Supplementary Information

Greenhouse gas implications of extending the service life of PEM fuel cells for automotive applications: a life cycle assessment

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S1 Acronyms

FCEV	Fuel cell electric vehicle
FEP	Fluorinated ethylene propylene
GDM	Gas diffusion medium
GHG	Greenhouse gas
GWP	Global warming potential
GWP100	Global warming potential over a time horizon of 100 years
HFC-23	Trifluoromethane
HFP	Hexafluoropropylene
PTFE	Polytetrafluoroethylene
TFE	Tetrafluoroethylene

S2 Life Cycle Inventory of gas diffusion medium (GDM)

This section presents the life cycle inventory of gas diffusion mediums (GDMs) coated with polytetrafluoroethylene (PTFE) and fluorinated ethylene propylene (FEP). The production of the GDMs requires three main steps: i) synthesis of the monomers: tetrafluoroethylene (TFE) for PTFE production, TFE and hexafluoropropylene (HFP) for FEP production; ii) polymerization; iii) preparation of the GDM: i.e., coating of the carbon cloth with the selected polymer, and thermal treatment. The input and output flows of the production of PTFE- and FEP-based GDMs are listed in Table S1 and Table S2, respectively. The sources and assumptions for the inventory are described in the following subchapters.

S2.1 Synthesis of monomers

The dataset representing the production of TFE is available in Ecoinvent 3.6 [1]. The inventory for HFP synthesis is modelled from data reported in the literature. The inputs include the precursor monomers (TFE and HFC-23), and energy (electricity, natural gas, and steam); the outputs include the produced HFP, the unreacted TFE, and emissions to air. As for the precursor monomer inputs, the amount of TFE is from Rodriguez et al. [2], whose work describes the production process of HFP via combined pyrolysis of trifluoromethane (HFC-23) and TFE, while the amount of HFC-23 is calculated stoichiometrically, assuming a HFC:TFE ratio of 1:4 [3]. Energy inputs were retrieved from Rodriguez et al. [2]. Since energy inputs are given in aggregate form for TFE and HFP production processes, a mass-based allocation is applied to divide the inputs between the two co-products. The co-monomer mass ratio is calculated from the annual production of the factory analyzed by Rodriguez et al. [2] assuming that the monomers produced corresponds to the needs of the factory. Both TFE and HFP are starting material for the production of the co-polymers FEP and Viton. At the same time, TFE is also used to produce the homopolymer PTFE. The mass concentration of HFP in FEP is calculated from stoichiometry (i.e., 5 mol. % [2]). Viton composition is modelled based on the information available in its patent [4] and in Rodriguez et al. [2]: 20% TFE, 33% HFP, and 47% vinylidene fluoride. Efficiencies of the polymerization processes reported by Rodriguez et al. [2] are finally used to calculate the amount of monomers needed. Greenhouse gas emissions are calculated from Rodriguez et al. [2] and the patent of HFP production via combined pyrolysis of TFE and HFC-23 [5]. The greenhouse gas emissions comprise CO_2, CO, H_2, CF_4, CF_3CF_3, CF_3CCCF_3, C_2F_3H, CF_3CHCF_2, $CF_3CF_2CFCF_2$ [5]. The total volume is calculated assuming that gas emissions represent the 1.8% (example 4 in ref. [5]), and the remaining volume is constituted by unreacted monomers and non-GHG emissions. Subtracting from the total the carbon dioxide emissions reported by Rodriguez et al. [2], the amount of the remaining emissions is estimated. However, since no information about the share of the various gases was available, the conservative hypothesis that all the remaining volume is constituted by the gas with the highest global warming potential over 100 years (i.e., hexafluoroethane with a GWP100 of 11,100 kg CO_2 eq. kg^{-1}) is adopted. A summary of the inputs and outputs to HFP production process is reported in Table S2.

S2.2 Polymerization

The energy consumption and efficiency of the PTFE and FEP polymerization processes are based on Rodriguez et al. [2]. The consumption of water, initiators, and surfactants are neglected since they are assumed to be the same for the two polymers.

S2.3 Preparation of the GDMs

The preparation of GDM consists in the immersion of the carbon cloth into a polymeric suspension, and in the subsequent thermal treatment. The amount of polymer on the carbon cloth material, i.e., 16 mg cm^{-2} (34 wt.%), is calculated by weighting the GDM before and after the coating treatment. The electrical energy for the thermal treatment of 1 cm^2 of GDM, measured in J cm^{-2}, is calculated as follows:

$$E = \frac{P \cdot \Delta t \cdot 60}{A_{GDM}}$$

Where:

- P [W] is the power of the oven used for the experiment, equal to 2,200 W.
- Δt [min] is the amount of time required for the treatment, given by the time needed to reach the sintering temperature (i.e., 350 °C for PTFE, and 260 °C for FEP) plus the holding time (i.e., 30 minutes). An initial temperature of 25° C and a temperature slope of 5 °C min^{-1} are assumed for both the materials.
- A_{GDM} [cm^2] is the total area of GDM that can be treated in the oven. This corresponds to 4 GDMs of 49 cm^2.

