

Mechanik, Werkstoffe und Konstruktion im Bauwesen | Band 73

Dunia Abdullah Agha

# 3D Paper Printing for the Built Environment

Optimization of the Material Behavior &  
Production Process to Reach Quality  
Integration and Dimensional Accuracy

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# **Mechanik, Werkstoffe und Konstruktion im Bauwesen**

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# 3D Paper Printing for the Built Environment

Optimization of the Material Behavior &  
Production Process to Reach Quality  
Integration and Dimensional Accuracy

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Institut für Statik und Konstruktion  
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Darmstadt, Germany

Vom Fachbereich 13 – Bau- und Umweltingenieurwissenschaften (Institut für Statik und Konstruktion) der Technischen Universität Darmstadt zur Erlangung des akademischen Grades eines Doktor-Ingenieurs (Dr.-Ing.) genehmigte Dissertation von Dunia Abdullah Agha aus Babel, Irak.

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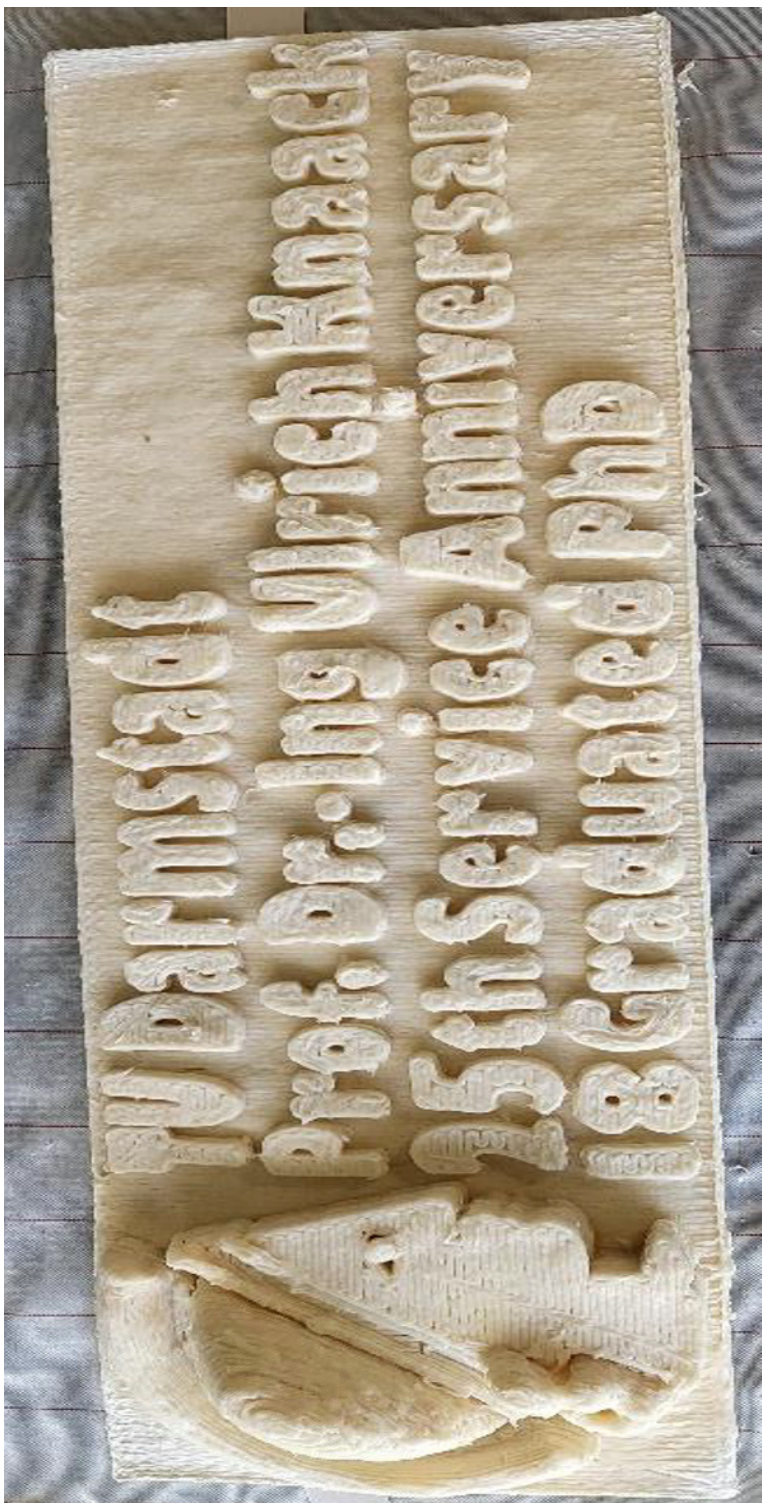
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Thank you.

Darmstadt, den April 2024

Dunia Abdullah Agha



3D printed paper model to congratulate my professor for his 25<sup>th</sup> service anniversary in 2022.

# **Dedication**

*Dedicated to my darlings - Ali and Fahad*



# Abstract

This research aims to produce and develop products from a new material formulation as bio-based paper material by using additive manufacturing (AM), in precise 3D printing. The approach of producing and developing these products is by optimizing material behavior and production process to reach a high level of quality and accuracy. Potential applications for the built environment of facade engineering are presented which fits the material properties and benefits the most from the complexity provided by the additive manufacturing technology.

