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Experimental feasibility and environmental impacts of compression molded discontinuous carbon fiber composites with opportunities for circular economy

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ABSTRACT

Compression molding of carbon fiber sheet molding compounds (CF-SMC) is a promising technology to reduce the waste of carbon fiber composites and to drive sustainable development in lightweight applications. Both, the production waste of prepreg scrap and the end-of-life waste of carbon fiber components can be used efficiently in the recycling process with an apparent sandwich structure of virgin and recycled materials. This contribution investigates the technology on the example of a structural automotive component (transmission crossmember). A one-shot compression molding process was developed which enables cost-efficient, large-scale manufacturing with cycle times of about two minutes. The process–structure–property–performance relationship was experimentally characterized with manufacturing studies and prototype tests. A multilayered hybrid structure of virgin and recycled carbon fiber materials maintains the manufacturability and the mechanical performance compared to primary CF-SMC. The environmental impacts were assessed within a comparative life cycle assessment (LCA). Different material and recycling scenarios were analyzed for the components produced. It revealed lower environmental impacts than an industrially used reference version in aluminum in most impact categories. The combination of the compression molding technology with multilayered hybrid carbon fiber composites reveals opportunities for circularity and the multistage use of industrial recyclates for varying quality requirements in lightweight applications.

1. Introduction

The increasing economic wealth of the global community has accelerated the demand and use of natural as well as synthetic resources. The growth rates are especially high in the carbon fiber industry due to the excellent specific material properties, durability, and corrosion resistance of carbon fiber reinforced polymers (CFRP) [1]. With the earlier generations of CFRP components reaching their end of service life, there is growing interest to determine alternative waste disposal and recycling methods. If no recycling methods are implemented on a commercial scale, the end-of-life and production waste of CFRP in the aeronautical and wind power sectors is expected to reach 527374 tons and 483000 tons, respectively, by 2050 (estimates before the Covid-19 pandemic) [2,3]. The accumulating production waste during typical manufacturing processes of CFRP components is estimated to be as high as 50% [1,4,5]. Currently, CFRP waste mainly ends up in landfill or incineration [4,6,7], and thus loses its intrinsic value considering the costly and energy-intensive production of virgin carbon fibers (vCF) [8–10]. Recycling and up-cycling strategies for CFRP are not yet established, but a lot of research in industry and academia is directed towards moving from a linear to a circular economy model for composites [1,10–21]. This move does not only stem from government initiatives and non-governmental organizations, but also increasing consumer awareness and preference for environmentally friendly products. The European Commission's strategy for a circular economy launched in 2018 [22] and the efforts observed from the World Economic Forum and Ellen MacArthur Foundation [23] are key examples of the transition to a circular economy. The underlying concept is to use limited resources in multiple product life cycles

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while reducing anthropogenic greenhouse gas emissions and other environmental impacts. This is challenging, especially when considering the economic aspects of production [10] to meet recycling quotes and climate and energy targets set by legislation. In addition to the development of cost-effective and energy-efficient recycling processes, effective waste separation and fiber grade sorting strategies need to be implemented in the supply chains of product production [5,10,24].

To understand if materials from recycled carbon fibers (rCF) have a better environmental performance than their virgin counterpart or existing metal solutions to be substituted, it is important to assess not only the production phase but the whole product life cycle [20,21,25]. Life cycle assessment (LCA) is a tool which provides a systematic framework that helps to identify, quantify, interpret and evaluate the environmental impacts of a product, function, or service in an orderly way. As a development tool for the future, LCA needs to meet the requirements of eco-design for successful product development. According to the ISO 14040 Environmental management – Life cycle assessment – principles and framework "LCA addresses the environmental aspects and potential environmental impacts (...) throughout a product's life cycle from raw material acquisition through production, use, end-of-life treatment, recycling, and final disposal (i.e. cradle-to-grave)" [26]. An interwoven product life cycle management and LCA can yield improved product development processes while simultaneously considering ecological and environmental issues [27,28]. Dedicated software tools (e.g. MSC Software's MaterialCenter, Hexagon) for state-of-the-art integrative simulation methods of CFRP, i.e., Integrative Computational Materials Engineering, increasingly support the use of LCA. Due to the compelling sustainable requirements, these simulation tools are involving novel modules, which are dealing with material compliance, material exchange, sustainability issues and allow the modeling of the entire life cycle of the materials and components within LCA simulations.

LCA has been applied to assess the environmental impacts for a range of CFRP applications with virgin and recycled fibers to substitute other materials in various industries, such as in aeronautics [21,29,30], in the automotive sector [9,24,25,31], in the wind power sector [32], or in construction [33,34]. Recycling of CFRP has been shown to have lower energy consumption and greenhouse gas emissions compared to the typical waste disposal methods of landfill or incineration [9,24]. Though, Li et al. pointed out that low-value recyclates with poor mechanical properties, i.e., from mechanical recycling, are a major barrier for economically viable products [9]. Thermal or chemical recycling processes can provide high retention of mechanical properties and thus produce high-value recyclates, with pyrolysis being the most mature technology with commercial-scale implementation [1,7,35].

However, LCA is always dependent on the application and cannot be used for an isolated material selection in the design process [20,24,36]. The substitution of materials, production processes, and services for better environmental performance is, therefore, an ongoing process and needs to be assessed constantly. Moreover, Tapper et al. highlighted that most LCA studies on CFRP recycling have been focused on fiber reclamation without including realistic re-manufacturing and that more research should be directed to recycling processes with adequate re-manufacturing to obtain high-performance materials [20].

Hagnell and Åkermo [10] presented an economic model to assess the competitiveness of recycled fibers and concluded high economic potential when used in the appropriate application. They also considered the typical fiber length reduction of current thermo-chemical recycling processes [1,7,35] and proposed a multi-stage closed-loop recycling approach from structural to non-structural applications. This means, that rCF from aeronautics might be utilized in automotive and/or in marine applications and, eventually, end up as powder fillers in thermoplastics. Thus, some sort of down-cycling from high-performance applications with longer fibers to applications with lower structural requirements is inherently involved.

Carbon fiber sheet molding compounds (CF-SMC) are discontinuous fiber reinforced composites and therefore attractive to incorporate rCF due to the typical fiber length reduction. CF-SMC are more suitable for high-volume applications than continuous fiber reinforced composites. They can be implemented in an industrial and cost-efficient compression molding production process. Three-dimensional shapes can be realized due to the discontinuous nature of the CF and thus good flow properties [37–39]. This means greater freedom of design with efficient functional integration and overcoming the common limitations of shell-like structures present with conventional laminate systems. CF-SMC materials consist either of chopped prepreg platelets or CF bundles dispersed in a thermoset resin, which are typically randomly oriented to form the semi-finished sheets. CF fractions usually amount to 40 vol% or above. These sheets can be cut into custom shapes, stacked, and finished in a one-shot compression molding process. CF-SMC feature both the high-performance characteristics of prepregs and the cheaper manufacturing possibilities of SMC [40–42]. Thus, the high strength to weight ratio and possible weight savings of 30% or more [38,40,43] reveal a viable and competitive alternative to replace existing steel and aluminum alloy solutions.

In contrast, secondary materials from CFRP recycling have limited processability and the final product performance is prone to high variations [1,7,35,44,45]. Depending on the industry and the specific quality requirements, these performance variations are either acceptable or not and need to be fully understood. Otherwise, the potential of rCF materials cannot be exploited, and they are not economically viable to replace primary composite materials or existing metal solutions in lightweight applications.

In this study, the focus is on CF-SMC to investigate the experimental feasibility by means of a real case study component in combination with LCA in a holistic eco-design approach to use recycled CFRP for high-value, structural applications. The case study component is a transmission crossmember of a middle-class car. A compression molding process of multilayered hybrid CF-SMC is developed to use the production waste of prepreg scrap and the end-of-life waste of CFRP components for the manufacture of lightweight parts. According to Ashby and Bréchet [46], the term hybrid material is understood here as the combination of properties of different materials to fill a hole in the property-space. SMC materials from rCF offer lower environmental impacts [9,24,31] but have impaired processing and mechanical properties [1,7,35,44,45]. A mix with virgin (primary) materials in a hybrid layup of CF-SMC forming an apparent sandwich structure maintains the desirable attributes of both on the component level. These are the good processability and the mechanical performance of the virgin material along with the better environmental performance of the recyclates.

