Multi-scale physico-chemical characterization of CEB/ANS bio-composites

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Abstract. In a vision to identify the non-linear behaviour of the compressed earth blocks (CEB) reinforced by the Argan nut shells particles (ANS) influenced by many parameters like the shape, the distribution and the quantity of the stabilizers, as well as the interactions between both phases: matrix and reinforcement. The use of numerical models seems to be indispensable. Yet, simulations of heterogeneous structures quickly become unaffordable by direct calculations on finite element software. Therefore, a homogenization of the experimental, analytical, and numerical macrostructure is performed. Thus, an overall micro-meso-macro approach to modelling the mechanical behaviour of CEB/CNA bio-composites has been established. It is mainly based on the notion of the representative elementary volume with two different structures (periodic structure and structure with a Poisson distribution). The numerical and analytical homogenization results were validated by the Young's modulus values resulting from the experimental compression test and the corresponding stress-strain curves.

1. Introduction

The behaviour of earthen building materials with respect to loading remains a very complex issue. Indeed, numerical modelling of these materials has to reliably reproduce local discontinuous behaviours as well as the global functioning of the structure [1,2] However, the scientific instructions concerning the soil, particularly with regard to its mechanical behaviour, are insufficient due to the heterogeneity of the different constituents of the soil (gravel, sand, silt and clay). The bio-fillers consist mainly of cellulose, hemicelluloses and lignin. The cellulose guarantees a protection of the earthen blocks from water attacks. The choice of organic material was made on the shell of argan tree nuts, presenting up to 26% cellulose and 34% hemicelluloses [3]. This mixture promises a good adhesion with the constituents of the soil and an improvement of its mechanical and thermal characteristics.

The overall vision of this chapter is to predict the macroscopic elastic behaviour of heterogeneous compressed earth blocks (CEB) reinforced with Argan nut shells (ANS) particles. Two multi-scale homogenization approaches are discussed throughout this study. A numerical approach based on the finite element method and an experimental approach based on stress-strain curves. The interest of the present work is to compare between the two approaches of Young’s modulus homogenization for CEB/ANS composites. The choice of Young’s modulus as an effective property will allow to model our bio-composite as a homogeneous material and thus to study its mechanical behaviour to static and dynamic loading (Fig. 1 [1]).

![Homogenization Principle](image)

Fig1 Explanatory diagram of the homogenization principle [1].

2. Experimental homogenization

The purpose of this section is to provide experimental data for the characterization of the building material used. The direct compression test is employed, strain gauges were attached to the blocks as shown in Figure 2. Thus, the Young’s modulus value and stress-strain curve are evaluated for CEB and CEB/ANS composites.

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Although the main parameter recommended to study soil behaviour is Young’s modulus, this property is relatively unstable and unpredictable for soil materials. In addition, experimental stress measurements using strain gauges influence the curves obtained [4]. The results of this experimental study will be used to verify the accuracy of numerical and analytical homogenization techniques.

The experimental stress-strain relationship of the non-stabilized CEB is presented in Figure 3, showing the elastic-plastic behaviour of the material. The Young's modulus dispersions of the non-stabilized soils according to the three tests showed a very low variability, on the order of 1.6%, with an average Young's modulus reaching 31 MPa.

Similarly, Figure 4 shows the experimental stress-strain curve of CEB stabilized at 2% ANS. The bio-composite keeps a brittle behaviour and the values of Young's modulus of CEB/ANS according to the three tests described a very small dispersion, about 2.8%; Thus, the average Young's modulus of CEB in presence of 2% in ANS takes a value of 32.7 MPa. In the same sense, direct compression tests were used to evaluate the Young's modulus values for the 4% and 6% ANS loading of CEB. The results obtained are presented in Figure 5.