The formulation consists of four components with water of ranges (79-68.2%) such as: cellulose of used range (12-16%), carboxymethylcellulose (CMC) of (2.4-6.2%), lecithin of (2.4-4.7%) and a filler of (2.23-15.6%) (such as chalk or undissolved starch or mgso<sub>4</sub>). Each ingredient has a specific function in the mix. Cellulose is the main structure for the mix. CMC is a hydrogel that provides the adhesive properties of the mix. Lecithin is a fatty substance that improves the extrudability of the compound. Filler increases the number of interparticle contacts and stabilizes the hydrogel, which means improved buildability. The used cellulose is native cellulose fiber (bleached pulp) from different sources (NBHK/NBSK) and with fiber length from 0.2 to 2.1 mm.

The 3D printer used is LUTUM -VormVrij® 3D clay printer version 2.1. The overall printing parameters are air pressure up to (0.3 - 0.85) Mpa, resolution (nozzle diameter of 0.6mm -1.2mm and 1.6mm), and printing speed in the range of (15 – 20) mm/s.

The methodology of the research is to try many mixture formulations to investigate the possible selection of homogeneous pastes with strong potential for AM by extrusion and to achieve buildability (higher height possible while maintaining shape retention and stability). The material would be able to support its own weight and have limited deformation of the printed part during room environmental drying. This was done by focusing on the adjustment and optimization controlled by rheological characterization and printing parameters settings to ensure optimal shape accuracy of 3D printed parts. A printing adjustment guideline and design constraints adapted to the developed paste were proposed.

For nozzle 1.6mm, the best formulation is Cellulose 15%, Aspen type, Water 69.2%, CMC 7.5%, Lecithin 2.8%, and the filler used is Starch of 5.6%.

For nozzle 1.2 mm, the best formulation is using cellulose 15.5%, aspen type, water 71.5%, CMC of 5.8%, lecithin 2.9%, filler used is starch of 4.3%. The nozzle 0.6mm, the best formulation is using cellulose 13.4%, aspen, water 78%, CMC 3.6%, lecithin 2.6%, and filler used is chalk of 2.3%.

The summery results lead to 3D paper printing of products with complex geometries with mechanical properties and a range of capabilities, such as

1. Safety aspect due to use of non-hazardous material,
2. 100% recyclable.
3. Large complex component.
4. Complex part design.
5. Self-supporting material that can carry and hold its own weight while maintaining shape and stability.
6. The buildability of height reached approximately 200 mm without buckling problems or collapsing during the drying process.
7. The wet density has a range of (7.27 - 8.08) kN/m<sup>3</sup> and the dry density is (4.02- 5.67) kN/m<sup>3</sup>.
8. Printing parameters: Air pressure up to 0.3 – 0.85 Mpa
9. Tensile strength up to 4.5 Mpa.
10. Adhesive strength reaches up to 20% of tensile strength.
11. Young's modulus reaches about 6 GPa.
12. Economic aspect of the material and the 1 kg of the material in the range of 10 €/kg.
13. Optimization of the material to achieve the lowest possible shrinkage of the extruded filament, it reached 17.4%.
14. Dimensional accuracy of a 3D printed part after drying achieved 14.8%.

To achieve this goal, this work focused on

- Paste formulations based on short and long cellulose fibers, CMC, lecithin, and filler (indissoluble potato starch, chalk, or Mgso<sub>4</sub>).
- Room drying.
- Use of 3D printer (LUTUM -VormVrij® 3D clay printer version 2.1).

Keywords: 3D printing, paper, cellulose fibers, bio-based material, 3D paper structure.

# Zusammenfassung

Das Ziel dieser Forschung ist, Produkte aus einer neuen Materialformulierung als biobasiertes Papiermaterial mittels additiver Fertigung (AM) im präzisen 3D-Druck herzustellen und zu entwickeln. Der Herstellungs- und Entwicklungsprozess dieser Produkte erfolgt durch Optimierung des Materialverhaltens und des Produktionsprozesses, um eine hohe Qualität und Genauigkeit zu erreichen. Mögliche Anwendungen für die gebaute Umwelt im Bereich der Fassadentechnik werden vorgestellt. Diese sind auf die Materialeigenschaften abgestimmt und profitieren am meisten von der Komplexität, die die additive Fertigungstechnologie bietet.

Die Formulierung besteht aus vier Komponenten mit Wasser wie Cellulose, Carboxymethylcellulose (CMC), Lecithin und einem Füllstoff (wie Kreide oder ungelöste Stärke oder  $\text{mgso}_4$ ). Jeder Inhaltsstoff hat eine bestimmte Funktion in der Mischung. Cellulose ist die Hauptstruktur der Mischung. CMC ist ein Hydrogel, das für die Klebeeigenschaften der Mischung sorgt. Lecithin ist ein Fettstoff, der die Extrudierbarkeit der Mischung verbessert. Füllstoff erhöht die Anzahl der Kontakte zwischen den Partikeln und stabilisiert das Hydrogel, was zu einer besseren Verarbeitbarkeit führt. Bei der verwendeten Zellulose handelt es sich um native Zellulosefasern (gebleichter Zellstoff) aus verschiedenen Quellen (NBHK/NBSK) und mit einer Faserlänge von 0,2 bis 2,1 mm.

Als 3D-Drucker wird der LUTUM -VormVrij® 3D-Clay-Drucker Version 2.1 verwendet, wobei die allgemeinen Druckparameter Luftdruck bis zu (0,3 - 0,85) Mpa, Auflösung (Düsendurchmesser von 0,6 mm -1,2 mm und 1,6 mm) und Druckgeschwindigkeit im Bereich von (15 - 20) mm/s sind.