This concept and recycling strategy are addressed in this work by assessing the following three aspects:

- the manufacturability of the prototypes during compression molding,
- the structural performance of the manufactured prototypes, and
- the product life cycle for various recycling scenarios of CF-SMC compositions.

A prototype mold for the transmission crossmember was realized to gain insights into the material's process–structure–property–performance relationship. Manufacturing studies and structural prototype tests were performed for a benchmark to compare the performance with the primary CF-SMC material. Different material and recycling scenarios were modeled through a comparative LCA to assess the related environmental impacts. The industrially used aluminum version serves as basis for comparison. The transmission crossmember was defined as the functional unit and the whole product life cycle was evaluated, from the extraction of raw materials and energy to end-of-life treatment (cradle-to-grave). All the necessary material and energy data to conduct the LCA was collected during the production of the CF-SMC prototypes. Nevertheless, it must be emphasized that due to the complexity of

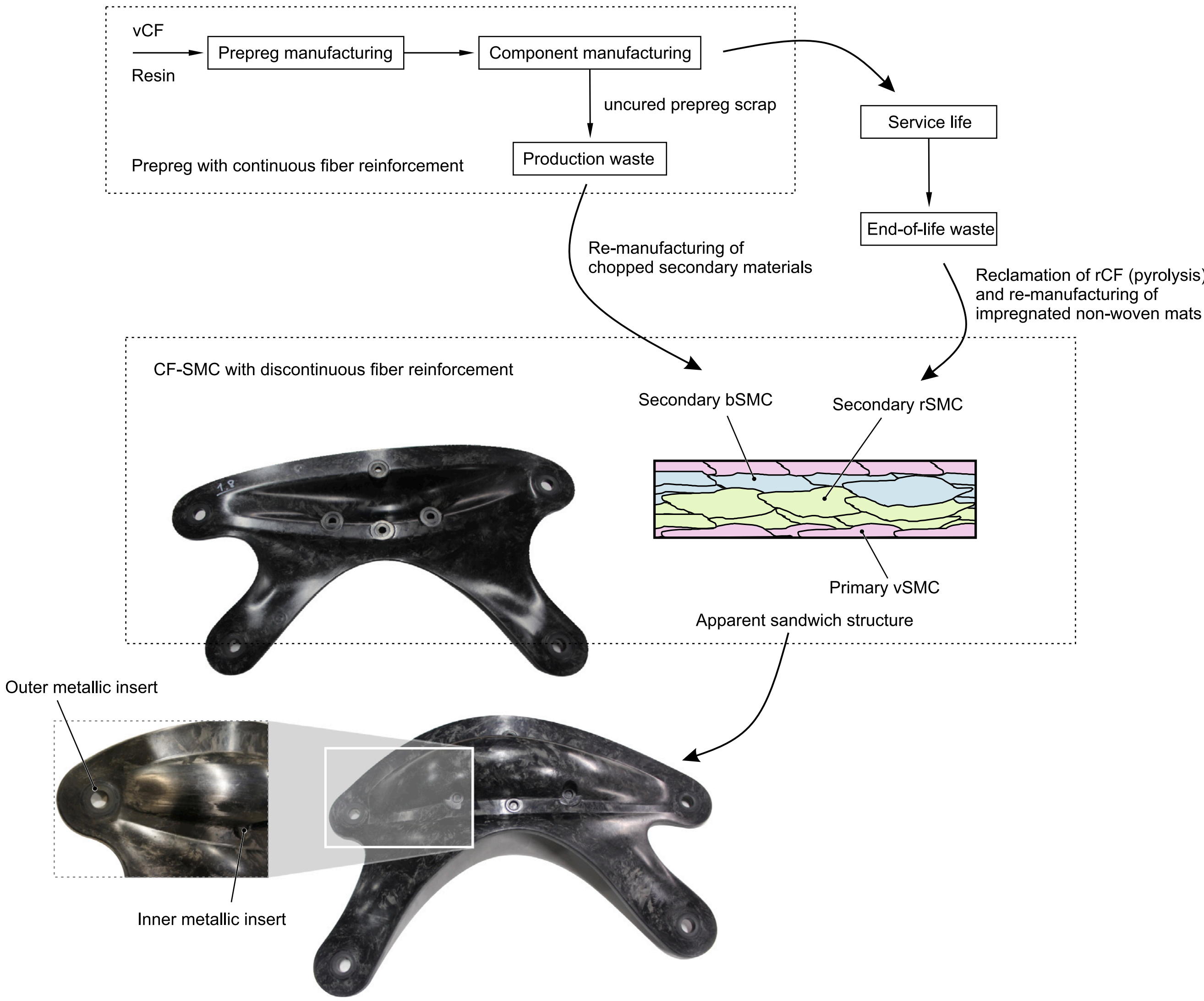


Fig. 1. Schematic workflow of the recycling routes followed in this work with the multilayered hybrid material structure and the automotive transmission crossmember prototype.

Table 1
Material data for the used CF-SMCs (average values are given ± standard deviation).

Acronym	Density (g/cm^3)	Fiber volume fraction (vol%)	Fiber length (mm)	Effective modulus (GPa)	Effective strength (MPa)
vSMC	1.56	56.6	50	39.48 ± 3.93	273.32 ± 46.61
bSMC	1.56	57.0	50	60.61 (est.)	464.64 (est.)
rSMC	1.42	33.5	12.7	28.02 ± 4.6	311.39 ± 44.32

the technical processes specific data cannot always be measured. In that case, generic data from the ecoinvent v3.5 database (ecoinvent Association, Zürich, Switzerland) was used.

2. Recycling scenarios and prototype design

2.1. Materials and automotive prototype

The schematic workflow of the different recycling routes demonstrated experimentally and within the LCA models by means of the transmission crossmember prototypes is presented in Fig. 1. The mix of primary materials (vSMC) and secondary recyclates (bSMC and rSMC) in a multilayered hybrid material structure of CF-SMCs guaranteed the manufacturability and the mechanical performance of the prototypes. A summary of the used CF-SMCs with respective material data is given in Table 1.

The primary material from vCF (vSMC) with the commercial-grade name HexMC®-i 2000 of Hexcel Inc (Stamford, CT, USA) serves as reference material in this study. Unidirectional (UD) prepreg tape is slit and chopped into discontinuous, rectangular platelets, which are randomly distributed to form the SMC semi-finished part. The prepreg platelets have nominal dimensions of 50 mm x 8 mm and an average cured ply thickness of 0.15 mm [47].

The secondary material from prepreg byproduct (bSMC) was obtained from uncured UD prepreg scrap of the industrial series production at Hexcel. Note that this uncured scrap is considered a byproduct from the primary production line rather than waste. The remanufacturing of the byproducts and the preforming were used in terms of a secondary production line of the transmission crossmembers. Thus, the primary and secondary production were coordinated to maintain the quality of the byproducts (same loading pre-history and fiber orientation compared to the primary prepregs). The UD material was used to produce prepreg stacks for BMW AG (Munich, Germany) for demanding automotive components with UD fiber reinforcement and the byproducts could be used in the secondary production of the CF-SMC transmission crossmembers. To form an SMC material with discontinuous platelets of UD alignment, the prepreg mats with the remaining scrap were cut transversely in 50 mm steps. Because of the unidirectionally aligned fibers of the secondary bSMC, it could be used in the outer layers of the hybrid layup to increase stiffness and strength. The aligned fibers with pre-defined orientation in the preform stack served thus as effective reinforcement for the main loading mode of bending in the transverse carrier section of the transmission crossmembers. It has been suggested by various authors [1,7,10,35] that alignment of recycled fibers is key to achieve high-value products and being economically viable.

The other secondary material consists of rCF (rSMC) and was produced by the company Carbon Conversions Inc (Lake City, SC, USA). Carbon Conversions uses a proprietary pyrolysis process to reclaim CF. Non-woven mats of the reclaimed, discontinuous fibers with random orientation were impregnated with epoxy resin on a prepreg line of Hexcel. It was placed as the core material, with the virgin material to be used only in the amount needed in an apparent sandwich structure.

