RECYCLING THERMOSET BIOBASED COMPOSITES: A CASE STUDY ON FLAX/FURAN COMPOSITES

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ABSTRACT

Composite materials are valued for their high stiffness, strength, and low density, offering durability, which makes them a popular choice for sustainable design. However, recycling composites at the End-of-Life (EoL) remains a challenge, particularly for thermoset composites. While bio-based composites reduce CO_2 emissions, and reduce dependence on fossil resources, they are not infinitely available.

This study used the Material Driven Design method to explore EoL reprocessing options for flax/furan composites, incorporating mechanical and user experience tests. A recycling process based on intentional delamination was identified, enabling recovery of oriented long fibre mats for re-use. To demonstrate its technical feasibility, a car dashboard for the Ecorunner car was designed and produced using recycled flax/furan composite material. The tensile strength of material patches ranged from 70-118 MPa, with Young's modulus of 9-24 GPa.

The dashboard production and user tests confirm the potential for multiple-use cycles of the flax/furan composite. This research demonstrates a novel recycling process for flax/furan composites, and shows how it can be integrated into industrial applications, offering valuable insights into the challenges and opportunities of recycling composite materials. As composite recycling methods continue to develop, these findings can stimulate further uptake of sustainable materials in the composite industry.

Keywords: Flax/Furan, Biobased, Natural fibre, Sustainability, Thermoset, Recycling

1. INTRODUCTION

The flax plant absorbs CO_2 during their growth. With the help of sunlight and water, the CO_2 is converted into carbohydrates and oxygen [1]. These flax fibres can be used to make different materials from linen to biobased composites. This means that these materials store CO_2 in the form of carbon hydrates and oxygen. If more CO_2 is stored than used, e.g. for production, during the lifecycle of the material, it can be said that this material is carbon negative [2].

With this in mind, we can understand that biobased materials are a sustainable choice when compared with fossil-based materials. However, it is important that the CO_2 that is captured in the material will be stored as long as possible [3]. Moreover, while bio-based composites reduce CO_2 emissions, and reduce dependence on fossil resources, they are not infinitely available. Therefore, it is important to look into the recycling opportunities for products made of biobased materials to ensure that this carbon storage can be extended.

Additionally, the durability of biobased composite material applications plays an important role in the acceptance and appreciation of users, which is essential for the successful implementation and long-term value retention of biobased composite materials. While recycling often leads to a decline in mechanical properties, it is not clear what the impact is on the experiential qualities. If these qualities change, could they be used to add new value, to enhance user engagement and acceptance?

This study will address the challenges of preserving mechanical and experiential properties by exploring opportunities to reprocess and extend the lifecycle of biobased composite materials.

2. RESEARCH SETUP & EXPERIMENTS

2.1 Materials

To study the recycling opportunities for biobased composites we used a flax/furan composite. The reinforcements were supplied by Eve-Reverse as fibre preforms consisting of scutched flax, processed in a unidirectional tile, including a natural stabilising agent to prevent fibres from splitting apart: the “Eve-tile”. The Eve-tile embraces the finite fibre length of natural fibres, reducing the energy required to make fibres continuous by eliminating processing steps like drawing, spinning, and weaving. Individual Eve tiles were stacked in a mould with overlapping edges, after which a resin was added, and the mould was sealed off with a vacuum bag.

A number of resin types can be used in combination with the Eve tile. For this study, the resin furan was selected because furan is a biobased thermoset polymer. Thermoplastic polymers are well known for having the advantage that they can be thermally processed, which gives opportunities for recycling. However, the lack of knowledge about the recycling possibilities for thermoset matrices makes it interesting to find out what is possible when combining flax with a thermoset polymer like furan. Additionally, furan has good fire and chemical resistance and is easy to process.

We tested two layups and two material conditions. Firstly, we tested uni-directional samples of five layers, this gave the maximal properties because all fibres are oriented in the direction of effort. Secondly, a multi-directional lay-up was tested to get more realistic values as multi-directional composites are more commonly used in existing products. The multi-directional lay-up has three layers in the direction of effort, and two layers perpendicular to the direction of effort. Lastly, the material was tested in first-cycle and recycled conditions, to evaluate the effect of the recycling process on the material properties, only the multi-directional material is recycled.

