

Supporting Information

Replacing Polyacrylonitrile by Kraft Lignin for Sustainable Carbon Fiber Manufacturing Mitigates Carbon Emission

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1. Environmental impact assessment of waste gases

The most commonly used method to assess environmental impacts in life cycle analysis (LCA) is to combine information about the degree of human activity (referred to as activity data) with coefficients of emissions per unit of activity¹⁻³. These coefficients are called equivalent factors, as given in Table S1.

In this study, we assume that the emission of waste gases is generated from the polyacrylonitrile (PAN) and lignin in carbon fiber production, including the emission of waste gases from thermostabilization and carbonization. To calculate each waste gas from the applied PAN, we defined the conversion rate (CR) of PAN conversion into each waste gas, which are listed in Table S2. In addition, the carbon loss of lignin was counted as the emission of CO₂. According to previous research, a carbon loss of 8% during thermostabilization was assumed⁴. Since as-prepared fibers generally have very low moisture content, the mass loss of carbon fibers can be attributed to the release of carbon in the form of CO₂ without the consideration of moisture releasing the consumption of PAN and lignin are listed in Tables S3-S5. The equations S1-S2 for calculating the waste gases generated for carbon fiber production are as below:

$$m_x = m_{PAN} * (CR_{Thermostabilization_x} + 0.92 * CR_{Carbonization_x}) \quad (\text{Equation S1})$$

$$m_{Lignin\ CO_2} = m_{Lignin} * (1 - yield) \quad (\text{Equation S2})$$

where m_x (kg) is the waste gas as HCN, CO₂, CO, NH₃, and CH₄ generated in carbon fiber production, which is calculated based on the weight of used PAN; $m_{Lignin\ CO_2}$ (kg) is the emission of CO₂ generated in lignin/PAN carbon fiber production, which is calculated based on the application of lignin; m_{PAN} and m_{Lignin} (kg) are the

consumption of PAN and lignin in carbon fiber production (Table S3-S5); $CR_{Thermostabilization_x}$ and $CR_{Carbonization_x}$ are the conversion rate of PAN conversion into the waste gases based on the weight of PAN applied in thermostabilization and carbonization, respectively, as given in Table S2; Carbon fiber yields were based on as-spun fibers, assumed as 59.4% and 51.3% in this study⁴.

Table S1. Environmental impacts of pollutant and equivalent factor.³

Environmental impact	Pollutant	Equivalent factor
GWP	CO ₂	1.00
	CH ₄	25.0
	N ₂ O	298
	NO	296
	SO ₂	1.00
AP	NO _x , NO ₃ ⁻	0.70
	NH ₃	1.88
	HCN	1.19
	P, PO ₄ ³⁻ , HPO ₄ ²⁻ , H ₂ PO ₄ ⁻ , H ₃ PO ₄	1.00
EP	NH ₄ ⁺ , NH ₃	0.14
	NO ₂ , NO _x	0.005
	1,4-DCB	1.00
HTP	CO	0.01
	NO _x , NO ₃ ⁻	1.20
ODP	CFC-11	1.00
	NO _x	0.03
	N ₂ O	0.42

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential; 1,4-DCB, 1,4-dichlorobenzene; CFC-11, chlorofluorocarbons-11.

Table S2. Conversion rate of PAN into the gases during thermostabilization and carbonization.²

Gas	CR _{Thermostabilization} (kg)	CR _{Carbonization} (kg)
HCN	0.03	0.121
CO ₂	0.102	0.068
CO	0.005	0.024
NH ₃	0.001	0.039
CH ₄	0	0.014

Note: CR, Conversion rate.

Table S3. Inputs and outputs for producing 1 kg 12 K lignin/PAN carbon fiber.

