STUDIES ON MECHANICAL SURFACE-FIBRILLATED AGRO-WASTE PINEAPPLE LEAF FIBRE REINFORCED POLYPROPYLENE COMPOSITES

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Studies on mechanical surface-fibrillated agro-waste pineapple leaf fibre reinforced polypropylene composites

by

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CERTIFICATE

This is to certify that the thesis entitled "**Studies on mechanical surface-fibrillated agro-waste pineapple leaf fibre reinforced polypropylene composites**" submitted by **Mr. Habibur Rahman** to the **Indian Institute of Technology Delhi** for the award of the degree of **Doctor of Philosophy** in the **Department of Textile and Fibre Engineering**, is a record of bona fide research work carried out by him. **Mr. Habibur Rahman** has worked under our guidance and supervision.

The results contained in this thesis are original and have not been submitted, in partial or full, to any other university or institute for the award of any degree or diploma.

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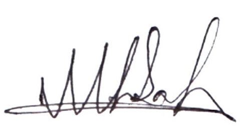

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Abstract

Agro-waste pineapple leaf fibre (PALF) was extracted from fresh pineapple leaves for use as a green reinforcement in polypropylene (PP) composites. There is a growing interest in the use of PALF as reinforcement in thermoplastic matrix composites to develop high-performance impact-resistant materials in technical applications. PP undeniably offers economic, ecological, and practical benefits compared to other thermoplastic polymers due to its affordability, environmentally friendly recycling process, and excellent thermal stability. Generally, chemical treatment is used to clean the sun-dried PALF, and the effluents of those chemicals are discharged into nature.

In this study, new mechanical approach (carding) was used to remove the remaining extraneous substances from the scratched and sun-dried PALF instead of using any chemical treatment. Optimum number of passages through carding process did not reduce the fibre length or deteriorate the mechanical properties of fibres and was found to be effective in improving the fibre surface roughness, leading to an improved fibre-matrix interface. The carding process also removed the non-cellulosic materials from PALF, resulting in an increase in the cellulose percentage. Four passages of PALF through carding successfully removed 12 wt.% of non-cellulosic materials.. The percentage of sticky fibres and dust content decreased, and fibre breakage increased with further increase in number of carding passages. The properties of individualized fibres were also influenced by the number of carding passages. The carding process reduced the diameter of PALF due to the removal of extraneous materials and increased the tensile strength and modulus. Scanning electron microscopy (SEM) and Fourier-transform infrared spectroscopy (FTIR) confirmed the elimination of non-cellulosic materials along with other gummy and waxy substances from the surface of fibrillated PALF due to the carding action.

The crystallinity % also increased after the carding process due to the removal of amorphous substances. As the reinforcement-matrix interfacial shear strength (IFSS) strongly influences the physico-mechanical properties of composites, the IFSS of carded PALF-reinforced PP fibre micro-bonded composites was also studied using the single fibre pull-out test (SFPT). PP in the form of fibres, which on melting transforms in to matrix, was used. PP fibres of two different fineness values were used to study their influence on composite properties. Composites with PALF, carded four times and finer PP fibre showed the highest IFSS. Additionally, the load extension curves and SEM images of the tensile pulled-out surface were investigated to study the interfacial adhesion between PALF and PP micro-droplets. The increased surface roughness of PALF due to the carding action resulted in an improvement in the IFSS of PALF-reinforced PP composites.

The high melt viscosity of PP hinders its penetration into the composite reinforcement. To overcome this difficulty, the individualized and fibrillated PALF and PP fibres of different fineness were blended (50% by weight) in the carding process to ensure homogeneous distribution of reinforcement and matrix in fibre form within composites. The blended carded sliver was further parallelized through a gill-drawing process, and subsequently, unidirectional composites were fabricated using the compression moulding technique. The drawn sliver was used to produce the composites in a compression moulding machine at 180°C for 15 minutes under 20 bar pressure. The consolidated samples were allowed to cool naturally to 100°C under constant pressure keeping inside the moulds. The fabricated composites were cut into different shapes according to the respective standards for the measurement of void percentage, surface water contact angle, thermal conductivity, dynamic mechanical analysis, tensile, flexural, short beam, and Izod impact strength, thermal conductivity, and water absorbency analysis. SEM was used to examine the fracture surface of the tensile strength-tested samples.

