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INTERNAL STRUCTURE AND WATER TRANSPORT IN THE COFFEE BEAN

Ramirez-Martinez A.^{1,2}, Benet J.C.¹, Cherblanc F.¹, Garcia-Alvarado M.A.², Rodriguez-Jimenes G.²

¹Laboratoire de Mécanique et Génie Civil, Université Montpellier 2, CNRS
Place Eugène-Bataillon 34095, Montpellier cedex 5, France
Tel.: +33 67149643

²Chemical and Biochemical Engineering Department, Instituto Tecnológico de Veracruz
Av. Miguel A. de Quevedo 2779, 91860 Veracruz, Ver. México
Tel.: +52 229 9345701, E-mail: ramirez@lmgc.univ-montp2.fr

Abstract: Microscope observations gave evidence of the coffee bean structure heterogeneity, whereas, coffee bean isotherms of different parts of grain show not significant difference excluding the parchment. Diffusion tests allowed determining the relationship between the diffusion coefficient and the water content. Therefore we propose an original method to determine the water transfer resistance in different parts of grain including the parchment. The set of results will permit to simulate a coffee grain drying and to have information about the zones at high water activity. This information would help to diminish the risk of fungus development.

Keywords: Grain structure, activity of water, diffusion coefficient, parchment.

INTRODUCTION

Coffee is the second most important commercial product in the world (Kouadio *et al.*, 2007). Morphologically, its beans are covered by different layers or structures (Fig. 1). At drying the external structures are eliminated and only the endocarp or parchment and the silver skin envelop the endosperm or grains. Coffee bean drying has also been identified as a step in coffee processing that could lead to the surface development of some fungus. For instance, *Aspergillus ochraceus* produces a toxin, named Ochratoxin A (OTA) that has carcinogenic and immunotoxic properties (Suárez-Quiroz *et al.*, 2004; Kouadio *et al.*, 2007; Taniwaki *et al.*, 2003; Palacios-Cabrera *et al.*, 2004; Frank, 2001).

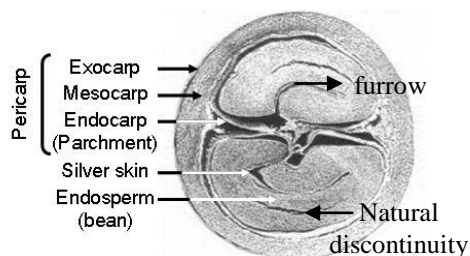


Fig. 1. Coffee bean structure (Nestle Corp.).

The structure of coffee bean has been extensively studied, usually in the transversal direction, finding that its cellular structure is different in different

regions of the coffee bean (De Castro and Marraccini, 2006; Sutherland *et al.*, 2004; Eira *et al.*, 2006). It suggests that the coffee bean has an anisotropic cellular structure, while the grain is often considered as an isotropic media when coffee drying is studied. One consequence of anisotropy of the structure could be the anisotropy of water distribution. Even when *Aspergillus ochraceus* develops principally at the surface of the bean, it can produce spores that could set at the endosperm (Fig. 1). It has been showed that water activity values less than 0.8 (Suárez-Quiroz *et al.*, 2004), prevents its development; however this is an average value. To evidence the homogeneity or heterogeneity of water content distribution throughout the grain will clarify the role of drying in this problematic. If a water content heterogeneity of coffee bean exists it should be important to determine possible regions where the fungus could grow and therefore produce the OTA (Suárez-Quiroz *et al.*, 2004; Kouadio *et al.*, 2007; Taniwaki *et al.*, 2003; Palacios-Cabrera *et al.*, 2004; Frank, 2001). By the other hand, if the heterogeneity of water distribution is confirmed, water activity should be considered at modelling instead of water content to better describe the process.

The heterogeneity of water content distribution has been studied by different authors using the same technique: Nuclear Magnetic Resonance (RMN). However, they lead to different results. Frank (2001) shows by images took by RMN that the grain water distribution is heterogeneous at a high value of water

activity (Fig 2). In the images the water content is higher in one side of the coffee bean than in the other. If Frank didn't propose an explanation for this water distribution this image suggests that this side should have a different composition or structure.

