EMBODIED ENERGY OF PYROLYSIS AND SOLVOLYSIS PROCESSES FOR RECYCLING CARBON FIBER REINFORCED POLYMER WASTE

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ABSTRACT

Composites are being increasingly used in aerospace, automotive, energy, gas storage, marine, infrastructure, sporting goods, and other secondary industries. The key drivers for composites is light weight, high specific strength, and durability. The composites market is projected to grow from $72.58 billion to $115.43 billion by 2022 (CAGR of 8.13% between 2017-2022) with carbon fiber demand alone projected to soar to 150,000 metric tons by 2020 globally, implying that much of this will reach their End-of-Life (EOL) stage in the coming years. Due to rising disposal cost of landfills and lack of space, it is important to diversify composite waste streams in order to address feasible options related to recovery, reuse, and remanufacture. The Institute for Advanced Composites Manufacturing Innovation (IACMI) has partnered with several industry collaborators to address the issue of carbon fiber reinforced polymer (CFRP) waste. This paper highlights the significance, complexities, application, and the embodied energy (EE) associated with two types of recycling processes for EOL CFRP. The cumulative energy demand (CED) method was applied to analyze the pyrolysis and solvolysis processes using the life cycle assessment (LCA) software SimaPro v.9.0.0.33 and the FRPC Energy Use Estimation Tool developed by the Oak Ridge National Laboratory. Data was sourced to model the amount of fiber, resin, and embodied energy that may be recovered from each recycling system considering 1 kg of carbon fiber-epoxy laminate as benchmark. It was found that a continuous natural gas furnace based pyrolysis system consumes a total of 52 MJ/kg by default and 42 MJ/kg while reusing syngas generated within the system as avoided energy. The supercritical solvolysis process considered for analysis presented a total embodied energy of 257 MJ/kg.

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1. INTRODUCTION

Carbon fiber (CF) composites have the capability of being used in ways that propel positive sustainability implications while considering manufacturing through the environmental, business, and humanitarian lens. For example, the Boeing 787 aircraft weight was significantly reduced by using CF composites, enabling fuel economy and cutting company costs in addition to reduced environmental emissions, ultimately translating to a cleaner supply chain. However, the end of life for these aerospace products still needs to be addressed for downstream processes. One aircraft contains an estimated 20 tons of carbon fiber composites [1]. Previous literature has suggested that landfilling CF composites is an environmentally friendly solution as these materials remain inert for long periods of time [2]. Despite this, landfilling may become an unreliable EOL option for CFRP waste due to several limiting factors due to limited space and rising costs. Current recycling methods available for CFRP available are categorized as: mechanical, pyrolysis, fluidized bed, and chemical [3]. In this study, the pyrolysis (thermal) and solvolysis (chemical) options are investigated.

1.1 Pyrolysis

Recycling CF composites comes with unique challenges when paired with thermoset resins due to their cross-linked molecular structure. Thermosets typically are not able to be reformed for second generation products and are not naturally biodegradable [4]. However, recycling via pyrolysis has emerged to address these challenges. Pyrolysis is the most widely adopted method in the industry for recycling CF composites, and therefore, one of the focus areas of this study. Figure 1 shows the general flow of the pyrolysis process when applied to EOL CFRP composites. Pyrolysis involves heating composites in an environment void of oxygen under an inert medium such as nitrogen at a set temperature range that decomposes the composite matrix into three components: gas, oil, and fibers [5]. A comprehensive LCA is needed to analyze the environmental impacts that rCF can have when replacing vCF. LCA was chosen to create a detailed understanding of the pyrolysis process that will account for the environmental burdens of the process along with implications of energy recovery.

