Moisture-Responsive Cellulose For 4D Printing

Cellulose Réactive À L'humidité Pour L'impression 4D

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ABSTRACT. Particularly focusing on 4D printing, a technology enabling objects to transform over time. We explore smart materials, emphasizing moisture-responsive variants crucial for 4D printing. Notably, cellulose emerges as key, offering renewable and sustainable bio-based filaments. We detail the meticulous preparation of cellulose from sugarcane bagasse, obtaining high-purity fibers essential for 4D printing. These filaments exhibit versatile stiffness and moisture responsiveness, crucial for hygromorphic structures. Our proposed method integrates a codesign approach tailored for 4D printing, utilizing fused filament fabrication and cellulose-filled filaments. Through this investigation, we uncover cellulose's potential in sensor technology and additive manufacturing, marking significant progress in responsive materials and 4D printing.

RÉSUMÉ. L'accent est mis en particulier sur l'impression 4D, une technologie qui permet aux objets de se transformer au fil du temps. Nous explorons les matériaux intelligents, en mettant l'accent sur les variantes sensibles à l'humidité, cruciales pour l'impression 4D. La cellulose apparaît notamment comme un élément clé, car elle offre des filaments biosourcés renouvelables et durables. Nous détaillons la préparation méticuleuse de la cellulose à partir de la bagasse de canne à sucre, ce qui permet d'obtenir des fibres d'une grande pureté, essentielles pour l'impression 4D. Ces filaments présentent une rigidité polyvalente et une réactivité à l'humidité, essentielles pour les structures hygromorphes. La méthode que nous proposons intègre une approche de conception de codes adaptée à l'impression 4D, utilisant la fabrication par filament fusionné et des filaments remplis de cellulose. Grâce à cette étude, nous découvrons le potentiel de la cellulose dans la technologie des capteurs et la fabrication additive, ce qui marque un progrès significatif dans le domaine des matériaux réactifs et de l'impression 4D.

KEYWORDS. Additive manufacturing, 4D printing, smart materials, moisture-responsive and cellulose.

MOTS-CLÉS. Fabrication additive, impression 4D, matériaux intelligents, réactifs à l'humidité et cellulose.

1. Introduction

One of the most revolutionary technologies in the world of industry is known as additive manufacturing (AM). It's used for prototyping, as well as the manufacture of a wide range of objects that are of high quality at a cheap cost. AM technologies have been divided into seven families, which are as follows: Selective Laser Sintering (SLS), Direct Metal Laser Sintering (DLMS), Fused Deposition Modelling (FDM), material jetting, binder jetting, Directed Energy Deposition (DED) and sheet lamination. Fused deposition modeling (FDM), which is one of its numerous processing techniques, is one of the most well-known techniques because of its widespread usage, cheap cost, environment-friendly and extensive application. The use of 3D printing has drawn significant interest in its ability to manufacture operational components for a diverse array of things. These technologies have found applications in several sectors such as the medical field, food industry, electrical components, aerospace, and soft robots, among others.

At the MIT Conference, Professor Skylar Tibbits was the one who first used the phrase "4D printing." He gave the following definition of 4D printing: "Using 3-D printer in the creation of objects that could transform their form whilst they are eliminated from the 3-D printer." To put it another way, 4d printing is nothing more than the worked 3D printing technique combined with the use of phase change materials (PCMs). This PCMs react to external stimuli and may change shape over time. As a direct consequence of this, it goes through change in the characteristics of the particular materials, which ultimately gives birth to the existence of a fourth dimension known as time [ALN 22] [ANT 22] [TIB 14] [ABE 23] [SPI

22] [CHR 21]. In this paper, we discuss the various types of smart materials, with a particular focus on moisture-responsive smart materials used in 4D printing. Additionally, we explore the preparation of cellulose and provide the perspectives of this research.

2. Smart materials moisture-responsive use in 4D printing

In this section we well discover the most frequent 4D printing smart materials they are moisture-sensitive as well as the most compatible material for our experience. Moisture-responsive materials have gained significant attention in recent years due to their many potential applications in several fields such as biomedicine, soft robotics, and environmental monitoring, among others.

