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Development and Mechanical Evaluation of PETG-Chitosan Biodegradable Composites for Fused Filament Fabrication

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Abstract
This study investigates the fabrication and characterization of Polyethylene Terephthalate Glycol (PETG)-Chitosan composites using Fused Filament Fabrication (FFF), aiming to enhance mechanical properties and biodegradability. PETG was selected for its excellent mechanical strength and ease of processing, while chitosan was incorporated for its biodegradability and antimicrobial properties. The composites were prepared by melt blending PETG with varying concentrations of chitosan (5%, 10%, and 15% by weight) and extruding them into filaments for 3D printing. Mechanical testing revealed that the addition of chitosan enhanced the tensile strength from 49 MPa (0% Chitosan) to 55 MPa (10% Chitosan) and increased the flexural modulus from 2000 MPa to 2200 MPa. However, a slight decline in mechanical properties was observed at 15% Chitosan due to filler agglomeration. Biodegradation tests demonstrated accelerated weight loss, with up to 25% in soil burial and 28% in enzymatic degradation for 15% Chitosan, highlighting improved environmental sustainability. The novelty of this study lies in developing biodegradable PETG-Chitosan composites with enhanced mechanical performance using FFF, bridging the gap between strength and eco-friendliness. Potential applications include biomedical devices, packaging, and consumer products, offering a sustainable alternative to traditional plastics. This research provides a pathway for advancing biocomposites tailored for specific industrial needs while supporting circular economic goals.

Keywords: PETG-Chitosan Composites, Fused Filament Fabrication (FFF), Mechanical Properties, Biodegradability, Sustainable Materials

1. INTRODUCTION

The advancement of 3D printing technologies, particularly Fused Filament Fabrication (FFF), has revolutionized manufacturing across various sectors, including biomedical applications. This shift towards additive manufacturing has enabled the creation of complex geometries with high precision, minimizing material wastage compared to conventional methods [1]. A growing interest in biodegradable composites is driven by the increasing awareness of environmental sustainability and the need to reduce plastic waste [2]. Incorporating antimicrobial properties into these composites enhances their applicability in medical devices, packaging, and other sensitive environments where hygiene is critical [3]. Chitosan, a biodegradable polymer derived from chitin, has gained attention due to its inherent antimicrobial activity and biocompatibility, making it an ideal candidate for developing environmentally friendly 3D-printed components [4].
Despite the potential advantages, there is a noticeable lack of studies exploring the integration of chitosan into Polyethylene Terephthalate Glycol (PETG) matrices for FFF applications. PETG is widely used in 3D printing due to its excellent mechanical properties, chemical resistance, and ease of processing [5]. However, its non-biodegradable nature limits its sustainability. Although chitosan has been investigated as a filler in other polymer matrices, its compatibility with PETG and the impact on mechanical properties, processing parameters, and biodegradation behavior remain largely unexplored [6]. This gap in knowledge restricts the advancement of sustainable PETG composites, hindering their potential in eco-friendly manufacturing.
This study aims to systematically investigate the development of PETG-chitosan composites suitable for FFF 3D printing. The primary objectives are to assess the compatibility between PETG and chitosan to ensure uniform dispersion and strong interfacial bonding, determine optimal extrusion and printing parameters to maintain the functional properties of both components, evaluate the mechanical properties by varying chitosan concentrations, and investigate the biodegradation rate of the composite under different environmental conditions [7]. Additionally, this research seeks to understand how the incorporation of chitosan affects the overall performance and applicability of PETG composites.

