

Synthesis and Characterization of Bio-Based Polymer from Arrowroot Peels (*Maranta arundinacea*) and Plantain Peels (*Musa Paradisiaca* L.)

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Abstract

This study investigates the development of bio-based polymers derived from plantain peels (*Musa paradisiaca* L.) (PPB) and arrowroot peels (*Maranta arundinacea*) (APB), as well as their copolymers (COP), as sustainable alternatives to non-degradable synthetic polymers. Biodegradable polymers offer unique physical, chemical, biological, biomechanical, and degradative properties, making them highly relevant for environmentally friendly applications. In this work, PPB, APB, and COP were synthesized and characterized through physico-chemical analysis, including moisture content determination, soil burial degradability tests, and various instrumental techniques: X-ray Diffraction (XRD), Fourier Transform Infrared (FTIR), Scanning Electron Microscopy (SEM) and Thermogravimetric Analysis (TGA). By varying glycerol content across samples (20, 15, 10, and 5 cm³), a reduction in moisture content was observed, with values ranging from 35.70% to 20.13%. A 30-day soil burial test revealed significant weight loss for PPB (100%), with moderate degradation for APB (2.17%) and COP (1.51%). XRD analysis indicated an amorphous phase across all samples, while FTIR spectra confirmed characteristic functional groups (OH, C-H, C=O, C=C, -CH₃, and C-O) consistent with successful polymer formation. TGA results showed that thermal stability decreased with glycerol content in the order of APB > PPB > COP. SEM images of samples with 5 cm³ glycerol displayed voids and cracks in APB and PPB, whereas COP exhibited a smoother and more uniform surface, depicting enhanced interfacial interaction and compatibility. These findings demonstrate that bio-copolymers COP offer increased moisture absorption and superior surface characteristics and also enhance biodegradability, making it promising candidate for eco-friendly applications in industries where sustainable and degradable materials are required.

Keywords: Arrowroot Peels, Plantain Peels, Biopolymer, Glycerol and Starch.

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INTRODUCTION

Synthetic petroleum-based polymers are essential in industries like packaging but pose significant environmental challenges due to their non-biodegradable nature and resistance to microbial degradation (Verma *et al.*, 2023). This has led to the accumulation of plastic waste and difficulties in waste management. In response, bio-based polymers have gained attention as sustainable alternatives to mitigate environmental pollution (Rizvi, 2024). Among these, polymers derived from natural sources, such as starch, offer promising solutions for replacing conventional plastics (Leja and Lewandowicz, 2010).

Polymers are large molecules composed of repeating monomer units linked by covalent bonds. Their classification into natural and synthetic types highlights their diverse properties and applications. Natural polymers, like proteins and polysaccharides, are abundant and biodegradable, whereas synthetic polymers, such as polyethylene and polystyrene, are engineered for durability but often lack biodegradability, contributing to environmental concerns (Verma *et al.*, 2023). Key properties, such as strength, flexibility, and thermal stability, depend on structural characteristics (e.g., linear, branched, or cross linked) (Molyneux, 2018). Innovations in polymer science now focus on biodegradable polymers like polylactic acid (PLA) and polyhydroxyalkanoates (PHA) to reduce plastic waste. However, challenges in mechanical performance and production costs persist, driving research into recycling and advanced bio-based alternatives (Samir *et al.*, 2022).

Biopolymers, produced by living organisms or synthesized from renewable resources, are eco-friendly materials increasingly used in packaging, agriculture, and biomedicine. Their biocompatibility and biodegradability make them a priority for sustainable applications (Kumari *et al.*, 2022). Common biopolymer categories include: Polysaccharides (e.g., cellulose, starch); widely used for packaging and medical applications (Kumari *et al.*, 2022), Proteins (e.g., collagen, gelatin); renowned for strength and flexibility, mainly applied in biomedicine (Liu, *et al.*, 2023), and PHA and PLA; microbially derived polymers with growing use in packaging and healthcare (Stublić, *et al.*, 2024). Despite their advantages, biopolymers face challenges such as limited mechanical strength, water sensitivity, and high production costs. Efforts to address these limitations involve polymer blending, additives, and advanced modification techniques (Patel & Sharma, 2023).

Starch, a carbohydrate polysaccharide found in plants like maize, cassava, and wheat, is an abundant and renewable biopolymer. Composed of amylose (linear) and amylopectin (branched) as shown in figure 1, its structure influences properties like crystallinity and strength, essential to starch's applications (Dong *et al.*, 2024). Starch biopolymers are characterized by their ability to form glycosidic bonds through polymerization, where glucose monomers connect via dehydration synthesis to form long chains. During processing, heat and plasticizers (such as glycerol) disrupt hydrogen bonds within starch granules, converting starch into thermoplastic starch (TPS), a moldable material with polymer-like behavior. This disruption enables amylose and amylopectin chains to reorganize, creating flexible and durable structures suitable for various applications (Diyana *et al.*, 2021).

These biopolymers find applications in: packaging for making biodegradable films and bags (Dirpan *et al.*, 2023); agriculture in controlled-release fertilizers and mulch films (Salimi *et al.*, 2024); biomedical for wound dressings, drug delivery, and scaffolds (Mirani, *et al.*, 2023); and food where it is used as stabilizers, thickeners, and emulsifiers (Quezada, *et al.*, 2024).