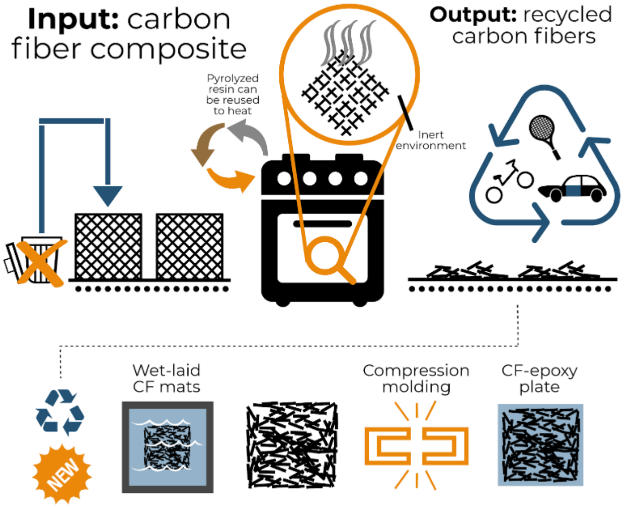

Figure 1. The pyrolysis process when applied to CFRP to promote circularity

1.2 Solvolysis

Solvolysis is a chemical process that uses a solvent to separate the resin and fibers of CFRP waste. Employing sustainable business practices while reducing cost applies to industries using CFRP. An analysis to quantify the embodied energy of 1 kg of CFRP undergoing solvolysis was also conducted. Solvolysis is a chemical process that can either be conducted at low or high temperatures. High temperature solvolysis is also referred to as supercritical solvolysis. This process uses a solvent at its supercritical temperature to separate resin and fiber from CFRP waste [6]. Addition of a catalyst or multiple solvents may be able to decrease operation conditions required [6]. The range of temperatures that supercritical solvolysis is conducted at is 375-650 °C and pressure ranges from 22-35 MPa [7]. Figure 2 displays the supercritical solvolysis route for the given CFRP part. The benefit of using supercritical solvolysis compared to the other methods is that the recovered fibers have similar properties to those of the virgin material [8]. Through the study Knight conducted, it was found that there was 100% retention of the single filament tensile strength and modulus [9]. The purpose of this study is to analyze the embodied energy of a supercritical solvolysis system using the life cycle assessment software SimaPro (© Copyright PRé 2020) to model this process.

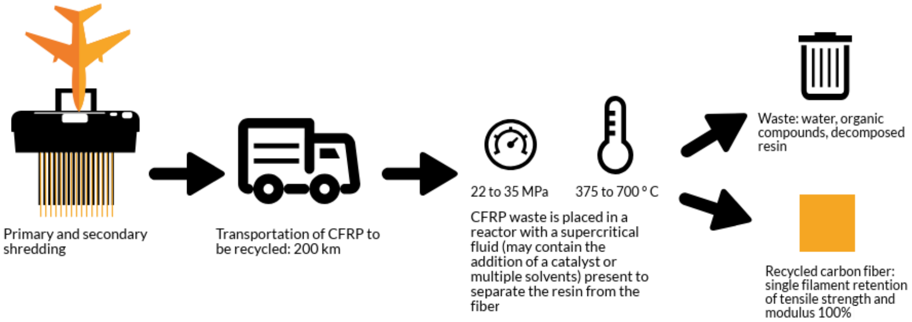

Figure 2. Supercritical solvolysis process considering EOL CFRP parts

2. EXPERIMENTATION

2.1 Materials and Methods

Information was obtained from a combination of scientific literature, databases within SimaPro (Ecoinvent 3 – allocation, cut-off by classification – unit; ELCD; Industry Data 2.0, US-EI 2.2; and USLCI) [10-12], and the FRPC Energy Estimator Tool [13] complemented by extensive industry interviews (company technicians, managers, and researchers) involved in pyrolysis and solvolysis composites recycling industry.

