



**STUDY OF 3D PRINTING FILAMENT MADE FROM RECYCLE
POLYPROPYLENE (PP) AND GROUND TYRE RUBBER (GTR)
TREATED WITH NaOH**



**BACHELOR OF MANUFACTURING ENGINEERING
TECHNOLOGY (PROCESS AND TECHNOLOGY) WITH
HONOURS**

2023



**Faculty of Mechanical and Manufacturing Engineering
Technology**



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Yusra Fitri Bin Yusoff

**Bachelor of Manufacturing Engineering Technology (Process and Technology) with
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WITH NaOH**

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Faculty of Mechanical and Manufacturing Engineering Technology

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2023

DECLARATION

I declare that this Choose an item. entitled “Study of 3D Printing Filament Made From Recycle Polypropylene (PP) and Ground Tire Rubber (GTR) Treated With NaOH” is the result of my own research except as cited in the references. The choosen item has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

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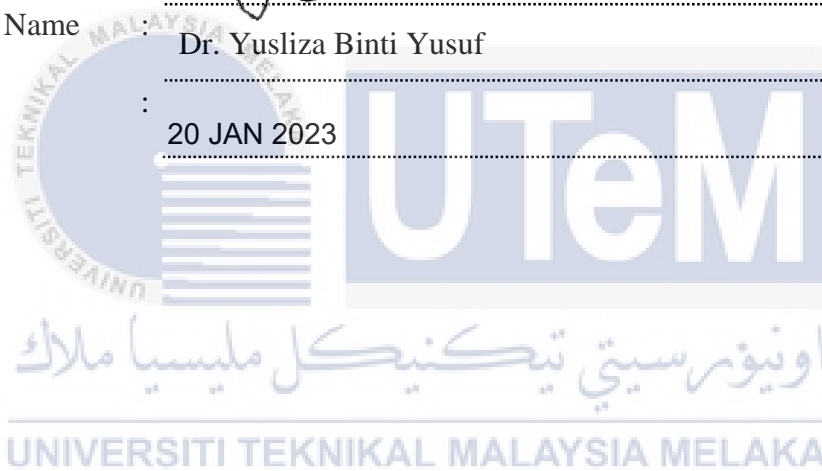
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APPROVAL

I hereby declare that I have checked this thesis and in my opinion, this thesis is adequate in terms of scope and quality for the award of the Bachelor of Manufacturing Engineering Technology (Process and Technology) with Honours.

Signature : 
Supervisor Name : Dr. Yusliza Binti Yusuf
Date : 20 JAN 2023



DEDICATION

Thank you, especially, to my wonderful family for their help in finishing this report. This research report is dedicated to my father, Yusoff bin Harun, who taught me that learning for the sake of learning is the best kind of learning. It is also dedicated to my mother, Rohani bt Othman, who taught me that even the most difficult task can be completed by taking one step at a time and to my lovely fiance, Nazirah Aqilah Binti Yunadi for your encouragement throughout this research.



ABSTRACT

The recycling and recovery of waste tyres is a severe environmental concern since vulcanised rubbers take several years to break down naturally cause the issue in waste management and environmental protection. On the other hand, using recycled plastic for 3D printer filaments can help reduce pollution and waste management challenges created by unwanted plastic. PP has similar properties to PLA, making it a potential material for deposition. Recycle Polypropylene (PP) exhibits good chemical resistance to a wide range of chemicals (acids, alkalis, and organic solvents), as well as fatigue resistance, environmental stress cracking resistance, detergent resistance, and ease of machining. Ground tyre rubber (GTR) was employed as a filler in a polymer matrix (composite material) for material manufactured utilizing the extrusion technique in this investigation. As a result, characterization of Ground Tyre Rubber (GTR) and Recycle Polypropylene (PP) as raw material for producing 3D printing filament is done. Then, analyzation on the microscopic, mechanical and water absorption properties of 3D printed specimen made from Ground Tyre Rubber (GTR) and Recycle Polypropylene (PP) at various composition to assess its properties. GTR that has been treated in homogenous NaOH (30g NaOH dissolves in 500ml water) before being combined with PP. GTR will be swirled to achieve consistent treatment before being soaked for 40 minutes, washed with distilled water, and dried for 24 hours at 60°C. GTR will then be subjected to particle size analysis to identify the particle size distribution in a substance. TGA and DSC will be used to study the thermal characteristics of both untreated GTR and treated GTR. To combine GTR and PP, a sintering method using a hot press machine will be used to create a solid material. It will then be crushed to a smaller size before being extruded using a single screw extruder to make the filament. By using those filaments, dog bone and rectangular shape samples have been printed by using 3D printing machine. For overall tensile test result, untreated PP/GTR has the highest tensile strength with the samples of 3% untreated GTR which is 18.3784 MPa compared to pure PP/GTR and PP/GTR treated with NaOH. Next for overall flexural test result, the highest flexural strength is 80.5253 MPa for 3% untreated PP/GTR sample followed by PP/GTR treated with NaOH samples with 61.2758MPa. For physical test, the percentage of water absorptions are inversely proportional to percentage of untreated GTR in PP/GTR samples. However, the result shows the percentage of water absorption for PP/GTR treated with NaOH samples is directly proportional to the percentage of GTR treated with NaOH as filler in PP/GTR filament. Thus, 3D printable eco-friendly PP/GTR filaments with low cost can be developed and used for 3D printing applications, contributing to reduce the impact of plastic and agricultural waste.

