



Physical and Mechanical Properties of Bio Based Natural Hybrid Composites

K. Sadashiva*, K.M. Purushothama

Department of Mechanical Engineering, Dr. Ambedkar Institute of Technology, Bengaluru, Karnataka. 560056, India

**Corresponding author, Email address: sadashiva41@gmail.com*

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Abstract: Nowadays, the researcher is more concerned with environmental issues and is focused on modern research using natural resources. Natural resources such as ramie and silk are combined with epoxy resin, and these materials are biodegradable and environmentally friendly, giving them more strength and a higher strength-to-weight ratio when compared to conventional reinforcements. The reinforcement and matrix are fabricated with dimensions of 300x300x3 mm³ by using a manual hand layup technique, which is a less expensive consumption technique. The laminate designations are four layers of silk (SSSS) and four layers of ramie fabric (RRRR) that were used for developing a natural composite, while for making the hybrid composite, two layers of silk and two layers of ramie were used to create the laminate, i.e., RSRS, SRRS, and RSSR. The composites of ramie fibre with epoxy laminates exhibited the highest interlaminar shear, impact strengths, and micro-hardness when compared to the silk fibre composite laminate, and moisture absorption is less in silk composites. The result revealed that the designations of RSSR laminate have enhanced the interlaminar shear, impact, and micro-hardness properties in the hybrid composite laminate group. The interlaminar shear properties of debonding and fracture of surface structure were carried out using scanning electron microscopy.

1. Introduction

The usage of synthetic fibres for strengthening or reinforcing engineering constructions has increased during the last few years (Xian *et al.* 2022; Khalita *et al.* 2018; Ehera *et al.* 2016) [1-3]. Regarding this application, a major difficulty is that synthetic fibres' fire resistance capability degrades abnormally when exposed to high temperatures [4-6]. An approach that gives excellent mechanical enactment is the hybridization of a composite comprised of a polymeric matrix reinforced with synthetic and natural fibres [7-9]. Compared to synthetic fibres, natural fibres have a lower specific gravity and are biodegradable [10]. Nevertheless, natural fibres commonly show compatibility issues with both fibre and polymer structures. It results from the hydrophobicity of the fibre and the incompatibility of the matrix, which can also be addressed by surface modification [11-13]. Natural fibers, mainly bast fibers, are an excellent alternative to synthetic fibres because they are widely available, simple to extract, low density, biodegradable, lightweight, and have a high specific strength [14-16]. The development of natural fiber-based composites in the automobile industry is commonly thought to replace the use of synthetic fibres in a variety of industries, including aerospace. By increasing toughness or impact resistance, hybridization provides a pathway for expanding the use of

composite materials, particularly in sophisticated applications [17]. Naveen et al were discovered that various weight percentages of the Kevlar/Cocos nucifera sheath-reinforced composite had different mechanical properties and that the Cocos nucifera sheath may be able to substitute the Kevlar fibre polymer composite [18]. Giridharan has evaluated the properties of a glass/ramie fiber-reinforced composite at different weight ratios. Ramie's qualities were improved when a trace amount of glass was added to the fibre, making it more affordable and environmentally friendly [19]. Yang et al have centered on the effects of ramie fibre that hasn't been altered Using melting hybrid technology, reinforced polypropylene can achieve good mechanical qualities. Thus, modified fibre has greater fibre reinforcement than ramie fibre that hasn't been treated. The polypropylene/ramie fibre lowers the thermal degradation temperature [20]. Conventional hand layup processes are being replaced by modern methods like resin infusion and vacuum-assisted resin transfer moulding (VARTM), in the manufacture of composite laminates. Because, material, labour and equipment costs are lower, parts are produced more affordably and with higher quality. A natural fiber-reinforced composite laminate with a low processing cost that is manufactured with bio-based epoxy resin has a high-volume percentage of fibre. The created composite is aimed at sectors of the economy that demand light weight, low costs, and low environmental impact [21-24]. Sanjeevi *et al.* investigated the impact of a hybrid natural fibre phenol-formaldehyde composite using the hand layup process. They have been observed at three distinct weight distributions: 25%, 35%, and 45%. The 35% composite material combination outperformed the other two manufactured composites in fiber-matrix bonding [25]. To investigate the load transfer process of composites in nano-hybrid shish-kebab structures, a shear-lag model was created. It exhibits clear impacts on morphological and elastic modulus [26]. Swamy et al. examined how areca fibre affected its strength. Areca-treated phenol-formaldehyde is excellent for structural applications and the packaging industry since it quickly absorbs a substantial amount of moisture and degrades slowly [27]. Joseph et al. reported that banana fibre enhances mechanical properties and interfacial strength when soaking in phenol formaldehyde when contrasted to glass fibres [28]. Sadashiva et al. investigated the natural hybrid composites made by the hand layup process and found that by the incorporation of ramie fibre with silk fiber, the flexural and interlaminar shear strengths have been increased with the addition of filler OMMT [29]. According to the previous research, there hasn't been much research on the mechanical assessment of thermoset-based hybrid composite laminates created with constituent materials including ramie, silk, and epoxy. The hybridization technique was used to produce inexpensive composites. The major goal of this work is to determine how the stacking order of hybrid composites made of ramie and silk fibres affects their mechanical characteristics, including interlaminar shear strength, impact resistance, and hardness. The cracked surface of the composite was examined morphologically using a scanning electron microscope.