The uni-directional, multi-directional and recycled samples were made with flax tape to represent the properties of Eve tiles. This is because the listed material tests need a large number of test samples, while making Eve-tiles in the current lab setup is still time-consuming. Flax tape however, is readily available in the desired size, shape and quantity. The main difference between Eve-tiles and flax-tape is that flax fibres in flax-tape are made continuous, and the Eve-tile is made while considering the finite fibre length of flax.

The first-cycle material was made by cutting flax tape to the required dimensions and evenly applying furan resin. These plies were b-staged in the oven at 50°C for 16 hours. All samples consisted of 5 plies; laminates were assembled in both uni-directional and multi-directional (0-90-0- 90-0) layups, put under a vacuum and cured in the oven for 4 hours at 115°C. The multi-directional material was recycled to obtain reusable fibre patches. This means that the recycled material has 3 layers in the direction of effort and 2 layers perpendicular to this direction. To reuse the patches in a second use cycle, for sample testing and demonstrating, the recycled patches were "glued" back together with a little bit of furan.

2.2 Material Driven Design method

To explore the reprocessing and design opportunities for the case material, we used the Material Driven Design methodology. Unlike traditional design methods, where material is selected based on a set of functional requirements set by the design, Material Driven Design takes the material as a starting point. This method helped to push the boundaries of traditional material development and product design by exploring the ‘unknown’ [4]. For this study, elements of the MDD method that are relevant to the scope of this project were be used.

2.2.1 Tinkering

To get to know the selected flax/furan material, we started with the first step of the MDD method: tinkering. Within the MDD method, this entails conducting a range of mini-experiments without having any assumptions. This activity aims to learn more about the qualities of the material, recognise the constraints, and identify its potential. The tinkering activities that were performed are: cutting with guillotine scissors, lasercutting, cutting with a stanley knive, bending, sheet forming, blending, granulating, boiling (for 2 hours), sanding, putting a hole in the sample, putting a sample in the oven (for 1 hour at 115 °C), putting it on hot metal, and putting it in water, ink, acetone and vinegar for 24 hours. The tinkering activities were performed with both first-cycle and recycled material.

2.2.2 Mechanical tests

Tensile and three-point bending tests were performed to create an overview of the technical properties of the first-cycle and recycled material. The density was measured using a densimeter.

Tensile tests were performed to measure the tensile strength and Young’s modulus of the material. The standard ISO 527-4 [5] was used, which gives information about test conditions for isotropic and orthotropic fibre-reinforced plastic composites. Three-point bending tests were performed to measure the flexural strength and flexural modulus. The standard NEN-EN-ISO 14125 [6] was used, which gives information about determining the flexural properties of fibre-reinforced plastic composites. Both tests were performed on the Zwick/Roell ZMART PRO with hydraulic grips, at room temperature.

2.2.3 User Experience tests

To find out how people perceive the first-cycle and recycled materials on sensorial, interpretive, affective and performative level, the Ma2E4 toolkit was used [7]. This toolkit provides vocabulary and structure, making it easier for participants to explain how they experience a specific material.

The user test was performed with the first-cycle and recycled material. Every participant performed both tests to maintain consistent contextual variables. It is important to note that the material given to the participants first was randomly chosen. Two participants started with the first-cycle material, and three with the recycled material. No context about the material was provided to the participants on beforehand.

This user test is a one-on-one activity performed in a private room with sufficient natural lighting. Participants' hands were recorded during the sessions to analyse their interactions with the material, and comments were noted. The user test consisted of questions being asked on a performative, sensorial, emotional, and interpretive level, the answers were noted in the user's booklet (Appendix 1). The test was done with 5 participants, this is a small sample size to retrieve qualitative insights. Among the 5 participants were two men and three women between 23 - 27 years old.

2.3 Shapeability

A brick pattern of two layers was designed to create a material with the length of the fibres in two directions (Appendix 2). To test this pattern and to determine the forming limits of the recycled material, it was formed into shapes of increasing complexity. Three shapes were selected: a sphere (bending radius ≈ 95 mm), half a cylinder (bending radius ≈ 5mm), and a conic shape (bending radius ≈ 1-20 mm). This way, a double curved shape, a single curved shape, and a curve with changing diameter was tested. The recycled patches were placed in the mould following the brick pattern using furan resin to bond the layers. A vacuum was created over the moulds, and the material was cured in the oven for 4 hours at 115 °C.