Ziel der Untersuchung ist es, verschiedene Mischungsformulierungen zu testen, um eine mögliche Auswahl homogener Pasten mit hohem Potenzial für AM durch Extrusion zu ermitteln und eine gute Verarbeitbarkeit (größere Höhe möglich bei gleichzeitiger Formstabilität) zu erreichen. Das Material sollte in der Lage sein, sein Eigengewicht zu tragen und eine begrenzte Verformung des gedruckten Teils während des Trocknens bei Raumtemperatur aufweisen. Dies wurde erreicht, indem man sich auf die Anpassung und Optimierung konzentrierte, die durch die rheologische Charakterisierung und die Einstellung der Druckparameter kontrolliert wurde, um eine optimale Formgenauigkeit der 3D-gedruckten Teile zu gewährleisten. Es wurde ein

Leitfaden für die Druckanpassung und Designbeschränkungen vorgeschlagen, die auf die entwickelte Paste abgestimmt sind.

Für eine Düse mit einem Durchmesser von 1,6 mm besteht die beste Formulierung aus 15 % Cellulose, 69,2 % Aspen, 69,2 % Wasser, 7,5 % CMC, 2,8 % Lecithin und als Füllstoff wird Stärke mit einem Anteil von 5,6 % verwendet.

Für die Düsengröße 1,2 mm ist die beste Formulierung: Zellulose 15,5 %, Aspen, Wasser 71,5 %, CMC 5,8 %, Lecithin 2,9 %, Füllmittel Stärke 4,3 %. Düse 0,6 mm, beste Formulierung mit Zellulose 13,4%, Aspen, Wasser 78%, CMC 3,6%, Lecithin 2,6%, Füllstoff Kreide 2,3%.

Die zusammengefassten Ergebnisse führen zum 3D-Papierdruck von Produkten mit komplexen Geometrien, mechanischen Eigenschaften und einer Reihe von Möglichkeiten, wie z.B.

1. sicher durch die Verwendung ungefährlicher Materialien
2. 100% recycelbar
3. große komplexe Teile.
4. komplexes Bauteildesign
5. Selbsttragendes Material, das sein eigenes Gewicht tragen und halten kann und dabei seine Form und Stabilität beibehält.
6. Die Bauhöhe erreicht ca. 200 mm ohne Knickprobleme oder Kollaps während des Trocknungsprozesses.  
Das Feuchtgewicht beträgt (7,27-8,08) kN/m<sup>3</sup> und das Trockengewicht (4,02-5,67) kN/m<sup>3</sup>.
8. Druckparameter: Luftdruck bis zu 0,3 - 0,85 Mpa
9. Zugfestigkeit bis zu 4,5 Mpa.
10. Haftfestigkeit bis zu 20 % der Zugfestigkeit.
11. Elastizitätsmodul erreicht ca. 6 GPa.
12. Ökonomischer Aspekt des Materials und 1 kg des Materials in der Nähe von 10 €/kg.
13. Das Material lässt sich optimieren, um die geringste Schrumpfung des extrudierten Filaments zu erreichen, die 17,4 % beträgt.
14. Die Maßhaltigkeit eines 3D-gedruckten Teils nach dem Trocknen erreichte 14,8%.

Zur Erreichung dieses Ziels konzentrierte sich diese Arbeit auf die Entwicklung von - Pastenformulierungen auf der Basis von kurzen und langen Cellulosefasern, CMC, Lecithin und Füllstoffen (unlösliche Kartoffelstärke, Kreide oder Mgso<sub>4</sub>).

- Trocknung im Weltraum.

- Verwendung eines 3D-Druckers (LUTUM -VormVrij® 3D Clay Printer Version 2.1).

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# List of Symbols

3D	Three-dimensional
AM	Additive Manufacturing
ASTM	American standards for testing material
B.R.	Beating Revolutions
CAD	Computer-Aided Design
CFR	Code of Federal Regulations
CMC	Carboxymethyl cellulose
CNC	Cellulose Nano Crystals
CNF	Cellulose Nanofibrils
DIC	Digital Image Correlation
DIN	Deutsches Institut für Normung
E	Elastic modulus
EC	Ethyl cellulose
ECF	Elemental Chlorine Free
FDM	Fuse Deposition Modeling - FDM Technology
FFF	Fused filament fabrication.
HPC	Hydroxypropylcellulose
HPMC	Hydroxypropylmethylcellulose
ISO	International Organization for Standardization
IGU	Insulated Glass Unit
kN	Kilo Newton- force unit
Mc	Methyl cellulose
mm	Millimeter
MPa	Megapascal
NBHK	Northern Bleached Hardwood Kraft
NBSK	Northern Bleached Softwood Kraft
PLA	Polylactic acid (one of the most used plastics in the AM)
RPM	Revolutions per minute
Rq	Surface roughness
STL	File Format of Stereolithography CAD Software
TCF	Total Chlorine Free
VIC	Visual Image Correlation (Non-Contact, Full-Field Strain & Deformation Measurement System)
W.f	Wood fibers
WRV	Wasserrückhaltevermögen (Water Retention Capacity)



3D paper printed object showing edge details for interlocking.



# 1 Introduction

Additive manufacturing can be a valuable tool for engineering paper materials for architectural and structural applications. As it deals with these possibilities:

2 In the concept, "AM represents a paradigm shift in design and product construction compared to traditional technology [Paul, C.P., and Jinoop, A.N. 2023], especially for paper material construction.