2. Materials and Methods

This experiment uses ramie and silk fabric from Vruksha Composite Guntur in Andhra Pradesh, India, as reinforcement materials. The physical characteristics of the fibres and epoxy are shown in Table 1. The epoxy resin with a grade of LY556 and a hardener of grade HY951 were obtained by Ultra nanotech India Pvt. Ltd. To prepare the matrix segment, epoxy and hardener were mixed in a 10:1 ratio. The composites of ramie and silk laminates were made using the manual hand layup process. The five different types of laminates, with dimensions of 300x300x3 mm³, with a total of four layers of fabrics made of ramie and silk, were prepared. The reinforcement layers were constructed using the weight-percent method. A metal mould was used in the fabrication of the laminates. The bottom and top surfaces of the mould were initially treated with a silicon spray. The fibre fabrics were layered one

after the other as per the required laminate designation mentioned in **Table 2**, with R referring to ramie fibre and S referring to silk fibre. The epoxy resin was applied with brushes in-between the fabric layers, while the roller action on the reinforcement allowed for easy escape of bubbles formed during the process. The mould is then dried for about 48 hours, and then the hybrid composite laminates were placed in an electric oven at 1100C for 4 hours to eliminate the moisture content. After removing the laminates from the oven, they were machined using a water jet cutting machine as per the ASTM standards for quasi-static testing. The key resources used in the present work are depicted in **Figure 1**.

Table 1: Physical characteristics of the fibers and epoxy

Description	Ramie fiber	Silk fiber	Epoxy
Density (g/cc)	1.5	1.38	1.1
Tensile strength (MPa)	1000	650	35 - 135
Tensile modulus (GPa)	61.4 - 128	16	3.4
Elongation (%)	2-4	18-20	1-8.5

Table 2: Laminate Designation

Sl no	Laminate Stacking Sequence
1	S+S+S+S
2	R+R+R+R
3	R+S+R+S
4	S+R+R+S
5	R+S+S+R

