# A Multiscale finite element model to predict the diffusional behaviour of biocomposites dedicated to structural applications

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# A MULTISCALE FINITE ELEMENT MODEL TO PREDICT THE DIFFUSIONAL BEHAVIOUR OF BIOCOMPOSITES DEDICATED TO STRUCTURAL APPLICATIONS

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**Abstract**
Predicting the in-service durability of a structure is a challenge for engineers, especially when water or moisture are involved. Therefore, a model-based approach which allows assessing the effect of this media on the material's behaviour is proposed in this study. Its first step consists in simulating the water diffusion in the material as a function of temperature and stress state. For composite materials, this simulation is complicated by the heterogeneous nature of the material and by its multiscale organization. The purpose of this research is to develop a water diffusion numerical model that accounts for the actual microstructure of a biocomposite. The material under study is a unidirectionally flax reinforced polyester composite dedicated to ship structures. Two observation scales are considered: the yarn scale composed of bulk matrix, single fibres and bundles, and the composite scale with bulk matrix and yarns. A so-called 'direct' model where geometry replicates accurately an example of a real microstructure was compared to a 'parametric' model which is built up from several key parameters assesed from a microscopic analysis of this composite. First results show that the parametric model leads to a reliable prediction of the water diffusion at the studied scales.

## 1. Introduction

Natural fibres reinforced composites (NFRC) are promising materials for the substitution of glass fibres reinforced composites (GFRC) for structural applications. They present several advantages over GFRC such as interesting specific mechanical properties, good damping ability and impact strength [1]. However NFRC remain rarely used in structural applications because of their poor durability in humid environment due to the hydrophilicity of natural fibres [2, 3]. Some authors have shown that static and dynamic mechanical properties of NFRC can be strongly impacted by water aging [4-6], while others demonstrate that NFRC fatigue behavior can be improved after humid ageing [7]. Interesting work on the chemistry approach has been performed to reduce the affinity of such fibre with water that lead to improve the long term behaviour of these biocomposites [8].
Predicting the modification of NFRC mechanical properties in humid environment would help designing structures exposed to real ageing conditions. A reliable computation is challenging since it requires knowing the evolution of mechanical properties of the constituents as a function of water content, but also the kinetics of water diffusion in a complex multiscale microstructure [9-11]. El Hadi et al. evaluated experimentally the diffusion kinetics in the three directions of flax/epoxy composites

[12]. This approach is interesting but limited to a specific composite material. The numerical approach proposed in this work integrates the impact of the flax fabrics morphology on the diffusional behaviour of the NFRC. A multiscale finite element model has been developed in order to predict the diffusion of water in a biocomposite based on the diffusional properties of its own constituents.

## 2. Materials and Methods

### 2.1. Composite Materials

The composite studied here is an unidirectional flax fibre reinforced unsaturated polyester (FFRC) produced by vacuum infusion. The flax fabric have been supplied by Fibre Recherche Developpement (Troyes, France), and the Enydyne® polyester resin by Cray Valley (Exton, USA). It is made of four 390g/$m^2$ plies with 29% wt. fibre. Its transverse multiscale microstructure is displayed in Figure 1. There are two scales of observation for the FFRC, the yarn scale with unitary fibres and bundles homogeneously dispersed into the matrix, and the composite scale with yarns arranged in staggered rows into the matrix.

