Photocured unsaturated polyester composites reinforced with glass and natural fiber used in the pipeline renovation

Małgorzata Krasowska[1), 2), *)] (ORCID ID: 0009-0008-3861-8099), **Rafał Oliwa**[3)] (0000-0003-1319-6199), **Mariusz Oleksy**[3)] (0000-0001-5515-8575), **Tomasz Galek**[4)] (0000-0001-7775-0908)

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Abstract: The vacuum bag method was used to obtain photocurable polyester resin composites, in which part of the glass fiber (3 wt%) in the glass mat was replaced with natural fiber (sisal, hemp and kenaf). The physicochemical properties of natural fibers (wetting angle, UV/VIS radiation transmittance) were determined. The influence of the hybrid filler on the structure (SEM) and mechanical properties of the composites was investigated in terms of their application in pipeline systems. The introduction of natural fiber reduced the stiffness and tensile strength of the composites. Due to the smallest wetting angle for polar and nonpolar liquids and the total UV/VIS radiation transmittance (425 nm) of sisal fiber, the best processing and mechanical properties were obtained for laminates reinforced with this fiber (tensile strength 257 MPa, flexural modulus 20.5 GPa).

Keywords: hybrid filler, photocured unsaturated polyester resin, pipeline renovation.

Fotoutwardzalne kompozyty poliestrowe wzmocnione włóknem szklanym i naturalnym stosowane w renowacji systemów rurowych

Streszczenie: Metodą worka próżniowego otrzymano kompozyty na bazie fotoutwardzalnej żywicy poliestrowej, w których część włókna szklanego (3% mas.) w macie szklanej zastąpiono włóknem naturalnym (sizal, konopie i kenaf). Oznaczono właściwości fizyko-chemiczne włókien naturalnych (kąta zwilżania, transmitancja promieniowania UV/VIS). Zbadano wpływ napełniacza hybrydowego na strukturę (SEM) i właściwości mechaniczne kompozytów pod kątem ich zastosowania w systemach rurociągowych. Wprowadzenie włókna naturalnego zmniejszyło sztywność i wytrzymałość na rozciąganie kompozytów. Ze względu na najmniejszy kąt zwilżania dla cieczy polarnych i niepolarnych oraz całkowitą transmitancję promieniowania UV/VIS (425 nm) włókna sizalowego najlepsze właściwości przetwórcze i mechaniczne uzyskano dla laminatów z jego udziałem (wytrzymałość na rozciąganie 257 MPa, moduł sprężystości przy zginaniu: 20.5 GPa).

Słowa kluczowe: napełniacz hybrydowy, fotoutwardzalne nienasycone żywice poliestrowe, renowacja systemów rurowych.

With concern for the environment and the reduction of a product's life-cycle carbon footprint (LCA) in mind, efforts to optimize the use of raw materials, including increasing the proportion of recycled materials and natural raw materials, are noted worldwide. Therefore, the use of natural fibers in composite materials is considered promising due to a number of advantages, such as availability, low cost, reusability, low density, biodegradability, and lack of toxicity [1, 2]. As a result, they are becoming increasingly important in the automotive, aerospace, and buildings [3, 4]. Despite this, polymer composites reinforced with natural fibers have some limitations in terms of their ability to carry heavy loads and achieve consistent properties [5, 6]. A promising research direction for the search for new composite materials, also in their application in pipeline construction and renovation, is the hybridization of reinforcement [7, 8]. The use of two types of fiber reinforcement enables obtaining the functional properties of the composites, also reducing the cost of the composite materials, which is difficult with a single type of reinforcement [9–13]. Hence, the hybridization of natural fibers with synthetic fibers is particularly useful due to the properties

1) Doctoral School of Rzeszów University of Technology, Al. Powstańców Warszawy 12, 35-959 Rzeszów, Poland.

2) Firma Handlowo-Usługowa INSTBUD Stanisław Boguta Spółka Jawna, Nieznanowice 50, 32-420 Gdów, Poland.