2.4 Demonstrator Product

During the exploration phase of this project, the tests mentioned above were conducted to create a list of requirements. These summarised the processing limits of the materials, their advantages and weaknesses and preliminary criteria for potential use cases. Based on these requirements, several ideas for bespoke reuse opportunities were identified, from which a demonstration case was selected. Using the developed recycling process, a prototype was produced and tested in-situ to evaluate and demonstrate the recycling potential of biobased composites.

3. RESULTS & DISCUSSION

3.1 Recycling process

The two most important findings of the tinkering activities were found during sheet forming and boiling a sample. Sheet forming the sample makes the different layers of the composite material split up, however the layers tended to break. Boiling the sample makes the material softer and more flexible, allowing the different layers to be separated without breaking. Combining these findings resulted in the recycling process intentional delamination, which will be used to produce the recycled material. The different steps of the recycling process are described in Table 1.

Recycling step	Details
1. Cutting the first-cycle material in desired patch dimensions	15 cm (length) x 6 cm (width)
2. Boiling patches	2 hours in boiling water
3. Airdrying	
4. Putting through sheet forming machine	*Layers start to separate*
5. Pulling the different layers by hand	*Complete separation*
6. Place two layers in the desired mould	Add a little bit of furan between two layers
7. Seal off the mould with a vacuum bag and cure in the oven	4 hours in the oven at 115°C

Table 1: Recycling steps

3.2 Tensile test outcomes

3.2.1 Tensile test results

Table 2 and Figure 1 show the results of the tensile test. The tensile strength of the Eve-tile and uni-directional samples show similar test results. However, the Young's modulus shows a drop of 45,53% when comparing the uni-directional samples to the ones previously tested by Eve Reverse.

The recycled samples are recycled multi-directional material samples. Comparing these to each other, the recycled samples show a 40.9% drop in tensile strength and a 22.7% drop in Young's modulus compared to the multi-directional samples.

	Tensile strength (MPa)	Specific strength (MPa*cm³/g)	Standard deviation (MPa)	Young's Modulus (GPa)	Specific Stiffness (GPa*cm³/g)	Standard deviation (GPa)	Fibre volume (%)	Resin volume (%)	Density (calc.) (kg/m³)	Density (meas.) (kg/m³)***
Eve-tile*	199,76	142,70	23,15	31,19	22,30	2,62	70,44	29,56	1411,32	1106,5
Uni-directional	170,602	125,679	11,394	16,992	12,518	0,318	52,48	47,52	1357,43	1092
Multi-directional	118,705	88,235	6,900	11,657	8,665	0,648	48,44	51,56	1345,32	1099
Recycled	70,152	53,151	9,755	9,009	6,826	0,576	39,95	60,05	1320	1070

Table 2: Results tensile test
**Eve-tile is a uni-directional sample tested previously by Eve Reverse.*
***Calculated based on material properties and dimensions*
****Measured with densimeter*

