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Numerical and Experimental Approaches to Characterize the Mass Transfer Process in Wood Elements

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Abstract: The scope of this paper is an experimental characterization of diffusion parameters for wood material. Based on a nonlinear mass transfer algorithm, the present study focuses on the need to capture experimental moisture profiles in the sample, along with its evolution in weighting during both the desorption and adsorption phases, especially when the moisture content of the samples is far from the equilibrium state inducing a great gradient between heart and exchange surfaces of specimen. These moisture profiles are derived by means of a gammadensimetry laboratory method based on the water adsorption of gamma rays. The determination of the diffusion parameters obtained through optimizing a simulation by means of implementing the mass transfer kinetics into a finite difference method. Both the diffusion coefficient and convective exchange coefficient are deduced by considering a Nelder-Mead simplex inversion method. This work highlights the efficiency of the approach dedicated to uncoupling nonlinear diffusion in the cross-sections from boundary conditions in terms of convective

exchanges and equilibrium moisture. Scale effects and boundary conditions are also investigated herein.

Keywords: Moisture content, Wood material, Mass transfer, Gammadensimetry, Diffusion model, Inverse methods.

1 - Introduction

In pursuit of sustainable development goals, greater numbers of timber elements are being used in many types of public construction like buildings and bridges. The demand for timber construction today is constantly rising, yet many technological obstacles still prevent the wider use of wood in the highly competitive field of construction materials.

When placed in outdoor conditions or heat spaces, these elements are subjected to moisture content variations induced by a non-stabilized equilibrium between wood and the climatic environment. The moisture content gradient, associated with the orthotropy of wood, induces hydric stresses amplified by the hyperstaticity level or the coupling with other materials, such as concrete or steel in the case of composite structures. The magnitude of these stresses is not well known at the design stage and not seriously taken into account in construction regulations. This mechanical state can induce a degradation in toughness and the appearance of cracks. In order to facilitate timber structure development, the issue of moisture content needs to be addressed by the scientific community.

This context requires in particular developing an *in situ* monitoring protocol. The control of moisture content mapping through a cross-section is currently limited by resistive methods that rely on the use of surface electrodes, which allow for the estimation, after calibration, of moisture to a depth of around 1 cm (Dubois *et al.*, 2006). This inadequate

method needs to be complemented by numerical simulations in order to deduce moisture content gradients and their evolution vs. time by taking into account temperature and humidity in the climatic environment (Manfoumbi, 2012; Manfoumbi, 2014). This approach however imposes several limitations. First of all, the diffusion properties introduced into numerical simulations must be correctly estimated by determining the nonlinear diffusion coefficients. Secondly, the simulation must integrate surface exchanges by considering the convective effects in terms of temperature and humidity. As a final step, specific thermo-hygro-mechanical behavior must be considered when estimating the thermo-hygro stresses (Dubois *et al.*, 2012; Colmars *et al.*, 2014).

The present work pertains to the first step and is intended to determine the diffusion parameters. The characterization protocol is typically based on the double weighing principle. For various sample geometries (at the centimeter scale), this technique consists of optimizing the diffusion parameters in accordance to their corresponding average moisture content during adsorption and desorption tests. The main difficulty lies in separating the effects of boundary conditions, including sorption hysteresis and surface humidity exchanges. Numerical algorithms, derived from a minimization step between experimental and numerical differences, do not always yield a unique realistic solution. Moreover, in the case of samples far from the equilibrium state, errors on the identifications of the diffusion properties may be done due to insufficient measurement points. In this case, the method doesn't allow the uncoupling between the diffusion to the heart and the surface exchanges between first fibers and the climatic environment.