Young's modulus values for the different ANS loadings were identified. Nevertheless, it must be emphasized that soil materials are nonlinear and inelastic, so a standard determination of Young's modulus can be used only for comparison between the three homogenization approaches and as an indicator of material stiffness, however, these values do not define the overall behaviour of the soil material.
3. Numerical homogenization

Experimental testing of the mechanical properties of materials remains a very long and expensive approach. Also, the installation of strain gauges for the measurement of the integrated deformation of CEB generates some disturbances on the measured effective properties. Therefore, from an experimental point of view and in order to obtain accurate measurements, numerical modelling must take place to support the experimental results and save time. Good modelling by the Finite Element (FE) method leads to reliable, accurate and adaptive results, while keeping its economic aspect.

This section will mainly deal with the comparison between a random model and a periodic one, in order to numerically characterize particle reinforced composites, while introducing a “hybrid” model in order to understand the origin of the discrepancies between the results of the two models. This modelling will be carried out at the mesoscopic scale with the assumption of 3D deformations while considering the heterogeneity of the material and the contact surface.

In this context, the modelling of the basic data is an important step, knowing that its accuracy influences the reliability of the homogenization calculations. Therefore, the modelling technique must consider the different aspects of morphology and phase distribution within the matrix. First, a discretization of the whole heterogeneous material into a set of heterogeneous cubic squares is carried out. The second step is then to determine a representative elementary volume (VER) and to specify a database for the multiple phases constituting the bio-composite, including: volume fraction, distribution and relocation of stresses and strains from the macro to micro model. Third, a global behaviour of each heterogeneous parcel is obtained through the classical homogenization method described in Chapter 1. Then, the effective property, of each heterogeneous parcel is enriched at the macro level in terms of behaviour. A calculation is then performed to determine the macroscopic mechanical properties.

The homogenization method is based on the numerical homogenization used in the work of Lejeunes and Bourgeois, Peillex, Mbodj, Kanit and Nhu) [5, 6, 7, 8]. This method consists of modelling the VER and extracting its effective properties. There are many boundary conditions to be set up, including: kinematic uniform boundary (KUBC), stress uniform boundary (SUBC), and periodic boundary conditions (PBC), etc.

The properties of the soil material have been calculated previously, and concerning the shell of the argan nut used, the parameters such as density and Young’s modulus were determined by Essabir [2]. The set of mechanical characteristics associated with the materials soil and ANS is illustrated in Table 1. In addition, for simplicity, the ANS particle is represented as a spherical inclusion with a diameter of 0.63 mm. Linking elements are used between the inclusion and the soil matrix to ensure continuity between these constituents. The microstructures of this material are the main objective of our study for the representation, the 3D numerical simulation and finally for the study of the size representativeness of the specimens.

| Table 1. Mechanical properties associated with soil and ANS materials [3,4] |
|---|---|---|---|---|
| E (MPa) | ν | Density (g/cm³) | Volume fraction (%) | Diameter d (mm) |
| Soil | 31 | 0.33 | 2.17 | —— —— |
| ANS | 8000 | 0.33 | 1.3 | 2, 4, 6 | 0.63 |

Fig. 5. The evolution of Young's modulus as a function of ANS particle content.
3.1 Periodic microstructure

3.1.1 Modelling process

The geometry chosen for the periodic model is rectangular with a single inclusion is placed in the centre of the microstructure. All simulations are performed by the finite element code ABAQUS with 10 processor cores. In a vision of simplification, periodic models have been implemented [10]. This technique for predicting material properties reduces to a VER with a single cell [11, 12]. The model is based on the KUBC homogenization method presented in the previous section. The heterogeneous modelling, for which the spherical inclusion is centred in the 4 mm3 volume square, will now be replaced by a homogeneous problem. In other words, the heterogeneous patch is replaced by a homogeneous cube enriched by an effective modulus obtained by the KUBC homogenization method. The enriched homogeneous patch is meshed with 27000 hexahedral elements with a mesh size of 0.033mm. In this sense, the cube will have the shape of a periodic cell with a geometry of quadratic symmetry.