There are 3 methods of synthesis of starch biopolymer which includes: physical modification where thermoplastic starch (TPS) is produced by gelatinization with heat and plasticizers (Cai *et al.*, 2024), chemical modification in which esterification enhance properties such as biodegradability and thermal resistance (Liu *et al.*, 2023), and polymer blending where combining starch with other polymers such as PLA, improves mechanical properties (Jayarathna, *et al.*, 2022).

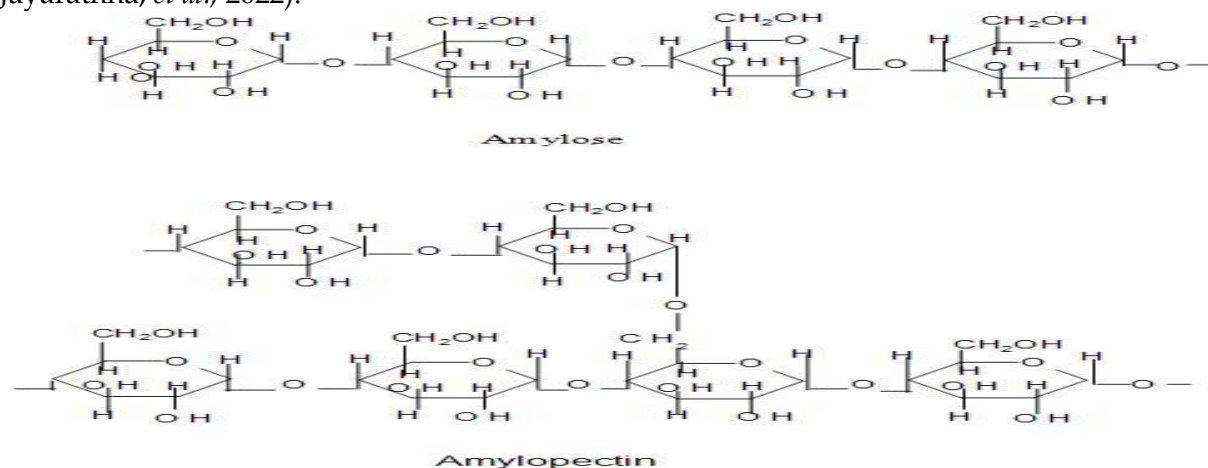


Figure 1: Structure of Amylose and Amylopectin (Otache, *et al.*, 2021).

Blends and composites, as discussed by Jayarathna *et al.*, (2022), employ strategies to enhance mechanical properties and water resistance. Hydrogels and aerogels, highlighted by Zhao *et al.*, (2024), find applications in active food packaging due to their high porosity, which facilitates substance delivery. Additionally, antimicrobial films that combine starch with essential oils, as explored by Zhao *et al.*, (2024), effectively improve food preservation.

This study aim to synthesize (through gelatinization and plasticization) and characterize starch-based biopolymers and co-polymers using arrowroot and plantain peels. The research introduces a comprehensive evaluation the effect of varying glycerol concentrations on physicochemical, thermal and biodegradability properties of the polymer. Unlike previous studies, this research emphasizes the synergistic blending of starches from two distinct sources, offering a novel approach to enhancing mechanical properties and environmental degradation rates. Additionally, it highlights the integration of waste reduction strategies with sustainable biopolymer production, aiming to reduce reliance on petroleum-based plastics while contributing to circular economy practices. Advanced characterization techniques (e.g. FTIR, SEM, TGA, and XRD) were employed to establish a detailed structure-property relationship, positioning the work as a significant advancement in bio-co-polymer technology for sustainable materials.

METHODOLOGY

Detection of presence of starch

Chemical Test for Starch was carried out by adding potassium iodide (KI) solution directly onto the AS and PS (Pesek *et al.*, 2022).

Sample Preparation of AS and PS

Arrowroot Starch (AS): Peeled, sliced, dried at 80 °C, pulverized, and sieved to 250 μm (Singh and Mohanty, 2007) and Plantain Starch (PS): Treated with sodium meta-bisulphite, boiled, air-dried then blended into paste (Sultan and Johari, 2017).

Biopolymer Production

APB were synthesized by heating starch with glycerol, water, and HCl at 100 °C, followed by molding and oven drying at 80 °C for 2 days (Singh and Mohanty, 2007). Similar procedure was used for the PPB and COP where the samples were mixed with glycerol, HCl and NaOH solution, followed by molding and it was allowed to oven dry at 80 °C for 3days (Sultan and Johari, 2017).

Characterizations

Soil Burial Test was performed according to ASTM G 160-12; samples were buried at a 15 cm depth, unburied after one month, cleaned, and weighed to calculate weight loss as a measure of biodegradability (Ashok *et al.*, 2018). SEM (PHENOM PRO X) at magnifications of 8000x–10000x was used to examine surface morphology (Yaradoddi *et al.*, 2022). Moisture uptake was evaluated using ASTM D5229, with weight changes recorded over a month. FTIR (Agilent Cary 630) identified functional groups in the biopolymers, operating in the 4000–650 cm⁻¹ range (Sultan and Johari, 2017). XRD analysis determined phase identification of the samples. TGA measured thermal stability across 0–900 °C in a nitrogen environment (Aksakal *et al.*, 2021).

