

Research Article

# Young's Modulus of Macroporous Bioceramics: Measurement and Numerical Simulation

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**Abstract** A biphasic calcium phosphate (BCP) powder is synthesized, mixed with various amounts of naphthalene particles, pressed and sintered at different temperatures to obtain ceramics with isolated macropores (from 3% to 52% of the specimen volume) and residual microporosity resulting from an incomplete sintering (from 2% to 45% of the ceramic matrix volume). Young's modulus is measured on a classical three-point bending setup. A good agreement is found between an existing analytical model and the experimental Young's modulus measurements, on the overall ranges of macro- and microporosity. The Young's modulus variation as a function of macroporosity is also calculated by a finite element method, by simulating the mechanical response of a periodic tri-dimensional repetition of elemental cubes containing various dispersions of macropores. The contribution of the shape and dispersion (in size and location) of the macropores on the decrease of Young's modulus as a function of porosity is simulated. Calculated trends are confirmed by experimental results.

**Keywords** biomaterial; modeling; modelling; apatite; microstructure; FEM; FEA

## 1 Introduction and model presentation

Biphasic calcium phosphates (BCP), used for bone substitution, are mixtures of Hydroxyapatite,  $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$  and  $\beta$ -tri-calcium phosphate,  $\text{Ca}_3(\text{PO}_4)_2$ . The best bone substitution behavior is obtained for materials exhibiting a double-scale porosity, combining isolated macropores of several hundreds of micrometers, and interconnected micropores of a few micrometers in size [1]. The former allow bone cells to develop in the material and the latter allow an impregnation of the ceramic by biological fluids. However, mechanical properties of materials decrease with porosity; in view of possible load-bearing applications, it is therefore necessary to investigate the influence of microstructure on strength and stiffness. In 2006, a first modeling approach was proposed to describe the evolution

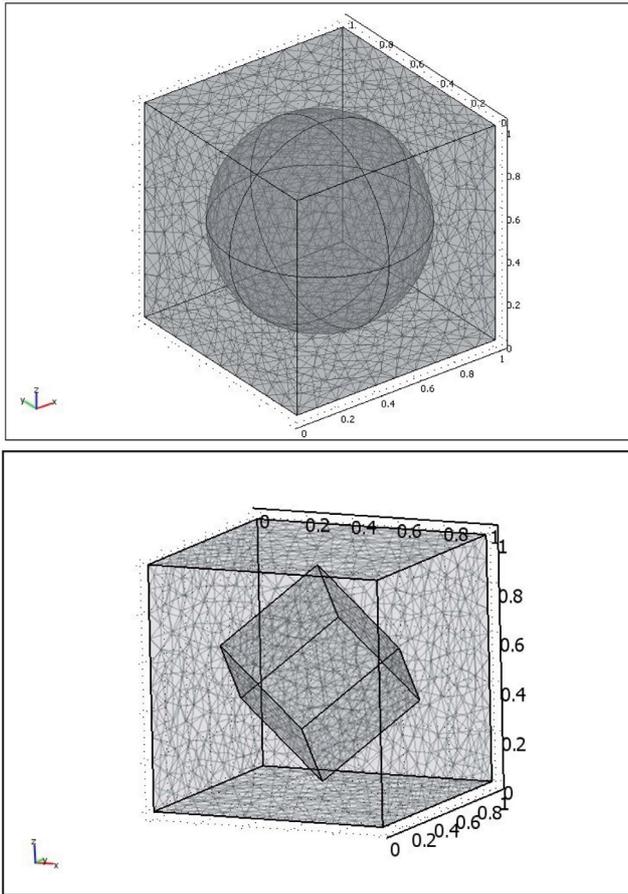
of mechanical properties of BCP ceramics with both macroporosity and microporosity [2]. In the case of Young's modulus, the developed model writes as follows:

$$E = E_0 \cdot \left[ N_C (1 - p_{\text{micro}}) - (N_C - 1) (1 - p_{\text{micro}})^{2/3} \right] \cdot (1 - p_{\text{macro}})^m \quad (1)$$

with  $p_{\text{macro}}$  the overall macroporosity,  $p_{\text{micro}}$  the local microporosity of the ceramic matrix between macropores,  $m$  a parameter depending on the morphology of macropores, and  $N_C$  the mean coordination number, that is, the average number of closest neighbors for each matrix grain.  $E$  and  $E_0$  are, respectively, Young's modulus of the porous ceramic and of the fully dense material (i.e. with  $p_{\text{micro}} = p_{\text{macro}} = 0$ ). Nevertheless, the model could not be completely validated due to a lack of experimental data over a wide range of porosities. One of the aims of the present study is to validate the model by experiment. The other aim is to investigate the possibility to use finite element simulation to describe the influence of macroporosity on Young's modulus, and to study the effect of microstructural parameters on stiffness.

