Development and Optimization of Polypropylene-Chitosan Composites with Conductive Fillers for Improved Biodegradability

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Abstract

Addressing the environmental concerns associated with non-biodegradable plastics, this study focuses on the development of biodegradable polypropylene-based composites using chitosan nanoparticles and conductive fillers. Initially, five samples of polypropylene-chitosan composites were prepared: a control sample of neat polypropylene (AO) and four samples containing varying concentrations of chitosan nanoparticles—10%, 20%, 30%, and 40% (A1, A2, A3, and A4, respectively). A 90-day soil biodegradation test was conducted to determine the optimal chitosan content for biodegradability. The sample containing 30% chitosan nanoparticles (A3) exhibited the highest degradation rate, leading to its selection for further experimentation. Subsequent tests examined the effect of conductive fillers—activated carbon and graphene—on the biodegradability of the optimized A3 sample. Nine new composites were formulated by adding activated carbon and graphene at concentrations of 5%, 10%, 15%, and 20%. Results showed that the incorporation of graphene significantly enhanced biodegradability, with the sample containing 20% graphene (C4) achieving the greatest weight reduction, indicating a near-complete breakdown by the end of the 90-day period. While activated carbon also improved degradation, its impact was less pronounced compared to graphene, particularly at higher concentrations. These findings suggest that the combination of 30% chitosan nanoparticles with 20% graphene offers an optimized biodegradable composite, balancing environmental degradation with material stability. This study concludes that

graphene, particularly at higher concentrations, is a promising additive for optimizing the biodegradability of polypropylene composites, offering a pathway to environmentally friendly alternatives to conventional plastics.

Keywords: *Biodegradable composites; Polypropylene-chitosan blend; Activated Carbon; Conductive fillers in polymers; Plastic wastes.*

1.0 INTRODUCTION

The environmental burden of plastic waste has become a significant global concern, with conventional plastics contributing to soil and water pollution due to their long degradation times [1, 12, 31, 6]. Polypropylene, a widely used polymer due to its durability and versatility, is particularly problematic because of its resistance to biodegradation, leading to accumulation in landfills and natural habitats [4, 38, 15]. As a result, there has been growing interest in developing biodegradable alternatives or modifications to existing polymers that can reduce their environmental footprint without compromising functionality [17, 29].

One approach to enhancing the biodegradability of polypropylene involves blending it with biodegradable additives, such as chitosan, a natural polysaccharide derived from chitin. Chitosan has been shown to promote microbial colonization and facilitate the breakdown of synthetic polymers when incorporated into composite materials [28, 2]. Its biodegradability and compatibility with other polymers make it a promising candidate for the development of eco-friendly composites [21, 14, 27, 9].

Another strategy involves the use of conductive fillers like activated carbon and graphene to modify the polymer matrix. Conductive fillers are not only capable of enhancing the mechanical and electrical properties of composites, but they also influence the material's interaction with environmental factors, such as moisture and microbial communities [17, 19, 4]. Studies suggest that conductive fillers can increase the surface area available for microbial colonization and may facilitate electron transfer, potentially accelerating degradation rates [5, 16].

Among conductive fillers, activated carbon has been widely studied for its role in enhancing polymer degradation due to its high surface area and ability to adsorb moisture, promoting microbial activity [32, 3, 8]. However, recent research has highlighted the potential of graphene—a two-dimensional nanomaterial with exceptional conductivity and surface properties—as a superior filler for biodegradable composites. Graphene's ability to form conductive networks within the polymer matrix can increase the rate of microbial degradation, particularly in soil environments [35, 24].

Despite the promising results associated with conductive fillers, questions remain regarding the optimal type and concentration required to maximize biodegradability while preserving the desired material properties [18]. This study aims to address these gaps by investigating the biodegradability of polypropylene-based composites modified with chitosan nanoparticles and varying concentrations of either activated carbon or graphene. The research seeks to identify an

optimal filler configuration that balances biodegradability with mechanical performance, contributing to the development of sustainable materials that mitigate the environmental impact of conventional plastics.

