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Posted Date: 17 June 2025

doi: 10.20944/preprints202506.1470.v1

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Article

Exploitation of Plastic and Olive Solid Wastes for Accelerating the Biodegradation Process of Plastic

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Abstract: Recently, plastic and agricultural waste have gained attention as sustainable alternatives. Despite efforts to recycle these materials, much still ends up in landfills, raising environmental concerns. To optimize their potential, these wastes ought to be transformed into value-added products for diverse industrial applications. This work focused on producing thin composite material films using olive oil solid waste called JEFT and recycled plastic bottles. JEFT was cleaned, dried, and processed mechanically via ball milling to produce nano- and micron-sized particles. Composite films were produced via melt compounding and compression molding with rapid cooling process for controlled crystallinity and enhanced flexibility. Their density, water absorption, tensile strength, thermal stability, water permeability, functional groups, and biodegradation were comprehensively analyzed. Results indicated that 50% JEFT in recycled plastic accelerated thermal deterioration (42.7%) and biodegradation (13.4% over 60 days), highlighting JEFT's role in decomposition. Peak tensile strength (≈ 32 MPa) occurred at 5% JEFT, decreasing at higher concentrations due to agglomeration. Water absorption and permeability slightly increased with JEFT content, with only a 1% rise in water permeability for 50% JEFT composites after 60 days. JEFT maintained the recycled plastic's surface chemistry, ensuring stability. The findings of this study suggest that EFT/r-HDPE films show potential as greenhouse coverings, enhancing crop production and water efficiency while improving plastic biodegradation, offering a sustainable waste management solution.

Keywords: recycled plastic; biodegradation; waste management; olive pomace waste; bio-composite materials; thermal stability; biodegradable plastic; packaging materials

1. Introduction

The olive oil industry has experienced significant growth over the past two decades due to rising demands and favorable market conditions [1]. The process of oil extraction, i.e., separating the oil from olives, releases significant greenhouse gases (GHGs) and produces considerable amounts of solid and liquid waste by-products. Commonly referred to as olive pomace, olive cake, olive oil residue, or olive husk, this solid waste comprises seeds and residual pulp and accounts for up to 30-40% of the output from this process [2,3]. The annual global production of olive pomace is approximately 1,300 million tons and is projected to reach 2,200 million tons by 2025 [4]. This waste consists of a lignocellulosic matrix ($\approx 31\%$ cellulose, $\approx 23\%$ hemicelluloses, and $\approx 26\%$ lignin), along with phenolic compounds, acids, and oily residues. Consequently, olive oil waste can be viewed as an economic asset that can be transformed into valuable products, offering sustainable solutions to the persistent problem of waste disposal [5].

Multiple studies have explored the utilization of olive oil waste or olive pomace, derived from continuous or discontinuous extraction processes, in diverse applications such as combustion, biorefinery, livestock feeding, and soil amendments [6,7]. For instance, Najafi et al. [8] explored the potential of olive waste in a multi-product biorefinery, employing various pretreatment methods on different olive waste components for bioconversion into biogas, bioethanol, and lignin. The study demonstrated the energy recovery potential from olive waste by observing increases in methane and bioethanol yields from specific pretreatments and residual oil extraction for biodiesel production.

Ameixa et al. [9] explored the potential of black soldier flies to bio-convert olive pomace into animal protein. The results indicated that higher levels of olive pomace in diets extended larval development times and impacted feed conversion rates, yet improved the fatty acid profile of the larvae, making them richer in monounsaturated fats. These findings suggest that black soldier flies can effectively convert olive pomace into a sustainable feed alternative, enhancing protein content and altering fatty acid compositions beneficially. Moreover, Alaoui et al. [10] reviewed the environmental impact of olive oil production, emphasizing the potential of olive pomace as a sustainable soil amendment. Olive pomace was found to improve soil fertility, structure, and moisture capacity, particularly in Mediterranean regions with poor soil conditions. Similarly, Ameziane et al. [11] investigated the effects of using various concentrations of olive pomace on the growth of fava beans and lentils in Morocco. The results showed that olive pomace, particularly at 10, 15, and 25% concentrations, significantly improved seed germination and various growth parameters for both crops, such as leaf number, stem size, leaf area, dry matter weight, seedling vigor, and germination rate compared to control soil. However, limitations were encountered due to the high moisture and chlorine content of the waste. Therefore, managing these large quantities of residues presents substantial challenges due to their high phytotoxicity, potential as pollutants, and the costs associated with proper disposal treatments [12,13]. For example, a substantial amount of olive oil waste is burned in fields post-harvest to clear land for subsequent crops, a practice necessitated by labor constraints, yet resulting in nutrient loss and increased carbon dioxide (CO₂) emissions [14,15].

The olive industry has started expanding into new climatic regions outside the Mediterranean basin, which is driven by growing demand for extra virgin olive oil [16]. This is exemplified by the Kingdom of Saudi Arabia (KSA), where olive cultivation has escalated to over 5 million trees, producing 80,000 tons annually in Al-Jouf and Tabuk regions [17]. This surge in production leads to considerable agricultural waste, posing environmental challenges. However, a study by Khdaif et al. [18] suggested that while olive oil wastes do not affect soil properties, there is a critical need for regulations and new technologies to manage and utilize these wastes effectively in industrial applications.

