ADDIS ABABA UNIVERSITY

COLLEGE OF TECHNOLOGY AND BUILT ENIVRONMENT

SCHOOL OF CHEMICAL AND BIO ENGINEERING

Process Optimization for the Production of Bio-composite Using Waste Plastic (HDPE) and Coffee Waste (Silver Skin)

A Thesis submitted in Partial Fulfillment of the Requirements for the Degree of Master of Science in Chemical Engineering (Environmental Engineering Stream)

By

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Approval page

This is to certify that the thesis prepared by Ms. Redeat Habtamu, entitled "**Process Optimization for the Production of Bio-composite using Waste Plastic (HDPE) and Coffee Waste (Silver Skin)**," has been submitted as a partial fulfillment of the requirements for the degree of Master of Science in Chemical Engineering (Environmental Engineering Stream). It complies with the regulations of the university and meets the accepted standards with respect to content, quality, and originality.

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Declaration

I declare that this thesis, entitled "**Process Optimization for the Production of Bio-composite using Waste Plastic (HDPE) and Coffee Waste (Silver Skin)**," is the outcome of my own research work towards my MSc degree. It has not been previously submitted, either in whole or in part, to any other institution for any degree or qualification. All source of information used have been properly acknowledge through appropriate referencing.

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Abstract

This study investigated the production and optimization of the recycled High-Density Polyethylene (HDPE) and coffee silver skin (CSS) Bio-composites as a sustainable approach for valorizing plastic and agricultural wastes. Coffee silver skin (CSS), the main by-product obtained after coffee beans roasting and has no commercial value, can be potentially used as filler to develop a sustainable Bio-composite using recycled High-Density Polyethylene (rHDPE) as polymer matrix. Chemical treatment of CSS significantly improved cellulose content, structural order, thermal stability, and surface morphology, providing a high-quality reinforcement material. In this research work, CSS was grounded and fractionated in to three different sizes (100µm, 300µm, and 500µm) with varying filler ratio (10, 20, and 30wt.%), the Composites were fabricated through compounding and consolidation processes. Fabrication process was optimized for tensile strength of the resulted composite, achieving a maximum of 5.99MPa, under conditions of a particle size of 285.82µm, 10.44% of filler ratio, and a temperature of 175.97℃. Under this optimized processing conditions composite from treated CSS achieved 6.71MPa of tensile strength. Thermal, morphological and physicochemical properties of the bio-composite also investigated. The results show that the fabricated composite own suitable for replacing the conventional building material by sustainable Bio-composite through waste valorization align with the principles of circular economy.

Key words: CSS, Sustainable Bio-composite, Circular Economy, HDPE, Optimization, Tensile Strength

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List of Abbreviations and Acronyms

Abbreviation and Acronyms	Full Term
ANOVA	Analysis of Variance
BBD	Box-Behnken Design
CSS	Coffee silver Skin
CP	Composite
DTG	Derivative Thermogravimetric
GHG	Greenhouse Gas
HDPE	High-Density Polyethylene
rHDPE	Recycled High-Density Polyethylene
ISO	International Organization for Standardization
MAPE	Maleic Anhydride grafted Polyethylene
NFRP	Natural Fiber Reinforced Plastic
NFPCs	Natural Fiber Polymer Composites
PBAT/PHBV	Poly (butylene adipate-co-terephthalate) / Poly (3- hydroxyl butyrate -co- hydroxyvalerate
PLA/PBS	Poly Lactic Acid) / Poly Butylene Succinate
PE	Poly Ethylene
PSW	Plastic Solid Waste
RSM	Response Surface Methodology
SDGs	Sustainable Development Goals
WPC	Wood Plastic Composite

1. Introduction

1.1. Background

In recent years, environmental concerns has arisen, this fact together with the overall depletion of fossil fuels, recycling and waste upgrading etc. act as leading force for the development of new and an alternative material, which are based on fossil fuel those materials made from renewable resources are promising. This situation has been particularly marked in the field of polymers and polymer-based composites; research has been focused on the use of low environmental impact polymer matrices and reinforcing fibers (Dominici et al., 2019; Carbonell-verdú et al., 2015; Teuber et al., 2015). A key concern the use of fossil fuel-based plastics that are persistent and non-degradable in the environment, it remains for a long time, only break down into smaller and smaller pieces as a result, they accumulate rather than decompose, which cause pollution on natural environment (Geyer et al., 2017 ; Ghazvini et al., 2022).

After stagnation in 2020 due to the Covid-19 pandemic, the global plastics production increased to 430.9 million tonnes in 2024. 89.8% of the world plastics production was fossil based. Post-consumer mechanically recycled plastics, chemically recycled and bio-based/bio attributed plastics respectively accounted for 9.5%, 0.1 and 0.6% of the world plastics production; HDPE, accounting for 12.5% from the total production of plastics in 2024, (*Plastics the Fast Facts 2025*). However, with the one-time-use attitude towards plastic items in conjunction with the current levels of plastic production and low recovery rate (Esterhuizen & Kim, 2022). Countries around the world are struggling to manage current volumes of plastic waste and plastic pollution. However, the petrochemical industry announced over $204 billion U.S. in investment, leading to a projected acceleration in virgin plastic production. In 2016, 19 to 23 million metric tons, or 11%, of plastic waste generated globally and entered in aquatic ecosystems. Considering the ambitious commitments currently set by governments, annual emissions may reach up to 53 million metric tons per year by 2030 (Borrelle et al., 2020).

Plastics are mainly of two types, thermosetting and thermoplastics: thermosetting plastics or engineered polymers, like polyurethanes, phenolic resins, and acrylonitrile butadiene styrene (Kibria et al., 2023). Thermoplastics are a family of plastics that can be melted when heated and hardened when cooled. The most common Thermoplastics are: Polyethylene Terephthalate (PET), Polypropylene (PE), Low-Density Polyethylene (LDPE), High-Density Polyethylene (HDPE), Polystyrene (PS), Expanded polystyrene (EPS), Polyvinyl-chloride (PVC), Polycarbonate, Polypropylene (PP); Polylactic acid (PLA) and Polyhydroxyalkanoates (PHA). High-Density Polyethylene (HDPE): Quite special compared to the other types, HDPE has long virtually unbranched polymer chains which makes them really dense and thus, stronger and thicker than PET. HDPE is commonly used as the grocery bag, opaque milk bottles, juice containers, shampoo bottles, detergent bottles, oil jerry cans, toys and medicine bottles (Singh & Singh, 2020).

It is important to note that thermoplastics contribute to the total plastic consumption by about 80%, and are used for typical plastics applications such as packaging (Gwada et al., 2019). According to World Bank group report, plastics comprise about 5-12% of the world's total waste generation. Africa contributes share of 17.2% with the Middle East and North Africa accounting for 8.3% and 8.9% for sub-Saharan Africa (SSA). SSA is also reported to have 80-90% of plastic waste being inadequately disposed (Ncube et al., 2021; Gwada et al., 2019). In today's world, 'clean-up' of those plastics waste by recycling then managed by authorities or organizations has become a challenging phenomenon to a global extent (Kibria et al., 2023). While a small percentage is recycled, only 9% and 5% of waste plastics were recycled in Canada and USA, respectively, 12% was incinerated, and 79% of plastics waste being sent to landfills, detrimental effects include the long degradation times and the release of toxins into the environment (zian zhang, 2023). Mechanical recycling is one of the most commonly used approaches to tackle the plastics residues problem. The challenges of the polymer recycling industry are related to difficulty to predict and obtain consistent, uniform and stable mechanical properties depending on the source/origin of the material and the recycling process used (Ncube et al., 2021). Incineration, widely adopted since the late 1990s, releases carcinogenic substances and other pollutants into the atmosphere while generating large amounts of carbon dioxide and GHG emissions, thereby increasing environmental burden and offering limited treatment efficiency (Gandhi et al., 2021).

These challenges require sustainable waste valorization strategies as alternatives to incineration and landfilling, which generate secondary pollutants and waste valuable resources. Likewise, chemical recycling methods such as pyrolysis and de-polymerization remain limited as optimal environmental solutions due to their high investment (Ayana, Ha, et al., 2024). These have prompted exploring alternatives like thermo mechanical recycling and valorizing them to more sustainable, which can reduce post-consumer waste before considering incineration or landfilling (Sommerhuber et al., 2015). Therefore, the producers, policy makers and researchers are now under pressure to take responsibility and find solutions for this challenge; this is why several investigations focused on recycling polymers/plastics to convert the conventional linear economy of the plastics industry into a circular one (Aurrekoetxea et al., 2001; Sommerhuber et al., 2015; Lorenzo et al., 2017; Vidakis & Petousis, 2021).

Simultaneously, the coffee industry is one of the industries with most waste and by-product generation, since coffee is the third most consumed drink after water and tea (Dominici et al., 2019) and the world's second most valuable traded commodity, behind only petroleum (Blinová et al., 2017). The annual production of coffee waste is projected to surpass 23 million tons (Petaloti & Achilias, 2024). More than 90 % of the coffee cherries biomass ends up as waste during coffee processing (Assefa et al., 2025), depending upon the method of coffee cherries processing, roasting and brewing solid residues like pulp husk, silver skin and spent are obtained during roasted coffee manufacturing, over 50 wt. % of the fresh coffee cherry is discarded and treated as waste (Murthy & Madhava Naidu, 2012).

Coffee silver skin (CSS) is papery skin, a thin tegument contained in the external layer of the green coffee beans, and represents, approximately 4.2 wt. % of the beans. CSS is the main by-product obtained after coffee beans roasting and has no commercial value (Dominici et al., 2019). For every ton of roasted coffee beans, an average of 8 kg of silver skin is generated (Assefa et al., 2025). As result, substantial quantities of coffee silver skin waste are produced annually. However, it usually finds its end-of-life in controlled landfills or it is subjected to incineration which is a source of contamination due to its high organic content and presence of chemical compounds such as caffeine, tannins and polyphenols (Janissen & Huynh, 2018). Despite this, a potential alternative to minimize this huge amount of CSS waste is its use as filler/reinforcement in composite materials (Dominici et al., 2019).

It could be a useful by-product for industrial applications because, unlike coffee pulp or used coffee grounds, it has low moisture content and no perishable characteristics (Hejna, 2021; Gigante et al., 2020). The growing body of research on the utilization of natural plants and agricultural waste products is aimed at addressing the dual objectives of waste reduction and the discovery of novel carbon resources (Petaloti & Achilias, 2024). Utilizing coffee silver skin as a raw material in bio-composite production not only address the waste problem from the coffee industry but also create a more sustainable circular economy where waste is repurposed in to valuable material.

Natural fiber filled polymer composites are denoted as green-composites or Bio-composite. Bio-composites are materials, made by incorporating natural fibers as reinforcement into a polymer matrix, using low-value agricultural waste for developing polymer composites provides economic and environmental advantages (Kuram, 2022). The simultaneously enhances the recycling economy prior to energy recovery, reduces the environmental carbon footprint, and linear economy of single use thermoplastics reinforce with lignocellulosic like natural fiber: the high molding capability, thermal process ability, and moisture resistance of plastics demonstrate superior good balance on mechanical properties, low density, and cost reduction making them suitable for various applications such as building, aerospace, automotive, packaging sectors show a clear interest on this material (Dominici et al., 2019 ; Ayana, Ha, et al., 2024).

However, the primary limitation is their incompatibility between hydrophobic thermoplastics and polar hydrophilic natural fibrous particle, due to the high density of hydroxyl and carbonyl groups in cellulose, hemicellulose, and lignin (Ayana, Ha, et al., 2024). Addressing this challenge through chemical treatments to the fillers to remove non cellulosic components of the filler and modify the fiber surface, thereby improving surface roughness and enhancing interfacial bonding with the polymer matrix, is major focuses of the current research.

As per the literature, only a few articles have been reported on using CSS as filler in developing bio-composites (Totaro et al., 2019), investigated the effect of filler loading on the thermo mechanical properties of CSS filler-reinforced PLA/PBS-based particulate composites. (Hejna et al., 2021) studied how the processing, physic-mechanical and thermal performances of polyethylene-based composites were affected by the incorporation of CSS at different loading.

Most reported studies have focused primarily on biodegradable polymer matrices such as Polylactic acid (PLA) and poly butylene succinate (PBS), mainly packaging applications. However, investigations on the incorporation of CSS into conventional thermoplastic matrices, particularly high-density polyethylene (HDPE) remain scarce, since filler size plays a crucial role in defining the compatibility between fillers and polymers, but sufficient literature on the effect of filler size on the properties of polyethylene-based composite is not available, as well as addressing interfacial compatibility and processing optimization, are still limited.

In this context, this study aims to develop a sustainable bio-composite using recycled high-density polyethylene (HDPE) as the matrix and agro-industrial by-product coffee silver skin as a filler. The experiments, which methodically assess the effects of filler particle size and loading levels on the composite mechanical properties with application of experimental design techniques. Furthermore, the optimal formulation chosen based on its enhanced tensile strength was evaluated for thermal stability, melting behavior and chemical properties which provided a deeper insight into its performance characteristics and a close approximation of possible applications.

1.2. Statement of the Problem

The accumulation of both HDPE plastic and coffee silver skin wastes highlight a dual environmental issue, especially from high-density polyethylene (HDPE) present a persistent environmental pollution due to non-biodegradability and the difficulty associated to disposal, Moreover, agro-industrial residues such as coffee silver skin (CSS) a waste material generated in large quantities during coffee roasting are also discarded, leading to represents the dual waste problem is a missed opportunity for resource recovery. Despite efforts to recycle plastics, the recycling rates remain low due to the technical challenges involved, particularly in the case of thermoplastics like HDPE (Elkoun et al., 2023 ; Alrazen et al., 2025). None of the commonly used plastics are biodegradable, as a result they accumulate, rather than decompose in landfills or the natural environment, causing long - lasting ecological damage (Geyer et al., 2017).

In parallel, the coffee industry produces large amounts of waste, including coffee silver skin, which is often disposed of in landfills or incinerated (Dominici et al., 2019). These waste management challenge emphasize the need for sustainable alternative that can repurpose agricultural residues in to valuable products and the Potential applications of the coffee industry by-products in polymer technology (Hejna, 2021). The production of bio-composites using waste materials like HDPE and coffee silver skin offers an innovative solution to both of these issues. However, the challenge lies in optimizing the production process to ensure that the resulted composite exhibits thermal stability, mechanical, and thermochemical properties required for industrial application.

The combination of HDPE with CSS has been rarely explored, recent studies have begun investigating coffee silver skin as a reinforcing filler, (Kumar et al., 2024) ; (Petaloti & Achilias, 2024), and it has been successfully incorporated into biodegradable polymer matrices with demonstrable effects on thermal and mechanical behavior, indicating its potential as a sustainable filler with appropriate treatment methods, implying that coffee silver skin can effectively reinforce bio-based polymer matrices like PLA/PBS. The specific combination of HDPE and coffee silver skin has not been fully explored, yet limited attention has been given to the synergistic effects of combining HDPE, a major source of plastic waste with CSS, an underutilized by-product of coffee processing.

This research marks a step forward in the production of bio-composites using waste plastic and agricultural by-products and is part of one strategy on sustainability by converting waste materials into something useful while protecting the environment. By designing innovative composites, this strategy not only recycles waste, but also embeds the values of a circular economy in efficient use of resources.

1.3. Objectives

1.3.1. General Objective

The general objective of this research is to optimize selected operational parameters on the production of bio-composite using waste plastic (HDPE) and coffee waste (silver skin).

1.3.2. Specific Objectives

- To remove non-cellulosic components of coffee silver skin by alkaline hydrogen peroxide treatment method and characterize the physicochemical property.
- To statistically investigate and optimize the effect of selected processing parameters on the mechanical property (Tensile strength) of the resulted composites.
- To compare the tensile strength of the optimized composites fabricated from treated and untreated coffee silver skin.
- To examine the thermal stability, thermal characteristics, chemical structure, morphological and physical property of the composites prepared under optimum conditions.

1.4. Significance of the Study

The primary aim of this research is for addressing two critical environmental challenges: plastic waste accumulation and agro-industrial waste disposal, diverting waste HDPE and coffee silver skin in to advancing sustainable material development providing economic benefit/resource valorization by reducing waste and lowering reliance on virgin resources and promoting circular economy principle. The optimized bio-composite production process ensures that maximizing the properties of material while minimizing resource and energy consumption. The identification of optimum processing parameters, including filler ratio, particle size and temperature facilitates efficient bio-composite production while minimizing the environmental impact of waste disposal, mitigate pollution and resource depletion. This research will contribute to the growth body of knowledge on natural fiber-reinforced composites, providing valuable insights into the use of unconventional waste materials like coffee silver skin as reinforcement in polymer matrices.

The study will propose an innovative solution to reduce waste while creating value-added products without compromising the product quality and performance of the fabricated composites. The development of alternative material continuously evolving, and essential for industries looking to reduce their carbon foot print and dependency on virgin resources, particularly in sectors construction and interior application are interested in bio composite due to their potential for lightweight materials with good mechanical and thermal property.

Additionally, demonstrating the practical and scalable application bio-composite in manufacturing may inform future regulation and policies, support sustainable industrial practice, encourage industries to adopt greener practices and driving the shift toward a more sustainable economy. The findings could pave the way for further research into the use of other agricultural and coffee byproducts in bio-composite production, thereby promoting a more circular economy.

1.5. Scope of the Study

This study focuses on the development and optimization of bio-composites based on recycled high-density polyethylene (rHDPE) reinforced with coffee silver skin (CSS) as lignocellulosic filler. Both untreated and chemically treated CSS were investigated to evaluate the effect of surface modification on filler characteristics and composite performance. They were characterized using relevant analytical techniques prior to composite fabrication. The fabrication process was carried out with two major processes; compounding and consolidation process by varying three processing parameters, and the influence of these variables on tensile strength was analyzed and optimized using RSM with a BBD. The optimized composites were further characterized in terms of mechanical, thermal, chemical, morphological, and physical properties through tensile testing, Thermogravimetric Analysis (TGA), Differential scanning calorimeter (DSC), Fourier transform infrared spectroscopy (FTIR), Scanning electron microscope (SEM), and water absorption analysis.

2. Literature Review

2.1. Overview of plastic waste problem

Plastic waste has become one of the most pressing environmental issues in modern times, primarily due to the exponential increase in plastic production and its low biodegradability, over 8.3 billion metric tons of plastic have been produced globally since 1950, as of 2015 approximately 6300Mt of plastic waste had been generated, approximately 12% was incinerated with only 9% being recycled and the remaining 79% was disposed of in landfills, oceans, or other environments, posing significant ecological and health risks (Geyer et al., 2017) (Alrazen et al., 2025). Plastics degrade very slowly in the environment, gradually fragmenting into micro plastics that contaminate water, soil, and air, posing risks to both wildlife and human health (Wright & Kelly, 2017). The environmental impacts of plastic waste are widespread. In marine environments, plastic debris can physically harm organisms through ingestion or entanglement, damage habitats, and facilitate the transport of invasive species (Jambeck et al., 2015). On land plastics can alter soil structure and microbial communities, leading to reduced soil fertility and negatively affecting agricultural productivity. Plastics present a wide range of problems when they enter the environment. They may clog sewers, provide breeding ground for mosquitoes and disease-causing pests, get ingested, choke and/or entangle animals, leak toxic materials, and find their way into the human food chain (Ncube et al., 2021). Plastic pollution is a burgeoning threat to the sustainability of our planet. The world is responding at an already impressive scale, with grassroots action, national-level product bans, public-private partnerships for investment in waste management infrastructure, innovative alternatives to leakage-prone plastic products, and greater transparency in the trade of plastic waste (Single- et al., 2020).

2.2. High-Density Polyethylene (HDPE) and HDPE Waste

High-Density Polyethylene (HDPE) is a semi crystalline thermoplastic polymer produced through the polymerization of ethylene monomers, characterized by a linear molecular structure with minimal branching. This structural configuration results in high density, excellent chemical resistance, good tensile strength, low moisture absorption, and favorable process ability. Due to these properties, HDPE is extensively used in packaging materials, pipes, containers, household products, and industrial applications (Andrady & Neal, 2009). However, the widespread consumption of HDPE products has led to a substantial generation of post-consumer and post-industrial HDPE waste (Alrazen et al., 2025). Although HDPE is technically recyclable and commonly processed through mechanical recycling routes, recycled HDPE (rHDPE) often exhibits some degree of thermal and mechanical property degradation due to repeated processing and contamination. Consequently, valorization strategies such as incorporating rHDPE into composite materials reinforced with bio-based fillers have gained increasing attention as sustainable approaches to extend its service life and enhance material performance while mitigating plastic waste accumulation.

In 2024, 89.8% of the world plastics production was fossil based and the global plastics production increased to 430.9 million tonnes, 12.1% HDPE approximately 50 million ton was produced (*Plastics the Fast Facts 2025*, 2025). Three types of plastic waste management landfill, incineration, and recycling of mixed polymers, were studied. Plastics being sent to landfills, detrimental effects include the long degradation times and the release of toxins into the environment. Incineration another common method, it is not an efficient treatment method, it contribute significant environmental burden by creating GHG emission (Elkoun et al., 2023). Recycling HDPE can help mitigate environmental impacts by reducing the volume of plastic waste and conserving resources. The recycling process for HDPE typically involves collection, sorting, cleaning, compatibilization to reduce contamination by incompatible polymers and reprocessing into pellets, which can then be used in manufacturing new products (Hopewell et al., 2009).

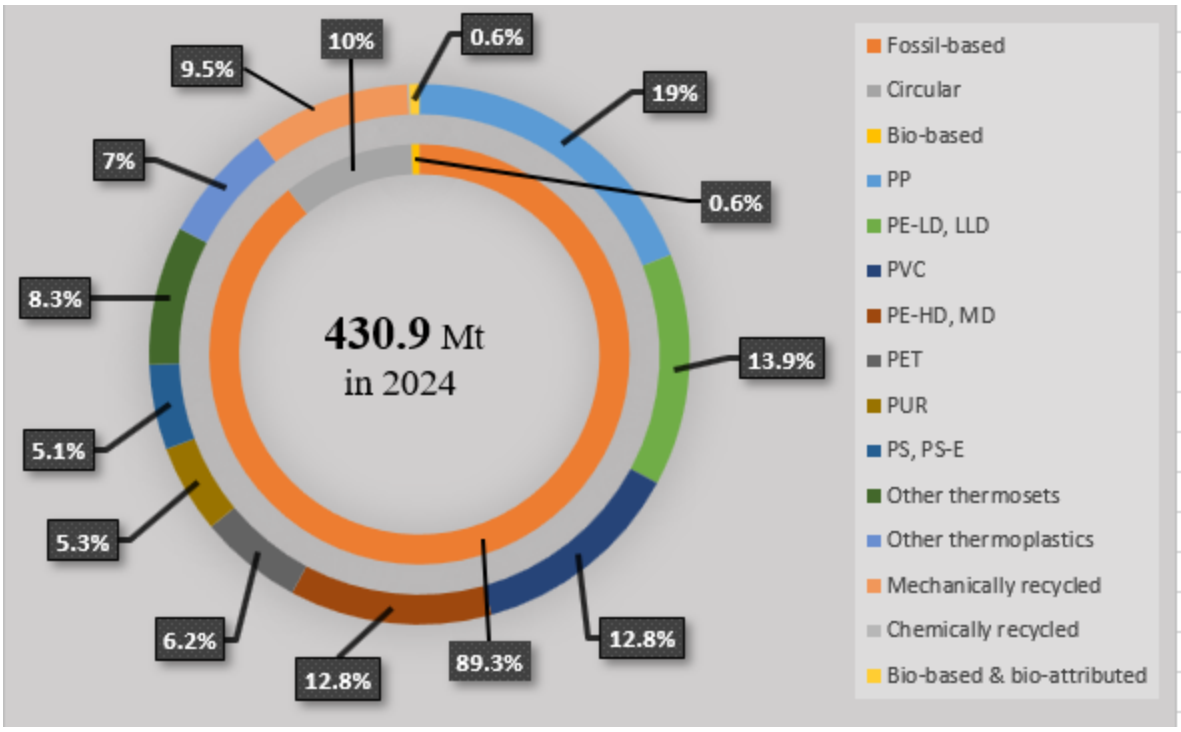


Figure 2.1: Distribution of global plastic production by type source : (*Plastics the Fast Facts 2025*, 2025)

2.3. HDPE Recycling Methods

2.3.1. Mechanical Recycling

Mechanical recycling, also known as secondary recycling, is the process of recovering plastic solid waste (PSW) for the re-use in manufacturing plastic products via mechanical means, It was promoted and commercialized all over the world back in the 1970s, A number of products found in our daily lives come from mechanical recycling processes, such as grocery bags, pipes, gut-tars, window and door pro les, shutters and blinds, etc. Recycling PSW via mechanical means involves a number of treatments and preparation steps to be considered (Andrady & Neal, 2009).

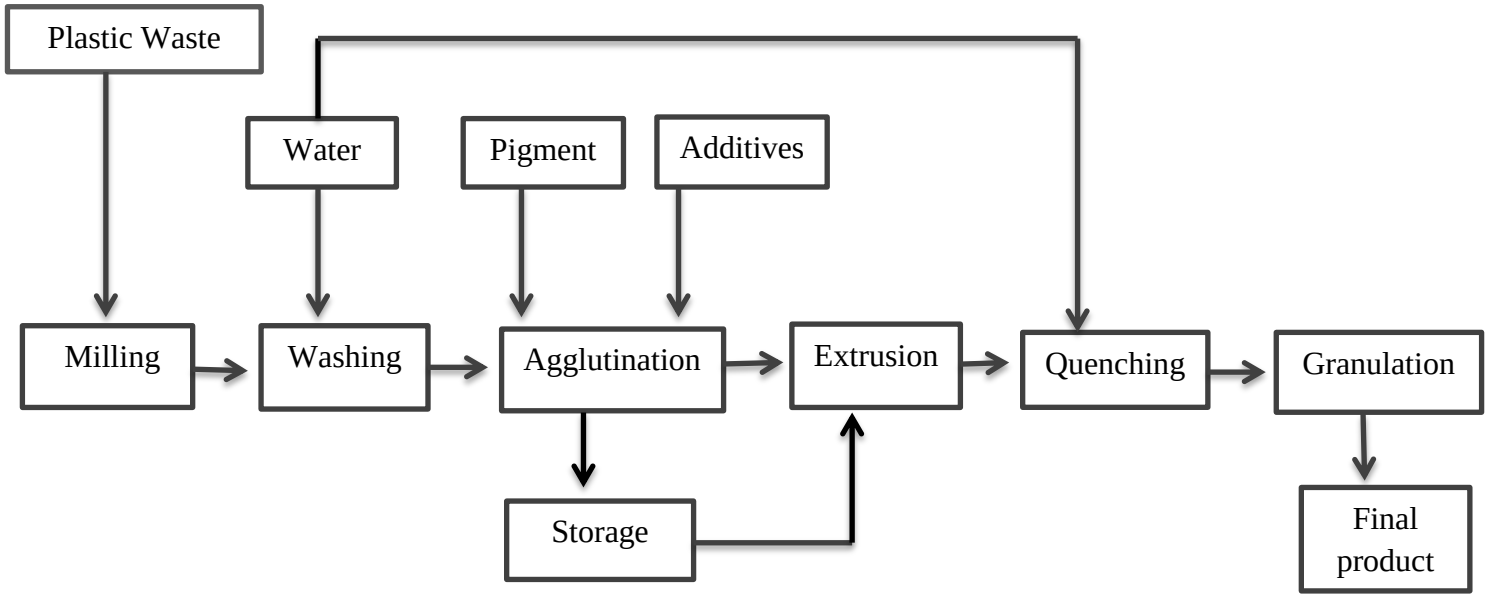


Figure 2.2: Mechanical Recycling steps source: (Al-Salem et al., 2009)

Mechanical recycling, the most common method for HDPE, involves shredding the plastic into flakes and melting it down to be formed into new products. However its widespread use, HDPE is difficult to recycle due to its high melting temperature and the degradation of its mechanical properties such as reduced tensile strength and stiffness over multiple recycling cycles due to thermal and mechanical stress during the process (Al-Salem et al., 2009).