## 2 Materials and methods

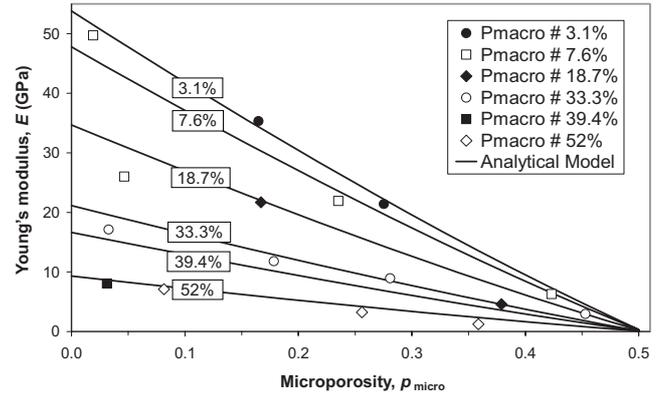
A calcium deficient apatite (CDA) of general formula  $\text{Ca}_{10-x}(\text{HPO}_4)_x(\text{PO}_4)_{6-x}(\text{OH})_{2-x}$  is first synthesized by hydrolysis of  $\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$  in an aqueous solution of  $\text{NH}_4\text{OH}$  heated at 70 °C and maintained under stirring for 6 h. The initial pH and the reactant stoichiometry are chosen to insure a final Ca/P ratio around 1.52 ( $x = 0.87$ ). The solution is then filtered and dried overnight at 120 °C. A porogen (naphthalene particles with a diameter ranging from 200  $\mu\text{m}$  to 633  $\mu\text{m}$ ) is then mixed with the CDA powder in a Turbula shaker-mixer. The resulting mixture is then cold isostatically compacted under a pressure of 140 MPa. After sublimation of naphthalene at  $\sim 80$  °C and a plateau of 1 h at 400 °C to eliminate residual traces of water or of porogen, the material is pressureless sintered in air for 8 h



**Figure 1:** Examples of simulated porous structures, and the corresponding meshes.

at temperatures from 950 °C to 1150 °C. Such temperatures also provoke a complete transformation of the starting CDA into BCP. The resulting ceramics exhibit macroporosities ranging from 3% to 52% (measured by image analysis on polished cross-sections) and microporosities from 2% to 45% (deduced from apparent density and macroporosity). Parallelepipedic bars ( $\sim 10 \text{ mm} \times 10 \text{ mm} \times 50 \text{ mm}$ ) are cut out from the blocks. For each material, several specimens are tested on a three-point bending setup. The deflection is measured with a precision lower than a micrometer thanks to a linear variable differential transformer (LVDT) sensor.

For finite element simulation, the porous material is assimilated to a tri-dimensional repetition of elemental cubes (Figure 1) of a continuous microporous ceramic matrix containing one or a few macropores of various shapes, sizes and distributions. Macroporosity is varied by changing the relative pore/cube size ratio. Simulated compression tests are then performed on the cubes by a finite element method (Comsol Multiphysics software) using periodic strain boundary conditions. The elastic response of the material to a uniaxial strain is simulated.



**Figure 2:** The measured Young modulus of the BCP as a function of the microporosity; solid lines represent the calculated variation of the analytical model for the mean of the macroporosities of each selected sample.

The computed reaction force is used to calculate the mean stress on the cube section and, once divided by the imposed mean strain, gives the apparent Young modulus.

