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Mechanical characterization and homogenization approach of a bio-composites based on PP reinforced with Nut-shells of Argan

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Abstract**:** *The morphology characterization and computational methods favored numerical simulation and design of microstructures. Indeed, the multiscale approaches enable us to determine the Young's modulus of materials. In this paper, the objective is to develop a two-dimensional microstructure of bio composites containing natural particles. The bio composite is made of polypropylene matrix mixed with natural fillers (Argan nut shells and bio-polymer polypropylene as matrix.) A modelization was made by the Zebulon calculation code on three different ranges, each contains four microstructures of different fractions of the inclusions 10%, 15%, 20% and 30%, and each microstructure contains 3 sizes of the inclusions (20% of small size, 60% of medium and 20% large). This numerical result is compared with experimental data.*

Keywords: *Argan nut shell particles, polymer composite, mechanical properties, computational homogenization, representativity*

I. Introduction

In recent decades composite materials based on polymer matrix have attracted great interests for both industrial applications and fundamental research to the high cost of the petroleum derived products, environmental hazard and to public concern for engy security [1] [2], a growing effort has emerged in the research of polymer composites reinforced with natural fillers from renewable natural resources (bio-fillers) instead of the synthetic fillers (carbon or glass). Composite materials occupy an increasingly large in multiple applications, starting with the automobile and transportation, aeronautics, radio, computers and furniture. These composites are typically made by combining two different materials both in shape by mechanical or chemical, to try to improve their performance. Both the composite components are the matrix and the reinforcements. In general, composite made of a bio-polymer [3], and a reinforcement of bio-fibers are often the most used in many areas of human activity materials. Their use is justified for their biodegradability, the opportunity to find everywhere in nature, whether in the plant world or in the animal world, they produce and recycle naturally on earth for millions of years (biodegradable materials and renewable by culture).

Numerical simulations have been often used to predict the mechanical behavior of bio composite microstructures. Indeed, the finite element method (FEM) can be used to analyze the macroscopic elastic properties and plays an important role for numerical homogenization of bio composites. This approach is used by Silva et al. [4] who compared between numerical and experimental analyses of bio composites fabricated with epoxy resin and unidirectional sisal and banana fibers. Behzad and Sain [5] focus on the use of finite element in micromechanics of polymer reinforced with natural hemp fibers. Globally, a correct accordance between numerical and experimental results is shown. Recently, an experimental characterization of a new biocomposite (PP/ASAN) made of polypropylene (PP) mixed with aggregates of shells of argan nut (ASAN) was achieved by Essabir et al. [6].

In this paper, the effective elastic properties of PP/ASAN are estimated from the available 2D images using FEM.. This bio composite the main objective of our investigation for digital representations, 2D visualizations, numerical computations and the representativeness of used samples. The finite element (FE) mesh associated with the 2D image was realized using multi-phase element techniques. In order to quantify the precision of the found numerical estimates. The obtained results by numerical homogenization technique are compared with experimental data and analytical approaches.

Material And Experimental Details

1. Material

The bio-composite used in this study is composed of a polymer matrix PP (of Exon Chemical products) with a density of 0.9 g / cm3 and a melting temperature of 165 ° C) and natural particles (nut-shells of Argan). **Fig. 1** schematic stages of the development of bio-composite based of the nut-shells of Argan with a polypropylene matrix

