Faculty of Mechanical Technology and Engineering

ANALYSIS OF MECHANICAL BEHAVIOUR OF 3D PRINTED POLYLACTIC ACID – POLYCARBONATE-URETHANE FOR ARTICULAR CARTILAGE MATERIAL

Muhammad Nur Akmal bin Kazim

Master of Science in Mechanical Engineering

2024

ANALYSIS OF MECHANICAL BEHAVIOUR OF 3D PRINTED POLYLACTIC ACID – POLYCARBONATE-URETHANE FOR ARTICULAR CARTILAGE MATERIAL

MUHAMMAD NUR AKMAL BIN KAZIM

A thesis submitted
in fulfilment of the requirements for the degree of Master of Science
in Mechanical Engineering

Faculty of Mechanical Technology and Engineering

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2024

DEDICATION

To my beloved wife, mother, father and also a tribute to my late grandfather.

ABSTRACT

Polylactic acid (PLA) and polycarbonate urethane (PCU) are widely utilized materials in the medical field, particularly for articular cartilage applications. These materials exhibit remarkable strength, durability, and high wear resistance. However, the current artificial implants made lack of porous properties required for optimal lubrication. Thus, the purpose of this study is to analyse the mechanical behaviour of 3D printed polylactic acid – polycarbonate urethane for artificial cartilage material. A polymer blend of PLA-PCU with the composition of 90% PLA and 10% PCU was extruded using twin-screw extruder. The Fused Filament Fabrication (FFF) technology was employed to print the specimen. Subsequently, the response surface methodology was utilized to analyse the experimental data and develop an effective empirical prediction model. A hardness test using a Shore D durometer and a compression test using Instron UTM5 were carried out. The study's results showed that the generated model was not significantly related to the absorption rate but was highly significant for the surface roughness response parameter. Detailed analysis of the results revealed that layer thickness was the most significant factor affecting surface roughness where higher layer thickness produced higher surface roughness, while PCU composition affected absorption rate. The optimal printing process parameters for achieving the desired surface roughness (2.5400 µm) and absorption rate (0.0470%) were obtained by setting the PCU concentration to 10 wt.%, layer thickness to 0.1 mm, nozzle speed to 15 mm/s, and extruding temperature to 195°C. The study results demonstrated that the addition of PCU to PLA reduced the hardness level of the specimens. Interestingly, specimens with the same element exhibited almost the same hardness level despite being fabricated using different layer thicknesses. Moreover, the results have shown that compared to 3D-printed PLA, adding PCU to PLA reduced the compressive strength and Young's modulus of the 3D-printed polymer blend. The thickness of the printing layers has an impact on the mechanical characteristics of 3D-printed specimens as well as specimen with lower layer thickness is stronger and stiffer. It has also been found that higher layer thickness resulting higher hardness-to-elasticity ratio, which is an essential factor for proper load transfer. The cross-sectional surface area of the compression-moulded specimen is smoother than that of the 3D-printed specimen, which contributes to distinct sorts of failure modes for both specimens, according to the SEM image analysis. These findings suggest that PLA-PCU composite materials could be a suitable option for medical applications that require high wear resistance, strength, and durability.