2.1.1 Pyrolysis machinery specification and processing information

Figure 3 represents the culmination of information gathered to create a realistic industry schematic of the pyrolysis furnace starting with the sorting process. Transportation, size reduction of the composite part, and post-processing steps were added to create a complete pyrolysis recycling system. This process was then used as the baseline for SimaPro calculations. Machinery specifications were obtained from scientific articles or directly from the manufacturers. Steps considered in the LCA are as follows; primary shredding, secondary shredding, sorting, furnace, scrubber, hydrocarbon cracker, combing, precision cutting, and transportation.

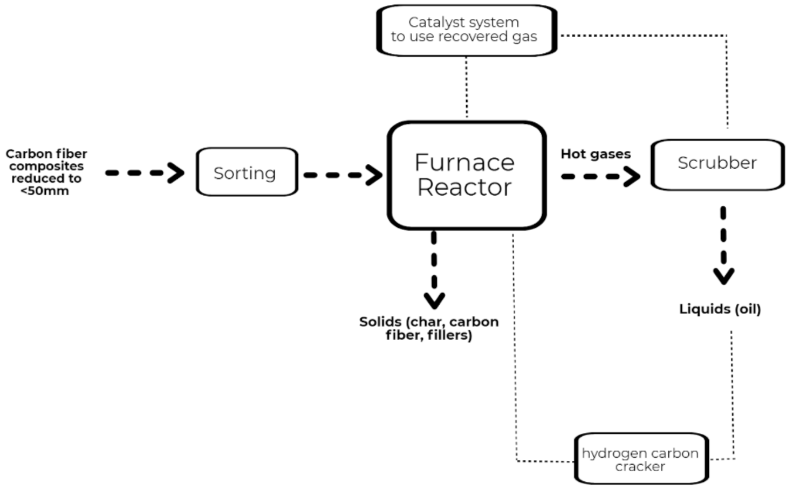

Figure 3. Pyrolysis furnace system boundaries

A 50:50 CF-epoxy composite was taken as the representative specimen for the pyrolysis process. The processes used to make the virgin CF-epoxy composite include total transportation after raw material extraction (as transportation for this life stage is accounted for in the material's embodied energy values), which was assumed to be 200 km, and manufacturing process involving wet-layup followed by compression molding.

2.1.2 Pyrolysis process estimations and assumptions

A number of general assumptions were made due to the proprietary nature of some information relating to the pyrolysis furnace design and company innovations.

1. The furnace was assumed to be a continuous design capable of processing 3800 lb every 1.25 hours, run at a frequency that recycles 1200 metric tons of CF per year.
2. Epoxy breaks down into oil and gas in the furnace, with exact ratios taken from a laboratory study that used a small static bed reactor running at 600 °C. These were then extrapolated to industry scale [14].
3. Environmental impacts of individual gases formed from the epoxy breakdown were not considered due to their containment and reuse within the system.
4. Regarding post-processing of recycled CF, combing and carding were calculated based on industry information considering a throughput of 3000 lb/hr.
5. Scrubber values were based on a standard cyclone design recommended by the Environmental Protection Agency [15, 16].
6. For furnace systems that had the ability to reuse oil, data from hydrocrackers were taken, however any catalytic system with the ability to break apart oil molecules could theoretically be used.
7. In order to obtain syngas densities, temperatures taken for individual chemical densities ranged from 500-600 °C.

It was understood that the pyrolysis process energy would consume a substantial amount of energy as a result of being a large continuous process. However, the syngas that is created within the furnace may be reused within the same process in the place of natural gas as fuel to run the same system as avoided energy. These scenarios are found to be most prevalent within the industry currently and have been considered for analysis.

