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ORIGINAL ARTICLES

# Fabrication, characterization and mechanical behaviour of *Tamarindus indica* fruit fibre-reinforced polymer composites

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## Abstract

In this work, a study was undertaken to explore the possibility of using leftover tamarind fruit fibres as reinforcement in PLA and HDPE matrix. PLA and HDPE polymers form minimum 75% of the total polymers used in the composites. PLA and HDPE was mixed with natural fibres (5 wt.%, 10 wt.%, 15 wt.%, 20 wt.% and 25 wt.%,) individually and also as a hybrid filler to enhance its mechanical properties. Characterizations, mechanical behaviour and microscopic investigation were performed to understand the excellent mechanical properties and good chemical resistance of the prepared composites, which demonstrated potential suitability for semi-structural applications.

**Keywords:** *polymer composites, Tamarindus indica fruit fibre, mechanical properties, sustainability, morphological analysis.*

**Data Availability:** Research data is available upon request from the corresponding author.

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## 1. Introduction

Growing global interest in the usage of environmentally friendly bio-waste materials-based polymer composites with the necessary mechanical properties that has been spurred by increased environmental consciousness[1]. Agro bio-waste fillers are a promising potential replacement for synthetic fibre polymer goods in environmental limits due to their wide availability and cost-effective processing[2]. Natural fillers in a polymer matrix can offer substantial benefits over typical fillers used in composites, and their use is growing globally due to the influence of low cost, low density, renewable, biodegradable, desirable qualities, and environmental friendliness. Nagarjun et al.[3] showed the mechanical characteristics of PLA composites with tamarind and date seed Micro fillers. The composites were made using the compression moulding method. The seed filler reinforcement greatly enhanced the tensile strength of the PLA matrix, according to the tensile data. Both tamarind and palm particle reinforcements nearly increased the flexural and impact strength of PLA matrix. Stalin et al. [4] experimented the usage of tamarind seed filler (TSF) as reinforcement in vinyl ester composites. The composite plates have been fabricated by compression moulding machine with TSFs of varying wt% from 5 to 50 as reinforcement material, and their properties such as tensile, flexural, impact, hardness, water absorption, heat deflection tests, and thermo gravimetric analysis are studied. On jute poly-lactic acid resin composite, Ramachandran et al.[5] performed different experiments such as Impact (IZOD and CHARPY tests), Differential Scanning Calorimeter test, Fourier Transform Infrared test, and Optical Imaging. The testing revealed that the results were comparable to synthetic composites such as polyester and epoxy. Mofokeng et al.[6] studied morphology, thermal and dynamic mechanical properties, and degradation patterns. SEM micrographs of the composites demonstrate more intimate contact and better interaction between the fibres and PLA than PP. The presence of hydrogen bonding interaction between PLA and the fibres was validated by Fourier-transform infrared (FTIR) spectroscopy data, which demonstrated the presence of enhanced interaction. The thermal stability of both polymers improved with increasing fibre content, with PP showing a more substantial improvement. Curcumin-loaded electro spun Poly (lactic acid) (PLA) composite membranes were created by Chen et al.[7]. Curcumin was loaded with varying concentrations of 1, 3, and 5 wt percent to investigate its anticoagulant properties as a drug-eluting stent. Fourier Transform Infrared (FTIR) spectroscopy was used to examine the structure of the composite membrane, and the results indicated that both

