THE RECYCLING OF OMC'S CARBON REINFORCEMENT BY SOLVOLYSING THERMOSET MATRIX. A WAY OF SUSTAINABILITY FOR COMPOSITES

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Abstract

Originally developed for high-tech applications, carbon fibre/thermoset matrix composites have been increasingly used in leisure and sports industries, for several years. But the carbon reinforcement is the most expensive constituent, and also the most environmentally impacting in the elaboration of a composite part. To this day, no end-of-life solution or recycling process efficiently exists. This paper aims at demonstrating that recovering the carbon reinforcement is possible, technically and economically speaking. Moreover, it is particularly the basis for a life cycle analysis that assesses benefits and environmental challenges of this recycling loop based on the reinforcement recovery by a solvolysis of the organic matrix. Lastly, the lack of data to consider the better end-of-life option (reuse, recycling, energy recovery and material valorisation) will be underlined.

1. Introduction

Organic matrix composites (OMCs), and more specifically carbon fibre/thermoset matrix composites were originally developed for high-tech applications in aeronautic and aerospace fields. However for several years, these materials have been increasingly used in automotive and leisure and sports industries. In these sectors, one may seek aesthetic criterions or a simple feeling of high technology, more than high technical properties. Thus, constituents' characteristics, and specifically reinforcement's (*e.g.* Young's modulus, tensile strength, etc.), are considered as a secondary matter and may be oversized regarding the function of the product. This is particularly true for non-structural decorative parts (*e.g.* with a carbon look finish), for which the reinforcement is the most expensive constituent, and where glass fibres, very much cheaper, cannot be used.

As a consequence for these scopes, alternative solutions to technical and economic inadequacies have to be found, but kept in line with the environmental impact of the product. Indeed, it has been shown that the carbon reinforcement is environmentally the most impacting constituent in a carbon/epoxy composite's elaboration process (cf. analysis figure 1) [1]. First, recycling end-of-life composites (even restricted to the reinforcement recovery) could lead to reduce some anthropogenic impacts, by decreasing the use of first-generation

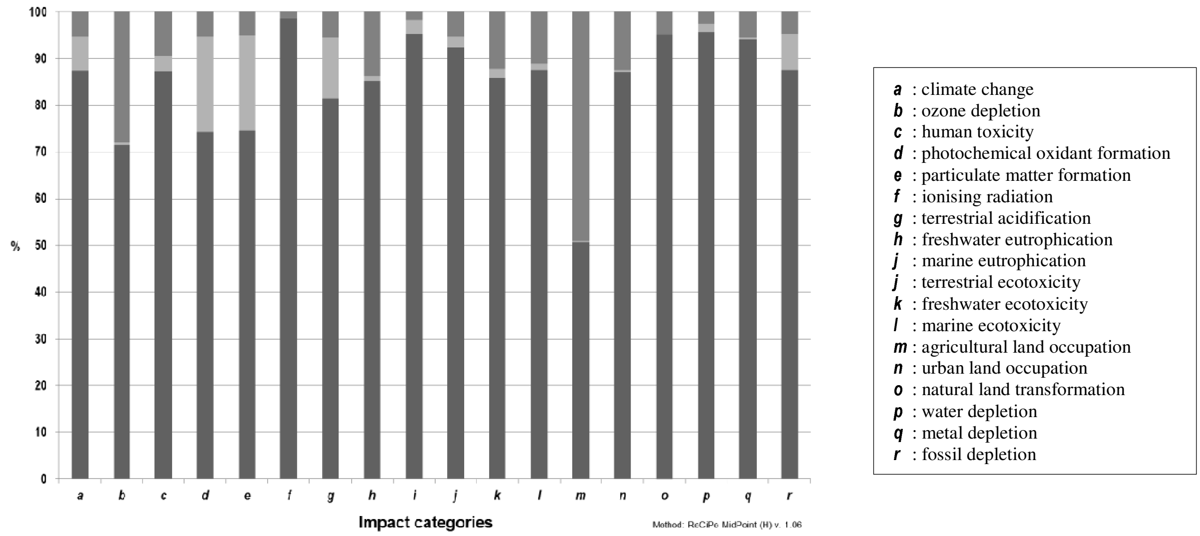

Figure 1. Environmental impacts due to the carbon reinforcement (*dark grey*), the epoxy matrix (*light grey*) and the injection moulding process (*intermediate grey*), while processing a 1 kg carbon/epoxy composite part. The analysis is based on Duflou *et al.* data [1].

raw materials (mainly petroleum) for their production. Besides, it would help design engineers to balance energy efficiency and cost, by opening new opportunities for developing second-generation composites, firstly dedicated to the manufacture of medium or low loaded parts. Lastly, recycled carbon fabric could widen the range of reinforcements on the marketplace, between first-generation carbon and glass fibres.