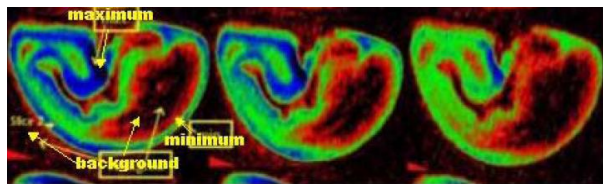


Fig. 2. Coffee water distribution obtained by NMR (Frank, 2001).

On the other hand, Toffanin et al (2000) reveals that the water distribution in rehydrated coffee is homogeneous. They also calculate the diffusion coefficient in different zones of coffee bean finding different values, attributing this behaviour to the presence of mucilage at the natural discontinuity of bean (Fig. 1) whom would restrict water movement.

Even though it has been made efforts to determine the distribution of water content, this is not clear yet. To understand the precise effect of water distribution, it seems essential to model the water transfer in a grain taking into account the heterogeneity of structure and of its properties.

This communication aims to clarify the geometry of different parts of the grain using observations. Then it presents a study of the relationship between water activity and water content in different parts of the grain. Finally, we propose an experimental study of water diffusion coefficient in the whole grain and from a part of the grain. These informations will allow us to simulate the drying process.

INTERNAL STRUCTURE

Coffee samples observations

Often, the coffee cuts reported in literature are made only in the transversal direction. In this sense, coffee cellular heterogeneity has been reported (Sutherland et al., 2004, De Castro and Marraccini, 2006). In order to have a more information about the cellular distribution, cuts in the longitudinal direction were made. For the observation of coffee cuts, coffee beans were cut longitudinally and transversally, then they were stained by immersion in safranin for 30 min, followed by washings with ethanol of different concentrations, and left in contact with fast green for 10 min, finally they were washed in pure ethanol and observed in an optical microscope. Both, transversal and longitudinal cuts were made at different parts in coffee bean, showing a difference when the cuts made near the border that in the middle part (Fig. 3, 4, 5 and 6) and confirming the heterogeneity of cell structure in both directions.

In the other hand, another structure, the furrow, was

studied. For this purpose, images were taken by a stereoscope Olympus SZ-CTV in fresh and dried state.

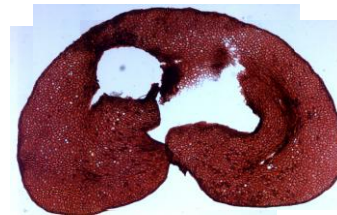


Fig. 3. Transversal coffee bean cut near the border.

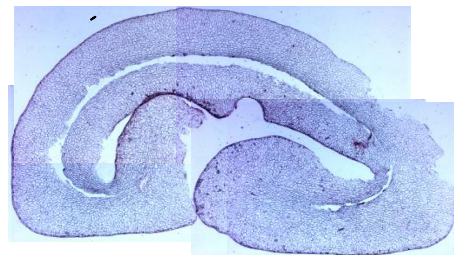


Fig. 4. Transversal coffee cut at the middle part of the endosperm.



Fig. 5. Longitudinal coffee cut near the border.

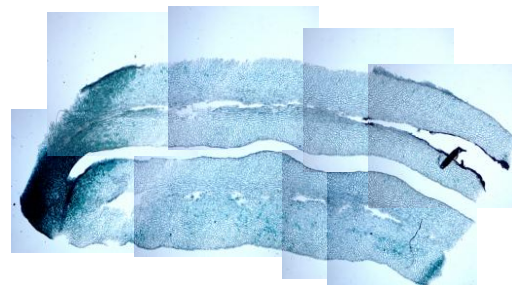


Fig. 6. Longitudinal coffee cut at the middle part of the endosperm.

At fresh state, the furrow seems filled with mucilage, more viscous at this point (Fig. 7). As the coffee bean becomes dryer, it can only be observed the silver skin, and an empty space (Fig. 8) that should be full of air. Since the water diffusion in air is higher, this configuration (silver skin+air) should also be considered as a boundary condition.

WATER ACTIVITY IN DIFFERENTS PARTS OF COFFEE BEAN

In order to probe the heterogeneity of coffee bean throughout the grain, isotherms were realised for different parts. These parts were obtained from a sample taken at the center of the grain by using a

cylindrical stainless steel cutting tool (Fig. 9) which reveals two principal parts (part 1 and part 2).



Fig. 7. Image showing the presence of mucilage at the space near the furrow in a fresh stage.



