

Non-Conventional Recycling Process of Hemp/Carbon Hybrid Laminates via Thermo-Mechanical Disassembly

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Abstract. The increasing use of fibre-reinforced composites raises critical issues related to sustainability and end-of-life management, particularly for thermoset-based systems. In this work, a non-conventional thermo-mechanical recycling strategy is proposed for hemp/carbon hybrid laminates, aiming at the recovery and reuse of intact reinforcement plies without destructive fibre-matrix separation. Full carbon, full hemp, and two hybrid laminate configurations with different stacking sequences were manufactured, recycled through controlled thermo-mechanical disassembly, and reprocessed into new laminates. The flexural and interlaminar shear behaviour of virgin and recycled materials was investigated to assess the influence of the recycling process on mechanical performance.

Introduction

Over the last decades, fibre-reinforced composite materials have experienced a steadily increasing adoption across several industrial sectors due to their high strength-to-weight ratio, outstanding design flexibility, and excellent structural performance. In particular, polymer matrix composites reinforced with carbon fibres have progressively replaced metallic materials in high-value applications such as aerospace, automotive, and sports equipment, where weight reduction represents a primary design requirement. However, the growing awareness of environmental issues, together with increasingly stringent regulations on sustainability and end-of-life management, has highlighted the limitations of these solutions, fostering strong interest in the development of more sustainable composite materials capable of reducing environmental impact without significantly compromising mechanical performance [1].

In this context, the use of natural fibres as reinforcement in composite materials represents a particularly promising strategy, owing to their renewability, low energy demand during production, and potential contribution to the reduction of CO₂ emissions. Nevertheless, fully “green” composites reinforced exclusively with vegetal fibres still exhibit significant application limitations when high structural performance is required. The intrinsic variability of natural fibre properties, their sensitivity to moisture, and their inferior mechanical characteristics compared to synthetic fibres confine their use mainly to non-structural or semi-structural applications [2–4].

To overcome these limitations, a solution widely explored in the literature is the adoption of hybrid composites, in which natural fibres and high-performance fibres are combined within the same polymer matrix [5, 6]. Hybridisation enables the exploitation of the advantages of different reinforcement types while mitigating their respective drawbacks. In particular, carbon/natural fibre hybrid composites allow the high stiffness and strength of carbon fibres to be combined with the enhanced ductility, energy dissipation capability, and potential damping behaviour of vegetal fibres, resulting in an effective compromise between mechanical performance, cost, and sustainability.

Previous studies [7, 8] have demonstrated that, through careful design of the stacking sequence, it is possible to tailor the flexural and shear behaviour as well as the damage tolerance of hemp/carbon hybrid laminates, in some cases approaching the performance of full carbon laminates while benefiting from a partially bio-based content.

Beyond the sustainability of raw materials, another critical issue affecting composite materials, especially those based on thermosetting matrices, concerns end-of-life management and the limited recyclability of such systems, which remains a significant barrier to circularity due to the irreversible cross-linked structure of thermosets that prevents remelting or conventional reprocessing (e.g., CFRP and GFRP) [9]. Historically, the end-of-life problem of composites was considered marginal owing to their relatively limited early diffusion; however, the exponential growth in the use of carbon and glass fibre-reinforced polymers is leading to a significant increase in composite waste volumes, while traditional disposal routes such as landfilling and incineration are increasingly unsustainable and restricted by regulation, prompting the need for cost-effective recycling strategies [10]. Current recycling technologies for thermoset composites can be broadly classified into three main categories: mechanical, thermal, and chemical recycling [11, 12]. Mechanical recycling, based on cutting and grinding operations, produces fibre-rich and resin-rich fractions that are mainly reused as fillers in new materials, but it results in a severe reduction of fibre length and integrity. Thermal processes, such as pyrolysis, allow the recovery of relatively clean fibres but require high temperatures, entail high energy consumption, and can cause significant degradation of fibre mechanical properties. Similarly, chemical processes, such as solvolysis, enable the recovery of fibres with properties closer to virgin ones, but they present critical issues related to solvent use, processing time, and by-product management. Overall, these strategies are generally characterised by a “down-cycling” approach, in which the value of the recovered material is lower than that of the original composite. Moreover, in previous studies, the authors introduced and validated a novel, non-conventional thermo-mechanical recycling strategy specifically designed for full carbon fibre-reinforced thermoset composites, demonstrating the feasibility of separating and recovering intact reinforcement plies without resorting to destructive fibre-matrix separation techniques [13].

