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Development of composite sheet from leather waste and plant fibres - Value added sustainable product

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This study focuses on preparing composite material from leather fibres (LF) in combination with palm fibres (PF) using a natural binding agent for footwear applications. The raw materials are characterized for their mechanical, morphological, chemical and thermal properties to ensure their suitability and compatibility for composite preparation. Composite sheets are fabricated with varying proportions of palm and leather fibres (PF: LF) such as 50:50, 60:40, 70:30 and a control with 100% LF. Prepared sheets are tested for their tensile strength, tear strength, flexing, abrasion resistance, water absorption, desorption and other properties. Results reveal that sample with 50:50 ratio offers the best performance, making it suitable for product fabrication. Subsequently, the composite sheet is fabricated into footwear accessories. The study highlights the sustainable use of solid leather waste and natural fibres.

Keywords: Composites, Footwear component, Leather fibre, Natural fibre, Palm fibre

1 Introduction

The footwear industry utilizes various materials and processes to produce a wide range of products, from sandals to specialized safety footwear. Approximately 40 different components are used in shoe construction like leather, synthetic materials, rubber and textiles. Consequently, this industry generates enormous solid wastes which are difficult to manage[1,2]. Conventionally, 20-30% of leather is discarded as waste during footwear production[3]. The different types of solid leather wastes generated include leather trimmings, leather dust etc. and these pose threatening effects on the environment as well as on human health during disposal. Converting leather waste into value-added products is an effective step toward eco-friendly environment and industry sustainability[4,5], transforming solid wastes into usable composite materials for footwear components.

Natural fibre-reinforced composites are increasingly popular when it comes to waste management. Composites can be defined as materials that consist of two or more chemically and physically different phases separated by a distinct interface[6]. Many natural fibres, such as flax, hemp, jute, palm, ramie, roselle, mesta, rattan, agave, manila, barley, rye coir, kapok, cotton, sisal, abaca, maize, rice, bamboo, pineapple, plantain, etc., are being used for several years in the fabrication of composites reinforced with polymer materials like polyester, polyethylene, polypropylene, polyurethane, or polyamides, due to their added advantages of abundant availability, cost-effectiveness, enhanced strength characteristics, lightweight properties, degradability, eco-friendly, energy dissipation and so on[7]. They enhance the existing properties in the materials in the matrix structure[8].

Palm fibres, one of the most abundant natural fibres, are of potential interest to support industrial sustainability by producing alternatively economically eco-friendly materials[9]. Even though various parts of palm tree (*Borassus flabellifer*), such as trunk, petiole, stalk, bunch, fruit, etc. are used for making multiple products like fibre boards, bags, biofuel, oils, waxes, wines, roofs, mats, composites, etc., large quantities of palm biomass often remain underutilized[10].

Hence, an attempt has been made in the present work to combine leather waste fibres and palm fibres with natural binders to produce a composite material. It is decided to choose natural rubber latex from *Hevea brasiliensis*, as a binder for composite preparation, to create an eco-friendly product along with leather and palm fibres[11]. The natural rubber obtained is pure poly-cis-1.4 isoprene (contains more than 99.9% of cis 1.4 structural units)[12]. This value-

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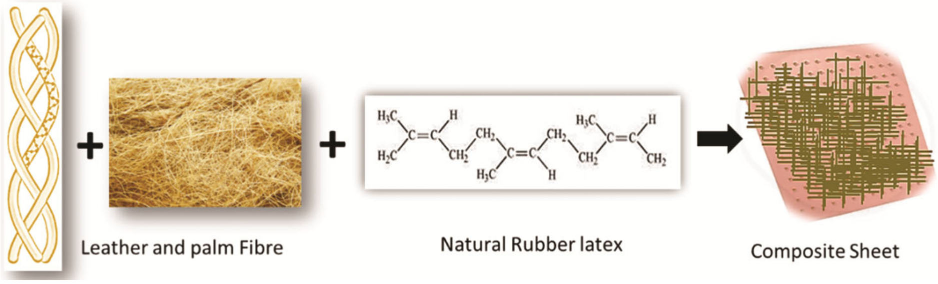

Fig. 1 — Preparation of reinforced composite sheet combining leather wastes with natural fibre

added product will lead to remarkable improvements in the life cycle of natural raw materials and sustainable use of wastes, contributing to sustainability, improving ecology and economic efficiency, and reducing the "pressure" of waste on the environment.

