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Experiments on Mechanical Properties of Bio-Composite Produced from the Shell of Argan Nuts

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ABSTRACT

The argan tree (Argania Spinosa) grows mainly in Morocco. This forest species, also called iron tree, covers about 830000 ha of Moroccan territory. Virtues of this argan oil are multiple: uses for cooking, medicines, or cosmetics. The objective of this work is the valorization of the argan tree, and especially in its shell, which remains poorly exploited and which is sold by the premises at low prices for use as a fuel in the Moorish baths and bakeries. Argan shell is the reinforcement of this bio-composite. For this, the shell is ground, sieved and separated into five different diameters d. Then, the preparations of the three ranges, each of which is composed of proportions (20%, 60%, 20%) of increasing diameter of particles. Each particle range is used as reinforcement in a matrix of urea formaldehyde and water as a non-polluting solvent. Using a traction machine, the Young module and the breaking stress of the three biomaterial ranges were determined. Our results show that the Young module of first range (R1) remains superior to that of other two ranges and also has a high breaking voltage. Then the bending measurements were made by the three-point machine MTS EM Flexion. Our results show that the R1 has a greater bending stress than the other two ranges.

1. INTRODUCTION

The argan tree is considered a valuable resource in Morocco due to its various uses. The argan fruit is used to produce argan oil, which has numerous health and cosmetic benefits. The oil is also used in traditional Moroccan cuisine. In addition, the argan tree provides an important source of income for local communities, particularly women who are involved in the production of argan oil.

The protection of the argan tree and its ecosystem is crucial for sustainable development and the preservation of biodiversity. The argan biosphere reserve was established to protect the argan forest and its biodiversity, and to support the sustainable use of its resources by local communities.

Argania Spinosa (L.) Skeels, or iron trees, is a perennial, endemic and emblematic tree that grows in the center and south-Western Morocco with an area of more than 830000 ha of forest and about 20 million trees. It is estimated that the argan tree can live up to 250 years and produce fruit only at 8 years. For reference, the olive tree can do so from the age of 2 years.

The recognition of International Argan Day by the United Nations highlights the importance of the argan tree and its ecosystem, and raises awareness about the need to protect and preserve this valuable resource for future generations.

The Argan tree also plays a crucial role in the subsistence of local populations by providing firewood and woodwork materials as well as a source of food for animals, especially goats. Argan fruits are composed of two parts: the kernel, which represents around 3% of the fruit and is used for Argan oil production, and the rest, which is in the form of waste [1, 2].

In Morocco, annual waste production is estimated at 128,000 tons, but the Argan tree remains a valuable source for local populations due to its gastronomic, medicinal, and cosmetic benefits [3].

Composite materials offer new possibilities to industrial and design professionals, allowing them to combine functions, shapes, and materials in increasingly high-performing systems. These materials consist of a polymer matrix and reinforcing fibers, which is one of the goals of many researchers worldwide.

The world production of argan oil is 100% Moroccan. One of the parts can exploit in the argan is its shell which represents 80% of its weight.

As a potential biomaterial, the use of ANS as a biomaterial can have several benefits, including reducing waste in the production of argan oil, providing a sustainable and renewable source of material, and potentially offering a cost-effective alternative to traditional materials. Additionally, the mechanical properties of ANS, as demonstrated in our experiments, show potential for use in industries such as packaging and construction.

The development of innovative uses for argan products, including ANS, can contribute to the sustainable development of the region and the preservation of the argan tree ecosystem [4].

Additionally, Urea formaldehyde UF resins provide good mechanical and physical properties, such as high bonding strength, good dimensional stability, and resistance to moisture, heat, and chemicals. However, the use of UF resins has been criticized due to their formaldehyde emissions, which can have negative health effects. Therefore, efforts have been made to reduce formaldehyde emissions from UF resins, such as the use of alternative resins and additives.

UF resins, used in this work, are widely used adhesives in several fields, including applied plywood, particle board, medium density fibreboard, oriented chip boards and other artificial boards [5, 6] due to their low cost, moderate drying temperature and water solubility (clean solvent).