The present work aims to study the mechanical behaviour of the composite CEB/ANS, in particular to deduce the origin of the various instability regimes. Also, the influence of the boundary conditions of the material on its mechanical behaviour will be highlighted. With the help of a simple calculation, and as illustrated on figure 6, a periodic structure in the form of a cube with dimensions included in the interval [1.87 mm; 2.35 mm] with a single inclusion of ANS will be studied, this morphology will accept the application of kinematic and periodic boundary conditions.

The simulations are carried out with a deformation equal to 0.2 mm and a cube of dimension 2x2 mm. Numerical homogenization by FEM requires specific boundary conditions on the VER contour supporting the macroscopic deformation components. Homtools is a computational interface offering the possibility to implement the homogenization theories KUBC, SUBC and PBC, in small perturbations and also in large deformations.

No special post-processing for the calculation of the effective properties of the VER is necessary. Through the reference points, the shear modulus (G) and the compression wave modulus (M, P-wave modulus) are calculated according to the three directions of the triplet (X,Y,Z). Thus, the equation (1) allows to obtain the values of the Young’s modulus (E). This operation is optimized thanks to the PYTHON scripts allowing to automate the Young’s modulus data.

\[
E = \frac{G(2M-4G)}{M-2G}
\]  

3.1.2 Results and discussion

A mesh convergence study was performed. Different mesh sizes were used, from coarse to fine meshes. The evolution of Young’s modulus is plotted against the total number of elements in the heterogeneous plot. The results of homogenization of CEB stabilized at 2% ANS are shown in Figure 7.

![Fig.6. Microstructure with a spherical inclusion (a) 3D inclusion (b) porous microstructure. (c) matrix-inclusion. (d) mesh](image-url)
The periodic conditions give a good estimate of Young's modulus from $\approx 4000$ finite elements. Now the KUBC conditions, convergence is obtained using 24000 finite elements, a number six times larger than that of the PBC. Moreover, in the EL Houdaigui project [4], the convergence of properties is obtained from volumes with 100 grains with PBC, and 2000 grains for KUBC and SUBC. From these results, it is possible to consider that the size of the VER depends on the desired characteristic, the composition of the material and the phase contrast. The homogenized Young's modulus for the PBC and KUBC boundary conditions was calculated. Although the convergence of the periodic boundary conditions calculation is progressing faster, the use of the KUBC boundary conditions is important because the convergence of the two calculation methods implies that the selected VER are indeed representative of the macrostructure of the CEB/ANS bio-composite.

### 3.2 Random microstructure

Although the results of the periodic microstructure are reliable, parameters, such as phase distribution and boundary conditions, representative of real bio-composites are not integrated in this investigation. In this sense, a recourse to random microstructures took place for the continuation of the study [13, 14], thus, a comparative analysis between the two microstructures will be adopted [14, 15]. In what follows, the proposed strategy is based on the KUBC boundary conditions due to its ease and convergence.

#### 3.2.1 Homogenization process

The developed methodology is based on a three-dimensional two-phase random elastic microstructure. The random distribution of the ANS inclusions in the matrix is performed according to the Poisson process with positive repulsion distance to eliminate any risk of interconnection between the inclusions. The influence of microstructure on the elastic properties of CEB is investigated. Subsequently, the post-processing of the effective property will be reduced to three elementary loading cases along the directions (X, Y, Z) and several scenarios of ANS particle distribution, according to a Poisson process, will be treated while varying the size of the subdomains, in order to highlight statistical values of the Young's modulus of the CEB/ANS composite. The structures were modeled and mapped on the Digimat software (Fig. 8). Then, these models were imported into the calculation code Zebulon to perform a numerical homogenization in linear elasticity with kinematic boundary conditions. Thus, the whole of these calculations made it possible to determine the effective macroscopic properties of the composite. All the numerical models were subjected to a deformation of 0.5 mm in its three dimensions (X, Y, Z).

