Impact of the Manufacturing Processes of Aromatic-Polymer-Based Carbon Fiber on Life Cycle Greenhouse Gas Emissions

Kaito Sakamoto 1, Kotaro Kawajiri 2,*, Hiroaki Hatori 3 and Kiyotaka Tahara 2

Abstract: Carbon fibers (CFs) are promising lightweight materials to reduce vehicle fuel consumption. However, the most widely used polyacrylonitrile (PAN)-based CF production process consumes a considerable amount of energy. A novel production process for CFs from aromatic polymers (APs) is proposed as an alternative. In this study, the greenhouse gas (GHG) emissions from PAN-based CFs, from APs using the classical benzidine method, and from APs using the coupling method on a cradle-to-gate basis, were analyzed. The results indicate that the AP CFs with the classical benzidine method generated 11% fewer GHG emissions compared with the conventional PAN CFs. Emissions were further reduced by 42% using a large-tow production process. As the classical benzidine method for manufacturing CFs from APs uses a monomer synthesized via benzidine, which is carcinogenic, we examined a different synthetic route using the coupling method for monomer synthesis to avoid the benzidine intermediate. The GHG emissions from the AP CFs manufactured by the coupling method showed a 51% increase compared with PAN-based CFs, indicating a trade-off between GHG emissions and carcinogenicity. However, with proper chemical management, the classical method of CF manufacturing from APs via benzidine showed reduced GHG emissions.

Keywords: LCA; carbon fiber; GHG; aromatic polymer

1. Introduction

Reducing greenhouse gas (GHG) emissions generated by the transportation industry is an urgent issue. According to the Intergovernmental Panel on Climate Change, approximately 23% of all GHG emissions originate from road transport [1]. A lightweight design that can reduce automobile weight is currently considered one of the most promising approaches to improve fuel efficiency [2].

Carbon-fiber reinforced polymers (CFRPs) are materials for lightweight design in automobile applications because of their outstanding properties, such as high specific strength and stiffness, low density, high corrosion resistance, and good design flexibility [3]. Although the use of lightweight materials such as CFRPs in automobile bodies can improve fuel efficiency by reducing fuel consumption and tailpipe emissions during driving, lightweight materials may consume more energy and produce more environmental emissions during other life phases than traditional materials such as steel [4,5]. For instance, carbon fibers (CFs), which are necessary for the formation of CFRPs, are not considered environmentally friendly. To produce CFs from polyacrylonitrile (PAN), which is currently the most widely used raw material for CFs, 198–595 MJ/kg of energy is necessary [6], and its environmental burden is estimated to be 24.00 kg-CO\textsubscript{2} eq/kg [7]. Because the GHG intensity of standard steel is 2.41 kg-CO\textsubscript{2} eq/kg [7], using CFRPs in an automotive application requires a significantly greater mileage to justify its usage compared with steel, considering...
its environmental impact over its entire life cycle. However, no study has conclusively proven that the utilization of CFRPs in automobiles reduces overall GHG emissions [8]. Thus, considering the numerous benefits of CFRPs, reducing the environmental impact of CF production, and thereby reducing the lifecycle of the GHG impact of lightweight automobiles comprising CFRPs is an urgent issue.

One possible solution is the use of lignin, which is the world’s most commonly available natural aromatic polymer (AP) and accounts for approximately 30% of the organic carbon in the biosphere [9,10]. Recently, lignin has been recognized as a potential renewable raw material for carbon fibers. It is being evaluated as a replacement for polyacrylonitrile (PAN) in CFs [11,12]. However, ascertaining and mitigating the overall environmental impact of a potential replacement is crucial. In the bioeconomy, the tool often used to compare conventional products and bio-based alternatives is life cycle assessment (LCA) [13]. LCA is a standardized tool used to model the entire life cycle of a product or system, from resource extraction to final waste management [14,15], and can be used to identify opportunities to mitigate the environmental impact of a product throughout its life cycle [16]. LCA has been applied to assess wood-based alternatives to fuels [17], chemicals [18], and materials [19]. However, the literature on the production or use of AP CFs is limited [20].

AP fibers can transform into CFs without a gas-phase stabilization step in the production process, which is necessary for conventional PAN fibers. Gas-phase stabilization in conventional PAN-fiber processing consumes about 48% of the energy required for the entire process [21]. This is because APs have a high flame resistance performance to avoid melting as the carbonization reaction proceeds. Therefore, CFs can be produced more efficiently using APs than conventional PAN [22]. In addition, the use of APs also facilitates large-tow (LT) conversion. This is because AP fibers can be carbonized with a high carbonization yield, and it is possible to suppress the damage to the fibers owing to the small generation of pyrolysis gas and cavities in the fibers during carbonization.