2.1.3 Solvolysis solvent and equipment selection

A variety of solvents may be used during solvolysis of CFRP. However, the scenario chosen for analysis follows the work of Knight where 150 kg of CFRP was analyzed with 700 kg of water as the solvent [9]. This scenario represents a functional unit of 1 kg CFRP with a 50:50 ratio of carbon fiber to epoxy. Throughout literature review, power ratings of solvolysis boilers and reactors were not provided for the desired industrial scale. Therefore, a high pressure reactor that could maintain the required temperature and pressure of supercritical water and a volume of 2 m^3 was chosen [17]. These properties are based on the work of Knight where an industrial sized reactor was defined as 1.93 m^3 [9]. An average distance of 200 km was considered for transportation of the CFRP waste. Some studies state that the solvent after solvolysis may be reused [18]. However, was not considered in this study as the pathway of such processes were not clearly specified.

3. RESULTS AND DISCUSSION

3.1 Life Cycle Assessment

Embodied energy is the total amount of energy that is found within the supply chain of a given product, process or service [19, 20]. A Life Cycle Assessment (LCA) is the concept that caters to all of the environmental impacts associated with a certain product or process [21, 22]. The Cumulative Energy Demand (CED) method [23, 24] was the method that was used to calculate the embodied energy of both pyrolysis and solvolysis processes for a given functional unit of 1 kg CFRP scrap adhering to the ISO 14040 and 14044 standards [25, 26]. Embodied energy was calculated using the LCA software SimaPro [10] with some values referenced from the FRPC Energy Estimator Tool, developed by the Oak Ridge National Laboratory [13]. The goal of the study was to perform a gate-to-gate embodied energy analysis considering both the pyrolysis and solvolysis recycling technologies. Medium voltage US electricity grid dataset from the SimaPro US-EI library DATASMART, provided by Long Trail Sustainability, 2015 [11] was considered for modeling.

3.1.1 Pyrolysis process impacts

In regards to the pyrolysis model, the most energy intensive step in the pyrolysis process was found to be that of the furnace, with transportation, size reduction, and post-processing steps accounting for a total of only 1.168 MJ/kg. To calculate the EE of the pyrolysis furnace system, values for sorting, furnace, and scrubber, were combined from Table 1. A sensitivity analysis was run based on the ability of the furnace to reuse gas to offset energy needed to heat the system. The most advanced pyrolysis process has the ability to avoid energy consumption by using the syngas *and oil* produced to power the same system. However, this case is not considered for the study. The most energy intensive scenario was the default scenario for the pyrolysis furnace where neither gas nor oil were reused to power the process with a furnace EE of 51.02 MJ/kg (Table 1). The most common system in industry today is a pyrolysis furnace with the capacity to only capture and reuse the syngas (excluding oil) created to power the same furnace. This type of system considering an energy offset (as avoided electricity) resulted in furnace EE of 41.32 MJ/kg (Table 2).

Table 1. Embodied energy per process step within a natural gas based furnace pyrolysis process. Default scenario (without avoided energy consideration) results reflect a total EE of 52 MJ/kg

Impact category	Unit	Step 1: Primary Shredding	Step 2: Secondary Shredding	Step 3: Sorting	Step 4B: Furnace Continuous-Natural Gas	Step 5: Scrubber w/o avoided product	Step 6: Hydrocracker w/o avoided product	Step 7: Combing	Step 8: Precision cutting	Transport
Total	MJ	1.65E-01	4.36E-01	2.69E-01	5.06E+01	7.01E-07	1.92E-01	6.48E-05	4.35E-05	5.67E-01
Non renewable, fossil	MJ	1.14E-01	3.02E-01	1.87E-01	4.73E+01	4.85E-07	1.33E-01	4.48E-05	3.01E-05	5.63E-01
Non-renewable, nuclear	MJ	4.44E-02	1.17E-01	7.25E-02	2.84E+00	1.89E-07	5.16E-02	1.74E-05	1.17E-05	3.60E-03
Non-renewable, biomass	MJ	1.26E-09	3.34E-09	2.07E-09	9.00E-08	5.38E-15	1.47E-09	4.96E-13	3.33E-13	1.30E-07
Renewable, biomass	MJ	1.83E-04	4.83E-04	2.98E-04	7.17E-02	7.76E-10	2.12E-04	7.17E-08	4.81E-08	9.29E-05
Renewable, wind, solar, geothermal	MJ	2.90E-03	7.68E-03	4.74E-03	9.79E-02	1.23E-08	3.38E-03	1.14E-06	7.65E-07	1.24E-04
Renewable, water	MJ	3.32E-03	8.78E-03	5.42E-03	2.51E-01	1.41E-08	3.86E-03	1.30E-06	8.75E-07	3.19E-04
		Size reduction		Pyrolysis				Post-processing		Transportation
		0.601		51.019				0.0001		0.567