To overcome this inconsistency and to highlight how the choice of the experimental measurements may influence the identification of the diffusion parameters, the present paper provides an additional experimental test on larger samples (at the decimeter scale), thus making it possible to generate a moisture content profile measured with the

gammadensimetry technique. Three boundary condition tests have been taken into account in order to demonstrate the relevance of this method and its robustness. The first section will describe the experimental set-up through the selected sample geometries and boundary conditions. This section will be completed by a presentation of the gammadensimetry technique and its specific application in water profile measurements during wetting or drying phases.

The numerical model and the determination of the diffusion parameters will be discussed in a second section. A nonlinear Fick's law will be coupled with an inverse method based on the downhill simplex method, which in turn allows optimizing the diffusion parameters in accordance with average moisture content measurements and water profiles determined by gammadensimetry method.

The last section will focus on experimental results. The method robustness will be validated by exhibiting the effects of boundary conditions and geometric dimensions.

2 – Materials and methods: experimental set-up and numerical model

2.1 Materials and hydric loading

The primary target of the experimental tests was to monitor moisture content evolution vs. time through various boundary conditions and geometries, using large samples at high relative humidity increments. As shown in Fig 1, three configurations were proposed considering moisture diffusion along the longitudinal direction. More precisely, the samples had been cut out along the anisotropic directions (i.e. Longitudinal L along the tree stem, Radial R perpendicular to the rings, and Tangential T parallel to the rings), with moisture diffusing along the longitudinal direction.

0.6	5.1	9.5	Initial moisture content (%)
97	97	86	Relative Humidity (%RH)

Fig. 1 Moisture boundary conditions and sample geometries for the three cases

Under these conditions, only one or two faces were exposed to the external environment. Other faces were covered by two Parafilm layers to ensure unidirectional gas exchanges along the grain direction. Two geometries for the wood specimen, machined in Douglas fir (*Pseudotsuga menziesii* (Mirb.) Franco), were considered. For samples (a) and (b), their height equaled 200 mm (longitudinal direction) with a 70 mm x 70 mm cross-section (radial and tangential directions). In reality, specimens (a) and (b) were one in the same and had been repeatedly dried between the two experiments. This option allowed us to focus on the effect of boundary conditions. Sample (c) was the largest with a 320-mm height and a 95 mm x 95 mm cross-section, for the purpose of specially exploring ultimate scale and moisture loading effects.

For one thing, all specimens were conditioned in a dry environment using several equilibrium moisture contents for w : less than 1% for sample (a), around 5% for sample (b), and 9% for sample (c). The moisture content w is typically defined as the ratio of the water mass M_{H_2O} to the dried or solid mass of wood M_s . Under a constant temperature of approx. 19°C (+/-1°C), samples were placed in desiccators over saturated salt solutions, which served to impose a constant relative humidity of around 97% RH for samples (a) and (b), and roughly 86% RH for sample (c). Average moisture content is basically determined by measuring weight during the sorption test.

2.2 - Gammadensimetry method

Gammadensimetry is a non-destructive method used to determine the density or moisture content of civil engineering materials (Da Rocha *et al.*, 2001; Villain and Thierry, 2006; Ferraz and Aguiar, 1985). This technique is based on the absorption of gamma rays emitted by a radioactive source (in our case Cesium Cs¹³⁷) and follows Lambert's law. To determine moisture content at various heights, it is assumed that the wood material is composed of three phases, namely solid s (dried wood), water w (bound water) and air a , such that:

$$\ln\left(\frac{N_o}{N}\right) = \mu_s \cdot \rho_s \cdot x_s + \mu_w \cdot \rho_w \cdot x_w + \mu_a \cdot \rho_a \cdot x_a \quad (1)$$

where N_o is the number of incidental photons in the air, N the number of photons crossing the sample of thickness X (m), and μ_i , ρ_i and x_i the mass absorption coefficients (m²/kg), densities (kg/m³) and thicknesses (m) of phases i ($i \in \{s, w, a\}$), respectively. Let's remark that the air phase density and its mass absorption coefficient

can both be neglected. If the gamma ray crosses a thickness X , the following can be expressed:

$$X = \sum_i x_i \quad (2)$$

The moisture content $w(t)$ is typically defined such that:

$$w = \frac{M_{H_2O}}{M_s} = \frac{\rho_w \cdot x_w}{\rho_s \cdot x_s} \quad (3)$$

