



The economic and mechanical potential of closed loop material usage and recycling of fibre-reinforced composite materials

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ABSTRACT

This paper presents a novel recycle value model derived from the retained mechanical performance of retrieved fibres in fibre-reinforced composites. The proposed recycle value model was used to perform an economic analysis for establishing the future closed-loop material usage of fibre-reinforced composite materials. State-of-the-art recycling of carbon and glass-reinforced thermosets was adopted and resulted in a proposed recycling hierarchy in order to achieve a more sustainable environment and raw material cost reduction. The recycle value model showed that approximately 50% material cost reductions can be achieved at comparable mechanical performance by using recycled fibre instead of virgin fibre in appropriate applications. From the aspect of lightweight design this cost reduction provides the designer with new material choices, appropriate for lower cost and diverse stiffness designs. The proposed closed-loop hierarchy documents the importance of further improvement of fibrous material recycling, including sorting according to mechanical performance, in order to identify application areas previously not utilised and to maximise material sustainability and value throughout the material's lifetime.

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1. Introduction

Fibre-reinforced composite materials, such as carbon-reinforced epoxy, are used extensively for demanding high-stiffness applications in aeronautical and aerospace adaptations where their low structural weight potential leads to increased fuel efficiency, and thereby to reduced usage phase costs, and environment benefits (Kaufmann et al., 2011; Timmis et al., 2015; European Commission, 2014a). More recently, the potential to reduce these usage phase costs and the overall environmental impact has attracted also the automotive industry. For example, BMW uses resin-transfer-moulded (RTM) carbon-fibre reinforced plastic (CFRP) components in their i3-and i8-series (Jacob, 2013, 2012). Indeed, it has been shown that a 10% reduction of the structural weight of a vehicle can lead to improved fuel economy and a fuel consumption decrease of 6–8% (Fontaras and Samaras, 2009; Chu and Majumdar, 2012). However, CFRP is an expensive material system and previous work performed by the authors (Hagnell and Åkermo, 2016, 2015; M Karlsson (now M K Hagnell), 2013) has shown that material costs are considerable in composite production, representing between

20 and 60% of the total production cost depending on annual production volume. The recent development of lignin-based carbon fibres (Mannberg, 2017; Baker and Rials, 2013; Li et al., 2017) and bio-based fibres such as kenaf (Wu et al., 2017), flax and hemp (Pil et al., 2016) illustrates the interest, and need for, low-cost composite materials when moving beyond aeronautical applications.

One challenge caused by the use of traditional composite materials is their recycling. Recyclability is an issue of particular importance to the vehicle industry as it is faced with sustainability legislation such as the ELV Directive (2000/53/EC), where it is stated that by January 2015, a minimum of 95% of a new vehicle by average weight is to be recycled at its end-of-life (EoL). In addition, 85% of the waste involved is to be re-used (European Commission, 2014b, c). These are challenging targets, as the current recycling degree by average weight of a vehicle is about 75% (Cholake et al., 2017). Indeed, if the interest in low density fibre-reinforced composites continues to grow (Mathes, 2018) and translates into the wider introduction of composites into more adaptations, the issue of their EoL recyclability, as well as the actual volume of composite materials to be recycled, will become even greater (Shuaib and Mativenga, 2016; Pimenta and Pinho, 2011; Witik et al., 2013; Nilakantan and Nutt, 2015).

To date, most composites met their EoL either in landfills (Dayi et al., 2016; Ribeiro and de Oliveira Gomes, 2015; Rybicka et al.,

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Nomenclature

α	Composite efficiency factor
ν_f	Fibre volume fraction
ρ	Density
r_p	Percentage reclaimed recyclate
B	Base recyclate value
b	Lowest resale constant
BFRP	Bio (natural) fibre reinforced plastic
BMC	Bulk moulding compound
C-HM/IM/HS	Carbon fibre grades; high modulus/intermediate modulus/high-strength
CFRP	Carbon fibre reinforced plastic
CSM	Chopped strand mat
E	Young's modulus of composite along fibre direction
E-glass (EG)	E-grade glass fibre
EoL	End of life of component

GFRP	Glass fibre reinforced plastic
GFW	Glass fibre waste
LCA	Life-cycle analysis
LCC	Life-cycle costing
LCFRP/LC	Carbon fibre based on lignin precursor
m	Retained mechanical performance factor
P	Recycling processing cost
PAN	Polyacrylonitrile (carbon fibre precursor)
PEEK	Polyether ether ketone (thermoplastic polymer matrix)
Pitch	Petroleum pitch (carbon fibre precursor)
RTM	Resin transfer moulding
RV	Recyclate resale value
S-glass (SG)	S-grade glass fibre
SD	Semi-directional reinforcement
SMC/ASMC	Sheet moulding compound/advanced
UD	Uni-directional reinforcement

2015) or in incineration for energy recovery (Oliveux et al., 2015), strategies which fully neglect the mechanical potential of reclaimed fibres. This is not only unwise from the perspective of mechanical potential loss but also from that of value loss and in relation to the high raw material cost of the virgin materials (Hagnell and Åkermo, 2015, 2016). Recycled high-cost, high-stiffness aerospace-grade polyacrylonitrile (PAN) - and petroleum pitch (pitch) - based carbon-fibre for example, has high potential for reuse even for fairly demanding applications, as it is less costly to recycle than to produce in virgin form (Carberry, 2008). To that end, an EoL component ought, in itself, represent a certain material value; and not be treated as simple waste.

The idea of utilising waste as a resource forms the basis of the circular economy (Veleva et al., 2017), echoed in the European Commission "Roadmap to a resource efficient Europe" (European Commission, 2011), as well as in strategies such as zero waste (ZW) (Singh et al., 2017) and zero waste manufacturing (ZWM) (Singh et al., 2017; Veleva et al., 2017). Indeed, the act of recycling and re-using material waste instead of landfilling has shown multiple benefits with regards to reduced energy consumption and reduced green-house emissions through life-cycle-analyses (LCA) (Y.S.Song et al., 2009; Jank et al., 2017; Eckelman et al., 2014; C.J.O'Reilly et al., 2016; Li et al., 2016), improved mechanical performance in non-structural and semi-structural adaptations (Cholake et al., 2017; Novais et al., 2017; S.Cousins et al., 2019) as well as a good method to reduce the amount of land claimed as landfill (Ferreira et al., 2014).

Recently, there has been some focus on the development of fibre reinforcement retrieval through pyrolysis (ELG Carbon fibre Ltd, 2016; Holmes, 2018), fluidised bed techniques (F.Meng et al., 2017) and chemical treatments such as solvolysis (Dauguet et al., 2015). However, industrial pyrolysis often recycles composites in bulk, with little sorting of carbon fibre grades, resulting in non-continuous, randomised or filler-based reinforcement mats (ELG Carbon fibre Ltd, 2016; Holmes, 2018). Solvolysis and fluidised bed techniques, on the other hand, are promising with regards to decreased energy consumption and the potential to also recover some useful matrix, however these processes are still mainly available on laboratory scales (Dauguet et al., 2015; F.Meng et al., 2017). Nevertheless, the introduction and development of composite recycling methods illustrate and enable a shift in production and recycling of the material system, towards achieving a closed-cycle-material loop also for composite materials.