RESULTS AND DISCUSSION

Moisture Absorption

Table 1 shows the results obtained from the moisture absorption of the two samples biopolymer (APB and PPB) and copolymer (COP) using varying amounts of glycerol. It can be inferred that biopolymers synthesized with 20 cm³ of glycerol absorbed more moisture to those containing 5, 10, and 15 cm³ of glycerol. This trend indicates that moisture absorption increases with higher glycerol concentrations in the biopolymers. The findings are consistent with FTIR analysis, where higher moisture absorption corresponds to a more intense O-H stretching band, reflecting the hydrophilic nature of both glycerol and starch. These hydrophilic properties enhance the interaction between glycerol and moisture, leading to increased absorption (Abidin *et al.*, 2021). Additionally, PPB demonstrated higher moisture absorption than APB and COP across all glycerol concentrations (5, 10, 15, and 20 cm³).

Table 1.0: Percentage moisture absorption at different glycerol concentrations

Samples + volume of glycerol	Number of days													
	2	4	6	8	10	12	14	16	18	20	22	24	26	28
APB 5 cm ³	16.14	18.50	20.08	20.08	20.08	20.47	20.87	20.87	21.27	21.27	21.27	21.65	21.65	21.65
APB 10 cm ³	21.74	23.73	26.76	27.09	27.42	27.42	27.76	27.76	28.09	28.09	28.43	28.78	28.76	28.76
APB 15 cm ³	24.89	28.44	30.66	30.66	30.66	30.66	30.66	30.22	30.22	30.22	29.78	29.78	29.78	29.78
APB 20 cm ³	30.00	31.90	32.86	32.91	33.24	33.30	33.38	33.46	33.55	33.67	34.87	33.94	33.94	33.94
PPB 5 cm ³	12.66	12.99	13.46	12.64	10.79	10.53	-	-	-	-	-	-	-	-
PPB 10 cm ³	12.26	18.64	20.51	21.01	21.66	22.79	-	-	-	-	-	-	-	-
PPB 15 cm ³	24.86	28.45	27.62	25.97	26.55	26.55	-	-	-	-	-	-	-	-
PPB 20 cm ³	32.61	35.87	33.69	33.69	34.78	35.87	-	-	-	-	-	-	-	-
COP 5 cm ³	22.26	25.53	25.37	22.26	21.11	18.99	17.68	17.84	18.17	18.49	19.15	19.80	20.13	20.13
COP 10 cm ³	21.12	26.62	28.17	24.08	23.24	22.80	22.68	22.68	23.24	23.52	23.66	23.94	24.23	24.36
COP 15 cm ³	20.12	25.58	28.71	30.27	30.27	29.88	30.47	30.66	30.66	31.05	31.45	31.45	31.84	32.31
COP 20 cm ³	17.16	23.27	25.05	25.84	27.02	28.01	28.59	28.79	30.18	31.95	33.73	35.05	35.70	35.70

FTIR Analysis

The FTIR analysis revealed the presence of key functional groups in both AS and PS, including hydroxyl (O-H), alkyl (C-H), carbonyl (C=O), aromatic (C=C), methyl (-CH₃), and ether (C-O) groups, as shown in Figure 2 (a-h), corresponding characteristic wave-numbers around 3339.7–3297.6 cm⁻¹, 2944.6–2922 cm⁻¹, 1636.3–1625.1 cm⁻¹, 1438.8–1364.2 cm⁻¹, and 1155.5–1148.0 cm⁻¹ respectively. These functional groups confirm the presence of hydroxyl groups, hemi-cellulose, cellulose, and non-cellulosic components such as lignin and pectin in the raw materials before plasticization. This finding aligns with the observations reported by Abidin *et al.*, (2021).