The table 1 comparing various moisture-responsive materials used in 4D printing, it is clear that different materials have different properties that make them more or less suited to certain uses. Cellulose is well suited for usage in biomaterials and textiles despite its delayed reaction time because to its superior mechanical characteristics, for all that reason it is the optimal choice for our experiment to make a humidity sensor.

3. Cellulose Bio-based filaments

In order to use additive manufacturing to create a humidity sensor, we should make a filament that has moisture responsiveness. According to literature reviews, cellulose-bio-based filaments are the most suitable. These innovative filaments combine cellulose powder with various plastics, resulting in materials with versatile stiffness and moisture responsiveness. We will use a codesign approach that has been developed specifically for 4D printing of hygromorphic structures using fused filament fabrication. We are integrating the development of biobased cellulose-filled filaments, which vary in stiffness and hygroresponsiveness, together with the strategic design of mesoscale structures within printed elements. Specifically, we detail the formulation of a filament palette that we will achieved by blending cellulose powder in mass ratios ranging of 0, 10 and 30% [ZIA 22].

No	Material	Advantages	Disadvantages	Applications
1	Cellulose	Renewable, biodegradable, low cost	High degree of crystallinity, limited water responsiveness	Packaging, textiles, sensors
2	Silk fibroin films	Biocompatible, biodegradable, high tensile strength	Limited water responsiveness, expensive	Biomedical applications, sensors
3	Smart polyurethane (PU)	Excellent mechanical properties, high water responsiveness	Non-renewable, potential toxicity concerns	Actuators, drug delivery systems
4	Polyethylene glycol diacrylate (PEGDA)	Highly tunable water responsiveness, simple modification process	Limited mechanical strength, potential toxicity concerns	Sensors, drug delivery systems
5	Poly (N-isopropylacrylamide- co-acrylic acid) (pNIPAM- AAc)	Excellent water responsiveness, reversible actuation	Limited mechanical strength, potential toxicity concerns	Microgrippers, actuators
6	Double cross-linked polymer	High water responsiveness, excellent mechanical stability	More complex synthesis process	Artificial muscles, soft robotics

Table 1. A survey of the most popular moisture responsive materials and their uses in a variety of applications [ALN 22].

4. Materials and Equipment

The fabrication of the cellulose-bio-based filaments requires a natural source of cellulose, like plants. There are a lot of plants that we can use for preparing the cellulose, like wood, grass, vegetables, etc. We selected sugarcane juice waste as our source for cellulose extraction owing to its high extraction efficiency, its potential for environmental preservation and organic waste reduction, and its widespread availability in Morocco. After the collection of the sugarcane juice waste to prepare the cellulose, we need the following materials and equipment:

- Sugarcane bagasse (30 g)
- Tap water
- Sodium hydroxide (NaOH) solution (4%)
- Sodium hypochlorite (NaClO) solution (4%)
- Deionized water
- Oven
- Crushing or milling equipment
- Containers for washing and soaking
- pH meter or pH strips
- Heating source for maintaining 65°C

5. Preparing Cellulose

The cellulose preparation process consists of the following six steps:

5.1. Collection and Initial Processing:

The bagasse underwent several processing steps to extract cellulose. Initially, we began by collecting sugarcane waste and crushing it into powdery particles with lengths of several millimeters. In this experience we will used 30 grams of sugarcane bagasse.

5.2. Washing:

After the collection and make the initial processing of sugarcane bagasse, we then washed the powdered bagasse using ordinary tap water at room temperature (21c°), until all soluble impurities are removed. After that we let the washed bagasse dry in the air.

5.3. Alkali Treatment:

This step helped break down lignin and hemicellulose components, facilitating cellulose extraction. Next, we prepare a 4% sodium hydroxide (NaOH) solution, add the dried sugarcane bagasse powder to it, and heat the mixture at 65°C for 3 hours to complete the alkali treatment.

5.4. Bleaching:

Following the alkali procedure, we utilized a 4% sodium hypochlorite (NaClO) solution to bleach the fibers for an added three hours. This process is vital in eliminating any residual lignin and escalating cellulose purity.