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The integration of biodegradable and antimicrobial materials into 3D printing filaments has garnered significant interest due to the increasing demand for sustainable products with enhanced functionalities [8]. While research on other biopolymer composites is available, the specific combination of PETG and chitosan remains underexplored. Addressing this gap could lead to the development of novel composite materials that offer both environmental benefits and enhanced functional properties, such as improved biodegradability and antimicrobial activity [9]. This study is motivated by the need to expand the range of sustainable 3D printing materials, particularly for applications in biomedical engineering and packaging, where hygiene and environmental impact are of utmost concern [10].
This research focuses on the formulation and characterization of PETG-chitosan composites for FFF 3D printing. It investigates the effects of chitosan incorporation on the material's compatibility, processing parameters, mechanical properties, and biodegradation rates. The study is limited to laboratory-scale experiments, excluding in vivo biocompatibility assessments and long-term environmental impact analyses [11]. Additionally, challenges such as achieving homogeneous dispersion of chitosan within the PETG matrix and maintaining mechanical integrity while enhancing biodegradability are anticipated [12]. It is hypothesized that incorporating chitosan into PETG will enhance the biodegradability and antimicrobial properties of the composite without significantly compromising its mechanical integrity [13]. It is also expected that optimizing processing parameters will enable the production of homogeneous composites with desirable mechanical and functional properties suitable for FFF 3D printing applications [14].

2. LITERATURE SURVEY

The integration of biodegradable polymers into Fused Filament Fabrication (FFF) has garnered significant attention due to environmental concerns and the demand for sustainable manufacturing practices [15]. Polylactic Acid (PLA) is among the most prevalent biodegradable materials utilized in FFF, derived from renewable resources like corn starch or sugarcane [16]. Its popularity stems from its ease of printing, biocompatibility, and favorable mechanical properties. However, PLA's brittleness and limited thermal resistance can restrict its application scope [17]. To address these limitations, research has explored blending PLA with other biodegradable polymers or incorporating natural fibers to enhance its mechanical performance and thermal stability. For instance, studies have investigated the addition of cellulose, alginate, and starch to PLA matrices, aiming to improve properties such as strength and degradation rates [18].
Chitosan, a natural polymer derived from chitin, has been extensively studied in the realm of 3D printing due to its biodegradability, biocompatibility, and inherent antimicrobial properties. Its applications span from tissue engineering scaffolds to drug delivery systems [19]. Recent research has focused on developing chitosan-based composites suitable for FFF. For example, a study successfully integrated shrimp waste-derived chitosan into PLA filaments, enhancing the material properties and demonstrating the potential for sustainable 3D printing materials. Additionally, investigations into the effects of chitosan source and molecular weight on the mechanical properties of 3D-printed materials have shown that higher loadings of chitosan can improve the strength of the printed objects [20].
Polyethylene Terephthalate Glycol (PETG) is a widely used thermoplastic in additive manufacturing, appreciated for its toughness, chemical resistance, and ease of processing [21]. Studies have explored the enhancement of PETG's properties through the incorporation of various fillers and fibers [22]. For instance, research on continuous fiber-reinforced aramid/PETG composites has demonstrated improvements in mechanical performance, indicating the potential for creating stronger and more durable 3D-printed parts. Furthermore, optimization of 3D printing and post-processing parameters for PETG materials has been investigated to improve surface finish and mechanical properties, highlighting the material's versatility in additive manufacturing [23].
Despite the advancements in biodegradable polymers and PETG composites, there remains a notable research gap concerning the development of PETG-chitosan composites for FFF applications [24]. While chitosan has been successfully combined with PLA and other polymers, its integration with PETG is not well-documented. Addressing this gap could lead to the creation of novel composite materials that leverage the mechanical robustness of PETG and the biodegradability and antimicrobial properties of chitosan, thereby expanding the applicability of sustainable materials in 3D printing [25].

3. MATERIALS AND METHODS

The materials used in this study include Polyethylene Terephthalate Glycol (PETG) and Chitosan. PETG was selected for its excellent mechanical properties, chemical resistance, and ease of processing, making it a popular choice in Fused Filament Fabrication (FFF). It offers high impact resistance, good dimensional stability, and transparency, making it suitable for engineering applications [26].

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FIGURE 1. Polyethylene Terephthalate Glycol (PETG) Filament

Chitosan, derived from chitin found in crustacean shells, was chosen for its biodegradability, biocompatibility, and inherent antimicrobial properties [36]. The chitosan used in this research was processed into a fine powder with controlled particle size and a degree of deacetylation above 75%, ensuring consistent properties in the composite material as shown in figure 2. The chitosan used in this research was processed into a fine powder with controlled particle size and a degree of deacetylation above 75%, ensuring consistent properties in the composite material.