ANALISIS KELAKUAN MEKANIKAL CETAKAN 3D ASID POLILAKTIK – POLIKARBONAT-URETANA SEBAGAI BAHAN ARTIKULAR TULANG RAWAN

ABSTRAK

Polilaktik asid (PLA) dan polikarbonat uretan (PCU) adalah bahan yang digunakan secara meluas dalam bidang perubatan, terutamanya untuk aplikasi kartilaj artikular. Bahan-bahan ini menunjukkan kekuatan, ketahanan dan rintangan haus yang luar biasa. Walau bagaimanapun, implant tiruan terkini yang diperbuat mempunyai kurang sifat berongga yang diperlukan untuk pelinciran yang optimum. Oleh itu, tujuan kajian ini adalah untuk menganalisis kelakuan mekanikal polilaktik asid – polikarbonat uretan yang dicetak dalam 3D sebagai bahan kartilaj tiruan. Campuran polimer PLA-PCU dengan komposisi 90% PLA dan 10% PCU diekstrud menggunakan pengekstrudan berulang. Teknologi Fused Filament Fabrication (FFF) digunakan untuk mencetak spesimen. Seterusnya, Response Surface Methodology (RSM) digunakan untuk menganalisis data eksperimen dan membangunkan model ramalan empirik yang berkesan. Ujian kekerasan menggunakan durometer Shore D dan ujian mampatan menggunakan Instron UTM5 dilakukan. Keputusan kajian menunjukkan bahawa model yang dihasilkan tidak berkait secara signifikan dengan kadar penyerapan tetapi sangat signifikan untuk parameter respons kekasaran permukaan. Analisis terperinci keputusan menunjukkan bahawa ketebalan lapisan adalah faktor paling signifikan yang mempengaruhi kekasaran permukaan di mana ketebalan lapisan yang lebih tinggi menghasilkan kekasaran permukaan yang lebih tinggi, manakala komposisi PCU mempengaruhi kadar penyerapan. Parameter proses pencetakan optimum untuk mencapai kekasaran permukaan yang diinginkan (2.5400 µm) dan kadar penyerapan (0.0470%) diperoleh dengan menetapkan kepekatan PCU kepada 10 wt.%, ketebalan lapisan kepada 0.1 mm, kelajuan nozzle kepada 15 mm/s, dan suhu penyemperitan kepada 195℃. Keputusan kajian menunjukkan bahawa penambahan PCU kepada PLA mengurangkan tahap kekerasan spesimen. Spesimen dengan elemen yang sama menunjukkan tahap kekerasan yang hamper sama walaupun diperbuat menggunakan ketebalan lapisan yang berbeza. Selain itu, keputusan menunjukkan bahawa berbanding dengan PLA yang dicetak dalam 3D, penambahan PCU kepada PLA mengurangkan kekuatan mampatan dan modulus Young bagi campuran polimer yang dicetak dalam 3D. Ketebalan lapisan pencetakan mempengaruhi ciri-ciri mekanikal spesimen yang dicetak dalam 3D dan spesimen dengan ketebalan lapisan yang lebih rendah adalah lebih kuat dan kaku. Didapati juga bahawa ketebalan lapisan yang lebih tinggi menghasilkan nisbah kekerasan kepada keanjalan yang lebih tinggi, yang merupakan faktor penting untuk pemindahan beban yang betul. Luas permukaan keratan rentas spesimen acuan mampatan adalah lebih licin daripada spesimen cetakan 3D, yang menyumbang kepada jenis mod kegagalan yang berbeza untuk kedua-dua spesimen, menurut analisis imej SEM. Penemuan ini mencadangkan bahawa bahan komposit PLA-PCU boleh menjadi pilihan yang sesuai untuk aplikasi perubatan yang memerlukan rintangan haus, kekuatan dan ketahanan yang tinggi

ACKNOWLEDGEMENTS

In the Name of Allah, the Most Gracious, the Most Merciful

I express my heartfelt gratitude to Allah the Almighty, who is not only my creator but also my unfailing pillar of strength, the source of my inspiration, wisdom, knowledge, and understanding. Without His guidance and support, I would not have been able to complete this thesis successfully. I would like to extend my appreciation to the Universiti Teknikal Malaysia Melaka (UTeM) for providing me with the research platform to pursue my studies. I am also grateful to the Malaysian Ministry of Higher Education (MOHE) for the financial assistance provided during my studies.

I am especially thankful to my main supervisor, Prof. Dr. Mohd Fadzli bin Abdollah, Dean of the Faculty of Mechanical Technology and Engineering at UTeM, for his invaluable support, advice, and inspiration throughout my research journey. His unwavering patience and guidance, along with his priceless insights, will always be remembered. I would also like to express my gratitude to my co-supervisor, Dr Hilmi bin Amiruddin, from the Faculty of Mechanical Technology and Engineering at UTeM, for his continuous support throughout my studies.