	Item	Unit	Value
Inputs	Electricity	kWh	21.79
	Lignin	kg	0.84
	Polyacrylonitrile	kg	0.84
	Nitrogen	kg	0.094
	Methanol	kg	0.089
	Dimethyl sulfoxide	kg	0.92
Outputs	Waste water	kg	717.7
	HCN	kg	0.119
	CO ₂	kg	0.481
	CO	kg	0.023
	NH ₃	kg	0.031
	CH ₄	kg	0.011

Note: Consumption of all inputs were collected from real-life data, and the emission of outputs was evaluated according to the references. The yield of carbon fiber assumed as 59.4%.

Table S4. Inputs and outputs for producing 1 kg 12 K lignin/PAN carbon fiber.

	Item	Unit	Value
Inputs	Electricity	kWh	26.12
	Lignin	kg	0.97
	Polyacrylonitrile	kg	0.97
	Nitrogen	kg	0.109
	Methanol	kg	0.102
	Dimethyl sulfoxide	kg	1.07
Outputs	Water	kg	831.0
	HCN	kg	0.138
	CO ₂	kg	0.635
	CO	kg	0.026
	NH ₃	kg	0.036
	CH ₄	kg	0.013

Note: All inputs and outputs were evaluated according to the references. The yield of carbon fiber assumed as 51.3%.

Table S5. Inputs and outputs for producing 1 kg 12 K PAN carbon fiber.

	Item	Unit	Value
Inputs	Electricity	kWh	21.79
	Polyacrylonitrile	kg	1.68
	Nitrogen	kg	0.094
	Methanol	kg	0.089
	Dimethyl sulfoxide	kg	0.92
Outputs	Water	kg	717.7
	HCN	kg	0.238
	CO ₂	kg	0.277
	CO	kg	0.046
	NH ₃	kg	0.062
	CH ₄	kg	0.022

Note: All inputs and outputs were evaluated according to the references. The yield of carbon fiber assumed as 59.4%.

In this study, the activity data of generated waste gases are shown in Tables S3-S5 and the relevant environmental impact factors and equivalent coefficients are shown in Table S1. GWP, AP, EP and HTP generated by waste gases from PAN were calculated by using the equations S3-S6. GWP generated by waste gases from lignin is calculated by using equation S7. GWP, AP, EP and HTP generated by waste gases from PAN and lignin were calculated by using the equations S8-S11.

$$G_{PAN\ gas} = EG_{CO_2} * m_{CO_2} + EG_{CH_4} * m_{CH_4} \quad (\text{Equation S3})$$

$$A_{PAN\ gas} = EA_{NH_3} * m_{NH_3} + EA_{HCN} * m_{HCN} \quad (\text{Equation S4})$$

$$E_{PAN\ gas} = EE_{NH_3} * m_{NH_3} \quad (\text{Equation S5})$$

$$H_{PAN\ gas} = EH_{CO} * m_{CO} \quad (\text{Equation S6})$$

$$G_{Lignin\ gas} = EG_{CO_2} * m_{Lignin\ CO_2} \quad (\text{Equation S7})$$

$$G_{Gas} = G_{PAN\ gas} + G_{Lignin\ gas} \quad (\text{Equation S8})$$

$$A_{Gas} = A_{PAN\ gas} \quad (\text{Equation S9})$$

$$E_{Gas} = E_{PAN\ gas} \quad (\text{Equation S10})$$

$$H_{Gas} = H_{PAN\ gas} \quad (\text{Equation S11})$$

Where $G_{PAN\ gas}$ (kg CO₂-eq), $A_{PAN\ gas}$ (kg SO₂-eq), $E_{PAN\ gas}$ (kg PO₄-eq), and $H_{PAN\ gas}$ (kg 1,4-DB-eq) are the GWP, AP, EP, and HTP generated by waste gases from PAN applied in thermostabilization and carbonization for carbon fiber production, respectively; $G_{Lignin\ gas}$ (kg CO₂-eq) generated by waste gases from lignin applied in thermostabilization and carbonization for carbon fiber production; G_{Gas} (kg CO₂-eq), A_{Gas} (kg SO₂-eq), E_{Gas} (kg PO₄-eq), and H_{Gas} (kg 1,4-DB-eq) are the GWP, AP, EP, and HTP generated by waste gases from thermostabilization and carbonization for