All materials were provided by Hexcel (Hexcel Composites GmbH & Co KG, Neumarkt, Austria). The CFs were pre-impregnated with the same grade of epoxy resin, Hexcel's HexPly® M77. It is a fast-curing resin suitable for automotive applications where short production cycle times and low energy are necessary. The degree of quasi-isotropy due to random fiber orientation for vSMC and rSMC was characterized by means of tensile tests. The tensile test results are given in Appendix A. While vSMC showed overall quasi-isotropic properties, rSMC featured a rather pronounced anisotropy of mechanical properties due to the re-manufacturing process of the reclaimed fibers. It is worth mentioning that rSMC has higher strength than vSMC, even though the fiber volume fraction and stiffness are smaller. This can be explained by the different fiber grades and the nature of their morphology. The non-woven mats of rSMC are made from recycled aerospace-grade fibers, which are stronger than the industrial-grade fibers used in vSMC. However, the semi-laminated meso-scale morphology of the prepreg platelet based vSMC allows for higher fiber volume fractions with less fiber distortions and thus results in higher stiffness [40,48,49]. Average values are reported in Table 1. For bSMC, estimated values are provided for the effective modulus and strength. The values are based on a knockdown factor comparing high-flow (high degree of fiber orientation) and low-flow (in-plane random fiber orientation) CF-SMC specimens from an experimental study by Martulli et al. [50] and thus represent rather conservative estimations.

The case study component of this work, the transmission crossmember, represents a structural automotive component (Fig. 1). The transmission crossmember supports the transmission and is connected to the vehicle frame. It was designed according to the performance specifications and design space requirements of an OEM. The design target was to comply with a minimum load carrying capacity for structural performance in terms of the maximum load under the critical load case. The nominal wall thickness of the component is 8 mm. The main dimensions are 520 mm x 340 mm x 100 mm. Aluminum inserts were co-molded to ensure adequate joining and load introduction with the surrounding chassis (four outer inserts) and the transmission (four inner inserts). The inserts were hard coat anodized to prevent galvanic corrosion.

2.2. Scenarios and material configurations

Six different scenarios of the transmission crossmember were investigated in this work and are summarized in Table 2. Each scenario represents a nominal material configuration with varying weight fractions of secondary materials and assumptive recycling strategies for modeling the LCA. The CF-SMC scenarios are divided into two groups. The first group (V, B-1 and B-2) consists only of materials produced from vCF with the aim to evaluate how the use of bSMC from prepreg byproduct can lower the environmental impacts compared to the primary vSMC. While the second group (R-1 and R-2) considered the use of rSMC from rCF in a multilayered layup with the materials of the first group.

A systematic experimental study was carried out to assess the multilayered hybrid concept of primary and recycled materials in terms of manufacturability and structural performance. Three material configurations (V, B-1 and R-1) of CF-SMC were manufactured with the same net-shaped preform geometry (resulting in weld lines) and mechanically tested. Reference prototypes (V) were produced of the primary material vSMC for experimental benchmark comparison. Moreover, a second preform geometry was employed in the case of (V) to avoid weld lines. This is interesting in terms of product engineering because weld lines reduce the strength significantly, but a change of the preform also affects the in-mold material flow and thus the component stiffness.

The LCA modeling of the six different material configurations were based on realistic and idealistic scenarios of assumed compositions to demonstrate the possible environmental potential of using the recyclates in industrial production. The modeling assumes that the same product function is satisfied without changing the geometry and processing. The transmission crossmember in aluminum (A), which was used commercially in the vehicle by the OEM, served as the basis for the comparison of the environmental impacts. The aluminum version (A) was made of die-cast aluminum (2149 g) and polyurethane acoustic foam (340 g).

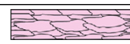
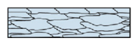
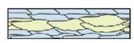
3. Methodology

3.1. Process development

The transmission crossmember features eight aluminum inserts, which could be integrated into a one-shot process (Fig. 2a). No additional manufacturing steps were necessary to mount the inserts. Special hold-down devices with spring elements were integrated into the moving mold side to seal the inserts and to prevent material flow into the insert holes. The steel tool was manufactured by Alpex (Alpex Technologies GmbH, Mils, Austria). Two diaphragm pressure sensors were used in the cavity to monitor the pressure during compression molding. The mold was heated with oil. The hydraulic press Engel v-duo 700 (Engel Austria GmbH, Schwertberg, Austria) with a maximum clamping force of 7000 kN was used for the manufacturing of the prototypes.

Net-shaped preforms were automatically cut with an NC cutter from the SMC rolls (or from the prepreg cutoffs for bSMC) and stacked to obtain the predefined charge weight. The stack together with the aluminum inserts was then transferred into the heated mold at temperatures ranging from 135 °C until 150 °C. The transfer can be handled automatically by a robot with suitable end-of-arm tooling. As soon as the material was located inside the mold, the machine closed at maximum speed and the cavity was evacuated. Vacuum evacuation of the mold was important to fill the mold, especially for complex geometries. Although, the curing process initiates at the contact interfaces between the material and the hot mold, cross-linking mainly happens after mold filling [51]. After closing, the mold compacted the CF-SMC material in speed-controlled and parallelism-controlled mode with 20 mm/s until a set switch-over point at which the stack was compressed by 20% of its original thickness. The integrated pressure sensors in the cavity were used to guarantee that the material was sufficiently compacted at this point (about 8 bar). At this switch-over point, the material was heated to the processing temperature. After material heating, a force-controlled molding at a set maximum force value and time with parallelism-control took place until final curing. After curing at maximum force, the finished part was ejected. Finishing was necessary due to the slight flash of resin at the shear edge. This

Table 2
Recycling scenarios and material configurations of the transmission crossmember analyzed in the experimental part and/or in the LCA (B-1, R-2 and A are only included in the LCA, average values are given ± standard deviation).

Scenario	Material configuration	Preform	Experimental (process and structural)		LCA (cradle-to-grave)	
			Weight fraction of secondary material	Component weight (g)	Assumed weight fractions of secondary material	Applied allocation of environmental impacts
V	CF-SMC, from primary prepreg (vSMC) only, serve as reference prototypes; with and without weld lines		0w% (100w% vSMC)	1273.09 ± 5.40	0w% (100w% vSMC)	–
B-1	CF-SMC, mix from primary prepreg (vSMC) and prepreg byproduct (bSMC); with weld lines		6.7w% bSMC 17.1w% bSMC 27.5w% bSMC	1290.01 ± 8.78 1287.07 ± 6.88 1284.13 ± 7.09	60 w% bSMC	50:50 allocation bSMC
B-2	CF-SMC, from secondary prepreg byproduct (bSMC) only		–	–	100w% bSMC	50:50 allocation bSMC
R-1	CF-SMC, from primary prepreg (vSMC) and rCF (rSMC); with weld lines		16.5w% rSMC	1279.07 ± 13.69	16.5w% rSMC 30.0w% rSMC 40.0w% rSMC	Cut-off rSMC
R-2	CF-SMC, from prepreg byproduct (bSMC) and rCF (rSMC)		–	–	83.5w% bSMC and 16.5w% rSMC	50:50 allocation bSMC; Cut-off rSMC
A	Aluminum, the industrial version consisted of die-cast aluminum and polyurethane acoustic foam		–	(2489)	30.0w% primary and 70w% secondary aluminum	50:50 allocation from end-of-life recycling

