

Hygroscopic Properties of Green Coffee Transported by Sea

M. Ruszkowska¹, P. Dmowski¹ & K. Urbanowicz²

¹ *Gdynia Maritime University, Gdynia, Poland*

² *Mega Coffee, Kwidzyn, Poland*

ABSTRACT: The aim of research was to evaluate the hygroscopic properties of green coffee beans determining its quality during sea transportation. The research material consisted of seven samples of unroasted bean coffee from different countries of origin (Kenya, India, Ethiopia, Columbia and Guatemala) obtained directly from the coffee producer. The water content of green coffee must not exceed 12% as it then has a tendency to grow mould and become musty. Coffee beans require particular temperature, humidity/moisture and possibly ventilation conditions and therefore, to explore and predict the behaviour during transport of green coffee its equilibrium moisture content must be determined for a range of transport temperatures and relative humidity levels. The present paper focuses on the evaluation of the hygroscopic properties of green coffee from different countries of origin based on isotherms of water vapour sorption and characteristic selected parameters of the surface microstructure determining transport conditions and the microbiological stability of this cargo. While assessing the quality and transport durability of green coffee beans, based on the parameters of monomolecular layer capacity and the specific surface area of sorption, it was found that especially samples of coffee from Guatemala and Ethiopia were the least susceptible to changes in the transport conditions.

1 INTRODUCTION

Coffea Arabica and *Coffea Canephora* var. *Robusta* there are two main species of coffee. About 60% of the world's coffee production is Arabica and 40% is Robusta. The largest Arabica coffee producers are situated in Brazil, Colombia and Ethiopia. In turn, the largest producers of Robusta are Viet Nam, Uganda and India [11]. According to Goneli et al. [6], in the world there are two main methods of processing coffee beans: dry method and wet method. The applied process significantly influences the selection of appropriate the coffee beans storage and transport conditions.

According to the data presented by the International Coffee Organisation (ICO), global coffee

production has increased by more than 60% since the 1990s. The value of annual coffee exports during last 30 years has increased more than four times from 8.4 billion USD in 1991 to 35.6 billion USD in 2018. The global coffee trade is characterized by two main markets: the commodity market, which mainly offers standard-quality bulk coffee produced in high volumes and the specialty market, which offers higher prices for coffee exporters handling lower volumes of higher-quality coffee beans [11].

Coffee is one of the most popular beverages consumed in the whole world. According to data presented in the Coffee Development Report [11], Europe's coffee consumption in years 2019/20 was estimated 55.09 million bags and has grown by an average annual rate of about 1.3% from 38.41 million

bags in year 1990/91. The European Union (EU) is by far the largest coffee consumer globally and accounts for 82% of regional demand within the continent. In 2019/20, coffee consumption in the EU amounted to 45.05 million bags. Among the EU members, Germany, France, and Italy are the three largest consumer countries, accounting for 45% of total consumption in whole Europe. Around 70% of the European Union coffee imports include unprocessed coffee imported mainly from Brazil, Viet Nam and Honduras. More than 90% of coffee is still shipped in green form. However, it must be stressed that, since the beginning of 2020, the Covid-19 pandemic has exacerbated the situation of producing countries affected by low prices and volatility. The global spread of the virus, in addition to its dramatic effect on public health, has resulted in supply chain disruptions and influenced global demand for coffee.

Coffee is transported from producer to consumer nations in bulk or in sacks, mostly in containers holding to 22 tonnes. During the transport even well-dried coffee contains a great deal of water (about 12%) that, as long as it remains evenly distributed, do not poses the problem. However, temperature fluctuations, that may occurred during sea transport, can cause condensation of water. And as a consequence of re-wetting fungal and moulds may growth [7]. Because coffee beans are sensitive to moisture they are usually shipped in jute or sisal bags which allows free circulation of air [2]. The net weight of bags depend on the country of origin of coffee beans. Coffee bags from Africa are generally 60 kg but bags from Central America, especially from Colombia may weigh 69 kg. Over 90% of the European coffee imports are already transported in ventilated containers. In sea transport, it is very important to assign the cargo to a particular class of storage climate conditions (SCC group). It is carried out based on the requirements that cargo during the transport process. In sea transport coffee like tea requires particular temperature, humidity and ventilation conditions. Green coffee belongs to the group of loads with the third biotic activity, for which respiration is essentially suspended, but biochemical and microbiological processes are continue. Green coffee belongs to the group of loads with the third biotic activity, for which respiration is essentially suspended, but biochemical and microbiological processes are continue. Inadequate ventilation may result in fermentation and rotting of the coffee beans. It is the result of increased CO₂ levels and to low supply of atmospheric oxygen [33].

