Development of Sustainable Hybrid Breathing Materials and Their multiple Applications in Polymer Composites

Lu Tong

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Content

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List of Publications

The following peer-reviewed journal articles have been published as a result of the work undertaken as part of this thesis:

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The following peer-reviewed journal articles have been published but not as a part of this thesis:

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List of Abbreviations

AMP	Amplitude of C scan
ANOVA	Analysis of Variance
ASTM	American Society for Testing and Materials
CFRP	Carbon fibre reinforced plastic
DTG	The first derivative of thermogravimetric
EMS	Expandable microspheres
EP	Epoxy
FM	Jute/Polyester fibre mat
FoM	Figure of merit
GFRC	Glass fibre reinforced composites
HNB	Hybrid nonwoven breather
ISO	International Organization for Standardization
JN	Jute nonwoven
LCA	Life cycle assessment
N_2	Nitrogen
NFRC	Natural fibre reinforced composites
PET	Polyester
PLA	Polylactic Acid
PN	Polyester nonwoven
PVA	Polyvinyl alcohol

RF	Ramie fabric
R_{max}	Maximum weight loss rate of thermogravimetric analysis
SEM	Scanning electron microscopy
STL	The sound transmission loss
$T_{5\%}$	The onset temperature of thermogravimetric analysis
TG	Thermogravimetric
TGA	Thermogravimetric analysis
T_{max}	Major weight loss temperature of thermogravimetric analysis
TOF	Time of Flight of C scan
TPS	Transient plane source

Abstract

Sustainable manufacturing is crucial for addressing global challenges such as climate change and resource depletion. This research focuses on sustainable manufacturing in the context of composite materials, which have significant potential in various applications but also generate substantial waste during production. The aim of this study is to develop eco-friendly and sustainable materials for composite manufacturing, reduce waste generation, and explore recycling possibilities.

The research begins by fabricating jute/polyester hybrid breathing materials through the needle-punched method, specifically designed for use as breathers in composite manufacturing. These materials exhibit high permeability even under demanding high-temperature and high-pressure conditions. The feasibility of using these hybrid breathers in the composite manufacturing process is investigated, and their performance is compared with commercially available breathers. Results indicate that the hybrid breathing materials have higher permeability and can be reused multiple times while maintaining a significant portion of their original permeability.

Furthermore, a circular economy strategy is proposed to address the challenge of waste management for end-of-life breathers. The used hybrid nonwoven breathers are recycled and repurposed as reinforcing materials in the

production of new composites. The mechanical and damping properties of these recycled composites are evaluated, demonstrating their potential as alternatives to synthetic materials in semi-structural applications.

To extend the use of renewable resources across a wider range of applications, hybrid breathing materials reinforced syntactic foams with epoxy/expandable microsphere matrix were developed. Jute/polyester fibre mats serve as the structural core, while ramie fabric is used for the outer layer. Adjusting the reinforcement geometry and microsphere content allows for varied density. The natural fibre reinforcements effectively improve the mechanical and sound insulation properties of the foams than the control group. The foam presents potential for lightweight, multi-functional applications.

Overall, this thesis contributes to sustainable composite manufacturing by developing jute/polyester hybrid breathing materials, exploring the reuse of end-of-life breathers, and fabricating fibre-reinforced epoxy foams.

Chapter 1

Introduction

1.1 Background

Sustainability has received increasingly attention in recent years due to the global challenges posed by climate change and resource depletion (He et al., 2021, Pimenov et al., 2022). As shown in Figure 1.1, sustainable manufacturing revolves around the concept of life-cycle thinking, which entails the consideration of environmental and social impacts across the entire lifespan of a product (Sadiku et al., 2023). This approach emphasizes the importance of minimizing resource consumption and reducing environmental impact at every stage of the process, ranging from the procurement of raw materials to manufacturing, product usage, and eventual disposal. By acknowledging these factors, sustainable manufacturing aims to proactively address resource efficiency and environmental responsibility throughout the entire product life cycle (Huang and Badurdeen, 2018).

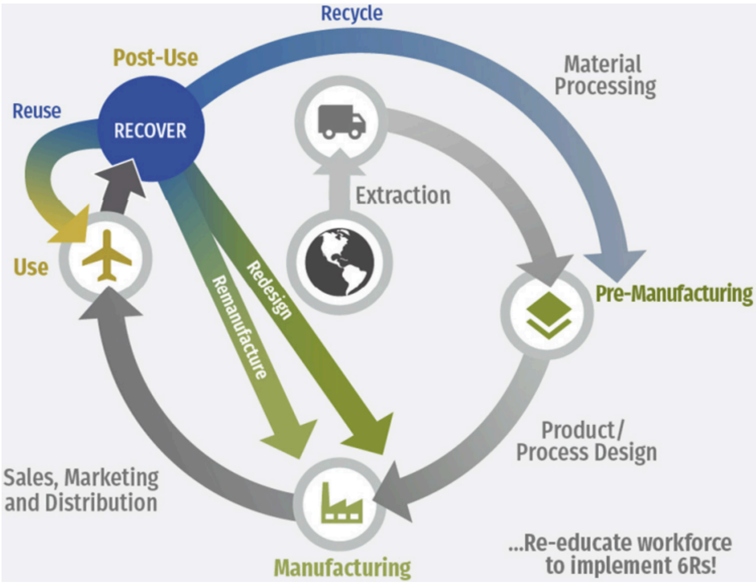


Figure 1.1 Sustainable manufacturing utilizing a closed-loop system (Badurdeen et al., 2017).

Composite materials, with their superior strength-to-weight ratio, corrosion resistance, and design flexibility, hold significant potential across diverse applications like construction, automobile components, electronics, civil infrastructure, and medical implants (Khalid et al., 2021). Nevertheless, the conventional production processes associated with composites drives a large number of wastes, including end-of-life composite products and manufacturing rejects (Witik et al., 2013). Studies have been conducted on the recycling of end-of-life composite materials by using mechanical, thermal, and chemical techniques (Oliveux et al., 2015, Yang et al., 2012a). Insufficient attention has been given to the waste generated during composite manufacturing, such as bagging film, breathers, release films, etc. These consumables do not appear in the final product, and their presence is easily overlooked. Most of these consumables, predominantly made from unsustainable, non-degradable petroleum-based materials, are disposed of through landfill or incineration, leading to environmental pollution and resource wastage.

To reduce the production and use of petroleum-based materials, eco-friendly and sustainable materials are increasingly preferred for the development of new products that they can reduce the carbon footprint (Gonçalves et al., 2021, Soutis et al., 2019). Natural fibres offer a promising alternative to synthetic fibres in virous applications, owing to their renewable and biodegradable nature, lightweight, noble mechanical properties and low cost (Khalid et al., 2021). Besides, the utilization of waste and recycled materials, either individually or in combination with other materials, offers significant benefits for sustainable development (Casadesus et al., 2019, Dominguez-Candela et al., 2021, Kratz et al., 2017).

As the increasing importance of waste management and the growing focus on the circular economy, utilizing production waste, adding value to it, and maximizing material efficiency are imperatives. To make better use of resources, circular economy has been proposed creates a closed-loop system where waste is minimized, and resources are used in a sustainable and efficient manner (Velenturf and Purnell, 2021). As shown in Figure 1.2, the European Union promotes efficient waste management through the five-step waste hierarchy including prevention, preparing for reuse, recycling, recovery, and disposal. Waste should be prevented before it occurs and dumping it in landfills should be the last resort. Although incineration has the advantage to recover energy, it has drawbacks, such as toxic gases, fly ash, and the need to process solid residue (Chen et al., 2019). Reuse and recycling are desirable options to minimize landfill space, incineration pollution, and energy consumption (Smith and Ball, 2012).

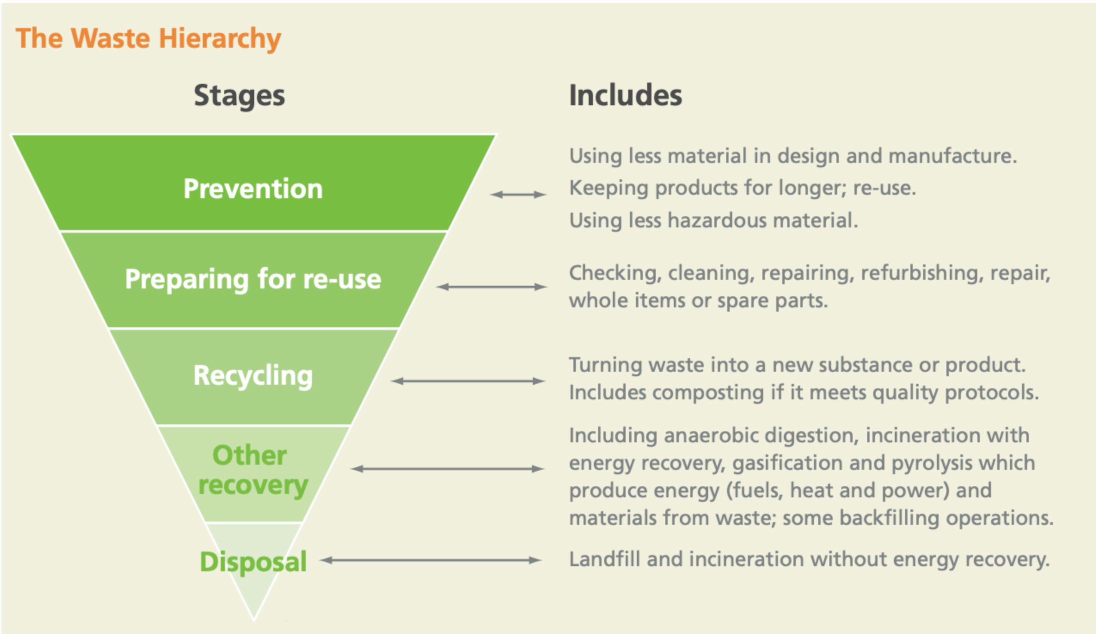


Figure 1.2 EU waste framework (DEFRA (2011) Government Review of Waste Policy in England 2011).

1.2 Aims and objectives

This thesis introduces a research investigation focused on the advancement of sustainable composite manufacturing. In particular, the utilization of jute fibres as a substitute for synthetic nylon or polyester fibres in breathers is explored. Additionally, the study employs recycling and reuse strategies to effectively utilize breathers at the end of their lifecycle, thereby enhancing material use efficiency and minimizing the consumption of petroleum-based resources. The details are as follows:

The research in Chapter 3 aims to investigate evaluate the feasibility and performance of jute/polyester hybrid breathing materials in high-pressure and high-temperature conditions, with a focus on reducing plastic waste and improving permeability of commercial breathers. The research investigates the sensitivity of the hybrid materials to moisture and replicates real-life breather usage in composite manufacturing through humidity and hot press conditions. Morphology characteristics of the breathing materials are observed using scanning electron microscopy. The study also examines moisture absorption, air permeability, and tensile properties of the materials. Finally, the research proposes the recycling use of these breathing materials as composite reinforcement for sustainable practices.

Propose a circular economy strategy for multiple applications of waste hybrid nonwoven breathers to address the challenge of waste management at the end of their life cycle. Recycle and reuse the used breathers in three ways: Directly as reinforcing material; In hybrid combinations with glass fibres for the production of new composites; In hybrid combinations with ramie fibres for the production of new composites. Investigate the thermal, mechanical, and damping properties of the composites incorporating the reused hybrid nonwoven breathers.

Developing and characterizing syntactic foams reinforced with natural fibre for lightweight and multi-functional applications, while exploring the effects of reinforcement geometry and microsphere content on density, mechanical properties, and sound insulation capabilities.

1.3 Structure of thesis

The thesis is organized into the following eight chapters:

Chapter 1 gives a brief introduction to the research background, aims and objectives, and structure of this thesis.

Chapter 2 provides an overview of the literature related to sustainable manufacturing in the context of the present study. It focuses on the challenges in polymer composite manufacturing, sustainable manufacturing methods, and sustainable practices in composite manufacturing. It introduces the application of breathers in composite manufacturing. The influencing parameters and the future requirements for the design of breathers are discussed. Additionally, it discusses the utilization of renewable and sustainable resources in the composite industry, including natural fibres, recycling or waste materials, and biobased polymers. Furthermore, it also introduces composites derived from renewable and sustainable resources, highlighting their potential but also acknowledging the limitations of biobased composites.

Chapter 3 starts the investigation into sustainable composite manufacturing by exploring the development of jute/polyester hybrid breathing materials as alternatives to conventional nylon or polyester breathers. The investigation includes verifying their suitability for high-temperature and high-pressure composite curing conditions, assessing their sensitivity to moisture, and evaluating their permeability and tensile properties. Additionally, the chapter proposes a recycling and reusing approach for the breathing materials as reinforcement in composite applications.

Chapter 4 proposes a circular economy strategy to explore the recycling possibilities of waste hybrid nonwoven breathers. The study begins by utilizing hybrid nonwoven breathers in the autoclave forming of carbon fibre-reinforced plastics. Non-destructive and mechanical tests are conducted to evaluate the quality of the formed composites. Subsequently, the used breathers are recycled and reused in three ways: as reinforcing material directly, in combination with glass fibres, and in combination with ramie fibres for the production of new composites. To estimate the structural damping properties of the composites, the product of specific modulus and loss factor is used as the figure of merit. The chapter concludes that hybrid nonwoven breathers can be effectively employed as replacements for synthetic materials in semi-structural applications.

Chapter 5 presents the fabrication of syntactic foams using expandable microspheres reinforced with natural fibres, specifically jute/polyester fibre mats and ramie fabric outer skins, bonded together with an epoxy resin binder. The chapter highlights the fabrication and characterization of syntactic foams reinforced with natural fibres and demonstrates their potential as high-performance materials with advantages in mechanical properties, sound insulation, and lightweight applications.

Chapter 6 summarises research efforts into preparation and properties of jute/polyester hybrid breathing materials, their use as breathers in composite manufacturing, the reuse of end-of-life breathers, and the fabrication of fibre-reinforced epoxy foams. The chapter emphasizes the potential of hybrid breathing materials in sustainable composite manufacturing and suggesting future research direction.

Chapter 2

Literature Review

2.1 Introduction

Sustainability has become a critical issue in the manufacturing industry, and composite manufacturing is no exception. Composite materials are widely used in various industries due to their unique properties, such as high strength, lightweight, and durability. However, the manufacturing process of composites generates a lot of waste, leading to concerns about sustainability. In this literature review, the current state of research on sustainability manufacturing and related practice is investigated. Breathers, consumables in composite manufacturing, are discussed along with their influencing parameters and the future requirements for sustainable manufacturing. Specifically, the emphasis lies on renewable and sustainable resources such as natural fibres, biobased polymers, and biocomposites. To optimize the use of these materials, composite made from renewable and sustainable resources are reviewed for the fabrication of natural fibre reinforced composites and hybrid composites. We also highlight the challenges and opportunities associated with bio-based composites.

2.2 Sustainable composite manufacturing

2.2.1 Sustainable manufacturing

Sustainable development was first coined in the 1980 World Conservation Strategy report of the International Union for Conservation of Nature (IUCN), which stated that progress must consider social and ecological implications in addition to economic ones (Nature and Fund, 1980). In recent years, the concept of sustainable development has received increasing global attention from both public and business sectors as the environmental impact of human activities, the rising demand for resources, and the urgent need to ensure a sustainable future (Wang et al., 2015). More than one hundred countries around the world have pledged to reduce their carbon emissions and reach carbon neutrality by 2050 or 2060 (Chen, 2021).

The manufacturing of polymer composites has traditionally been associated with significant environmental impacts, including the consumption of natural resources, energy, and water, as well as the generation of waste and greenhouse gas emissions. The increased production and use of composite materials in various industries will inevitably lead to a corresponding increase in the amount of composite scrap that is generated. The disposal of this waste will need to be managed appropriately to prevent environmental harm and to optimize the reuse of the material. The issues with the composite industry are that the use of non-renewable resources, such as fossil fuel base matrixes and fibres; the high energy requirements of the production process; the potential for environmental pollution from the disposal of composite materials. Therefore, sustainable manufacturing is a broad concept that encompasses many aspects, including reducing energy consumption and costs, minimizing environmental impact, ensuring operational safety, and protecting the health of workers (Haapala et al., 2013).

To address these challenges, sustainable manufacturing methods have been developed that aim to minimize the environmental impact of polymer composite production. The circular economy is a viable alternative to the existing linear system, wherein product is produced, used and disposed of. Circular economy practices are integral to delivering various sustainable development goals (Velenturf and Purnell, 2021). Circular economy is a concept aimed at making better use of resources and reducing waste by minimizing resource exploitation and maximizing waste prevention. Circular economy seeks to contribute to sustainability by optimizing social, environmental, technical, and economic values of materials and products in society. The European Union promotes efficient waste management through the five-step waste hierarchy including prevention, preparing for reuse, recycling, recovery, and disposal. Waste should be prevented before it occurs and dumping it in landfills should be the last resort. Although incineration has the advantage to recover energy, it has drawbacks, such as toxic gases, fly ash, and the need to process solid residue (Chen et al., 2019). Reuse and recycling are desirable options to minimize landfill space, incineration pollution, and energy consumption (Smith and Ball, 2012).

In composite industry, sustainable manufacturing refers to the production of materials using economically feasible methods that conserve energy and natural resources, while minimizing detrimental effects on the environment. To adopt the concept of sustainable manufacturing, it is crucial to commence the planning and decision-making procedures in the initial stage of material design and development. This phase is of utmost importance since it determines the majority of a material's impact on sustainability (He et al., 2021). When designing and manufacturing composites for sustainable development, several factors need to be taken into consideration. These factors include the choice of materials, the manufacturing process, and the end-use of the composite.

Sustainable and eco-friendly materials are based on renewable resources, such as plants and agricultural waste, and they are designed to be biodegradable and compostable, which have the potential to revolutionize industries, from automotive to construction, and reduce our reliance on fossil fuels(He et al., 2021). Additionally, incorporating recycled plastic materials into composites reduces dependence on virgin Petro-based materials (Martikka et al., 2019).

2.2.2 Sustainable practice in composite manufacturing

The relationship between manufacturing technologies and sustainability can be positively influenced by government intervention, market demand, and competition pressure (Foo et al., 2019, Bhanot et al., 2017). Global engineering companies need more research and support for sustainable development in the process of composite manufacturing in terms of ecology, economics, politics, and culture (Kireitseu, 2017). In the recent decade, research into sustainable manufacturing is rapidly developing worldwide. It is observed that the mainly part of environmental impacts was significantly influenced by the initial design of a product (Jha, 2015). Manufacturers should recognize the potential positive impact of adopting green supply chain practices on various dimensions of performance and take proactive steps to integrate these practices into their operations (Charanjit et al., 2021).

The concept of Industry 4.0 was introduced by the German government to describe the current trend of automation and data exchange in manufacturing technologies and processes. The objective of Industry 4.0 is to make manufacturing more sustainable by making the entire manufacturing process chain more efficient, visible, and autonomous (Jamwal et al., 2021). Wang et al. developed a composite sustainable manufacturing practice and performance framework to assess the effectiveness of composite sustainable manufacturing practices and their contribution to overall sustainability improvement in Chinese auto-parts companies. The study found that lean, green and social practices can improve sustainability key performance indicators (triple bottom line performance). The study also formulates an optimisation model to help firms select a few sets of practices among the composite sustainable manufacturing practices that would aid them in achieving their targets (Wang et al., 2015). Regression and correlation analysis were used to identify the most important green manufacturing success factors and their impact on organizational performance (Rehman et al., 2016). The artificial neural network simulation can be used to predict and validate green manufacturing models, which are valuable for industries that are looking to improve their environmental, operational, and financial performance through green manufacturing initiatives. The study by K. Madan et al. provides insights into the challenges and opportunities of sustainable manufacturing in India. The findings suggest that promoting 6R concepts, which encompass reduce, reuse, recycle, recover, redesign, and remanufacture, is a promising approach for improving the sustainability of manufacturing in India (Madan Shankar et al., 2017).

In the future development of sustainable composite manufacturing, the shift to a circular economy is essential to address the environmental impact linked with traditional polymer composite production. This transformation aims at resource efficiency and waste reduction. Sustainable manufacturing involves the integration of eco-friendly materials from renewable sources, with careful planning initiated in the material's design phase. Crucially, adherence to the principles of the 6Rs (reduce, reuse, recycle, recover, redesign, and remanufacture) is imperative for optimizing the entire manufacturing process chain, promoting sustainability within the industry.

2.3 Breathers in composite manufacturing

2.3.1 Introduction to breathers

Breather is a nonwoven fabric used in the composite manufacturing processes, especially in the vacuum forming and autoclave forming techniques. Figure 2.1 shows a primary lay-up method of the vacuum forming process. The laminate is placed on a mould and covered with a release film to prevent the breather and laminate adhesion. The breather is placed between the vacuum bag and the release film to maintain the air path throughout the bag to the vacuum source and ensure all air is extracted. It can also cushion any sharp radius and protect the vacuum bag from puncture. The vacuum bag can seal the arrangement around the edges against the mould with sealant tape. A vacuum source is then used to pump out until the vacuum level achieves around 0.95 bar. The sealed package is then placed in a circulating air oven for curing. Higher stresses on the composite part can be applied by an autoclave (Fernández et al., 2003) or hydraulic presses. When the laminate is cured in a vacuum forming process, the compaction pressure is crucial for a good quality composite part (Nugroho et al., 2018). Nonwoven breathing materials with a porous structure allow air and volatile to pass through to maintain continuous pressure on laminate. The soft texture of breathers can extrude excess resin, thus forming good conformity around a composite part.

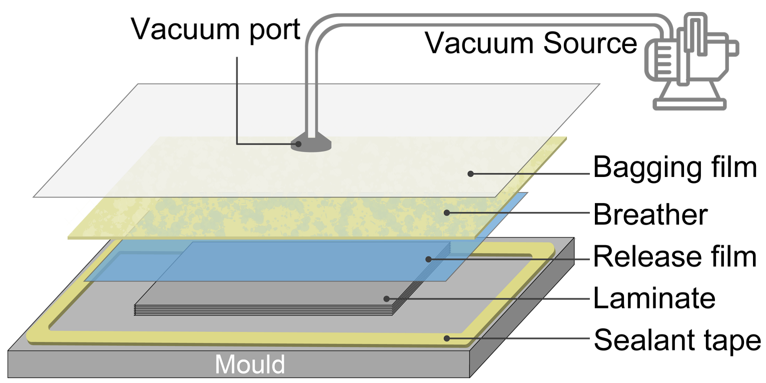


Figure 2.1 Vacuum forming scheme.

Breathers are normally fabricated by needle punching which allows for the production of nonwoven fabrics with varying thicknesses, densities, and surface characteristics. Needle punching is a rapid and cost-efficient manufacturing process, ideal for large-scale production (Zobel and Gries, 2010). Its versatility enables the production of breathers with tailored properties, suitable for various needs in composite manufacturing. The fabrication of needle punching nonwoven fabrics operates on the principle of transforming a section of horizontally oriented fibres or filaments into vertical tufts by employing barbed needles (Pourmohammadi, 2013). The resulting nonwoven fabric exhibits a three-dimensional structure due to the random entanglement of fibres, contributing to its bulkiness and a wide range of pore sizes. Breathers with this structure are advantageous for efficiently discharging gas from the material and absorbing excess resin during composite production.

2.3.2 Considerations for breathers in composite manufacturing

The main consideration for breathers is their air permeability, which plays a crucial role in managing air extraction of composite parts throughout the manufacturing process, ultimately influencing the overall quality of the composite material. Permeability can be influenced by several factors including porosity, bulk density and fibre orientation (Zhu et al., 2015, Dedov et al., 2020). Porosity refers to the empty spaces or voids between the fibres in the fabric. An increase in porosity tends to increase the air permeability. Bulk density is the measure of how closely packed the fibres are in the fabric. Higher bulk density results in lower permeability. The orientation of the fibres in the fabric also affects permeability. Fibres that are predominantly oriented in the transverse direction of the fabric tend to have higher permeability than in the longitudinal direction.

Furthermore, breathers used in various applications are influenced by factors such as fibre composition, areal density, and heat resistance. The specific types of breathers with different fibres vary based on industry standards, manufacturers, and intended applications. Common fibre types used in breathers for composite manufacturing include polyester, polypropylene, nylon, and other synthetic materials. The areal densities of commercially available synthetic fibre breathers range from 100 g/m^2 to 400 g/m^2. Lightweight breathers are suitable for manufacturing simple shape composites and low-pressure curing processes, while heavyweight breathers can be used in higher pressure conditions to minimize trapped volatiles and protect the vacuum bag from puncture by sharp objects. The maximum temperature range of breathers varies from approximately 190 ℃ to 230 ℃, depending on the softening point of the fibres used. In order to cater to even higher processing temperature conditions, glass fibre breathers have been developed for use at temperatures up to 400 ℃.

Drapability, the ability to conform to different shapes, surfaces, and contours (Koncar, 2019), is important for breathers in composite manufacturing, as it directly affects the effectiveness of the curing process and the quality of the final product. Breathers exhibiting exceptional drapability can easily conform and shape themselves to the geometry of intricate or irregular-shaped moulds. As the nonwoven breather is directly processed by single fibres, the drapability of breathers is closely related to the type of fibres, fibre arrangement, flexibility, and resilience (Hearle et al., 1964). Manufacturers may incorporate specific design considerations and techniques to enhance the drapability of breathers, ensuring optimal performance during composite manufacturing processes.

