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**Official URL:** <https://doi.org/10.1016/j.resconrec.2018.01.024>

# **To cite this version:**

Vo Dong, Phuong Anh<sup>-2</sup> and Azzaro-Pantel, Catherine<sup>-2</sup> and Cadene, Anne-Laure *Economic and environmental assessment of recovery and disposal pathways for CFRP waste management.* (2018) Resources, Conservation and Recycling, 133. 63-75. ISSN 0921-3449

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# Economie and environmental assessment of recovery and disposai pathways for CFRP waste management

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**ARTICLE INFO ABSTRACT** 

*Keywords:*  **Carbon fibre reinforced polymer Waste management Recycling Economie assessment GWP** 

**The high cost and energy intensity of virgin carbon fibre manufacturing constitute a challenge to recover substantial value from carbon fibre reinforced polymers (CFRP). The objective of this study is to assess the environmental and financial viability of several waste management processes for CFRP. Life cycle costing and environmental assessment models are developed to quantify the financial and environmental impacts of waste treatment pathways comparing a panel of recycling techniques that are now available (grinding, pyrolysis, microwave and supercritical water) and that can be used to substitute different grades of both carbon and glass fibres by recycled carbon fibres at competitive prices compared to landfill and incineration. GWP assessment promotes recycling activities by recovery of carbon fibre due to the high avoided impacts from substitution of virgin fibre, thus highlighting the high interest of recycling over conventional production for environmental purpose. Fibre recovery rate and recycling capacity are pivotai to decrease the unit cost of recycled fibre as well as GWP impacts. The advantages and drawbacks of each technique are analysed through economic and environmental indicators, to better understand the network configuration for optimisation purpose of waste management pathway in a holistic viewpoint.** 

# **1. Introduction**

**Due to their low density and high performance of physico-chemical properties, Carbon Fibre Reinforced Polymer Composites (CFRP) are increasingly used in structural applications to replace more conventional materials (steel, aluminium, alloys ... ) for the design of lighter products. According to Black (2012), the global demand of carbon fibres was expected to exceed production capacity in 2015 and if growth**  remains at this rate, a huge amount of waste will be generated. The **benefits of CFRP recycling are threefold: first, it is necessary to limit the accumulation of waste second, recycling could be a fibre supply solution in order to meet future demand (Black, 2012) and third, recycling could be expected as a Jess energy-intensive operation with lower environmental impact than the traditional way to produce virgin CFRP, due to the bypass of some operation steps. Carbon fibre manufacturing is an energy intensive process (183-286 MJ/kg of carbon fibre, (Song**  **et al., 2009)) that transforms the precursors with poorly ordered structure into a nearly perfect graphite structure in carbon fibre (CF) and generates environment and human health impacts due to emissions from the oxidation and carbonization fumaces, such as HCN, NH3, NOx .. , (Grzanka, 2014).** 

**Composites recycling is a difficult process due to the heterogeneous nature of the matrix and the reinforcement, especially in the case of thermoset composite (Pickering, 2006). Only few commercial recycling operations for main strearn composite materials are available due to technological and economic constraints. The utilisation of recycled carbon fibres (RCF) in industry generates some challenges due to their lower quality than virgin carbon fibres (VCF) (McConnell, 2010) and variability affecting many factors such as, length distribution, surface quality (adhesion of fibre and matrix), as well as their origin (different grades of fibres are found at various composite scraps from different manufacturers) (Oliveux et al., 2015a). This explains why the Jack of** 

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**https:/ /doi.org/10.1016/j .resconrec.2018.01.024** 

Abbreviations: BMC, bulk moulding compound; CEPCI, chemical engineering plant cost index; CF, carbon fibre; CFC, carbon fibre composite; CFRP, carbon fibre reinforced polymer; D, depreciation; FRP, fibre reinforced polymer; FU, functional unit; GF, glass fibre; GFRP, glass fibre reinforced polymer; GHG, green house gas; GLARE, glass laminate aluminium reinforced epoxy; GWP, global warming potential; GWPA, GWP impact of substituted products; GWPP, GWP impact of process; GWPTOT, GWP total of the system; LCA, life cycle assessment; LCC, **life cycle cost; LP, linear programming; MFA, material flow analysis; MILP, mixed integer linear programming; NPV, net present value; OC, operation cost per mass unit of waste; PAN,**  polyacrylonitrile; RCF, recycled carbon fibre; RGF, recycled glass fibre; SCW, supercritical water; SMC, sheet moulding compound; TC, total annual costs; TRL, technology readiness level; **UCF, average unit cost per mass unit of recovered fibre; UCW, average unit cost per mass unit of waste; VCF, virgin carbon fibre; VGF, virgin glass fibre** 

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markets, high recycling cost, and lower quality of the recyclates versus virgin materials still currently constitute major commercialisation barriers for composite recycling (Yang et al., 2012).

Current waste policies served as an incentive to develop composite recycling solutions, including general policies (The European Directive on Landfill of Waste (Directive 1999/31/EC, 1999)) and applicationspecific legislation (e.g., the End-of-life Vehicle (Directive 2000/53/EC, 2000)).

In parallel, several recycling technologies have been developed for composite materials over the past decades. In particular, the recycling of thermoset composites is receiving a lot of attention due to the technical difficulties to separate the thermoset matrix from the reinforcement materials (Yang et al., 2012). Different recycling techniques of FRP have been studied and developed in order to improve the recycling yield and the properties of the recovered fibre by three main types of techniques: (1) Mechanical techniques in which fibre and matrix are separated by shredding (grinding technique) (Pannkoke et al., 1998; Kouparitsas et al., 2002; Ogi et al., 2005, 2007, Palmer et al., 2009, 2010; Howarth et al., 2014) or high voltage pressure (electrodynamic fragmentation) (Müller, 2013; Mativenga et al., 2016) without chemical reactions; (2) Thermal techniques in which matrix is decomposed by heat (conventional pyrolysis, fluidised bed) (Fenwick, 1996; Kennerley et al., 1998; Pickering et al., 2000; Yip et al., 2001; Cunliffe et al., 2003; Gosau et al., 2006; Jiang et al., 2008; Meyer et al., 2009; L6pez et al., 2012, 2013) or microwave radiation (microwave) (Lester et al., 2004; Akesson et al., 2013; Obunai et al., 2015) into heat or residual liquid; and (3) Solvolysis techniques in which matrix is decomposed by chemical reactions in water or in other organic liquids at atmospheric pressure or supercritical conditions (Allred et al., 2001; Hyde et al., 2006; Pifiero-Hemanz et al., 2008a,b; Jiang et al., 2009; Nakagawa et al., 2009; Yuyan et al., 2009; Bai et al., 2010; Kamimura et al., 2010; Feraboli et al., 2012; Knight et al., 2012; Morin et al., 2012; Onwudili et al., 2013; Oliveux et al., 2013, 2015b; Okajima et al., 2014; Yildirir et al., 2014). Other recycling solutions can be found such as electrochemical (Sun et al., 2015) and biotechnological (Hohenstein Institute, 2015) techniques but they are less mature than other ones for CF recovery.