Consequently, while the effectiveness of non-conventional recycling approaches has been mainly demonstrated for full carbon laminates, their application to hybrid composites containing natural fibres remains a largely unexplored scenario. The presence of vegetal fibres, characterised by a different thermal and mechanical response compared to synthetic fibres, introduces additional challenges related both to the disassembly process and to the preservation of mechanical performance after recycling. At the same time, the possibility of recovering and reusing plies containing natural fibres opens new perspectives for extending the service life of hybrid composites with reduced environmental impact.

Considering these factors, the present study aims to extend the current knowledge on hemp/carbon hybrid composites by introducing a non-conventional recycling methodology based on a thermo-mechanical disassembly approach as a tool for the recovery and reuse of reinforcements [14]. To this purpose, full hemp, full carbon, and different hybrid laminate configurations were manufactured, and their recyclability and the reuse of the recovered individual plies were investigated. The mechanical properties of recycled laminates were then compared with those of the corresponding virgin materials, with particular focus on flexural and shear behaviour. The objective of this work is to assess the extent to which the thermo-mechanical recycling process affects the key mechanical properties of these systems and to evaluate the potential of recycled hybrid laminates as sustainable structural solutions capable of combining adequate mechanical performance with reduced environmental impact.

Materials and Methods

Materials and composite manufacturing.

The composite laminates investigated in this study were manufactured using carbon fibre and hemp fibre fabrics as reinforcements combined with a SX10 epoxy matrix (supplied by Mates Srl). The carbon fibre reinforcement consisted of a woven fabric with an areal density of 200 g/m² (supplied

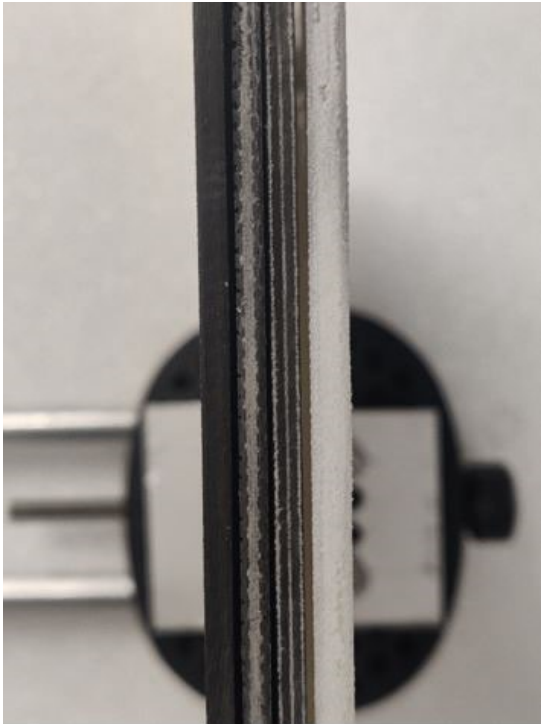
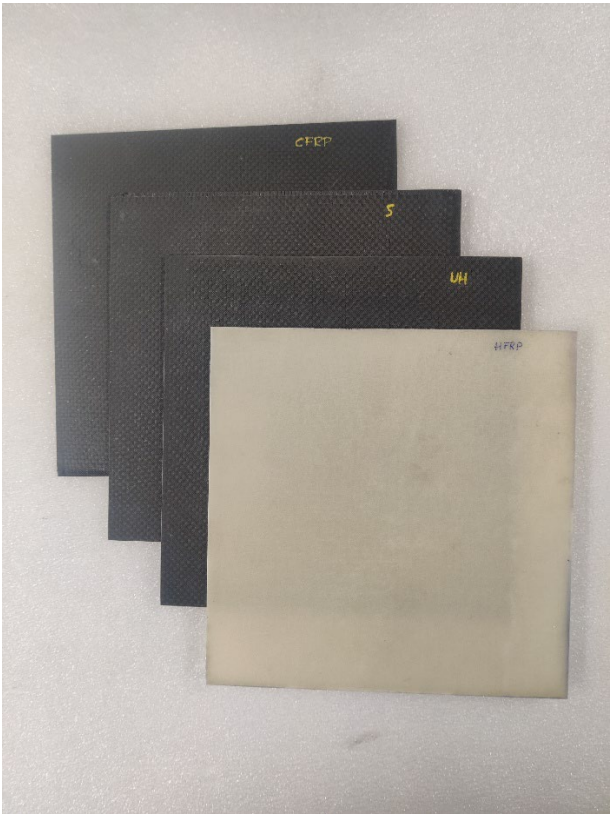
by Toray international Srl), while the woven hemp fabric with an areal density of 160 g/m² (supplied by Maeko Srl). Before the impregnation, the hemp fabrics were dried in a climatic chamber for 12 h at 60 °C and a relative humidity of 20% to reduce moisture content and improve impregnation quality.