However, aromatic polymers are typically produced from benzidine, which is carcinogenic. This necessitates the investigation of other production methods that possess lower health risks. In addition, the environmental impact of such a method remains unknown. In this study, we assessed CFs with PBI–BBB (polybenzimidazole–poly (bis-benzimidazobenzophenanthroline)) copolymer fibers as one of the AP CFs in terms of its practical use and the toxicity of its raw materials.

Various efforts have been made to reduce the GHG emissions of CFs. Karp et al. [23] presented a production process of renewable acrylonitrile using 3-hydroxypropionic acid from glucose with an engineered Escherichia coli strain. Kawajiri et al. [24] found that the production of CFs using a flame-resistant precursor of PAN could eliminate the gas-phase stabilization step and reduce GHG emissions. The results showed that flame-resistant fibers and carbonization steps account for a large percentage of GHG emissions. Moretti [25] conducted an extensive literature review of lignin-related LCAs using the Scopus database (www.scopus.com) on 8 July 2020 and identified 62 papers that presented relevant information. Among these, the sole study on LCA for AP CFs was conducted by Das [26], who compared the primary energy and CO$_2$ emissions from CFRPs using CFs derived from conventional textile-based PAN with those of CFRPs derived from renewable-based lignin using two manufacturing methods: the programmable powdered preforming process (P4) and sheet molding compound (SMC). The result showed that CFs derived from lignin resulted in 22% fewer GHG emissions than those derived from conventional PAN fibers. However, past studies have not conducted an LCA of CFs derived from APs as alternatives to benzidine.

The objective of this study was to estimate the cradle-to-gate GHG emissions of the production process of CFs manufactured from an AP (AP CF) and those manufactured from conventional PAN. We evaluated two production processes for AP precursors using the classical method of benzidine and the coupling method. The novelty of this study is as follows: The values for the GHG emissions from the raw materials necessary to form AP CFs (TAB [3,3′,4,4′-tetraaminobiphenyl] and NTCA [1,4,5,8-Naphthalenetetracarboxylic acid])
are not currently available in the IDEA_v2.1.3 database [7], which we use to estimate GHG emissions. Therefore, these values were first estimated using various sources. Based on these values, we estimated the GHG emissions from the AP precursors and AP CFs. Using the aforementioned process, we identified, compared, and assessed the environmental impact of CFs based on PAN, of APs using the classical method with benzidine, and of APs using the coupling method.

2. Materials and Methods

2.1. CF Production Processes

2.1.1. PAN CF Production Process

The PAN CF production process is as follows: PAN-based CFs are manufactured using multistage processes. The starting material for PAN CFs is acrylonitrile. After polymerization of acrylonitrile to obtain PAN, the main processes are spinning, gas-phase stabilization, low-temperature carbonization, high-temperature carbonization, surface treatment, and thermal oxidation. The heating process conditions, such as gas-phase stabilization and carbonization, depend on the expertise of each engineer or scientist, and the heating parameters and time vary in the literature. Gas-phase stabilization generally occurs at temperatures between 200 and 300 °C over tens of minutes, whereas carbonization occurs at temperatures between 1200 °C and 1600 °C over several minutes.

2.1.2. Aromatic Polymer CF Production Process

APs are composed of aromatic amines and carboxylic acids. The PBI–BBB copolymer is composed of TAB (3,3′,4,4′-tetraaminobiphenyl) as an aromatic amine and TPA (terephthalic acid) and NTCA (1,4,5,8-Naphthalenetetracarboxylic acid) as carboxylic acids. There are several types of manufacturing methods in the TAB production process [27]. In this study, we chose the classical benzidine method [28] and the coupling method using granular copper [27]. The classical method using benzidine has good potential to lower GHG emissions because the reaction processes are simple. Benzidine is a carcinogenic substance [29]. Its use is often regulated by law, especially in developed countries. However, this coupling method has low carcinogenic risk. The production processes of TAB, TPA, and NTCA are summarized in the Supporting Information (SI). The production processes of the AP and PAN CFs are shown in Figure 1.