PLA and curcumin were present in the composite membrane without any chemical reaction. In the recycling of polyethylene terephthalate, Aldas et al.[8] discovered certain biopolymers that were regarded as pollutants (PET). The results revealed that PET-PLA and PET-PHB miscibility is good. However, due to the high processing temperatures employed in PET recycling, PHB is damaged; in the meantime, PET and TPS are poorly miscible, which is reflected in the microstructure. Sachin et al.[9] developed a composite that can be utilised as a substitute for standard plywood in the automotive, airline, and railway industries for furniture, building infrastructure, and interior components. PLA and NWF were used to create a new bio composite, which was then tested for mechanical properties. Natural fibres can be employed as reinforcement in polymers made from renewable raw materials, according to Oksman et al.[10]. Flax fibres and polylactic acid were used as the materials (PLA). PLA is a lactic acid-based thermoplastic polymer that has primarily been utilised in biodegradable items such as plastic bags and planting cups, but it can also be used as a matrix material in composites in theory. Maheswari et al.[11] experimented with tamarind fibres recovered by the water retting technique from ripened fruits. The hand lay-up technique was used to construct composite samples using these fibres as reinforcement and unsaturated polyester as matrix. Jo et al.[12] investigated ABS-based automotive console boxes with better environmental friendliness using composites made of acrylonitrile-butadiene-styrene copolymer (ABS) and poly(lactic acid). Nuthong et al.[13] found that adding fillers or reinforcements to PLA improves its impact characteristics. The brittleness of PLA polymer necessitates the modification for more practical usage. Alternative reinforcements in PLA composites included bamboo fibre, vetiver grass fibre, and coconut fibre. Untreated and flexible epoxy-treated composites with varying reinforcing amounts were injection moulded. With increasing fibre content, the impact strength of natural fibre reinforced PLA composites dropped[13,14]. Natural fibre composites, as investigated by Siregar et al.[15] in the field of materials, have piqued many people's interest due to their fundamental biodegradability property. As a result, unlike synthetic fibre, pineapple leaf fibre is not only biodegradable but also environmentally benign. Somashekhar et al.[16] investigated coconut shell powder, which has several advantages over other materials, including low cost, renewable, high specific strength to weight ratio, low density, low abrasion on machine, and environmental friendliness.

## 2. Materials and Methods

### *2.1 Matrix*

The virgin PLA pellets and HDPE pellets were purchased from the local merchant Augment 3Di, Coimbatore District, Tamil Nadu State, India and Kings Polymer, Coimbatore District, Tamil Nadu State, India. As mentioned by the seller, PLA has the melt flow index of 10-30 g/10 min with density of 1.24 g/cm3 at 190 °C. 65, 120 and 180 °C are the glass transition, crystallization and melting point temperatures, respectively[3]. Both PLA and HDPE pellets were kept in a dry air oven before processing to remove the moisture. During injection moulding, both PLA and HDPE were melted at their melting point temperature. PLA is a polyester manufactured from fermented corn, cassava, maize, sugarcane or sugar beet pulp. These renewable resources sugar is fermented and converted to lactic acid, which is subsequently converted to polylactic acid or PLA. HDPE is the most environmentally friendly of all plastics, emitting no toxic gases into the atmosphere.

### *2.2 Fibre*

The tamarind fruit fibre (TFF) of around 2.5 kg was collected from Salem District, Tamil Nadu State, India. The collected fibres were treated with water. After cleaning with water, the fibres were dried for 6 hrs in direct sunlight in order to remove the water content by means of evaporation. Moreover, the remaining moisture content is then removed by drying it in a hot air oven. The dry fibres were next processed into a powder form (25–60 m) in a local flour mill at 500 rpm for 1 hr. 450 gms of TFF powder were finally obtained after the grinding process.

### *2.3 Composite fabrication*

Injection moulding is used to create various combinations of specimens (S1 to S18), as displayed in Table 1. The moulding process is carried out at Perumal Injection Moulding Company in Coimbatore District, Tamil Nadu State, India. The detailed process flow diagram of fabrication and testing of composites is shown in Figure 1. The machines hopper is filled with PLA pellets, HDPE pellets and TFF powder, as and when needed. Followed by material loading, the machine gets heated-up, melts and mixes the materials. The obtained paste is then injected into the die and specimen plates were created accordingly. A rectangular die with the dimensions of 147 cm × 98 cm × 4 cm is used in this study. A total of 18 plates were made, out of which pure PLA = 1, pure HDPE = 1, PLA + HDPE = 1, TFF powder + PLA = 5,

**Table 1.** Combination of specimens and their corresponding weight ratios.

| Combinations | Specimen label | TFF (wt.%) | PLA (wt.%) | HDPE (wt.%) |
|---|---|---|---|---|
| Pure PLA | S1 | - | 100 | - |
| Pure HDPE | S2 | - | - | 100 |
| PLA + HDPE | S3 | - | 50 | 50 |
| TFF + PLA | S4 | 5 | 95 | - |
| | S5 | 10 | 90 | - |
| | S6 | 15 | 85 | - |
| | S7 | 20 | 80 | - |
| | S8 | 25 | 75 | - |
| TFF + HDPE | S9 | 5 | - | 95 |
| | S10 | 10 | - | 90 |
| | S11 | 15 | - | 85 |
| | S12 | 20 | - | 80 |
| | S13 | 25 | - | 75 |
| TFF + PLA + HDPE | S14 | 5 | 47.5 | 47.5 |
| | S15 | 10 | 45.0 | 45.0 |
| | S16 | 15 | 42.5 | 42.5 |
| | S17 | 20 | 40.0 | 40.0 |
| | S18 | 25 | 37.5 | 37.5 |