For the development of the construction with paper material, additive manufacturing is the solution. This is due to several factors:

1. 3D paper printing allows for more freedom and complexity in design, enhancing construction with operational flexibility. With 3D printing, intricate shapes, patterns, and custom designs can be easily created, allowing for greater architectural and engineering possibilities. On the other hand, paper-cardboard sections are limited in terms of design options due to the limited shapes of the sections from pressed paper or corrugated paper, such as layered sheets, tubes, etc. and limited techniques such as sheet layering by additive and subtractive manufacturing, milling, and folding. These sections and techniques do not provide a wide range of complexity. The inherent stiffness and bendability of the material can also limit design options, making it difficult to achieve certain desired shapes or functionalities. Therefore, compared to additive manufacturing technology, it's laborious and unproductive to produce such complex geometries [H. Bruck, Y. Chen, and S. K. Gupta, 2021].
2. Paper-cardboard elements generally have limited structural strength and durability compared to 3D printed paper parts. 3D printing allows the creation of intricate and complex geometries that can be reinforced with internal structures or by designing the print orientation to control the failure mechanism and support mechanisms, increasing their load-bearing capacity and overall durability. In addition, paper-cardboard sections have limited performance, requiring many joints and the need for bonding between layers as an adhesive bond, which localizes failure and increases the possibility of debonding, compared to 3D paper-printed sections which have full bonding between layers. This makes 3D paper printed parts more suitable for applications requiring higher structural integrity. [Tan, X., et. al., 2021], [Chen, Y., & Wang, X., 2019] and [Paoletti, P., et. al., 2020].
3. With its ability to accurately reproduce complex geometries with better aesthetics, 3D paper printing is ideal for precise and efficient manufacturing. These characteristics lead to higher production efficiency and limited material wastage compared to manual paperboard mounting.

Furthermore, 3D printing allows to produce consistent and uniform profiles, ensuring a more acceptable quality control [Paoletti, P., et. al, 2020] and [Kotnik, T., & Dolšak, B., 2019].

### 3 The concept of “its properties help to perform the functionalities.”

Paper material offers various functionalities that make it fit for engineering purposes. 3D printed paper material is highly flexible and can be easily molded or formed into different shapes and geometries. It is capable of complex geometries, making it useful for the creation of intricate structures and architectural models. Paper material has natural insulating and sound absorbing properties, both thermal and acoustic. It can be used as an insulating element in buildings, where the reduction of heat transfer and the transmission of noise are important. For acoustic purposes, according to measurements of the sound absorption coefficient, both loose and mixed with the glue that builds the structure [A. Trematerra<sup>1</sup> and I. Lombardi, 2017], cellulose-based paper material is considered a good system for absorbing sound in a simple, economic, and environmentally friendly way.

In addition, when used as an internal architectural part for acoustic purposes, the material also helps as a thermal insulator, could be in contact with humans, because it is not harmful to health, recyclable, renewable, widely available, low cost, and light.

### 4 The concept “ waste and recyclability”

Paper is a renewable and recyclable material that contributes to environmental sustainability by being an environmentally friendly choice for engineering applications. It can contribute to sustainable design practices because of its smaller carbon footprint than many other materials. Conversely, plastics used in 3D printing, especially non-biodegradable plastics, can contribute to environmental problems such as waste and pollution. Increasing bans on plastic products will increase paper production due to waste issues, even though paper is less durable and stiff than plastic. Paper is considered to have the advantage of decomposing much faster than plastic, so it is less likely to be a source of litter or a threat to wildlife. In other words, it's more widely recyclable and environmentally friendly [Lamberti, F.M., et al 2020], and [Hopewell J, et al 2009]

## 1.1 Research Variables

The factors and characteristics that have been measured, manipulated, or controlled in a research study are the following:

- Paste formulation.

This describes the process of developing a mixture consisting of solid particles dispersed in a liquid medium. Manipulating the paste formulation and the proportions of the ingredients provides valuable insight into the effect of each component within the binder formulation in providing adhesion characteristics for 3D-printed paper materials.

It helps provide a vision for optimizing these factors to achieve the required paper 3D printing aspects.

The specific components and proportions of the paste formulation were varied according to the intended operation, such as viscosity and rheological properties, and application, such as providing sufficient strength as a self-supporting material to print large slender elements. This was done to ensure extrudability, buildability, and adequate adhesion. The variables investigated were:

- Selection of raw materials and ingredients

The necessary ingredients were identified to give the paste the desired properties.

- Proportions of components in the mix formulation

The appropriate proportions of each ingredient were determined based on the desired functionality, compatibility and performance required, depending on viscosity, rheological properties, and printability.

This was done by adding thickeners, adhesion components and adjusting the concentration of the liquid medium. Finally, the formulated paste is tested to ensure that it meets the desired specifications, such as viscosity, extrudability, buildability, stability, and other relevant parameters. It's then essential to re-design and make the necessary adjustments if the results are not within the required range.

- Nozzle size

To investigate different resolutions, the extrusion process was studied for different nozzle resolutions (1.6 mm, 1.2 mm, 0.6 mm). Extrudability and buildability were considered, and the compound formulation was improved to gain better performance.

- Printing parameters

These refer to the settings and parameters that are adjusted to control the printing process and achieve the desired results, such as print speed, layer height, infill setting, flow rate and retraction settings. These parameters were varied and redesigned according to the results of the structural integration to control the dimensional accuracy of the 3D paper printed elements.

- 3D models design

The design of complex 3D models and the printing results were studied to examine the efficiency of printing and to discover the challenges of 3D printing with paper material that could be encountered for the purpose of redesigning and modifying the model and to see the printability and limitations. The dimensional accuracy and structural integration for each 3D model is examined.