3) Department of Polymer Composites, Faculty of Chemistry, Rzeszow University of Technology, Al. Powstańców Warszawy 12, 35-959 Rzeszów, Poland.

4) Faculty of Mechanics and Technology, Rzeszów University of Technology, ul. Kwiatkowskiego 4, 37-450 Stalowa Wola, Poland.

*) Author for correspondence: m.krasowska@in-liner.pl

of both materials. Hybrid polymer composites reinforced with natural and synthetic fibers are increasingly being used in industry because of their environmental impact and the promotion of sustainable development [14–16]. In particular, this applies to polyester-glass composites, which have been successfully modified with natural fibers such as hemp [17, 18], kenaf [19, 20], sisal [21], jute [22], palm [23–25], coconut [26, 27], basalt [28], bamboo [29], banana [30] and flax [31]. It is worth emphasizing that material combinations, layer sequences, weight percentages, and fiber orientation are key factors determining the properties of hybrid composites. Therefore, the hybridization of natural fibers allows improvement of the functional properties of composites [32–34] but often their mechanical properties are often lower compared to composites reinforced only with glass fibers [35, 36]. For these reasons, it is important to appropriately select fibers in hybrid composites to obtain suitable functional properties, considering their target application. For fiberglass sleeves with a photo-cross-linked polyester resin matrix used for the construction and renovation of sewer networks, transparency is also important to ensure a uniform degree of cross-linking throughout the composite. However, there is no information in the literature on the effect of hybrid reinforcement of glass fiber and natural fiber on the mechanical properties of composites with a matrix of photocured polyester resins. Thus, one of the goals of this study was to determine the effect of hybrid reinforcement of glass fiber with different natural fiber on the mechanical properties of photocured unsaturated polyester resin.

EXPERIMENTAL PART

Materials

The polymer matrix used was a commercial, photocurable unsaturated polyester-styrene resin (UP) Polimal 129-1 (IZO/NPG, Sarzyna Chemicals, Nowa Sarzyna, Poland). Commercial grade Tex 2400 glass mats (ECR, Jushi Co. Ltd., Tongxiang, China) were used. The glass mat consisted of a mesh made of glass fiber on which chopped fiber was applied, and then everything was sewn with polyester threads. The chopped fiber constitutes about 33 wt% of the glass mat. Sisal fiber (SF) was supplied by Fibresco Ltd. (Lodz, Poland) and Wimar WM Ltd. (Krasnik, Poland), kenaf fiber (KF) by Wilhelm G Clasen Ltd. (Dhaka, Bangladesh) and hemp fiber (HF) by Ekotex Ltd. (Namyslow, Poland). The photoinitiator system Omnirad 184 (1-hydroxycyclohexylphenyl ketone) and Omnirad 819 [bis(2,4,6-trimethylbenzoyl)-phenyl phosphine oxide] was provided by IGMResins (Waalwijk, The Netherlands).

Composites preparation

The polyester composites were prepared using the vacuum bagging method. Five layers of 300 × 300 mm ECR glass mat were successively impregnated with an unsaturated polyester resin with the addition of a photoinitiator system using a grooved roller. The impregnated laminate was then successively covered with perforated release film and a breather layer cloth, respectively, before being placed in a vacuum bag to remove excess resin and remove air bubbles. Hybrid glass/natural fiber composites were prepared in the same way using a glass mat, in which part of the glass fiber (3 wt%) was replaced with chopped natural fiber. Natural fiber (3 wt%) was used to ensure transparency and a uniform structure of obtained laminates – higher natural fiber content resulted in delamination of the laminates.

The laminates obtained were cured in a UV chamber using the following conditions: UV lamp (Prokasro – UV, Strahler, Germany) with a power of 600 W, dosage of 11 mW/cm^2, distance of the radiation source from the sample of 38 cm and curing time of 20 min. After hardening, the composites were cured at room temperature for 24 hours.