2.2. Functional Unit and System Boundary

The functional unit was 1 kg of CF. We estimated the GHG emissions from the PAN and AP CFs according to the system boundary indicated by the dotted line in Figure 2. As shown in the figure, there were two steps in the CF production process. One is precursor-fiber preparation, and the other is CF production.

2.3. Inventory Analysis

The inventory analysis was performed in three steps. First, the GHG emissions from the raw materials were estimated. The values of the GHG emissions from TPA are listed in the IDEA_v2.1.3 database. For materials that are not listed in the IDEA_v2.1.3 database, these values were estimated by consultation with experts and by referencing patent documentation [27,28,30,31]. Based on this analysis, the GHG emissions from the AP precursors were estimated next. Subsequently, the GHG emissions from the entire CF production processes using the classical method via benzidine and the coupling method were estimated and compared with those from the PAN CFs.
2.2. Functional Unit and System Boundary
The functional unit was 1 kg of CF. We estimated the GHG emissions from the PAN and AP CFs according to the system boundary indicated by the dotted line in Figure 2. As shown in the figure, there were two steps in the CF production process. One is precursor-fiber preparation, and the other is CF production.

2.3. Inventory Analysis
The inventory analysis was performed in three steps. First, the GHG emissions from the raw materials were estimated. The values of the GHG emissions from TPA are listed in the IDEA_v2.1.3 database. For materials that are not listed in the IDEA_v2.1.3 database, these values were estimated by consultation with experts and by referencing patent documentation [27,28,30,31]. Based on this analysis, the GHG emissions from the AP precursors were estimated next. Subsequently, the GHG emissions from the entire CF production processes using the classical method via benzidine and the coupling method were estimated and compared with those from the PAN CFs.

2.4. Assumptions
The following assumptions were made in this study:

• Ratio of reused solvent: 95% [32];
• Ratio of reused catalyst: 95%;
• Carbonization efficiency: PAN CF = 50% and AP CF = 75%;
• Possibility of heat reduction was fixed;
• Power-reduction ratio by LT (large tow): 75% of RT (regular tow);
• Sizes of RT and LT models were 12 k and 48 k, respectively;
• Weight ratio of PBI to PBN was 1:1.

The GHG emissions of the entire CF production process are the sum of the GHG emissions from the precursor-fiber preparation and the CF production. As redundant heat energy was recovered from each CF production process, a process called heat recuperation was added, and the recovered energy was assigned a negative GHG value as a GHG credit. The scale of production was measured in tons per year (TPY). The energy consumption of the production process of CFs was based on Harper’s data [33]. Because the inventory analyses of raw materials and precursor-fiber preparation were based on patent information, lab-scale production was undertaken. In this study, the production scale of the precursor fiber was 2000 kg/batch, and CF production was 1500 TPY. To fill the gap between the lab- and mass-production scales, the scaling factor method was used [34]. Processes other than heating, such as creel and tensioner in Figure 1, were summarized as “others”, and exhaust-gas treatment and heat recovery were also considered in the inventory. Heat recovery was adopted as the fixed value of heat recuperation, regardless of the PAN CFs and AP CFs. These terms appear in the GHG emissions from the CFs in Figure 3.

Figure 1. Production process of PAN and AP CFs.

Figure 2. System boundaries of CF production process.

2.4. Assumptions
The following assumptions were made in this study:
• Ratio of reused solvent: 95% [32];
• Ratio of reused catalyst: 95%;
• Carbonization efficiency: PAN CF = 50% and AP CF = 75%;
• Possibility of heat reduction was fixed;
• Power-reduction ratio by LT (large tow): 75% of RT (regular tow);
• Sizes of RT and LT models were 12 k and 48 k, respectively;
• Weight ratio of PBI to PBN was 1:1.

The GHG emissions of the entire CF production process are the sum of the GHG emissions from the precursor-fiber preparation and the CF production. As redundant heat energy was recovered from each CF production process, a process called heat recuperation was added, and the recovered energy was assigned a negative GHG value as a GHG credit. The scale of production was measured in tons per year (TPY). The energy consumption of the production process of CFs was based on Harper’s data [33]. Because the inventory analyses of raw materials and precursor-fiber preparation were based on patent information, lab-scale production was undertaken. In this study, the production scale of the precursor fiber was 2000 kg/batch, and CF production was 1500 TPY. To fill the gap between the lab- and mass-production scales, the scaling factor method was used [34].

