

Article

Mechanical Performance of 3D-Printed Cornstarch–Sandstone Sustainable Material

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Abstract: The objective of this research is the improvement of the physical properties of artificial sandstone in order to obtain a printable construction material whose composition and structure is suitable for the design, study and construction of environmentally friendly architectural structures. To investigate the suitability of the researched material for 3D printing and determine the physical and mechanical properties of the starch-based sandstone 3D-printed material, both cylindrical and cellular samples were designed. The 3D-printed artificial starch–sandstone material was found to have satisfactory mechanical properties.

Keywords: 3D printing; starch-based sandstone; mechanical properties; bio composite structure material; hierarchical honeycomb structures; finite element analysis



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1. Introduction

Three-dimensional printing is a method of manufacturing structures of objects by successively adding layers of material using a computer-aided design (CAD) model. The CAD is used to create a digital database for the construction. This method is often referred to as additive manufacturing, manufacturing of free solid form (Solid Freeform Fabrication) and rapid prototyping and construction with successive layers of the material (Layer-Based Manufacturing) [1,2]. Three-dimensional printing is seen as a revolutionizing technology for the development and manufacture of innovative products. The 3D printing of cement-based and clay-based mortars has had a significant effect on many applications in the construction and architecture industries [3,4]. As building materials acquire progressively more flexible characteristics, architectural design is expected to enable the change of building processes through the production of geometrically complex structures using the technology of 3D printing. The supremacy of 3D printing technology in relation to traditional construction techniques lies in the development of new manufacturing methods and the use of new innovative materials.

Although during the last three decades there has been considerable research on the 3D printing of polymer and metal materials, research on the technology of prosthetic construction with fluid building materials is still rather limited [4,5]. There are significant technical and scientific challenges related to the invention of fluid construction materials suitable for 3D printing, such as the composition of the raw material and the material temperature control during and after the 3D printing process [3,6]. In addition, as has already been pointed out in previous studies, the reliability of some materials (such as natural clay and concrete) is significantly reduced due to defects that arise during the 3D printing process. Usually, due to their low strength, such materials are mixed with chemical additives, which improve their physical and mechanical properties [7,8]. The

geometrical structures printed with the 3D printing technique are generally suitable for several structural applications, such as decorative structural elements and temporary light structures, provided that innovative materials with improved properties are used [9].

The technology of 3D printing is also gradually attracting investment in the building sector. The number of research projects and scientific publications on the utilization of 3D printing technology in the construction industry has increased dramatically since 2013, highlighting the role of building-scale 3D printing technology as a key driver of economic development [5,10–15]. Until now, interest in 3D printing has focused mainly on conventional concrete structures. It was observed that, compared to traditional construction methods, the application of 3D printing technology in concrete-based construction activities has several advantages, such as cost reduction and construction speed increase. This leaves room for the more efficient use of human and technological potential. In addition, with 3D printing technology, conventional molds (forms) are not used to cast the concrete, resulting in the liberation of architectural design from the limitations of the formwork and in the decreased consumption of natural resources and energy [16]. Judging from the aforementioned advantages, 3D printing technology can solve a number of problems that traditional construction faces, such as the time and cost required to realize a project and the environmental impact (pollution, waste of non-renewable natural and energy resources, etc.) [17]. In the last decade, research into the technology of 3D-printed concrete (3DPC) with the applications of robotics, artificial intelligence, and automatic control has contributed decisively to the upward trend of building and construction activities.

With the application of 3D printer technology in the near future, it will become possible to collectively manage and address the housing problem created by migration flows and extreme weather events, on a global scale. Starch and cellulose are low-cost and environmentally friendly renewable natural materials. Thanks to their advantages, recyclable natural building materials have the potential to gradually replace the conventional (cement-based) ones. This is of utmost importance in order to effectively face the impact of climate change and global warming [18–25].

In the present study, a combination of the aforementioned procedures was performed for the preparation of polymer cylinder specimens and cellular hierarchical structures using an artificial sandstone structure. The latter was formed by incorporating particles of sand and cellulose into the polymer matrix of the gelatin starch and the films which are based on plasticized native corn starch [18–20,24,25]. The three-dimensional printing of the artificial sandstone samples was made with the Liquid Deposition Modeling (LDM) method by extruding a structural viscous fluid with high viscosity. The material used to print the shell samples was a paste which consisted of sand, cellulose, gelatinized starch and water [21–23]. In this research, an innovative and environmentally friendly building material was developed and manufactured based on sand and non-edible corn starch and cellulose, which can also be obtained from alternative forms of waste management and processing. Additionally, common sand is found in abundance in the natural environment and does not require energy consumption, in contrast to the production of aggregates derived from rock mining and crushing, which contribute to deforestation and climate change. By using cellular hierarchical structures, either in functional components or in structural structures, increased robustness and resistance to external loads can be achieved. In this way, components or structures can be manufactured with significantly reduced weight, while maintaining their robustness and strength. In recent years, with the development of the construction industry, the additive method of construction has begun to be used for the construction of building structures. The combination of the use of the sandstone material and the cellular geometry could lead to the construction of robust yet complex, low-weight, environmentally friendly architectural structures.