These variables are explored within the chapters, as shown in Fig. 1.2.

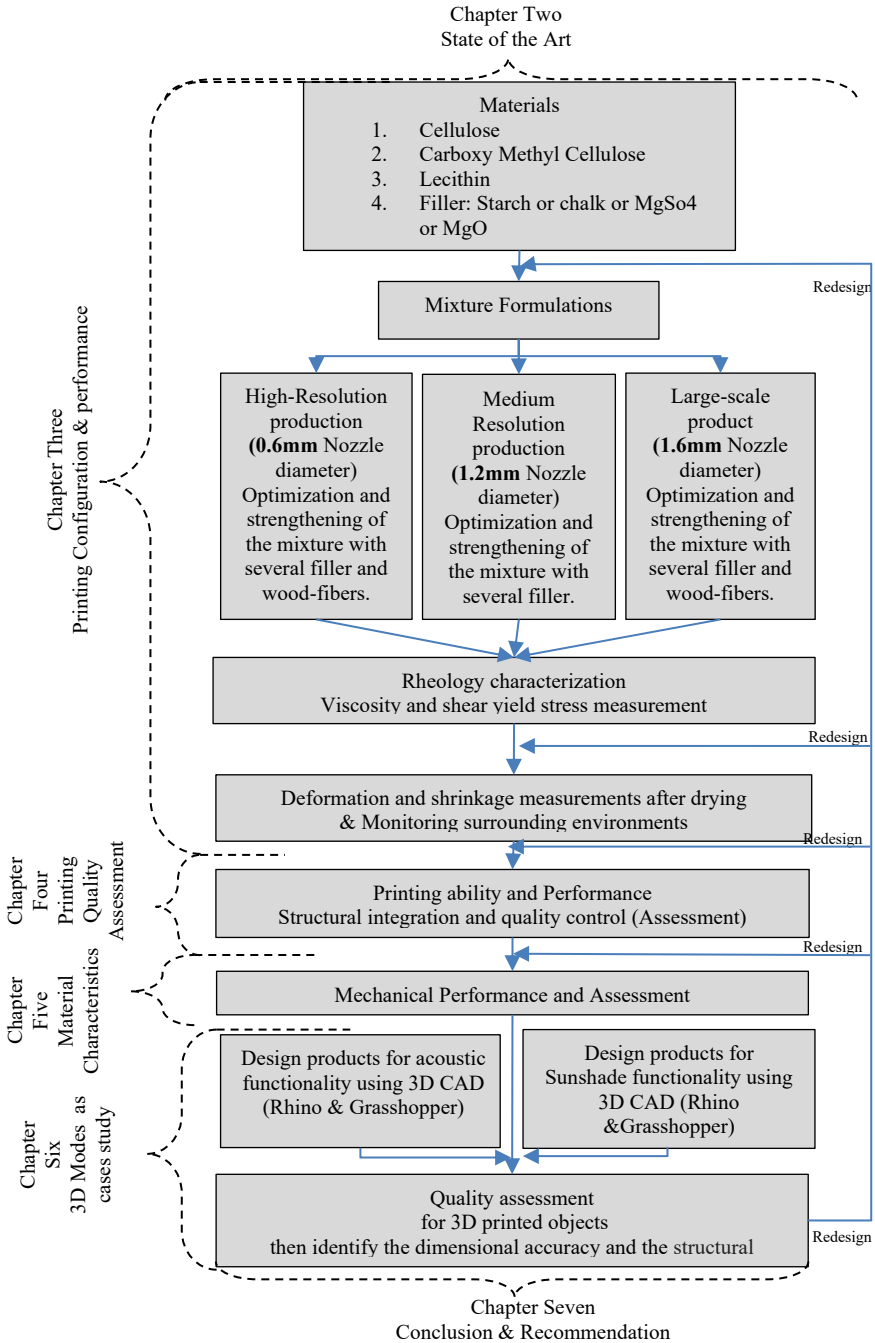


Figure 1.2 Scheme of research variables studied within chapters.

## 1.2 Research Objectives

The research objectives include:

1. To consider the implementation of advanced manufacturing technology for the production of elements as an innovative formulation for the paper material from a cellulose-based material. In order to be able to produce a complex geometry without self-supporting material.
2. To find paper pastes that fit the nozzles (1.6 mm, 1.2 mm, 0.6 mm) and achieve extrudability, buildability, and adequate adhesion.
3. To optimize the production process to improve product quality, production process efficiency, shrinkage control, and dimensional requirements.

These objectives were achieved by:

- Trial and error to find the best compound formulations and measure the rheological characterizations, both viscosity and shear yield stress.
- By trial and error to find the best printing parameters.
- By measurement of deformation and shrinkage after drying
- By measuring the mechanical performance.
- By creating 3D models and using them to specify the details and challenges of the printing performance and then evaluate the results.

## 1.3 Research Questions

The formulation of the paper paste was designed, then, the geometries are printed using a 3D printer with the designed paper paste. From this point, the investigation is carried out to examine the readiness of this design paste to follow the requirements of the production process in terms of printability, buildability, structural integration, and fulfill the manufacturing of a complex geometry.

Redesign of the paper paste is required and printing parameters in case of not achieving these challenges. The quality of these printed elements is investigated in terms of the printing outcomes and challenges to introducing the design into a 3D paper-printed model in terms of structure integration and production efficiency, and dimensional accuracy.