In an ideal, circular closed-cycle-material loop, recycled

reinforcement is coupled with appropriate structural design after each use cycle. Potential benefits of such a closed-loop would be many including improved recyclability, resource-efficiency, energy-conversion and, most likely, reduced raw fibre reinforcement costs. The act of retrieving fibres could therefore exert an overarching impact on the design envelope available, enabling the further use of composite materials in newer applications such as in the automotive industry where composites have been under-utilised due to the relationship between material cost and mechanical properties. As an added benefit, the structural reinforcing potential of recycled fibre reinforcement could mitigate some of the concerns raised by the vehicle industry regarding cost of recycling, essentially transforming recycling to resource harvesting.

LCA research (Witik et al., 2013; Jank et al., 2017; Y.S.Song et al., 2009; Dong et al., 2018; Li et al., 2016) have demonstrated that recycled traditional composite materials are efficient from the perspective of lifetime energy consumption, both in use and in recycling. Recent research on recycling of CFRP materials have become exhaustive, for example, (Dong et al., 2018) presented a detailed, combined life cycle cost (LCC) and LCA evaluation that compared the pure processing costs of different recycling strategies and discussed collisions between environmental targets and production cost considerations. However, the LCA-based research generally does not focus on the mechanical and structural potential of the recycled fibres. In contrast, a recent, combined LCC- and LCA-research publication (Meng et al., 2018), addressed some of the mechanical potential of fluidised-bed-recycled carbon fibres for automotive applications through an introduction of a design material index that compares the structural potential of alternative materials to that of mild steel. Complementary assessments of the mechanical and economic potential of recycled composite materials directly connected to an exemplified conceptual, future circular closed-cycle-material loop could however improve the level of knowledge and interest in recycled composite materials and their circular material flow. Moreover, these assessments could provide a basis for identifying potential applications for the recycled materials which, as indicated by (Pimenta and Pinho, 2012), is of key importance to enabling the re-introduction of recycled composite materials.

Although there is research on the actual industrial impact of a future circular closed-loop-material flow (Pimenta and Pinho, 2011; Palmer et al., 2009; Naqvi et al., 2018), proposed applications of the recycled materials are generally not structural and their potential impact on overarching design envelopes have not been discussed at

great length.

In this paper the authors have analysed and discussed the material cost reduction potential of introducing recycled fibre-reinforced composite materials into structural applications. As material recycling ideally is a iterative process where the act of recycling is repeated after each material use, the material cost reduction was researched on a cyclic basis where a proposed recycle value model estimated a conceptual material value for each recycling generation. The conceptual material value was expressed as a function of retained mechanical stiffness and recycle yield. Through the established material value for each cycle, it became possible to predict the material value sustainability and material cost impact as well as suggest appropriate number of recycling cycles and industry applications. Indeed, the generic form and predicted general trends of the proposed model meant the research become of interest to a wider audience beyond that of a composite material engineering. Ultimately, the proposed model and circular closed-cycle-material loop provided important insights for industry and researchers regarding the coupling between mechanical potential, raw material cost and recycled composite materials.

2. Outline

In this paper, a recycle value model based on state-of-the-art composite material recycling strategies has been proposed and applied towards evaluating the industry impact of a future circular material recycling flow of fibre-reinforced composite materials. This paper include

- The research scope, limitations, material systems as well as defined material flow.
- Literature study on state-of-the-art recycling of fibre-reinforced composites with justifications regarding choice of appropriate recycling method depending on respective fibre system.
- Definition of proposed recycle value model and data points of respective fibre system.
- Results on estimated recycle value depending on recycling generation, overarching raw material cost reductions for estimated full circular material flow cycle and estimated impact on lightweight design.

Finally, the presented research includes discussions and conclusions on key findings.

3. Scope and limitations of economic analysis and proposed recycle value model

This research study proposed a recycle value model on fibre-reinforced composites that has been implemented towards evaluating material value sustainability and the impact of a future circular, closed-loop, material flow. A number of strategic limitations and assumptions have been applied that dictates the applicability and validity of the proposed recycle value model. Given the intention to review a future cyclic material recycling flow, proposed recycle value model was implemented using fibre system data drawn from industry and published literature. The research was carried out at an overarching level to provide a wider basis for further discussion with implications for lightweight design and potential future composite applications. The reinforcing fibre materials researched ranged from structural to semi-structural applications and were traditional PAN- and pitch-based carbon as well as lignin-based carbon and glass. The connective matrix of the fibre-reinforced composite was epoxy. This resin was chosen not only due to its mechanical properties and frequent industrial use

but also as it represents the worst-case recycling scenario as a thermoset is not re-mouldable. With regards to the state-of-the-art recycling strategies studied, only economically viable methods for each fibre-reinforcement type were considered in order to limit the scope of the paper. Although other, more recent, recycling strategies may prove more economically efficient than those currently studied, methods with sufficient technical readiness level were preferred in order to be able to use industry-relevant cost data. Finally, it has been assumed that the use of composite materials will grow, resulting in sufficient volumes of EoL-components to properly feed proposed circular closed-cycle-material loops.

4. The material systems researched and their reinforcement potential

The stiffness effect of a composite is directional and governed by fibre system used, volume fraction and type of reinforcement. An approximative Young's modulus along the fibre direction can be calculated using the modified, rule of mixtures (Zenkert and Battley, 2011) according to

$$E = \sum_i \alpha v_f E_f \quad (1)$$

where α is a composite efficiency factor related to possible packing of fibres in respective reinforcement type, v_f is the fibre volume fraction and E_f is the Young's modulus of the fibre reinforcement. Mechanical properties and representative costs of fibre materials and grades researched are given in Table 1. The fibre orientations with associated approximative volume fractions and reinforcement efficiency factors are given in Table 2.

5. Composite wastes and potential overarching material flow

In addition to EoL component composite waste, there is also production waste, or scrap, generated during component manufacture (Nilakantan and Nutt, 2015). The recycling of both of these types of waste would generate reclaimed material that could supply a new production with raw material, as illustrated in Fig. 1.

Waste in the form of dry reinforcement scrap, prepreg cuts and resin of virgin-material quality can, given an appropriate new component size and an efficient patching method, be directly returned to production. Waste such as out-of-date prepreg rolls, on the other hand, must first be treated, reclaiming the fibre reinforcement, before they can be reused in new production. As such, this type of waste follows the same recycling process as EoL components. Because focus of the authors has been to analyse the mechanical and economic impact of recycling and not the retrieval and patching of virgin materials, virgin-material quality production waste has not been researched. However, the value of virgin-material quality production waste could be assumed to be higher than that of recycled production waste material as it need not be

Table 1

Mechanical properties and cost data of considered fibre reinforcements (Carberry, 2008; Baker and Rials, 2013; Cripps, 2018; Mannberg, 2017; Hexcel, 2019; Mårtensson, 2016; Li et al., 2017).

Fibre type	E_f [GPa]	kg cost [€/kg]	ρ [kg/m ³]
High-modulus carbon (CHM)	400	60	2000
Intermediate-modulus carbon (CIM)	300	30	2000
High-strength carbon (CHS)	250	20	2000
Ideal lignin carbon (iLC)	172	4	2300
Lignin carbon (LC)	120	4	2300
S-glass (GS)	89	20	2700
E-glass (GE)	80	2.5	2700

Table 2
Reinforcement type data (Zenkert and Battley, 2011).