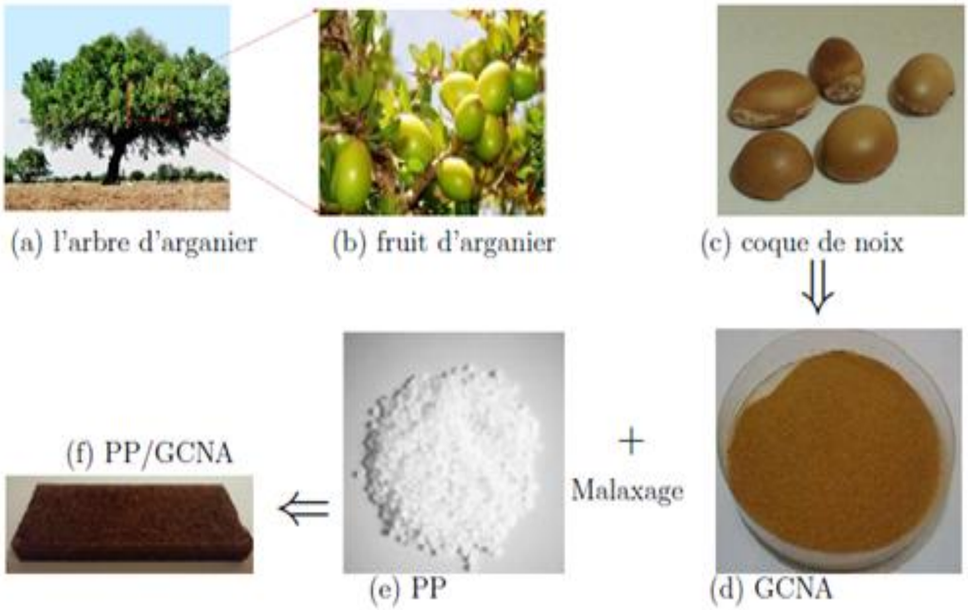

Fig.1. The materials and the overall process of making bio-composite.

II. Experimental procedure

2.1 Preparation of the granulometry of particles

Particle size analysis allowed us to separate the NA particles in three ranges size depending on the diameter (D) of the particles. Three series of samples (Polypropylene (PP) reinforced by NA particles)were prepared:

Series	
1	10, 15, 20 and 25 wt. % of NA particles belonging to the first range.
2	10., 15, 20 and 25 wt. % of NA particles belonging to the second range
3	10, 15, 20 and 25 wt. % of NA particles belonging to the third range.

2.2. Matrix preparation

The polypropylene was modified by adding a compatibilizer, 8 wt.% of a linear block copolymer based on styrene and butadiene. The resulting composites are denoted PP-D1152. The compatibilized PP was extruded in a twin screw extruder (LEISTRITZEXTRUSIONS TECHNIK GMBH, Germany),with the main screws rotating at 125 rpm while the side-feeding screw used for compatib ilizer was set at 40 rpm. The extruder barrel was heated with the following profile of 200, 200, 200, 200, 180, 180, 180, and 180C [7] from hopper to the die, respectively .

2.3 Specimen and test machine

Tensile tests are menus on flat test pieces according to ISO 527-3 [6] using a universal tensile testing machine (INSTRON 8821S, USA) at a 3mm pulling rate / min using a cell load of 5 kN without strain**.**

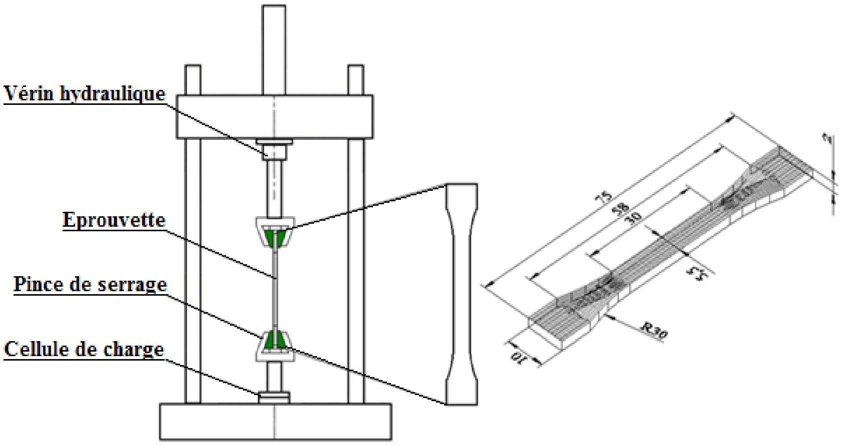

Fig. 2 Universal tensile testing machine and Specimen

To traverse this technique the following characteristics are determined: Young's modulus E(MPa), characterizing the rigidity of the material and according to the ISO standard, it is determined as the slope of the linear portion of the stress-strain curve and this in the deformation zone between 0.0025 and 0.005.

III. Test results

The test results show that the Young's modulus increases progressively with increasing concentration of NA to a maximum and then decreases. On the other hand, all bio composites have a Young's modulus greater than that of the pure PP (1034MPa), which shows that NA act as reinforcement. [2]

In this study it was observed that the size (diameter) of the particles significantly influences the mechanical strength properties of bio-composites. The Young's modulus decreases with increasing particle diameter. The results show that bio-based composite particles in Groups 1 and 2 reach their maximum Young's modulus at 20%, with a gain of 42.65% and 25.72, respectively, against the bio-based composites large particles (group 3) give a maximum value of 1129,1MPa 15%.Big particles (diameter) result in poor dispersion / distribution of particles through the reduction of the wettability between the particles and the matrix.