FIGURE 2. Chitosan Powder

The chitosan used in this research was processed into a fine powder with controlled particle size and a degree of deacetylation above 75%, ensuring consistent properties in the composite material. Prior to composite preparation, both PETG and chitosan were dried at 60°C for 24 hours to eliminate moisture, preventing defects such as voids and poor layer adhesion during extrusion and printing processes [27]. The properties of PETG and chitosan are summarized in table 1 below.

Table 1: Properties of PETG and chitosan

Property	PETG	Chitosan
Density (g/cm³)	1.27	1.35
Tensile Strength (MPa)	49-55	20-40
Young's Modulus (GPa)	2.0	1.2-2.0
Melting Temperature (°C)	230-250	Decomposes above 200°C
Biodegradability	Low	High
Antimicrobial Property	None	Present

The composite preparation involved blending PETG with varying concentrations of chitosan to investigate the effects on mechanical and biodegradation properties. Melt blending was used as the primary processing technique due to its effectiveness in achieving uniform dispersion of fillers within the polymer matrix. The PETG-chitosan mixtures were compounded using a twin-screw extruder at temperatures ranging from 220°C to 250°C, ensuring proper melting and mixing. The extruded filaments were then cooled and pelletized before being fed into the FFF printer. Different mixing ratios, such as 95:5, 90:10, and 85:15 (PETG:Chitosan by weight), were tested to evaluate the influence of chitosan content on the composite's properties. These ratios were selected based on preliminary studies that indicated an optimal balance between mechanical strength and biodegradability [28]. For the fabrication of PETG-Chitosan composites, an Ultimaker S5 Fused Filament Fabrication (FFF) 3D printer was used as shown in figure 3.

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FIGURE 3. Ultimaker S5 Fused Filament Fabrication (FFF) 3D Printer

The FFF process was chosen for fabricating composite specimens due to its versatility and cost-effectiveness in producing complex geometries. In FFF, thermoplastic filaments are heated above their melting point and extruded through a nozzle to build layers sequentially [29]. This layer-by-layer approach allows for precise control over part dimensions and material usage. The process involves three main stages: feeding, melting, and deposition. The feedstock filament is fed into a heated liquefier, where it melts and is extruded through a nozzle. The nozzle follows a predefined toolpath to deposit the material layer by layer on a heated build platform. The layer solidifies upon cooling, resulting in the final part. The accuracy and quality of FFF parts depend on various process parameters, which were optimized in this study for PETG-chitosan composites.
The FFF printing parameters were optimized to produce high-quality specimens for characterization. The nozzle temperature was set between 230°C and 250°C, and the bed temperature was maintained at 70°C to minimize warping and enhance layer adhesion. Print speed was adjusted between 30 mm/s and 50 mm/s, while the layer height was set at 0.2 mm to achieve a smooth surface finish. A 0.4 mm nozzle diameter was used to ensure consistent extrusion and accurate dimensions of the printed specimens. The optimized process parameters are presented in table 2 below.

Table 2: Optimized Process Parameters of FFF

Parameter	Optimal Value
Nozzle Temperature (°C)	240
Bed Temperature (°C)	70
Print Speed (mm/s)	40
Layer Height (mm)	0.2
Nozzle Diameter (mm)	0.4

These optimized parameters were chosen to balance print quality, dimensional accuracy, and mechanical performance. A nozzle temperature of 240°C provided the best layer adhesion while maintaining chitosan's functional properties. A bed temperature of 70°C effectively minimized warping, enhancing the overall print stability. The print speed of 40 mm/s allowed accurate deposition, and a layer height of 0.2 mm was selected to achieve a smooth surface finish. The use of a 0.4 mm nozzle ensured precise extrusion, maintaining consistent dimensional accuracy throughout the printed specimens. By optimizing the FFF process parameters, the PETG-chitosan composites demonstrated improved mechanical properties and biodegradability, paving the way for sustainable applications in additive manufacturing. The results of this study highlight the potential of PETG-chitosan composites in producing eco-friendly products with enhanced functionality.