Several studies have been conducted on the use of urea-formaldehyde adhesives in composite materials production. The performance evaluation of these adhesives using mechanical tests was carried out in the study by Ferra et al [7]. The effects of particle acidity and the amount of hardener on the physical and mechanical properties of particleboard composites bonded with urea-formaldehyde were examined in the study by Akyüz et al [8]. Sajeeb et al. [9] evaluated the mechanical properties of natural fiber reinforced urea-formaldehyde melamine resin composites. To improve the performance of urea-formaldehyde adhesives, Duan et al. presented a new composite sealing material using a modified urea-formaldehyde resin with cement [10]. Lastly, polymer nanocomposites of wood impregnated with melamine-formaldehyde acrylamide and gum polymer were explored in the study by Hazarika et al [11]. These studies, conducted by different authors, highlight diverse approaches to improving the performance of urea-formaldehyde adhesives in composite materials production.

The development of new materials based on renewable natural resources is a rapidly growing field of research due to the increasing demand for sustainable and eco-friendly products. This includes the use of biomaterials, such as plant-based or animal-based materials, as well as biodegradable and compostable materials, to replace traditional materials made from non-renewable resources. The use of renewable resources not only reduces environmental impact but also contributes to the development of a circular economy, where waste is minimized and resources are reused and recycled [12, 13].

Several studies have been conducted on the physicochemical properties of argan nut shells in order to enhance and conserve biodiversity.

In a previous study by Essabir et al. [14], the thermal and mechanical properties of a polypropylene matrix $(C_3H_6)_n$ reinforced with ANS were investigated. The results showed that the addition of ANS to the polypropylene matrix reduced its thermal stability, and that the composite's decomposition temperature decreased as the particle size of ANS increased.

Another study [15] investigated the use of ANS particles in agriculture during summer as pyrolyzed biomass to enhance water retention and increase nutrient density in soil.

Recently, Babty et al. [16] characterized the swelling behavior of a biomaterial based on argan nut shells in the presence of water. The authors first sorted and sieved the ground ANS, then mixed the resulting powder with urea formaldehyde to prepare three biomaterials with different ranges of shell grain sizes. They also included two types of red and beech woods in the study. The biomaterials and woods were then immersed in water for 15 days, with mass measurements taken every 24 hours. The authors found that the swelling coefficient of the larger biomaterial size distribution was lower than that of the smaller distribution. However, the red wood and beech wood showed the highest coefficients.

Another study by the same authors (Babty et al.) [17] investigated the thermal conductivity of the three ranges of the same biomaterial. The experiment was conducted using a thermal conductivity analyzer (λ-Meter EP500e). The results indicated that ANS particles with glue had a thermal conductivity that decreased with an increase in size distribution, while argan powder without glue had a thermal conductivity that decreased with a decrease in size distribution.

Akhzuz et al. [18] suggested the use of compressed earth blocks (CEB) made from argan nut shells (ANS) as building materials in less developed countries, in order to enhance the durability of stabilized earth blocks against water damage. They conducted a study on the water absorption behavior and examined different cement proportions to evaluate the chemical stabilization effect on the new material.

Boujibar et al. [19] created an energy storage capacitor using an activated carbon electrode made from argan nut shells. The authors were able to achieve a high number of micropores and a sufficient number of mesopores, resulting in a specific surface area of 2251 m^2/g and a total pore volume of 1.04 cm^3/g. The activated carbon electrode also exhibited excellent ion conduction in the electrolytes.

Veigel et al. [20] investigated the use of an adhesive reinforced with nanocellulose in the production of particleboards and oriented strand boards. The study found that the exceptional mechanical properties of nanocellulose enhance the mechanical properties of these composites. A similar vein study conducted by Laaziz et al. [21] explores the production and properties of bio-composites based on polylactic acid and argan shell. The use of these two materials allows for the production of biodegradable and renewable composites, while reducing agricultural waste. These studies highlight the importance of using bio-based materials in the production of environmentally friendly composite materials.

We aim to conduct a mechanical analysis of ANS-based biomaterials that have been homogenized and bonded using formaldehyde in this study.

2. MATERIALS AND METHODS

2.1 Material development

The shell of argan nuts is crushed by a hammer crusher. which made it possible to obtain different sizes (s) of grains, then three ranges are prepared with its different sizes with percentages presented in Table 1.

Table 1. The three ranges are based on diameters and percentages

	Particle sizes (mm)				
Range	**0<d<0.5**	**0.5<d<1.0**	**1.0<d<1.7**	**1.7<d<2.5**	**2.5<d<3.7**
R1	20 %	60 %	20 %	-	-
R2	-	20 %	60 %	20 %	-
R3	-	-	20 %	60 %	20 %

Biomaterials are materials made from organic or inorganic substances that can be used in various industrial applications. In the case of this specific biomaterial, previous studies have determined the percentages of the different components that constitute it.