R-Ramie fabric, S-Silk fabric

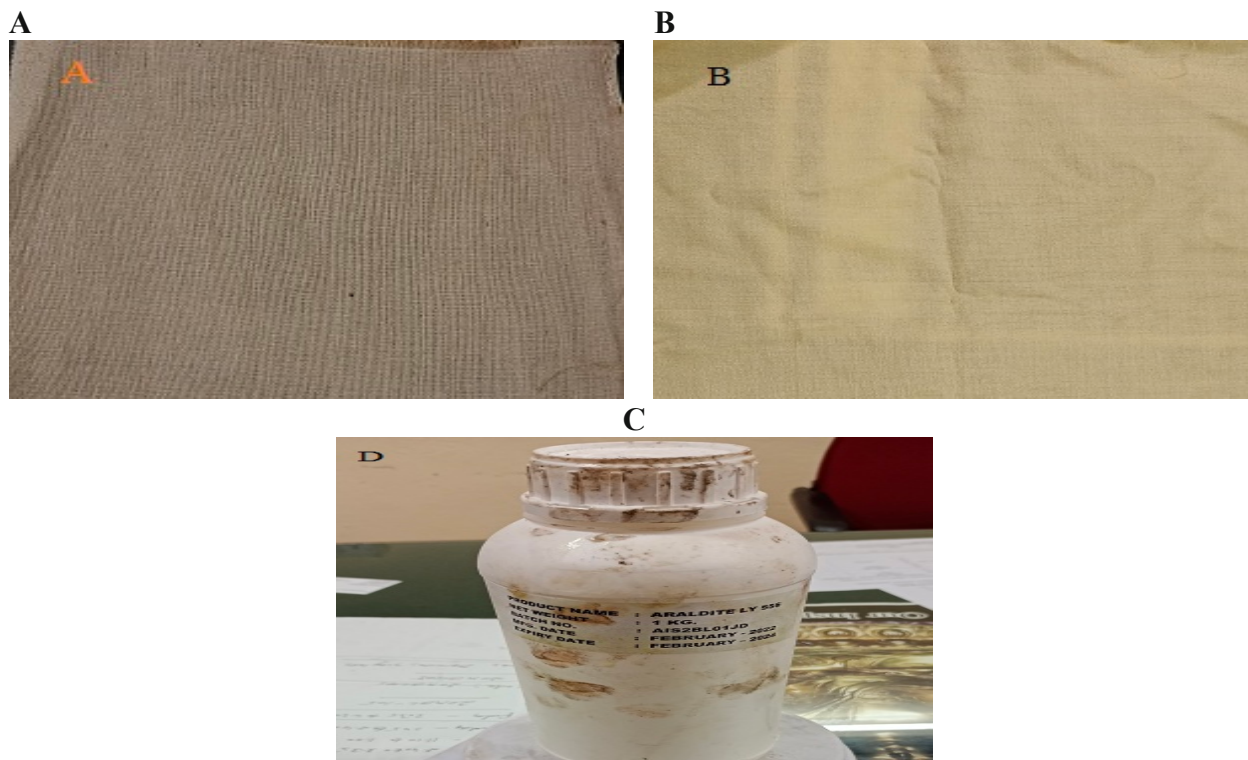


Figure 1 : A. Ramie Fabric B. Silk fabric C. Epoxy matrix

2.1 Interlaminar shear strength (ILSS)

By conducting relatively brief span-length shear tests on composite samples at room temperature, the shear strength value is determined. ASTM standard D2344 specifies how to conduct the test on the universal testing apparatus. This test uses equipment with a cross-head speed of 1.5 mm/min on a small specimen measuring 45x6x4mm³. As the loading cylinder exerts a downward force on the specimen, it is subjected to normal (bending) and transverse shear stress. The use of a straight beam is anticipated to reduce bending forces, which will lead to interlaminar shear failure with breaking in a horizontal direction between the laminates. Each type of composite had three identical specimens examined, and the composite's average value is shown as a property.

2.2 Impact strength

The impact strength of the laminates is determined by subjecting each of the specimen with dimension 63×12.7×3mm³ (ASTM D-256 standard) onto the grippers of the impact testing machine. The quantity of energy absorbed by these specimens (which is the bonding strength of the reinforcement and matrix with filler) at the failure is noted in terms of energy, i.e., Joules.

2.3 Micro-Hardness test

The digital Shore-D hardness durometer is used to measure the specimen's hardness. The durometer with a step of 0.5 HD has a range of 0–100 HD and is hence used to find out the hardness of rubber, polymers, as well as plastics. While testing, the durometer is used to press onto the exterior part, where the indenter pin will get penetrated onto the sample, thus displaying digitally the resistance to indentation. [Cicala et al. \[24\]](#) Suppose the value of HD is more than 60, it is found that it is a good resilience material or else a poor resilience material.

2.4 Moisture Absorption Test (MAT)

The water absorption test will be carried out by immersing the laminate in normal water as well as in distilled water for nearly 30 days. However, the laminates were machined enough as per the ASTM D570 standard, for the dimensions of 30×28×3 mm³. The weights of the laminate were frequently measured at intervals of 10 days. The water molecules should be removed from the surface of the specimen using the cotton cloth and weighed with a digital balance. The percentage of moisture absorption was calculated by using [Equation \(1\) \[25\]](#):

$$\text{Moisture absorption (\%)} = \frac{W_m - W_n}{W_n} * 100 \quad (1)$$

Where W_m is the final weight of the laminate after 10 days of immersion and W_n is the initial weight of the laminate before immersion.