Reinforcement type	Volume fraction v_f [-]	Efficiency factor α [-]
Chopped strand mat (CSM) or non woven	0.3	0.2
Advanced SMC (ASMC)	0.55	0.375
Medium semi-directional (MSD)	0.6	0.6
Semi-directional (SD)	0.6	0.7
Uni-directional (UD)	0.6	1.0

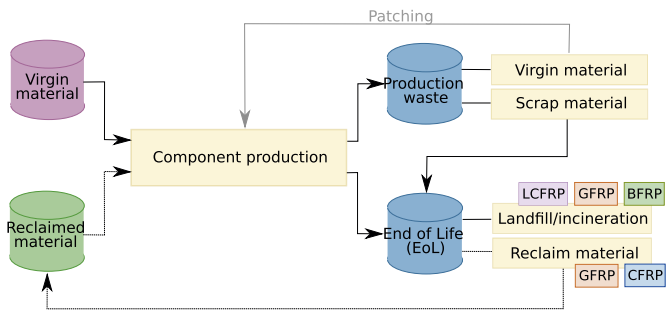


Fig. 1. Waste produced during production, production waste, and in the form of EoL has the potential to feed a new component production with reclaimed material.

recycled.

6. Recycling of thermoset composite fibre-reinforced materials

6.1. Traditional landfill and incineration

Most of today's recycled composite materials are placed either in landfill or incinerated (Rybicka et al., 2015). Although, the latter option generates energy and therefore is a recycling method of sorts, the Waste Framework Directive (European Commission, 2008) does not classify incineration as recycling. Instead they define the term as “Recycling means any recovery operation by which waste materials are reprocessed into products, materials or substances whether for the original or other purposes. It includes the reprocessing of organic material but does not include energy

recovery and the reprocessing into materials that are to be used as fuels or for backfilling operations”. In the scope of this paper, costs for landfill and incineration were used for comparative purposes, see Fig. 2.

6.2. State-of-the-art recycling methods of thermoset composite fibre reinforced materials

Recycling methods for thermoset composites are numerous and can generally be categorised as either mechanical, thermal or chemical recycling (Pickering, 2006). As the object of this paper is to couple fibre reclamation with structural adaptations, recycling processes were categorised in relation to possible material reclamation after recycling; as either of low level or high level material reclamation, see Fig. 2. Note that although recycling through solvolysis (Dauguet et al., 2015) and fluidised bed (F.Meng et al., 2017) are promising both in relation to cost and energy required, they are not covered within the scope of this paper as they are currently not available on an industrial scale. Only recycling methods of particular interest for this research, as defined in Fig. 2, are further described in section 6.2.1 - 6.2.3. Please refer to literature for more details on other recycling methods (Pickering, 2006; Pimenta and Pinho, 2011; Oliveux et al., 2015).

6.2.1. Pyrolysis of composite fibre-reinforced materials

The fibre reinforcement is freed as the composite matrix is converted to gases, tar and char through pyrolysis, a thermochemical conversion process occurring in organic materials at elevated temperatures (450–700 C°) (Samuelsson et al., 2017). The lower temperature in the range is sufficient when converting polyester resins while the higher temperature (at least 500 C°) is required when converting epoxies or high-temperature thermoplastics such as PEEK. Consequently, in order to reclaim the fibre reinforcement, the temperature must be high enough to convert enough resin, but low enough to not significantly affect the mechanical properties of the reinforcement (Pickering, 2006; Pimenta and Pinho, 2011; Oliveux et al., 2015). Pyrolysis is today performed on a commercial level (ELG Carbon fibre Ltd, 2016). In general, different quality and property levels of each fibre type are reclaimed in bulk within the same process, returning fibres of different properties in the same batch (Oliveux et al., 2015). For this reason, sorted pyrolysis and laboratory-scale pyrolysis report reclaimed fibres of higher quality than that available in industrial

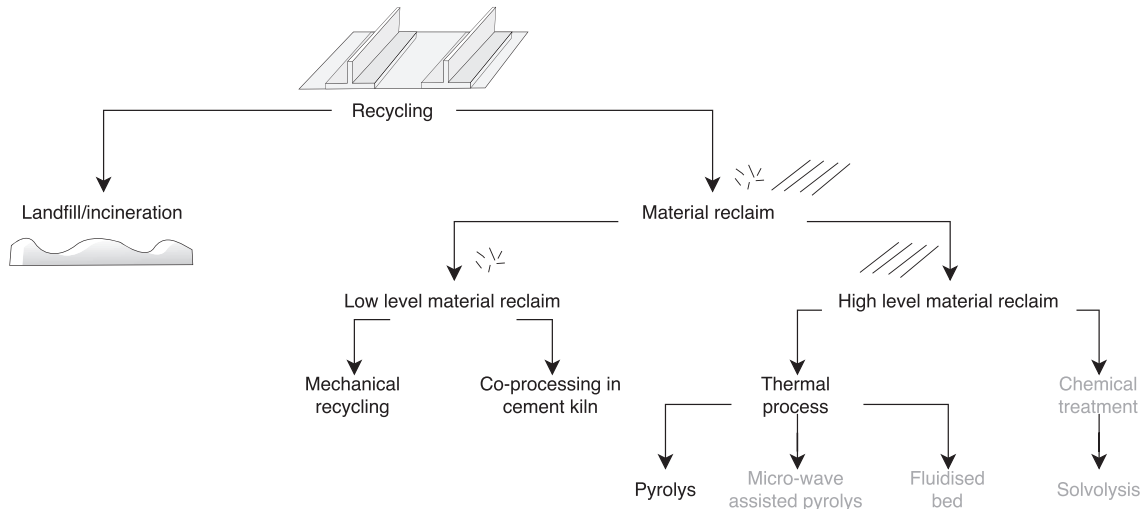


Fig. 2. Recycling strategies sorted on level of materials reclaimed with methods of choice printed in bold print.

production.

6.2.2. Mechanical recycling of composite fibre-reinforced materials

In mechanical recycling, the structure to be recycled is reduced in size in order to be repurposed as, for example, filler material. This process generally uses several size-reduction steps. First the structure is dismantled and crushed into sizes of about 50–100 mm, which enables the removal of foreign objects such as metallic inserts. Then the pieces are further reduced using high-speed milling to final size, which in its smallest particle form reach sizes of less than 50 μm (Pickering, 2006). Note that the recyclate retrieved is comprised of both fibre and thermoset material. Consequently, particles of the smaller size retrieved generally contain more polymeric material while the coarser powders contain more fibrous material (Pickering, 2006; Oliveux et al., 2015; Pimenta and Pinho, 2011).

6.2.3. Co-processing of composite fibre-reinforced materials in cement kilns

In co-processing, the composite waste material is recycled in cement kilns. In this process, the minerals of the composite contribute to the raw material of the cement and the resin contributes to the cement production as combustion fuel, which reduces the amount of fossil fuel required by as much as 16% (EuCIA, 2011; Pickering, 2006; Oliveux et al., 2015). There is much discussion underway about whether or not co-processing is a recycling method, or if it too close to incineration. However, the latest decision within the European Union is that as 2/3 of the material is recycled as cement raw material, the majority if the waste is reused in another form, making this a true recycling method (EuCIA, 2011).

7. Economically-feasible recycling methods

An appropriate selection of recycling strategy depends on the fibre reinforcement and matrix of a composite structure. Here, economically-advantageous recycling strategies for each fibre reinforcement system researched are described and justified.

