

Multiscale modelling and optimisation strategies for better understanding of biopolymer mechanical properties: review of recent contributions

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Abstract – Some biopolymers are considered as interesting natural materials for the replacement of oil-based products. Others are known as complex food systems for which texture properties determine the sensory perception. For both applications, the need to understand the mechanical behavior of biopolymers is essential. Control of texture, improvement of mechanical performance are some of the issues for which a growing research interest is noticed. The understanding of the mechanical properties cannot be considered without valuable information about the microstructure. For such materials, structural heterogeneities, which affect the behavior, appear to be dependent on several interacting scales. It is the aim of this work to shed more light on the possibility to use optimization techniques to either identify or optimize the mechanical properties of starch-based materials at different scales. We take the example of the starch cellular material, which proved to be necessitating a multiscale approach. The context of the present study is related to the investigation of elasticity behavior of cellular materials.

Key words: Biopolymers, Cellular materials, Composites, Elasticity, Microstructure, Optimization

1 Introduction

Cellular materials are known to allow interesting functional properties such as resistance to heat transfer, impact, noise propagation, etc. The structure of cellular materials plays a key role in the determination of their mechanical performance. One of the well known structural effect is the amount of void. Gibson and co-workers [1] have determined a correlation between the elastic parameters and the ratio of void in arbitrary cellular materials with periodic arrangement of cells. Such correlation writes as follows for the elastic modulus

$$E_Y = C\rho^n \quad (1)$$

where E_Y is the modulus of elasticity, ρ is the relative density, i.e., density of the cellular materials over that of the dense material, C and n are constants to be determined.

Some other contributions fed the concerned literature with series of studies describing the role of other structural features such as cell size distribution, defects, etc [2-5].

Size effects have also been described leading to several limitations for the generalisation of the effective properties of cellular materials [6]. The main limitation is related to the contribution of free surfaces which affect significantly bulk properties if the cell size is comparable to the specimen size. Biopolymer cellular materials such as bread crumb [7] add another structural complexity (Fig. 1), which is related to the constitutive material. In fact, such material appears to be a composite with a randomly structured phases [8, 9]. Mechanical properties as well as microstructure of biopolymer

cellular materials have been widely studied in the past decades [10-14].

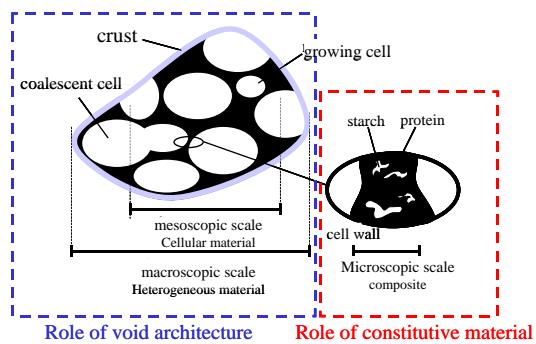


Fig. 1. Biopolymer cellular material within the multiscale approach (Example of the bread structure).

Finite element approach has been utilised to predict elastic parameters of cellular materials taking into account the explicit void architecture [15]. The technique is based on the conversion of voxels belonging to the solid phase to solid elements. This approach contrasts with other numerical approaches such as the effective medium theory [16, 17]. In the following is a description of the main advances realised on the assessment of biopolymer cellular material properties with respect to structural descriptors. Two main sections are considered. The first one is dedicated to the prediction of the mechanical properties of the struts, being considered as a composite material. The second section is dedicated to the optimisation of biopolymer cellular materials taking advantage of the architecture and the predicted strut behaviour.

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2 Biopolymer-based composites

Fig.2 shows a typical microstructure of a starch-based composites revealed using SCLM (Confocal laser scanning microscopy) [18]. The dark gray levels represents starch whereas the light ones are relative to zein (protein). Because of phase compatibility, during the forming process, diffusion of both phases leads to a high level of percolation. In order to determine the effective properties we need to assess the following aspects:

- Mechanical behavior of intrinsic materials;
- Nature of the interface and its corresponding effect on the effective properties;
- Sensitivity of the mechanical behavior to the microstructure features.

