

Effect of Modified Waste Bio-Filler for Sustainable Reinforced Polymer Composite for a Circular Economy

Raza, Ali; Mushtaq, Asim^{*+}; Saleem, Ahmed

Polymer & Petrochemical Engineering Department, NED University of Engineering & Technology, Karachi, Sindh, Pakistan.

ABSTRACT: Circular economy aims to create a more sustainable and resilient economic system by closing the loop on material flows, reducing environmental impact, and leading to a restorative and regenerative approach to production and consumption. The study focuses on repurposing recycled expanded polystyrene (R-EPS), commonly found as disposable utensils, and incorporating spent tea leaves powder (STLP), a globally popular beverage byproduct. The study seeks to enhance bio-composite properties through STLP integration with surface treatment and modifications. The materials to be used, R-EPS and STLP, are collected through waste management and pre-treated with organic surfactants before being dried in an oven at elevated temperatures to remove any moisture present. STLP is subsequently dispersed within the R-EPS polymer matrix in two-roll mill, followed by crushing and compression molding to produce ASTM standard samples. The samples of untreated and treated R-EPS/STLP are tested for both mechanical and thermal properties. The results indicate improvements in various properties. Tensile strength, modulus, flexural strength, and impact strength increased by 30%, 40%, 17%, and 23% respectively. Elongation and hardness experienced a decrease of 33.3%, and 9.9%. The optimal enhancement is observed with the addition of 3 wt. % of the treated STLP into the polymer matrix. Moreover, thermogravimetric analysis shows an increase in thermal stability with reinforcements significant rise of 10°C. Water absorption test revealed no significant water absorbance capability in the composite, whether incorporating untreated or treated STLP in (R-EPS). Fourier transform infrared spectroscopy (FTIR) are used to analyze the morphological characteristics and chemical bonding in treated R-EPS/STLP. The results reveal that the chemically treated STLP exhibits compatibility with the R-EPS and can be used to fabricate a bio-composite polymer for various industrial packaging applications. Therefore, this research highlights the broader implications by demonstrating the potential of repurposing waste materials into value-added products with enhanced properties.

KEYWORDS: Circular economy, expanded polystyrene, spent tea leaves, surface treatment, bio-fillers mechanical strength, thermal stability, water absorption.

INTRODUCTION

The circular economy is a green approach that focuses on minimizing environmental impact through efficient recycling, optimizing resource usage, and encouraging the reduce, reuse, and recycle principles for waste management [1]. Recycling plastic waste and reducing food waste is a key aspect of a circular economy, as it has the potential to lower the demand for new resources and minimize landfill waste. The idea of sustainability has led to a focus on reducing waste, promoting the reuse of materials, and encouraging recycling of products. The world population was approximately 7.7 billion in 2019, and it is predicted to reach 9.7 billion by 2050 [2, 3]. This upsurge in the population will result in a 50% rise in global food demand. In the European Union, the household and manufacturing sectors jointly generate about 89 million tonnes of food waste annually, with households contributing 42% and manufacturing contributing 38% to this total [3, 4]. Plastic films are popular for food packaging due to their distinguishing characteristics like ease of processing, transparency, flexibility, low weight, and cost-effectiveness, and mark the final phase of food processing. The primary materials used for these films are synthetic polymers such as low-density polyethylene (PE-LD), high-density polyethylene (PE-HD), polypropylene (PP), polyethylene terephthalate (PET), polyvinyl chloride (PVC), ethylene vinyl alcohol (EVOH), and polystyrene (PS). PE-HD and PE-LD are commonly used in the production of flexible and rigid food packaging films [5, 6]. These materials are characterized by their affordability, low water vapor permeability, excellent tear resistance with high elongation at break, and good thermal stability. Expanded polystyrene (EPS), also known as thermocol, is a material made from pre-expanded polystyrene beads. It has many useful advantages like being lightweight, rigid, and easy to shape. EPS is used in various applications like Trays, dishes, bowls, and containers, thermal insulation, packaging, and building walls and roof materials are common examples [4, 6]. In the context of the global annual production of approximately 15 million metric tons (MMT), the majority of EPS finds its way into landfills. Due to its wide usage and inadequate recycling rates, EPS is recognized as a significant contributor to the solid waste issue. EPS has emerged as a favored option for food takeout containers due to its lightweight, excellent thermal insulating properties, and auspicious impact resistance [7, 8]. An excessive amount of waste from food packaging is being thrown into the environment, leading to pollution due to its non-biodegradable nature, causing long-lasting harm to the environment and nature. Ensuring the recycling or reutilization of plastics already in circulation is essential. This helps mitigate the growing occurrence of unplanned or intentional discharge of these materials into the environment. By doing so, we have the potential to successfully fight and reduce environmental pollution through effective actions and strategies [9, 10]. The process of recycling involves various modifications and transformations, such as mechanical processing, chemical processing, or thermal treatment, to extract valuable feedstock from a previously used polymer. This recovered feedstock can be reintroduced into the industry for reuse. Nowadays, there are various methods of recycling plastic, ranging from simple techniques to more advanced processes, categorized from primary to quaternary levels [11, 12].

Presently, there exist four distinct methods for recycling plastics: The categories of plastic recycling methods consist of primary, and secondary recycling (mechanical recycling), tertiary recycling (chemical recycling), and quaternary recycling (energy recovery). Both primary and secondary recycling are types of mechanical recycling for plastics. This involves sorting, washing, crushing, melting, and remolding to produce new products that replicate the characteristics of the initial plastic [13, 14]. In mechanical recycling with the application of thermal conduction and shearing within an extruder under the influence of shear forces, the molecular chains of polymers

change breaking into smaller segments (chain scission), developing side branches (branching), or forming connections between chains (crosslinking) [15, 16]. As a consequence of this chain scission, the polymer's chain length is reduced, resulting in reduced mechanical properties and making it more difficult to process. Quaternary recycling, known as incineration, is a widely used method in the processing industry to manage mixed waste plastics. It involves burning waste plastics to produce heat, which can be used for electricity generation. The high heating value of these plastics, ranging from 20 to 40 MJ/kg, this value is comparable to that of crude oil (45 MJ/kg) [2, 4]. Incinerating plastic waste for energy recovery is a common practice in some countries to save landfill space and offset fossil fuel-based energy production. However, it is controversial because of its inefficiency in recovering energy and the risk of releasing harmful compounds such as dioxins, furans, and greenhouse gases that contribute to global warming [16, 17]. Tertiary recycling, also known as chemical recycling, provides a dynamic and evolving solution to address the global plastic challenge. Chemical strategies like glycolysis and thermolysis widen the range of recyclable plastics by employing depolymerization techniques to convert post-consumer polymers back into their original monomers, oligomers, or novel chemical feedstocks. Chemical strategies face difficulties, particularly with certain polymers that are hard to depolymerize into their original monomers through reverse synthesis reactions. Additionally, certain chemical recycling methods, like the pyrolysis process that operates at high temperatures without oxygen, require even more significant energy input. Consequently, this process often generates complex mixtures of oils, waxes, and hydrocarbon gas, which then require additional processing [18, 19].