2.3.2. Chemical Recycling

It involves breaking down the plastic in to its original monomers or other useful chemicals through chemical process, chemical recycling, although less widely used, breaking HDPE down into its monomers, leading in total de polymerization to the monomers, or partial degradation to other secondary valuable material (Achilias et al., 2007). chemical recycling of waste HDPE has potential challenges such as, high cost, energy consumption, complexity and environmental impact (Manzuch et al., 2021). as a result, much of the HDPE produced ends up in landfills or the natural environment, contributing to the growing plastic pollution crisis (Jambeck et al., 2015). However, the potential of HDPE recycling, challenges remain. Several advancements in HDPE recycling aim to overcome these barriers. For instance, the introduction of compatibilizer additives that enhance the blending of HDPE with other materials has improved the mechanical properties of recycled HDPE composites (Manzuch et al., 2021).

2.4. Utilization of Agro-industrial waste: Coffee waste (Coffee silver skin)

Coffee is one of the most important agricultural commodities in the world; it was traditionally developed as a colonial cash crop. An estimated 11 million hectares of the world's farmland are dedicated to coffee cultivation (Blinová et al., 2017). According to International coffee organization annual review: World coffee production for the 2023/24 coffee year is estimated at 178.0 million bags, comprising 102.2 million bags of Arabicas and 75.8 million bags of Robustas (Annual Review, 2024). Uganda and Ethiopia are the major suppliers of African coffee during the first half of the 2023/2024 coffee year, contributing 46.3% and 42.8% of the total exports respectively (Fatimatuzzahro, Nadie, 2016). Coffee is produced from harvested coffee cherries, after their processing, which can be classified as pre- (wet or dry processing) and post-roasting (Sherefa et al., 2026).The annual production of coffee waste is projected to surpass 23 million tons. Its by-products mainly come from the extraction of shells and mucilaginous components from coffee fruits, and their composition varies based on the specific processing methods used, including wet or dry processing, as well as subsequent stages like roasting and brewing. Solid coffee residues include coffee pulp, coffee husks, silver skin, and spent coffee (Petaloti & Achilias, 2024).

a, Husk b, Pulp c, Silver skin d, Coffee spent ground

Figure 2.3: Major coffee by-products source: (Blinová et al., 2017)

Silver skin is the thin covering of the coffee beans' outer layer, which remains on the bean after their processing, and its name is derived from its silverish color. It is generated as the only by-product from roasting process, and for this reason, it is accumulated in the roasting factories and not in the coffee processing factories, where the coffee pulp, mucilage, and parchment of husk are obtained (Assefa et al., 2025).

It is a residue with high concentration of soluble dietary fiber (86% of total dietary fiber) and having high antioxidant capacity, the main components of these fibrous tissues are cellulose and hemicellulose. Glucose, xylose, galactose, mannose, and arabinose are the monosaccharaides present in coffee silver skin along with proteins (Murthy & Madhava Naidu, 2012).

Table 1.1: Typical composition of coffee silver skin source: (Petaloti & Achilias, 2024)

Component	Coffee Silver skin (Content, % Dry Mass)
Cellulose	17.9-23.8
Hemicellulose	7.5-16.9
Holocellulose	28.6-40.5
Lignin	28.6-31.0
Ash	4.5-7.6
Lipids	11.8-18.7
Protein	2.1-5.8

Coffee silver skin, it has no commercial value, so that, it usually finds its end-of-life in controlled landfills or it is subjected to incineration which is a source of contamination, due to its high organic content and presence of chemical compounds (Janissen & Huynh, 2018). From the technological point of view, coffee silver skin should be considered one of the most promising by-products of the coffee industry. Because of the very low moisture content, it is not as perishable as other by-products and does not often require quite energy-consuming drying processes, the majority of lignocellulosic materials is the relatively high content of proteins (~17–18%), which may act as plasticizers or, due to the presence of functional groups, provide additional possibilities for interfacial adhesion with polar polymer matrices. Therefore, the development of the potential applications of CSS in polymer technology, requiring low levels of moisture of raw materials, could be very beneficial for the coffee market (Hejna, 2021).

2.5. Introduction of Bio-composite

Bio-composites are materials composed of natural fibers or organic fillers embedded in a polymer matrix. Due to environment and sustainability issues, this century has witnessed remarkable achievements in green technology in the field of materials science through the development of bio-composites. These materials have gained significant attention due to their environmental benefits, including reduced reliance on non-renewable resources and lower greenhouse gas emissions compared to traditional composites (Faruk et al., 2012). The use of natural fillers/reinforcements into polymeric matrices could lead to multiple advantages, such as clear cost reduction, lightness and good balance on mechanical properties, and a marked low environmental impact as well, due to the use of renewable material, natural fillers/reinforcements are majorly lingo-cellulosic materials and, therefore, they show an extremely high hydrophilicity which results in poor material cohesion and, subsequently, poor mechanical performance for Improvement of interface adhesion in NFRP (Natural fiber reinforced plastic) or WPC (wood plastic composite) can be achieved by several strategies (Dominici et al., 2019).

2.6. Interaction between polymer matrices and natural fiber

The interaction between the polymer matrix (HDPE) and the natural fiber (CSS) is critical in determining properties of the final bio-composite. Natural fillers/reinforcements are majorly lingo cellulosic materials and, therefore, they show an extremely high hydrophilicity. This results in poor adhesion with the usually hydrophobic polymer matrix. In addition, the hydrophilic nature of the filler/reinforcement also leads to aggregate formation (poor particle dispersion), which results in poor material cohesion and, subsequently, poor mechanical performance by lowering the hydrophilicity of the filler/reinforcement (Montanes & Balart, 2018). Improvement of interface adhesion in NFRP or WPC can be achieved by several strategies, such as selective surface modification of fillers, modification of the polymer matrix by additives and using compatibilizer to provide increased interactions among the polymer-particle interface (Chan et al., 2018). For surface modification, chemical pretreatments to the fillers commonly used for alkali treatments to remove non-cellulosic components like lignin Bleaching with hydrogen peroxide can be carried out subsequently to further purify the cellulose and hemicelluloses, leading to a change in the fiber color to white.

Furthermore, it enhances the surface area of the fibers, promoting better contact and interaction between the filler and matrix (Petaloti & Achilias, 2024), thereby enhancing the dispersion of the bio-fillers within the polymer matrix.

2.7. Composite Fabrication Processing Method

The fabrication of thermoplastic composites typically involves two sequential stages: Homogenizing compounding and consolidation process, both of which strongly influence the microstructure and final properties of the composite material. In compounding process, the polymer matrix and reinforcement are mixed under controlled temperature and shear conditions to promote homogeneous filler dispersion and adequate matrix-filler wetting, often achieved using twin-screw extruders due to their efficient mixing and distributive capabilities (Berzin et al., 2020). However, laboratory-scale approaches such as melt blending using internal or batch mixers are also reported in the literature, where the polymer is melted and mechanically mixed with fillers under controlled conditions to achieve a homogeneous composite blend. The selection of compounding technique depends on equipment availability, material characteristics, and research scale (Vincent et al., 2021). After compounding, the homogenized material is processed through consolidation techniques including compression molding or extrusion compression molding, which facilitate melt flow, void reduction, and structural densification to shape the final composite product. The selection and control of processing conditions in both compounding and consolidation stages are crucial in thermoplastic composite manufacturing, as they determine the extent of filler dispersion, interfacial bonding, and ultimately the mechanical performance of the composite (Oksman et al., 2014).

2.7.1. Compounding Process

Compounding is a primary step in thermoplastic composite fabrication in which the polymer matrix and reinforcement are melt-mixed under controlled thermal and mechanical conditions to achieve homogeneous filler dispersion and improved interfacial interaction. Twin-screw extrusion is widely used for this purpose due to its enhanced distributive and dispersive mixing capabilities, which facilitate uniform dispersion of fibers or particles within the polymer (Hietala & Oksman, 2018).

The design of co-rotating twin-screw extruders allows for adjustable screw configurations that generate controlled shear forces necessary to de agglomerate fillers and reduce fiber bundles into smaller fragments, thereby improving composite homogeneity (Berzin et al., 2017). In thermoplastic compounding, melt blending is widely used as an alternative to continuous extrusion; it involves melting the polymer and mechanically blending it with fillers to form a composite, and it is compatible with laboratory-scale mixers such as batch internal mixers or low-shear mixers (Vincent et al., 2021). During compounding, critical processing parameters such as screw speed, feed rate, and barrel temperature influence the extent of fiber breakage, residence time, and dispersion efficiency, ultimately affecting composite morphology and mechanical performance (Melt Processing of Bio-plastic Composites via Twin Screw Extrusion and Injection Molding (Taylor et al., 2013).

2.7.2. Consolidation Process

The consolidation stage in thermoplastic composite fabrication involves applying heat and pressure to a compounded melt in order to eliminate voids, enhance polymer flow, and densify the structure into final part. Compression molding is the most commonly employed consolidation technique for thermoplastic composites due to its simplicity, ability to process high filler contents, and suitability for flat or moderately complex geometries (Sormunen, 2019). During compression molding, the compounded charge is placed between heated platens where elevated temperature reduces matrix viscosity and applied pressure promotes resin flow, intimate contact between phases, and void reduction, ultimately increasing composite integrity and mechanical performance Key processing parameters such as molding temperature, applied pressure, dwell time, and cooling rate influence the degree of consolidation and void content; inadequate control of these parameters can lead to residual porosity, weak interface bonding, and reduced mechanical properties (Jaafar, 2019). For composites made with recycled polymers and natural fillers, achieving sufficient consolidation is particularly important because non-uniform pressure or temperature gradients can exacerbate filler agglomeration and hinder polymer penetration into void spaces, affecting final performance (Sormunen, 2019).

2.8. Factors Affecting Composite Fabrication

Several factors determine the performance and structural integrity of thermoplastic composites during fabrication. Numerous studies have shown that variables such as filler characteristics, filler loading, mixing time, pressure and processing temperature significantly affected the fabrication process.in this study, the influence of three factors, namely particle size, filler ratio, and temperature are studied.

2.8.1. Effect of Particle Size

Particle size of lignocellulosic fillers significantly influences the microstructure and performance of thermoplastic composites. Reduced particle size increases the specific surface area for matrix-filler interaction, which enhances stress transfer and improves mechanical properties such as tensile strength and modulus (Migneault et al., 2009). Smaller particles also tend to promote more uniform dispersion within the polymer matrix, reducing stress concentration sites and minimizing void formation during processing (Yang et al., 2004), However, excessively fine particles less than 90µm, may increase melt viscosity and promote agglomeration due to strong inter particle interactions, which can negatively affect process ability and composite homogeneity (Delviawan et al., 2019). Larger particle sizes may lead to poor interfacial bonding and reduced mechanical performance due to limited surface contact between the filler and polymer matrix. Therefore, optimization of filler particle size is essential to balance dispersion quality, interfacial adhesion, and processing efficiency in bio-based thermoplastic composites.

2.8.2. Effect of Filler Ratio

Filler ratio is one of the most influential parameters affecting the mechanical, thermal, and morphological properties of thermoplastic composites. As the filler content increases, a higher number of filler particles are dispersed within the polymer matrix, which can enhance stiffness and improve load transfer due to increased interfacial interaction between the filler and matrix. At moderate filler loadings, generally enhances stiffness and elastic modulus due to the higher proportion of rigid reinforcement within the polymer matrix (Migneault et al., 2009).

However, excessive filler content lead to poor dispersion, increased agglomeration, and insufficient matrix wetting due to weak interfacial bonding, which can reduce tensile strength and elongation at break (Yang et al., 2004), Higher filler loading particularly above 30wt.%, also increases melt viscosity during processing, potentially affecting flow behavior and consolidation quality (Gupta et al., 2004). Hence, an optimum filler ratio is required to achieve a balance between improved mechanical properties and acceptable processing behavior. In this study, filler loadings in the range between 10-30 wt.% were selected to evaluate their effect on the performance of the fabricated composites.

2.8.3. Effect of Temperature

Processing temperature during consolidation strongly influences the melt flow, filler impregnation, and mechanical performance of thermoplastic composites. In natural fiber-reinforced polymer systems, increasing temperature within an appropriate range enhances polymer melt viscosity reduction, which facilitates matrix penetration into the filler network and improves interfacial bonding, contributing to better consolidation and mechanical properties. For example, studies have shown that processing composites with higher temperatures can improve tensile modulus but may reduce tensile strength if the temperature exceeds optimal levels due to changes in rheological behavior and filler-matrix interactions (Salleh et al., 2014). However, excessive thermal exposure can induce degradation of thermally sensitive natural fiber components such as hemicellulose and pectin at temperatures typically observed near or above 180°C, leading to diminished reinforcement integrity and lower overall composite performance (Mattlet et al., 2023). Selecting an appropriate consolidation temperature such as 140-180°C commonly applied for HDPE-based composites is critical to balance sufficient polymer flow and wetting with minimal thermal degradation of the natural fillers.

2.9. Applications

Bio-composites are considering one of the major materials utilized as a part of green materials at this time. It could be categorized, with regard to their application in the building market into two principle products: first, structural bio-composites, which include bridge as well as roof structure, and second, nonstructural bio-composites which include window, exterior construction, composites panels, and door frame (Mohammed et al., 2015).

HDPE-based bio-composites reinforced with natural fillers have attracted considerable attention across a range of application sectors due to their favorable balance of mechanical performance, durability, and cost effectiveness. One of the most prominent application areas is in wood-plastic composites (WPC) used for construction and building materials such as decking, siding, fencing, and outdoor furniture, where enhanced stiffness and resistance to environmental degradation are critical (Ribeiro et al., 2023).

In the automotive industry, natural based composites have been explored for interior panels, trim components, and under-the-hood applications due to their lightweight nature and improved impact resistance when appropriately reinforced; the car manufacturer in Europe has done various researches to increase the application of Natural fiber polymer composites (NFPCs) in automotive industry, especially in car interior such as seat backs, parcel shelves, boot lines, font and rear door lines, truck lines, and door-trim panels (Mohammed et al., 2015). Furthermore, Packaging is the largest application for plant-based plastics, consumer goods, and household products benefit from the eco-friendly profile of bio-composites, especially when recyclability and reduced carbon footprint are priorities (Shogren et al., 2019).

2.10. Summary of Previous Research

Recent studies have extensively investigated thermoplastic composites reinforced with lignocellulosic fillers as sustainable alternatives to conventional petroleum-based materials. Several researchers have reported that incorporating natural fibers or agricultural residues into thermoplastic matrices such as HDPE can improve stiffness, reduce material cost, and enhance environmental sustainability (Mohammed et al., 2015) ; Ribeiro et al., 2023).

For example, Zhang et al., (2020), used bio char derived from agricultural waste to reinforce HDPE investigating excellent properties of the composite, they obtained in 50% and 60% bio char added the composite achieved tensile strength of 29.05MPa with good thermal and flame retardant properties.

In addition to the selection of suitable fillers, several studies have reported the importance of chemical modification of lignocellulosic materials to improve their compatibility with thermoplastic matrices. Chemical treatments such as alkaline modification are commonly applied to natural fillers to remove surface impurities and enhance adhesion between the filler and polymer matrix. Likewise, Petaloti & Achilias, (2024), reported that CSS as a reinforcing filler in bio-based polymer matrix, the alkali-treated CSS improved the tensile strength by 9.3% compared with untreated CSS filler due to surface modification. Another researchers, Dominici et al., (2019), used chemically treated CSS as reinforcing agent in bio-based polyethylene matrices, reported that tensile strength slightly increased in alkali treated CSS matrix composite. Furthermore, Cecilia et al., (2023), investigated HDPE with rice husk composites and reported a tensile strength of 15.25 MPa through the incorporation of MAPE compatibilizer to improve the interfacial adhesion between the hydrophobic HDPE matrix and the hydrophilic lignocellulosic filler.

Various processing techniques, including melt compounding and compression molding, have been widely used to fabricate natural fiber-reinforced thermoplastic composites, with processing parameters such as filler content, particle size, and processing temperature significantly influencing composite performance (Yang et al., 2004 ; Shogren et al., 2019). For instance, Kumar et al., (2024) employed a single screw extruder for compounding of CSS and PLA/PBS polymer matrixes by varying filler size of CSS between (150–212 µm and 212–425 µm) and loading (10, 20, 30 wt.%) at a temperature of 165℃, followed by injection molding machine for consolidation process. They reported that CSS fillers can improve the mechanical properties of bio-PBS composites. Based on their report, 10 wt.% of filler content of smaller filler size has shown optimum tensile strength and composites fabricated with CSS fillers with lower filler size and loading have shown better thermal stability and mechanical properties.

2.11. Research Gap

Although considerable research has been conducted on natural fiber reinforced thermoplastic composites, most studies have focused on commonly used lignocellulosic fillers such as wood flour, rice husk, bamboo fiber and other agricultural residues. In contrast, relatively limited attention has been given to the utilization of coffee processing by-products, particularly coffee silver skin, as reinforcement material in thermoplastic composite systems. Considering the large quantity of coffee processing waste generated worldwide, the valorization of coffee silver skin as a functional filler in polymer composites represents a promising yet underexplored research area.

In addition, the interfacial compatibility between lignocellulosic fillers and thermoplastic matrices remains one of the major challenges that significantly influence the overall performance of bio-composites. Due to the inherent hydrophilic nature of lignocellulosic materials and the hydrophobic characteristics of thermoplastic polymers such as HDPE, weak interfacial bonding often occurs, leading to poor stress transfer and reduced mechanical properties. To address this limitation, various chemical modification techniques, particularly alkali (NaOH) treatment, have been widely employed to enhance the surface characteristics of natural fibers. While these treatments are known to improve filler-matrix adhesion, existing studies have largely focused on commonly used natural fibers, with comparatively limited attention given to coffee processing by-products such as coffee silver skin to investigate the influence of such treatments on the physicochemical properties of coffee silver skin and its subsequent interaction with recycled HDPE matrices.

Comprehensive characterization of modified fillers, including their thermal behavior, structural properties, and surface morphology, is essential to better understand their role in composite fabrication. Ayana et al., (2022) fabricated PE-based composites reinforced with treated and untreated bamboo fiber and observed improved mechanical properties of the composites from alkali surface treatment of bamboo compared to raw fiber product. However, the study focused only on material characterization and did not evaluate process optimization on composite performance.

Similarly, Kumar et al., (2024) and Petaloti & Achilias, (2024), investigated coffee silver skin as a reinforcing filler in bio-based polymer composites and demonstrated its potential for improving material properties. But, the studies did not establish on optimum processing conditions and primarily focused on bio-based thermoplastic polymers not HDPE.

Further research is required to investigate the processing conditions, effect of filler modification, and performance characteristics of rHDPE composites reinforced with coffee silver skin. A systematic evaluation of the effects of filler particle size, filler loading, and processing temperature, combined with statistical optimization approaches, can contribute to the development of sustainable and high-performance bio-composite materials derived from thermoplastic and agro-industrial wastes.

3. Materials and Methods

3.1. Materials

3.1.1. Raw Materials

Post-consumer packaging High-Density Polyethylene (HDPE) waste was collected from local waste stream serve as raw material for the production of bio-composite. Additionally one of the major coffees processing industry by-product which is coffee silver skin was collected from local roastery factor, and subjected for further treatment and analysis.

3.1.2. Chemicals and Reagents

In this study, all chemicals and reagents used were of analytical grade and they were used without additional purification. Distilled water was used throughout the experimental work for washing collected waste HDPE and raw coffee silver skin prior to processing, rinsing the material after chemical treatment and bleaching steps, and immersing molded composite specimens during the water absorption test. It was also employed as a solvent for the preparation of all chemical solutions. Sodium hydroxide (NaOH) pellets, with purity level of (99.9%) were used for the alkali treatment of CSS to treatment and modify the fiber surface. Hydrogen peroxide (H_2O_2, 5% w/v) was utilized and applied as a bleaching agent during fiber modification. Acetic acid solution (5% v/v) was prepared from glacial acetic acid (99%) for neutralization following alkali treatment. motor oil as lubricant for composite fabrication process.

3.2. Research Frame work and Experimental work flow

The following diagram presents the research framework and experimental workflow adopted in this study.

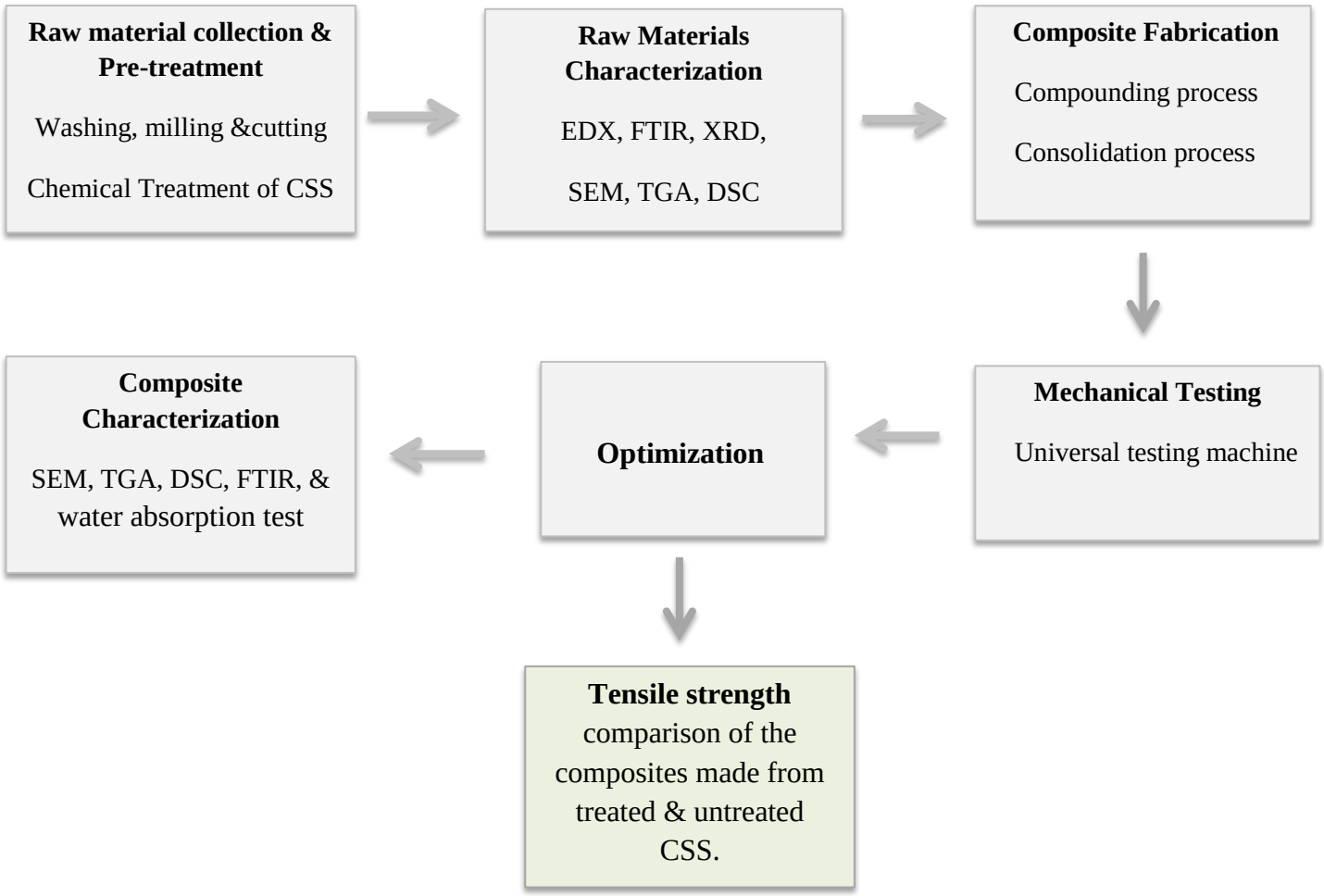


Figure 3.1: Process flow diagram of experimental work flow

3.3. Raw Material Treatment and Sample Preparation

3.3.1. Pre-processing of CSS

Raw coffee silver skin was initially used approximately 1.5kg, primarily cleaned to remove visible impurities and repeated washing with distilled water to eliminate dust and soluble residues, then dried an oven at 80℃ for 24h and stored in a dissector. After drying the dried coffee silver skin ground by electrical grinder, following this ground CSS was fractionated using a vibratory sieve shaker with mesh size of 500µm, 300µm, and 100µm. The sieved particles were separated and stored in labeled zipper polyethylene bags according to their particle size range. 150gram of CSS was prepared from each size category for subsequent chemical treatment.

3.3.2. Chemical Treatment of CSS

Coffee silver skin was treated using alkali treatment followed by acetylation and hydrogen peroxide bleaching to reduce lignin content and extract cellulose. The chemical treatments involve functional groups interacting with bio-filler structures to remove non-cellulosic elements and purify surfaces, altering compositions, these pretreatments such as alkali, bleaching, and acetylation, aim to enhance the properties and performance of the fillers, ultimately improving the quality of the final product. Sodium hydroxide is commonly used for alkali treatments to remove non-cellulosic components like lignin (Imran et al., 2024). In this study each CSS size fraction underwent alkali pretreatment using an 8% sodium hydroxide (NaOH) solution at a solid-to-liquid ratio of 1:10 (g/mL).

The samples were soaked and stirred in the NaOH solution at room temperature for 24h, this method was adopted from the procedure described by (Petaloti & Achilias, 2024),who used a similar alkaline process for coffee silver skin alkali treatment. After soaking, the treated CSS was filtered using vacuum filtration with appropriate filter papers; following this the samples were washed extensively with distilled water until the PH became 10, then placed in 5% acetic acid solution for 30 minute with stirring, to neutralize any remaining hydroxide, then filtered from neutralized solution and washed by distilled water, finally the chemical-treated particles were dried in an oven at 65 °C for 24h.

The delignified of coffee silver skin was subjected to a bleaching process using a 4% NaOH solution in combination with 5% hydrogen peroxide (H_2O_2). The bleaching process using hydrogen peroxide in alkaline medium studies (Behrouzian et al., 2016); the treated particles were placed in 4% aqueous solution of sodium hydroxide solution at a temperature of 90℃ in a water bath with continuous stirring for 1h. In the first bleaching stage, 500mL of 5% hydrogen peroxide were gradually added to the solution while stirring, then the particles changed color, they were cooled to room temperature, separated from the solution through filtration, afterward washed with distilled water repeatedly. Due to incomplete whitening in the first bleaching stage, a second bleaching process was applied, and then the second bleaching was performed with same procedure as first one.