As consumables, breathers serve their purpose during the curing process but are not a part of the final composite product. They are removed after the curing process and rapidly scrapped into industrial waste once used and need to be incinerated or landfilled. The increased focus on sustainable development and the rising costs of waste disposal have encouraged manufacturers to prioritize greener breathers in the composite Manufacturing.

2.4 Renewable and sustainable resources

2.4.1 Natural fibres

Natural fibres have gained significant attention in various industrial and consumer applications as they have been found to possess comparable preference and, in some cases, even better specific properties than synthetic fibres (Wambua et al., 2003). Based on the origin, natural fibres can be divided into plant, animal, and mineral fibres as shown in Figure 2.2 (Kozlowski and

Mackiewicz-Talarczyk, 2020a). Plant fibres come from the bast or stem, seed hair, leaf, or husk of plants that contain cellulose as their main structural component, such as cotton, flax, hemp, jute, abaca, coconut, wheat, cane, grass, and bamboo. Animal fibres include wool, mohair, and silk are primarily composed of protein. Mineral fibres are those that are produced by geological processes, such as asbestos. They were previously used in composites but are now avoided due to health concerns and are banned in many countries. When it comes to strength and stiffness, plant fibres generally offer higher performance compared to readily available animal fibres. This is why plant-based fibres are more suitable for use in composites with structural requirements, and they receive significant attention in research and reviews. The term "natural fibre" in this thesis refers specifically to fibres derived from plants, as the text focuses on principles relevant to plant materials.

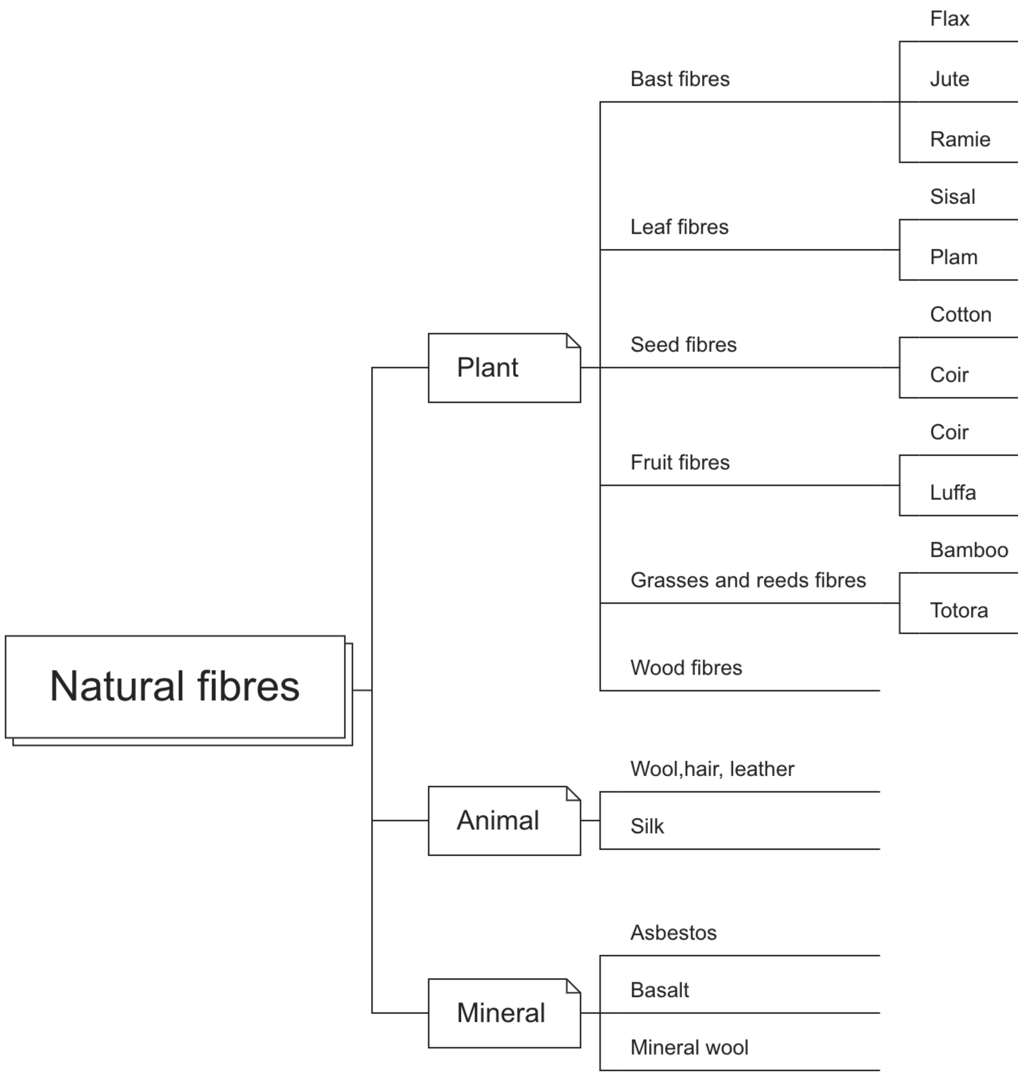


Figure 2.2 Classification of natural fibres.

Table 2.1 presents the advantage and disadvantage of natural fibres. Natural fibres are renewable and biodegradable materials. They have lower density, resulting in a higher specific strength and stiffness compared to synthetic fibre composites (Karimah et al., 2021). Additionally, natural fibres have a very low cost, which reduces the overall manufacturing cost of products (Al-Oqla and Sapuan, 2014). Despite their advantages, natural fibres have drawbacks such as susceptibility to environmental degradation, reduced mechanical properties, and low tolerance to high temperatures during manufacture when compared to synthetic fibre composites (Pickering et al., 2016). Natural fibres have limited thermal stability, with a permitted temperature up to 200°C. Beyond this temperature, the fibre will degrade and shrink. Natural fibres exhibit higher water absorption (24 h) compared to petroleum-based synthetic fibres, with a range of 7% to 20% (Stokke et al., 2013).

Table 2.1 Advantage and disadvantage of natural fibres.

Advantage	Disadvantage
Lower Density	Inconsistent Properties
Biodegradability	Poor Compatibility
Low Cost	Moisture Absorption
Simple Extraction	Low Microbial Resistance
Safe for Workers	Limited Thermal Stability
Non-Corrosive	
High Strength-to-Weight Ratio	
Good Mechanical Properties	

Plant fibres is one of the most widely used natural fibres. They are mainly composed of cellulose, lignin, hemicellulose, pectin, wax and minerals. The cellulose content is a major determinant of the mechanical properties of plant fibres that higher cellulose content translates to better mechanical properties in these fibres (Gibson, 2012). However, several factors such as planting area, extraction method, growth stage, and harvest season of plant fibres can significantly affect their mechanical properties, leading to substantial variation in their properties (Baley et al., 2020, Divya et al., 2022). Table 2.2 list the mechanical properties of some common natural fibres as well as E-glass fibre (Pickering et al., 2016, Nurazzi et al., 2021). In comparison to glass fibre, natural fibres tend to possess lower strength and stiffness. However, in terms of specific properties, natural fibres exhibit superior performance. Specifically, natural fibres can attain a higher specific Young's modulus, and their specific tensile strength can rival that of lower-strength E-glass fibres (Bledzki and Gassan, 1999, Yan et al., 2014, Pickering et al., 2016).

Table 2.2 Mechanical properties of plant fibre and E-glass fibre.

Fibres	Density (g/cm3)	Tensile strength (MPa)	Tensile modulus (GPa)	Elongation at break (%)
Bamboo	1.25	140-230	11-17	-
Ramie	1.5	400-938	44-128	2.0-3.8
Flax	1.5	345-1830	27-80	1.2-3.2
Hemp	1.5	550-1110	58-70	1.6
Jute	1.3-1.5	393-800	10-55	1.5-1.8
Sisal	1.3-1.5	507-855	9.4-28	2.0-2.5
Cotton	1.5-1.6	287-800	5.5-13	3.0-10
Coir	1.2	131-220	4-6	15-30
E-glass	2.5	2000-3000	70	2.5

The choice of suitable fibre reinforcements and matrices is crucial, requiring consideration of their mechanical and physical properties, alignment with application needs, adaptation to environmental conditions, and ensuring cost-effectiveness (Al-Oqla and Hayajneh, 2021, Al-Oqla, 2023). For example, bamboo fibres are known for their high strength and durability, making them ideal for producing furniture and flooring materials. Hemp fibres, on the other hand, have high tensile strength and are commonly used in the production of automotive and construction materials. Cotton fibres are widely used in the textile industry due to their softness and absorbency. Jute fibre stands as the second-largest producer after cotton. It is initially utilized as an industrial raw material for crafting packaging materials supplanted the cultivation of flax and hemp in Europe (Kozlowski and Mackiewicz-Talarczyk, 2020a). Jute fibre is a kind of coarse fibre with high modulus, and low breaking elongation, can be used to fabricate porous nonwoven structure (Kozlowski and Mackiewicz-Talarczyk, 2020b, Al-Oqla and Al-Jarrah, 2021).

2.4.2 Recycling or waste materials as resources

The use of recycled and waste materials is an increasingly important environmental initiative that helps to conserve natural resources, reduce pollution, and release the pressure of landfill space. Agricultural and forest industry waste materials, such as wood chips, wheat straw, stalks, and corncobs, are potentially valuable sources of fibre that can be used as a supplement or direct substitute for wood fibre in the manufacture of composites (Panthapulakkal and Sain, 2007a, Väisänen et al., 2016). Those low-cost lignocellulosic sources can lower manufacturing costs and increase material stiffness. Composites made from cotton, coconut, and sugarcane waste fibres are a promising alternative to traditional materials for acoustic panels (Hassan et al., 2020).

In textile industry, post-industrial textile waste is a significant problem, and much of it is landfilled due to a lack of recycling infrastructure. Studies identified three ways to reduce textile waste, and models for industrial textile waste management was created that integrates analysis of fabric composition, production processes, and material flows (Rapsikevičienė et al., 2019, Dissanayake and Weerasinghe, 2020). Waste materials obtained from the fabric industry of denim and jute have the potential to be used as sound absorbers, with their acoustical properties comparable to commercial Glasspool (Raj et al., 2020). Projections indicate a significant increase in wind turbine blade waste production on a global scale in the coming years. Waste wind turbine blades can be repurposed for a variety of uses, including architectural applications, consumer goods, and industrial filler materials (Jensen and Skelton, 2018, Joustra et al., 2021).

2.4.3 Biobased polymers

The issue of "white pollution" caused by discarded plastics has become increasingly severe in recent years, leading to global efforts to develop green industries that utilize clean and renewable bio-based polymers as an alternative to petroleum-based plastics. Plastic can come from either renewable or non-renewable resources, and they can be categorized as biodegradable or non-biodegradable. Biobased plastics are made only from renewable materials, while biodegradable plastics can contain a mix of renewable and non-renewable materials. Figure 2.3 outlines the classification of bioplastics, which can be categorized into three types: biodegradable and bio-based, biodegradable and fossil-based, and non-entirely or partly bio-based and biodegradable plastics (Sidek et al., 2019).

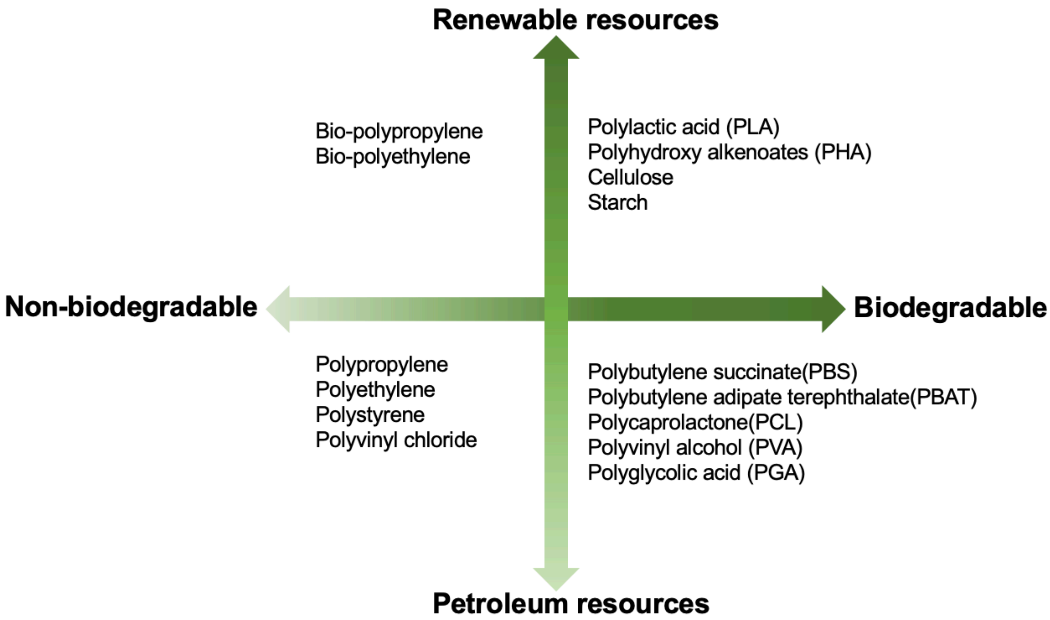


Figure 2.3 Classification of bioplastic on the basis of resources and biological degradation.

Polymers derived from sustainable sources can be categorized into three main types: polysaccharides-based bioplastics, protein-based bioplastics, and microbial-based bioplastics. Polysaccharide-based biopolymers are cellulose, chitin and chitosan, starch, hyaluronic acid, dextrin, alginate, carrageenan and various gums (Aaliya et al., 2021). Starch-based plastics are the most prevalent and can be produced easily with simple tools and can be tailored to specific needs by adding additives like glycerine and sorbitol. Cellulose-based plastics, such as cellulose acetate, can also be used but are more expensive and require extensive modifications. Protein-based plastics, derived from soy proteins and other sources, have been used since the early 1900s but can be costly and water-sensitive. Blending with existing biodegradable polyesters can overcome these challenges, and other proteins like wheat gluten and casein show promise for use in bioplastics.

On the basis of reaction to changes in temperature, biopolymer matrices can be categorized into thermosets and thermoplastics. Polypropene and polylactic acid are two examples of thermoplastic resin in plant fibre reinforced bio-composites uses (Hao et al., 2013, Siakeng et al., 2018). Biobased thermosets have gained significant attention in recent times for the development of components, surpassing biobased thermoplastics (George et al., 2020). The reason behind this shift in focus is that thermosets, such as epoxy resin, polyurethane, and unsaturated polyester, cannot be melted or recycled due to the strong cross-link bonds present in their polymer chains. Consequently, thermoset bioplastics that can rapidly decompose after being discarded are better suited to meet the requirements of environmental protection.

The development of biodegradable plastic materials presents a viable alternative to reduce the amount of non-degradable waste sent to landfills, while fostering a sustainable and pollution-free environment that is of paramount importance to both consumers and industries. The use of bioplastics is gaining prominence in diverse industrial applications, especially in the areas of medicine, agriculture, and product packaging. Biodegradable plastics have become increasingly popular in the medical field for various applications, such as surgical implants, sutures, and drug delivery systems. Poly-(glycolic acid), poly(L-lactic acid), and their copolymers are commonly used for surgical sutures due to their strength and flexibility (Tian et al., 2012). Biopolymer bone fixation implants dissolve entirely, promoting undisturbed dynamic bone healing without requiring additional surgery. Biopolymer nanoparticles offer enhanced biocompatibility and absorption for drug delivery systems. Commonly used biopolymers for drug delivery include poly-(hydroxy acid) and poly-(ortho ester). Polyurethanes are also commonly used for grafting scaffolds that represent artificial blood vessels due to their wear-resistant, flexible, and blood-compatible properties.

In the agricultural domain, the use of biopolymers in mulches has several benefits, such as promoting plant growth, hindering the formation of weeds, maintaining optimum soil temperature, and reducing the need for water and soil nutrients. The biopolymer films made from specific materials, such as polyvinyl chloride, low-density polyethylene, polyvinyl alcohol, or polybutylene, are used for mulch manufacturing, and these films are designed to undergo degradation only when the crop-growing period is completed. The same trait is observed in polycaprolactone, which is used to make biodegradable agricultural plant containers (Averous, 2004).

Polysaccharide-based biopolymers such as starch, cellulose, and copolymers of polyesters and polyethylene are being used to develop potential packaging films. Natural fillers like starch are also blended into synthetic polymers to enhance the degradation properties of packaging laminates. Biopolymer-based packaging has found applications in various sectors such as groceries/compost bags, storage containers, disposable plates, cups, cutlery, and textiles.

Polylactic acid (PLA) is a biodegradable thermoplastic that is derived from renewable resources such as corn starch, sugarcane, or tapioca roots. It is a type of polyester that is produced by the polymerization of lactic acid, which is itself derived from the fermentation of sugars. The exceptional mechanical and barrier characteristics that make it an excellent choice for manufacturing a wide range of biomaterials for use in industries such as textiles, packaging, biomedical, and automotive manufacturing (Farah et al., 2016). Research has shown that PLA composites reinforced with natural fibres have demonstrated significant advantages, including renewability, recyclability, excellent durability, strong processability, high-specific resilience, and compostability (Getme and Patel, 2020).

The development of sustainable polymers aims to create commercial goods that are environmentally friendly and can replace traditional petroleum-based materials. A significant challenge in this endeavor is the identification of platform chemicals or building blocks that can be easily prepared from abundant feedstocks without competing for resources with food crops or causing ecological disruptions (Zhu et al., 2016).

2.5 Composite derived from renewable and sustainable resources

2.5.1 Bio-composites

Composites are materials made by combining two or more constituent materials that have different properties, resulting in a heterogeneous structural material mixture. One of the key advantages of composites is that they can be engineered to have specific properties based on the requirements of the application. By selecting and combining different constituent materials, composites can exhibit properties such as such as strength, stiffness, durability, or light weight, making them a valuable and versatile class of materials for various applications. Bio-composites are a specific type of composite material that incorporates naturally derived constituents, either in the form of fibres or resins. Figure 2.4 illustrates the manufacturing of bio-composites from raw materials to the final composite products (Mohanty et al., 2018). Basing on the fibres and matrix systems used, bio-composites can be categorized into three types, namely partially biobased, fully biobased, and hybrid bio-composites. The resulting composites can either be thermoplastic or thermoset depending on the type of polymer resin selected. Various processing techniques, such as compounding, mixing, extrusion, injection moulding, compression moulding, and resin transfer moulding, have been extensively developed and proven successful in producing composites with consistent quality. However, to cater to the demands of new application areas, there is a need for intensive research on innovative technologies and process solutions to achieve high-strength engineering composites (Faruk et al., 2012).

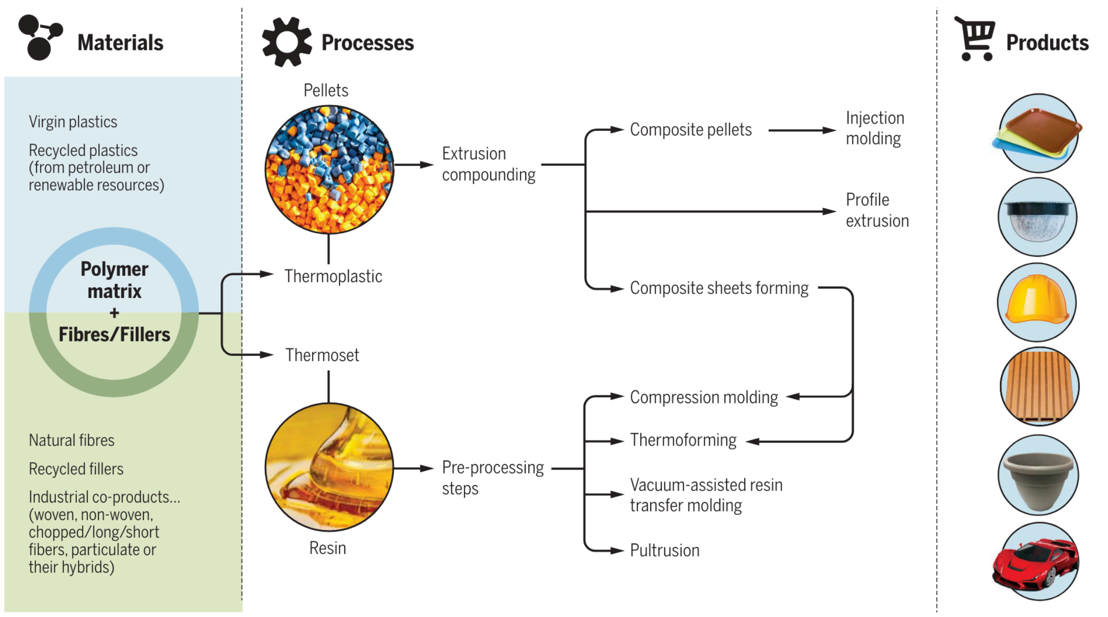


Figure 2.4 Scheme of Bio-composite products manufacturing (Mohanty et al., 2018).

2.5.2 Natural fibre reinforced composites

Natural fibre reinforced composites (NFRCs) are a type of bio-composite material that replaces traditional synthetic reinforcement fibres like glass and carbon with natural fibres. The environmentally friendly, economic, and lightweight advantages of natural fibres have demonstrated their ability to replace costly synthetic fibres over time in composites (Wambua et al., 2003). NFRC have been found to possess specific properties that are, in some cases, even better than those of glass (Wambua et al., 2003). Shah et al. conducted a comparison of the tensile properties between natural fibre reinforced composites and glass fibre composites as shown in Figure 2.5 (Shah, 2014). It states that NFRCs exhibit tensile modulus values that are comparable to those of glass fibre reinforced composites (GFRCs), but their strength is noticeably lower than that of GFRCs. However, when taking density into account, NFRCs demonstrate a more favourable comparison by achieving higher specific stiffnesses than GFRPs, although GFRPs still maintain the highest specific strengths.

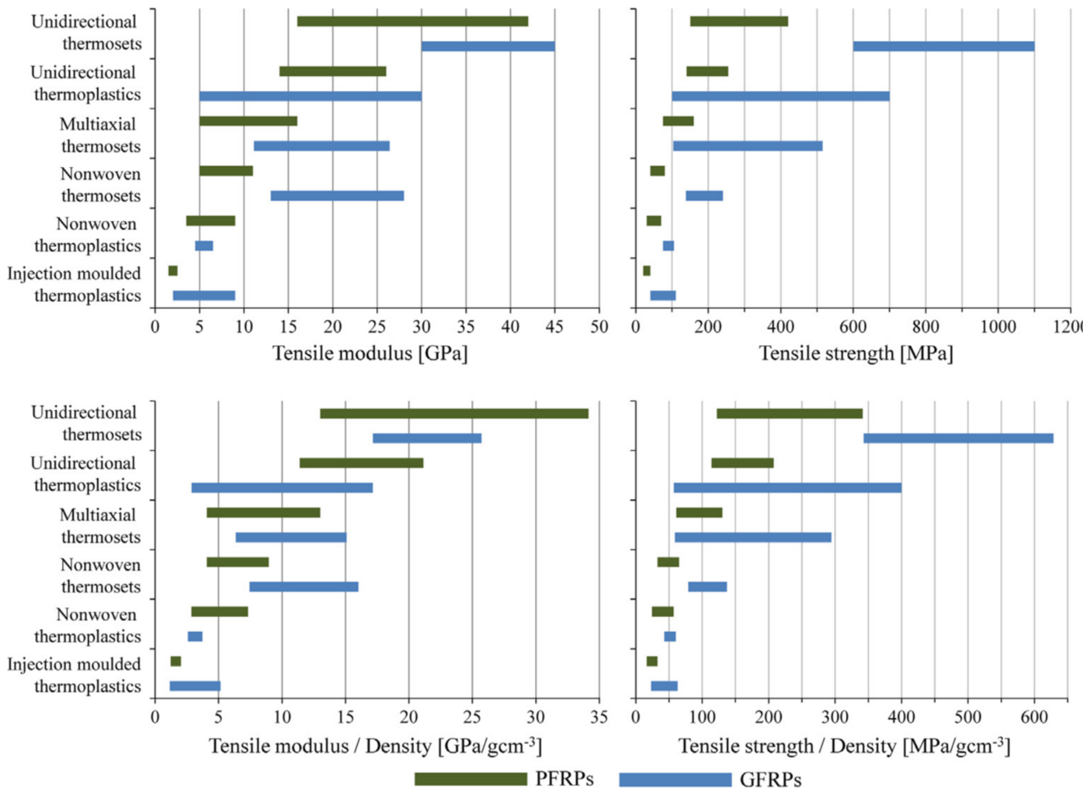


Figure 2.5 Comparison of the tensile properties between natural fibre and glass fibre composites (Shah, 2014).

Several factors can greatly influence the mechanical properties of natural fibre composites, including fibre orientation, fibre volume fraction, treatment method, and physical properties of the fibres (Shalwan and Yousif, 2013, Pickering et al., 2016). Composites exhibit the best mechanical properties when the fibres are aligned parallel to the direction of the applied load. This alignment allows for effective load transfer and optimal utilization of the fibre’s properties(Shibata et al., 2008). Similarly, the structure of reinforcement plays a crucial role in determining the mechanical performance of composites. Woven structures, which involve fibres interlaced in a regular pattern, offer improved tensile strength due to the efficient load transfer along the aligned fibre directions. On the other hand, mat structures, where fibres are randomly distributed, tend to exhibit lower tensile strength (Ratim et al., 2012). As the fibres provide a stiff, reinforcing skeleton for the matrix, the stiffness of a composite material is directly proportional to the volume fraction of fibres in the matrix. It is note that there is an upper limit to the amount of fibres that can be incorporated. As the fibre volume fraction approaches the upper limit, the amount of matrix material may become insufficient to fully surround the fibres, limiting further increases in stiffness and strength (Shibata et al., 2008).