All this has to be led in line with European directives that already force industries to improve their products' recyclability (*e.g.* in automotive industry [2]). However, making viable this new recycling sector requires overcoming users' reluctances by ensuring the second-generation semi-product's validity from economic and environmental aspects. Therefore, we carried out a life cycle assessment (LCA), in which the resource efficiency and potential environmental challenges of the carbon/epoxy composites' recycling process are analysed.

2. Materials and methods

2.1. Carbon/epoxy manufacturing: available data

Duflou *et al.* [1] have some life cycle assessment based information on the environmental impacts due to the manufacturing of semi-structural composite parts in a car, as an alternative to steel. It leads to lighten vehicles, therefore to reduce life-cycle air emissions, beyond the benefits of plug-in vehicles [3]. Indeed in a conventional car, the use phase has the greatest environmental impact due to high fuel consumption (directly related to the mass of the vehicle). In its lighter alternative version, the manufacturing phase becomes predominant [4]. This is mainly due to the carbon fibre manufacturing, as previously seen in our analysis based on data from Duflou *et al.* [1] (figure 1). Hence, it might be of real interest from a sustainability point view to propose the use of recycled fibres as a way forward to limit the environmental impacts of the composite parts in lighter cars.

Due to the fact that the carbon reinforcement is the most impacting constituent in a carbon/epoxy composite's elaboration process (figure 1) [1], recycling end-of-life composites (even restricted to the reinforcement recovery) could lead to reduce some anthropogenic impacts, by decreasing the use of first-generation raw materials (mainly petroleum) for their production. Besides, it would help design engineers to balance energy efficiency and cost, by opening new opportunities for developing second-generation composites, firstly dedicated to the manufacture of medium or low loaded parts. Lastly, recycled carbon fabric could widen the range of reinforcements on the marketplace, between first-generation carbon and glass fibres.

All this has to be done in line with European directives that already force industries to improve their products' recyclability (*e.g.* in automotive industry [5]). However, making practicable this new recycling sector requires overcoming users' reluctances by ensuring the second-generation semi-product's validity from economic and environmental aspects. Therefore, we carried out a life cycle assessment (LCA), in which the resource efficiency and potential environmental challenges of the carbon/epoxy composites' recycling process are analysed.

2.2. Life cycle assessment: goal and scope

Every stage of the lifecycle of the composite part has to be modelled in a LCA, from the extraction and treatment of raw materials, to the product end-of-life treatment, following the usual steps defined by the ISO 14040 standards [6]. Results are assigned to impact categories like climate change or ionizing radiation (cf. figure 1's caption). The *environmental profile* consisting of the indicator results, provides information on the environmental issues associated with the inputs and outputs of the product system under study.

As previously mentioned, we focus on carbon/epoxy composites. The deposit of materials to be recycled consists possibly in end-of-life aeronautic parts, but most likely to date, in composite offcuts. The composite part chosen for the LCA is assumed to be processed in Europe, with Japanese carbon reinforcement. Its mass is supposed to be 1 kg. Thus, we aim at studying the interest of recycling such material that is to say more generally, the environmental feasibility of the recycling process.

2.3. Life cycle inventory

The following analysis is still based on Duflou's *et al.* data [1] that have been recalculated relative to the mass of the chosen product, *i.e.* 1 kg.

In our case study, the use phase is not taken into account. Indeed, to the best of our knowledge, the only input data that can be taken into account concern transport operations. Like so, as rather classically, the present simulation led to show that this factor did not contribute much to the overall impacts (less than 5%).

Regarding the product's end-of-life, two scenarios have been modelled:

- the first one consists in burying the composite part, that is what is currently done;
- the second one consists in the recovery of the carbon reinforcement. We focus on the process mastered by the Institute for Solid State Chemistry - Bordeaux (ICMCB). We consider:
  - (i) an aqueous solvolysis of the matrix by water under supercritical conditions (temperature around 400 °C and pressure about 25 MPa);
  - (ii) and a hydrothermal oxidation of the effluent to clear matrix components from water, at the end of the solvolysis process.

This technology allows the fibre to be recovered, using energy, water and oxygen, and emitting only water and carbon dioxide. Therefore, it is a real (but partial) recycling, and not a simple material valorisation [7].