2. Materials and Methods

2.1. Materials

For the development of a starch-based 3D printable paste, commercial corn starch of approximately 23% amylose and 12 wt% moisture content was used. The levels of moisture were determined using the method of drying the starch of a constant mass [26–28]. Corn starch powder was provided by Nestle Hellas S.A, Athens, Greece, and was mixed with commercial quartz sand provided by Kourasanit (Thessaloniki, Greece). Additionally, cellulose was provided by Sigma Aldrich, St. Louis, MO, USA, in order to be mixed in the polymer matrix of gelatinized starch.

2.2. Methods

2.2.1. Preparation of Starch–Sand-Based Paste

For the development of a structural material suitable for 3D printers, the starch was gelatinized before the printing process, in order to bind the (critical) amount of water required to print the material samples. The calculation of the critical water quantity considers the water needed to prepare the artificial sandstone specimens, as well as the water expelled from the fresh specimen material when the latter is forced by the piston to exit the nozzle during printing. The appropriate ratio of components of the fresh mixture of the studied material was obtained by experimental tests, with a constant ratio of starch: sand $\approx$ 1:5 [21–23] and with various contents of cellulose and water. To make the artificial sandstone samples suitable for printing on a 3D printer, water was added at a ratio of approximately 25 and 35 wt% in the dry mixture of starch, cellulose and sand. The starch grains were heated in water to a critical temperature range of 62–72 °C in order to swell and gelatinize, forming a thick paste. The fresh mixture, consisting of gelatinized starch, sand particles, cellulose and water, was heated to about 100 °C and mixed until fully homogenized. The final sample with the appropriate texture for 3D printing consisted of 33 wt% water.

Preliminary tests have shown that the optimal cellulose content for the most optimal mechanical properties was around 10% (on dry starch). Lower percentages of cellulose did not give the plasticity required for the successful 3D printing of the geometry, while higher percentages resulted in excessive fluidity of the printed paste of the material due to the high absorption of water by the hydrophilic cellulose and starch.

According to previous research [21–23], the starch was gelatinized before the printing process in order to bind the (critical) amount of water required to print the samples of the material. The drying of the samples was performed in the ambient conditions of the laboratory or in a microwave (not conventional) oven, with some modifications of the heating process. A flowchart of the manufacturing process is illustrated in Figure 1.

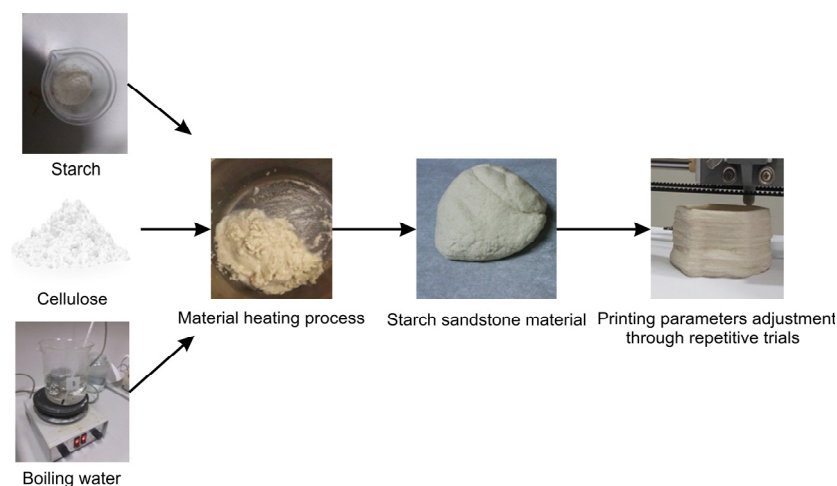


Figure 1. Flowchart of the manufacturing process of the current study.

2.2.2. Geometric Characteristics of Cellular Structures

Both cylindrical and cellular specimens were designed in Solidworks™ 2022 software. Cellular structures can be used as an infill pattern during the 3D printing process. Therefore, it is essential to investigate the mechanical properties of regular and first-hierarchy cellular structures. The first-order hierarchy design allows for the specimens to have a repeating pattern with a unit cell that can be easily replicated and studied. This facilitates the investigation of the mechanical behavior of the specimen under different loading conditions, and enables researchers to develop a better understanding of the structure–property relationships of the material. Additionally, the two levels of hierarchy were manufactured to assess the feasibility of 3D printing starch-based materials using complex designs as advanced constructions. The cellular structures designed and fabricated had a constant relative density $\rho = 0$. The length of the side of the base hexagon was $a = 40$ mm, and the thickness of the side of the hexagon was chosen to be $t = 8$ mm. The structural arrangement of the cellular hierarchical structures can be defined as the ratio of the length of the hexagon of the first hierarchy divided by the length of the base hexagon, $\gamma_1 = b/a$, as shown in Figure 2. The first hierarchy had $\gamma_1 = 0.3$ and $t = 5$ mm. As expected, the thickness of the side of the hexagon decreased when increasing the hierarchy in order to keep the overall relative density constant. After fabrication, the specimens had external dimensions of 150 mm high and 94 mm wide.

