Recyclability of Additively Manufactured PEI-Carbon Fiber Composites: Assessment of Environmental Footprint and Mechanical Performance

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Keywords: *Composites; Recycling; Circular Economy; Mechanical Properties; Life Cycle Assessment.*

1. Abstract

The global commitment to achieving carbon neutrality by 2050 places significant constraints on material selection and life-cycle management in the aerospace industry. High-performance thermoplastic composites reinforced with short fibers, such as carbon fiber-reinforced polyetherimide (PEI-CF), offer a high strength-to-weight ratio and sustainability benefits. As large volumes of composite materials reach end-of-life, their revalorization through recycling becomes important within a circular economy framework. However, their degradation during recycling limits effective end-of-life valorization. This research aims to quantify circularity trade-offs by assessing mechanical performance and environmental impacts from virgin material through two cycles of recycling. The study combines experimental characterization with life cycle assessment (LCA) conducted in accordance with ISO14040, using OpenLCA to model raw material extraction, processing, transport, additive manufacturing, recycling, and end-of-life scenarios. Manufacturing and recycling were performed in a closed-loop approach using a high-temperature 3D printer, a plastic shredder, and a filament maker. Filaments of 1.75 mm-diameter were produced using a 3DEVO FM2 with process parameters optimized for PEI-CF. Preliminary LCA results for 1 kg of PEI-CF undergoing the printing process of one cycle of recycling indicate total emission of 20.1 kg CO_2 eq. Raw material (extraction, production, and transportation) contributes to 18.1 kg CO_2 eq. while printing process accounts for 1 kg CO_2 eq., each recycling cycle for 0.5 kg CO_2 eq., and end-of-life transport to landfill and treatment for 0.5 kg CO_2 eq. Recycling represents less than 3% of total emissions, which is negligible and enables a longer material lifetime. However, these results do not consider the potential changes in material properties. If recycling-induced degradation results in a significant decline in mechanical performance, the material may no longer meet the requirements of high-performance structural aerospace applications beyond a limited number of recycling cycles, leading to a practical transition from laboratory-scale closed-loop recycling to industrial downcycling or cascading uses. Mechanical performance for virgin and recycled PEI-CF is analyzed using tensile and Izod impact testing.

Material characterization includes melt flow index measurement, rheology, and differential scanning calorimetry. This integrated approach provides a quantitative basis for assessing material circularity trade-offs and an effective recycling process for thermoplastic composites in aerospace applications.

2. Introduction

Composites materials, consisting of a matrix and reinforcement, are widely used in the aerospace industry due to their high weight-to-strength ratio [1]. As these materials represents majority of aircraft structural weight, their environmental impacts and end-of-life (EoL) management have become a challenge [2]. In the context of global decarbonization targets, the transition toward sustainable aerospace systems requires not only lightweight materials but also effective strategies for managing their full life cycle. High-temperature composites, such as carbon fibre-reinforced polyetherimide (PEI-CF), are promising due to their high glass transition temperature (Tg) and high thermal stability for demanding applications [1]. In parallel, additive manufacturing (AM), particularly material extrusion processes such as fused filament fabrication (FFF), enables the processing of these materials offering reduced material waste and increased design flexibility compared to conventional manufacturing routes[3], [4]. Furthermore, unlike thermosets, thermoplastic matrices can be reprocessed through melting, which opens opportunities for mechanical recycling and closed-loop material strategies [5]. However, repeated processing may induce material degradation, leading to a decline in mechanical performance and limiting their effective reuse, in aerospace applications where strict performance requirements must be maintained; Moving to downcycling in a less demanding industry.

This introduces a critical trade-off between environmental benefits and functional requirements, which remains insufficiently quantified for high-performance composites such as PEI-CF. To address this gap, this study evaluates the circularity potential of PEI-CF through an integrated approach combining experimental characterization and life cycle assessment (LCA). By analyzing both environmental impacts and mechanical performance across a cycle of recycling. The work aims to provide a quantitative basis for assessing the feasibility of closed-loop recycling strategies in high-performance aerospace applications.