5.5. Neutralization and Washing:

To ensure complete cleansing, we extensively washed the bleached fibers with deionized water until neutral pH that is below 7 is achieved after bleaching them.

5.6. Drying:

We dried bleached fibers in an oven at an approximate temperature of 70°C for precisely twenty-four hours to eradicate moisture content. This process produces a stable bulk dry matter that is perfect for subsequent analysis and characterization. Figure 1 describes the steps in preparing cellulose from sugarcane waste.

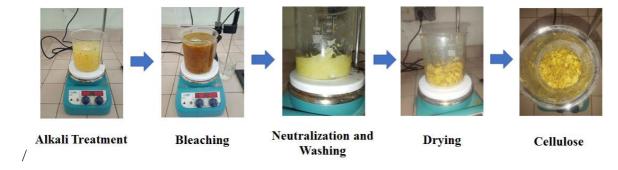


Figure 1. Steps for Preparing Cellulose.

6. Results

From extracting cellulose from sugarcane bagasse, we got 14.6805 grams of cellulose which corresponds to about 48.94% relative to the initial quantity of 30 grams used for bagasse. This substantial yield indicates a very efficient extraction process that optimizes use of resources and minimizes wastage.

The extracted cellulose underwent extensive characterization to evaluate its purity and composition. Analytical techniques showed it had a high degree of purity with minimal leftover lignin and hemicellulose. For applications at an advanced level, this high purity is necessary.

The positive findings from these investigations imply that such cellulose can be utilized to create a filament based upon it which may be highly useful in 4D printing. They are eco-friendly as they are renewable and biodegradable materials thus aligning well with the sustainability agenda for manufacturing responsible products. Their ability to adapt with humidity fluctuation makes them useful in developing innovative hygromorphic structures.

7. Conclusions

Cellulose derived filaments present for the 4D printing a great potential in terms of sustainability and adaptability. If we look carefully at our research, sugarcane bagasse cellulose has been successfully extracted and processed into moisture-responsive filaments that indicate its possible use as a renewable and biodegradable material in advanced manufacturing technologies.

Our next step in research will be to make improvements on this process in order to create humidity sensors and also explore other areas where it can be applied. This is a monumental advancement within the area of materials that respond to external stimuli and additive manufacturing, towards future innovations of smart and sustainable technologies.

References

[ALN 22] AL NAHARI, B. B. M. A., ZARBANE, K., & BEIDOURI, Z., « Smart Materials Moisture-Responsive Use in 4D Printing. In Casablanca International Conference on Additive Manufacturing », *Cham: Springer Nature Switzerland*, pp. 277-285, November 2022,

[ANT 22] ANTAR, I., OTHMANI, M., ZARBANE, K., EL OUMAMI, M., & BEIDOURI, Z., « Topology optimization of a 3D part virtually printed by FDM », *Journal of Achievements in Materials and Manufacturing Engineering*, 112(1), 2022.

[TIB 14] TIBBITS, S, « 4D printing: multi-material shape change», Architectural Design, 84(1), 116-121, 2014.

[ABE 23] ABEROUMAND M., SOLTANMOHAMMADI K., RAHMATABADI, D., SOLEYMAN E., GHASEMI I., BANIASSADI M. & BAGHANI M. « 4D printing of polyvinyl chloride (PVC): a detailed analysis of microstructure, programming, and shape memory performance », *Macromolecular Materials and Engineering*, 2200677, 2023.

[SPI 22] SPIEGEL C. A., HACKNER M., BOTHE V. P., SPATZ J. P., & BLASCO E. « 4D printing of shape memory polymers: from macro to micro », *Advanced Functional Materials*, 2110580, 2022.

[CHR 21] CHRYSANTHOS MARAVEAS, ILKER S. BAYER B, THOMAS BARTZANAS. « 4D printing: Perspectives for the production of sustainable plastics for agriculture », *Biotechnology Advances*, 2021.

[ZIA 22] ZIA U. A., MUHAMMAD YASIR KHALID, ALI ZOLFAGHARIAN, MAHDI BODAGHI., « 4D bioprinting of smart polymers for biomedical applications: recent progress, challenges, and future perspectives », *Reactive and Functional Polymers*, 105374, 2022.