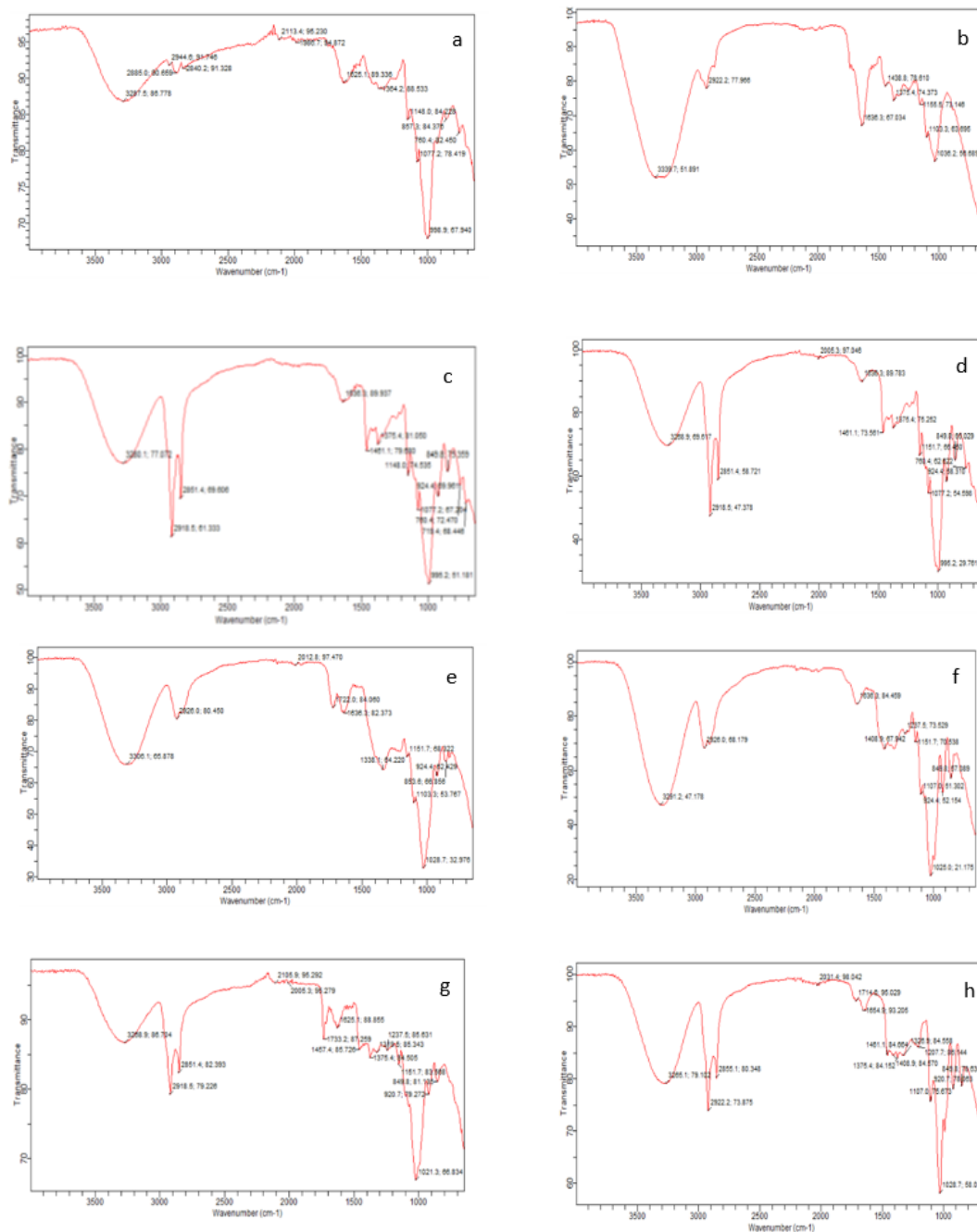


Figure 2: FTIR analysis of; AS and PS (a, b), APB 5 and 20 cm³ (c, d), PPB 5 and 20 cm³ (e, f), COP 5 and 20 cm³ (g, h)

Post-plasticization analysis (figure 2, c-h) showed that PPB exhibited a higher intensity of hydroxyl groups compared to APB and COP. This is evidenced by broader and more prominent O-H stretching peaks at 3306.1 cm^{-1} for PPB, compared to 3280.1 cm^{-1} for APB and 3291.2 cm^{-1} for COP. The increase in hydroxyl group density is significant as it enhances the hydrophilic nature of the material, leading to greater moisture absorption. Furthermore, the abundance of hydroxyl groups facilitates biodegradation, as these groups serve as active sites for microbial attack, accelerating the breakdown of the polymer matrix. This correlation between hydroxyl group intensity and functional properties emphasizes the role of chemical structure in determining the materials environmental and performance attributes.

Scanning Electron Microscopy

SEM micrographs revealed notable structural differences across the samples as depicted in Figure 3. In APB and PPB with 5 cm^3 glycerol (Figure 3 a and c respectively), voids and cracks were observed. These defects are attributed to the lower glycerol concentration, which limits the effective plasticization of the starch, resulting in less cohesive polymer structures (Boonsuk *et al.*, 2020). Conversely, samples with 20 cm^3 glycerol exhibited fewer defects due to enhanced plasticization, which improved the material's structural integrity.

For COP 20 cm^3 , the presence of voids was linked to the inclusion of arrowroot starch (AS) in the bio-co-polymers. AS, being more brittle than plantain starch (PS), compromises the structural uniformity of the material (Cano *et al.*, 2015). However, the smooth surface observed in COP 5 cm^3 indicates improved compatibility and better interfacial interactions between the polymer components. This enhanced compatibility makes COP a more viable option compared to single bio-polymers, particularly for applications like packaging, where surface uniformity and structural stability are critical.

De Almeida *et al.*, (2021) similarly reported that higher glycerol concentrations in polymer blends, such as those using arrowroot starch and polyvinyl alcohol, promote homogeneous morphologies with low surface roughness. Their findings align with this study, emphasizing the role of glycerol in enhancing compatibility and reducing defects.

The SEM observations align with results from other analytical techniques. The amorphous morphology identified in XRD analysis for higher glycerol concentrations reflects increased amorphousness, which corresponds to the defect-free surfaces in SEM. Additionally; TGA results confirm that this reduction in crystallinity due to glycerol decreases the material's thermal stability. This trade-off is indicative of increased flexibility and susceptibility to degradation at elevated temperatures, reinforcing the multifunctional role of glycerol in modifying the material's properties.