Processes other than heating, such as creel and tensioner in Figure 1, were summarized as “others”, and exhaust-gas treatment and heat recovery were also considered in the inventory. Heat recovery was adopted as the fixed value of heat recuperation, regardless of the PAN CFs and AP CFs. These terms appear in the GHG emissions from the CFs in Figure 3.

Additionally, we considered the conversion of LT to AP CFs. The size of the LT model in this study was assumed to be 48 k, and that of the control model was 12 k (RT). Thus, there was a quadruple input difference between the LT model and the control; therefore, we estimated the energy consumption of LT by comparing a 1500 TPY production scale with a 3000 TPY production scale that uses two tows. In the 3000 TPY/1500 TPY case, the energy consumption of a double weight ratio was calculated; therefore, the value of energy consumption of LT was calculated by squaring the value of the 3000 TPY/1500 TPY case. The GHG emissions from the fiber body were estimated in three steps. First, the amount of waste from the fiber body was calculated by comparing the input weight of the polymer fiber with the output weight of the produced CFs. Second, the amount of carbon contained in the waste was calculated by comparing the amount of waste with the chemical formula of the polymer fiber. Finally, we calculated the amount of CO$_2$, assuming that all carbon determined above was completely burned.

3. Results
3.1. GHG Emissions from the Raw Materials

The GHG emissions from the raw materials (TAB and NTCA) are shown in Figure 4. The difference between the classical method via benzidine and the coupling method comes from the raw materials used in TAB. Therefore, the figure includes TAB with the classical method using benzidine as well as with the coupling method. The details of each stage are provided in the Supporting Information (SI).
Additionally, we considered the conversion of LT to AP CFs. The size of the LT model in this study was assumed to be 48 k, and that of the control model was 12 k (RT). Thus, there was a quadruple input difference between the LT model and the control; therefore, we estimated the energy consumption of LT by comparing a 1500 TPY production scale with a 3000 TPY production scale that uses two tows. In the 3000 TPY/1500 TPY case, the energy consumption of a double weight ratio was calculated; therefore, the value of energy consumption of LT was calculated by squaring the value of the 3000 TPY/1500 TPY case. The GHG emissions from the fiber body were estimated in three steps. First, the

**Figure 3.** GHG emissions from CFs. (a) GHG emissions from CFs by process. (b) GHG emissions from CFs by material.
amount of waste from the fiber body was calculated by comparing the input weight of the polymer fiber with the output weight of the produced CFs. Second, the amount of carbon contained in the waste was calculated by comparing the amount of waste with the chemical formula of the polymer fiber. Finally, we calculated the amount of CO2, assuming that all carbon determined above was completely burned.

3. Results

3.1. GHG Emissions from the Raw Materials

The GHG emissions from the raw materials (TAB and NTCA) are shown in Figure 4. The difference between the classical method via benzidine and the coupling method comes from the raw materials used in TAB. Therefore, the figure includes TAB with the classical method using benzidine as well as with the coupling method. The details of each stage are provided in the Supporting Information (SI).

Figure 4. Cont.
Figure 4. Cont.
Figure 4. GHG emissions from the raw materials by stage and material. (a) GHG emissions from NTCA by stage and material. (b) GHG emissions from TAB by stage and material for classical method with benzidine intermediate. (c) GHG emissions from TAB by stage and material for coupling method.

The GHG emissions from NTCA, from TAB by the classical method, and from TAB by the coupling method were found to be 11.26, 12.15, and 33.23 kg-CO$_2$ eq/kg, respectively. The GHG emissions from TPA were 2.41 kg-CO$_2$ eq/kg from IDEA_v2.1.3. A comparison of the GHG emissions from TPA with the classical and coupling methods shows that the coupling method emitted 2.7 times more GHGs. In the coupling method, 10.02 kg-CO$_2$ eq/kg of p-dichlorobenzene alone was emitted.

3.2. GHG Emissions from the Precursors

The GHG emissions from the AP precursors obtained by the classical method and the coupling method and of the PAN precursors, according to stage and material, are shown in Figure 5.
The GHG emissions from NTCA, from TAB by the classical method, and from TAB by the coupling method were found to be 11.26, 12.15, and 33.23 kg-CO$_2$ eq/kg, respectively. The GHG emissions from TPA were 2.41 kg-CO$_2$ eq/kg from IDEA_v2.1.3. A comparison of the GHG emissions from TPA with the classical and coupling methods shows that the coupling method emitted 2.7 times more GHGs. In the coupling method, 10.02 kg-CO$_2$ eq/kg of p-dichlorobenzene alone was emitted.