The main question that the whole dissertation will cover is:

**How to produce 3D paper printed models with better quality and dimensional accuracy?**

The quality here indicates by the nice outcomes and referred to uniform lines with smooth extruded outlines and have acceptable aesthetics with geometrical integration and considerable accuracy. The research carried out successive procedures that answer this main question and clarified by each chapter until reaching the goal in the final chapter, which producing a 3D paper functionalized model.

Each process has been considered as a millstone that filters the successive steps and separates the paths according to the product requirements. Therefore, the following sub-questions will guide the principles for these millstones.

**- Chapter one:**

Why is AM needed to produce paper structures in the field of engineering for the built environment?

What are the main objectives achieved through the research?

**- Chapter two:**

What are the overall parameters that influence the quality of the 3D printed objects?

What are the previous studies that have investigated the use of cellulose as multiple forms within AM?

**- Chapter 3:**

What is the formulation and mix design of the pastes/mixtures in terms of (changing the type of cellulose, type of filler, proportions of the components and using the wood fibers)?

What are the optimal mixtures suitable for the nozzles in terms of rheological properties to meet extrudability and buildability?

What are the solutions used in this study to minimize shrinkage?

How can the quality of 3D paper printed objects be improved and optimized in terms of the production process?

**- Chapter 4:**

What are the results of the evaluation of the printing performance and the assessment of the printing quality?

What are the defects of the 3D paper printing results?

**Chapter 5**

What is the mechanical behavior of the material from different aspects such as filament orientation, adhesive strength, nozzle size and strengthening the paste with wood fibers?

What are the proper print parameters that give better mechanical performance for solid objects?

**- Chapter 6:**

What are the potential applications for 3D paper models?

What are the results of evaluating the quality and dimensional accuracy of the 3D models?

**- Chapter 7:**

What are the most valued results of this research?

What are the design limitations for 3D printed paper models from this research?

What are the avenues opened by the endpoints of this research?



## 1.4 Thesis Structure

The thesis is divided into six chapters:

**Chapter One** presents the main ideas of the research, including explicit statements of significance specific to the topic studied, and illustrates the main problems and questions of the research.

**Chapter Two** reviews the literature in the context of the research in parts:

Part I presents a thorough examination of cellulose resources, covering types, production processes, resulting products and their impact in the field. In this section, the specific type of cellulose used in the research is identified and the different types are distinguished according to the production process. Part II focuses on the use of cellulose as a paper-based material within additive manufacturing (AM) technology. It provides a detailed overview of its various applications and discusses primary strategies to overcome the challenges involved. It also explores conceptual frameworks aimed at unlocking the potential of paper-based materials to achieve specific goals within AM processes. The motivation behind the research and its main objectives and contributions are summarised. The **third chapter** describes in detail the approaches followed for all the stages of the experimental work, from the material used, the mix design of the compounds, the mixing procedure, to the manufacturing by 3D printing. The rheological behavior and shrinkage measurements for all the mixes used are also provided. In addition, it offers solutions and optimizations to the problems of printing performance, geometric accuracy and structural integration of 3D paper printed objects.

**Chapter four** shows an evaluation that has been carried out to provide guidelines to produce 3D paper printed objects.

**Chapter five** presents the mechanical properties and material response for 3D paper printing in terms of stress-strain behavior, Young's modulus and failure mode for the experimental work conducted in chapter three.

**Chapter six** presents the details of the evaluation of the 3D paper printed models. Identification for the printing efficiency and the effect factors of different printing results for these 3D models were also presented in terms of structural integration, and efficiency of printed geometry and dimensional accuracy. The 3D models were designed to satisfy the functionality of 3D printed paper models as potential applications for shading elements by optimizing the passive solar radiation energy, and for acoustic purposes for interior partitions or cladding acoustic panels with aesthetic and complex geometries.

**Chapter seven** summarizes the conclusions drawn from the present study and the design considerations in the main of the evaluations and limitations for: the printing performance and mechanical performance for 3D paper printed objects for the building environment. The chapter will then give prospective recommendations for future work.

