

Moisture sorption properties of matcha tea powder as affected by high humidity storage

Libor Červenka^{1*}, Jiří Palarčík², Vojtěch Zeman¹, and Sali Muriqi¹

¹ Department of Analytical Chemistry,
The University of Pardubice, CZ–532 10 Pardubice, Czech Republic

² Institute of Environmental and Chemical Engineering,
The University of Pardubice, CZ–532 10 Pardubice, Czech Republic

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The effect of high relative humidity (RH) storage on the subsequent moisture sorption of matcha tea powder was examined. The sample of interest was stored above silicagel (control), at 52.9 and 75.3 % RH for two months at 25 °C. Moisture adsorption and desorption experiments were performed using a dynamic vapor system, and the sigmoid-shaped (type II) isotherms obtained with the aid of multi-Freundlich equation. Hysteresis loop disappeared for powder samples previously exposed to high humidity environment. According to Caurie and multi-Freundlich equations, higher critical values of a_w , monolayer content, number of monolayers, and surface area of adsorption were observed for the samples stored under high RH. The change in the particle size distribution was also discussed.

Keywords: Matcha tea; Moisture sorption; Particle size

Introduction

Matcha tea is a special kind of green tea prepared from the leaves of *Camellia sinensis* grown in a shady place, which results in a longer maturation time and thus in the increased quality of the final product [1]. After harvesting, the leaves are steamed, dried, and ground to produce a powder with fine particles.

* Corresponding author, ✉ libor.cervenka@upce.cz

Matcha tea powder is usually used for the preparation of beverages or as an active ingredient in various food products [2,3]. It represents a good source of substances with antioxidant properties including chlorogenic, sinapic, and gallic acid plus selenium [4,5].

Food powder is a form that allows one a wider use in various product preparations. Better solubility in water, the formation of homogeneous mixtures with other ingredients, and better efficiency in the extraction of biologically active substances are the main advantages. On the other hand, the powder is more sensitive to storage temperature and humidity than the unground counterparts. The smaller the particles the larger the surface area, which facilitates the adsorption of moisture from the surrounding environment [6]. This behaviour can be described using the moisture sorption isotherm. Moisture sorption is known to depend on temperature, the particle size, and the main composition of a food material [7]. There are scarce data available on the effect of storage humidity on moisture sorption. In a study by Ho et al. [8], spray-dried camel milk powder stored at different relative humidity has been reported to show changes in its solubility properties. Similarly, fruit powders kept under various humidity conditions showed significant changes in solubility and wettability during rehydration [9].

Knowledge about the moisture sorption isotherm of food powder is a key to understanding the moisture behaviour under a specific condition. The experimental data are usually fitted by various mathematical models to obtain an additional valuable information. The critical water activity, $a_{w,c}$ is an important parameter at which the material undergoes a transition from the glassy to the rubbery state [10]. A multi-Freundlich equation has been proposed for the estimation of $a_{w,c}$, when the total moisture content M was assumed to be given as a sum of the moisture content in the monolayer (M_m) and multilayer (M_M), which can be formulated by the following expression

$$M = M_m + M_M = K_m a_w^{n_m} + K_M a_w^{n_M} \quad (1)$$

where n_m and n_M are the monolayer and multilayer adsorption coefficients, and K_m and K_M are the distribution coefficients. Surface sorption (i.e. monolayer) dominates at relatively low values of a_w (i.e. $K_m a_w^{n_m} > K_M a_w^{n_M}$). In contrast, multilayer sorption occurs at relatively high a_w values, particularly if the excess of water in the fluid phase is to drive the condensation (i.e. $K_m a_w^{n_m} < K_M a_w^{n_M}$). The parameter $a_{w,c}$ is estimated as a cross-over between monolayer- (i.e. surface) and multilayer (i.e. bulk) water adsorption. Monolayer values (M_d), density of adsorbed water (c_d), and specific surface can be estimated from the linearized form of Caurie equation [11]:

$$\ln \frac{1}{M} = -\ln(c_d M_d) + \frac{2c_d}{M_d} \ln \frac{1-a_w}{a_w} \quad (2)$$

A plotting of $\ln\left(\frac{1}{M}\right)$ against $\ln\left(\frac{1-a_w}{a_w}\right)$ followed by linear regression analysis gives an estimation of specific surface of sorption (A , m²/g) and number of adsorbed layers (N) according to the equations:

$$A = \frac{54.45}{\text{Intercept}} \quad (3)$$

$$N = \frac{2}{\text{Intercept}} \quad (4)$$

The main objective of this research was to examine the effect of various humidity storage conditions on moisture sorption properties and the particle size distribution of matcha tea powder.