Methods

Contact angle of natural fibres

The contact angle and surface tension of the natural fibres were determined using an optical goniometer, DSA308 (Krüss GmbH, Hamburg, Germany) at 23°C and 45% humidity. The test consisted of depositing drops of a polar liquid (water) and a nonpolar liquid (diiodomethane) on the fiber surface. Based on the obtained contact angles, the surface tension at the solid-liquid interface was calculated.

UV/VIS transmittance of natural fibres

Due to the UV curing of the composites, the transmittance of the natural fibres at this wavelength (410–430 nm) was used as a critical parameter. The analysis was conducted using a Shimadzu UV2600 spectrophotometer (Shimadzu, Kyoto, Japan). Barium sulphate was used as a reference sample. Tests were carried out for samples containing 2, 3 and 5 wt% fiber.

Mechanical properties of natural fibres

The mechanical properties of the fibres were determined in a static tensile test according to PN-EN ISO 9163 using samples in the form of a bundle of fibres with a linear mass of 1000 tex and a gauge length of 10 mm. Tensile properties were determined.

Morphology of polyester composites

Microscopic analysis of the cross section of the composites was conducted using a Tescan SEM-EDS MIRA3 (Tescan, Brno, Czech Republic) scanning electron microscope. The samples were subjected to vacuum copper spraying by Quorum Q150T ES spray device (Quorum, Laughton, UK) using a spray/deposition current of 60 mA, a spray time of 30 s, and a sprayed layer thickness of approximately 12.5 nm.

Mechanical properties of polyester composites

The tensile and flexural properties of the laminates were evaluated using a Shimadzu AG-X/MST-X/X-TYPE 50 kN (Shimadzu, Kyoto, Japan) (class 0.5) testing machine in accordance with EN ISO 527-2 and EN ISO 178, respectively. The crosshead speed for tensile and flexural tests were 5 mm/min and 10 mm/min, respectively. The samples were conditioned for 24 h at 23±2°C. The dimensions of the tensile and flexural specimens were 250 × 25 × 4 mm and 60 × 15 × 4 mm, respectively.

RESULTS AND DISCUSSION

Natural fibers characterization

UV/VIS spectra of the fibers are presented in Figs. 1-4. The spectra were analyzed in the range 200–800 nm. The transmittance at 425 nm wavelength showed that the glass fiber concentration had no effect on absorption capacity (99.99%). In the case of natural fibers, near-full radiation transmittance was also observed. As the fiber content increased, the transmittance decreased from 99.99% to 99.30%. The exception is hemp fiber, the transmittance of which at 5 wt% content is 95.42%. Also, considering the polar liquid contact angle, sisal fiber had the highest processing capacity for photocured laminates among natural fibers (Table 1). The contact angle for this fiber was below 90°, whereas for the hemp and kenaf fibers it was above 98° and 102°, respectively. For a nonpolar liquid, the lowest contact angle was also obtained for the sisal fiber. This indicates good wettability of the material, which in turn has an impact on the permeability of the sisal fiber-modified glass mat. This is because unsaturated polyester resins have polar and nonpolar centers, depending on the degree of condensation and the content of hydroxyl groups. Furthermore, the presence of the sisal fiber should not hinder the photopolymerization process of the hybrid composite, as the optical properties of the materials are similar and allow the assumption that the resulting laminate will be homogeneous.

Table 2 presents tensile properties of natural fibers. Sisal fiber shows the highest tensile strength and stiffness. However, it also exhibits a significant deformation capacity.