2.0 MATERIALS AND METHOD

2.1 Sample collection

A commercially available plastic wastes, a derivative of polypropylene was used in this study. The research objectives narrowed the preliminary resin selection to commodity polymer, which are commonly available, and have excellent application to the fuel cell environment. Fillers from chitosan nanoparticle, derived from chitin, were used in conjunction with polypropylene matrix to form electrically conductive bio composites. The choice of fillers for this research is based on their ability to impart high conductivity to the composites, effective biodegradability while still maintaining a relatively low cost and high availability.

Polypropylene waste used in this study was obtained from Federal University of Technology, Akure (FUTA) community. It was washed thoroughly and shredded to pellets (sizes 1.69mm and 3.21mm) at ZL Alliance Global, Alagbaka Akure, Nigeria. Banana pseudo stem waste was obtained from a banana farm located at FUTA South gate Akure, Nigeria. Chitosan was obtained from a sea food company in Lagos, Nigeria with average size of 80 mm and degree of DE acetylation (DD) of 81%.

2.2 Sample Preparation

2.2.1 Preparation of Nano chitosan

Nano Chitosan CNP was prepared using Daramola *et al* [10] modified method. 30g of Chitosan powder was dissolved in 2liters of 2% acetic acid. It was stirred for 24h at 60^0C and TPP solution was prepared by dissolving 73.4g of Sodium Tri-Poly-Phosphate (TPP) in 2liters distilled water. Chitosan solution was then added to Sodium Tri-Poly-Phosphate (TPP) solution in drops to give a final ratio of 1:1 (CS:TPP). The resultant sample (Chitosan Nanoparticle CNP) was then filtered and washed severally with distilled water which was made to stand for a day before been filtered with a sintered glass. Obtained samples were then oven-dried for 48hrs and grinded to powder.

2.2.2 Composite Fabrication

The prepared nano chitosan was mixed with the polypropylene pellets in varying proportions (A0, A1, A2, A3, A4) to create different composite formulations; where A0 (PP/unfilled) was used as the CONTROL 1. The optimized sample of PP/CNP was labeled X, which serve as CONTROL 2, a constant used for X/Activated carbon (AC) and X/Graphene (Gr) bio composite blend. X/AC and X/Gr bio composites were prepared by blending the optimized sample (X) obtained above with 5%, 10%, 15% and 20% of each of the conductive fillers (Gr and AC).

For the blending, solvent casting method was employed. Polypropylene (PP) was dissolved in 0.067g/ml Xylene at 164oC using a magnetic stirrer hot plate. The mixing was performed using a mechanical stirrer to ensure uniform dispersion of the chitosan nanoparticles within the

polypropylene matrix. The mixing time was standardized to 30 minutes to achieve a consistent blend. Fillers (CNP) were added to the dissolved PP. It was allowed to cool down and was poured into a mold. The resulted blend; bio composite was sundried to drain the organic solvent left in the sample.

Table 2.1: Blend formulation for PP/CNP, PP/CNP/AC and PP/CNP/Gr composite sample

Designation	Sample	Designation	Sample	Designation	Sample
A0	PP	X	PP/CNPopt	X	PP/CNPopt
A1	PP/CNP10%	B1	X/AC5%	C1	X/Gr5%
A2	PP/CNP20%	B2	X/AC10%	C2	X/Gr10%
A3	PP/CNP30%	B3	X/AC15%	C3	X/Gr15%
A4	PP/CNP40%	B4	X/AC20%	C4	X/Gr20%