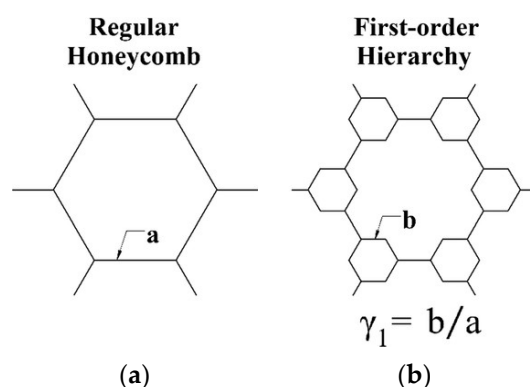


Figure 2. Cellular structures with (a) zero and (b) first hierarchy.

2.2.3. Specimen Fabrication

For the LDM fabrication of the specimen, the Z-Morph 2.0 SX printer with a thick paste extruder was used, as seen in Figure 3, while a Voxelizer 3 was used for the slicing procedure. At this point, it is worth noting that for this particular head the software does not accept the 3D file (e.g., stl file). Instead, a two-dimensional vector file (e.g., dxf, svg) must be imported, whose lines are the path the header will follow during printing. This peculiarity of the software requires that the entire path per layer be drawn manually. After the appropriate file has been designed, it is imported into the software and the print parameters are set. A 2 mm diameter nozzle was used to fabricate the specimens. In addition, the temperature of the printer bed was set at 85 °C in order to achieve partial drying of the specimens during printing. The printing parameters for the test specimens were set so that there was a smooth and continuous flow of material during printing. The 3D-printed specimens and structures are shown in Figure 4. Specifically, the number of layers was set to 13 with a height of 1 mm each, a path width of 8 mm and a printing speed of 10 mm/s. All specimens were printed at room temperature conditions.

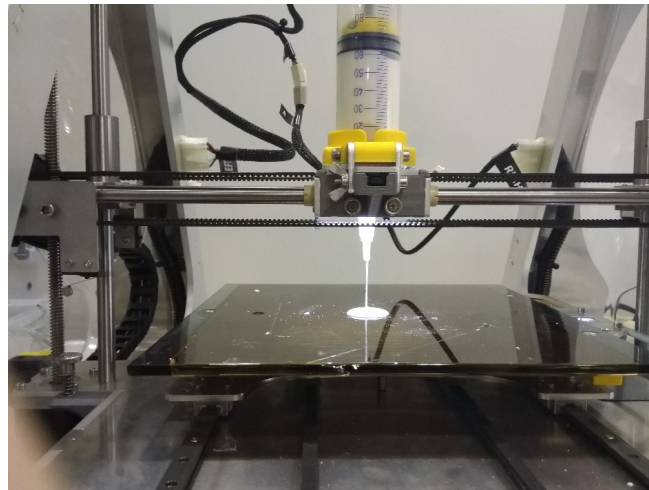
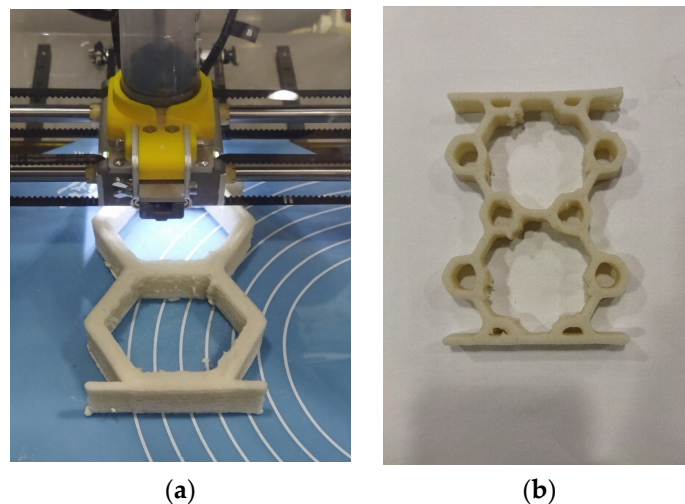


Figure 3. Three-dimensional printing of starch-based sandstone material through LDM process.



(a)

(b)

Figure 4. Three-dimensional printing process using the starch-based sandstone of (a) zero and (b) first level hierarchy.

2.2.4. Drying Process

After 3D printing the artificial sandstone samples, two drying methodologies were tested in order to find the experimental procedure that gives the best mechanical properties. For the reliability of the experiment, five cylindrical specimens with a diameter of 29 mm and a height of 12.5 mm were printed for each methodology (Figure 5). The samples of the first methodology were placed in the free air to dry for at least 72 h, in the ambient conditions of the laboratory at a temperature of about 26 °C. The drying of the second samples was undertaken by heating them in a microwave oven, 30% of the time in defrost mode and the rest of the time at full operating power [21,23]. The printed samples were stored under controlled conditions of temperature (≈ 23 °C) and relative humidity ($\approx 55\%$) for at least one week, in order to be equilibrated.