Life cycle assessment of FRP/CFRP has also received a lot of attention in order to study the environmental benefits of these composites that can be gained from the use of more conventional materials (Takahashi et al., 2002; Duflou et al., 2009; Suzuki and Takahashi, 2005; Song et al., 2009; Das, 2011; Witik et al., 2011, 2012). However, these studies focused mostly on the production and utilisation phases of such materials. The step of waste treatment is poorly studied and generally limited to one technique, e.g. recycling by microwave (Suzuki and Takahashi, 2005; Das, 2011) or recovery energy by incineration (Witik et al., 2011).

The literature analysis reveals that the majority of works reported are devoted to the development of a specific CFRP recycling process or to a specific recycling pathway. As highlighted in (Job et al., 2016), the challenge is now to develop appropriate business models, integrating with existing waste management supply chains and with associated capital investment, to enable commercialisation of what is technically proven. The proposed works aim at considering the whole waste management supply chain model in order to compare the potential benefit of each recovery pathway not only from an environmental viewpoint but also from an economic one.

For this purpose, the independent assessment of each pathway through its inputs and outputs under economic and environmental which is the prerequiste for system modelling is carried out in this study to identify the typical features, as well as the advantages and weaknesses of each recycling/recovery pathway. The composite waste treatment technologies that have been identified in the dedicated literature whatever their technology readiness level (TRL), i.e. landfill, incineration, co-incineration, mechanical recycling, pyrolysis, microwave and supercritical water, are ail assessed in this study with economic and environmental indicators in an exhaustive and complementary way. Various indicators which represent the different viewpoints of the involved stakeholders will also be discussed.

This paper is organized as follows. First, a brief literature review on the Life Cycle perspective situates (see Section 2) the research focus within the scope of CFRP recycling/recovery pathways. The methods and tools that will be used throughout this study for the development of the framework for CFRP waste management and the assessment of economic and environmental will be addressed in Section 3. The analysis and results are presented in detail in Section 4. Finally, Section 5 will conclude this study on CFRP waste management and offer perspective for CFRP waste supply chain deployment and optimisation.

# **2. Literature review on life cycle perspective of CFRP recycling pathways**

The literature analysis reveals that some articles have discussed the environmental impacts of transitioning from conventional materials to FRPs, as determined by Life Cycle Cost (LCC) and Life Cycle Assessment (LCA). The work reported in Hedlund-öström (2005) that applied LCC and LCA is focused on waste treatments of End-of-life CFRP and other composites involving grinding, fluidised bed and incineration. As LCC and LCA of waste treatment phase depend on the recovered products, not surprisingly, the choice of the replaced material between virgin carbon fibre (VCF) and virgin glass fibre (VGF) is particularly significant for result interpretation. Incineration may have a higher advantage than recycling if the recycled carbon fibre is used to replace low value material, such as glass fibre. In reality, the characteristics of the recycling process may impact the quality of recovered fibre output, besides the type of origin fibre in waste. The studies on CFRP recycling techniques have thus reinforced the need of in-depth investigations on the structure of CFRP waste treatment (Hedlund-Åström, 2005; Witik et al., 2013; Li et al., 2016)

Witik et al. (2013) studied the environmental impacts (climate change, resources, ecosystem quality and human health) of three waste treatment options, i.e., pyrolysis, incineration and landfilling. A quantitative mode! for the determination of equivalent quantities of VCF and VGF, which are replaced by RCF to achieve mechanical performance equivalent to virgin material in Sheet Moulding Compound (SMC) through the tensile modulus. However, the utilisation of RCF in polymer matrix is a complex process depending on numerous criteria apart from the tensile modulus. Although the market of RCF has not been mature due to the uncertainty of their mechanical properties compared to VCF, their potential applications are numerous, not only in reinforcement purpose (Bulk Moulding Compound (BMC), Sheet Moulding Compound (SMC), thermoplastic composites, concrete ... ), but also in other applications which do not depend much on mechanical properties of materials such as electrical and electronic products, e.g. electromagnetic shield (Wong et al., 2010).

Li et al. (2016) carried out a study on LCC and environmental assessment (GWP, energy use, final disposai waste) for End-of-life CFRP in automotive with three options (landfilling, incineration and mechanical recycling) within regulations of UK and EU. In this hypothetical case, a landfill tax can be viewed as a useful tool to shift CFRP waste from landfill to incineration because of the low GWP impacts and energy use in landfilling. Recycling benefits depend on the displacement factors of VCF by recycled fibre and on the recycling rate in order to balance the energy-intensive recycling process. However, grinding process in mechanical recycling degrades fibres on reducing their length and cannot separate cleanly fibre and matrix from the composite (Kouparitsas et al., 2002; Palmer et al., 2009). Increasing recovery rates can improve environmental and financial performance of the mechanical recycling pathway: in the base case, only 40% of CF present in CFRP waste is assumed to be recoverable. Considering higher recovery rates is hypothetical for (Li et al., 2016).