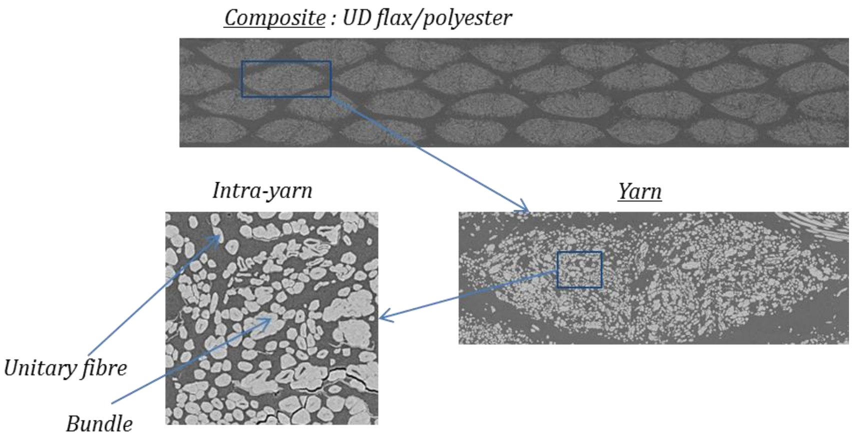


**Figure 1.** Multiscale microstructure of the unidirectional flax fibre reinforced polyester.

Samples of 250 x 25 mm were cut from composite sheets using a diamond blade. Thickness ranged from 2.7 to 3.4 mm. Small samples of polyester (15 x 8 x 3 mm) were also produced for this study. All samples were then stored in a climatic room at 23°C and 50% rh before testing.

### 2.2. Aging methods

Samples were immersed into water for more than 6 months at 30°C while their weight uptake was monitored by gravimetric analysis. The relative water uptake $W$ was determined according to equation (Eq. 1) :

$$W = 100\ (w_t - w_0)/w_0 \qquad (1)$$

Where $w_t$ is the sample mass at time t and $w_0$ the initial mass.

### 2.3. Image analysis

Before immersion, cross sections of samples were polished and coated with a thin layer of graphite to be observed in an environmental scanning electron microscope FEI Quanta 200 ESEM. Images were then treated with the analysis software Aphelion® to statistically determine the morphological parameters of fibres, bundles and yarns presented in Figure 2. 50 images have been studied at the yarn scale and 15 images at the composite scale. Feret ratio, defined as the ratio between Feret length and Feret width, was used to evaluate the shape factor of a particle.

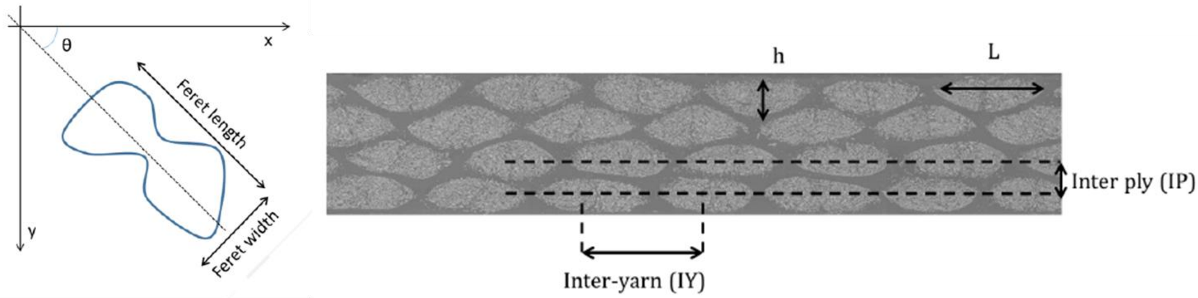


**Figure 2.** Definition of statistical parameters at yarn and composite scale.

## 3. Modelling strategy

To predict the water diffusion in the FFRC, the two steps process proposed in this study has been implemented as a numerical Finite Element model (FEM) in the software Comsol Multiphysics®. As the composite studied is unidirectionnally reinforced, it was chosen to model the 2D diffusion behaviour in the cross section transverselly to fibres. Firstly, the diffusional behaviour of yarns is determined based on the diffusion properties of flax and polyester of a representative geometric model. These properties can then be used in the upper composite scale model along with polyester diffusion properties to evaluate the global multilayer material properties (grey box in Figure 3). In this first approach, the diffusion is supposed to be Fickian in the matrix, fibres, yarns and composite as most FFRC are shown to follow this law [13, 14].