ABSTRAK

Kitar semula dan mendapatkan semula tayar buangan adalah kebimbangan alam sekitar yang teruk kerana getah tervulkan mengambil masa beberapa tahun untuk rosak secara semula jadi menyebabkan isu dalam pengurusan sisa dan perlindungan alam sekitar. Sebaliknya, menggunakan plastik kitar semula untuk filamen pencetak 3D boleh membantu mengurangkan pencemaran dan cabaran pengurusan sisa yang dicipta oleh plastik yang tidak diingini. PP mempunyai sifat yang serupa dengan PLA, menjadikannya bahan yang berpotensi untuk pemendapan. Kitar Semula Polipropilena (PP) mempamerkan rintangan kimia yang baik terhadap pelbagai jenis bahan kimia (asid, alkali dan pelarut organik), serta rintangan keletihan, rintangan retak tekanan persekitaran, rintangan detergen dan kemudahan pemesinan. Getah tayar tanah (GTR) digunakan sebagai pengisi dalam matriks polimer (bahan komposit) untuk bahan yang dihasilkan menggunakan teknik penyemperitan dalam penyiasatan ini. Hasilnya, pencirian Ground Tyre Rubber (GTR) dan Recycle Polypropylene (PP) sebagai bahan mentah untuk menghasilkan filamen cetakan 3D dilakukan. Kemudian, analisis sifat mikroskopik, mekanikal dan penyerapan air spesimen cetakan 3D yang diperbuat daripada Getah Tayar Tanah (GTR) dan Polipropilena Kitar Semula (PP) pada pelbagai komposisi untuk menilai sifatnya. GTR yang telah dirawat dalam NaOH homogen (30g NaOH larut dalam 500ml air) sebelum digabungkan dengan PP. GTR akan dipusingkan untuk mencapai rawatan yang konsisten sebelum direndam selama 40 minit, dibasuh dengan air suling dan dikeringkan selama 24 jam pada suhu 60°C. GTR kemudiannya akan tertakluk kepada analisis saiz zarah untuk mengenal pasti taburan saiz zarah dalam bahan. TGA dan DSC akan digunakan untuk mengkaji ciri terma kedua-dua GTR yang tidak dirawat dan GTR yang dirawat. Untuk menggabungkan GTR dan PP, kaedah pensinteran menggunakan mesin penekan panas akan digunakan untuk menghasilkan bahan pepejal. Ia kemudiannya akan dihancurkan kepada saiz yang lebih kecil sebelum disemperit menggunakan penyemperit skru tunggal untuk membuat filamen. Dengan menggunakan filamen tersebut, sampel tulang anjing dan bentuk segi empat tepat telah dicetak menggunakan mesin cetak 3D. Untuk keputusan ujian tegangan keseluruhan, PP/GTR yang tidak diuji mempunyai kekuatan tegangan tertinggi dengan sampel 3% GTR yang tidak dirawat iaitu 18.3784 MPa berbanding PP/GTR tulen dan PP/GTR yang dirawat dengan NaOH. Seterusnya bagi keputusan ujian lenturan keseluruhan, kekuatan lenturan tertinggi ialah 80.5253 MPa untuk 3% sampel PP/GTR yang tidak dirawat diikuti oleh PP/GTR yang dirawat dengan sampel NaOH dengan 61.2758MPa. Untuk ujian fizikal, peratusan penyerapan air adalah berkadar songsang dengan peratusan GTR yang tidak dirawat dalam sampel PP/GTR. Walau bagaimanapun, keputusan menunjukkan peratusan penyerapan air untuk PP/GTR yang dirawat dengan sampel NaOH adalah berkadar terus dengan peratusan GTR yang dirawat dengan NaOH sebagai pengisi dalam filamen PP/GTR. Oleh itu, filamen PP/GTR mesra alam cetak 3D dengan kos rendah boleh dibangunkan dan digunakan untuk aplikasi percetakan 3D, menyumbang untuk mengurangkan kesan sisa plastik dan pertanian.