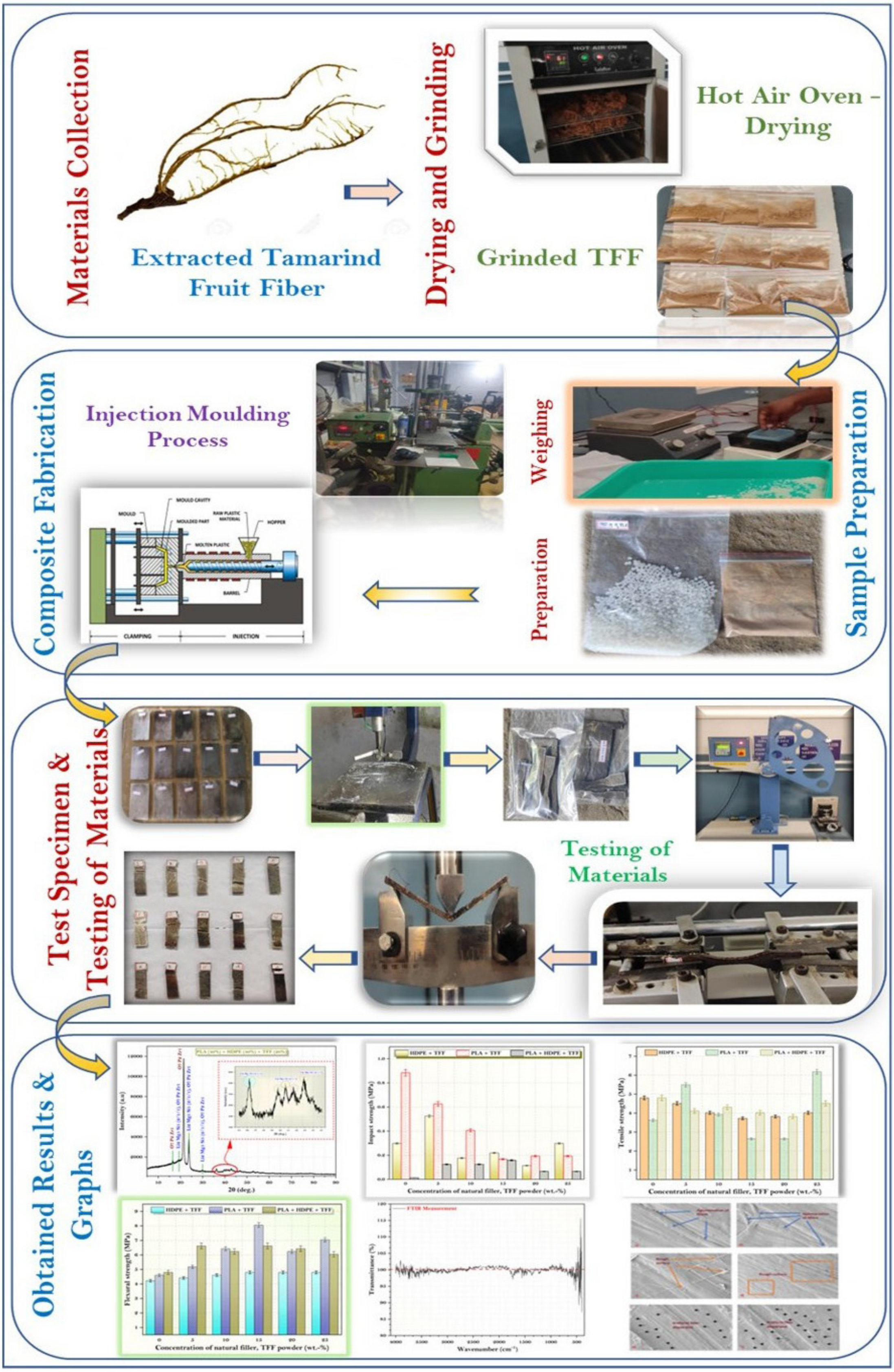


**Figure 1.** Overall process flowchart and obtained results.

TFF + HDPE = 5, TFF + PLA + HDPE = 5. The different weight ratios for all the specimens are mentioned in Table 1.