Materials and methods

Sample preparation

Imbio matcha tea powder (Vieste group, Hradec Králové; Czech Republic) was purchased at a local store. The moisture content and water activity were immediately monitored after opening the package. The former parameter was determined as the change in mass after drying at 103 °C to a constant weight (Kern DLB 160-3A, Kern & Sohn, Balingen; Germany). Water activity was measured at 25 °C using AquaLab TDL (Mettler Group, Pullman, WA, USA). The results were expressed as the mean of three replicates.

An experimental environment with various relative humidity levels (RH) was prepared using desiccators containing silicagel (control), and the MgNO₃, and NaCl slurry, which corresponded to 52.9 and 75.3 % relative humidity, respectively. Approximately 2.0 g of matcha tea powder was spread in a plastic plate (40 mm in diameter) and placed on each desiccator for two months at 25 °C. Salt slurries and silicagel were regularly checked and refreshed when necessary.

Determination of moisture sorption

The dynamic vapor sorption technique (DVS Intrinsic Plus, Surface Measurement System, Wembley; UK) was applied to the determination of the sorption isotherm at 25 °C. Approximately 15 mg of the sample was placed on an aluminium plate hanged on a sensitive analytical microbalance (mass resolution ±0.1 µg) in a closed chamber. The relative humidity of the space surrounding the sample was controlled by air stream (200 ccm) passing through a reservoir with deionized

water. Before analysis, the sample was exposed to dry air (<0.1 % RH) for 180 min to obtain a dry mass. Thereafter, an adsorption process took place from 0 to 85 % RH in the 10 % steps. The mass change of the sample was monitored every 20 s at each RH level until being less than 2.0 µg/min for 20 min. The same procedure was applied to the desorption process. The results were expressed as equilibrium moisture content (EMC) in mg/g of dry mass.

Analysis of particle size distribution

The particle size distribution of matcha tea powder was measured by Morphologi 4 (Malvern Panalytical, Malvern; UK) by dispersing the dry powder (5.0 mm³) on a glass plate. Three magnifications (5×, 20×, and 50×) were used for the measurement. The particle size was expressed as lower (D_{10}) and upper (D_{90}) decils and a median (D_{50}) of the cumulative weight. It means that, for example, 10 % of particles are smaller than the value of D_{10} (software Morphologi, v. 10.10). A span was calculated according to the equation,

$$\text{Span} = \frac{D_{90} - D_{10}}{D_{50}} \quad (5)$$

which reflects the uniformity of the particle size distribution (value approaching 0 for an ideally uniform particle size distribution). The particles were also classified based on their size into four groups as: very small (0–5 µm), small (5–10 µm), medium (10–50 µm), and large (>50 µm). The content of particles in each group was expressed as a percentage of overall particle content. The measurement of particle size was done in duplicate.

Statistical analysis

Non-linear regression (Levenberg-Marquardt iteration algorithm) was applied to obtain parameters of multi-Freundlich (Eq. 1) using OriginPro 9 (OriginLab Corp., Northampton, MA, USA). The goodness of fit of the experimental data was evaluated by the relative percentage deviation (E , %) between experimental (M_e) and predicted (M_p) moisture content using the formula:

$$E = \frac{100}{n} \sum_{i=1}^N \frac{|M_e - M_p|}{M_e} \quad (6)$$

The model is acceptable if the E value is below 10 % [12]. The parameters of Caurie's equation (Eq. 2) were obtained by linear regression with coefficient of determination (R^2) as the indicator of goodness of fit of the experimental data.

Results and discussion

A typical sigmoid-shaped adsorption and desorption isotherms are depicted in Fig. 1 (A–C). Samples stored in an environment of high relative humidity absorbed more moisture compared to the sample held in a dry condition (Table 1). This behaviour was more evident at low a_w values (0.20), where EMC of 30.20 and 33.67 mg/g was observed for samples stored at 53.9 and 75.4 % RH, respectively.