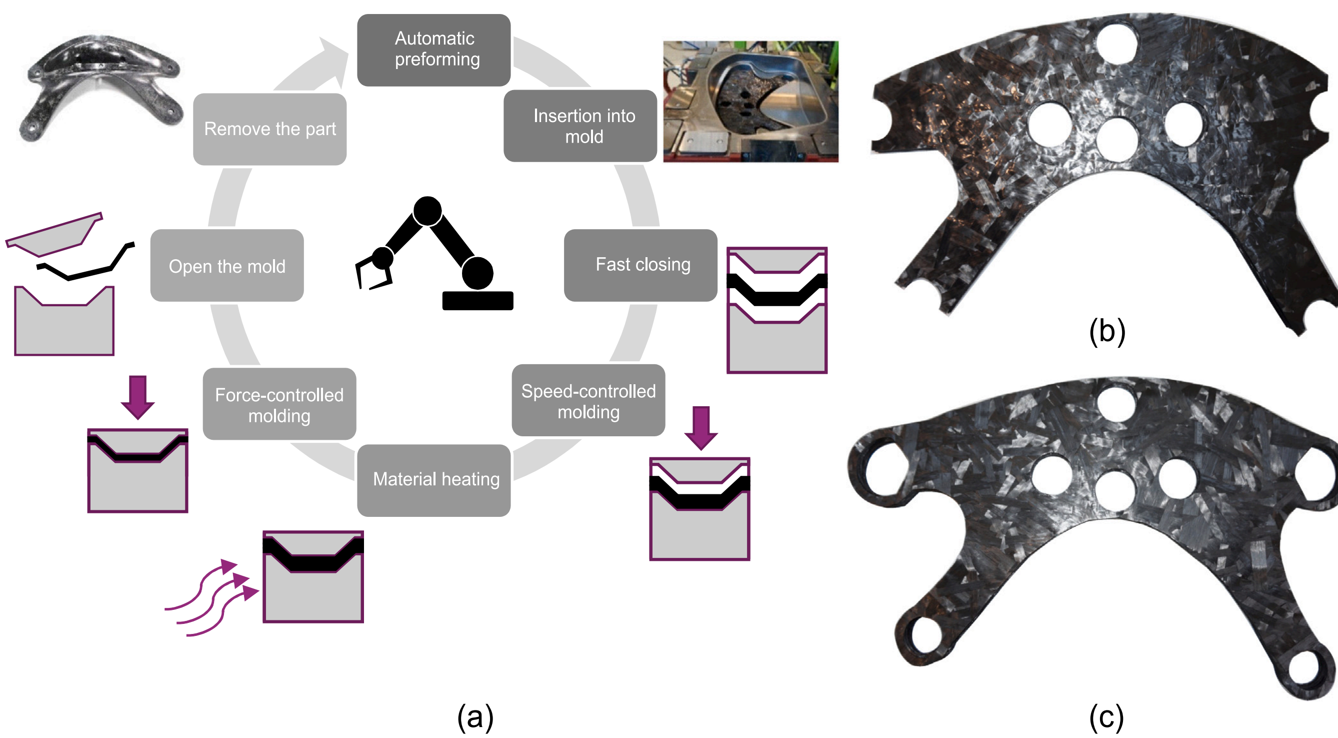


Fig. 2. (a) One-shot compression molding process with aluminum inserts; preforms for the transmission crossmember (b) with weld lines and (c) without weld lines.

was done by trimming with a knife. The weight of the parts remained constant over the production cycles. The ejectors were located at the metal inserts to avoid marks on the part's surface and maintain the smooth surface finish.

Transmission crossmember prototypes were manufactured with three different material configurations (cf. Table 2). The net-shaped preforms for the transmission crossmember were adapted and optimized based on preliminary trials so that the cavity pressure distribution was evenly balanced throughout the mold. All prototypes were manufactured in a low-flow compression molding setup with a coverage of 90% of the mold's cavity. Two different net-shaped preforms were selected for testing. A geometry with weld lines (Fig. 2b) was used for all material configurations (V, B-1 and R-1). This preform with open outer holes (establishing weld lines) was suitable for automatic handling together with the metal inserts by a robot. Otherwise, reliable handling and production stability were not achieved because of the tight tolerances around the inserts which led to misalignments and tilting of the inserts when picking the preform and placing it into the mold. Reliable and reproducible placement and positioning of the stack was important since minor variations can affect the flow behavior during mold filling significantly [52]. However, with these preforms, weld lines result from the material flow around the outer inserts when the two flow fronts collide. Weld lines are unfavorable for the mechanical performance, and thus a second preform geometry

Table 3
Process conditions for the prototype manufacture.

Temperature (°C)	Material heating (s)	Clamping force (kN)	Time of force-controlled molding (s)	Cycle time (s)
135	20	5000	360	405
150	20	5000	90	135

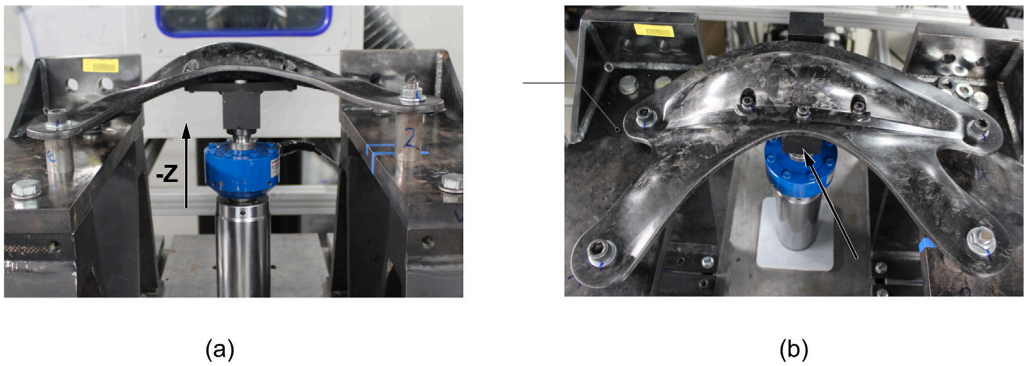


Fig. 3. Component testing set-up for the transmission crossmember: (a) critical load case in -Z direction and (b) an arrow indicating the transmission dummy.

was designed to avoid weld lines (Fig. 2c). The difference between the two preform geometries was assessed by means of the reference configuration (V).

Prototypes of the different material configurations were produced at a processing temperature of 135 °C. The material was heated for 20 s at the defined switch-over point. In the subsequent force-controlled molding stage a clamping force of 5000 kN was necessary to fill the mold. The cycle time was 405 s considering a set time of 360 s in the force-controlled molding stage for a mold temperature of 135 °C. Cycle times of 135 s could be achieved with optimized process parameters at 150 °C mold temperature with force-controlled molding for 90 s. A cycle time study with respective mechanical component tests was conducted to ensure that the curing time was sufficient. The process conditions for the prototype manufacturing are summarized in Table 3.

3.2. Prototype testing

The three material configurations (cf. Table 2) were compared in a benchmark study with the reference prototypes (V). The effect on the structural integrity of the different preform geometries with and without weld lines was investigated based on the reference material. The transmission crossmember prototypes were mechanically tested after OEM requirements on the laboratory scale. The component testing set-up (Fig. 3) features the same outer fixation points as in the vehicle body. The component performance under monotonic, quasi-static loading was investigated using the critical load case in -Z direction (Fig. 3a). A transmission dummy (Fig. 3b) connected to the actuator of the servo-hydraulic test bench introduces the load from the bottom.

The test bench was an MTS 852 damper test system of MTS Systems Corporation (Eden Prairie, MN, USA). During testing, the displacement was measured by the internal LVDT of the machine via the piston movement, and the reaction force was captured by a load cell. The quasi-static component tests were conducted at a testing speed of 2 mm/min and room temperature (23 ± 1 °C). At least three prototypes were tested per material configuration. For (B-1) three prototypes were only tested for the highest weight fraction of bSMC. For the lower weight fractions, two prototypes were tested. The tests were stopped when the reaction force dropped by 25% after surpassing the maximum recorded load.

3.3. Comparative Life Cycle Assessment (LCA)

3.3.1. Scope of the study

Six different scenarios with realistic and idealistic recycling assumptions of the transmission crossmember were studied with the comparative cradle-to-grave LCA (cf. Table 2). The commercially employed industrial version in aluminum served as the basis for comparison (A). For the aluminum option the ecoinvent 3.5 dataset "market for aluminum, cast alloy [GLO]" was chosen for the LCA model. It consists of 30 w% primary aluminum and 70 w% secondary aluminum, whereby about 42 w% is secondary aluminum from external sources and about 28 w% from internal sources. That includes estimations for transports. The LCA study refers to the methodology of the ISO 14040 and 14044 and it was modeled and conducted using the software Umberto LCA+ (ifu Hamburg GmbH, Hamburg, Germany) and the database ecoinvent v3.5. The manufacturing of primary and secondary materials along with the part production is discussed in Appendix A with a flow chart of the different processes (cf. Fig. S2).