![Fig.8. Homogenization process of CEB/ANS bio-composites](image)
The multi-scale method consists in obtaining a convergence in the value of the Young's modulus of the composite according to different volumes of the cube, while keeping the same content of inclusions [5, 16]. In this vision, the number of inclusions, $N$, will be increased until the VER of each of the studied bio composites is obtained. For each volume, different realizations, $n$, will be investigated. In these realizations, it is important that the models have various morphologies thanks to a random distribution of the inclusions in the matrix in an optics of representativeness of the results. Table 2 below presents the number of these realizations as a function of the dimension of the cube edge $a$.

| $a$  | 2.2 | 2.5 | 3   | 3.5  | 5   | 10  |
|------|-----|-----|-----|------|-----|-----|
| $N$  | 2   | 3   | 5   | 7    | 21  | 168 |
| $n$  | 30  | 21  | 15  | 15   | 15  | 5   |

### 3.2.2 Results and discussion

First, the results of the variation of Young's modulus as a function of VER size, directions (X, Y, Z) and ANS particle distribution are shown in Figures 9-14.

- **Fig. 9.** Variation of Young's modulus with VER size ($a=2.2$ mm) for five realizations of the 2% CEB/ANS bio-composite with random particle distribution
- **Fig. 10.** Variation of Young's modulus with VER size ($a=2.5$ mm) for five realizations of the 2% CEB/ANS bio-composite with random particle distribution
- **Fig. 11.** Variation of Young's modulus with VER size ($a=3$ mm) for five realizations of the 2% CEB/ANS bio-composite with random particle distribution
- **Fig. 12.** Variation of Young's modulus with VER size ($a=3.5$ mm) for five realizations of the 2% CEB/ANS bio-composite with random particle distribution
- **Fig. 13.** Variation of Young's modulus with VER size ($a=5$ mm) for five realizations of the 2% CEB/ANS bio-composite with random particle distribution
- **Fig. 14.** Variation of Young's modulus with VER size and 2% CEB/ANS realizations.

The soil and ANS phases are considered isotropic. Initial simulations (Figures. 13, 14) have characterized the behavior of the composite material, at periodic boundary conditions, as isotropic.
Similarly, Figure 15 represents the variation of Young's modulus according to the number of inclusions. The dispersions between the Young's modulus according to the three dimensions obtained vary between 0.5% and 19%. For some morphologies, very low variation, of the order of 1%, was obtained. The results obtained here validate the isotropy of the CEB/ANS bio-composite, thus the marked influence is due to the presence of cut particles at the boundaries of the homogenization subdomain and the consideration of their effect on the boundary conditions.

![Figure 15. Variation of the Young's modulus according to the number of inclusions](image.png)

The Young's modulus values taken according to Figure 16 are:
- 31 MPa for unfilled CEB
- 32.45 MPa for CEB/ANS 2%.
- 33.94 MPa for CEB/ANS 4%.
- 34.01 MPa for CEB/ANS 6%.

The understanding of the mechanical behavior of materials is necessary to optimize the number of experimental tests. These properties have a direct application in calculation codes in order to predict new mechanical behaviors of characterized materials.

4. Comparative study between the two approaches

Some works like Bledzki and Gassan [17] have set up information about natural fiber reinforced polymer composites and its applications. This study has been extended by Faruk [18] for the years between 2000 and 2010. Generally, these bio composites are rigid, resistant, less dense and have the possibility of recycling compared to other commonly used inorganic fillers. These advantages place natural filler bio composites among the best performing materials offering economic and environmental benefits [19]. However, it should be noted that there are several difficulties associated with the integration of these fillers into polymer matrices, in particular, filler-matrix incompatibility. This problem can be a disadvantage when chemical means to improve compatibility are not available.