The laminates (250 mm x 250 mm) were manufactured by combining hand lay-up and vacuum-assisted press forming, following a consolidated procedure already adopted in previous studies [6]. In detail, the impregnated fabrics were manually stacked inside the mould according to the desired stacking sequence and subsequently cured under vacuum and pressure.

Four types of virgin laminates were produced and used both as reference materials and as starting structures for the recycling process: full hemp laminates (H), full carbon laminates (C), and two hemp/carbon hybrid configurations, referred to as S and U, characterized by different ply distributions through the laminate thickness. All laminates, shown in Fig.1, were manufactured with the same total number of plies (i.e., 15) in order to allow a meaningful comparison of mechanical properties. All stacking sequences are labelled in Table 1.

Table 1. Main laminate characteristics.

Label	Reinforcement	Stacking sequence	Thickness [mm]	Fibre volume fraction [%]
C	Carbon	[C ₁₅] 	2.87	58.1
S	Carbon/Hemp	[C ₆ H ₃ C ₆] 	3.00	55.1
U	Carbon/Hemp	[CHC ₅ HC ₅ HC] 	3.00	55.1
H	Hemp	[H ₁₅] 	3.70	43.2



C S U H

a)

b)

Fig. 1. Virgin laminates (a) and their cross section (b).

Recycling process and Manufacturing of recycled laminates.

The recycling of the virgin laminates was performed using a thermo-mechanical disassembly approach aimed at recovering individual plies while preserving fibre integrity and maintaining the matrix bonded to the fibres. Unlike conventional recycling technologies, this method does not rely on matrix degradation or destructive fibre separation but exploits the increase in ductility of the thermosetting matrix when heated above its glass transition temperature.

Specifically, the virgin laminates were heated in a muffle furnace to 200 °C for 5 min. The selected temperature was high enough to promote epoxy matrix softening and facilitate thermo-mechanical disassembly, while the limited exposure time prevented the onset of significant thermal degradation. Subsequently, the laminates were subjected to controlled mechanical deformation using a manual roll-bending machine with a roll diameter of 38.5 mm, as shown in Fig.2. The cyclic application of bending deformation promoted progressive interlaminar delamination, enabling the separation of individual plies or small ply stacks without inducing macroscopic fibre damage. When necessary, the heating and bending steps were repeated until complete laminate disassembly was achieved. Particular attention was paid to the adjustment of the roll gap and the number of bending passes in order to maximize delamination while preserving the structural integrity of the recovered plies. At the end of the process, the individual plies appeared mechanically intact, flexible, and suitable for reuse in subsequent restacking and consolidation steps.