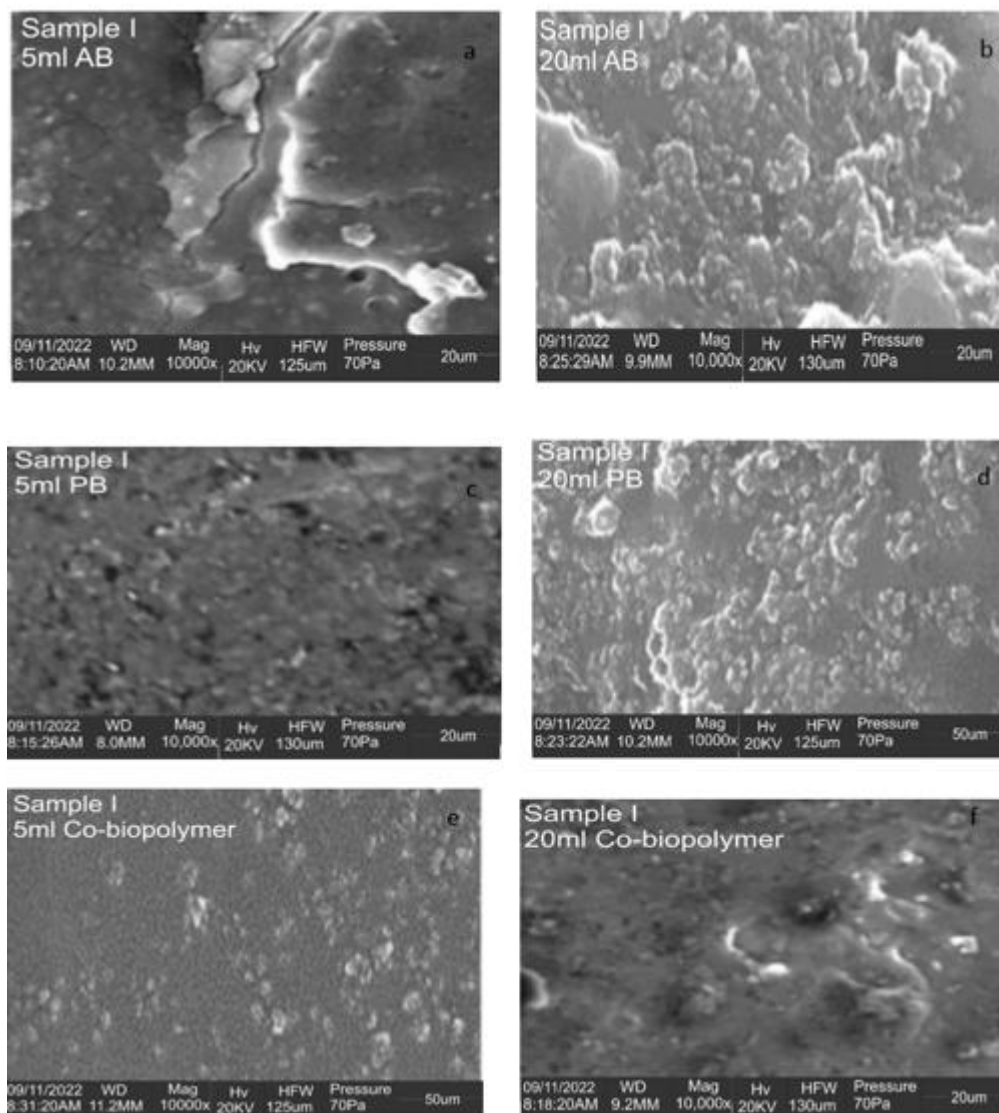


Figure 3: SEM micrograph of APB 5 and 20 cm³ (a, b), PPB 5 and 20 cm³ (c, d), COP 5 and 20 cm³ (e, f).

Thermogravimetric Analysis (TGA)

Thermogravimetric analysis of the biopolymer samples revealed three distinct weight-loss stages: drying, pyrolysis, and decarbonization. These stages provide an understanding into the thermal behavior of the samples and the influence of glycerol concentration on their stability.

1. Drying Stage (27–200°C)

The initial weight loss in this region is due to the evaporation of physically and chemically bound water, reflecting the hydrophilic nature of the biopolymers (Valencia *et al.*, 2012). The extent of weight loss varied among the samples. APB (5% weight loss): This relatively low loss suggests moderate moisture content and hydrophilicity. PPB (7% weight loss): Higher moisture loss compared to APB indicates a greater affinity for water due to increased hydroxyl groups. COP (15% weight loss): The highest loss reflects the combined hydrophilicity of AS and PS in the copolymer, further enhanced by the glycerol's hygroscopic nature. For samples with higher glycerol concentrations (20 cm³), the weight loss in this stage was consistently greater than in samples with lower concentrations (5 cm³).

Glycerol enhances hydrophilicity, thereby binding more moisture, which evaporates during heating (Valencia *et al.*, 2012).

2. Pyrolysis Stage (200–500°C for APB and COP, 250–500°C for PPB).

This stage is characterized by significant thermal degradation due to the breakdown of polymer backbones, such as starch, cellulose, and other non-cellulosic components. APB: Degradation commenced at 200°C, indicating lower thermal stability. This is due to fewer hydroxyl groups available for cross-linking, leading to earlier decomposition. PPB: Decomposition started at a higher temperature (250°C), suggesting better structural integrity and thermal resistance, likely due to stronger polymer-glycerol interactions. COP: Decomposition started at 200°C but exhibited a broader range, signifying the heterogeneous nature of the copolymer. The presence of AS contributes to its brittleness, while the glycerol improves flexibility, balancing its thermal behavior. Higher glycerol concentrations in all samples resulted in increased weight loss during this stage. Glycerol acts as a plasticizer, disrupting the crystalline regions of the polymers, thereby reducing their thermal stability and increasing degradation.