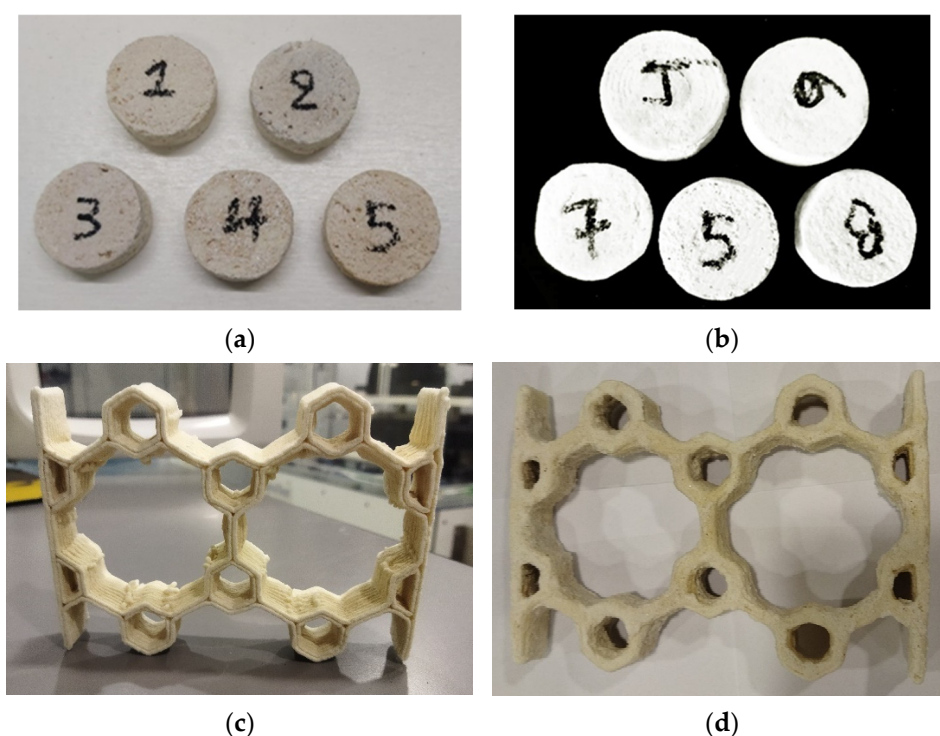


Figure 5. Cylinder specimens dried in (a) microwave oven and (b) air, and cellular specimens dried in (c) microwave oven and (d) air.

2.2.5. Mechanical Properties

The compression properties of the composite starch–sandstone material were determined using an M500-50AT (Testometric, Rochdale, UK) universal testing machine which was equipped with a 50 kN load of cell with a loading rate of 10 kN/min. In order to advance the calculation accuracy of the compression tests, a finite element analysis (FEA) was performed. After experimentation, there was a calculation of the load–displacement curves for the slow dried honeycomb specimen so as to directly be in comparison with the computer-generated data from the FEA. Ansys® Academic Research Mechanical, Release 2023 was used for the finite element analysis, and a static structural module was selected to simulate the static loading.

2.2.6. Morphology of the Samples

The morphology of the obtained starch–sand composite samples was examined with the scanning electron microscopy (SEM) technique using a JEOL JSM-840A, Tokyo, Japan, scanning electron microscope (SEM). Micrographs of the starch/sand-based specimens were obtained using an OLYMPUS SZX9 stereo microscope, operating in the optical mode of a 10× and 15× zoom range.

3. Results and Discussion

3.1. Mechanical Properties of Cylinder Specimens

The mechanical properties and, mainly, the compressive strength of the starch–sand-based material shown in Figure 6 depend on the degree of compaction, i.e., on the dry density of the material. If the artificial sandstone is prepared by casting the fresh material into molds, it is possible to achieve the maximum dry density of the material. This is achieved by compressing the material while pouring the fresh mixture into the molds, thereby increasing the degree of compaction and the compressive strength of the solid shell [21–23]. In the case of 3D printing, however, the deposition of the material is carried out freely by the printing extruder and the material is not compressed, since that would cause a change in the geometry of the object. Therefore, during the 3D printing process,

some gaps are created between the sand grains and air is trapped between the successive layers of the molten material. As a result, the strength of the 3D-printed samples is reduced.