An alternative to LCA and LCC is cost-benefit analysis (CBA) (Leu

and Lin, 1998; Morrissey and Browne, 2004; Ali et al., 2013; Farel et al., 2013; Karrnperis et al., 2013). A very interesting contribution is proposed in Farel et al. (2013). These authors have developed a framework for performance evaluation through a cost and benefit analysis of a future End-of-Life Vehicle (ELV) glazing recycling. Technical and economic details of activities have been discussed. The main barriers and potential solutions have been identified from field observation and expert interviews. The consistency and complementarity of LCA and LCC vs CBA assessment methodologies has been presented in detail in Hoogmartens et al. (2014). Traditionally, first, LCA and LCC can be viewed as product related assessments while CBA mostly focuses on projects or policies (Ness et al., 2007). Second, LCA and LCC focus on the whole life cycles of the assessed products while CBA, focusing on the lifetime of a particular project, makes the lifetime of used products secondary. A third key aspect relates to the use of a reference scenario. LCA and LCC are comparative assessment tools that compare products while CBA is typically used for autonomous project evaluation. This reason motivates the use of a combined LCA-LCC approach in this study. lt must also be emphasized that an approach combining environmental assessment and life cycle cost analysis has been recently identified to play a crucial role in identifying suitable waste management strategies to address the emerging waste burden of end-of-life and manufacturing scrap CFRP materials and to determine its beneficial uses in automotive sector or in other applications (Meng et al., 2017).

In that context, the main innovation of the work that is targeted here is to develop a methodological framework for the design and deployment of CFRP waste supply chain considering multiple criteria based on economic and environmental assessment and to highlight the endogenous variables including the characteristics of each waste treatment option as well as the exogenous ones (type of CFRP waste, deposit waste, transport distance, market) that will be further studied in the modelling and optimization of the global supply chain embedding a11 the recovery/recycling pathways of CFRP options.

# **3. Materials and methods**

# *3.1. Studied system*

The system boundary considered is presented in Fig. 1. Ali the impacts or benefits are assessed from the beginning to the end of operation leading to different recovered products until there is no waste left to be treated. Two options conceming carbon fibre recovery are considered: Recovery Pathways and Non-Recovery Pathways. The techniques in the former category allow carbon fibre recycling. In the latter one, although carbon fibre cannot be directly recycled, either energy or materials recovery may be obtained by incineration or co-incineration techniques. Ail the studied techniques will be presented in detail in Section 3.3. The choice of the techniques that are considered within the scope of this work is based both on recent literature review showing the current trends of CFRP recycling and on interviews with experts representing the major stakeholders of the CFRP supply chain (aeronautics, automotive, recycling industries, local and regional administrations, etc.). Technical and economic details of activities have been discussed and the main barriers and potential solutions have been identified from interviews and active survey carried out by the industrial leader of the ANR SEARRCH project (ALTRAN) (http://www. agence-nationale-recherche.fr/Project-ANR-13-ECOT-0005). Interviews with stakeholders have been led in order to take into account their current constraints and concems. This bottom-up approach has been performed in order to build a practical tool, that can be used by current and future actors in the CFRP recycling sector, and in the longer term, beyond the composites, for stakeholders of the recycling sector.

An average composite waste of CFRP type composed of 65 wt% of carbon fibre and 35 wt% of matrix has been considered. The studied carbon fibre is assumed to be produced from Polyacrylonitrile (PAN) precursor. The formulation of the composite will not be further developed and 100% of matrix is assumed to be composed by Bisphenol A epoxy resin without filler.

As CFRP is the composite of polymer matrix, it is not classified as an inert waste regarding organic substances for matrix. In waste management, CFRP can be considered as either non-hazardous waste or hazardous waste depending on matrix properties. Prepreg, which is an uncured composite, is considered as hazardous waste (PlasticsEurope, 2006). The cured composite is considered as a non-hazardous waste if it does not involve any hazardous substance in its formulation.

# *3.2. Methodological framework*

Since the products from the studied waste treatment techniques are different in both type and yield, the functional unit (FU) defined for this study is 1 kg of waste to be treated by one of the proposed technology. Within the boundary of the studied system, three phases of CFRP waste management are assessed: plant construction, operation, and applications for recovered products. These three steps are studied complementally through economic assessment and environmental assessment.



**Fig. 1. Boundary of the studied system.** 

#### **Table 1**

### **Framework of economic model for Recovery Pathways.**



### *3.2.1. Economie assessment methods*

According to literature, composite recycling suffers from financial instability due to the low value of recovered products and the Jack of market (Yang et al., 2012). In this context, an economic model for CFRP waste management is developed here in order to study the profitability of recycling techniques. A classical period of 10 years is considered to study the economic feasibility of the project.

**lt** must be emphasized that the price of CFRP waste has been set equal at zero even though it can be considered as a raw material. Even if this assumption cannot be viewed as a penalizing one, it can be justified here in order to promote the deployment of the market of the recycled fibre.

- The **Non-Recovery Pathways** are considered as outsourcing services of the system, their costs are therefore estimated on the basis of the current fees charged by the government or the concerned industry.
- For **Recovery Pathways,** the contribution of variable costs, fixed capital costs and capital depreciation has been determined using classical methodologies for early estimates as reported in Anderson (2009) (see Table 1). A linear 10 year-depreciation is considered. The investment cost is estimated from the classical six-tenths rule for a fixed capacity of waste input (Seider et al., 2009). The utility costs including electricity, natural gas, and water have been extrapolated from literature data. The source and amounts of utilities depends on the recycling techniques that will be presented in the next section. Labour cost has been estimated from legislation (Eurostat, 2015a) (legal working hours of 1607 h per year with an hourly cost of 34.3 $\epsilon$ ). The recycling plants are assumed to be medium scale with 4 people for operating labour. This assumption will be valid for ail recycling plants whatever the process and the capacity used.

Three economic indicators are considered in this study (Table 2):

- 1. Operation Cost per mass unit of waste (OC) is the cost of input utilities required by each recycling technique.
- 2. Average Unit Cost per mass unit of waste (UCW): for Non-Recovery techniques, this indicator corresponds to the total fees charged by government or the concerned industry; for Recovery pathways, this indicator is the breakeven point charged to an amount of waste through a 10-year horizon time of recycling plant for Recovery pathways. lt corresponds classically to a zero value of Net Present Value (NPV) of the project calculated by Eq. (3.1) with a discount rate ( $\beta$ ) of 10%. This assumption mays be considered as severe but may prevent from economic difficulties that may be encountered from deployment to mature sales if the size of the market is not as large as expected (Yang et al., 2012).
- 3. Average Unit Cost per mass unit of recovered fibre (UCF) is

#### **Table 2 Economie indicators.**



computed on a different basis. lt only concerns Recovery Pathways, which is the average cost of recovered fibre during 10-year horizon time so that recycling plants can cover ail their manufacturing cost and begin to have profit.