Finally, the sample color changed to white filtered from the solution and washed with extensive distilled water until neutral pH was achieved and oven dried at 65℃ for 24h. Following to this the dried sample was milled by electrical grinder and sieved to obtain particle size fractions of 500µm, 300µm, and 100µm, selected for composite fabrication.

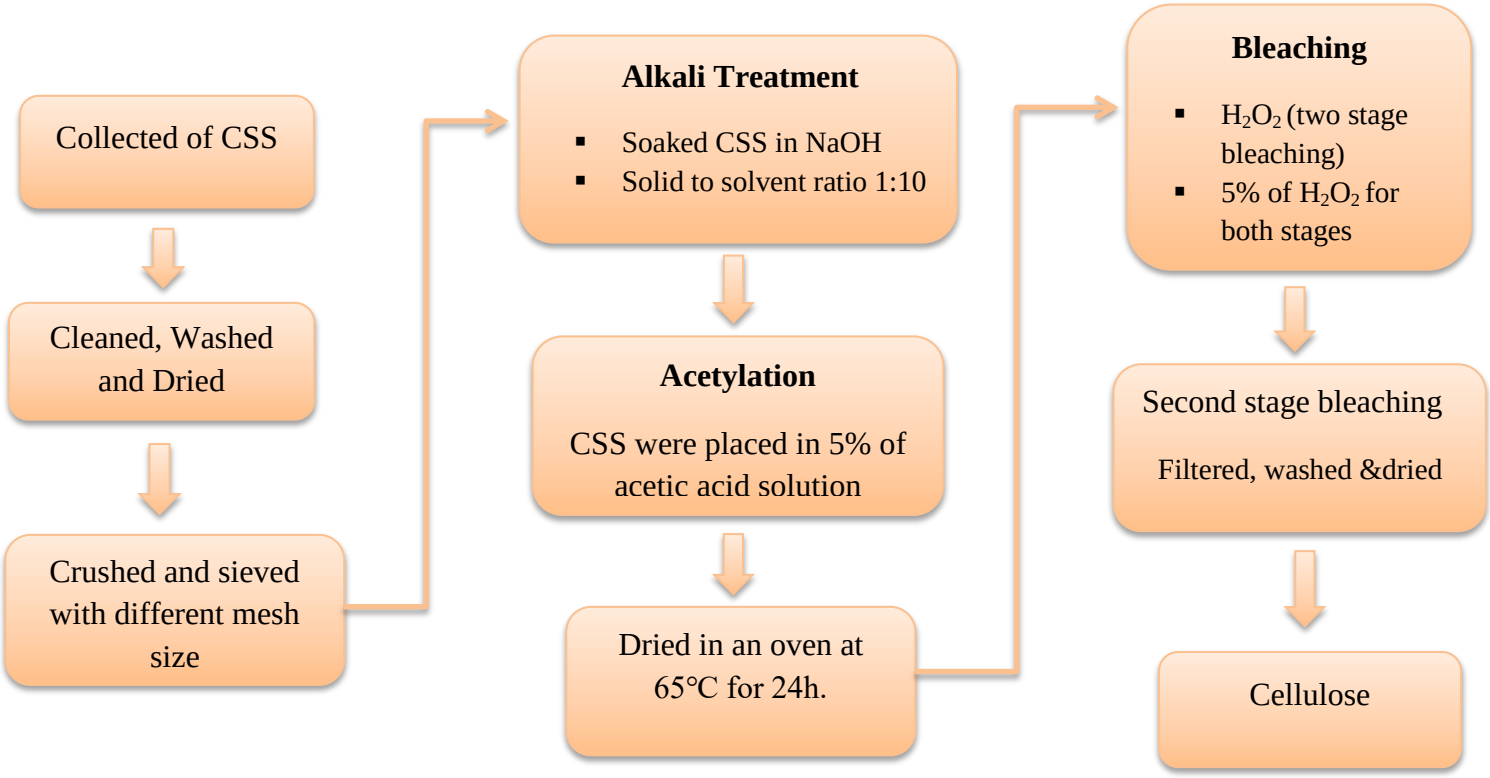


Figure 3.2: Schematic diagram of chemically treated CSS preparation process

3.4. Characterization of CSS

Six different characterizations were conducted to evaluate the physicochemical and thermal characteristics of both untreated and treated coffee silver skin to assess the effects of chemical treatment which were provide baseline information for composite fabrication. Elemental composition, crystalline structure, chemical functional groups, surface morphology, thermal stability, and thermal characteristics of the materials were characterized using Energy-dispersive X-ray spectroscopy (EDX), X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), Thermogravimetric analysis (TGA), and Differential scanning calorimeter (DSC).

3.4.1. Energy-Dispersive X-Ray Spectroscopy (EDX) Analysis

The elemental composition of untreated and chemically treated coffee silver skin was analyzed using energy-dispersive X-ray spectroscopy (EDX) coupled with a scanning electron microscope. The analysis was performed to identify the presence and relative distribution of major elements and to evaluate changes in elemental composition resulting from chemical treatment. EDX measurements were conducted under high-vacuum conditions, and the acquired spectra were used for qualitative and semi-quantitative elemental analysis of the samples.

3.4.2. X-Ray Diffraction (XRD) Analysis

Crystallographic structure and phase composition of the CSS both treated and untreated were identified by X- ray diffraction (XRD). The sample was scanned in 2θ angles to receive the XRD pattern. This method is the analysis of the characteristic peaks of diffraction that allow knowing on the one hand the crystallinity of the material and the other the organization of the atomic structure. With regard to chemical treatment, it helps differentiate amorphous and crystalline regions and identify structural changes.

3.4.3. Fourier Transform Infrared Spectroscopy (FTIR) Analysis

The functional groups of raw and treated coffee silver skin surface were identified by using Fourier Transform Infrared Spectrometer. The spectra were recorded by wave numbers in the range of 400 - 4000 cm^{-1}. Each peak in the spectrum defines a specific functional group.

3.4.4. Scanning Electron Microscope (SEM) Analysis

Scanning electron microscope analysis was performed to study the surface morphology of the coffee silver skin before and after chemical treatment. Prior to imaging, the sample were coated for a better image resolution. The analysis was done under high vacuum by applying an accelerating voltage of 5.0 kV.

3.4.5. Thermogravimetric Analysis (TGA)

The thermal stability of the treated and untreated coffee silver skin was investigated through TGA. The analysis took place under an inert atmosphere. The apparatus used was TGA-s 1000 Thermogravimetric analyzer. A 3mg sample was measured and heated from room temperature of 27.80 to 800°C gradually to examine its thermal profile.

3.4.6. Differential Scanning Calorimeter (DSC) Analysis

It was conducted to evaluate the thermal behavior of untreated and chemically treated CSS. 3mg of each sample were placed in aluminum pans and heated in the DSC instrument under a nitrogen atmosphere. The samples were subjected to control heating over a specified temperature range at a constant heating rate to record their thermal transitions and heat flow characteristics.

3.5. HDPE Matrix Preparation and Characterization

High-density polyethylene (HDPE) was used as the polymer matrix for composite fabrication, Post-consumer packaging HDPE waste were also collected from local waste stream, cleaned and manually chopped into smaller flakes using scissors, then washed with distilled water and dried in open air to eliminate residual moisture that could adversely affect melt processing and interfacial bonding, for facilitate compounding process of the two materials. The dried rHDPE was then stored in sealed containers at room temperature to prevent moisture uptake and was used directly in the subsequent composite fabrication process.

3.5.1. Scanning Electron Microscope (SEM) Analysis

The microstructural features of the samples were then observed using a scanning electron microscope at appropriate magnifications. The obtained micrographs were used to analyze the surface characteristics and morphological structure of the rHDPE material.

3.5.2. Thermogravimetric Analysis (TGA)

The purity and composition of the neat HDPE was investigated through TGA. The analysis took place under an inert atmosphere. The apparatus used was TGA-s 1000 Thermogravimetric analyzer. A 3 mg sample was measured and heated at room temperature of 27.80 to 800°C gradually to examine its thermal profile.

3.5.3. Differential Scanning calorimeter (DSC) Analysis

Differential Scanning Calorimetry (DSC) is a thermal analysis technique widely used to characterize the thermal transitions and heat flow behavior of rHDPE as a function of temperature. In DSC analysis, the difference in heat flow between a sample and an inert reference is measured while both are subjected to a controlled temperature program.

3.5.4. Fourier Transform Infrared Spectroscopy (FTIR) Analysis

FTIR was used to characterize recycled HDPE before composite fabrication in order to confirm the presence of characteristic polyethylene functional groups and assess possible chemical degradation. The spectra were recorded by wave numbers in the range of 400-4000 cm^{-1}.

3.6. Composite Fabrication

The fabrication of rHDPE and CSS composite involves compounding and consolidation processes. For this fabrication process first performing homogenous mixing of the two materials, to ensure uniform dispersion of coffee silver skin particles within the HDPE matrix, a melt-mixing approach commonly employed for thermoplastic-based composites was adopted. Such methods are widely reported to enhance filler distribution, interfacial interaction, and dimensional stability, particularly when particle size and filler loading are varied as processing parameters.

In this study, the compounding procedure was adapted from the method reported by (Kumar et al., 2024) with necessary modifications due to equipment limitations. Instead of a conventional laboratory-scale single-screw extruder, a manually operated oil pressing machine equipped with a single rotating screw was employed to facilitate mechanical mixing. Although the system lacked an internal heating unit, external thermal energy was supplied using a controlled hot-air gun to soften the rHDPE matrix during processing. The temperature was maintained approximately at 165°C for a residence time of 15min to promote polymer melting and uniform incorporation of CSS particles. This modified melt-mixing approach enabled effective dispersion of the filler within the polymer matrix while maintaining process controllability and reproducibility under the available laboratory conditions/equipment.

Following this, reduced size of compounded rHDPE:CSS granulates was consolidated using a custom-fabricated compression molding plate. The consolidation procedure was adapted from the method reported by (Ayana et al., 2024), with modification introduced due to absence of a laboratory-scale computerized hot-plate compression molding system. In this study, consolidation was carried out under constant applied pressure and holding time, while the molding temperature was varied as a processing parameter according to the experimental design. Accordingly, a total of 15 formulations were prepared through compounding and consolidation processes, followed by four additional experimental runs conducted after optimization.

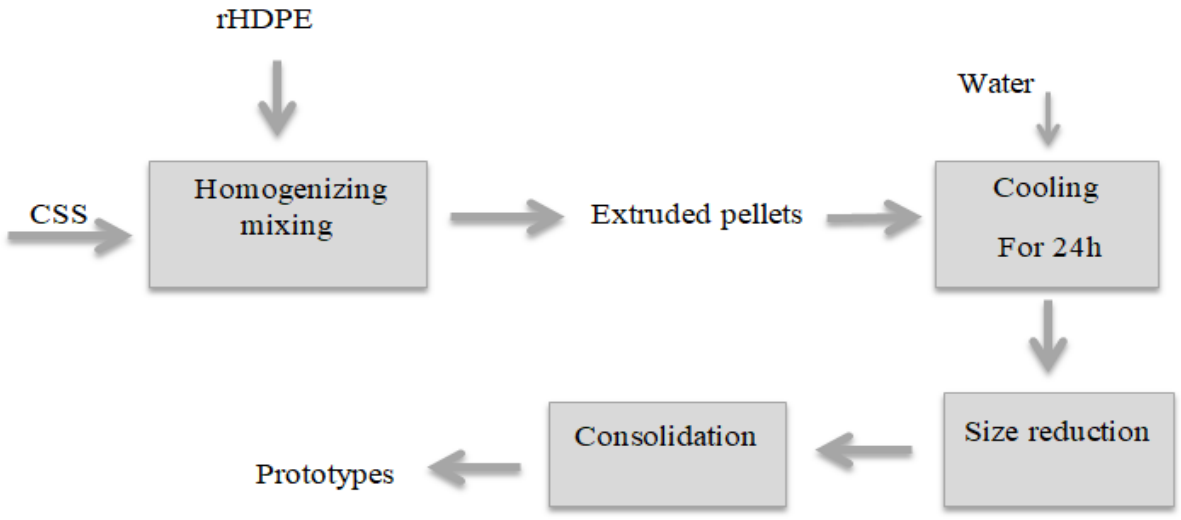


Figure 3.3: Schematic diagram of composite fabrication process

3.6.1. Compounding Process

The rHDPE and oven dried CSS particle blended with three different filler sizes, Large (500μm), Medium (300μm) and Small (100μm) in loading ratio of (10, 20, 30 wt. %) by using a single screw oil pressing machine as the main processing equipment. Before initiating the compounding process, motor oil was used as a lubricant for the pressing chamber and moving parts of the machine, the oil press to minimize frictional resistance and to facilitate smooth sample release after molten mixing, then the machine was preheated approximately for 5min to enhance the efficiency of melting and compounding, thereby improving the overall yield and homogeneity of the composite. Following preheating, the rHDPE pellets were fed through the hopper under continuous external heat supply. After 5min, CSS was added and mixing was continued for 15min.

The machine contained a rotating hand-like structure connected to a single internal screw that revolved within the pressing chamber manually, facilitating the mixing and compounding of the two materials. As the screw rotated, the feed materials were gradually melted and uniformly mixed to form a homogeneous blend. This mixing and compounding process was maintained for 15 minutes at temperature of 165°C. Upon completion, the molten composite began to exit through the outlet of the chamber and was immediately directed into a water container to allow rapid cooling and solidification, which enhanced the strength and stability of the extruded pellets. The extruded pellets stay in the water container for 3-4h until it become cold and back to at room temperature, the maintain heat need to be remove and become cool making better for next process. The cooled pellets were then open air-dried for 24h to remove residual moisture. Finally, the dried composite pellets were ground using a cutting machine and double conical solid mixer to achieve uniform particle size distribution and preparing them for the next consolidation process. This approach provided an effective alternative to conventional extrusion, enabling successful mixing and preliminary formation of rHDPE and silver skin bio-composite materials. Additionally, no chemical coupling agents were incorporated during this process in order to minimize chemical consumption, maintain low-cost, and reduce environmental impacts.

3.6.2. Consolidation Process

A manual compression mold was custom-fabricated in a mechanical workshop with standard dimensions of 8 cm × 2 cm × 4 cm. The molding setup consisted of a stove used as the heating source, a thermometer for temperature monitoring, lubricating oil to prevent adhesion between the mold and the sample, and a water container for cooling. The molding process was carried out at three different temperature conditions (140,160 and 180°C) to examine the effect of temperature on composite formation. Initially, the molding plate was placed on the stove and preheated until the surface temperature reached approximately 140°C, as monitored by the thermometer. Once the desired temperature was attained and stabilized, the plate need to be put oil before introducing the powder form of the composite, then size reduced composite granules was carefully weighed and evenly spread onto the lower plate. Before placing the composite material, a thin layer of lubricating oil was applied to facilitate easy removal after molding. The material was then heated for about 15min to ensure uniform softening. Subsequently, the upper plate, preheated separately for about 2min, was positioned on top of the lower plate, and the assembly was manually compressed for 5min, the applied pressure was not instrumentally controlled. After compression, the entire mold was immediately immersed in a water container for 20min to promote rapid cooling and improve dimensional stability. Once cooled, the plates were gently separated to release the molded composite specimens. This procedure was repeated until 19 prototype samples were successfully fabricated. Although the process required significant manual effort and precise temperature control, all prototype composites were produced successfully with consistent shape and structure.

3.7. Composite Characterization

The fabricated rHDPE:CSS composite were analyzed to evaluate their mechanical performance, thermal, morphological, chemical structure and water absorption test also evaluated the fabricated composite. The tensile and flexural tests were assessed by a UTM and texture analyzer machine. TGA was used to assess the thermal stability of developed composites, and DSC was study their thermal transition behavior and crystallization characteristics**.** SEM images of all the fractured samples indicate the microstructural features of CSS fillers in matrices.

FTIR analysis was used to identify the functional groups present in rHDPE:CSS composites and to examine possible chemical interactions between the HDPE matrix and CSS filler. Water absorption test Measures the percentage of water absorbed by the material over time, which is important for applications in humid or wet environments. It directly affects its performance, usability, and compliance with standards for intended applications.

3.7.1. Mechanical Testing

The mechanical testing of the fabricated composites samples evaluated to determine their strength and suitability for practical applications. Since these composites are intended for use in load-bearing and semi-structural applications as furniture components and interior construction panels, deal their behavior under tension and bending is essential. Tensile and flexural tests were primarily selected for evaluation methods. Both are important performance indicators for wood polymer composites exposed to practical service conditions. The key mechanical properties which describe the material's load-bearing capacity, stiffness, rigidity and ductility of the composite material based on stress-strain measured during testing. Both the tensile and flexural tests were conducted at a strain rate of 0.05–0.25%, which is commonly applied in polymer composite studies to ensure stable and accurate strength measurement. Tensile property was further used as the response variable for process optimization using RSM, while flexural test results are presented in the Appendix for supplementary evaluation of the composite performance.

3.7.1.1. Tensile Test

The tensile properties of the rHDPE:CSS bio-composites were evaluated according to the ISO 527-2 standard, which specifies the procedure for determining the tensile strength, tensile modulus, and elongation behavior of plastics and polymer composites under uniaxial loading. This standard was selected because it is widely applied to rigid and semi-rigid thermoplastic composites**,** ensuring the generation of reliable, reproducible and comparable mechanical property data for HDPE-based bio-composites. The specimens were 15 samples prepared by compression molding and then cut into a dumbbell-shaped geometry. Following the dimensional proportions and stress concentration principles specified in the ISO standard in reference to ISO 291, the tensile specimens were conditioned at (23 ± 2) °C and (50 ± 5) % relative humidity for

at least 48 h prior to testing in order to minimize the influence of moisture on the mechanical properties. Tensile tests were conducted using a Universal Testing Machine (UTM, Model: ETM-50, Manufacturer: China) equipped with a load cell of 50kN. The specimens were clamped firmly at both ends, and under uniaxial tension at a constant cross-head speed of 5 mm/min. During testing, the applied load and corresponding displacement were continuously recorded by the testing software and subsequently converted into stress-strain curves within a strain rate range of 0.05% to 0.25%**.** The tensile strength, tensile modulus, and elongation at break were determined from the resulting stress-strain data.

3.7.1.2. Flexural Test

The flexural properties of the rHDPE:CSS bio-composites were determined according to ISO 178, which specifies method for measuring the flexural strength and flexural modulus of the material under bending loading. This standard is widely applied to polymer composites reinforced with natural fillers and provides reliable, reproducible and comparable flexural performance data for HDPE-based bio-composites; therefore, it was selected for this study. Flexural testing was carried out using a Texture Analyzer (Model: TA Plus, Lloyd Instruments, England) equipped with a 5 KN load cell and fitted with a three-point bending fixture. The test specimens were positioned on two supports, and the support span length (L) was set to 16 times the specimen thickness (L=16t) in accordance with ISO 178 requirements to ensure uniform stress distribution during bending. The test was conducted at a constant crosshead speed of 5mm/min, corresponding to a strain rate between 0.05% and 0.25%, thereby ensuring quasi-static loading conditions; the applied load and mid-span deflection were continuously recorded using the machine's data acquisition software. The flexural strength and flexural modulus of the composites were calculated from the resulting load-deflection curves.

3.7.2. Scanning Electron Microscope (SEM) Analysis

By using Scanning Electron Microscopy (SEM) to examine the interface between the rHDPE and coffee silver skin at a microscopic level to assess the quality of bonding and dispersion of CSS within the HDPE matrix. Poor bonding can lead to weak mechanical properties, so SEM analysis is crucial to understanding material performance.

3.7.3. Thermogravimetric Analysis (TGA)

Assess the thermal stability and decomposition temperature of the bio-composite. Thermogravimetric Analysis (TGA) will be used to evaluate the thermal stability and decomposition behavior of the bio-composite, a small sample of the composite is heated at a controlled condition. The analysis will focus on key temperature ranges: moisture loss (50°C-150°C), filler decomposition (200°C-400°C), and polymer degradation (300°C-700°C). This ensures the composite is thermally stable for furniture and interior applications.

3.7.4. Differential Scanning Calorimetry (DSC) Analysis

Differential scanning Calorimetry (DSC) was used to evaluate the thermal transitions and crystallization behavior of the fabricated composite. The samples were heated and cooled under a nitrogen atmosphere at a controlled heating-cooling rate, and the melting temperature (Tm), crystallization temperature (Tc), melting enthalpy (ΔHm), crystallization enthalpy (ΔHc), and degree of crystallinity (Xc) were determined from the resulting thermo grams.

3.7.5. Fourier Transform Infrared Spectroscopy (FTIR) Analysis

Fourier Transform Infrared (FTIR) spectroscopy was employed to characterize the functional groups present in the fabricated composite containing coffee silver skin filler. The analysis was conducted using an FTIR spectrometer in the wavenumber range of 4000-500 cm^{-1}.The obtained spectra were analyzed to identify characteristic absorption bands of the polymer matrix and lignocellulosic filler, as well as to evaluate possible interfacial interactions within the composite material.

3.7.6. Water Absorption Test

This test assesses how much water the bio-composite absorbs, which is important for applications in humid or wet environments, measuring the percentage of water absorbed by the material over time. Prepare specimens according to standardized dimensions (ASTM D570 for plastics). Immerse the samples in water at room temperature (23°C) for a specified duration for 24, 48 and 72 h.

3.8. Experimental Design and Optimization Approach

This study focused on three independent variables, which are temperature, filler ratio, and particle size. The variables were selected from the previous studies (Gozdecki et al., 2025a; Kumar et al., 2024; Ayana et al., 2022; Hejna et al., 2021; Sohn et al., 2019; Sarasini et al. 2018). Tensile strength was selected as the primary response variable for the experimental design and optimization process; other mechanical properties, including tensile modulus, flexural strength and flexural modulus, were measured for supplementary analysis are presented in the Appendix. Box-Behnken Design (BBD) was employed for the optimization of Tensile strength. BBD is a structured statistical method to model complex interactions with a limited number of experimental runs. This design enhances efficiency compared to trial-and-error approaches, particularly suitable for experimental studies material resources, time and energy consumption must be minimized. In addition, it helps to identify how different factors interacts each other to influence the responses and the development of a predictive model for the response variable. The design matrix included 15 experimental runs. Among 15 runs, center points are repeated to estimate experimental error. The responses were analyzed using Design-Expert software. In the design expert, ANOVA was conducted to assess the significance of each factor. This study investigated the influence of three processing parameters: Temperature, particle size and Filler ratio on the tensile strength of fabricated bio-composite, also the effect and interaction among these variables were analyzed to determine the optimal processing condition for enhanced material properties. The independent variables was assessed at three levels where the molding temperature ranged between 140°C to 180°C, particle size of CSS ranged between 100 and 500µm, and Filler ratio was set from, 10wt.% to 30wt.% of CSS loading.

Table 3.1: Factors and levels in the BBD for Tensile strength

Factors	**symbol**	**units**	**Levels**		
			Low (-1)	Medium (0)	High (+1)
Temperature	A	°C	140	160	180
Particle Size	B	µm	100	300	500
Filler Ratio	C	wt.%	10	20	30

3.8.1. BBD Matrix for Three Factors

The BBD experimental matrix, which consists 15 experimental runs with Tensile strength response presented in table 3.2.

Table 3.2: Design matrix for Tensile strength by BBD

	Factor 1	**Factor 2**	**Factor3**	**Response**
Run	A:Temperature °C	B: Particle size µm	C: Filler Ratio %	Tensile Strength MPa
1	180	300	30	*
2	160	500	10	*
3	160	300	20	*
4	160	500	30	*
5	160	100	30	*
6	180	300	10	*
7	160	300	20	*
8	180	500	20	*
9	180	100	20	*
10	140	300	10	*
11	140	100	20	*
12	160	300	20	*
13	140	300	30	*
14	140	500	20	*
15	160	100	10	*

A mathematical relationship was established between these factors and the corresponding response, which was the tensile strength. The tensile test of the composite evaluated and optimized through 15 experimental runs. After the completion of the experimental runs, the data was imported into Design Expert Software.

Therefore, the significance of the model was assessed using ANOVA. The ANOVA examined indicators such as the coefficient of determination (R^2 value), probability (p-value), and Fisher value (F-value).

3.9. Optimization of Tensile strength

The optimization approach was developed to address one of the specific objectives of this study: evaluating the effects of processing parameters on the tensile strength of the fabricated composite. All variables were systematically varied based on an interaction model predicted by the RSM through a Box-Behnken design was employed these effect and to determine the optimal combination of variables that maximizes tensile strength. This approach enables the development of a second-order polynomial regression model that describes the relationship between the response and the independent variables within the experimental domain. Analysis of variance (ANOVA) was used to assess the statistical significance and adequacy of the developed model. Based on the validated model, numerical optimization was performed to identify the optimal combination of processing parameters that maximizes tensile strength. Following numerical optimization, confirmatory experimental runs were conducted at the predicted optimal conditions to validate the developed model. In addition, composites were fabricated using treated CSS under the optimized processing parameters, and their tensile strength was experimentally determined. The predicted and experimental results were compared to assess model accuracy, while the influence of CSS treatment was further evaluated through comparison with composites produced from untreated CSS.

4. Result and Discussion

4.1. Characterization of CSS

4.1.1. Energy-Dispersive X-Ray Spectroscopy (EDX) Analysis

The EDX spectrum of untreated coffee silver skin (CSS 2) is dominated by strong carbon and oxygen peaks as shown in Figure 4.1(a), confirming the lignocellulosic nature of the biomass, Carbon (C) accounts for 67.33 wt. %, while oxygen (O) contributes 27.86 wt. %, indicating that raw CSS is primarily composed of organic constituents such as cellulose, hemicellulose, and lignin, which are rich in carbon-oxygen functional groups. The C:O ratio is consistent with previously reported EDX results for coffee-processing residues and reflects the polymeric backbone of polysaccharides and aromatic lignin structure (Zarrinbakhsh et al., 2016). Minor inorganic elements, including calcium (Ca) and sulfur (S), are also detected in low concentrations. Calcium is present at 4.58 wt. %, which can be attributed to naturally occurring mineral content absorbed by the coffee plant from soil and retained during processing. In addition, all silver skin samples exhibited sulfur content ranging from 0.2% to 0.29% (Papamatthaiakis et al., 2025).