### 2.4 Characterization techniques

According to ASTM D 3039, tensile tests were performed on samples with dimensions 115 mm × 19 mm × 4 mm. The universal testing machine (UTM) with a load cell capacity of 100 kN was used for all the tests. During testing, the gauge length was set to 25 mm and the cross-head speed was kept at 5 mm/min. The displacement across the cross-head was measured using an external LVDT device. Five samples of each composition were examined and the average value was reported as the corresponding specimen's tensile property.

The flexural characteristics of the test specimens were assessed using a three-point bending test. The test was carried out on samples with dimensions 115 mm × 12.7 mm × 4 mm, as specified by ASTM D 790. The support span was adjusted to 50 mm and the test was carried-out at a cross-head speed of 5 mm/min. Five samples were tested and the average flexural value was reported in all the cases.

The Charpy impact test was carried-out to determine the impact energy. The load is applied via an impact strike from a hefty pendulum hammer discharged from a fixed height location. The test material or specimen is placed at the bottom. The pendulum impacts the test piece and fractures it at the notch when it is released. The pendulum continues to swing lower than its original height. Simple calculations can be used to calculate the energy absorbed at the fracture. The technique can be used on both short and long fibre composites. The Charpy test is a defined method for determining how much energy a test material absorbs during a break. The test specimen is 4 mm thick as per ASTM D 4812. It is commonly used in industry since the results are inexpensive and rapid. The average value was calculated using five sample specimens for the Charpy test in all the conditions.

FESEM is a sophisticated microscope that provides higher magnification and the ability to view very tiny features at a lower voltage than conventional SEM. An instrument named CARL ZEISS (USA), model: sigma with Gemini column was used to observe the interaction between the TFF powder and the matrix (PLA and HDPE) with an accelerated voltage of 10 kV. The composite samples were coated with gold before the examination in order to avoid charging during the experiments. The sample portions were magnified as much as 3500 times during the examination.

FTIR testing is an analytical testing procedure that uses infrared radiation to identify organic and some inorganic chemicals (IR). It also identifies unfamiliar solid, liquid, or gaseous components. It mainly determines the presence of surface contamination on a material and, in some situations, quantify it. It is carried out in order to identify the chemical functional groups in the composite.

XRD analysis is used to detect the crystalline phases contained in a material and so reveal chemical composition information by studying the crystal structure. The phases are identified by comparing the obtained data to that in reference databases.

## 3. Results and Discussions

### 3.1 Tensile behaviour

The addition of fillers (TFF powder) to the matrix has been demonstrated to improve the corresponding tensile characteristics (Figure 2). The tensile strength of the neat PLA matrix is improved by 2.55 MPa or 70.26 percent, when TFF powder was added. When the TFF powder concentration was 25 wt.-% with 75 wt.-% of PLA matrix polymer, the highest tensile property is achieved. Crack initiation and propagation at the inter-laminar area determines the strength of composites made with fillers in general. The addition of fillers reinforces the matrix material in this location, preventing crack start and allowing for excellent tensile strengths. The aggregation of fillers may have caused the tensile property to deteriorate. Overall, strong interfacial bonding has allowed stresses to flow from the matrix to the fibre, which increases the strength and stiffness.