3. Literature Review

3.1. LCA of Composites

Global commitments to carbon neutrality and the transition toward a circular economy, integrating life-cycle thinking into material design and manufacturing processes is increasingly recognized as essential [6]. In this context, LCA is a widely adopted methodology for quantifying environmental impact across the life cycle of materials, from raw material extraction to EoL treatment, thereby supporting decision-making toward more sustainable systems.[7], [8]. Composites increasingly considered as promising solutions due to their high performance and potential for recyclability.

While recyclability has been demonstrated for several thermoplastics, including commodity and engineering polymers[5][9] the behavior of high-performance composites under repeated processing remains insufficiently documented. In particular, degradation mechanisms such as polymer chain scission, thermos-oxidative effects, and deterioration of fiber-matrix interfaces can occur across recycling cycles [10].

This lack of knowledge limits the assessment of their true circularity potential. However, despite their suitability for closed-loop recycling, repeated thermal and mechanical processing can induce degradation mechanisms, including polymer chain scission and deterioration of the fiber-matrix interface, ultimately affecting mechanical [10]. This introduces a fundamental trade-off between environmental benefits, such as reduced resource consumption and emissions, and the retention of functional properties required for high-performance applications. LCA has been done for composites and AM studies, such as in the boat industry [11], comparing recycling and conventional methods [12], the automotive industry [13], or for case studies of aircraft component materials [14]. Although LCA studies have highlighted the potential environmental advantages of recycling composite materials, they often neglect the coupled evolution of mechanical properties, which is critical for assessing their true circularity potential.

3.2. Additively Manufactured PEI-CF and Characterization

Kim et al. [15] study demonstrated the feasibility of processing PEI-based composite via FFF, highlighting their thermal stability and suitability for high-temperature aerospace applications. Their study primarily focuses on material formulation and initial performance, without addressing behavior beyond the first processing cycle. A recent work on polyetherketonekethone-polyethermide (PEEK-PEI) blend processed by FFF shows that high-performance composites reinforced with 30% wt.% carbon fibers can achieve a Young's modulus of up to 13GPa for optimized compositions (80/20 PEEK/PEI), along with an increase in glass transition temperature of up to 37 °C [16]. The study also reports the presence of voids ranging from 1 to 50 µm and porosity levels reaching ~19.7%, which can adversely affect mechanical performance. While these results demonstrate the potential of PEEK–PEI blends for high-temperature AM, the analysis remains limited to initial processing and characterization. The effects of repeated processing and recycling on material properties and environmental performance are not addressed. Furthermore, recent reviews on large-format material extrusion AM show that, while the technology enables high deposition rates, reduced costs, and the production of large-scale polymer components, it remains constrained by defects such as porosity, weak interlayer bonding, and anisotropic mechanical behavior [17]. These process-induced limitations directly influence mechanical performance and may be exacerbated under repeated processing. However, their implications for multi-cycle recyclability and integration within life cycle assessment frameworks remain largely unexplored.

4. Method and Results

4.1. High-Temperature Printing and Recycling

In this study, the mechanical recycling of the PEI-CF is investigated through a closed-loop, laboratory-scale approach. Initially, 1.75mm diameter filaments of PEI-CF are processed via FFF using a high-temperature Hylo™ printer manufactured (AON3D). This printer is used to produce impact Izod specimens in accordance with ASTM D256, using a 0.4 mm-diameter nozzle. After mechanical testing, these specimens were shredded using a 3DEVO granulator and subsequently reprocessed into filaments of identical diameter (1.75 mm) using a Filament Maker FM2 machine by 3DEVO. This procedure is aimed to be repeated to simulate successive recycling cycles, ensuring consistent processing conditions across iterations.

To minimize humidity-induced defects and porosity in this amorphous, the PEI-CF is dried under vacuum at 110 °C for at least 24h prior to each processing step (printing and recycling). The FM2 is a single-screw extruder equipped with four independently controlled heating zones. The selected processing temperature is around 320 °C, between the glass transition temperature (Tg) and the fusion temperature, and below the typical extrusion nozzle temperature of PEI-CF in AM, to ensure adequate melt flow while limiting thermal degradation. Due to the inherent brittleness of PEI-CF during reprocessing, particular attention is given to filament handling at the die exit. A heated guiding tube is implemented downstream of the nozzle to maintain the filament temperature during cooling and prevent abrupt thermal gradients. This configuration mitigates thermal shock as the material transitions from 320 °C to room temperature of 25 °C, thereby improving filament integrity and enabling continuous spooling without fracture.