2.5 Scanning Electron Microscope

The morphology of the composite material will be examined utilizing scanning electron microscopy. However, scanning electron microscopy is utilized to analyse the interfacial properties, internal cracks, and inward design of the broke surfaces of the laminate. The conducting material will be coated on the surface of the laminate preceding to the SEM assessment of the surfaces.

3. Results and Discussion

3.1 ILSS

The interlaminar shear strength was investigated in the bending test methods with a shorter beam length. The goal of this experiment was to determine the bonding between the matrix and the

reinforcement as well as the fracture load of the specified specimens. The interlaminar shear strengths for the various laminates were shown in Figure 3. The ILSS epoxy laminates had a shear strength of 26.66 MPa due to the epoxy matrix being purely glassy. The maximum ILSS obtained in laminate RRRR, which is purely a natural ramie with a strength of 98.39 MPa, is due to the fibre stiffness to oppose shearing in the laminates and is thus improved in the laminates. The pure silk SSSS laminates of ILSS have a strength of 43.56 MPa, which is due to the smooth surface of the fabric on which it shears the laminates. The hybridised composite of laminates showed an RSSR that showed an ILSS of 91.21, which was the highest strength in the hybrid composite when compared to the other two laminates. The hybrid laminates RSRS and SRRS show moderate strengths of 76.59 MPa and 69.67 MPa, respectively.

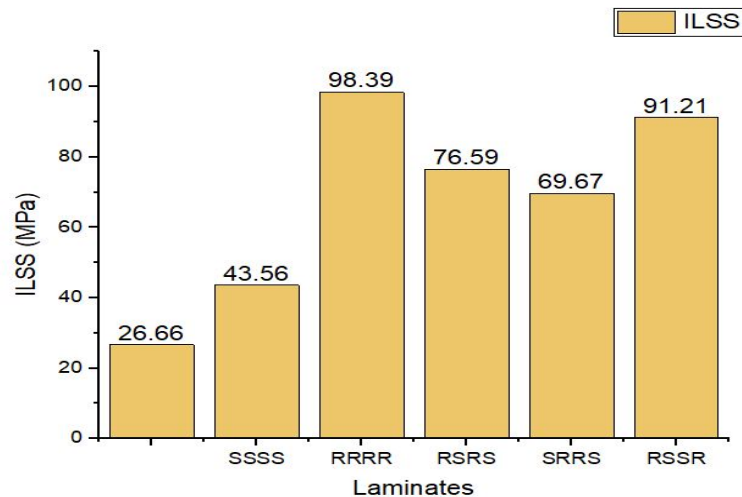


Figure 3: ILSS of different laminates

3.2 Impact strength

The bonding strength of the fibre and the matrix, along with the filler material, will be determined by the impact studies. Figure 4 shows the impact strength (kJ/m²) of the different laminates. The test on the impact relies on many variables, like fibre-matrix bonding and the nature of the fibre material. Equation 2 gives us the calculation of the impact strength. Because the epoxy laminate was purely a glassy state of matrix, it has an impact strength of 13.12 kJ/m². The laminate RRRR filler has a higher impact strength of 51.68 kJ/m² when compared to other laminates. The reason is due to the stiffness of the Ramie fabric. The pure silk fibre reinforcement laminate SSSS has a lower impact strength. This is due to the presence of hemicellulose, which reduced the impact strength. Silk and Ramie reinforced composite laminates (RSSR) had a reasonable impact strength of 33.65 kJ/m². Furthermore, when the number of fabric layers was increased in the hybrid composites, it led to an increase in the impact strength of 23.80 kJ/m² in RSRS and 28.50 kJ/m² in SRRS. The gain in strength was attributed to the fact that the reinforcement material increased the compatibility and adhesive nature between the matrix phases.

$$\text{Impact strength (I.S)} = \frac{\text{Impact Energy in joules}}{\text{Area of cross section in m}^2} \quad (2)$$

3.3 Micro-Hardness test

Figure 5 illustrates the micro-hardness of different laminates. The laminate RRRR, which consists of only ramie fibre layers, exhibits 82 as the highest hardness number. The SSSS laminate, which was used to make pure silk yarns, had a hardness value of 66, which was lower than the ramie laminate.

The natural silk fibres incorporated into the composites showed a sleek hardness, which is due to the softness of the material. When considering the hybrid laminates, which have a slightly increased hardness value, this shows resistance to the deformation in the material as well as the indentation on the laminate, resulting in effortless material deformation. The hybrid composite laminates show a hardness value of 72, 68, and 71, respectively.