3. Decarbonization Stage (500–900°C)

In this final stage, residual char decomposes, marking the material's carbonization:

The APB, PPB, and COP samples exhibited similar trends in this stage, with the primary difference being the total residual char. Lower glycerol concentrations yielded more residual char due to reduced polymer breakdown during earlier stages. For higher glycerol concentrations, the samples exhibited less residual char, reflecting more extensive thermal degradation in earlier stages.

The TGA results point out the significant role of glycerol concentration in influencing the thermal behavior of the biopolymers: Low glycerol concentrations (5 cm³): These samples exhibited less weight loss across all stages, indicating higher crystallinity and reduced thermal degradation. However, they were more brittle, as evidenced by lower compatibility and greater defects observed in SEM. High glycerol concentrations (20 cm³): These samples displayed higher weight loss, reduced thermal stability, and lower char residues. The increased plasticization effect of glycerol improved flexibility but compromised thermal resistance, as corroborated by the amorphous morphology in XRD analysis and smoother surfaces in SEM.

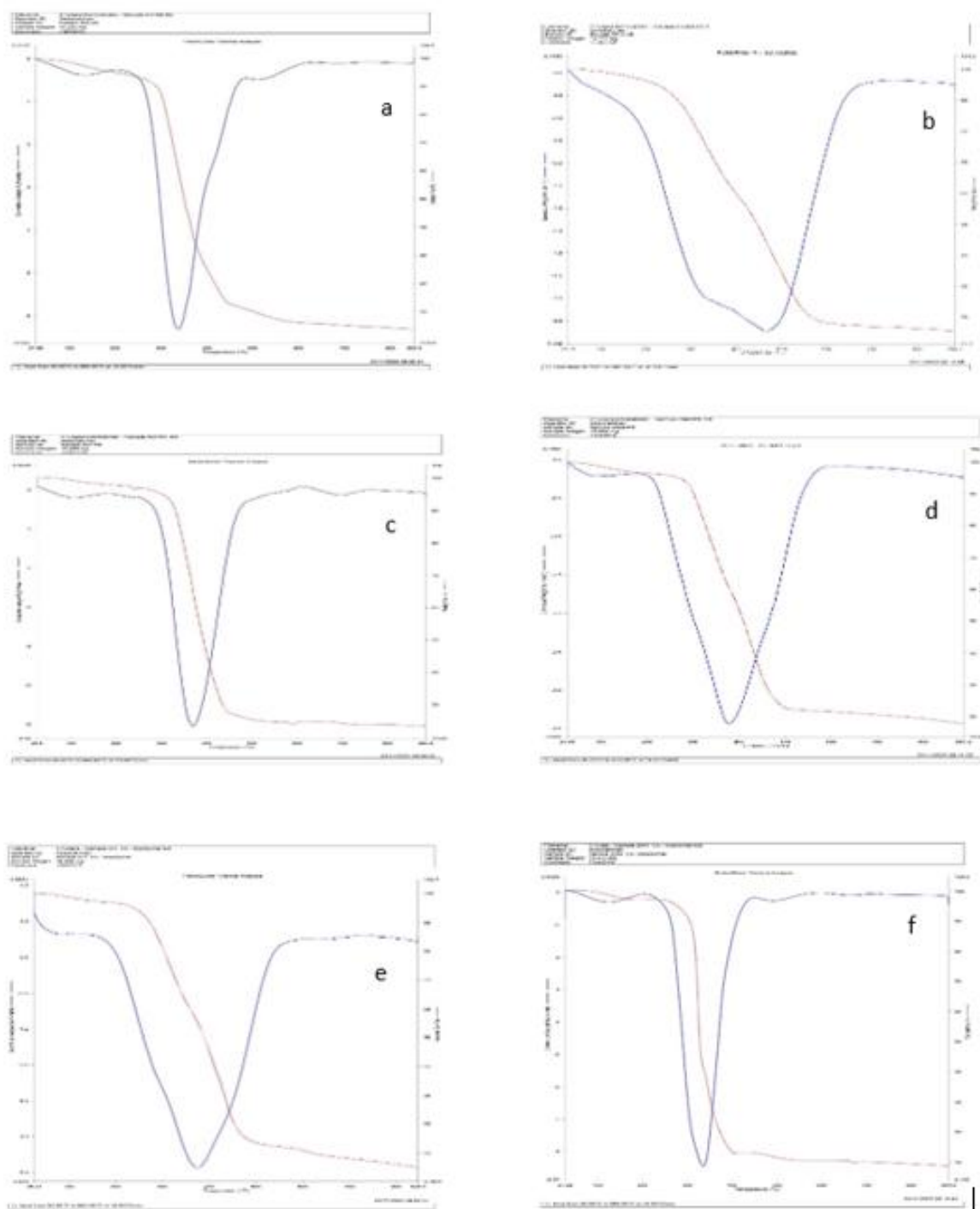


Figure 4: Thermograph of the surface of APB 5 and 20 cm³ (a and b), PPB 5 and 20 cm³ (c and d), COP 5 and 20 cm³ (e and f).