### 3.2 Impact strength

PLA was discovered to have higher impact strength than composites containing TFF powder fillers (Figure 3). This could be explained by PLA's brittle nature at room temperature. The reduced impact strength was owing to

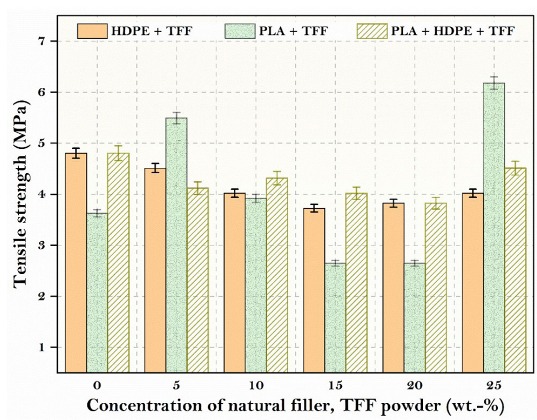


**Figure 2.** Tensile behaviour of TFF reinforced PLA/HDPE-based composites.

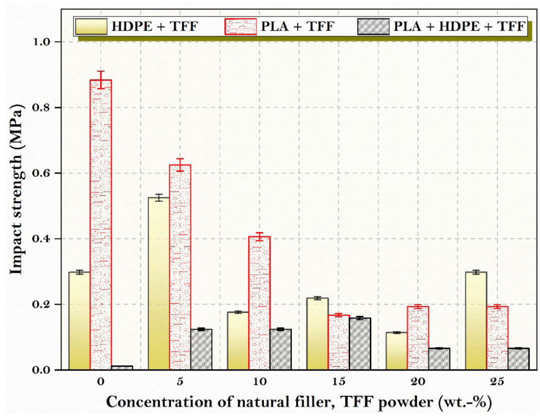


**Figure 3.** Impact behaviours of TFF reinforced PLA/HDPE-based composites.

inadequate interfacial adhesion between the filler and the matrix, as well as the presence of larger voids, which caused specimens to break prematurely. The composites used in this investigation have 15 wt.-% TFF fillers and a PLA matrix. The impact strength of polymer was around 81 percent lower than that of plain PLA. The impact strength of HDPE + PLA composites was found to be lower than composites added with TFF, which could be due to HDPE + PLA's ductile behaviour at room temperature. The composites with 15 wt.-% TFF fillers and an HDPE + PLA matrix polymer exhibit 31 percent better impact strength than the pure HDPE + PLA. Composites with 5 wt.-% TFF fillers and an HDPE matrix polymer displays 76 percent higher impact strength than pure HDPE.

### *3.3 Flexural behaviour*

Figure 4 shows the influence on the addition of micro fillers to the clean matrix which results in improved flexural characteristics. Flexural strength of HDPE is 4.22 MPa, PLA is 4.61 MPa and PLA + HDPE is 4.8 MPa. TFF + PLA, TFF + HDPE and TFF + PLA + HDPE composites all about doubled the flexural strength of its corresponding neat matrix. The flexural strength of PLA with TFF filler is enhanced by 13.64 percent or 4.8 MPa. The PLA composite with TFF filler has a higher flexural property due to its good size, which fills the spaces in the composites and effectively resists force. With 15 wt.-% TFF filler concentration and 85 wt.-% PLA matrix polymer, the greatest flexural strength of 8.06 MPa is achieved, which is 75 percent higher than pure PLA. The increase in flexural strength illustrates the superior matrix-filler absorption property.