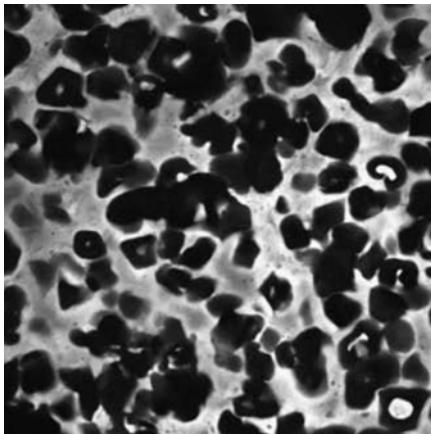


Fig. 2. Randomly structured microstructure of a typical biopolymer composite using SCLM. Image size about 140x140 μm^2 .

In order to proceed with the first aspect, both macroscopic and microscopic investigations can be considered to determine the behavior of intrinsic materials. Macroscopic testing would, of course, require designing composites with similar feature to what is observed at the microscopic scale. Thermomoulding is a way of designing such materials, allowing the control of the phase ratio in the full range [19]. Extracting the mechanical parameters of the composite requires optimization technique to identify the optimal set of parameters fitting the observed response. For such purpose, three-point bending tests are performed on strips ($38 \times 10 \times 1 \text{ mm}^3$). Finite Element model is implemented to simulate sample bending under the same testing conditions (Fig. 3a). To each solid element is associated a material behavior that takes into account elasticity and plasticity stages. In our case, a bilinear model is considered because it describes the material behavior with a small number of mechanical parameters, namely Young's modulus, yield stress and tangent modulus [20]. If the simulation is performed in the first linear stage (Fig. 3b), one mechanical parameter is tuned in order to predict the optimal elastic modulus. The objective function can be written as follows

$$R = \frac{1}{N} \sum_{i=1}^N (F_{\text{Exp}}(\delta_i) - F_{\text{Num}}(\delta_i)) \quad (2)$$

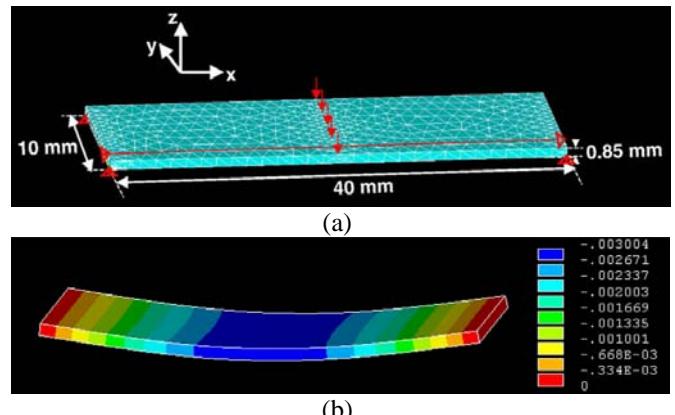


Fig. 3. (a) Loading conditions for the simulation of three-point bending test. (b)

Where F_{exp} and F_{num} are the experimental and numerical forces corresponding to a given deflection δ_i , respectively. N is the total number of deflection points for which the matching is searched.

The objective function is minimized by assuming a Newton-Raphson iterative scheme:

$$E_Y(t) = E_Y(t-1) - \alpha \frac{R(E_Y, t-1)}{R'(E_Y, t-1)} \quad (3)$$

where E_Y is the modulus of elasticity. R' is the first derivative of the objective function. t refers to the iteration level and α scales the rate of convergence.

The predicted results show that the modulus of elasticity is in the range 0.5 – 1.5 GPa for a composite containing 70% of starch [20].