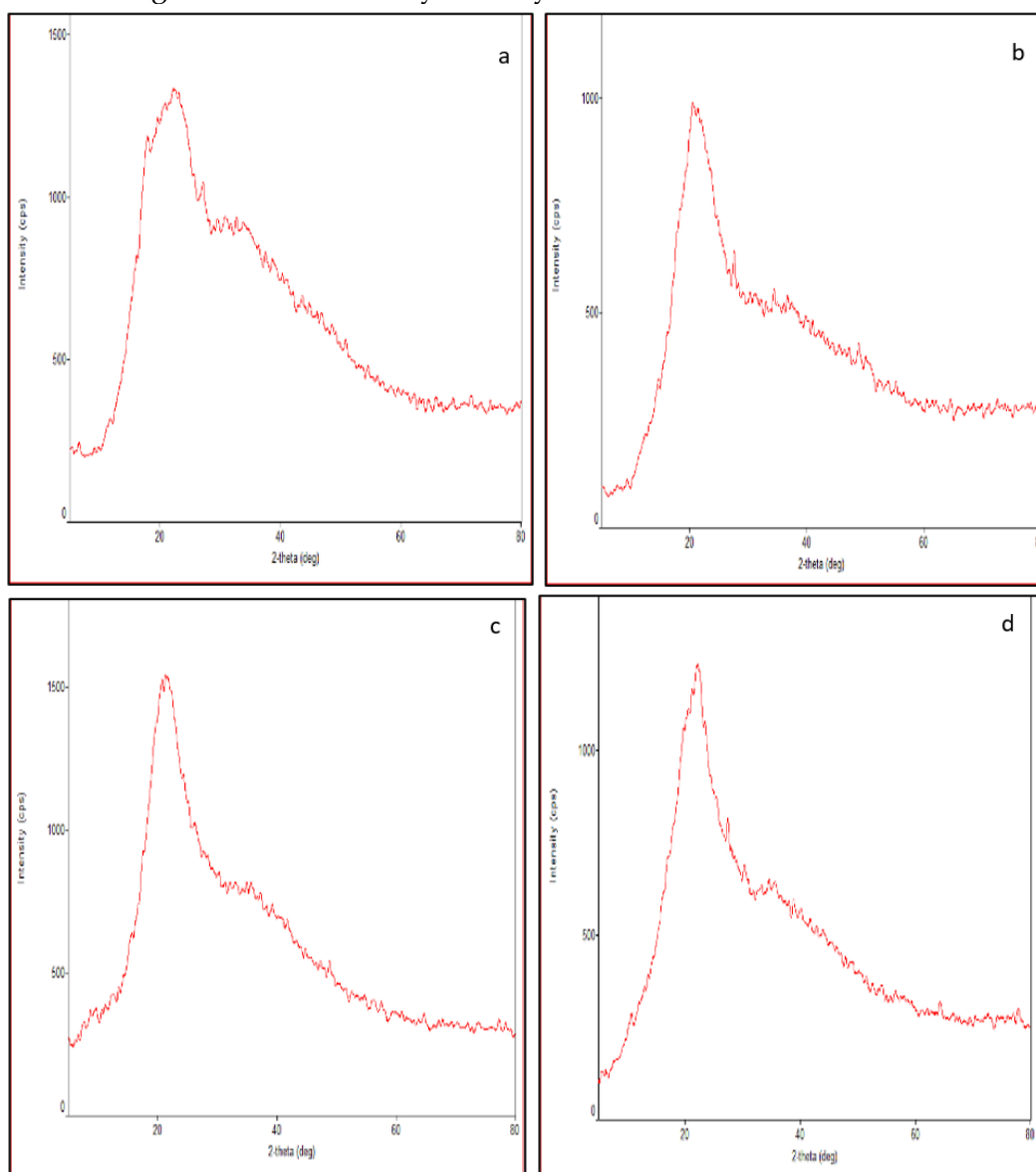
X-Ray Diffraction (XRD) Analysis

The XRD patterns of APB, PPB, and COP samples indicate predominantly amorphous structures, characterized by broad, low-intensity peaks. This amorphization is attributed to glycerol's role as a plasticizer, which disrupts starch crystallinity by reducing intermolecular hydrogen bonding during processes like gelatinization and film casting (De Almeida *et al.*, 2021). All samples with low glycerol content (5 cm³) depict broad peaks with slightly higher intensity suggesting residual semi-crystalline regions, as lower glycerol levels fail to completely disrupt starch crystallinity. On the other hand, in samples with high glycerol

content (20 cm³), the peaks become significantly broader and less intense, indicating enhanced amorphization as higher glycerol content increases molecular disorder (Jiang *et al.*, 2016; Yan *et al.*, 2015).

PPB exhibits a slightly more amorphous structure, possibly due to differences in matrix composition. While COP displays the most amorphous nature, particularly with 20 cm³ glycerol, likely due to the combined polymer matrix enhancing glycerol's impact on structural disruption.

According to the literature, increased amorphization enhances material flexibility making it ideal for packaging. Improved homogeneity may reduce permeability which correlates with TGA findings, where reduced crystallinity leads to lower thermal resistance.