Various studies have explored the impact of chemical and physical treatments on the mechanical properties of NFRCs, emphasizing the need for improved composite materials without porosity and brittleness, which depend on filler-resin compatibility (Adekomaya and Majozi, 2019, Reddy et al., 2020, Kalia et al., 2009). To promote the expansion of research on sustainable treatment approaches, particularly with a focus on environmentally friendly options, it is essential to sustain efforts in direction of matrix viscosity and the development of mathematical equations to estimate the presence of surrounding air in the resin (Adekomaya and Majozi, 2019).

Moisture absorption is one of the most undesirable factors in NFRCs because moisture can cause the fibres to swell, which can create voids and micro gaps between the fibres and the matrix. These defects reduce the interfacial adhesion between the fibre and matrix and further influence the strength, durability, and performance of composites (Moudood et al., 2019, Dayo et al., 2020). In the initial stage, the composites show a rapid increase in water absorption until they reach a saturation point, after which there is no further increase in water uptake (Dhakal et al., 2007). The water absorption of NFRCs poses a significant limitation for their application, as plant fibres tend to absorb more water in comparison to synthetic fibres.

2.5.3 Hybrid composites

With the need to develop new materials with improved strength, damping, and temperature resistance, the concept of “hybridization” is widely implemented. An overview of the main results of natural fibre hybrid composite research is provided in Table 2.3. The synergistic combination of natural and synthetic fibres mitigates the disadvantages of a homogeneous fibre type (Reddy et al., 2018, Bachmann et al., 2018). High-performance synthetic fibres can be used in composites to boost mechanical and thermal properties as well as reduce some of the variability associated with plant fibres. Natural fibre hybrid composites improve damping and reduce the density of the final laminate (Ashworth et al., 2016). As a result of hybridization, balanced thermal, mechanical, damping as well as density of composites can be achieved, attracting more applications in infrastructure, automotive, sports, furniture, etc. (Sapuan et al., 2020, Shah et al., 2019).

Table 2.3 Summary of hybrid composites, their brief descriptions and key findings.

Reinforcement	Matrix	Key findings	Ref.
Jute fibre/ Pineapple leaf fibre/ Glass fibre	Epoxy resin	The higher fibre content in composites with Jute, Pineapple leaf fibre and Glass fibre hybrid fibres increased tensile and flexural properties	(Reddy et al., 2018)
Woven flax//E-glass fibres	Epoxy resin	The hybridization of flax and E-glass fibres increased damping. However, the tensile properties were compromised.	(Cihan et al., 2019)
Sugar palm fibre /woven E-glass fibre	Unsaturated polyester resin	By combining glass fibre with treated sugar palm fibre, hybrid composites for structural applications have improved thermal properties.	(Nurazzi et al., 2020)
Jute fibre/Carbon fibre	Low viscosity epoxy resin	The tensile modulus of hybrid composite was almost twice that of jute fibre composite. Loss factors for jute fibre composite and hybrid composite were considerably higher than carbon fibre reinforced composite.	(Ashworth et al., 2016)
Kenaf/Carbon Fibre	Epoxy resin	The thermal stability of plain woven kenaf hybrid composites is higher than that of the satin design. The addition of carbon fibre improves the thermal stability of the composite.	(Aisyah et al., 2019)

Flax fibres /Recycled carbon fibre	Epoxy resin	Hybrid materials made from flax and recycled carbon fibre allow the characteristics of composites to be customized.	(Bachmann et al., 2018)

2.5.4 Applications and limitations of bio-based composites

Bio-composite have gained significant popularity in several industries, particularly in the automotive, aviation, and construction sectors. The automotive industry has made notable advancements in adopting bio-composites as alternatives to synthetic composites on a significant scale. These sustainable composites are already being used in a wide range of auto parts, such as trim panels, seat backs, packaging trays, spare wheel covers, headliners, dashboards, and air baffle components (Akampumuza et al., 2017). The use of bio-composites allows the automotive industry to reduce its environmental impact by utilizing low-cost alternatives that incorporate non-traditional fibres, fillers, and matrix systems. Some of these alternative materials include "waste" agro-forestry co-products, waste rubber, cork, and recycled waste materials (Malnati, 2018, Pegoretti et al., 2014). By using these materials, the automotive industry can significantly reduce its environmental impact. Natural fibre composites have been extensively researched and both reinforcement and matrices are being substituted with sustainable materials (Du et al., 2014). In addition to the substitution of traditional reinforcement and matrices with sustainable materials, there is ongoing research on the use of plastics derived from renewable resources, such as PLA and Bio-PA, for automotive applications. Furthermore, there is increasing attention being paid to the incorporation of recycled content with virgin plastics in composite material fabrication. These efforts aim to promote a more sustainable future for the industry.

However, the wider acceptance of bio-composite materials in different applications depends on various factors. One key consideration is moisture repellence, as plant fibres used in bio-composites tend to absorb moisture from the surrounding environment, leading to dimensional instability. This can pose challenges in certain applications. Additionally, plant fibres, including biofibres, have low thermal stability and tend to degrade at temperatures around 200°C, which limits their usage in high-temperature applications that require materials with good thermal stability, such as engineering plastics. The incompatibility of certain plant fibres with specific polymers can also limit the range of materials that can be used to develop bio-composites.

To overcome these challenges and ensure the wider acceptance of bio-composite materials in a wide range of applications, it is crucial to continue investing in the development of sustainable composites. Research efforts should focus on addressing the issues of moisture repellence, structural stability, and flame-retardant properties. Additionally, exploring new fibre treatment and modification techniques, as well as investigating the compatibility of different plant fibres with various polymers, can expand the range of materials available for bio-composite development. (Loureiro and Esteves, 2019).

The management of end-of-life situations for bio-composites and other polymer matrix composites is gaining attention. While a circular economy approach that emphasizes reusing and recycling plastics is being promoted, thermal recycling (i.e., incineration) is also considered as a solution (Aaliya et al., 2021). Circular economy approach that emphasizes reusing plastic composites and plastics in general can be achieved through feedstock recycling or cascaded use. In addition to established methods such as collection and remoulding for thermoplastics, these strategies can be used to maximize the use of plastics and minimize waste. Recycling thermoset polymers poses a greater challenge compared to other types of polymers. The vast array of composites used makes it difficult to recycle these materials in a meaningful way. However, standardization could be a viable solution to increase recycling rates.

It is important to note that the term "bio-based" does not necessarily mean "sustainable" in the case of composites. Sustainability encompasses various aspects such as the production process, materials used, and end-of-life treatment applied. Therefore, a comprehensive approach that considers the entire life cycle of bio-composites is essential to promote a truly sustainable future for the automotive industry.

Chapter 3

Development of Hybrid Breathing Materials for Sustainable Composite Manufacturing

3.1 Overview

In order to reduce the plastic waste generated from the composite manufacturing process and improve the permeability of commercial breathers in high-pressure and high-temperature conditions, jute/polyester hybrid breathing materials were fabricated by the needle-punched method. The thermogravimetric analysis and the related thermal properties of hybrid breathing materials were carried out to verify the feasibility of materials used in the high-temperature environment. Breathing materials were then exposed to humidity atmosphere and hot press conditions respectively. These conditions were chosen to investigate the effect of moisture on breathing materials and replicate the using conditions of breathers in the composite manufacturing process. The scanning electron microscope was used to observe the morphology characteristics of breathing materials before and after use. The moisture absorption, air permeability and tensile properties of breathing materials were evaluated and compared with the unconditioned specimens. The results showed that hybrid breathing materials were sensitive to moisture due to the hydrophilic groups in jute fibre. The permeability of hybrid breathing materials was higher than commercially available breathers at a similar areal weight. The permeability of breathing materials increased with the increase of porosity. Humidity and hot press processing conditions influenced the permeability due to the change of porosity. The hybrid breathing materials had a higher permeability retention rate. They could be reused at least three times, and their permeability could be maintained above 56% of the original data. Higher jute fibre content resulted in higher tensile strength. The tensile strength of the hybrid breathing material was higher in wet conditions. The reuse of the breathing material led to an increase in tensile strength. Finally, the recycling use of breathing materials as composite reinforcement was proposed.

3.2 Introduction

Breathers are made of synthetic fibres such as polyester and nylon fibres. They are designed into different sizes, thicknesses, and areal weights to meet the temperature and pressure requirements in the composite manufacturing process. As consumables, breathers are not a part of the final composite product. They are removed after the curing process and rapidly scrapped into industrial waste once used and need to be incinerated or landfilled. However, the increase in landfill tax and the limitation on the emission of pollutants from waste incineration have led to increased waste disposal costs.

The output of composite materials is expected to grow continuously in newer aircraft, automotive, marine, wind energy, nuclear engineering and sports industries (Mrazova, 2013, Akampumuza et al., 2017). The process of composite manufacturing will generate approximately 30% - 50% of waste relative to the initial quantity (Rybicka et al., 2015). Almost all plastic products are not biodegradable, and they may cause the impact of micro plastics in the natural environment. Short-life plastics are not sustainable given limited landfill capacity and declining fossil fuel reserves (Thompson et al., 2009, Geyer et al., 2017). Composite manufacturers are facing the pressure of government regulation and the concern of the environment from society. They have to take environmental issues seriously. It is necessary to reduce the plastic waste generated from composite production and develop a sustainable composite manufacturing mode. The concept of sustainable production is also in line with various countries and companies to achieve the goal of carbon neutrality in 2050 (Otto et al., 2020).

In addition to the environmental impact, commercial polyester breathers cannot channel air under high-temperature and high-pressure conditions. Although polyester fibre has a high melting point, its soften point is relatively low. When a breather works at a temperature exceeding 120°C, fibres are likely to fuse together. The pressure forces breather to compress into a paper-like sheet and restricts airflow.

As synthetic fibre products have the above shortcomings, natural fibre can be used as an environmentally friendly substitute for synthetic fibre. Generally, natural fibre requires less energy for production, has low carbon dioxide emissions, and has performance comparable to synthetic fibres (Sreekumar and Thomas, 2008, Mohanty et al., 2018, El Nemr, 2012). Moreover, natural fibre will not soften at high temperatures, which makes up for the weakness of polyester breathers with poor air permeability under high-temperature and high-pressure conditions. Jute fibre is one of the cheapest nature fibres in the world. The production of jute fibre products will have a considerable price advantage compared with synthetic fibre products. The jute plant can increase soil fertility and consume carbon dioxide in the air by its high carbon dioxide assimilation rate (El Nemr, 2012). However, the utilization of jute fibre can introduce some challenges. The main disadvantages are the susceptibility to moisture uptake(Sreekala and Thomas, 2003) and poor thermal properties (Vaisanen et al., 2017). Those properties need to be paid attention to and reasonably avoided.

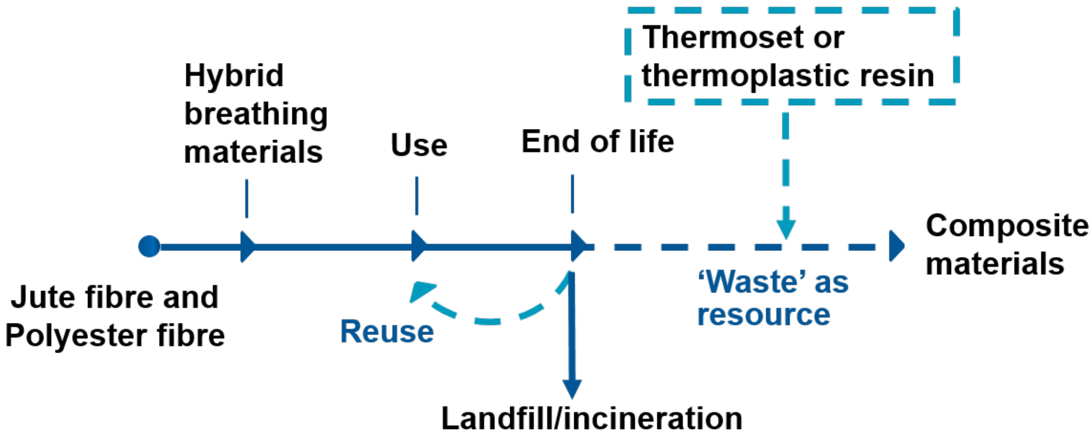


Figure 3.1 Flow chart of sustainable composite manufacturing.

For the goal of sustainable manufacturing, products need to be derived from renewable resources, having recycling capability and biodegradable properties (Mohanty et al., 2002). Figure 3.1 presents an ideal way to reuse and recycle breathing materials in sustainable composite manufacturing. The solid line shows the currently use process of commercial breathers, and the dashed line represents a sustainable production model. Reuse of breathing materials allows composite materials to be produced with fewer consumables, reducing the production costs and producing less waste. The end-of-life breathing materials can be combined with the thermoset or thermoplastic resin to prepare composite materials and to be further used in the secondary structure.

In this work, jute/polyester hybrid breathing materials were prepared by the needle-punched method to replace the traditional synthetic fibre breathers in the composites manufacturing process. The thermal properties were also investigated to understand the feasibility of applying materials in high-temperature conditions. The influence of humidity and hot press on the breathing materials was investigated by studying air permeability and tensile property before and after the sample conditioning.

3.3 Materials and methods

3.3.1 Materials

Jute fibre and polyester fibre with the properties shown in Table 3.1 were used to prepare hybrid breathing materials. Suppliers furnished data on diameter, strength, and density for fibres. The data of diameter, strength, and density were provided by suppliers. Jute fibre was supplied by Yunfeng Textile Co., Ltd, Suixi, China. The polyester fibre was supplied by Jiaxing Chemical Fibre Products Co., Ltd, Xianghe, China. Fibres from suppliers were used directly without treatment.

Table 3.1 Properties of jute fibre and polyester fibre.

Raw Materials	Length (mm)	Diameter (μm)	Strength (MPa)	Density (g/cm^3)
Jute Fibre	60	67.10	449	1.48
Polyester fibre	64	38.39	76	1.39

3.3.2 Preparation of jute/polyester hybrid breathing materials

The manufacture of jute/polyester hybrid breathing materials involved four steps: opening, carding, over-lapping, and needle punching. Jute fibre was manually opened and mixed with polyester fibre at 40wt% and 60wt% fibre content and fed together into the nonwoven machine system, consisting of carding, over-lapping, and needle punching sections. The fibre mixture was broken up into single fibre in the opening section. The individual fibre in the fibre mixture was further combed to be parallel, and the isotropy of fibre was reduced significantly and output in the form of fibre web at the end of the carding section. The web was then transferred to the over-lapping section, where the web was cross lapped to equalize the lamination in perpendicular and longitudinal directions. The over-lapped lamination was subsequently delivered to the needle punching section, where punching depth, punching density and punching time was set at 7-8mm, 70 punches/cm^2 and two times respectively. The preparation process and brief description can be found in Figure 3.2.

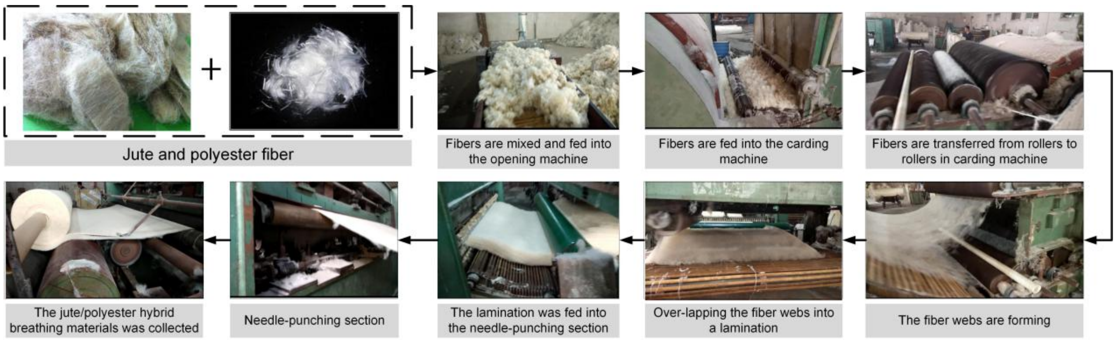


Figure 3.2 Nonwoven processing line.

Table 3.2 Parameters of hybrid breathing materials and commercial breathers.

Sample ID	Areal Weight (g/m^2)	Weight composition (%)		Thickness (mm)
		Jute	Polyester	
JN125	125 ± 5	40	60	3.36 ± 0.08
JN315	315 ± 8	40	60	5.89 ± 0.07
JN340	340 ± 6	60	40	6.05 ± 0.14

PN150	150 ± 3	-	100	3.02 ± 0.05
PN340	340 ± 4	-	100	4.92 ± 0.06

Table 3.2 shows the physical properties of prepared hybrid breathing materials and control samples. The control samples were polyester breathers, and they were bought from Sino Composite Co., Ltd in Beijing, China. The breathing materials containing jute fibre were named jute nonwoven fabric (JN). The two polyester breathers were named polyester nonwoven fabric (PN). Different JN and PN breathing materials were identified by their areal weight. According to the areal weight, breathing materials were divided into two groups. The lightweight group includes JN125 and PN150. The heavyweight group contains JN315, JN340 and PN340.

3.3.3 Sample conditioning

3.3.3.1 Humidity

The raw fibres, prepared breathing materials and polyester breathers were first placed in an electric blast drying oven at 103 °C until the weight is constant, recording the dry weight. Then the dried samples were exposed to weathering at an 80% relative humidity and 20 °C atmosphere in a weather chamber. The weight of the specimen was measured after 24 hours.

3.3.3.2 Hot press aging

Specimens of breathing materials were applied in four different temperature and pressure conditions (Table 3.3) to simulate the using condition of breathers in the production of composite. They were placed on a mould and sealed under a vacuum bag film by the sealant tape. The pressure of 1bar was applied to the sample by a vacuum pump (the actual pressure ranged from 0.95 to 1 bar). The temperature was controlled by an electric blast drying oven. The pressure of 7 bar and the relative temperature were achieved by an autoclave, as shown in Figure 3.3. The processing time for each experiment was two hours. Before each test, breathing materials were placed in an electric blast drying oven at 80°C for at least two hours.

Table 3.3 Hot press aging conditions.

Conditions	Pressure (Bar)	Temperature (°C)	Time duration (h)
1	1	130	2h
2	1	180	2h
3	7	130	2h
4	7	180	2h

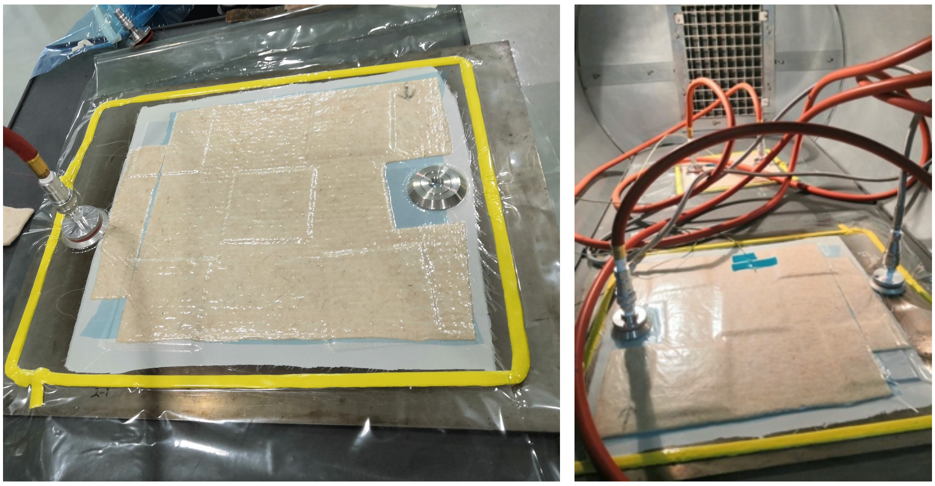


Figure 3.3 Application of JN315 in the autoclave.

3.3.4 Characterization and Measurement

The moisture absorption was calculated by the weight difference between the saturated water absorption specimen and dry specimen with the following equation:

$$Moisture\ absorption = \frac{M_s - M_d}{M_d} \times 100 \quad (3.1)$$

Where M_s and M_d are the mass of samples in saturated moisture absorption and dry conditions respectively.

The thermogravimetric (TG) analysis of fibres and breathing materials was analysed by an SDT Q600 instrument. Specimens with around 10mg were placed in an alumina crucible. They were heated to 800 °C at a rate of 10 °C / min, under a 50ml / min nitrogen flow. In order to eliminate the influence of moisture on materials, the test program is set to record data after the quality of the specimen was stable at 100 °C. Thermal conductivity, thermal diffusivity and specific heat of breathers were tested by National Quality Supervision and Inspection Centre. The test utilized the transient plane source (TPS) technique employing a double-sided hot disc sensor which is a combined heat source and resistance thermometer. In this method, a consistent power is applied to the sensor, and the temperature within the sensor is continuously monitored. By analysing the temperature evolution within the sensor, the thermal properties of the sample can be accurately calculated. Thermal conductivity is defined as ability of material to conduct heat. It is quantified in watts per square meter of surface area, considering a temperature gradient of one Kelvin per unit thickness of one meter. Thermal diffusivity is the rate of temperature spread through a material. Specific heat quantifies the energy needed to elevate the temperature of one cubic meter of a material by one Kelvin.

The permeability of breathing materials was measured as specified in ISO 9273(15) by the YG461E digital fabric air permeability meter. The air pressure drop was set as 100Pa. The porosity of breathing materials was calculated by the following equation:

$$Porosity = 1 - \frac{D_b}{D_f} \tag{3.2}$$

Where D_b and D_f are the density of breathing material and the average fibre density respectively. D_f is determined by the rule of mixture, which involves multiplying the density of each type of fibre by its corresponding volume fraction and then adding up these individual values.

The areal density was determined following ISO 9073(1), computed as the ratio of mass to area. The size of test samples was 500 cm^2, and a total of 5 samples were examined. Variability of areal densities arises from challenges in precisely controlling weight during manufacturing processes. The density of breathing material is calculated by dividing the areal weight by the thickness. The thickness of breathing materials was measured following ISO 9073(2).

Tensile strength and strain of jute fibre in dry and wet conditions were performed according to ASTM D3822 with 12mm gauge length and at a displacement rate of 1mm/minute. The fibre was mounted between a pair of plastic tabs with V alignment features and secured by a UV-curing adhesive. The diameter of each fibre was measured by the laser scan micrometre LSM 6200 at three points along the gauge length before proceeding with the test. The loading assembly ALS1500 was used to determine the tensile strength and strain of jute fibre.

The tensile tests of hybrid breathing materials in the machine direction were carried out using an MTS E45 universal testing machine according to ISO 9273(3) at an extension rate of 100mm/min. The width of the specimen was 50mm, and the gauge length was 200mm.

The morphology of breathing materials was determined by a Zeiss Sigma V.P. scanning electron microscope (SEM). Samples were pre-coated with gold before being observed.

3.4 Results and discussion

3.4.1 Morphology of hybrid breathing materials

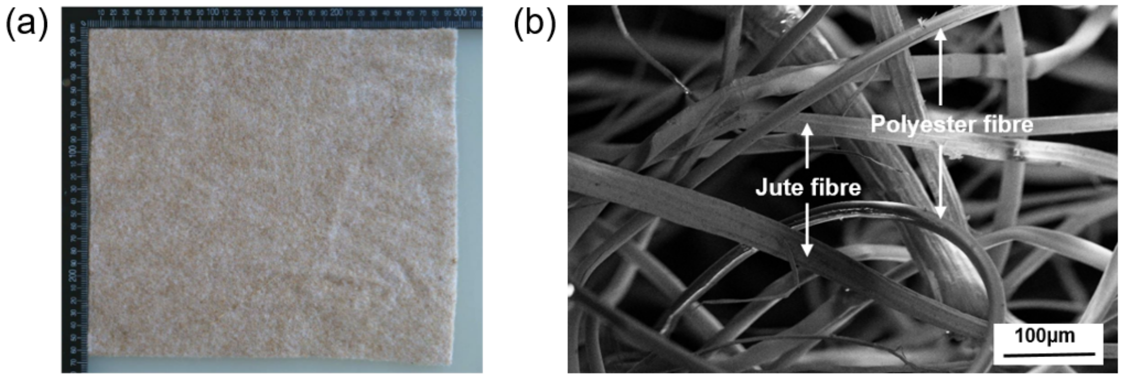


Figure 3.4 Digital image (a) and the SEM image (b) of JN315.

The image of JN315 is shown in Figure 3.4(a), which is a light brown and white nonwoven fabric. It can be observed from the SEM image in Figure 3.4(b) that fibres in the breathing material have a random arrangement, and they are entangled with each other to form a porous structure. Compared with polyester fibre, jute fibre has a larger diameter, and the diameter of the different jute fibres is not consistent. The surface of the polyester fibre is smooth and even, while the surface of jute fibre is relatively rough, with longitudinal lines in the fibre length direction.