In parallel with olive oil waste, plastic waste has become a significant environmental challenge in the KSA, where large amounts of plastic are produced and discarded annually. Since the introduction of plastics, these materials have played a crucial role in enhancing modern lifestyles due to their versatility and durability. However, their widespread use has led to an alarming accumulation of non-biodegradable waste in landfills. A study by Islam et al. [19] estimated that KSA generates over 800,000 tons of plastic waste from water bottles annually, with approximately 500,000 tons ending up in landfills, contributing to 2.6 million tons of CO₂ emissions per year. To mitigate this issue, KSA has implemented regulatory measures banning non-biodegradable plastics, particularly in packaging materials, as such composites usually demonstrate a strong antioxidant potential [20,21]. In response, several plastic manufacturers have begun developing biodegradable and cost-effective plastic alternatives to reduce environmental pollution and shorten the lifespan of plastic waste in its final disposal stage [22–24]. Among the most common plastic waste materials are polyethylene terephthalate (PET) bottles and polyethylene in different grade i.e., high and low density, a major post-consumer waste stream with high potential for mechanical recycling. PET recycling has evolved to a high standard, enabling the reuse of post-consumer materials in various applications. When collected properly, PET bottles can be recycled in a closed-loop system, reintroduced into new bottles (bottle-to-bottle), or diverted into alternative applications, such as textile fibers (bottle-to-fiber) and thermoformed sheet products (bottle-to-sheet). Despite the availability of several recycling paths for PET bottles, certain bottle types, such as milk bottles, remain challenging to repurpose due to material separation complexities during the recycling process [25].

To address the growing concerns surrounding both olive oil and plastic waste, researchers have explored innovative solutions that integrate these materials into sustainable applications. Past studies have investigated the incorporation of olive pomace waste into polymer composites, emphasizing fabrication techniques, characterization methods, and potential industrial applications. For instance, Hamida et al. [26] explored the incorporation of olive stone flour into polystyrene composites at different weight ratios (0–30%). The findings indicated that as the filler content increased, the hardness of the composites rose, while both tensile strength and elongation at break diminished, primarily attributed to inadequate filler dispersion and interfacial bonding. Similarly, olive stone flour was tested as a filler in polyvinyl chloride and polypropylene composites [27,28]. Their findings revealed

that while the addition of olive stone improved stiffness and reduced water absorption, it also reduced tensile strength and impact resistance, indicating the need for coupling agents to enhance adhesion. From a broad perspective, the primary challenge faced when integrating natural lignocellulose materials into polymers is interfacial adhesion. Hejna et al. [29] compared olive stone, date seed, and wheat bran as bio-composite fillers in poly(ϵ -caprolactone) matrices, showing that olive stone-reinforced composites had the highest hardness and improved thermal stability compared to the other fillers. Additionally, previous research has examined the chemical composition of olive pomace waste and its economic feasibility for waste management [30,31].

A review by Valvez et al. [32] highlighted the potential of olive stone residues as fillers in polymeric composites, improving stiffness but limiting strength; however, further efforts are required to fully understand their mechanical behavior, particularly in terms of fracture and fatigue properties. Given the pressing need for cost-effective and sustainable solutions using locally available waste resources, this study focuses on the development of novel polymeric composite films as potential greenhouse covering and packaging materials. These composite films, derived from olive oil solid waste and bottle wastes, aim to enhance biodegradation under KSA's atmospheric conditions. The developed composite materials were subjected to a comprehensive characterization process, evaluating their physical, chemical, thermal, and mechanical properties, including density, water absorption, tensile strength, thermal degradation, water permeability, surface chemistry, and biodegradation performance. The obtained findings are expected to provide valuable insights for decision-makers, supporting the implementation of sustainable waste management strategies and promoting the adoption of bio-based composite materials in industrial applications.

2. Materials and Methods

2.1. Substrate Sampling

The composite materials were prepared using milk bottles as the recycled polymer matrix, as shown in Figure 1a. These bottles are made from high-density polyethylene (HDPE) (according to recycling symbols), hereinafter abbreviated as r-HDPE. The olive oil waste was sourced from a local olive oil processing plant in the Al-Jouf region in 2021 and used as the reinforcing agent, as shown in Figure 1b. Prior to use, the olive oil waste underwent purification, which involved soaking in distilled water overnight to remove excess oil and dirt. In this study, the purified olive oil waste is referred to as JEFT. There is no coupling agent used in this work.



Figure 1. Material used in this study: a) milk bottle and b) as-received JEFT residue.

2.2. Preparation of Composite Films

To prepare the composite films, the JEFT samples were first soaked, washed, and then dried at 70°C for 24 hours until a stable weight was achieved. The dried waste was then subjected to mechanical treatment using a ball-milling machine to obtain finer particles. The treated samples were stored at 25°C for further utilization, including fabrication and characterization. Figure 2 presents images of JEFT residue with different particle sizes after mechanical treatment. Before any processing or characterization, both JEFT and r-HDPE were dried again in a vacuum oven at 70°C for 24 hours to ensure moisture removal.

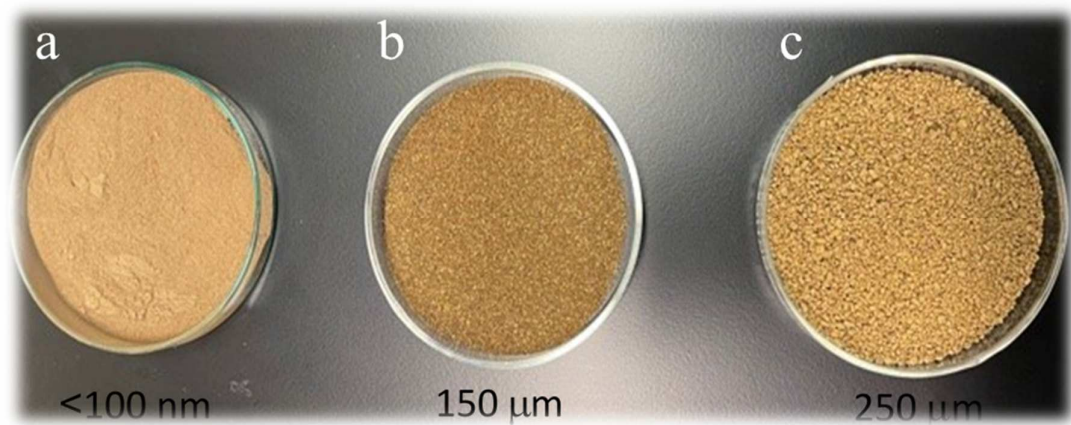


Figure 2. Images for different particle sizes of JEFT filler after the treatment.