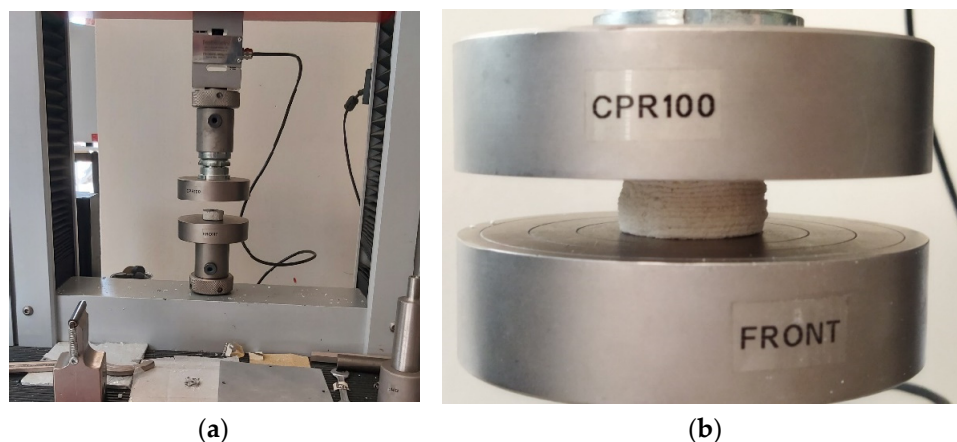


Figure 6. Starch-based, 3D-printed cylindrical specimen, (a) right before and (b) during compression test.

The experimental tests showed that the 3D-printed samples left in free air to dry had a higher compressive strength compared to the printed samples which were dried in the microwave oven, making the deposition of one layer of the printed material on top of the other quite successful. During the rapid drying process in the microwave oven, a large increase in the heating rate was caused with a corresponding increase in the pressure inside the material (the shell) due to the conversion of the trapped moisture into superheated steam. This apparently favors the formation of cracks within the material, resulting in air-dried, 3D-printed samples having (generally) higher compressive strength compared to microwave-dried printed samples. Figure 7 shows examples of the compressive stress–strain curves of the 3D-printed samples prepared with the two drying methods. The experimental data show that, with the air-drying methodology, the compressive strength of the samples is almost three times higher compared to the fast microwave drying methodology (9.6 MPa and 3.2 MPa, approximately).

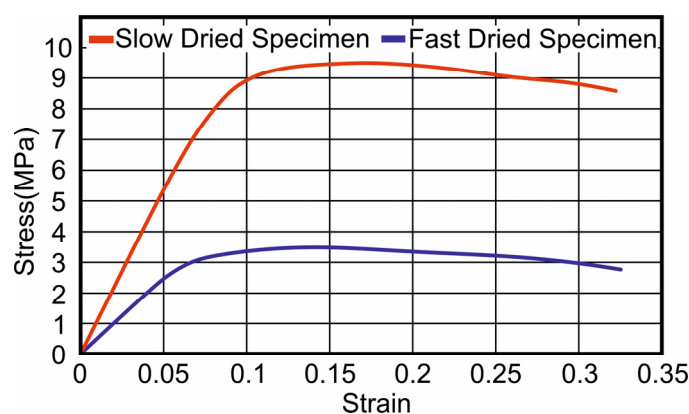


Figure 7. Typical stress–strain graphs from compression tests.

3.2. Mechanical Properties of Honeycomb Samples

Two different hierarchical honeycomb structures were 3D printed in order to study their mechanical performance: HC0, which represents the zero level of hierarchy (regular honeycomb), and HC1, which represents the first level of hierarchy. Figures 8 and 9 illustrate the typical load–displacement response of each hierarchical honeycomb structure under compressive loading. In Figure 8, the ultimate compression load for the honeycomb

specimens dried in air was measured to be above 150 N and 325 N for the regular honeycomb and the first order hierarchy, respectively. However, the ultimate compression load was reduced for the fast, microwave-dried specimens, as shown in Figure 9.

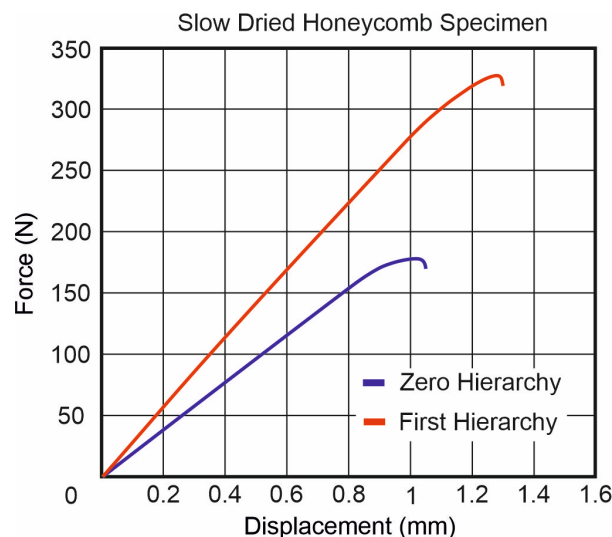


Figure 8. Force-displacement graphs from compression testing of zero and first level hierarchy slow-dried honeycomb structures.