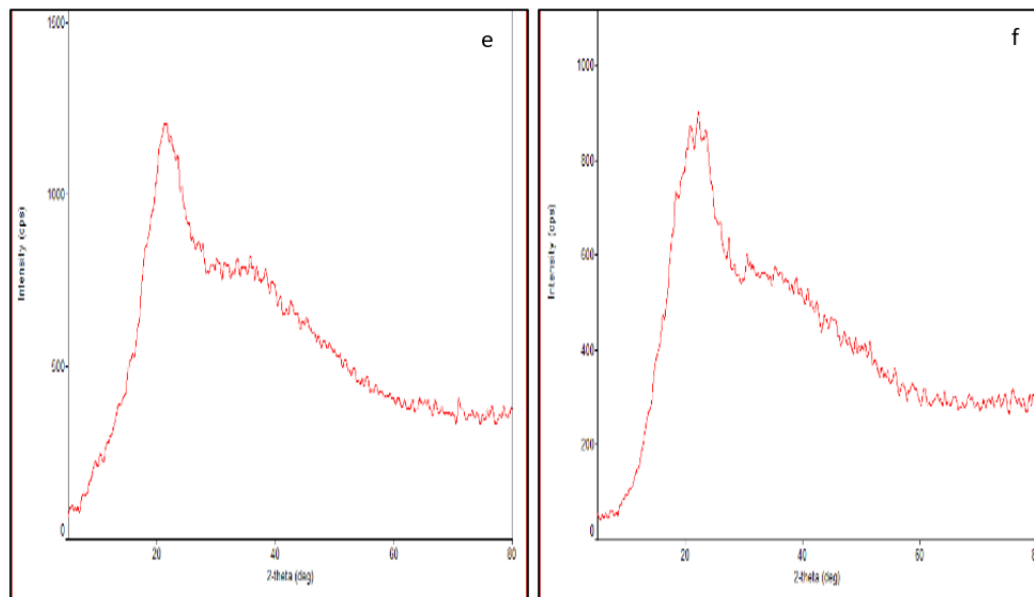


Figure 5: XRD pattern of the surface of APB 5 and 20 cm³ (a, b), PPB 5 and 20 cm³ (c, d), COP 5 and 20 cm³ (e, f).

Polymer Degradation by Soil Burial Test

From the result obtained (Table 2), 30-day soil burial test revealed significant weight loss for PPB (100%), with moderate degradation for APB (2.17%) and COP (1.51%). It is shown that PPB loses more weight than APB and COP. This degradation property is due to high moisture absorption associated with PS making it more favorable for microorganism to act on the material which in turn causes degradation. Moisture is the critical factor influencing degradation by making the material susceptible to fungal attack (Stark and Mutuana, 2021; Verhey, *et al.*, 2001). PPB showed moisture absorption of 35.87%. This is the reason for higher degradation observed in PPB when compared with APB and COP with moisture absorption of 33.94% and 35.70% respectively.

Table 2: Soil burial degradation of APB, PPB and CO
Detection of Presence of Starch

S/N	Sample	Initial weight (g)	Final weight (g)	Degradation (%)
1	APB 5 cm ³	3.07	3.05	0.65
2	APB 10 cm ³	2.37	2.35	0.84
3	APB 15 cm ³	3.11	3.07	1.29
4	APB 20 cm ³	3.69	3.61	2.17
5	PPB 5 cm ³	0.87	-	100
6	PPB 10 cm ³	2.88	-	100
7	PPB 15 cm ³	3.79	0.30	92.08
8	PPB 20 cm ³	2.14	0.35	83.65
9	COP 5 cm ³	4.66	4.51	3.22
10	COP 10 cm ³	5.90	5.80	1.69
11	COP 15 cm ³	7.20	7.15	0.69
12	COP 20 cm ³	4.64	4.57	1.51

Chemical Test for Starch: potassium iodide (KI) solution was added directly onto the AS and PS and a black color was observed as seen on Figure 6 below, which indicates the presence of starch in both the AS and PS (Pesek *et al.*, 2022).



Figure 6: Starch test on AS and PS

Moisture absorption increases with glycerol concentration; PPB absorbs the most due to higher hydroxyl group density. FTIR confirms functional groups responsible for hydrophilicity and biodegradability; PPB shows the strongest O-H peak. SEM shows structural integrity differences; higher glycerol enhances plasticization and reduces defects. TGA reveals thermal instability due to amorphization at higher glycerol levels. XRD shows that higher glycerol disrupts crystallinity, enhancing flexibility but reducing thermal resistance. And Soil degradation shows that PPB degrades faster due its hydrophilic nature, while APB and COP degrades slower due to lower moisture absorption. These findings underscore the multifunctional role of glycerol and the inherent differences in starch compositions, supporting their potential applications in flexible, biodegradable materials.

CONCLUSION

In this study, we successfully synthesized and characterized bio-based polymers from arrowroot and plantain peels using varying glycerol concentrations (5-20 cm³). Higher glycerol levels increased moisture absorption (up to 35.87%) due to the hydrophilic nature of glycerol and starch, enhancing biodegradability but reducing thermal stability. Plantain bio-polymers exhibited superior biodegradability (100% wt loss in 30 days), attributed to higher moisture absorption facilitating microbial activity. FTIR, TGA, XRD, and SEM analysis confirmed the successful polymer formation, amorphous structures, and the influence of glycerol on material properties. Compared to synthetic polymers, these bio-polymers offer significant environmental benefits, including rapid biodegradability, but are limited in moisture-sensitive and high- temperature applications. Potential uses include biodegradable packaging, agricultural mulch films and compostable food containers.

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