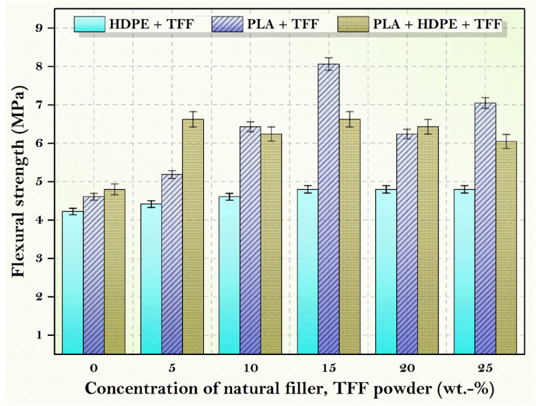


**Figure 4.** Flexural behaviours of TFF reinforced PLA/HDPE-based composites.

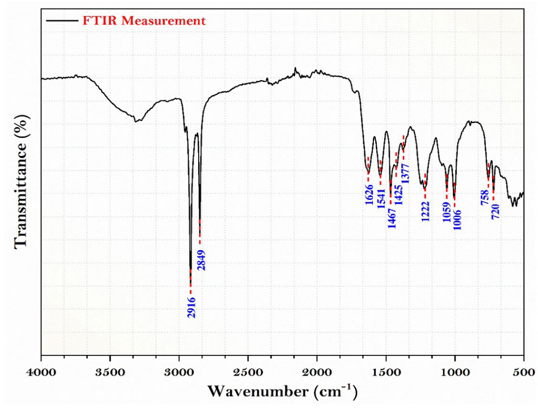


**Figure 5.** FTIR analysis.

### *3.4 FTIR analysis*

FTIR test is the straightforward method used to obtain an infrared spectrum of liquid, solid and gas absorption or emission. It can classify unidentified materials, determine sample quality and quantity the chemical component present in a mixture and so on. The stretches of different functional groups such as hydroxy isoxazole $C_4H_3NO_4$ stretch, amide CO (Ninhydrin) stretch, polyetherimide (PEI) stretch are produced via the polycondensation reaction between bisphenol-A dianhydride such as tetracarboxylic dianhydride (produced from the reaction of bisphenol A and phthalic anhydride) and a diamine such as m-phenylene diamine and ethyl. The above composite result (Figure 5) is derived from the spectrum generated during an FTIR test and digitally cross-checked against established reference from libraries and databases to determine the type of substance.

### *3.5 Morphological analysis*

Figure 6A-6F illustrates the surface of TFF + PLA with 15 wt.-% filler concentration, which has the best tensile strength. Increased wettability is due to strong adherence between the filler and the matrix. It is due to the reinforcement and matrix's increased interfacial contact. In the TFF + PLA composite, the filler dispersion was consistent, which helped to improve the tensile properties. In contrast, increasing the filler content in HDPE + TFF causes the fillers to clump together and separate from the matrix. It generates a discontinuity between the matrix and the filler, which aids crack development and reduces the tensile performance. This is the main reason behind deteriorating the composite mechanical properties. Figure 7A and Figure 7B shows the chemical composition of TFF/PLA/HDPE. The elements spectrum of the TFF was obtained using a SEM with an EDAX instrument, Nano XFlash Detector model. The obtained peak demonstrates that carbon (85.82 percent) and oxygen (14.18 percent) make up the majority of the weight. The crystal structure of the PLA (40 wt.-%) + HDPE (40 wt.-%) + TFF (20 wt.-%) specimen was determined using XRD. The powdered particle is made up of several tiny and randomly oriented particles that are exposed to monochromatic X-ray radiation. Figure 8 depicts the appropriate XRD patterns of the specimen produced. The following peaks show the composites crystalline solid structure: 16.56°, 19.21°, 21.53°, 23.87°, 30.03°, 36.27°, 39.71°, 40.66°, 41.56°, 42.87°, 44.02°, 46.89°, 48.98°, 52.97°, 54.78, 57.31°, 61.64°, 74.36°, 78.46°, 36.27°, 39.71°, 40.66° and 41.00o. However, at 21.53°, the most intense peak of Li2 Mg1 Si1 can be seen, with a peak height of 10486.13 cts and a relative intensity of 100 percent. The discovered compounds Lithium Magnesium Silicide (2/1/1) (Li2 Mg1 Si1) with cubic crystal structure and Zirconium Diphosphate (O7 P2 Zr1) with orthorhombic crystal structure provide the crystal structure of PLA + HDPE + TFF composite.