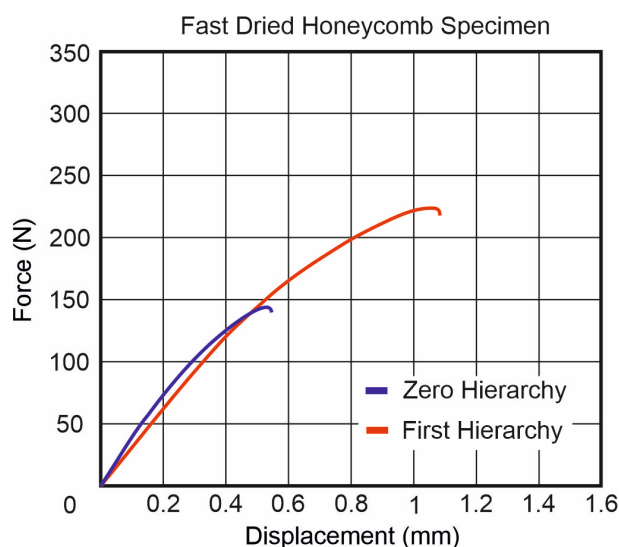


Figure 9. Force-displacement graphs from compression testing of zero and first level hierarchy fast-dried honeycomb structures.

FE analyses were performed for two levels of hierarchy of honeycomb structures. The results of the FE analysis, along with the stress–strain curves, are demonstrated in Figure 10. The change in strength between zero and first level was investigated in order to determine the stress–strain behavior of 3D-printed hierarchical honeycombs. This was accomplished by curve fitting the compressive experimental results with FEA generated data. A computational model was developed using the commercial code ANSYS, performing a static structural analysis. Assumptions of material values stress, strain and the moduli of the bilinear stress–strain curves of the honeycombs were used in the FE model. An imposed displacement was step-applied, and the reaction force was calculated at the bottom of each hierarchical honeycomb structure, at a fixed boundary condition. The input value of this displacement was acquired using the measured compression tests. Given these

deformation values, force data were calculated in FEA and contrasted with the measured forces. If the calculated force values did not fit with the experimental, then the initial values of stress, strain and moduli were approximated and the FE model was solved again. The mesh consisted of hexahedrals for the honeycomb and hierarchical structures, as well as for the upper and bottom plates. In order to ensure the mesh-independent response, a convergence study was performed. Based on the convergence results performed for an elastic response of the honeycomb structures, a minimum element size of 1.00 mm was considered to be adequate to obtain acceptable accuracy in the calculated responses. In Figure 10a,b, the change in terms of strength can be easily illustrated for the regular honeycomb compared to the first level of hierarchy. Regarding the elastic modulus, it was calculated by the initial slope at the elastic region. As shown in Figure 10c, it can be realized that the elastic modulus was not significantly affected by the increasing level in hierarchy. This can be attributed to the fact that the relative density was considered constant.

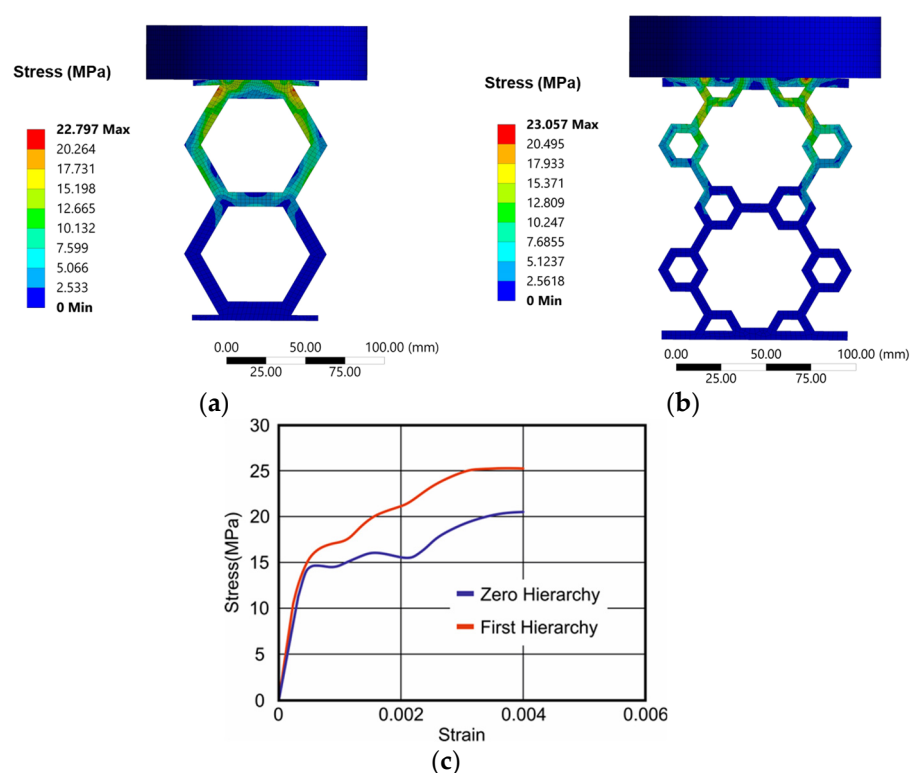


Figure 10. FE stress distribution of (a) HCO and (b) HC1 structures, and (c) comparison of the stress–strain curves between the two levels of hierarchy.