Even though completely replacing synthetic plastics with biodegradable materials might not be realistic, using more biodegradable options can still help protect the environment's challenges. Food packaging accounts for a substantial 40% of all plastic consumption, with nearly 20% being utilized for disposable items like bottles, sheets, and cups. This poses a significant challenge in the global efforts to reduce waste. Between 1988 to 2005, there was a staggering 37% surge in solid waste, with packaging accounting for a significant 31.2% of the overall solid waste generated [1, 20]. To improve packaging techniques by the utilization of bio-additives to develop biodegradable packaging that not only safeguards food but also improves its shelf life. Biopolymers provide the advantage of being compatible with synthetic polymers, enhancing their functionality and offering improved protection. Nevertheless, it is important to note that biopolymers frequently fail to meet the minimum standards necessary for food packaging, and their performance still falls behind that of conventional materials [3, 4]. For mitigating various challenges associated with biopolymer-based packaging materials it is essential to enhance the industrial viability of biopolymers. An innovative approach to utilizing Expanded Polystyrene (EPS) is by transforming it into hybrid composites through the incorporation of inorganic fillers, but inorganic fillers are typically derived from non-renewable resources and are not biodegraded to any food packaging, which can contribute to waste and more pollution. However, in response to these limitations, manufacturers have fabricated composite materials using a range of fillers for specific applications [21, 22]. Biofillers or natural fillers derived from renewable resources have several advantages over inorganic fillers in composite manufacturing, including sustainability, biodegradability, cost-effectiveness, improved mechanical properties, compatibility with the matrix, and versatility. These advantages make them an effective option for the development of new and improved composite materials [17, 23]. Hybrid composites in the polymer industry are becoming more popular because they use organic fillers that provide better qualities than inorganic fillers. These fillers offer improved properties, making the composites more desirable for various applications. In the past, fillers were mainly used to reduce

material costs and enhance the performance of composites, focusing on aspects like strength, stiffness, and rigidity. However, there is now a growing interest in improving these composite resist water vapor transmission. Much literature highlights that limited exploration has been done considering different bio fillers used, such as Vinyl ester composites reinforced with 30% KPF fibers exhibited enhanced mechanical properties, such as a peak tensile strength of 125 MPa, a 12% improvement in impact energy absorption, and an elevated thermal decomposition temperature of 399°C. 5 wt% silane treatment to natural cellulosic *Kigelia africana* fruit fiber (KAF) resulted in an enhanced crystallinity index of 70.22% and a significant increase in tensile strength, reaching 490.77 MPa. Silane treatment of fish tail palm fibers improved cellulose content (72.51% at 9% silane), enhanced crystallinity (62.5% at 5% silane), and confirmed hemicellulose removal by FTIR. Tensile strength also significantly increased by 7.3%, 12%, and 6.6% for fibers treated with 1%, 5%, and 9% silane, respectively. Silane treatment enhances fish tail palm (FTP) fiber strength in a vinyl-ester matrix. The 30% silane-treated composite demonstrates increased tensile strength (109 MPa) and flexural strength (138 MPa), with a notable 61.3% rise in crystallinity, reinforcing FTP fiber structure [24-27]. More similar *Acacia Caesia* Bark, *Zea* agricultural residue, black limestone, tamarind nut powder, pinecone particles, almond shell flour, date palm leaflets, sawdust and rice husk, and banana leaves in terms of their performance in improving tensile, thermal, increasing modulus, impact on mechanical, impact strength and percent elongation, water barrier properties, elongation at break and impact-absorbed energy [28-34].

To be considered spent tea leaves powder as fillers in a polymer matrix has many benefits, such as being sustainable, cost-effective, biodegradable, improving mechanical and thermal properties, and having versatile applications. These advantages make the tea leaves a promising option for the development of new and Hybrid composite. It is imperative to prioritize and invest resources in finding sustainable and eco-friendly alternatives to food packaging, specifically in the context of tea leaves waste, to promote a more circular and sustainable future [35-38].

This research aims to produce a bio-composite utilizing EPS and STLP with the objectives of reducing the carbon footprint and promoting waste recycling within the circular economy. By incorporating bio fillers spent tea leaves powder into conventional EPS and promote recycling. The utilization of STLP to prepare a renewable bio-polymer composite filled with EPS has been investigated for the first time in this study. The mechanical properties of the composite films were assessed under load, and thermogravimetric analysis (TGA) was conducted to determine their thermal stability. The interaction between the filler and matrix was examined using Fourier transform infrared (FT-IR) spectroscopy. Additionally, the morphology of the composite films was observed using optical microscopy (OM) and scanning electron microscopy (SEM).

EXPERIMENTAL SECTION

Expanded Polystyrene Waste

Recycled expanded polystyrene, found in the form of utensils and containers, poses a significant challenge in terms of degradability, contributing substantially to global pollution. R-EPS being non-hygroscopic makes it easier to remove the contaminants by simply washing thoroughly using surfactant and water and the R-EPS were dried at room temperature to eliminate any moisture content, the presence of moisture could potentially impact the proper mixing of materials and in subsequent stages, it might lead to issues during the production.

Spent Tea Leaves Powder (STLP) Waste

Spent Tea Leaves Powder commonly found in households and hotels, is collected in large quantities after use. These STLPs undergo an initial cleaning process with a surfactant to remove any residual fats or particles of milk and are then washed multiple times with distilled water to ensure the removal of any particles that might have accumulated during their previous use. Grinding STLP to decrease their particle size and then sieving them to achieve a finely powdered form with specific particle sizes predetermined before their combination with R-EPS.

Waste Collection

The biggest aspect of this study is the usage of waste-expanded polystyrene (R-EPS) to address global pollution and reduce carbon emissions. EPS, commonly used in packaging for it has good insulating properties, was obtained easily from local markets, educational institution canteens, and scrap collectors in the form of cups and food boxes. Tea, one of the most consumed beverages in the world, particularly in Southeast Asia, is a natural source that can be easily found in bulk and is a good option as a natural filler due to its easy availability. The STLP in our project was collected directly from various commercial tea stations and canteens.

Sample Preparation

The schematic diagram shown in Figure 1 explain the process of chemical modification and the steps involved in sample preparation.