The profit from by-products (filler, oligomers) is not considered in total revenue to estimate the values of UCW and UCF of Recovery pathways to avoid the interference on fibre recycling. These two indicators reflect the different economic viewpoint of the involved stakeholder from waste owners with UCW to clients of recycled fibre with UCF.

$$
NPV = -INV + \sum_{i=1}^{10} \frac{(REV - TC) \times (1 - a) + D}{(1 + \beta)^i}
$$
  
with NPV: Net Present Value (3.1)

H: the horizon time of recycling plant (10 years)

t: the year index a: tax rate (34%)  $\beta$ : discount rate (10%) INV: Investment cost REV: Annual Revenue of process D: Depreciation  $(D = \frac{INV}{H})$ 

TC: Total annual costs 
$$
(TC = D + \sum_{i=1}^{8} Cost_i)
$$

*3.2.2. Environmental assessment methods* 

Besides the impacts released from operation activities, the impacts related to plant construction have been considered as insignificant compared to the operating phase: this assumption has been considered for valid for a lot of chemical processes (Morales Mendoza, 2013). The benefits obtained from recovered products have of course been included in environmental assessment with the avoided impacts. Three indicators involving GWP are computed:

- 1. GWP impact of process (GWPP) encompasses ail the activities of waste management
- 2. GWP impact of substituted products (GWPA) includes the GWP impacts from the utilisation of recovered products to replace virgin materials. In this study, a quantity of recovered products is assumed to replace the equivalent quantity of virgin materials (1:1 ratio). This assumption is proposed in order not to limit the applications of recovered fibre by mechanical properties as proposed in Witik et al.  $(2013)$ . The GWPA for an amount of recovered products is therefore equal to GWP impacts of production of the same quantity of virgin products which the recovered products replace;
- 3. Finally, GWP total of the system (GWPTOT) which take into account impacts from both activities and substitution effect: *GWPTOT* = *GWPP* - *GWPA*



Fig. 2. Materials flows in the studied system (Hedlund-Åström, 2005; Suzuki and Takahashi, 2005; Palmer et al., 2010; Akesson et al., 2013; Knight, 2013; Witik et al., 2013; Howarth **et al., 2014).** 

# *3.3.* Mode! *data*

Ali the studied technologies are assumed to be available to treat CFRP waste. The mass and energy balances of each pathway in the modelled system are summarised in Fig. 2 and Table 1. Based on literature and on Ecoinvent datatbase v2.2 with ReCiPe impact assessment method implemented in SimaPro v7.3, model data have been collected for being used in economic assessment and environmental assessment. The involved variables, their numerical value, and the source they come from can be found in Fig. 2 and Tables 3 and 4. The typical features of the studied waste treatment techniques will be shortly presented in the two following subsections.

# *3.3.1. Non-Recovery pathways*

3.3.1.1. Landfill. Landfill can be defined as a specific underground storage of waste when there is no available recycling technique for this kind of waste. In this study, landfilling is considered as a disposai pathway, not as a kind of storage. Therefore, once landfilled, the potential recovered products from waste are lost. The composite waste that is likely to be landfilled is considered as non-hazardous solid waste.

No specific process for composite landfilling is defined in Simapro

v.7.3 databases, e.g. Ecoinvent 2.2. The landfilling of plastics mixture in sanitary landfill process, which is the closest option to composite landfilling solutions regarding the similar organic chemical nature of polymeric composite and plastics, has been adopted in order to evaluate GWPP of CFRP waste landfilling. The impacts from losing the recyclable fibre in CFRP waste are considered in order to avoid neglecting the lost potential in landfilling. These lost impacts are evaluated at negative GWPA of production for the equivalent quantity of VCF as the quantity of carbon fibre presented in landfilled CFRP waste. According to GPIC et al. (2003), the fees of composite landfill are around 76-90  $\epsilon$ /tonne. The same order of magnitude for landfill charge in France in 2015, i.e., 95 €/tonne has been found in Fischer et al. (2012). This value is used in this study for economic assessment.

*3.3.1.2. lncineration.* Incineration is a thermal process, which allows recovering energy in heat resulting of waste combustion. Heat can be used either directly or converted into electricity. In this scenario, the process is assumed to be auto-thermal; heat and ash by-product released from the process are estimated at  $32 \text{ MJ}$  and  $8 \text{ wt\%}$  of input waste respectively according to Witik et al. (2013); the emission of combustion is based on the work of Hedlund-Åström (2005). Heat is

#### **Table 3 Data of Unit Cost and GWP impact in the modelled system.**



then converted to electricity with an efficiency of 35% (Antonini, 2012). Ash by-product is landfilled as an inert waste. The cost of general waste incineration is about 92€/tonne in France in 2015 according to Fischer et al. (2012). The UCW of this route includes this charge as well as the cost of ash landfilling.

*3.3.1.3. Co-incineration.* As incineration and co-incineration are both based on combustion of waste, the quantity of heat and ash produced in co-incineration is assumed to be similar to the respective value involved in incineration technique. However, co-incineration allows material recovery in addition to energy recovery. Indeed, in co-incineration, waste is used as a substituted fuel involved in clinker fabrication where coal is normally used as a fuel and the products of waste combustion, i.e. heat and ash, are completely valorised in co-incineration. Heat released from combustion of CFRP waste can substitute the same amount of heat from coal combustion in fumace. Ash is also mixed with the raw materials of clinker. According to Halliwell (2006), the cost of treatment of co-incineration of composite waste charged by the cement industry is around  $1 \in \ell$ , This cost is considered as UCW for this technique.

# *3.3.2. Recovery pathways*

The techniques that have been investigated here have been selected as they are representative of the existing processes: grinding, pyrolysis, microwave, and supercritical water (SCW). These techniques have attracted a lot of attention from academic and industry and have reached a sufficient level of maturity of development. Grinding process is the simplest recycling technique with only energy requirement but the recovered products cannot be used in high-valued applications due to the strong degradation of recovered fibre and unclear separation of fibrematrix. Pyrolysis which is the most successful industrialised technique for clean CF recycling with high retention of mechanical properties yet requires high energy consumption. Microwave, another thermal technique, can recycle CF with Jess energy than pyrolysis and lead to potential recovery of matrix. SCW is the recycling technique in trend because of the utilisation of water, a cheap and low-hazardous risk raw material compared to organic solvents, but this technique requires a large amount of energy to operate at supercritical conditions.