Composites characterization

SEM microphotographs of the cross section of the composites show the glass and natural fibers stacking

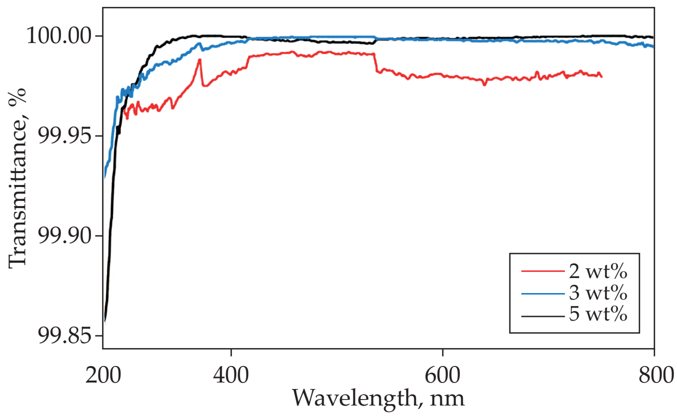

Fig. 1. UV/VIS spectra of glass fiber

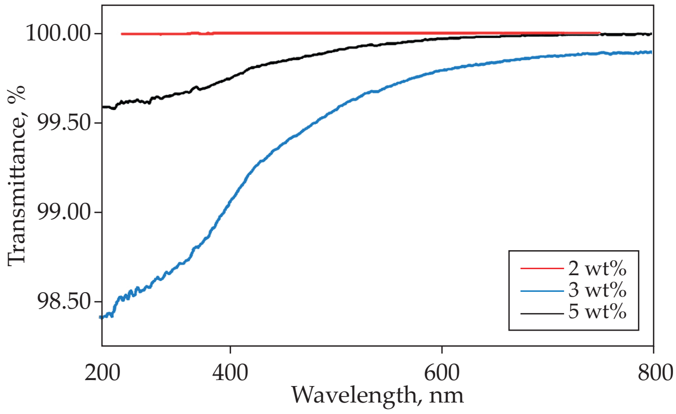

Fig. 2. UV/VIS spectra of sisal fiber

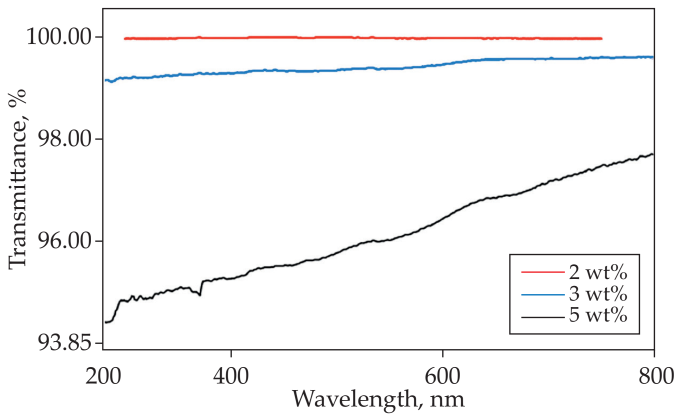

Fig. 3. UV/VIS spectra of hemp fiber

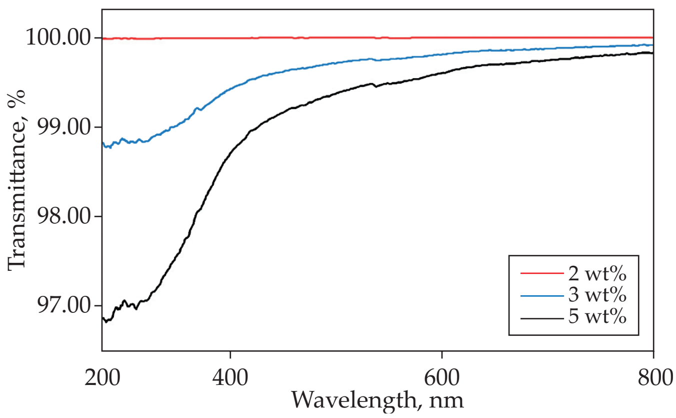

Fig. 4. UV/VIS spectra of kenaf fiber

T a b l e 1. Contact angle of natural fibers

Fiber	Contact angle, °		Surface tension mN/m
	water	diiodomethane	
Sisal	86.4±6.7	64.6±6.9	30.3±6.7
Hemp	98.5±9.3	65.5±7.5	26.5±6.1
Kenaf	102.8±3.9	67.7±5.5	24.7±3.7