4.2. LCA

A LCA is conducted in accordance with ISO 14040 standards to provide a preliminary evaluation of the environmental impacts associated with the PEI-CF recycling scenario. The goal of the study is to quantify the environmental impacts associated with 1 kg of PEI-CF across its life cycle, from raw extraction to EoL, including one cycle of recycling. The analysis is performed at a laboratory scale using OpenLCA software.

The system boundary encompasses the following stages: raw material extraction and production, transportation to the laboratory, AM (printing), mechanical recycling, and EoL (final disposal). The life cycle inventory (LCI) combines primary and secondary data sources. Primary data are collected directly from the experimental setup, including material consumption, auxiliary inputs, and energy consumption for each processing step (AON3D printer, 3DEVO shredder, and filament maker) was calculated using equipment power rating and active processing times. Secondary data are obtained using the Ecoinvent v3.12 database, and for the material, patent data were used. Four main processes are defined within the system: (i) **Raw extraction production,** including extraction, transformation process, transports, and upstream activities of the supply chain; (ii) **AM**, corresponding to FFF process controlled in this study; (iii) **Recycling**, including shredding and filament re-extrusion of the PEI-CF within the system; (iv) **EoL,** representing the final disposal of the PEI-CF into a plastic treatment and waste facility in Montreal, including its waste transport collection. This structured approach enables a consistent comparison of environmental impacts across the life cycle while linking experimental recycling performance to sustainability metrics. Environmental impacts, including climate change, are calculated using the ReCiPe 2016 Hierarchist (H) method in OpenLCA, expressed in kg CO_2 eq.

4.3. Impact Izod Tests

The impact resistance of the PEI-CF material is evaluated using Izod impact tests on FFF-produced specimens. Virgin material (cycle 0) is tested to establish a baseline for mechanical performance. Specimens are printed under controlled conditions using a nozzle temperature of 390 °C, a PEEK build plate temperature of 210 °C, and a heated chamber of 210 °C to ensure thermal stability during deposition. To assess the influence of printing-induced anisotropy, specimens are printed in two raster orientations: 0° and 90°. For each orientation, at least five specimens are tested to ensure the statistical significance of the results. The pendulum used is 5.5 J. This approach enables the evaluation of directional dependence in impact behavior. At this stage, only cycle 0 has been tested. Impact characterization of recycled material (cycle 1 and subsequent cycles) is planned to quantify the evolution of the repeated heating process and the mechanical performance of recycling process.

5. Results and Discussion

5.1. LCA results and discussion

Table 1. presents the LCA results for the life cycle processing and recycling of 1kg of PEI-CF within the defined system boundaries. These results indicate that the raw material extraction step (i) dominates the contribution to the overall carbon footprint, representing approximatively 90% of total emissions. This is consistent with the high energy demand associated with the production of high-performance thermoplastic composites. In contrast, the recycling process (iii) contributes only a marginal share of the total impact with only 2.5% of the overall emission. The electricity grid mix is the market for medium-voltage electricity of the laboratory, situated in Quebec. As a result, the incorporation of recycling enables the extension of material service life while adding a limited environmental burden. The total emission associated with one cycle of recycling is 20.1 kg CO_2 eq., with an incremental contribution of approximatively 0.5 kg CO_2 eq. per additional cycle, compared to 19.6 kg of CO_2 eq. only with a virgin linear material process. These results highlight the potential of circular economy strategies to reduce the environmental burden per functional use of the material.

Table 1. LCA of 1kg of PEI-CF

Steps	Kg. CO_2 eq.	% of CO_2 emission
(i) Raw extraction	18.1	90
(ii) Printing	1	5
(iii) Recycling	0.5	2.5
(iv) EoL	0.5	2.5
TOTAL	20.1	100

However, these environmental benefits must be evaluated in conjunction with the evolution of material properties.

5.2. Impact Izod results and Discussion

The Izod impact resistance of PEI-CF specimens was evaluated as a function of printing orientation and first-layer processing conditions. Table 2 summarizes the results obtained for 0 ° and 90° raster orientations.