Although the recycling yield of carbon fibre in CFRP waste has not reached 100%, the recent results obtained are promising (Oliveux et al., 2015a). In this study, we consider that CF can be ideally recycled at 100% yield by pyrolysis, microwave and SCW to study the maximum benefit that can be potentially obtained without introducing a bias in the analysis since the recycling yield of CF may vary in different works.

For CFRP based on bisphenol A epoxy resin, the residuals are constituted of phenol derivatives principally. Due to the complexity of oligomers mixture, the residuals from decomposition of matrix are simplified to be reused as phenol in this study.

Technical, economic and environmental data have been collected regarding CFRP applications. Yet, in case of the Jack of data, those relative to GFRP will be used. The majority of recycling techniques on fibre recovery from FRP waste have been developed for both GFRP and CFRP because of the similarity of these two polymeric composites, e.g. (Kennerley et al., 1998; Pickering et al., 2000; Yip et al., 2001; Jiang et al., 2008) for fluidised bed, or (Lester et al., 2004; Akesson et al., 2013; Obunai et al., 2015) for microwave, etc.

The general reviews on global composite recycling have shown show that there are still few recycling sites for FRP/CFRP waste, with



**Data of lnvestment Cost for Recovery Pathways.** 





**Fig. 3. Economie assessment of the studied pathways.** 

little available information (capacity, technique, location). The recycling capacity of the studied techniques is set at the best performance of the current FRP/CFRP recycling industry reported in Pimenta and Pinho (2011): 4000 t/year for grinding, 2000 t/year for thermal recycling (pyrolysis and microwave) and 1000 t/year assumed for chemical recycling (SCW).

*3.3.2.1. Grinding.* The principle of this technique is to separate fibres from matrix by a grinding process. After mechanical process and sieving, the obtained products are constituted by a mixture of matrix and fibre. They are separated into different fractions in function of fibre proportion and length (Kouparitsas et al., 2002; Palmer et al., 2010). Palmer et al. (2009) have shown that two products are assumed to be recovered from the composite waste, i.e., a matrix-rich powder product (29 wt%), used as filler, and a fibrous fraction (71 wt%). The process energy is estimated at 0.27 MJ/kg by Hedlund-Âstrôm (2005) which is in agreement with the value proposed by Howarth et al. (2014) carried out at industrial scale.

In this work, the mechanical technique is based on ERCOM process which operates at industrial scale by using a mobile shredder and hammer mill. The plant has a capacity of 4000 t/year with a mobile shredder of value of 200,000€ (Halliwell, 2006). The capital cost of hammer mill is presented in detail and has been assumed to be one third of the value of shredder (Schutte Buffalo Hammermill, 2016).

*3.3.2.2. Pyrolysis.* In this study, the pyrolysis is modelled as a combustion process of the matrix (35 wt% of CFRP waste) environmental impacts. No energy recovery from thermal decomposition of matrix has been assumed. The total energy used in pyrolysis has been estimated at about 30 MJ/kg composite (Witik et al., 2013).

In general, pyrolysis for composite recycling requires a minimum amount of 10 million € for capacities ranging from 20,000 to 80,000 t/ year (Krawczak, 2012). An average value (50,000 t/year) has been assumed and used to estimate the corresponding capital cost of the studied capacity by six-tenths rule.

*3.3.2.3. Microwave.* The process energy is estimated at 10 MJ/kg according to Lester et al. (2004) and Suzuki and Takahashi (2005). According to Lester et al. (2004), oligomers from the decomposition of polymeric matrix can be obtained by this technique. Another study on GFRP (Akesson et al., 2013) has shown that besides the recovery of solid product, i.e. glass fibre, the thermoset matrix (unsaturated polyester resin) is decomposed into pyrolysis oil and gas with 56 wt% and 44 wt% of quantity of matrix in waste respectively. These yields

will be used to estimate the quantity of oligomers and the emission of  $CO<sub>2</sub>$  released from 35 wt% of matrix in the studied CFRP waste through this process. The pyrolysis oil, which is composed of various aromatic substances, is assimilated to phenol in this model. The gas fraction which is composed of a rich amount of CO and  $CO<sub>2</sub>$  with low presence of methane and other hydrocarbons reported in the study of Akesson et al.  $(2013)$  is assumed to be exclusively composed of  $CO<sub>2</sub>$  considering a total oxidation.

No information of investment cost on FRP recycling is yet available. This later is estimated based on the BRC process for tyres scrap treatment (Appleton et al., 2005), that is 9,400,000  $\pounds$  for a capacity of 50 000 t/year. The investment cost of the BRC process reported in 1990s is updated from 1995 to 2014 by Chemical Engineering Plant Cost Index (CEPCI).

*3.3.2.4. Supercritical water.* In supercritical condition (temperature > 374 °C and pressure  $> 221$  bar), the polymer matrix can be decomposed into different oligomers and the carbon fibre is recovered in supercritical water. This technique has been industrialised for hazardous waste treatment since 1980s (Marrone, 2013). For composite application, although it has received a lot of attention from academics and industry (Oliveux et al., 2015a), supercritical water for CFRP waste is still at pilot scale. As information of this process is still limited, data used for assessment are based on the work of Knight (2013). For an amount of 1 kg of CFRP  $(35 wt\%$  matrix) waste, the process requires 2.61 kWh of electricity, 1.64 m <sup>3</sup>of natural gas, 3.5 kg of pure water for solvent and 72.07 t of cooling water. CFRP waste is assumed to be entirely recovered with 100% yield of carbon fibre and matrix (in the form of oligomers). A capital cost value of 5,770,000\$ for 150 kg/hour of capacity has been adopted from in Knight (2013) (i.e. 1000 t/year plant).

# **4. Results and discussion**

# *4.1. Economie assessment*

Fig. 3 presents the values of OC, UCW and UCF for a11 the studied CFRP waste techniques. Based on UCW indicator, it must be first emphasized that not surprisingly, the fibre recycling techniques are not cost-competitive compared to landfill and incineration routes. These options (requiring around 0.1  $\epsilon$ /kg of waste) are the most competitive ones for CFRP waste treatment without consideration of profits from recoverable products in waste. This indicator may reflect the viewpoint of the waste producer who will be referred as the « waste owner » who may have no economic interest to reuse or stock waste and have to

#### **Table 5**

**Price ranges of carbon fibres and glass fibres in market.** 



select one of the existing techniques in order to remove waste at minimal cost. So, this may suggest that if no regulation is imposed, landfill and incineration will continue to be the dominant economic choice in CFRP waste management at current costs despite there is no mass recovery in these options.