Diffusion coefficient $D_f$ and water uptake at saturation $Ws_f$ of flax fibres have been taken from literature [15]. Based on water uptake experiments on polyester samples, an inverse method applied to the FE diffusion model of a parallelepipedic 3D geometry allowed assessing the matrix diffusion parameters ($D_m$, $Ws_m$).

In order to create the FEM geometries both at the yarn scale and at the composite scale, two approaches have been carried out and compared. Firstly, a so-called "direct" approach consisted in building a geometry with the exact contour of fibres and bundles (resp. yarns) at the yarn scale (resp. composite scale). This model is easy and fast to implement in Comsol Multiphysics® and does not require a thourough image analysis, it is however difficult to handle because of a heavy mesh (up to 1 million elements). The other approach consists in building a parametric model based on the parameters determined by image analysis at the two scales. Digimat® software is used to create the yarn scale geometry containing elliptic unitary fibres and bundles with random orientation. This geometry is then imported in Comsol Multiphysics® to be meshed. For the composite scale model, parameters are selected randomly and used to build an elliptic geometry in Comsol Multiphysics®. The yarn diffusion properties identified by an homogeneization procedure in the yarn scale model are then implemented in the upper composite scale model (Figure 3)
The size of the model at the yarn scale is a critical parameter. Three parametric square models of 200, 400 and 700µm side have been studied in order to determine the size of a representative elementary surface (RES) from the diffusion physics point of view. Ten models of each size have been drawn and diffusion was simulated for each case. Water absorbed is plotted as a function of square root of time

and the dispersion on the water uptake at saturation and the dispersion on the slope at origin are evaluated and plotted as a function of the surface size (Figure 4).