2.3 Biodegradability Assessment of the composites

The PP/CNP bio composite samples obtained were evaluated for its biodegradability using soil burial, a modified method in Adelaja and Babaniyi [17]. The biodegradability of the composites were evaluated by measuring the percentage weight loss over time. 3kg of the soil sample was used for the soil degradation test. PP/CNP bio composites were weighed and buried into already filled pots with soil 2cm beneath the surface under laboratory conditions. The soil was regularly irrigated with clean water to maintain a stable humidity. After a predetermined degradation time (30days, 60days and 90days), the sample were removed from the soil, cleaned with water and dried. The initial mass (Mi) and final mass (Mf) of the samples were recorded, and the percentage mass loss was calculated using the formula in equation 1,

$$\%\, Mass\ loss\ = \frac{Mi-Mf}{Mi} \times 100 \qquad [1]$$

Where Mi – initial mass and Mf - final mass

3.0 RESULTS AND DISCUSSION

3.1 Influence of Chitosan nanoparticle on Polypropylene

The soil biodegradability test is a critical assessment method used to evaluate the degradation behavior of materials in soil environments. This test is particularly relevant for polypropylene-chitosan nanoparticle (PP-CNP) composites, as it provides insights into their environmental impact and potential for sustainable applications. The results of the soil biodegradability test, presented in Table 3.1 and 3.2, indicate a significant weight loss for the PP-CNP composite samples over a 90-day period. The weight loss is a crucial parameter for assessing biodegradability, as it reflects the extent of material breakdown in a specific environment.

In the first phase, five samples with varying concentrations of chitosan (10% to 40%) were evaluated, and the result is presented in table 3.1. The neat polypropylene sample (A0) exhibited the least weight loss, retaining a final weight of 2.61 g after 30 days, 2.40 g after 60 days, and 1.84

g after 90 days. This minimal degradation can be attributed to the hydrophobic nature of polypropylene, which limits its susceptibility to microbial attack and environmental degradation [20]. The results suggest that without the incorporation of biodegradable materials, polypropylene retains its mass more effectively, leading to slower degradation rates.

Table 3.1: Soil biodegradability result of the PP-CNP composite samples.

Sample		**Final weight after being buried (WA)**		
	Initial Weight (WB)	**30days**	**60days**	**90days**
A0	3g	2.61	2.4	1.84
A1	3g	2.54	1.74	1.09
A2	3g	2.44	1.68	1.02
A3	3g	2.42	1.69	1.04
A4	3g	2.31	1.64	1.01

In contrast, the composite samples containing chitosan nanoparticles demonstrated a more pronounced weight loss, indicating enhanced biodegradability. For instance, Sample A1, which contains 10% chitosan nanoparticles, showed a final weight of 2.54 g after 30 days, decreasing to 1.74 g after 60 days, and further to 1.09 g after 90 days. This trend aligns with the findings of Gopi *et al*. [13], who reported that the incorporation of biodegradable materials such as chitosan into polymer matrices significantly enhances the overall biodegradability of the composites. Chitosan, a natural polymer derived from chitin, is known for its biocompatibility and biodegradability, making it an ideal candidate for improving the environmental performance of synthetic polymers.

The enhanced biodegradability observed in the PP-CNP composites can be attributed to several factors. Firstly, the presence of chitosan nanoparticles facilitates microbial colonization and enzymatic degradation, leading to a more rapid breakdown of the composite material in soil environments [31]. Chitosan's hydrophilic nature also allows for better water absorption, which is crucial for microbial activity, thereby accelerating the degradation process.

The results for Samples A2, A3, and A4, which contain increasing percentages of chitosan nanoparticles, further support the conclusion that higher chitosan content correlates with improved biodegradability. For example, Sample A2, with 20% chitosan, exhibited a final weight of 2.44 g after 30 days, decreasing to 1.68 g after 60 days, and 1.02 g after 90 days. This pattern suggests that the enhanced hydrophilicity and porous structure of chitosan nanoparticles promote greater microbial activity and moisture retention, thereby facilitating the degradation process [34]. The increasing weight loss trend with higher chitosan nanoparticle content indicates that chitosan plays a significant role in promoting the degradation of the polypropylene-chitosan composites in the soil environment.

Moreover, the biodegradation process is influenced by various environmental factors, including soil composition, temperature, moisture content, and the presence of microorganisms. The soil used in this test was assessed for its composition and microbial profile, which are critical for understanding the degradation behavior of the composites. The microbial activity present in the soil interacts with the chitosan component of the composites, leading to enhanced degradation

rates. Chitosan, being a natural polysaccharide, is known to be susceptible to microbial degradation, as highlighted by Silva *et al*. [31]. The presence of specific microbial strains capable of degrading chitosan can further enhance the biodegradation process, underscoring the importance of soil biodiversity in the degradation of composite materials.