The related reference flows of the different material and recycling scenarios differ regarding the particular weights of the transmission crossmembers. The average weight of the transmission crossmember for the aluminum version (A) was 2489.00 g. For the CF-SMC scenarios, the average weights were 1273.09 g (V, B-1 and B-2) and 1279.07 g (R-1 and R-2, including 205.11 g rSMC). The average weights were determined from the manufactured CF-SMC prototypes. The theoretical reduction of environmental impacts with increased use of recycled material was assessed based on the scenario (R-1) with two additional weight fractions of 30 w% and 40 w%. The analysis includes all environmental aspects and potential impacts from raw material acquisition and manufacturing, transports, the weight of the component during the use phase and the respective end-of-life treatment following a cradle-to-grave approach. Regarding the end-of-life treatment, the aluminum transmission crossmembers (A) were assumed to be recycled and in the case of the CF-SMC scenarios incineration was chosen as industrially feasible end-of-life route. Transport data were collected and included in the LCA model for the CF-SMC scenarios. Specific transport data for the aluminum version (A) was not known or collected but was an implicit part of the corresponding ecoinvent v3.5 processes. The geographical and temporal system boundaries relied on the ecoinvent v3.5 data sets, including a time frame from 1990 to 2018. Regarding the energy mix, data sets for Europe were chosen to compare all scenarios. However, the pyrolysis data regarding the recycling process for the secondary used rCF was not available and lies therefore outside the system boundaries of the study.

3.3.2. Inventory analysis

In addition to the collection of primary data for the production of materials and prototypes, secondary data from the ecoinvent v3.5 databases was used. The material and energy data for the manufacturing of the steel tool was also considered in the LCA study. Based on talks with the manufacturer, it was assumed that this tool can be used for the production of 100000 transmission crossmembers. Moreover, the environmental impacts in terms of component weight resulting during the use phase of the transmission crossmember in the passenger car over a life cycle distance of 168000 km were included in the LCA study. Whereas specific and generic data were used for all CF-SMC scenarios, only generic data was available for the aluminum scenario.

For all ecoinvent v3.5 datasets, the system model APOS (allocation at the point of substitution) was chosen [53–55]. Regarding the byproduct in the scenarios (B-1), (B-2) and (R-2) the 50:50 rule was applied, as it is assumed that this byproduct is a residual material and the transmission crossmember production was not the initial purpose of the production of this material. The 50:50 rule was also used for environmental credits that arise from recycling. This is the case in scenario aluminum (A), where credit for end-of-life recycling was calculated. Moreover, there were also credits included for the CF-SMC scenarios as a result of the recycling of material losses. Credits were calculated for the recycling of the material loss of the aluminum inserts and the steel in the tool manufacturing process. Additionally, the cut-off rule was applied for rCF in the scenarios (R-1) and (R-2) because they are considered waste material from another industry and therefore, their environmental impacts are excluded in the LCA analysis.

3.3.3. Impact assessment

A midpoint, as well as an endpoint method, was used for calculating the LCA results. For the analysis of the results at the midpoint level the international reference life cycle data system (ILCD) 1.0.8 2016 method was chosen. Moreover, for the ILCD 1.0.8 2016 midpoint method there are resident equivalents available that serve as normalization factors [56]. Regarding the analysis at the endpoint level, the ReCiPe Endpoint (H, A) method was chosen. For the current LCA study the hierarchist perspective (H), which relies on scientific consensus and is neither seen as an optimistic nor pessimistic approach with an average weighting (A) was chosen for the endpoint analysis [57,58].

4. Results and discussion

4.1. Prototype manufacture

The manufactured reference transmission crossmembers (V) for the experimental benchmark comparison had an average component weight of 1273.09 g with a standard deviation of 5.40 g. The total weight of the eight integrated aluminum inserts was 37.90 g. The component weight remained stable throughout the production cycles with low standard deviations for all material configurations (Fig. 4a). The difference in terms of the average component weight between the material configurations was below 1.4%. The highest standard deviation of about 1.1% occurred for the component weights of the (R-1) prototypes.

The secondary bSMC made of prepreg byproduct cut each 50 mm showed good flow properties, similarly to the commercial reference material. This simplified the recycling of industrial prepreg scrap and might be suitable for series production due to time and cost savings. It was mixed with primary vSMC in a multilayered hybrid material layup with varying weight fractions of bSMC (B-1). The prototypes were manufactured with three different weight fractions of bSMC (6.7 w%, 17.1 w%, and 27.5 w% respectively). Note that the supply of the prepreg byproduct for this study was limited, but preliminary manufacture trials showed that the good flow properties allow higher weight fractions up to 100 w% for complete mold filling.

The recycling material rSMC exhibited insufficient flow properties due to the mats with random, entangled long fibers. Thus, the sole use of such a recycled mat material is not suitable for real applications with complex geometries. However, it was possible to produce transmission crossmember prototypes when using rSMC in a multilayered hybrid structure. As core material with outer layers of primary vSMC it could be processed well without changing the process parameters or the preform geometry and mold coverage (R-1). The (R-1) prototypes had 16.5 w% of rSMC. Due to limitations in the tool's shear edge design and the relatively low aerial weights of rSMC, a higher weight fraction could not be processed. Adaptions of the tool are practically possible and would allow higher weight fractions of the recycled material. It is therefore interesting to assess the theoretically possible benefits of lower environmental impacts for scenarios with higher amounts of rSMC in the comparative LCA.

The locations of the two pressure sensors integrated into the mold cavity are shown in Fig. 4b. Representative clamping force and cavity pressure curves of exemplary prototypes for the different material configurations at a tool temperature of 135 °C are given in Fig. 4c and Fig. 4d. An overshoot force of about 5500 kN was recorded in the force-controlled molding stage before reaching a constant value of the set clamping force of 5000 kN. As a result of the thick-walled geometry, relatively high pressures were measured in the cavity. The cavity pressure reached maximum values of up to 300 bar. A pronounced overshoot pressure with a sharp drop and subsequent further pressure increase was observed for all material configurations. The cavity pressure decreased after reaching a second peak with the progressing cure of the epoxy resin. Because of the inherently stochastic nature of the used CF-SMC materials, the measured pressure curves were subject to strong variation between individual prototypes. The randomly distributed discontinuous CFs lead to a large spread in terms of stack thickness, areal weight, fiber orientation, and defect distribution in the preforms. As a result, the local flow conditions vary significantly.

4.2. Component test results

A representative load–deflection response (Fig. 5a) and typical fracture patterns and failure positions (Fig. 5c) for a quasi-static component test of the transmission crossmember with weld lines are shown on an exemplary prototype of (V). As expected, fracture occurred at the weld lines. After reaching the maximum load of 23.59 kN, a sudden drop in the load–deflection curve indicated a brittle fracture process. Macroscopic cracks were observed optically at the first fractured weld line. However, the damage was localized in the vicinity of the outer fixation point and it did not lead to ultimate failure. The component was able to bear additional loads at considerable deflection values. The test was stopped after fracture at the third weld line at a deflection of about 25 mm. The measured load was around 70% of the maximum recorded load at this deflection value.

The weld lines were weak spots in the structure and determined the strength of the compression molded CF-SMC components. Strength reductions of 60 to 80% [59] and 48 to 88% for high-flow and up to 72% [60] for low-flow compression molded CF-SMC specimens with weld lines compared to the pristine material were observed by other authors. On the component level, strength reductions of 20 to 40% compared to a weld line-free configuration [59] and 422% difference in strength due to variation of the charge pattern with different weld line scenarios [39] were reported. In this study, a reduction of component strength of 9.4% (considering the maximum load) was found between the preform designs either with weld lines or without weld lines for the reference scenario (V). Direct comparisons are not possible, but it is obvious that an optimized process and part geometry with proper tooling and preforming are key for improved structural performance. It is of particular importance to gain deep insights into the process–structure–property–performance relationship on the component-level