Poor wettability causes particle-particle interactions that give rise to particle agglomerations which increases the stress concentration around these agglomeration point [2.8]. On the other hand with increasing particle size of their surface area decreases, which leads to poor affinity between the particles and the matrix.

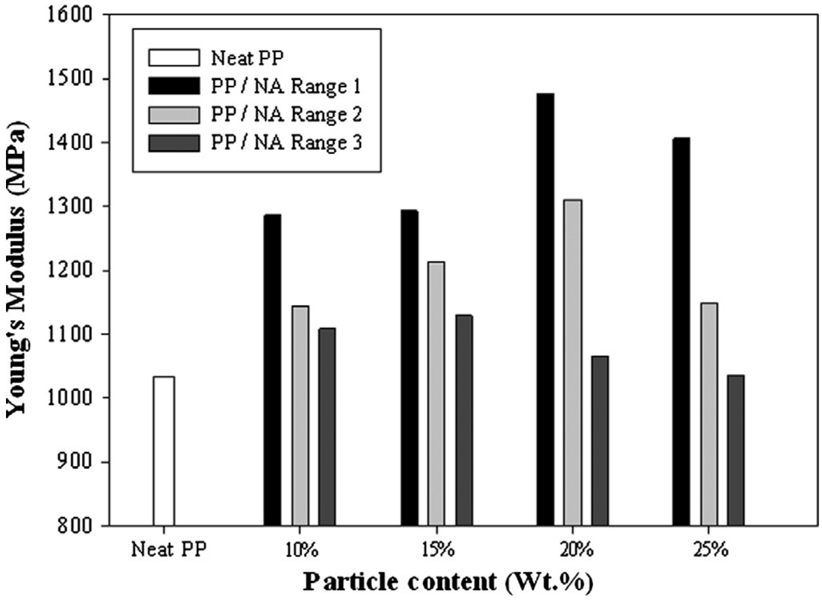

Fig.3 The Young's modulus of neat PP and PP/NA composites at various loading content

A linear increase in the Young's modulus is recorded by increasing the charge of the particles, up to a maximum value, then a drop in rigidity with the addition of particles.
For the 3rd series, the Young modulus reaches a maximum value to 15%, which shows that the Young's modulus E decreases with increasing particle size.

IV. Morphological analysis

Particle size has an important role in the mechanical properties of bio-composites. Particle morphology of Argan nut shells is evaluated by electron microscope (Fig.4).As shown in the figure, the particles have an irregular shape and a porous structure. The porous structure leads to an apparent density much lower (1.3g/cm3) than that of inorganic fillers (about 2.8 g / cm3) [6]. Therefore, with this morphology Nut-shells of Argan particles can be dispersed more uniformly in the polymer matrix.
Dentifying the three main constituents of the microstructure:

- The polymer matrix that is linear coating the particles NA
- The smallest constituents of bio-composite and the agglomeration phase produced by the association of several particles.
- Pores characterized by white spots.

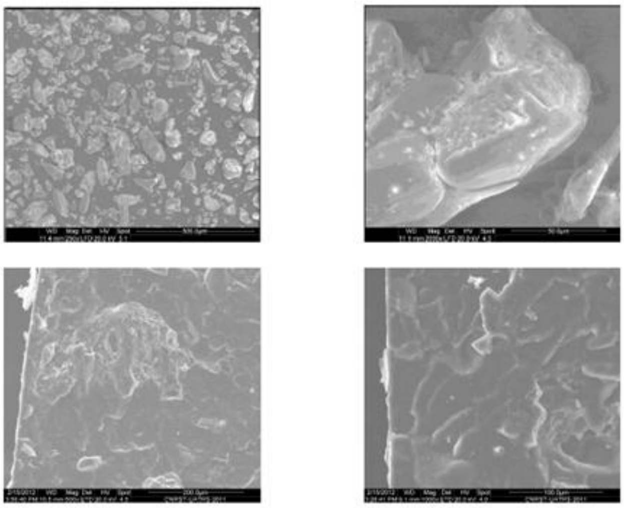

Fig.4. Snapshots of bio-composite PP / NA obtained by microscopic observation.

Numerical Homogenization

The objective of this part is to check first is that the size of the specimens in tension used to predict the behavior of our materials is broad enough to predict the overall mechanical behavior and also make a numerical modeling with multiscale finite element which will be developed using the Zebulon calculation code. In this part, we will assume two phase structure: PP matrix and inclusion NA without introducing the effect of porosity, assuming the elastic behavior.