To indirectly assess the residual resin content and to evaluate possible matrix redistribution or loss induced by the recycling process, each recovered ply was individually weighed. The mass of recycled plies was then compared with that of the corresponding virgin plies, allowing an estimation of resin retention after thermo-mechanical disassembly.

The recovered plies were reused to manufacture recycled laminates, preserving the same reinforcement configurations as the original virgin laminates (H, C, S, and U) without introducing additional reinforcing or matrix materials.

The production of recycled laminates was performed by following the same procedure adopted for the manufacturing of virgin laminates.

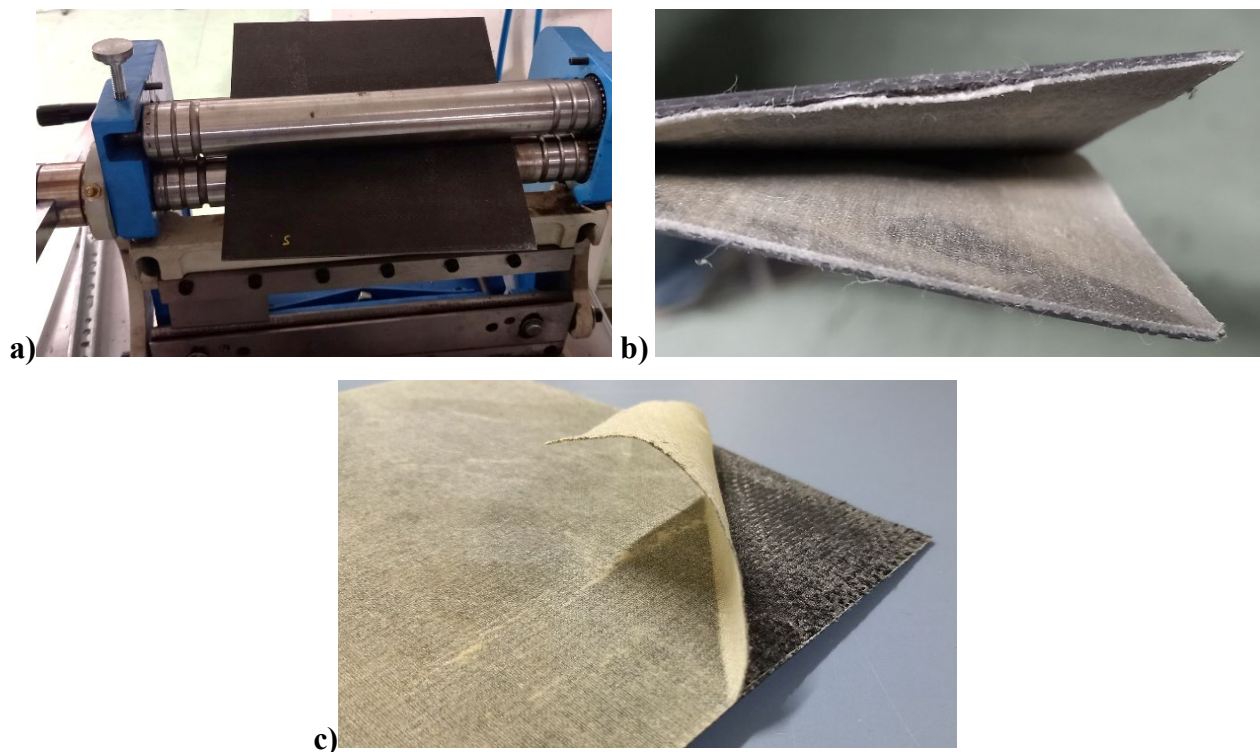


Fig. 2. Thermo-mechanical disassembly of a composite laminates: (a) roll-bending system used, (b) the start of delamination and the separation between two hemp plies (c) and among carbon fibre ply and hemp ply.

Mechanical characterization.

The mechanical characterisation of both virgin and recycled laminates was carried out by means of three-point bending tests and interlaminar shear strength (ILSS) tests, in order to evaluate the effect of the recycling process on the main structural properties of the materials.