To fabricate the composite materials, the JEFT and r-HDPE components were manually pre-mixed to ensure uniform JEFT particle distribution within the recycled plastic before compounding. The melt compounding technique was then employed to develop JEFT/r-HDPE composite samples with various loading ratios: 0/100, 5/95, 10/90, 15/85, 20/80, 25/75, and 50/50 (JEFT/r-HDPE). The compounding process was conducted using a Minilab co-rotating twin-screw extruder, operating with a screw speed of 100 rpm at 180°C over 10 minutes. After extrusion, the composite materials were cooled in an ice-water bath and then dried before undergoing molding. The extruded composites were then processed into films (~0.1 mm thick) using compression molding at 190°C under a pressure of 50 kN for 5 minutes, followed by an additional 2-minute compression at the same temperature and pressure. This was followed by rapid cooling at a rate of ~20°C/min and subsequent drying in a vacuum oven at 40°C for 24 hours. Figure 3 presents different JEFT/r-HDPE composite film samples developed in this study. The prepared films were then stored for characterization and analytical testing.

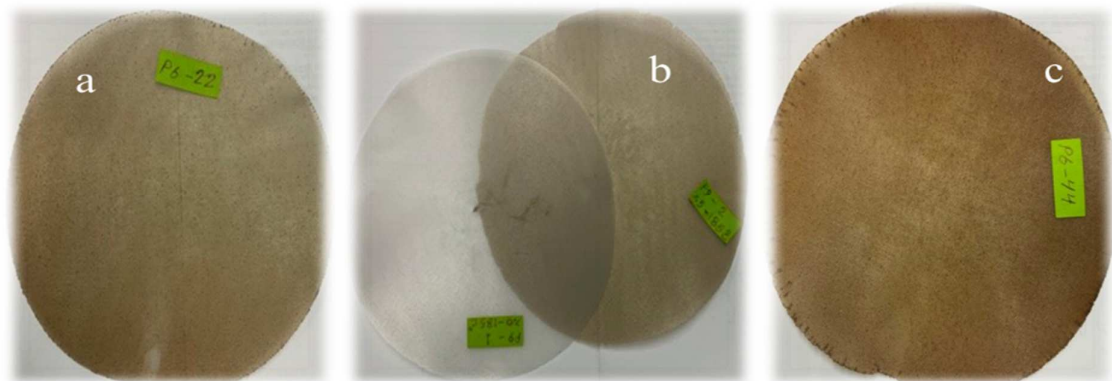


Figure 3. Different JEFT/r-HDPE composite samples.

2.3. Analytical Testing

2.3.1. Density

The bulk density of the composite material films was determined according to ASTM D792 -13 by using Archimedes' principle. About 1.0 g of each composite specimen was cut and immersed into the cylinder containing distilled water. Afterward, displacement of water volume was observed, and the density of the JEFT/r-HDPE composites was calculated where ρ is the density (g/cm^3), m is the mass (g), and v is the volume (cm^3).

$$\rho = \frac{m}{v} \quad (1)$$

2.3.2. Water Absorption

The water absorption of the JEFT/r-HDPE composites was analyzed as per ASTM D570-98. Prior to testing, the samples were oven-dried at 50°C to remove any residual moisture. The dried samples were then immersed in distilled water at 25°C for 24 hours and subsequently removed at regular time intervals. After removal, the samples were wiped with tissue paper to remove excess surface water and weighed using a high-precision scale every 24 hours for a period of two weeks. The recorded weight changes were used to calculate the percentage of water absorption, allowing for the assessment of the composites' moisture uptake behavior over time using the following formula:

$$W_b = \left[\frac{M_0 - M_t}{M_t} \right] \times 100\% \quad (2)$$

where W_b is the water absorption (%), M_0 is the initial dried mass (g), and M_t is the final wet mass (g).

2.3.3. Tensile Test

The tensile properties of the JEFT/r-HDPE composites were evaluated according to the ASTM D638-19 standard using a universal testing machine (Instron 5544, USA) equipped with a 5 kN load cell. Dog bone-shaped specimens of each composite formulation were tested by keeping the cross-head speed of about 1 mm/min at room temperature while keeping the humidity at $50 \pm 5\%$ for 24h. The tensile stress at yield, tensile strength, and elongation at break were measured and analyzed to assess the mechanical performance of the developed composites.

2.3.4. Thermal Characterization

The thermal behavior of the composites was analyzed using a thermogravimetric analyzer (TGA 1, PerkinElmer, USA). Each sample was heated gradually from 25°C to 800°C at a heating rate of 10°C/min. The thermal decomposition temperatures and weight loss were determined from the weight-temperature curves to assess the thermal stability of the composites. The thermogravimetric test was conducted under a nitrogen atmosphere to prevent oxidative degradation during heating.

2.3.5. Permeability Test

The permeability of the composite films was evaluated following the ASTM E96-95 standard under ambient chamber conditions, with a relative humidity of $35\% \pm 5\%$ and room temperature. Three circular specimens ($\sim 30 \text{ cm}^2$ in area) were cut from each sample. Each specimen was placed over a cup containing 40 mL of distilled water, sealed using silicone rings, and then enclosed in hermetically sealed jars to maintain controlled test conditions (Figure 4). The cups were periodically weighed using an analytical balance, and the water permeability values were calculated using regression analysis of the weight loss versus time data. The weight loss was determined by subtracting the loss through the sealing from the total weight loss. Three replicates of each film were tested to ensure reliability.