2 Materials and Methods

Finished leather trimmings, buffing dust and product wastes were collected from leather product industries. The palm fibres and natural rubber latex used were of commercial grade.

2.1 Preparation of Palm and Leather Fibres

Palm fibres were washed and cleaned with water and a 2% detergent solution, then sun-dried for one week to remove moisture completely. The fibres were chopped to 3-5 mm length and ground using a pulverizing machine (Sturtevant, SDL868, USA). The pulverized fibres were then sieved to obtain grades of varying particle size with 1-2 mm diameter. Leather trimming wastes were cut into small pieces and converted into leather fibre using the pulverizing machine. The average fibre size is 1-3 cm in length and 0.3-0.8 mm in diameter[13].

2.2 Preparation of Composites

The obtained leather fibres (LF) and palm fibres (PF) were converted into composite sheets using natural rubber latex (dry rubber content 60%) as a binding agent (Fig. 1). Composite sheets were fabricated with PF and LF ratios of 50:50, 60:40, and 70:30, designated as samples B, C, and D, respectively, while sample A (control) contained 100% LF. According to Table 1, these fibres were mixed. A solution of natural rubber latex and water (100 mL: 50 mL) was added to the mixed fibres along with 1% ethylene glycol. The mixture was agitated using a high-speed mechanical stirrer at 20°C and the resulting homogenous mixture was poured onto a steel frame mould measuring 200 mm x 150 mm, spreading it evenly to achieve uniform thickness. The mould was kept in a hot air oven at 90±2°C for 20 min, followed by 40±2°C for 8 h and then cooled. The sheets were roller-pressed to obtain a uniform surface and stored in a conditioning chamber at 20±2°C and 65% relative humidity. The measured thickness of the sheets was 2.5-3 mm. Different compositions were studied to develop composites and then characterized for their functional properties.

Table 1 — Compositions of composite samples

Sample code	Leather fibres (%)		Palm fibres (%)
	Leather dust	Leather trimmings	
A	70	30	0
B	35	15	50
C	25	15	60
D	20	10	70

2.3 Characterization

The different composite samples were prepared and characterized for their functional properties, including tensile strength, elongation at break, stitch tear strength, flexing resistance and water absorption/desorption. They were also subjected to structural and thermal analyses[14].

2.3.1 Mechanical Properties

Tensile strength, elongation at break and tear strength of LF/PF composites were measured using a Universal Testing Machine in accordance with SATRA TM43 and SATRA TM30 standards. The testing machine with a uniform separation speed of the jaws at 100±20 mm/min was used. The flexural endurance of composites was also tested to determine the resistance of the material to cracking, according to ASTM D6182 standard. At set intervals, the number of flexing cycles were recorded, noting any damage to the specimen. Water absorption and desorption properties of the composite sheets were analyzed according to SATRA TM 171 test method, to determine the rate at which the material absorbs and desorbs water. The mass of the test specimen before and after the water immersion for standard time was recorded and measured. The water uptake (mass gained) was measured by subtracting the initial weight from the final weight and then the percentage was calculated. These tests were conducted at an ambient temperature of 28°C and relative humidity of 65%.

2.3.2 Thermal Properties

The thermal characteristics of composite specimens were analyzed using Netzsch DSC 200 PC Differential Scanning Calorimeter. The samples were subjected to nitrogen atmosphere between 30 to 850°C at a heating rate of 10°C/min to measure the thermal properties.

2.3.3 SEM Analysis

The structural and morphological characteristics of the composite sheets were examined using Scanning Electron Microscopy (SEM). The analysis was conducted using a TESCAN/CLARA Scanning Electron Microscope. Prior to imaging, the leather specimens were coated with a thin layer of gold to enhance conductivity, using an Edwards E 306 Sputter Coater. The gold-coated samples were then analyzed under the microscope to capture detailed surface morphology and structural features.

*2.3.4 FTIR Analys*is

Fourier Transform Infrared (FTIR) spectroscopy was employed to identify the presence of specific functional groups in the composite samples. The samples for FTIR analysis were prepared by finely grinding the material with potassium bromide (KBr) and compressing the mixture into pellets. The analysis was performed using a JASCO FT/IR-4000 series spectrometer, which provided detailed information on the functional group composition and chemical structure of the samples.

3 Results and Discussion

3.1 Mechanical Properties

The prepared composites are proposed to be used in footwear applications. Mechanical properties such as tensile strength, elongation, tear strength and flexing, water absorption and desorption, are evaluated to understand their behavior. Tensile strength ensures the material's durability when an external force is applied. The results of the mechanical properties for different proportions of leather fibres and palm fibres are presented in Table 2.