The contribution of natural reinforcements in bio composites has been the subject of several experimental research works. For example, Crespo [20] studied the elastic behavior of a new thermoplastic bio composite based on almond shell. During this study, the authors evaluated the effect of the proportion of aggregates on the elastic and morphological properties of the bio composite. It is noted here that the Young's modulus of the bio composite increases with the increase of the almond shell content. It also appears that the proportions of the filler influence
the morphology of the bio composite, as well as the dispersion of the filler in the matrix. The same approach was used by Kaci [21] concerning the characterization of a mixture of a polyethylene matrix and the olive shell. The results obtained also show that an increase in the proportion of the filler influences the Young's modulus of the bio composite. On the other hand, other works are interested in studying the elastic behavior of bio composites using numerical methods and multiscale analysis.

Indeed, the finite element method is the most widely used and plays an important role in the homogenization of this type of material. This approach was used by Silva [22] who compared the results of numerical simulation with those obtained by experimental characterization of epoxy resin reinforced with sisal and banana fibers. In their work, Behzad and Sain [23] present a review of the use of the finite element method for estimating the effective properties of hemp fiber reinforced bio composites. After comparing the simulation results with the experimental data, it appears that this method is robust and can be used to describe the macroscopic behavior of natural reinforcement materials. Overall, a good agreement between the numerical simulation results and the experimental ones was observed.

Until now, experimental methods are not sufficient to estimate the macroscopic properties of heterogeneous materials. For this reason, researchers should discuss around the use of imaging and design techniques. Many contributions offering direct links between real 3D images and FE calculations have appeared [24]. This is justified by the fact that physical properties generally depend on phase morphology.

In this part, a comparative study of the microstructure of a bio composite based on the soil. This bio composite, named CEB/ANS, consists of a matrix of soil (CEB) loaded by aggregates of the shell of Argan tree nuts (ANS). The validation of the results is done by comparing the Young's modulus obtained numerically with the value measured during the crushing of the Soil/ANS specimens (Fig. 17). The agreement between the numerical results and the experimental data decreases with the increase of the particle rate. This finding is explained by the fact that the experimental models reveal a better interfacial adhesion between the ANS particles and the soil matrix, while the numerical results showed that the interfacial adhesion between the natural reinforcements and the matrix decreases at a filler content higher than 4%.

This study revealed that the ANS grains are rather spherical, and the bio-composite has good interfacial adhesion between the ANS particles and the CEB matrix. Thus, the use of the natural reinforcement provided by the ANS particles improves the mechanical properties of the CEB.

5. Conclusion

The behavior of filled soil composites, despite the progress made in the understanding of some phenomena related to this material, remains a very complex issue, especially by the contacts that occur between its different phases during this process. In general, although the contact materials are heterogeneous, homogeneous assumptions have been considered. Yet, the scale of material heterogeneity must be incorporated for better predictions. Therefore, this section examines this dimension of material heterogeneity.

The approach is based on homogenization methods, at the micro level, allowing to obtain contact laws or homogenized coefficients reflecting the heterogeneities (mainly surface). Thus, these elements will be introduced in the global model of the studied system. Basically, the contact surface is discretized into a set of zones and each cell is enriched with the contact evolution laws or the homogenized coefficient. The advantage of the homogenization method is that the surface is always flat and does not require a refined mesh, which reduces the computation time. Periodic homogenization was chosen first to determine the mechanical properties of the composite, using different boundary conditions (PBC and KUBC). Thus, the correct value of the Young's modulus was found by a mesh convergence. Subsequently, a random distribution was developed based on the representative elementary volume study method. The two aspects were compared, and a good correlation of the results was found.
Indeed, and independently of the numerical approach used for the calculation of the VER, the different boundary conditions are supposed to lead to the same value of the homogenized property. Moreover, the reliability of the results is conditioned by the size of the VER and more precisely by the distribution of the heterogeneities in the matrix, and by their sizes for a specific volume fraction. For the numerical homogenization by periodic structure, a post-processing procedure by the FE method of a heterogeneous medium via the PYTHON programming has been developed. It aims at extracting the effective elastic properties of the studied biocomposites. As for the various analytical methods of calculation of the effective properties of the homogeneous material, by terminal or by estimation, they constitute asymptotic calculations in the sense that they are only applicable to volumes of material supposed to be very large.