carbon fiber production, respectively; EG_{CO_2} and EG_{CH_4} (kg CO₂-eq/kg) are the equivalent coefficients of GWP of CO₂ and CH₄, respectively (Table S1); EA_{NH_3} and EA_{HCN} (kg SO₂-eq/kg) are the equivalent coefficients of AP of NH₃ and HCN, respectively (Table S1); EE_{NH_3} (kg PO₄-eq/kg) is the equivalent coefficients of EP of NH₃ (Table S1); EH_{CO} (kg 1,4-DB-eq/kg) is the equivalent coefficients of HTP associated with CO (Table S1); m_{CO_2} , m_{CH_4} , m_{NH_3} , m_{HCN} , and m_{CO} (kg) are the emissions of CO₂, CH₄, NH₃, HCN, and CO produced in carbon fiber production as calculated based on the using of PAN, respectively (Tables S3-S5); $m_{Lignin CO_2}$ (kg) is the emissions of CO₂ produced in carbon fiber production as calculated based on the using of lignin.

2. Environmental impact of PAN replaced by lignin in carbon fiber production

The intensity of GWP, AP, ODP, EP, and HTP generated from the production of 1 kg carbon fiber (CF) were calculated by using the Equations. S12-S16 as below:

$$GWP_{CF} = (\sum C G_R * m_R + G_{gas}) \quad (\text{Equation S12})$$

$$AP_{CF} = (\sum C A_R * m_R + A_{gas}) \quad (\text{Equation S13})$$

$$ODP_{CF} = \sum C O_R * m_R \quad (\text{Equation S14})$$

$$EP_{CF} = (\sum C E_R * m_R + E_{gas}) \quad (\text{Equation S15})$$

$$HTP_{CF} = (\sum C H_R * m_R + H_{gas}) \quad (\text{Equation S16})$$

where GWP_{CF} (kg CO₂-eq), AP_{CF} (kg SO₂-eq), EP_{CF} (kg PO₄-eq), HTP_{CF} (kg 1,4-DB-eq), and ODP_{CF} (kg CFC-11-eq) are the intensity of GWP, AP, EP, HTP, and ODP generated in carbon fiber production, respectively; G_{gas} (kg CO₂-eq), A_{gas} (kg SO₂-eq), E_{gas} (kg PO₄-eq), and H_{gas} (kg 1,4-DB-eq) are the GWP, AP, EP, and HTP generated by waste gases for carbon fiber production, respectively; $C G_R$ (kg CO₂-eq/kg), $C A_R$ (kg SO₂-eq/kg), $C E_R$ (kg PO₄-eq/kg), $C H_R$ (kg 1,4-DB-eq/kg), and $C O_R$ (kg CFC-11-eq/kg) are the characterization factor of inputs and outputs related to GWP, AP, EP, HTP, and ODP, respectively (Table S6); m_R (kg) is the related input and output for 1 kg carbon fiber production (Tables S3-S5);

Table S6. Characterization factors of inputs and outputs used for calculating the environmental impacts of carbon fiber production.

Item	Unit	Acidification	Eutrophication	Human toxicity	Ozone depletion	Climate change	References
Electricity	kWh	4.31E-03	1.11E-03	1.70E-01	3.13E-08	5.30E-01	S5, S6
Lignin	kg	6.62E-03	5.09E-04	4.16E-03	3.92E-08	6.02E-01	S7, S8
Polyacrylonitrile	kg	1.59E-02	4.05E-03	7.45E+00	5.33E-07	5.04E+00	S9, S10
Nitrogen	kg	4.35E-03	1.02E-03	1.21E-02	3.07E-08	5.18E-01	S6
Methanol	kg	6.49E-03	5.48E-04	1.14E-01	1.40E-07	3.81E-01	S6
Dimethyl sulfoxide	kg	5.40E-02	2.00E-03	1.08E+00	1.76E-07	1.27E+00	S11
Waste water	kg	5.20E-06	1.59E-07	2.44E-04	9.55E-11	8.02E-04	S12, S13

Note: The item represents 1 kWh electricity, 1 kg other consumptions or 1 kg waste water.