Fig. 8. Longitudinal cut of coffee bean in a dried state.

The part 2 can also be divided into three fractions (4, 5 and 6) where the silver skin is also present (5) and the furrow (6). Including the coffee bean extremities left when the sample is taken and the whole grain, five isotherms were obtained: parchment, part 1, part 2, and whole grain.

The isotherms were determined by the standard salt solutions at 35°C (Table 1) and in the case of whole bean, water activity values between 0.92 and 0.98 were also obtained by a new mechanical method (Ouoba et al., 2010) which allows to determine high water activity values at equilibrium in a very short time (one day), without risk of moulding growth.

Table 1. Salts used for determining the sorption isotherm of different parts of the coffee bean.

Salt solutions	a_w at 35°C
Lithium chloride	0.12
Potassium acetate	0.21
Potassium carbonate	0.43
Sodium nitrite	0.62
Sodium chloride	0.75
Potassium chloride	0.84
Potassium sulphate	0.96

For the five sections studied, the isotherms are given in Fig. 10. No significant differences between the isotherms for the whole grain, the extremities of grains, and there of the samples taken from the central part (part 1 and part 2) were found at water activities lesser than 0.84. However there is a large difference between the values of the parchment and the other parts at high water activity values. This difference may be attributed to its lignocellulosic composition, different from that of the other parts. Eventhough the water activity curves have the same trend for the other parts, water contents at water

activity of 0.96 are different. Mechanical method, which allows to determine accurate high water activity values can be used to confirm this behaviour. The high activity water area of the isotherm is interesting to consider because is at high values of water activity that *Aspergillus ochraceus* could develop.

DIFFUSION COEFFICIENT

To study the diffusion coefficient, two methods were used. The first one uses the whole grain, the second uses a sample from the internal structure of the grain.

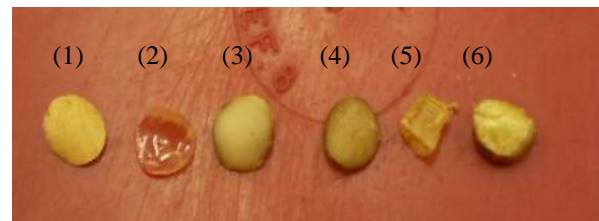
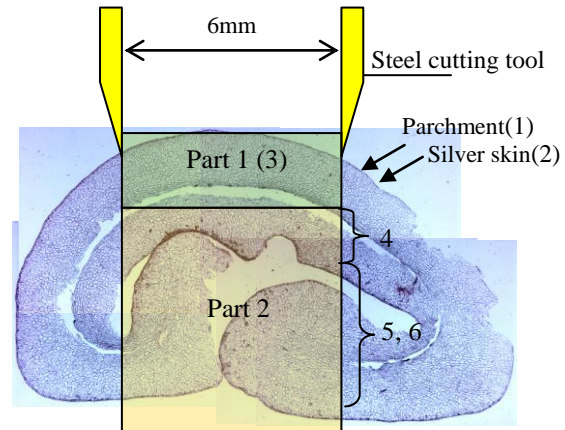


Fig. 9. The different parts of the coffee sample: (1) parchment, (2) the silver film covering part 1; The Part 1 form a single piece (3); (4), (5) and (6) are all part of Part 2.

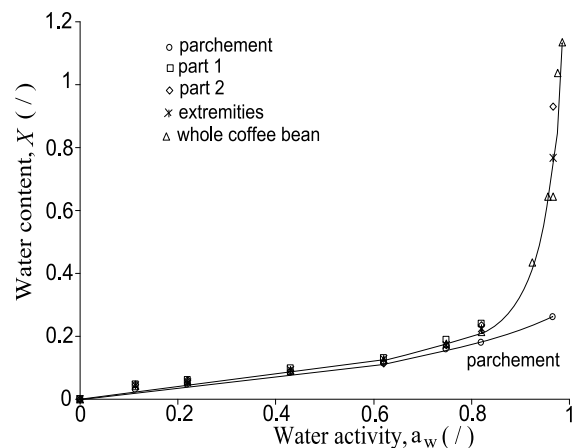


Fig. 10. Coffee bean isotherm at 35°C.

