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Moisture sorption isotherms and isosteric heat of sorption of leaves and stems of lemon balm (*Melissa officinalis* L.) established by dynamic vapor sorption

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ABSTRACT

The equilibrium moisture contents (MC) of leaves and stems of lemon balm (*Melissa officinalis* L.) were determined separately at temperatures of 25, 35 and 45 °C over a stepwise increase of relative humidity (RH) ranging from 3 to 90% by an automatic, gravimetric analyzer (DVS system). Equilibrium was achieved within 6 h for most of the target values of relative humidity. The equilibrium moisture content of leaves was significantly higher than that of stems ($p < 0.05$). Differences in moisture sorption capacity between the leaves and stems can be attributed to chemical composition and structure of the tissues. Five three-parameter moisture sorption models (modifications of Chung–Pfost, GAB, Halsey, Henderson and Oswin) were tested for their effectiveness to fit the experimental sorption data. The modified Oswin equation was found to be the best model to describe the adsorption isotherms of both leaves and stems of lemon balm. The recommended MC values of leaves and stems for microbial safe storage at 25 °C were 0.124 and 0.113 kg water per kg dry solids, respectively. The net isosteric heat of sorption was computed from the predicted sorption data by applying the integrated form of the Clausius–Clapeyron equation.

1. Introduction

Lemon balm (*Melissa officinalis* L.) is a perennial herb of the family Lamiaceae, cultivated for its characteristic lemon-scented leaves. It is implemented for several purposes in the food and pharmaceutical industries due to its active ingredients. The essential oil of *M. officinalis* is a well-known antibacterial and antifungal agent (Mimica-Dukic, Bozin, Sokovic, & Simin, 2004) while rosmarinic acid has been identified as the main compound related to its antioxidative and antiviral activity (Wang, Provan, & Helliwell, 2004).

The most common preservation method for medicinal plants is by convective hot-air drying because it not only reduces the moisture content to the microbial safe water activity for storage but also allows the quick conservation of the medicinal qualities of the plant material in an uncomplicated manner (Müller, 2007). The drying process of herbs usually includes low or high mechanization processing levels using flat-bed or conveyor belt dryers respectively (Heindl & Müller, 1997). After drying, the material is packed and kept at indoor climate conditions in the storage facilities until further processing. For the optimization of storage stability, knowledge of the equilibrium relationship between the moisture content (MC) in the plant material and the relative humidity (RH) of the surrounding air at a given temperature is required to prevent decline of quality by microbial or enzymatic activity (Karel, 1975). The moisture adsorption data can be further analyzed to provide a theoretical interpretation of food microstructure and physical interaction between water molecules and the solid matter of a foodstuff (Rizvi, 1986).

The measurement of moisture sorption isotherms usually relies on the static gravimetric method using thermally stabilized desiccators filled with saturated salt solutions as described in a study conducted by Spiess and Wolf (1987). Although this method is still commonly employed, automated humidity generating instruments have been introduced to the market for the continuous determination of sorption isotherms in a dynamic system. For instance, the Dynamic Vapor Sorption (DVS) method is designed to measure the equilibrium moisture content of a material at any desired relative humidity and selected temperatures in a short period of time. It has been applied to measure the moisture sorption properties of cellulosic reinforcement fibers (Kohler, Dück, Ausperger, & Alex, 2003), microcrystalline cellulose (Kachrimanis, Noisternig, Griesser, & Malamataris, 2006), lactose (Vollenbroek, Hebbink, Ziffels, & Steckel, 2010), wood (Hill, Norton, & Newman, 2010)

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and natural fibers (Xie et al., 2011). However, limited attempts have been performed to examine the moisture sorption behavior of foodstuffs (Desmorieux & Decaen, 2006; Roca, Broyart, Guillard, Guilbert, & Gontard, 2007; Ziegleder, Amanitis, & Hornik, 2004) and especially of agricultural products (Argyropoulos, Alex, & Müller, 2011b) using a DVS system.

Recently, comprehensive reviews on sorption characteristics of foodstuffs have been reported in the literature (Al-Muhtaseb, McMinn, & Magee, 2002; Basu, Shivhare, & Mujumdar, 2006). Among the several empirical, partially theoretical and theoretical equations for moisture sorption isotherms (van den Berg & Bruin, 1981), the modified equations of Oswin (Chen, 1988; Oswin, 1946), Halsey (Halsey, 1948; Iglesias & Chirife, 1976a,b), Henderson (Henderson, 1952; Thompson, Peart, & Foster, 1968), Chung–Pfost (Chung & Pfost, 1967a,b; Pfost, Maurer, Chung, & Milliken, 1976) and Guggenheim–Anderson–de Boer (GAB) (van den Berg, 1984; Jayas & Mazza, 1993) models have been commonly applied to fit the equilibrium MC/RH data of various agricultural products and foods.