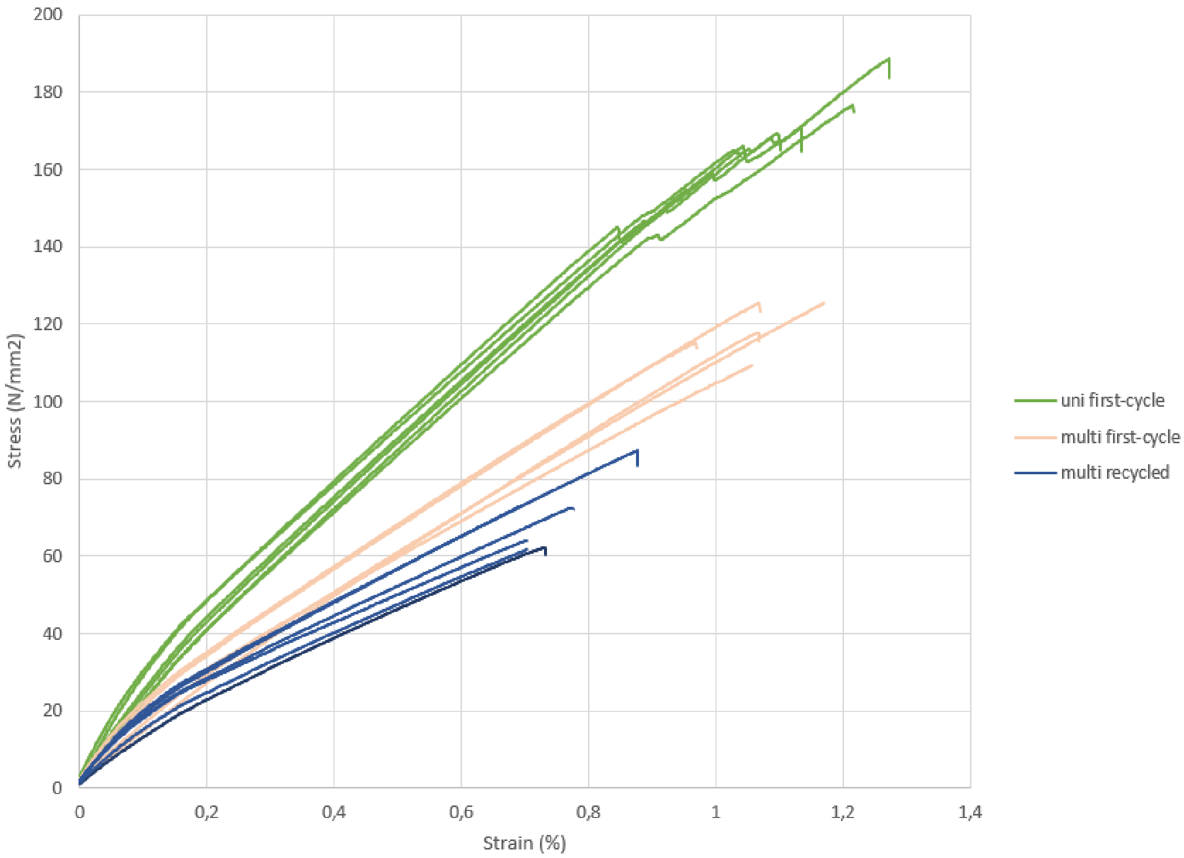

Figure 1: Tensile test results

3.2.2 Tensile test discussion

Small differences between the first-cycle and Eve-tile samples could be explained by variation in fibre volume fraction, which is difficult to control when laminating by hand. The tensile strength of the flax tape is lower than that of the Eve-tile. This could be because the tape uses continuous fibres, while the Eve-tile contains fibres with a finite length, meaning fewer fibre ends along the sample. The Young's modulus of the uni-directional samples was much lower than the values of Eve Reverse. This could be caused by a combination of factors like fibre quality, harvest conditions, or the binder.

The recycled samples show that some stiffness remains after recycling, but the strength of the material is reduced a lot (-40,9%). Many samples failed at the transition between the narrow and wide parts of the dogbone, so it is unclear whether the actual failure point was reached.

3.3 Three-point bending test outcomes

3.3.1 Three-point bending test results

Table 3 and Figure 2 show the results of the three-point bending test. The results from the flexural test show lower values for the uni-directional samples compared to the Eve-tile samples. The flexural strength and modulus are 43% and 48% of the Eve-tile values.

The recycled samples are recycled multi-directional material samples. Comparing these to each other, the recycled samples show a 24.2% drop in flexural strength and a 24.5% drop in flexural modulus compared to the multi-directional samples.

	Flexural strength (MPa)	Specific strength (MPa*cm^3/g)	Standard deviation (MPa)	Flexural Modulus (GPa)	Specific Modulus (GPa*cm^3/g)	Standard deviation (GPa)	Fibre volume (%)	Resin volume (%)	Density (calc.) (kg/m^3)**	Density (meas.) (kg/m^3)***
Eve-tile *	406,90	290,64	54,10	36,80	26,29	2,00	70,44	29,56	1411,32	1106,5
Uni-directional	174,975	128,901	10,577	17,625	12,984	1,252	52,480	47,52	1357,43	1092
Multi-directional	139,998	104,063	4,024	14,622	10,869	0,645	48,441	51,56	1345,32	1099
Recycled	106,097	80,385	25,091	11,045	8,368	2,617	39,95	60,05	1320	1070