Three-point bending tests were performed in accordance with ASTM D790 using a universal MTS Exceed E43 universal testing machine equipped with a 50 kN load cell. Specimen dimensions, span length, and crosshead speed were selected as a function of laminate thickness and reinforcement type, with the aim of minimising shear effects and ensuring a reliable evaluation of flexural behaviour. Three specimens for each sample configuration were tested with a span-to-depth ratio of 16:1 was adopted.

The flexural stress σ_f was then evaluated according to Eq. 1, where P is the load in (N), L is the support span in (mm), b and d are respectively the width and the thickness of the specimen in (mm)

$$\sigma_f = \frac{3}{2} \frac{PL}{bd^2} \quad (1)$$

Interlaminar shear strength tests were conducted according to ASTM D2344 using a short-beam configuration to emphasise shear stresses between adjacent plies. For each configuration, three specimens were tested using a span-to-depth ratio of 5:1. The shear stress τ was evaluated according to Eq. 2:

$$\tau = 0.75 \frac{P}{bd} \quad (2)$$

Results and Discussion

Virgin laminates.

The flexural behaviour of the virgin laminates is strongly affected by the reinforcement type and stacking sequence. The carbon laminate (C) exhibits the typical response of CFRP systems under three-point bending, characterised by a linear elastic region with high stiffness followed by an abrupt catastrophic failure. From the experimental results, laminate C shows a flexural modulus of approximately 55 GPa and a flexural strength close to 680 MPa, confirming the high stiffness and strength but limited damage tolerance typical of carbon fibre composites. In contrast, the hemp laminate (H) displays a markedly different behaviour. The stress-strain curve is characterised by a much lower slope and a progressive, ductile response without a clear catastrophic failure. The flexural modulus of H is approximately 7 GPa, while the flexural strength reaches about 130 MPa, corresponding to reductions of nearly -80% in stiffness and -90% in strength compared to the carbon laminate. However, the hemp laminate reaches significantly higher strain levels, exceeding 0.03 mm/mm, highlighting its superior deformation capability and ductile nature, in agreement with the behaviour reported by Pinto et al [6].

The symmetric hybrid configuration (S), with hemp plies positioned at the laminate mid-plane, exhibits a flexural response that combines the advantages of both reinforcements. The initial linear portion of the stress-strain curve is characterised by a flexural modulus comparable to that of the pure carbon laminate. This confirms that placing hemp layers close to the neutral axis does not significantly affect the elastic stiffness of the laminate. The flexural strength of configuration S is around 610 MPa, corresponding to a moderate reduction (around -10%) compared to laminate C. At the same time, the maximum strain increases to about 0.022 mm/mm, indicating a more progressive damage evolution and enhanced deformation capability with respect to the carbon reference. The uniform hybrid configuration (U), characterised by a symmetric distribution of hemp layers throughout the thickness, shows a further modification of the flexural response. The flexural modulus decreases to approximately 43 GPa, reflecting the greater influence of hemp layers located away from the neutral axis. The flexural strength is reduced to about 460 MPa, while the maximum strain at failure remains close to 0.015 mm/mm.

Compared to configuration S, laminate U exhibits lower stiffness and strength but still maintains significantly better mechanical performance than the hemp laminate, confirming the effectiveness of hybridisation in balancing performance and ductility. In Fig. 3, typical stress-strain curves for each sample configuration are plotted.