According to expression (1), thicknesses x_s and x_w at experimental time t are written as follows:

$$x_w(t) = \frac{(\ln \frac{N_0}{N})_t - (\ln \frac{N_0}{N})_{t=0}}{\mu_w \rho_w} + x_w(t=0) \quad (4)$$

$$x_s(t) = x_s(t=0) = \frac{(\ln \frac{N_0}{N})_{t=0}}{\mu_s \rho_s + \mu_e \rho_s w_{(t=0)}} \quad (5)$$

Due to swelling of the wood material under a sorption process, the water content may be underestimated given that the thickness x_s at time t may be overestimated in the relation (3). Nevertheless, the error on the moisture content due to the swelling effects in the transverse plane (RT) is in the same magnitude order that the estimated swelling strains. So, it can be neglected in regards to the dispersion of the measurements in link with the further discussion about the possible uncertainties. In relation (5) therefore, it may be assumed that thickness x_s crossed by the gamma ray does not vary during the sorption process to simplify the determination of the moisture content. Moreover, the moisture content profile was assumed to be homogeneous in the sample at time $t = 0$. The calculus applied also had to integrate the Parafilm thickness in accordance with expression (5).

For the various phases, the densities were: $\rho_w = 1,000 \text{ kg/m}^3$ and $\rho_s = 1,520 \text{ kg/m}^3$. The mass absorption coefficient of each phase was calculated from its corresponding chemical analysis, while the elementary mass absorption coefficients for the radiation energy of Cs^{137} are given, for instance, in the tables published by the U.S. National Bureau of Standards. For the water phase, the mass absorption coefficient was: $\mu_w = 8.57 \cdot 10^{-3} \text{ m}^2/\text{kg}$. As for the dried wood (solid) phase, it was assumed that wood is composed of cellulose, hemicellulose and lignin, thus leading to a mass absorption coefficient $\mu_s = 8.17 \cdot 10^{-3} \text{ m}^2/\text{kg}$. More precisely, it was considered that the dried wood phase contained 50% carbon, 6% hydrogen and 44% oxygen in mass terms. Slight variations in these percentages however do exert a small impact on the final mass absorption coefficient value.

As shown in Fig 2, the moisture content profiles required placing the specimen in a plate that was vertically moved by a robot through the gamma ray beam at a 5-mm step. According to diffusion kinetics, it was assumed that moisture content is homogeneous along the radial direction for each cross-section and solely dependent on the longitudinal position of the gamma ray. Under these conditions, for each gamma ray position, the average moisture content in the cross-section was measured. Regular profile measurements were conducted throughout the experimental period (2 to 10 months). Several profiles were measured (2 for samples (a) and (b), 3 for sample (c)), yet only the averaged profile will be considered for the purpose of identifying diffusion parameters.

The viability of the gamma-ray method has been shown by Ferraz and Aguiar (Ferraz and Aguiar, 1985) for determining density and moisture content of wood samples. Relative deviations (absolute errors) between the traditional gravimetric method and the gamma-ray method have been estimated. The differences between the two methods are estimated up to 3% for samples with moisture content MC between 9 % and 30 % (relative error

inferior to 20%). The exactness of the gamma-ray method is limited mainly by errors made in the determinations of N and N_0 as a consequence of random disintegration; the errors made in the determination in the other parameters (eg. x_s) are negligible. Moreover, in the case of the use of the Cesium Cs^{137} radioactive source, the sensitivity of the method may be low because of the small differences between the mass absorption coefficients of water and dried wood (solid) phase. But despite these possible absolute errors, it is possible to obtain an average moisture content profile for large samples subjected to hydric loading.