T a b l e 2. Tensile properties of natural fibers

Fiber	Maximum load cN	Specific strength cN/tex	Young modulus GPa	Elongation at break %
Sisal	41307±1773	41.3±1.8	26.6±4.7	5.4±1.0
Hemp	23742±5180	23.7±5.2	21.0±1.8	5.1±1.8
Kenaf	33794±3188	33.7±3.2	21.0±1.9	3.9±1.3

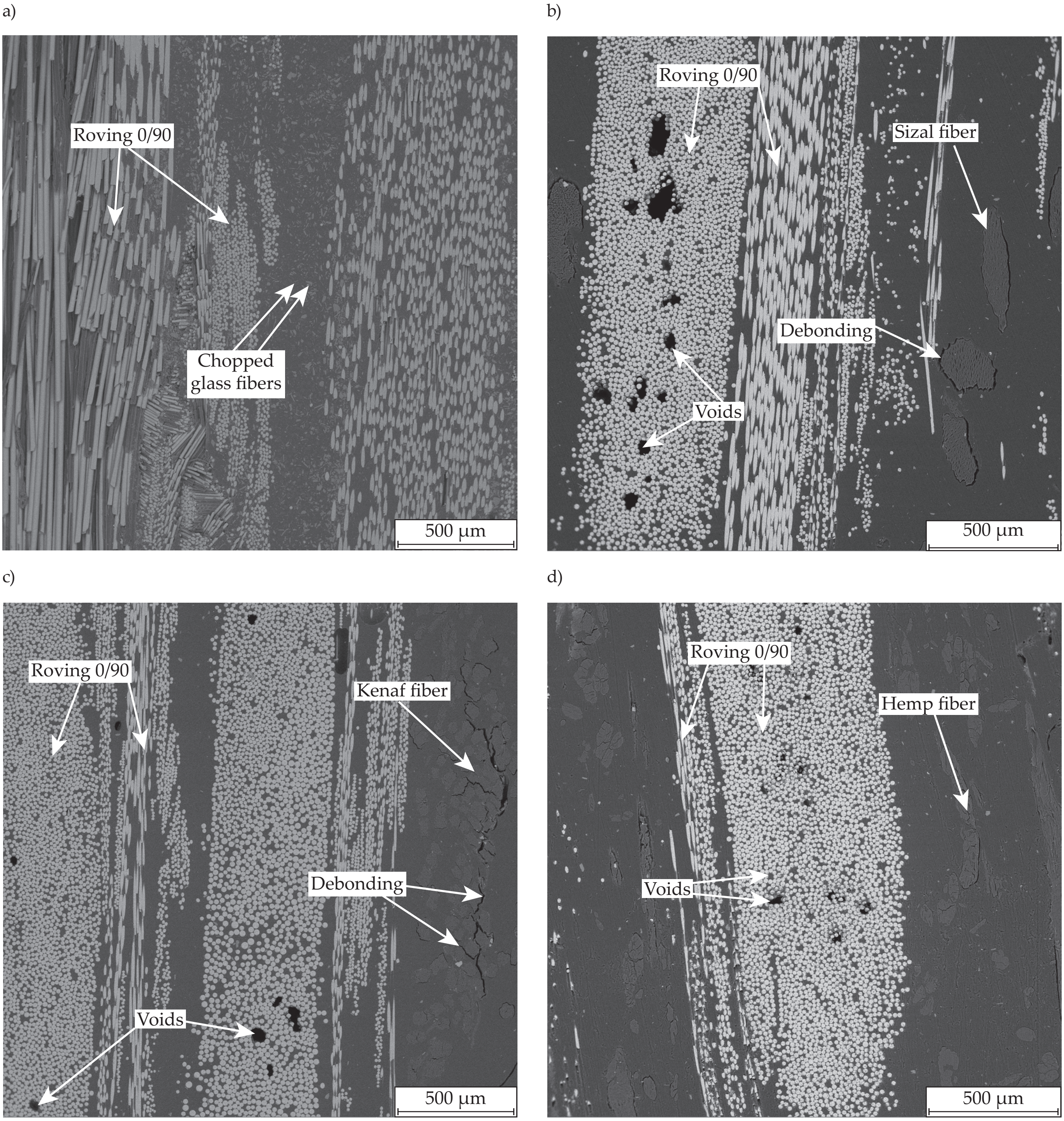

Fig. 5. SEM microphotographs of the composites: a) GF, b) GF/SF, c) GF/KF, d) GF/HF

in the respective layers (Fig. 5). In all composites, there are layers of glass roving stacked in the 0- and 90-degree directions, and a layer of loose chopped fibers consisting of glass and natural fibers (Fig. 5). With the roving layer [0], voids are visible, which are caused by poor percolation of the resin between tightly packed fibers in the composites – there is a fiber-to-fiber contact. In the case of a layer consisting of chopped fibers, regions of higher resin content and clusters of natural fibers with a clearly irregular shape typical of these fibers can be observed. Debonding between the natural fiber and the resin was also observed, indicating their weaker interaction with the matrix compared to the glass fiber. This is due to the hygroscopic properties of natural fibers [21, 37].

Table 3 shows tensile and flexural properties of the composites. Hybrid filler decreased flexural properties. The lowest flexural modulus and flexural strength were observed for the composite with kenaf fiber (GF/KF). The highest flexural modulus showed the composite with sisal fiber (GF/SF). This is consistent with the highest Young's modulus of the sisal fiber. Furthermore, a significant increase in tensile strength (≥ 200%) was recorded for the GF/SF and GF/KF composites compared to the GF composite. The GF/SF hybrid filler can be used in linings for the renovation of UV/VIS-cured pipelines.

T a b l e 3. Mechanical properties of the composites

Property	Type of filler			
	GF	GF/SF	GF/HF	GF/KF
Flexural modulus, GPa	21.15±1.51	20.57±1.06	16.67±0.74	12.32±1.40