Table 2. Embodied energy per process step within a natural gas based furnace pyrolysis process. When considering avoided electricity from syngas reuse, the total EE is 42 MJ/kg

Impact category	Unit	Step 1: Primary Shredding	Step 2: Secondary Shredding	Step 3: Sorting	Step 4B: Furnace Continuous-Natural Gas	Step 5: Scrubber	Step 6: Hydrocracker w/o avoided product	Step 7: Combing	Step 8: Precision cutting	Transport
Total	MJ	1.65E-01	4.36E-01	2.69E-01	5.06E+01	-9.70E+00	1.92E-01	6.48E-05	4.35E-05	5.67E-01
Non renewable, fossil	MJ	1.14E-01	3.02E-01	1.87E-01	4.73E+01	-6.71E+00	1.33E-01	4.48E-05	3.01E-05	5.63E-01
Non-renewable, nuclear	MJ	4.44E-02	1.17E-01	7.25E-02	2.84E+00	-2.61E+00	5.16E-02	1.74E-05	1.17E-05	3.60E-03
Non-renewable, biomass	MJ	1.26E-09	3.34E-09	2.07E-09	9.00E-08	-7.43E-08	1.47E-09	4.96E-13	3.33E-13	1.30E-07
Renewable, biomass	MJ	1.83E-04	4.83E-04	2.98E-04	7.17E-02	-1.07E-02	2.12E-04	7.17E-08	4.81E-08	9.29E-05
Renewable, wind, solar, geothermal	MJ	2.90E-03	7.68E-03	4.74E-03	9.79E-02	-1.71E-01	3.38E-03	1.14E-06	7.65E-07	1.24E-04
Renewable, water	MJ	3.32E-03	8.78E-03	5.42E-03	2.51E-01	-1.95E-01	3.86E-03	1.30E-06	8.75E-07	3.19E-04
		Size reduction		Pyrolysis				Post-processing		Transportation
		0.601		41.323				0.0001		0.567

3.1.2 Solvolysis process impacts

Table 3 outlines the embodied energy during every step of the solvolysis process including the energy required to achieve supercritical condition, to power the reactor and amount of deionized water that is used within the system. The values for size reduction, solvolysis, post-processing and transportation values were kept constant as per the pyrolysis process. Values for reuse were not modeled as this is not clear from published literature. It is seen that within the solvolysis process, the maximum amount of energy is consumed at the stage where supercritical water needs to be reached, with EE of 255.16 MJ/kg.

Table 3. Embodied energy results of the supercritical solvolysis process. Total EE associated with the solvolysis process of 1 kg CFRP using supercritical water is 257 MJ/kg