Finally, and contrary to what one might think, many of the homogenization methods described above are commonly used in the study of composite materials. Some of them allow the determination of the effective properties of heterogeneous materials, by estimation, by scaling via bounds, or by numerical calculation. The use of these methods is effective at the center of the material and limited at the edges.

References
1. A. Elmoumen, "Prediction of heterogeneous material behavior based on numerical homogenization: modeling, visualization and morphological study", (2014)
2. T. Kanit, S. Forest, I. Galliet, V. Mounoury, D. Jeulin, Determination of the size of the representative volume element for random composites: Statistical and numerical approach" International Journal of Solids and Structures, (2003)
3. H. ESSABIR, "Bio-composites based on argan nutshell: Implementation, characterization and modeling of mechanical behavior", (2015)
4. F. EL Houdaigui, S. Forest, A. Gourgues, D. Jeulin, On the size of the representative volume element for isotropic elastic polycrystalline copper. IUTAM, (2007)
5. S. Lejeunes and S. Bourgeois. An abaqus toolbox for computing effective properties of heterogeneous media. In 10e colloque national en calcul des structures, (2011)
6. V.-H. Nhu. Numerical dialogues between tribological scales. PhD thesis, (2013)
7. T. Kanit, S. Forest, I. Galliet, V. Mounoury, D. Jeulin, Determination of the size of the representative volume element for random composites: Statistical and numerical approach. Int. J. Solids Struct. 40, 3647-3679, (2003)
8. Kanit, T., N’Guyen, F., Forest, S., Jeulin, D., Reed, M., Singleton, S., Apparent and effective physical properties of heterogeneous materials: Representativity of samples of two materials from food industry. Comput. Meth. Appl. Mech. Engng. 195, 3960–3982, (2006)
9. M H. Houben, M H. Guillaud, "Traité de construction en terre", CRATerre
10. R. Christensen [et al.], Solutions for effective shear properties in three phase sphere and cylinder models. J. Mech. Phys. Solids 27, 315-330. 33, 75, 175, (1979)
11. M. Jiang, I. Jasiuk and M. Ostoja-Straszewski, Apparent thermal conductivity of periodic two- dimensional composites, Computational Materials Science- 3: Vol. 25. -pp. 329-338, (2002)
12. M A A Cavalcante, H Khatam and M J Pindera, Homogenization of elastic-plastic periodic materials by FVDAM and FEM approaches - An assessment, Composites Part B- 6: Vol. 42. - pp. 1713-1730, (2011)
13. G. Chen [et al.] On the statistical determination of strength of random heterogeneous materials, Composite Structures - Vol. 149. - pp. 220-230, (2016)
14. T. Leidi [et al.] Computing effective properties of random heterogeneous materials on heterogeneous parallel processors, Computer Physics Communications- 11: Vol. 183. - pp. 2424-2344, (2012)
15. L. Bouaoune [et al.]: Random versus periodic microstructures for elasticity of fiber reinforced composites [Article] // Composites Part B - Vol. 103. - pp. 68-73, (2016)
16. A. El Moumen, "Effect of the interconnection of inclusions on the effective properties of a heterogeneous material", 21St French Congress of Mechanics, (2013)
17. Bledzki, A. K., Gassan, J., Composites reinforced with cellulose based fibers. Prog. Polym. Sci 24, 221-274, (1999)
18. O. Faruk [et al.], Biocomposites reinforced with natural fibers: 2000-2010. Prog. Polym. Sci 37, 1552-1596, (2012)
19. S. Mir [et al.], Improvement of physico-mechanical properties of coir-polypropylene bio composites by fiber chemical treatment. Materials and design 52, 251-257, (2013)