STUDY ON CREEP BEHAVIOUR OF PLANT FIBRE/EPOXY COMPOSITES

DEBARATI BHATTACHARYYA

DEPARTMENT OF TEXTILE AND FIBRE ENGINEERING

INDIAN INSTITUTE OF TECHNOLOGY DELHI

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by

Debarati Bhattacharyya

Department of Textile and Fibre Engineering

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Dedicated to my parents, parent in-laws, sister, nephew, husband, who have always been a constant source of support and encouragement in my life.

Certificate

This is to certify that the thesis entitled "Study on Creep Behaviour of Plant Fibre/Epoxy Composites" being submitted by Debarati Bhattacharyya to the Indian Institute of Technology Delhi for the award of the degree of Doctor of Philosophy is a record of bona fide research work carried out by her. Debarati has worked under my guidance and supervision and has fulfilled the requirements for submitting this thesis, which to our knowledge has reached the requisite standard. The results contained in this thesis are original and have not been submitted, in part or full, to any other university or institute for the award of any other degree or diploma.

Dr. Vijaykumar Narayandas Baheti

Associate Professor

Department of Textile and Fibre Engineering

Indian Institute of Technology Delhi

Hauz Khas, New Delhi-110016

India

Date:

Place: New Delhi, India

Declaration by the Author

This thesis comprises my original work and contains no material previously published or written by another person except where due reference has been made in the text. I have clearly stated the contribution of others in jointly authored works, which I have included in my thesis.

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Debarati Bhattacharyya

Abstract

Recently, the plant-based textile structural composites gained importance because of their greater strength-to-weight ratio, high corrosion resistance, and lower maintenance. The plant fibres are gradually replacing the synthetic fibres as reinforcements for the development of environment friendly products (door trim panels, engine and transmission enclosures, etc.) However, the plant fibres have few limitations over synthetic fibres especially in textile structural composites (subsea equipment, airplane and the oil gas industry, etc.), where they need to demonstrate superior long term mechanical performance to cyclic (fatigue) as well as long term constant load (creep) situations in harsh environments of varying temperature and humidity levels. This could result in dimensional instability and even structural failure of composites due to poor creep properties. In this thesis, we try to understand the creep behaviour of plant fibre-based textile structural composites during their service lifetime and exposure to aggressive environments conditions.

In the first study, the surface of jute fabrics was modified by alkali, ozone, and plasma treatments to remove the non-cellulosic materials and thereby improve their adhesion with epoxy matrix. The performance of surface treatments was evaluated based on surface morphology, mechanical properties, hydrophilicity, etc. The ozone surface treatment was found to remove lignin and increase hydrophilicity of jute fibres to greater extent as compared to other treatments, however with higher tendency of defibrillation and fibre rupture.

In the second study, the effect of alkali, ozone, and plasma surface treatments on the variability of the tensile strength of jute fibres was characterized by the Weibull analysis. It was found that the shape parameter (i.e. Weibull modulus) of surface treated jute fibres was higher than that of the untreated jute fibres. The plasma and ozone treatments of jute fibres

showed best results in terms of high Weibull modulus, low standard deviation and higher coefficient of correlation (R^2).

In the third study, the creep behaviour of jute fabric/epoxy composites at different environment temperatures was studied using several creep models (i.e., Burger's model, Findley's power law model, and Coupling Model). Ozone-treated samples demonstrated higher creep resistance than alkali- and untreated samples over most of the temperature range, whereas alkali-treated fibres exhibited superior behaviour at lower temperatures. Furthermore, the ozone treated composites shown higher elastic deformation occurring instantaneously at elevated temperatures of 70 °C and 100 °C, whereas less amount of deformation occurring due to the viscous regions as compared to alkali-treated jute fabric composites. The various creep models suggested that the improvement in fibre/matrix interface is very important to ensure longer life of natural fibre-based composites.

In the fourth study, the effect of surface treatment and subsequent separation of individual cellulose micro/nano fibrils from jute, corn husk and sisal plant fibres were studied. The ozone pre-treatment followed by pulverization in ball milling was selected for this purpose. The effect of ozone treatment was examined from morphological, physical, and mechanical properties using FTIR, XRD, SEM, TGA, etc techniques. The tensile strength of the fibres weakened gradually as the treatment time of ozonisation increased. The maximum drop in tensile strength was seen in case of corn husk fibres followed by sisal and jute fibres after the ozone treatments. The tensile strength was reduced by 70 %, 60 %, and 47 % in case of corn husk, sisal, and jute fibres respectively after 15 min of ozone treatment. When pulverization of plant fibres was carried out in ball milling, the ozone treatment was observed beneficial for easier defibrillation of cellulose micro/nano fibrils. The pulverization of jute, corn husk and sisal fibres produced the particles with diameter of 610 nm, 380 nm, and 530 nm after 15 min of ozone treatment respectively.