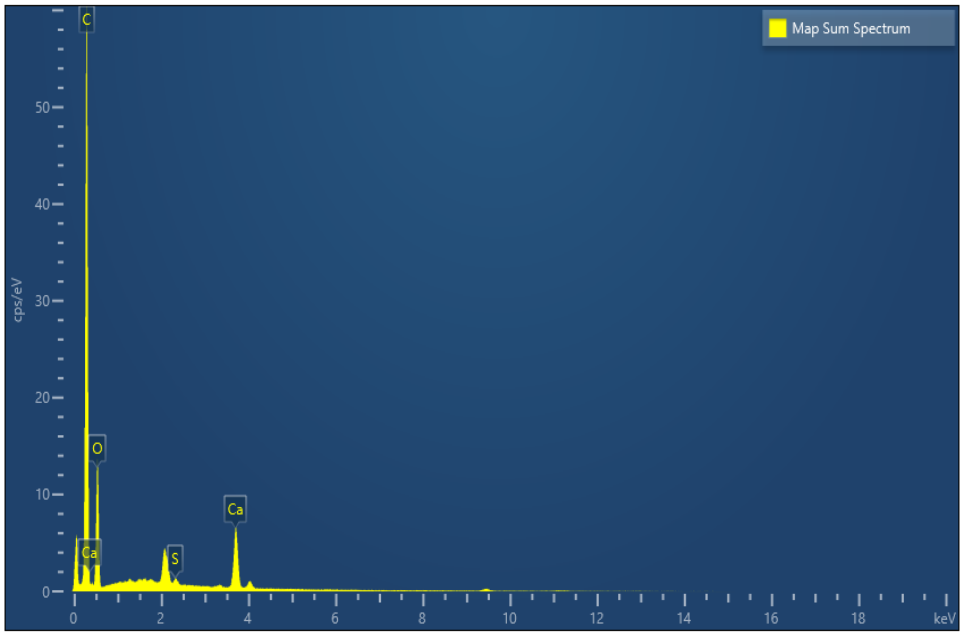


Figure 4.1(a): EDX of untreated coffee silver skin (CSS 2)

Sulfur appears in small amounts 0.23 wt. %, associated with sulfur-containing proteins or residual inorganic compounds inherent to the biomass. The absence of heavy or toxic elements suggests that raw CSS is environmentally relevant and suitable as sustainable filler for polymer matrix-based composite.

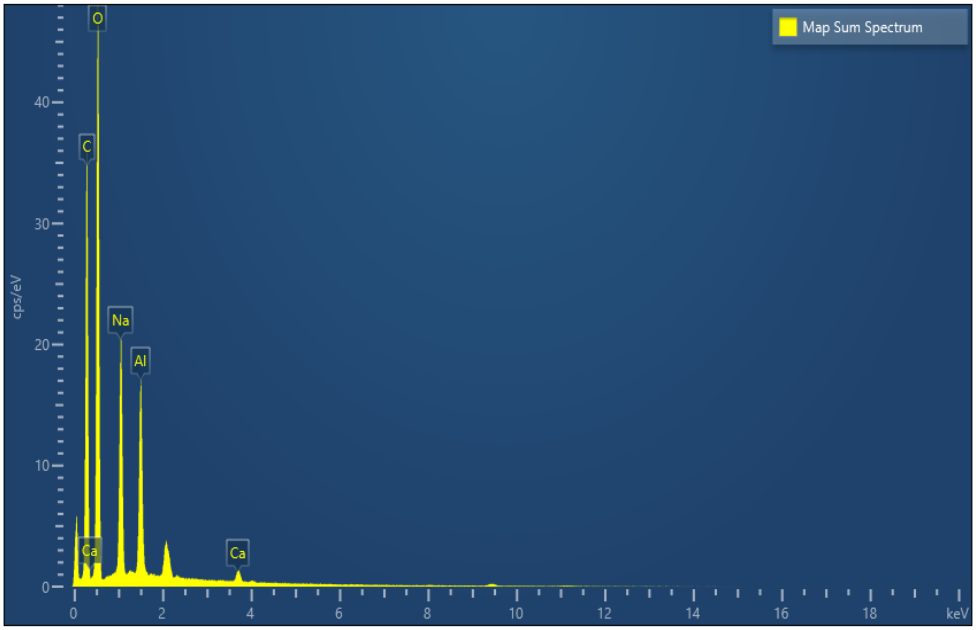


Figure 4.1 (b): EDX of treated Coffee silver skin (CSS 1)

Figure 4.1(b) presents the EDX spectrum of alkali-treated coffee silver skin (CSS 1), and Figure 4.1(a) corresponds to the untreated sample (CSS 2). Due to an effective surface modification of coffee silver skin, a clear change in elemental composition is observed. In the treated CSS, carbon and oxygen remain the dominant elements; but their relative proportions change substantially compared with the untreated CSS. The carbon content decreases from 67.33 wt. % in untreated CSS to 44.75 wt. % after treatment, while oxygen increases markedly from 27.86 wt. % to 44.54 wt. %. This shift shows the partial removal of non-cellulosic components such as lignin, waxes, and extractives during alkali treatment, accompanied by increased exposure of oxygen-rich functional groups associated with cellulose and hemicellulose. In treated CSS spectrum there is clear appearance of sodium (Na) peaks, corresponding to 6.37 wt. %. This confirms the interaction of NaOH with the lignocellulosic biomass surface and indicates residual sodium species after chemical treatment.

In contrast, calcium content is significantly reduced from 4.58 wt. % in untreated CSS to only 0.50 wt. % in treated CSS, suggesting the leaching or removal of mineral impurities during alkali washing. The existence of aluminum (Al) at 3.84 wt. % in the treated CSS assigned to trace inorganic residues introduced during processing/sample handling, as EDX is highly sensitive to surface-associated elements. This EDX results demonstrate that chemical treatment alters the surface elemental composition of CSS by reducing mineral impurities, introducing sodium (Na) species, and increasing the relative oxygen (O) content. These compositional changes enhance the interfacial adhesion between CSS and the rHDPE polymer matrix by improving surface reactivity and compatibility, which can contribute to improve the fabricated composite mechanical performance.

4.1.2. X-Ray Diffraction (XRD) Analysis

X-ray Diffraction (XRD) is used to determine the crystallographic structure, phase composition and crystallinity of the samples. Cellulose is crystalline in nature due to hydrogen bonding interactions and Van der Waals forces between adjacent molecules contrary to hemicellulose and lignin, which have an amorphous structure (Alghooneh et al., 2017). The diffraction angles (2θ) and peak intensities provide valuable information about the material's crystalline phases; inter planar spacing and degree of crystallinity. The crystallinity index is calculated using the peak area method, for understanding their structure-property relationships and potential applications (Woldie et al., 2025). The cellulose molecule is known to have crystalline and amorphous regions, Crystalline regions are responsible by a high tensile strength and represent cellulose less accessible to chemical attacks due to hydrogen strong interactions between the microfibers (F. Huang & Ragauskas, 2013). However, hemicellulose and other constituents of CSS exhibit an amorphous structure more easily degradable and susceptible to chemical attacks (Ballesteros et al., 2014).

As shown in Figure 4.2, after chemical treatment the coffee silver skin sample (CSS1) displays an increase in peak intensity compared to the untreated sample (CSS2). This enhancement in intensity implies a reduction in the amorphous content, due to the removal of non-crystalline components such as hemicellulose and lignin, leading to an increase in the relative crystallinity of cellulose.

Alkali treatments are known to remove amorphous regions and improve the crystalline structure of biomass, as observed in other lignocellulosic materials (Woldie et al., 2025).

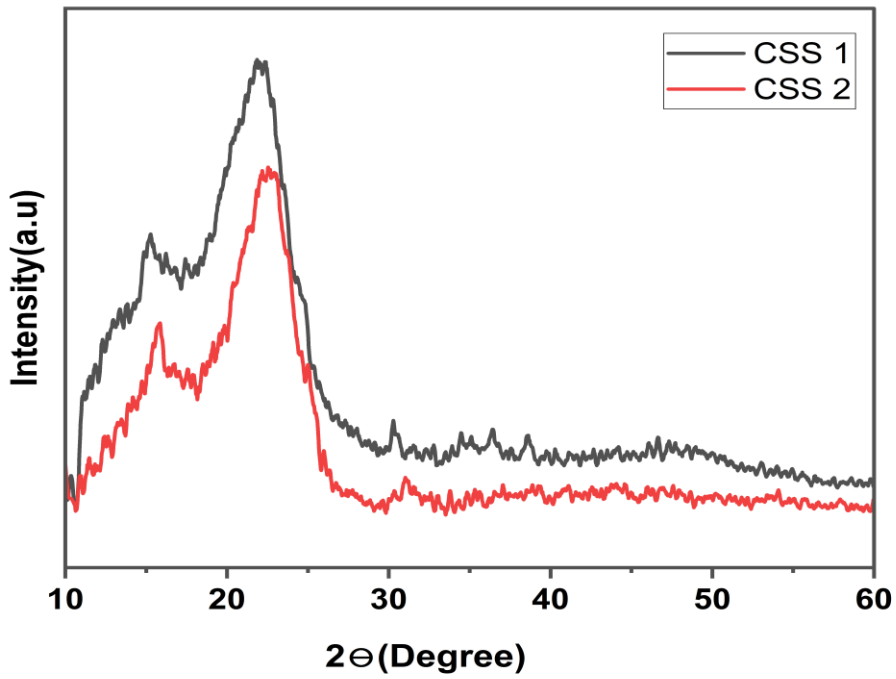


Figure 4.2: XRD Diffraction of CSS 1(Treated silver skin) and CSS 2 (Untreated silver skin)

The major crystalline peak occurred at 2θ = 23°, which represents the cellulose crystallographic plane and the second lower intensity at 2θ=15° with index of crystallinity 54% and 45% using segal empirical method, this improvement is attributed to the removal of amorphous constituents of CSS during alkali treatment (Alghooneh et al., 2017) and (Mateus et al., 2021) reported similar angles for the same crystallographic planes when analyzing coffee residues; the decrease in background noise and amorphous hump in the treated coffee residue sample also aligns. The higher crystallite index of treated CSS compared to untreated CSS is an important characteristic that contributes to their unique properties, such as improved mechanical strength, thermal stability, and barrier performance, desirable for applications including construction, furniture and interior design, which align for this study. The increased crystallinity 54%, observed after alkali treatment of CSS suggests improved filler rigidity and is expected to enhance interfacial interaction and reinforcement efficiency within the rHDPE matrix for polymer composite.

4.1.3. Fourier Transform Infrared Spectroscopy (FTIR) Analysis

FTIR technique was employed to analyze the functional groups and fingerprints of coffee silver skin before and after treatment on their wavelengths. The functional group typically seen above 1500 cm^{-1} in the infrared spectrum and finger prints were identified at wave length below 1500 cm^{-1}.

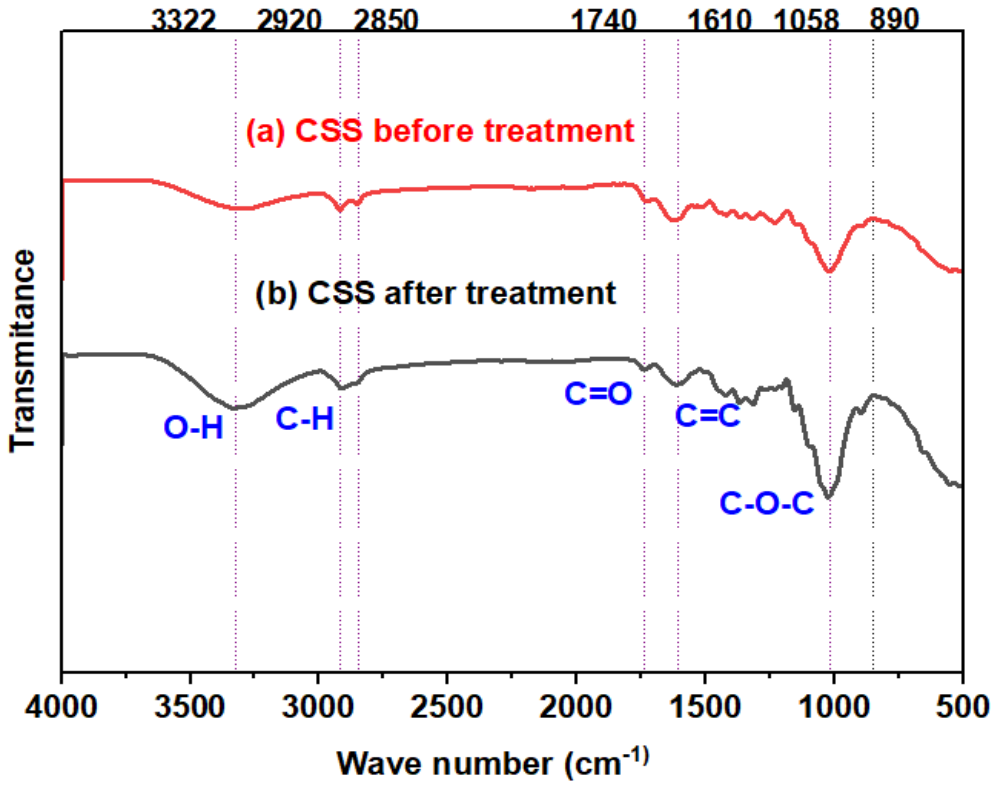


Figure 4.3: FTIR Spector of (a) raw CSS, (b) chemically treated CSS

The peak at around 3322 cm^{-1} the broad band corresponds to the O-H and N-H stretching vibrations, indicating the presence of hydroxyl groups associated with cellulose, hemicellulose and lignin with small contribution of N-H stretching of protein components in the lignocellulosic material (Kumar et al., 2024). After Chemical treatment of the coffee silver skins the intensity of the band around 3322 cm^{-1} slightly reduced and disruption of hydrogen bonds between the hydroxyl groups. The sharp bands at 2920-2850 cm^{-1} are presented aliphatic and alkyl (C-H) groups of cellulose and it attributed to asymmetric and symmetric CH stretching, associated with the methyl group in caffeine,

while the first band is reported to be related to lipids (Petaloti & Achilias, 2024). The latter band is reported in many caffeinated beverages and is usually associated to the methyl group in caffeine, while the first band is reported to be related to lipids, the presence of caffeine is confirmed by the carbonyl stretching at 1640 cm^{-1}. Such band is also associated to chlorogenic acid or adsorbed water (Totaro et al., 2019).

Additionally, the absorption band at 1740 cm^{-1}, corresponding to C=O stretching from ester and carbonyl groups in hemicellulose, is partially reduced, confirming hemicellulose removal during treatment (Gonçalves et al., 2021). The peaks at 1610 cm^{-1} and 1058 cm^{-1}, assigned to C=C aromatic skeletal vibrations and C-O-C stretching, respectively, indicate structural changes in lignin, hemicellulose and cellulose. These findings imply that the chemical treatment reduced lignin and hemicellulose content, enhancing the cellulose-rich nature of the material, the hemicelluloses group was partially removed from the surface after the NaOH treatment, as evidenced by the decreased carbonyl peak at 1653 cm^{-1} (Dominici et al., 2019) . These findings suggest that the chemical treatment partially reduced lignin and hemicellulose content, enhancing the cellulose-rich nature of the material.

The absorption bands at 890 cm^{-1} and 1546 cm^{-1} represent the presence of cellulose and lignin in CSS fillers, respectively. The sharp peak at 1058 cm^{-1} (C-O-C stretching) is associated with hemicellulose and cellulose. After chemical treatment of CSS the C-O-C stretching of cellulose and hemicellulose peak increase which represent lignin is successfully removed from the material. The broad band between 1100 cm^{-1} and 890 cm^{-1} is instead related to the C-O stretching and C-H rocking vibrations of cellulose (Sarasini et al. 2018). The absorption band at 890 cm^{-1} increased after alkali treatment of CSS, it represent the presence of cellulose improve from the above graph.

4.1.4. Scanning Electron Microscope (SEM) Analysis

The surface-structure morphology of raw and treated coffee silver skin was examined by scanning electron microscope; Figure 4.4(a–d) presents SEM image of untreated coffee silver skin (CSS 2) at different resolutions. The raw CSS illustrated a flake-like surface morphology with longitudinal fibrous structures and smooth surface (Sarasini, Tirillò, Zuorro, Ma, et al., 2018). From image (a) shows the overall surface morphology of untreated CSS, characterized by an irregular, flake-like structure with compact and heterogeneous regions. The surface appears densely packed, indicating the presence of lignin, hemicellulose, waxes, and other extractives that coat the lignocellulosic biomass of coffee silver skin and maintain the native structure. At higher magnification images (b) and (c), the untreated CSS exhibits agglomerated fibrous structures embedded within the matrix. Individual fibers are not clearly separated; imply that the cellulose network remains bound by non-cellulosic components. This compact and connected morphology reflects the structure of raw coffee silver skin, where the binding materials block fiber separation and limit surface exposure.

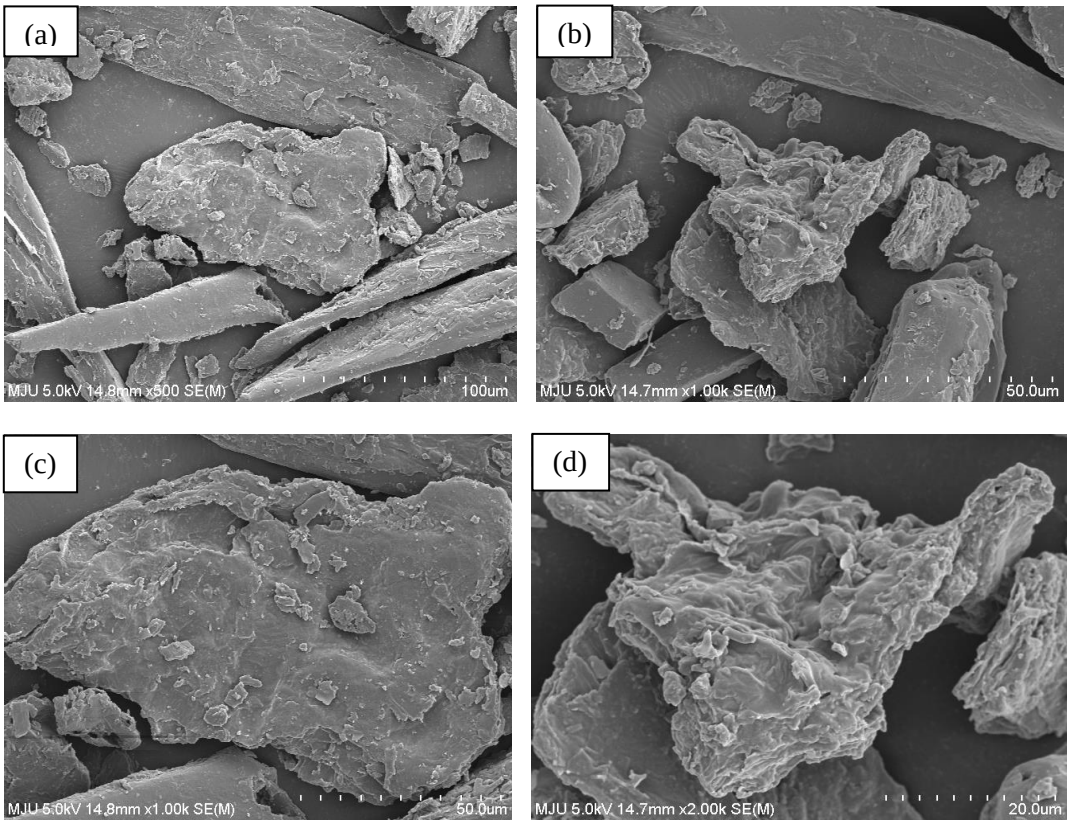


Figure 4.4: SEM images for CSS 2 at magnifications of: (a) 500, (b) 1.00k, (c) 1.00k, (d) 2.00

Image (d) reveals surface irregularities and adhered particles on the CSS surface, accompanied by limited porosity and an absence of fiber separation. These images indicate minimal disruption of the raw lignocellulosic structure prior to chemical treatment, which indicate cellulose fibers remain largely encapsulated within the lignin-hemicellulose matrix and that surface modification has not yet occurred. The limited porosity and absence of fibrillation imply restricted surface area and reduced interfacial accessibility, which can affect interfacial bonding between CSS biomass with HDPE polymer matrices. Similar surface characteristics have been reported for untreated agricultural residues and coffee by-products prior to chemical modification (Woldie et al., 2025) (Gonçalves et al., 2021) (Silverskin & Qiu, 2021).

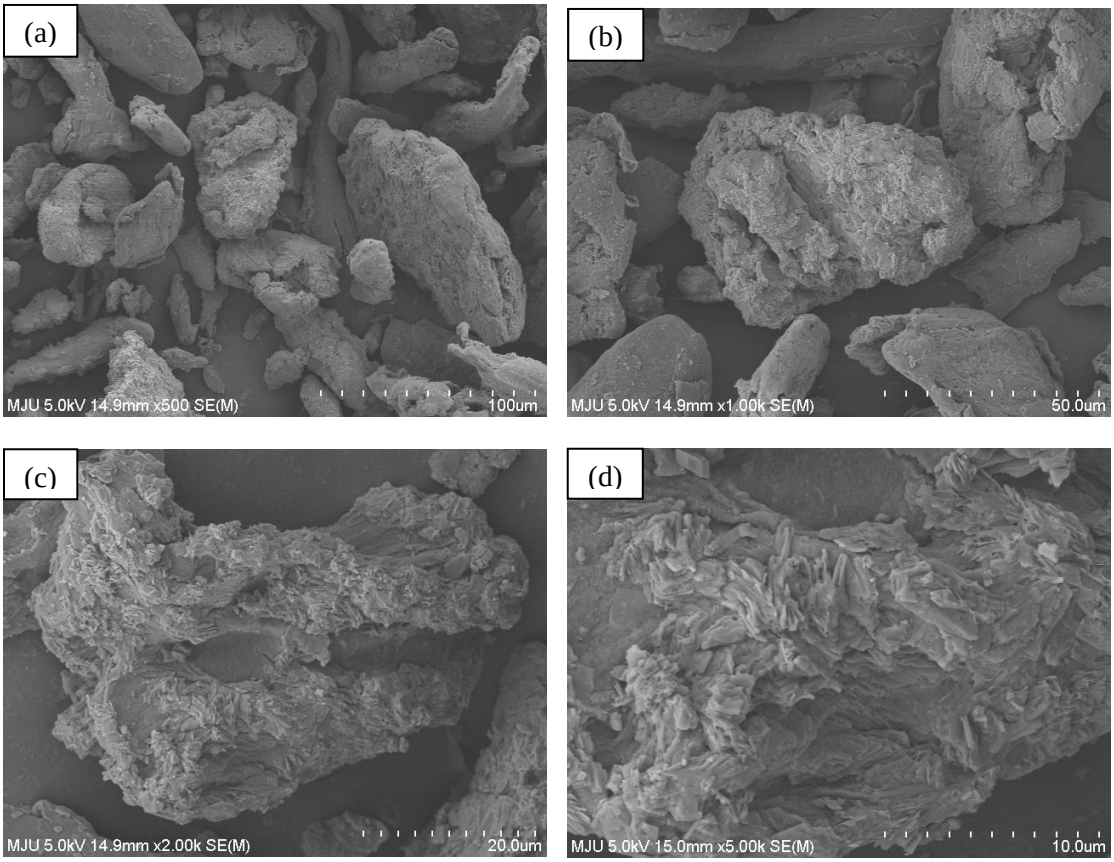


Figure 4.5: SEM images for CSS 1 at magnifications of: (a) 500, (b) 1.00k, (c) 2.00k, (d) 5.00k

After chemical treatment (Alkali treatment & Bleaching) the dense coffee silver skin was destroyed by removing impurities, hemicellulose and lignin as observed from Figure 4.5(a-d), image (a) illustrates the surface morphology of chemically treated CSS, where a noticeable transformation from a dense rigid to a more soft and roughened structure is observed.

Compared to the untreated CSS, the treated sample exhibits partial removal of surface layers, resulting in increased surface heterogeneity and the exposure of underlying fibrous features. At higher magnification images (b) and (c), the treated CSS shows improved fiber separation and a more open fibrous network. The cellulose fibers appear less agglomerated, with visible grooves and inter-fiber spacing, indicating partial removal of lignin and hemicellulose due to chemical treatment; it also increased surface roughness and enhanced fiber exposure of treated CSS.

Image (d) displays distinct surface roughness, micro-cracks, and localized porosity on the treated CSS surface. The absence of bonded particles and the presence of separated fiber features indicate significance surface modification via chemical treatment. These morphological characteristics denote an increase in effective surface area, CSS filler is interacted more with rHDPE matrix, possibly enhancing stress transfer within the composite. There is a clear difference in surface morphology is observed between untreated and treated CSS.

The untreated CSS exhibited a longitudinal, smooth, and flake-like surface morphology with embedded and agglomerated fibrous structures, limited porosity; however, the treated CSS shown a roughened and porous surface with improved fiber exposure, and visible fibrillation due to surface modification, which makes suitable physical interaction with the matrix along with increased contact surface area (Petaloti & Achilias, 2024).These changes verify the effectiveness of chemical treatment in removing non-cellulosic components mainly lignin and hemicellulose to enhance surface accessibility of CSS for fiber-matrix interaction and improved mechanical properties of the resulting bio-composite.

4.1.5. Thermogravimetric Analysis (TGA)

The thermal stability of treated and untreated coffee silver skin was assessed using Thermogravimetric analysis as shown in Fig. 4.6, it was found that a three-step thermal degradation and weight loss process for each sample. The first stage degradation occurred between 40-110°C for treated coffee silver skin (CSS 1), resulted in a weight loss of 5.87 % , for untreated coffee silver skin (CSS 2) accounted a weight loss of 5.2% between 40-130°C,this initial degradation is due to the evaporation of moisture and other volatile compounds (Kumar et al., 2024).

The slightly higher mass loss and narrower temperature range observed for treated CSS indicate enhanced moisture release due to structural modification and increased surface accessibility induced by chemical treatment. In the second stage presented significant weight loss: CSS 1 at 360°C a weight loss of 41.95% and CSS 2 exhibited 34.68% of a weight loss at 340°C, on this stage primarily to the thermal decomposition of hemicellulose and cellulose components which exhibited significant difference between treated and untreated CSS (Gonçalves et al., 2021).