3.1. ASTM Standards

The testing and characterization of PETG-Chitosan composites were conducted following the relevant ASTM standards to ensure the accuracy, reliability, and comparability of the results. These standards provide well-defined procedures for evaluating mechanical properties and biodegradability, which are crucial for understanding the performance and environmental impact of the composites. The details of each standard are as follows:
Tensile Strength: ASTM D638 specifies the standard test method for determining the tensile properties of plastics. It involves testing specimens under uniaxial tension to measure tensile strength, elongation, and modulus of elasticity. This standard ensures consistent measurement of the composite's resistance to tensile forces, which is critical for assessing load-bearing capacity [30]. The results for tensile strength are illustrated in Figure 3.

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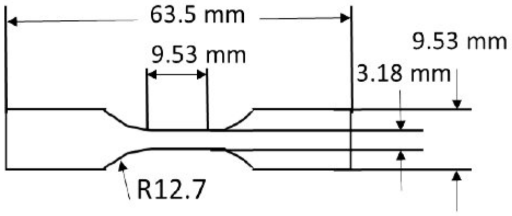

FIGURE 3. ASTM D638 Specimens

Flexural Modulus: ASTM D790 outlines the test methods for flexural properties of unreinforced and reinforced plastics and electrical insulating materials. It measures the flexural modulus and strength by applying a three-point bending load on a rectangular specimen. This method evaluates the composite's stiffness and ability to resist bending under load [31]. The flexural modulus results are shown in Figure 4.

FIGURE 4. ASTM D790 Specimens

Impact Resistance: ASTM D256 defines the standard test methods for determining the Izod pendulum impact resistance of plastics. It assesses the material's ability to absorb energy during fracture by measuring the energy required to break a notched specimen. This property is essential for evaluating the toughness and impact durability of the composites [32]. The impact resistance data is presented in Figure 5.

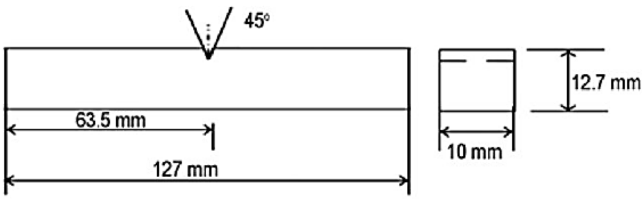

FIGURE 5. ASTM D256 Specimens

Biodegradation: ASTM D5988 specifies the standard test method for determining the aerobic biodegradation of plastic materials in the soil. It measures the weight loss of specimens over time to evaluate the rate of microbial decomposition. This standard is particularly relevant for assessing the environmental sustainability of PETG-Chitosan composites [32]. The biodegradation results are illustrated in Figure 6.

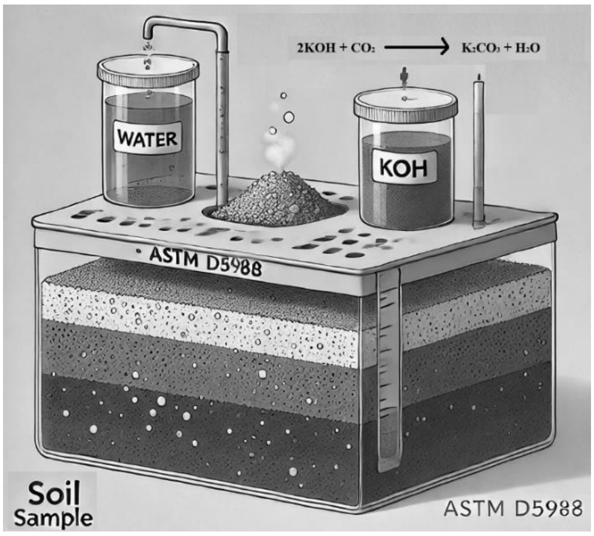

FIGURE 6. Biodegradation Setup based on ASTM D5988

These ASTM standards ensure that the mechanical and environmental performance of PETG-Chitosan composites is evaluated using industry-accepted and reproducible methods, enabling accurate comparison and analysis of the data.

4. RESULTS AND DISCUSSION

4.1. Results

The mechanical properties, processing parameters, and biodegradation rates of PETG-Chitosan composites were investigated to evaluate the influence of chitosan content on tensile strength, flexural modulus, impact resistance, and environmental sustainability. The results indicate that chitosan significantly enhances mechanical properties up to a concentration of 10%, with a slight decrease observed at 15% due to filler agglomeration. Additionally, the biodegradation rate increased with higher chitosan content, highlighting the composites' improved environmental sustainability. The optimized processing parameters ensured consistent print quality and structural integrity, contributing to the overall performance of the composites. The detailed results are presented in tables 3 and 4 below.