7.1. PAN/pitch-based carbon fibre-reinforced composites

Carbon-fibre reinforcement made from PAN or Pitch precursors are produced in different grades, resulting in different mechanical properties and production costs. In general, both precursor carbon fibre types are high-cost, ranging from 15 to 60 €/per kg as presented in Table 1, with the lower cost representing lower stiffness fibres designed for automotive and general industrial applications. As a result of the high level of mechanical properties and high cost of PAN/Pitch carbon fibres, higher cost fibre reclaiming methods such as pyrolysis can be justified. The cost of carbon fibres reclaimed through pyrolysis is reported to range from 8 to 12 €/per kg for fibres (ELG Carbon fibre Ltd, 2016) and 17–29 €/per kg for fabrics (Carberry, 2008; ELG Carbon fibre Ltd, 2016), which are effectively less than that of the cost of virgin fibres. Some researchers even estimate cost per kg of pyrolysis as low as 1 €/per kg (Dong et al., 2018). Furthermore, the mechanical properties of carbon fibres are generally fairly well contained after pyrolysis-temperatures of up to 550°C. The stiffness can be reclaimed at about 80–100% while the fibre strength is affected more (Pimenta and Pinho, 2012). Indeed, some authors report nearly no effects of pyrolysis on stiffness and strength (Longana et al., 2016).

About 10% of currently-produced CFRP waste corresponds to production waste (Pimenta and Pinho, 2012). The rest is EoL components. The carbon fibres in production waste have the potential to be reclaimed in the same form as its previous architecture (Pimenta and Pinho, 2012; Meredith et al., 2012), making uni-

directional-quality components possible. However, pyrolysis recycling of EoL waste generally first implies cutting up of the parts into pyrolysis-sized lengths (a few cm (Samuelsson et al., 2017)) to achieve full resin removal. These retrieved fibres are therefore chopped, returning CSM- or at the most ASMC-quality if sufficient fibre alignment is possible. Moreover, the fibre length inevitably decreases with each recycling generation. As the fibre lengths become too short for efficient alignment, the fibrous fractions can be used in SMCs or as fillers in thermoplastic resins (Oliveux et al., 2015). When used in thermoplastic resins the carbon fibres have travelled full circle, being present in a component that can simply be reheated and remoulded to appropriate new parts. This generation flow is illustrated in Fig. 3.

7.2. Lignin-based carbon fibre-reinforced composites

Carbon-fibre reinforcements made from lignin precursors are bio-based and as such their incineration or landfilling does not emit any further CO₂-emissions. However, if the fibre system is used in conjunction with an epoxy matrix, pyrolysis might still be required to recycle or reduce the emissions of the resin. In either scenario, the recycling is unlikely to produce much of reclaimed material.

7.3. Glass fibre-reinforced composites

Glass-fibre reinforcements are generally fairly inexpensive and perform at low-to medium stiffness levels. Due to the low cost of the fibres, the cost of material reclamation must be low if the recycling is to be economically viable. Glass fibres lose as much as 50% of its mechanical properties if heated in a pyrolysis process beyond 400 °C (Oliveux et al., 2015). Such a decrease in mechanical properties, coupled with the low cost of virgin glass fibre, makes high-level material reclamation of GFRP not economically viable. Mechanical recycling to filler is possible, but is not cost-efficient when considering the low cost of other, comparable, fillers such as calcium carbonate or silica (Oliveux et al., 2015; Pickering, 2006). Instead, recycling in cement kilns, where the glass fibre composite is reclaimed as cement base material and energy supply, has been promoted (EuCIA, 2011; Oliveux et al., 2015; Pickering, 2006). More recently however, the mechanical recycling and, more importantly, sorting of glass fibre composites into fibre and resin-rich bundles

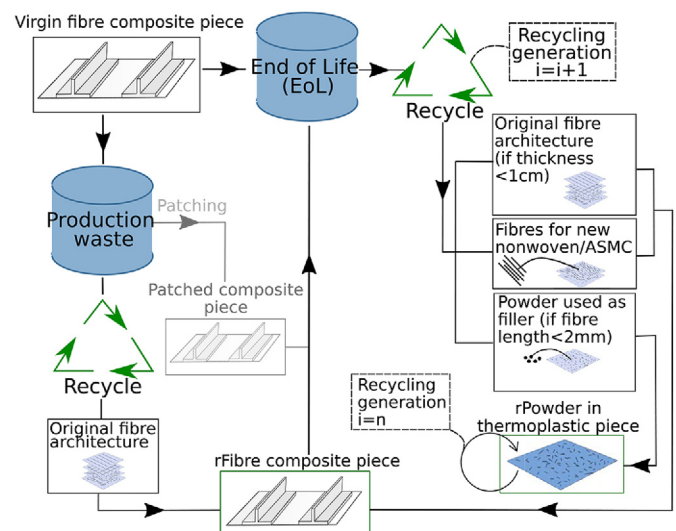


Fig. 3. The recycling flow of a composite part as a function of fibre length, which in turn is dictated by recycling method and generation i ($i_{end} = n$).

for use in bulk moulding compounds (BMC) has been shown to be mechanically viable (Palmer et al., 2009). As the recycling strategy of mechanical recycling and sorting of fibre and resin-rich bundles presents higher material reclamation levels than co-processing in a cement-kiln, the potential of this strategy was further researched by the authors in this paper.

8. The proposed recycle value model: establishing the economic material value at each recycling loop

The proposed recycle value model has been formulated on an iterative form for each recycling cycle (i). It was suggested that the economic value of the recycled material (RV_i) is a function of the material value in its previous recycling step (RV_{i-1}), the retained mechanical performance (m), percentage of reclaimed recycle yield (r_p), recycling processing cost (P) and a final base recycle value (B) according to the following assumptions and expressions.

The mechanical performance factor ($0 \leq m < 1$) was introduced to account for recycling-induced heat or chemical fibre degradation effects. Apart from the process-induced fibre degradation, the recycling generally reduces possible fibre length. Depending on the size of the component where reclaimed fibres are reintroduced however, the fibre length need not always limit the mechanical performance. For example, if reclaimed fibres are reintroduced in a new component where the average required fibre length is below that of the reclaimed fibres, optimal fibre length and reinforcing effect could still be achieved. Therefore, the mechanical performance as a function of fibre length reduction was not included in the factor m . Instead, the mechanical performance as a function of fibre length was calculated for representative fibre orientations and volume fractions in Tables 1 and 2 using Eq. (1) together with a degraded mechanical stiffness that involved the proposed mechanical performance factor (mE). Ultimately, by removing the effect of length-reduction from the proposed recycle value model, a full range of reclaimed fibre types, from short fibre to full fibre architecture and UD-reinforcement, has been reviewed on a conceptual basis.

The reclaimed recycle yield ($0 \leq r_p \leq 1$) was introduced to account for potential material loss when recycling.

The recycling processing cost, P , was for sake of simplicity considered to be a fixed value, depending on recycling method and material type given by published data from suppliers. Although it is a simplification to neglect to model recycling costs as a function of facility size, the method was deemed sufficient for posed conceptual research scope. Note that in an early recycling cycle when reclaiming full fibre architectures a high mechanical performance is followed by higher recycling processing costs, as indicated in Table 3, as larger EoL sections dictates a more tedious and complex recycling process.