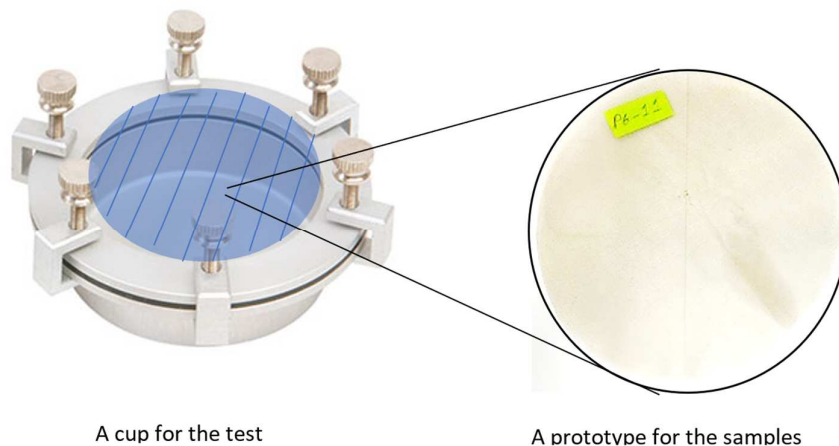


Figure 4. Permeability cup photo and prototype of the investigated samples.

2.3.6. Functional Group Characterization

Fourier-transform infrared (FTIR) spectroscopic measurements were conducted using a Nicolet 5700 FTIR spectrometer (Thermo Electron Corporation). The samples were prepared by mixing with dried potassium bromide (KBr) (Fisons Scientific) to form KBr pellets for analysis. Prior to mixing, the samples were dried at 120°C overnight to remove any residual moisture. The dried samples were then ground and mixed with KBr (1 mg sample with 220 mg KBr) for several minutes until a fine powder was obtained. The FTIR spectra were recorded over the 400–4000 cm⁻¹ wave number range to identify functional groups present on the film surfaces. This analysis was performed to assess the modifications in JEFT after acid treatment.

2.3.7. Biodegradation Investigation

The soil burial test is a widely used method for assessing the biodegradability of polymeric materials [33,34]. In this study, the test was conducted in a flower container filled with farmland sand, sourced from a private garden in Riyadh, where the samples were exposed to natural weather conditions, including temperature variations, sunlight, humidity, and wind. Rectangular samples (20 × 40 × 0.1 mm³) were cut, washed with distilled water, and then dried overnight at 25°C before testing. The dried samples were weighed and buried at a depth of ~10 cm in the soil for 60 days, starting from April 1, 2021. The garden was watered daily in the morning to simulate real environmental conditions. The biodegradability of the samples was assessed by calculating the percentage weight loss over the burial period, using the following formula:

$$W_L = \left[\frac{W_0 - W_t}{W_t} \right] \times 100\% \quad (3)$$

where W_L is the weight loss (%), W_0 is the initial weight (g), and W_t is the final weight (g).

2.3.8. Scanning Electron Microscopy (SEM)

Scanning Electron Microscopy (SEM) is an extensively utilized imaging method that delivers high-resolution surface morphology through the scanning of samples with a focused electron beam. This study necessitates a thorough analysis of microstructures and surface features. In this study, the fractured surface morphology of the samples was examined using a scanning electron microscope model (SEM, *Jeol JSM-6010PLUS/LV, Peabody, USA*) operated at an acceleration voltage of 15 kV. Before imaging, the composite samples were coated with a thin layer of gold to enhance conductivity. A small amount of JEFT was dispersed in ethanol, then sonicated for about 15 min and dropped on the conductive carbon tape attached to the sampling stub.

3. Results and Discussions

3.1. Density Measurements

Figure 5 illustrates the density values of composite material films, highlighting the influence of JEFT filler concentration on the r-HDPE matrix. Generally, the density of the composites does not exhibit a significant increase as the JEFT content rises from 5 to 50 wt.%. However, a gradual decrease in density is observed between 10 and 50 wt.% JEFT content. This trend can be attributed to the higher JEFT content, which increases the bulkiness of the composite material. The increased bulkiness expands the overall volume, ultimately leading to a reduction in composite density. To the best of the author's knowledge, no prior studies have specifically measured the density of JEFT-reinforced polymer composites. However, Banat [35] conducted a review listing the measured properties of olive pomace-reinforced polymer composites, providing a relevant reference for comparative analysis.

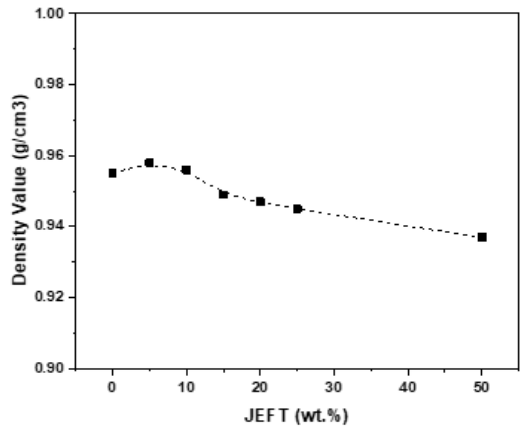


Figure 5. Density values of the JEFT/r-HDPE composites.

3.2. Moisture Content behaviors

Figure 6 illustrates the water uptake behavior of the composite films as a function of exposure time under various immersion conditions and JEFT loadings. The results indicated that water absorption increases with higher JEFT filler content, particularly after 24 hours of immersion. During the initial stages of absorption, the water content increased gradually. It is expected that with extended immersion time, a saturation plateau will be reached at JEFT loadings exceeding 50 wt.%. This trend is well-documented in natural fiber-reinforced polymer composites, where the hydrophilic nature of natural fillers contributes to increased water absorption. These observations agree with previous published studies that reported similar behavior [36,37].

