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MOISTURE SORPTION THERMODYNAMIC PROPERTIES OF BERMUDA GRASS

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Abstract

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Thermodynamic properties such as differential enthalpy and differential entropy are important tools for analyzing post-harvest handling and drying of grasses such as Bermuda grass. This particular grass is widely used as a feedstock or as pellets for energy production. Thus, this study was initiated with the aim of modeling moisture sorption thermodynamic properties of Bermuda grass with potential benefits of minimizing energy costs and optimizing drying kinetics. Moisture sorption thermodynamic properties of Bermuda grass were studied using gravimetric methods under three temperature levels, namely, 20, 30 and 40°C. Based on findings of related previous studies, the Modified Halsey model was used to obtain the thermodynamic properties of the grass. It was found that, within the ranges considered here, both properties were strong functions *of equilibrium moisture content* and decreased substantially (by about 97%) with equilibrium moisture content. Specifically, differential enthalpy of the grass decreased exponentially with increase in moisture content from 1912 kJ/kg at 0.03 equilibrium moisture content (decimal w.b.) to 62.8 kJ/kg at an equilibrium moisture content of 0.27. Similarly, differential entropy decreased also exponentially from 7.93 kJ/kg.K to 0.26 kJ/kg.K at the same respective levels of equilibrium moisture content. Enthalpy-entropy compensation was validated by the linear relationship between the two. Finally, it was found that the moisture sorption process for Bermuda grass is enthalpy-driven.

Key words: Bermuda grass; equilibrium relative humidity; moisture sorption isotherms; postharvest; thermodynamic properties

List of Abbreviations: ΔH_d – differential heat of sorption (also known as net isosteric heat of sorption); ΔS_d – differential entropy; T_β – isokinetic temperature; w.b. – wet basis; T_{hm} – harmonic mean temperature; *ERH* – equilibrium relative humidity; T – temperature; EMC – equilibrium moisture content; ΔG – Gibb's free energy change; Q_s – isosteric heat of sorption; R^2 – correlation coefficient; MSE – mean square error

Introduction

Bermuda grass (*Cynodon dactylon*) is a popular, fast growing turf grass grown in warm climates around the world. In hot *arid* regions such as Jordan and Middle East, the grass is grown for it's high tolerance to extreme dry and hot weather conditions. The cut grass is produced in large amounts from recreational sites and is used as a feedstock or as a source of energy by direct incineration in the form of pellets. However, several problems are associated with the handling and utilization of cut Bermuda grass such as self-ignition that results from internal heat developed in the grass and the uncontrolled bio-degradation associated with microbial and allergic particles emission to the environment. In an attempt to handle such problems, Bermuda grass has to be dried efficiently (Çağlar, 2013). It is established that

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high initial water content of the grass increases costs of drying, transportation and storage. While handling and storage of dried grass is easier and safer as compared to wet biomass. The high costs associated with the drying process remain a challenge. Nevertheless, optimization of the drying operation can help find feasible and cost-effective post-harvest treatment of the grass. Establishing the relationship between temperature and relative humidity through moisture sorption isotherms and thermodynamic analysis are of great importance for efficient handling, drying and storage practices. Due to its hygroscopic nature, Bermuda grass can easily adsorb or desorb humidity (Rizvi and Benado, 1984; Yang and Cenkowski, 1993; Aviara and Haque, 2001; Seiiedlou et al., 2010; Lago and Norena, 2015).

Thermodynamic analysis of moisture sorption phenomenon has been used to understand drying energy requirements, microstructure and chemical-physical interactions taking place at the water–solid interface. It is known that water in biomass exists as bound rather than free water at the water-solid interface. Since energy needs for dehydration are largely dependent on the strength by which water is bound to the solid surface, energy requirement in biomass dehydration is significantly larger than that for free unbound water. As such, drying process can be an energy-intensive process. Therefore, thermodynamic analysis can be a useful tool in determining the amount of theoretical energy needed to achieve a desired final moisture level for stable storage of biomass (Acharjee et al., 2011; Argyropoulos, 2012).

Some of the important thermodynamic functions include the differential heat of sorption (also known as net isosteric heat of sorption (ΔH_d)), the differential entropy (ΔS_d), and the isokinetic temperature (T_β). *It worth noting that ΔH_d can be used to express the strength of sorption between water and solid surfaces and, therefore, the energy needed for drying process. In contrast, ΔS_d may be used to relate the number of available sorption sites to a specific energy level, while T_{hm} can be utilized to determine the thermodynamic nature of the sorption process* .The isokinetic temperature represents the temperature at which all chemical and biochemical reactions proceed at the same rate, while the Gibb's free energy change is used to determine if the sorption process was spontaneous or not (Rizvi and Benado, 1984; Polachini, 2016).