Fig 2 Gammadensimetry profiles A and B for samples (a) and (b) along the longitudinal direction (for sample (c), three profiles A, B and C have been considered)

Ultimately, the average moisture content profiles provided by gammadensimetry and weighing must allow for the characterization of diffusion properties. The next sections will propose an inverse method technique based on a mass transfer algorithm, along with a downhill simplex method that serves to minimize the difference between numerical model output and experimental measurements. This work will be limited to a uniaxial approach due to moisture content homogeneity within the cross-sections.

2.3 - Diffusion model

Many models are proposed or developed to describe diffusion in wood: Fickian models, multi or not Fickian models, which can take into account dependence of diffusion coefficient on moisture content and take into account the surface exchanges. In the general case, the choice of the mass transport model deals with the necessity (or not) of separation of bound liquid, free water and vapor (Krabbenhof 2003; Perre and Turner, 2008). In the present work, we are not in drying case from high water content where liquid, bound water and vapor cohabit, but below the saturation fibers point: in the hygroscopic domain, only bound water and vapor have to be considered. In this case, there is a common model with only one variable for both forms of water (Jakiela et al., 2008; Rozas et al., 2009; Olek et al., 2011; Da Silva et al., 2011) or model with separated variables (Frandsen et al., 2007, Krabbenhof 2003, 2004). . Then, differences between models can come from the choices for the treatment of the convective boundary conditions: relaxation term (Olek et al., 2016), hydrous surface exchange coefficient. It can be noticed that there is no consensus on the diffusion models which may have to be used and on the convective boundary conditions. Our assumptions and geometries allow us to use the common model which is described below.

The mass transfer process over time t is typically modeled using Fick's second nonlinear law. Fick's coefficients and the surface emission and diffusion coefficients are all described as phenomenological parameters since they can only be grasped or conceptualized through experience. According to isothermal conditions, the mass transfer process integrates a relationship focused on non-linearity between moisture content level and orthotropic diffusion tensor. From the perspective of a uniaxial representation characterized by the x coordinate, the time/space differential equation can be written as follows (Perre and Degiovanni, 1990; Merakeb *et al.*, 2006):

$$\frac{\partial w}{\partial t} = \frac{\partial}{\partial x} \left(D_w(w) \cdot \frac{\partial w}{\partial x} \right) \quad (6)$$

The mass transfer process is principally driven by the diffusion property D_w . In wood materials, this property is admitted to be nonlinear with respect to moisture content, in accordance with the following form (Droin Josserand *et al.*, 1989):

$$D_w(w) = D_o \cdot \exp(k \cdot w) \quad (7)$$

where D_o is the anhydrous moisture diffusion coefficient, and k a constant reflecting the nonlinear effect.

According to boundary conditions, solving Fick's equation requires knowledge of the surface moisture content, denoted w_Ω . So, if S designates the hydrous surface exchange coefficient, then the diffusion equation can integrate the following boundary limits:

$$D_w(w) \frac{\partial w}{\partial x} = S \cdot (w_e(t) - w_\Omega(t)) \quad (8)$$

where w_Ω is the surface moisture content. To take into account the exchanges between water vapor molecules in air and bound water molecules in wood, it is commonly considered to employ a virtual exchange layer defined by an instantaneous equilibrium between external fibers and water vapor in the neighboring climatic environment. This equilibrium is usually driven by sorption hysteresis and defined by the virtual moisture content value, called w_e (Lasserre, 2000; Merakeb *et al.*, 2009).

Moreover, our problem can be reduced to characterizing D_o , k , S and w_e . The mass transfer subroutine is implemented as part of the finite element method. However, the problem is further reduced to a uniaxial configuration, and expressions (6) through (8) are solved using a finite difference method based on a time discretization based on an explicit Euler scheme (Zhou *et al.*, 2011; Liu and Simpson, 1999).