Lastly, the research team headed by O. Mantaux, Institute of Mechanics and Mechanical engineering of Bordeaux, has developed a prototype for packaging these second-generation fibres in an attractive form for users (*i.e.* designers). Data that match this remanufacturing stage have not been taken into account yet in this very first LCA. However, this energy input is assumed to be very weak, compared to those involved in the first-generation reinforcement process, and even to the fibre recovery stage. As a consequence, the life cycle only loops after the manufacturing of the first-generation carbon reinforcement, with no specific additional remanufacturing.

2.4. Life cycle assessment: software, database and method

The LCA is performed with the SimaPro software (v. 7) [8], the EcoInvent database [9] and the ReCiPe Midpoint (H) method [10]. As previously mentioned, in the recycling stage, the avoided material is the reinforcement. In other words, the production of a new raw material with non-renewable resources (*i.e.* first-generation carbon reinforcement) is avoided.

3. Results and discussion

3.1. Environmental validation

Let assume the two end-of-life options for the 1 kg carbon/epoxy composite part: landfill of the whole part, or recycling of the only reinforcement by aqueous solvolysis of the matrix.

Environmental impacts of both scenarios have been assessed, and compared in figure 2. One may note that a negative impact does not mean that the recycling process would be in itself an environmental benefit; this analysis only highlights the impacts avoided by the reinforcement recovery instead of a new manufacturing stage. Thus, according to the ReCiPe Midpoint (H) method, figure 2 shows that the fibre recycling almost offsets the whole impacts due to the manufacturing stage, in spite of the electricity consumption of the recycling process.

If we compare the environmental impacts of the composite part throughout its lifecycle (end-of-life included), the interest of the recycling option is also clearly shown (cf. figure 3). The environmental gain is on average about 80%. Moreover, as a case in point, the climate change impacts (referenced *a* on figure 3) even become zero in the case of a French electricity country mix-based analysis. Indeed, it is mainly nuclear energy sourced; however, it primarily results on ionizing radiation impact (*f*).

As a conclusion, the interest of recycling the only reinforcement is clearly shown on the environmental point of view. Actually, it especially leads to almost offset the whole environmental impacts of the manufacturing stage.

3.2. Economic validation

A market study has been recently carried out. It shows that there will always be relevant uses for recycled reinforcements or for semi-products based on second-generation fibre, whatever their mechanical characteristics are, and as long as their price remains reasonable. The integration of recycled carbon fibre is only interesting if the mechanical performances/price

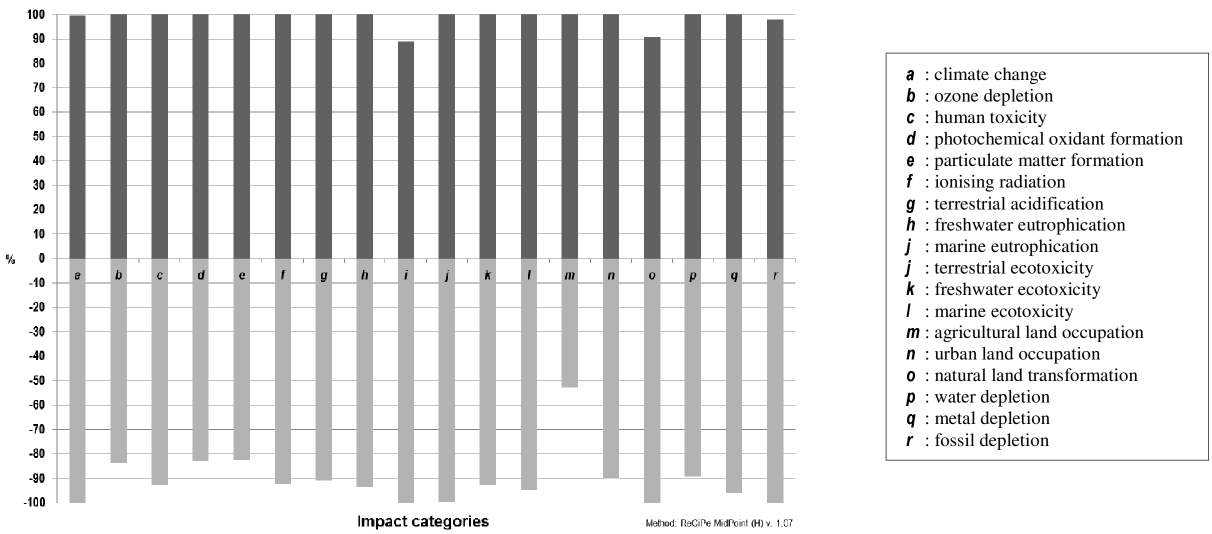

Figure 2. Compared impact assessment of two end-of-life scenarios for a 1 kg carbon/epoxy part: landfill of the whole part (*dark grey*) and reinforcement's recycling (*light grey*). The analysis is based on the ReCiPe Midpoint (H) method.