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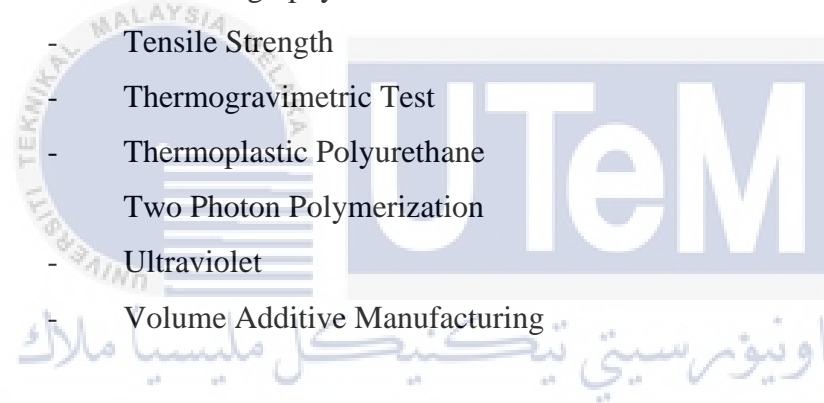
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LIST OF SYMBOLS AND ABBREVIATIONS

3D	-	3-Dimensional
ABS	-	Acrylonitrile Butadiene Styrene
AM	-	Additive Manufacturing
ASA	-	Acrylic Styrene Acrylonitrile
CB	-	Carbon Black
CNTs	-	Carbon Nanotubes
CAD	-	Computer Aided Design
CLIP	-	Continuous Liquid Interface Polymerization
DSC	-	Differential Scanning Colometry
DLP	-	Digital Light Processing
DIW	-	Direct Ink Writing
DP	-	Direct Printing
EB	-	Elongation at Break
EU	-	European Union
FDM	-	Fused Deposition Modeling
FFF	-	Fused Filament Febrication
GTR	-	Ground Tire Rubber
HDPE	-	High-Density Polyethylene
LDPE	-	Low-Density Polyethylene
ME3DP	-	Material Extrusion 3D Printing
PP/GTR	-	Mixture of Polypropylene and Ground Tyre Rubber as filler
NR	-	Natural Rubber
NBR	-	Nitrile Rubber
PC	-	Polycarbonate
PE	-	Polyethylene
PET	-	Polyethylene Terephthalate
PETG	-	Polyethylene Terephthalate
PLA	-	Polylactic Acid
PP	-	Polypropylene

PP-gMAH	-	Polypropylene with maleic anhydride
PS	-	Polystyrene
PVC	-	Polyvinyl Chloride
RP	-	Rapid Prototype
rPP	-	Recycle Polypropylene
RTR	-	Recycled Tire Rubber
SLS	-	Selective Laser Sintering
SPI	-	Society of the Plastics Industry
NaOH	-	Sodium Hydroxide
SBR	-	Sterene-Butadiene Rubber
SLA	-	Stereolithography
TS	-	Tensile Strength
TGA	-	Thermogravimetric Test
TPU	-	Thermoplastic Polyurethane
TPP	-	Two Photon Polymerization
UV	-	Ultraviolet
VAM	-	Volume Additive Manufacturing



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CHAPTER 1

INTRODUCTION

1.1 Background of Study

In recent years, environmental concerns about plastic waste pollution have prompted a gradual shift from linear to circular economy models, which allow for the production of sustainable plastics with numerous socioeconomic and environmental benefits (Arrigo et al., 2022; Gnanaraj et al., 2018; Mikula et al., 2021). The basic components of this new economic paradigm are the extension of material life and the recovery of materials at the end of their service life. The development of effective recycling strategies for polymer-based materials is critical, because continuous re-use of wastes and scraps should ideally eliminate the use of virgin resources and promote the complete valorization of material wastes (Arrigo et al., 2022).

Three-dimensional (3D) printing is a rapidly evolving technology that can create anything a computer tells it to from raw materials. It has been around for quite some time, but it may still sound futuristic (Iunolainen, 2017). A 3D printer system is one that allows a 3D object to be printed such a way that each portion or object element has a user-defined or user-selected material parameter, such as varying elastic deformation (Materal, 2016; Zhang, 2021). For a 3D printer system to run successfully, it needs filaments to be projected to a desired design. Filaments is a raw material such as plastic or rubber that are already through some treated process to become a string or wired outcome shapes that can be used for a 3D printer. Some of the commonly used filaments is acrylonitrile butadiene styrene (ABS) or polylactic acid (PLA) (Iunolainen, 2017; Materal, 2016; Savu et al., 2019).