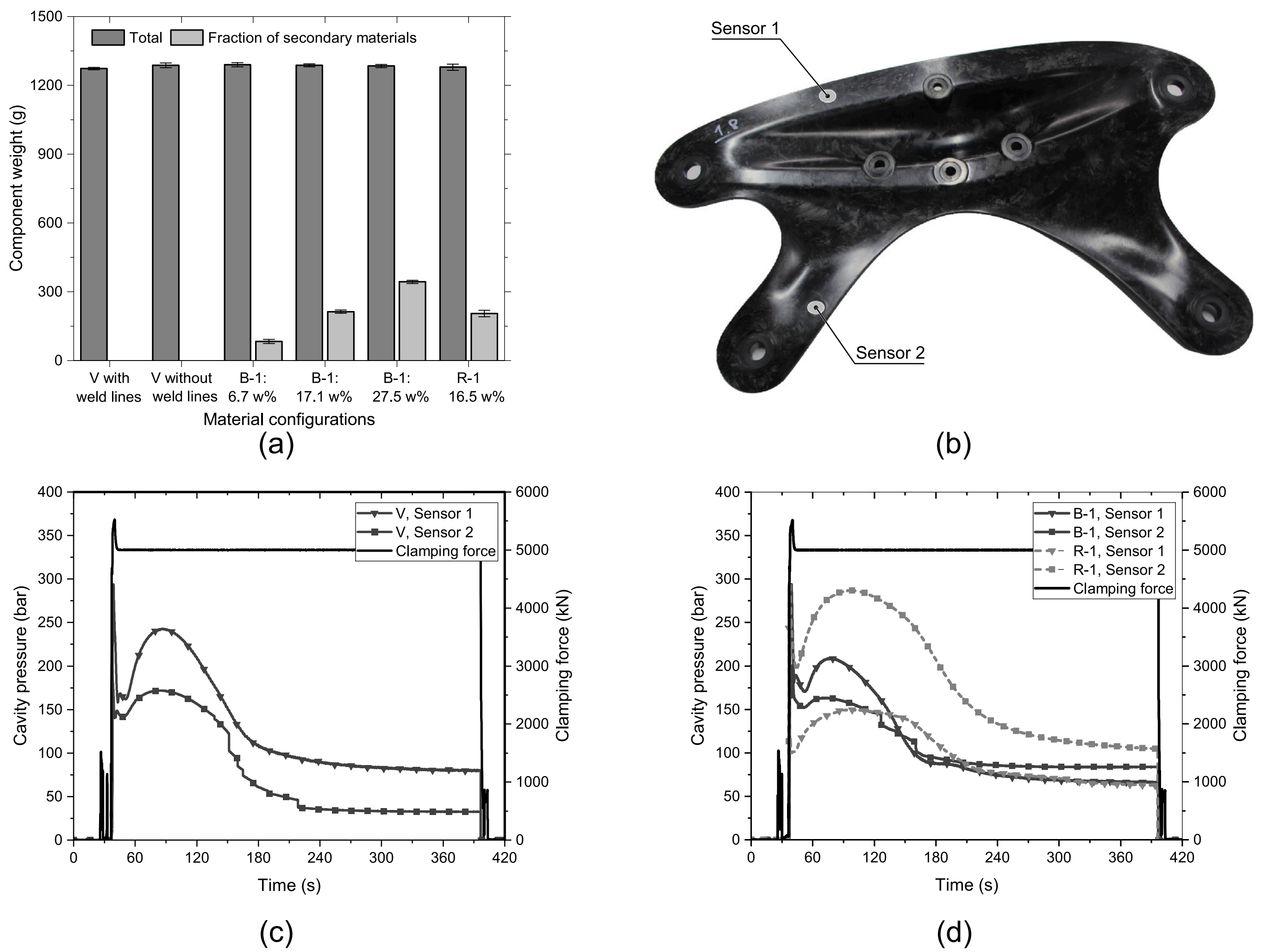


Fig. 4. (a) Component weights of the manufactured transmission crossmember prototypes; (b) pressure sensor locations; (c, d) representative clamping force and cavity pressure curves from exemplary prototypes for the different material configurations at a tool temperature of 135 °C.

since standardized material tests on the coupon level are not representative due to the typical fiber lengths in the range of 25 to 50 mm of CF-SMC materials [61–64].

Furthermore, weld lines caused higher scattering in terms of the component strength. The complex flow at the free surfaces and the stochastic initial meso-structure with its evolution during compression molding [65,66] induce local variations in the colliding flow fronts. Consequently, weld lines differ significantly from one part to another in their geometrical configuration, surface area, and defect distribution, which affects the crack growth resistance. The average maximum load observed for (V) with weld lines was 23.79 kN with a standard deviation of 2.22 kN. Higher maximum loads with less scattering were observed in the quasi-static component tests for (V) using the preform design without weld lines (Fig. 5b). The maximum load was 26.26 kN ± 0.22 kN. The macroscopic cracks initiated in this case not at the outer, but at the inner inserts and grew across the component transverse carrier section. At the inner insert, sharp radii were required by the limitations of the design space, thus resulting in macroscopic stress concentrations.

All the manufactured material configurations were compared in a benchmark study with the primary scenario (V) serving as a reference. The criteria of comparison were the linear component stiffness and the maximum value of the recorded force (Fig. 5a). The results were normalized to the data of the reference configuration (V) with weld lines (Fig. 6).

The benchmark study revealed that at least the same quasi-static component performance in terms of mean values could be achieved when using secondary materials made from prepreg byproduct (B-1) or rCF (R-1) in a multilayered hybrid material structure. The differences to the reference configuration with weld lines (V) were generally below 6%. Only (B-1) with 6.7 w% of bSMC had peak values with +15.6% for linear stiffness and +9.8% for the maximum load. Aligned fiber orientations in a layer (B-1) even yielded an improvement in linear component stiffness with a reduction of its standard deviation at least by half. The maximum load values were not considerably improved by these methods, since the component strength was primarily determined by the weld lines. High scattering of the maximum load values with standard deviations in a range from 9.3 to 21.4% was observed. As mentioned above, the scattering can be reduced drastically when using a preform design without weld lines (V). The component stiffness, though, was significantly reduced by changing the preform geometry.

4.3. Results of the LCA

The results of the ILCD midpoint method for the impact categories for the first group of the CF-SMC scenarios from vCF and