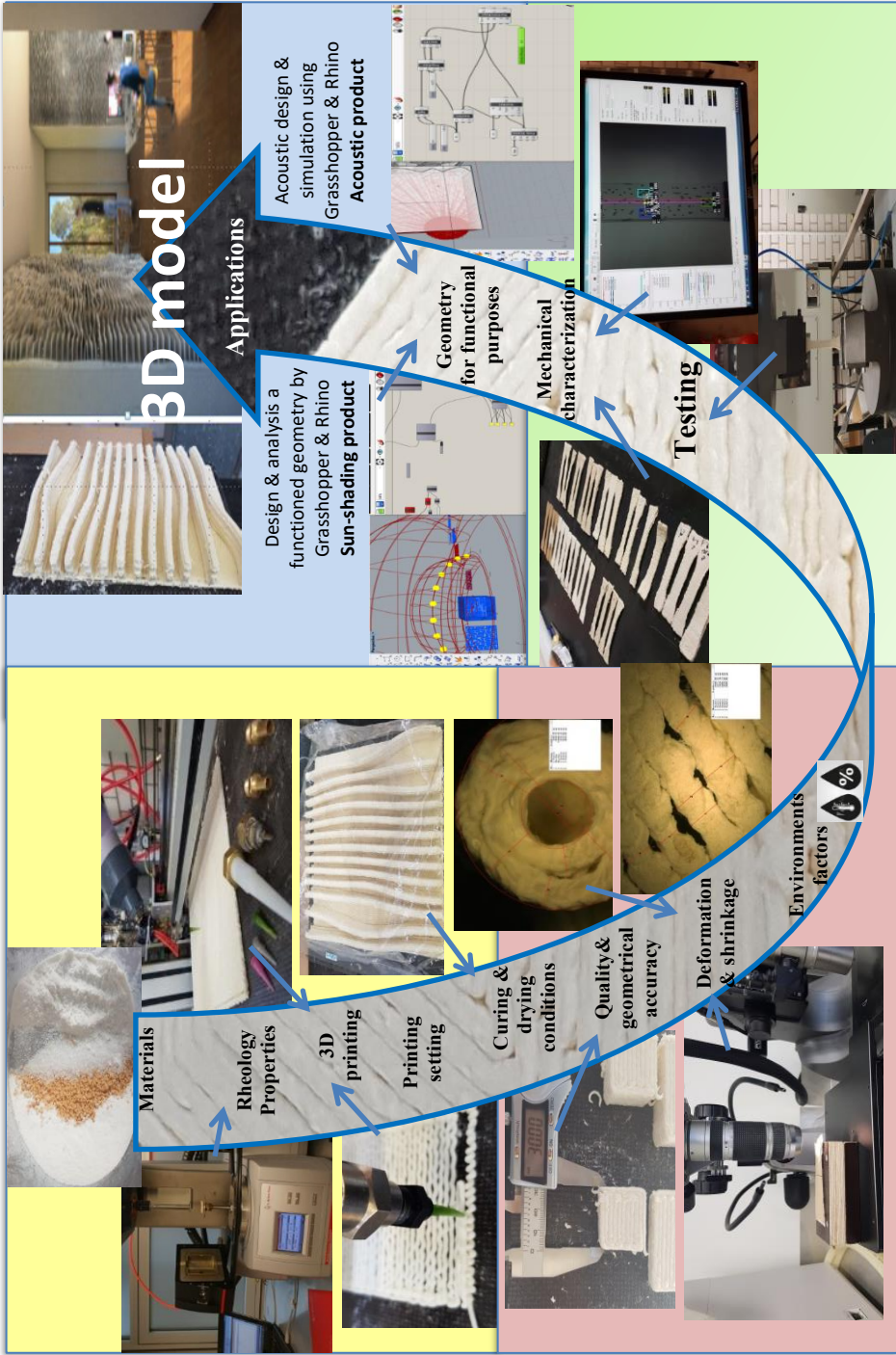
## 1.5 Methodology

The goal of this research focuses on producing 3D paper printed structures that achieve functionality for the built environment. The production approach in the process must be developed and optimized to reach these customized products with better quality and desired accuracy. The method for presenting this approach signifies following several steps till reach the goal. These steps will be representing sectors such as: optimizing a mixture according to the extrusion ability and buildability and controlling the deformation and shrinkage after drying. Furthermore, for dealing with buckling issues, large-scale printing, and for manufacturing objects with better material properties.

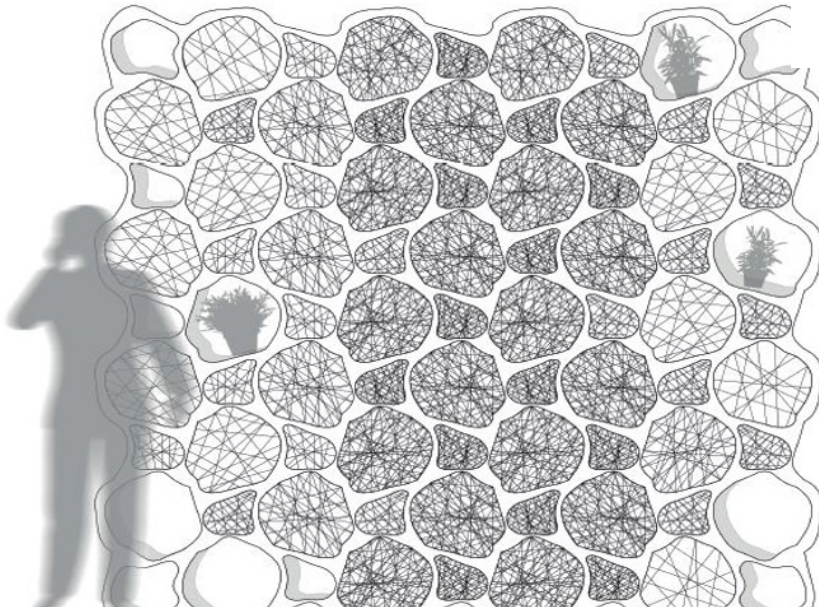
The investigation of the different sectors involved will be based on engineering principles for manufacturing a product within additive manufacturing. These engineering principles will be represented with the engineering process that matches the objectives of this dissertation. The objectives clarify an essential planned system that covers the essence of all the contents of this research. There are several parts of the mentioned system that are not regular for the common additive manufacturing since this research investigates a new input material for AM technology, which are the mixtures that have been studied from mainly cellulose. This is considered something novel and modern in the fields of research and industry. Therefore, some steps and additional major sectors are being linked to the followed engineering process that fit the interest of objectives of the dissertation.

The engineering process has been followed described in Fig. 1.1 for design, optimization, and manufacturing with the quality control that would achieve the structural integration and performing the purposes and functionality that it has been designed for. Redesign is necessary to make a loop in the engineering process for optimizing and finding the most optimal solution that performs an outrun of the issue. The redesign represents in the current investigation by moderating the values for the same variable or other input parameters that are the basis for the design case for each sector.