Determination of effective diffusion coefficient in the whole grain

In the case of water transfer in the coffee bean, most studies obtain the value of effective diffusivity from

experimental data by fitting a linearization of the equation of Fick. One hypothesis is that the coffee bean has a homogeneous structure (Corrêa et al., 2006; Hernández et al., 2008; Sfredo et al., 2005).

Hernandez et al., (2008) proposed a model for coffee bean mass transfer. In this study, a prolate spheroidal geometry is taken into account. The equation proposed in this work is:

$$\frac{X - X_e}{X_o - X_e} = 0.78 \exp\left(-12.3 \frac{D_{ef} t}{L^2}\right) \quad (1)$$

In order to measure the effective diffusion coefficient, tests were conducted in a plant pilot fixed bed dryer with transversal air flow of $0 - 2 \text{ m s}^{-1}$, 80°C of maximum temperature and with a 0.0024 m^2 transversal section drying chamber. Tests were carried at two temperatures: 35°C and 45°C and air velocity of 1.5 m s^{-1} . These temperatures are similar to those present at solar drying where the risk of OTA development is higher (Frank, 2001; Paulino de Moraes and Luchese, 2003; Kouadio et al., 2007; Taniwaki et al., 2003). In each test, we evaluated the kinetics of drying weight loss and determining the moisture content at the end. The results obtained are shown in Fig. 11.

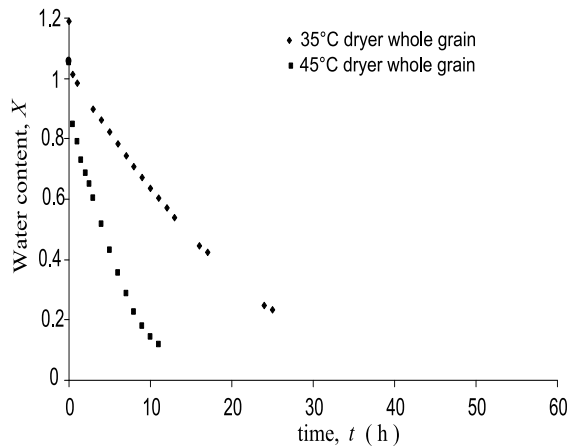


Fig. 11. Comparison of kinetics of dried complete coffee beans at 35°C and 45°C .

Equation 1 was used to determine the effective diffusion coefficient from the data obtained at drying test (Fig. 11). The diffusion coefficient values obtained at 35°C and 45°C are, 3.27×10^{-11} and $6.75 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$ respectively.

Determination of diffusion coefficient of a sample from internal structure of the grain

To determine the water diffusion in the endosperm we use a method where the solution was calculated by Crank (1975) for a plane sheet of thickness L . The boundary conditions are a no-flow condition at one side and on the other side the water content is imposed at X_e the water equilibrium content. Also external mass transfer must be negligible. In the initial state, the water content X_o is assumed to be uniform:

$$\frac{X - X_e}{X_o - X_e} = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left[-\frac{(2n+1)^2 \pi^2 D t}{4L^2}\right] \quad (2)$$

In this case the graph of the logarithm of dimensionless water content versus time gives a straight line whose slope is proportional to the diffusion coefficient D .

In order to measure the local diffusion coefficient and to study the effect of the silver skin, coffee samples were obtained as shown in Fig. 9. From these samples we took part 1 where silver skin was removed in some of tests. The average thickness of these samples was 0.9 mm . The samples thickness made difficult to carry an experiment using forced convection, so the tests were carried as described below.

After being taken from coffee bean, the samples with and without silver skin were glued on an aluminum plate. To overcome the variability of the product, 10 samples were placed at each plate. The schema of the experimental device is given in Fig. 12. All tests were carried at 35°C .

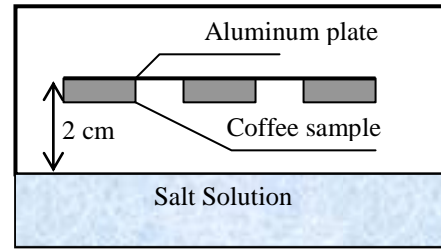


Fig. 12. Diagram of the experimental device used to determine diffusion coefficient at different water contents.