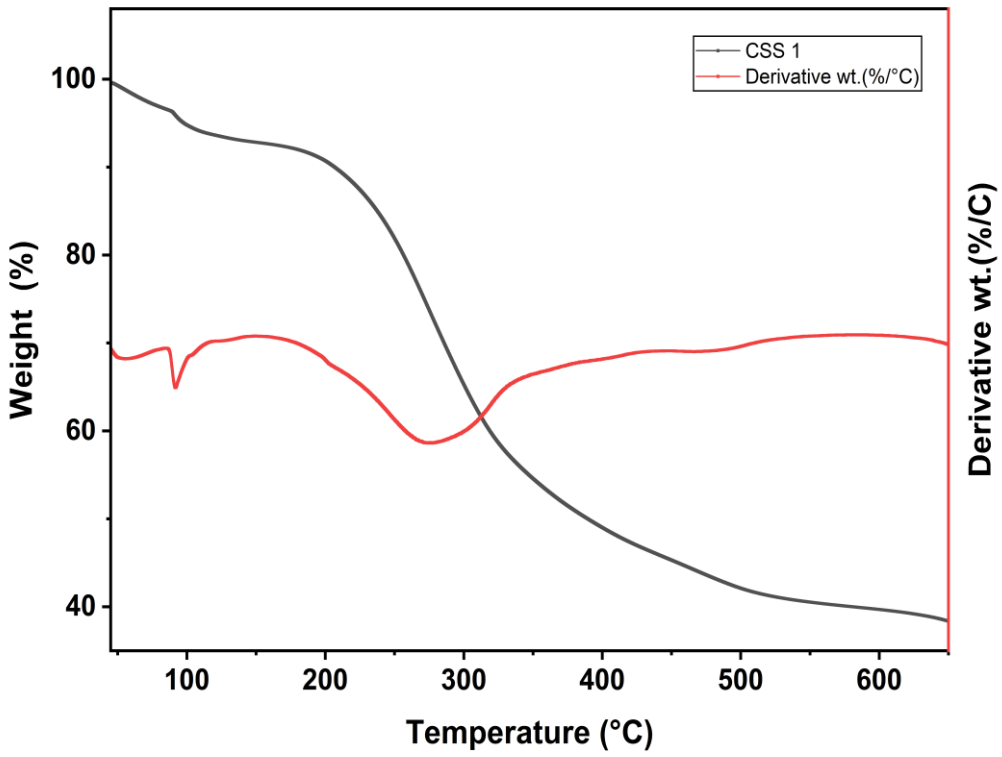


Figure 4.6: TGA and DTG of treated Coffee silver skin (CSS 1)

Untreated CSS showed marked weight loss, it consistent with the high content of thermally volatile polysaccharides especially hemicellulose and amorphous cellulose fractions. This is also supported by the sharp and intense DTG peak raw CSS (Gigante et al., 2020), indicating a rapid degradation rate and the presence of a large proportion of easily degradable polysaccharide components while, treated CSS shown a reduced weight loss of about 41.95% with the main degradation completed by 360°C, attended by a broader and less intense DTG peak.

The decrease in weight loss and change of the DTG profile attributed to the partial removal of hemicellulose, disruption of amorphous regions, and an increase in the relative crystallinity of cellulose induced by chemical treatment, which decrease the fraction of easily degradable components of CSS and alter the degradation kinetics of the remaining cellulose.

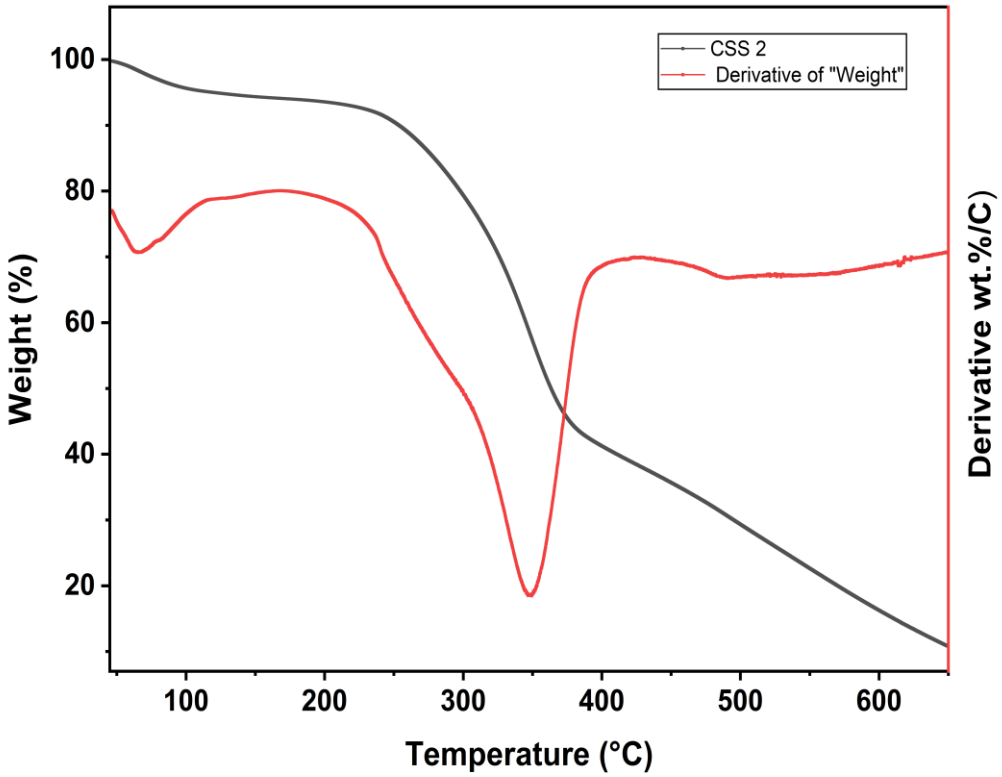


Figure 4.7: TGA and DTG graph of untreated coffee silver skin (CSS 2)

The third degradation stage, occurring at higher temperature between 370-600°C untreated coffee silver skin (CSS 2) exhibited 31% of a weight loss which reflect the gradual thermal breakdown of lignin and associated aromatic structures. However, treated coffee silver skin (CSS 1) showed a significantly lower weight loss of about 13.88 % over the temperature range of 400-600°C, accompanied by a reduced DTG intensity. This behavior indicates enhanced char stability and increased fixed carbon content in treated CSS, which attributed to lignin condensation and structural rearrangement induced by chemical treatment. The reduced mass loss at elevated temperatures proves the enhanced thermal stability of chemically treated CSS (Sarasini et al., 2018.), is favorable for its application as reinforcing filler in polymer composites processed at high temperatures.

The slower thermal degradation and weight loss of CSS 1 compared to CSS 2, which approved an excellence thermal stability with decomposition temperature the shift in degradation behavior, together with the lower degradation rate observed also in DTG profile that indicate it can withstand heat without significant degradation. Its high crystallinity additionally contributes to mechanical stability making them suitable for application as reinforcing filler in polymer composite.

4.1.6. Differential Scanning Calorimetry (DSC) Analysis

The DSC curves of treated (CSS1) and untreated coffee silver skin (CSS2) are shown in Figure 4.8, both samples show a broad thermal transition without a clear melting peak, which is characteristic of lignocellulosic CSS biomass composed primarily of cellulose, hemicellulose, and lignin component. A small endothermic feature observed at the lower temperature region at 75-115℃ attributed to the removal of physically absorbed moisture and volatile components existing within the biomass structure.

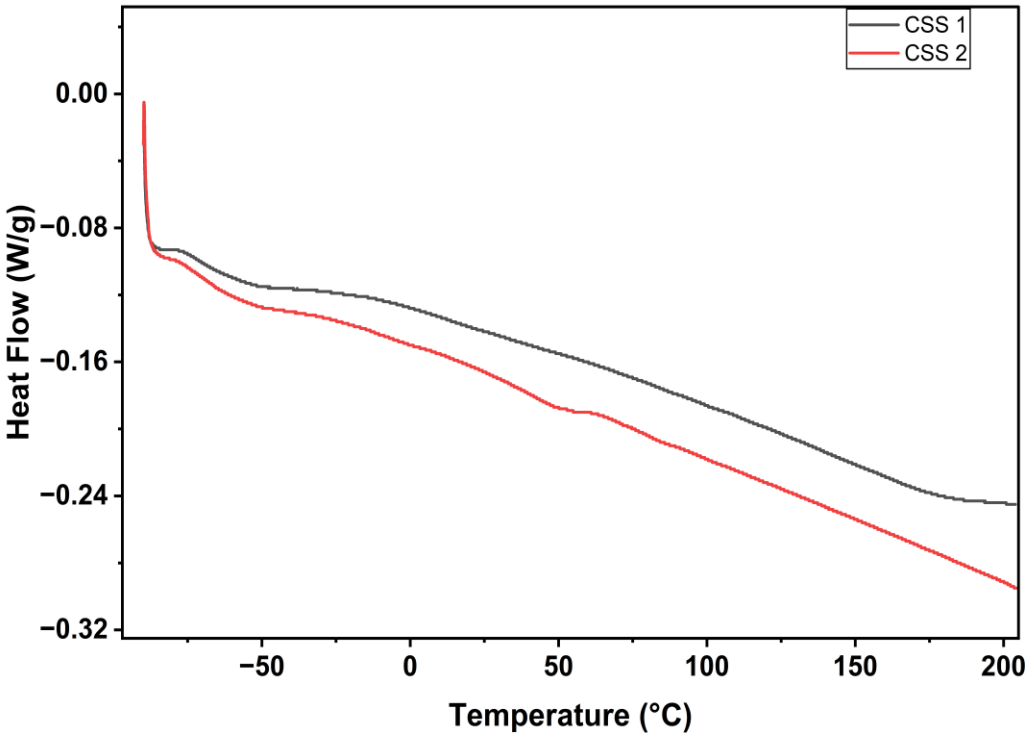


Figure 4.8: DSC curve of Alkali-treated (CSS 1) and untreated coffee silver skin (CSS 2)

Similar thermal behavior has reported for various lignocellulosic agricultural residues, where moisture evaporation and structural shift taken place before major thermal degradation (Yang, 2007) (Poletto et al., 2012). In comparison, the treated CSS sample exhibits a slightly different heat flow profile compared with untreated CSS. The reduction in heat flow intensity associated with the removal of hemicellulose, surface impurities, and other extractives during the chemical treatment process. Alkali (NaOH) treatment is widely reported to alter the internal structure of lignocellulosic fibers by partially dissolving hemicellulose and lignin components, which influence on their thermal response (Móczó et al., 2020). This structural modification improves the compatibility between the CSS filler and rHDPE polymer matrix, thus enhancing the interfacial interaction during composite fabrication.

4.2. BBD and Model Prediction

Table 4.1 presents the experimental runs from the Box-Behnken Design (BBD) that investigated the effects of Temperature, CSS particle size, and filler ratio on the tensile strength response of the rHDPE:CSS bio-composite. The experimental results show distinct trends associated with each factor, as well as their interactions, confirming the sensitivity of tensile performance to fabrication conditions.

Processing temperature exhibited a significant influence on tensile strength, with intermediate temperatures generally producing superior mechanical performance. Composites processed at 160°C consistently achieved higher tensile strength values, reaching up to 5.72-5.96MPa in Runs 3, 6, 7, and 12. In contrast, samples fabricated at lower temperatures (140°C) showed comparatively reduced strength in several cases, due to insufficient polymer melt flow and incomplete wetting of CSS particles. Similarly, excessively high temperatures (180°C) resulted in lower tensile strength in some runs, implying matrix over-softening and reduced interfacial integrity. These observations indicate that an optimal processing temperature is required to balance melt viscosity, dispersion, and stress transfer.

CSS particle size also played a critical role in determining tensile strength. Composites reinforced with medium-sized particles (300µm) generally shown higher tensile strength than those fabricated with finer (100µm) or coarser (500µm) particles. For instance, at comparable temperatures and filler ratios, samples with 300µm particles achieved tensile strengths above 5.5MPa, whereas those containing 500µm particles often showed values below 4.2MPa. The reduced performance at larger particle sizes attributed to poor interfacial contact and stress concentration effects, while very fine particles have agglomerated, limiting effective load transfer within the matrix.

The filler ratio demonstrated a nonlinear effect on tensile strength**.** Moderate filler loadings (approximately 20% CSS) produced the highest tensile strength values across multiple runs, indicating effective stress distribution and matrix-filler interaction at this level. Conversely, higher filler contents (30% CSS) resulted in a distinct reduction in tensile strength, with values dropping as low as 1.37MPa (Run 4). This decline is due to insufficient matrix enclosure and increased particle-particle interactions, which weaken the composite structure. Lower filler contents (10%) showed moderate strength values, indicating that excessive dilution of reinforcement limits mechanical enhancement.

Generally, the effect of individual factors was further influenced by their interactions; the BBD results denote that tensile strength is maximized at intermediate temperature, moderate particle size, and balanced filler loading, underlining the necessity of simultaneous optimization of all processing variables, highlighting the importance of optimizing processing parameters to enhance mechanical performance of rHDPE:CSS bio-composite.

Table 4.1: BBD experimental runs with factor levels and response data

	Factor 1	Factor 2	Factor 3	Response	
Run	A:Temperature °C	B:Particle size µm	C: Filler Ratio %	Tensile strength MPa	
				Actual	Predicted
1	180	300	30	3.02	2.93
2	160	500	10	4.10	4.01
3	160	300	20	5.1	5.31
4	160	500	30	1.37	1.60
5	160	100	30	2.50	2.59
6	180	300	10	5.96	6.19
7	160	300	20	5.72	5.31
8	180	500	20	3.24	3.10
9	180	100	20	3.63	3.63
10	140	300	10	5.19	5.28
11	140	100	20	3.88	4.02
12	160	300	20	5.72	5.31
13	140	300	30	5.50	5.27
14	140	500	20	4.12	4.12
15	160	100	10	3.68	3.45

4.3. Statistical Analysis for Tensile strength

4.3.1. Sequential Model Sum of Squares

The appropriate regression model for the tensile strength response was evaluated using the Sequential Model Sum of Squares (Type I). The linear model did not significantly improve the explanation of variation relative to the mean model (F = 1.41, p = 0.2912), indicating that linear terms alone were insufficient to describe the tensile strength behavior. Similarly, the 2FI model did not significantly enhance model performance compared with the linear model (F = 0.6651, p = 0.5966), showing that simple interaction terms did not provide additional explanatory power.

In contrast, the quadratic model produced a substantial improvement over the 2FI model, with a highly significant F-value of 31.52 and a p-value of 0.0011. This indicates that the inclusion of squared terms markedly increased the model's capability to capture curvature and nonlinear effects in the response surface. As a result, the quadratic model was identified as the most statistically appropriate and was suggested by the software. The cubic model was aliased (F = 0.4933, p = 0.7227), meaning that insufficient experimental degrees of freedom prevented independent estimation of all cubic terms.

Table 4.2: Sequential model of sum of squares for response model selection

Sources	Sum of square	Df	Mean of square	F - value	P - value	
Mean vs Total	255.28	1	255.28			
Linear vs Mean	6.45	3	2.15	1.41	0.2912	
2FI vs Linear	3.34	3	1.11	0.6651	0.5966	
Quadratic vs 2FI	**12.72**	**3**	**4.24**	**31.52**	**0.0011**	**Suggested**
Cubic vs Quadratic	0.2860	3	0.0953	0.4933	0.7227	Aliased
Residual	0.3866	2	0.1933			
Total	278.46	15	18.56			

4.3.2. ANOVA

The analysis of variance (ANOVA) for the quadratic model demonstrated that the fitted model was highly significant in describing the tensile strength of the produced rHDPE:CSS bio-composite. The model showed a strong comprehensive effect, with an F-value of 23.59 and a corresponding p-value of 0.0014, indicating that the selected processing variables and their polynomial components collectively provided a meaningful prediction of the response. Examination of the individual linear terms showed that temperature (A) had a statistically significant influence (p = 0.0285), suggesting that variations in processing temperature directly affected polymer-filler interactions and consequently the tensile performance.

Table 4.3: ANOVA for the quadratic model fitted to the experimental data.

Source	Sum of square	Df	Mean Square	F-value	P-value	
Model	**23.03**	**9**	**2.56**	**23.59**	**0.0014**	**significant**
A -Temperature	1.01	1	1.01	9.30	0.0285	
B - Particle Size	0.0925	1	0.0925	0.8524	0.3982	
C- Filler Ratio	5.35	1	5.35	49.29	0.0009	
AB	0.0992	1	0.0992	0.9148	0.3828	
AC	2.64	1	2.64	24.35	0.0043	
BC	0.6006	1	0.6006	5.54	0.0653	
A^2	0.1596	1	0.1596	1.47	0.2793	
B^2	11.92	1	11.92	109.94	0.0001	
C^2	1.32	1	1.32	12.14	0.0176	
Residual	0.5423	5	0.1058			
Lack of Fit	**0.2860**	**3**	**0.0953**	**0.7441**	**0.6169**	**Not significant**
Pure Error	0.2563	2	0.1281			
Cor Total	23.57	14				

The Filler ratio (C) showed a highly significant effect ($p = 0.0009$), highlighting the important role of filler loading in the composite strength. In contrast, particle size (B) did not exhibit a significant linear effect within the tested range.

Among the interaction terms, only the AC interaction was significant ($p = 0.0043$), indicating that the influence of temperature on tensile strength depended on the chosen Filler ratio level. This interaction highlights the importance of balancing thermal energy input with filler concentration to achieve optimal dispersion and interfacial bonding. The quadratic terms B^2 and C^2 were significant ($p = 0.0001$ and $p = 0.0176$, respectively). It's exposing clear curvature in the response surface, especially with respect to particle size and Filler ratio. This verifies that the tensile strength did not vary linearly across the factor ranges but followed a nonlinear trend usually observed in composite systems. The non-significant lack-of-fit ($p = 0.6169$) further validated that the quadratic model adequately shown the experimental data, making it suitable for predictive analysis and optimization.

According to the ANOVA result and model adequacy tests, a quadratic model was selected and A Second-order polynomial regression model was developed, to describe the relationship between the processing parameters and tensile strength of the fabricated bio-composite. The intercept represents the predicted tensile strength at the center levels of all factors. The linear coefficients indicate the individual effects of temperature, particle size, and Filler ratio, while the interaction terms describe their combined influence on tensile strength. The presence of quadratic terms confirms a nonlinear relationship between the processing parameters and the response.

$$\text{Tensile Strength (MPa)} = 5.31 - 0.3550A - 0.1075B - 0.8175C - 0.1575AB - 0.8125AC - 0.3875BC + 0.2079A^2 - 1.80B^2 - 0.5971C^2$$

Where, A is Temperature (°C), B is particle size (μm), C is Filler ratio (%).

4.3.3. Fit Statistics

Fit statistics were used to evaluate the overall quality and reliability of the model. An observed high R^2 value of 0.9770 indicates that approximately 97.8% of the variation in tensile strength efficiency explained by the developed model. This strong explanatory power was further supported by an Adjusted R^2 of 0.9356, which accounts for the number of terms in the model and remains high. In addition, the developed model illustrated predictive power with a predicted R^2 value of 0.7814. The predicted R^2 showed reasonable agreement with the Adjusted R^2, as their difference was less than 0.2, which regarded as a positive indicator of model reliability.

The Adequate Precision, an important measure of the signal-to-noise ratio and model quality, was 17.0786. The ratio significantly exceeds the acceptable ratio of 4. This indicates that a satisfactory signal can be obtained, validating the model's suitability for exploring the design space. Other measures, such as a standard deviation of 0.3293 and a coefficient of variation (C.V. %) of 7.95, were also observed. Together, these fit statistics provide strong evidence for an excellent model fit, good predictive capability, and overall reliability in representing the tensile strength.

Table 4.4: Fit Summary for the developed model

Std. Dev.	0.3293	R^2	0.9770
Mean	4.14	Adjusted R^2	0.9356
C.V. %	7.95	Predicted R^2	0.7814
		Adequate Precision	17.0786

4.2.4. Diagnostic Plots

For assessing the accuracy of the developed model, three diagnostic plots that correspond to the most important regression assumptions were chosen. The plots are ideal for the demonstration of the model's reliability.

4.2.4.1 Normal Probability Plot of Residuals

The normal probability plot of residuals was used to assess the model assumption. This plot provides a visual analysis of whether the residuals, (identified as the differences between the observed and predicted values), follow a normal distribution. The best fit for the data is the residual points align along a straight diagonal line across the plot. A close alignment with this line indicates that the residuals are normally distributed, which is a vita assumption for the validity of ANOVA and other statistical analyses. As shown in Figure 4.9, the residual points are observed to lie reasonably close to the straight line. Therefore, it indicates that the normality assumption is satisfied and confirms the suitability of the model for subsequent analysis.

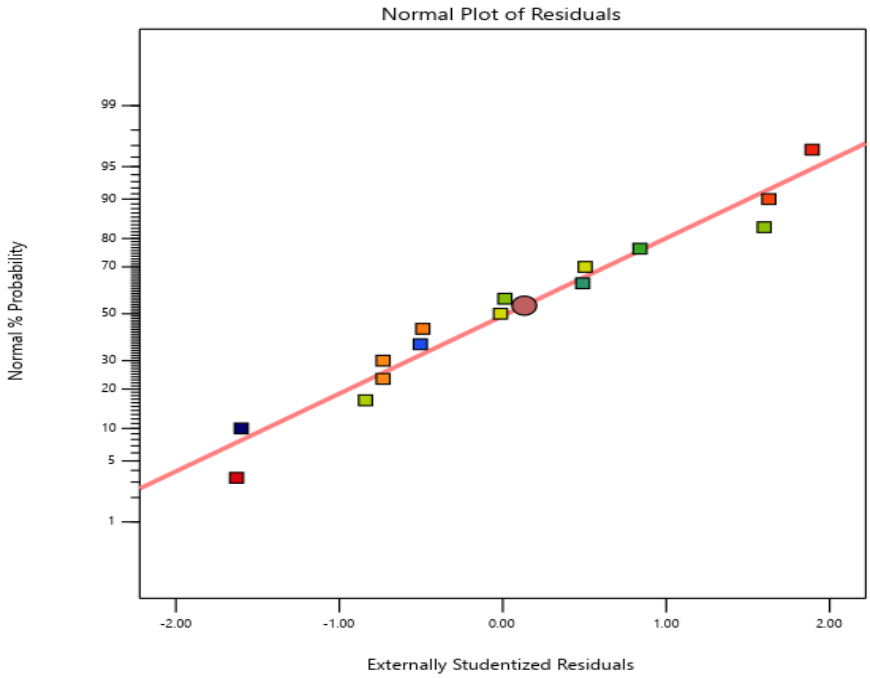


Figure 4.9: Normal probability plot of residuals for Tensile strength

4.2.4.2 Residual value Vs Run plot

The residuals versus run number plot was analyzed to evaluate the independence of errors and to identify any systematic trends during the experimental runs. In this plot, the externally studentized residuals are randomly dispersed around the zero-reference line across the entire run order. Irregular, cyclic behavior, and time-related trend is observed; it indicates that the residuals are independent of the run sequence. In addition, all residual values lie within the boundaries, indicating the absence of exceptional observations. The random distribution of points involves that the experimental conditions were stable throughout the runs and that it have no invisible variables affected the response. Therefore, this plot confirms that the assumption of independence of residuals is satisfied, supporting the reliability of the experimental design and the validity of the developed statistical model.

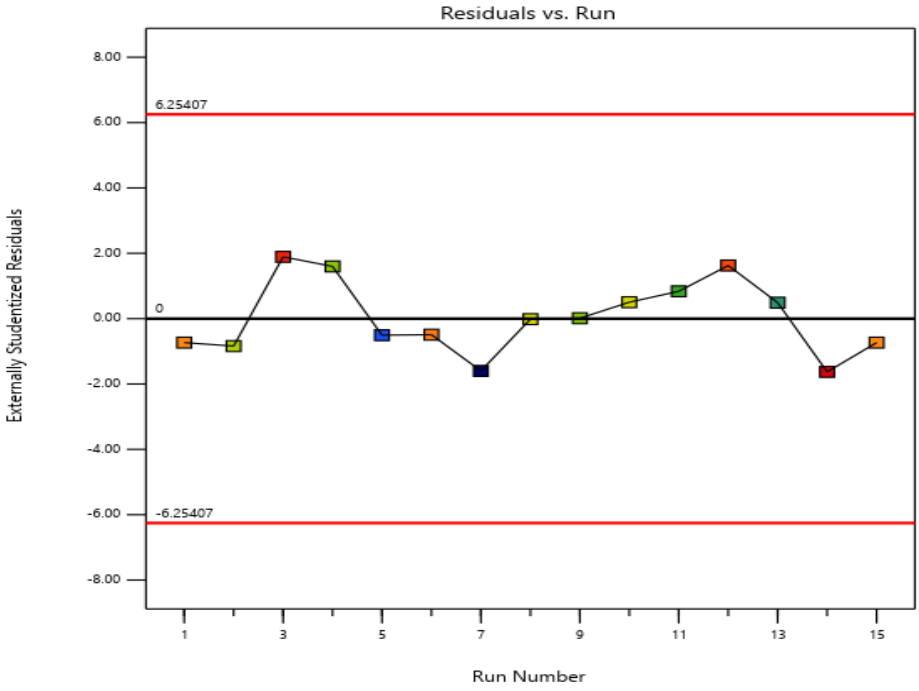


Figure 4.10: Residual value Vs Run values for Tensile strength

4.2.4.3. Predicted Values Vs Actual Values Plot

The performance of the model was also assessed visually using the Predicted versus Actual plot shown in the Figure 4.11. This plot is a very simple way of showing how closely the predictions of the model matched up to what was actually observed during the experiments. In principle, if the model was perfect, all of the data points would be located on the 45-degree line, which is perfectly aligned with predicted value equaling actual value. For the plot that resulted from this study, the majority of the data points were seen to be very close to the ideal, straight-line relationship. The validity of the predicted values was found to be very closely aligned to the actual tensile strength as measured during the experiments.

The scatter of data points around the dashed line appeared fairly consistent again confirming some reliability to the model along the available range of tensile strength. Overall evidence was very promising; a developed model with good predictive capabilities, and reliable estimation for tensile strength of the produced bio composite.

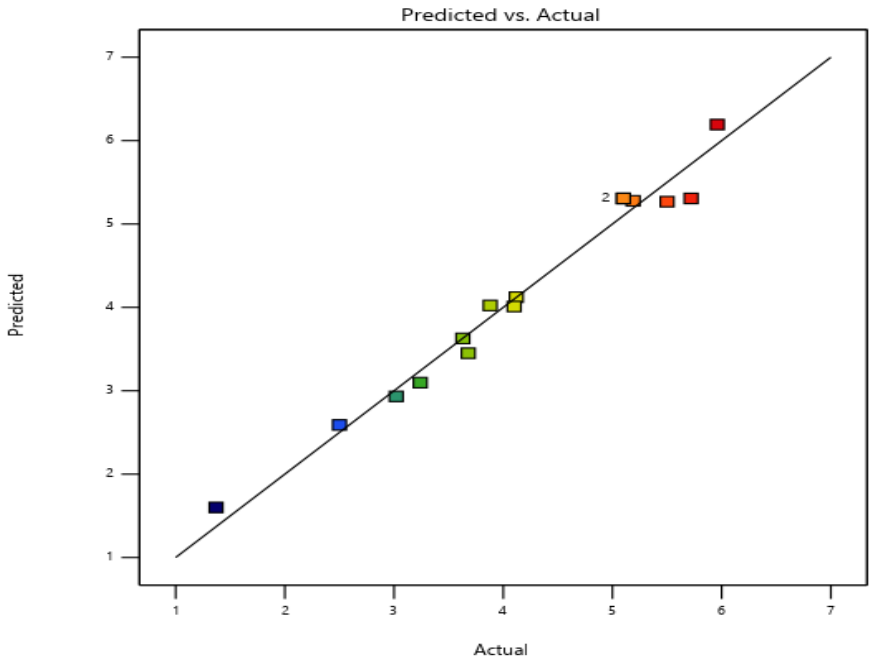


Figure 4.11: Actual vs predicted values for Tensile strength

4.2.5. Combined Effects of the Independent Variables

The complex associations among the different experimental factors and tensile strength were shown using a set of three-dimensional (3D) response surface plots as put in Figure 4.12. Response surface plots are unique features of RSM, and they perform the function of demonstrating how two factors change while a third factor is held constant and how all three factors influence the response, including the identification of maximum or minimum operating conditions. The first 3D surface plot a, the tensile strength affected by A: temperature (°C) and B: particle size (µm), while holding factor C: Filler ratio constant at 20%. The surface exhibited that tensile strength is strongly influenced by the interaction between temperature and particle size. Strength increases with temperature up to about 160–170°C, where improved HDPE flow enhances interfacial bonding.