Table S1 Inventory of PTFE-based gas diffusion medium

Polymer production: polytetrafluoroethylene (PTFE)			
Inputs	*Amount*	*Unit*	*Remarks*
Tetrafluoroethylene (TFE)	7.20E+01	g	Dataset ecoinvent 3.6: Tetrafluoroethylene {RER}| production
Electricity	3.32E+02	kJ	Dataset ecoinvent 3.6: Electricity, medium voltage {Europe without Switzerland}| market group for
Heat	1.19E+03	kJ	
Output	*Amount*	*Unit*	*Remarks*
Polytetrafluoroethylene (PTFE)	6.00E+01	g	Intermediate product
Gas Diffusion Medium production: PTFE-based GDM			
Inputs	*Amount*	*Unit*	*Remarks*
Polytetrafluoroethylene (PTFE)	6.00E+01	g	
Electricity	1.08E+02	kJ	Electricity for thermal treatment
Output	*Amount*	*Unit*	*Remarks*
Gas diffusion medium (PTFE-based GDM)	1.00E+00	cm^2	Final product

Table S2 Inventory of FEP-based gas diffusion medium

Monomer production: hexafluoropropylene (HFP)			
Inputs	*Amount*	*Unit*	*Remarks*
Tetrafluoroethylene (TFE)	3.24E+00	g	Dataset ecoinvent 3.6: Tetrafluoroethylene {RER}∣ production
Trifluoromethane (HFC-23)	1.26E+00	g	Dataset ecoinvent 3.6: Trifluoromethane GLO∣production
Electricity	8.29E+01	kJ	Dataset ecoinvent 3.6: Electricity, medium voltage {Europe without Switzerland}
Heat from steam	8.12E+01	kJ	Dataset ecoinvent 3.6: Heat, from steam, in chemical industry {RER}∣ steam production, as energy carrier, in chemical industry
Heat from natural gas	1.62E+02	kJ	Dataset ecoinvent 3.6: Heat, district or industrial, natural gas {RER}∣ market group for
Outputs	*Amount*	*Unit*	*Remarks*
Carbon dioxide (CO_2)	8.29E-03	g	Emission to air
Tetrafluoromethane (CF_4)	2.56E-02	g	Emission to air
Hexafluoropropylene (HFP)	6.00E+00	g	Intermediate product
Polymer production: fluorinated ethylene propylene (FEP)			
Inputs	*Amount*	*Unit*	*Remarks*
Tetrafluoroethylene (TFE)	7.80E+01	g	Dataset ecoinvent 3.6: Tetrafluoroethylene {RER}∣ production
Hexafluoropropylene (HFP)	6.00E+00	g	
Electricity	5.40E+02	kJ	Dataset ecoinvent 3.6: Electricity, medium voltage {Europe without Switzerland}∣ market group for
Heat	4.32E+02	kJ	Dataset ecoinvent 3.6: Heat, from steam, in chemical industry {RER}∣ market for heat, from steam, in chemical industry
Output	*Amount*	*Unit*	*Remarks*
Fluorinated ethylene propylene (FEP)	6.00E+01	g	Intermediate product
Gas diffusion medium production: FEP-based GDM			
Inputs	*Amount*	*Unit*	*Remarks*
Fluorinated ethylene propylene (FEP)	6.00E+01	g	
Electricity	8.62E+01	kJ	Dataset ecoinvent 3.6: Electricity, medium voltage {Europe without Switzerland}∣ market group for
Output	*Amount*	*Unit*	*Remarks*
Gas diffusion medium (FEP-based GDM)	1.00E+00	cm^2	Final product

S3 Supplementary results

This section presents the results of the two scenarios analysed: scenario A (both PTFE- and FEP-based vehicles are disposed of after 150,000 km), and scenario B (PTFE-based vehicle disposed of after 150,000 km, and FEP-based vehicle disposed of after 233,180 km). Table S3, Table S4 and Table S5Table show the global warming potential (GWP) difference between a FEP-based fuel cell electric vehicle (FCEV) and a PTFE-based FCEV for the different case studies and hydrogen production pathways analysed in scenario A. Table S6, Table S7, Table S8, Table S9, Table S10 and Table S11 present the GWP reduction linked to the substitution of PTFE with FEP in the vehicle's GDM for scenario B.