In hot environments, the cut Bermuda grass with high initial moisture content is prone to fast deterioration. Consequently, drying is required for proper handling and use of the grass. Hence this work involves a detailed analysis of water sorption isotherms thermodynamics of Bermuda grass in an attempt to optimize the drying operation. Although a detailed analysis of moisture sorption isotherms was discussed in a previous work for the authors, no previous work was found on the thermodynamic analysis of Bermuda grass. The objectives of this study were to determine these thermodynamic properties which can then be used to design and optimize the drying equipment and processes for Bermuda grass.

Materials and Methods

Sample preparation

Two kg of freshly cut Bermuda grass were collected from recreational parks in Irbid area (Irbid, Jordan). Samples were first oven-dried at 105°C to 6% (w.b.) moisture and then fine ground with a hammer mill using a 4 mm sieve. Finally, the samples were stored in 2-L hermetically-sealed containers at 4^{o}C until further use.

Experimental procedure

The moisture sorption isotherm curves were determined using the gravimetric method. In the process, 0.5 g of Bermuda grass were placed inside aluminum cups in a glass desiccator with 7 saturated salt solutions having water activity values ranging from 8% to 90% (Table 1). All stock solutions were prepared from laboratory grade salts by adding the required distilled water to attain a slightly over-saturated solution. As shown in Table 1, three temperature levels were used, specifically, 20, 30 and 40°C which were so chosen to represent the normal handling and storage temperatures in Jordan climate. All measurements were made inside a thermostatic chamber to maintain constant temperature conditions throughout the whole experiment. The samples were regularly checked until reaching a constant weight and all measurements were obtained in triplicates.

Table 1
Equilibrium relative humidity (ERH) of the seven saturated salt solutions used in this study

Saturated salt solution	Chemical formula	ERH (%)		
		20°C	30°C	40°C
Sodium Hydroxide	NaOH	8.9	7.6	6.3
Magnesium chloride	$MgCl_2$	33.0	32.4	31.6
Potassium carbonate	K_2CO_3	43.2	43.2	41.0
Sodium bromide	NaBr	59.1	57.3	56.0
Potassium Iodide	KI	69.9	67.9	69.1
Potassium chloride	KCl	86.0	84.0	83.0
Barium Chloride	BaCl	89.0	89.0	89.0

Thermodynamic properties

Based on a previous study that indicated the modified Halsey equation was the best to represent the moisture sorption isotherm in Bermuda grass (Al-Mahasneh, 2014). Thermodynamic properties of Bermuda grass in this current study were derived from the modified Halsey equation. Several important thermodynamic parameters were calculated including the differential heat of sorption (net isosteric heat of sorption) or differential enthalpy (ΔH_d), the differential entropy of adsorption (ΔS_d), enthalpy-entropy compensation, and harmonic mean temperature (T_{hm}). It's worth noting that ΔH_d can be used to express the strength of sorption between water and solid surfaces and, therefore, the energy needed for drying process. In contrast, ΔS_d may be used to relate the number of available sorption sites to a specific energy level, while T_{hm} can be utilized to determine the thermodynamic nature of the sorption process (Polachini, 2016; Fasina, 1997).

The differential heat of sorption (ΔH_d) and the differential entropy of sorption (ΔS_d) were obtained by applying the Clausius-Clapeyron equation. They were determined from the slope of the graph of the natural log of equilibrium relative humidity (*ln ERH*) vs temperature reciprocal ($1/T$) at different constant equilibrium moisture contents (EMC) as follows (Igathinathane, 2008):

$$Ln(ERH)_m = -\left(\frac{\Delta H_d}{RT}\right) + \left(\frac{\Delta S_d}{R}\right) \quad (1)$$

The compensation theory which proposes a linear relationship between differential enthalpy and entropy was also investigated to determine whether the drying process is enthalpy or entropy driven using the following relationship (Fasina, 1997):

$$\Delta H_d = T_\beta \Delta S_d + \Delta G \quad (2)$$

Finally, the harmonic mean temperature (T_{hm}), was obtained using the expression (Iglesias, 1976):

$$T_{hm} = \left(\frac{m}{\sum_{i=1}^{m} \frac{1}{T}}\right) \quad (3)$$

and was compared to the isokinetic temperature, T_β, to determine whether the sorption process was enthalpy or entropy driven. Gibb's free energy change, ΔG, was used to indicate the sorbent affinity to water; a negative ΔG for a spontaneous water sorption process while a positive ΔG for a nonspontaneous process (Polachini, 2016; Fasina, 1997).