The moisture sorption isotherms of various leafy materials with medicinal and therapeutic properties have been studied by employing mainly the static gravimetric method for *Olea europaea* L. (Bahloul, Boudhrioua, & Kechaou, 2008; Nourhène, Neila, Mohammed, & Nabil, 2008), *Citrus x hystrix* DC. (Phoungchandang, Srinukroh, & Leenanon, 2008), *Citrus reticulata* B. (Jamali, Kouhila, Mohamed, Idlimam, & Lamharrar, 2006), *Citrus x aurantium* L. (AitMohamed, Kouhila, Jamali, Lahsasni, & Mahrouz, 2005), *Dysphania ambrosioides* L. (Jamali, Kouhila, Mohamed, Jaouhari et al., 2006), *Maytenus ilicifolia* Mart. Ex Reissek (Cordeiro, Raghavan, & Oliveira, 2006) and *Eucalyptus globulus* Labill. (Kouhila, Kechaou, Otmani, Fliyou, & Lahsasni, 2002). Soysal and Öztekin (1999) investigated the equilibrium moisture content of several medicinal and aromatic plants including some herbs of the mint family (Lamiaceae) such as *Origanum majorana* L., *Thymus vulgaris* L. and *Mentha x piperita* L. Additionally, the authors reported another work about the isosteric heat of sorption for selected medicinal and aromatic plants (Soysal & Öztekin, 2001) by dividing them into three major groups. It was found that the magnitude of the sorption heat was dependent on the species and type of the plant group. Moreover, the moisture sorption isotherms of *Mentha viridis* L. and *Salvia officinalis* L. (Kouhila, Belghit, Daguenet, & Boutaleb, 2001), *Mentha crispa* L. (Jin Park, Vohnikova, & Pedro Reis Brod, 2002), *Rosmarinus officinalis* L. (Timoumi & Zagrouba, 2005) have also been documented, however, limited experimental moisture sorption data for lemon balm *M. officinalis* L. can be retrieved from the literature (Argyropoulos, Alex, & Müller, 2011a).

Therefore, the objectives of the present work were (i) to investigate the moisture sorption behavior of leaves and stems of lemon balm using a dynamic vapor sorption apparatus at temperatures typically found in storage and processing of medicinal plants (ii) to find the most appropriate mathematical model to describe the experimental sorption data and (iii) to estimate the isosteric heat of sorption.

2. Materials and methods

2.1. Material

Herbs of lemon balm (*M. officinalis* L.) cultivar Citronella were collected before flowering from an organic farm in Magstadt, approximately 20 km west of Stuttgart (Germany). The material was obtained by cutting the herb manually to a height of about 20 cm above the ground. Prior to experiments, the leaves were picked manually from the stems. The samples were dried in a high precision hot-air laboratory dryer (Argyropoulos, Heindl, & Müller, 2011) at a temperature of 40 °C, maintaining 10 g/kg of specific humidity and an air velocity of 0.2 m/s (Argyropoulos & Müller, 2011) until constant mass was achieved. The dried leaves and stems were kept packed in aluminum coated polyethylene bags and stored at ambient conditions in the laboratory before the determination of moisture sorption isotherms.

2.2. DVS apparatus

The adsorption isotherms of the samples were determined by DVS at the Reutlingen Research Institute, Reutlingen University, Germany using a DVS-1000 gravimetric moisture sorption analyzer (Surface Measurement Systems Ltd., London, U.K.). The instrument essentially consists of a Cahn microbalance with two sample crucibles made of quartz and a humidification system in a temperature controlled chamber. One of the crucibles is used as a reference whereby the other contains the sample to be analyzed. A stream of dry and wet nitrogen gas flows along the crucibles. The relative humidity of the mixture is regulated by two electronic mass flow controllers. The apparatus is computer controlled, allowing pre-programming of stepwise variation of relative humidity at set temperature and continuous measurement of temperature, humidity and mass during the sorption process.

2.3. Experimental procedure

Pre-dried samples of leaves and stems with masses of 12.67 ± 1.63 and 16.11 ± 1.12 mg respectively were used for the experiments. The adsorption isotherms were determined at temperatures of 25, 35 and 45 °C by exposing the material to different values of relative humidity within the range of 3 and 90%. Each sample was first dried by exposure to dry nitrogen until a constant weight of the sample was reached. The dry reference mass was established for both tissues within 16 h. Then, the relative humidity was subsequently increased stepwise whereby the sample weight was equilibrated at each step. Mass, temperature and humidity data were recorded in 2 min time intervals. Equilibrium was considered to have been reached when the change in mass was less than 0.001 mg/min. To obtain the adsorption isotherms, the moisture content of the sample, expressed in kg water per kg dry solids, was calculated at equilibrium of each relative humidity step. Three consecutive measurements for each material and temperature were performed at a total of fourteen target values of relative humidity.

2.4. Mathematical description of sorption isotherms

The five three-parameter moisture sorption equations tested for their accuracy to fit the experimental sorption data are listed below. The models are presented in terms of moisture content, X_e dry basis (kg/kg d.b.), water activity a_w (equilibrium RH/100), temperature T (°C) and a, b, c as model constants.

Modified Chung–Pfost

$$X_e = \frac{-1}{a}\ln\left[-\frac{(T+b)}{c}\ln(a_w)\right] \tag{1}$$

Modified Oswin

$$X_e = (a+b\cdot T)\left[\frac{a_w}{1-a_w}\right]^{1/c} \tag{2}$$