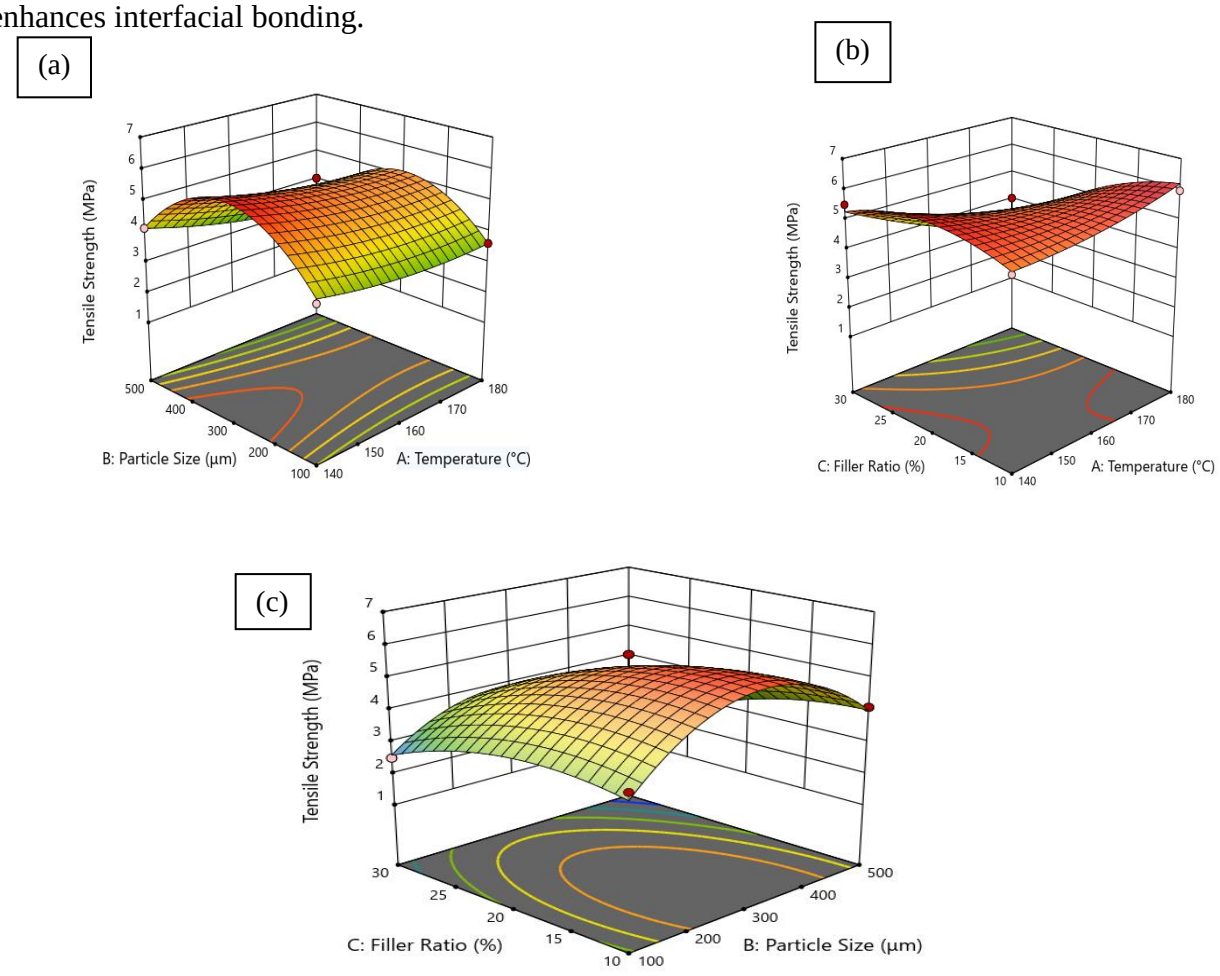


Figure 4.12: 3D surface plots for combined effects of factors on Tensile strength: (a) temperature and particle size, (b) temperature and Filler ratio, (c) particle size and Filler ratio

Medium particle sizes (250–350µm) output the highest tensile strength, while very small or large particles reduce performance. At higher temperatures and large particle sizes, tensile strength declines slightly, indicating possible thermal degradation and poor filler-matrix interaction.

The second plot b, shows tensile strength with A: temperature (°C) and C: Filler ratio (%) with a constant of factor B: particle size at 300µm. this surface illustrates the combined effect of temperature and Filler ratio on tensile strength. Tensile strength increases steadily with temperature, reaching its highest values above 170°C, as shown by the orange-to-red regions. However, the Filler ratio shows a moderate effect: lower and mid-range ratios (10-20%) yield relatively lower tensile strength, while higher ratios improve performance. The curved surface and non-parallel contour lines indicate a noticeable interaction, where high tensile strength is achieved primarily at elevated temperatures and higher filler ratios.

The third surface plot was c, representation of tensile strength with B: particle size (µm) and C: Filler ratio (%). keep constant the factor of A: temperature at 160°C, Tensile strength increases with moderate particle size, reaching higher values around 300-400µm, as indicated by the yellow-to-red regions. likely, the Filler ratio exhibits a moderate, curved influence: very low and very high ratios reduce strength, while intermediate levels yield improved performance. The curved surface and elliptical contour lines highlight the interaction between the two factors, indicating that maximum strength occurs both on mid-range Filler ratios and particle size.

4.2.6. Optimization and Experimental Validation of Tensile Strength

After the study of the interaction effect of operating parameters, statistician optimization of those process parameters was carried out to investigate the optimum condition for maximizing mechanical performance of the rHDPE-coffee silver skin bio-composite. As discussed above, The interaction effect of tensile strength of the bio-composite material increase with increasing temperature and particle size but decrease with increasing Filler ratio. To achieve the best conditions for maximize tensile strength such type of fluctuation should be optimized. The process parameters were adjusted within the range studied and the goal was to maximize the tensile strength.

From Table 4.5 provides a summary of the optimization process conditions, including the ultimate goals and the lower and upper limits of the factors affecting the response (Tensile strength). The optimum process conditions were selected based on the highest value of the tensile strength that obtained from the solutions generated using design expert software.

Table 4.5: optimization criteria for factors and response

Factor / Response	Goal	Lower Limit	Upper Limit
A: Temperature	is in range	140	180
B: Particle Size	is in range	100	500
C: Filler Ratio	is in range	10	30
Tensile Strength	Maximize	1.37	5.96

According to the optimization process, a Temperature of 175.97°C, particle size of 285.82 µm, and Filler ratio of 10.44% was identified as optimal condition for maximal tensile strength of 5.99MPa. To validate this model prediction, two confirmatory experiments were conducted. The individual conformations run for tensile strength were recorded as 5.98MPa and 6.03MPa. The average of these two experimental runs of tensile strength was 6.01MPa. A close agreement was observed between the predicted and experimentally verified tensile strength, with a 0.33% variance, this is a promising agreement validated the model's predictive capability.

Table 4.6: Predicted and observed values of factors and response in the optimization

Factors	Temperature (°C)	Particle Size (µm)	Filler Ratio (%)	Tensile Strength (MPa)
Predicted	175.97	285.82	10.44	5.99
Observed (1)	176.00	286.00	10.00	5.98
Observed (2)	176.00	286.00	10.00	6.03
Observed (Average)	176.00	286.00	10.00	6.01

4.3. Effect of Chemical Treated CSS on Tensile Performance of Bio-composites

The effect of alkali treatment of coffee silver skin (CSS) on the tensile performance of the resulting bio-composites were evaluated for address the specific objective of comparing treated and untreated reinforcements, bio-composite specimens were fabricated under the optimized processing conditions for tensile strength using both alkali-treated and untreated CSS. The tensile strength results obtained from these specimens were then compared to assess the influence of fiber surface modification on the mechanical performance of the composite. The tensile strengths of hardwood fibers reinforced HDPE composites increased gradually, and up to a maximum at 25% of fiber loading by volume, also the bio-composite tensile strengths were increased after surface modification (Ku et al., 2011).

These chemical treatments involve functional groups interacting with bio-filler structures to remove non-cellulosic elements and purify surfaces, promoting better contact and interaction between the filler and matrix, ultimately improving mechanical performance of the final product (Dominici et al., 2019 ; Ayana et al., 2022 ; Petaloti & Achilias, 2024). The improvement in tensile performance observed for the composite reinforced with chemical-treated CSS is consistent with the FTIR analysis of treated and untreated fillers. From Figure 4.3 a noticeable reduction in the intensity of peaks associated with lignin and hemicellulose components was observed, indicating partial removal of amorphous regions. In addition, the relative enhancement of hydroxyl-related functional groups suggests increased surface reactivity of the treated CSS.

Table 4.7: Tensile strength of Bio-composites reinforced with treated and untreated CSS

	Run	Tensile Strength (MPa)	Mean Tensile Strength (MPa)
Untreated CSS	1	5.98	
	2	6.03	6.01 ± 0.04
Treated CSS	1	6.68	
	2	6.73	6.71 ± 0.04

Based on Table 4.7, representation of tensile strength of the fabricated bio-composite reinforced with untreated and treated coffee silver skin under the optimized processing conditions. The observed improvement of mechanical performance of the fabricated rHDPE:CSS bio-composite assigned the interfacial bonding between the treated CSS and the HDPE matrix, improving better contact and interaction between the filler and matrix, also enhancing the dispersion of the bio filler, enabling more efficient stress transfer during tensile loading than the untreated lignocellulosic natural filler of CSS with HDPE polymer matrix. This result implies a substantial improvement in tensile strength due to the chemical treatment of CSS, which enhanced the interfacial adhesion and compatibility between the CSS filler and the rHDPE matrix.

4.4. Characterization of Composites

4.4.1. Scanning Electron Microscope (SEM) Analysis

Scanning electron microscope (SEM) was employed to examine neat rHDPE and the fracture surfaces of the rHDPE:CSS composites after tensile testing. The SEM analysis was conducted to examine the fracture morphology of neat rHDPE and rHDPE:CSS composites, focusing on filler dispersion, fiber-matrix interfacial adhesion, and the dominant failure mechanisms responsible for tensile behavior of the composites. The observed fracture morphologies were used to establish a correction between microstructural features and the tensile performance of the fabricated composites.

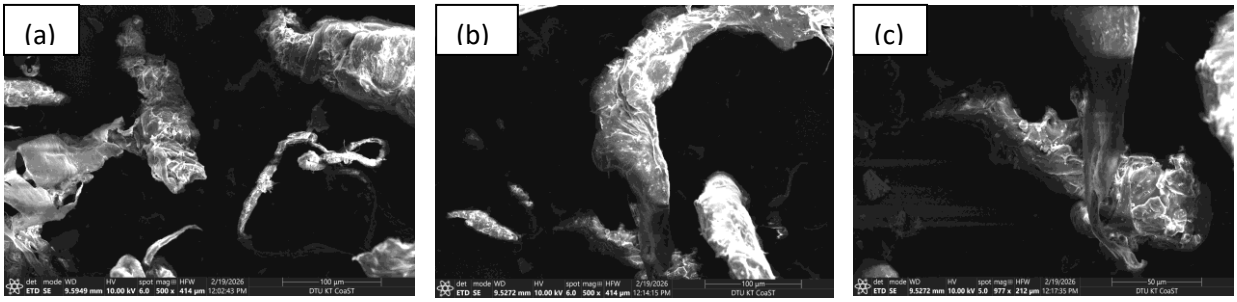


Figure 4.13: SEM image of neat rHDPE (a-c)

Figure 4.13 (a–c) shows SEM micrographs of neat rHDPE with lower and higher magnification, fracture surfaces show relatively smooth and homogeneous morphology, indicating ductile matrix failure without reinforcement particles.

The absence of interfacial voids and filler pull-out confirms that failure occurred primarily through matrix yielding. It serves as a reference for assessing the structural changes introduced by CSS incorporation.

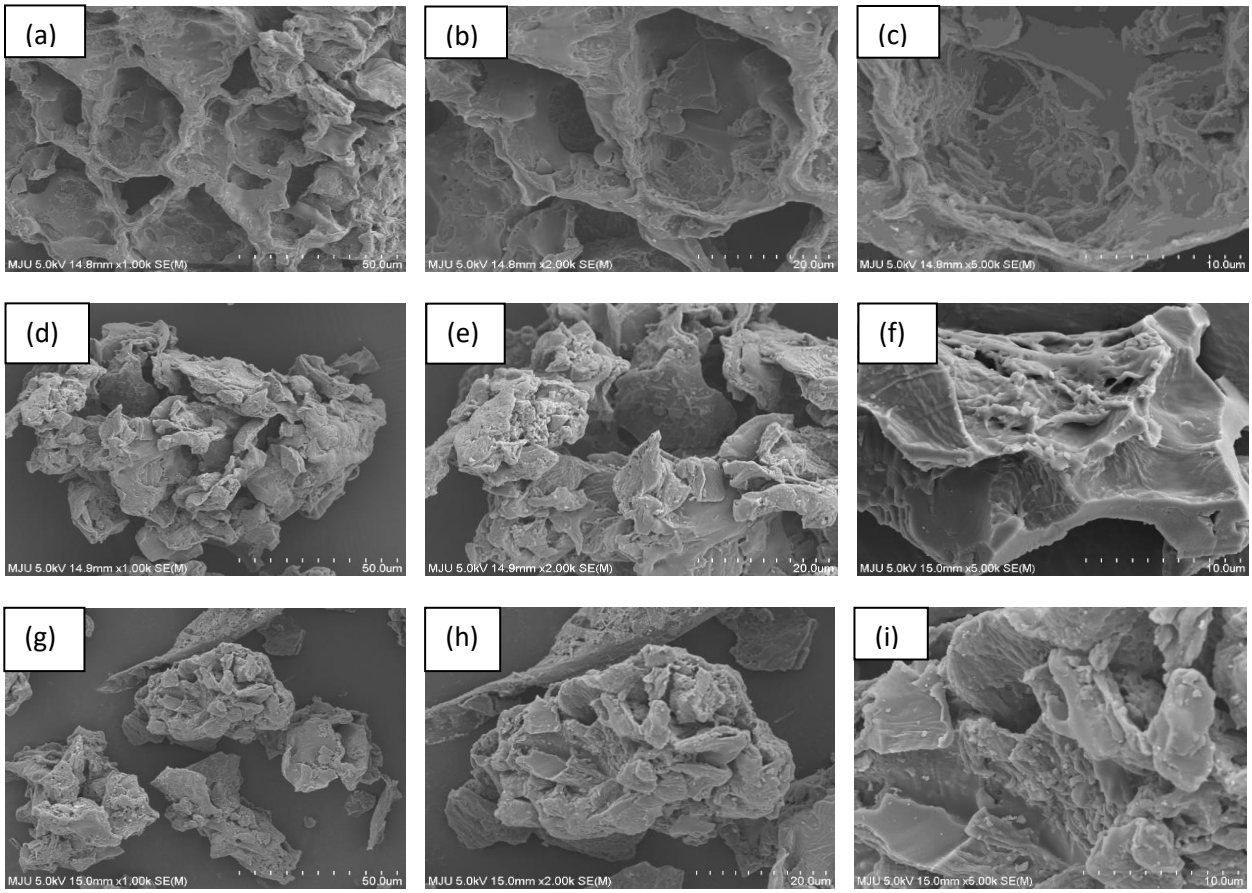


Figure 4.14: SEM images of rHDPE/CSS composites (a-c) rHDPE/10CSS, (d-f) rHDPE/20CSS, and (g-i) rHDPE/30CSS at magnification of: 1.00k, 2.00k and 5.00k

Figure 4.14 (a-c) shows the SEM micrographs of the fracture surface of the rHDPE/10CSS composite with medium filler size after tensile testing at different magnifications. At lower magnification image (a), the fracture surface appears relatively rough but continuous, with CSS particles well embedded within the HDPE matrix. The filler distribution is normally uniform, and no extreme agglomeration is observed. This morphology indicates good matrix continuity and implies that the low filler loading allowed effective wetting of the CSS particles by the rHDPE matrix. At intermediate magnification (b), the filler-matrix interface becomes more evident. filler particles remain largely anchored within the rHDPE matrix, with limited interfacial gaps/voids (L. Huang et al., 2016).

Only minor filler pull-out is observed, indicating that interfacial de-bonding was not the dominant failure mechanism. This suggests that mechanical interlocking and interfacial contact between CSS and rHDPE were sufficient to enable stress transfer during tensile loading. At higher magnification image (c), the fracture surface shows stretched polymer ligaments and localized plastic deformation of the recycled HDPE matrix nearby the CSS particles. The presence of matrix fibrillation indicates ductile fracture behavior, confirming that the polymer matrix participated actively in load bearing prior to failure. The limited extent of clean filler pull-out denotes the presence of effective filler-matrix interaction at this filler loading.

Figure 4.14 (d–f) presents SEM micrographs of the fracture surfaces of the fabricated rHDPE/20CSS composite with medium filler size at lower magnification image (d), the fracture surface exhibits a rough and heterogeneous morphology, indicating ductile fracture behavior of the rHDPE matrix. CSS particles are generally embedded within the polymer matrix, with limited but noticeable filler agglomeration. The fairly uniform distribution of filler at this loading indicates that stress transfer from the matrix to the filler occurred during tensile deformation. At higher magnifications (e) and (f), partially exposed CSS particles, interfacial gaps, and localized voids are observed. These features indicate that interfacial de-bonding occurred during tensile loading, although the limited extent of clean filler pull-out implies partial mechanical interlocking between the filler and rHDPE matrix (Gozdecki et al., 2025b).

The presence of stretched polymer ligaments and plastically deformed regions around the CSS filler indicates that the matrix contributed significantly to load bearing prior to fracture. However, the fracture surfaces of the rHDPE/30CSS with medium filler size (300µm); composites (g–i) show different morphology. At lower magnification (g), broad filler agglomeration and discontinuous matrix structure are shown, (Petaloti & Achilias, 2024).The increased concentration of CSS particles leads to reduced matrix continuity, which promote stress concentration and premature crack initiation under tensile loading (Saccani & Sisti, 2022).

At higher-magnification images reveal pronounced filler pull-out, larger interfacial voids, and micro cracks propagating along the filler–matrix interface. The advanced change in fracture surface morphology from rHDPE/10CSS to rHDPE/30CSS tightly correlates the observed tensile behavior.

At low filler loading (10 wt.%), uniform dispersion, good matrix continuity, and distinct matrix plastic deformation promoted effective stress transfer, resulting in optimal tensile strength under the investigated conditions. At intermediate loading (20wt.%), increased filler participation enhanced reinforcement effects but also introduced localized agglomeration and interfacial de bonding, leading to reduced ductility. further, excessive filler loading (30 wt. %) leads to agglomeration, interfacial de bonding, and brittle fracture, explaining the deterioration in tensile properties. These results confirm that the tensile performance of the rHDPE/CSS composites is strongly governed by filler dispersion and interfacial integrity, as evidenced by SEM analysis.

4.4.2. Thermogravimetric Analysis of rHDPE and its composite (TGA)

The thermal degradation behavior of neat rHDPE and rHDPE:CSS composites was evaluated using TGA and DTG analyses, to assess the influence of coffee silver skin (CSS) incorporation, on thermal stability of the fabricated composites. The first started in the temperature range of 50-130°C, primarily with the removal of physically adsorbed moisture and low-molecular-weight volatile compounds. Neat rHDPE showed a minor weight loss of around 0.4%, indicating the absence of moisture and volatile components within this temperature range the hydrophobic nature of rHDPE also supported by the nearly flat TGA curve and the absence of a distinct DTG peak, (Sameni et al., 2018), indicating excellent thermal stability under mild heating conditions.

In comparison, the rHDPE:CSS composite (CP) showed a slightly weight loss of approximately 0.67%, attributable to the presence of coffee silver skin as a lignocellulosic filler. Notably, the moisture-related weight loss of the composite remained below 1 %, which is substantially lower than that observed from raw CSS (6.7%) in the same temperature range; the reduction indicates that CSS within HDPE matrix effectively limit moisture retention and good interfacial synergy between them. The weak DTG response further confirms the limited presence of free or bound moisture in the composite system.

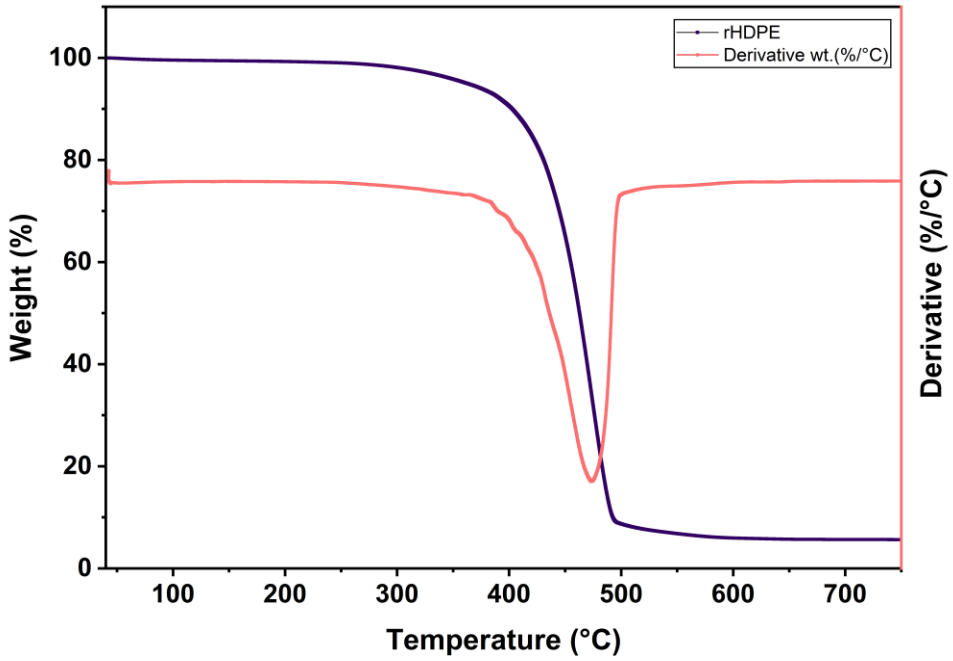


Figure 15: TGA and DTG graph of rHDPE

The second degradation stage occurring between 200-370°C for rHDPE is characterized by minor but measurable weight loss prior to the main polymer decomposition. It showed a limited weight loss of 4.8%, which assigned to the initial scission of weak polymer chain segments, removal of residual low-molecular-weight fractions and early oxidative the primary degradation of the polyethylene backbone. The absence of a sharp DTG peak in this region indicates that rHDPE remains thermally stable within this temperature range. The composite displayed a slightly higher weight loss of 7.2% at 200-350°C, with the onset of degradation occurring at a slightly lower temperature. This additional weight loss is mainly with the thermal decomposition of lignocellulosic components in CSS, particularly hemicellulose and amorphous cellulose fractions. which are to degrade within this temperature range, (Taylor et al., n.d.). The broader DTG response observed for the composite confirms overlying degradation processes of the biomass filler and the polymer matrix. Despite this increase, the comparatively low mass loss indicates that the composite maintains adequate thermal stability up to 350°C, supporting its suitability for typical thermoplastic processing conditions.

In the third stage degradation, represents the thermal decomposition of the polymer matrix and is characterized by rapid weight loss. For rHDPE, at temperature range of 375-480°C, with a maximum weight loss of 69.8%, similar degradation pattern was reported with some variation (Ayana, Ali, et al., 2024), corresponding to the random scission of polyethylene chains and the formation of volatile hydrocarbon products. This behavior is reflected by the sharp and intense DTG peak, indicating rapid degradation kinetics. It is confirming that this temperature region governs the thermal stability limit of the HDPE matrix, (Kord et al., 2017).

Similarly, the rHDPE:CSS composite shown its main degradation stage between 370-480°C, with a comparable weight loss of approximately 67.8%.The slight reduction in weight loss relative to neat rHDPE attributed to the presence of thermally stable lignin-derived char and inorganic residues originating from CSS, which contributes to higher residual mass at elevated temperatures. Furthermore, the interactions between rHDPE polymer matrix and the treated CSS filler restrict polymer chain mobility and suspend volatilization to a limited range. The affinity in degradation behavior between the composite and neat rHDPE indicates that the incorporation of CSS does not significantly compromise the thermal decomposition profile of the polymer matrix, while slightly enhancing char formation at high temperatures.

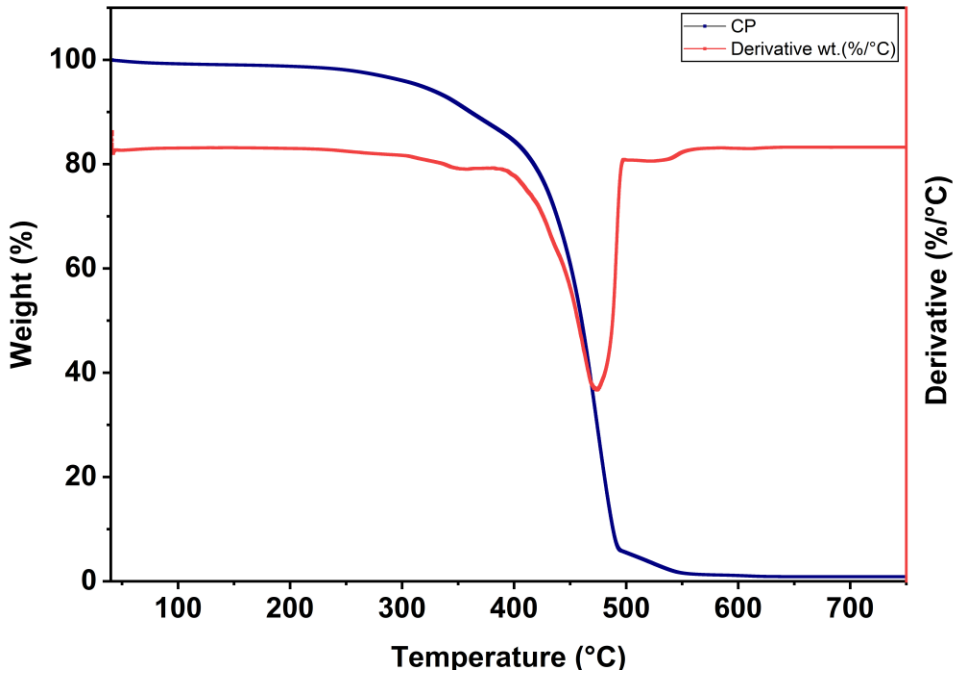


Figure 16: TGA and DTG of fabricated composite

The fourth degradation stage, occurring between 480–700°C for both rHDPE and the rHDPE: CSS composite (CP) in weight loss of 18.5% and 19.3% respectively.it corresponds to the slow decomposition of remaining carbonaceous residues and thermally stable fragments formed during the main degradation stage. The close agreement between the two values implies that the presence of CSS does not significantly alter the final-stage thermal degradation. Any minor differences assigned to the decomposition of lignin-derived char and inorganic constituents from CSS, which contribute to the formation of thermally stable residue. These results confirm that the HDPE:CSS composite possesses adequate thermal stability for typical thermoplastic processing and subsequent applications, its suitability for further mechanical and structural performance evaluation.