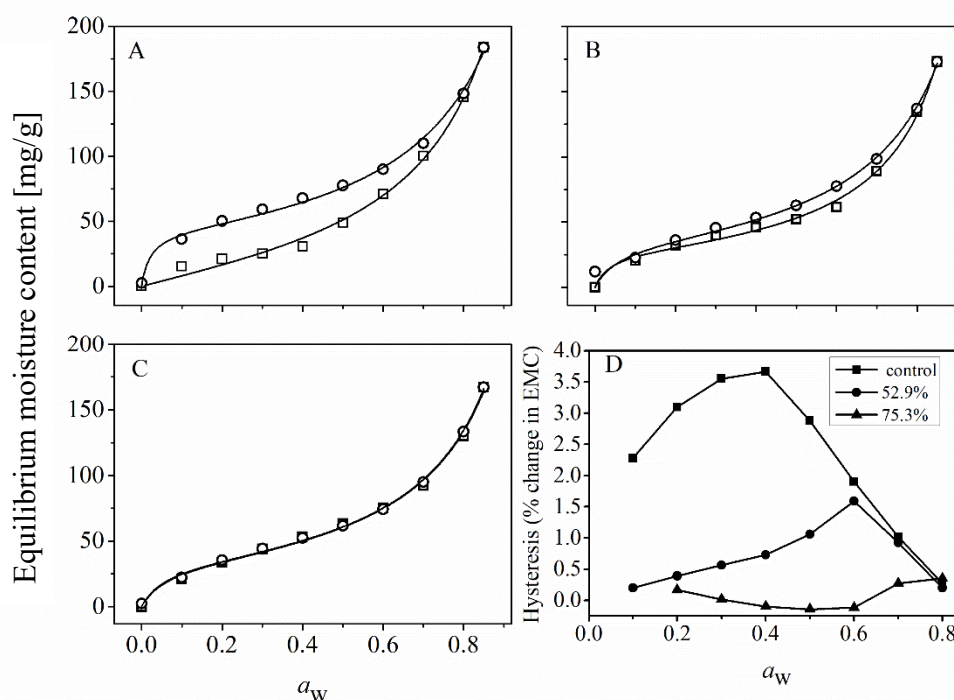


Fig. 1 Moisture adsorption (\square) and desorption (\circ) isotherms (A–C), and hysteresis (D) of matcha tea powder at 25 °C

At the same a_w level, control sample adsorbed lower amount of moisture resulting in EMC of 16.47 mg/g. On the other hand, control sample showed higher amount of moisture at 0.80 a_w during adsorption (145.22 mg/g) compared to samples exposed to higher humidity levels for 2 months, i.e., 132.20–133.47 mg/g).

Table 1 Equilibrium moisture content [mg/g] of matcha tea powder stored under high humidity levels

a_w	Relative humidity [%]		
	control*	53.9	75.3
0.20	16.47	30.20	33.67
0.50	51.08	53.08	60.50
0.80	145.22	132.20	133.47

* stored above freshly dried silicagel

Equilibrium moisture content was higher during desorption process in the whole range of a_w values for the control of the sample. This phenomenon was previously described as sorption hysteresis, and observed in various food products [13,14]. The difference between adsorption and desorption isotherms can be explained by the interaction of solid matrix with the adsorbed moisture. When the relative humidity increases, some additional porous region can be formed via the swelling up of capillaries. Increase of air's vapor pressure results in movement of water molecules towards to the pore interior. Prior to desorption, the pores are initially full of liquid which can escape only when the vapor pressure of the surrounding air is lower than this one occurred inside the pores. The food material may contain pores with different dimeters and water molecules cannot freely diffused, when trapped in pores with small diameter, from interior to the surface [13]. It is evident that hysteresis loop was smaller for matcha tea powder kept at 52.9 % RH (Fig. 1B) and disappeared in case of sample stored at 75.3 % RH (Fig. 1C). Analyses of Fig. 1D showed that hysteresis magnitude had decreased with the increase of storage humidity. The maximal 3.6–3.7 % of EMC difference between adsorption and desorption at 0.30–0.40 a_w was examined for the control sample, whereas only 0.6–0.7 % of EMC difference was found for the powder sample hold under 52.9 % RH.

Multi-Freundlich equation (Eq. 1) was fitted to the moisture sorption data, showing an excellent goodness of fit ($E = 0.77–4.38\%$). As can be seen in Fig. 1, all the sorption isotherms have a sigmoidal shape, characteristic of a type II curve, according to the IUPAC classification [15]. The similar pattern was observed for various kinds of food products, such as paddy rice grains, melon seeds, or yerba mate leaves [13,14,16].