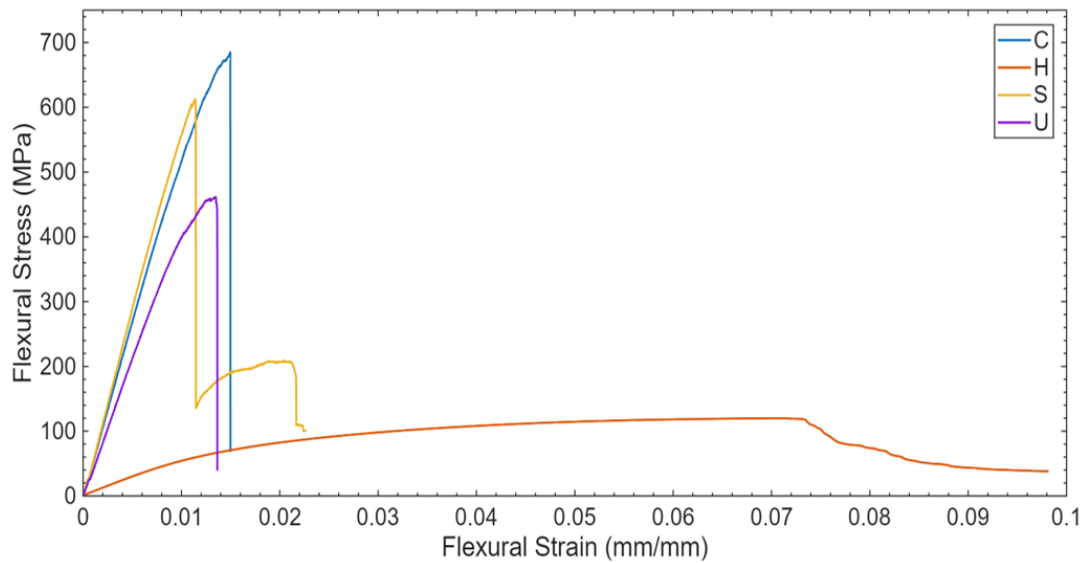


Fig. 3. Typical flexural stress-strain curves for each sample configuration under inspection.

The interlaminar shear behaviour of the virgin laminates further highlights the effect of hybridisation on damage mechanisms, as shown by the stress-displacement curves of Fig.4.

The carbon laminate (C) exhibits the highest ILSS value, reaching approximately 42 MPa, with a sharp peak in the shear stress-displacement curve followed by an abrupt load drop, indicative of a brittle interlaminar failure.

The hemp laminate (H) shows a significantly lower ILSS, with a maximum shear stress of approximately 15 MPa, corresponding to a reduction of about -64% compared to the carbon laminate. The shear response of H is characterised by a smooth and progressive curve, associated with combined shear and flexural deformation rather than a purely interlaminar failure.

Both hybrid configurations (S and U) exhibit an ILSS of approximately 37 MPa, slightly lower than that of the carbon laminate (-12%). The shear stress-displacement curve closely resembles that of laminate C, indicating that the shear load is still mainly sustained by the carbon rich regions of the laminate, while the hemp layers do not significantly compromise interlaminar strength.

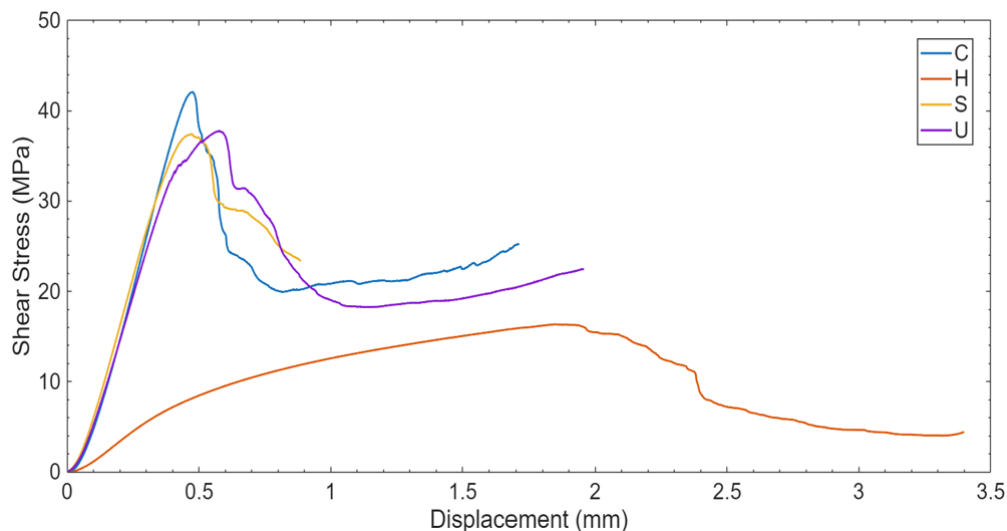


Fig. 4. Typical ILSS stress-displacement curves for each sample configuration under inspection.