3.4.2 Moisture absorption of hybrid breathing materials

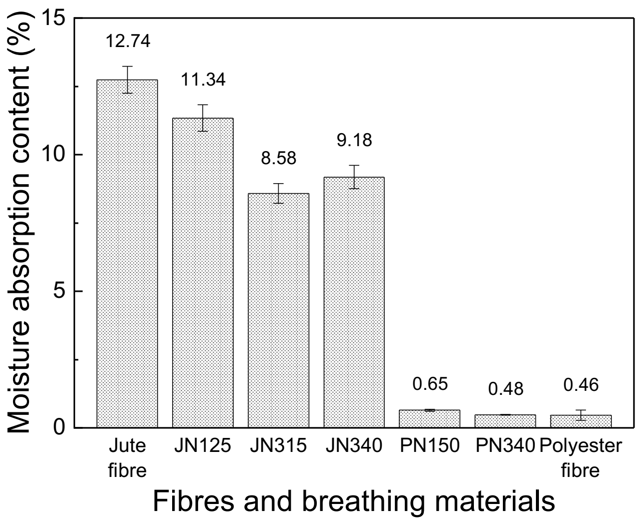


Figure 3.5 Saturated water absorption of fibres and breathing materials.

The moisture absorption of fibres and breathing materials was tested to show the hydrophilic character of different materials. All the hybrid breathing materials are sensitive to moisture. This difference can be attributed to the high moisture uptake of jute fibre with 12.74%. There are many hydroxyl groups on the structure of jute fibre, which makes jute fibres hydrophilic (Doan et al., 2006). A high moisture absorption content of hybrid breathing materials may cause application issues and needs to be dried before use.

When the fibre content is the same, the moisture uptake of the lightweight breathing material JN125 and PN150 is higher than heavyweight breathing material JN315 and PN340 respectively. This result can be explained by the nonwoven structure of breathing materials. When processing methods are the same, the heavyweight nonwoven fabric is easier to achieve a compact structure with a lesser number of voids, which limits the structure to hold more water(Debnath and Madhusoothanan, 2010).

3.4.3 Thermal properties of hybrid breathing materials

Table 3.4 Thermal properties of JN125 and PN150.

Sample	Thermal Conductivity (w/mk)	Thermal Diffusivity (mm^2/s)	Specific Heat (MJ/m^3K)
JN125	0.06060	0.84100	0.07206
PN150	0.04500	0.55880	0.08053

The thermal properties of thermal conductivity, thermal diffusivity and specific heat may affect the ability of heat transfer, the rate of temperature spread and the ability to absorb or release heat in curing process, thereby affecting the quality of the composite materials. A comparison of JN125 and PN150 among those thermal properties is shown in Table 3.4. The thermal conductivity and thermal diffusivity of JN125 are higher than PN150, while the specific heat of JN125 is lower than that of PN150.

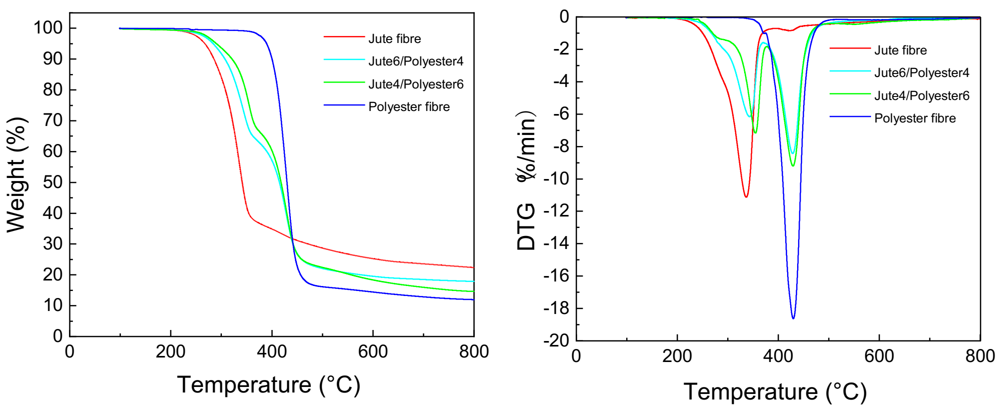


Figure 3.6 TG and DTG curves of pristine fibres and hybrid breathing materials.

Table 3.5 Data obtained from the TG test in N_2.

Sample	$T_{5\%}$ (°C)	T_{max1} (°C)	R_{max1} (%/min)	T_{max2} (°C)	R_{max2} (%/min)	Residue at 800°C (%)
Jute Fibre	267	336	11.19	-	-	22.43
Jute6/Polyester4	282	341	6.17	427	8.42	17.83
Jute4/Polyester6	290	354	7.10	427	9.24	14.62
Polyester Fibre	388	428	18.59	-	-	11.97

Figure 3.6 shows the TG and DTG curves of fibres and breathing materials, and the critical information is presented in Table 3.5. The onset degradation temperature ($T_{5\%}$) of jute fibre is 267 °C. The weight loss can be attributed to the degradation of lignin (Raghavendra et al., 2013). The prominent thermal degradation with a rapid weight loss of 60% can be observed from the temperature range between 259°C and 355°C. The maximum weight loss (T_{max1}) is at 336°C with the maximum loss rate (R_{max1}) of 11.19 %/min. The decomposition of lignin, hemicellulose and cellulose takes place in this stage(Manfredi et al., 2006). The weight loss keeps a constant rate until 800°C, remaining 22.43% char residue. For the polyester fibre, weight loss starts from 388°C. The prominent weight-loss stage occurs from 380°C to 460 °C, losing 70% of the weight. The char residue of jute fibre is 11.97% at 800°C. The jute/polyester hybrid breathing materials have a higher $T_{5\%}$ than the jute fibre but lower than that of the polyester fibre. They have two weight loss stages. The weight loss in the lower temperature range is related to the jute fibre, and

the higher one is attributed to the polyester fibre. Compared with pure jute fibre and polyester fibre, the R_{max} and T_{max} of jute/polyester hybrid breathing materials are much lower.

The polyester breathers can be applied in a broader range of temperature than hybrid breathing materials because of the higher onset degradation temperature of polyester fibre. However, the curing temperature of composite fabrication is mostly below 200°C. We use the temperature of 200°C as a reference temperature to determine whether the breathing material can be used in the production of composite materials. It can be seen in Figure 3.6 and Table 3.5 that hybrid breathing materials do not reach their degradation temperature at 200°C. Thus, the thermal stability of hybrid breathing materials meets the temperature requirements in composite materials production.

3.4.4 Permeability of hybrid breathing materials

3.4.4.1 Relationship between Porosity and permeability

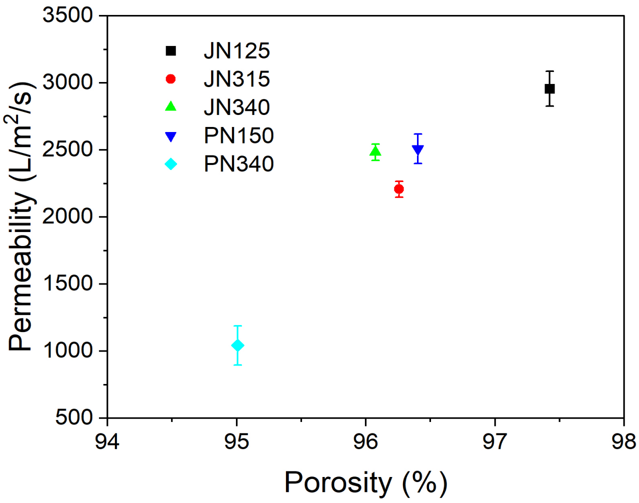


Figure 3.7 Permeability and porosity of breathing materials.

The air permeability of nonwoven fabric depends on the fabric density, thickness and porosity (Burlatsky et al., 2009, Payen et al., 2012, Pradhan et al., 2016). Due to the detailed information of pore size and pore shape cannot be provided by density, the porosity of different breathing materials was calculated and the relationship between permeability and porosity of different breathing materials are shown in Figure 3.7. It is noting that the permeability of all breathing materials raises with the increase of porosity. The lightweight breathing materials, such as JN125 and JN 150, have a relatively higher permeability than others. This is because the decrease in areal weight results in less entanglement of fibres and the structure becomes less consolidated(Debnath, 2011). The air is thus easier to go through the lightweight breathing materials. Among the heavyweight breathing materials, JN340 has the highest permeability of 2483 L/m^2/s, while PN340 has the lowest performance with 943 L/m^2/s. The air permeability of JN 315 is slightly lower than JN340. Those differences may be caused by the various average fibre diameters in different breathing materials. Nonwoven fabric with larger average diameter fibres has a higher air permeability (Pradhan et al., 2016, Payen et al., 2012). JN340 with 60wt% jute fibre has a higher average fibre diameter than that of JN315 with 40wt% jute fibre and PN340 with 100% polyester fibre.

3.4.4.2 Effect of moisture absorption on permeability

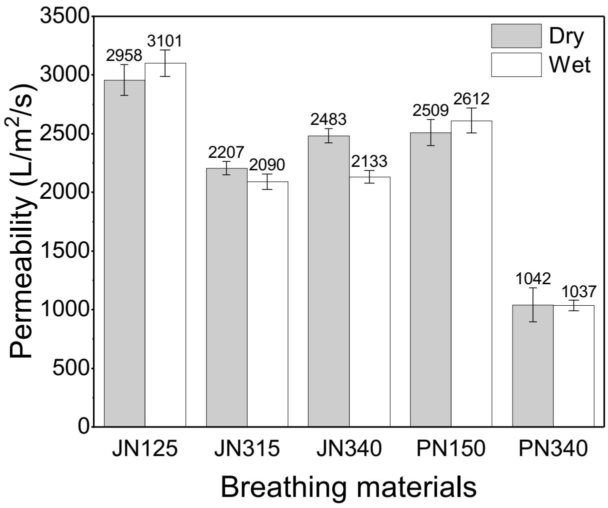


Figure 3.8 Permeability of breathing materials in dry and wet conditions.

The permeability of breathing materials before and after absorbing water is presented in Figure 3.8. For the hybrid breathing materials, the permeability of JN315 and JN340 is lower in the wet condition, while that of JN125 is higher under the wet state. This is possible because of the change of jute fibre fineness. When the jute fibre absorbs water in wet condition, the fibre swells and its diameter become larger (Ashori and Sheshmani, 2010, Masoodi and Pillai, 2012). For the heavyweight breathing materials, the swelled fibre is likely to occupy the voids in the nonwoven structure and restrict the airflow. However, the lightweight breathing materials are thinner in thickness. The wet jute fibre swells and stretches the fibre apart, thereby making the pores of the material larger. The increase in porosity leads to higher air permeability, which is consistent with the analysis in 3.3.1. For the polyester breathers, the permeability of PN340 with lower water absorption has basically not changed. The increase of permeability in PN150 may be attributed to the moisture in the structure of nonwoven fabric increases the porosity.

3.4.4.3 Effect of hot press aging on permeability

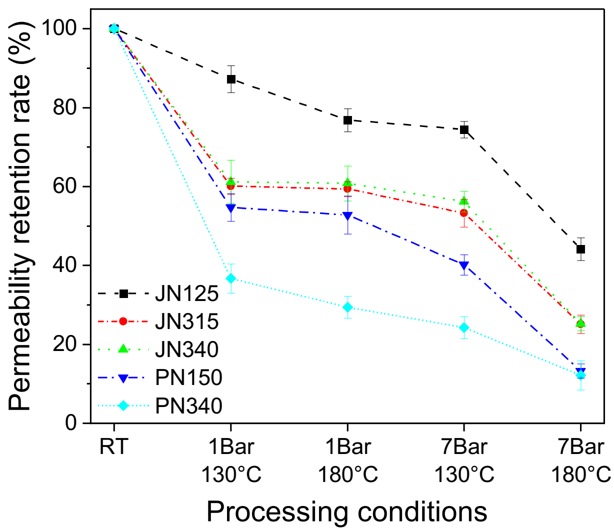


Figure 3.9 Permeability of breathing materials after applying in different conditions.

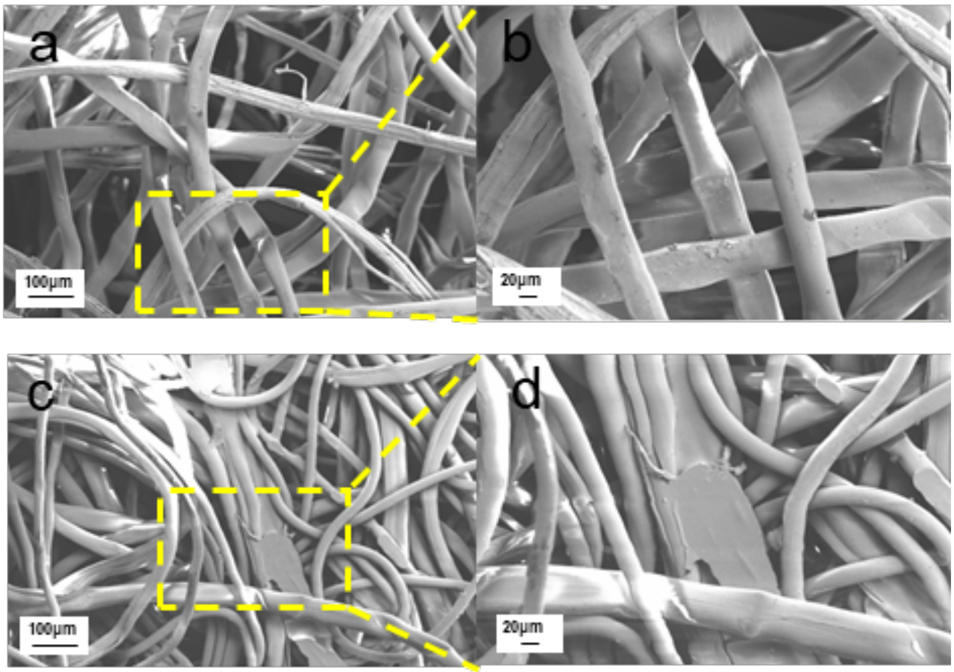


Figure 3.10 SEM images of JN315 (a and b) and PN340 (c and d) after applying in the 7bar, 180°C condition.

In order to investigate the influence of the hot press aging on breathing materials, the permeability retention rate of each breathing material is calculated by dividing the permeability before use by the permeability after use. The results are plotted in Figure 3.9. Under the different conditions, the permeability retention rate of all the breathing materials decreased to varying degrees. It is evident that the higher temperature and pressure treatment will lead to more significant permeability reduction. The reduction can be explained by equation 3.2. In equation 3.2, the porosity drops with the increase of the fabric density. The hot press aging condition forces the thickness of breathing materials to decrease, leading to the increasing density of the nonwoven structure and thus the porosity drop. For the breathing materials with the same fibre content, the permeability retention rate of the lightweight breathing material is higher than that of the heavyweight breathing material. However, under high pressure, the lightweight breathing materials are unable to exhaust the air from the laminate well in the actual manufacturing process.

It is evident from Figure 3.9 that the permeability retention rate of hybrid breathing materials is higher than that of polyester breathers in different processing conditions. Figure 3.10(a, b) show the morphologies of hybrid breathing materials JN315 after using in the 7bar,180°C condition. Compared with the pristine polyester fibre in Figure 3.4, the cylindrical structure of the fibre is flattened and deformed. Figure 3.10(c, d) show the deformation of PN340. It is seen that polyester fibres are not only deformed but also fused together. The existence of the jute fibre in hybrid breathing materials separates polyester fibre and prevents the fusion of polyester fibres. Therefore, hybrid breathing materials can keep their permeability better than polyester breathers.

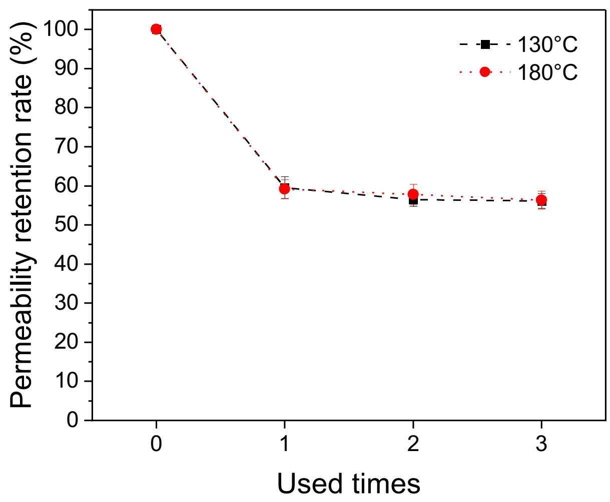


Figure 3.11 Permeability retention rate after reuse of JN315.

After used under 1 bar pressure, the permeability retention rate among hybrid breathing materials was at least 59%. The JN315 was reused three times under the 1 bar, 130°C and 1 bar, 180°C conditions to verify whether it can be used repeatedly instead of being discarded directly. The permeability retention rate versus used times of JN315 is plotted in Figure 3.11. It is seen that the effect of temperature on the permeability is not apparent when the pressure is 1 bar. After the first-time use, there is a significant drop of permeability, reducing around 40% of permeability in both 130°C and 180°C conditions. The subsequent two times of use have a relatively week effect on the permeability. After the third time use, the permeability of JN315 remains around 56% of pristine permeability in 130°C and 180°C conditions. This data is still higher than the permeability retention rate of PN340 with 37% at 130°C and 29% at 180°C after first-time use.

3.4.5 Tensile property of hybrid breathing materials in different conditions

3.4.5.1 Tensile property of hybrid breathing materials

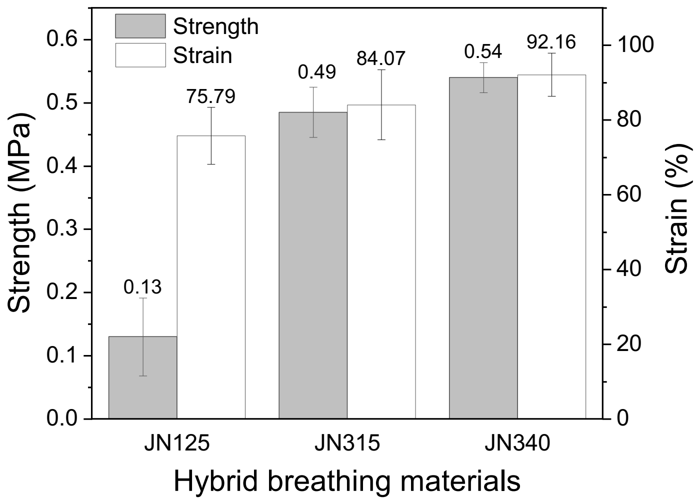


Figure 3.12 Breaking strength and strain of hybrid breathing materials.

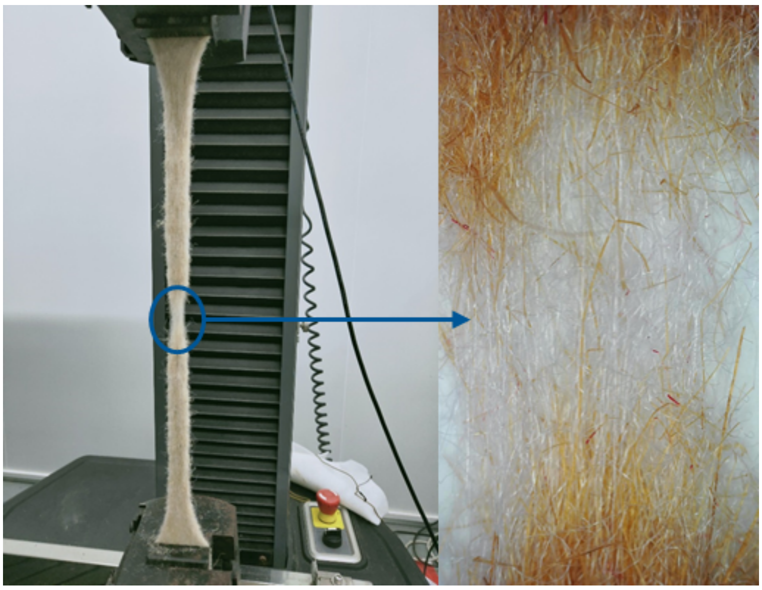


Figure 3.13 Fracture image from the tensile test of JN315.

Figure 3.12 shows the breaking strength and strain of hybrid breathing materials. When the fibre content is the same, the strength of the lightweight breathing material (JN125 with 0.13MPa) is much lower than that of the heavyweight breathing material (JN315 with 0.48MPa). This is probably due to the less entanglement of fibres in the structure with lower areal weight. For the heavyweight breathing materials, the breaking strength of JN340 is higher than that of JN315. The higher jute fibre content in JN340 improves nonwoven structure consolidation (Debnath, 2011). Besides, the tensile strength of the jute fibre is higher than the polyester fibre (Table 3.1). The breaking strain of JN315 and JN340 is 84.7% and 92.16% for each. The strain of JN125 is relatively low, with 75.79%. Figure 3.13 presents the image of fracture in the tensile test of JN 315. When the sample is stretched, the original crimped fibre is gradually straightened, and the fibre tends to be parallel to the direction of stretching. At the fracture, most of the jute fibre was broken while the polyester fibre still exists. It can be deduced that the breaking of jute fibre provides the tensile strength of hybrid breathing materials.

3.4.5.2 Effect of moisture absorption on tensile properties

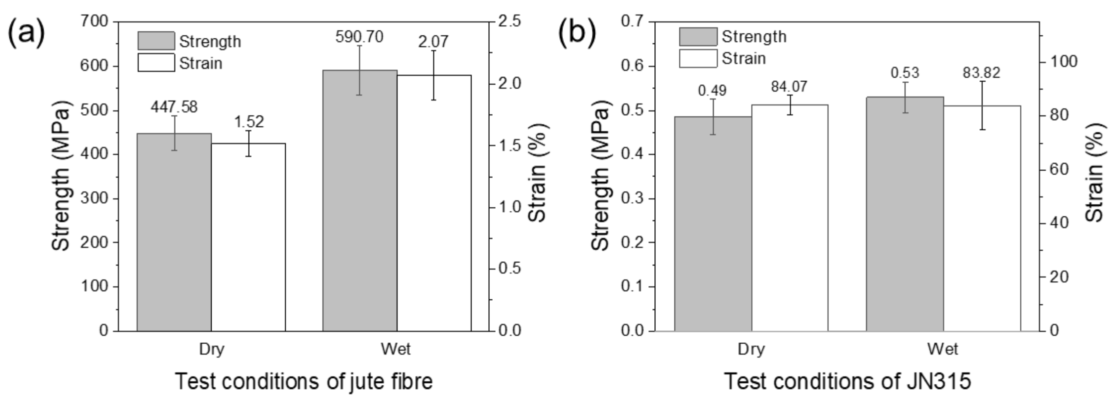


Figure 3.14 Tensile properties of jute fibre (a) and JN315 (b) in the dry and wet conditions.

The tensile properties of jute fibres and JN315 in the wet and dry conditions are shown in Figure 3.14. In the wet condition, the strength of jute fibre is 590.70 MPa, and it is higher than that in dry condition with 448.58 MPa. The higher tensile strength of wet jute fibre leads to the increased tensile strength of the JN315 in the wet condition. The tensile strain of JN315 is much higher than that of jute fibre, and the breaking strain is almost unchanged in wet and dry conditions.

3.4.5.3 Effect of hot press aging on tensile properties

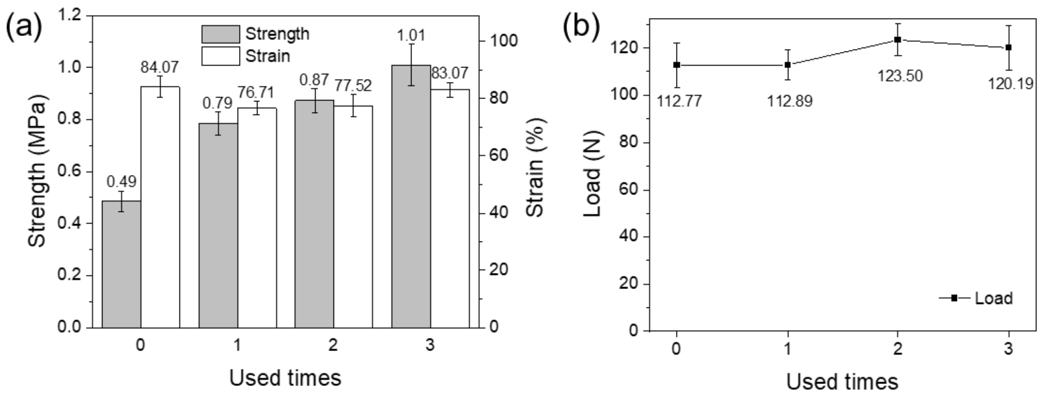


Figure 3.15 Tensile properties (a) and breaking load (b) of JN315 after three used times (1bar, 180 °C).

The tensile stress and strain, as well as the breaking load of JN315 are plotted versus using times in Figure 3.15. With the increase of using times, the breaking strength of JN315 raises from 0.49 MPa in pristine condition to 1.01 MPa after three times of use. It is worth noting that there is only a slight change in the breaking load, increasing from 112.77 N in the initial state to 120.19 N after three times of use. Considering the strength is mainly affected by the breaking load and the cross-sectional area, the increase of strength is attributed to the reduction of the cross-sectional area caused by the hot press aging.

3.5 Conclusions

In the present study, the jute/polyester hybrid breathing materials were prepared by the needle-punched method to be used as breathers in the process of composite manufacturing. Thermogravimetric analysis proved that hybrid breathing materials were stable below 200 °C, and it meets the curing temperature requirements of most composite manufacturing. The effect of humidity and hot press aging was investigated by testing the moisture absorption rate, air permeability and tensile property of hybrid breathing materials. Hybrid breathing materials were sensitive to moisture than polyester breathers. The moisture absorption increased with the increase of jute fibre content and reduced with the increase of the areal weight. Moisture could influence the permeability of breathing materials due to the changes in the jute fibre diameter. The permeability of breathing materials raised with the increase of porosity. Lightweight breathing materials had a higher permeability than heavyweight breathing materials. When the areal weights were similar, breathing materials with higher jute fibre content had higher permeability.