Table 3: Results three-point bending test
**Eve-tile is a uni-directional sample tested previously by Eve Reverse.*
***Calculated based on material properties and dimensions*
****Measured with densimeter*

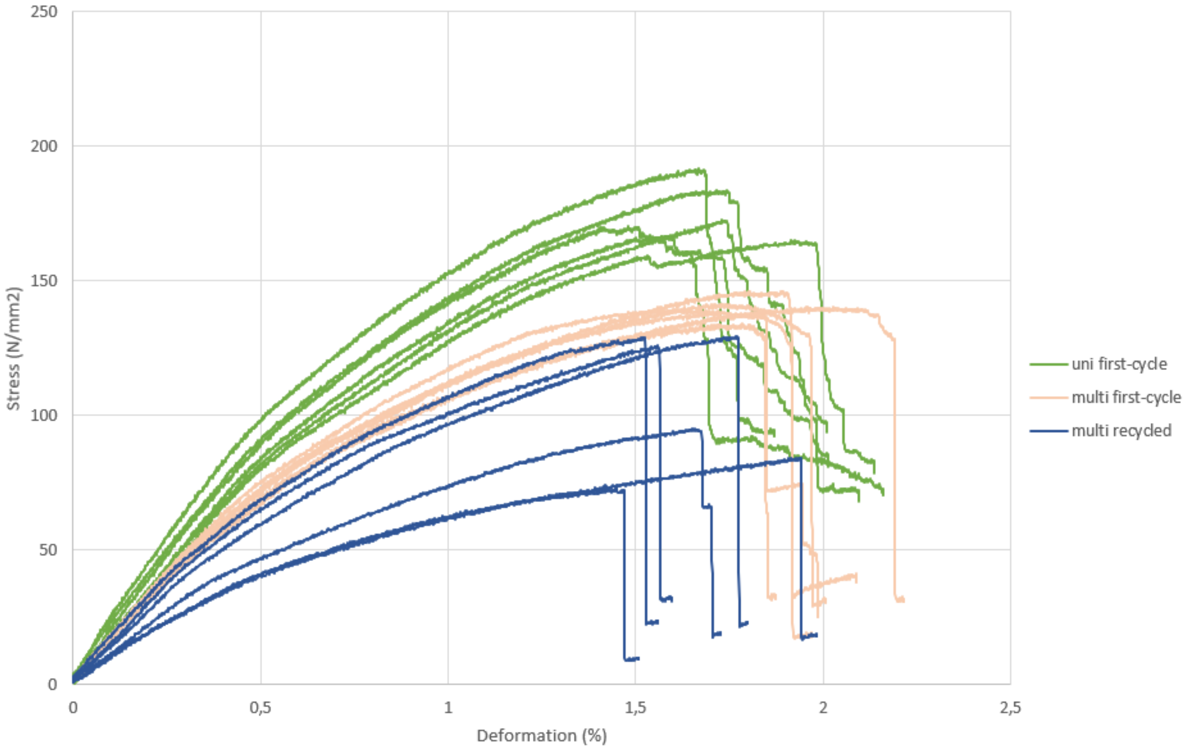

Figure 2: Three-point bending test results

3.3.2 *Three-point bending test discussion*

The lower strength and stiffness of the uni-directional samples compared to the Eve-tile samples, could be caused by inexperience during sample making. For example, some air might have been trapped. This idea is supported by the fact that the measured density is lower, and the samples look different visually.

The recycled samples show more variation in test outcomes than the other sample types, which is shown by a higher standard deviation, and this is also visible in the flexural stress-strain curves. The larger spread makes the material less suitable for applications where mechanical performance needs to be reliable.

The drop in properties could be caused by a combination of factors: a lower fibre volume fraction due to extra resin, porosity, and degradation during boiling and drying.

Table 4 shows a theoretical recycled value based on observed reductions. Properties of recycled Eve-tiles are estimated to be between 70.15–118.05 MPa for tensile strength, 9.01–24.11 GPa for Young's modulus, 106.10–234.86 MPa for flexural strength, and 11.05–23.66 GPa for flexural modulus.