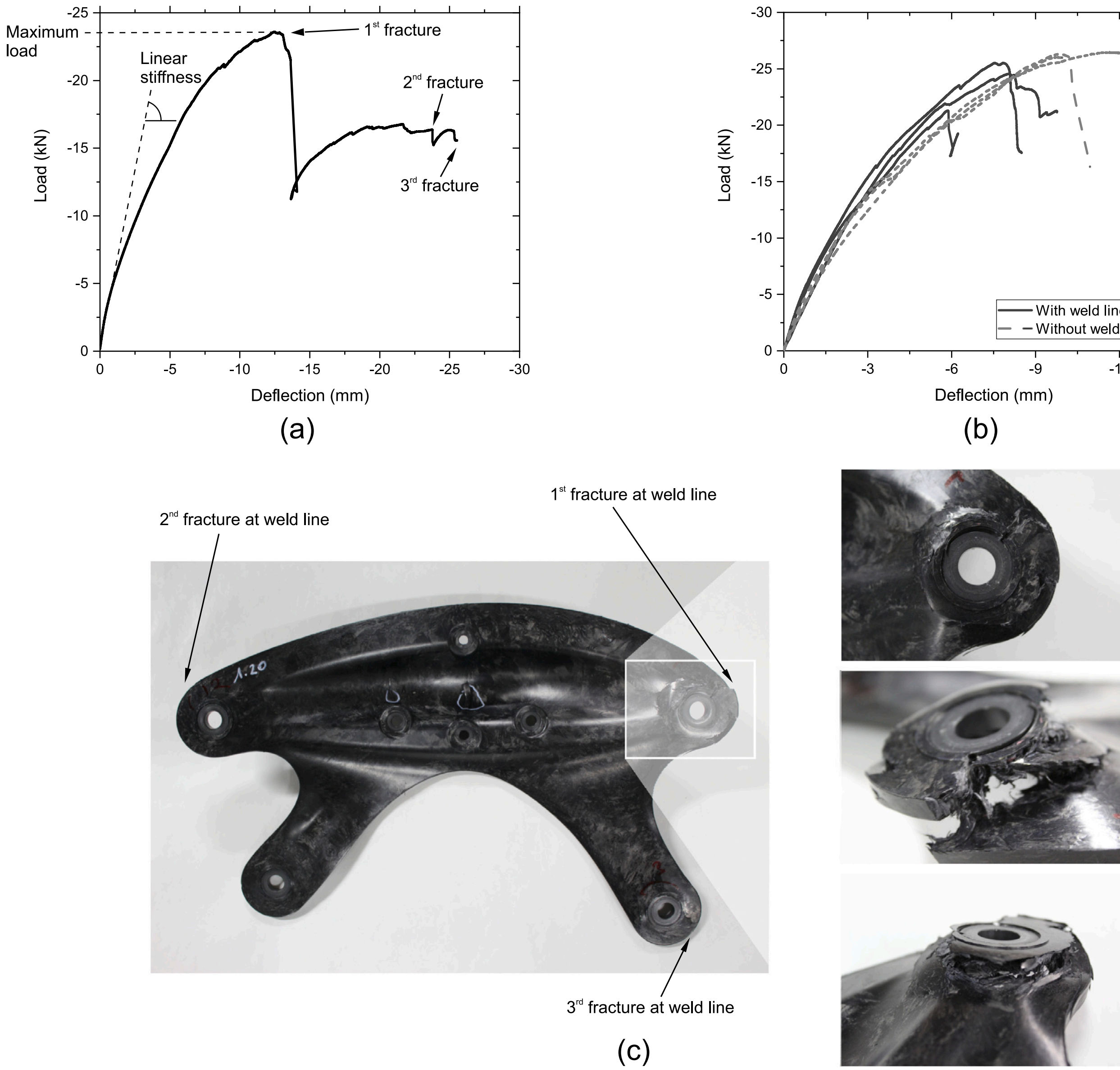


Fig. 5. (a) Load–deflection response for a representative quasi-static component test of an exemplary transmission crossmember with weld lines (V) and criteria of comparison for the benchmark study; (b) comparison of the quasi-static component tests with and without weld lines (V), tests were stopped after reaching the maximum load; (c) fracture patterns and failure locations for the representative quasi-static component test (V).

prepreg byproduct are shown relative to the aluminum scenario (A) (Fig. 7). The environmental impact categories are climate change, GWP 100a; freshwater and terrestrial acidification (ecosystem quality); freshwater ecotoxicity (ecosystem quality), freshwater eutrophication (ecosystem quality); marine eutrophication (ecosystem quality); terrestrial eutrophication (ecosystem quality); carcinogenic effects (human health); ionizing radiation (human health); non-carcinogenic effects (human health); ozone layer depletion (human health); photochemical ozone creation (human health); respiratory effects, inorganics (human health); land use (resources); and mineral, fossil and renewables (resources). All CF-SMC scenarios provide better results than the industrially used aluminum version (A). The impact category ionizing radiation (human health) is the only exception. In this impact category, the transmission crossmember consisting of aluminum performs better than the transmission crossmember made of primary vSMC (V). However, the other two CF-SMC scenarios (B-1 and B-2) have also less environmental impacts in this category than the aluminum version. The differences between the results of the CF-SMC scenarios arise from the assumption of the reuse of the residual material as a byproduct. Therefore, scenario (B-2), CF-SMC from prepreg byproduct provides the best results when looking at all impact categories (without considering the CF-SMC scenarios with rSMC).

The specific CF-SMC scenario with rSMC had less environmental impacts than the corresponding CF-SMC scenario from the first group with the same prepreg manufacturing assumption throughout all impact categories. This means that (R-1) has less environmental impacts than (V), but it does not perform better than (B-1) and (B-2) using prepreg byproducts. When the CF-SMC scenarios are ranked according to the ILCD 1.0.8 2016 midpoint method (cf. Table S2 given in Appendix A) starting with the highest environmental impacts, the following order results:

V > **R-1*** > **B-1** > **B-2** > **R-2***

Note that the specific pyrolysis data was not included in the LCA models of the CF-SMC scenarios with rSMC (marked by "*"). Potential

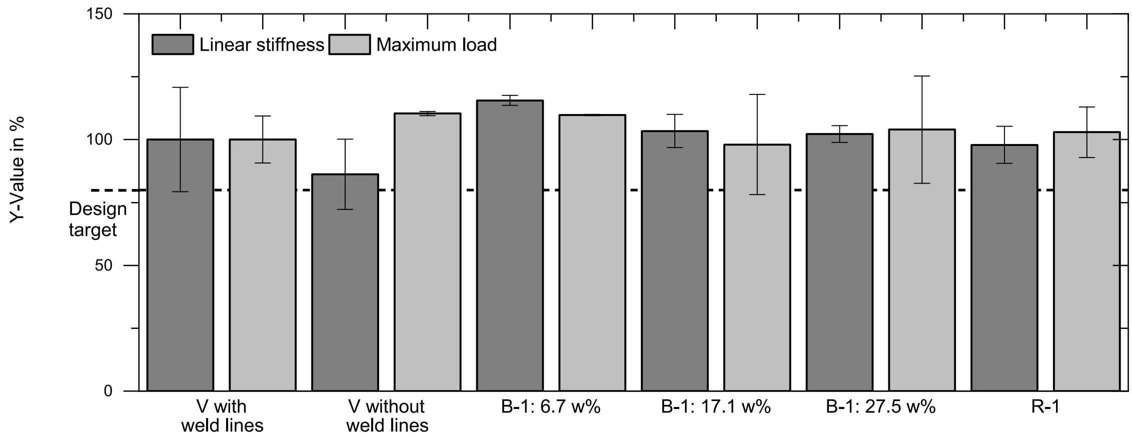


Fig. 6. Results of the benchmark study normalized to the reference configuration of the primary CF-SMC material (V).

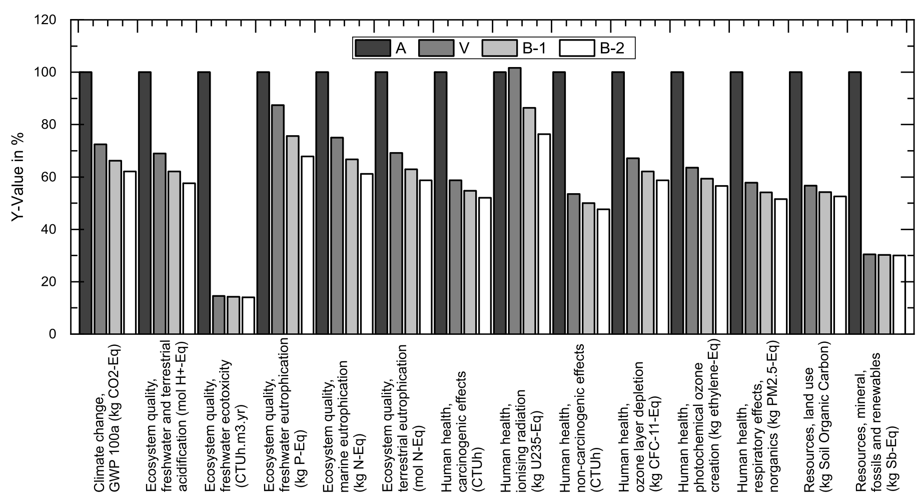


Fig. 7. Aluminum (A) versus CF-SMC scenarios (V, B-1 and B-2) with the ILCD 1.0.8 2016 midpoint method, for one transmission crossmember as a functional unit.

environmental impacts and/or environmental credits of the pyrolysis process in the CF recycling were therefore not considered. Hence, these scenarios tend to be ecologically better off. It was reported, though, that pyrolysis has lower energy demands and impacts and is therefore a better alternative to replace vCF [24,31].

Using the ReCiPe Endpoint (H, A) method it was additionally analyzed which life cycle phase leads to the most environmental impacts. The aluminum version (A) was compared to the CF-SMC scenario with the most "environmental points", CF-SMC (V) from primary prepreg (Fig. 8a and Fig. 8b). In both figures, the recycling credits are not included. As mentioned, it was not possible to directly include the transport data for the aluminum version. Thus, there are no "environmental endpoints" for the transports phase. Most of the environmental impacts are assigned to the consumer use phase. In this phase, the different results of the two scenarios are only defined by the different weights of the transmission crossmembers. Hence, the advantage in the consumer use phase that comes with a lighter weight of the CF-SMC transmission crossmember becomes obvious.

The results of the three damage categories ecosystem quality, human health and resources as well as the endpoint result, that aggregates the results of the three damage categories to one endpoint category, are shown in Fig. 8c. The analysis of the results of the three damage categories shows that the aluminum version (A) of the transmission crossmember had more "environmental points" than the CF-SMC scenarios (V), (B-1) and (B-2). The environmental differences between the three CF-SMC scenarios, represented by ReCiPe endpoints, result from the different assumptions concerning the prepreg manufacturing with the use of prepreg byproduct.