The 0° specimens, printed without a brim and with a first-layer extrusion speed of 30 mm/s, exhibited an impact strength of 5357 J/m. In contrast, the 90° specimens, printed with a brim (2 lines with a reduced first-layer extrusion speed of 15 mm/s) to enhance bed-adhesion and reduce warping to promote better interlayer bonding, showed a higher impact strength of 6123 J/m. Although the speed and brim variables were modified, the combination of improved adhesion and slower deposition in the 90° configuration likely reduced defects at the specimen base. Although additively manufactured composites typically exhibit weaker properties when loaded perpendicular to the filament direction, these processing conditions appear to compensate for this effect, highlighting the strong influence of printing parameters on impact performance as we can see in Figure 1.

Table 2. Impact Izod Results of PEI-CF (cycle 0)

Orienrtation	0°	90°
Brim	no	Yes (2)
First layer extrusion speed (mm/s)	30	15
Izod Impact (J/m)	5357	6123

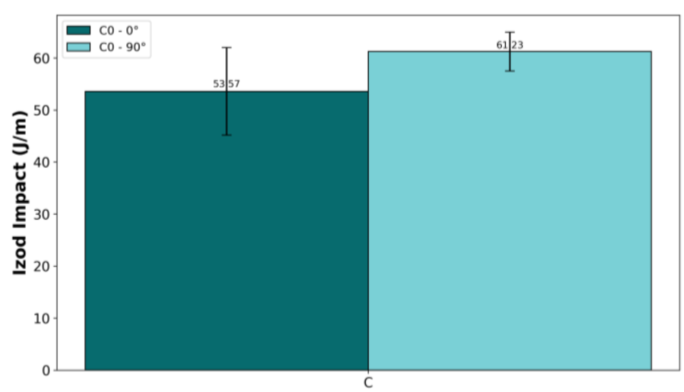

Figure 1. PEI-CF Cycle 0 Izod Impact test Comparison between 0° and 90° raster orientations

The current analysis does not yet account for potential degradation in mechanical performance induced by repeated thermal and mechanical processing. Ongoing work focuses on the characterization of material behavior across multiple recycling cycles using differential scanning calorimetry (DSC), melt flow index (MFI), dynamic mechanical analysis (DMA), and rheological measurements. This combined approach will enable a comprehensive assessment of the trade-off between environmental performance and material functionality, which is critical for evaluating the true circularity potential of PEI-CF in high-performance applications.

6. Conclusion

This study evaluates the circularity potential of PEI-CF through a combined LCA and experimental approach. The results show that raw material production dominates the environmental impact, contributing approximately 90% of total emissions, while recycling adds only ~2.5% per cycle, corresponding to an incremental impact of about 0.5 kg CO_2 eq. to the total emission of 20.1 kg CO_2 eq. These results confirm that circular-economy activities, such as recycling, can extend material lifetimes with a limited additional environmental burden.

From a mechanical perspective, Izod impact results reveal a clear influence of processing conditions, with impact resistance increasing from 5357 J/m (0°) to 6123 J/m (90°) under optimized first-layer parameters (reduced extrusion speed and use of a brim). This highlights the critical role of printing parameters in controlling defect formation and crack initiation, particularly in high-performance thermoplastic composites.

However, the environmental benefits identified through LCA must be balanced against potential degradation of PEI-CF induced by repeated thermal and mechanical processing. The evaluation of mechanical performance across multiple recycling cycles is currently ongoing. Overall, this work emphasizes the need to couple environmental and mechanical assessments to determine the true circularity potential of PEI-CF in high-performance applications.

7. Acknowledgement

The authors acknowledge the financial support from the Natural Sciences and Engineering Research Council of Canada (NSERC Alliance, #ALLRP 581752 – 23) and Prima Quebec (Le Pôle de Recherche et d'innovation en Matériaux avancés du Québec, #R25). The authors would also like to thank our industrial partners, Safran Group and Dyze Design INC., for their financial upport as well as their technical support, specifically from Mr. Alexandre Hatt, Mr. Jean-Sebastien Carrier, Mr. Maxime Darblade and Mr. Lionel Ridosz.

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