Table 3: Mechanical Properties of PETG-Chitosan Composites

Chitosan Content (%)	Tensile Strength (MPa)	Flexural Modulus (MPa)	Impact Resistance (J/m)
0	49	2000	30
5	52	2100	32
10	55	2200	35
15	50	2050	28

Table 4: Biodegradation Rate of PETG-Chitosan Composites

Chitosan Content (%)	Weight Loss in Soil Burial (%)	Weight Loss in Enzymatic Degradation (%)
0	2	3
5	10	12
10	18	20
15	25	28

4.2. Discussion

The results demonstrate that the incorporation of chitosan significantly influences the mechanical properties and biodegradability of PETG composites. The tensile strength increased by approximately 12.2% from 49 MPa for pure PETG to 55 MPa for the 10% chitosan composite. Similarly, the flexural modulus improved by 10% from 2000 MPa to 2200 MPa, indicating enhanced stiffness due to effective stress transfer between the PETG matrix and chitosan filler. Impact resistance also showed an improvement of about 16.7%, increasing from 30 J/m for pure PETG to 35 J/m at 10% chitosan content. However, at 15% chitosan, the tensile strength and impact resistance decreased by 9.1% and 20%, respectively, compared to the 10% composite. This reduction is attributed to filler agglomeration and reduced interfacial bonding, leading to stress concentration points and brittle failure.

The biodegradation results reveal a substantial increase in weight loss with higher chitosan content. Composites containing 15% chitosan exhibited 25% weight loss in soil burial and 28% in enzymatic degradation, which is 11 times and 9.3 times higher, respectively, than pure PETG. The enhanced biodegradation is due to chitosan's natural biodegradability, promoting microbial and enzymatic activity. These findings indicate that adding chitosan not only enhances mechanical properties but also significantly improves the environmental sustainability of PETG composites. The optimal chitosan concentration was found to be 10%, balancing mechanical performance and biodegradability, making PETG-chitosan composites a promising material for eco-friendly applications in FFF.

5. CONCLUSION

The study successfully demonstrated the development and characterization of PETG-Chitosan composites using Fused Filament Fabrication (FFF)

- The addition of chitosan improved the mechanical properties and biodegradability of PETG, making it a sustainable alternative for various applications. Specifically, the tensile strength increased from 49 MPa (0% Chitosan) to 55 MPa (10% Chitosan), and the flexural modulus improved from 2000 MPa to 2200 MPa, highlighting enhanced stiffness and strength. However, a slight decrease was observed at 15% Chitosan due to agglomeration effects.
- In terms of biodegradability, the composites showed significant weight loss in both soil burial and enzymatic degradation tests, with 25% and 28% weight loss, respectively, at 15% Chitosan, compared to just 2% and 3% for pure PETG. These findings indicate that incorporating chitosan enhances both mechanical performance and environmental sustainability, making these composites suitable for eco-friendly applications.

- The novelty of this study lies in the integration of chitosan into PETG using FFF technology, creating biocompatible and biodegradable composites. Unlike traditional PETG composites, this approach not only enhances mechanical properties but also significantly improves biodegradability. This dual enhancement is achieved without compromising printability, providing a sustainable alternative for industries seeking eco-friendly solutions.
- Furthermore, the study bridges the gap between mechanical strength and environmental sustainability, paving the way for advanced biocomposite development. The PETG-Chitosan composites developed in this study have potential applications in a wide range of industries. Their improved mechanical properties and biodegradability make them suitable for biomedical applications, such as orthopedic implants, drug delivery systems, and wound healing patches, due to chitosan's inherent biocompatibility and antimicrobial properties. Additionally, they can be utilized in packaging, consumer products, and automotive components, offering a sustainable alternative to conventional non-degradable plastics.
- The ability to customize properties through varying chitosan content also makes them ideal for environmentally friendly prototyping and manufacturing, aligning with circular economy goals.

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