It is evident from the Table 2 that sample B, with a ratio of 50:50 (PF/LF), shows appreciable tensile strength compared to other samples. The presence of both collagen fibres and cellulose in equal proportions in the composite result in a uniform network between palm and leather fibres, which enhances strength[15]. Sample A, consisting of 100% leather fibres, performs reasonably well in tensile strength while samples C and D, with a relatively higher composition of palm fibres as compared to leather fibres, exhibit decreased tensile strength due to insufficient interfacial bonding between the leather fibres and palm fibres.

Elongation is a measure of deformation of a material before it breaks when subjected to a tensile load. The data clearly shows that sample A has the highest elongation property, and this might be attributed to collagen, a flexible material and main constituent in leather which provides the elastic property. In samples C and D, a drastic decrease in elongation is observed with a decrease of leather fibre content[16].

Tear strength is the resistance of a material against tearing, which is necessary for fabricating footwear products. Table 2 indicates that the tear strength values are appreciable for combinations of samples having both leather fibres and palm fibres. The prime structural component in palm fibres, cellulose, provides adequate tear strength to the composites. Sample A results in a comparatively lower tear strength due to the random orientation of collagen fibres[17].

The basic requirement of footwear is the ability to flex the foot during walking and running without deforming. Shoes with high flexibility allow faster, smoother movement and help prevent foot deformation. From Table 2 it is observed that flexing resistance is maximum for sample B compared to sample A. In addition, with increased palm fibre content in samples C and D, flexural index values were decreasing. This decrease at higher palm fibre

Table 2 — Mechanical properties of composite samples

Sample code	Tensile strength MPa	Elongation at break, %	Tear strength N/mm	Flexing index	Water absorption %	Water desorption %
A	2.8±0.9	153.0±30	4.0±0.07	3.0±0.02	72.0±3	85.0±5
B	3.0±0.3	81.0±10	10.0±0.2	4.0±0.07	15.0±1	88.0±5
C	2.5±0.7	23.0±2	7.0±0.2	1.0±0.09	31.0±3	100.0±10
D	2.3±0.4	29.0±2	8.0±0.2	1.0±0.09	27.0±3	96.0±8

loading may be due to the interruption caused by hydrophilic lignocellulose in palm fibres[18].

Water absorption and desorption refer to the water retention and transport properties that affect the final product's durability. Footwear should have high desorption properties to remove sweat and moisture from the feet while being worn, and low absorption properties to respond minimally to external moisture sources such as rain or chemical spills. The results in Table 2 show that sample A has the lowest water desorption values compared to other samples, while the highest water desorption tendency is observed for sample C. Regarding water absorption properties, control sample A exhibits the highest value and this may be attributed to leather's inherent water permeability. Samples C and D show higher absorption values, attributable to the lignocellulosic materials - cellulose, hemicellulose and lignin - which contains -OH groups that attract water molecules[19].

The results reveal that sample B achieves the intended values of higher desorption and lower absorption rates. Therefore, sample B demonstrates better functional properties, indicating its suitability for fabricating footwear and its components.

3.2 SEM Analysis

The SEM images of various composites are given in Fig. 2. The morphology of the samples helps assess