Special thanks to my wife, Dr Nor Amirah Afiqah binti Mohamad Abu Bakar (MBBS) for giving me an insight on articular cartilage injuries and treatment from a medical view. My sincere thanks go out to all the lecturers, assistant engineers, and friends who have taught and supported me during my academic journey. I am also deeply grateful to my beloved parents, who have been my pillar of strength and source of encouragement throughout all my endeavours. Finally, I would like to extend my gratitude to all the individuals who have provided me with assistance, support, and inspiration along the way. Thank you for your endless support, love, and prayers.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

µm	-	Micrometre
3D	-	Three dimensional
ANOVA	-	Analysis of variance
ASTM	-	American Society of Testing Materials
°C	-	Degree Celsius
CAD	-	Computer assisted drawing
CCD	-	Central composite design
DOE	-	Design of experiment
FDA	-	Food and Drug Administrative
FFF	-	Fused filament fabrication
FIDEC	-	Fibre and Biocomposite Development Centre
GAGs	-	Glycosaminoglycans
H	-	Hardness
HA	-	Hydroxyapatite
mm	-	Millimetre
MTIB	-	Malaysia Timber Industry Board
PCL	-	Polycaprolactone
PCU	-	Polycarbonate urethane
PEG	-	Polyethylene glycol

PLA	-	Polylactic acid
Ra	-	Average roughness
RSM	-	Research surface method
SEM	-	Scanning electron microscopy
SLA	-	Stereolithography
SLS	-	Selective laser sintering
UTM 5	-	Universal testing machine 5
wt.%	-	weight percentage

LIST OF PUBLICATIONS

JOURNAL PAPER

2. **Kazim, M. N. A.**, Abdollah, M. F. Bin, Amiruddin, H., Liza, S., and Ramli, F. R., 2023. Mechanical Properties of Additively Manufactured New Polymer Blend (Polylactic Acid-Polycarbonate Urethane) with Varying Printing Layer Thickness. *Journal of Materials Engineering and Performance*, 8 May 2023. https://doi.org/https://doi.org/10.1007/s11665-023-08261-1
3. **Kazim, M. N. A.**, Abdollah, M. F. Bin, Amiruddin, H., Liza, S., and Ramli, F. R., 2022a. Surface Quality and Absorption Properties of Polymeric Composite (PLA-PCU) Fabricated Using 3d Printing for Articular Cartilage Application. *Jurnal Tribologi*, 3 May, 169–185. https://doi.org/10.2139/ssrn.4093626

CONFERENCE ATTENDED

1. **Kazim, M. N. A.**, Abdollah, M. F. Bin, Amiruddin, H., Liza, S., and Ramli, F. R., 2022b. Hardness level and compressive properties of polymeric composite (PLA-PCU) fabricated using 3D printing with different layer thickness for articular cartilage application. *Proceedings of SAKURA Symposium on Surface Technology and Engineering Material 2022*, 26–28.

2. **Kazim, M. N. A.**, Abdollah, M. F. Bin, Amiruddin, H., Liza, S., Ramli, F. R., and Umehara, N., 2021. Multi-objective optimization of 3D-printed ABS using response surface methodology. *Proceedings of ICE-SEAM 2021*: Special Edition, 1–3.

CHAPTER 1

INTRODUCTION

1.1 Introduction

This chapter gives an overview of the research work carried out for finding out the mechanical properties of polymer blend of PLA-PCU. It begins with a description of the research background. Then, it was followed by the problem statement and the objectives of the research. Next, the scope of the research was also stated and the thesis outline was shown. This chapter is closed with a summary.

1.2 Background

Polycarbonate-urethane (PCU) and polylactic acid (PLA) are up-and-coming materials in the field of artificial joints due to their exceptional mechanical stability, durability, and resistance to wear and corrosion. However, current artificial meniscus implants made with thermoplastic lack the porous properties required for optimal performance. Thus, the use of 3D printing technology is crucial for the fabrication of porous artificial samples. Improving the mechanical properties of 3D-printed specimen and exploring its mechanical behaviour could result in longer product lifespans. This behaviour includes the properties and mechanisms of the contacting surfaces, the environment, and the printing parameters. Therefore, the aim of this study is to investigate the impact of printing process parameters on the surface roughness of 3D printed PLA-PCU and to explore its mechanical behaviour. The research methodology comprises several distinct phases, including an analysis of printing process parameters on the surface roughness and absorption properties of the fused filament 3D printed PLA-PCU. Before mechanical testing, the surface roughness, absorption properties and porosity of the samples will be evaluated under Ringer's solution. Next, the mechanical properties of the samples, such as the hardness, will be assessed using micro indentation method, while the compression test will be carried out using a compression machine. The results will be compared with those of moulded specimens. It is expected that the printing process parameters will impact the surface roughness, absorption properties, and mechanical behaviour of 3D printed PLA-PCU. Consequently, this study aims to provide new scientific knowledge on the behaviour of 3D-printed PLA-PCU, thereby improving its mechanical performance. Such an approach would be advantageous for patients experiencing knee pain due to deteriorating cartilage.