Representative Volume Element (RVE)

Since no microscopic image is large to represent the entire bio-composite, the only way to quantify the size of the RVE is to work with the technical sub-volumes, developed in [9], by cutting the samples. The strategy used by Kanit et al. for the reals images of the ice cream was applied by ElMoumen [9] for the study of real images of PP / GNA. Indeed statistically RVE is defined by Willot Jeulin [10] and ElMoumen et al. [11] as the volume for which only one embodiment is sufficient to deduce a macroscopic behavior. The statistical method developed by ElMoumen [9] shows that the statistical RVE has an approximate value of V = 9.5 cm3, roughly a cube of length 2.2 cm. This statistical RVE is the minimum volume to have the representation of the results, is greater than that used by Essabir et al. [6] for the experimental study. The experimental samples have a volume of 1.6 cm3, roughly a cube of length 1.2 cm. To have a representative of the macroscopic behavior of our bio-composite and for calculating the macroscopic Young's modulus, the RVE proposed in this study is proposed well above the minimum static RVE.

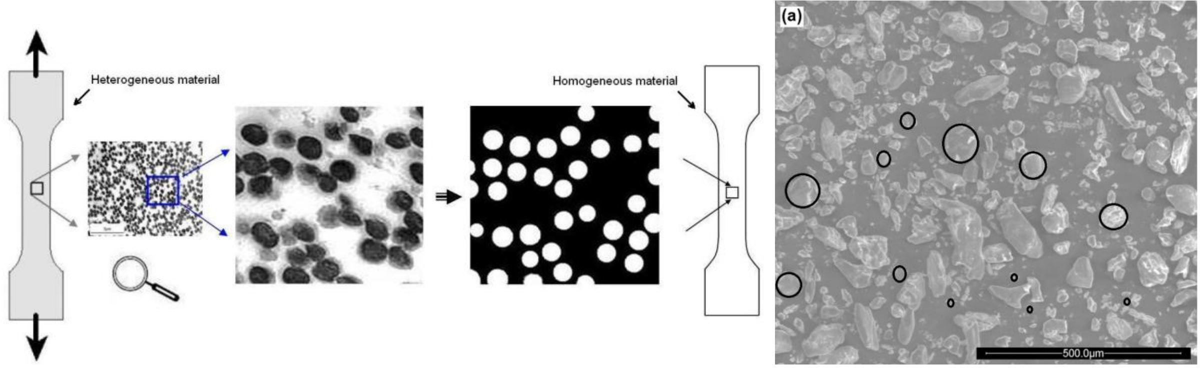

Fig.5 Concept of equivalent morphology and RVE

The microstructure generation

Traction tests were conducted on three bio-composite ranges. The microstructures were generated by the random Poisson process. This process is well suited to random and isotropic microstructures. The distribution of particles in the volume of the microstructure is generated in the following way: Mi spheres centers are generated randomly in space according to a Poisson process. For microstructures without inter particle connection, **a** distance of a repulsion to prevent contact.

Table 1: NA particles distribution

Range 1		**Range 2**		**Range3**	
Sizes(μm)	Loading (wt.%)	Sizes(μm)	Loading (wt.%)	Sizes(μm)	Loading (wt.%)
D<125	20	125<D<160	20	160<D<250	20
125<D<160	60	160<D<250	60	250<D<315	60
160<D<250	20	250<D<315	20	315<D<360	20

Table 2 illustrates the conversion of the weight fraction volume fraction using equation 1.

Table 2: Volume fractions for the three ranges

f_m		**10%**	**15%**	**20%**	**25%**
f_v		11,11	16,67	22,22	27,78
f_v**(Ranges)**	20%	2,222	3,334	4,444	5,556
	60%	6,666	10,002	13,332	16,668
	20%	2,222	3,334	4,444	5,556