Based on the assumptions above, the proposed recycle value was estimated according to

$$RV_i = f(RV_{i-1}) = mr_p RV_{i-1} - P \quad (2)$$

$RV_i \geq B$

where RV_0 = Virgin material kg cost

As indicated, in the first recycling step when virgin fibre EoL or production waste is recycled, the initial value (RV_{i-1}) is the same as the cost of the virgin material. Given that the recycling process performed degrades reclaimed fibres for each recycling cycle ($m < 1$) and loose material through recycle loss ($r_p < 1$), eventually, the combined impact of the mechanical performance factor, recycle yield and the recycling processing cost makes continued high-level recycling unsustainable. At that point, a low-level EoL recycling stage was considered to have been reached where returned material was of low mechanical value fit for non-structural applications. There, the recycle value approached the base recycle value (B), which represented the potentially lowest resale value of reclaimed fibre material, or the value of a final fibre form at the end of the circular closed-loop cycle. Normally, this would be a powder or particle form and was therefore defined as

$$B = br_p - P_{end} \quad (3)$$

where b is a representative lowest resale constant that varies on material type and P_{end} is the final stage recycling cost. In fact, as one possible recycling route in any recycling generation would be to retrieve material in its final, powder or particle, fibre form, a recycled material could be considered to always own its inherent base recycle value. Such an assumption would predict a consistently higher recycle value, as the recycle value would correspond to the sum of that predicted by Eq. (2) and its final base recycle value B . Therefore, the formulated recycle value predicted by Eq. (2) presents a conservative estimate.

9. Implementation of the proposed recycle value model

Data points for fibre systems studied are presented in Table 3. The justifications and reasoning behind the individual data points are described in detail for each fibre material system respectively.

9.1. PAN/pitch-based carbon fibre data

The PAN/pitch carbon fibres are fully freed from the epoxy resin through pyrolysis at a recycling cost in the range of 10–23 €/kg depending on reinforcement type (ELG Carbon fibre Ltd, 2016), where the higher cost is required for fabrics and full fibre architectures while the lower cost is sufficient for lower level reclamation. Although pyrolysis has been reported to retrieve the fibres at little mechanical degradation (ELG Carbon fibre Ltd, 2016; Pimenta and Pinho, 2012; Meredith et al., 2012), as a conservative measure, it is assumed that the thermal process induces a 5–10% drop in overall mechanical properties (Pimenta and Pinho, 2012). It is assumed that the recycling process is designed to retain all recycled fibres in order to maximise recycling value. The value of the final material form, b , is set to 5 €/per kg, half that of reported value of recycled carbon fibre (ELG Carbon fibre Ltd, 2016), as the final material form is achieved through mechanical recycling. Given that some retailers offer milled carbon fibres at 20–25 €/per kg (easycomposites, 2017; Haufler Composites, 2019), a higher final material form could potentially have been assumed. However, retailer-offered carbon fibres were milled from virgin, or high-level

Table 3
Recycling value data points. Omitted values (–) are a result of assumed reclaimed recycle yield of that specific fibre system.

Fibre type	Retained mechanical performance m [-]	Reclaimed recycle yield r_p [-]	Recycling cost P [€/kg]	Final fibre resale constant b [€/kg]	Final stage recycling cost P_{end} [€/kg]
PAN/Pitch based carbon	0.8–0.95	1.0	10–23	5.0	0.3
Lignin based carbon	–	0	–	–	–
Glass	1.0	0.72	0.3	0.5	0.3

pyrolysis-treated carbon fibres while the final material form considered herein was more conservatively assumed to have been achieved through mechanical recycling with no prior or secondary pyrolysis.

9.2. Lignin-based carbon fibre data

Lignin-based carbon fibres are conservatively assumed to be incinerated through pyrolysis, returning no material of value ($r_p = 0$). Even if this assumption may seem to be severe it is worth noting that even if 100% of the fibres were retrievable at fully-retained mechanical performance, the low cost of the initial raw material significantly limits the resale value, Eq. (2), making their contribution negligible.

9.3. Glass fibre data

Glass fibre composites are mechanically recycled through grinding to proper size and are then sorted by fibre size using cascade air classification (Palmer et al., 2009). Simple grinding and milling has been reported to cost approximately 0.3€/per kg (Shi and Zheng, 2007). Given that the cascade air classifiers are generally used in large-scale industry such as agriculture and mining, it is safe to assume that its application in a recycling plant ought to be performed in a cost-sensitive manner. Therefore, it is assumed that the recycling cost on an industrial level should be dominated by that of the mechanical recycling and fall close to that of 0.3 €/per kg. It is assumed that the effect of mechanical recycling on the mechanical properties of glass fibres is limited to that of fibre length reduction, therefore, the retained mechanical performance factor equals 1. As the fibres are milled there will be recycle that can be reprocessed or scrapped, (Palmer et al., 2009) with a recovery level of 72% useful recycle in a first recycling generation. The value of the final material form b is assumed to be 0.5€/per kg, half that of milled virgin glass fibre (EC Fibreglass, 2018). Potentially, an even lower final material form value could have been assumed as research (Palmer et al., 2009) has demonstrated that only about 10% of the reinforcing fibres of a BMC can be exchanged with that recycled fibres in order to retain its mechanical properties.

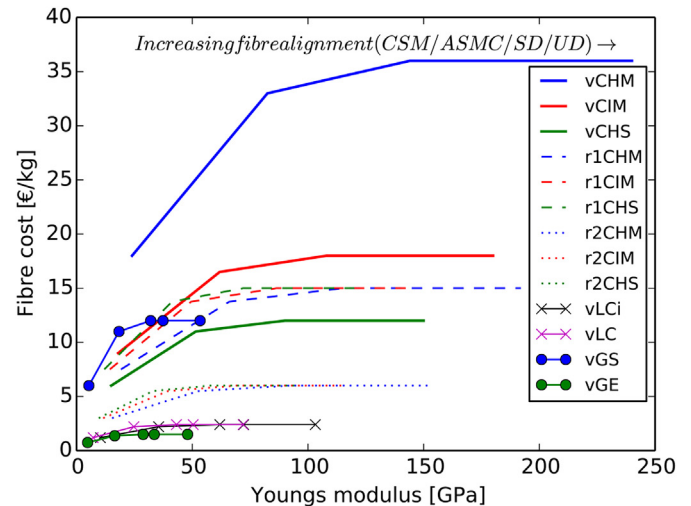
10. The resulting closed-cycle-material loop

Material reclamation potential and initial raw material value dictates the resulting closed-cycle-material loop hierarchy of each fibre system researched.

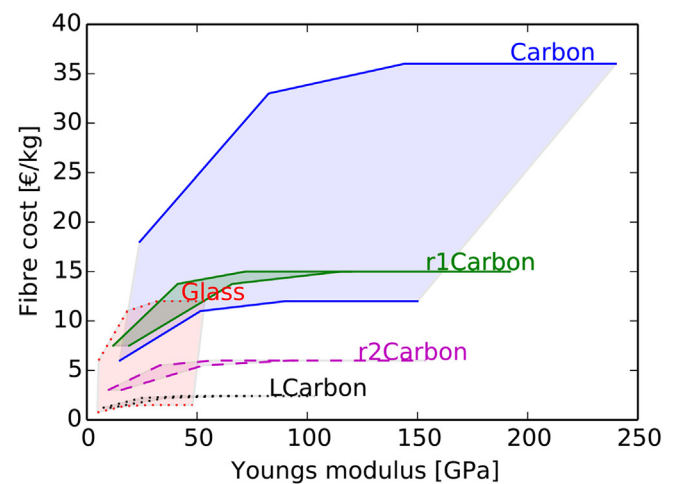
10.1. Raw material costs

Raw material costs, or raw material values, of a composite depends on volume fraction, length and fibre alignment. Given data supplied in Tables 1 and 2, Fig. 4a illustrates the relationship between fibre cost per kg for increasing Young's modulus with increasing fibre alignment as governed by Eq. (1). Note that the fibre cost for the recycled carbon fibre material is the resale value given by Eq. (2). The cost and mechanical performance potential of recycled carbon, first generation (r1) and second generation (r2) are shown using the conservative assumption that the recycled fibres experience a 20% drop in Young's modulus. It is important to note that the highest modulus achievable for the recycled carbon corresponds to recycled uni-directional reinforcement retrieved from production waste. For such cases,

In order to address the outer application boundaries of each fibre system, Fig. 4a is simplified into the presentation of the limiting costs, shown in Fig. 4b. This type of boundary-based graph does not present all data points but is useful for discussing the



(a) Full detail level.