Taking into consideration the required transport conditions, coffee is classified in the SC VI group of cargo. This group includes cargoes with a low and medium water content from 1.5% to 30% in dry mass. [28]. In general, temperatures during transport of green coffee beans should range between 10 and 20°C, because there is a connection between fluctuations in the ambient temperature and the formation of condensation water in the container. During the transport from a hot climate (e.g. Brasil) to a cold climate (e.g. Poland) the high temperature gradients are possible and therefore. It is very important that the temperature inside of container with coffee should not drop below zero Celsius degrees, because a sudden drop in temperature leads to higher probability of

condensation water formation in the container and directly on the cargo and thus causes considerable cargo losses. Therefore the preventing the formation of condensation water in the hold is essential and constitutes the number one priority during sea transportation. Hence, the cargo in the container should be protected from dripping water by the use of appropriate mats. To ensure adequate ventilation, a sufficient distance the spacing from the cargo is also necessary. As per the sorption isotherm for green coffee beans, with 8.5 – 10% water content are at equilibrium with relative humidity of 50 – 65%. If coffee beans have an excessively high moisture content, there is a risk of mustiness, mould growth and post or overfermentation [9, 34].

According to Baptestini et al. [1] temperature and relative humidity of the environment surrounding the product are the primary parameters to be considered during transport. Due to its low moisture content, roasted and ground coffee may absorb moisture from the environment, causing clumping and thus increasing economic losses. The key metric for assessing water sorption during transport and storage are the sorption isotherms.

Taking into account all the above facts, it is necessary to know the relationship between the air temperature and relative humidity, and desirable conditions for transport the product. To obtain this information, sorption isotherms are indispensable, which are efficient tools to determine thermodynamic interactions between the water and food components. [6].

Given these reasons the objective of this study was to determine the adsorption isotherms of green bean coffee. Additionally, this study sought to determine the thermodynamic properties of water sorption as a function of water activity (a_w).

2 RESEARCH MATERIAL AND METHODS

The research material included seven green coffee bean samples imported to Poland by sea from India (samples I, II and V), Ethiopia (samples III and IV) and one sample from Columbia (VI) and Guatemala (VII). The samples of green coffee bean were processed in two different ways: dry method (Ethiopia IV and India V) or natural form and wet method (India I, II, Ethiopia III, Columbia VI and Guatemala VII).

The tested green coffee bean samples were subjected to a preliminary analysis by the determination of the initial water content. Each sample's moisture content was determined by oven-drying method as recommended for ISO standard ISO 6673 - 2005: Green coffee – Determination of loss in mass at 105 degrees Celsius. For water activity (a_w) measurement, AquaLab 4TE apparatus with an accuracy of ± 0.003 at 20°C was used (AS4 2,14.0 2017, Decagon Device Inc., Pullman, WA, USA).

Isotherms of adsorption were determined by the static method, based on moisture equilibrium between the tested product samples and the atmosphere of defined relative humidity, adjusted by means of salt

solutions [5]. The determination of adsorption isotherms was carried out at $20^{\circ}\text{C}\pm 1^{\circ}\text{C}$ in a range of water activity $a_w=0.07\div 0.98$. Time fixture equilibrium was 30 days. Based on the initial weight of the product and the growth or loss of water content equilibrium water content was calculated and sorption isotherms were plotted.

For the purpose of mathematical description of empirically determined sorption isotherms and performance of sorption feature characterisation of the tested green coffee bean samples, sorption isotherms rearrangements were made by using the BET equation (1). The water activity (a_w) ranged from 0.07 to 0.98 [22].

$$V = \frac{V_m C a_w}{(1 - a_w) [1 + (C - 1) a_w]} \quad (1)$$

where:

a_w – water activity [-],

V – adsorption [g H₂O/100g d.m.],

V_m – monolayer water content [g H₂O/100g d.m.],

C – constant energy [kJmol⁻¹].