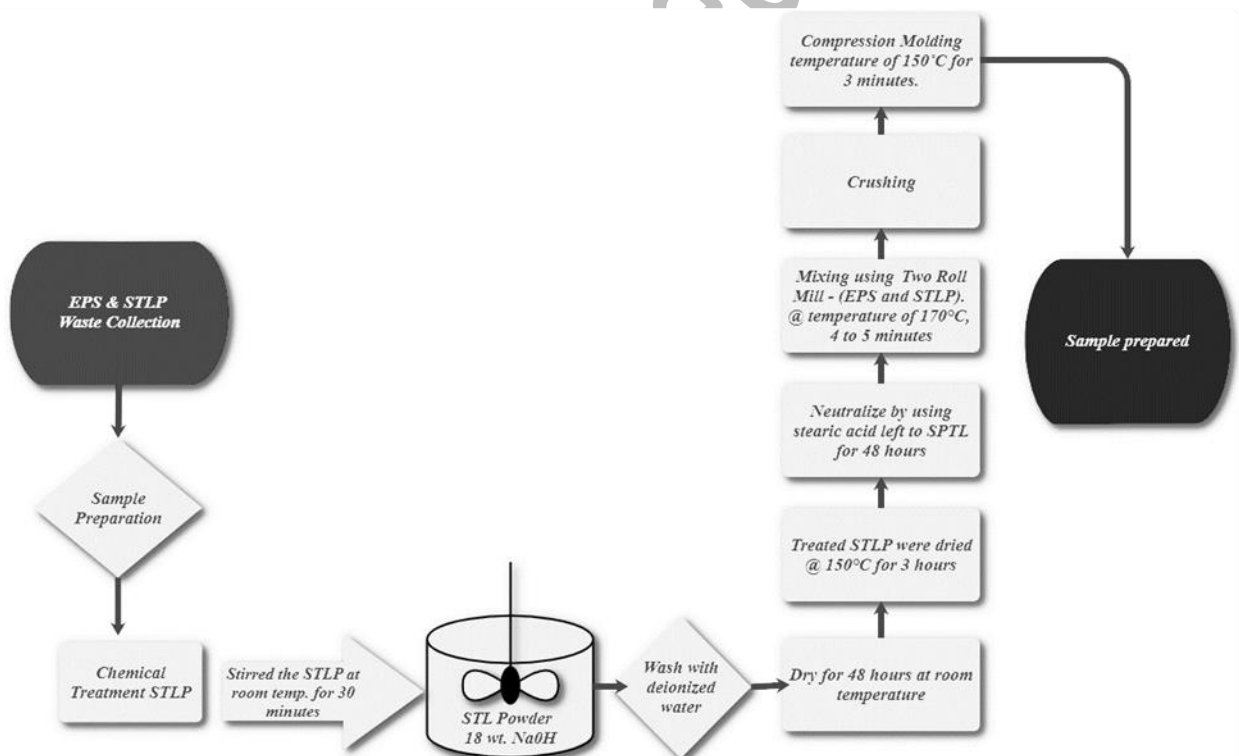


Fig. 1. Chemical modification and sample preparation

Drying

STLP is hydrophobic, it tends to absorb moisture. Moisture is not beneficial during the production. It might cause the formation of voids. To completely remove the moisture the STLP sample is sieved and oven-dried in a

Forced Convection Oven (Model: Jsof-100) for 4 hours at a temperature of 80°C until the weight of the tea sample becomes constant.

Two Roll Mill Mixing

The waste samples of R-EPS and STLP were mixed according to pre-determined weight percentages, using a Two Roll mill from Labtech Engineering Co. Ltd. (model: LRMR-S-110/W), at a temperature of 170°C and a 0.40 mm gap between the rollers. The samples are allowed to mix for a period of 4 minutes to 5 minutes to achieve proper mixing. For each batch, the STLP was dispersed into the R-EPS Matrix and passed through the rollers multiple times until adequate mixing. The material was peeled off and cooled at room temperature until hardened samples were obtained. Samples of different filler compositions were produced.

Crushing

The combined samples of different compositions produced through the two-roll mill are subsequently crushed with the help of a Hammer mill from 'Christy-Turner' (model: 3922C) at room temperature and turned into flaky powder which was then utilized for further sample preparation using the compression press.

Compression Molding

Compression molding is a closed molding process used to fabricate various composite products. It involves placing raw materials in pallet or powder form are placed within a mold cavity. This cavity is formed by applying heat and pressure. The material is first pre-heated in a mold between two plates at a temperature of 150°C for a short span of 3 minutes in a laboratory press molding. It is then suddenly compressed at the same temperature and time, at approximately 100 bars pressure to obtain standardizing testing samples of various compositions of about 0, 0.5, 1, 3, and 5 wt. % respectively.

Chemical Modification

It is an effective method for enhancing the properties of the composite by removing the hemicellulose and lignin from the natural filler. This process results in better chemical bonding, contributing to the overall improvement of the composite's characteristics. This achievement is attained through the treatment with NaOH (Sodium Hydroxide). In the First step, the sample was placed in a flat-bottomed round flask along with a solution containing 18% NaOH. Stirred the sample at room temperature for 30 minutes and filtered using paper. The modified STLP goes through a wash with deionized water. Subsequently, the sample was left to dry for 48 hours at room temperature.

To neutralize by using stearic acid, in the second step, as shown in Figure 2, a solution is made by mixing 3.98 grams of stearic acid in 200 mL of toluene in a beaker. The stearic acid was completely dissolved in toluene by thoroughly mixing the solution with a magnetic stirrer. STLP was added to this solution and left to settle in a hood for 48 hours. Afterward, the STLP was separated from the solution using filtration by filter paper and a funnel. The wet STLP were dried in a forced convection oven at around 150°C for 3 hours. Once dried, the tea leaves were sealed in an airtight container to prevent moisture contact. The chemical modifications, enhancing polar groups, have significantly improved the properties.



Fig. 2. Chemical modification (NaOH-Toluene Treated STLP)

Scanning Electron Microscope (SEM) Test

SEM, an essential technique in polymer research, allows for the examination of filler morphology and distribution within the polymer matrix, providing valuable insights into their structure and arrangements. SEM analysis, which uses high-resolution imaging, is useful for inspecting various materials for surface fissures, flaws, contaminants, or corrosion. It has also the capability to identify agglomeration and non-uniform dispersion filler. As the electron beam scans the surface, it triggers the emission of secondary electrons and results in the backscattering of some incident electrons. By measuring the intensity of these secondary or backscattered electrons and comparing them to the scanned beam, a contrasted image is generated, and the surface characteristics of the composite are shown on the screen. This research going on to analyze the distribution of STLP within EPS, encompassing both treated and untreated variations. EPS/1 wt. % STLP as well as chemically treated EPS/1 wt. % STLP composites were examined. The images were obtained on a micro-scale which gives a clear understanding of the porosity and surface properties of the composites.