The composite made with four times carded PALF, demonstrated the best physico-mechanical performance among the samples, while those made with finer PP fibres as matrix forming fibres also contributed positively. The thermal insulation performance of the composites improved with shorter PALF carded five times and with coarser PP fibres, but at the same time, the water absorbency of these composites also increased, indicating reduced water resistance of such composites.

The drawn sliver PALF-PP fibres were converted into thermal-bonded sliver by melting the outer layer to keep the reinforcement fibres unidirectional in composite fabrication at 180, 190, 200, and 210°C. The tensile performances, bending rigidity, and weavability of the thermal-bonded slivers were evaluated. Then, the unidirectional composites of the thermally bonded slivers were fabricated according to the above-mentioned process, and their properties were analysed. The thermal-bonded sliver was treated at 200°C, and composites fabricated with this sliver showed the best feasible physico-mechanical properties. Based on the results of this investigation, PALF and PP fibres can be chosen to produce thermal-bonded sliver with improved fibre alignment, as well as the final composites in accordance with the specifications of the intended use.

The thermal-bonded sliver treated at 200°C was used to make cross-laid (0/90°), 1/1 plain, 2/1 twill, and 3/1 twill structures with the same EPM (ends per meter) and PPM (picks per meter) but different patterns of interlacements. These structures were compression moulded into composites according to the same process used earlier. The tensile failure surface morphology and the interlaminar failure of short-beam shear strength (SBS) tested samples were analysed by SEM and micro computed tomography (μCT) images, respectively. The composite sample fabricated with cross-laid structures showed the best mechanical properties and a lower void percentage due to the lower order of interlacements.

According to the findings of this research, PALF and PP fibres can be converted in to thermal-bonded sliver to produce good quality textile-structured composites with aligned fibres. These composites can be tailor-made to have desired specifications, as a viable alternative to conventional and costly materials for protective applications.

Saar (Hindi)

कृषि अपशिष्ट अनानास पत्ती रेशा (PALF) को ताज़ी अनानास की पत्तियों से निकालकर पॉलीप्रोपाइलीन (PP) समिश्रों में हरित सुदृढीकरण के रूप में उपयोग किया गया। तकनीकी अनुप्रयोगों के लिए उच्च-प्रदर्शन एवं आघात-प्रतिरोधी सामग्री विकसित करने हेतु थर्मोप्लास्टिक मैट्रिक्स समिश्रों में PALF के उपयोग में निरंतर रुचि बढ़ रही है। PP अपनी कम लागत, पर्यावरण-अनुकूल पुनर्चक्रण क्षमता तथा उत्कृष्ट तापीय स्थिरता के कारण अन्य थर्मोप्लास्टिक पॉलिमरों की तुलना में आर्थिक, पारिस्थितिक और व्यावहारिक लाभ प्रदान करता है। सामान्यतः धूप में सुखाए गए PALF की सफाई के लिए रासायनिक उपचार किया जाता है, जिसके अपशिष्ट पर्यावरण में छोड़ दिए जाते हैं।