The sorption properties research results were developed using Microsoft Office Excel 365 computer programs. The choice of the classic BET model was determined by the recommendation of the International Union of Pure and Applied Chemistry (IUPAC) [32].

The fitting of empirical data to the BET equation was characterised based on the value of determination coefficient (r^2), fitting of standard error (FitStdErr) and statistics value F, calculated also using Jandel. Table Curve 2D v 5.0,1 software [13].

In order to determine the suitability of the model for the description of obtained adsorption isotherms, analysis of the mean square error (RMS) expressed in % was performed, which was calculated based on the equation:

$$RMS = \sqrt{\frac{\sum \left(\frac{z w_e - z w_p}{z w_e} \right)^2}{N}} * 100 \quad (2)$$

where:

$z w_e$ – experimental activity, [g H₂O/100g d.m.],

$z w_p$ – computed activity, [g H₂O/100g d.m.],

N – the observation number.

Based on the water content estimated in the monolayer adsorbed at a temperature lower than the boiling temperature and the so called water cross - section", the specific surface area of adsorbent was calculated according to the equation (3) [22]:

$$a_{sp} = \omega \frac{V_m}{M} N \quad (3)$$

where:

a_{sp} – sorption specific surface, [m²/g d.m.],

N – Avogadro number, [6.023×10²³ molecules /mol],

M – water molecular weight, [18 g/mol];

ω – water setting surface, [$\omega = 1.05 \cdot 10^{-19}$.m²/molecule].

3 RESULTS AND DISCUSSION

The determination of water content and activity are the basic criteria for assessing the quality and evaluation of products transported by sea. The level of water content and activity of green coffee beans is mostly influenced by the method of post-harvest treatment of the grain, storage and transport conditions. Green coffee beans, as tea leaves, constitute very hygroscopic plant material and they are very susceptible to water. During the green coffee beans transportation process, the relative humidity of the surrounding air could be increased, then beans of coffee tend to absorb water. On the other hand, as the relative humidity drops, green coffee beans tend to desorb water. Very important is also to along with water sorption, the thermodynamic sorption properties of the material should be monitored by assessing the water activity (a_w). According to Baptestini et al. [1], the thermodynamic properties of agricultural products is a key resource that can provide information for assessing the effect of a_w on product storage and transportation and they can aid in the understanding of the adsorbed water properties and the study of physical phenomena that occur on product surfaces. Table 1 presents the mean water content and water activity of green bean coffee samples.

Table 1. Corresponding water content and water activity values for green bean coffee samples

Product	Mean water content [g/100 g d.m.]	SD	Water activity [-]	SD
I (India)	9.8712	0.0003	0.6563	0.0015
II (India)	8.8775	0.0001	0.6218	0.0012
III (Ethiopia)	8.8795	0.0014	0.5797	0.0012
IV (Ethiopia)	8.8685	0.0003	0.5973	0.0037
V (India)	10.3785	0.0006	0.6876	0.0007
VI (Columbia)	10.6678	0.0010	0.6816	0.0003
VII (Guatemala)	8.3880	0.0007	0.5846	0.0027

Abbreviation: SD –/standard deviation

Source: Own correlation (n=3).

Table 1 indicates that the obtained values of the initial water content of all samples of coffee was consistent with the literature data determining the water content in coffee and did not exceed the recommended content of 8-12% [10, 29]. Based on the obtained results it can be assumed that the water content was determined primarily by the country of origin and different processing procedures. Despite the water content in accordance with the ICO standards, coffee beans from India (I, II and V), as well as coffee beans from Columbia (VI), were characterized by a high level of water activity, which could indicate a relatively low degree of water bonding with the dry matrix of green coffee beans raw [20, 21, 24].

The high values of the initial water activity in all green coffee samples could indicate low microbiological stability of the tested products. The probable reason for this was the adsorption of water on the surface of coffee beans resulting from the phenomenon of condensation occurring as a result of the daily temperature fluctuation during sea transport.