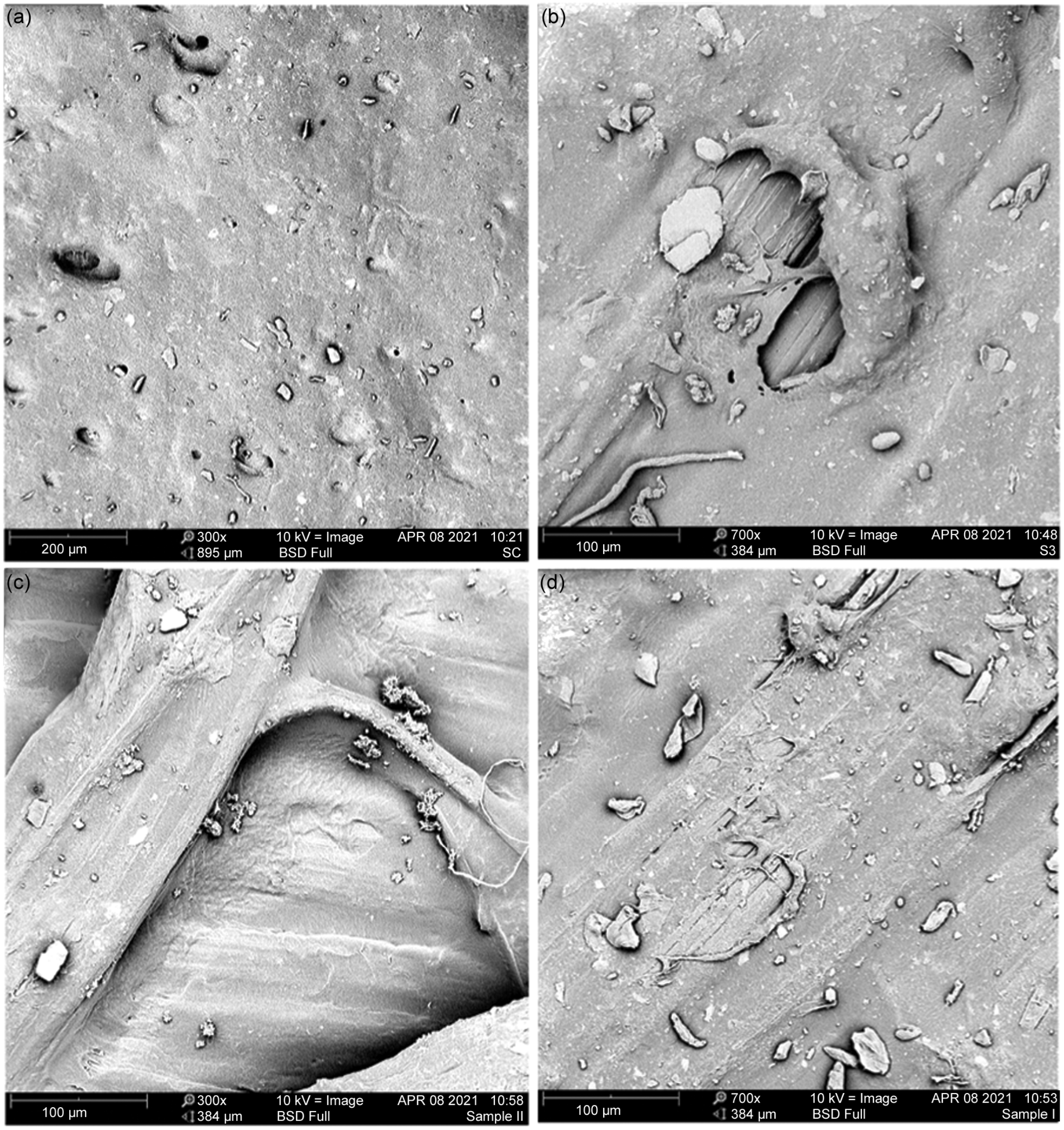

Fig. 2 — SEM images of composite samples with PF/LF of (a) 0:100, (b) 50:50, (c) 60:40 and (d) 70:30

the distribution of fibres within the composite matrix. Additionally, morphological changes provide better predictions of fibre interactions with the leather matrix and the fracture behavior of the composite samples[17]. Figure 2 (a) displays a homogeneous matrix structure, while Fig. 2 (b) indicates a well-combined distribution of palm and leather fibres. In Figs 2 (c) & (d), there is a mixture of fractured fibres and coagulated rubber solids, which results from poor adhesion between the matrix and palm fibres. Other than the pure leather matrix, the surfaces of all other matrices appear irregular, with some voids due to fibre pull-out, confirming the presence of weaker bond interactions[20].

3.3 FTIR Analysis

Figure 3 shows the FTIR spectra of the composite samples, confirming the presence of functional groups characteristic of the composite materials. The main functional groups in palm fibres and leather fibres include hydroxyl (OH), N-H, C-H and carbonyl (C=O) groups. The spectra in Figs 3 (b), (c) and (d) show a peak around 1020 cm^{-1}, indicating the stretching of unconjugated C=O groups in polysaccharides and lignin, confirming the presence of palm fibres[21]. The peak at 1371 cm^{-1} is probably due to CH_2 bending in cellulose, while the peak at 2853 cm^{-1} is due to the stretching of methyl and methylene C-H groups in polysaccharides. The peak at 3288 cm^{-1} indicates the presence of OH groups[22].