Table S7. Life cycle impact for the production of 1 kg 12 K lignin/PAN carbon fiber.

Item	AP [kg SO ₂ -eq]	EP [kg PO ₄ -eq]	HTP [kg 1,4-DB-eq]	ODP [kg CFC-11-eq]	GWP [kg CO ₂ -eq]
Electricity	9.40E-02	2.42E-02	3.71E+00	6.82E-07	1.22E+01
Lignin	5.57E-03	4.28E-04	3.50E-03	3.30E-08	5.07E-01
Polyacrylonitrile	1.34E-02	3.41E-03	6.27E+00	4.49E-07	4.24E+00
Nitrogen	4.11E-04	9.64E-05	1.14E-03	2.90E-09	4.90E-02
Methanol	5.76E-04	4.86E-05	1.01E-02	1.24E-08	3.38E-02
Dimethyl sulfoxide	5.01E-02	1.85E-03	1.00E+00	1.63E-07	1.18E+00
Waste gas	2.00E-01	4.22E-03	2.74E-04	0.00E+00	7.51E-01
Waste water	3.73E-03	1.14E-04	1.75E-01	6.85E-08	5.76E-01
Carbon fiber	3.67E-01	3.44E-02	1.12E+01	1.41E-06	1.95E+01

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. Waste gas, represent the emissions of CO₂, CH₄, NH₃, HCN, and CO by the applied polyacrylonitrile and lignin in carbon fiber production. The yield of carbon fiber assumed as 59.4%.

Table S8. Life cycle impact for the production of 1 kg PAN carbon fiber.

Item	AP [kg SO ₂ -eq]	EP [kg PO ₄ -eq]	HTP [kg 1,4-DB-eq]	ODP [kg CFC-11-eq]	GWP [kg CO ₂ -eq]
Electricity	9.40E-02	2.42E-02	3.71E+00	6.82E-07	1.22E+01
Polyacrylonitrile	2.67E-02	6.83E-03	1.25E+01	8.97E-07	8.48E+00
Nitrogen	4.11E-04	9.64E-05	1.14E-03	2.90E-09	4.90E-02
Methanol	5.76E-04	4.86E-05	1.01E-02	1.24E-08	3.38E-02
Dimethyl sulfoxide	5.01E-02	1.85E-03	1.00E+00	1.63E-07	1.18E+00
Waste gas	4.00E-01	8.44E-03	6.96E-04	0.00E+00	8.19E-01
Waste water	3.73E-03	1.14E-04	1.75E-01	6.85E-08	5.76E-01
Carbon fiber	5.75E-01	4.16E-02	1.74E+01	1.83E-06	2.33E+01

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. Waste gas, represent the emissions of CO₂, CH₄, NH₃, HCN, and CO by the applied polyacrylonitrile in carbon fiber production. The yield of carbon fiber assumed as 59.4%.

3. Key contributions of emission for lignin/PAN carbon fiber

Table S9. Contribution analysis from processing of 1 kg lignin/PAN carbon fiber.

Item	AP [kg SO ₂ -eq]	EP [kg PO ₄ -eq]	HTP [kg 1,4-DB-eq]	ODP [kg CFC-11-eq]	GWP [kg CO ₂ -eq]
Dope	8.15E-02	8.91E-03	7.76E+00	7.35E-07	7.46E+00
Spinning	2.97E-02	6.70E-03	1.18E+00	2.65E-07	3.73E+00
Thermostabilization	7.21E-02	1.05E-03	1.59E+00	2.94E-07	5.13E+00
Carbonization	1.87E-01	9.06E-03	7.45E-01	1.39E-07	2.96E+00

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. The yield of carbon fiber assumed as 59.4%.

Table S10. Contribution analysis from production factors of 1 kg lignin/PAN carbon fiber.