Hybrid breathing materials could maintain their air permeability better than polyester breathers under high-temperature and high-pressure conditions. After three times of use, the permeability of hybrid breather JN315 was still around 56%. The tensile strength of the hybrid breathing materials increased after being processed in the humidity condition, while the breaking strain nearly unchanged. The repeating use of breathing materials increased their tensile strength. Therefore, it can be concluded that hybrid breathing materials could be used as breathers, and they could maintain relatively high permeability under high-temperature and high-pressure conditions. They were available to be reused at least three times under the pressure of 1 bar. The end-of-life hybrid breathing materials were possible to be further utilized as raw materials to produce composite materials. High moisture absorption of hybrid breathing materials requires low humidity storage conditions or drying before use.

Chapter 4

Reuse of Hybrid Breathing Materials for Sustainable Composite Manufacturing

4.1 Overview

Jute/polyester hybrid breathing materials were developed as an alternative to the conventional nylon or polyester breather to reduce the use of synthetic polymers and associated waste generated during composite manufacturing. However, end-of-life breathers still face the challenge of waste management. In this study, a circular economy strategy for multiple applications of waste hybrid nonwoven breathers was thus proposed to study their recycling possibilities. Hybrid nonwoven breathers were first used to facilitate the autoclave forming of carbon fibre-reinforced plastics, and non-destructive and mechanical tests were performed to determine the quality. The used breathers were then recycled and reused in three ways: directly as reinforcing material and in hybrid combinations with glass fibres and ramie fibres to produce new composites. Results of quality checks show that the hybrid nonwoven breather is a viable alternative to commercial breathers. The thermal, mechanical, and damping properties of the composites incorporating reused material were investigated. The hybrid nonwoven breather/Glass fibre composites show superior thermal and mechanical properties as well as improved damping properties. The hybrid nonwoven breather/Ramie fibre composites have the advantages of low density and high loss factor reaching up to 0.1572. The product of specific modulus and loss factor was then used as the figure of merit to estimate the structural damping properties of the composites. Both glass fibre and ramie fibre hybrid composites presented a higher figure of merit with 0.86 and 0.83 GPa/(g/cm3), respectively. Therefore, hybrid nonwoven breathers can be adapted to replace synthetic materials in semi-structural applications.

4.2 Introduction

It is well-known that certain composite manufacturing routes drive a large volume of waste material, including offcuts of fabrics and prepregs, single-use consumables such as breathers, peel-plies, bagging membranes, end-of-life products and manufacturing rejects (Stelzer et al., 2022, Rybicka et al., 2015). Sustainability is a critical consideration for manufacturing organizations(Machado et al., 2020), and this importance will likely increase over time as resources become scarce and emerging economies introduce their regulatory frameworks (Smith and Ball, 2012, Pimenov et al., 2022). The pressure for waste management reinforces the need for utilizing previous waste materials. The European Union encourages that waste should be prevented before it occurs and dumping it in landfills should be the last resort. Although incineration has the advantage of partial energy recovery, it is also associated with harmful emissions, fly ash, and the need to process solid residue (Chen et al., 2019). Re-use and recycling are desirable options to minimize landfill space and incineration pollution. Besides, the use of virgin materials and associated resources will be reduced (Dissanayake and Weerasinghe, 2020). Several studies have been reported on the reuse of end-of-life composites using mechanical, thermal, and chemical techniques (Oliveux et al., 2015, Yang et al., 2012b, Hasan et al., 2019). However, little attention has been paid to the ancillary consumables such as bagging films, breathers, release films, etc. Such consumables do not feature in the final product; thus, their presence is easily overlooked. Most of these consumables are made from non-degradable petroleum-based materials with few options for disposal beyond incineration or landfill.

Jute/polyester hybrid breathers were developed in our previous study, reducing the non-renewable content by partially replacing polyester fibres with jute fibres (Tong et al., 2021). It was found such *green* breathers had higher initial permeability and residual permeability of 56% after 3 cycles of compression moulding under a pressure of 1 bar. It is foreseen that after certain cycles, this *green* breather will reach its permeability limit which cannot act as a breather anymore. How to deal with end-of-life *green* breathers will become our next interest.

A good way to deal with these waste breathers is to repurpose and turn them into new products that meet quality standards. Using waste hybrid breathers as components of composites could be a means to mitigate problems that arise when recycled materials with relatively low properties are used. It is proven that waste materials as reinforcement reduce the use of plastics while maintaining or improving their technical properties (Civancik-Uslu et al., 2018). Cotton/Polyester reinforced epoxy composite derived from nonwoven wastes has been reported to be added into epoxy resin and exhibits a modest improvement in Young's modulus, specific tensile strength, and impact resistance compared with epoxy resin. Nonwoven waste has been proved to be a promising candidate to replace up to 25wt% epoxy resin to save resin usage whilst achieving mechanical performance improvement (Baccouch et al., 2020). However, because these recycled composites do not provide significant advantages in terms of availability, durability, quality, and functionality, they face challenges in expanding their end-user market.

Hybridization can be a promising approach to tailor the properties of composite materials to meet specific requirements in various applications. However, research on hybrid composites is currently conducted primarily using pristine materials. Applying waste materials, especially low-performance materials, to hybrid composites has yet to be explored fully. Besides, developing sustainable industries requires biopolymer composites (Rababah and Al-Oqla, 2020). Bio-based matrix will be a demanding goal for composites to reduce the proportion of non-renewable resources. The development of hybrid composites offers new possibilities for reusing and combining waste materials into new sustainable products.

In this paper, a circular economy approach and multiple application strategies were used to design a route for such purpose: analysing the feasibility of hybrid nonwoven mats as a breather in composite manufacturing and utilizing the used breather as the reinforcement to develop hybrid functional composites. As shown in Figure 4.1, jute/polyester hybrid nonwoven breathers were used for autoclave processing in composite manufacturing. Initial feasibility was verified by comparing the permeability with that of a commercially available breather, and then the quality of the final composite was examined. Following initial use, the hybrid nonwoven breathers were then reused in three ways to explore their added value:

1) As a lightweight structural filler for epoxy matrix composites.

2) Hybridized with glass fibre to improve the mechanical properties of composites

3) As a functional component, combined with ramie fibre and rosin-based epoxy to produce environmentally friendly green composites.

The thermal, mechanical and damping properties of the composites were studied in detail to investigate the reusability of the hybrid nonwoven breathers.

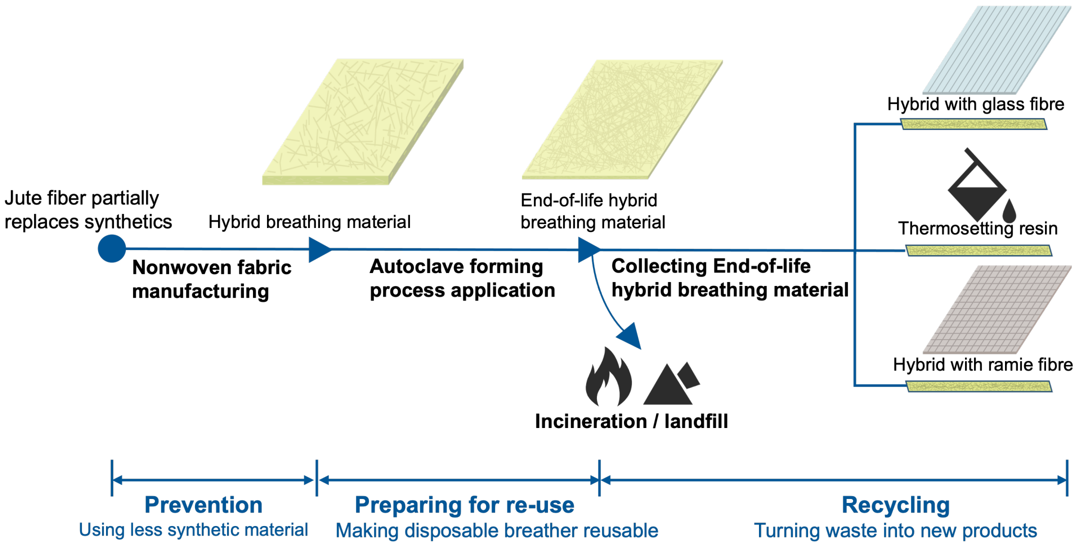


Figure 4.1 Scheme of sustainable composite manufacturing and application, and the three utilization ways of recycling of the hybrid nonwoven breathers.

4.3 Materials and methods

4.3.1 Materials

Jute/polyester hybrid nonwoven breathers (HNBs) with an area density of 315 g/m^2 were produced by needle punching. Jute fibre and polyester fibre were mixed at the fibre content of 40 wt% and 60 wt% in HNBs.

In the autoclave forming process, commercial polyester breather Airweave® N10 with the areal density of 339g/m^2 (Airtech International Inc.) was used as a comparative sample. Unidirectional carbon fibre-epoxy prepreg 5228A/CCF300 was provided by AVIC Composite Corporation Ltd. Beijing, China. The initial fibre volume fraction in the prepreg was 55%, the fibre area density was 145g/m^2, and the resin area density was 83 g/m^2.

Following fabrics and epoxy resins were used with recycled HNBs to manufacture composites. Unidirectional glass fabric（EL200/ec468）with an areal density of 1200 g/m^2 was obtained from CPIC Co., Ltd, Chongqing, China. Plain weave ramie fabric with an areal density of 123g/m^2 was obtained from Hunan Huashengdongting Ramie Co., Ltd, Yueyang, China. Properties of fibres and fabrics used in this study were listed in Table 4.1 and Table 4.2, respectively. Detail properties of jute/polyester hybrid nonwoven breather can be found in chapter 3 and referenced as JN315. Properties of fibres were obtained from suppliers. The determination of areal density and tensile properties followed the standard outlined in section 3.3.4. Araldite® LY 1572 epoxy and hardener 3846 were purchased from Huntsman Corporation, the United States. Rosin-based epoxy resin AGMP3600 was provided by AVIC Composite Corporation Ltd. Beijing, China.

Table 4.1 Properties of fibres in jute/polyester hybrid nonwoven breather, ramie fabric, and unidirectional glass fabric.

Fibre types	Length (mm)	Diameter (μm)	Strength (MPa)	Density (g/cm^3)
Jute fibre	60	67	449	1.48

Polyester fibre	64	38	76	1.39
Ramie fibre	/	/	/	1.50
Glass fibre	/	16	≥805	2.30

Table 4.2 Properties of jute/polyester hybrid nonwoven breather, ramie fabric and unidirectional glass fabric.

Reinforcements	Areal density (g/m^2)	Tensile force at break (N)	Tensile elongation (%)
Jute/polyester fibre mats	315	113	84.07
Ramie fabric	123	650	3.75
Glass fabric	1200	/	/

4.3.2 Methodology

Figure 4.2 presents the methodology of this study. HNBs were initially used to facilitate the autoclave forming of CFRP. Mechanical and non-destructive tests were done to check the quality of CFRP. Used HNBs were collected and reused as reinforcement within composites. The thermal, mechanical and damping tests were conducted to determine the properties of composites. Obtained data were analysed and critically discussed.

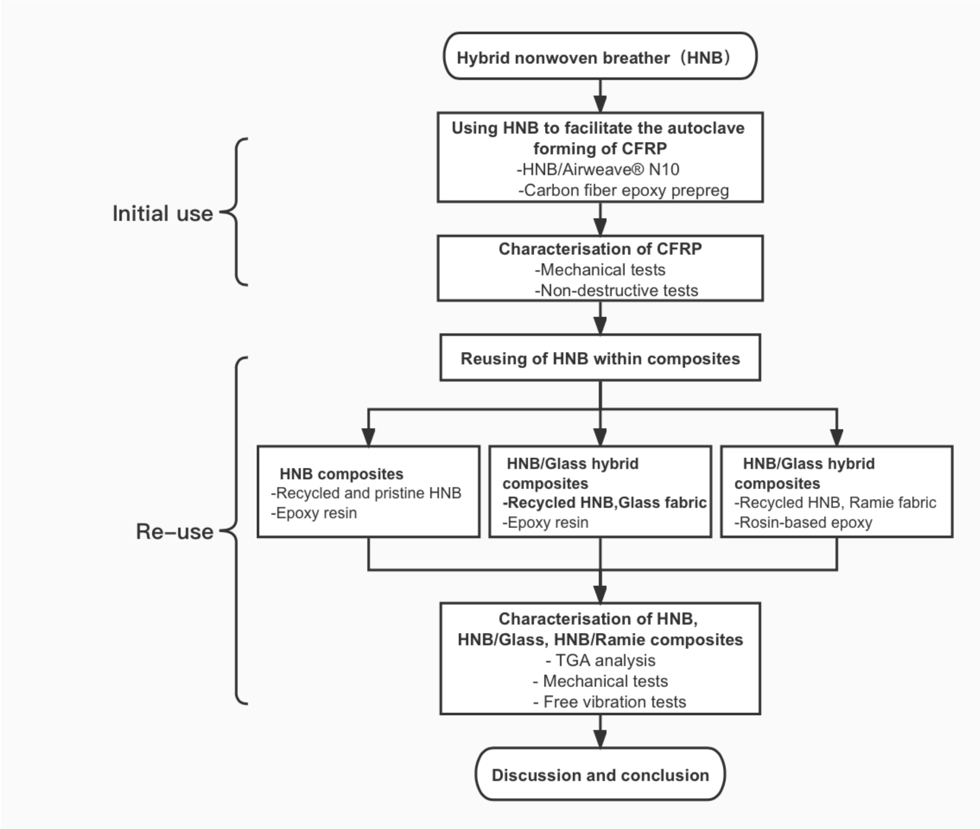


Figure 4.2. Flow diagram of study methodology.

4.3.3 Evaluation of the viability of hybrid nonwoven breathers

Fabrication of carbon fibre reinforced plastic.

Two carbon fibre reinforced plastic (CFRP) laminates were formed simultaneously using the conventional polyester control breather and the HNB in an autoclave as shown in Figure 4.3(a). Unperforated release film was applied between breathers and performed laminates to prevent breathers from being impregnated. A vacuum (around 0.9 MPa) was applied to the whole assembly at room temperature, and the curing conditions are illustrated in

Figure 4.3(b). Two thicknesses of laminates with the stacking sequence of $[0]_{28}$ and $[0]_{8}$ were fabricated, respectively.

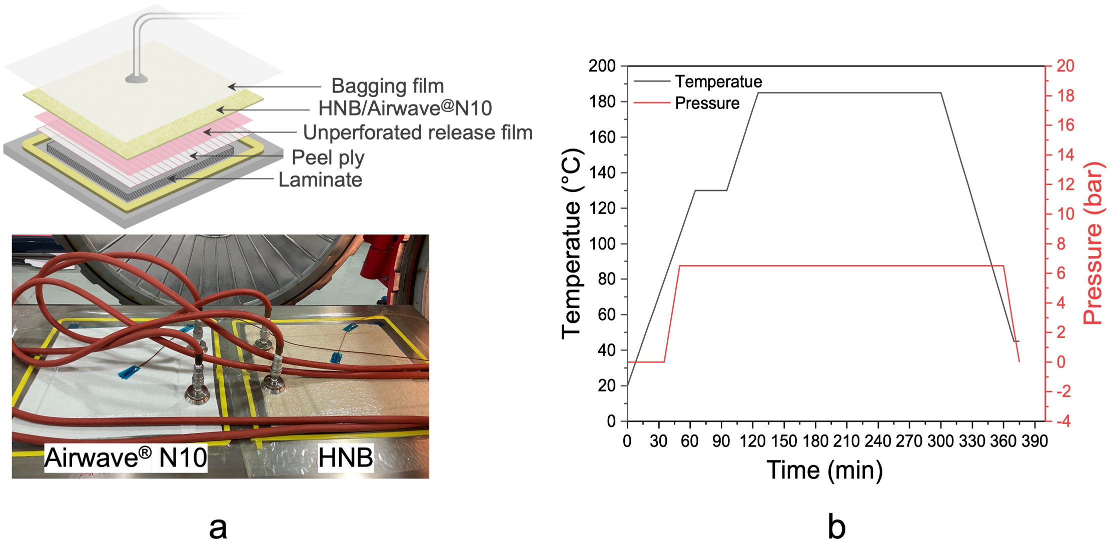


Figure 4.3 Images of CFRP laminates fabrication with Airwave® N10 and HNB (a), and the curing procedure in an autoclave (b).

Characterization of CFRP.

Non-destructive tests of CFRP laminates were performed using a PAC UPK-T36 ultrasonic scanner. The tensile, flexural, compressive and shear properties of the prepared laminates were tested using a universal test machine (MTS E42, MTS Systems Corporation, USA). The test standard, sample size as well as sample number were listed in Table 4.3. The data obtained from the mechanical tests were compared using the Student's paired t-test for the comparison of specimens in each test.

Table 4.3 Test standard and sample detail of CFRP.

Properties	Test standard	Sample size(mm^3)	Sample number
Tensile	ASTM D3039	250*15*1	6
Flexural	ASTM D7246	150*12*3.5	6
Compressive	ASTM D6641	180*13*3.5	6
Shear	ASTM D2344	24*8*3.5	6

Characterization of HNB.

The end-of-life HNBs were collected after the composite is cured in an autoclave (as described in 2.2). The recycled HNBs without resin stained were selected for the reusing process. ISO9073 (2) was followed to measure the thickness of breathing materials. The testing pressure was 0.5 kPa. The transverse permeability of HNB and Airwave® N10 before and after use was determined using a YG461E digital fabric air permeability meter (Baien Instrument Co., Ltd, Wenzhou, China) under a pressure difference of 100 Pa. Table 4.4 shows the difference in thickness and transverse permeability between the HNB and conventional control breather.

Table 4.4 Comparison between the HNB and Airwave® N10 breather before and after being used in an autoclave.

Breather Type	Areal density (g/m^2)	Permeability($L/m^2/s$)		Thickness(mm)	
		Pristine	After use	Pristine	After use
Airwave® N10 (Control)	339	1738	33	3.6	0.59
HNB	315	2207	165	5.89	0.92

4.3.4 Re-use of hybrid nonwoven breathers within composites

4.3.4.1 Fabrication of composites

HNBs were first used as fillers to prepare HNB-reinforced epoxy composite samples via vacuum infusion. One layer of pristine HNB and two layers of recycled HNBs were infused with epoxy, respectively. The absolute pressure within the vacuum bag was approximately 0.1 MPa (full vacuum). Cast epoxy resin was prepared in a 2 mm steel mould as a control sample. All the samples were cured under a full vacuum at 80 °C for 5 hours.

HNB/Glass fibre composites were also fabricated via vacuum infusion. The HNBs and glass fabrics were stacked in different sequences as shown in Table 4.3. The glass fibre plies were stacked with angle-ply orientations of 0°, and the HNB plies were stacked along the machine direction. The infusion and curing procedures were identical to those used with HNB composites.

To prepare the HNB/Ramie fibre composites, the recycled HNBs and ramie fabrics were first pre-impregnated with rosin-based epoxy to prepare prepregs. The resin was mixed with acetone in the ratio of 1:4 and brushed evenly on the HNBs and ramie cloth. After drying in a vacuum oven at 30 degrees for 24 hours, prepregs with a resin volume fraction of 45% were obtained. After that, each layer was stacked as shown in Table 4.3. A stainless-steel mould with a thickness of 1.1 mm or 2.2 mm was used to control the thickness of the composites. The whole assembly was placed between two steel trays covered with Teflon sheets and cured in a press of 2 MPa at 90 °C for 30 minutes, then 130 °C for two hours with the same pressure. To form the porous structure, HNBs applied in the RHR* and RHRHR* samples were not impregnated with resin.

4.3.4.2 Characterization of composites

The density was measured by dividing the weight by the volume of the composite. The volume was calculated based on the dimensions of the samples. The volume of each component in composites was established by dividing its mass by the average fibre density, and subsequently, the volume fraction for each component was computed by dividing the obtained volume by the total volume of the composite. The stacking sequence, thickness, density, and volume fraction of different laminates are shown in Table 4.5. Figure 4.4 shows the cross-section images of HNB/Glass fibre composites and HNB/Ramie fibre composites.

Table 4.5 Designation of composites for multiple applications of HNBs.

Composite samples	Stacking sequence	Thickness (mm)	Density (g/cm^3)	Volume fraction (%)		
				HNB	Glass fibre	Ramie fibre
HNB composites	H(Pristine)	2.21	1.16	13	/	/
	HH	1.85	1.18	32	/	/

HNB/Glass fibre composites	GHHG	3.41	1.51	15	30	/
	HGGH	3.20	1.49	17	34	/
	GG	1.83	1.85	/	60	/
HNB/Ramie fibre composites	RHR*	1.10	0.68	34	/	27
	RHR	1.10	1.01	34	/	27
	RHRHR*	2.20	0.62	34	/	17

H denotes the HNBs, G the glass fibre fabric and R represents the ramie fabric;

In the RHR* and RHRHR* laminates, the HNBs were not pre-impregnated with resin.

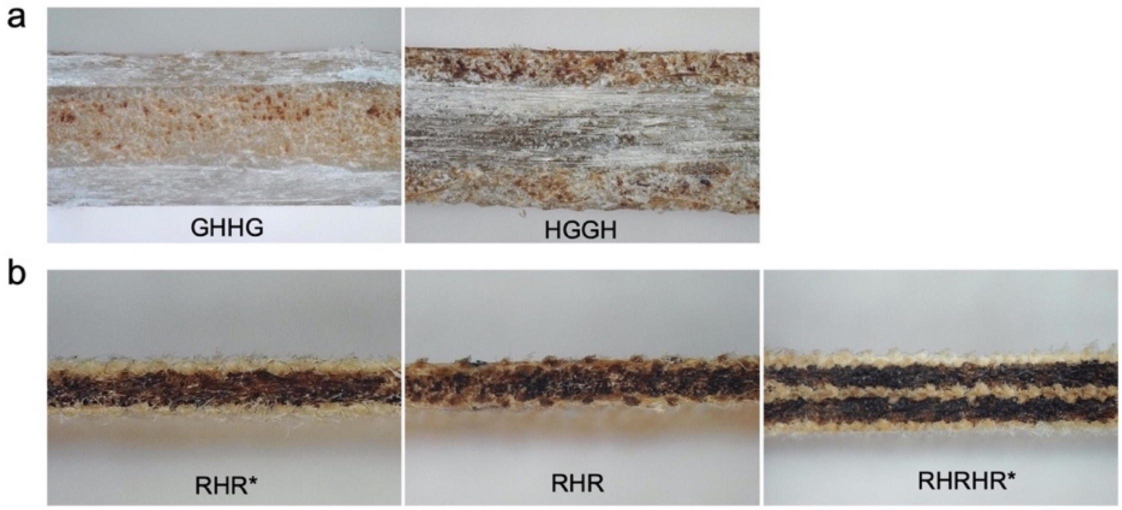


Figure 4.4 Cross-section images of HNB/Glass fibre composites (a) and HNB/Ramie fibre composites (b).

The thermal stability of prepared composites was analysed by performing the thermogravimetric analysis (TGA) with an SDT Q600 analyser (TA Instruments). Specimens with a weight of 10 mg were placed in an alumina crucible. To remove the influence of moisture, specimens were first equilibrated at 100 °C and heated by 10 °C/min under a 50 ml/min nitrogen flow to 800 °C.

The tensile properties of the composites were performed on a universal testing machine (MTS E42, MTS Systems Corporation, USA) according to ASTM

D3039 with a speed of 2 mm/min. The length and width of the tensile specimens were 180 mm x 20 mm. The flexural test was conducted following ASTM D790 standards with a span-to-depth ratio of 32:1 and a crosshead speed of 1 mm/min. The impact strength of composites was tested according to ASTM D256. The tests were conducted along the machine direction of the HNB. Tests were performed on at least five specimens at room temperature in each case to determine the average value.

Free vibration tests were done on cantilever beam specimens to determine the damping properties according to ASTM E756. The dimension of the specimens was 200mm × 10mm. The fixed length was 20 mm. An impact hammer (PCB piezotronics, 086C03 model) was used to excite specimens near the clamped end. An accelerometer (PCB piezotronics, 352C65 model) was used to measure the output response. A dynamic analyser (Data Physics QUATTRO) was used to digitize the excitation and response signals. The loss factor η was calculated by taking the values of the successive peaks from the vibration response graph and substituting them in Equations 4.1 and 4.2.

$$\delta = \frac{1}{n} ln \frac{g_i}{g_{i+n}} \quad (4.1)$$

$$\eta = \frac{1}{\pi} \delta \quad (4.2)$$

Where δ is the logarithmic decay rate, n corresponds to the number of periods observed, g_i represents the first observed peak in acceleration and g_{i+n} represents the (i+n)th peak in acceleration.