In the FTIR spectrum of leather fibres shown in Fig. 3 (a), a peak around 3730 cm^{-1} appears due to OH stretching vibrations of water molecules present in leather. Peaks near 3300 cm^{-1} is characteristic of NH stretching vibrations in proteins, and peaks between 2900 cm^{-1} and 2800 cm^{-1} signify CH stretching vibrations of CH_2 groups[23]. The peak at 1723 cm^{-1} is due to C=O stretching of carbonyl groups, while the peaks around 1654 cm^{-1},1550 cm^{-1} and 1445 cm^{-1}

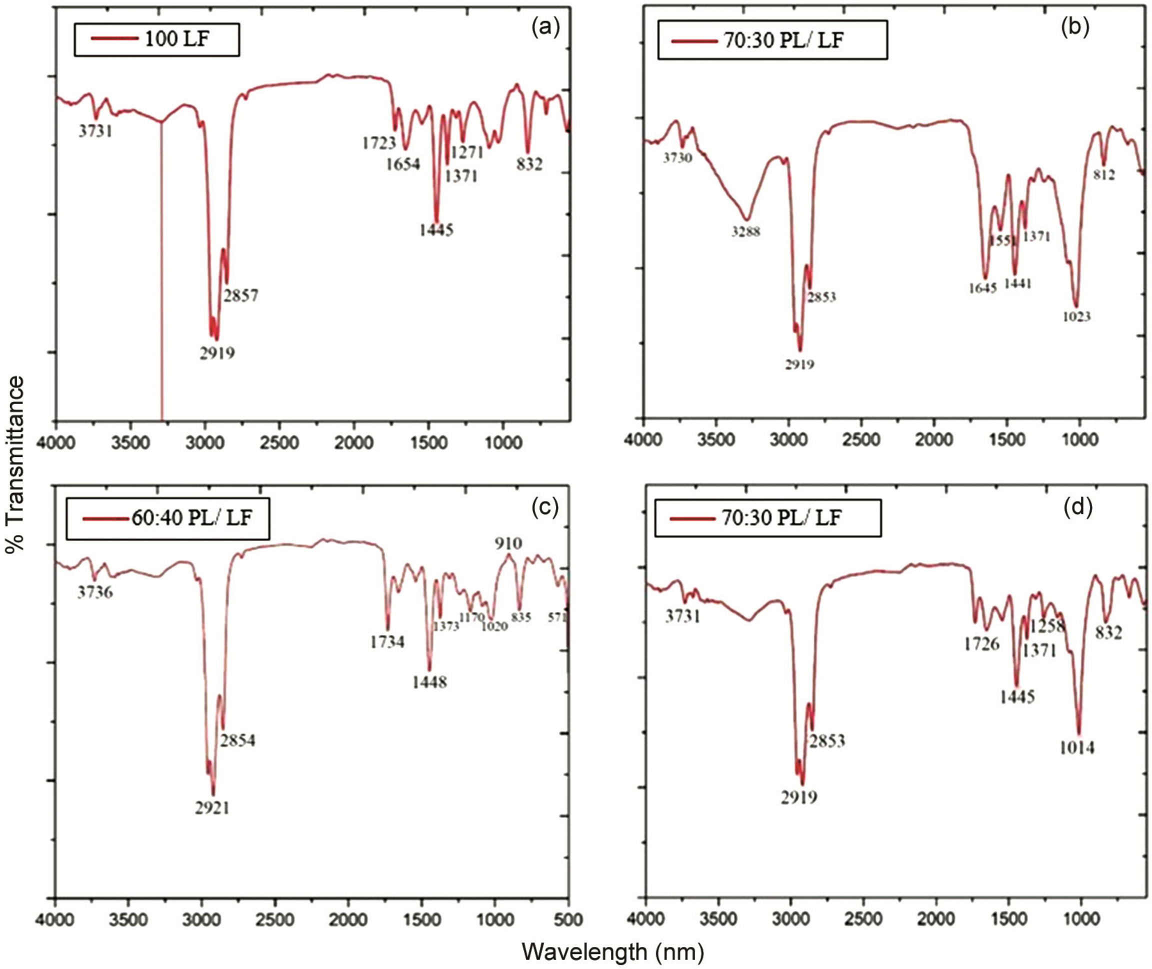

Fig. 3 — FTIR spectra of composite samples with PF/LF of (a) 0:100, (b) 50:50, (c) 60:40 and (d) 70:30