इस अध्ययन में रासायनिक उपचार के स्थान पर एक नवीन यांत्रिक विधि (कार्डिंग) का उपयोग किया गया, जिससे खुरचे और धूप में सुखाए गए PALF से शेष अशुद्धियों को हटाया गया। कार्डिंग की उपयुक्त संख्या ने रेशा लंबाई या यांत्रिक गुणों को प्रभावित नहीं किया, बल्कि रेशा सतह की खुरदरापन बढ़ाकर रेशा-मैट्रिक्स अंतरफलक को बेहतर बनाया। कार्डिंग प्रक्रिया से गैर-सेल्यूलोज पदार्थ हटे, जिससे सेल्यूलोज प्रतिशत में वृद्धि हुई। चार बार कार्डिंग करने पर लगभग 12 भार प्रतिशत गैर-सेल्यूलोज पदार्थ हट गए। आगे कार्डिंग बढ़ाने पर चिपचिपे रेशों और धूल की मात्रा कम हुई, जबकि रेशा टूटन बढ़ी। कार्डिंग प्रक्रिया के कारण रेशा व्यास में कमी आई तथा तन्य शक्ति और मापांक में वृद्धि हुई। स्कैनिंग इलेक्ट्रॉन माइक्रोस्कोपी (SEM) और फूरियर ट्रांसफॉर्म अवरक्त स्पेक्ट्रोस्कोपी (FTIR) से यह पुष्टि हुई कि कार्डिंग क्रिया के कारण रेशा सतह से गोंद, मोमी तथा अन्य गैर-सेल्यूलोज पदार्थ हट गए। अमोर्फस पदार्थों के हटने से क्रिस्टैलिनिटी प्रतिशत में भी वृद्धि हुई।

चूंकि सुदृढीकरण-मैट्रिक्स अंतरफलक कतरनी शक्ति (IFSS) समिश्रों के भौतिक-यांत्रिक गुणों को प्रभावित करती है, इसलिए कार्डेड PALF सुदृढ़ित PP सूक्ष्म-बॉन्डेड समिश्रों का IFSS एकल रेशा पुल-आउट परीक्षण (SFPT) द्वारा अध्ययन किया गया। दो विभिन्न महीनता वाले PP रेशों का उपयोग किया गया। चार बार कार्डेड PALF और अधिक महीन PP रेशों वाले समिश्रों में सर्वाधिक IFSS प्राप्त हुआ। कार्डिंग से बढ़ी रेशा सतह खुरदरापन ने अंतरफलक आसंजन को बेहतर किया।

PP की उच्च गलन श्यानता के कारण समिश्र में उसका समुचित प्रवेश बाधित होता है। इस समस्या के समाधान हेतु PALF और विभिन्न महीनता वाले PP रेशों को कार्डिंग प्रक्रिया में 50% भार अनुपात में मिश्रित किया गया, जिससे समिश्र में समान वितरण सुनिश्चित हो सके। इसके बाद गिल-ड्रॉइंग प्रक्रिया द्वारा रेशों को समानांतर किया गया और संपीड़न मोल्डिंग तकनीक द्वारा एकदिशीय समिश्र तैयार किए गए। 180°C तापमान और 20 बार दबाव पर 15 मिनट तक मोल्डिंग की गई। नमूनों को दबाव के तहत 100°C तक ठंडा किया गया। विभिन्न मानकों के अनुसार उनके रिक्तता प्रतिशत, जल संपर्क कोण, तापीय चालकता, गतिशील यांत्रिक गुण, तन्य, मोड़, लघु-बीम तथा इज़ोड आघात शक्ति एवं जल अवशोषण का परीक्षण किया गया। SEM द्वारा टूटन सतह का विश्लेषण भी किया गया। चार बार कार्डेड PALF से बने समिश्रों ने सर्वोत्तम भौतिक-यांत्रिक प्रदर्शन प्रदर्शित किया, जबकि महीन PP रेशों का उपयोग भी लाभकारी रहा। पाँच बार कार्डिंग तथा मोटे PP रेशों के साथ तापीय इन्सुलेशन बेहतर हुआ, परंतु जल अवशोषण भी बढ़ा, जिससे जल प्रतिरोध में कमी आई। ड्रॉन स्लिवर PALF-PP रेशों को 180, 190, 200 और 210°C पर बाहरी परत को पिघलाकर तापीय-बॉन्डेड स्लिवर में परिवर्तित किया गया, जिससे रेशों को एकदिशीय बनाए रखा जा सके। इन स्लिवरों के तन्य गुण, मोड़ कठोरता और बुनाई योग्यता का मूल्यांकन किया गया। 200°C पर तैयार स्लिवर से बने समिश्रों ने सर्वोत्तम गुण प्रदर्शित किए।