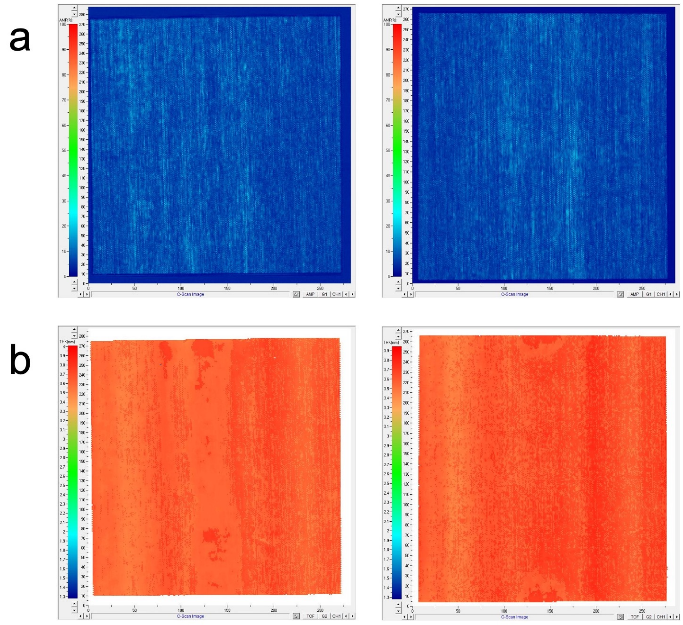
The fracture surfaces of specimens after tensile tests were studied with a Zeiss Sigma V.P. scanning electron microscopy (SEM). Before the SEM imaging, specimens were coated with a 5nm layer of gold by a Leica SCD 500 gold sputter.

The analysis of variance (ANOVA) was used to determine the effects of design variables at 95% confidence levels.

4.4 Results and discussion

4.4.1 Viability of hybrid nonwoven breathers

The two CFRP laminates were C-scanned prior to the mechanical tests. Figure 4.5 shows the scan results for the CFRP laminates produced by HNBs and Airweave® N10 breathers. Amplitude (AMP) C-scans indicate that the amplitude of both laminates is greater than 90%. C-scans of Time of Flight (TOF) indicate that both laminates are generally flat and uniform in thickness.



CFRP fabricated with Airweave® N10

CFRP fabricated with HNB

Figure 4.5 AMP (a) and TOF (b) C-scan results of $[0]_{28}$ CFRP laminates.

Table 4.6 Mechanical properties of CFRP laminates formed with Airweave® N10 and HNB.

Mechanical properties		Airweave® N10	HNB	P
Tensile	Strength (MPa)	1690.87±114.93	1608.61±72.73	1.69E-01
	Modulus (GPa)	219.29±11.17	202.41±10.08	1.24E-01
Flexural	Strength (MPa)	1445.25±104.99	1460.57±31.26	7.39E-01
	Modulus (GPa)	134.55±2.02	134.09±2.42	7.28E-01
Compressive	Strength (MPa)	748.11±28.32	760.34±33.74	5.12E-01
	Modulus (GPa)	110.65±11.71	111.30±11.67	9.25E-01
Shear	Strength (MPa)	110.60±1.36	105.63±3.00	5.92E-02

The mechanical strength and modulus of the two laminates were tested and the results are shown in Table 4.6. The results show that the performance of CFRP composites fabricated with N10 and HNB breathers slightly varied although the differences in all of the static properties mentioned were statistically insignificant according to the Student's t-test results ($P > 0.05$).

4.4.2 Re-use of hybrid nonwoven breathers as the single filler

4.4.2.1 Compression among epoxy, and pristine and recycled HNB reinforced composites.

Table 4.7 Mechanical and damping properties of HNB composites and cast epoxy samples.

Sample	Tensile		Flexural		Loss
	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Factor (η)
Epoxy	38.30±1.59	0.61±0.04	86.50±2.44	2.25±0.11	0.0738
H(Pristine)	40.47±2.81	0.92±0.08	63.21±0.49	2.54±0.14	0.0814
HH	54.72±1.33	1.39±0.23	84.09±0.82	3.69±0.28	0.0983

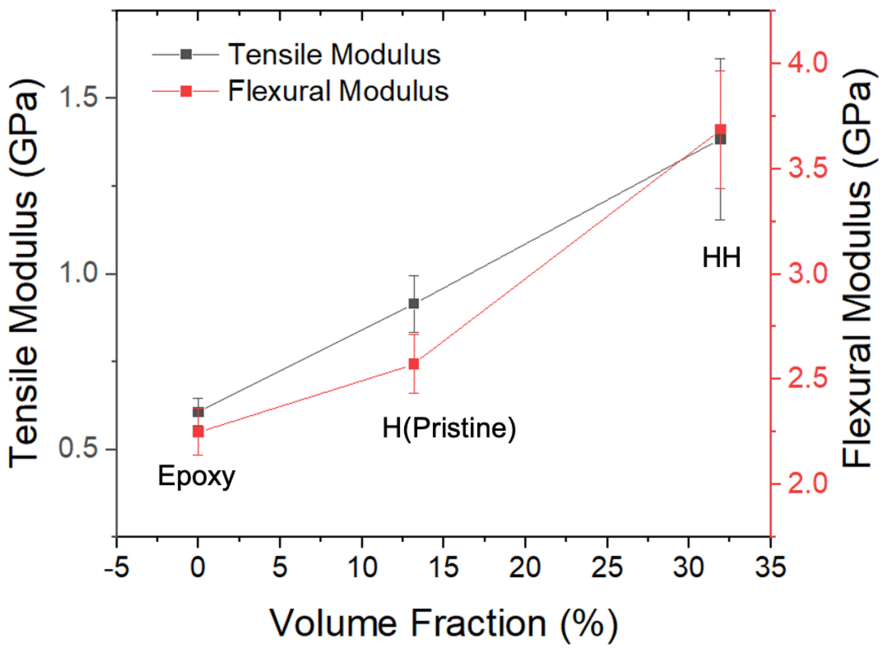


Figure 4.6 The effect of the HNB volume fraction on the tensile and flexural modulus.

Table 4.7 shows the differences among the pristine HNB composite (H(Pristine)), recycled HNB composite (HH) and cast epoxy samples in terms of mechanical and damping properties. The epoxy sample demonstrated a tensile strength of 38.30 MPa. Although the H(Pristine) composite sample had a slightly higher tensile strength of 40.47 MPa, the improvement is not statistically significant (P>0.05). In comparison, the HH composite sample exhibited a 43% higher tensile strength (54.72 MPa) than the neat epoxy sample. It is reported in the literature that the tensile strength of jute fibre composites was between 40 MPa and 80 MPa (Raghavendra et al., 2013, Sezgin and Berkalp, 2016, Reddy et al., 2018). The results of this study are close to the general value, while the polyester fibre in the HNB reduces the overall volume fraction of jute fibre in the composite, resulting in less strength (Baccouch et al., 2020). The flexural strengths of H(Pristine) and HH composite samples were 63.21 MPa and 84.09 MPa, respectively. In comparison to the neat epoxy sample with 86.50 GPa, the flexural strength was thus not improved via the incorporation of jute fibres. The tensile modulus of the H(Pristine) and HH samples was 0.92 GPa and 1.39 GPa, respectively, which is superior to the modulus for epoxy with 0.61 GPa. It is seen from Figure 4.6 that there is a linear relationship between the volume fraction of HNB and tensile modulus. The flexural modulus also increases with the volume fraction, although some non-linearity is evident. The differences between the H(Pristine) and HH composite results are due to the change of state of the breather after the first use. According to the previous study, the breaking load of HNB exhibited little change after the first use, while its porosity decreased as the structure became densified (Tong et al., 2021). Therefore, under the same pressure, recycled HNB absorbs less resin, and thus the HNB volume fraction in the HH composite is higher, leading to a higher modulus. Generally, the mechanical properties of natural fibre reinforced composites increase as the fibre volume fraction increases (Esmaeili et al., 2014). However, the lower flexural strength of the H(Pristine) and HH composite samples compared to the neat epoxy can be attributed to insufficient reinforcement to withstand bending forces. In addition, weak interfacial bonding of fibre and matrix reduces the stress transfer between fibres and epoxy (Hu et al., 2020, Doan et al., 2012).

The epoxy sample had the lowest loss factor at 0.0738. The value for the H(Pristine) composite sample was slightly higher than that of epoxy. The highest loss factor was found in the HH composite sample with 0.0983, which also had the highest fibre volume fraction. Note that the introduction of HNB appears responsible for the increase in loss factor. The intrinsic structure of the jute fibre, such as the lumen and the heterogeneous cell wall contributes to energy dissipation (Ashworth et al., 2016, Le Guen et al., 2014). In addition, the poor interfacial bonding between the fibres and epoxy may also contribute to the increase in the loss factor (Cihan et al., 2019, Essabir et al., 2013).

4.4.3 Re-use of hybrid nonwoven breathers by hybridization

4.4.3.1 Thermal properties of hybrid nonwoven breathers reinforced composites

The thermal stability of hybrid composites investigated by TGA is shown in Figure 4.7 and Table 4.8. The onset temperature ($T_{5\%}$) of HNB/Ramie fibre composites was relatively lower at a temperature range of 300-310°C and that of HH composite was 319°C. In the literature, the decomposition of lignin occurred first at low temperature (150°C-170°C), followed by the decomposition of hemicellulose and cellulose components at a temperature range of 200 – 400°C (Raghavendra et al., 2013, Zeriouh and Belkbir, 1995). The biomass content in HNB/Ramie fibre composites is higher than that in HH composites, resulting in a lower $T_{5\%}$. The degradation of polyester generally takes place between 350-450°C (Papageorgiou et al., 2014, Trask and Beninate, 1986). The GHHG and HGGH composites containing glass fibres had higher $T_{5\%}$ than the HH composite as well as an increased percentage of residues. As glass fibre has high thermal stability, it prevents degradation and absorbs heat in the matrix, thereby prolonging degradation (Panthapulakkal and Sain, 2007b). It is noted that all composites lost major weight (T_{max}) at around 370°C from degradation and volatilization of epoxy along with fibres. A similar finding has been reported by other researchers as well (Boopalan et al., 2013). As a result, the maximum thermal degradation rate (R_{max}) obtained from DTG results for the corresponding T_{max} is related to fibre volume fraction. The higher fibre volume fraction of composites leads to a reduced R_{max}.

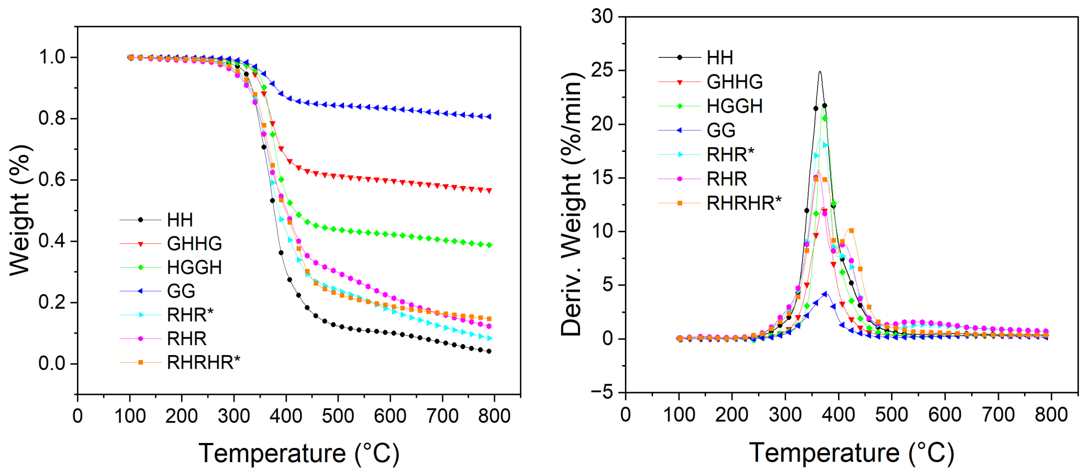


Figure 4.7 TGA and DTG results of all seven composites.

Table 4.8 The characteristic temperature of composites obtained from TGA and DTG curves.

Sample	$T_{5\%}$ (°C)	T_{max} (°C)	R_{max} (%/min)	Residue (%)
HH	313.97	367.7	24.69	10.88
GHHG	335.6	371.96	12.17	56.67
HGGH	330.67	369.17	15.74	39.85
GG	351.43	375.14	4.20	80.66
RHR*	304.32	374.16	17.99	8.36
RHR	300.32	362.69	15.54	12.37
RHRHR*	308.87	364.8	15.4	14.76

4.4.3.2 Mechanical properties of hybrid nonwoven breathers reinforced composites

Table 4.9 Mechanical properties of HNB/Glass fibre, Glass fibre and HNB/Ramie fibre composites.

Sample	Tensile		Flexural		Impact
	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	strength (KJ/m^2)
GHHG	503.10±13.95	20.40±0.39	695.90±46.64	33.55±0.16	405.45±28.55
HGGH	551.15±19.18	21.80±1.23	353.03±26.20	9.25±0.42	397.71±67.20
GG	837.52±14.77	36.93±0.038	954.72±14.70	31.82±0.62	/
RHR*	32.47±8.09	1.95±0.05	46.69±4.12	4.64±0.53	30.08±1.35
RHR	61.17±2.08	2.59±0.13	79.33±4.29	4.88±1.21	17.47±1.20
RHRHR*	17.75±1.19	0.45±0.10	13.27±0.86	1.73±0.12	27.36±2.86

The tensile properties, flexural properties, and impact strength of HNB/Glass fibre, Glass fibre and HNB/Ramie fibre composite samples are listed in Table 4.9. It is observed that adding two plies of glass fabric in the HH composite sample enhanced its tensile strength approximately ten-fold, reaching 503.10 MPa for the HGGH composite sample and 551.15 MPa for the GHHG composite sample, influenced by the fibre volume fraction (Cihan et al., 2019). The tensile modulus of the GHHG and HGGH composite samples also increased to 20.40 GPa and 21.80 GPa, respectively, while that of the HH composite sample was only 1.39 GPa. Unsurprisingly, the stacking sequence does not have a significant impact on tensile strength and modulus, which is consistent with the previous research on the stacking sequence of composites

(Sezgin and Berkalp, 2016). Component materials and their relative loadings determine the tensile properties of hybrid composites (Li et al., 2017).

The tensile failure modes of GHHG and HGGH composite samples are different. In the GHHG composite sample, delamination is observed between the glass fibre and jute fibre layers, followed by the break of glass fibres. This can be explained by the lower elongation of glass fibres than that of jute fibres at failure (Wambua et al., 2003). When it comes to the HGGH composite sample, the jute fibre layer stacked on the surface is the first to break. The fracture surface morphology of HNB and Glass fibre layers after tensile testing is shown in Figure 4.8. All the samples display brittle fibre failures or the fibre pull-out as the dominant failure modes. In the HNB layer, both the jute and polyester fibres are pulled out with a significant length, which reveals poor fibre-matrix bonding. When the glass fibre layer fails, the fibre exhibits a clean fracture with some fragmentation of the epoxy matrix.

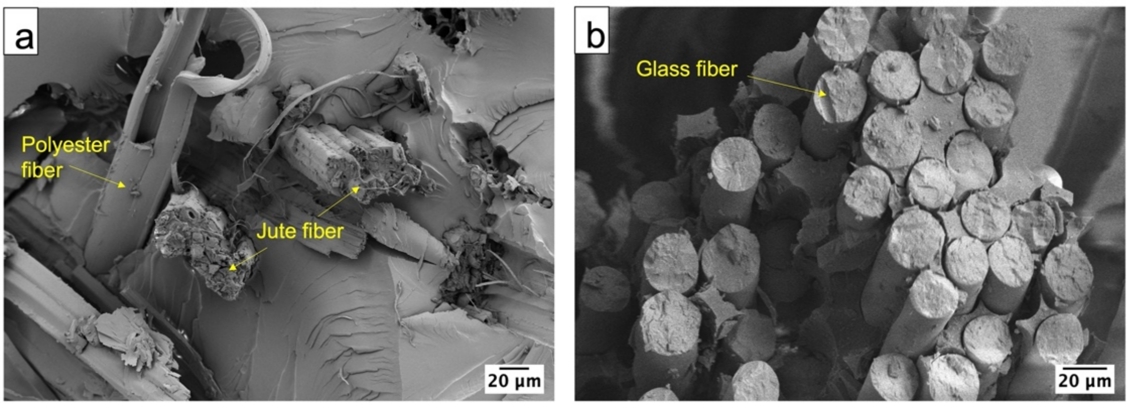


Figure 4.8 SEM images of the tensile fracture surface of (a) the HNB layer and (b) the Glass fibre layer.

The stacking sequence has a substantial effect on flexural strength and modulus. The flexural strength of the GHHG composite sample reached approximately 73% of the GG composite sample, while that of the HGGH layup was only 37% of the GG composite sample. As might be expected the flexural strength of hybrid composites is primarily determined by the z-position of the high-strength fibres (Khalil et al., 2016). In flexural tests, the failure usually begins at the outer layer and then gradually extends to the inner layer of laminates. As a result, the performance of the outer surface layer dominates the overall flexural properties of the hybrid composites (Selver et al., 2018). However, the influence of stacking order on the impact strength is less apparent. The impact strength of the GHHG and HGGH composite samples were similar at 405.45 and 397.71 KJ/m^2, respectively.

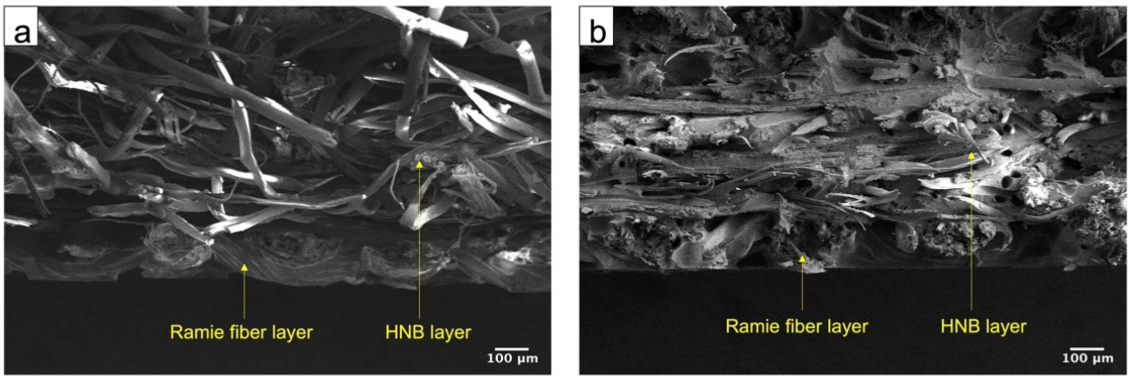


Figure 4.9 SEM images of the tensile fracture surface of the RHR* (a) and the RHR (b) samples.

The tensile strength of the RHR* composite sample was 32.47 MPa, approximately half of the strength of the RHR at 61.17 MPa. The RHRHR* composite sample exhibited the lowest tensile strength with 17.75 MPa. The lowest tensile modulus was also found in the RHRHR* composite sample with 0.45 GPa, while that of the RHR sample was the highest at 2.59 GPa. Figure 4.9 shows the tensile fracture surface of the RHR* and RHR samples. There is an evident difference between the two samples in the state of the HNB layer. In the RHR* sample, jute and polyester fibres are not entirely impregnated with resin and the fibre pull-out extends over a considerable length (around 0.7 mm). Large voids (around 0.01mm^2) can be seen in the HNB layer. In the RHR composite sample, the HNB layer is well impregnated, and the pull-out length is shorter than that in the RHR* sample. The strength and modulus will decline as the porosity increases (Madsen et al., 2009, Santulli et al., 2002). Accordingly, the tensile strength and modulus of the RHR* sample with 25% porosity were lower than those of the RHR sample. As the RHRHR* sample has the highest porosity (40%), its strength and modulus are the lowest of all three samples. The lower fraction of ramie fibre in the RHRHR* sample may also be responsible for the low mechanical properties since the plain weave structure has better fibre alignment than the nonwoven structure (Alavudeen et al., 2015). The flexural strength and modulus follow a similar trend to the tensile properties. The RHR sample gave the highest flexural strength with 79.33 MPa and flexural modulus with 4.88GPa. The flexural strength of the RHRHR* sample was reduced to 13.27 MPa. Its modulus was also the lowest at 1.73

GPa. The impact strength of the RHR sample was 17.47 KJ/m^2, while that of the RHR* sample was 72% higher. The RHRHR* composite sample had a slightly lower impact strength with 27.36 KJ/m^2 than that of the RHR*. After the impact test, the RHR sample exhibited a clean fracture, while the RHR* and the RHRHR* specimens failed to separate.

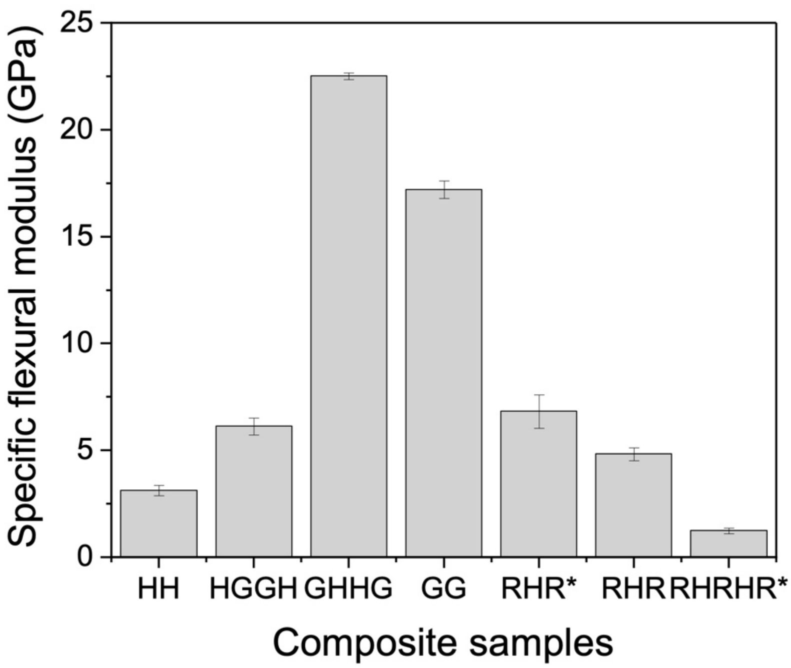


Figure 4.10 Specific flexural modulus of HNB, HNB/Glass fibre, and HNB/Ramie fibre composite samples.

The specific flexural modulus of the various composite laminates is compared in Figure 4.10. The hybrid GHHG composite sample emerges as the highest performing combination and exceeds the performance of the monolithic glass laminate GG. Despite variations in thickness among composites, differences in thickness do not exert a significant impact on the flexural modulus (Tanna et al., 2023). The specific modulus of the RHR* sample is the highest among the HNB/ramie fibre composite combinations and exceeds that of the glass cored layup (HGGH).

4.4.3.3 Damping properties of hybrid nonwoven breathers reinforced composites

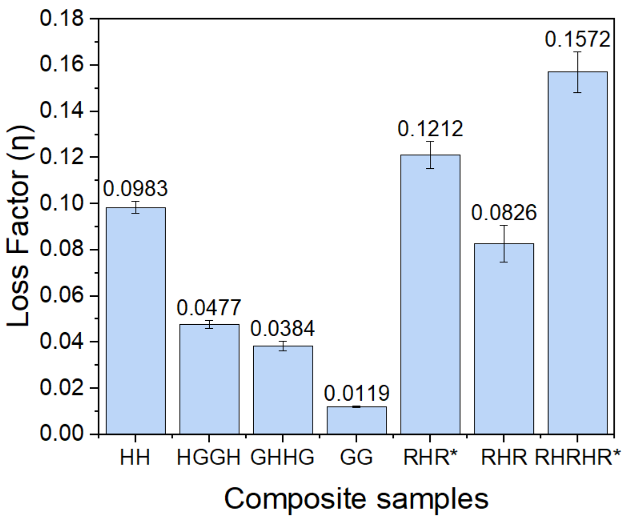


Figure 4.11 Loss factor comparison of HNB, HNB/Glass fibre, and HNB/Ramie fibre composite samples.

The vibration testing results of prepared composite samples are summarized in Figure 4.11. As a result of the manufacturing process and variations in volume fraction among samples, there are some differences in thickness among samples. However, according to Crane (Crane and Gillespie, 1991), the damping loss factor was found to be less sensitive to specimen thickness. The HH sample as discussed in 3.2.1 gave a high damping performance because of the high volume fraction of HNB. In contrast, the energy dissipation mechanism of the glass fibre composite is primarily determined by the epoxy phase (Cihan et al., 2019). Thus, the difference in loss factor between the HH and GG composite samples is rather large - the GG sample representing only 12% of the HH loss factor. The hybridization of the HNB and glass fibre results in an intermediate loss factor, lying between the HH and GG results. The loss factor of the GHHG layup sample was 0.0384. This is similar to the constrained damping structure (Ni et al., 2015) since the mid-plane HNB layer acts as an internal damping layer. The loss factor of the HGGH layup is 24% higher than the GHHG sample, which is attributed to the outer surface layer of HNB. Several studies have reached a similar conclusion since energy is primarily dissipated by the outermost plies (Assarar et al., 2015, Ben Ameur et al., 2018, Cihan et al., 2019).

The loss factors of all HNB/Ramie fibre composite samples were higher than those of the HNB/Glass fibre composite samples. Among the three HNB/Ramie fibre composite samples, the RHR composite sample had the lowest loss factor (0.0826), whereas the RHR* sample was 47% higher. The RHRHR* sample exhibited the highest loss factor at 0.1572. The low resin content of the RHR* and RHRHR* samples generally results in greater porosity and poor adhesion between the fibres and matrix, leading to more energy dissipation (Hadiji et al., 2020).

4.4.4 Comparison of different hybrid composites

The radar chart in Figure 4.12 illustrates the differences in thermal, mechanical and damping properties among HNB, glass, ramie fibre composites and their hybrids. There are axes aligned with the best values on the outside of the figure, indicating that better materials are represented by large swept-out areas. HNB/Glass fibre composites dominated the area on the bottom of the figure where mechanical and thermal properties are shown. Glass fibres with high thermal stability absorb heat and prevent the degradation of composites. Glass fibres also play a role in increasing the modulus of a composite as they are relatively stiffer than HNBs. However, this decreases the loss factor of HNB/Glass fibre composites since the microstructure of synthetic fibres has little energy dissipation. The density and damping properties of the HNB/Ramie composite are shown more prominently on the top of the figure. The porous structure in RHR* and RHRHR* composites improves energy dissipation ability and reduces the density of composites.