Moisture content can be the principal parameter for assessing the current status of a coffee lot. Transport of coffee can be considered as an extension of storage of coffee but at the same time introduces practical challenges in meeting the storage strictures of avoidance of re-wetting and maintaining the temperature on the constant level. Even well-dried coffee contains a great deal of water that, as long as it remains evenly distributed, poses no problem. However, the fluctuations of temperature can cause condensation and local re-wetting and lead to fungal and their metabolites - toxins (especially Ochratoxin A) outgrowth. According to ICO, Ochratoxin A is a heat-stable mould metabolite produced by a few species *Aspergillus* especially *Aspergillus ochraceus*, which which develops in environments with temperature ranged from 8 to 37°C with the optimum temperature between 24 and 31°C. The minimum water activity for its development amounts to 0.76 at 25°C, with the optimum a_w between 0.95 and 0.99. Although *Aspergillus ochraceus* develops from a water activity of 0.76 then the optimal level of a_w to the toxins produced in green coffee beans is between 0.85, and 0.97. Ochratoxin A (OTA) can occur in raw and roasted coffee beans. Additionally the toxins produced by these fungi survive roasting and could present a potential hazard for health. [8, 30].

Additionally, excess water can cause the hydrolysis of some aroma compounds, the adverse effects of oxygen, and polymerization of the aromatic compounds contained in coffee.

The characteristics of the green coffee beans hygroscopic properties were determined by comparing the position of water vapour sorption isotherms (Fig. 1). An sorption isotherm is a graphic representation of the relationship between water mass per dry matter and water activity in a constant temperature and describes the state of dynamic equilibrium of the system. Knowledge of the shape of isotherm allows to evaluate the mechanism determining the water binding process in the product, identify the product sensitivity to moisture and predict changes during the storage of product. Results obtained from equilibrium moisture studies are important for knowing how a material absorbs and loses moisture during storage, and for defining the transport conditions in order to maintain the best quality of product [12, 31]. Sorption isotherms are also efficient tools to determine thermodynamic interactions between water and food components. [6, 17, 20].

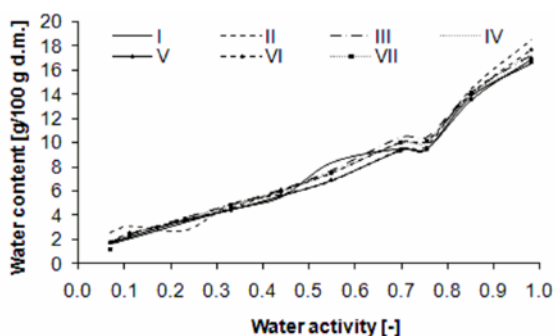


Figure 1. The sorption isotherm of water vapor product I - VII, determined after 30 days of storage in environment with $a_w=0.07-0.98$. Source: Own correlation.

Sorption isotherms determined empirically in the tested green coffee beans were characterized by sigmoidal in shape and, the classification by Brunauer showed similarity to the isotherms of type II. These results are similar to those found by Dmowski and Ruskowska [4], Menkov [15, 16] and Pałacha and Karwowski [23], working respectively with black tea leaves (*Camellia sinensis* and *assamica*), lentils (*Lens esculenta*), chickpeas (*Cicer arietinum*) and bean (*Phaseolus vulgaris*).

The sigmoidal shape of the sorption isotherms is related to the occurrence of the range of monolayer adsorption in the low water activity ($a_w<0.3$), the multilayer adsorption range ($0.3<a_w<0.75$) and the capillary condensation area ($a_w>0.75$). Developed sorption isotherms in the tested coffee samples, characterized by continuity in the course of the entire range of water activity ($0.07\div 0.98$) with increasing water activity increased the water content in the analyzed green coffee beans.

Based on the analysis of the course of empirically determined isotherms, in the range of water activity $a_w = 0.07\div 0.55$, in green coffee beans from India (II), Ethiopia (III and IV) and Guatemala (VII), water desorption process was found, while above the range of environmental water activity an adsorption process was found.

It can be assumed that such a course of desorption and adsorption process in the tested green coffees was determined primarily by the water content in the tested products and its state related to the chemical composition of products.