### 3 Results and discussion

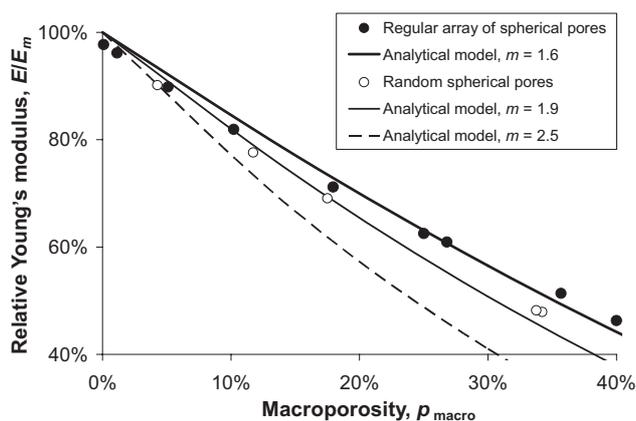
#### 3.1 Experimental results

The measured Young modulus is plotted in Figure 2 as a function of microporosity, for different values of macroporosity. Also plotted are solid lines representing evolutions calculated using the model (see (1)). The following values of model parameters have been determined by fitting:  $E_0 = 57 \text{ GPa}$ ,  $m = 2.5$ ,  $N_C = 4.8$ . A good agreement is obtained between the analytical model and the measured values (Figure 2), which tends to confirm the validity of the model to describe the variation of Young's modulus as a function of both macroporosity and microporosity.

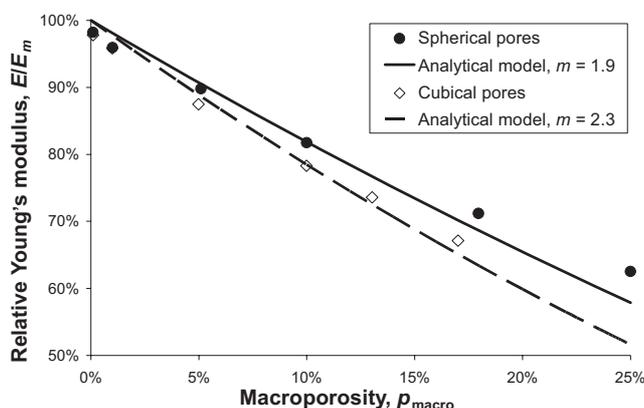
#### 3.2 Finite element simulation

Some of the results are shown in Figure 3, where the relative Young modulus  $E/E_m$  (with  $E_m$  the modulus of the ceramic matrix) is plotted as a function of macroporosity for different distributions in pore size and location. Also plotted, as a guide for the eyes, are solid lines representing evolutions calculated with the macroporosity dependence of (1), as  $E/E_m = (1 - p_{\text{macro}})^m$ . The first evolution to look at is the one represented by filled circles, corresponding to spherical pores of identical size regularly spaced in the material volume on a simple cubic array. This variation can be reasonably well described by the equation with  $m = 1.6$ , which is somewhat lower than the experimentally determined value of  $m = 2.5$ .

The second set of simulations is also represented in Figure 3, corresponding to a random distribution of eight spherical pores in one elemental cube. The introduction



**Figure 3:** The simulated relative Young modulus of the BCP as a function of macroporosity; solid lines represent the best description of the calculated values by the analytical model.



**Figure 4:** The simulated relative Young modulus of the BCP as a function of macroporosity; the curves represent the best possible description of the calculated values by the analytical model.

of randomness in the macroporous structure (pore size randomly distributed and pores randomly scattered in the elemental cube) decreases the mechanical response of the material. The value of the  $m$  parameter of the analytical model increases from 1.6 to 1.9 as the disorder increases, getting closer to the experimental value of 2.5. This is consistent with the hypothesis of Wagh et al. [3], suggesting that the parameter  $m$  would be representative of the “tortuosity” of the porous structure. In this respect, increasing the randomness of the porous structure is consistent with an increase in “tortuosity”. Also, the even more complex macroporous structure of the actual ceramic is consistent with the higher (experimental) value of  $m = 2.5$ .

A third set of simulations corresponds to a change in pore shape, where spherical pores are replaced by cubical

pores with their long diagonal being parallel to an edge of elemental cube (see Figure 1). The cubical pores concentrate the local stresses on their edges and corners (more than the spheres do on their surface), inducing a faster decrease in Young’s modulus with porosity (Figure 4). This also corresponds to a change in the apparent parameter  $m$ , which increases from 1.9 for spherical pores to 2.3 for cubical pores, getting even closer to the actual value of 2.5. This rather high experimental value can thus be considered to be the result of the combination of shape effects (naphthalene particles are polygons) as well as distribution effects (in size and location).

#### 4 Conclusions

The analytical model for Young’s modulus variation as a function of the two porosity types, taken separately, can be used to describe the experimental measurements. The simulation study revealed the contribution of changes in macroporosity morphology (in shape and distribution) to the decrease of stiffness. The relevance of the finite element simulation method to predict the mechanical influence of macroporosity is partially confirmed by this study. These first results tend to show that finite element simulation is an acceptable method to investigate a priori the influence of porosity on mechanical properties. In the future, this tool will be used to simulate the behavior of materials with more complex morphologies and to design optimized microstructures, which will then need to be fabricated and tested.

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