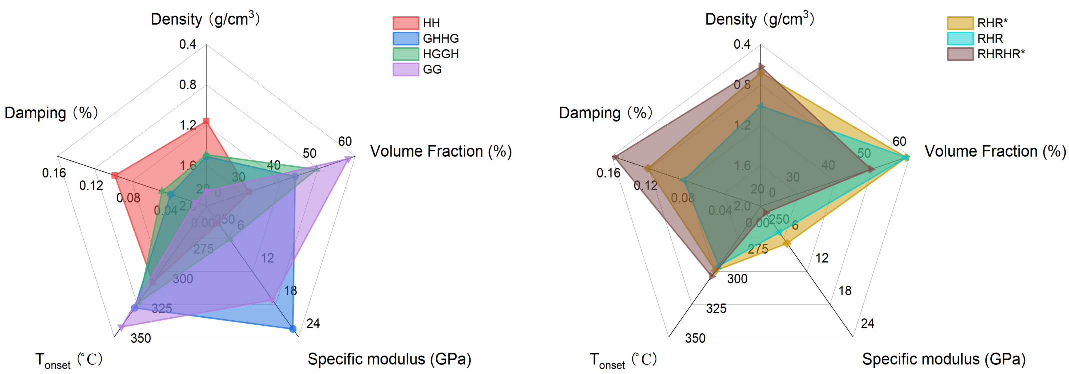


Figure 4.12 Summary of the findings on radar charts.

4.4.5 Structural damping properties of different hybrid composites

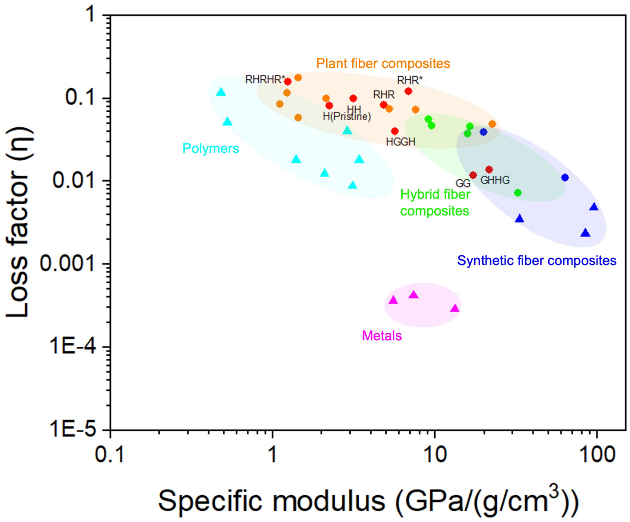


Figure 4.13 Loss factor-specific modulus plot of some common materials and the composites in the present study.

The low density is one of the most attractive characteristics of natural fibres. Therefore, the specific modulus was used to compare the different natural fibre combinations. Figure 4.13 compares the loss factor vs. specific modulus plot for some common materials at room temperature together with the data of the present study. The specific modulus was calculated based on the average density of the materials reported in the literature. The circles represent the test data obtained by modal tests (Cihan et al., 2019, Senthilrajan and Venkateshwaran, 2019, Cheung et al., 2009, Zhang et al., 2019), and the triangle represents the data collected from literature by DMA tests at 1 Hz (Ni et al., 2015, Liu et al., 2021). The metals exhibit a low loss factor, while the polymers are all significantly higher. The loss factor of fibre-reinforced composites is in the middle value. Composites made of synthetic fibres have the advantage of a high specific modulus. The loss factor, however, is relatively low for these structural fibre composites ranging from 0.002 to 0.04. Natural fibre composites typically exhibit higher damping properties, while a gap remains for materials with intermediate modulus and damping properties.

The product of Young's modulus and loss factor is usually used to estimate structural damping properties(Ni et al., 2015, Lakes, 2002, Yaw Attahu et al., 2022, Gao et al., 2018). Furthermore, the product of specific Young's modulus and loss factor is defined as the figure of merit (FoM) for structural damping. Figure 4.14 shows the computation of FoM for the HNB, HNB/Glass fibre, and HNB/Ramie fibre composite samples, respectively. The FoM of the GG and RHRHR* samples is found the lowest, whereas GHHG and RHR* samples provide the highest FoM values arising from the contribution of the HNB interlayer, even when using recycled HNB. Overall, the test results suggest that HNBs can be useful in tailoring the structural damping properties of laminated composites to meet certain engineering needs.

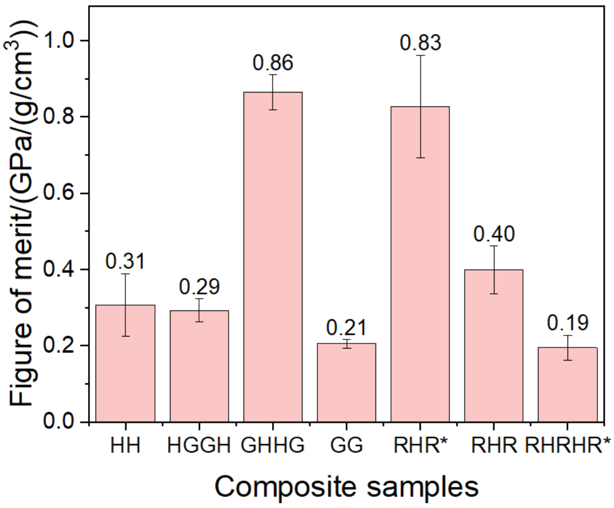


Figure 4.14 The figure of merit of the composites in the present study.

4.5 Conclusions

This study offers insight into sustainable composite manufacturing through the application of the jute/polyester hybrid nonwoven mat as a *green* breather, and the re-use the end-of-life hybrid nonwoven breathers (HNBs) to reduce manufacturing waste, improve material utilization, and foster a circular economy strategy in multiple composite applications. The HNBs were initially applied in an autoclave to channel air during the manufacturing of CFRP composites. It is evident from the mechanical and C-scan results that the HNBs are viable alternatives to commercial synthetic fibre breathers. The used HNBs were reused as reinforcement to manufacture HNB, HNB/Glass fibre and HNB/Ramie fibre composite samples with different layout designs. The HNBs improve the mechanical and damping performance of epoxy. The HNB/Glass composite samples have a significant increase in thermal stability and mechanical properties. The stacking sequence has little influence on the tensile and impact properties, whilst the improvement in flexural properties is evident when the glass fibre is placed at the outer layer. The HNB/Ramie fibre composites get boosted damping properties, achieving 0.1572 in the RHRHR* composite sample. Concerning the figure of merit, the HNB hybrid composites yielded good structural damping properties compared with competing materials. The low density, high modulus, and high damping properties of the end-of-life HNBs can thus be adapted to a variety of semi-structural applications such as aerospace or automotive parts, and construction material.

Chapter 5

Design and Development of Jute/Polyester Nonwoven Reinforced Foams for Enhanced Mechanical and Acoustic Capabilities

5.1 Overview

Syntactic foams comprised of expandable microspheres reinforced with natural fibre were fabricated using an epoxy resin binder. Jute/polyester nonwoven mats were used as a skeleton structure with ramie fabric outer skins. By adjusting the reinforcement geometry and the content of microspheres, the density was varied from 0.26 to 0.56 g/cm^3, resulting in diverse mechanical and sound insulation behaviours. The flexural modulus of fibre mats reinforced foams exceeded unreinforced foams but was lower than that of pure epoxy. The addition of ramie fabric face sheets enhanced both the flexural strength and modulus of composite foams and the specific modulus of composite foams was around twice that of pure epoxy. The sound insulation properties of the epoxy foams were higher than pure epoxy due to the increased density and modulus provided by the reinforcements. This novel lightweight epoxy foam holds promise as a multi-functional material with applications in various systems, including vehicles, buildings, and aircrafts.

5.2 Introduction

The development of lightweight composites in engineering applications has become increasingly important due to the growing demand for energy efficiency and emission reductions (Wu et al., 2018). Syntactic foam, a hollow particle filled polymer composite has strong potential for weight-critical applications (Sankaran et al., 2017, Afolabi et al., 2020). The combination of hollow microspheres and matrix materials offers an excellent opportunity to optimise the microstructural morphology of syntactic foams and thereby their thermo-mechanical properties. Such microsphere-structured materials have attracted growing interest and have potential to replace conventional materials in a range of applications, from domestic to aerospace and defence, due to their superior thermal insulation (Xue et al., 2019), dielectric properties (Chan et al., 2012), noise attenuation properties (Xu et al., 2017, Xue et al., 2019) and good mechanical properties (Kumar and Ahmed, 2014, Wang et al., 2014). Expandable microspheres typically consist of a thermoplastic shell encapsulating a volatile hydrocarbon liquid (Jr and Tetreault, 1971). When heated, the liquid core vaporizes, and the thermoplastic shell softens and expands due to the increasing internal gas pressure. Compared to other hollow microspheres, expandable microspheres have relatively low density, excellent foaming properties, and low prices. They can be used in a wide range of matrix types and formulations since the foaming process is not closely correlated with the curing reaction.

Table 5.1 Average SLT values of common barrier materials (Federal Highway, 2000, Öztürk et al., 2012).

Material	**Details**	**Thickness (mm)**	**Surface density (Kg/m^2)**	**STL (dB)**
Concrete	Light Weight Concrete	100	151-161	36
		150	244	39
	Dense Concrete	100	244	40
	Concrete Block	200	151	34
Woods	Fir, Pine, Redwood	12-50	8.3-32.7	18-24
	Plywood	12-25	8.3-16.1	20-23
	Particle Board	13	/	20
Metal	Aluminium	1.5-6	4.4-17	23-27
	Steel	0.5-1.3	4.9-10	18-25
	Lead	1.5		28
Composite	Aluminium-Faced Plywood/ Particle Board	19	/	21-23
	Plastic Lamina on Plywood/Particle Board	19	/	21-23
Miscellaneous	Glass（Safety Glass）	6	7.8	22

Masonite	13	/	20
Fibre Glass/Resin	6	/	20

In addition to weight reduction, syntactic foams can be formulated to function in complex environments (Sankaran et al., 2017), including the possibility of multifunctional applications. One important application of hollow filler material is sound insulation. Table 5.1 provides typical sound transmission values for commonly used materials. It is seen that conventional materials with high density and thickness are typically better at reducing sound transmission (Herrera and Recuero, 2010). Thin, lightweight soundproof materials are of great interest to both industry and academia. Closed-cell structured fillers such as hollow glass microsphere and carbon or silicon nanotubes have been proven to have good sound insulation properties in polymer composites due to the energy absorption within the hollow microspheres (Kim et al., 2013, Shi et al., 2017, Zhang et al., 2021). However, these materials can be intrinsically expensive or energy-intensive to fabricate. The use of microspheres within syntactic foams can improve sound attenuation, but this often comes at the cost of decreased mechanical properties. Balancing the trade-off between sound attenuation and mechanical properties in syntactic foams is challenging but formulation parameters, such as matrix composition, microsphere size, wall thickness, and volume fraction, can all be adjusted to tailor the mechanical properties of synthetic foams (Curd et al., 2021). Wouterson and Wong et al. demonstrated that the tensile properties of synthetic foams were significantly improved by 3 wt% short carbon fibre reinforcement (Wouterson et al., 2007). However, fibre length effects mean that continuous fibres are more effective, for example a PVC composite foam reinforced with long aramid fibres had significantly improved properties over unreinforced PVC foam (Huang, 2009). Reaction injection moulding has been utilized with a blowing agent to infiltrate the mat and foam the core. Recently, recycled and renewable materials have gaining increased interest as alternatives to petroleum-based materials (Song et al., 2020, Cho et al., 2022). Natural fibre reinforced materials also have potential in a variety of structural applications due to their low density and high specific modulus (Pickering et al., 2016, Takagi, 2019) and their enhanced sustainability compared to synthetic fibres and here we explore their potential uses with composite foams.

In the present study, epoxy resin was chosen as the matrix material due to its widespread use in construction and automotive applications, as well as its high strength and compatibility with fillers (John and Reghunadhan Nair, 2022). The aim was to develop natural fibre reinforced epoxy/expandable microsphere foams and evaluate their mechanical performance and their potential as noise insulation materials. Three types of composite foams were fabricated: epoxy foams, mat reinforced foams and mat reinforced foams incorporated with face sheets, which were loaded with various contents of expendable microspheres. The density, component volume fraction and porosity of the prepared foams were varied. The mechanical properties and sound attenuation characteristics of these materials are characterized with respect to structure and expandable microsphere content and properties are compared to pure epoxy control.

5.3 Materials and methods

5.3.1 Materials

Expancel 093DU120 thermally expandable microspheres (EMS) with an average density ≤ 6.5 kg/m^3 were supplied by AkzoNobel (Sweden). The EMS initiates foaming at a temperature of 120°C. Upon heating, these expandable foam microspheres swiftly expand from an initial diameter of 28-38 μm to a maximum diameter of 120 μm. Araldite® LY 1572 epoxy and hardener 3846 were sourced from Huntsman Corporation, USA. Jute/polyester 40/60 fibre mats with an areal density of 315 g/cm^2 were produced by needle punching. The thickness of the fibre mats was 5.89 mm. The average length of jute fibre was ~ 67 mm and that of polyester fibre was ~ 38 mm. See Ref. (Tong et al., 2021) for further details regarding the manufacture of the fibre mat. Plain weave ramie fabric with an areal density of 123 g/m^2 was obtained from Hunan Huashengdongting Ramie Co., Ltd, Yueyang, China and used as received. Table 5.2 shows density of fibres used in this study. Digital images of the fibre mat and ramie fabric are shown in Figure 5.1.

Table 5.2 Density of fibres in jute/polyester fibre mat and ramie fabric.

Fibre type	Density (g/cm3)
Jute fibre	1.48
Polyester fibre	1.39
Ramie fibre	1.50

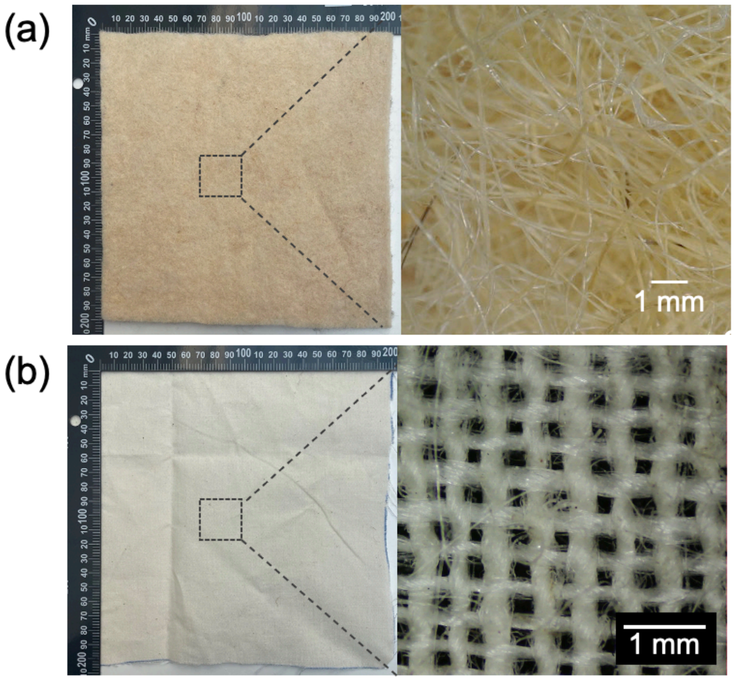


Figure 5.1 Digital images of (a) jute/polyester fibre mat, and (b) ramie fabric.

5.3.2 Fabrication of epoxy foams

Pure epoxy foams, fibre mat reinforced foams, and fibre mat/ramie fabric reinforced foams were fabricated as indicated in Figure 5.2. To form the epoxy/expandable microspheres foams, up to 6 wt.% of expandable microspheres were first added to the epoxy matrix (67 wt.% epoxy and 33 wt.% hardener) and mechanically mixed. The resulting slurry was poured into a

PTFE-lined stainless-steel mould with a dimension of 300 × 300 × 5 mm. The mould was covered with a stainless-steel plate and placed in a preheated hot press at 130 °C and a pressure of 1 MPa was applied for 20 min. Upon heating, the microspheres expanded to fill the mould, with the resin dispersed among the expanding microspheres. Foaming and curing occurred successively during this period.

To fabricate the jute/polyester mat reinforced foams (EMS/FM/EP) and jute/polyester mat/ramie fabric reinforced foams (EMS/FM/RF/EP), the jute/polyester mat and ramie fabric were cut to match the mould dimension. They were placed in the mould after the epoxy slurry was poured into it, depressing the reinforcement sufficiently to become infiltrated. The foaming and curing processes were as described above. The foam was evenly dispersed among the fibres of the jute/polyester mat and connected the top and bottom ramie fabrics. In EMS/FM/RF/EP foams, the jute/polyester mats ensured a uniform distribution of fibres in the matrix. As for EMS/FM/RF/EP foams, the jute/polyester mat supported the top and bottom of the ramie fibre fabric. In the foaming process, the ramie fabric skin and the jute/polyester mat core are filled with the same matrix forming an integral core-skin structure without the need for adhesives.

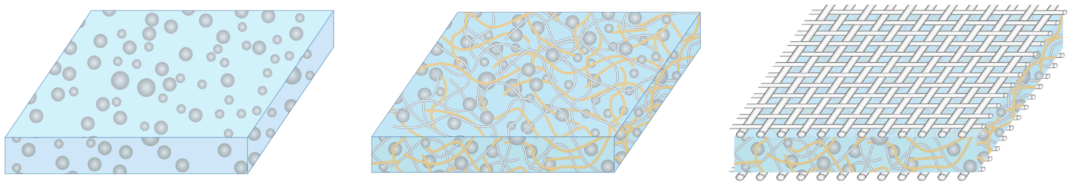


Figure 5.2 Schematics of epoxy/expandable microspheres foam (a), jute/polyester fibre mat reinforced foam (b), jute/polyester fibre mat reinforced foam with ramie fabric face sheets (c).

5.3.3 Measurements and characterization

5.3.3.1 Density and porosity measurement

The density of composite epoxy foams (D_f) was calculated based on ISO 845. Specimens with a minimum volume of 100 cm^3 were cut without deforming the original cell structure. The density was determined by dividing the volume by the mass.

The porosity (P) of epoxy foams was calculated by the following equation:

$$P = \left(1 - \frac{D_f}{D_a}\right) \times 100 \quad (5.1)$$

Where D_a is the average density of the constituent epoxy resin and fibres.

5.3.3.2 Flexural tests

Three-point flexural tests were performed on an MTS E42 universal test machine (MTS Systems Corporation, USA) in accordance with ASTM D 790. The span length was maintained constant at 80 mm and the specimen dimension was 13 × 5 × 120 mm. At least five specimens of each syntactic foam type were tested. The crosshead displacement rate was maintained at 2 mm/min.

5.3.3.3 Morphology analysis

After flexural testing, the fracture surface was cut from the specimen. A sputter coating of gold was applied to the fracture surface before SEM examination. The morphological characteristics of specimens were assessed by a Phenom pro scanning electron microscopy supplied by Lambda Photometrics Ltd, UK. The acceleration voltage was configured to 15 kV, and a backscattered electron detector was employed. The working distance was set around 8mm.

5.3.3.4 Sound insulation tests

The sound insulation performance of epoxy foams was measured by its sound transmission loss (STL) using a four-microphone impedance tube (BSWA Technology Co., Ltd) in accordance with ASTM E2611. The samples were machined into discs with diameters of 29.8 mm and 99.8 mm. The thickness of all the specimens was 5 mm. The 30 mm and 100 mm diameter tubes were used for the test frequency range of 63 Hz - 1600 Hz and 1000 Hz-6300 Hz, respectively. The average STL was calculated from the 1/3 octave values. A single-range curve was produced by combining the low and high frequencies using VA-Lab software. Each sample was tested three times to determine the average test curve.

Figure 5.3 illustrates the measurement setup. Sound waves are generated by the speaker and four microphones are used to measure the sound levels. *A* represents the incident sound wave, *B* represents the reflected sound wave, *C* represents the panel-transmitted sound wave, and *D* represents the panel-reflected sound wave. The STL is calculated by a transfer function method as follow:

$$STL = 10 \times log_{10} \frac{1}{\tau} \tag{5.2}$$

Where τ is the ratio of the transmitting sound energy to the incident sound energy.

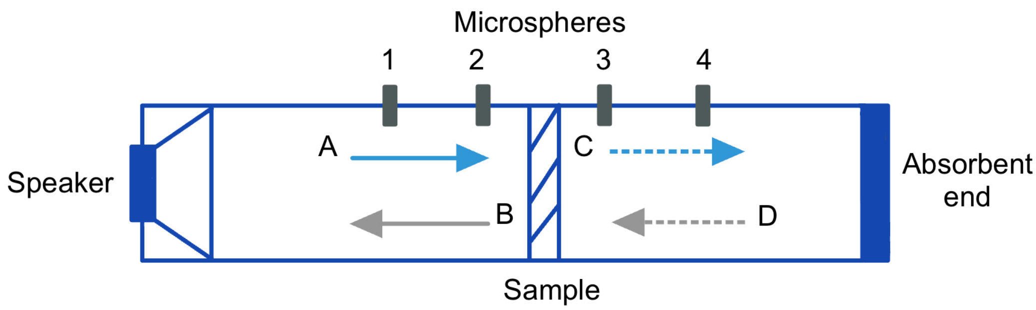


Figure 5.3 Scheme of four-microphone impedance tube for measuring sound transmission loss.

5.4 Results and discussion

5.4.1 Structure and density of foams

Table 5.3 Porosity and density of pure epoxy and epoxy composite foams.

Sample code	EMS content	Porosity (%)	Density (g/cm^3)	Weight saving potential (%)
EP	/		1.2	/
EMS/EP	2%	59	0.49	59
	4%	69	0.37	69
	6%	78	0.26	78
EMS/FM/EP	2%	57	0.53	56
	4%	66	0.42	65
	6%	75	0.31	74
EMS/FM/RF/EP	2%	54	0.56	53
	4%	63	0.46	62
	6%	72	0.35	71

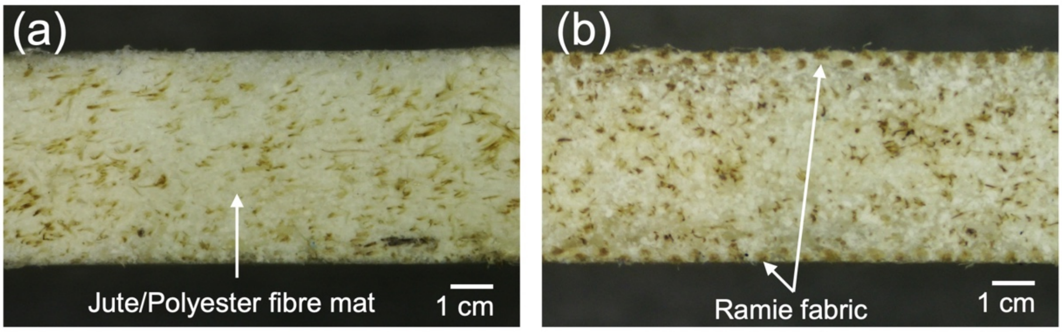


Figure 5.4 Digital images of the cross section in the thickness direction of (a) EMS/FM/EP and (b) EMS/FM/RF/EP foams.

The density and porosity of the different samples are summarized in Table 5.3. In comparison to pure epoxy resins, the epoxy foams offer substantial weight reductions, with weight portions decreasing from 78% to 53%. With increasing EMS content, the foams exhibit rising porosities. Unsurprisingly the 6%

EMS/EP foam had the highest porosity and the lowest density. Compared with EMS/EP foams, the density of jute/polyester mats reinforced epoxy foams (EMS/FM/EP) increased around 8-19%. By adding jute/polyester mats, a portion (4%) of volume is occupied by fibres, resulting in a lower porosity than EMS/EP foams with the same EMS contents. When adding ramie fabrics as face sheets, 3% volume fraction of EMS/FM/RF/EP foams was occupied by the fabric. The density of samples was 14-35% higher than that of equivalent EMS/EP foams. Figure 5.4 displays cross-sectional images of EMS/FM/EP and EMS/FM/RF/EP foams in the thickness direction. The cross-sections of jute fibres, polyester fibres, and ramie yarn can be seen in the figure. It is observed that the jute/polyester fibres are evenly distributed within the foams, and the ramie fabric layer is distributed uniformly on the surface of the EMS/FM/RF/EP foam.