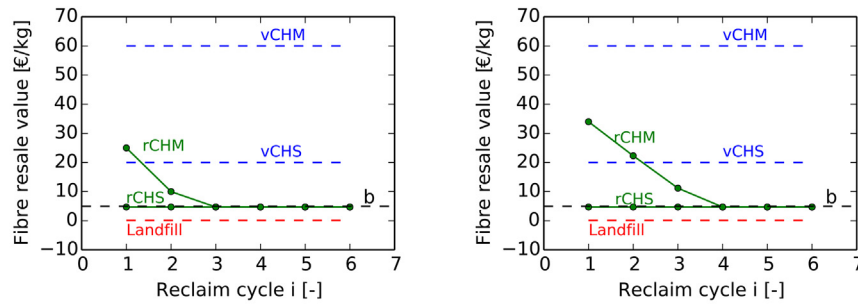
(b) Generic level.

Fig. 4. Fibre reinforcement kg cost with increasing reinforcement performance according to Eq. (1) given data according to Tables 1 and 2. Fibre reinforcement costs for the recycled materials (r1C in figure) were predicted using Eq. (2) assuming a retained mechanical performance of 80% for recycled carbon fibres.

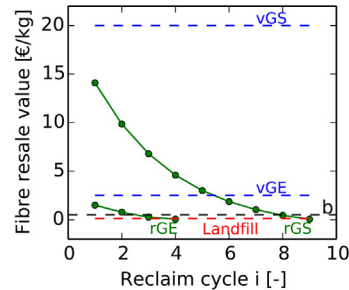
overall limits and potential of each fibre material on a generic level.

10.2. Predicted value potential of each recycling generation for researched composite materials and recycling methods

Based upon Eq. (2), the value potential of each recycling generation is given in Fig. 5a and b for carbon fibres and 5c for glass fibres. For reference purposes, the cost of landfilling (Shi and Zheng, 2007) is given in each figure. Virgin and recycle value of higher and lower grade fibres are presented to show the value span. It was proved that it is economically sound to recycle rather than landfill both fibre systems. High-modulus carbon and high-cost S-glass fibres have high potential resale values, while the lower-bound high-strength carbon and E-glass fibres have lower resale values. High-



(a) Carbon fibre at 80% retained mechanical property. (b) Carbon fibre at 95% retained mechanical property.



(c) Glass fibre.

Fig. 5. Resale value of fibre systems researched for each recycling generation as predicted by Eqs. (2) and (3).

level recycling preserves the value of the high-cost material fibres, carbon and glass, for several generations.

The high-modulus carbon fibre should be recycled in fibrous form and reintroduced as fibre-reinforcement 2–3 times. After three cycles its value approached the lowest resale value b . A maximum of three cycles can also be considered reasonable from the viewpoint of pyrolysis recycling, in which the retrievable fibre length decreases for each cycle. Indeed, (Longana et al., 2016) reports that if starting from a chopped-fibre-reinforced part a maximum of two cycles are possible before the fibres become too short to ensure stable mechanical stiffness as dictated by volume fraction. For high-strength carbon fibres however, only one recycling generation is economically viable, and this material can be recycled into powder form and reintroduced as filler material in the first recycling generation.

For S-glass, 6–7 recycling cycles were predicted before reaching the lowest resale value b . However, it is important to note that this number of cycles is dependent on the initial cost of the fibre. The initial cost of S-glass is generally tied to mechanical parameters other than simply stiffness as the difference between E-glass and S-glass in stiffness is low, and this means that retaining these other parameters throughout each cycle must be recorded and reflected in the mechanical property efficiency factor (m) in Eq. (2). Altering the value of this efficiency factor radically changes the predicted number of recycling cycles. For example, if it is assumed that GFRP is recycled with a mechanical efficiency of 0.8 instead, 4 recycling cycles are predicted while a mechanical efficiency of 0.5 immediately predicted a recyclate value equal to the lowest resale constant.

10.3. Introduction of recyclate value potential into raw material cost

Subtracting predicted recyclate value from the raw material cost of a specific material gave the new, adjusted, raw material cost ranges in Fig. 6. This subtraction effectively shifts the cost of all fibre

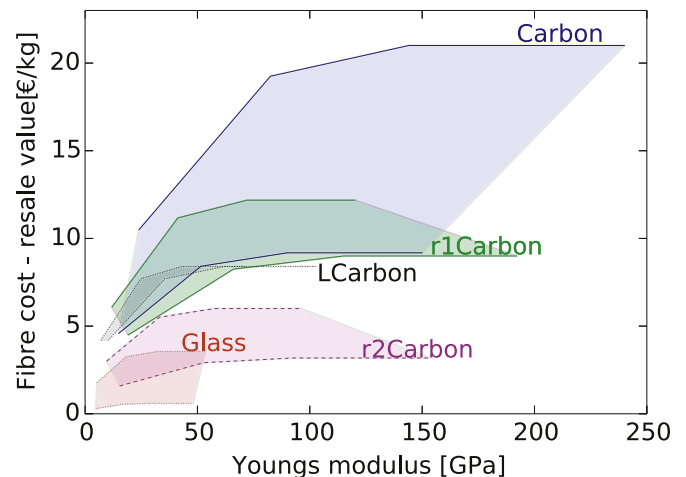


Fig. 6. Fibre cost ranges shift, or causes reductions in cost, when the recyclate value is subtracted from representative raw material cost ranges.

systems. The highest material cost effect is present in the scenario of reclaiming and reintroducing uni-directional fibres with ultimate directionality. This could be done if reclaiming fibres from out-dated prepreg rolls or, potentially, in a scenario where the component to be reclaimed is thin and need not be significantly chopped to achieve full pyrolysis which enables the retrieval of fibres of significant length. Given such a scenario, virgin carbon fibre material cost decreases by 43–58% for $0.8 \leq m \leq 0.95$, or a mean of 50%. Lignin-based carbon fibre material costs on the other hand increases by 300%, which is a result of its low initial raw material value in relation to potential pyrolysis costs. Indeed, lignin-based carbon fibre may be too severely punished by the assumption that the epoxy matrix needs to be removed via pyrolysis. If instead it is assumed that the full composite can be

incinerated, as the lignin-based fibres have a CO₂-neutral footprint, the cost of the fibre system will most likely be of the same level or lower than the cost presented in Fig. 4b.

With regard to glass fibre composites, the two highest stiffness points presented in Fig. 6 are hypothetical figures. This is because these data points correspond to stiffness achieved when using UD or ASMC reinforcement which is not achievable when recycling fibres through mechanical recycling. These hypothetical data points were given to illustrate that the use of a gentler recycling process where fibre length and quality are sustained ought to result in a better recycle value. Furthermore, the value-versus-stiffness-trend depends on whether S-glass or E-glass is used, and results in material cost reductions of 32% and 45% respectively. Please note that the primary reason for choosing S-glass is not stiffness, as the difference between S- and E-glass is fairly low (~10%), but other governing properties. Consequently, in this application, where stiffness is the resale criterion, it is deemed appropriate to use the figure of 45% material cost reduction when discussing glass fibre recycling potential.