	Tensile strength (MPa)	Young's Modulus (GPa)	Flexural strength (MPa)	Flexural Modulus (GPa)	Fibre volume (%)	Resin volume (%)	Density (meas.) (kg/m³)
Eve-Tile	199,76	31,19	406,90	36,8	70,44	29,56	1106,5
Uni-directional	170,602	16,992	174,975	17,625	52,48	47,52	1092
Multi-directional	118,705	11,657	139,998	14,623	48,44	51,56	1099
	-40,9%	-22,7%	-24,2%	-24,5%			
Recycled	70,152	9,009	106,097	11,045	39,95	60,05	1070
Theoretical value	118,05 (59,1%)	24,11 (77,3%)	308,37 (75,8%)	27,80 (75,5%)	(Percentage of Eve-tile values)		

Table 4: Overview of the test and theoretical values

Compared to other materials, the recycled composite is stronger and stiffer than most wood and plastic, but weaker than traditional composites. However, in practice, the performance may be lower due to overlapping patches and more uncertainty in the recycled material.

3.4 User Experience Outcomes

The glossy finish and dark brown color of the first-cycle material (Figure 3) make the material to be perceived as elegant but strong, luxurious and finished. The dark brown color also gives a modern, boring and nostalgic experience. The sample is perceived lightweight to its looks.

Figure 3: First-cycle material sample

The recycled material (Figure 4) has different fibre patches, which makes the material perceived as natural but also fragile and unfinished. The patches evoke curiosity, fascination, surprise and confusion. Lastly, they make the material look shapeable.

Figure 4: Recycled material sample

The first-cycle material is perceived as luxurious and elegant yet somewhat boring. In contrast, the recycled material is described as unfinished, but its texture and patches spark curiosity, fascination and surprise. While some experiential qualities go down, new positive qualities have also emerged. In the design process, it is crucial to incorporate these new qualities while minimising the negative ones.

3.5 Shapeability

The shapeability has been tested by forming the material around different shaped moulds (Figure 5). The single curved mould with small diameter worked good. The material was able to replicate the small radius of the mould perfectly. The mould with changing diameter worked good as well. The material was able to follow the changing diameter of the mould really well. The sphere mould did not work out as intended. The material formed over the spheric mould shows some creases over the length of the fibre patches and some patches slide down, which made that only one layer of material was left and small holes were visible between the patches of this one layer.

Figure 5: Shapeability test results

When designing the demonstrator product with the recycled material it is important to keep in mind these shapeability limitations.

3.6 Demonstrator Product

To test the procedures and demonstrate the technical feasibility of the recycled material, a car dashboard for the Ecorunner car is designed and produced (Figure 6). Producing the demonstrator product resulted in a lot of interesting findings about the recycling method and production of the material. The final result also shows some challenges and opportunities for further development of the material, process and design opportunities.

Figure 6: Demonstrator product

3.6.1 Recycling method

Four batches of first-cycle material are made. One batch was b-staged longer than 16 hours, which gave the material a darker look. For this specific batch, the recycling method did not work as intended. The separated patches broke before complete separation, probably because the material was brittle. The recycling went very smoothly for all other batches, and a lot of recycled material was made this way. Over time, the patches curled up, probably due to moisture. The curled-up patches were not a problem for production. The outside layers of the laminate are curled up because they are asymmetrical. The middle layers are symmetrical and stayed flat. Some curled-up patches were placed underneath a heavy plate to make them flat again. It is important to completely airdry the patches before storing them. A few patches were not completely dry before stacking them on top of each other. The moisture caused mould to develop on these patches.

3.6.2 Production

It was difficult to follow the pattern as it was intended. The brick pattern in one layer was easy to maintain, but placing the second layer accurately was a challenge due to the complex shape of the dashboard. Rolling the furan on the patches made it hard to control how much resin was added to each patch. Estimating the total amount added is possible, but it's unclear how it spreads across the patches. A challenging part compared to placing first-cycle material is that the patches don’t stick to the mould. The outsides are dry, so the patches move around easily when placing them, causing holes and misplacement, which gives a messy look. To keep the patches in place, heat resistant blue masking tape was used. This can be removed after curing in the oven, but gives limitations for scaling up. It was also challenging to position the vacuum foil onto the screen section of the mould, likely because the foil crease had too small a radius compared to the depth of the indent. If such a small radius is required, the indent should be closer to the surface. Otherwise, the radius should be bigger or the indent itself increased in size.