Tensile Testing

The developed hybrid composite samples were subjected the tensile tests analysis following the ASTM D638-10 standard at room temperature (25°C). Using a Zwick/Roell Universal testing machine equipped with a 5 kN load cell, the tests were performed on dumbbell-shaped samples with varying filler weights, ranging from 0 to 5 wt. %. The samples, with filler loading, gradually increased from 0.5 wt. % to 5 wt. % were subjected to tests at a cross-headed speed of 5mm/min. Each sample had a neck width of ± 13.5 mm, a thickness of ± 3.5 mm, and a gauge length of 50 mm as illustrates in Figure 3 (a, b, c). For optimal grip, the samples were positioned in the machine to ensure effective jaw contact at both ends. Each test was run three times, and the average result from these trials was considered as the outcome.

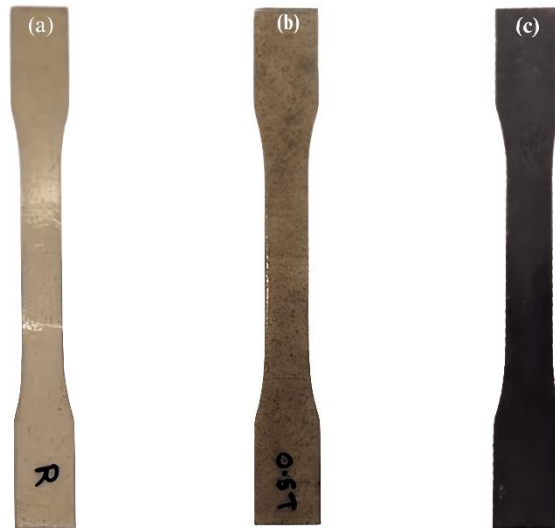


Fig. 3. a) R-EPS containing 1 wt. % Untreated STLP b) R-EPS containing 3 wt. % treated STLP c) R-EPS containing 5 wt. % treated STLP

Izod Impact Test

The amount of energy absorbed by the composites was conducted through Izod testing by the ASTM D4812-99 standard. The test was then performed following the standards and procedure for impact analysis with specimens sized 65 mm × 13 mm × 3.2 mm as depicted in Figure 4 (a, b).. Samples with varying filler weights, ranging from 0 to 5 wt. % of the EPS/STLP composites were produced in the shape of strands through compression molding. This measures a plastic resistance to fracture under stress. An impact force is generated by a moving hammer on standard equipment to fracture samples with a single swing of the pendulum, measuring the force produced. The outcome of this test measures the energy absorbed per unit of specimen width.



Fig. 4. a) R-EPS containing 3 wt. % treated STLP b) R-EPS containing 5 wt. % treated STLP

Shore-D Hardness Test

Hardness characterizes a material property, signifying its capacity to resist localized deformation. In the domain of polymer materials, various hardness tests apply to differently shaped indenters for evaluation. The Shore hardness test employs pin-shaped indenters, while the Barcol hardness testing uses a Barcol indenter. The specifications followed for performing the Shore-D hardness test are by the ASTM D-2240. In the hardness test, 10 indentation points can be measured on composite surfaces. The experiment involved testing various samples

with different compositions of STLP, including both untreated and treated specimens. Indenter was used to apply a consistent load of approximately 4 kg specified force and time conditions onto samples, measuring penetration force in the context of hardness. The thickness of the test samples is typically 6.4 mm. The hardness values are calculated using a scale.

Flexural Test

Flexural strength determines a material's resistance to deformation under load. The material's bend before permanently deforming is indicated by its flexural modulus. In the 3-point bend test, a specimen gets positioned between two support points, and force is exerted on a third point, the findings of the flexural test are obtained using the ASTM D790-10 standard test, which was performed on EPS, 0.5 wt. %, 1 wt. %, 3 wt. %, and 5 wt. % STLP composite specimens according to the standard shape of 3.2 x 12.7 x 125mm for each composite material. The dimension of the sample is depicted in Figure 5. The span length was kept at 50 mm while a speed of 5mm/min was used. The examination is terminated when the sample deflects by 5% or fractures before 5%.



Fig. 5. R-EPS containing 3 wt. % treated STLP

Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) is a method that analyzes how the mass of a sample changes over time as the temperature changes. In this study, both treated and untreated STLP composite and EPS materials were analyzed using TGA equipment. The temperature range used was from 50°C to 650°C, and the analysis was conducted in a nitrogen atmosphere, with the temperature increasing at a rate of 10°C per minute. The TGA-generated graphs, called thermograms, were obtained for the EPS as well as for the treated and untreated STLP. Additionally, derivative curves (DTG) were derived from these thermograms. The main focus of this research was to explore the thermal stability of recycled EPS, EPS/3 wt. % untreated STLP, and EPS/3 wt. % treated STLP. These composites were thoroughly investigated using the thermal analyzer, and the results were presented in the form of primary thermographs to gain a comprehensive understanding of the composites' thermal properties.

Fourier Transform Infrared Spectroscopy (FTIR) Test

FTIR spectroscopy is an effective analytical method used to identify functional groups within molecules to characterize the bonding present in a sample after chemical modification and evaluate the quality of polymeric products. This technique works by measuring the absorption of infrared radiation by the sample, certain bonds within the molecules absorb energy, causing them to vibrate. The remaining energy not absorbed by the sample is transmitted through it. The output observed at the detector in the form of a spectrum represents the unique molecular fingerprint of the material. By analyzing the peaks and patterns in this spectrum, researchers identify the specific functional groups and types of bonds present in the molecule at unique characteristic frequencies. The chemical composition of STLP before and after NaOH treatment was obtained using a Spectrum at FTIR spectrometer (Thermo-Fisher Scientific). The spectra were obtained from 5-time scans of the sample. Each spectrum was recorded in the range of 400 – 4000 cm^{-1} with a resolution of 4 cm^{-1} .

Water Absorption Test

The water absorption rate has two main purposes: firstly, as an indicator of how much water is consumed by composites and how it influences its characteristics, secondly it ensures that the product maintains consistent quality and uniformity. Water absorption tests were performed according to ASTM D570. The developed STLP-reinforced EPS composites were immersed in distilled water at room temperature as shown in Figure 6 (a, b). Their weight was measured before and after immersion in distilled water. Water absorption was obtained as a percentage of weight increase to the original weight. Absorption of moisture has a significant impact on the degradation of the mechanical properties of composites. Water penetrates the composites and ends up either in the matrix or at the filler matrix interphases.