The weight loss observed in the composite samples can also be attributed to the physical and chemical interactions between the chitosan nanoparticles and the polypropylene matrix. The incorporation of chitosan may disrupt the crystalline structure of polypropylene, making it more accessible to microbial attack and enzymatic degradation [11]. This disruption can lead to an increase in the surface area available for microbial colonization, further enhancing the biodegradation process.

Consequently, the soil biodegradability test results demonstrate that the PP-CNP composites exhibit varying degrees of biodegradability, with the presence of chitosan nanoparticles significantly enhancing the degradation process, with the sample containing 30% chitosan (A3) showing the highest degradation rate. This sample was selected as the optimal formulation for further testing. The findings underscore the potential of utilizing biodegradable materials in composite formulations to mitigate environmental impact and promote sustainability in material design. By adjusting the chitosan nanoparticle content, it is possible to tailor the composite's biodegradability to meet specific environmental considerations, paving the way for the development of more sustainable materials in various applications.

3.2 Influence of the carbon fillers on the PP-CNP Biocomposite

In the second phase, the soil biodegradability test results for the nine samples indicate varying levels of biodegradation over 90 days, influenced by both the type and concentration of conductive fillers incorporated into the polypropylene and chitosan nanoparticle matrix. The result is presented in Table 3.2. The primary component of these samples is polypropylene (PP), which is inherently resistant to biodegradation. However, the addition of chitosan nanoparticles and conductive fillers (activated carbon and graphene) appears to impact the rate of biodegradation.

Sample AX, which contained only polypropylene and chitosan nanoparticles without any conductive filler, served as the control to benchmark biodegradation. The reduction in weight from 3.00 g initially to 1.04 g after 90 days indicates a moderate level of biodegradability.

Table 3.1: Soil biodegradability test results for the PP-CNP Composites

Sample	Initial Weight (WBB) (g)	Weight after 30 days (WA 30 days) (g)	Weight after 60 days (WA 60 days) (g)	Weight after 90 days (WA 90 days) (g)
AX	3	2.42	1.69	1.04
B1	3	2.26	1.51	0.87
B2	3	2.29	1.44	0.81
B3	3	2.16	1.46	0.88
B4	3	2.02	1.04	0.79
C1	3	2.21	1.13	0.21

C2	3	2.05	1.21	0.16
C3	3	2.04	1.07	0.11
C4	3	2.01	1.1	0.07

This result aligns with the well-established understanding that polypropylene, a synthetic polymer, is inherently resistant to biodegradation due to its high molecular weight and crystalline structure (Xu and Wang, 2018). However, the presence of chitosan nanoparticles, a biodegradable biopolymer, likely facilitated some microbial interaction and degradation, though not to the same extent as the samples with conductive fillers [19].

The inclusion of activated carbon as a conductive filler in samples B1 to B4 significantly influenced biodegradability. As the filler concentration increased from 5% to 20%, the final weights of the samples decreased progressively from 0.87 g (B1) to 0.79 g (B4). This trend suggests that higher concentrations of activated carbon improve microbial activity by providing additional surface area and sites for microbial colonization, thereby enhancing degradation rates [22]. Notably, B4 (20% activated carbon) exhibited the highest degradation among the activated carbon-containing samples, suggesting that a higher percentage of this filler optimizes microbial interaction without excessively reinforcing the composite structure, which could impede degradation [37].

However, while the degradation was significant with activated carbon, the rate of weight loss plateaued between the 60 and 90-day intervals, suggesting that a saturation point may be reached with activated carbon concentrations beyond 15-20%. This plateau could indicate that the activated carbon was not entirely accessible to microbes, or that the microbial population stabilized after depleting easily degradable materials [23].