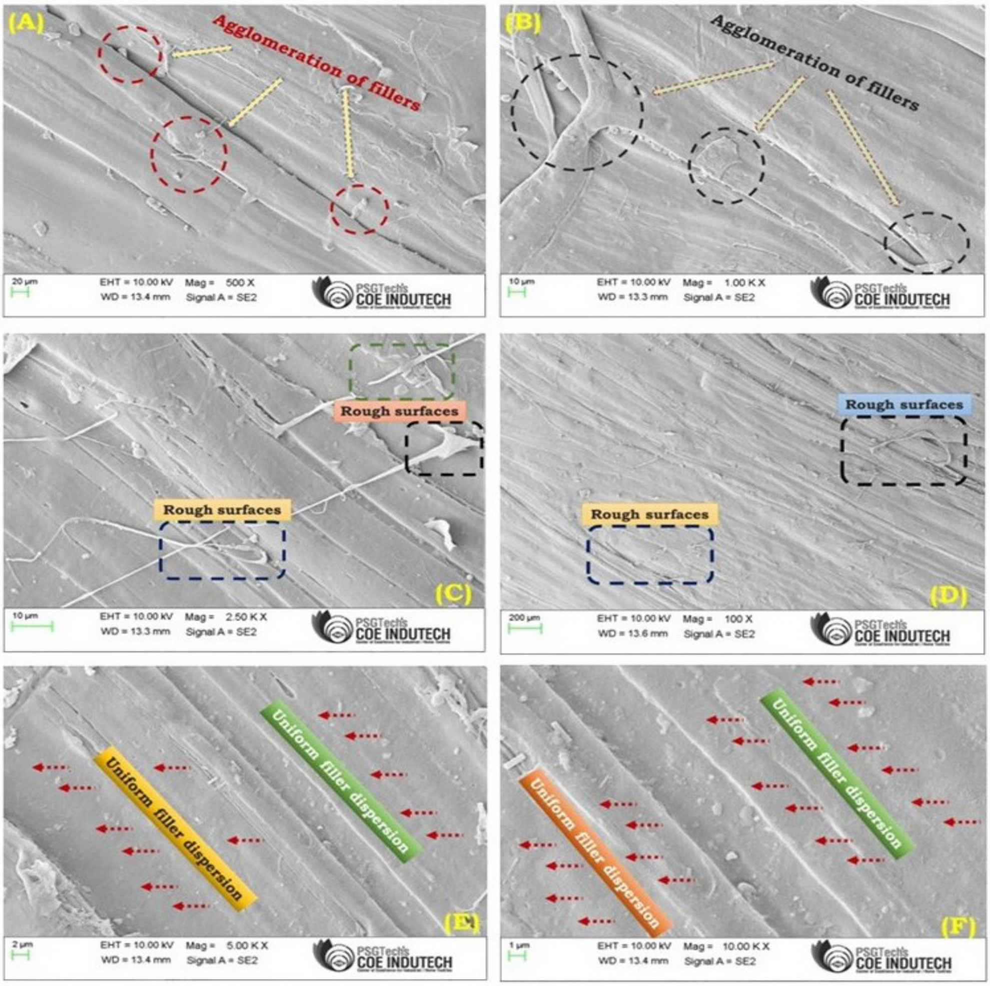


**Figure 6.** FESEM micrographs showing morphological features of the composite, highlighting agglomeration of fillers (A & B); rough surfaces (C & D); and uniform filler dispersion (E & F) at various magnifications.

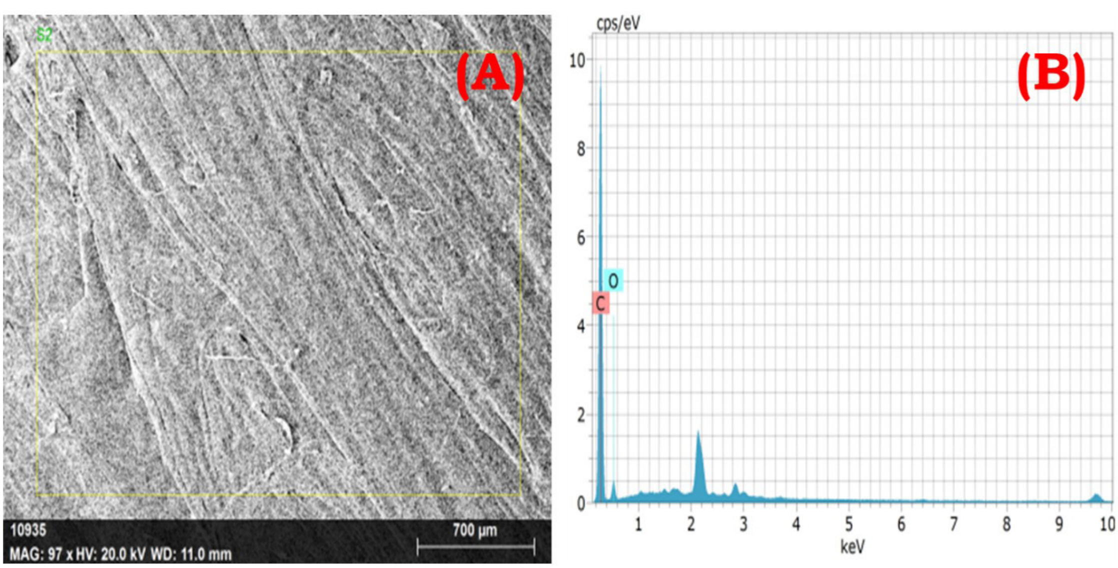


**Figure 7.** (A) FESEM image showing the surface morphology of the composite at low magnification; and (B) corresponding EDS spectrum with chemical composition of the TFF.