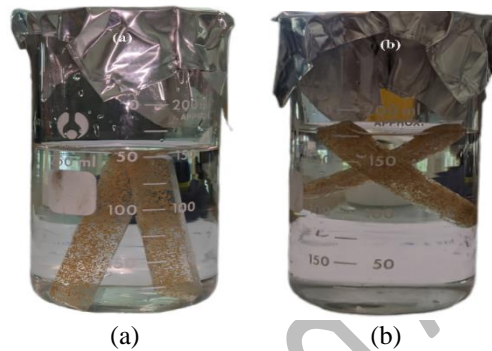


Fig. 6. a) R-EPS containing 1 wt. % of both untreated and treated STLP in a single beaker b) R-EPS containing 3 wt. % of both untreated and treated STLP in a single beaker

RESULTS & DISCUSSION

Scanning Electron Microscopy

Figure 7 presented low and high-magnification SEM images of R-EPS loaded with 1 wt. % of both untreated and chemically treated STLP. The composite with a 1 wt. % filler percentage was chosen to reflect the visible variations in surface adherence and particle size. The particle size distribution is visualized to be below $30 \mu\text{m}$ and is dispersed uniformly [4, 39]. SEM images appear to be completely devoid of gaps and spaces, demonstrating that the surface of the composite is smooth and impermeable to water. Moreover, Figure 7 (a, b) shows STLP on the surface due to poor adhesion that may lead to water absorption, later clarified in the water absorption test. However, STLP is a hygroscopic material but no bounded moisture content or swelling is observed in either case. In contrast, from Figure 7 (c, d) chemically treated STLP shows superior surface adhesion with the polymer matrix. Additionally, as compared to untreated STLP, chemical treatment induces fibril structure modification to the STLP, facilitating improved interlinking with R-EPS.

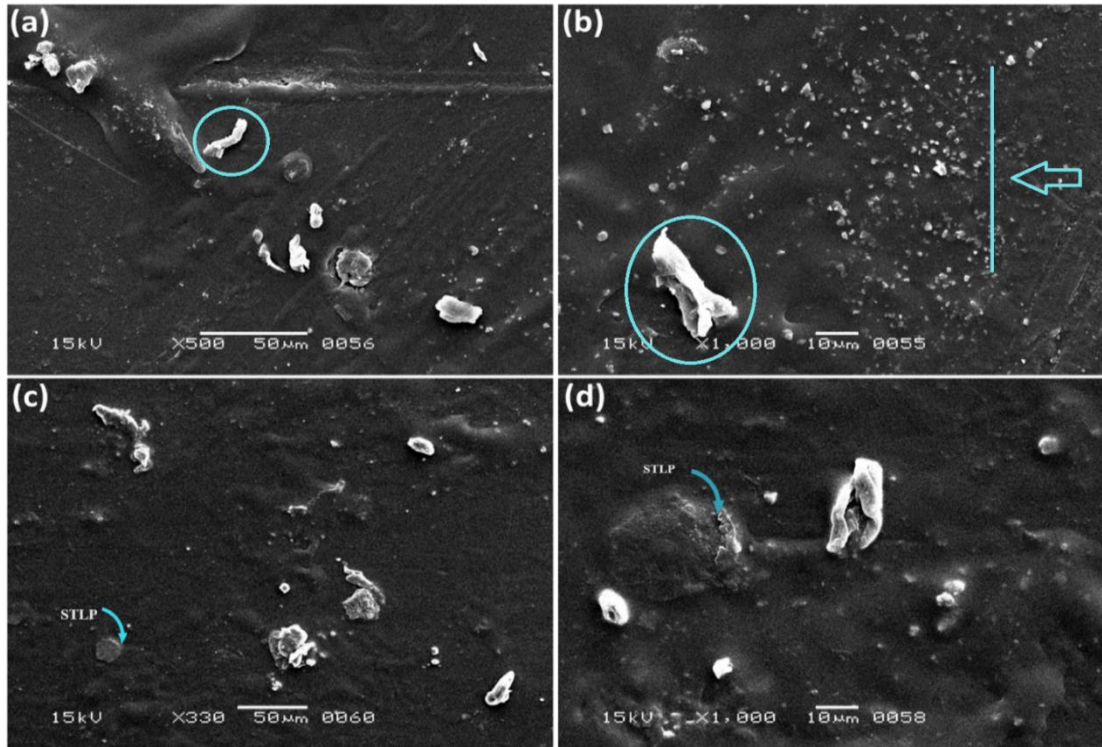


Fig. 7. SEM images of R-EPS composite with (a-b) 1 wt. % of untreated STLP and (c-d) 1 wt. % of treated STLP

Tensile Strength

Figure 8 illustrates how the tensile strength changes with different amounts of filler added. In comparison, untreated samples exhibit lower tensile strength than the samples with treated STLP across various filler percentages. This difference is primarily due to the absence of interfacial bonding in the untreated sample. As previously stated FTIR results removal of hemicellulose and lignin and treatment of the surface has enhanced the contact between the reinforcement and the matrix, resulting in better filler integration with the matrix and therefore an increase in the tensile strength [19, 30]. The tensile strength of R-EPS was measured to be 27.05 MPa. Initially, on increasing the content of filler in the matrix to 3 wt. %, there is an increase of 22.5% in tensile strength. However, a further increase in filler loading leads to a decrease in strength due to an increase in the volume fraction of the filler content and binding in the matrix, creating uneven stress distribution [11, 23].

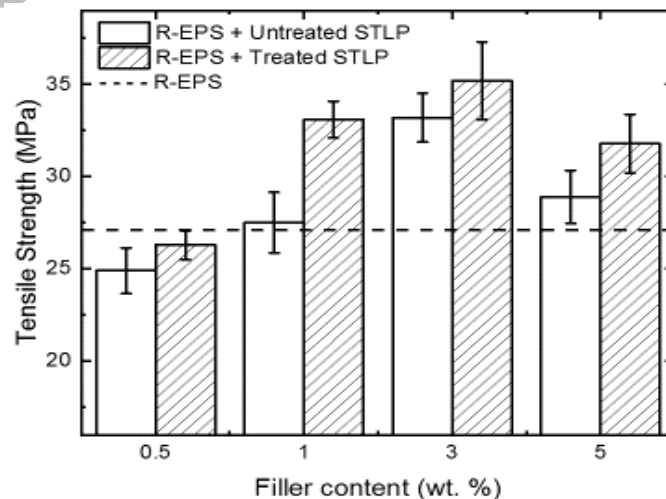


Fig. 8. Tensile strength of R-EPS, R-EPS with untreated and treated STLP composite

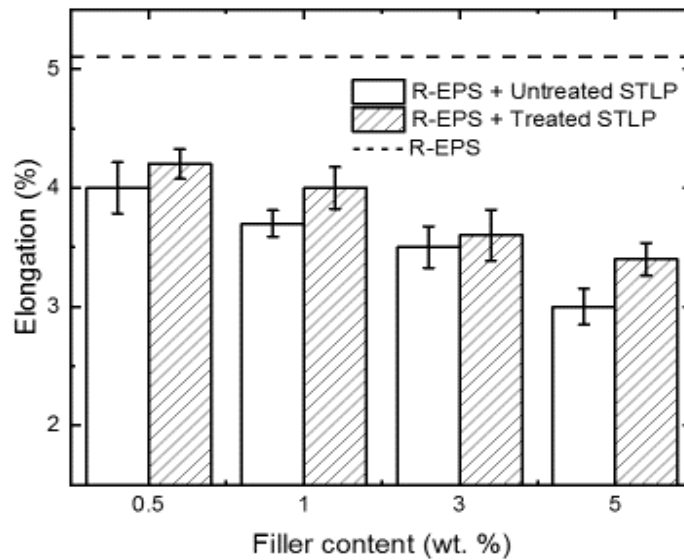


Fig. 9. Elongation at break of R-EPS, R-EPS with untreated and treated STLP composite