3.3. Morphology of the Samples

The main problems of the mechanical properties of 3DPC were the reduced strength of the interlayer and the lack of homogeneity of the material's physical properties in all directions (anisotropy). Reduced interlayer strength is a problem not only in printed concrete but also in cast concrete, which exhibits poor bonding strength when the material is cast in discrete layers [7,29,30]. In the case where the technology of the prosthetic construction of the object is applied, air is trapped between the successive layers of the extruded material and a weak bond is created between the different layers of the material. This contributes to the reduction in the strength of the 3DPC and to the degradation of its mechanical properties [31]. The strength of the inner bonds can be affected by various factors, such as the extrusion speed of the paste, the environmental conditions, the curing conditions of the material, etc. Various research efforts have been conducted to enhance the bonding strength between the different layers of 3DPC, by adding a thin layer of paste to bond the layers [32,33] or a thin layer of polymer [34]. However, due to the chemical

treatment of the material, the environmental impact is significant, without any substantial improvement in the mechanical properties of the concrete. The weak bond strength between the layers of 3DPC creates anisotropy of the mechanical properties of the material, which can lead to the degradation of concrete strength [7,28,35–39]. In contrast, the printed samples of artificial sandstone present, from visual inspection, a relatively solid structure with a small number of voids and cracks or other material failures compared to the printed samples of the 3DPC. The latter can also be seen from cross-sectional photos of samples in Figure 11 taken with the OLYMPUS SZX9 stereoscope, showing small distortions, cracks and voids.

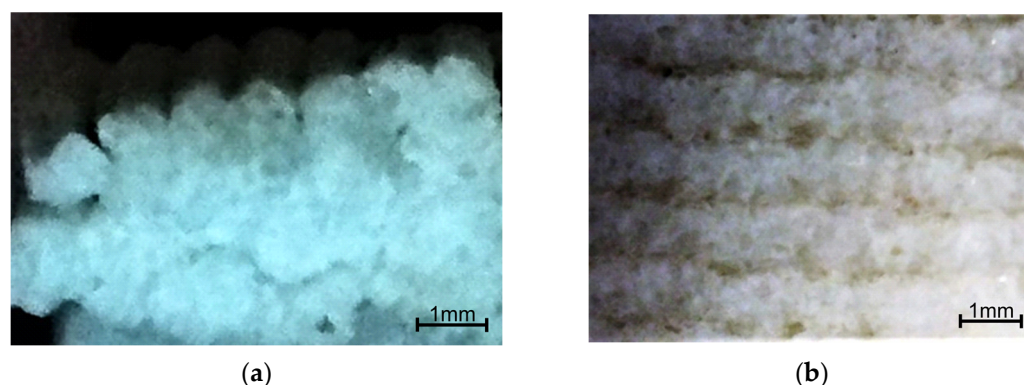


Figure 11. Snapshots of the successive layers of the 3D-printed geometry of the starch-based artificial sandstone paste. Stereoscopic photographs of the solid structure of the material in sections made on a cylindrical specimen: (a) LENS X10 and (b) LENS X15.

The morphology of the obtained starch–sand–cellulose 3D-printed samples was also examined using scanning electron microscopy (SEM). As it can be seen in Figure 12, where thermoplastic starch and cellulose surround sand grains, specimens are characterized by compact morphology. The SEM analysis clearly illustrated that the studied mixture of thermoplastic starch, cellulose, sand and water resulted in a solid, starch-based 3D-printed material with a certain amount of pores. This porous structural morphology of the 3D-printed specimens negatively affected the mechanical properties of the obtained material, in agreement with previous publications. The durability of the obtained 3D-printed geometries was significantly degraded under wet conditions. This factor is a major limitation in the studied material. Previous research has suggests various methods to reduce the hydrophilic morphology of similar materials (starch–sand-based materials), such as the use of several low-cost coatings such as waxes and oils in order to increase the hydrophobicity of this type of specimen [21–23].

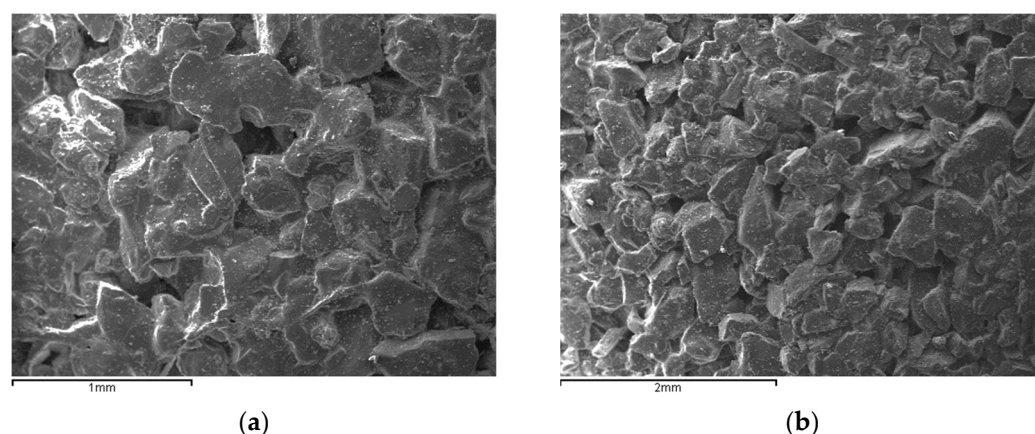


Figure 12. Structural characteristics and morphology of the samples with scanning electron microscopy, SEM: (a) 1 mm and (b) 2 mm.