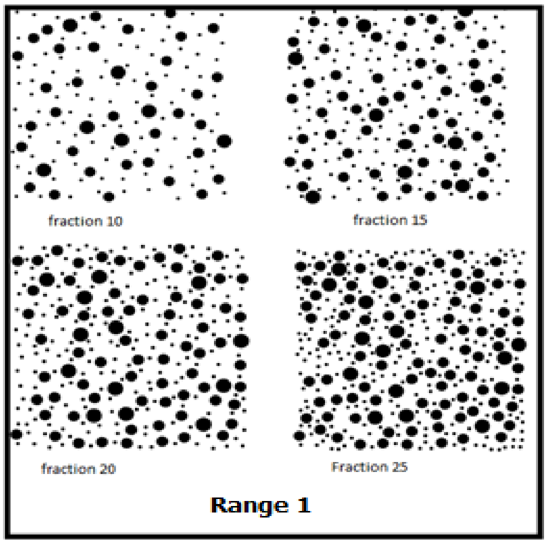

Fig.6. Example of Virtual images generated for different fractions for range1

Finite element meshing

The finite element (FE) mesh associated with the image of 2D microstructures is obtained using the so-called multi-phase element technique. This technique provide the convergence of elastic properties with a small number of FE. It is adopted for elastic behavior of composite materials, see Barbe et al. [12]. For illustration, an example of the obtained 2D meshes using this technique is presented in Fig.7.

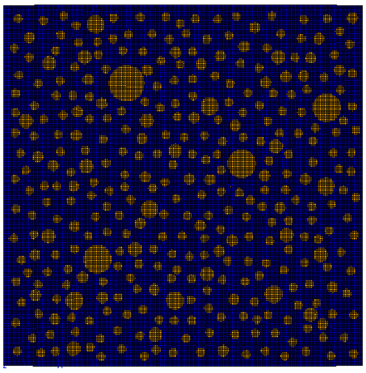

Fig.7 2D FE meshes of simulated microstructures

For numerical computation, the traction load is applied on one face. The opposite one is fixed and all the other faces are free of forces. For example, a tensile loading in one direction is described as follows:

$$\bar{u}\{face(x=0,y,z)=0\};\ \bar{w}\{face(x=0,y,z)=0\} \quad (1)$$

$$\bar{v}\{face(x=0,y,z)=0\};\ \bar{u}\{face(x=L,y,z)=d\} \quad (2)$$

where : $\bar{u}$; $\bar{v}$ and $\bar{w}$ are the applied displacements in the x, y and z-directions, L is the microstructure length and d is the imposed displacement.

A simple uniaxial tensile is applied on the real microstructure of PP/ASAN and the homogenized effective Young's modulus is determined

The elastic properties of the bio-composite of the constituents are shown in Table 4.

Table .4 : Elastic properties of PP-NA components.

PP-NA	E (MPa)	ν	K (MPa)	μ (MPa)
Matrix (PP)	1300	0,4	464,28	2166 ,67
Inclusions (NA)	8000	0,3	3076,92	6666,66

V. Numerical Homogenization Results And Discussions

The obtained numerical results for different particles volume fractions are presented in Tab. 5. These results are compared with analytical bounds and experimental data.. The numerical results were plotted in Fig. 8 then compared with results from other analytical approaches. The particle sizes have a minor effect on mechanical properties.

Table .5 : The results of the Young's modulus of bio composite

Fraction des inclusions (%)	**E(GPa)**		
	Range 1	Range 2	Range 3
10%	1,83495	1,8432	1,87302
15%	1,992	1,98848	2,0172
20%	2,17908	2,178	2,3029
25%	2,37875	2,373577	2,3054

Table .6. Analytical values of Young's modulus and Poisson coefficients PP-NA

Fraction of PP-CNA	**Reuss E**		**Voigt**		**Estimation ofGuth-Gold**
	E(GPa)	ν	E(GPa)	ν	$E^{GUTH-GOLD}$(GPa)
10%	1,970	0,39	1,419	0,387	1,808
15%	2,305	0,385	1,4867	0,381	2,200
20%	2,640	0,38	1,561	0,375	2,683
25%	2,975	0,375	1,644	0,369	3,258

The test results presented in the Fig.8 and 9 show that the Young's modulus increases progressively with increasing concentration of NA to a maximum and then decreases. On the other hand, all bio-composites have a Young's modulus greater than that of the pure PP (1,034GPa), which shows that NAC act as reinforcement. Comparing numerical results to those experimental analytical, they fall outside terminals HS, only the Young's modulus calculated by Voigt [13] is the closest experimental values. The divergence of numerical and experimental results can be explained by the made firstly that the experimental samples used by Essabir et al. (2013) [6], have a volume of 1.6 cm3, approximately a 1.2cm cube of length, much less than the minimum statistical RVE which is V = 9.5 cm3. On the other hand the poor wettability causes particle-particle interactions that give rise to particle agglomerations which increases the stress concentration around these agglomeration point.