3.2. GHG Emissions from the Precursors

The GHG emissions from the AP precursors obtained by the classical method and the coupling method and of the PAN precursors, according to stage and material, are shown in Figure 5.

The GHG emissions from the AP precursors from the classical method, from the AP precursors from the coupling method, and from the PAN precursors were 20.49, 37.63, and 10.08 kg-CO$_2$ eq/kg, respectively. In the precursor production process, AP polymerization occupied 98% in the classical method and 99% in the coupling method. Consequently, the GHG emissions from the AP precursors from the coupling method were larger than those from the classical method because the polymerization process is directly related to the GHG emissions from the raw materials. As the GHG emissions from TAB from the coupling method were 2.7 times larger than those from the classical method, the GHG emissions from the AP precursors followed the same trend.

3.3. GHG Emissions from AP and PAN CFs

The GHG emissions from the CFs using PAN, the APs using the classical method (RT), the APs using the classical method (LT), and the APs using the coupling method are shown in Figure 3. The GHG emissions from the AP CFs by the coupling method were based on RT.

The GHG emissions from the CFs from PAN, from the APs (RT) and APs (LT) from the classical method, and from the APs from the coupling method were 26.94, 23.53, 15.53, and 40.68 kg-CO$_2$ eq/kg, respectively. Compared with the conventional PAN-based CFs, GHG emissions from the AP CFs from the classical method (RT) were 11% lower, and those from the AP CFs from the classical method (LT) were 42% lower. GHG emissions from the AP precursors were 20.49 kg-CO$_2$ eq/kg and those from PAN were 10.08 kg-CO$_2$ eq/kg. However, the GHG emissions from the AP CFs from the CF production process were insignificant. This is because AP CFs do not require gas-phase stabilization during the CF production process, owing to the flame resistance of the AP precursors. For quantitative comparison, it must be noted that the gas stabilization process consumed 11.15 kg-CO$_2$ eq/kg. This property of AP CFs enables the production of LT CFs. The GHG emissions from the AP precursors by the classical method were identical for RT and LT. However, the CF production process of LT was much more efficient than that of RT as the scale effect was in place. The GHG emissions from the AP CFs from the coupling method were 51% higher than those from the PAN CFs. This was due to the large amount of GHG emissions from the AP precursors, which were 37.63 kg-CO$_2$ eq/kg.

Regarding GHG emissions from material, GHG credit from heat recuperation was 10.64 kg-CO$_2$ eq/kg in the PAN, the AP (RT) classical method, and the AP coupling method. That from the AP (LT) classical method was 10.27 kg-CO$_2$ eq/kg. The corresponding value of the AP (LT) classical method was marginally smaller as less energy was used due to better production efficiency. Therefore, less heat was recovered. It must also be noted that the largest GHG contributors in AP CFs and in PAN CFs are the AP precursors and gas-phase stabilization, respectively.
The analysis showed that CFs obtained by the AP classical method, especially in LT, were superior to CFs derived from PAN. However, benzidine used in the classical method is carcinogenic. Thus, it can be assumed that the coupling method, which is free from carcinogenic risk, would be preferable as a production method. However, the
AP CFs produced by the coupling method resulted in 51% higher GHG emissions than the PAN CFs and 162% higher GHG emissions than the AP CFs (LT) from the classical method. However, there are several guidelines against benzidine usage regarding safety protection [35,36]. With this guidance properly integrated into the production process, AP CFs using the classical method can be a promising raw material to achieve the aim of producing lightweight automobiles.