On the other hand, elongation at break of R-EPS with untreated and treated STLP composite displays a consistent decrease across various filler loading, as illustrates in Figure 9. For composite with untreated STLP a 21% decrease in elongation takes place with inclusion of 0.5 wt. % of filler loading. Similarly, for composite with treated STLP a 17% decrease in elongation was observed under the same filler content. This decline may be because the polymer matrix inherently tends to exhibit brittleness, especially when fillers are incorporated [2, 16]. Therefore, the presence of the STLP in the composite tends to resist plastic deformation and further restrict the mobility and deformability of the polymer matrix, resulting in a reduction of elongation at break [36, 40].

Elastic Modulus

The variation in the results of the elastic modulus for different fillers loading of both untreated and treated STLP in the composite is depicted in Figure 10. As filler loading is increased to 3 wt. %, the elastic modulus shows a remarkable increase and reaches a threshold value of 1170 MPa with a maximum increase of 39.1% in elastic modulus for treated STLP content. However, untreated samples show a maximum increase of 25.8% in elasticity. In treated samples, alkali treatment filler enhances the interfacial bonding with the polymer matrix so that the stress is distributed effectively between the matrix and dispersed phase. However, upon further increasing the filler loading, there is a decrease in the elastic modulus of the composites.

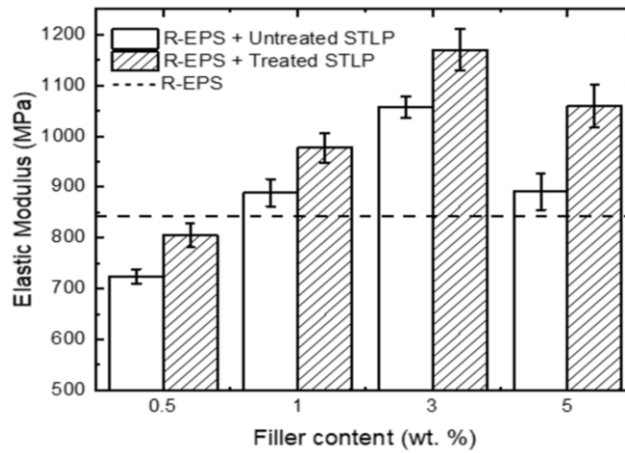


Fig. 10. Elastic Modulus of R-EPS, R-EPS with untreated and treated STLP composite

Izod Impact Strength

The Izod impact strengths of composites with adding different wt. % of filler loadings is shown in Figure 11. The unnotched impact strengths show improvement initially then diminish on increasing the filler loading. Maximum impact strength was observed at 1 wt. % which is 33.2% higher than the R-EPS, which stands at 7.392 kJ/m². On increasing filler loading, particles may act as stress concentrators, leading to localized areas of elevated stress [12, 39]. Moreover, an increase in the volume fraction of filler material in the composite alters the overall deformation behavior of the composite, leading to a significant decrease in the energy absorption before fracture. It can be seen that the resultant values of treated samples are slightly higher than the composite with untreated STLP due to bonding between dispersed and matrix phases [17, 23].

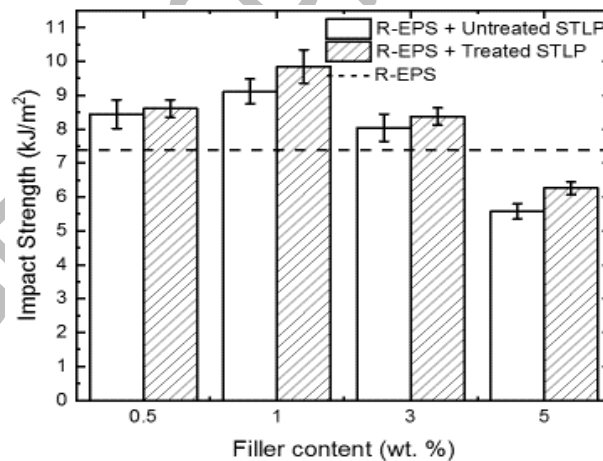


Fig. 11. Izod Impact test of R-EPS, R-EPS with untreated and treated STLP composite

Shore-D Hardness

Figure 12 illustrates the indentation hardness of the composite for both untreated and treated filler loading. As the loading of filler particles increases in the polymer matrix, the volume fraction of filler increases resulting in a decrease in the hardness of the composite. Moreover, the weakness of the filler strength itself reduces the hardness of the composite. It is observed that the values of filled composite have lower hardness as compared to R-EPS and an exponential decrease of 4% in hardness is attributed to each additional loading up to 5 wt. %.

Specifically, however, there is no significant difference in the indentation values for treated and untreated samples [31, 39].

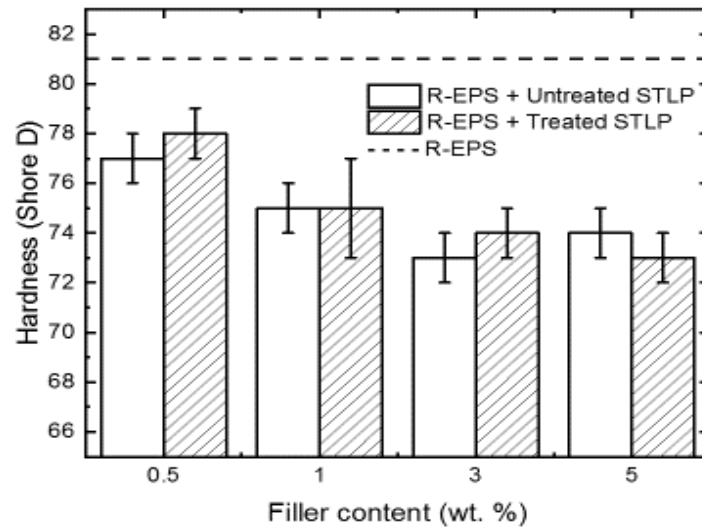


Fig. 12. Hardness Test of R-EPS, R-EPS with untreated and treated STLP composite

Flexural Strength

Figure 13 illustrates the results of the flexural strength testing reveal a significant impact of filler loading on the mechanical performance of the composite. As the filler loading increases, there is a noticeable and progressive increase in the flexural strength of the composite material. One primary contributor to the observed increase in flexural strength of the composite with treated STLP is the enhanced reinforcement effect imparted by the fillers and improved interfacial bonding. This reinforcement provides additional support to the composite material, resisting deformation and enhancing its ability to withstand bending loads. The results also highlight the optimization of filler loading at 3 wt. % to achieve superior flexural strength with a cumulative improvement of 10.7% for untreated and 16.8% for treated samples compared to the unfilled polymer matrix. However, the filler particles at 5 wt. % and more, when concentrated may introduce stress concentrations or disrupt the uniformity of the matrix, potentially compromising the material's overall flexibility [23, 31].