1.3 Problem Statement

It is interesting to note that polycarbonate-urethane (PCU) is being considered as a promising material for artificial joints due to its mechanical stability, durability, and resistance to wear and corrosion (Vrancken et al., 2013). However, it is currently not porous enough to sustain joint lubrication through a weeping mechanism, which works as a sponge that absorb the lubricant, that is an essential function of natural cartilage (Elsner et al., 2010). To address this issue, the fused filament 3D printing technique can be used to create a porous structure in polymeric blend of PLA-PCU. This would enable the material to support joint lubrication by allowing the passage of synovial fluid through the implant, thus reducing friction and wear.

PLA is widely used in the medical industry due to its excellent physical, chemical, and mechanical properties. It finds applications in 3D printing, tissue engineering, orthopaedics, and medication delivery (DeStefano et al., 2020). While current artificial cartilage implants made with PLA lack porosity (Elsner et al., 2010), additive manufacturing shows promise in creating porous implants at a lower cost providing it can be produced in-house, making it highly feasible and accessible (Martelli et al., 2016). However, PLA's stiffness is higher than that of natural cartilage (Farah et al., 2016; Gabarre et al., 2014) resulting higher wear and tear risk. PCU, with similar stiffness to cartilage, is gaining popularity in the medical field for prosthetic components like knee cartilage (Inyang and Vaughan, 2020; Vrancken et al., 2013). However, PCU requires high-end 3D printers for production and has the potential for wear due to its low stiffness (Miller et al., 2017; Sivakumar et al., 2021). Past research had proved that increasing the stiffness of material can enhance its wear resistance (Friedrich, 2018). Further research is needed to determine the tribological and material properties of PCU for artificial cartilage implants (AbdelGaied et al., 2015; Inyang and Vaughan, 2020).

Combining PLA and PCU materials in a polymeric blend is hypothesized to enhance the printability of PCU, potentially reducing PLA costs and stiffness. The fused filament fabrication (FFF) process, a recognised 3D printing technique, can be used for various purposes (Norani et al., 2021; P. Zhang et al., 2020). FFF involves depositing filament layer by layer to create a model from a CAD file (Rahmati and Vahabli, 2015). Analysing and improving the mechanical properties of the 3D printed PLA-PCU blend especially the rate of absorption, porosity, surface roughness, compression strength and hardness can extend the lifespan of the product.

Overall, this research on the behaviour of 3D printed PLA-PCU is a significant step towards the development of improved artificial joints, and it has the potential to revolutionize the field of orthopaedics.

1.4 Objectives of Research

1. To analyse the effect of printing process parameters (percentage of PCU, layer thickness, printing speed and extrusion temperature) on surface roughness and the absorption properties of 3D printed PLA-PCU.

2. To investigate the mechanical properties (surface roughness, the rate of absorption, hardness, compression strength and hardness to elasticity ratio) of both 3D printed and moulded PLA-PCU, in correlation with their optimal printing parameters.

1.5 Scope of Research

1. Create a set of design of experiment (DOE) by using research surface methodology (RSM) in DesignExpert.
2. Fabrication of specimen using fused filament fabrication and compression moulding. The material used for the specimen fabrication is PLA-PCU blend.
3. The testing involves are absorption, porosity, surface roughness, compression and hardness test. The absorption and porosity test will be performed under Ringer's solution.

1.6 Thesis Outline

Based on the previously presented objectives and on the proposed approach before, this dissertation is made up of five (5) chapters, the contents of which is summarized as follows:

Chapter 1. Introduction. In this chapter, an overview of the research that will be conducted is presented. The chapter covers various aspects, including the background of the study, research problems, objectives, scope, contributions, and importance of the research. In summary, this chapter provides an essential overview of the research that will be conducted, outlining its background, research problems, objectives, scope, contributions, and importance. It serves as a foundation for the rest of the study, providing the reader with a clear understanding of what the research is about and why it is significant.