Interface properties can be extracted from local measurements such as by nanoindentation [21]. Following the idea that the mechanical properties of biopolymers are sensitive to water content, viscoelasticity is investigated using a coupling between experimental and modeling techniques [22]. A grid of indents is performed near starch/zein interface to follow the change of mechanical properties across the interface. The experimental force – depth of penetration curves are compared to the equivalent numerical response taking into account a viscoelastic model. Such a model is described using three parameters, namely decay factor (β), initial (E_{YI}) and final moduli (E_{YF}). These parameters are related using the expression [22]

$$E_Y = E_{YF} + (E_{YI} - E_{YF}) e^{-\beta t} \quad (4)$$

The constitutive law is implemented in the Finite Element (FE) model using the meshing shown in Fig. 4.

Considering several combinations of the mechanical parameters (β , E_{YI} , E_{YF}), FE runs result in series of predicted force – depth of penetration curves. Identification of the experimental behavior is performed based on two criteria (F_{min} , F_{max}). These represent particular force values corresponding to the deepest penetration during loading and the final penetration during unloading, respectively.

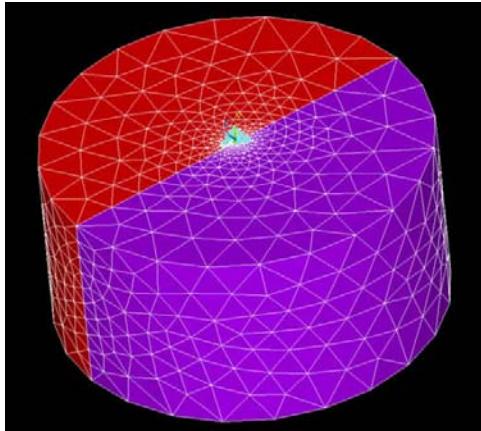


Fig. 4. Solid meshing of the composite and the indent to simulate nanoindentation at the interface of a starch/zein composite.

The application of an automatic fitting procedure gives the exact form of the objective functions relative to the force criteria.

$$R_{\text{MIN}} = \sqrt{(F_{\text{MIN}}^{\text{EXP}} - F_{\text{MIN}}^{\text{NUM}})^2} \quad (5)$$

where R_{MIN} is the objective function corresponding to the criterion F_{MIN} . EXP and NUM subscripts refer to the numerical and experimental responses, respectively.

An equivalent expression is derived for R_{MAX} . The fitting procedure well shows that different expressions are predicted for starch and zein phases. These write

$$F_{\text{ZEIN}}^{\text{ZEIN}}_i = aE_{YI} + bE_{YF} + c\beta + d \quad (6)$$

$$F_{\text{STARCH}}^{\text{STARCH}}_i = \exp(aE_{YI} + bE_{YF} + c\beta + d) \quad (7)$$

where i takes one of the following indices MIN or MAX.

Fitting parameters a , b , c and d are fully determined in each case with a correlation factor ranging from 0.85 to 1.00.

Equations (6) and (7) are used to determine, for each indent conditions, the optimal mechanical parameters. The predicted results show that both phases have similar mechanical properties. The relative difference between the intrinsic materials is 17%, 6% and 2%, for E_{YI} , E_{YF} and β , respectively.

Small difference between the intrinsic phase properties contrasts with the large deviation (up to 50%) of the numerical results with regard to those experimental conditions related to the interface indentation [22]. Such discrepancy is explained by the weak adhesion between constituents.

In order to determine the effective properties of the considered composite, FE simulation must take into account the weak adhesion effect. Such effect can be either approximated as an interphase [8] (third phase) which has intermediate properties with respect to intrinsic materials, or by considering imperfect interface action [23].

The former approximation allows the prediction of optimal interphase properties taking advantage of real microstructures and known intrinsic material properties. For such a purpose, SCLM images are converted into finite element meshes (Fig. 5). When an interface unit is detected, the sur-

rounding region is attributed material properties different from the intrinsic materials. The interphase material is thus characterized by two main parameters (Young modulus E_i and thickness t).