2.4 - Downhill simplex method

The downhill simplex method is introduced here to characterize diffusion properties (Nedler, 1965). This method consists of minimizing the objective function in using only function values and without calculating the derivative, so as to represent the gap between the moisture content evolution during experimental time and model results from the standpoint of least squares by means of adjusting diffusion parameters, such as diffusion coefficient D_o , nonlinear parameter k , hydrous surface exchange S and equilibrium moisture content w_e . For these four unknowns, the method considers a geometric polyhedron, called simplex, composed of 5 nodes, with each node representing a unique solution. Based on a hierarchy of errors for each solution, this algorithm reduces the geometric surface until convergence is reached. The vertices of this polyhedron will undergo geometric transformations in moving towards a global minimum (Lagarias *et al.*, 1998). The simplex algorithm iterations correspond to simple algebraic operations on the polygon vertices for elementary geometric transformations (reflection, contraction and expansion) (Kelley, 1999). Each iteration transformation solely depends on a series of comparisons between objective function values corresponding to the calculated points and values of the polygon vertices to replace the worst vertex (maximum) by the new fixed point. The polygon is thus reflected, extended and reduced depending on the function shape until its optimum corresponds to a complete reduction of the simplex yielding the optimal solution. As demonstrated by Kouchade (2004), this method is well adapted to incomplete experimental curves.

The mass transfer process however performs two competing effects, in accordance with expressions (6) and (8). Based solely on the average moisture content evolution, the downhill simplex method does not distinguish the surface effect characterized by S and w_e from the mass diffusion characterized by D_o and k . When limited to the average

moisture content data, the minimization method can actually yield an infinite number of solutions. The use of water profiles provided by gammadensimetry could ensure an acceptable level of separation between surface and mass effects.

3 - Results and discussion: moisture content profiles of sorption tests and identifications of diffusion parameters

This section proposes comparing the two methods for identifying diffusion parameters by taking into account both moisture loading and sample geometry. This methodology will be applied to samples (a) and (b) and then to sample (c). The diffusion parameters will be discussed afterwards.

3.1 - Identification of diffusion parameters for sample (a): reference case

Sample (a) is characterized by just one surface of moisture content exchange, with initial drying in an oven at 103°C. As shown in Fig. 3 after 81 days of adsorption, the average moisture content increased to only 14%, i.e. far below the equilibrium moisture content estimated to lie around 22% at about 20°C, according to Loulou (2013) and Merakeb *et al.* (2009). The equilibrium moisture content value is highly sensitive to both the moisture and thermal loading; hence, the level of precision is approx. +/-2%, especially for a high Relative Humidity loading.

Diffusion parameters are normally characterized using a simple weighing of small samples corresponding to average moisture content information over time; this procedure is referred to as standard identification. For this standard identification, the equilibrium state is normally raised, leading to a possible identification of the diffusion parameters. In our particular case, let's set the equilibrium moisture content value w_e as unknown. In our example therefore, this approach is being compared on five weighing measurements (at 3, 10, 18, 25 and 81 days) with a so-called coupled identification using crossing

weights and gamma-ray measurements.

The comparison between these two identification methods is presented in Fig. 3 and Fig. 4, in accordance with identified diffusion parameters (see Table 1). The nonzero values obtained for the non-linearity coefficient and the rather low hydrous surface exchange coefficient serve to validate the hypotheses of nonlinear diffusion and exchanges by hydrous convection.

Tab 1 Diffusion parameters for sample (a)

Identification method	$D_0 \left(m^2 \cdot s^{-1} \right)$	k	$S \left(m \cdot s^{-1} \right)$	w_e
Standard identification	$4.50 \cdot 10^{-9}$	1.12	$5.03 \cdot 10^{-8}$	20,6%
Coupled identification	$3.27 \cdot 10^{-9}$	1.94	$4.73 \cdot 10^{-8}$	22,5%

Results highlight the sensitivity of identification by means of the inverse method. Fig. 3 illustrates that in terms of average moisture content evolution, various diffusion parameters can yield the same results, especially within the experimental time frame.