3.4. Discussion

As research into the 3DPC technique is still at an early stage, there are still several problems that need to be addressed. The weak strength of the bonding of the successive layers of the material, when the technology of the additive manufacturing of the geometry is applied, can be observed during the process of 3D printing with concrete. To a lesser extent, the phenomenon of anisotropy was also observed during the 3D printing of artificial sandstone. Additionally, in cases of printing highly detailed 3D geometry, failure phenomena appeared during the drying of the material [40]. This is a serious problem that has been observed in both 3DPC and printed ceramics and is of great concern to researchers, since the increase in cracks due to the shrinkage of those materials during drying reduces the durability of the specimens [41].

The shrinkage (either plastic or dry) of the material can be attributed to the exposure of a large area of the freshly printed material to the environment, since no mold is used during the printing process. In general, the occurrence of plastic shrinkage is due to the rapid evaporation of water from the surface of the material, which renders the material susceptible to shrinkage. Thus, plastic cracks appear when the shrinkage stress is higher than the corresponding tensile stress, which is very low in this type of material. Normally, for more conventional methods, there are already many proposed solutions which effectively limit the shrinkage of the material and deal with the extensive cracks that appear during conventional concrete printing. One of the methods of dealing with these problems is to chemically treat the concrete and mix it with cellulose fibers [42]. However, the applicability of those experimental procedures in 3DPC is limited. As a result, careful steps should be taken in order to reinforce the artificial sandstone paste based on starch and cellulose, with the use of appropriate methods. Three-dimensional printing technology with the full automation of production is expected to contribute to the development and production of innovative commodities and to the improvement of all the phases of industry operation [5]. The 3D printing of construction materials such as concrete and cement-based and clay-based mortars is used in many applications related to the construction industry and architecture design [9,42–45]. With the development of 3D printing technology, environmentally friendly materials can be important building blocks for smart, energy-efficient and sustainable cities, as well as contributors to the mitigation of the adverse effects of climate change.

The main challenge of applying 3D printing technology in construction is the application of construction materials from conventional casting methodology to successful 3D printing. Due to the way the geometrical structures are made from successive layers of the material, the physical and mechanical properties of 3D-printed materials differ from their respective ones for conventional building. While concrete needs to be chemically treated before it can be used for 3D printing, fresh, artificial starch-based sandstone is a material suitable for immediate printing. However, additional laboratory tests as well as extensive theoretical and experimental research is needed to improve the composition and structure of artificial sandstone, so that it acquires the physical and mechanical properties of a durable building material. Nevertheless, the improved mechanical properties and the good adhesion between the printing layers make starch-based artificial sandstone a suitable material for printing environmentally friendly architectural structures, compared to 3D-printed concrete. For instance, Wolfs et al. [37,38] found that the strength of 3D-printed concrete was about 20–22 KPa. Later studies, such as Panda et al. and Ding et al. [17,36], found an improved strength of 3D-printed concrete of about 35–50 KPa. Efforts have been made to enhance the bond strength between the different layers and the very low values of the compressive strength of 3DPC or other similar materials suitable for 3D printing [45–48], such as by adding a thin layer of paste to bond the layers [32,33], or a thin layer of polymer [34]. However, due to the chemical treatment of the material, the burden on the quality of the environment is even greater. Similar phenomena have been observed in the case of the 3D printing of ceramic materials, which, in order to obtain their optimal mechanical properties, must be heated to very high temperatures [11].

4. Conclusions

Cellulose, starch and sand were used in the present research for the development of a starch-based sandstone material suitable for 3D printing. The study has shown that the 3D-printed artificial starch–sandstone material was found to have good mechanical properties. These findings can be easily attributed to the uniform morphology and 3D printing layer-to-layer adhesion shown by the starch sandstones. Additionally, the bonds between the successive layers of the printed material appeared stronger in the artificial starch–sandstones. The 3D-printed sandstone specimens showed a compact structure, without significant gaps, cracks or other material failures. Furthermore, the test results show that the compressive strength of the air-dried specimens was almost three times higher than that of microwave-oven-dried specimens. The improved mechanical properties and good adhesion between the printing layers make the artificial starch sandstone an innovative construction material suitable for 3D printing. However, further tests in the future should study the long-term reliability of such materials under cyclic loading regimes, while different applications need to be investigated and explored, such as 3D printed blocks and assemblies and small architectural structures.

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