Results and Discussion

Moisture sorption isotherms and model fitting

The saturated salt solutions used for obtaining moisture sorption isotherms are shown in Table 1. The time needed for Bermuda grass samples to reach a constant weight varied between 8 and 17 days. Figure 1 shows the measured moisture sorption isotherms, MSI, for Bermuda grass at the three different temperatures used, namely, 20°C, 30°C and 40°C. It was observed that the relationship between equilibrium moisture content, EMC, and equilibrium relative humidity, ERH, followed sigmoidal type II isotherms (Arabhosseini et al., 2010). According to Brunauer (1945), classifications of isotherms, the type II isotherms are typical for intermediate moisture materials such as agricultural products.

It was found that the Halsey equation provides the best moisture sorption relationship with a correlation coefficient, R^2 of 0.973 and a mean square error, MSE of 2.18. The equation that best represents the fit can be written as follows:

$$EMC = \left[\frac{-\exp(1.76 + 0.013T)}{\ln(ERH)}\right]^{\left(\frac{1}{1.313}\right)} \quad (6)$$

The increase in EMC with temperature may be attributed to the strong influence of temperature on the physiochemical properties of the grass which resulted in reducing the number of active moisture binding sites. At higher temperatures, molecules of bound water are activated to higher energy levels resulting in an increase in their distance apart causing a decrease in their attraction forces and leading to decreased EMC values (Mohamed et al., 2005). The decrease in sorption energy with EMC was a result of water tendency to initially occupy the highly active polar sites on the solid surface that corresponds to the strongest binding energy. A progressive filling of the less available sites corresponding to lower activation energies will occur thereafter. A similar trend was reported for flax straw, hemp stalks and reed canary grass (Nilsson et al., 2005), and for corn Stover, wheat straw, big bluestem, and sorghum stalk (Theerarattananoon et al., 2011).

Thermodynamic properties

The variation of EMC with temperature is shown in Figure 1. The figure shows that EMC increases with temperature which may be attributed to the fact that temperature increase results in more activated water molecules due to the higher energy level and therefore they tend to break away from the binding sites of the solid material (Belghith 2015, Hossain et al., 2001).

The modified Halsey equation was found to provide the best fit of moisture sorption isotherms among all the moisture sorption isotherms models tested according to the values of MSE and R^2. This finding is in line with other previous studies on grasses and leafy plant products which reported that the Modified Halsey equation provides the best fit of moisture sorption isotherms for flax straw, hemp stalks and reed canary grass (Nilsson et al., 2005), corn stover (Igathinathane, 2008), and switch grass and prairie cord grass (Karunanithy, 2013).

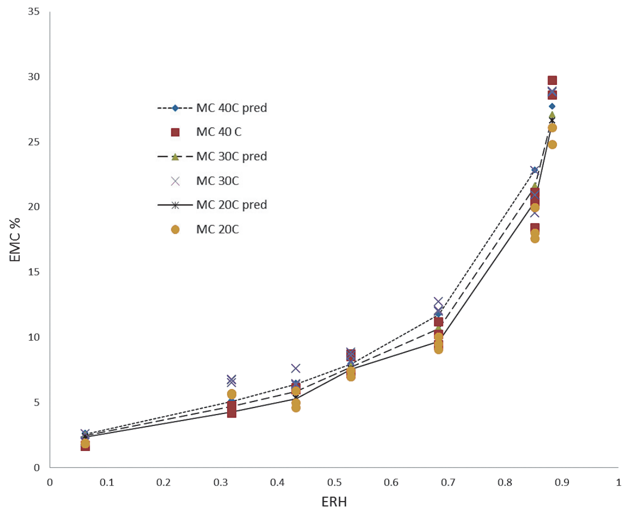

Fig. 1. Moisture sorption isotherms of Bermuda grass using Halsey model

Thermodynamic properties of Bermuda grass were obtained using the modified Halsey equation by plotting the linear relationship between ln(ERH) and 1/T as shown in Figure 2. The variation of differential enthalpy (ΔH_d) with EMC is shown in Figure 3, which readily shows that ΔH_d decreases exponentially with EMC. In particular, Figure 2 shows that ΔH_d decreased from 1912 kJ/kg to 62.8 kJ/kg, corresponding to an increase in EMC from 0.03 and 0.2.