Recycling process.

The thermo-mechanical recycling process led to a partial disassembly of the virgin laminates, as the complete separation into individual plies was not achievable for all configurations. In particular, the full hemp laminate (H) was the only configuration that allowed the complete disassembly into all 15 individual plies. This behaviour is directly related to the lower interlaminar shear strength characterising the hemp laminate, which facilitates delamination under thermo-mechanical loading.

Conversely, for carbon and hybrid laminates (C, S and U), the disassembly process resulted in a combination of single plies and small blocks consisting of 2–3 plies still bonded together, due to their higher interlaminar cohesion. For the manufacturing of recycled laminates, only fully disassembled single plies were used, while multi-ply blocks were discarded, in order to ensure a controlled and repeatable restacking process.

Each recovered ply retained a certain amount of residual epoxy resin on its surface, quantified in terms of mass fraction. Specifically, for the recycled carbon plies, the mass fraction of residual resin was 33%, whereas for the recycled hemp plies it was 54%. Details of the recycled laminates are reported in Table 2. The recycled laminates were manufactured using the same processing route and curing parameters adopted for the virgin laminates, with additional epoxy resin introduced to ensure proper impregnation and bonding. As a consequence, the recycled laminates exhibited an increased thickness and a corresponding reduction in fibre volume fraction, as reported in Table 2.

Table 2. Main recycled laminate characteristics.

Label	Thickness [mm]	Fibre volume fraction [%]
rC	4.0	42
rS	4.4	38
rU	4.3	38
rH	4.9	33

Properties of recycled laminates.

The flexural stress-strain curves of the recycled laminates (Fig. 5) reveal a significant modification of the mechanical response compared to the virgin counterparts, mainly associated with changes in material used and fibre volume fraction.

The recycled carbon laminates (rC) exhibits a lower stiffness compared to the virgin carbon laminate, with a flexural modulus of approximately 33 GPa, corresponding to a reduction of about 40%. The flexural strength is also reduced, reaching values close to 275 MPa, compared to approximately 680 MPa for the virgin laminate. Despite this reduction, the stress-strain curve of rC shows a markedly more progressive post-peak behaviour, characterised by multiple load drops and an extended deformation regime, indicating enhanced damage tolerance and energy dissipation capability induced by the recycling process.

The recycled hemp laminate (rH) maintains a very compliant and ductile flexural response. The flexural modulus remains low, around 6 GPa, while the flexural strength reaches approximately 108 MPa. Compared to the virgin hemp laminate, rH exhibits very similar behaviour, with a truly minimal loss of properties; this aspect distinguishes it from all other types of recycled composites. The recycled symmetric hybrid laminate (rS) shows a flexural modulus of approximately 31 GPa, close to that of rC, while the flexural strength reaches about 255 MPa. Although these values are significantly lower than those of the virgin S laminate, the stress-strain curve displays a progressive damage evolution with multiple stress drops and a long post-peak plateau, indicating effective stress redistribution and enhanced deformation capability.

The recycled uniform hybrid laminate (rU) exhibits a flexural modulus of approximately 25 GPa and a flexural strength close to 245 MPa. Compared to rS, rU shows a further reduction in stiffness, consistent with the more uniform distribution of hemp layers through the thickness. Nevertheless, the flexural response remains stable and progressive, with extended strain levels exceeding 0.05 mm/mm,

highlighting the high damage tolerance of the recycled hybrid configuration. It was therefore observed that, unlike the virgin specimens, although a reduction in mechanical properties is evident, the differences between the various hybrid configurations and the full carbon laminate are much less pronounced.