Fig. 3 Results of standard and coupled identification in terms of average moisture content evolution for sample (a)

The coupled identification was performed using five profiles (at each weighing time), but only three profiles have been presented for the sake of clarity. The profile results displayed in Fig. 4 however reveal slight differences. As explained previously, it can be observed that the gammadensimetry measurements present dispersions around an average profile value. Despite this dispersion (and some anomalous points), it is still possible to compare the experimental and numerical profiles. Experimentally, we can observe that the moisture content first increases on the exposed face and only in the first quarter of the sample (day 3), then the moisture gradient decreases while the moisture content increases up to the whole sample and the sealed face (day 81). Numerically, the main difference between the profiles from the standard identification (that does not take into account the experimental profiles) with those from the coupled identification concerns the higher moisture content values on the exposed face as seen experimentally.

Fig. 4 Results of standard and coupled identification, in terms of moisture content profiles for sample (a)

As regards the identified diffusion parameters, let's note a slight difference in exchange surface coefficient of around 6% and a difference in the anticipated value of w_e (9%). The main differences actually pertain to the diffusion parameter characterization and its property of non-linearity. According to expression (7), the identification method produces a great difference (of up to 25%) in the diffusion parameter evolution vs. moisture content (Fig. 5).

Fig. 5 Diffusion coefficient vs. moisture content within the hygroscopic domain given by the two identifications performed on the results of sample (a)

According to a homogeneity assumption on the sample however, parameters D_0 and k should be intrinsic material properties, while the surface exchange coefficient is somewhat intrinsic to the experiment and its exposure conditions. The small correction due to the coupled identification on its value has a significant effect on the diffusion parameters values and we can expect a better identification of this lasts.

3.2 - Diffusion parameters identification for sample (b): effect of boundary conditions

In the following discussion, both boundary and initial conditions will be the focus of an investigation considering two surface exchanges and various initial moisture contents. According to Figure 1, this second experiment will concentrate on sample (b). To investigate the decoupling between the intrinsic diffusion coefficient (D_0 and k) and the surface exchange coefficient S , sample (b) is to be the same specimen as sample (a), yet the initial conditions and exchange conditions have been modified. After the first experiment, the sample is once again dried until a homogeneous moisture content of 5% is obtained and two faces are in contact with the environment. As shown in Fig. 6 after less than 80 days of adsorption, the average moisture content has already risen to 19%, i.e. not far below the expected equilibrium moisture content.

Fig. 6 Results from both the standard and coupled (recovery from sample (a)) identification, in terms of average moisture content evolution for sample (b)

These same approaches are then proposed in using diffusion coefficients from the coupled identification of sample (a) in order to calibrate equilibrium moisture content and a

second surface exchange coefficient S' that characterizes the additional surface (assumption of a moisture content exchange asymmetry: surfaces and exposition being the same, we can nevertheless not be sure that surface states, with wood rings, are so). It can be noticed that the asymmetry may be not measured by the gamma-ray method owing to moisture content dispersion. According to the diffusion properties listed in Table 2, the average moisture content evolution and moisture content profiles can now be plotted (Figs. 6 and 7).

Tab 2 Diffusion parameters for sample (b)

Identification method	$D_o \left(m^2 \cdot s^{-1} \right)$	k	$S \left(m \cdot s^{-1} \right)$	$S' \left(m \cdot s^{-1} \right)$	w_e
Standard identification	$1.24 \cdot 10^{-9}$	2.58	$1.43 \cdot 10^{-7}$	$2.72 \cdot 10^{-7}$	20.1%
Coupled identification (recovery from sample (a))	$3.27 \cdot 10^{-9}$	1.94	$4.73 \cdot 10^{-8}$	$1.86 \cdot 10^{-7}$	19.2%