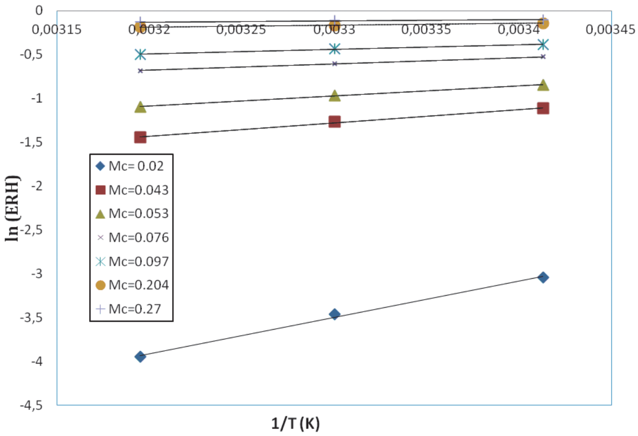

Fig. 2. Relationship between ln (ERH) and 1/T for Bermuda grass

The relationship between ΔH_d and EMC was fitted using the following equation:

$$\Delta H_d = 11.25(EMC)^{-1.313} \tag{7}$$

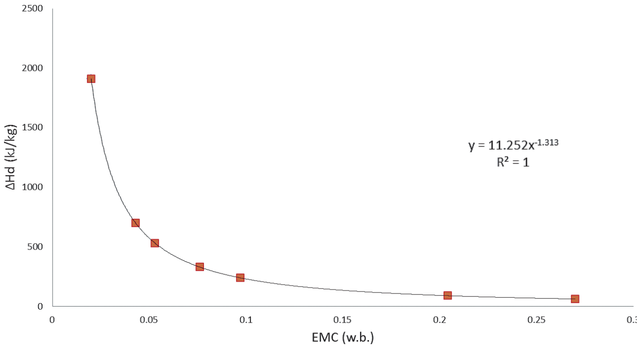

Fig. 3. Variation of differential enthalpy with EMC for Bermuda grass

The decrease in differential enthalpy (ΔH_d) with EMC suggests the presence of a much stronger binding energy between sorption sites and water molecules at lower EMC levels compared to that in the higher EMC side. The decrease in ΔH_d from 1912 kJ/kg at 0.02 EMC to 62.8 kJ/kg at 0.2 EMC indicates a 30-fold increase in the theoretical drying energy in the EMC range of 0.2 and 0.03. Fortunately, the needed EMC end point for Bermuda grass is normally close to 0.1, which corresponds to a ΔH_d of 231 kJ/kg. This value corresponds to about 370% of the energy requirement for water at 0.2 EMC. It should be observed also that ΔH_d represents the amount of binding energy above the latent heat of vaporization of free water, h_{fg}, of 2200 kJ/kg. The sum of differential enthalpy and latent heat of vaporization is termed as isosteric heat of sorption (Q_s). Q_s at a specific EMC can be a useful indicator of the state of adsorbed water and, therefore, a measure of chemical, physical and microbial stability of the biological material under specific storage conditions. ΔH_d can be also a useful tool in investigating the variation of affinity of water particles to a solid surface and, therefore, the corresponding amount of theoretical energy needed during dehydration. It was reported that higher values of ΔH_d at lower EMC could be a result of the strong interaction between water molecules and food solids hydrophobic groups (Wang and Brennan, 1991), while other findings indicated that it could be a result of chemical sorption affinity of the polar sites and strong hydrogen bonding among food particles (Lim et al., 1995). Similar trends were reported for sugar beet root (Iglesias et al., 1976), alfalfa pellets (Fasina, 1997), sesame hulls (Al-Mahasneh et al., 2010), and cassava bagasse (Polachini, 2016). Moreover, a similar trend was observed for differential entropy as differential entropy (ΔS_d) was also found to decrease exponentially with EMC. Specifically, Figure 4 shows that ΔS_d decreased from 7.9 kJ/kg K to 0.26 kJ/kg K when ERH increased from 0.03 to 0.27.

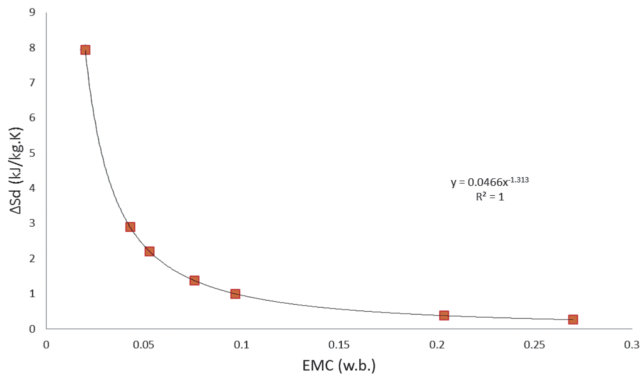

Fig. 4. Variation of differential entropy with EMC for Bermuda grass

The relationship between ΔS_d and EMC was fitted as follows:

$$\Delta S_d = 0.0466(EMC)^{-1.313} \quad (8)$$

This result is in agreement with those reported for alfalfa pellets (Fasina, 1997), sesame hulls (Al-Mahasneh et al., 2010), and cassava bagasse (Polachini, 2016).