A conceptual framework for the variables related to the research was illustrated by a visual representation for the successive parameters that impact the followed approach, as shown in Fig. 1.3.



**Figure 1.3** The Methodology and the research path.



3D String shade wall (a), and 3D paper printed model for the string wall part.  
This 3D model is presented at BAMP exhibition – Biennale in Venedig 30. Juni 2021.



## 2 State of the Art

Additive manufacturing has indeed brought about a significant evolution in product development and manufacturing processes. This evolution is continuously expanding as new technologies, materials, and applications are developed [Redwood, B. 2017]. This is qualifying AM at high industrial levels using different materials. The number of materials used by AM is constantly increasing, giving the potential to implement more adopted materials that AM can handle and treat according to their properties and functionality. [Mahmood A, 2022]. Cellulose, a natural polysaccharide found in the cell walls of plants, has gained significant interest in 3D printing applications due to its abundant availability, renewable nature, and biodegradability. The incorporation of cellulose-based materials into 3D printing processes offers several advantages and opens new possibilities in various fields. Cellulose-based materials used in AM can include cellulose acetate, cellulose nanocrystals (CNC), cellulose derivatives and cellulose-based composites. These materials can be processed into filaments, powders, or liquids suitable for various 3D printing techniques [Xiaoyu Bi, Runzhou Huang, 2022].

This chapter reviews and explores the state of the art for various aspects of the production of AM paper products from cellulosic materials using different additive manufacturing technologies, including design and manufacturing processes, material properties and material formulation development.

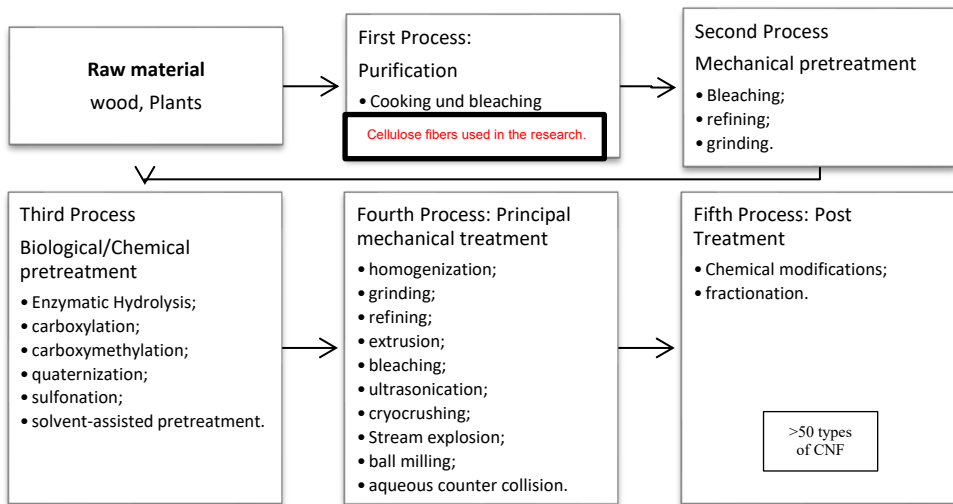
### 2.1 Cellulose

The cellulose forms that are adequately available in the market are lignocellulose, bleached pulp, dissolving pulp. However, to understand the uses and implementation of cellulose within AM, first must review its structure, types and its functioning, and its available forms. Cellulose is a versatile biopolymer with a variety of formulations. The most common types of cellulose are:

1. Native cellulose (cellulose fiber)

It refers to the natural form of cellulose found in plants. Cellulose fibers are the main load-bearing component in trees and plants due to the high modulus of its crystalline part, which can reach 140 GPa [Daniele R., 2019]. The type of cellulose used in the study is the pulp cellulose fibers from the first purification stage, as indicated in Fig. 2.1.

Cellulose fibers are used for industries such as papermaking and textiles. It's used also in food additives, pharmaceuticals and cosmetics that is typically found in younger plant tissue [Brethauer, S. et. al, 2020] and [Gauss Ch. et al, 2021].



**Figure 2.1** Wood fibers treatments and up to five stages to produce CNF [Thibaut, C. 2020].

## 2. Nanocellulose

- Nanocrystals (CNC): are produced by breaking down cellulose fibers into smaller crystalline particles. It has unique characteristics, including high strength, high aspect ratios and excellent optical clarity. CNC is used in nanotechnology, biomaterials, coatings, and polymer reinforcement.
- Cellulose Nanofibrils (CNF): are long and slender structures derived from cellulose fibers. They have high strength. CNFs are used in a wide range of applications including films, coatings, gels and as reinforcement in nanocomposites.
- Bacterial cellulose (BC) is a gel-like substance composed of interconnected cellulose nanofibers produced by certain types of bacteria [Chen et al., 2011; Moon et al., 2011; Torres et al., 2012].

## 3. Cellulose Derivatives

Cellulose Microfibril can be chemically modified to produce cellulose derivatives with specific properties. Industries such as pharmaceuticals, food additives, adhesives and personal care products use these derivatives. The types of available and widely used cellulose derivatives are shown in Fig. 2.2.

### - Carboxymethylcellulose (CMC)

It is a water-soluble polymer derived from cellulose fibers by chemical modification.

The production of CMC generally involves: 1) cellulose extraction through pulping. 2) Alkali treatment with NaOH. 3) Carboxymethylation by monochloroacetic acid (-CH<sub>2</sub>COOH) or its sodium salt with a catalyst. 3) Neutralization and washing: 4) Drying and Finishing. [S. Slotte, 2021] and [Gauss et al, 2021].