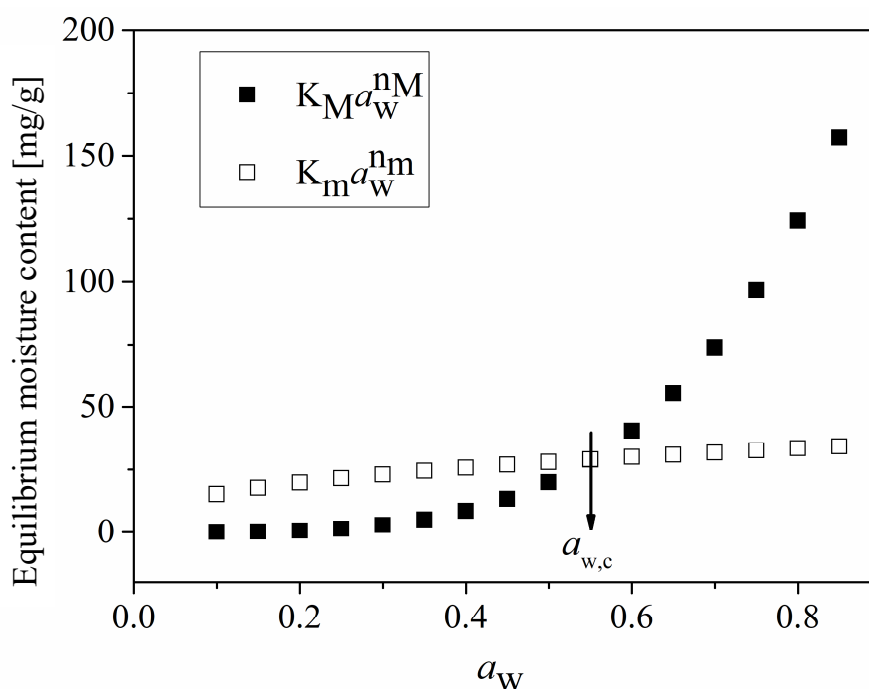


Fig. 2 Illustration of the fitting of the adsorption isotherm by the multi-Freundlich equation (Eq. 1)

The parameters of multi-Freundlich adsorption isotherm were used only to find the critical a_w values, i.e., the point above the multilayer moisture sorption occurred. The intersection of the curves $K_m a_w^{n_m}$ and $K_M a_w^{n_M}$ is depicted in Fig. 2, and it can be seen that the assumption of $K_m a_w^{n_m} > K_M a_w^{n_M}$ is valid below 0.550 a_w (i.e. monolayer adsorption prevails) for matcha powder sample kept above the dry silicagel. The samples previously stored at 52.9 and 75.3 % RH resulted in $a_{w,c}$ values of 0.783 and 0.868, respectively. It also corresponded with the effect of the surface adsorption and numbers of monolayers obtained by linear regression of Caurie equation (Eq. 2). The surface area of adsorption increased from 80.61 (control) to 105.01 and 108.44 m^2/g for matcha tea samples stored at 52.9 and 75.3 % RH, respectively. Number of monolayers was also higher for samples stored at high relative humidity in comparison with the control (Table 2).

Table 2 Properties of sorbed water for matcha tea powder at 25 °C using Caurie equations (2–4)

Caurie	Relative humidity level [%]		
	control*	52.9	75.3
M_d [mg/g]	20.19	23.32	23.85
C_d [g/cm ³]	0.68	0.60	0.60
S	0.68	0.52	0.50
A [m ² /g]	80.61	105.01	108.44
N	2.96	3.86	3.98
R^2	0.97	0.97	0.99

*stored above freshly dried silicagel;

M_d , monolayer moisture content; C_d , density of adsorbed water; S , slope of the linear relationship between $\ln\left(\frac{1}{M}\right)$ and $\ln\left(\frac{1-a_w}{a_w}\right)$ (Eq. 2); A , surface area of adsorption (Eq. 3); N , number of adsorbed layers (Eq. 4)

It may suggest one that a swelling of particles had occurred and new sorption sites were exposed to an interaction with water molecules when being stored at a higher relative humidity environment. The monolayer value also slightly increased from 20.19 mg/g (control) to 23.32–23.85 mg/g for the samples previously exposed to higher humidity. Further, the density of water reflects the interaction between molecules of water, i.e., the closer the intramolecular distances the greater forces acting between them and therefore giving rise to a higher density of water [11]. The interaction between water molecules and adsorbate may reduce the intramolecular distance, thus increasing the density of water. Matcha tea powder held in an environment with higher humidity showed a weaker interaction between water molecules and adsorbate (0.60 g/cm³) in comparison with the control (0.68 g/cm³).