Enthalpy–entropy compensation theory was investigated by plotting differential enthalpy against differential entropy as shown in Figure 5.

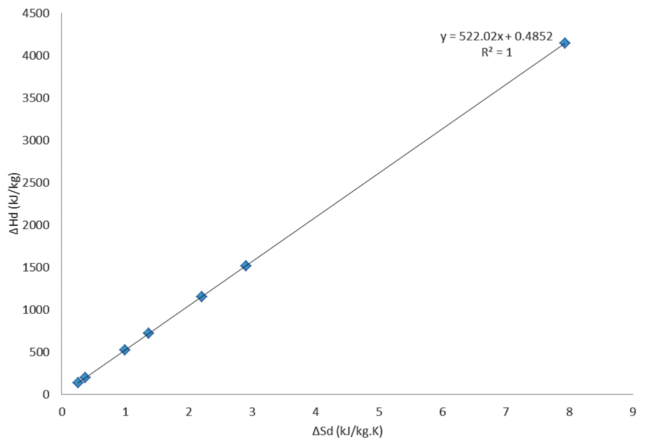

Fig. 5. Linear relationship between differential enthalpy and differential entropy for Bermuda grass

A straight line relationship was obtained with $R^2 = 1$, as follows:

$$\Delta H_d = 522.02\Delta S_d + 0.485 \quad (9)$$

The linear relationship suggests that the changes in enthalpy are accompanied by corresponding changes in both Gibbs free energy and entropy. Gibbs free energy demonstrates values close to zero as moisture content increases. This indicates that dehydration process becomes more spontaneous as Bermuda grass is adsorbed by multilayer water molecules (Bahar, 2017; Polachini, 2016). The slope of this equation represents the isokinetic temperature (T_β) in Kelvin (K), which is the temperature at which all reactions proceed at the same rate (Leffler, 1955). To validate the enthalpy-entropy compensation theory, T_β was compared with the harmonic mean temperature, T_{hm}, which was calculated from Eq. 5 and found to be 302.9 K. The linear relationship between differential enthalpy and differential entropy showed that isokinetic temperature (T_β =522K) was significantly higher than the harmonic mean temperature (T_{hm} =303K) suggesting that enthalpy-entropy compensation theory in our case is valid. Furthermore, since T_β value was much greater than T_{hm}, the sorption process was found to be enthalpy- rather than entropy–driven (Leffler, 1955). Similar results were observed for corn stover (Igathinathane, 2008) and cassava bagasse (Polachini, 2016).

Based on the above, it may be concluded that the Halsey model is a very good predictor of Bermuda grass sorption data with an R^2 of 0.98 and MSE of 2.18. In addition, the linear relationship between differential enthalpy and entropy indicates that higher energy is necessary to remove a specific amount of water at lower moisture contents which may be due to decreased number of available sorption sites followed by the reduced interactions between Bermuda grass and water molecules in the multilayer moisture region. This phenomenon explains the "more spontaneous" water removal process when Bermuda grass contains higher water contents. Moreover, the water adsorption process of Bermuda grass is enthalpy-driven as evidenced by the higher values of isokinetic temperature (T_β = 522°K) compared to harmonic temperature (T_{hm} = 302°K).

Conclusions

Based on the findings of this study, the adsorption isotherms of the grass showed increasing equilibrium moisture content with higher equilibrium relative humidity and lower temperature. It was found that the Halsey model is a very good predictor of Bermuda grass sorption data based on which it may be stated that thermodynamic properties, namely, differential enthalpy and differential entropy, showed substantial reductions with equilibrium moisture content in the range examined. Moreover, the linear relationship between differential enthalpy and entropy indicates that higher energy is necessary to remove a specific amount of water at lower moisture contents which may be due to decreased number of available sorption sites followed by the reduced interactions between Bermuda grass and water molecules in the multilayer moisture region. This phenomenon explains the “more spontaneous” water removal process when Bermuda grass contains higher water contents. Finally, it was concluded that water adsorption process of Bermuda grass is enthalpy-driven as evidenced by the higher values of isokinetic temperature (T_β = 522°K) compared to harmonic temperature (T_{hm} = 302°K).

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