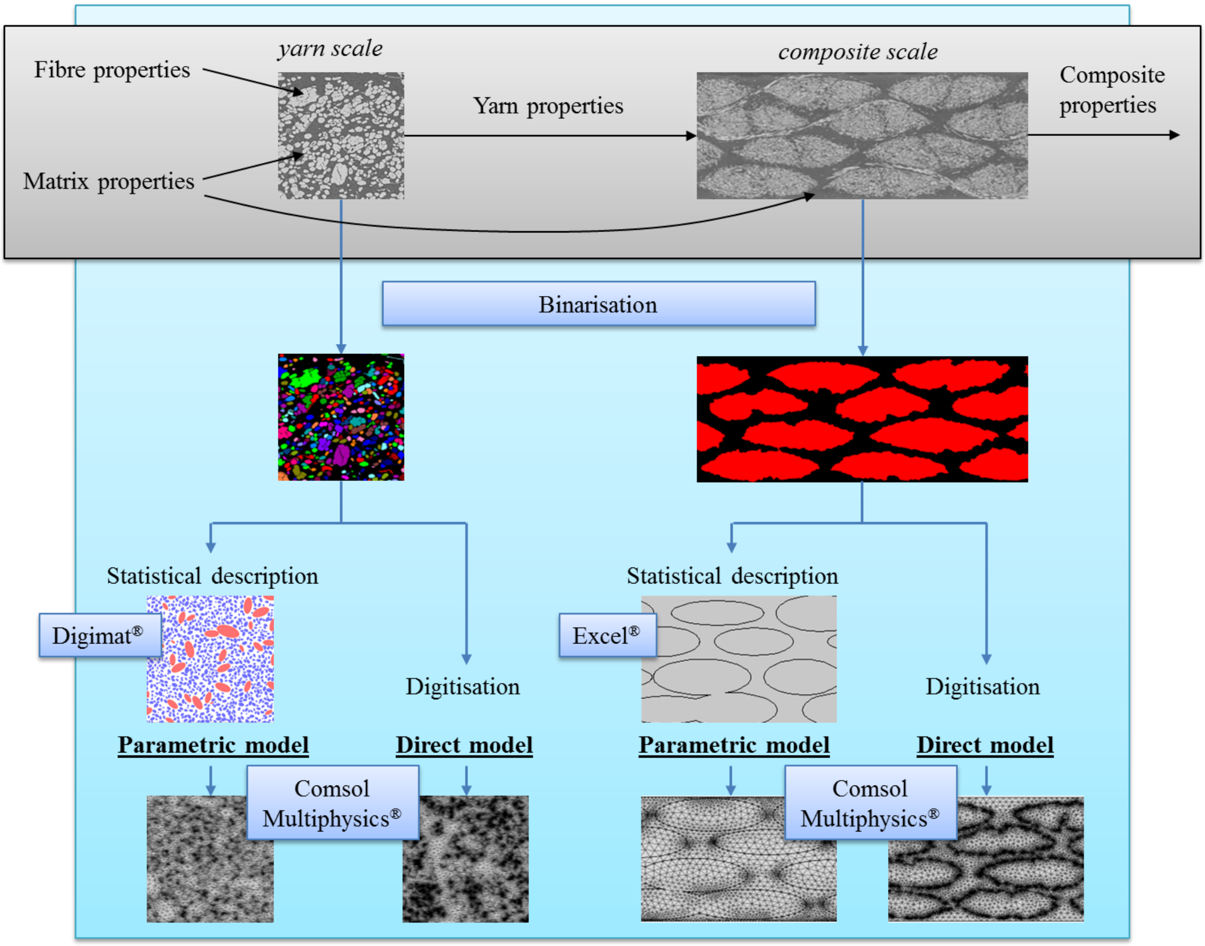


**Figure 3.** Schematic representation of the modeling strategy.

To model the Fickian diffusion of water, it is necessary to respect the continuity of parameters such as water uptake or local concentration at the interface between two components. This implies, in the case of a composite with two component absorbing water, to simulate the evolution of the undimensionned parameter $W_i/Ws_i$ (i represent the fibre or the matrix) which is continuous at interfaces as demonstrated by Liu and al. [16] and Yoon and al. [17].

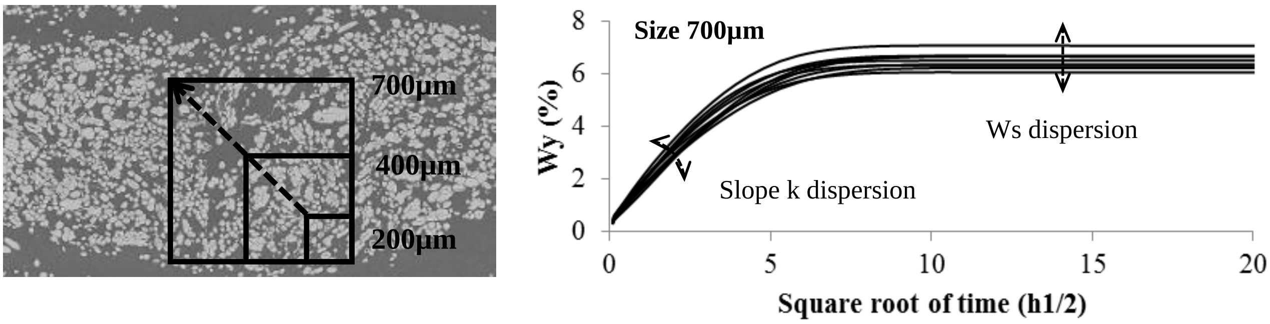


**Figure 4.** Impact of model size on the dispersion of absorption results at yarn scale.

Once the RES size determined, diffusion simulations were carried out at yarn scale on 10 'direct' models of that size and on 10 'parametric' models. The same work was done at composite scale on 5 'direct' and 'parametric' models. Average absorption curves were then plotted and compared to evaluate the representativeness of this parametric approach.