Since the tool did not allow more recycling material, the amount of rSMC (205.11 g) with a weight fraction of 16.5 w% was relatively small in the investigated CF-SMC scenarios (R-1). It is assumed that if it would be possible to increase the amount of recycling material technologically, the environmental impacts could further be reduced. Therefore, two additional LCA scenarios were modeled for the CF-SMC scenario (R-1) with weight fractions of rSMC of 30 w% and 40 w%.

The results with the three different recycling ratios for (R-1) were calculated at the midpoint (given in Appendix A, cf. Fig. S6, Fig. S7 and Table S3) as well as at the endpoint level (Fig. 8d) and were compared to scenario (V), where no rSMC was used. The results at the midpoint and endpoint level show the decrease of environmental impacts that comes with a higher proportion of rSMC. The reduction of environmental impacts is limited due to the fact that the fiber volume fraction of rSMC is relatively low and that the main impacts arise from the use phase. The share of the CF production in scenario (V) makes up about 52.16% of the raw material & manufacturing phase, but only 19.71% of the total product life cycle in terms of

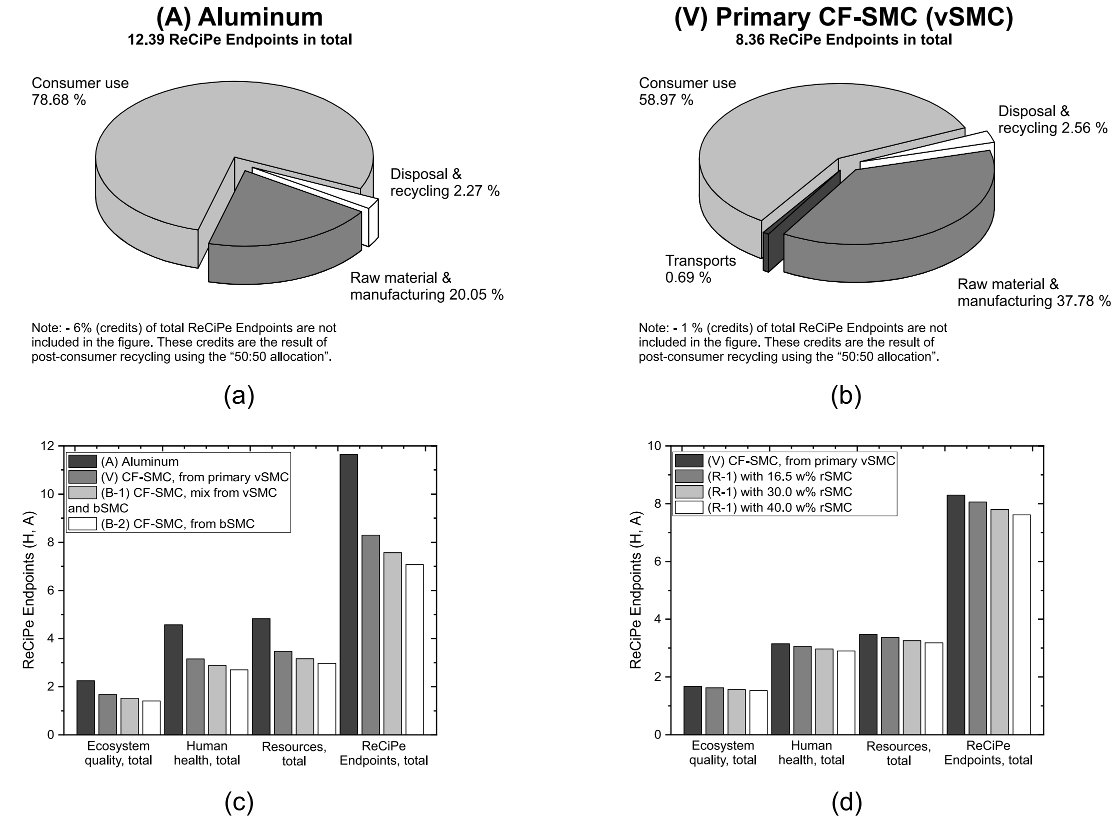


Fig. 8. (a) Aluminum (A) (2489.00 g) and (b) primary CF-SMC (V) (1273.09 g) in life cycle phases using the ReCiPe Endpoint (H, A) method, in % of the total ReCiPe Endpoints; (c) Aluminum versus CF-SMC scenarios with the ReCiPe Endpoint (H,A) method; (d) (V) versus (R-1) with 16.5 w%, 30.0 w% and 40.0 w% of rSMC with the ReCiPe Endpoint (H, A) method.

total ReCiPe endpoints. In contrast, the resin production amounts to 6.26% and 2.37%, respectively. However, as mentioned above, specific data for the pyrolysis recycling process were not included. It would be interesting for future research not only to get more detailed information concerning the recycling process, but also to find technical and/ or life cycle design options, where the cut-off allocation is applied for the specific recycling process.

5. Conclusions

The research findings demonstrated the potential of using secondary prepreg byproducts and end-of-life recyclates in the production of CF-SMC transmission crossmembers for middle-class cars. The holistic eco-design approach for material appropriate product engineering was comprised of prototype manufacturing, structural testing and LCA modeling. The focus was on commercially available materials and recycling scenarios for a feasible industrial production. Various material configurations with varying fractions of recyclates were compared in an experimental benchmark study with the primary CF-SMC material and the difference was shown to be low (below 6%) in terms of component stiffness and maximum load. The hybrid layup of virgin and recycled materials in an apparent sandwich structure did not only maintain manufacturability of the prototypes, but also the structural performance. In that way material waste can be reduced without sacrificing the capability of cost-efficient large-scale production.

The flow behavior of the impregnated non-woven mats of rCF (rSMC) was insufficient and showed virtually no material flow. Hence, it was placed as the core material to be overmolded by the primary CF-SMC (vSMC). Furthermore, the component stiffness was improved by the use of the secondary byproduct material (bSMC) due to the aligned fiber orientation. Such high stiffness/strength layers could be used to tailor anisotropy to improve product reliability despite the higher variability in the material properties of recyclates. Using the industrial prepreg scrap from a primary in a secondary production line simplified the recycling process and the re-manufactured bSMC showed good flow properties comparable to the primary material. This supports a cascadic use of industrial recyclates for varying quality requirements in the production of lightweight applications and might be suitable for series production due to time and cost savings.

According to the midpoint and endpoint method within the LCA model, the environmental impacts were reduced with increasing fraction of prepreg byproduct and rCF. All CF-SMC scenarios revealed lower environmental impacts in most categories than the industrial version in aluminum. The impact category ionizing radiation was the only exception, in which the aluminum version had fewer impacts than the primary CF-SMC (V) scenario. It has to be highlighted that the data used to estimate the environmental impacts of the aluminum scenario were generic due to the lack of specific LCA data. Note also, that the amount of rSMC in the prototype manufacturing was practically limited and the specific pyrolysis data was not available for LCA modeling.

In future work, it should be studied in detail if higher weight fractions can be processed and what environmental impacts and/ or credits arise from the pyrolysis process. This must be an ongoing process in the design efforts of lightweight applications, since current research activities are directed towards improving the recycling processes for CFRP materials. However, this work shows a potential path towards real closed-loop recycling in the life cycle of CFRP within a circular economy. The overall goal is to reuse the recycled carbon fiber materials in structural components to retain their high value. To achieve this, the ratio of the amount between higher performing (virgin or equivalent secondary material from uncured production waste) and lower performing (recyclates from end-of-life waste) materials can be adjusted to meet the quality requirements depending on the industrial application.

CRediT authorship contribution statement

Philipp S. Stelzer: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Project administration. **Umut Cakmak:** Conceptualization, Writing – original draft, Project administration, Funding acquisition. **Lisa Eisner:** Data curation, Formal analysis, Writing – original draft. **Leonhard K. Doppelbauer:** Data curation, Investigation, Formal analysis. **Imre Kállai:** Methodology, Investigation, Formal analysis. **Gernot Schweizer:** Data curation, Investigation, Resources, Writing – original draft. **Heinz K. Prammer:** Methodology, Writing – original draft, Supervision. **Zoltan Major:** Conceptualization, Methodology, Writing – original draft, Resources, Funding acquisition, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.compositesb.2022.109638.

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