So far, in the world of 3D printing there is no problem with existing filaments that are commonly used such as PLA and ABS. However, there are better materials to replace commonly used materials such as polypropylene (PP) and thermoplastic polyurethane (TPU) (Iunolainen, 2017; Materal, 2016). Now we can use plastic and rubber as our raw materials for filament and these materials can be easily obtained while bringing benefits to the world. This is because plastic and rubber can be obtained from recycled waste materials that can be made into filament. Therefore, the selection of this material can solve the main issues of environmental protection and waste management (Arrigo et al., 2022; Materal, 2016).

Besides, the material extrusion 3D printing (ME3DP) based on fused deposition modelling (FDM) is currently thought to be a highly adaptable and efficient additive manufacturing technique for developing components with complex geometries using computer-aided design (Banerjee et al., 2019). The material that are chosen for the experiment that is Polypropylene (PP). Polypropylene is a polymer that is synthesised catalytically from propylene. PP was chosen because its main advantage is its high temperature resistance (Banerjee et al., 2019; Carneiro et al., 2015; Novak & Maddah, 2016b). In addition, because of its low cost, excellent processability, mechanical properties, and recyclability, PP is widely used for a variety of commodity and industrial applications such as trays, bottles, funnels, pails and instrument jars that must be sterilised (cleaned) frequently for use in a clinical setting (Banerjee et al., 2019; Novak & Maddah, 2016b).

Moreover, another material chosen for this experiment is ground tyre rubber (GTR), which is obtained from recycled tyre rubber. However, poor bonding between GTR and most polymer matrices results in low mechanical strength and durability of the resulting compounds, causing a significant challenge and limiting the blends' performance (Fazli & Rodrigue, 2020a). To solve this issue, a variety of techniques, including GTR surface modification, devulcanization, dynamic vulcanization, and the addition of compatibilizing

agents, have been used to improve GTR-matrix interface adhesion, resulting in improved homogeneity and processability, as well as mechanical and long-term (durability) compound properties. As a result, in this study, GTR will have its surface modified using sodium hydroxide (NaOH) solution. As a filler in a polymer matrix, ground tyre rubber (GTR) was used as an ink material (composite material) for material extrusion in a 3D printing process (Alkadi et al., 2019; Fazli & Rodrigue, 2020a). The amount of a filler in a matrix can be proportional to various mechanical properties of composite materials, such as tensile strength, compression strength, and abrasion resistance (Alkadi et al., 2019). As a result, the mechanical properties of the GTR can be controlled to achieve the desired results. GTR was considered because of its positive environmental and economic impacts. The GTR manufacturing process is both environmentally friendly and economically beneficial, as it eliminates some of the processes required to reclaim raw rubber (Alkadi et al., 2019).

By choosing these two materials, many environmental issues can be reduced because the raw materials are obtained from recycled materials. It will also greatly reduce the cost of getting raw materials because they are considered waste for others while the waste can be made into a good product which is 3D printer filament.

1.2 Problem Statement

Plastic and rubber are one of the causes that are polluting the world. Scientists estimate that there are approximately 100 million tonnes of plastic waste floating in the world's oceans, with each piece taking up to 1,000 years to decompose (Iunolainen, 2017). Thus, the issue of plastic and rubber recycling has become one of the leading issues of environmental protection and waste management. By using recycled plastic and rubber for 3D printer filaments, the filaments can overcome the pollution issues that are caused by unwanted plastic and rubber and the waste management issue. It is like giving a second life

for the recycled material to serve the world again. Numerous manufacturers are currently producing reclaimed ABS, PLA, and PET filaments (Arrigo et al., 2022; Iunolainen, 2017; Mikula et al., 2021).

While ABS and PLA are the most commonly used thermoplastics in the 3D printing industry, Polypropylene (PP), despite being one of the most widely used plastics in the world with a wide range of industrial and consumer applications, is only rarely used for 3D printing at the moment. For polypropylene (PP), filament that are made from PP can be considered as flexible filaments would be extremely beneficial, as it will be a long time before we run out of this type of plastic waste. Each modification affects the behaviour of the polymer during the 3D printing process (Iunolainen, 2017; Savu et al., 2019). Therefore, by combining with GTR as a filler for the composite, the properties of GTR might influence the final outcome combined PP and GTR filament.

For ground tire rubber (GTR) composition like the properties for example (particle size), and processing like manufacturing method and pre-treatment of GTR when used as a filler added to a rubber matrix must be considered to obtain the appropriate composite for a given application (Alkadi et al., 2019; Fazli & Rodrigue, 2020a). Therefore, an investigation for different ratios of GTR to polymer matrix have to be conducted to confirm the printability of the ink. On conducting the investigation, there two types of GTR that will be used that is virgin and modified.

So, to modify the mechanical properties of these materials, sodium hydroxide (NaOH) is used as the chemical substance to treat the materials. NaOH is a corrosive white crystalline solid that readily absorbs moisture until it dissolves. The selection of NaOH is to investigate the effect of modification on the mechanical and physical properties of PP and GTR.