According to these studies, the biomaterial is composed of 50% argan shell powder, 25% urea-formaldehyde, and 25% water. Argan shell powder is a byproduct of argan oil production and is often considered waste. However, it has been found to be a useful resource for the production of biomaterials due to its mechanical properties and renewability. Urea-formaldehyde is a synthetic resin widely used in the production of biomaterials due to its ability to bind with natural materials and form strong polymers. Water is added to facilitate the formation of the resin and make it easier to work with.

These component percentages were determined as a result of extensive studies on the mechanical properties and characteristics of biomaterials, to ensure an optimal combination for the production of durable and high-performing biomaterials.

A powder-based adhesive of pre-catalyzed urea and formaldehyde resin, along with water as a non-toxic solvent, is utilized in this study. To prepare each range of ANS, the ANS is homogenized with the powder glue, gradually adding the percentage of water until achieving a homogeneous mixture. For the tensile study, the size mold is (200x80x4) mm^3, and for the bending study, another size mold (200x80x40) mm^3 is used for each range. The preparation process is conducted at room temperature and normal pressure. After preparation, the three ranges are left in the room for 72 hours to dry and ensure perfect cohesion of different components.

Figure 1. Biomaterial test according to ASTM D638-0 for bending tests

For bending tests, the test pieces dimension is (200x80x40) mm^3 (Figure 1).

2.2 Bending test

The bending tests of the test pieces of the three biomaterial ranges are carried out on MTS EM Flexion three-point machine.

The experiment involves placing the sample between two supports of the MTS EM bending machine at a distance of AB = 160 mm, and then applying a load F in the middle of the beam at AC = 80 mm, as shown in Figure 2.

Bending tests are used to determine the flexural strength and stiffness of a material. In this test, the material is subjected to a bending load, which causes it to deform. The amount of deformation is measured and used to calculate the flexural strength and stiffness of the material.

In a three-point bending test, the material is placed on two supports, with a load applied at the midpoint between them. This causes the material to bend, with the maximum bending stress occurring at the center of the specimen.

The MTS EM Flexion three-point machine is a commonly used machine for conducting bending tests. It is important to ensure that the test pieces are properly prepared and placed in the machine to ensure accurate and consistent results. It is also important to record and analyze the data carefully to determine the flexural strength and stiffness of the material.

The initial quasi-static tests involved applying a displacement of F load at a rate of 0.1 mm/s to determine the stress at which our composite biomaterial ruptured. We will now present the results for the three ranges of our biomaterials.

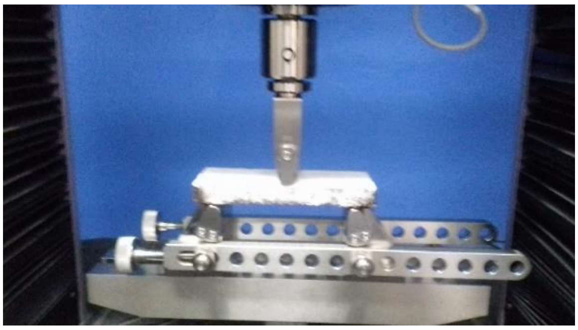

Figure 2. Position of a test piece in the MTS EM Three-point bending machine

2.3 Tensile test

For the tensile study of the three ranges, the test pieces are cut using a laser cutting machine according to the standards set by [ASTM D638-03]. The dimensions of these test pieces are (200x80x4) mm^3.

Figure 3. Biomaterial test according to ASTM D638-0 for tensile testing

The test pieces used are Argan Nut Shell Biomaterial Dumbbell Test Pieces (Figure 3), which are manufactured according to the ASTM D638-03 standard.

The "Zwick-Roell" machine is controlled by the "Test Expert" software, which allows for the results to be saved in a file. The mechanical tensile tests can be carried out by imposing either a constant effort or a constant displacement of the crosshead. During the stress and strain testing, the data numerical acquisition is recorded until the test piece is broken.

The tensile test measures the behavior of a material when subjected to an increasing load. The test piece is typically in the form of a dumbbell-shaped specimen, as described in ASTM D638-03, which is placed in the machine and pulled apart until it breaks (Figure 4). The force required to break the specimen is recorded, and from this data, the stress and strain of the material can be calculated.