		AP	EP	HTP	ODP	GWP
Item		[kg SO ₂ -eq]	[kg PO ₄ -eq]	[kg 1,4-DB-eq]	[kg CFC-11-eq]	[kg CO ₂ -eq]
Inputs	Chemicals	5.10E-02	2.00E-03	1.01E+00	1.78E-07	1.26E+00
	Materials	1.89E-02	3.84E-03	6.27E+00	4.81E-07	4.74E+00
	Electricity	9.39E-02	2.42E-02	3.70E+00	6.82E-07	1.21E+01
Outputs	Waste water	3.73E-03	1.14E-04	1.75E-01	6.85E-08	5.75E-01
	Waste gas	1.99E-01	4.22E-03	2.73E-04	0.00E+00	7.51E-01

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. The yield of carbon fiber assumed as 59.4%.

4. Sensitivity analysis

4.1 Different spinning tows of 6 K, 12 K, and 24 K precursor fibers

Table S11. Life cycle impact for the production of 1 kg 6 K lignin/PAN carbon fiber.

Item	AP [kg SO ₂ -eq]	EP [kg PO ₄ -eq]	HTP [kg 1,4-DB-eq]	ODP [kg CFC-11-eq]	GWP [kg CO ₂ -eq]
Electricity	1.50E-01	3.86E-02	5.91E+00	1.09E-06	2.10E+01
Lignin	5.57E-03	4.28E-04	3.50E-03	3.30E-08	5.07E-01
Polyacrylonitrile	1.34E-02	3.41E-03	6.27E+00	4.49E-07	4.24E+00
Nitrogen	8.22E-04	1.93E-04	2.29E-03	5.80E-09	9.79E-02
Methanol	5.76E-04	4.86E-05	1.01E-02	1.24E-08	3.38E-02
Dimethyl sulfoxide	5.01E-02	1.85E-03	1.00E+00	1.63E-07	1.18E+00
Waste gas	2.00E-01	4.22E-03	2.74E-04	0.00E+00	7.51E-01
Waste water	3.73E-03	1.14E-04	1.75E-01	6.85E-08	5.76E-01
Carbon fiber	4.24E-01	4.89E-02	1.34E+01	1.82E-06	2.84E+01

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. Waste gas, represent the emissions of CO₂, CH₄, NH₃, HCN, and CO by the applied polyacrylonitrile and lignin in carbon fiber production. The yield of carbon fiber assumed as 59.4%.

Table S12. Life cycle impact for the production of 1 kg 24 K lignin/PAN carbon fiber.

Item	AP [kg SO ₂ -eq]	EP [kg PO ₄ -eq]	HTP [kg 1,4-DB-eq]	ODP [kg CFC-11-eq]	GWP [kg CO ₂ -eq]
Electricity	6.16E-02	1.59E-02	2.43E+00	4.50E-07	7.35E+00
Lignin	5.57E-03	4.28E-04	3.50E-03	3.30E-08	5.07E-01
Polyacrylonitrile	1.34E-02	3.41E-03	6.27E+00	4.49E-07	4.24E+00
Nitrogen	2.06E-04	4.82E-05	5.72E-04	1.45E-09	2.45E-02
Methanol	5.76E-04	4.86E-05	1.01E-02	1.24E-08	3.38E-02
Dimethyl sulfoxide	5.01E-02	1.85E-03	1.00E+00	1.63E-07	1.18E+00
Waste gas	2.00E-01	4.22E-03	2.74E-04	0.00E+00	7.51E-01
Waste water	3.73E-03	1.14E-04	1.75E-01	6.85E-08	5.76E-01
Carbon fiber	3.35E-01	2.60E-02	9.89E+00	1.18E-06	8.10E+00

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. Waste gas, represent the emissions of CO₂, CH₄, NH₃, HCN, and CO by the applied polyacrylonitrile and lignin in carbon fiber production. The yield of carbon fiber assumed as 59.4%.