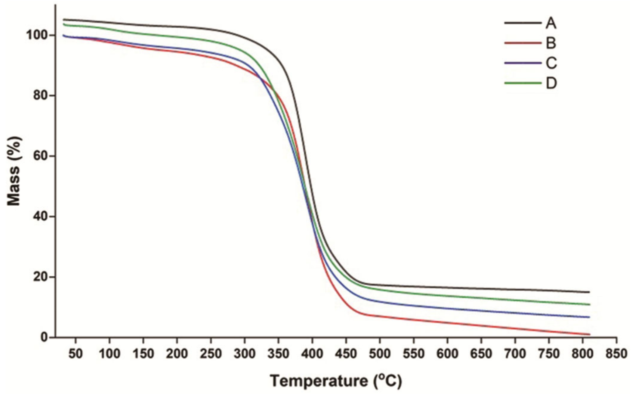

Fig. 4 —Thermogravimetry analysis

represent amide I, amide II, and amide III groups, respectively[17,24,25].

The shift in wavenumber from 1654 cm^{-1} to 1645 cm^{-1} in sample B is due to the strong hydrogen bond between the NH group of collagen in leather fibres and the carbonyl group in palm fibres. The disappearance of the peak at 1723 cm^{-1} is due to the high concentration of C=O groups in both leather fibres and palm fibres, weakening the vibration of the C=O group [Fig. 3(b)].

Similarly, for the composites, peaks around 3730 cm^{-1}, 2920 cm-1 and 2853 cm^{-1} correspond to OH, NH and CH stretching vibrations, while peaks around 1730 cm^{-1}, 1445 cm^{-1} and 1371 cm^{-1} indicate C=O stretching and C-H bending vibrations. Peaks around 1050 cm^{-1} are due to C-O bonds in cellulose and hemicellulose. Similar peaks to those of leather fibres are observed in composite samples [Figs 3 (c) & (d)].

3.4 Thermal Properties

Thermal degradation and yield efficiency of composite samples are assessed through TGA analysis, presented in Fig. 4. Decomposition begins around 300°C for all samples and continues till 500°C, leaving a residue of about 10-15%. This significant weight loss is due to the thermal degradation of hemicellulose, cellulose and collagen[19,26,27]. Hemicellulose, which is comprised of polysaccharides, degrades at a faster rate than cellulose due to its lower thermal stability[28]. Final decomposition occurs around 800°C, resulting in 3-10% residue. The results show that all the samples follow a three-step weight loss pattern, irrespective of composition.

In sample A, where LF content is maximum, the amide bonds contribute to the initial decomposition observed at a highest initial decomposition temperature (IDT) (317°C) compared to other blends. While in sample B (50:50), the equal presence of PF and LF results in a slightly lower IDT (303°C) due to hemicellulose in PF and hydroxyl groups and hydroxyproline in LF. The IDT further reduces to 292°C and 289°C in samples C (60:40) and D (70:30), respectively as LF content reduces, leading to fewer amide bonds and reduced IDT. The results suggest that as amide bond content, increases, the thermal stability also enhances.

Table 3 — Comparison of insole quality standard with composite sample

Parameters	Standard requirements	Composite sample B
Tensile Strength, N/mm^2	Min. 4.0	4.0
Flexing Index	Min. 2.7	4.0
Water absorption, %	Min 35	14.0
Water desorption, %	Min. 40	89.0

3.5 Fabrication of Insole from Composite

Among the composite samples with varying proportions, sample B possesses better physical and mechanical properties and is therefore chosen for insole fabrication[29]. The selected composite sample for product making is analyzed for insole applications before fabrication (Table 3). The composite sample B possesses comparable properties with respect to standard requirements. The composite sheet thickness ranges between 2.2-2.4 mm, and it is attached to a thin layer of fabric (0.8-1.0 mm of spun cloth) as a protective layer using adhesive. Then the insole is attached to the shoe and assessed for its application, meeting the desired functional requirements[30,31].

4 Conclusion

In this study, an effort is made to utilize the leather wastes from leather and leather and palm product industries to create composite sheets for footwear fabrication. The results show that the LF 50: PF 50 composite sheets exhibit the highest mechanical and thermal stability. Optimum incorporation of palm fibres improves functional properties and has a marked effect on water desorption tendency. SEM analysis confirms composite's cohesive structure, which is necessary for product fabrication. TGA analysis supports the required degradation temperature. This work effectively demonstrates the utilization of replenishable plant fibres and leather fibres to develop environmentally and economically beneficial lifestyle products.

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