3.6.3 Final result

The more complex the shape, the more difficult it is to follow the pattern and the more difficult it is to ensure an even material distribution. It was challenging to ensure the vacuum was in the sharp edge of the screen. The result is curved edge. When designing with this specific type of recycled material, it is recommended to use shapes with smoother transitions. The radius of this screen was too small for the depth of the screen. However, the conic shape on the bottom of the dashboard is an example of a shape that would be easy to follow for the material. Now that it is clear that achieving a smooth surface on a more complex shape is not possible, other applications than a car dashboard might be more suitable. Applying a coating to the sides will help to lock in the open fibres.

3.6.4 Opportunities

The production of the dashboard and development of the recycling method highlighted several opportunities for the future use of recycled biobased composites. One key opportunity is the ability to extend the lifecycle of composite materials through reuse. Using the separating layers method, first-cycle products can be taken apart and reshaped into new patches that are suitable for creating new applications. The dashboard demonstrates that recycled composite material can still be used in a functional and aesthetically pleasing product.

This project also shows the potential of integrating recycling strategies early in the development of new materials. Designing with end-of-life in mind could make reuse much easier in the future. For example, cutting first-cycle materials into specific shapes before curing might simplify the recycling process. There is also potential to optimise patch size and shape for better flexibility during production. Smaller patches or tailored geometries could make forming complex shapes easier and reduce overlapping. These kinds of improvements could support scalability and broaden the use of recycled composites across different applications.

3.6.5 Challenges

At the same time, this project uncovered several challenges that need to be addressed for recycled biobased composites to become a reliable alternative. The most important issue is the reduced mechanical performance of the recycled material. Although the material still retains some stiffness, its tensile and flexural strength is lower. To improve this, it’s important to understand which part of the recycling process causes the most fibre damage. Steps like boiling, drying, or curing may need adjustment to better protect the fibres.

Another challenge is the unknown effect of overlapping patches on mechanical performance. In real products, patches are placed on top of each other to cover a surface, but how this affects overall strength has not been tested. Industrial products also often include coatings or lightweight core materials, which weren't part of this study. These could make recycling more difficult or require extra pre-treatment like sanding or removal before separation.

The recycled material used in this project did not experience a full product life. In reality, materials used for several years may have aged or absorbed moisture, which could influence their properties. Future research should look at how these factors affect recyclability. Finally, no LCA was conducted during this project due to its early stage. However, to validate the sustainability of this recycling method, its environmental impact needs to be assessed and compared to other recycling methods. Addressing these challenges will be essential for making recycled biobased composites a viable and scalable solution.

4. CONCLUSIONS

This research explored the end-of-life potential of thermoset biobased composites through a case study on flax/furan materials. By applying the Material Driven Design method, a recycling process based on intentional delamination was developed, allowing for the recovery and reuse of long fibre mats. These recycled patches were used to produce a car dashboard, demonstrating the feasibility of multiple-use cycles for flax/furan composites.

The material tests confirmed a measurable reduction in mechanical properties after recycling, with an average drop of around 25–40%, depending on the test. Despite this, the recycled material still showed sufficient stiffness and strength for various applications and performed better than many natural materials. User experience tests revealed that although some qualities of the first-cycle material were lost, the recycled material gained new characteristics such as a natural look, shapeability, and a more expressive aesthetic, offering opportunities to design for emotional engagement.

The production of the dashboard provided valuable insights into the practical challenges and design constraints when working with recycled biobased composites.

Overall, this study shows that with careful material and process design, recycled flax/furan composites can be successfully reprocessed into new, secondary applications. Further research into fibre degradation, overlapping effects, and long-term ageing is necessary, alongside environmental assessments, to validate and scale this approach. By addressing these challenges, recycled biobased composites have the potential to play a meaningful role in the transition towards circular and sustainable materials.

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APPENDIX

Appendix 1: User's Booklet

Appendix 2: Brick Pattern