In the fifth study, the creep performance of ozone treated jute fabric/epoxy composites was studied at different environment temperatures after incorporation of 1, 3 and 5 wt. % of ozonized and pulverized cellulose particles. The incorporation of cellulose particles was found to lower the creep deformation of jute fabric/epoxy composites. The creep resistance of 5 wt.% filler loaded composites was found superior, especially at higher temperatures. The higher elastic deformation and slower viscous deformation was found in case of filler loaded composites at elevated temperatures of 70 °C and 100 °C. The various creep models were used to estimate the durability of composites by validating the obtained creep results. The coupling model predicted the creep behaviour of composites more accurately in addition to explanation on the creep mechanism.

सारांश

हाल ही में पौधों पर आधारित वस्त्र संरचनात्मक सम्मिश्र ने अपने उच्च बल-भार अनुपात, अधिक क्षरण प्रतिरोध तथा न्यूनतम रखरखाव के कारण महत्त्व प्राप्त किया है। कृत्रिम रेशों के स्थान पर पर्यावरण-अनुकूल उत्पादों (जैसे दरवाज़े के पैनल, इंजन एवं ट्रांसमिशन आवरण आदि) के विकास हेतु पौधों के रेशों का उपयोग बढ़ रहा है। तथापि, इन रेशों की कुछ सीमाएँ भी हैं, विशेषकर वस्त्र संरचनात्मक सम्मिश्रों में (जैसे सबसागर उपकरण, विमानन, तथा तेल-गैस उद्योग), जहाँ दीर्घकालिक यांत्रिक गुणधर्मों—चक्रीय थकान तथा विरूपण व्यवहार का कठोर परिवेश (भिन्न तापमान व आर्द्रता) में स्थायित्व आवश्यक होता है। इस कारण विरूपण गुणधर्म की कमी आयामी अस्थिरता अथवा संरचनात्मक विफलता उत्पन्न कर सकती है। प्रस्तुत शोध-कार्य में पौध-आधारित वस्त्र संरचनात्मक सम्मिश्रों की विरूपण प्रवृत्ति को उनकी कार्याविधि एवं आक्रामक वातावरणीय स्थितियों के अंतर्गत समझने का प्रयास किया गया है।

प्रथम अध्ययन में, जूट वस्त्र की सतह को क्षार उपचार, ओज़ोन उपचार तथा प्लाज़्मा उपचार द्वारा संशोधित किया गया जिससे गैर-सेलूलोसिक पदार्थ हटाकर एपॉक्सी मैट्रिक्स से आसंजन सुधारा जा सके। सतह संशोधन का आकलन सतही संरचना, यांत्रिक गुणधर्म एवं जलाभिलाषिता के आधार पर किया गया। ओज़ोन उपचार ने लिग्निन हटाने व जलाभिलाषिता बढ़ाने में सर्वाधिक प्रभावी परिणाम दिए, किंतु इसके साथ रेशा-वियोजन तथा रेशा-विच्छेदन की प्रवृत्ति भी अधिक पाई गई।

द्वितीय अध्ययन में, उपरोक्त सतह उपचारों के बाद जूट रेशों की तन्यता शक्ति की परिवर्तनशीलता का मूल्यांकन वेइबुल विश्लेषण से किया गया। सतह-उपचारित रेशों का वेइबुल गुणांक असंशोधित रेशों से अधिक पाया गया। प्लाज़्मा एवं ओज़ोन उपचारित रेशों ने उच्च वेइबुल गुणांक, निम्न मानक विचलन तथा उच्च सहसंबंध गुणांक (R^2) प्रदर्शित किया।

तृतीय अध्ययन में, विभिन्न तापमानों पर जूट वस्त्र/एपॉक्सी सम्मिश्रों के सरीजन व्यवहार का परीक्षण बर्गर मॉडल, फाइंडले की घात नियम मॉडल तथा युग्मन मॉडल द्वारा किया गया। ओज़ोन-उपचारित नमूनों ने

प्रायः सम्पूर्ण तापमान सीमा पर सर्वाधिक विरूपण प्रतिरोध दर्शाई, जबकि क्षार-उपचारित रेशों का प्रदर्शन निम्न तापमान पर श्रेष्ठ पाया गया। उच्च तापमान (70 °C तथा 100 °C) पर ओज़ोन-उपचारित सम्मिश्रों में अधिक तात्कालिक लोचदार विकृति तो देखी गई, किंतु श्यान विकृति अपेक्षाकृत कम पाई गई। अध्ययन से स्पष्ट हुआ कि रेशा/मैट्रिक्स अंतरफलक में सुधार प्राकृतिक रेशा-आधारित सम्मिश्रों की दीर्घायु हेतु अनिवार्य है।