## 4. Results

### 4.1. Composite morphology

Results of the image analysis are gathered in Table 1. They are used to create the parametric model at the yarn and composite scales. At the yarn scale, the total reinforcement represents 46% of the surface, in which 34% are single fibres and 12% are bundles. Yarns represents 63% of each samples cross section. The overall fibre reinforcement surface is 29%.

**Table 1.** Statistical parameters of the composite morphology at two scale level.

| Composite scale | | Yarn scale | | |
|---|---|---|---|---|
| | | | Unitary fibres | Bundles |
| IP (mm) | 0.81 ± 0.12 | Feret length (µm) | 19.0 ± 5.4 | 68.7 ± 19.6 |
| IY (mm) | 2.37 ± 0.21 | Feret ratio | 1.43 ± 0.23 | 1.7 ± 0.43 |
| H (mm) | 0.72 ± 0.12 | Fibres Surface area (%) | 34 ± 2 | 12 ± 2 |
| L (mm) | 2.18 ± 0.17 | | | |
| Yarn Surface area (%) | 63 ± 2 | | | |

### 4.2. Yarn scale

As explained in paragraph 3, the impact of the model size is critical and a RES has to be determined in order to perform simulation and comparison between 'parametric' and 'direct' models. The size of the RES has been determined by analyzing the evolution of the dispersion of two parameters, i.e. water uptake at saturation (*Ws*) and slope at origin of absorption curve (*k*), as a function of the model size (Figure 5).

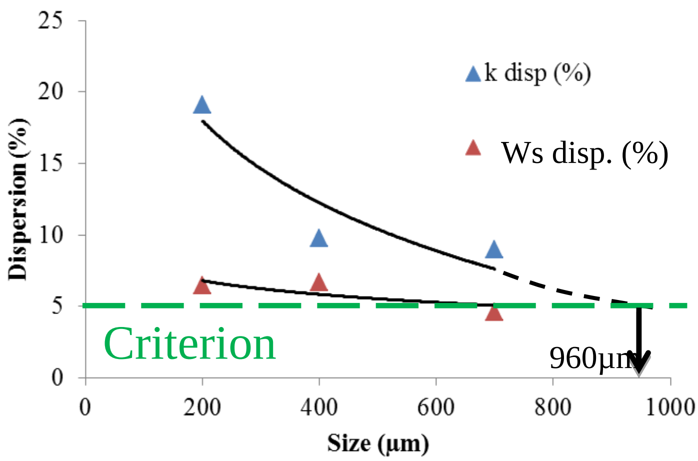


**Figure 5.** Determination of the diffusion representative surface at the yarn scale.

A criterion of 5% of dispersion was arbitrarily chosen below which it is considered that the size of the RES has been reached. An extrapolation of the logarithm function that best fits the evolution of these parameters gave a RES of 0,92mm$^2$ (square of 960µm side). This size is slightly lower than the average surface of yarns (1.1 mm$^2$) in the composite.

Diffusion parameters used in this numerical study are gathered in Table 2. The yarn properties were determined on a 2D homogeneous diffusion model by comparison with the average numerical sorption curve obtained with parametric models.

**Table 2.** Diffusion properties of the composite and its components.

| | Diffusion coefficient $D$ ($m^2$/s) | Water uptake at saturation $Ws$ (%) | Remark |
|---|---|---|---|
| Matrix | $8.10^{-13}$ | 0.9 | Experiments |
| Fibre | $1.10^{-10}$ | 14 | Litterature [15] |
| Yarn | $4,4.10^{-13}$ | 6.5 | Numerically identified |

Absorption curves at yarn scale are represented in Figure 6 for both 'direct' and 'parametric' models. They describe similar diffusion behaviours, thus validating our parametric approach. A Fickian behaviour is observed with an initial linear slope and a a final plateau. A simple calculation on a 2D homogeneous model allows identifying the yarn diffusion parameters in Table 2.