In optimizing S' we were able to find a relative good fit of the average moisture content evolution obtained from a dozen weighing measurements and the only two moisture profiles recorded at 7 and 15 days (with gamma measurements once again exhibiting dispersions) with higher moisture content values recorded on the second face. The coupled identification method leads to detecting clearer moisture exchange differences between the two faces, as presented by the moisture profiles in Fig. 7. The standard identification presents two main defaults, less in accordance with the experimental profiles: it quickly leads to high moisture content to both faces and in opposite the moisture content in the middle of the sample should remain low. At last, by considering the only weighing, it cannot easily estimate separated values for the two surface exchange coefficients.

Fig. 7 Results from standard and coupled (recovery from sample (a)) identification, in terms of moisture content profiles for sample (b)

The two profiles seem to be sufficient for determining the nonlinear characteristic of the diffusion process, as observed by comparing diffusion coefficient variations vs. the moisture content (Fig. 8) derived by both the standard and coupled identifications.

Fig. 8 Diffusion coefficient vs. moisture content within the hygroscopic domain, as provided by the identification methods performed on the results of sample (b)

As a matter of fact, this second experiment confirms the differences between coupled and standard identification techniques, with the latter most likely yielding less intrinsic results. In this case, the numerical profile with the coupled method allows for a better agreement with the experimental profile.

3.3 - Diffusion parameters identification for sample (c): scale effect

This experiment on sample (c) is based on the same single-face exposure adopted for sample (a). As specified in Figure 1, the dimensions and moisture conditions of sample (c) are different in that sample (c) has been previously conditioned in a dry chamber corresponding to the equilibrium moisture content around 9.5%. In terms of average moisture, the evolution of this sample is plotted in Fig. 9.

Fig. 9 Results of the coupled identification method with recovery from sample (a) and standard identification, in terms of average moisture content evolution for sample (c)

The experimental test was stopped after 220 days and a dozen weighing measurements, with the average water content equaling about 13%. At equilibrium, the moisture content might have been about 15% according to Loulou (2013). The three profiles measured at

14, 56 and 210 days are plotted in Fig. 10. The results of the standard and coupled identification methods are summarized in Tab 3.

Tab 3 Diffusion parameters for sample (c)

Identification method	$D_o \left(m^2 \cdot s^{-1} \right)$	k	$S \left(m \cdot s^{-1} \right)$	w_e
Standard identification	$9.33 \cdot 10^{-10}$	4.64	$2.64 \cdot 10^{-6}$	14.8%
Coupled identification (recovery from sample (a))	$3.27 \cdot 10^{-9}$	1.94	$2.35 \cdot 10^{-7}$	13.3%

Fig. 10 Results from the standard and coupled (with recovery from sample (a)) identification, in terms of moisture content profiles for sample (c)

Lastly, this third experiment again confirms the differences between coupled and standard identification as the numerical profiles with the coupled method allows, in this last case, for a better correlation with the experimental profiles: the moisture content evolution estimated on the exposed face is better predicted in the first moments (days 14 and 56), as the moisture content distribution in the main part (from position 0.1 to 0.3m) of the sample (until day 210). We can just note that the equilibrium moisture content seems to be a little weak regarding to the value recorded near the exposed face, this time better

represented by standard identification's last profile (days 210). More specifically, the two methods differ in terms of hydrous convection kinetics near the exchange surface. Significant differences are also observed in terms of diffusion coefficient evolution as a function of water content (Fig. 11).

