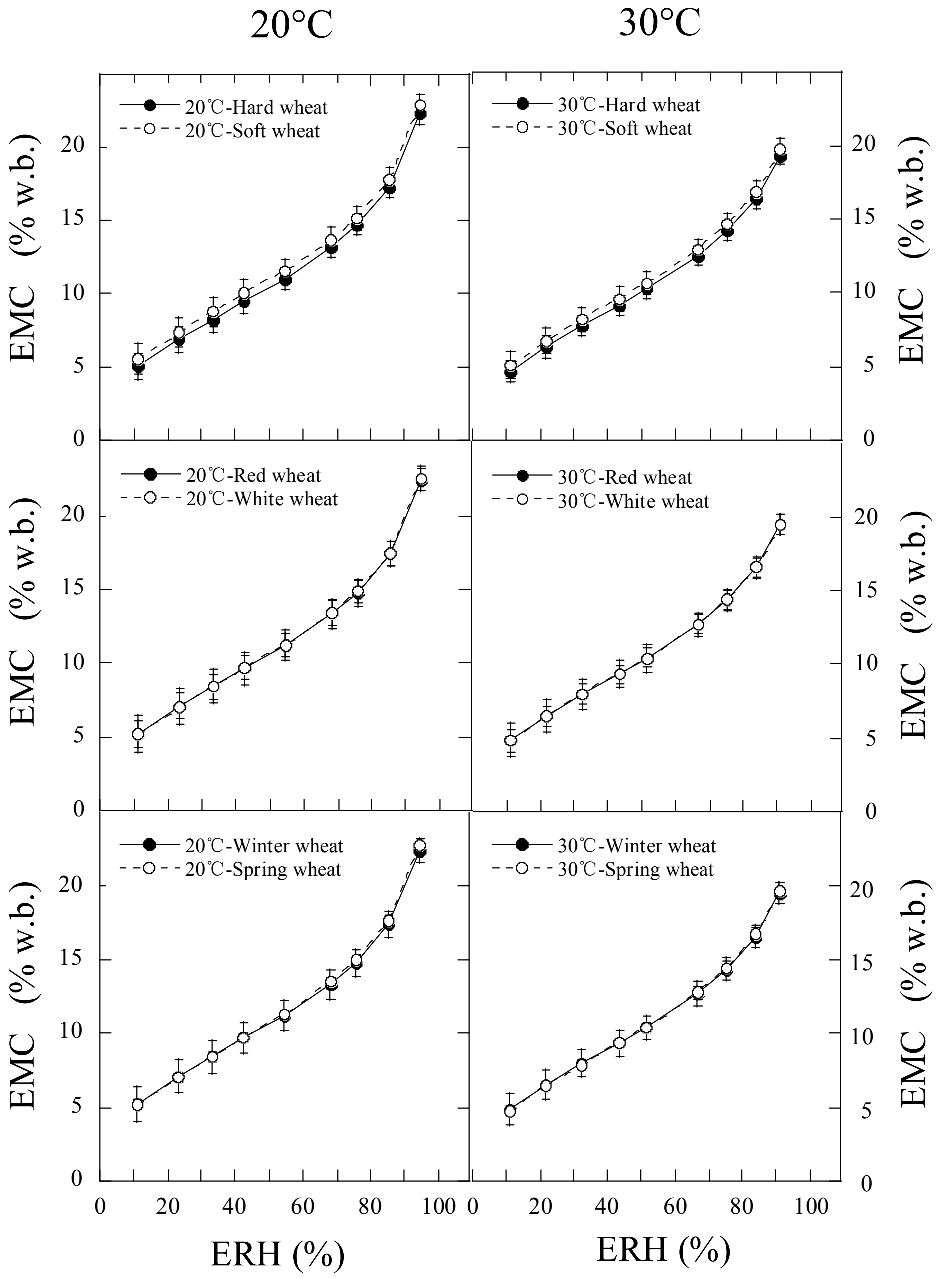

Figure 2. Comparison of the sorption isotherm of hard cv. soft wheat, red cv. white wheat, and winter cv. spring wheat at 20 and 30 °C predicted by the Modified Chung-Pfost equation