Flexural strength, MPa	559±42	434±76	483±25	203±37
Elongation at max load, %	2.8±0.3	2.6±0.5	3.1±0.2	2.0±0.0
Tensile strength, MPa	101±3	247±58	72±4	220±18
Elongation at break, %	3.6±0.5	6.3±0.9	3.0±0.1	5.3±0.8

CONCLUSIONS

In this study, the mechanical and physical properties of natural fibers (kenaf, sial, hemp) were analyzed and then used to modify glass fiber reinforced polyester composites. The effect of hybrid reinforcement (GF/natural fiber) on the performance properties of composites was investigated for their use in UV/VIS cured pipeline renovation linings. Among the natural fibers tested, sisal fiber showed the best photocuring properties, due to the lowest wetting angle between polar and nonpolar liquids. Furthermore, no significant effect of natural fiber content on absorption capacity - total transmission of radiation at 425 nm was found. The mechanical and physical properties of natural fibres were reflected in the results obtained for the hybrid polyester composites, as the composite with the addition of sisal fiber was characterized by the highest flexural modulus (a decrease of approx. 3% compared to the composite reinforced only with glass fiber). Furthermore, a significant increase (≥200%) in tensile strength was observed for the GF/SF composite compared to the GF composite. For better mechanical properties of hybrid composites, the interaction between natural fibres and the matrix would need to be improved, which is confirmed by SEM.

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Authors contribution

M.K. – writing-original draft, results, methodology, investigation, validation; R.O. – conceptualization, writing-original draft, references, validation; M.O. – conceptualization, supervision; T.G. – SEM results, visualization.

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Conflict of interest

The authors declare no conflict of interest.

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