5.4.2 Flexural properties of foams

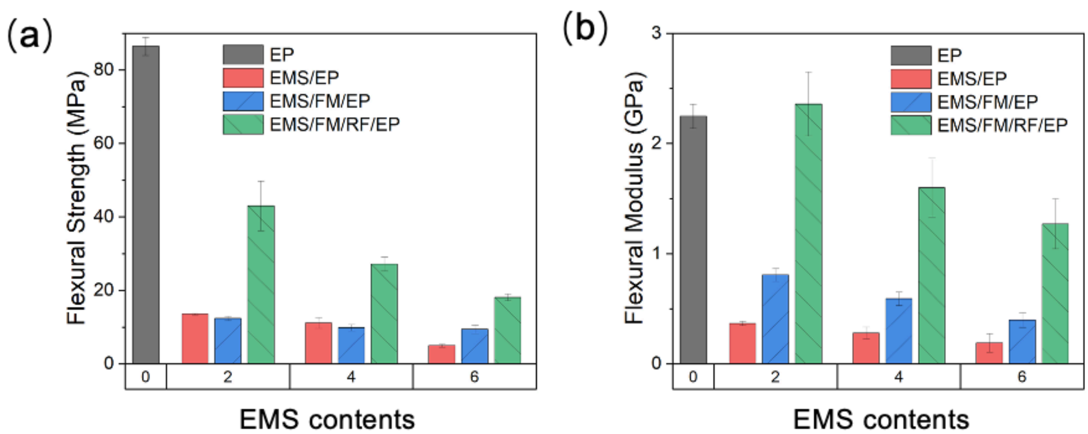


Figure 5.5 Flexural strength (a) and flexural modulus (b) of epoxy foams with different structures containing various amounts of EMS.

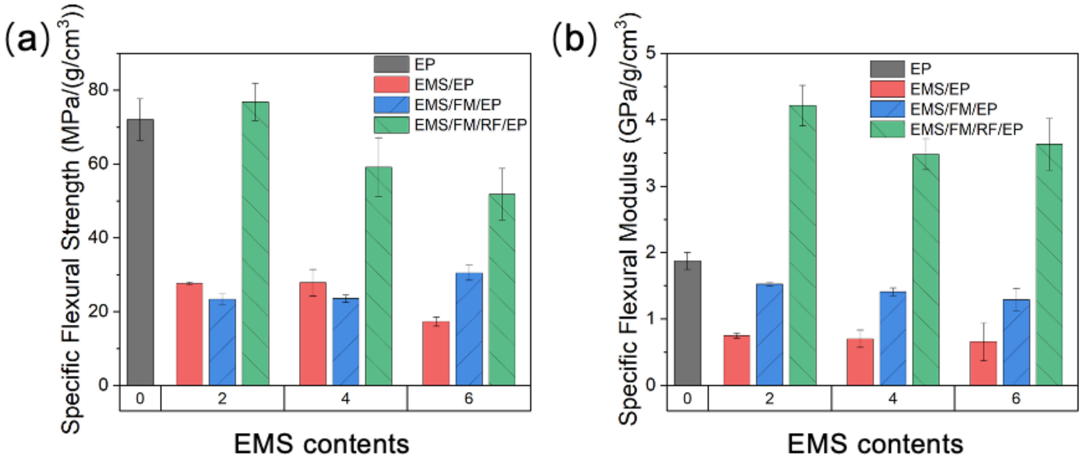


Figure 5.6 Specific strength (a) and specific modulus (b) of epoxy foams with different structures containing various amounts of EMS.

Figure 5.5 demonstrates the flexural properties of the different epoxy foams with various EMS contents and the pure epoxy control. The flexural strength and modulus for EMS/EP foams dropped dramatically as porosity was introduced and so the available cross section area was reduced, confirming that the resulting mechanical performance was profoundly affected by the foam structure. The flexural strength of EMS/EP foams with 2% EMS content decreased rapidly to 13.56 MPa. With increased EMS contents, the flexural strength decreased to 5.01 MPa for EMS/EP foams with 6% EMS content. Thus, the flexural strength was primarily influenced by the resin content of the composite foams. The flexural modulus of EMS/EP foams dropped to the lowest value of 0.19 GPa with 6% EMS content. Similar results have been found in the literature that the flexural strength decreases with the inclusion of foam structure (Cavasin et al., 2018). The voids introduced during foaming cause epoxy materials to lose a great deal of strength and modulus (Xue et al., 2019).

There are mainly three phases in EMS/EP foams including matrix, microspheres, and internal voids. During the loading process, voids in epoxy foams will initiate micro cracking which later coalesce into catastrophic fractures (Huang et al., 2016). The decline of flexural properties is also attributed to the existence of agglomerations in EMS/EP foams, which would increase the stress concentration of the microspheres locally (Yu et al., 2013). Under the same foaming condition, a higher EMS content would lead to proportionally more agglomerations, with a negative effect on flexural properties.

The addition of jute/polyester mats increases the flexural modulus of the composite epoxy foams, but the increase in flexural strength is only seen in materials with 6% EMS content. Fibres in the mats are loosely intertwined and randomly oriented, reducing both packing density and orientation efficiency (Norman and Robertson, 2003). Therefore only a few parts of fibre length is effective for discontinuous reinforcement due to the angles between fibres and the loading axis (Wouterson et al., 2007). The fibres can absorb energy transferred from the matrix and the fractured fibres are responsible for the enhanced flexural properties (Huang et al., 2016). On the other hand, weak fibre/matrix interfaces can also cause mechanical properties to be compromised. One possible explanation for the diminished flexural strength observed in 2% and 4% EMS/FM/EP foams, in contrast to EMS/EP foams, could be attributed to insufficient bonding between the epoxy and untreated fibres, potentially negating the reinforcing effects of the fibres (Doan et al., 2012). However, the reinforcement effect of fibre mats can vary in different densities of foams. In low density foams, the pore dispersion coefficient increased with the incorporation of fibre reinforcement (Li et al., 2023). The increased flexural strength observed in 6%EMS/FM/EP foams can be attributed to the enhanced distribution of pore structure facilitated by the fibre mat. The flexural modulus of jute/polyester mat reinforced epoxy foam with varying EMS contents increased by approximately two times than the EMS/EP foams. The strength and modulus of jute fibre measured in previous research were 449 MPa and 30 GPa, respectively (Tong et al., 2021). The improvements in both strength and modulus of jute/polyester reinforced foams were mainly attributed to the jute fibres (Biswas et al., 2015). As for EMS/FM/RF/EP foams, the flexural strength and modulus are higher than EMS/EP and EMS/FM/EP foams. The flexural modulus of 2% EMS/FM/RF/EP foam is even 5% higher than that of epoxy resin. Ramie fibres are known for having one of the highest values of Young's modulus and tensile strength among natural fibres, with strength ranging from 400 to 900 MPa and a modulus ranging from 30 to 60 GPa (Du et al., 2015). The presence of ramie fabric face sheets enhanced the flexural strength and modulus of composite epoxy foams. As shown in Figure 5.6, the specific bending modulus is reduced for the jute/polyester specimens but the materials incorporating ramie show a twofold improvement over the epoxy control due to the sandwich panel effect.

5.4.3 Morphology of foams

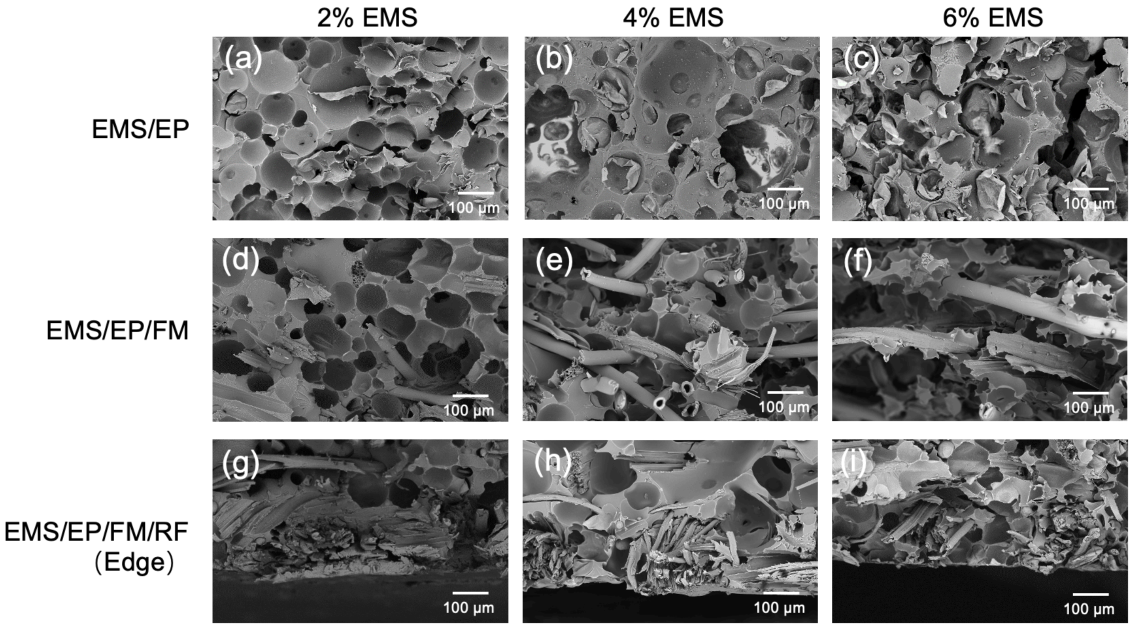


Figure 5.7 Typical SEM images of the fracture surface of various EMS content of epoxy foams with different structures: (a-c) EMS/EP, (d-f) EMS/FM/EP, (g-i) edges of EMS/FM/RF/EP specimens.

Figure 5.7 compares the morphology of prepared composite epoxy foams with different EMS contents. Due to the use of EMS blowing agents, every sample has a closed-cell structure. Some open pores are caused by the breaking of the microsphere shells. It was analysed by ImageJ that the mean diameter of 2% EMS/EP foams ranged from 80-130 μm and the average diameter was 100 μm. For samples with higher EMS content, small microspheres were dispersed among microspheres, and fractured microspheres were occupied by small microspheres. It is likely that samples with 4% EMS content produced more heat during the foaming process, resulting in higher local temperatures, and some microspheres expanded sufficiently to rupture and agglomerate within the foams (Yu et al., 2013). When the microsphere content was 6 wt.%, excessive microspheres collision, resulted in the thinning cell walls and more cases of microsphere rupture. Therefore, more debris is found in Figure 5.7 (c).

Figure 5.7 (d-f) shows the fracture surface of jute/polyester mat reinforced epoxy foams (EMS/FM/EP). The foamed microspheres are evenly dispersed among fibres and the length of the exposed fibres increased as the content of foamed microspheres increased. In the 2% EMS/FM/RF/EP foam, exposed jute and polyester fibres are short and some of jute fibres break in the foam matrix, indicating a strong interface between fibre and matrix. For the sample with 4% and 6% EMS content, gaps are observed between fibre and matrix, since the content of the resin largely determines the strength of the material. Therefore, debonding is more likely in higher EMS content samples, which reflects poor mechanical performance (Borges et al., 2017). Other studies have demonstrated that fibres embedded in foams may preferentially break rather than pull out during fracture events, again supporting suggesting evidence of effective interfacial bonding (Vaikhanksi and Nutt, 2003, Huang, 2009).

Figure 5.7 (g-i) shows a distinct region of skin of jute/polyester mat foams with ramie fabric face sheets, which indicates an integral bonding between ramie fabric and matrix. The thickness of the face sheets is around 0.2 mm. Fibres in the warp direction are broken, while those in the weft direction are pulled out from the matrix.

5.4.4 Sound insulation properties of foams

The trends of STL as influenced by sound frequency for three kinds of foam structures with different EMS contents are illustrated in Figure 5.8 (a-c), and the average STL value of all epoxy foams is shown in Figure 5.8 (d). STL is strongly correlated with frequency, and the curve can be divided into two regions. From 300-800 Hz, the first resonance frequency is achieved, where the high level of displacement causes high sound transmission, resulting in a minimum value of STL. At frequencies the STL is mainly controlled by the stiffness and resonance of materials. Above the first resonant frequency, the STL values generally increase, which is governed by the mass of materials and adheres to the mass law stating that the STL is inversely proportional to its areal density (Hao et al., 2013). The resonance frequency can be calculated as follows (Hopkins, 2012):

$$f_0 = \frac{1}{2\pi}\sqrt{\frac{k}{m}} \quad (5.3)$$

where f_0 represents the resonance frequency of the material, k is the spring stiffness of the material, and m is the mass of the material.

It is seen from Figure 5.8 (a-c) that the first resonant frequency of EMS/EP with 2% and 4% EMS contents and EMS/FM/EP foams was 630 Hz, while that of 6% EMS/EP foams decreased to 315 Hz. The value for EMS/FM/RF/EP foams shifted to 800 Hz. This is attributed to the twofold increase in stiffness and decrease in mass. Thus, the highest specific modulus coincides with the highest resonance frequency.

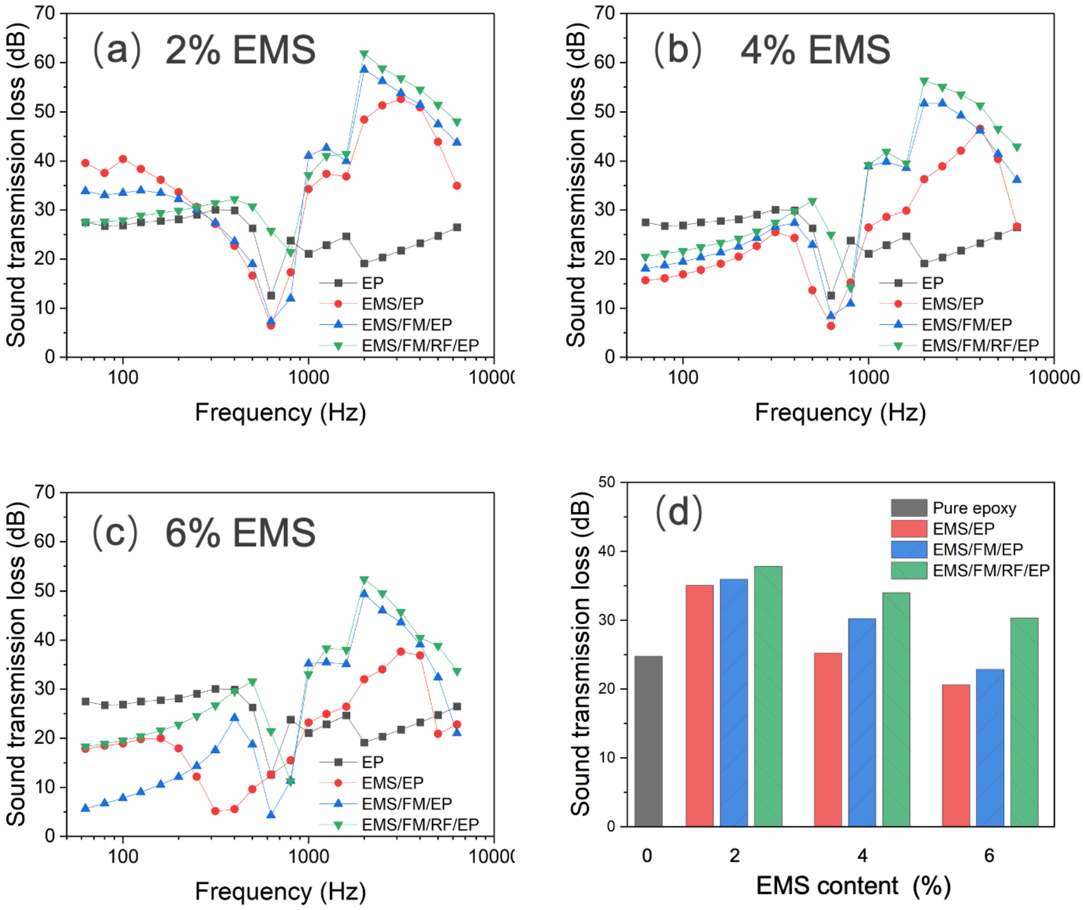


Figure 5.8 Effect of different structures on sound transmission loss of epoxy foams with 2% EMS content (a), 4% EMS content (b), and 6% EMS content (c), and average sound transmission loss value of all samples (d).

Figure 5.8 (a) shows that the sound transmission loss (STL) of epoxy foams with different structures is higher than that of pure epoxy resin, both in the low and high frequency ranges. The average STL value of pure epoxy resin is quite minor, at 24.82 dB. The average STL value of 2% EMS/EP foams increases to 32.57 dB, which is 31% higher than that of pure epoxy. Since sound energy is halved when the sound level is reduced by 3 dB (Ozen et al., 2019), the expandable microspheres provide satisfactory sound insulation properties for epoxy foams. This is contrary to the mass law in acoustics, and it is explained that the introduction of pore structures improves the sound insulation properties of materials (Xue et al., 2019, Shin et al., 2020).

The sound energy dispersion of foams with pore structures can be explained by three factors (Zhang et al., 2013, Shi et al., 2017, Fei et al., 2018). Firstly, by adding microspheres to the matrix, the crosslinking density is reduced, which improves damping properties. The matrix can thus absorb the mechanical vibration energy, dissipating some of the sound energy. Secondly, microspheres and epoxy matrix interfaces as well as microsphere and inner air interfaces can scatter, diffract and refract sound waves. Moreover, the narrow space within the microspheres prevents air from moving freely, dampening sound waves.

In addition to pore structures, the acoustic insulation capability of materials can be significantly influenced by their stiffness and mass. In stiffness control region, STL increase with the increase of the elastic modulus of material (Berry and Nicolas, 1994, Wang et al., 2011). In mass control region, the specimen vibrates in response to a sound wave, and vibration energy dissipation amplifies with the growing weight of the material during transmission (Hao et al., 2013). Therefore, as the EMS content increases to 4% and 6%, the density and modulus of the foams decreased, resulting in the decreased average STL value of 24.97 dB and 20.20 dB, respectively. It is seen from Fig. 9 (b-c) that the reduction mainly occurs in the low-frequency range. Similar results have been found in the literature that over a certain loading of microspheres, the STL actually decreases (Xue et al., 2019).

Fig. 9 (d) illustrates that composite foams containing fibre mat reinforcement exhibit higher STL values compared to EMS/EP foams with an equivalent EMS content in the matrix. Additionally, EMS/FM/RF/EP foams demonstrate higher STL values than both EMS/EP and EMS/FM/EP foams, with highest average STL value of 35.9 dB in 2% EMS/FM/RF/EP foam. The luminal structure of natural fibres has been shown to trap air pockets, thereby reducing the transmission of sound waves through the structure (Singh and Mukhopadhyay, 2022). Besides, both fibre mat and ramie fabric reinforcements increase the density of composite foams as well as the elastic modulus. Therefore, the average STL of EMS/FM/EP and EMS/FM/RF/EP foams increase on the basis of stiffness and mass law.

5.4.5 Performance comparison

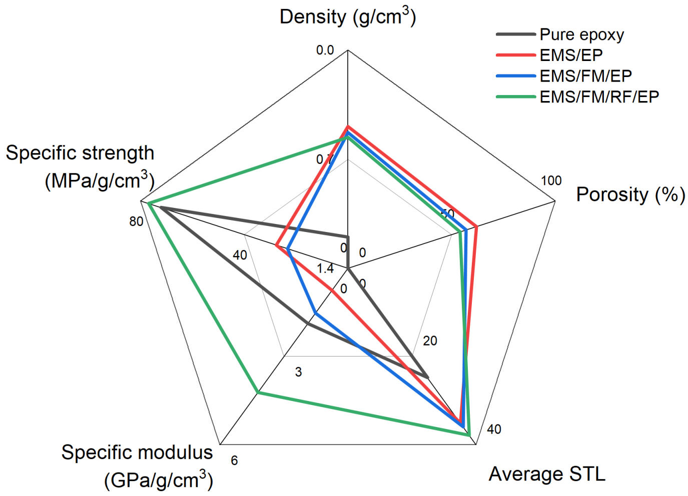


Figure 5.9 Radar chart for the overall properties of the natural fibre reinforced foams with 2% EMS content compared with pure epoxy.

In order to evaluate the mechanical, acoustic and structural performance between composite epoxy foams and pure epoxy, the radar chat of Figure 5.9 provides a comparison of the density, specific strength, specific modulus, average STL, and porosity. The chart shows that the three types of foams tested offer advantages in density and sound insulation due to the porous structure introduced by EMS. However, the pores negatively impact flexural strength and modulus which limits their use in applications requiring high mechanical performance. To address this, fibre mat reinforced foams can effectively increase flexural modulus while maintaining low density. For optimal performance, ramie fabric as face sheets offers a balance between high STL loss and mechanical performance. The epoxy foams introduced in this chapter stand out as a superior option, particularly in their exceptional sound insulation capabilities, low density, and thickness advantages when compared to traditional materials like concrete, steel, and wood. The low density, high sound attenuation and improved mechanical properties of the presented epoxy foams make them candidates for the construction of lightweight structural components where noise and weight reduction are critical, such as in the construction, automotive and aerospace industries.

5.5 Conclusions

In this study, three variants of epoxy-based composite foams, each with varying content of expandable microspheres (EMS) and reinforced with fibre mat (FM) and ramie fabric (RF), were produced through the mould compression method. The foams, designated as EMS/EP, EMS/FM/EP, and EMS/FM/RF/EP, contained expandable microspheres at 2 wt.%, 4 wt.%, and 6 wt.% in the matrix. The density of foams ranges from 0.26 g/cm^3 to 0.56 g/cm^3, which decreased with increasing EMS content, but increased with the addition of fibre reinforcements. When compared with pure epoxy, the porous structure of foams adversely affects the flexural properties. The flexural modulus was effectively improved by the fibre mat reinforcement. On the other hand, the enhancement of flexural strength only worked in the fibre mat reinforced foams with 6% EMS content. The use of ramie fabric as face sheets further increased the bending strength and modulus of the material. Microscopic analysis revealed the distribution of microspheres and the fibre-matrix interfacial bonding and the connection between the core and face sheet. The sound transmission loss was evaluated using the four-microphone impedance tube method. Experiments showed that expandable microspheres provide satisfactory sound attenuation, but this declined as the porosity increased. The average sound transmission loss of fibre reinforced epoxy foams (EMS/FM/EP and EMS/FM/RF/EP) was higher than EMS/EP foams with the same EMS contents, which is due to the improved density and modulus provided by the reinforcements. The results offer a novel approach to the fabrication of fibre reinforced epoxy foam with enhanced mechanical properties and sound insulation performance.

Chapter 6

Conclusion and Future Work

6.1 Overall summary

The thesis focuses on sustainable manufacturing in the field of composite materials, aiming to develop eco-friendly materials, reduce waste generation, and explore recycling possibilities. The study begins by fabricating jute/polyester hybrid breathing materials specifically designed for use as breathers in composite manufacturing. The results demonstrate that the hybrid breathing materials have higher permeability than commercially available breathers and can be reused multiple times while maintaining a significant portion of their original permeability. The study than proposes a circular economy strategy to manage end-of-life hybrid breathing materials. The used hybrid breathers are recycled and repurposed as reinforcing materials in the production of new composites. The mechanical and damping properties of these recycled composites are evaluated, showcasing their potential as alternatives to synthetic materials in semi-structural applications. In addition, the study fabricates the hybrid breathing materials reinforced epoxy foams with enhanced mechanical properties and sound insulation performance, offering potential applications in various industries. The findings highlight the potential of hybrid breathing materials in sustainable composite manufacturing through improving material utilization and exploring novel materials and fabrication techniques for enhanced composite properties.

6.2 Major contributions

This thesis contributes to advancing sustainable composite manufacturing, improving material utilization, and exploring novel materials and fabrication techniques for enhanced composite properties.

The study fabricated jute/polyester hybrid breathing materials through the needle-punched method, specifically designed for use as breathers in composite manufacturing. These materials exhibit high permeability even under demanding high-temperature and high-pressure conditions. They can be reused for at least three cycles under 1 bar pressure and can potentially be repurposed as raw materials for composite production, extending their lifecycle and promoting sustainability.

The study highlights the possibility of reusing end-of-life hybrid breathing materials as raw materials for composite production. This approach contributes to reducing manufacturing waste and fostering a circular economy strategy within the composite industry. The utilization of hybrid nonwoven breathers leads to improved mechanical properties and damping performance in epoxy-based composites.

The study introduces a novel approach to fabricating fibre-reinforced epoxy foam using expandable microspheres and fibre mat or ramie fabric reinforcement. This fabrication method results in foams with enhanced mechanical properties and sound insulation performance.

6.3 Future work

The jute/polyester hybrid materials cannot be directly discarded or decomposed due to the presence of non-degradable polyester fibre derived from petroleum-based sources. It is essential to develop all-green breathers using bio polymers that are suitable for a wide range of applications. These bio-based hybrid breathing materials should offer improved permeability, resistance to moisture, and high-temperature durability, addressing the limitations of the current jute/polyester materials.

The hybridization of end-of-life jute/polyester fibre mat with glass fibre and ramie fibre makes it possible to modify the characteristics of composites. The mixing ratio and distribution of the different fibres can be fully adapted to the requirements of the desired application. As a first attempt to reuse hybrid fibre mats, the study evaluated the use of epoxy, a widely used thermosetting matrix. The evaluation aimed to determine the feasibility of reusing hybrid fibre mats and explore the potential for fabricating hybrid composites in response to the growing demand for environmentally sustainable and recyclable products. Subsequent research in this area could evolve towards natural-origin thermoplastics to fabricate hybrid materials, in response to the growing demand for environmentally sustainable and recyclable products.

Finally, it is crucial to emphasize the importance of assessing environmental impacts. Merely substituting synthetic fibres with natural fibres does not guarantee a reduction in environmental impacts. To comprehensively evaluate the environmental impact of a breather throughout its life cycle, including factors such as resource consumption, energy use, greenhouse gas emissions, water pollution, and waste generation, a thorough assessment should be conducted. Life cycle assessment (LCA) is a technique for evaluating the environmental impact of a product or process, from the extraction of raw materials to the end of its life. It provides a comprehensive approach that encompasses production, use, and disposal. Therefore, further LCA assessment can be conducted to identify potential areas for improvement and guide decision-making towards more sustainable practices.

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