Chapter 2. Literature Review. In this chapter, a comprehensive review of existing literature pertaining to knee problems is presented. The review encompasses various aspects of knee problems, including treatment options available, the role of 3D printing technology in addressing knee problems, and the potential benefits of using 3D printed artificial implants over traditional treatment methods. Additionally, the chapter explores the advantages and disadvantages of two commonly used materials in 3D printing for knee problems, namely polylactic acid (PLA) and polycarbonate urethane (PCU). Through this literature review, readers can gain insights into the current state of knowledge and the latest developments in the field of knee problems, and can form a better understanding of the potential of 3D printing technology in improving treatment outcomes for patients with knee problems. The sources consulted for this review are cited appropriately in the reference list.

Chapter 3. Methodology. The methodology chapter provides a detailed account of the experimental setup used to manufacture and evaluate the mechanical properties of 3D-printed PLA-PCU specimens. This chapter includes a comprehensive description of the materials, equipment, and test procedures used in the study. The experimental setup was carefully designed to ensure accurate and reliable measurements of the mechanical properties of the specimens. The chapter outlines the steps involved in the manufacturing process, which includes the preparation of the materials, the use of the 3D printer, and the post-processing procedures. Additionally, the chapter provides detailed information on the test procedures used to evaluate the mechanical properties of the specimens. The results obtained from these tests are presented in a clear and concise manner, along with appropriate statistical analysis to support the findings. By providing a detailed account of the experimental setup and procedures used in the study, readers can gain a better understanding of the methodology used and can assess the validity and reliability of the results presented in the study.

Chapter 4. Results and Analysis. The results chapter presents a thorough analysis of the surface roughness, absorption properties, porosity, and mechanical properties of the 3D-printed PLA-PCU specimen. The chapter includes detailed discussions of the findings, which are presented in a clear and concise manner. The results of the study are supported by statistical data analysis, which provides a comprehensive understanding of the performance of the 3D-printed specimens. Furthermore, the chapter provides a comparison of the mechanical properties of the 3D-printed specimen with those of the moulded specimens. This comparison will enable to assess the benefits of 3D printing technology in improving the mechanical properties of artificial knee implants. By providing a detailed analysis of the results, this chapter provides valuable insights into the performance and potential of 3D-printed knee implants.

Chapter 5. Conclusion and recommendation. The conclusion chapter provides a concise summary of the main findings and conclusions of the study on 3D-printed PLA-PCU artificial knee implants. The chapter discusses the potential applications and implications of the results, highlighting the benefits of using 3D printing technology in improving the mechanical properties of knee implants. Additionally, the chapter suggests areas for future research and development of 3D-printed knee implants, which can further enhance their performance and applicability. The implications of the study's findings are discussed in the context of the broader field of knee implants and orthopaedic surgery, emphasizing the potential of 3D printing technology to revolutionize the treatment of knee problems. The chapter concludes with a call to action for researchers and practitioners to continue exploring the possibilities of 3D printing technology in the field of knee implants, and to collaborate on developing innovative solutions that can improve patient outcomes and quality of life. Overall, the conclusion chapter provides a comprehensive overview of the study's key findings and insights, and highlights the potential of 3D printing technology to transform the future of knee implant surgery.

1.7 Summary

This chapter discussed on the background, problem statements, research objectives, scopes of research and thesis outline. It is noted that PLA-PCU blend is a promising material to overcome the problems. Thus, this thesis will discuss the previous research on the treatment of cartilage injuries, and the mechanical properties of polymer blend of PLA-PCU to address the problem.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter discusses the use of 3D printing technology for articular cartilage replacement. It covers the anatomy and function of articular cartilage, as well as the causes of cartilage damage and degeneration. It then delves into the potential of 3D printing technology for cartilage replacement, before delving into various treatment options such as autografts, allografts, and synthetic materials.

Various types of 3D printing methods used for cartilage replacement, as well as previous research on 3D printed cartilage replacement materials, with a focus on PLA, PCU, and their blends. It also discusses the technical difficulties of using 3D printing for cartilage replacement, as well as the biological and clinical implications of 3D printed cartilage replacement materials.

Finally, the text compares the mechanical properties of PLA, PCU, and their blends to traditional cartilage replacement materials, and it investigates the potential for future research in this area to optimise the mechanical properties of 3D printed materials for cartilage replacement and tissue engineering applications.

2.2 Anatomy and Function of Cartilage

Articular cartilage is a type of connective tissue that covers the surfaces of bones at joints, allowing for smooth, friction-free movement. Chondrocytes, extracellular matrix, and