The material cost of the first generation recycled carbon fibre system reduces by about 30% when re-sale value is included and the material cost of the second generation of recycled carbon fibre system reduces about 18%.

11. Implications for material selection based on lightweight design and material cost

As the general driving force for implementing composite materials in a structure is weight reduction, it is of interest to investigate the impact of the recycle value potential on lightweight design. This can be done on a thematic level using the Ashby-methodology (Ashby, 1993, 2000; Ashby and Johnson, 2010), where the optimum multidisciplinary design can be determined using a combination of performance indices. Here, a tie performance index of ρ/E (Ashby and Johnson, 2010) is used as it represents a reasonable trade-off between mechanical properties and density and thereby, weight. A lower value of the performance index indicates greater mechanical performance per unit weight, meaning a more light-weight design. Adjusted raw material cost, as defined in Section 10.3 and Fig. 6, as a function of the weight-based performance index is presented in Fig. 7 for researched fibre systems. An optimal material minimizes both performance index and

raw material cost, as marked in the graph using a star. This direct comparison of weight-efficient performance and material cost shows that recycled materials indeed provide the designer with further material choices, choices which reduce both cost and weight, something the virgin fibre system (carbon/glass) does not achieve. In fact, given this, narrow, design space, the virgin material systems represent opposing objective functions with carbon fibres representing the lowest weight design solution and glass fibres representing the lowest cost solution while the recycled carbon fibres effectively fill the empty gap in between, providing the designer with a more optimal multi-objective design solution. This filled-in gap enables the use of composite materials in applications currently avoided due either to mechanical or cost reasons, as well as enabling the further optimization of current material usage.

For comparison, representative cost and stiffness data of aluminium and steel (Ashby and Johnson, 2010) are also given in Fig. 7. Note that the costs of these comparative materials are not adjusted with respect to potential recycle value, but are shown for comparative reasons in the scope of the material selection process. Both virgin and recycled CFRP have a lower performance index than steel. Recycled CFRP is of overlapping performance index with aluminium as decreasing fibre alignment is applied. GFRP and lignin-based carbon materials of a low fibre volume fraction and high randomness have a higher performance index than, for example, aluminium and carbon steel. The comparison to steel and aluminium show the importance of properly tailored fibre alignment and fibre length in composites in order to achieve lightweight performance.

12. Discussion

The authors have proposed a method for rating the economic value of recycled fibre reinforcement of common light-weight fibre-reinforced materials. The proposed method ties material values with mechanical performance of fibres retrieved and is based on a literature review of current state-of-the art, economically and industrially sound, recycling methods for fibre-reinforced composite materials. The proposed model and methodology have been founded on a number of limitations and assumptions, which in turn dictates the validity of the model. As such, further improvements and more detailed sensitivity analyses could be made on cost data and the parameters included towards verifying different aspects of the model. Such improvements could enable more specific, detailed future analyses as well as tailor the validity of the model to specific scenarios. Moreover, the model could be developed to include recycling costs of solvolysis and fluidised bed techniques, given data in recently published research (Dong et al., 2018; Meng et al., 2018). However, the simple form chosen was deemed sufficient and approachable with regards to the holistic analysis scope and the intended wider audience of this paper. Furthermore, the chosen modelling approach was purposely disconnected from the traditional, alternative LCA/LCC approach (Witik et al., 2013; Jank et al., 2017; Y.S. Song et al., 2009; Dong et al., 2018; Li et al., 2016). This to fully focus on the production phase, as opposed to the full structure lifetime scope of LCA analysis. This of course also means that the model only implicitly records environmental impact through the coupling of recycling cost to energy use. However, given the large body of work available demonstrating the reduced lifetime cost of recycled composite materials (Dong et al., 2018; Meng et al., 2018; Witik et al., 2013), it can be argued that the environmental impact is widely accepted, whilst the production cost impact is less often discussed as it represents a minor part of the full lifetime of a component. Indeed, for cost-sensitive applications such as automotive, marine and vehicle-bound transport, a low lifetime cost is sound however the component must still be

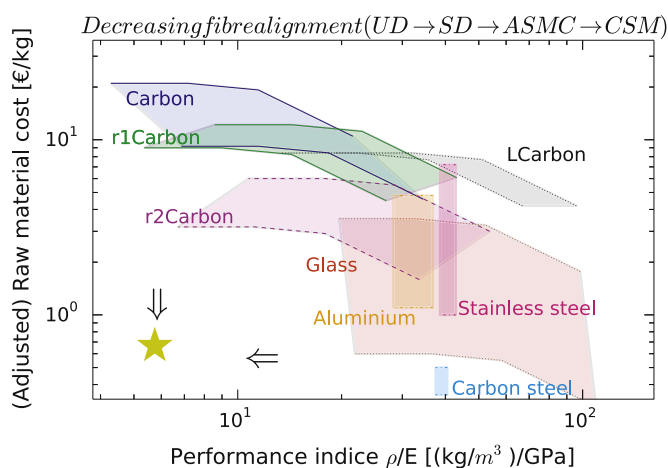


Fig. 7. Adjusted raw material cost as a function of tie performance index, illustrating weight efficiency of composite materials researched. Non-adjusted raw material cost ranges of comparative metals (aluminium, carbon steel and stainless steel) are drawn from data retrieved from Ashby and Johnson (2010).

competitive in production (Mårtensson et al., 2015). This makes the chosen research scope crucial to enable further use of composite materials, and to introduce the reduced environmental impact that comes with it.

The proposed model has been used to analyse the economic potential and impact of closed-loop recycling of carbon and glass fibre-reinforced composites on lightweight design. The potential of each fibre system addressed factors in different industries concerned with component weight. Drawing from the results, a final concluding figure was developed, listing the potential relevant applications of each fibre type and its recycle, see Fig. 8.

Fig. 8 shows that the re-introduction of recycled fibres into the production flow has the potential to be valuable to new industries, previously deterred by high fibre material costs for required mechanical property levels. Other authors have recognised this fact on a generic basis (Vieira et al., 2017; Pimenta and Pinho, 2011), and on more specific levels in automotive adaptations (Meng et al., 2018). However, the value of a closed cycle material loop could transcend individual industries, resulting in material transfer from one type of industry another. To exemplify, as in Fig. 8, a circular closed-loop material cycle of carbon fibres may start as a structural flying component. Then, be recycled as chopped fibres bordering ASMC-quality that are introduced into a structural press-formed automotive part. This is then recycled once more into non-woven, lower volume-fractions, SMC, and press-formed, now into a semi- or non-structural automotive or potentially marine component. Finally, the

fibres from the semi- or non-structural automotive component can be reclaimed and ground into a reinforcing powder used in an injection-moulded thermoplastic component, for example (Chen et al., 2014), for final use in a miscellaneous application outside the transport industry.

However, in order for this closed-loop, industry-transcending material flow to function, the infrastructure must be established so that material quality sorting and matching between industries is efficient and economical. In addition, transport costs and transport distances must be minimised. These recognised challenges of closed-material-cycles (Velis, 2018) are outside the scope of this paper but are nevertheless vital for the proposed potential closed material cycle to be realised.