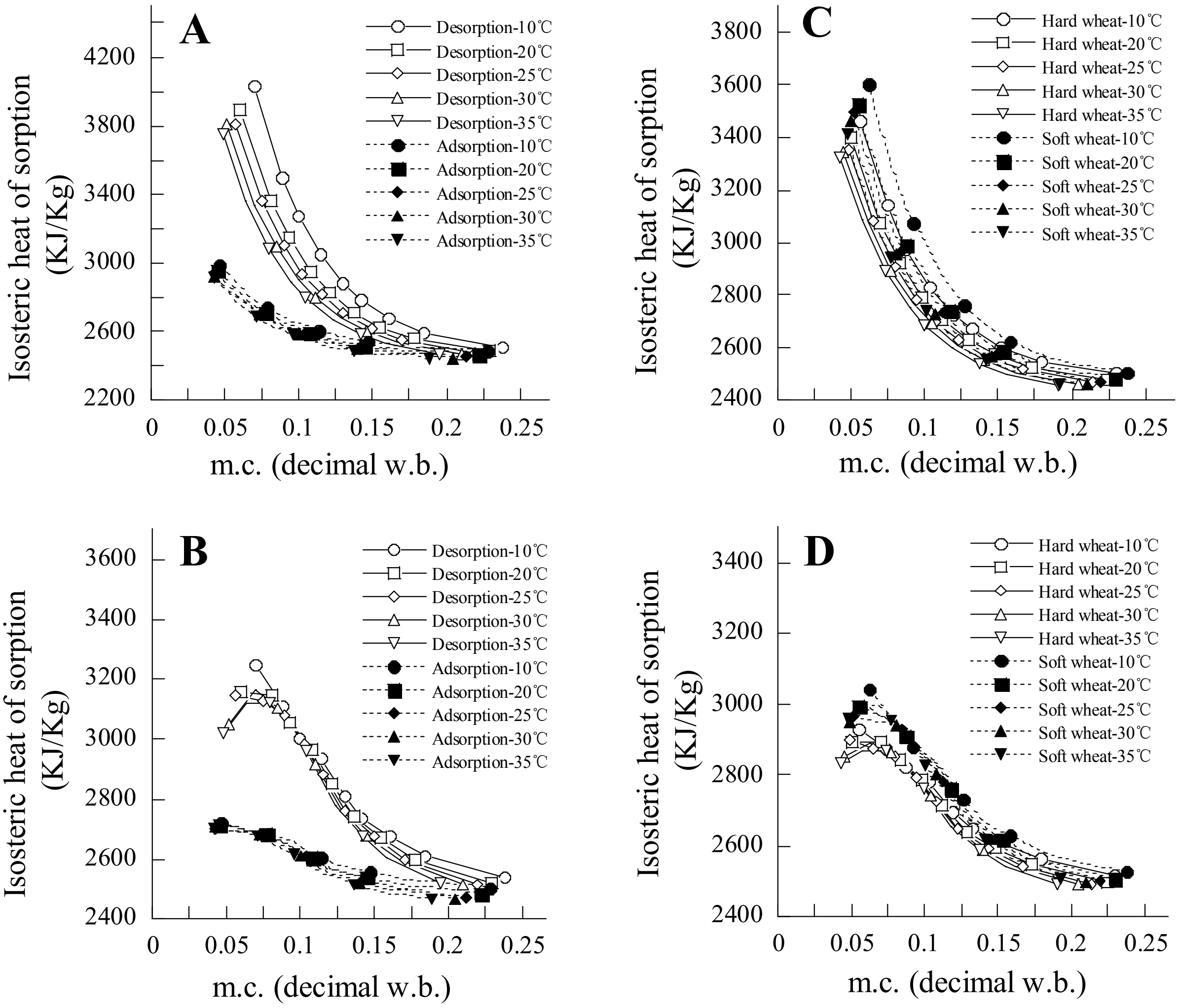

Figure 3. Comparison of isosteric heats of wheat desorption and adsorption, and the sorption isosteric heats of hard and soft wheat at different temperatures predicted by two models of MCPE (A and C), and MOE (B and D), respectively