Although it cannot recycle carbon fibre, co-incineration allows waste recovery considering both energy and material aspects, and prevents the use of coal in clinker production. With a charge of  $1 \in \ell$  kg of waste by cernent industry, co-incineration loses its economic interest compared to landfill or incineration and even to other fibre recycling techniques like grinding and microwave, despite its advantage on waste valorisation. However, if this technique is charged at the same fee of incineration, it is more interesting than incineration and landfill because this technique allows reducing the cost of ash landfilling in incineration by material recovery in clinker fabrication. However, the choice of non-recovery techniques is temporary and depends largely on the acceptance of recycled carbon fibre in market. When RCF become profitable, the non-recovery pathways will become obsolete for CFRP waste management.

Grinding that operates with low energy consumption, has the lowest value in Operation Cost in the Recovery pathways. Although the UCW of this technique is little higher than the cost of landfill and incineration, it is the cheapest one compared to the value of UCW for the three other recycling techniques due to simple equipment and high capacity. Pyrolysis and SCW which operate at high temperature or high pressure respectively, exhibit a high Operation Cost. This factor has an influence on the UCW indicator relative to these techniques, especially in SCW technique. It can clearly be observed that SCW leads to both the highest Operation Cost and UCW because of the conjunction of three factors, i.e., high utility cost, high investment and small capacity. Microwave that operates at the same capacity (2000 t/year) is more interesting than conventional pyrolysis regarding its lower Operation Cost and UCW principally due to energy reduction in microwave heating which requires only one third of energy used in pyrolysis. With UCW varying from 0.18 to 3.53€/kg of waste, the Recovery Pathways cannot compete with the Non-Recovery Pathways if there is neither market for recovered fibres nor regulation constraints.

In this context, UCF indicator is used to study the acceptable price range at which recovered fibres can be sold as well as their potential applications that can be determined in order to promote recycling and markets of recovered fibre. For this purpose, the UCF of recovered fibre from the Recovery Pathways will be compared with the average price of VCF, VGF and RGF in current market. This evaluation is pivotai to study the potential use of recovered fibre in classical applications of VCF, VGF and RGF by an economic viewpoint. The price of VCF may vary according to different grades on mechanical properties, precursors and production technique, etc.... from a price less than 20 \$/kg (low modulus) up to 2000 \$/kg (ultra-high modulus) (Chen, 2014). In a context where carbon fibre will be popularised in wide applications such as automotive, the production of carbon fibre from cheap precursor like lignin can reduce the manufacturing cost of CF at around 6.6 \$/kg (5.92 €/kg). According to Berreur et al. (2002), the ideal prices of carbon fibre are estimated at about 4.5-7.5 €/kg. Besides, the price of glass fibre is much lower than that of carbon fibre. The price of glass fibre is estimated at 1-3€/kg for general purpose and 3-30€/kg for high technology applications (Dupupet, 2008), while recovered glass fibre is sold at  $0.25 \cdot \sqrt{\text{kg}}$  (Job, 2013).

UCF for grinding (evaluated at 0.248 €/kg) exhibits a value that is very similar to the price of recovered glass fibre. The value of UCF for recovered fibre from thermal techniques is higher than the minimum price of VGF (1 $\epsilon$ /kg), but remains lower than the lowest price (i.e. 4.5 €/kg) of carbon fibre that is used for general applications (Berreur et al., 2002). Based on the assumptions of this study, the UCF of SCW is estimated at 5.43 €/kg which is the highest cost among the Recovery pathways and exceeds the threshold of  $4.5 \cdot \frac{1}{2}$  for carbon fibre price. Mechanical recycling has the least UCF cost, but carbon fibre cannot be cleanly separated from the matrix and the recovered products are usually used in low value applications. Although SCW has the highest UCF, the recovered fibres by this technique have the tensile strength which is slightly near the one of virgin fibres. This technique needs yet improvement to reduce investment cost and an expansion of capacity is required so that this process becomes more competitive than other recycling techniques such as pyrolysis or microwave.

Based on literature, Table 5 presents the price ranges of carbon fibre and glass fibre with different quality in market. The UCF value estimated in this study is yet lower than the data reported by Oliveux et al. (2015a): 13-19 \$/kg for RCF from thermo-chemical recycling and 5 \$/ kg (3.36€/kg) for ground CFRP. The observed gap can be explained by several factors: (i) the studied system does not consider exogenous factors (type of CFRP waste, transportation, conditioning process, packaging, etc.); (ii) average data and fixed capacity are used. However, the reported cost of recycled carbon fibre seems less attractive compared to the price of VCF from cheap precursors like lignin, i.e., 6.6 \$/kg, (Chen, 2014). It must be emphasized that, the recycled fibre costs have two competitors according to the targeted market, low-cost virgin CFRP for low value use and CFRP for high-value applications requiring carbon fibres of high-quality (e.g. aerospace applications).

Finally, it must be said that although the economic benefit that may result from the by-product release for some specific markets is not considered, the associated environmental benefit is taken into account via the concept of avoided impacts. The key factors from this economic assessment include recycling capacity and carbon fibre recovery that will be assessed in the sensitivity study section.

# *4.2. Environmental assessment*

Three indicators for the evaluation of GWP impacts are used in this assessment: GWPP, GWPA and GWPTOT (see Section 2). The obtained results are displayed in Fig. 4.