Samples C1 through C4, incorporating graphene as a conductive filler at similar concentrations (5% to 20%), showed a more pronounced degradation than the activated carbon samples. The final weights for these composites decreased markedly, from 0.21 g (C1) to 0.07 g (C4), demonstrating a nearly complete degradation of the composite material by the end of the 90-day period. Graphene's effect on biodegradability appears to be multifaceted: its large surface area, combined with its conductive properties, may facilitate electron transfer, boosting microbial metabolism and enhancing the breakdown of the composite matrix [16]. This effect becomes particularly evident in C4, where the highest graphene concentration of 20% resulted in the lowest final weight, signifying optimal biodegradability among all samples tested.

The superior performance of graphene over activated carbon as a filler could be attributed to its nanoscale dimensions, which allow for better integration within the polymer matrix, leading to a less rigid structure that is more susceptible to microbial attack. Additionally, graphene's interaction with environmental factors such as moisture and soil acidity might further enhance its biodegradability potential compared to activated carbon [25]. However, one downside of high graphene concentrations, as observed in the samples, is the potential for agglomeration, which could inhibit uniform microbial colonization if not adequately dispersed during composite formulation [26].

Comparatively, the results indicate that graphene is a more effective conductive filler for enhancing biodegradability than activated carbon. Among the tested samples, C4 (20% graphene) demonstrated the highest biodegradation rate, with only 0.07 g remaining after 90 days. This substantial weight loss suggests that graphene not only contributes to the disintegration of the polymer matrix but also does so more efficiently than activated carbon, likely due to better electron conductivity and integration with the polymer components.

For optimization, C4 stands out as the most promising sample, achieving the highest degree of biodegradability without compromising the structural integrity needed for practical applications. This observation is critical because an ideal biodegradable composite should balance sufficient stability during its intended use and rapid breakdown in soil environments post-disposal [33]. While B4 (20% activated carbon) showed effective degradation, it did not reach the same level of material breakdown as C4, indicating that graphene fillers are preferable for applications requiring optimized biodegradability.

4.0 Conclusion

This study demonstrates that the biodegradability of polypropylene-based composites is significantly influenced by the type and concentration of conductive fillers. This study highlights the potential of polypropylene-chitosan composites as environmentally friendly alternatives to conventional plastics. The research was conducted in two phases: an initial optimization of chitosan content in polypropylene, followed by an examination of the effects of conductive fillers—activated carbon and graphene—on the biodegradability of the optimized composite. In the first phase, five samples with varying concentrations of chitosan (10% to 40%) were evaluated, with the sample containing 30% chitosan (A3) showing the highest degradation rate. This sample was selected as the optimal formulation for further testing. In the second phase, the optimized A3 sample was modified with conductive fillers, specifically activated carbon and graphene at concentrations of 5%, 10%, 15%, and 20%, to assess their influence on biodegradability.

Over a 90-day period, graphene-filled composites (C1 to C4) consistently exhibited superior degradation compared to those containing activated carbon (B1 to B4). The highest degree of biodegradation was achieved by sample C4, containing 20% graphene, which resulted in a final weight of 0.07 g, highlighting graphene's effectiveness in enhancing microbial breakdown due to its conductive and nanoscale properties. In contrast, the control sample (AX) without conductive fillers showed the least biodegradation, underscoring polypropylene's inherent resistance to microbial activity. The results demonstrated that the addition of graphene significantly enhanced the biodegradation of the composite, particularly at higher concentrations. The sample with 20% graphene (C4) exhibited the greatest weight reduction after 90 days, indicating a near-complete breakdown. In contrast, although activated carbon improved biodegradation, its effectiveness plateaued at higher concentrations, suggesting that graphene's unique conductive and structural properties facilitate microbial activity more effectively.

These findings underscore the critical role of both chitosan and graphene in optimizing biodegradable composites. The combination of 30% chitosan and 20% graphene offers a promising

formulation, achieving a balance between initial material stability and accelerated environmental degradation. The study confirms that modifying polypropylene with biodegradable additives and conductive fillers can significantly enhance its decomposition, making it a viable solution to reduce plastic waste.

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