In wet-processed green coffee beans from India (I) and Colombia (VI) and dry-processed coffee from India (V), characterized by a high initial water content and water activity (Table 1), the desorption process involved monomolecular sorption ($a_w=0.07\div 0.33$) and the multilayer sorption range ($a_w=0.44\div 0.75$) (Fig. 1). After exceeding the water activity $a_w=0.75$, in all tested samples of green coffee beans I-VII, significant intensification of the adsorption phenomenon was found, expressed by an increase in the rate of water vapour absorption process, which indicated the initiation of capillary condensation. This phenomenon can be equated with exceeding the level of critical moisture, which determines the loss of product's ability to be stored further [20].

By comparing the course of sorption curves in a common frame of reference it was found that coffee beans from India II (wet method production) had a higher position, indicating greater hygroscopicity (starting from water activity $a_w=0.85$). According to the literature data, the course of sorption isotherms could probably be affected by the diverse chemical composition of the I-VI products, but above all by their structure (Fig. 1). When assessing the sorptive properties of lyophilized strawberries, Ciużyńska and Lenart [3], found that they have a sigmoidal shape characteristic of most food products, corresponding to type II isotherms. In turn, Moraga, Martinez-Navarrete and Chiralt [18] for freeze-dried strawberries obtained a sorption isotherm course typical of products with high sugar content. Probably, it is connected with the slow changes in equilibrium

water contents at low water activity and rapid increase of water activity above 0.5.

In order to determine the selected parameters of surface microstructure, the empirical data was subjected to the use of the Brunauer, Emmett and Teller equation BET (1). The course of sorption isotherms in the water activity range from 0.07 to 0.33 enabled to determine the parameters of BET equation (v_m , c_e) by assaying the degree of its fit (R^2 , RMS, SKO) to empirical data. The results were presented in Table 3.

According to the literature data, the mean square error (RMS) is lower than 10% and this indicates a good agreement of the model fit to the data in the studied range of water activity [20, 23]. Based on the research, it was found that the calculated RMS in green coffee beans I-VI ranged from 3.66% to 9.04%. This confirmed that the parameters of the BET model mathematically properly describe the process of water vapor sorption on the surface of evaluated green coffee beans. Only in the case of green coffee beans from Guatemala (VII), the RMS value exceeded 10% - indicating a smaller fit of the model.

Monolayer capacity (v_m) determined based on the BET equation, corresponds to a single layer of molecules adsorbed water vapour and is referred to as an indicator of the availability of polar water vapour independently, the component of which is a source of hydrophilic groups [14]. Theoretically, the water content of the layer corresponds with the optimum amount of water in the product and indicates the quality of the storage and transport stability. The monolayer moisture content is the safest moisture content for storage and transport purposes, because at a certain temperature it provides the increased time period with minimum quality loss. Values below monolayer moisture content slow down the respiration rate of the product and the action of pests [6]. On the other hand, the excess of water in relation to the monomolecular layer leads to achieving the critical humidity, the excess level of which may cause undesirable changes in the product, especially the development of microorganisms and toxins produced by fungi (OTA), that are very dangerous. [8, 14, 19, 26].

The determined values of monolayer v_m range from 3.06 g H₂O/100g for coffee originated from India to 4.77 g H₂O/100g for coffee from Guatemala (Table 3). The obtained results showed that a greater surface area of sorption was characterized by the Guatemalan coffee processed by wet method, thus coffee from this

region was characterized by the highest storage and transport stability.

According to Babtestini et al. [1] the C constant is associated with the chemical potential differences between the monolayer and superior layers. Constant energy gives information about the difference between the enthalpy of desorption monolayer and the enthalpy of vaporization of the liquid adsorbent. The results of constant c_e ($c_e \geq 2$) confirm the sigmoidal shape of the curve of adsorption and suggest that in the tested products only a process of physical adsorption was observed [27]. The lowest value of constant c_e was characteristic of the coffee from Guatemala, which indicated a lower amount of heat released from the product in the sorption process.