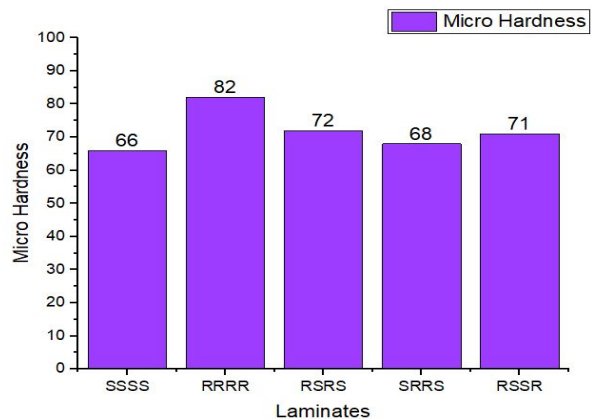
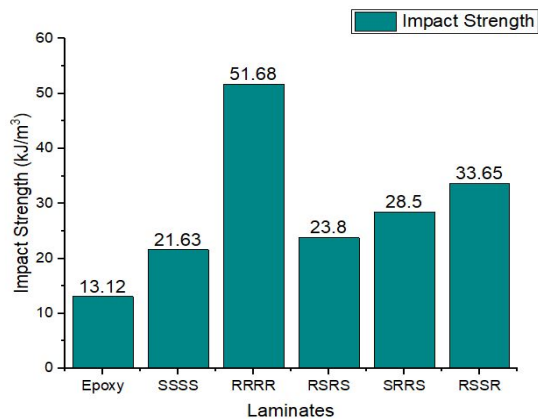


Figure 4: Impact strength of different laminates

Figure 5: Micro-Hardness values of different laminates

3.4 Moisture Absorption Test (MAT)

The specimens gain weight as a result of the water molecules absorbing them after immersion. The experiment continued for 30 days and was repeated every 10 days in both normal and distilled water. Tables 5 and 6 show the weights before and after the duration in both conditions. The results of the tests show that the water absorption percentage is higher in the RRRR laminate, which is made of pure natural ramie fabric. Water absorption was lowest in laminate SSSS, which contains four layers of natural silk fabrics. Because natural fibres are composed of cellulose and lignin, hybridised composites have a lower absorption capacity. In normal water, the specimen absorbs more than in distilled water.

Table 5: Moisture absorption Percentage in distilled water.

Laminates	Weight of the specimen before immersion (g)	Percentage increase in weights		
		Day 10	Day 20	Day 30
SSSS	3.766	5.72	9.46	13.35
RRRR	4.561	8.73	15.42	19.46
RSRS	4.235	7.38	13.18	18.26
SRRS	4.171	6.89	12.42	15.75
RSSR	4.323	8.97	13.75	16.33

Table 6: Moisture absorption Percentage in normal water.

Laminates	Weight of the specimen before immersion (g)	Percentage increase in weights		
		Day 10	Day 20	Day 30
SSSS	3.667	5.32	9.66	14.36
RRRR	4.577	8.93	15.73	19.98
RSRS	4.226	7.17	14.29	18.98
SRRS	3.972	7.28	12.73	15.80
RSSR	4.331	9.13	13.85	16.86

3.5 Scanning Electron microscope Analysis

A solid interface allows more load or stress transfer through the composite filaments, though a weak interface produces a helpless bond between the fibres and the matrix [17]. At the point when the power applied to the composite exceeded the interface bonding, weak bonding basically empowered the filaments to pull out and break, making the matrix split [18]. Case matrix break is presently the most predominant failure instrument, and it is in all likelihood associated with the brittle failure of SSSS composites delivered by huge fiber-to-fiber contact, as shown in **Figure. 6A** [19]. Another issue is inadequate fibre scattering, which causes unequal fibre breaking when the stacked fibres are broken since they are not symmetric and equal. The tensile properties of plain ramie composites are the most reduced (RRRR). However, due to its single fibre properties, the morphological construction of RRRR (**Figure. 6B**) shows a greater amount of fibre pull-outs, proposing a weak interfacial association among fibres and matrix, bringing about diminished tensile strength. The fibre breakage on the fractured surface shows the lower fibre pullout in the hybrid composite RSRS, as found in **Figure. 6C**. the tensile strength and modulus of hybrid composites have increased as the extent of ramie fibre inclusion has increased. Individual reinforced fibre characteristics have the biggest effect on hybrid composite properties [20]. Besides, various investigations have discovered that the mechanical properties of hybrid laminated composites might be fundamentally influenced by various stacking arrangements. This review supports previous findings that substituting stacked fibres in composite materials increases tensile strength [21]. This is because of the way that the top and base layers of a composite are skin-covered. Since the skin factor determines the rigidity of the essential loadbearing part [22], picking a high-strength material for the skin layer can bring about higher tensile strength. As an outcome, axial stress might be productively conveyed by the ramie fibre's external layers prior to being passed to the core layer.