Fig. 11 Diffusion coefficient vs. moisture content within the hygroscopic domain, as output by the two identification methods performed on the results of sample (c)

3.4 - Discussion

The difference between these two identification methods (standard and coupled), as compared for sample (a), may be explained in a series of remarks. First of all, because the standard identification is based on a global, or averaged, experimental moisture content monitoring, the number of diffusion parameters is capable of inducing self-optimization, in correlation with one measurement point per time interval. In this case, the surface exchange coefficient S can correct an error when characterizing the diffusion coefficient determination; moreover, as shown in Fig. 3, various identified parameters may be consistent with the time - average moisture content curve. Hence, the standard identifications for samples (a), (b) and (c) show significant differences on diffusion

coefficients (Fig. 12). The moisture content profiles given by gammadensimetry method make it possible to correct the specimen diffusion parameters along the moisture content gradient. All experimental time points represent a numerical constraint for the inverse method algorithm, thus facilitating the optimization of diffusion parameters. The unique couple of diffusion coefficients identified in the reference case (i.e. sample (a)), along with the proposed coupled method, is able to reproduce both average moisture content evolution and profiles for the two other study cases with different boundary conditions and geometric dimensions (samples (b) and (c)).

Fig. 12 Diffusion coefficient vs. moisture content within the hygroscopic domain, as provided by the two identification methods performed on the results of samples (a), (b) and (c)

Our proposed experimental method deals with robustness of identification of diffusion parameters, usually made with only weighing: with a spatial measurement of water content distribution we fill in a lack of information that could fault any sophisticated model. Indeed, this study has exposed the remediable flaws of the standard identification method, the weighing appearing insufficient with sparsely measurement points far from

equilibrium. Although characterization does allow for an acceptable modeling of the average moisture content evolution, it is not always possible to easily reproduce the profiles within the material. The balance between the hydrous exchange by convection and diffusion in the material can lead to a significant number of satisfactory datasets for the average moisture content. Moreover, should the experimental time frame prevent achieving moisture equilibrium, the diffusion kinetics errors can lead to an incorrect extrapolation of equilibrium moisture and induce errors in the estimation of sorption isotherms. In this case, a better characterization of diffusion properties, and more specifically convective parameters, at the beginning of the test can increase diffusion extrapolation accuracy. The moisture content profiles do yield better information for a better fit of the time moisture content evolution.

Although these profiles contain a high level of experimental noise, the coupled characterization method does enable constraining the minimization algorithm in order to limit the number of possible combinations, in terms of diffusion properties, by highlighting the moisture effects due to convective processes. Such a feature makes this method more accurate and robust with sets of parameters intrinsic to the studied material, thus offering, via the use of a diffusion model, a more realistic prognosis beyond experimental observations. At last, it may be useful for applications in the field of in-situ monitoring of structural timber elements and it will certainly be improved in the future thanks to more elaborated models.

4 - Conclusion and outlook

This work has proposed an experimental protocol for characterizing nonlinear diffusion properties. The coupling between a simple weighing of samples, corresponding to an average moisture variation identification, with a gammadensimetry technique has been

presented. Such a coupling allows for a realistic separation between diffusion kinetics through the cross-section and the moisture content surface exchange. Moreover, the nonlinear diffusion coefficient is better determined by taking into account the moisture content profiles given by the gamma-ray method. Scale effects, as well as the studied boundary and initial conditions, enable concluding the robustness of this approach in measuring some really intrinsic properties.

Moisture profiles have provided us with certain information during the start of adsorption; they allow for a better identification of the initial diffusion times in the vicinity of boundary elements. This additional information increases the model's potential to predict moisture content evolution vs. time. We are now able to propose other experimental protocols for characterizing diffusion without finding moisture equilibrium. In this specific case, this algorithm may be used to determine diffusion properties during moisture buffer tests in which outdoor humidity conditions are in a harmonic solicitation pattern.

Regarding the monitoring of timber structures, the diffusion behavior of elements must be known in order to predict long-term mechanical responses and durability of the structures. The gammadensimetry technique offers certain information about the moisture content gradient state; however, this method cannot be generalized to outdoor monitoring. A perspective on this work is the development of a non-destructive method to allow measuring moisture gradients in the cross-section. One solution consists of replacing the gammadensimetry method by a 2D or 3D resistive method coupled with a multiplexing technology and a numerical inversion method.

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