Impact category	**Unit**	**Step 1 -Primary Shredding**	**Step 2 - Secondary Shredding**	**Step 3 - Achieving supercritical condition**	**Step 4 - Reactor**	**Step 5D - Water (H20)**	**Step 6 - Combing**	**Step 7 - Precision cutting**	**Transportation**
Total	MJ	0.16	0.44	255.16	1.04	0.02	0.00	0.00	0.57
Non renewable, fossil	MJ	1.14E-01	3.02E-01	2.29E+02	7.22E-01	1.32E-02	4.48E-05	3.01E-05	5.63E-01
Non-renewable, nuclear	MJ	4.44E-02	1.17E-01	2.31E+01	2.81E-01	5.13E-03	1.74E-05	1.17E-05	3.60E-03
Non-renewable, biomass	MJ	1.26E-09	3.34E-09	6.68E-07	8.00E-09	4.51E-10	4.96E-13	3.33E-13	1.30E-07
Renewable, biomass	MJ	1.83E-04	4.83E-04	1.09E-01	1.15E-03	4.40E-04	7.17E-08	4.81E-08	9.29E-05
Renewable, wind, solar, geothermal	MJ	2.90E-03	7.68E-03	1.49E+00	1.84E-02	1.77E-04	1.14E-06	7.65E-07	1.24E-04
Renewable, water	MJ	3.32E-03	8.78E-03	1.73E+00	2.10E-02	4.55E-04	1.30E-06	8.75E-07	3.19E-04
		Size Reduction		Solvolysis			Post-processing		Transportation
		0.601		256.227			0.0001		0.567

4. CONCLUSIONS

Results indicate that in both recycling technology cases, transportation resulted in lower embodied energy (0.567 MJ/kg) compared to both size reduction (0.601 MJ/kg) and post processing (0.0001 MJ/kg) steps (Tables 1, 2, and 3).

4.1 Sustainability of pyrolyzed CF-epoxy composites

Industry is primarily concerned with the amount of energy and cost associated with recycling CF-epoxy composites. The current analysis shows that the most basic pyrolysis system with no reuse capability to offset energy will still have lower EE than that of vCF which has an EE range of 183-286 MJ/kg [27] and more recently some citing 1155-1166 MJ/kg [13]. This difference is even more significant while considering a pyrolysis process utilizing a natural gas furnace with the ability to offset energy demands. According to these results, reusing the same syngas produced during the process has potential to bring the overall EE of the process down by 19%. While considering pricing, the cost of vCF ranges from $33-66/kg depending on varying grades where rCF can be sold for a fraction of that cost based on the scale of the recycling facility ranging from anywhere between $5-15/kg [28]. This fact, along with the energy savings that is seen between vCF and rCF makes for a strong economic and environmental case in terms of recycling CF-epoxy composites via pyrolysis. Due to the extremely high EE of CF as a raw material, the energy required for the pyrolysis process itself is relatively low, resulting in reduced total EE for the recycled product as virgin manufacturing and procurement burdens are removed. Furthermore, if the pyrolysis process is optimized further by reusing both the syngas and oil, the EE of the recycled product could be further reduced.

4.2 Sustainability of CF-epoxy composites undergoing solvolysis

The embodied energy for achieving supercritical conditions is the source of the energy peak with respect to this process. Therefore, ways to improve the efficiency of this reaction or cutting down on the dwell time with the use of catalysts, and reusing solvents that are used for separation may alleviate this stress. Knight found that this process removed up to 99% of a highly cross linked resin with 100% retention of single filament tensile strength and modulus [9]. Supercritical solvolysis as a method for recycling CFRP is still a novel technology. Therefore, more research is required to analyze other environmental impacts associated with the process and how resins and solvents may be reused.

4.3 Closing Remarks

Pyrolysis has proven to be a sustainable pathway for CFRP EOL scrap. Although the process itself has scope for improvement, industry interviews suggest that pyrolysis optimization methods (syngas reuse, oil use, etc.) may become the norm in the coming years. In addition, supporting this sector of the composites industry has the potential to create supply for the ever-increasing demand of carbon fiber. In regards to supercritical solvolysis, using water as a solvent would help recover fibers that possess almost identical properties to those of virgin material. More research is needed to understand other environmental impacts such as greenhouse gas emission, acidification potential, respiratory effects, among others while considering the process, route for resin reuse, and applications for recycled carbon fiber. Future research publishing the effect of catalyst and solvent use in reducing process dwell time would substantially assist in understanding the energy consumption related to this significant reduction in cycle times.

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