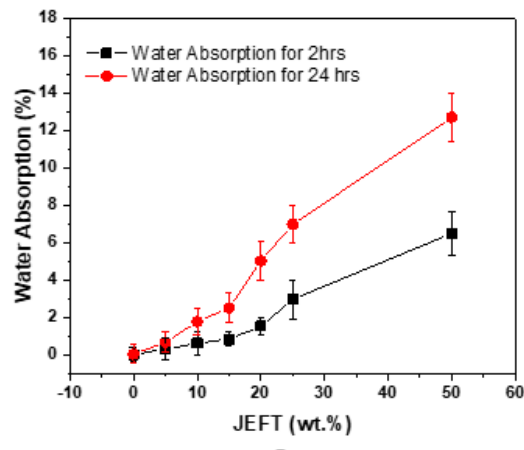


Figure 6. Water Absorption of the JEFT/r-HDPE composites with different amounts of JEFT.

3.3. Tensile Properties

Figure 7 demonstrates the tensile strength of the composite material. The tensile strength of JEFT/r-HDPE composites exhibited an initial increase at 5 wt.% JEFT content, followed by a gradual decline with further JEFT loading (blue curve). This decrease in strength is likely attributed to weak interfacial bonding between the r-HDPE matrix and JEFT filler, which becomes more pronounced as JEFT content increases. Additionally, a higher JEFT concentration may lead to increased void formation and poor dispersion, further contributing to the reduction in overall composite strength. Similar trends have been reported for olive pomace-reinforced polypropylene composites [30], as well as in other studies on polymer composites containing olive oil solid waste [31,35,37].

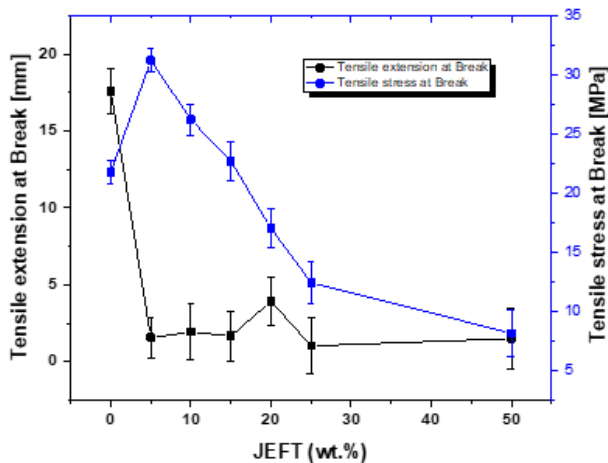
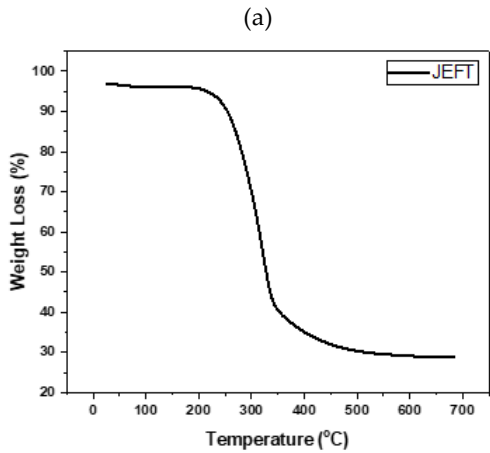


Figure 7. Tensile Properties of the JEFT/r-HDPE composites.

Similarly, the elongation properties of r-HDPE/JEFT composites tend to decrease as JEFT content increases (black curve). This decline is likely due to the presence of JEFT within the r-HDPE matrix, which disrupts the bonding and intermolecular structure of the polymer. As a result, the composite exhibits reduced flexibility, impairing its ability to stretch and return to its original form. Given this behavior, JEFT/r-HDPE composites may not be suitable for applications where elongation is a critical parameter. However, Sawalha et al. [38] reported enhancements in tensile properties when olive oil waste-reinforced recycled HDPE was subjected to carbonization at different temperatures. Their findings suggested that pre-treating JEFT could improve adhesion between JEFT and r-HDPE, leading to better dispersion and reduced agglomeration effects, ultimately enhancing the mechanical properties of the composite.

3.4. Thermal Properties

To reduce the effects of oxidative degradation, TGA scans were conducted under a nitrogen atmosphere to examine the thermal degradation behavior of pure JEFT and JEFT/r-HDPE composites. Natural fillers are highly sensitive to thermal degradation [39]; therefore, evaluating the thermal stability of the newly developed JEFT-based r-HDPE composites is crucial, as thermal degradation can significantly alter the color and properties of the final product. The TGA results for JEFT filler and its composites under a nitrogen atmosphere are presented in Figure 8, respectively. JEFT filler remained thermally stable up to approximately 200°C, with a 5% weight loss, primarily attributed to moisture evaporation. In contrast, r-HDPE exhibited greater thermal stability, undergoing thermal degradation at around 450°C, with less than 5% weight loss due to moisture release [4]. These findings highlight the significant difference in thermal behavior between JEFT and r-HDPE, reinforcing the importance of optimizing the composite’s thermal stability for potential industrial applications.