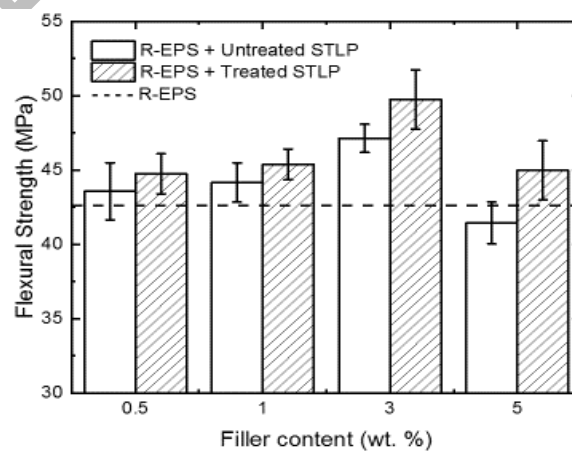


Fig. 13. Flexural strength of R-EPS, R-EPS with untreated and treated STLP composite

Thermogravimetric Analysis (TGA) Result

Thermogravimetric analysis (TGA) was carried out to study the thermal stability of recycled EPS, R-EPS containing 3 wt. % untreated STLP, as well as R-EPS containing 3 wt. % treated STLP composite as illustrates in Figure 14. No significant changes were observed at the initial stage, indicating no moisture content was present. Furthermore, no amount of volatile compound used during the chemical treatment was found either.

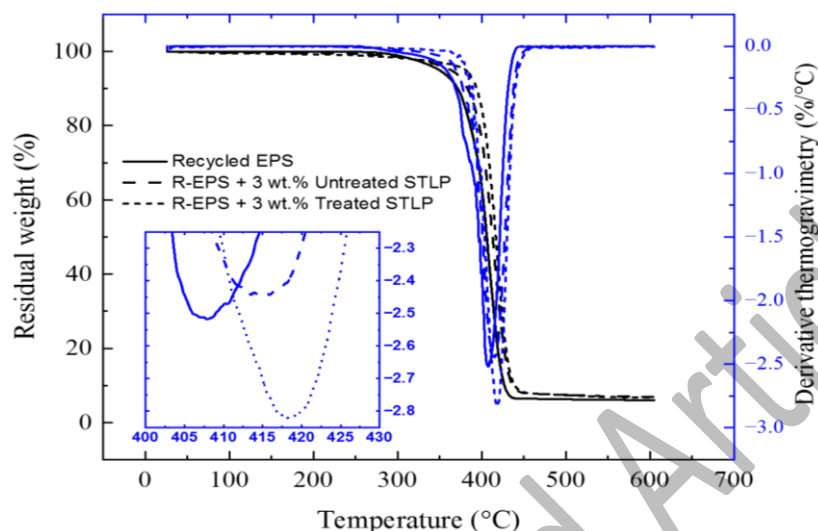


Fig. 14. Combined TGA and DTG Curve for Recycled EPS and R-EPS with 3 wt. % STLP untreated and treated composite

Moreover, to visualize better first derivative curves between temperature and residual weight are plotted to identify substantial changes during thermal analysis. From these derivative curves, temperature and degradation data are extracted and shown in Table 1. Upon observing the values, T-peak shifts to higher temperatures when filler loading is added to the R-EPS especially with treated STLP with maximum rate of decomposition at peak temperature. This suggests improved thermal stability due to the presence of fillers binding and hindering the decomposition. Moreover, filler incorporation influences the temperature for completion although energy for complete decomposition is decreased as shown in the area under the curve data. This is attributed to the improved thermal conductivity and heat dissipation capabilities introduced by the fillers and promoting the formation of a more protective and thermally stable char layer during degradation. However, initially STLP in the composite may hinder the polymer chains leading to early initialization of the decomposition, as can be seen with the onset temperature of both 3 wt. % filled composite. This data also suggests that chemical treatment significantly affects particulate at elevated temperatures [4, 9, 31].

Table 1. Degradation temperature-based data from TGA/DTG curves for composite with filler loading and treatment

Composite samples	T-onset (°C)	T-peak (°C)	T-offset (°C)	Maximum rate of decomposition temperature (%/°C)	Area under the curve during degradation (%)
R-EPS	271.218	407.225	441.598	2.62047	93.38035
R-EPS + 3 wt. % Untreated STLP	253.2315	414.251	547.301	2.50238	93.3144
R-EPS + 3 wt. % Treated STLP	253.551	417.909	548.359	2.89983	93.34053

Fourier Transform Infrared Spectroscopy (FTIR)

FTIR spectra of R-EPS, composite with 3 wt. % untreated STLP, and composite with 3 wt. % treated STLP composites were examined to identify the chemical composition and study the interaction of functional groups as presented in Figure 15. In the R-EPS spectrum, 3026.2 cm^{-1} (aromatic C-H stretching), and 2925.5 cm^{-1} (C-H stretching) were observed. Weak overtone peaks of the benzene vibrations are identified between $2000 - 1750\text{ cm}^{-1}$. The additional peak at 1602.1 cm^{-1} represents aromatic C=C bond stretching, while 1027.8 cm^{-1} indicates aromatic C-H deformation vibration. No additional chemical traces were identified, especially peaks at $3400\text{--}3500\text{ cm}^{-1}$ (O-H stretching).

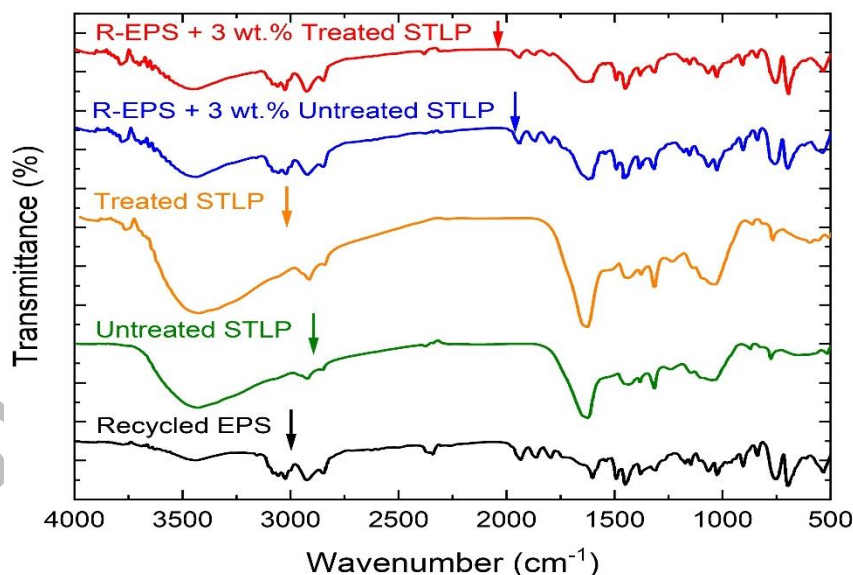


Fig. 15. FTIR spectra of recycled EPS, untreated and treated STLP, 3 wt. % untreated and treated STLP/R-EPS composite