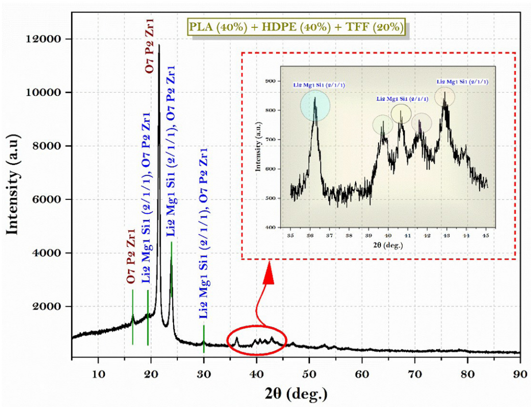


**Figure 8.** XRD analysis.

## 4. Conclusions

In this study, compression moulding was used to create the TFF/PLA, TFF/HDPE, and TFF/PLA/HDPE composites. The mechanical behaviour of the composites was studied as a function of the concentration of TFF filler. The experimental findings led to the following conclusions:

The tensile results revealed that the TFF filler reinforcement enhanced the tensile strength of the PLA matrix substantially. TFF/PLA attained a maximum tensile strength of 5.98 MPa. With a 15 wt.-% TFF filler concentration and 75 wt.-% PLA matrix polymer, the greatest flexural strength of 8.06 MPa was reached, which is 75 percent higher than pure PLA. The impact strength of composites showed an opposite trend to that of neat PLA which is mainly due to brittle nature of PLA at room temperature. Thus, the TFF/PLA composite with 15 wt.-% filler concentration exhibited the highest tensile strength among the studied samples. In the TFF+PLA composite, the filler dispersion was consistent, which helped to improve the tensile properties. The above composite result is derived from the spectrum generated during an FTIR test and digitally cross-checked against established reference from libraries and databases to determine the type of substance. The discovered compounds Lithium Magnesium Silicide (2/1/1) (Li2 Mg1 Si1) with cubic crystal structure and Zirconium Diphosphate (O7 P2 Zr1) with orthorhombic crystal structure provide the crystal structure of PLA + HDPE + TFF composite. As a result, composites have shown exceptional matrix absorption by the fillers, resulting in enhanced mechanical behaviour. The material is suitable for low to medium-duty applications.

## 5. Author's Contribution

- **Conceptualization** – Sreenivasaraja Nagarajan; Kathiresan Marimuthu; Prashanth Shanmugam; Moganapriya Chinnasamy
- **Data curation** – Sreenivasaraja Nagarajan; Kathiresan Marimuthu; Prashanth Shanmugam; Moganapriya Chinnasamy
- **Formal analysis** – Sreenivasaraja Nagarajan; Kathiresan Marimuthu; Prashanth Shanmugam; Moganapriya Chinnasamy
- **Funding acquisition** – Sreenivasaraja Nagarajan; Kathiresan Marimuthu
- **Investigation** – Sreenivasaraja Nagarajan; Kathiresan Marimuthu; Prashanth Shanmugam; Moganapriya Chinnasamy
- **Methodology** – Sreenivasaraja Nagarajan; Kathiresan Marimuthu; Prashanth Shanmugam; Moganapriya Chinnasamy
- **Project administration** – Kathiresan Marimuthu
- **Resources** – Sreenivasaraja Nagarajan; Kathiresan Marimuthu
- **Software** – NA.
- **Supervision** – Kathiresan Marimuthu
- **Validation** – Sreenivasaraja Nagarajan; Kathiresan Marimuthu; Prashanth Shanmugam; Moganapriya Chinnasamy
- **Visualization** – Kathiresan Marimuthu
- **Writing – original draft** – Sreenivasaraja Nagarajan; Prashanth Shanmugam
- **Writing – review & editing** –Kathiresan Marimuthu; Moganapriya Chinnasamy

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