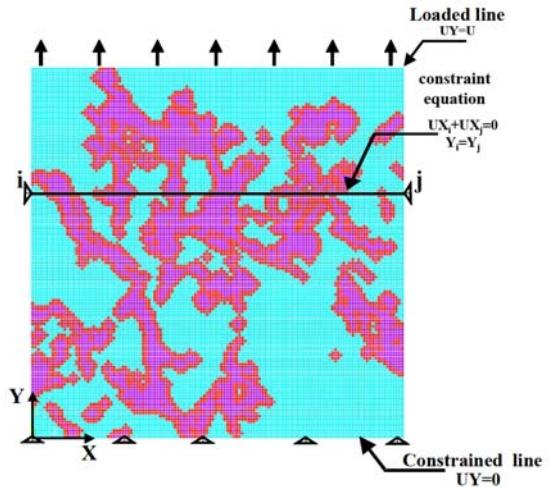


Fig. 5. Meshing and loading conditions for a composite microstructure under three-phase approximation.

The identification of the optimal interphase parameters requires experimental data that are available from three-point bending tests. The identification procedure is conducted by changing the interphase parameters (E_i , t) for each zein fraction considered in the experimental database. FE runs are performed to determine the effective modulus. Interpolation procedure is used to predict the optimal combination that allows the matching of the experimental and numerical moduli.

The predicted results show that the effective modulus is sensitive to the filler fraction and the ratio E_i/t [8]. The identification of the interphase parameters demonstrates that these are sensitive to the zein fraction. The interphase thickness varies between 0.31 and 1.24 μm , whereas the interphase modulus is found in the range 1.8 – 2.5 GPa.

It is worth mentioning that interphase approximation is mesh dependent. Interphase thickness less than the resolution of the image are not predictable. Moreover, interface action is not fully understood since the interphase region does not represent the true stress pattern near the interface. In order to overcome these difficulties, the implementation of the explicit interface behavior is suggested [23]. The model is based on the implementation of 1D interface elements characterized by tensile and shear effects. Indeed, to each element is associated a cohesive interface model characterized by the maximum traction, normal and shear separations. The simulation of tensile loading predicts different displacement patterns depending on the spatial distribution of phases. When the phase ratio is small, disconnected regions lead to local interface jumps [23]. When increasing further more the phase ratio, percolation of interfaces makes the interface effect stronger (Fig. 6). At this stage, the predicted effective properties are the smallest ones at the percolation threshold. Any adjustment of the mechanical parameters associated to the cohesive model leads to lower performance when the interface content increases. Such effect is not confirmed by the experimental testing [8, 9]. More complex variations are instead observed. For instance, different water content equi-

librums between the intrinsic phases change the interface properties. In order to determine the optimal interface parameters, neural computation and microstructure generation are exploited. Indeed, the search of optimal mechanical parameters cannot be decoupled from the microstructure information. Unfortunately, real microstructures such as that shown in Fig.2 are limited to allow independent following of all structural features. Microstructure generation is utilized to resolve such problem based on Monte Carlo technique [24]. The idea is to minimize interface energy and adjust generation parameters to control the interface content. Such algorithm requires several parameters to be tuned, which makes optimization difficult to perform without considering stochastic approaches. The idea behind neural network is to relate the generation parameters, interface and phase properties to the effective properties. By doing so, we are able to implicitly determine the shape of the objective function [24]. The predicted results show that tensile properties are more correlated to the maximum traction and the normal separation.

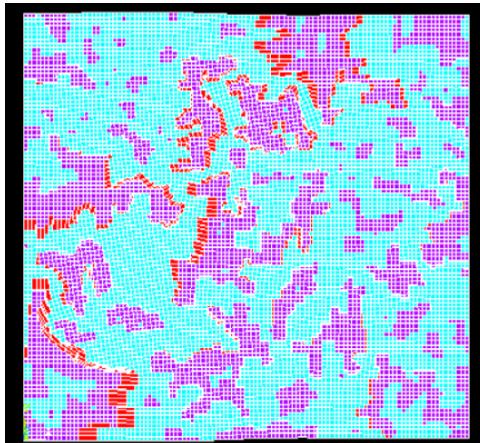


Fig. 6. Displacement pattern within a randomly structured composite showing interfacial displacement jumps.