4.4.3. Differential scanning calorimeter (DSC) analysis of rHDPE and its CP

The thermal transition and heat flow behavior of the two samples illustrating their thermal transition during heating (endothermic melting) and cooling (exothermic crystallization) are shown in figure 4.17. The observed thermal events are typical of semi-crystalline polyethylene-based materials and provide insight into the effect of filler incorporation on the crystalline behavior of the polymer matrix. The melting temperature (Tm) and crystallization temperature (Tc) of unreinforced rHDPE maintained its characteristics without major change in their molecular structures, reported by (Ayana, Ha, et al., 2024). During the heating cycle, rHDPE displays a distinct and sharp melting endotherm with a melting temperature (Tm) of 130.4°C, indicating advanced crystalline regions within the polymer matrix.

While, the composite sample exhibits a slightly lower melting temperature (Tm = 126.7°C) and a broader melting peak. This reduction and peak broadening imply restricted chain mobility and less perfect crystal formation due to the presence of the coffee silver skin filler, which disrupts the regular packing of rHDPE chains. The melting enthalpy (ΔHm) further confirms this behavior; rHDPE shows a high ΔHm of 126.7 J/g, whereas the composite shows a substantially lower value of 48.58 J/g, reflecting a reduced crystalline fraction in the composite system. Accordingly, the calculated degree of crystallinity (Xc) decreases markedly from 43.24% for rHDPE to 20.72% for CP, this related to the previously described case of the interaction of lignocellulosic filler as heterogeneous nucleate of the PE (Andrzejewski et al., 2020).

This significant reduction indicates that the filler acts primarily as a physical hindrance to lamellar growth rather than an effective nucleating agent, limiting the development of crystalline domains during solidification.

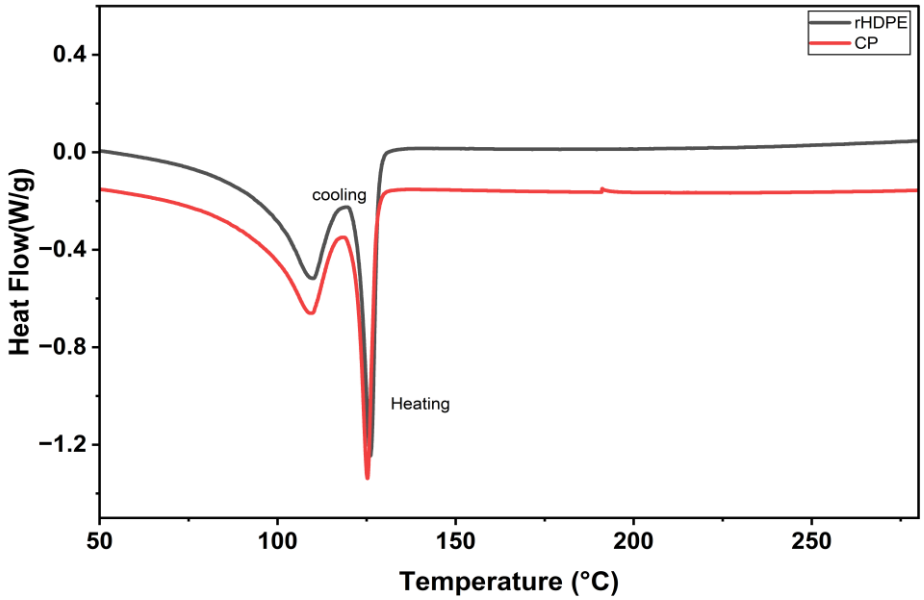


Figure 4.17: DSC curves of rHDPE and rHDPE/CSS composites

During the cooling cycle, the crystallization temperature (Tc) of rHDPE is viewed at 119.2°C, while that of the composite shifts slightly to a lower temperature (Tc=117.8°C). The crystallization enthalpy (ΔHc) also decreases from 78.6 J/g for rHDPE to 41.5 J/g for CP. It indicates suppressed crystallization kinetics in the composite. The lower Tc and ΔHc values imply delayed nucleation and slower crystal growth, caused by reduced polymer chain mobility and interfacial interactions between rHDPE and the lignocellulosic filler, reported by (Hejna et al., 2021). The DSC results illustrate that incorporation of coffee silver skin significantly changes the thermal and crystalline behavior of rHDPE by reducing melting and crystallization enthalpies, lowering crystallinity, and slightly shifting transition temperatures. These changes are consistent with previously study as reported, lignocellulosic-reinforced polyethylene composites and influence the mechanical performance and thermal stability of the composite material (Gozdecki et al., 2025a). Therefore, this result provides that the incorporation of coffee silver skin as a filler modifies the structural characteristics relationship of the HDPE matrix, which is reflected in both the mechanical and thermal performance of the fabricated composite.

4.4.4. FTIR analysis of rHDPE and its Composite

The FTIR spectra of recycled high-density polyethylene (rHDPE) and its composite (CP) were recorded in the range of 4000-500 cm^{-1}, to identify the functional groups present and to examine possible interfacial interactions between the rHDPE polymer matrix and lignocellulosic filler of CSS. The spectrum of rHDPE has shown the characteristic absorption bands of polyethylene. The prominent peaks at 2916 and 2848cm^{-1} are assigned to the asymmetric and symmetric stretching vibrations of methylene (-CH_2) groups, confirming the saturated aliphatic backbone structure of HDPE. These absorption bands are consistent with previously reported values for wood-plastic and lignocellulosic fiber-reinforced polyethylene composites,(Ayana et al., 2022). Supporting bands observed at 1460cm^{-1} correspond to CH_2 bending, while the peak at 1140cm^{-1} is related to C-C stretching vibrations within the polymer chain. The absorption at 720cm^{-1} represents CH_2 rocking, typically associated with long methylene sequences and the semi crystalline nature of polyethylene.

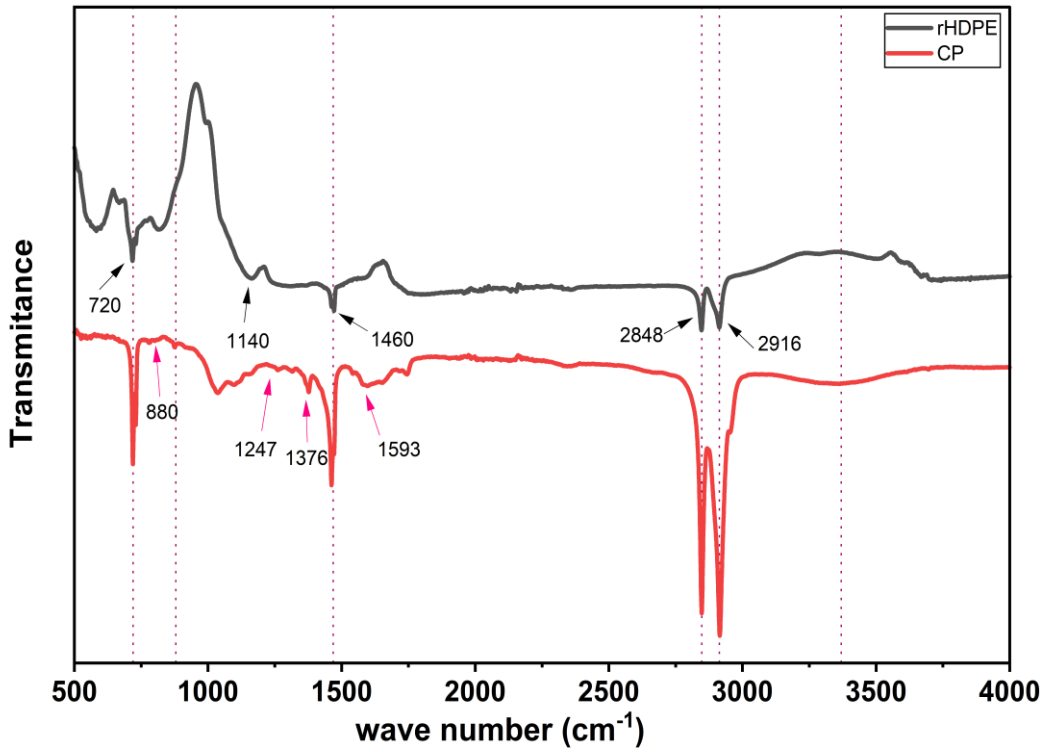


Figure 4.18: FTIR spectra of recycled HDPE and coffee silver skin reinforced composite

On the other hand, the composite spectrum showed several additional peaks initiating from lignocellulosic coffee silver skin filler constituents. A broad band around 3370cm^{-1} is assigned to O-H stretching vibrations of hydroxyl groups present in cellulose, hemicellulose, and lignin. The peak at 1593cm^{-1} corresponds to aromatic C=C stretching of lignin. Although bands at 1376 cm^{-1} and 1247cm^{-1} are attribute to C-H bending and C-O stretching of aryl-alkyl ether linkages, respectively.

The signal close 880 cm^{-1} indicates β-glycosidic linkages of cellulose representing a fingerprint indicator of polysaccharide structures and a minor weak peak, relatively around this area. It confirms the presence of this filler in the recycled PE waste (Hussein et al., 2020). The characteristic polyethylene methylene stretching bands at 2916cm^{-1} and 2848 cm^{-1} show in the composite spectrum without significant peak shifting, reported by (Cecilia et al., 2023), implying the chemical backbone of the rHDPE polymer matrix was saved through processing. The retention of polyethylene peaks without significant shifting and new bond formation hint that the matrix-filler interaction is mainly physical, reflecting limited interfacial compatibility between hydrophobic rHDPE and hydrophilic biomass CSS filler. The reduction in peak intensity observed in the 3600-3000 cm^{-1} region indicates improved interfacial interaction between the fiber and the polymer matrix, as also reported in previous study by (Zulkifli et al., 2015).

4.4.5. Water Absorption Test

The water absorption behavior of the optimized composites reinforced with treated and untreated coffee silver skin (CSS) was evaluated in accordance with ASTM D570 at different immersion times, due to the inherently hydrophilic nature of natural fillers; moisture uptake can adversely affect interfacial bonding and mechanical properties. The water absorption values of the composites with three different immersion times are summarized in Table 4.8 as mean ± standard deviation (n = 3).

Table 4.8: Water absorption of treated/untreated CSS composites at three immersion times

Immersion Time (h)	Untreated CSS Composite (%)	Treated CSS Composite (%)
24	1.3±0.22	0.66±0.38
48	2.96±0.69	1.83±0.56
72	5.6±1.33	3.38±0.75

The untreated CSS composite exhibited higher water uptake at all immersion times, reaching 5.6 ± 1.33% after 72 h, whereas the treated CSS composite showed significantly lower values, with a maximum water absorption of 3.38 ± 0.75% at the same immersion time. Error bars represent the standard deviation of three specimens, indicating acceptable repeatability of the measurements.

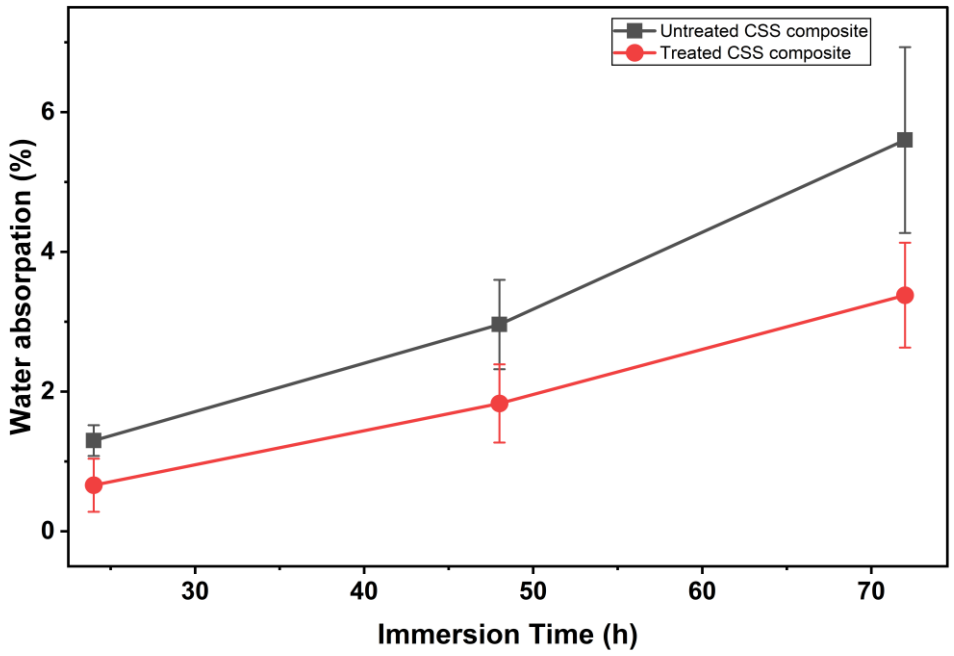


Figure 4.19: Water absorption behavior of treated & untreated CSS reinforced composites

The water absorption behavior of the optimized composites is closely related to the chemical structure of the coffee silver skin reinforcement, as evidenced by the FTIR analysis. As shown in Figure 4.19, both treated and untreated CSS composites displayed increasing water uptake with extended immersion time.

The untreated CSS composite consistently showed higher water absorption compared to composites from treated CSS. FTIR spectra of untreated CSS shown a stronger and broader -OH stretching band, indicating a higher concentration of hydrophilic functional groups such as hydroxyl groups associated with cellulose and hemicellulose (Kabir et al., 2012). These polar groups encourage moisture uptake through hydrogen bonding with water molecules. But, the treated CSS displayed reduced -OH band intensity in the FTIR spectrum, indicating partial removal of hemicellulose, lignin and surface impurities due to alkali treatment. This chemical modification reduces the hydrophilicity of the CSS, as reinforcement and enhances interfacial adhesion with the rHDPE matrix, consequently restricting water diffusion pathways. similar findings also reported for the Comparable reductions in water absorption in lignocellulosic-filled polymer composites (Amena et al., 2022) (Kruszelnicka et al., 2023). The higher standard deviation observed at longer immersion times, particularly for the untreated composite, implies increased heterogeneity in water diffusion behavior, caused by weak interfacial regions and microstructural defects. As a result, the treated CSS composite demonstrated improved moisture resistance and dimensional stability also with higher tensile strength compared to the untreated CSS composites.

4.5. Comparison with Previous Studies

The tensile strength obtained in the present study is comparable with previously reported natural fiber reinforced HDPE composites. As shown in Table 4.9 several studies demonstrated that the incorporation of lignocellulosic fillers, including coffee processing by-products, can influence the mechanical performance of polymer composites, such comparisons evaluate the consistency of the experimental findings and provide insight into the influence of filler type, treatment methods, and processing conditions on composites performance. Similar observations were reported for coffee waste and other agricultural residues used as reinforcement in polyethylene matrices and bio-based polymers, where filler treatment and matrix-filler interaction significantly affected tensile properties. The tensile strength achieved in this study is comparable to values reported for other polyethylene-based natural fiber composites.

The improvement noticed in the treated CSS composites in this study, assigned to enhanced filler-matrix compatibility resulting from alkali treatment, which advocates better stress transfer within the composite structure, as reported in other lignocellulosic composite materials.

Table 4.9: Comparison of this study with some previous works

Matrix Material	Filler Type	Conditions			Treatment	Tensile strength MPa	Reference
		Filler Load (%)	Particle size (µm)	Temperature (°C)			
rHDPE	CSS	10.4	285.8	175.9	Alkali-treated Untreated CSS	6.71±0.04 6.01±0.01	This study
Bio-PE	CSS	10	-	-	Alkali-treated	21.2±1.2	(Dominici et al., 2019)
PLA	CSS	10	< 200	-	Untreated CSS Alkali-treated	3.51 ± 0.61 36.26±4.89	(Petaloti & Achilias, 2024)
HDPE	CSS	20	400	170	-	15.6	(Hejna et al., 2021)
PLA/PBS	CSS	30	212-425	175	-	10.60±1.21	(Kumar et al., 2024)
rPE	Bamboo	30	200-600	180	Untreated bamboo Alkali-treated	14.57 17.47	(Plastic & Wpc, 2022)
Bio-polymer	Coffee Husk	30	125-500	170	-	28	(Gozdecki et al., 2025a)
rHDPE	Rice Husk	20	< 300	185	Compatibilizer (MAPE) used	15.25	(Ur et al., 2023)

5. Conclusions and Recommendations

5.1. Conclusions

The composite of waste rHDPE and CSS was developed through compounding and consolidation process. To determine the optimal processing conditions and improve their compatibility, CSS were subjected to alkali treatment and characterized by EDX, XRD, FTIR, SEM, TGA, and DSC, which verified that alkali treatment effectively removed impurities and reduced non-cellulosic components that improved fiber surface roughness and interfacial interaction with the rHDPE polymer matrix.

After composite fabrication, three processing factors were selected and optimized for tensile strength. The optimal processing conditions identified through BBD included a temperature of 175.97℃, a particle size of 285μm, and a filler ratio of 10.44%. With these optimal conditions, a maximum tensile strength of 5.99MPa was achieved. The tensile strength of optimized composites from treated CSS was shown a significant improvement than composites from untreated CSS, a mean tensile strength composite from treated CSS achieved 6.71MPa, the observed improvement of mechanical performance attributed better contact and interfacial bonding between treated CSS and the rHDPE matrix.

Additionally, the fabricated composites subjects in to thermal, chemical and morphological characterized by TGA, DSC, FTIR, SEM and water absorption test also investigated. Thermal analyses using TGA and DSC indicated that the composite maintained acceptable thermal stability and preserved the melting behavior of the polymer, implying its capability to withstand typical processing and service temperatures. SEM observations showed that filler dispersion was fairly uniform at lower filler loadings, while higher contents led to certain agglomeration and weaker interfacial adhesion. FTIR analysis confirmed the successful incorporation of coffee silver skin into the polymer matrix without altering the chemical backbone of rHDPE. Water absorption behavior reflected the hydrophilic nature of the lignocellulosic filler but remained within a reasonable range for composite materials. These findings advocate that the fabricated composite own suitable structural, thermal, and physical characteristics for applications such as interior panels, furniture components, and other non-load bearing construction materials.

5.2. Recommendations

In this study investigated the development and optimization of rHDPE:CSS composites and evaluated their mechanical, thermal, and morphological characteristics. Yet, to further improve the performance and practical applicability of the composites, the following recommendations are proposed for future research.

First, this study fabricated composites without the addition of chemical compatibilizer in order to minimize environmental and economic impacts. However, the inherent incompatibility between the hydrophilic lignocellulosic CSS filler and the hydrophobic rHDPE matrix limit the interfacial adhesion and general mechanical performance of the composites. Therefore, future studies should investigate the use of coupling agents to enhance filler-matrix interaction and furthermore, improve the mechanical properties of the developed composites.

Second, this research mainly focused on the optimization of tensile strength as a key mechanical property of the developed composites. Future studies should evaluate other mechanical properties, including flexural strength, modulus, impact resistance, and hardness.

Third, although surface modification of coffee silver skin was conducted in this study, future research further explore advanced treatments such as acid hydrolysis to produce Nano-cellulose. The use of Nano-cellulose as reinforcement may enhance filler dispersion and improve the overall reinforcement efficiency of the composite matrix.

Fourth, the fabrications of composites were conducted using an alternative method due to the limited availability of equipment. Since this process was relatively labor and time intensive, future studies should utilize extruder and computerized hot-press molding machine to better control condition such as pressure during composite fabrication.

In addition, this study was conducted at a laboratory scale and did not evaluate the economic feasibility of scaling up. Therefore, pilot or industrial-scale studies are needed to assess cost effectiveness, operational challenges, and practical applicability of the developed composites.

Lastly, furthermore investigation of long-term durability properties, weathering resistance and thermal aging, is recommended to evaluate the performance and stability of the developed rHDPE:CSS composites under real service conditions.

References

Achilias, D. S., Roupakias, C., Megalokonomos, P., Lappas, A. A., & Antonakou, V. (2007). Chemical recycling of plastic wastes made from polyethylene (LDPE and HDPE) and polypropylene (PP). *Journal of Hazardous Materials*, *149*(3), 536–542. https://doi.org/10.1016/j.jhazmat.2007.06.076

Al-Salem, S. M., Lettieri, P., & Baeyens, J. (2009). Recycling and recovery routes of plastic solid waste (PSW): A review. *Waste Management*, *29*(10), 2625–2643. https://doi.org/10.1016/j.wasman.2009.06.004

Alghooneh, A., Amini, A. M., & Behrouzian, F. (2017). Characterisation of cellulose from coffee silverskin. *International Journal of Food Properties*, *20*(11), 2830–2843. https://doi.org/10.1080/10942912.2016.1253097

Alrazen, H. A., Aminossadati, S. M., Mahmood, H. A., Kadhim, A., Arifin, K., Sharul, A., Dol, S., Jabbar, S., Jabbar, S., Algayyim, M., & Konarova, M. (2025). *A review of the pathways , limitations , and perspectives of plastic waste recycling*.

Amena, B. T., Altenbach, H., Tibba, G. S., & Hossain, N. (2022). *Investigation of Mechanical Properties of Coffee Husk-HDPE-ABS Polymer Composite Using Injection-Molding Method*.

Andrady, A. L., & Neal, M. A. (2009). Applications and societal benefits of plastics. *Philosophical Transactions of the Royal Society B: Biological Sciences*, *364*(1526), 1977–1984. https://doi.org/10.1098/rstb.2008.0304

Andrzejewski, J., Krawczak, A., Weso, K., & Szostak, M. (2020). *Rotational molding of biocomposites with addition of buckwheat husk filler . Structure-property correlation assessment for materials based on polyethylene (PE) and poly (lactic acid) PLA*. *202*(May). https://doi.org/10.1016/j.compositesb.2020.108410

Annual Review, I. (2024). *A message from the Chair*. *44*(0).

Assefa, B., Flaura, K., Kebede, S., Tsigkou, K., Assefa, B., & Hashim, S. (2025). Coffee processing waste : Unlocking opportunities for sustainable development Publication date : Publisher ' s PDF , also known as Version of record Coffee processing waste : Unlocking opportunities for sustainable development. *Renewable and Sustainable Energy Reviews*, *210*, 115263. https://doi.org/10.1016/j.rser.2024.115263

Aurrekoetxea, J., Sarrionandia, M. A., & Urrutibeascoa, I. (2001). *Effects of recycling on the microstructure and the mechanical properties of isotactic polypropylene*. *6*, 2607–2613.

Ayana, K. D., Ali, A. Y., & Ha, C. S. (2024). Wood Polymer Composites Based on the Recycled Polyethylene Blends from Municipal Waste and Ethiopian Indigenous Bamboo (Oxytenanthera abyssinica) Fibrous Particles Through Chemical Coupling Crosslinking. *Polymers*, *16*(21). https://doi.org/10.3390/polym16212982

Ayana, K. D., De Angelis, M., Schmidt, G., Krause, A., & Ali, A. Y. (2022). Valorization of Potential Post-Consumer Polyethylene (PE) Plastics Waste and Ethiopian Indigenous Highland Bamboo (EHB) for Wood Plastic Composite (WPC): Experimental Evaluation and Characterization. *Fibers*, *10*(10). https://doi.org/10.3390/fib10100085

Ayana, K. D., Ha, C. S., & Ali, A. Y. (2024). Comprehensive overview of wood polymer composite: Formulation and technology, properties, interphase modification, and characterization. *Sustainable Materials and Technologies*, *40*(June). https://doi.org/10.1016/j.susmat.2024.e00983

Ballesteros, L. F., Teixeira, J. A., & Mussatto, S. I. (2014). Chemical, Functional, and Structural Properties of Spent Coffee Grounds and Coffee Silverskin. *Food and Bioprocess Technology*, *7*(12), 3493–3503. https://doi.org/10.1007/s11947-014-1349-z

Behrouzian, F., Amini, A. M., Mohammad, S., & Razavi, A. (2016). Author ' s Accepted Manuscript. *Bioactive Carbohydrates and Dietary Fibre*. https://doi.org/10.1016/j.bcdf.2016.11.004

Berzin, F., Beaugrand, J., Dobosz, S., Budtova, T., & Vergnes, B. (2017). Lignocellulosic fiber breakage in a molten polymer. Part 3. Modeling of the dimensional change of the fibers during compounding by twin screw extrusion. *Composites Part A*. https://doi.org/10.1016/j.compositesa.2017.07.009

Berzin, F., David, C., & Vergnes, B. (2020). *Use of Flow Modeling to Optimize the Twin-Screw Extrusion Process for the Preparation of Lignocellulosic Fiber-Based Composites*. *7*(July), 1–7. https://doi.org/10.3389/fmats.2020.00218

Blinová, L., Sirotiak, M., Bartošová, A., & Soldán, M. (2017). Faculty of Materials Science and Technology in Trnava Review : Utilization of Waste From Coffee Production. *Research Papers*, *25*(40), 91–102.

Borrelle, S. B., Ringma, J., Law, K. L., Monnahan, C. C., Lebreton, L., McGivern, A., Murphy, E., Jambeck, J., Leonard, G. H., Hilleary, M. A., Eriksen, M., Possingham, H. P., & Rochman, C. M. (2020). Mitigate Plastic Pollution. *Science*, *1518*(September), 1515–1518. http://science.sciencemag.org/content/369/6510/1515

Carbonell-verdú, A., García-garcía, D., Jordá, A., Samper, M. D., & Balart, R. (2015). *Composites : Part B Development of slate fiber reinforced high density polyethylene composites for injection molding*. *69*, 460–466. https://doi.org/10.1016/j.compositesb.2014.10.026

Cecilia, Z., Ram, R., Franco-urquiza, E. A., & Carlos, S. (2023). *The Role of Coupling Agents in the Mechanical and Thermal Properties of Polypropylene / Wood Flour Composites*. 65–78.

Chan, C. M., Vandi, L., Werker, A., Laycock, B., Pratt, S., Halley, P., & Richardson, D. (2018). *Mechanical properties of poly (3-hydroxybutyrate-co- 3-hydroxyvalerate)/ wood fl our composites : Effect of interface modi fi ers*. *46828*, 1–10. https://doi.org/10.1002/app.46828

Delviawan, A., Kojima, Y., Kobori, H., Suzuki, S., Aoki, K., & Ogoe, S. (2019). The effect of wood particle size distribution on the mechanical properties of wood – plastic composite. *Journal of Wood Science*. https://doi.org/10.1186/s10086-019-1846-9

Dominici, F., García, D. G., Fombuena, V., Luzi, F., Puglia, D., Torre, L., & Balart, R. (2019). Bio-polyethylene-based composites reinforced with alkali and palmitoyl chloride-treated coffee silverskin. *Molecules*, *24*(17). https://doi.org/10.3390/molecules24173113

Elkoun, S., Robert, M., Mighri, F., & Diez, C. (2023). *Mechanical Recycling of Thermoplastics : A Review of Key Issues Recycling of Thermoplastics : A Review of Mechanical*. 860–883.