To accomplish the assumption of uniformity of water content in the initial state, the samples were set in the device of Fig. 12 in the presence of salts that regulate the activity at different values: 0.43, 0.50, 0.62, 0.75 and 0.84. The equilibrium of the samples was verified by the stability of the weight over time. Salt solutions were then replaced by a potassium acetate solution fixing water activity at 22% while the weight of the plates was measured over time. At the end of the experiment, the plates are placed in an oven to determine the dry weight and calculate the water content over time for each sample (Fig. 13). The water content at equilibrium was obtained from the desorption isotherm at a value of water activity of 0.22.

As the tests were also realised without forced convection, it should be verified the hypothesis of negligible external mass transfer. A general mass transfer equation can be applied at solid surface (Demirkol et al., 2006):

$$\phi_s = \alpha \left(\frac{P_s}{RT_s} - \frac{P_\infty}{RT_\infty} \right) \quad (3)$$

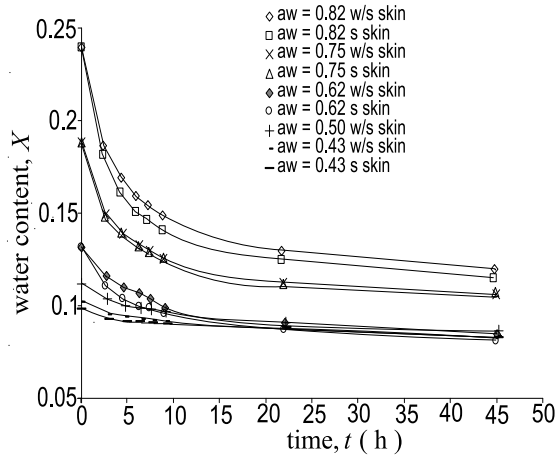


Fig. 13. Change in moisture content of samples with and without silver skin. Curves are in the same order than the labels; w/s skin represents the samples without silver skin and s skin, with silver skin.

Where R is the gas constant, P_s and T_s are the equilibrium vapor pressure and temperature on the surface of the sample, and P_∞ and T_∞ corresponds to the pressure and the temperature in the air and α is the external mass transfer coefficient in $m s^{-1}$. The flux at the surface can be calculated by:

$$\phi_s = \frac{m}{A} \frac{dX}{dt} \quad (4)$$

Where m is the dry mass of samples (0.3 g on average for the experiments), and A refers to samples surface ($2 \times 10^{-4} m^2$). The derivative value can be calculated from Figure 13. As the samples were equilibrated at $35^\circ C$, and after a short period of time they were set at devices at $35^\circ C$, T_∞ and T_s is taken at $35^\circ C$. As example, the values of the external mass transfer coefficient for the lowest and the highest pressure difference will be calculated. The lowest pressure difference ($a_w = 0.43$ at sample surface, $a_w = 0.22$ at air) corresponds to a vapor pressure at surface (P_s) about 2418 Pa and at the air about 1237 Pa. For the highest difference ($a_w = 0.84$) P_s corresponds to 4723 Pa. The calculated flux from equations 4 for the first one corresponds to $4 \times 10^{-7} kg m^2 \cdot s^{-1}$ and for the second to $8 \times 10^{-6} kg m^2 \cdot s^{-1}$ therefore mass transfer coefficient, calculated from equation 3 is 4.8×10^{-5} and $3.3 \times 10^{-4} m s^{-1}$, respectively. Biot number for mass transfer was determined to confirm that drying was diffusion controlled. The expression to determine Biot number is the following:

$$Bi = \alpha L / D \quad (5)$$

From external mass transfer calculated above and diffusion coefficients in the order of $10^{-11} m^2 s^{-1}$, we obtain Biot numbers higher to 100, allowing the use of equation 2 to calculate the diffusion coefficient. Moreover X_e samples value is very close to the value regulated by the saline solution ($a_w = 0.22$) and can be regarded as constant, which correspond to another assumption made to obtain equation 2.

Fig.14 represents the variation of diffusion coefficient of the part 1 (Fig. 9) with and without silver skin as a function of water content. This result confirms that the diffusion coefficient inside the grain depends on water content. Tests at higher values of water content should increase the value of diffusion coefficient. To confirm this, we envisage to make more experiences at higher values. By the other hand, we can note the influence of silver skin on diffusion coefficient. Even the diminution of the fold of diffusion coefficient values is lesser than 15%, the silver skin is found not only at the surface of the grain but also at the interior the grain (Fig. 7 and 8). Numerical calculations would reveal its final importance in water transfer.