The determined values of monolayer capacity (v_m) based on the BET model constituted grounds for the calculation of the sorption specific surface (Table 1). The highest value of monolayer capacity based on the BET model was determined in coffee from Guatemala (VII) - 167.7468 m²/g, and the smallest in coffee from India (II) - 107.6927 m²/g. Thus, it is possible to assume that the highest stability and storage stability determined by the value of monomolecular layer, was characteristic of the samples from Guatemala (VII). Based on the obtained results, it can be assumed that the differences in the microstructure of the product surface were determined by the different treatment process parameters used by the producers of the tested products and a different raw material composition of the tested green coffee beans. It can also be assumed that the process of technological processing as well as the process of transporting products by sea caused changes in the structure of the evaluated green coffee beans. Probably the products imported from India (I, II, V), Ethiopia (III, IV) and Colombia (VI) were characterized by a smaller number of active sites capable of attaching water molecules. According to Ociczek [21] and Ruskowska [25] the size of sorption specific surface is the product of total surface area and its affinity for water molecules determined by the distribution of functional groups.

The research also included the measurement of water activity of green coffee beans, performed after 30 days of storage in hygrometers with constant relative humidity covering the range of water activity between 0.07 and 0.98 (Fig 2.).

Table 3. BET equation parameters and specific surface of sorption [m²/g]

Product	v_m [gH ₂ O/100g]	c_e	R^2	RMS [%]	SKO	Specific surface of sorption [m ² /g]
I (India)	3.6806	8.7084	0.9944	8,5062	1,1668	129.3134
II (India)	3.0652	27.0375	0.8110	3,6630	1,3673	107.6927
III (Ethiopia)	3.9922	9.5233	1.0000	9,0408	1,4655	140.2632
IV (Ethiopia)	3.6904	11.5388	0.9971	4,7529	0,8982	129.6601
V (India)	3.5597	10.7666	0.9973	5,7973	1,0111	125.0669
VI (Columbia)	3.5918	11.6868	0.9974	8,5602	1,2313	126.1958
VII (Guatemala)	4.7745	4.7595	0.8778	28.5784	1,9110	167.7468

Abbreviation: R^2 - determination coefficient; FitStdErr - standard error; SKO - sum of squared deviations
Source: Own correlation.

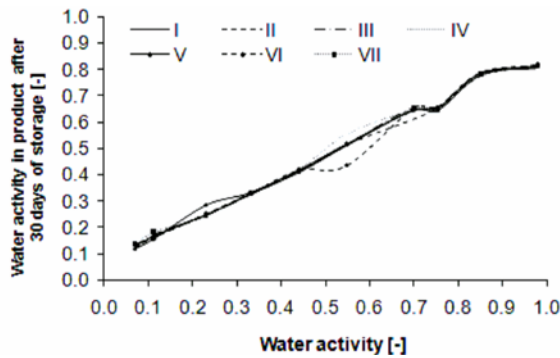


Figure 2. Water activity of I - VII green coffee, determined after 30 days of storage in the range of a_w from 0.07 to 0.98, at 20°C.

Source: Own correlation.

Fig. 2 provides that the greatest differences between environmental water activity and water activity measured in green coffee beans were observed in the environment with water activity ($a_w=0.75\div 0.98$). Adsorption processes dominated in this range of water activity. It was found that the time of storage and possible transport of raw materials were too short for the investigated raw materials to reach a state of equilibrium in higher environmental water activities.

Evaluation of the analyzed parameters is of fundamental importance to the correct transportation of the product, providing necessary information to design the climatic conditions such as temperature or relative humidity.

4 CONCLUSIONS

1. Under the obtained results, it can be concluded that the examined green coffees, despite having a similar initial water content in line with the Code of Practice guidelines, were characterized by a high level of initial water activity.
2. High water activity in the green coffee beans imported to Poland by sea from India (I, II and V), as well as coffee from Colombia, may indicate the process of mouldy grains and thus contribute to the formation of Ochratoxin A (OTA).
3. Assessing the quality and transport durability of green coffee beans, based on the parameters of the monomolecular layer capacity and the specific surface area of sorption, it was found that coffee imported from India (II) was the least susceptible to changes in the transport conditions.
4. Taking into consideration the quality, storage and transport stability of the tested green coffee beans, the coffees processed by wet method, imported from Guatemala (VII) and Ethiopia (III) have had the best sorption properties. These were the samples with the highest monomolecular layer capacity.

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