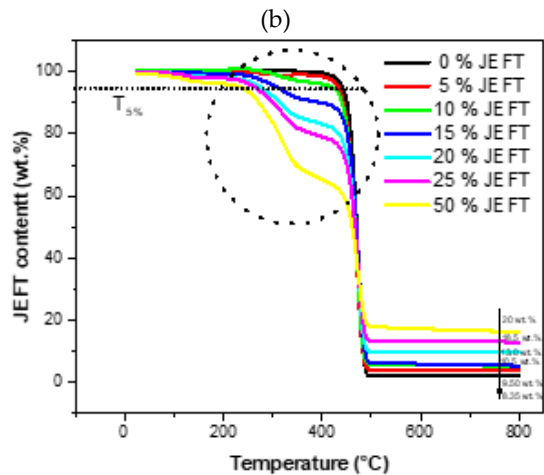


Figure 8. Thermogravimetric analysis curve of the a) JEFT filler and b) JEFT/r-HDPE composites.

The TGA results indicate that all composite samples exhibited minimal weight loss up to around 300°C. Beyond this temperature, a significant increase in weight loss occurs over a narrow temperature range, as reflected in the steep slopes of the TGA curves. However, the onset temperature for thermal degradation varies among the samples, as shown in Figure 8b.

A common method for evaluating thermal stability involves determining the temperature at 5% weight loss ($T_5\%$) and the thermal decomposition temperature. The $T_5\%$ values of the examined composites are presented in Table 1. The findings indicate that thermal stability decreases with increasing JEFT content in the r-HDPE matrix, likely due to the lower thermal resistance of JEFT compared to r-HDPE.

Table 1. Thermal degradation temperatures ($T_5\%$) for the JEFT/r-HDPE composites.

JEFT wt. %	$T_5\%$ (°C)
0	480.3 ±1.5
5	475.8 ±0.9
10	466.5 ±0.5
15	330.7 ±2.0
20	305.1 ±1.4
25	300.5±2.2
50	275.0±3.5

Generally, the thermal degradation of the JEFT/r-HDPE composites occurs in three distinct stages. The first stage, which takes place between 25 and 200°C, corresponds to the evaporation of moisture, a characteristic commonly observed in natural fiber-reinforced polymer composites. This stage is associated with minimal weight loss, primarily due to the release of absorbed water within the composite structure. The second stage, occurring between 200 and 350°C, is shorter and marks the degradation of the polymer matrix chains and hemicellulose, which are among the most thermally sensitive components in natural fillers. A maximum weight loss of approximately 30% was observed in composites containing 50 wt.% JEFT, indicating that hemicellulose degradation plays a significant role in the structural breakdown of the material. The third stage, which occurs between 350 and 500°C, is attributed to carbonization within the polymer and the thermal decomposition of lignin and cellulose. At this point, weight loss continues as the polymer matrix further degrades. Beyond 500°C, all composite samples exhibit no significant weight loss up to 800°C, indicating that the remaining material consists primarily of char residue (carbonaceous residue) produced during the earlier decomposition phases. As observed in Figure 8b, the amount of char residue increases with higher JEFT content, suggesting that JEFT incorporation contributes to greater thermal stability at elevated temperatures.

The thermal degradation behavior observed in this study aligns with previous findings. Banat et al. [40] and Kaya et al. [30] have reported similar three-step degradation processes in olive oil

pomace-reinforced HDPE and polypropylene composites, respectively. These results confirm that natural fiber-based polymer composites exhibit a multi-stage degradation process, influenced by the presence of moisture, hemicellulose decomposition, and final carbonization. Understanding these thermal degradation characteristics is crucial for optimizing the processing and application of JEFT/r-HDPE composites, particularly in high-temperature environments where thermal stability is a key performance parameter.

3.5. Permeability Results

The water permeability of polymer composite materials is a critical factor in various applications, including packaging, membrane technology, and protective coatings or sheets. The water permeation process in these materials is inherently complex, influenced by multiple factors such as the polarity of water molecules, their interaction with polymer chains, hydrogen bonding capacity, and the resulting effects on polymer swelling and chain mobility. To predict water permeation behavior, several theoretical models have been developed, incorporating parameters such as tortuosity, geometry, crystallinity, temperature, filler particle orientation, weight/volume fraction, and polymer chain confinement. Understanding water permeability is particularly important in this study, as the long-term objective is to develop composite films for greenhouse coverings and packaging applications, where the ability to control permeability is essential for functional performance in diverse environmental conditions [41].

Figure 9 shows the water permeability results for the composites. The water permeability of the investigated composite films was found to be very low, as expected for pure recycled bottle-based materials, but showed a slight increase with increasing JEFT loading. For instance, films containing 50 wt.% JEFT exhibited only a ~1% increase in permeability, indicating that water transport primarily occurs along the surface of JEFT particles.

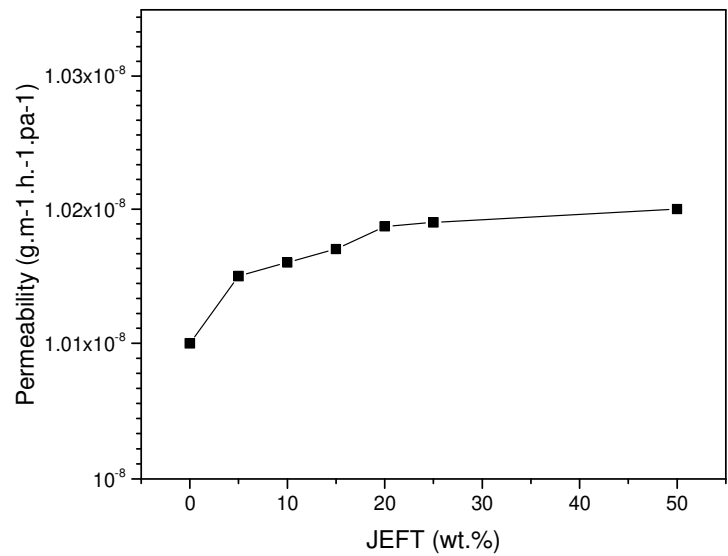


Figure 9. Water permeability of the JEFT/r-HDPE composites after 60 days.