Table S13. Contribution analysis for the environmental impacts for spinning tows of lignin/PAN carbon fiber.

Spinning tow	AP [kg SO ₂ -eq]	EP [kg PO ₄ -eq]	HTP [kg 1,4-DB-eq]	ODP [kg CFC-11-eq]	GWP [kg CO ₂ -eq]
6 K	4.23E-01	4.88E-02	1.33E+01	1.82E-06	2.83E+01
12 K	3.67E-01	3.43E-02	1.11E+01	1.41E-06	1.95E+01
24 K	3.34E-01	2.60E-02	9.89E+00	1.17E-06	1.46E+01

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. L/PAN CF, represent the lignin/PAN carbon fiber. The yield of carbon fiber assumed as 59.4%.

4.2 Sensitivity analysis of carbon emission for different carbon fiber yields

Table S14. Electricity consumption for producing 1 kg 12 K lignin/PAN carbon fiber.

Processing	Instrument	Electricity consumption (kWh)
Dope preparation	Blender	2.90
Spinning	Spinning machine	5.88
Thermostabilization	Pre oxidation furnace	9.39
Carbonization	Carbonization furnace	4.82
Total	n.a.	21.79

Note: n.a., not applicable. The yield of carbon fiber assumed as 59.4%.

Table S15. Electricity consumption for producing 1 kg 12 K lignin/PAN carbon fiber.

Processing	Instrument	Electricity consumption (kWh)
Dope preparation	Blender	2.90
Spinning	Spinning machine	6.81
Thermostabilization	Pre oxidation furnace	10.88
Carbonization	Carbonization furnace	5.51
Total	n.a.	26.12

Note: n.a., not applicable. The yield of carbon fiber assumed as 51.3%.

Table S16. Life cycle impact for the production of 1 kg 12 K lignin/PAN carbon fiber.

Item	AP [kg SO ₂ -eq]	EP [kg PO ₄ -eq]	HTP [kg 1,4-DB-eq]	ODP [kg CFC-11-eq]	GWP [kg CO ₂ -eq]
Electricity	1.13E-01	2.90E-02	4.44E+00	8.18E-07	1.38E+01
Lignin	6.45E-03	4.96E-04	4.05E-03	3.82E-08	5.87E-01
Polyacrylonitrile	1.55E-02	3.95E-03	7.26E+00	5.19E-07	4.91E+00
Nitrogen	4.76E-04	1.12E-04	1.32E-03	3.36E-09	5.67E-02
Methanol	6.67E-04	5.63E-05	1.17E-02	1.44E-08	3.91E-02
Dimethyl sulfoxide	5.80E-02	2.15E-03	1.16E+00	1.89E-07	1.37E+00
Waste gas	2.31E-01	4.89E-03	3.17E-04	0.00E+00	9.49E-01
Waste water	4.32E-03	1.32E-04	2.03E-01	7.94E-08	5.76E-01
Carbon fiber	4.29E-01	4.08E-02	1.31E+01	1.66E-06	2.23E+01

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. Waste gas, represent the emissions of CO₂, CH₄, NH₃, HCN, and CO by the applied polyacrylonitrile and lignin in carbon fiber production. The yield of carbon fiber assumed as 51.3%.

Table S17. Contribution analysis for the environmental impacts for carbon fiber yields of 1 kg 12 K lignin/PAN carbon fiber.

Carbon yield	AP [kg SO ₂ -eq]	EP [kg PO ₄ -eq]	HTP [kg 1,4-DB-eq]	ODP [kg CFC-11-eq]	GWP [kg CO ₂ -eq]
59.4%	3.67E-01	3.43E-02	1.11E+01	1.41E-06	1.95E+02
51.3%	4.29E-01	4.07E-02	1.30E+01	1.66E-06	2.23E+02

Note: GWP, global warming potential; AP, acidification potential; EP, eutrophication potential; HTP, human toxicity potential; ODP, ozone depletion potential. The yield of carbon fiber assumed as 59.4% and 51.3%.

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