On the structural potential of recycled fibres, current industry recycling of composite materials where fibre grades are, to some extent, mixed, is not ideal. In order to maximise the retrieved mechanical properties of the reinforcement more attention must be paid to developing efficient fibre and reinforcement grade sorting techniques, energy-minimised recycling methodology and techniques as well as actual manufacturing methods where recycled shorter fibres can be efficiently tailored and achieve directional reinforcement potential. Other researchers have recognised the need to develop further fibre grade sorting techniques (Job et al., 2016; ELG Carbon fibre Ltd, 2016), and there is on-going research into manufacturing techniques, such as HiPerDiF (High Performance Discontinuous Fibre), that can create directional

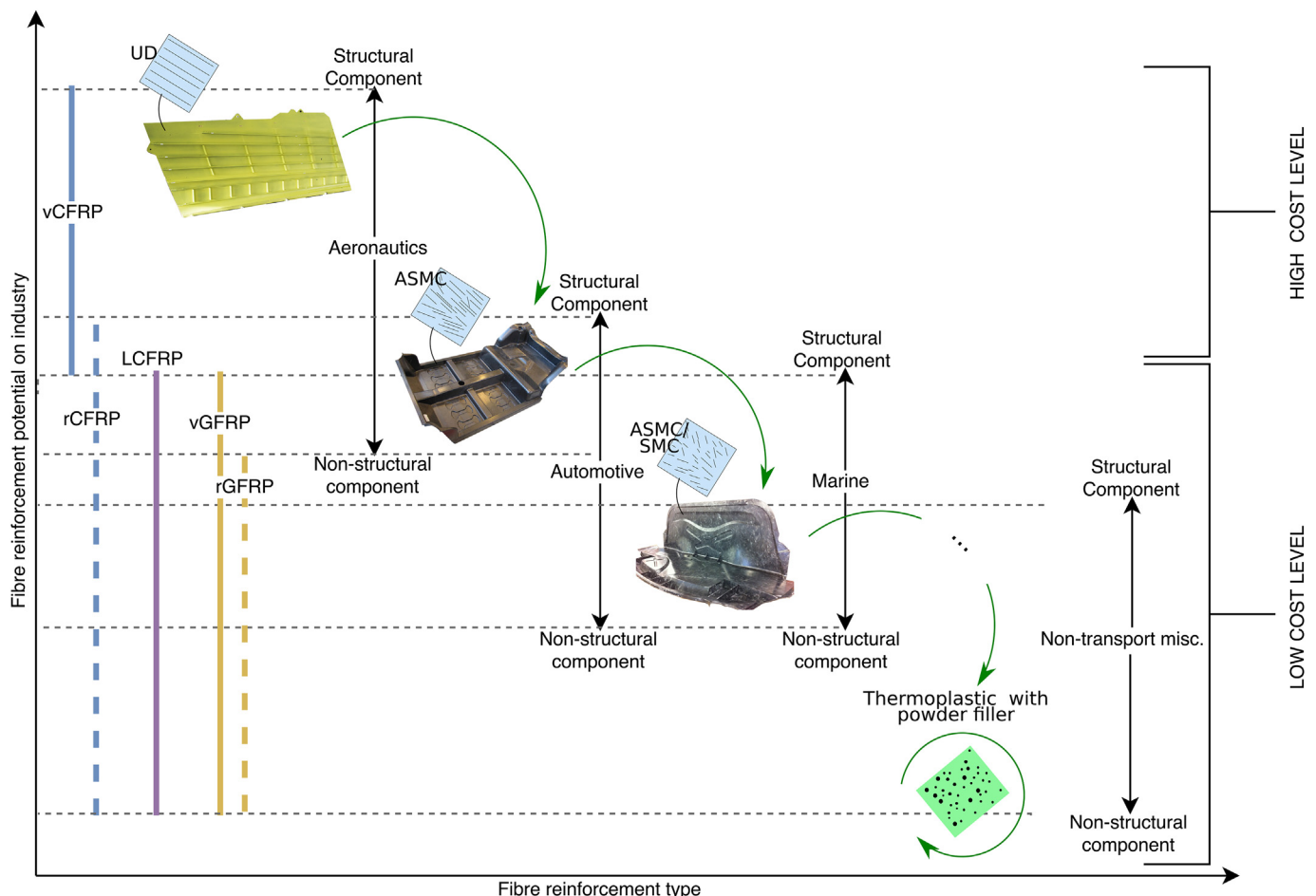


Fig. 8. Relevant applications for researched recycled composite materials together with a conceptual closed loop material flow of the transfer from a structural aeronautical component to a powder-filled thermoplastic component.

reinforcements from chopped fibres (Longana et al., 2016; J.Tapper et al., 2018).

The extent to which structural potential challenges can be addressed is very much governed by the supply and demand of actual EoL components and production waste available. Other researchers (Meng et al., 2018; Dong et al., 2018) have recognised issues with the availability of EoL components to feed a future supply of recycled materials. Moreover, in addition to fibre grade sorting issues there is the problem of disassembly of, for example, metallic inserts and other composite mixed materials (Dong et al., 2018). However, given the increased worldwide use of composite materials (Nilakantan and Nutt, 2015), increased availability of EoL components will enable the introduction of more efficient fibre grade sorting systems that better utilise the mechanical performance of fibre grades in a recycled form.

The results have further underscored the importance of reclaiming production waste. Data points of recycled material of highest mechanical property, meaning that of uni-directional fibre, correspond to either very efficient recycling where fibre quality and length is sustained, or that of production waste. This means that simply discarding production waste represents a high value loss, while reclaiming production waste may be highly beneficial in the form of a recycled material of high-level mechanical properties and low second-hand cost. Naturally, the reintroduction of production waste into the production loop presents challenges and costs in itself such as reinforcement sorting, grading and nestling of scrap pieces, however, given proposed results, it may well prove worthwhile, at least, within an individual facility.

An alternative use of proposed recycle model could be to optimize involved driving recycling parameters to improve the recycling efficiency. However, given the intention to investigate a cyclic recycling material flow and the fact that an efficient parameter and process optimization need to be performed in close collaboration with recyclers, such optimization was beyond the scope of this paper.

Finally, on a broader scale, the potential of the recycled composite materials in lightweight-performance-driven adaptations highlights the importance of further research into the recycling of composite materials for structural adaptations. In fact, results in this paper have justified future research by the authors (Hagnell, 2019) on the subject of the economic- and weight-potential of recycled composite materials in representative, weight-optimised, structural adaptations.

13. Conclusions

The authors proposed a recycled material value model based on retained mechanical performance of fibres retrieved. The results showed that recycled fibre materials return high value that can be used to help to mitigate the high raw material costs of common fibre-reinforcing systems. Some detailed conclusions on the fibre systems researched are:

- Reclaimed fibre length dictates potential reuse value of recycled carbon-fibre for reintroduction in higher-performing reinforcement designs.
- EoL and production waste produced from aeronautical-grade carbon fibre reinforcement should be recycled in reinforcing fibre form at least three times.
- Recycled high-level production and EoL carbon fibre waste can reduce material costs by about 50%
- Aeronautical-grade carbon fibres of generation four or more should be recycled into powder-form while lower-grade carbon fibres should be immediately recycled into powder-form.

- Lignin-based carbon fibres can ideally be incinerated to retrieve their energy value.
- Glass fibre reinforcement should be continuously recycled and used as fibrous and filler reinforcement in BMC-materials.
- Recycled production and EoL glass fibre waste can reduce material costs by about 45%.
- High-to medium level recycled CFRP are of similar raw material cost and lower weight performance as aluminium.

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