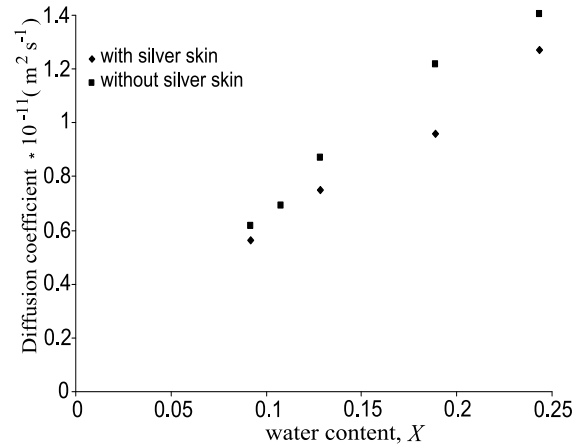


Fig. 14. Variation of diffusion coefficient as a function of water content.

Transfer coefficient in the parchment

Since the thickness of the parchment is low and its curved shape make difficult to fix it on the aluminum plates without creating cracks, it was not possible to determine the diffusion coefficient by the previous method. A specific experimental device has been realized. The parchment was fixed at the end of a tube (Fig. 16b) of 4 mm of internal diameter and placed in a cell (Fig. 16a) which has on its interior a saturated salt solution. Then, the cell is placed in a chamber with another saturated salt solution. With this configuration the sample acts as barrier (Fig. 15). The chamber is immersed in a thermo-regulated bath. The flux through the samples is equal to the weight change of the inner cell per time unit. Considering parchment thickness, we aim to determine a coefficient that links the flux per unit area and time (ϕ) with the difference in water activity on both sides of the parchment. The coefficient γ is defined by:

$$\phi = \gamma (a_{w2} - a_{w1}) \quad (6)$$

Figure 17 gives an example of weight change of the cell interior that is divided by the surface to get the flux ϕ . Knowing the difference in activity on both sides of the sample, we calculate the transfer coefficient by the equation 6. In this example we

obtain a coefficient γ of $0.11 \times 10^{-3} \text{ kg m}^{-2} \text{ s}^{-1}$ fixing a water activity difference of 0.21 at the inner cell and 0.85 at the external chamber.

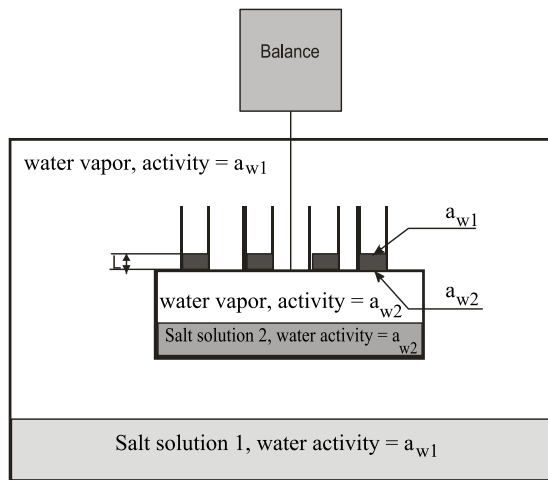


Fig. 15. Experimental device used for the determination of parchment diffusion coefficient.

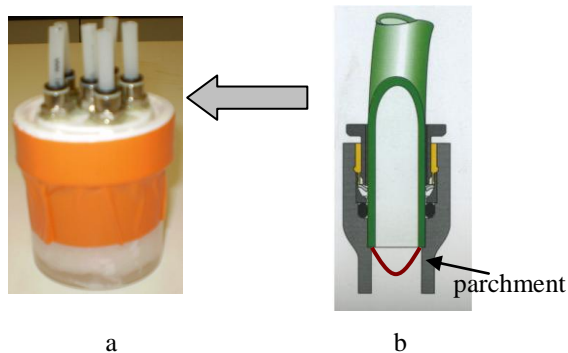


Fig. 16. Capsule and fixing sample system used in the determination of parchment diffusion coefficient.