The ILSS results further confirm the effectiveness of the recycling strategy. The recycled carbon (rC), symmetric hybrid (rS), and uniform hybrid (rU) laminates do not show significant variations compared to their virgin counterparts. All these configurations exhibit ILSS values of approximately 30 MPa, indicating only a moderate reduction with respect to the virgin laminates.

This behaviour suggests that the interlaminar adhesion in the recycled laminates is mainly governed by the new epoxy resin introduced during reprocessing, which effectively restores the bonding between adjacent plies despite the presence of residual resin on the recycled fibres.

A different trend is observed for the recycled hemp laminate (rH), which exhibits an ILSS of approximately 17 MPa, slightly higher than that of the virgin hemp laminate. This unexpected improvement can be attributed to the rough surface morphology of the recycled hemp plies, which enhances mechanical interlocking with the newly added epoxy resin. The increased surface roughness promotes a more effective mechanical anchoring mechanism, leading to improved interlaminar shear resistance.

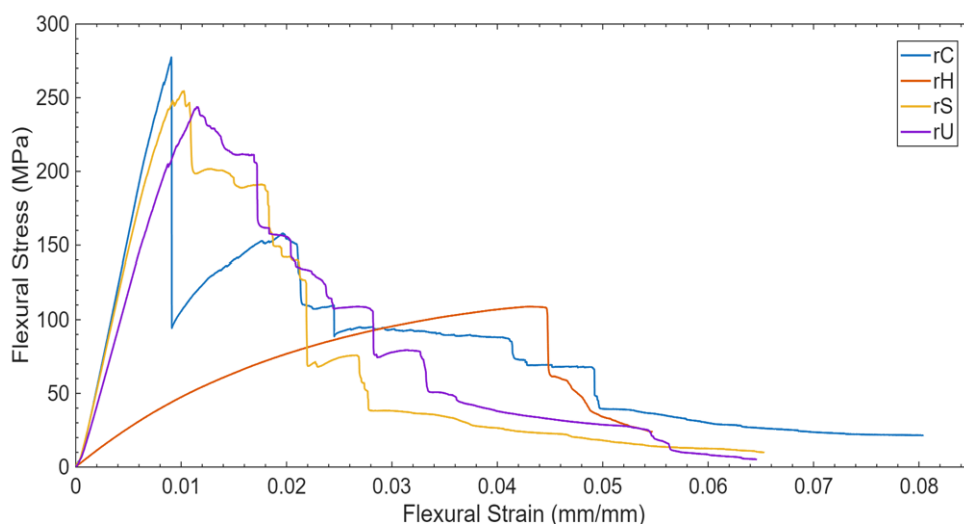


Fig. 5. Typical flexural stress-strain curves for each sample recycled sample configuration.

Conclusion

The proposed thermo-mechanical recycling approach represents a significant conceptual shift compared to conventional composite recycling technologies. Rather than relying on destructive fibre-matrix separation or matrix degradation, this strategy exploits the increased ductility of thermosetting matrices at temperatures above the glass transition to promote controlled laminate delamination. Through the application of suitably calibrated mechanical deformation, composite laminates can be disassembled into individual or partially bonded plies while preserving fibre integrity and fibre-matrix adhesion. The experimental results demonstrate that this approach can be successfully applied not only to full carbon laminates but also to hemp/carbon hybrid composites, and full hemp composites enabling the recovery of reusable reinforcements with limited damage. Although recycled laminates exhibit a reduction in stiffness and strength mainly associated with changes in fibre volume fraction and additional matrix content, they retain stable, progressive mechanical responses and satisfactory interlaminar shear properties. Notably, after recycling, the mechanical differences between hybrid configurations and full carbon laminates become less pronounced, highlighting the effectiveness of hybridisation in combination with the proposed recycling route.

Overall, the thermo-mechanical disassembly strategy allows the recovery of high-added value structural elements while reducing energy consumption and secondary waste generation, opening new perspectives for up-cycling thermoset-based hybrid composites and improving their overall sustainability.

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