Stress is the force acting on a material per unit area, while strain is the amount of deformation experienced by the material per unit length. The relationship between stress and strain is used to determine important mechanical properties of the material, such as its elastic modulus, yield strength, and ultimate tensile strength.

It is important to follow the ASTM D638-03 standard for the manufacture of the test pieces to ensure consistent and accurate results. The use of the "Zwick-Roell" machine and "Test Expert" software also helps to ensure accurate and reliable data collection.

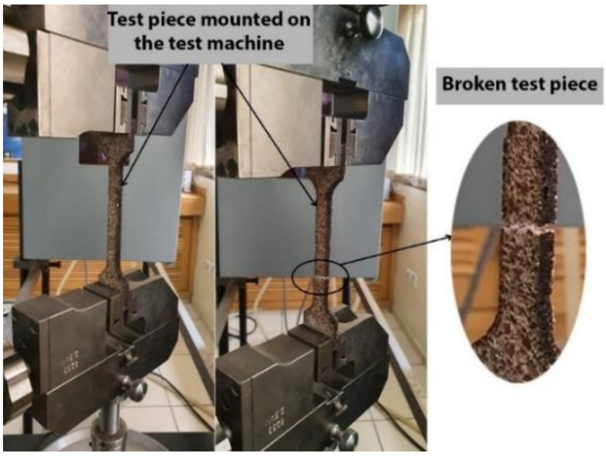

Figure 4. Mounting of tensile tests on dumbbell test pieces

3. RESULTS AND DISCUSSION

Data results are obtained directly from three samples.

Figure 5 shows the arrow evolution as an applied force function with a speed of 0.1 mm/s:

From the plot, it appears that the arrow is directly proportional to the applied force, indicating the linear behaviour of the material. This allows us to determine the Young module of the composite material, which is a measure of its stiffness. The Young module can be calculated as the slope of the linear portion of the curve in Figure 5.

Once the Young module is determined, the value of the breaking force of the material can be calculated. This is the force required to break the material. It is an important measure of material strength and is generally reported in units of force per unit area (N/mm^2).

It is important to ensure that the data obtained from the samples are accurate and representative of the material tested. Careful preparation and handling of samples, as well as proper calibration and operation of test equipment, can help ensure accurate and reliable results.

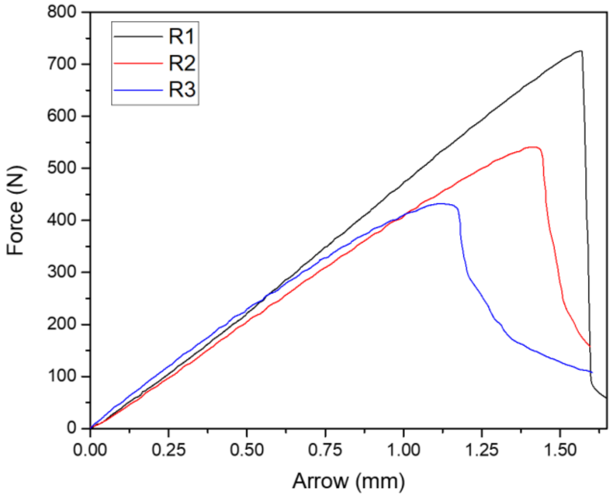

Figure 5. Arrow force - Range 1,2 and 3

Table 2. Characteristic of the three biomaterial ranges bending

Range	F$_{MAX}$ (N)	Arrow (mm)
R1	725.92	1.560
R2	541.62	1.350
R3	431.92	1.125

Table 2 shows that the three biomaterial ranges exhibited the same linear behavior before reaching their rupture stress, which enabled us to calculate their respective Young's modulus.

Furthermore, it was observed that R1 had a higher maximum breaking force than the second range (R2) and that its deflection was also greater than R2 and R3. These findings suggest that R1 is both stronger and stiffer than R2 and R3.

It was concluded that decreasing grain size can cause the material to become more fragile due to the presence of micro-cracks (internal defects) and macro-cracks (caused by manufacturing or design defects) which can make the material more prone to failure.

To draw accurate conclusions about the mechanical properties of biomaterials, it is crucial to analyze and interpret test data carefully. It is also important to consider the various factors that may impact the behavior and performance of the material to gain a better understanding of the test results.