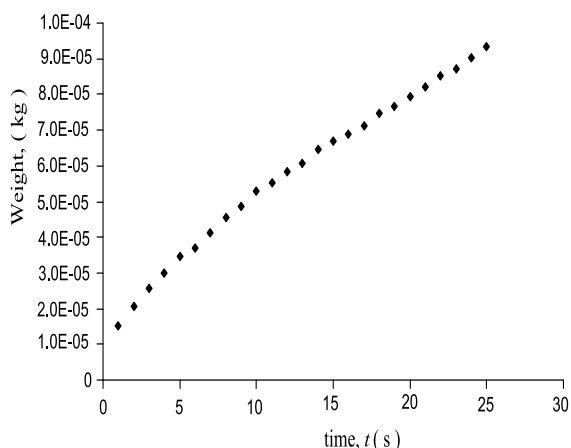


Fig. 17. Change over time of the weight of the cell with the parchment.

CONCLUSIONS

The coffee bean has a complex structure while it is often considered homogenous when modelled. It has been chosen to approach this problem by the accurate modelling of water transfer and a prediction of the

evolution of water content and activity within the grain during drying. Before modelling, it is necessary to gather information that will lead to formulate simplifying assumptions and to measure the parameters required for simulation. This communication is focused on these last two points by adopting an experimental approach.

Regarding the structure of grain, microscopic observations show a complex shape with discontinuities which constitute restrictions that can hinder the movement of water. The observations permit us to define the outer surfaces and internal structures that should constitute boundary conditions, however, doubts still remain about the nature of these conditions on the internal discontinuities in grain.

Experimental work is still needed to clarify this last point. On the other hand, knowledge of the kinetics of drying will define the surface water flux on the outer surfaces and then will lead to the determination of an external transfer coefficient.

The role of the parchment has been clarified. It seems that at high values of water content, the desorption isotherm is different from that of whole grain. This is certainly due to its composition that is essentially lignocellulosic and different from that of the endosperm. Knowledge of the transfer coefficient in the parchment, gives access, knowing the flux of water and the water activity of the air at the surface, to the water activity between the parchment and the endosperm. This activity can be expressed in term of water content at the surface, by the isotherm of the endosperm, to finally obtain the boundary condition at the endosperm.

The desorption isotherm of different parts of the grain suggests that the relationship between water content and water activity is the same throughout the grain. This result permits to formulate a hypothesis that greatly simplifies the modelling: the law of transfer for water can be expressed using the gradient of water content. Otherwise it would have been necessary to express it in terms of the gradient of water activity. The experiments realised with flat samples were carried out under the conditions of application corresponding to the solution of Fick's equation for a plane sheet. Under these conditions, it was possible to determine the variation of transfer coefficient as a function of water content. Considering that the tested values concern a relatively small range (between 9% and 23%) we intend to make experiences at water activities close to 1. From results obtained, we expect a large increase for the value of diffusion coefficient at high values of water activity in the grain which will rebound on the heterogeneity of water content and activity. Given the values obtained, it appears that the silver skin constitutes not a barrier but a restriction to water transfer and it should be considered because it is present not only in the surface but at the interior of

coffee endosperm.

The information reported in this paper permit us to consider an accurate simulation, based on verified hypothesis and with values derived from experience. For given boundary conditions, the model should provide the values of water activity in all parts of the grain. The complex configuration of internal discontinuities, the strong dependence of the transfer coefficient on the material constituting the grains and the dependence of water activity on water content, suggests that in the first minutes of drying, it exists a difference of water activity values throughout the grain, and these values could be higher than the critical water activity value where a toxin could develop. By varying the boundary conditions, the model should be able to provide the drying conditions at which the duration of these areas should not constitute a risk.

NOMENCLATURE

a	Activity	/
A	surface	m^2
Bi	Biot mass transfer number	/
D	Diffusion coefficient	$m^2 s^{-1}$
L	Width	m
m	Dry mass	kg
P	Pressure	Pa
R	Gas constant	$m^3 Pa kmol^{-1} K^{-1}$
T	Temperature	$^{\circ}C$
t	Time	s
X	Water content	/

Greek letters

α	External transfer	mass	$m s^{-1}$
γ	Resistance coefficient		$kg m^{-2} s^{-1}$
φ	flux		$kg m^{-2} s^{-1}$

Subscripts

0	Initial
e	Equilibrium
ef	effective
s	surface
w	Water
∞	Air phase

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