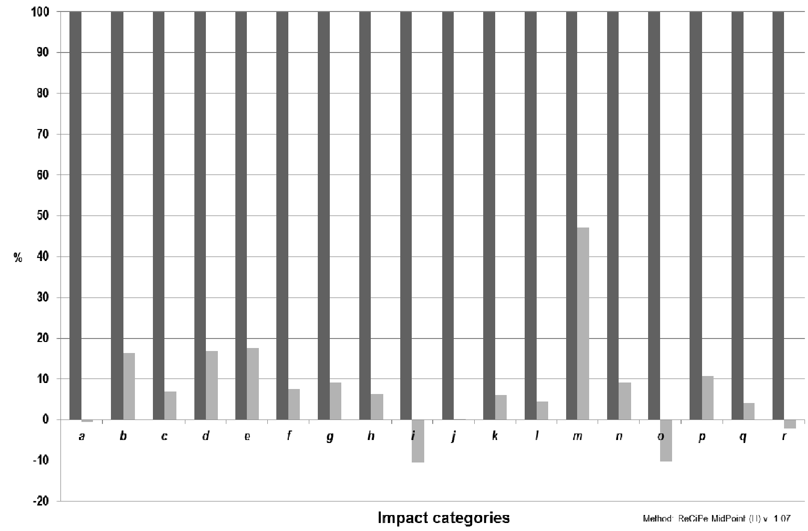

Figure 3. Comparison of the environmental impacts of a 1 kg composite part throughout its life cycle, and depending on the end-of-life option: dark grey refers to the landfill of the composite part, and light grey to reinforcement recycling (Caption: see above, figure 2)

ratio is higher than glass fibre's. Therefore, in light of the excellent second-generation reinforcement's mechanical properties [11], this ratio should be much higher than for new carbon fibres. Thus, the feasibility of the recycling will be provided if the second-generation semi-products price does not exceed 70 to 80% of the new ones.

The next step in the maturation of this technology is the development of a pilot scale facility for the recycling of carbon fibres from epoxy matrix based composite using the supercritical fluid technology.

3.3. End of life scenarios: a lack of information

To complete the environmental validity of the recycling of the carbon reinforcement, an overall study of several end-of-life scenarios is needed [12],[13],[14],[15].

For high-tech applications, the carbon fibre's reuse is not a valid option. Due to very specific and various shapes and the use of a heat-hardening matrix, composite parts are not reusable.

Incineration consists in burning the only matrix, while carbon fibres are kept unspoiled at these temperatures. Pyrolysis is only a way of recovering an unstructured (fluffy) carbon reinforcement architecture. Mechanical properties are also lost [12],[13]. Lastly, no environmental data are available for this process applied to the composite studied.

By grinding the whole composite, carbon fibres remain reusable, but they completely lose their mechanical properties. Recycling only comes down to *downcycling*, that is to say here to a just material valorisation. In order to model the environmental impacts due to this end-of-life option, we need to know what kind of material should be replaced by this recyclate. The problem lays on the fact that this composite is too young to be at its end of life and no real data exist on its future (only suppositions).

4. Conclusions

In the present context, the use of carbon/epoxy composite is ever increasing. Now, it is well known that those composites can be recycled [16], keeping good mechanical properties [11]. Anticipating that they may be subjected soon to regulation, it is essential to show if a composite recycling network can be set up, what would be both economically and environmentally favourable.

The recovery of the carbon reinforcement (constituent the most environmentally impacting in the composite manufacturing) by the aqueous solvolysis of the composite's matrix, leads to an average gain of about 80% for all eco-indicators, compared to the landfill end-of-life option.

Lastly, the recently developed remanufacturing process allows obtaining a semi-product easily usable. Consequently, from an economic point of view, the mechanical performances/price ratio of the second-generation carbon fibre should be higher than the virgin carbon fibres', or the glass reinforcement's. The next step in the maturation of this technology is the development of a pilot scale facility for the recovery of carbon reinforcement, using the supercritical fluid technology.

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