3 Biopolymer-based cellular materials

As introduced above, cellular materials are dependent on the void ratio and on the mechanical properties of the intrinsic material. Since the effect of the former is elucidated, optimization techniques are used to assess the contribution of several structural features on the elasticity behavior of biopolymer cellular material. In this particular case, the forming process (ex. fermentation, extrusion) results in an open void structure. The void content occupies up to 90 % of the cellular material depending on the processing conditions. In order to represent the effect of void content, void size distribution and cell coalescence, microstructure generation is considered. Among the possible ways of void creation, Random Sequential Addition (RSA) [25] is a sequential process in which voids are positioned based on a spacing criterion δ (Fig. 7a). Such criterion writes

$$\delta \geq r_{ij} - \sum R_i \quad (8)$$

where r_{ij} is the center-to-center distance between spherical voids, R_i is the radius of void i .

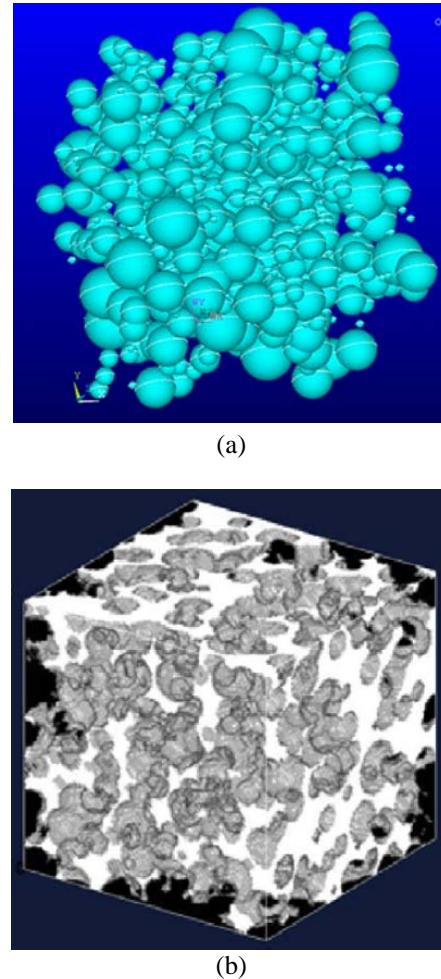


Fig. 7. (a) Generation of void structure using RSA (Random Sequential Addition). (b) Cellular material characterized by a void size distribution.

The void number is increased until the desired relative density is achieved. Note that any addition of voids would require that equation (8) being satisfied for all preceding voids. Size distribution is introduced by allowing R_i to be changed in equation 8 (Fig 7b). Gaussian size distribution can be considered with two control parameters: center and width of the void distribution [15].

3D images are converted into a FE model assuming that each voxel represents a solid element. Elastic properties of the intrinsic material are implemented. Uniaxial loading is simulated to allow the computation of the effective elastic parameters. The control of the effective properties, such as the elastic modulus requires the development of a hybrid strategy, which exploits a FE database representing the mechanical response for several combinations of input parameters. Neural computation is used to discover all causal correlations between the generation parameters, cellular structure and the mechanical parameters [26]. The generalization property of the optimized neural network predicts the iso-contours of the outputs as function of the input parameters in the hyperspace of the generation parameters.

4 Summary

The control of the mechanical behavior of biopolymer cellular materials requires a multiscale approach. Indeed, the effective properties are both affected by the cellular architec-

ture and the properties of the solid material. The latter belongs to a particular category of composites characterized by random distribution of phases, weak contrast between phases and a prevailing role of interface. Optimization can be used in two different situations to help assessing the mechanical behavior of biopolymer-based materials.

It can be used, firstly, for the identification of mechanical parameters in a micromechanical model such as for the determination of the interface/interphase properties. For this purpose, optimization is used to minimize the difference between experimental and numerical responses.

Optimization is also used, secondarily, to realize a sensitivity analysis in which the control of the mechanical parameters requires handling of a large number of input parameters. In such a way, large FE database is necessary to address the correlations in the space of input parameters. Neural network is used to implicitly encode such correlations. The optimal neural network is obtained thanks to a training procedure in which the objective function represents the difference between the predicted and the experimental/FE responses. Such a function is dependent on the internal parameters of the neural network.

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