Esterhuizen, M., & Kim, Y. J. (2022). Effects of polypropylene, polyvinyl chloride, polyethylene terephthalate, polyurethane, high-density polyethylene, and polystyrene microplastic on Nelumbo nucifera (Lotus) in water and sediment. *Environmental Science and Pollution Research*, *29*(12), 17580–17590. https://doi.org/10.1007/s11356-021-17033-0

Faruk, O., Bledzki, A. K., Fink, H. P., & Sain, M. (2012). Biocomposites reinforced with natural fibers: 2000-2010. *Progress in Polymer Science*, *37*(11), 1552–1596. https://doi.org/10.1016/j.progpolymsci.2012.04.003

Fatimatuzzahro, Nadie, H. E. P. (2016). Executive Summary Executive Summary Executive Summary. *South African Medical Journal*, *101*(2003), 16.

Gandhi, N., Farfaras, N., Wang, N. L., & Chen, W. (2021). *Life Cycle Assessment of Recycling High-Density Polyethylene Plastic Waste*. https://doi.org/10.32604/jrm.2021.015529

Geyer, R., Jambeck, J. R., & Law, K. L. (2017). Production, use, and fate of all plastics ever made. *Science Advances*, *3*(7), 25–29. https://doi.org/10.1126/sciadv.1700782

Ghazvini, A. K. A., Ormondroyd, G., Curling, S., Saccani, A., & Sisti, L. (2022). An investigation on the possible use of coffee silverskin in PLA/PBS composites. *Journal of Applied Polymer Science*, *139*(22), 1–10. https://doi.org/10.1002/app.52264

Gigante, V., Seggiani, M., Cinelli, P., Signori, F., Navarini, L., Amato, G., Lazzeri, A., Gigante, V., Seggiani, M., Cinelli, P., Signori, F., Vania, A., Navarini, L., Amato, G., & Lazzeri, A. (2020). Utilization of coffee silverskin in the production of Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) biopolymer-based thermoplastic biocomposites for food contact applications. *Composites Part A*, 106172. https://doi.org/10.1016/j.compositesa.2020.106172

Gonçalves, B. M. M., Camillo, M. de O., Oliveira, M. P., Carreira, L. G., Moulin, J. C., Neto, H. F., de Oliveira, B. F., Pereira, A. C., & Monteiro, S. N. (2021). Surface treatments of coffee husk fiber waste for effective incorporation into polymer biocomposites. *Polymers*, *13*(19). https://doi.org/10.3390/polym13193428

Gozdecki, C., Kociszewski, M., & Moraczewski, K. (2025a). *Containing Biopolymer and Waste Coffee HusksGreen Composite Based on a Polymer Mixture*.

Gozdecki, C., Kociszewski, M., & Moraczewski, K. (2025b). *Green Composite Based on a Polymer Mixture Containing Biopolymer and Waste Coffee Husks*.

Gupta, A., Singhal, R., & Nagpal, A. K. (2004). *Reactive Blends of Epoxy Resin (DGEBA) Crosslinked by Anionically Polymerized Polycaprolactam . II . Mechanical and Electrical Properties*. https://doi.org/10.1002/app.21476

Gwada, B., Ogendi, G., Makindi, S. M., & Trott, S. (2019). *Composition of plastic waste discarded by households and its management approaches*. *5*(1), 83–94. https://doi.org/10.22034/gjesm.2019.01.07

Hejna, A. (2021). Potential applications of by-products from the coffee industry in polymer technology – Current state and perspectives. *Waste Management*, *121*, 296–330. https://doi.org/10.1016/j.wasman.2020.12.018

Hejna, A., Barczewski, M., Kosmela, P., Mysiukiewicz, O., & Kuzmin, A. (2021). Coffee silverskin as a multifunctional waste filler for high-density polyethylene green composites. *Journal of Composites Science*, *5*(2), 1–13. https://doi.org/10.3390/jcs5020044

Hietala, M., & Oksman, K. (2018). Pelletized cellulose fibres used in twin-screw extrusion for biocomposite manufacturing: Fibre breakage and dispersion. *Composites Part A*. https://doi.org/10.1016/j.compositesa.2018.04.006

Hopewell, J., Dvorak, R., & Kosior, E. (2009). Plastics recycling: Challenges and opportunities. *Philosophical Transactions of the Royal Society B: Biological Sciences*, *364*(1526), 2115–2126. https://doi.org/10.1098/rstb.2008.0311

Huang, F., & Ragauskas, A. J. (2013). *Chemical Pretreatment Techniques for Biofuels and Biorefineries from Softwood*. https://doi.org/10.1007/978-3-642-32735-3

Huang, L., Mu, B., Yi, X., Li, S., & Wang, Q. (2016). Sustainable Use of Coffee Husks For Reinforcing Polyethylene Composites. *Journal of Polymers and the Environment*, *0*(0), 0. https://doi.org/10.1007/s10924-016-0917-x

Hussein, A. I., Ab-ghani, Z., Nazeer, A., Mat, C., Atikah, N., Ghani, A., Husein, A., & Rahman, I. A. (n.d.). *applied sciences Synthesis and Characterization of Spherical Calcium Carbonate Nanoparticles Derived from Cockle Shells*. 1–14.

Imran, A. I., Siregar, J. P., Cionita, T., Hadi, A. E., Setiyo, M., Ruzaimi, M., Rejab, M., Jaafar, J., & Fitriyana, D. F. (2024). *Advancements in sustainable material development : A Comprehensive review of coir fiber and its composites*. *4*(3), 415–454.

Jaafar, J. (2019). *A review of important considerations in the compression molding process of short natural fiber composites*.

Jambeck, J. R., Geyer, R., Wilcox, C., Siegler, T. R., Perryman, M., Andrady, A., Narayan, R., & Law, K. L. (2015). Entradas de residuos plásticos desde la tierra al océano. *Ciencia*, *347*(6223), 768–771. http://www.sciencemag.org/cgi/doi/10.1126/science.1260879%0Ahttps://www.sciencemag.org/lookup/doi/10.1126/science.1260352

Janissen, B., & Huynh, T. (2018). *Resources , Conservation & Recycling Chemical composition and value-adding applications of co ff ee industry by- products : A review*. *128*(July 2017), 110–117. https://doi.org/10.1016/j.resconrec.2017.10.001

Kabir, M. M., Wang, H., Lau, K. T., & Cardona, F. (2012). Composites : Part B Chemical treatments on plant-based natural fibre reinforced polymer composites : An overview. *Composites Part B*, *43*(7), 2883–2892. https://doi.org/10.1016/j.compositesb.2012.04.053

Kibria, M. G., Masuk, N. I., Safayet, R., Nguyen, H. Q., & Mourshed, M. (2023). Plastic Waste: Challenges and Opportunities to Mitigate Pollution and Effective Management. In *International Journal of Environmental Research* (Vol. 17, Issue 1). Springer International Publishing. https://doi.org/10.1007/s41742-023-00507-z

Kord, B., Ravanfar, P., & Ayrilmis, N. (2017). Influence of Organically Modified Nanoclay on Thermal and Combustion Properties of Bagasse Reinforced HDPE Nanocomposites. *Journal of Polymers and the Environment*, *25*(4), 1198–1207. https://doi.org/10.1007/s10924-016-0897-x

Kruszelnicka, I., Michałkiewicz, M., & Ginter-kramarczyk, D. (2023). *Spent Coffee as a Composite Filler for Wastewater Treatment*.

Ku, H., Wang, H., Pattarachaiyakoop, N., & Trada, M. (2011). A review on the tensile properties of natural fiber reinforced polymer composites. *Composites Part B: Engineering*, *42*(4), 856–873. https://doi.org/10.1016/j.compositesb.2011.01.010

Kumar, K. S., Gairola, S., & Singh, I. (2024). Waste Coffee Silverskin as a potential filler in sustainable composites: Mechanical, thermal, and microstructural analysis. *Industrial Crops and Products*, *210*(January), 118088. https://doi.org/10.1016/j.indcrop.2024.118088

Kuram, E. (2022). Advances in development of green composites based on natural fibers: a review. *Emergent Materials*, *5*(3), 811–831. https://doi.org/10.1007/s42247-021-00279-2

Lorenzo, V., Acosta, J., Orden, M. U. De, Urreaga, J. M., & Beltr, F. R. (2017). *Effect of simulated mechanical recycling processes on the structure and properties of poly (lactic acid)*. 1–7. https://doi.org/10.1016/j.jenvman.2017.05.020

Manzuch, Z., Akelyte, R., Camboni, M., & Carlander, D. (2021). *Chemical Recycling of Polymeric Materials from Waste in the Circular Economy*. *August*, 1–145.

Mateus, M., Camillo, M. D. O., Oliveira, M. P., Carreira, L. G., Neto, H. F., & Oliveira, F. De. (2021). *Surface Treatments of Coffee Husk Fiber Waste for Effective Incorporation into Polymer Biocomposites*.

Mattlet, A., Sicot, O., Schoors, L. Van, & Aivazzadeh, S. (2023). Influence of processing and matrix parameters on the manufacturing of unidirectional flax / polypropylene composites. *SN Applied Sciences*, *April 2022*. https://doi.org/10.1007/s42452-023-05457-x

Migneault, S., Koubaa, A., Erchiqui, F., Chaala, A., Englund, K., & Wolcott, M. P. (2009). Composites : Part A Effects of processing method and fiber size on the structure and properties of wood – plastic composites. *Composites Part A*, *40*(1), 80–85. https://doi.org/10.1016/j.compositesa.2008.10.004

Móczó, J., Purwaningsih, H., & Pukánszky, B. (2020). Alkali treatment of lignocellulosic fibers extracted from sugarcane bagasse: Composition, structure, properties. In *Polymer Testing*. Elsevier Ltd. https://doi.org/10.1016/j.polymertesting.2020.106549

Mohammed, L., Ansari, M. N. M., Pua, G., Jawaid, M., & Islam, M. S. (2015). *A Review on Natural Fiber Reinforced Polymer Composite and Its Applications*. *2015*.

Montanes, N., & Balart, R. (2018). SC. *Composites Part B*. https://doi.org/10.1016/j.compositesb.2018.04.017

Murthy, P. S., & Madhava Naidu, M. (2012). Sustainable management of coffee industry by-products and value addition - A review. *Resources, Conservation and Recycling*, *66*, 45–58. https://doi.org/10.1016/j.resconrec.2012.06.005

Ncube, L. K., Ude, A. U., Ogunmuyiwa, E. N., Zulkifli, R., & Beas, I. N. (2021). An overview of plasticwaste generation and management in food packaging industries. *Recycling*, *6*(1), 1–25. https://doi.org/10.3390/recycling6010012

Oksman, K., Hietala, M., Rollo, P., & Kek, K. (2014). *Extrusion Processing of Green Biocomposites : Compounding , Fibrillation Efficiency , and Fiber Dispersion*. *39981*, 1–9. https://doi.org/10.1002/app.39981

Papamatthaiakis, N., Barbero, A., Eemeli, L., Janne, E., Blas, J., & Yudego, M. (2025). *Hydrothermal Liquefaction of Coffee Silverskin and Spent Coffee Grounds : Bioenergy and Biochemical Potential*.

Petaloti, A.-I., & Achilias, D. S. (2024). The Development of Sustainable Biocomposite Materials Based on Poly(lactic acid) and Silverskin, a Coffee Industry By-Product, for Food Packaging Applications. *Sustainability*, *16*(12), 5075. https://doi.org/10.3390/su16125075

Plastic, W., & Wpc, C. (2022). *Valorization of Potential Post-Consumer Polyethylene (PE) Plastics Waste and Ethiopian Indigenous Highland Bamboo (EHB) for Wood Plastic Composite (WPC): Experimental Evaluation and Characterization*.

Plastics the Fast Facts 2025. (2025). *3*(October), 2025.

Poletto, M., Zattera, A. J., Forte, M. M. C., & Santana, R. M. C. (2012). Bioresource Technology Thermal decomposition of wood : Influence of wood components and cellulose crystallite size. *Bioresource Technology*, *109*, 148–153. https://doi.org/10.1016/j.biortech.2011.11.122

Ribeiro, L. S., Stolz, C. M., Amario, M., Ana, L., & Haddad, A. N. (2023). *Use of Post-Consumer Plastics in the Production of Wood-Plastic Composites for Building Components : A Systematic Review*.

Saccani, A., & Sisti, L. (2022). *An investigation on the possible use of coffee silverskin in PLA / PBS composites*. *January*, 1–10. https://doi.org/10.1002/app.52264

Salleh, F., Hassan, A., Yahya, R., & Azzahari, A. D. (2014). Composites : Part B Effects of extrusion temperature on the rheological , dynamic mechanical and tensile properties of kenaf fiber / HDPE composites. *COMPOSITES PART B*, *58*, 259–266. https://doi.org/10.1016/j.compositesb.2013.10.068

Sameni, J., Jaffer, S. A., & Sain, M. (2018). Thermal and mechanical properties of soda lignin/HDPE blends. *Composites Part A, September*. https://doi.org/10.1016/j.compositesa.2018.09.016

Sarasini, F., Luzi, F., Dominici, F., Maffei, G., Iannone, A., Zuorro, A., Lavecchia, R., Torre, L., Carbonell-verdu, A., Balart, R., & Puglia, D. (n.d.). *Effect of Different Compatibilizers on Sustainable Composites Based on a PHBV / PBAT Matrix Filled with Coffee Silverskin*. 1–16. https://doi.org/10.3390/polym10111256

Sarasini, F., Tirillò, J., Zuorro, A., Ma, G., Lavecchia, R., Puglia, D., Dominici, F., Luzi, F., Valente, T., & Torre, L. (2018). *Industrial Crops & Products Recycling co ff ee silverskin in sustainable composites based on a poly hydroxyvalerate) matrix*. *118*(March), 311–320. https://doi.org/10.1016/j.indcrop.2018.03.070

Sarasini, F., Tirillò, J., Zuorro, A., Maffei, G., Lavecchia, R., Puglia, D., Dominici, F., Luzi, F., Valente, T., & Torre, L. (2018). Recycling coffee silverskin in sustainable composites based on a poly(butylene adipate-co-terephthalate)/poly(3-hydroxybutyrate-co-3-hydroxyvalerate) matrix. *Industrial Crops and Products*, *118*(November 2017), 311–320. https://doi.org/10.1016/j.indcrop.2018.03.070

Sherefa, A., Tsigkou, K., & Kebede, S. (2026). Biomass and Bioenergy Sustainable valorization of coffee pulp : Evaluation of the pretreatment effect on the phenolic compounds recovery and anaerobic digestion of the residual biomass. *Biomass and Bioenergy*, *209*(January), 108969. https://doi.org/10.1016/j.biombioe.2026.108969

Shogren, R., Wood, D., Orts, W., & Glenn, G. (2019). Plant-based materials and transitioning to a circular economy. *Sustainable Production and Consumption*, *22*(xxxx), 1–22. https://doi.org/10.1016/j.spc.2019.04.007

Silverskin, C., & Qiu, M. (2021). *Morphological Changes and Component Characterization of Coffee Silverskin*. 1–12.

Singh, P., & Singh, O. P. (2020). Plastics Addiction : Humanity's Existential Crisis. *Akgec International Journal of Technology*, *11*(1), 41–47.

Single-, A., Resolutions, A., & Litter, M. (2020). *mitigate plastic pollution*. *1518*(September), 1515–1518.

Sohn, J. S., Ryu, Y., Yun, C. S., Zhu, K., & Cha, S. W. (2019). Extrusion compounding process for the development of eco-friendly SCG/PP composite pellets. *Sustainability (Switzerland)*, *11*(6). https://doi.org/10.3390/su11061720

Sommerhuber, P. F., Welling, J., & Krause, A. (2015). Substitution potentials of recycled HDPE and wood particles from post-consumer packaging waste in Wood – Plastic Composites. *WASTE MANAGEMENT*. https://doi.org/10.1016/j.wasman.2015.09.011

Sormunen, P. (2019). *Compression Molded Thermoplastic Composites Entirely Made of Recycled Materials*. https://doi.org/10.3390/su11030631

Taylor, P., Gunning, M. A., Geever, L. M., Higginbotham, L., Killion, J. A., Lyons, J. G., Gunning, M. A., Geever, L. M., Killion, J. A., & Lyons, J. G. (2013). *Polymer-Plastics Technology and Engineering Melt Processing of Bioplastic Composites via Twin Screw Extrusion and Injection Molding Melt Processing of Bioplastic Composites via Twin Screw Extrusion and Injection Molding*. *December 2014*, 37–41. https://doi.org/10.1080/03602559.2013.844825

Taylor, P., Krehula, L. K., Katan, Z., & Pti, A. (n.d.). *Journal of Wood Chemistry and Weathering of High-Density Polyethylene-Wood Plastic Composites Weathering of High-Density Polyethylene-Wood*. *February 2014*, 37–41. https://doi.org/10.1080/02773813.2013.827209

Teuber, L., Osburg, V., Toporowski, W., Militz, H., & Krause, A. (2015). Wood polymer composites and their contribution to cascading utilisation. *Journal of Cleaner Production*. https://doi.org/10.1016/j.jclepro.2015.04.009

Totaro, G., Sisti, L., Fiorini, M., Lancellotti, I., Andreola, F. N., & Saccani, A. (2019). *Formulation of Green Particulate Composites from PLA and PBS Matrix and Wastes Deriving from the Coffee Production*. *0123456789*.

Ur, A., Shah, R., Jalil, A., Sadiq, A., Alzaid, M., Naseem, M. S., Alanazi, R., Alanazi, S., Alanzy, A. O., Alsohaimi, I. H., & Malik, R. A. (2023). *Effect of Rice Husk and Wood Flour on the Structural , Mechanical , and Fire-Retardant Characteristics of Recycled High-Density Polyethylene*.

Vidakis, N., & Petousis, M. (2021). *Sustainable Additive Manufacturing : Mechanical Response of High-Density Polyethylene over Multiple Recycling Processes*. 1–14.

Vincent, G. A., Bruijn, T. A. De, Wijskamp, S., Iqbal, M., Rasheed, A., Drongelen, M. Van, & Akkerman, R. (2021). Composites Part C : Open Access Characterisation and improvement of the quality of mixing of recycled thermoplastic composites. *Composites Part C: Open Access*, *4*(October 2020), 100108. https://doi.org/10.1016/j.jcomc.2021.100108

Woldie, W. A., Shibeshi, N. T., & Kuffi, K. D. (2025). Optimization of cellulose nanocrystals extraction from teff straw using acid hydrolysis followed by ultrasound sonication. *Carbohydrate Polymer Technologies and Applications*, *9*(February), 100707. https://doi.org/10.1016/j.carpta.2025.100707

Wright, S. L., & Kelly, F. J. (2017). *Plastic and Human Health: A Micro Issue?* https://doi.org/10.1021/acs.est.7b00423

Yang, H. (2007). *Characteristics of hemicellulose , cellulose and lignin pyrolysis*. *86*, 1781–1788. https://doi.org/10.1016/j.fuel.2006.12.013

Yang, H., Kim, H., Son, J., Park, H., Lee, B., & Hwang, T. (2004). *Rice-husk flour filled polypropylene composites ; mechanical and morphological study*. *63*, 305–312. https://doi.org/10.1016/S0263-8223(03)00179-X

Zarrinbakhsh, N., Wang, T., Rodriguez-uribe, A., Misra, M., & Mohanty, A. K. (2016). *com Characterization of Wastes and Coproducts from the Coffee Industry for Composite Material Production*. *11*(Cc), 7637–7653.

Zhang, Q., Zhang, D., Xu, H., Lu, W., Ren, X., Cai, H., Lei, H., Huo, E., Zhao, Y., Qian, M., Lin, X., Villota, E. M., & Mateo, W. (2020). Biochar filled high-density polyethylene composites with excellent properties: Towards maximizing the utilization of agricultural wastes. *Industrial Crops and Products*, *146*(January), 112185. https://doi.org/10.1016/j.indcrop.2020.112185

zian zhang, V. H. and D. R. (2023). *Blending Recycled High-Density Polyethylene HDPE (rHDPE) with Virgin (vHDPE) as an Effective Approach to Improve the*.

Zulkifli, N. I., Samat, N., Anuar, H., & Zainuddin, N. (2015). Mechanical Properties and Failure Modes of Recycled Polypropylene/Microcrystalline Cellulose Composites. *JOURNAL OF MATERIALS&DESIGN*. https://doi.org/10.1016/j.matdes.2014.12.053

Appendices

Appendix A: Proximate Analysis of Raw Coffee Silver Skin

Table A.1: Proximate Analysis Results of CSS

Analysis Parameter	Standard Method	Value (%)
Moisture content	Moisture analyzer	9.86
Ash Content	ASTM D3174	7.41
Volatile Matter	ASTM D3175	85.45

Appendix B: Additional Mechanical Properties of Fabricated Bio-composites

Table B.1: Tensile and Flexural Properties of Fabricated Bio-composites from BBD Runs

	Factor 1	Factor 2	Factor 3	R - 1	R- 2	R-3
Run	A:Temperature °C	B:Particle size µm	C: Filler Ratio %	Tensile Modulus MPa	Flexural Strength MPa	Flexural Modulus MPa
1	180	300	30	249	5.9	532.9
2	160	500	10	160	4.95	203.56
3	160	300	20	182	6.82	264.5
4	160	500	30	343.2	4.78	964.53
5	160	100	30	354	8.36	226.2
6	180	300	10	27	6.2	493.73
7	160	300	20	182	6.82	264.5
8	180	500	20	143.8	5.03	663.75
9	180	100	20	243	18.31	87.6
10	140	300	10	136	7.87	126.8
11	140	100	20	223	16.98	85.2
12	160	300	20	165	7.88	265
13	140	300	30	352	6.2	493.73
14	140	500	20	336	5.02	560.3
15	160	100	10	177.7	20.42	68.5

Table B.2: ANOVA for Tensile modulus Model

Model	1.254E+05	9	13930.69	50.31	0.0002	**significant**
A-Temperature	18451.20	1	18451.20	66.63	0.0004	
B-Particle Size	27.01	1	27.01	0.0975	0.7674	
C-Filler Ratio	79500.78	1	79500.78	287.09	< 0.0001	
AB	11257.21	1	11257.21	40.65	0.0014	
AC	9.00	1	9.00	0.0325	0.8640	
BC	11.90	1	11.90	0.0430	0.8439	
A^2	53.43	1	53.43	0.1930	0.6788	
B^2	15086.30	1	15086.30	54.48	0.0007	
C^2	1259.71	1	1259.71	4.55	0.0861	
Residual	1384.61	5	276.92			
Lack of Fit	1191.95	3	397.32	4.12	0.2013	**not significant**
Pure Error	192.67	2	96.33			
Cor Total	1.268E+05	14				

Table B.3: ANOVA for Flexural Strength

Model	380.26	9	42.25	30.97	0.0007	**significant**
A-Temperature	0.0001	1	0.0001	0.0000	0.9954	
B-Particle Size	291.01	1	291.01	213.29	< 0.0001	
C-Filler Ratio	14.72	1	14.72	10.79	0.0219	
AB	0.4356	1	0.4356	0.3193	0.5965	
AC	0.1444	1	0.1444	0.1058	0.7581	
BC	15.72	1	15.72	11.52	0.0194	
A^2	0.0694	1	0.0694	0.0509	0.8305	
B^2	52.91	1	52.91	38.78	0.0016	
C^2	2.70	1	2.70	1.98	0.2188	
Residual	6.82	5	1.36			
Lack of Fit	5.58	3	1.86	3.00	0.2602	**not significant**
Pure Error	1.24	2	0.6209			
Cor Total	387.08	14				

Table B.4: ANOVA for Flexural Modulus

Model	9.521E+05	9	1.058E+05	58.64	0.0002	**significant**
A-Temperature	1613.69	1	1613.69	0.8946	0.3877	
B-Particle Size	4.553E+05	1	4.553E+05	252.41	< 0.0001	
C-Filler Ratio	3.781E+05	1	3.781E+05	209.60	< 0.0001	
AB	2552.78	1	2552.78	1.42	0.2876	
AC	1245.03	1	1245.03	0.6902	0.4439	
BC	86192.15	1	86192.15	47.78	0.0010	
A^2	1462.01	1	1462.01	0.8105	0.4092	
B^2	22127.59	1	22127.59	12.27	0.0172	
C^2	6029.05	1	6029.05	3.34	0.1271	
Residual	9019.45	5	1803.89			
Lack of Fit	8028.59	3	2676.20	5.40	0.1602	**not significant**
Pure Error	990.86	2	495.43			
Cor Total	9.611E+05	14				

Table B.5: Fit Summary for the developed model of Tensile Modulus

Std. Dev.	16.64	**R^2**	0.9891
Mean	218.25	**Adjusted R^2**	0.9694
C.V. %	7.62	**Predicted R^2**	0.8461
		Adeq Precision	23.3113

Table B.6: Fit Summary for Flexural Strength

Std. Dev.	1.17	**R^2**	0.9824
Mean	9.18	**Adjusted R^2**	0.9507
C.V. %	12.73	**Predicted R^2**	0.7621
		Adeq Precision	16.8054

Table B.7: Fit Summary for Flexural Modulus

Std. Dev.	42.47	**R^2**	0.9906
Mean	325.35	**Adjusted R^2**	0.9737
C.V. %	13.05	**Predicted R^2**	0.8640
		Adeq Precision	26.2969

Appendix C: Photographic Documentation of Laboratory Procedures

Raw coffee silver skin

Grounded Coffee silver

Alkali-Treatment

Filtration

After 1st & 2nd stage bleaching

Dried treated CSS

Size reduced rHDPE

Compounding of CSS & rHDPE

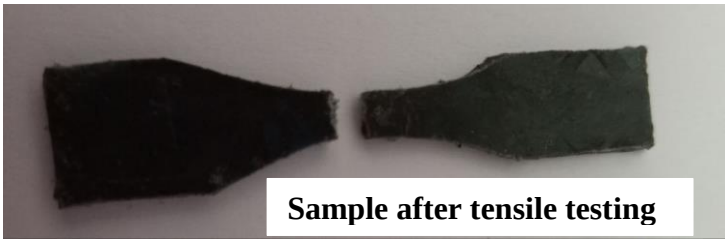
Extruded pellets

Size reduced pellets

Molded prototype

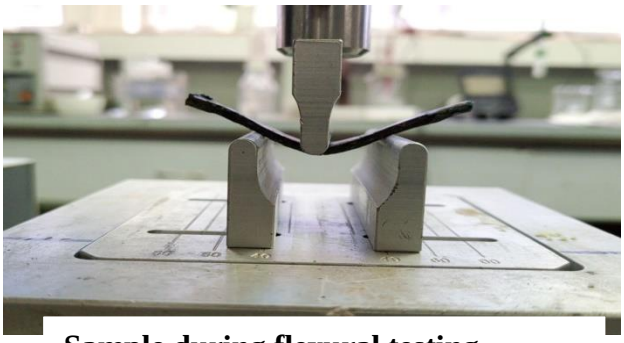
Dumb-bell shape

Universal Testing Machine



Sample after tensile testing

Texture Analyzer Machine



Sample during flexural testing

Figure C.1: Experimental procedures' photograph confirmations