चतुर्थ अध्ययन में, सतह उपचार एवं तत्पश्चात् जूट, मक्का भूसी तथा सिसल रेशों से व्यक्तिगत सेल्यूलोज़ सूक्ष्म/नैनो रेशिकाएँ पृथक्करण का अध्ययन किया गया। इसके लिए ओज़ोन प्री-उपचार एवं गेंद पीसाई का प्रयोग किया गया। फ़ोरियर रूपांतर अवरक्त वर्णक्रमिकी, *एक्स-रे विवर्तन, स्कैनिंग इलेक्ट्रॉन माइक्रोस्कोपी,* तथा *थर्मोग्रैविमेट्रिक विश्लेषण,* आदि तकनीकों द्वारा सतही एवं यांत्रिक गुणधर्मों का मूल्यांकन किया गया। ओज़ोन उपचार समय बढ़ने पर तन्यता शक्ति क्रमशः घटती गई, जिसमें अधिकतम कमी मक्का भूसी (70%), तत्पश्चात सिसल (60%) एवं जूट (47%) में 15 मिनट उपचार उपरांत देखी गई। साथ ही, ओज़ोन उपचार ने रेशिकाओं के आसान वियोजन में सहायक भूमिका निभाई।

पंचम अध्ययन में, ओज़ोन-उपचारित जूट वस्त्र/एपॉक्सी सम्मिश्रों में 1, 3 तथा 5 भार प्रतिशत ओज़ोनित एवं गेंद-पीसाई सेल्यूलोज़ कणों को सम्मिलित कर विभिन्न तापमानों पर विरूपण प्रदर्शन का परीक्षण किया गया। कणों की उपस्थिति से सरीजन विकृति में कमी देखी गई। 5 भार प्रतिशत युक्त सम्मिश्रों ने विशेषतः उच्च तापमानों पर श्रेष्ठ सरीजन प्रतिरोध प्रदर्शित किया। इन नमूनों में उच्च लोचदार विकृति तथा धीमी श्यान विकृति पाई गई। युग्मन मॉडल ने सरीजन व्यवहार का सर्वाधिक सटीक पूर्वानुमान प्रस्तुत किया एवं विरूपण तंत्र की व्याख्या भी की।

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Abbreviations

ASTM	American Society for Testing and Materials
AJF	Alkali treated jute fabric
AJY+E	Alkali treated jute yarn +epoxy
BNC	Bacterial Nanofibrillated Cellulose
CGAR	Compound Annual Growth Rate
CNC	Nanocrystals
CNF	Nanofibre
EPI	Ends per Inch
E_0	Intial Modulus
E_e	Equilibrium Modulus
FTIR	Fourier Transform Infrared Spectroscopy
FESEM	Fourier Scanning Electron Microscope
HDPE	High Density Polyethylene
K/S	Colour Strength
L	Lightness
MCC	Microcrystalline Cellulose
m	Weibull Modulus
NFC	Nanofibrillated Cellulose
NCC	Nanocrystalline Cellulose
NaBr	Sodium Bromide

NaClO	Sodium Hypochlorite
NPP	Neat Polypropylene
O_3	Ozone
OJF	Ozone treated fabric
OJY+E	Ozone treated jute yarn + epoxy
OTJF, OTCHF OTSF	Ozone treated jute fibres, corn husk fibres and sisal fibres
PBAT	Polybutylene adipate terephthalate
PPI	Picks per Inch
PVC	Polyvinyl Chloride
PJF	Plasma treated fabric
PVA	Polyvinyl Alcohol
PJY+E	Plasma treated jute yarn +epoxy
R^2	Coefficient of Determination
RHA	Rice Husk Ash
RPP	Recycled polypropylene
SEM	Scanning Electron Microscopy
TEMPO	2,2,6,6-tetramethylpiperidine-1-oxyl
TGA	Thermogravimetric Analysis
UTM	Universal Testing Machine
UJF	Untreated Jute Fabric

UJY+E Untreated jute yarn + epoxy

XRD X-ray Diffraction

List of Symbols

%	Percentage
χ_c	Crystallinity
γ_l^d, γ_l^p	Surface energies of liquid dispersive and polar components
γ_s^d, γ_s^p	Surface energies of solid dispersive and polar components
γ_s	Surface free energy of solid
$F(x)$	Cumulative distribution function of Weibull distribution
$R(x)$	Probability of survival of the variable x, reliability
τ	Interfacial shear force
$\varepsilon(t)$	Strain at a given time
ε_0	Instantaneous strain.
σ_0	Applied stress
E_M , E_K	Elastic modulus of Maxwell Spring and Kelvin Spring
η	Viscosity of dashpot
ε_M	Constant value of creep with time,
ε_K	Earliest saturated value of creep in short time
ε_∞	Long-time value of creep
η_M , η_K	Represents the viscosities of the Maxwell and Kelvin dashpots

$\boldsymbol{D(t)}$	Creep compliance dependent on time
$\boldsymbol{D_0}$, $\boldsymbol{D_e}$	Initial and Equilibrium compliances
$<$, $>$	Less than, Greater than