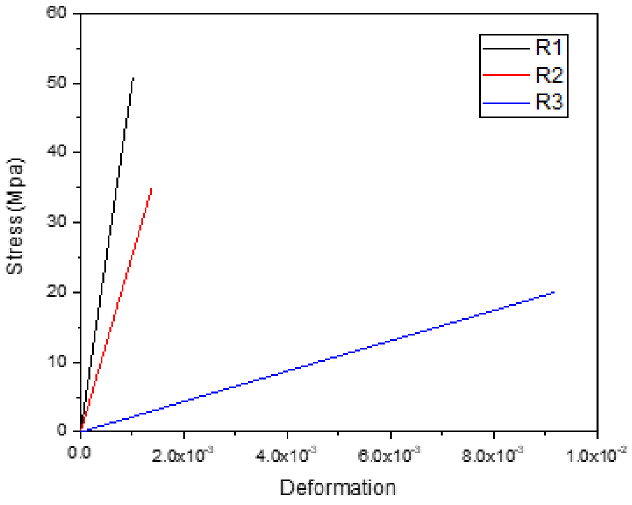

Figure 6. Stress-deformation - Range 1,2 and 3

Figure 6 displays the stress evolution as a function of the deformation of the biomaterials in the three ranges. We can observe that the deformation is proportional to the applied stress, following Hooke's law. It is important to note that while this law is strictly true only for ceramics, glass, most minerals, and the hardest metals, it also applies to the biomaterials being tested in this study.

Based on our findings, we can conclude that the third range biomaterial is weaker and more prone to fracture compared to the second and first ranges. Additionally, the slope of the stress-strain curve for the third range (R3) is smaller than that of the second range (R2) and the first range, indicating that it is less stiff and more deformable. This information can be useful in understanding the mechanical properties and behavior of the biomaterials and can guide further research and development in this field.

Table 3 presents the compiled values of Young's modulus, deformation, and stress at the point of fracture for the three ranges.

Table 3. Mechanical properties of the three ranges

Range	Young E Module (GPa)	Deformation ε_r	strain σ_r (MPa)
R1	50	1.016×10^{-3}	51
R2	25	1.380×10^{-3}	35
R3	21	9.190×10^{-3}	20

Upon analyzing the data, it can be concluded that the first range exhibits a higher Young's modulus and fracture stress in comparison to R2 and R3, whereas the deformation of the first range is significantly lower than that of R2 and R3. This can be attributed to the higher compactness of the first range biomaterial.

4. CONCLUSIONS

This study focused on investigating the mechanical properties of a new biomaterial derived from argan nut shells. To achieve this, we prepared samples of the biomaterial that met ASTM D638-03 experimental standards and conducted bending experiments using the MTS EM Three-point bending machine. Our results showed a linear relationship between applied stress and deformation for all three ranges until the point of rupture. The breaking stress of R1 was found to be greater than that of R2 and R3. We also observed that larger grain size corresponded to lower bending resistance, which we attribute to the compactness of the biomaterial ranges and the presence of glue.

The test specimens were prepared and cut according to the [ASTM D638-03] standard for the tensile strength analysis. Traction experiments were performed on the three ranges of the biomaterial using the Zwick-Roell machine. The stress-deformation data obtained from the tests were used to calculate the Young's modulus and stress at rupture for each of the three biomaterial ranges. Our results indicated that the Young's modulus of the first range was higher than the other two ranges, but the deformation εr of the first range remained very low compared to that of the second and third ranges. This observation is attributed to the compactness of the biomaterial ranges and the presence of glue in our biomaterial.

Indeed, the mechanical properties of this new biomaterial based on argan nut shells make it a promising material for various industrial applications. In addition to packaging and construction, it could also be used in the automotive industry, furniture manufacturing, and even in biomedical applications such as prosthetic implants. Further research can also explore its thermal and acoustic properties, as well as its environmental sustainability and potential for large-scale production. This could pave the way for the development of more sustainable and eco-friendly materials for various industries.

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NOMENCLATURE

ANS	Argan Nuts Shell
R1	Range 1
R2	Range 2
R3	Range 3
E	Young Module (GPa)
CEB	Compressed Earth Block

Greek symbols

ε	Deformation
l	Thermal conductivity, W.m^{-1}. K^{-1}
σ	Strain (MPa)