The thermal techniques, i.e. pyrolysis, co-incineration and incineration are the pathways that exhibit the highest values for GWPP impacts. The combustion in pyrolysis involves the decomposition of the polymeric part, so that a lower GWPP impact is released than the one resulting from the combustion of the entire composite in incineration and co-incineration. Co-incineration induces slightly lower impacts than incineration because it does not need ash landfilling like incineration. For the other techniques with no or very low GHG emissions, the GWPP impacts depend majorly on the consumption of utilities in the process. Conceming GWPP impacts, the processes can be ranked in increasing order, that is, mechanical recycling, landfill, microwave, SCW. Although microwave and pyrolysis belong to thermal recycling, the recovery of oligomers from matrix in microwave reduces the GWP impacts compared to pyrolysis by avoiding the combustion of



**Fig. 4. Environment assessment of the CFRP waste treatment techniques.** 

# the entire matrix.

GWPA assessment is pivotai to study the outcome of waste treatment activities. If only the GWP impacts of the activities are assessed in the system, the potential benefit from materials recovery by recycling techniques or the Joss of materials in landfill can be under-evaluated. The materials that can be replaced by the recovered products that can be generated by each technique are presented in detail in Section 4. Despite its low GWPP impacts, landfill has high GWPTOT impacts since landfilling activity loses the recycling potential of carbon fibre in CFRP waste. In spite of a higher UCW cost, the interest of co-incineration over incineration is shown through GWPA evaluation. The benefit from recovery of entire CFRP waste on energy and material in co-incineration allows compensating over the GWP impacts produced in the process (GWPP), so that GWPTOT impacts become negative. Yet due to the specific situation of France that is explored in the study, the heat recovery from electricity conversion in incineration is not very profitable towards GWP impacts: the avoided impacts are too low to compensate ail GWPP of this technique since the GWPA impacts of incineration are evaluated from GWP from an electricity mix in France which is produced principally from nuclear power (75%) and others (hydropower - 12%, hard coal - 4%, natural gas - 4% and imported - 2%) (Itten et al., 2012).

The GWPA evaluation of recycling techniques depends strongly on replaced materials. The production of VCF is extremely energy intensive and so emits much higher GHG than the production of glass fibre or of the other recovered products (limestone, phenol). Therefore, the avoided impacts from replacement of VCF by RCF constitute an important contribution of GWPTOT for the studied techniques, which recycle carbon fibre cleanly such as pyrolysis, microwave and supercritical water. The effect of the low-value applications of recovered products from mechanical recycling (glass fibre and limestone) is indeed recognised in the GWPA assessment. This technique is the least interesting option among the recycling pathways despite its low GWPP impacts. The recovery of by-products in addition to carbon fibre constitutes a key advantage for microwave and supercritical water. However, a variant of pyrolysis process equipped with a section for recovery of condensable decomposed polymeric matrix from the incomplete oxidation could exhibit similar GWPTOT performances with microwave and supercritical water.

For ail the studied recycling techniques, the GWPP impact is low enough so that the avoided impact from the recovered products compensates for GWPP impacts and GWPTOT is negative. GWPA impact assessment promotes the implementation of recovery pathways while the market of recycled carbon fibre is not yet mature.

To evaluate the potential benefit of recovered products, ail the studied indicators, i.e., GWPA GWPP, UCW and UCF are complementary indicators in the study of the whole CFRP recycling system from plant deployment to waste recovery.

# *4.3. Sensitivity analysis*

The study results are sensitive to a number of key parameters, including recycling capacity and carbon fibre recovery rate and the material type replaced by recovered fibre through the variation of UCF and GWPTOT. Sensitivity analysis results are presented here.

# *4.3.1. Capacity of recycling techniques*

The economic assessment has highlighted that UCF depends on the installed capacity of the recycling techniques: UCF varies in function of capacity due to waste quantity input and the capital cost. This study is aimed to analyse the impact of this factor on UCF of each technique. Three levels of recycling capacity have been selected, i.e., 1000, 2000 and 4000 t/year that correspond to small, medium and large range of FRP recycling industry.

Not surprisingly, an increase in recycling capacity reduces the UCF of recovered fibre (Fig. 5). The UCF of grinding for three scales (lower than the UCF of other techniques) are all lower than  $1 \in \ell$  and even down to  $0.25 \cdot \frac{1}{2}$  (kg. This result promotes the use of grinding in the classical applications of glass fibres, even in the lowest grade (recovered glass fibre) with a threshold of 0.25  $\epsilon$ /kg. However, the UCF values for pyrolysis, microwave and SCW are all greater than  $0.25 \, \epsilon / kg$ . The recovered fibre from these techniques cannot be reused in the same grade as recycled glass fibre. For the recovered fibre from pyrolysis and microwave, the application range may include at least the substitution of the general purpose grade of glass fibre with their UCF range from 1.6–2.4 €/kg (pyrolysis) and 1–1.9 €/kg (microwave). With a capacity range of 1000- 4000 t/year, the range of UCF of SCW is around of 1-3 €/kg of general purpose glass fibres. The UCF value are 5.4, 4.4 and 3.8 €/kg for 1000, 2000 and 4000 t/year respectively which are lower than the price of VCF from lignin (5.9  $\epsilon$ /kg, (Chen, 2014)). The recovered fibres from this technique are thus competitive with limestone or low grade of glass fibre. Yet some recent studies have highlighted the high retention of properties of carbon fibre that can be obtained by this recycling technique (Oliveux et al., 2015a) so that the reuse of recycled carbon fibres from SCW is promising.

# *4.3.2. Carbon fibre recovery rate*

The impact of carbon fibre recovery rate in recycling techniques is



**Fig. 6. Sensitivity study of Economie Assessment by Carbon Fibre recovery rate.** 



**Fig. 7. Sensitivity study of Environmental Assessment by Carbon Fibre recovery rate.** 

now studied with UCF for economic assessment (Fig. 6) and GWPTOT for environmental assessment (Fig. 7). This parameter will be varied from 10% to 100% in a fixed capacity of 2000 t/year for ail the recovery pathways. In this scenario, the recovered fibre fraction which can be used as carbon fibre applications is characterized by a carbon fibre recovery rate  $(y)$  of total recovered fibre quantity; the remaining part of recovered fibre  $(1-\gamma)$ , which cannot be used as carbon fibre, is

considered to substitute glass fibre. The UCF indicator is evaluated by considering the profit from by-products (filler, oligomers, low-valued fraction of recovered fibre  $(1-\gamma)$ ).

For economic assessment, three ranges of carbon fibre price are determined by the minimum ideal cost that the industry aims to reach, i.e., 4.5 €/kg according to Berreur et al. (2002) and the lowest price of VCF from lignin (the cheapest precursor for carbon fibre) (i.e., 5. 9 €/kg, (Chen, 2014)): 0-4.5 €/kg, 4.5-5.9 €/kg and above 5.9 €/kg. These three ranges are separated by the dotted lines of  $4.5 \in /kg$  and  $5.9 \in /kg$ in Fig. 6. The UCF values in the first range can be viewed as the most competitive prices to substitute virgin carbon fibre by recycled carbon fibre. The second one can be considered as a kind of "safe" price that recycled fibre can be accepted to replace conventional carbon fibre. The recycled carbon fibre with an UCF above the cost of lignin-based carbon fibre (5.9 $\varepsilon$ /kg) may have difficulties to win over this carbon fibre type from an economic viewpoint.