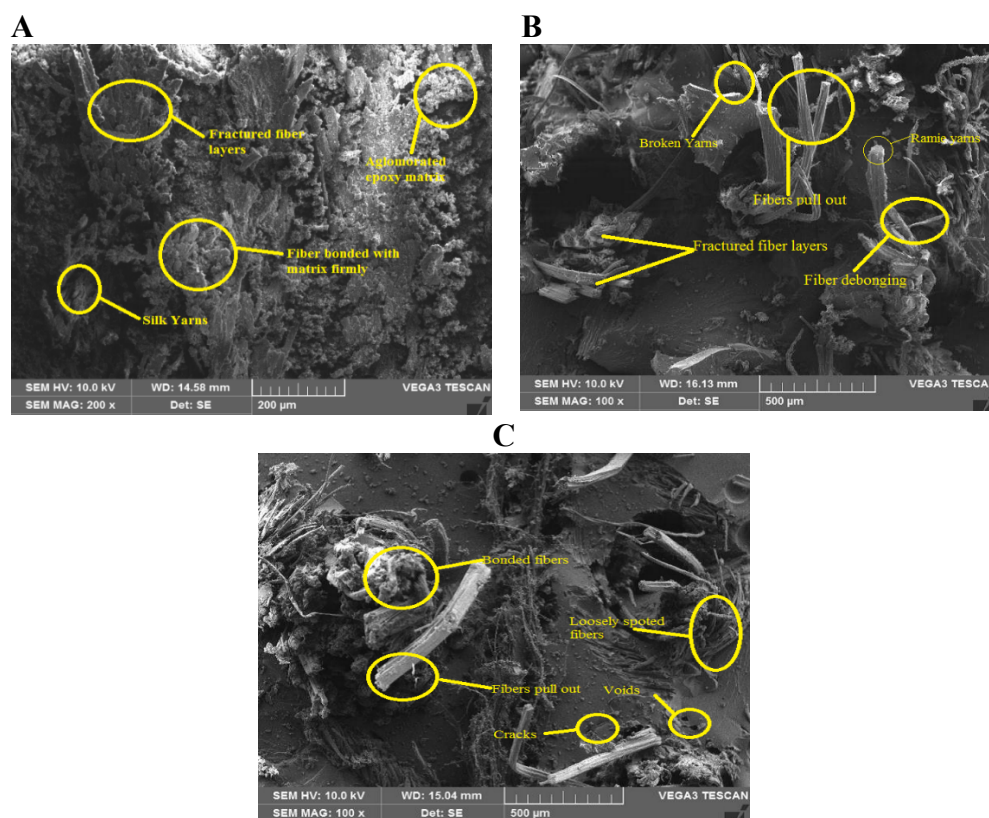


Figure 5: Tensile fractured SEM Images., **A.** Silk laminate **B.** Ramie laminate **C.** Hybrid laminates.

Conclusion

In this study, experimental work has been done in accordance with ASTM standards to analyse the hybridization effect on ramie and silk fiber-reinforced epoxy resin polymer composites made by a manual hand layup procedure. RSSR laminate has the highest shear strength when compared to other laminates composed of two layers of silk and two layers of ramie, and RSSR impact strength has interpreted the highest value of hybrid composite. The moisture absorption content is higher in normal water than in distilled water, and laminate SSSS has a lower moisture absorption compared to other composites. The fracture specimen of ILSS was analysed by SEM. As a result, this hybridization of materials has to be used in medium structural applications, like car door panels, computer parts, and railway interior design materials. The current study can be improved by including filler materials made of natural fibres to increase the scope of applications for polymer composites.

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