4. Discussion

The results of the analysis in this study were compared with similar ones from previous studies. The studies referenced were Das [26], Kawajiri [24], and Murphy [37]. According to these studies, the GHG emissions from CFs from PAN were 31.00, 29.50, and 30.29 kg-CO$_2$ eq/kg, respectively. These estimates are consistent. GHG emissions from flame-resistant CFs were 21.12 kg-CO$_2$ eq/kg [24]. The GHG emissions from CFs from APs were 24.00 [26], 25.35 from the APs with the classical benzidine method, and 40.46 kg-CO$_2$ eq/kg from the APs with the coupling method. In the absence of a statement regarding the AP CF production details in Das [26], we assumed that the classical benzidine method was used. The results of each analysis are shown in Figure 6. The analysis indicates that the GHG emissions from PAN-based CFs were higher than those from AP CFs using the classical method. The GHG emissions from flame-resistant CFs were lower than those from AP-based CFs. This can be attributed to the difference in GHG emissions during CF-precursor preparation. GHG emissions from flame-resistant CF precursors were 15.69 kg-CO$_2$ eq/kg [24], and those of AP-based CF precursors were 20.49 kg-CO$_2$ eq/kg (classical method) and 37.63 kg-CO$_2$ eq/kg (coupling method). The values of GHG emissions among PAN-based CFs and CFs using the AP classical method were consistent. Our study, as well as the studies by Kawajiri [24] and by Das [26], included GHG credit in the analysis; however, the study by Murphy [37] does not mention the GHG credit. Among the few studies available regarding the GHG emissions from AP CFs using the classical method, our analysis result was consistent with the previous ones. Therefore, it is safe to conclude that we can reduce the GHG emissions from CFs derived from APs by approximately 20% compared with those from CFs derived from PAN. The only exception is CFs based on the AP coupling method. The GHG emissions from CFs produced by the coupling method are high because their precursor preparation emits 37.63 kg-CO$_2$ eq/kg. Our analysis corroborates the findings of Das [26], who mentioned that the precursor production step accounts for approximately 35% of the total energy consumption in carbon-fiber production. Therefore, to reduce the overall GHG emissions caused by the coupling method, a new approach is proposed to reduce the GHG emissions from the precursor fabrication. An additional point of interest is the analysis of the GHG emissions from flame-resistant CFs. However, this is difficult because it remains limited to lab-scale production. Moreover, only one relevant study is available at present; therefore, more analysis is required to draw any conclusions.
Production processes of AP CFs exhibit good potential in reducing GHG emissions from the production process of CFs using AP fibers. A summary of the analysis results is as follows:

1. Production processes of AP CFs exhibit good potential in reducing GHG emissions compared with those from PAN CFs. Furthermore, these processes omit gas-phase stabilization, which is the bottleneck in the conventional production of CFs, thereby improving the performance of carbonization and reducing the GHG emissions from the fiber body by 11%.

2. GHG emissions from APs were identified for the first time by the coupling method. The observed value of 40.46 kg-CO$_2$ eq/kg was approximately 25% higher than that of PAN-based CFs and approximately 40% higher than that of CFs derived from APs using the classical method. Although CFs derived from APs from the coupling method are free from carcinogenic risk, this analysis raises a question regarding the utilization of the coupling method as far as GHG emissions are concerned. The use of APs in the coupling method requires a reduction in GHG emissions during precursor fabrication.

3. The trade-off between GHG emissions and carcinogenic risk was confirmed by comparing the classical and coupling methods in TAB production. One solution is to use the benzidine process under control without leakage of hazardous substances.

4. Our analysis results were consistent with the previous studies of GHG emissions for AP and PAN CFs. GHG emissions from AP CFs were approximately 20% lower than those from PAN CFs. This supports our analysis results. Since there are no data for GHG emissions from the coupling method, we need to wait for a new analysis or until the GHG coefficients of TAB and NTCA are defined in the IDEA_v2.1.3 database.

As shown in the above summary, AP CFs have great potential for reducing the GHG emissions from CFs. In particular, with the proper management of benzidine, a large reduction in GHG emissions is expected.

**Author Contributions:** Formal analysis, K.S.; investigation, K.S.; writing—original draft, K.S.; project administration, K.K.; supervision, K.K., H.H. and K.T.; writing—review & editing, H.H. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was funded by the New Energy and Industrial Technology Development Organization (NEDO) grant number JPNP14014. The APC (Article Publishing Charge) was funded by the National Institute of Advanced Industrial Science and Technology (AIST).
Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: This study is based on the results obtained from a project commissioned by NEDO. We thank Michio Kobayashi for his efforts in improving the quality of the manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