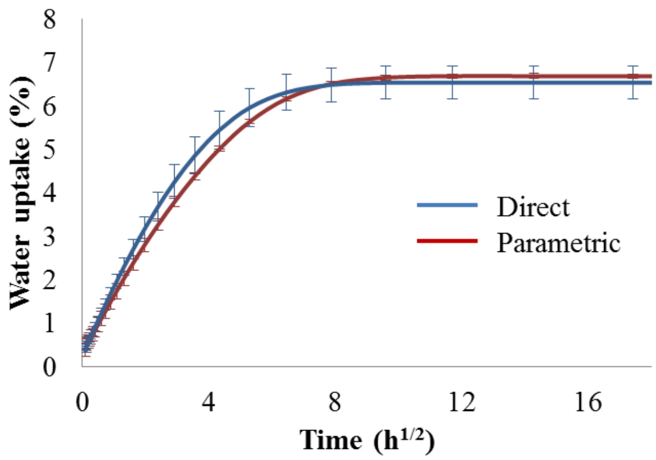


**Figure 6.** Absorption curves at yarn scale for 'direct' and 'parametric' model.

## 4.3. Composite scale

Numerical water uptake as a function of square root of time is represented in Figure 7 for both 'direct' and 'parametric' models at composite scale using the yarn and matrix diffusion properties displayed in Table 2. A similar behaviour is observed for both numerical approaches validating the choice of parameters at the composite scale. These results are then compared to experimental values. A significant deviation is observed between numerical and experimental results. This result is expectable since fibre and matrix data were both measured on bulk materials and since authors have shown that fibre and matrix properties can be altered in a composite laminate [9].

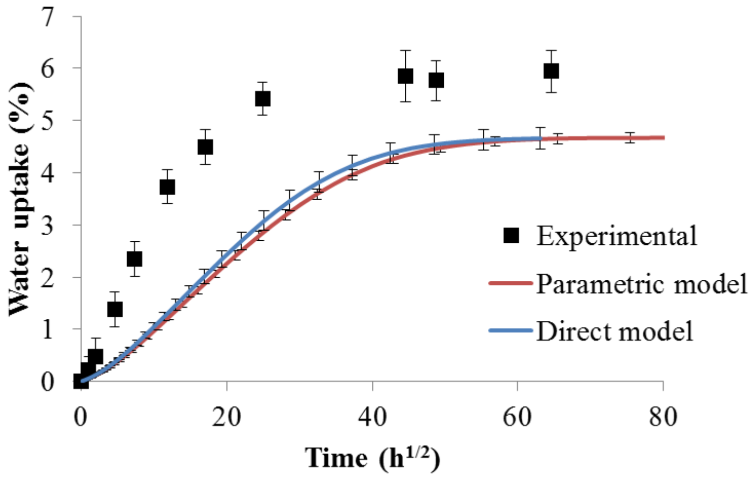


**Figure 7.** Absorption curves at composite scale for 'direct' and 'parametric' model compared with experimental water uptake for composite samples.

## 5. Conclusion

This work is a first attempt to develop a numerical model that can help predicting the durability of natural fibre reinforced composites. A thorough image analysis of the composite has been performed and two sets of parameters have been chosen to build simplified geometries of the composite at the yarn scale and at the sample's cross section scale. These parametric models have been compared with 'direct' models representing the actual microstructure of samples, and have been shown to give similar results for a set of materials properties experimentally measured by the authors or taken from the literature. An important deviation is observed when comparing the numerical water uptake for an entire composite sample and the experimental measurements. This can be explained by the fact that resin and fibres diffusion properties in the composite and in the bulk are probably very different.
An identification of the yarn properties could be performed using the composite scale model and then the fibre properties using the yarn scale model. Such approach is valid if one considers that the matrix is not modified in the composite.
Concerning the model itself, a very strong simplification of the real morphology of reinforcing elements has been performed using ellipsis to represent the complex contour of fibres and yarns. A significant improvement would consist in using statistical approaches to create more realistic objects as proposed by Mattrand et al [18].

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