The density of values lower than that for pure water might be due to the presence of relatively higher amount of lipids (4.1–8.2 %) in matcha tea powder [4]. The storage of matcha tea samples at higher-humidity environment influenced their particle size distribution. The substantial changes of particle size distribution were observed after two months of storage at higher humidity. Very small particles and small particles decreased from 14.5 and 49.6 % (control) to 5.2 and 38.4 % (sample stored at 52.9 % RH) and to 6.7 and 36.7 % (sample stored at 75.3 % RH), respectively. The content of medium particles increased from 35.6 % to 55.8 and 56.0 % for matcha tea powder kept at 52.9 and 75.3 % of RH, respectively (Fig. 3). This behaviour can be attributed to the formation of aggregates during storage at higher humidity.

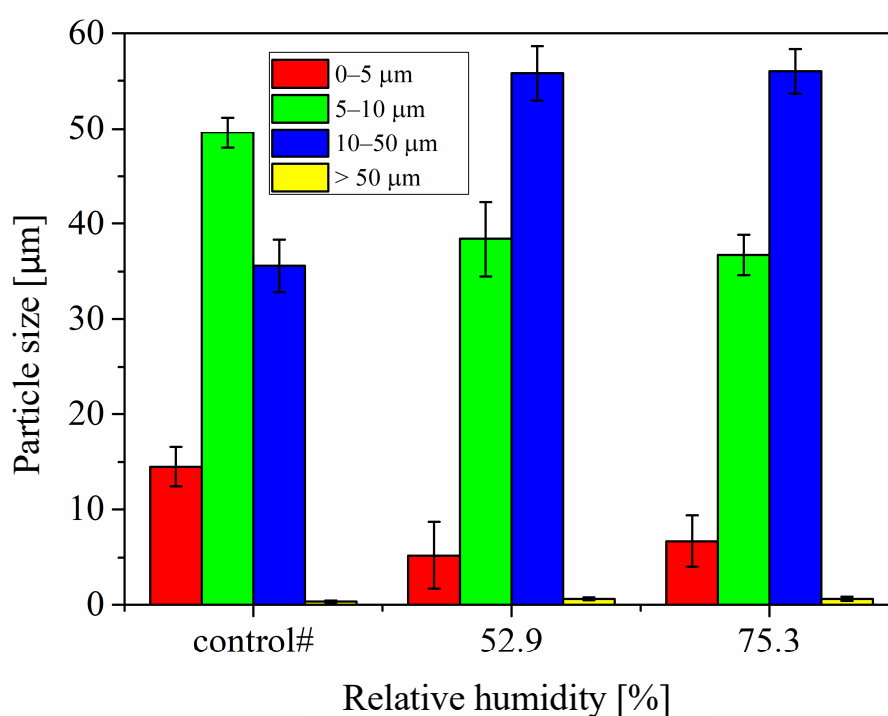


Fig. 3 Particle size distribution of matcha tea powder after 2 months of storage at various relative humidities (# stored above freshly dried silicagel)

The increase of moisture content (and water activity) of food powder resulted in the change of flowability by formation of liquid bridges between solid particles [17]. When the powder matrix is saturated with liquid, the dissolution of the particles may occur, and thus aggregates can be formed. Gaudel et al. [9] reported on the increase of span values for various fruit powders stored at 0.35 a_w , which was in accordance with our study. The span values increased from 1.27 (control sample) to 1.93 and 2.46 for samples stored at 52.9 and 75.3 % of RH, respectively, for matcha tea powder.

Conclusions

Moisture sorption isotherms of matcha tea powder were of a sigmoid shape, and multi-Freundlich equation fitted well to the experimental data. Hysteresis loop was apparently observed for a sample stored in a dry environment. The hysteresis gradually disappeared for samples previously kept at 52.9 and 75.3 % of relative humidity for two months. Those matcha tea powder samples also exhibited higher monolayer values, number of monolayers, and surface area for adsorption, but lower density of adsorbed water. It also corresponded with higher critical a_w values obtained from multi-Freundlich equation. After two months of storage at higher relative humidity, very small and small particle contents decreased, while medium particle content increased in comparison to that of the control sample. The distribution of particles exhibited an increase of heterogeneity suggesting us the formation of aggregates. Despite the above-summarised results, a detailed study is needed yet to fully elucidate the role of high humidity on the functional properties of matcha tea powder.

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