References
1. IPCC. The Intergovernmental Panel on Climate Change, Transport. 2018. Available online: https://www.ipcc.ch/site/assets/uploads/2018/02/ipcc_wg3_ar5_chapter8.pdf (accessed on 8 September 2021).
2. Cui, X.; Zhang, H.; Wang, S.; Zhang, L.; Ko, J. Design of lightweight multi-material automotive bodies using new material performance indices of thin-walled beams for the material selection with crashworthiness consideration. Mater. Des. 2011, 32, 815–821. [CrossRef]
3. Landi, D.; Vita, A.; Germani, M. Interactive optimization of the resin transfer molding using a general-purpose tool: A case study. Int. J. Interact. Des. Manuf. 2019, 14, 295–308. [CrossRef]
4. Liu, Z.; Wang, J.; Zhang, L.; Bao, H. Lifecycle assessment of automotive hoods made of aluminum alloy and glass mat reinforced thermostatic. J. Heifei Univ. Technol. 2012, 35, 433–438.
5. Dubreuil, A.; Bushi, L.; Das, S.; Tharumarajah, A.; Gong, X. A Comparative Life Cycle Assessment of Magnesium Front End Autoparts; Society of American Engineers World Congress: Detroit, MI, USA, 2010.
6. Meng, F.; Mckechnie, J. Towards a circular economy for end-of-life carbon fiber composite materials via fluidised bed process. In Proceedings of the 21st International Conference on Composite Materials, Xi’an, China, 20–25 August 2017.
7. IDEA_v2.1.3 Database. Advanced Industry Science and Technology. 2017. Available online: https://en.aist-riss.jp/ (accessed on 5 August 2020).
8. Kawajiri, K.; Kobayashi, M.; Sakamoto, K. Lightweight materials equal lightweight greenhouse gas emissions?: A historical analysis of greenhouse gases of vehicle material substitution. J. Clean. Prod. 2019, 253, 119805. [CrossRef]
9. Boerjan, W.; Ralph, J.; Baucher, M. Lignin Biosynthesis. Annu. Rev. Plant Biol. 2003, 54, 519–546. [CrossRef] [PubMed]
10. Ragauskas, A.J.; Beckham, G.T.; Biddy, M.J.; Chandra, R.; Chen, F.; Davis, M.F.; Davison, B.H.; Dixon, R.A.; Gilna, P.; Keller, M.; et al. Lignin valorization: Improving lignin processing in the biorefinery. Science 2014, 344, 1246843. [CrossRef] [PubMed]
11. Libre, L. Lignin Based Carbon Fibres for Composites. 2016. Available online: libre2020.eu (accessed on 20 November 2021).
12. GreenLight. GreenLight-Cost Effective Lignin-Based Carbon Fibres for Innovative Light-Weight Applications. 2016. Available online: https://greenlight-project.eu (accessed on 25 November 2021).
13. Giuntoli, J.; Commission, E.; Bulgheroni, C.; Commission, E.; Marelli, L.; Commission, E.; Sala, S.; Commission, E. Brief on the Use of Life Cycle Assessment (LCA) to Evaluate Environmental Impacts of the Bioeconomy. Technical Report. 2019. Available online: https://publications.jrc.ec.europa.eu/repository/handle/JRC109817 (accessed on 1 January 2021).
14. ISO. Environmental management—Life cycle assessment—Requirements and guidelines. Int. Stand. Org. ISO 2006, 14044. Available online: https://scholar.google.com.hk/scholar?hl=en&as_sdt=0%2C5&q=Environmental+management%E2%80%94Life+cycle+assessment%E2%80%94Requirements+and+guidelines.&btnG= (accessed on 1 January 2022).
15. ISO. Environmental Management—Life Cycle Assessment—Principles and Framework. In Proceedings of the Technical Committee ISO/TC, Geneva, Switzerland, 14–15 March 2006; Volume 207.
16. Hermansson, F.; Janssen, M.; Svanström, M. Allocation in life cycle assessment of lignin. Int. J. Life Cycle Assess. 2020, 25, 1620–1632. [CrossRef]
17. Janssen, M.; Xiros, C.; Tillman, A.M. Life cycle impacts of ethanol production from spruce wood chips under high gravity conditions. Bio Technol. Biofuels 2016, 9, 53. [CrossRef] [PubMed]
18. Aryapratama, R.; Janssen, M. Prospective life cycle assessment of bio-based adipic acid production from forest residues. J. Clean. Prod. 2017, 164, 434–443. [CrossRef]
19. Hermansson, F.; Janssen, M.; Gellerstedt, F. Environmental evaluation of Durapulp biocomposite using LCA: Comparison of two different applications. J. For. 2016, 5, 68–76.
20. Janssen, M.; Gustafsson, E.; Echardt, L.; Wallinder, J.; Wolf, J. Life cycle assessment of lignin-based carbon fibers. In Proceedings of the 14th Conference on Sustainable Development of Water, Environment and Environment Systems (SDEWES), Dubrovnik, Croatia, 1–6 October 2019.
21. Maghe, M.; Creighton, C.; Henderson, L.C.; Huson, M.G.; Nunna, S.; Atkiss, S.; Byrne, N.; Fox, B.L. Using ionic liquids to reduce energy consumption for carbon fibre production. J. Mater. Chem. A 2016, 4, 16619–16626. [CrossRef]
22. Irisawa, T.; Hatori, H.; Soneda, Y.; Kodama, M. Polybenzimidazole Carbon Fiber and Method for Manufacturing Same. U.S. Patent US 20,170,152,612 A1, 1 June 2017.
23. Karp, E.M.; Eaton, T.R.; Nogue, V.S.; Vorotnikov, V.; Biddy, M.J.; Tan, E.C.D.; Brandner, D.G.; Cywar, R.M.; Liu, R.; Manker, L.P.; et al. Renewable acrylonitrile production. Science 2017, 358, 1307–1310. [CrossRef] [PubMed]
24. Kawajiri, K.; Sakamoto, K. Environmental impact of carbon fibers fabricated by an innovative manufacturing process on life cycle greenhouse gas emissions. *Sustainability* 2021, *under Review*. [CrossRef]