200°C पर तैयार तापीय-बॉन्डेड स्लिवर का उपयोग करके क्रॉस-लेड (0/90°), 1/1 प्लेन, 2/1 ट्विल और 3/1 ट्विल संरचनाएँ तैयार की गईं। समान EPM और PPM के साथ इन संरचनाओं को संपीड़न मोल्डिंग द्वारा समिश्रों में परिवर्तित किया गया। SEM और सूक्ष्म-सीटी (μCT) विश्लेषण द्वारा इनके विफलता व्यवहार का अध्ययन किया गया। क्रॉस-लेड संरचना वाले समिश्रों में न्यूनतम रिक्तता तथा सर्वोत्तम यांत्रिक गुण पाए गए। इस अध्ययन के आधार पर यह निष्कर्ष निकाला जा सकता है कि PALF और PP रेशों का उपयोग करके तापीय-बॉन्डेड स्लिवर द्वारा उच्च गुणवत्ता वाले, संरचित और अनुकूलन योग्य समिश्र विकसित किए जा सकते हैं, जो सुरक्षात्मक अनुप्रयोगों के लिए पारंपरिक एवं महंगे पदार्थों का प्रभावी विकल्प सिद्ध हो सकते हैं।

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ABBREVIATION

PALF - Pineapple Leaf Fibre

PP - Polypropylene

% - Percentage

wt.% - Weight Percentage

Vol.% - Volume Percentage

FTIR - Fourier-Transform Infrared spectroscopy

SEM - Scanning Electron Microscopy

IFSS - Interfacial Shear Strength

SFPT - Single Fibre Pull-out Test

TS – Tensile Strength

TM – Tensile Modulus

FS – Flexural Strength

FM – Flexural Modulus

IS – Impact Strength

EPM - Ends Per Meter

PPM - Picks Per Meter

SBS – Short-Beam Shear Strength

μCT – Micro Computed Tomography

FAOSTAT - Food and Agriculture Organization Statistics

NaOH – Sodium Hydroxide

HCL – Hydrochloric Acid

MAgPP - Maleic Anhydride-grafted-PP

MFI - Melt Flow Index

ATR - Attenuated Total Reflectance

XRD - X-ray Diffraction

AFM - Atomic Force Microscopic

USA – United States of America

UD – Unidirectional

ASTM – American Society for Testing and Materials

FRPC - Fibre Reinforced Polymer Composite

°C - Degree Centigrade

W - Watt

K - Kelvin

W/mK – Watt per Meter Kelvin

θ - Theta

g/cm^3 - Grams per Centimeter cubed

NFRC - Natural Fibre Reinforced Composite

NFRTC - Natural Fibre Reinforced Thermoplastic Composite

2D, 3D - Two-dimensional, Three-dimensional

FSC - Film Stacked and Compressed

TBS – Thermal-bonded Sliver

IR - Infrared

MPa - Mega-Pascal

GPa - Giga-Pascal

μm - Micro meter

PLA - Poly-Lactic Acid

kHz - Kilo-Hertz

m/min - Meter per Minute

cm – Centimeter

m – Meter

m^2 – Square Meter

DSC - Differential Scanning Calorimetry

TGA - Thermogravimetric Analysis

cN - Centi-Newton

kN - Kilo-Newton

mm - Mili-Meter

g – Gram

D - Denier

g/den - Grams per Denier

kg - Kilogram

m/s – Meter per Second

J – Joule

Ref. - Reference