Both untreated and treated STLP spectrum of resembled lignocellulose with peaks at 3430.1 cm^{-1} (bonded O-H groups), 2924.3 cm^{-1} (Aliphatic C-H groups), and shoulder band at 1625.2 cm^{-1} is attributed to carbonyl (C=O) stretching of the carboxyl (-COOH) group. Peaks within the range of 1148.3 to 1053.1 cm^{-1} may signify C-O

stretching, and those between 652.6 and 780.1 cm^{-1} could represent aromatic substituted groups, although this determination may be less accurate due to their location in the fingerprint region. FTIR research showed that the STLP possessed lignocellulose's chemical content. However, the intensity of O-H and C=O functional groups are reduced for treated STLP, suggesting reduced hemicellulose and lignin interaction with the polymer matrix after post-treatment with stearic acid [4, 39]. Moreover, the presence of toluene was not found in either STLP sample. Combinational peaks are observed with different intensities of absorbance at hydroxyl and carbonyl groups bonding for composite with untreated and treated STLP, which shows the inclusion of filler in the polymer matrix.

Water Absorption Test Result

Figure 16 illustrates that treated STLP sample generally have less water absorption compared to the untreated STLP samples. It can be seen that compared to R-EPS, the composite seems to have absorbed water, however quantitatively it is not significant enough. Furthermore, as interpreted from the functional groups in FTIR spectra, due to chemical modification composite with treated STLP appears to be effective in resisting water absorption, with a more significant impact at the higher filler loading. Specifically, the reduction in water absorption is more pronounced at 3 wt. % (0.31% to 0.15%) compared to 5 wt. % (0.61% to 0.27%). SEM images as discussed previously also support this notion as the composite was found to be impermeable having a smooth surface [11, 31].

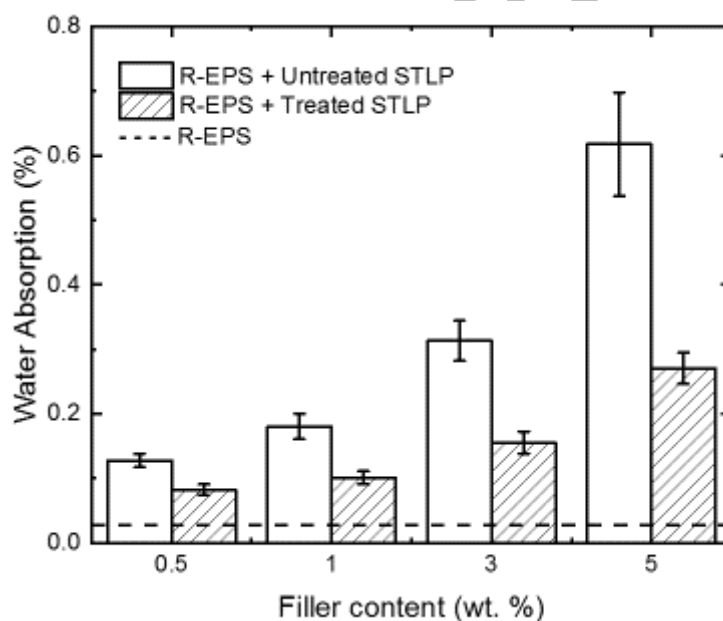


Fig. 16. Water absorption of R-EPS, R-EPS with untreated and treated STLP composite

CONCLUSION

In conclusion, this scientific research paper presents a comprehensive exploration of the circular economy, focusing on the application of bio-filler composites as an alternative to conventional polymer composites. The study emphasizes the pressing issue of plastic waste accumulation globally and underscores the growing significance of bio-filler in addressing this environmental challenge. STLP as a bio-filler is made more compatible with the surface treatment using NaOH as the strong base and is then incorporated within the R-EPS polymer matrix. Mixing is performed in a two-roll mill to avoid agglomeration of the STLP within the matrix, followed by

crushing the blend into fine powder and compression molded into the desired ASTM sample. Mechanical and thermal characterizations reveal the influence of untreated and treated filler content on the properties of the reinforced composite. The incorporation of fillers into the composite led to significant enhancements in mechanical properties, with maximum increases observed in tensile strength (29.8%), modulus (39.1%), flexural strength (16.8%), impact strength (23.3%), and thermal stability (rise of 10°C). However, a concurrent decrease in elongation (33.3%) and hardness (9.9%) was detected compared to the recycled plastic, indicating a trade-off between improved strength and reduced ductility and hardness in the filled composite accompanied by a noteworthy improvement in thermal stability. Treated samples show improved properties as compared to untreated samples. The SEM images confirm the smooth surface and show minimal void fraction, indicating the composite is resistant to moisture absorption, in contrast to STLP being hygroscopic material. Water absorption tests further supported the composite's impermeability, showing minimal moisture absorption of up to 0.6% for composite with 5 wt. % of filled content and even less with treated STLP. Moreover, reduced hydroxyl group intensity of absorption peak from FTIR of treated STLP corresponding to the untreated STLP results also confirms the capability of improving the wettability of filler and matrix adhesion. With analysis of all the results, it was found that the optimum filler content is 3% by weight of the treated STLP within the polymer matrix for reinforced properties. Despite these positive outcomes, the research acknowledges room for improvement, particularly in terms of chemical bonding between the filler and matrix. Consequently, recommendations for future enhancements include the utilization of coupling agents and chemical treatments. Moreover, the incorporation of an elastic material could improve the impact strength of the composite creating possibilities for more applications. Hence, this study concludes that this composite material not only achieves cost-effectiveness with enhanced properties through the consumption of an economical bio-filler and reducing polymer material but also aligns with circular economy principles in responsible consumption and production by recycling and reusing waste plastic.

Abbreviation

Symbol	Description
ASTM	American Society for Testing and Materials
EPS	Expanded Polystyrene
R-EPS	Recycled Expanded Polystyrene
STLP	Spent Tea Leaves Powder
TGA	Thermogravimetric Analysis
DTG	Derivative Thermogravimetric Analysis
FTIR	Fourier Transform Infrared Spectroscopy
SEM	Scanning Electron Microscopy

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this manuscript.

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