Gårdebjer et al. [33] reported that the presence of hemicellulose contributes to higher permeability in composite films, which could explain the observed trend. Several approaches have been explored to enhance the permeability of polymer composites, such as reducing the filler volume fraction, modifying the filler, or developing hybrid composites [34]. For example, Jin et al. [42] suggested that incorporating graphene nanomaterials can improve the permeability of polymer nanocomposites due to their high aspect ratio and large surface area, which enhance reinforcement effects. Additionally, improving adhesion properties and filler dispersion between JEFT and r-HDPE could enhance the water barrier properties of the composite films. JEFT agglomeration may act as a stress

concentrator, leading to the formation of microcracks, which could eventually merge and cause fractures or failures in the JEFT/r-HDPE composites, as observed in the tensile properties (Figure 7).

3.6. Functional Group Analysis

Compared to the nearly featureless spectrum observed for all JEFT/r-HDPE composites, Figure 10 reveals three strong absorption peaks around 3000 cm^{-1} , 1500 cm^{-1} , and 750 cm^{-1} , corresponding to the hydroxyl (-OH), carbonyl (C=O in carboxyl, -COOH), and C-H functional groups, respectively. This indicates that all composite samples exhibit similar functional groups, suggesting that JEFT does not significantly alter the surface chemistry of the r-HDPE matrix. This characteristic is beneficial for applications such as greenhouse covering materials, as it ensures chemical stability with no undesired changes to the surface properties of the films.

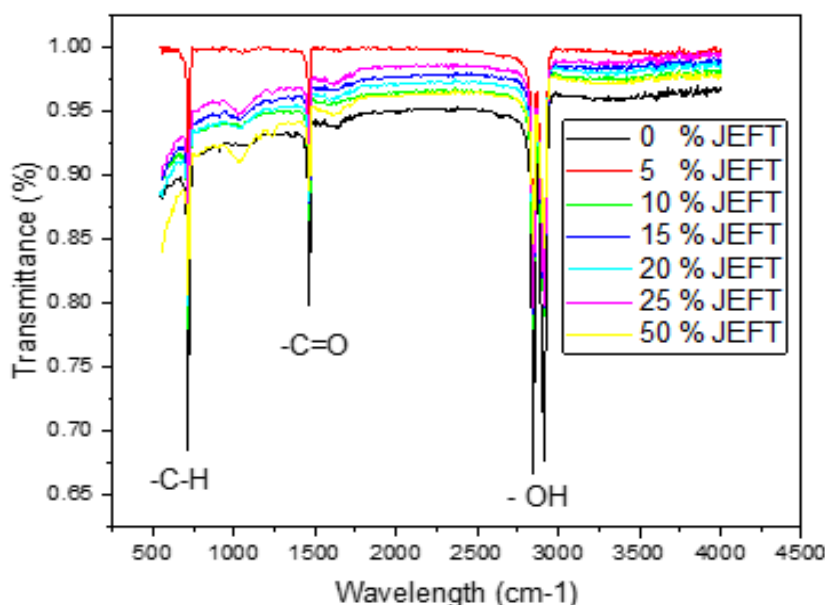


Figure 10. Functional Groups Spectra of the JEFT/r-HDPE composites.

A similar presence of functional groups has been reported in comparable polymer composites and various agricultural waste-based materials [43]. Additionally, previous studies have highlighted that the key constituents of JEFT offer excellent potential for natural ultraviolet (UV) protection in composite films [44,45]. This reinforces the suitability of JEFT-based materials for outdoor applications, particularly where UV resistance and environmental stability are critical factors.

3.7. Biodegradation Analysis

The weight loss of buried composite films serves as a key indicator of the biodegradation process of the investigated samples. To better understand the biodegradation mechanism and the factors influencing this process, biodegradation can be defined as the natural breakdown of a material into its basic components through microbial activity and environmental interactions [46,47]. Table 2 presents the weight loss of JEFT/r-HDPE composite samples as a function of burial days. The weight loss percentage of the composite films after 60 days of soil burial varied among the samples. The r-HDPE exhibited significantly lower weight loss compared to the JEFT/r-HDPE composites, indicating that JEFT enhances the biodegradability of the material. The highest recorded weight loss was observed in samples containing 50 wt.% JEFT, which degraded by 13.4% over 60 days, whereas pure r-HDPE exhibited only 0.1% degradation during the same period.

Table 2. Weight loss of JEFT/r-HDPE composite samples as a function of burial days.

JEFT wt. %	Initial weight (g)	Final weight (g)	Weight loss (%) in 60 days
0	1.096	1.094	0.1
5	1.172	1.161	0.9
10	1.154	1.140	1.2
15	1.093	1.078	1.4
20	1.066	1.043	2.1
25	1.089	1.011	7.0
50	1.190	1.031	13.4

The biodegradation process in soil burial conditions is a biochemical conversion, where micro-organisms break down complex polymer structures into simpler molecules through natural degradation pathways. A comprehensive review by Shah et al. [48] explored the mechanisms, influencing factors, and key parameters involved in the biodegradation of plastics, further emphasizing the role of organic fillers like JEFT in accelerating polymer degradation. These findings suggest that incorporating JEFT into r-HDPE enhances the material’s environmental sustainability, making it a promising alternative for biodegradable composite applications.