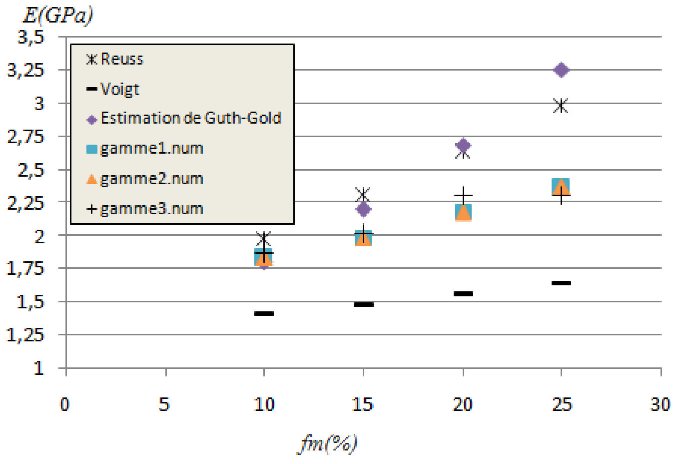

Fig.8. Numerical Young's modulus versus the volume fraction of the bio-composite PP-NA

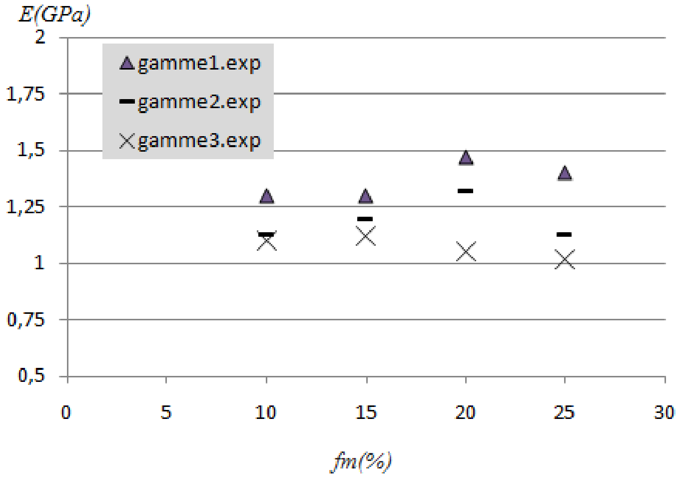

Fig.9.Variation of Young's Experimental modulus based on the volume fraction of the bio-composite PP-NA

Experimenters justify this behavior as the incompatibility load-matrix (interface problem). The presence of pores observed microscopically under an irregular shape lead to an apparent density much lower on the one hand but also favors the stress concentration around the porosity and may explain the structural rigidity drop. The figures show that the numerical results are bounded analytical results contrary to the experimental results, it is due to several factors that influence on the rigidity and quality of such materials, the porosity is considered detrimental to the composite material high performance. All authors agree to affirm that below a certain volume percentage of between 0.5% and 1% depending on the material studied, porosity has no influence on the behavior of the part if iso distributed. However, for higher void ratio, the mechanical properties of the part are significantly affected is the material becomes very sensitive to the presence of gaseous inclusions. This means that existing tools, which discretize a mesoscopic scale, do not take into account the influence of the manufacturing process on the constitution of the structure.

VI. Conclusion

This work is devoted to the study of the microstructure Two-dimensional (2D) of a bio-composite responsible for natural aggregates. The bio-composite is made of a polypropylene matrix (PP) reinforced with natural aggregates hull of argan nuts (GNA). The aggregates hull are used as a filler to improve the behavior of polymers. The results show that the concentration of particles and the particle size influences the morphological properties, structural and mechanical bio-composites. The particle size has a greater influence on the mechanical properties, because the bio-composite particles having the smaller diameter have the best mechanical properties. The test results show that the Young's modulus increases progressively with increasing concentration of NA to a maximum and then decreases. On the other hand, all bio-composites have a Young's modulus greater than that of the pure PP (1,034GPa), which shows that NA act as reinforcement. Comparing numerical results to those experimental analytical, they fall outside terminals HS, only the Young's modulus calculated by Voigt [3] is the closest experimental values. The divergence of numerical and experimental results can be explained by, firstly that the experimental samples used by Essabir et al. (2013) [2], have a volume of 1.6 cm3, approximately a 1.2cm cube of length, much less than the minimum statistical RVE which is V = 9.5 cm3. On the other hand the poor wettability causes particle-particle interactions that give rise to particle agglomerations which increases the stress concentration around these agglomeration point. The incompatibility load-matrix and the presence of pores observed by microscope can explain the structural rigidity drop. Homogenization in two step in this case can address this issue and taking into account the assumed variant porosity between 2-5% of the total volume fraction.

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