25. Moretti, C.; Corona, B.; Hoefnagels, R.; Vural-Gürsel, I.; Gosselink, R.; Junginger, M. Review of life cycle assessments of lignin and derived products: Lessons learned. *Sci. Total Environ.* 2021, 770, 144565. [CrossRef] [PubMed]

26. Das, S. Life cycle assessment of carbon fiber-reinforced polymer composites. *Int. J. Life Cycle Assess.* 2011, *16*, 268–282. [CrossRef]

27. Yasuda, H.; Domitrovich, O.V. Method of Manufacturing 3,3′,4,4′-Tetraaminobiphenyl. U.S. Patent US 7,601,873 B2, 13 October 2009.

28. Clyde, O.H.; Wilmington, D.; Benner, R.G. Process for the Manufacture of Benzidine. U.S. Patent US 2,194,938 A, 23 April 1938.

29. United States Environmental Protection Agency. Integrated risk Information System (IRIS). 1987. Available online: https://iris.epa.gov/static/pdfs/0135_summary.pdf (accessed on 1 January 2022).

30. Sato, T.; Ito, I.; Takeda, K. Production Process of 1,4,5,8-Naphthalene Tetracarboxylic Acid. U.S. Patent US 5,354,899 A, 11 October 1994.

31. Bennett, C.W.; Wilson, B.R.; Charles, B.; Hilton, A.J.E.; Kenneth, G. Davenport Method for the Production of 3,3′,4,4′-Tetraaminobiphenyl. U.S. Patent US 5,041,666 A, 20 April 1991.

32. Geisler, G.; Hofstetter, T.B.; Hungerbuhler, K. Production of fine and specialty chemicals: Procedure for the estimation of LCIs. *Int. J. LCA* 2004, *9*, 101–113. [CrossRef]

33. Harper International. Processing advancements within reach for achieving significant reductions in carbon fiber cost of manufacturing. In Proceedings of the JEC Europe 2013, ICS Carbon Conference, Paris, France, 12–14 March 2013.

34. Kawajiri, K.; Goto, T.; Sakurai, S.; Hata, K.; Tahara, K. Development of life cycle assessment of an emerging technology at research and development stage: A case study on single-wall carbon nanotube produced by super growth method. *J. Clean. Prod.* 2020, *255*, 120015. [CrossRef]

35. U.S. Department of Health and Human Services. Preventing Health Hazards from Exposure to Benzidine Congener Dyes. 1983. Available online: https://www.cdc.gov/niosh/docs/83-105/default.html (accessed on 8 August 2021).

36. Sigma-Aldrich. Safety Data Sheet Version 4.6. Safety Data Sheet (t3db.ca). 2014. Available online: https://www.google.com.hk/url?sa=t&rct=j&q=&esrc=s&source=web&cd=&cad=rja&uact=8&ved=2ahUKEwjClr-dosz2AhVSsVYBHziVB3wQFnoECAUQAQ&url=http%3A%2F%2Fwww.t3db.ca%2Fsystem%2Fmds%2Fdata%2F000%2F000%2F000%2F0026.pdf%3F1413587672&usg=AOvVaw1WnNYXE6j_tCSZcJMnLYCA (accessed on 1 January 2022).

37. Murphy, T. The New Face of CAFÉ. *Ward’s Autoworld* 2008, *34*, 36–40. Available online: https://trid.trb.org/view/858441 (accessed on 1 January 2022).