In this sensitivity study, the profits from by-products included in UCF evaluation cannot cover ail the recycling costs.

Logically, an increase in carbon fibre recovery rate reduces the UCF for recovered carbon fibre fraction. Whatever the value of carbon fibre recovery rate, the UCF exhibits the highest value for SCW, followed in decreasing order by pyrolysis, microwave and grinding. This can be explained by high operation cost and investment cost in SCW technique. For low carbon fibre recovery rates (10% and 20%) of SCW, the estimated costs of recycled carbon fibre is higher than the price of the virgin PAN carbon fibre (15.5–19.5 €/kg, (Chen, 2014)). This could suggest to adopt recycled carbon fibre from SCW in carbon fibre market if the carbon fibre recovery rate of this technique reaches around 60% and preferably 80% from which UCF is below 4.5  $\epsilon$ /kg.

In the thermal recycling techniques, the recovery of oligomers allows reducing largely the UCF of microwave, which has moderate operation cost, compared to the UCF of pyrolysis, which does not recover any by-products and requires high energy for operation. Grinding is the most modest technique for which UCF values are always below 4.5€/ kg, from 2.1€/kg to 0.43€/kg at 10% and 100% carbon fibre recovery rate respectively. Even at very low yield of recycled carbon fibre (e.g. 10%), this technique can still offer low prices for utilisation of recycled fibre in carbon fibre applications. For the most expensive techniques, i.e. SCW and pyrolysis, a **high** carbon fibre recovery rate is important to get competitive prices of recycled carbon fibre.

In the assessment of GWP impacts, the GWPTOT values of ail recycling techniques are negative due to the high value of avoided impacts from replacement of virgin materials by recovered products. Furthermore, the high gap in GWP impacts between carbon fibre production (31 kg  $CO_2$  eq./kg, (Das, 2011)) and glass fibre production  $(2.6 \text{ kg } CO<sub>2</sub>/\text{kg }$  (Kellenberger et al., 2007)) promotes yield increase for recycled carbon fibre instead of using recovered fibre for substitution of glass fibre in order to gain important avoided GWP impacts.

Less GWP impact results from pyrolysis among the recovery pathways because of the high energy consumption, the combustion of matrix and the absence of by-products recovery. By contrast, grinding with low energy input has the most significantly reduced GWP impacts, especially at high carbon fibre recovery rates. Although grinding is the most environmental friendly process, the use of fibre fraction at high yield is difficult due to an important degradation of fibre properties through this process. For microwave, the oligomers recovery makes this technique attractive with similar GWPTOT with the low-energy technique, i.e. grinding, at low carbon fibre recovery rates (10% and 20%). However, the oligomers yield released from SCW is higher than from microwave, the avoided impacts of the additional oligomers in SCW compensate for the gap in GWPP between microwave and SCW. From 90% of carbon fibre recovery rate, GWPTOT of SCW is lightly lower than microwave.

# **5. Conclusion**

The objective of this study was to study the potential benefits for CFRP waste management in economic and environmental viewpoints. Multiple pathways are assessed ranging from the options which cannot recover fibre in composites (i.e., landfill, incineration, co-incineration) to the recycling techniques (i.e., grinding, pyrolysis, microwave and supercritical water). In this study, fibre quality is indirectly taken into account through the knowledge of the involved recycling process and the substitution market of the recycled fibre.

The cost and GWP assessments of the modelled pathways show two main trends:

- 1. The Non recovery techniques apart from incineration, i.e. landfill and incineration are the cheapest options but have high GWP impacts due to the Joss or the low value of recovered products.
- 2. The techniques with high yield of recovery require more capital, especially supercritical water, than other pathways, but allow important reduction of GWP impacts by consideration of the avoided impacts.

These results highlight the potential conflicts between economic and environmental indicators as there is no technique having both low cost and GWP impacts.

The economic assessments show highly potential for substitution of VCF /VGF by recycled carbon fibres. The prices of recovered fibres from the recycling techniques are found to be competitive compared to the prices of virgin fibres. However, the reutilisation of RCF in different markets of glass fibres and carbon fibres depend on recycling technologies, plant scale, and recovery rate. Due to the simplicity of the involved process, RCF from grinding can be sold at a low price, about  $1 \in \mathcal{C}$ kg at low capacity (1000 t/year). Even with low substitution rate of carbon fibre (10%) at moderate capacity (2000 t/year), grinding can be competitive (2.1  $\epsilon$ /kg) for carbon fibre market. However, in the advanced recycling technologies, high recycling capacity and high carbon fibre recovery rate are required to overcome both the price of virgin fibre and recycled fibre from cheaper techniques. Indeed, recycled fibres from SCW are not competitive in recycled glass fibre market due to the very high treatment cost (over 3.5€/kg of fibre) even at high capacity of 4000 t/year.

Considering the avoided impacts, GWP assessment clearly promotes recycling activities by recovery of carbon fibre and avoids utilisation of Non recovery routes. This assessment also shows the high interest of recycling over the conventional production of carbon fibre and glass fibre with negative GWP impacts. Yet, waste treatment techniques are complex processes which produce not only GHG emissions but also noise pollution, human toxicity impacts, etc., so that a complete LCA assessment is needed to have a complete cartography of the environmental impacts.

Besides, the CFRP waste streams are composed not only of the cure composite that is considered here but also of the uncured production composite (prepreg) and of the End-of-life waste which may contain metallic inserts or other contaminants. Each waste stream may require specific treatment so that the choice of the technique depends on waste composition and on the market for recovered fibre. The modelling of the whole system embedding ail the different sources for CRFP waste and options for recycling process via (Linear Programming)/MILP (Mixed Integer Linear Programming) formulation is a perspective of the proposed work. The objective is to design a CFRP waste management system which is a good compromise between economic and environmental issues with the variability of waste flows and the different waste treatment techniques.

# **Acknowledgement**

The authors would like to thank the French National Research Agency (ANR) for the financial support of the SEARRCH project (Sustainability Engineering Assessment Research for Recycled Composite with High value) : see the link for more detail, http://www. agence-nationale-recherche.fr/Project-ANR-13-ECOT-0005.

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