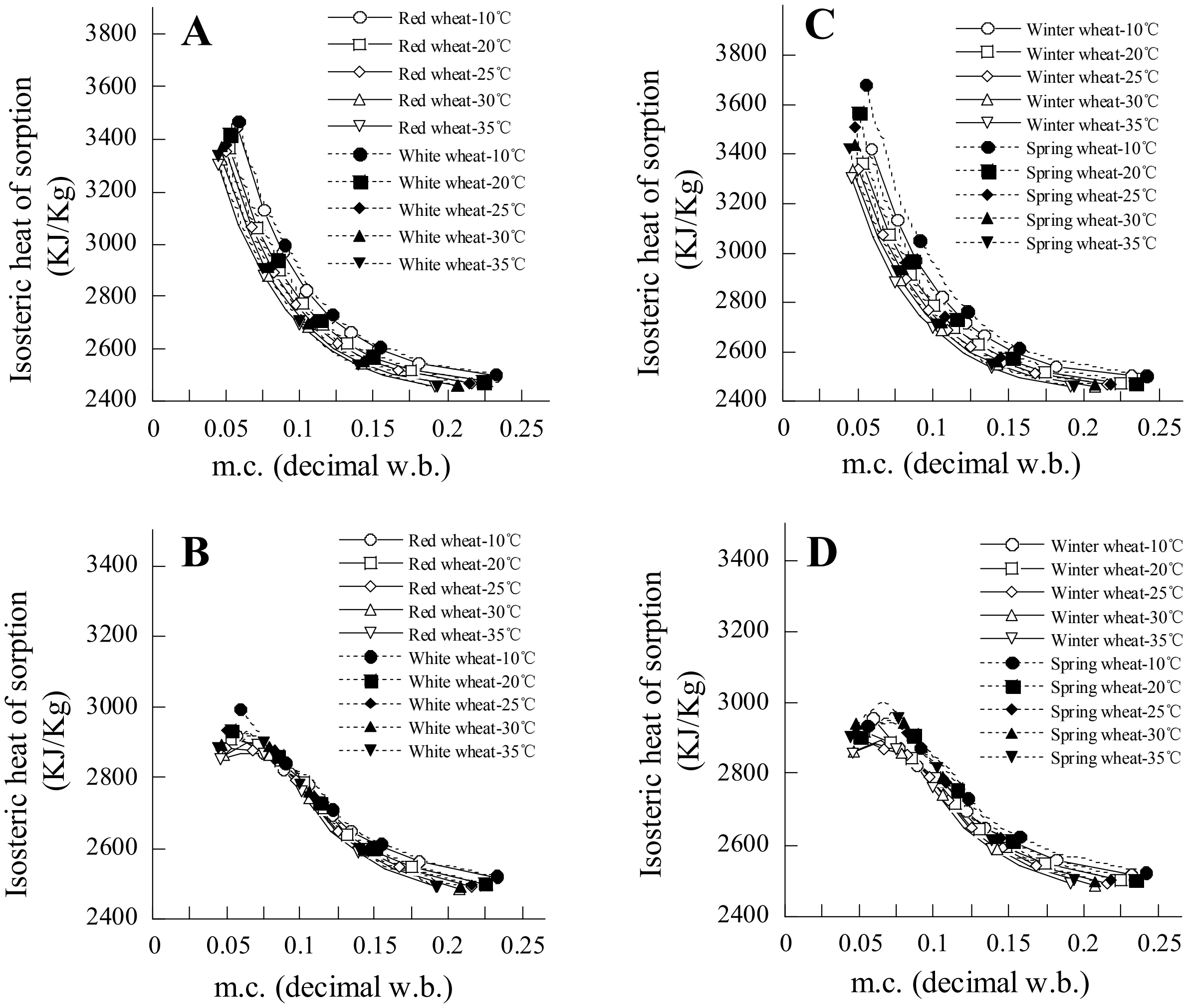

Figure 4. Comparison of the sorption isosteric heats between red and white wheat, and between winter and spring wheat at different temperatures predicted by two models of MCPE (A and C) and MOE (B and D), respectively