3.8. Morphology Observation

Figure 11 displays the SEM images of purified JEFT, r-HDPE, and the fractured surfaces of JEFT/r-HDPE composite film samples mixed with different amounts of JEFT filler. The figure revealed different fractographic features. The sample of neat r-HDPE had flat surface morphology with wrinkle-like and ductile behavior features with free air voids, as shown in Figure 11a. This was due to the asymmetrical interaction between polymeric chains. In general, a rough fracture surface indicates more ductility with the deformation of the matrix during tensile loading, whereas a smooth surface indicates more brittle fracture and lower fracture toughness. On the other hand, Figure 11b represents the morphology of the JEFT, which apparently showed irregular particle shapes. The assumption that spherical particle shapes can be obtained is unrealistic, as particles JEFT typically exhibit complex shapes. The interfacial tension between polymer and filler plays a crucial role in determining phase morphology.

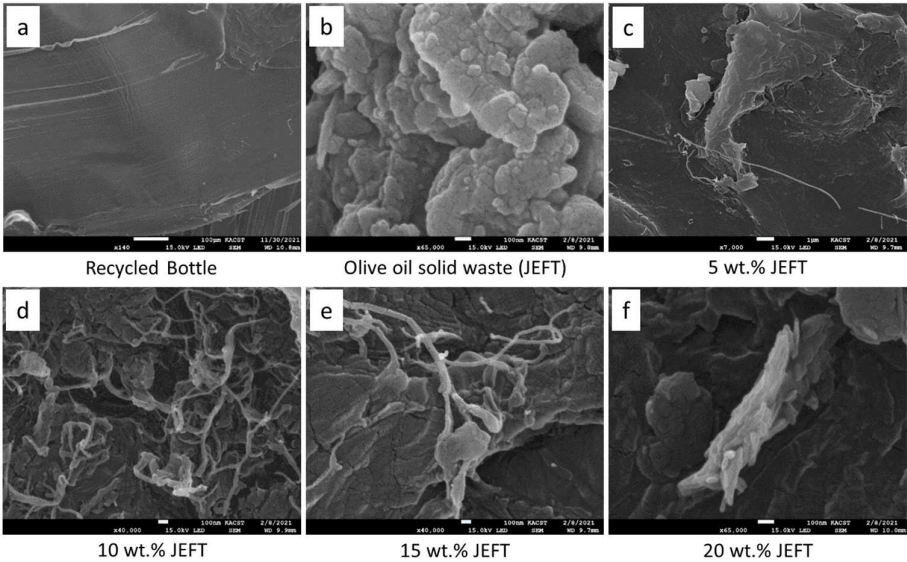


Figure 11. SEM images of pure epoxy composites, SEM images of a) r-HDPE, b) JEFT, c), d, e, and f) JEFT/r-HDPE composites with different amounts of JEFT, i.e., 5, 10, 15 and 20 wt.%, respectively.

The morphology of the filler at all JEFT loading levels (Figure 11(c,d,e,f)) shows slight variations compared to that of the neat r-HDPE polymer matrix. Low level pullout was noted at a low loading

of JEFT (5 wt.%), indicating a relatively strong tensile stress at break performance of the r-HDPE composite in comparison to pure r-HDPE. While, at medium and high JEFT loading (10, 15, and 20 wt.%), more JEFT pullout, debonding, and more agglomerations states were observed. This was probably due to poor adhesion between JEFT and r-HDPE polymer matrix. The presence of some agglomerations due to JEFT debonding is more pronounced in the r-HDPE composite containing 20 wt.% of JEFT amount. These morphological behaviors seem to agree with the results obtained in the tensile test in this study. These composites explain the lower mechanical strength and modulus with respect to unfilled r-HDPE samples. As shown in Figure 7, the tensile strength of the r-HDPE composite initially increases up to 5 wt.% followed by a decline due to aggregation and poor adhesion between the matrix and fillers at higher loading. Banat et al. [35] observed a similar agglomeration effect at 25 wt. % of olive oil waste in an HDPE matrix. They suggested that to enhance the compatibilization between olive waste filler and HDPE polymer matrix, ~ %5 coupling agent should be used for successive polymer composites for packaging application. Furthermore, it has already been established that unstable interfacial adhesion between natural filler and polymer matrix often results in fiber pull-out, fiber-fiber delamination, and matrix debonding Sawalha et al. [38]. Comparative arguments were presented by other researchers who utilized olive stone as a filler for recycled high-density polyethylene and found that adhesion between the HDPE matrix and the olive stone waste was improved by using compatibilizers [49].

4. Conclusions

This research presents an economical and sustainable solution for managing both agricultural and plastic waste in KSA by developing composite materials from olive oil solid waste (JEFT) and recycled plastic bottles (r-HDPE). The composite films were fabricated using a melt compounding process, followed by compression molding and rapid cooling, to produce thin and flexible films suitable for agricultural applications like greenhouse covering materials. The developed films were subjected to different types of characterization to investigate their properties and performance. Increasing JEFT content resulted in lower composite density, contributing to lightweight materials. However, water absorption and permeability increased slightly with higher JEFT concentrations. Additionally, thermal degradation and biodegradation rates were faster in JEFT-containing films compared to pure plastic films, indicating improved environmental sustainability. Functional group analysis further confirmed that JEFT did not alter the surface chemistry of the r-HDPE matrix, maintaining the integrity of the polymer structure. According to the literature and these observations, the results highlight the potential of JEFT/r-HDPE composites as a sustainable bio-based material with good biodegradability and UV resistance, making them suitable for general packaging and greenhouse covering applications aimed at improving water and energy efficiency. The developed partially green JEFT/r-HDPE polymer composite may serve as an alternative to traditional polymers in general packaging applications.

Future research should focus on pre-treating JEFT, adding coupling agents like the maleic anhydride grafted polyethylene, and optimizing the JEFT/r-HDPE ratio to enhance interfacial adhesion and improve the mechanical and functional properties of the final composite materials.

Acknowledgments: This work was financially supported by King Abdulaziz City for Science and Technology (KACST), Saudi Arabia.

Conflicts of Interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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