S3.1 Scenario A

Table S3 Global warming potential difference between a FEP-based fuel cell electric vehicle and a PTFE-based one for the different case studies considered in scenario A, in the case of hydrogen produced via electrolysis with EU electricity mix. Positive values indicate that the FEP-based vehicle has higher impacts.

Case	**Power density**	**Initial fuel consumption**	**GWP difference between FEP- and PTFE-based FCEVs**			
			GDM production	H_2 consumption	Life cycle	Life cycle (F.U.=1vkm)
	W/cm²	*kgH₂/100 km*	*kgCO₂ eq./150,000 km*	*kgCO₂eq./150,000 km*	*kgCO₂eq./150,000 km*	*gCO₂eq./km*
Low consumption	0.64	0.58	1.41	-192.51	-191.10	-1.27
	0.91	0.58	0.99	-192.51	-191.52	-1.28
High consumption	0.64	1.15	2.80	-377.62	-374.82	-2.50
	0.91	1.15	1.97	-377.62	-375.65	-2.50

Table S4 Global warming potential difference between a FEP-based fuel cell electric vehicle and a PTFE-based one for the different case studies considered in scenario A, in the case of hydrogen produced via electrolysis with renewable electricity mix in scenario A. Positive values indicate that the FEP-based vehicle has higher impacts.

Case	**Power density**	**Initial fuel consumption**	**GWP difference between FEP- and PTFE-based FCEVs**			
			GDM production	H_2 consumption	Life cycle	Life cycle (F.U.=1vkm)
	W/cm²	*kgH₂/100 km*	*kgCO₂ eq./150,000 km*	*kgCO₂eq./150,000 km*	*kgCO₂eq./150,000 km*	*gCO₂eq./km*
Low consumption	0.64	0.58	1.41	-22.36	-20.95	-0.14
	0.91	0.58	0.99	-22.36	-21.36	-0.14
High consumption	0.64	1.15	2.80	-43.85	-41.06	-0.27
	0.91	1.15	1.97	-43.85	-41.89	-0.28

Table S5 Global warming potential difference between a FEP-based fuel cell electric vehicle and a PTFE-based one for the different case studies considered in scenario A, in the case of hydrogen produced via steam reforming in scenario A. Positive values indicate that the FEP-based vehicle has higher impacts.

Case	Power density	Initial fuel consumption	GWP difference between FEP- and PTFE-based FCEVs			
			GDM production	H_2 consumption	Life cycle	Life cycle (F.U.=1vkm)
	W/cm²	*kgH₂/100 km*	*kgCO₂ eq./150,000 km*	*kgCO₂eq./150,000 km*	*kgCO₂eq./150,000 km*	*gCO₂eq./km*
Low consumption	0.64	0.58	1.41	-233.91	-232.50	-1.55
	0.91	0.58	0.99	-233.91	-232.92	-1.55
High consumption	0.64	1.15	2.80	-458.82	-456.03	-3.04
	0.91	1.15	1.97	-458.82	-456.86	-3.05

S3.2 Scenario B

Table S6 Life cycle GWP difference between the FEP-based FCEV and the PTFE-based one for the low consumption case (initial fuel consumption = 0.58 kgH_2/100 km) in scenario B, in the case of hydrogen produced via electrolysis powered by EU electricity mix.

GWP of vehicle production and EoL			GWP of hydrogen consumption		Life cycle GWP		
	PTFE	FEP	PTFE	FEP	PTFE	FEP	Difference
kgCO₂eq.	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*
12000 [6]	80.0	51.5	78.3	83.9	158.3	135.4	-23.0
14475 [7]	96.5	62.1	78.3	83.9	174.8	146.0	-28.9
15540 [8]	103.6	66.6	78.3	83.9	181.9	150.5	-31.4
16000 [9]	106.7	68.6	78.3	83.9	181.9	150.5	-31.4
14504[a]	96.7	62.2	78.3	83.9	175.0	146.1	-28.9

[a] Average of the four values retrieved from the literature.

Table S7 Life cycle GWP difference between the FEP-based FCEV and the PTFE-based one for the high consumption case (initial fuel consumption = 1.15 kgH_2/100 km) in scenario B, in the case of hydrogen produced via electrolysis powered by EU electricity mix in scenario B.

GWP of vehicle production and EoL			GWP of hydrogen consumption		Life cycle GWP		
	PTFE	FEP	PTFE	FEP	PTFE	FEP	Difference
kgCO₂eq.	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*	*gCO₂eq./km*
12000 [6]	80.0	51.5	155.3	166.3	235.3	217.8	-17.5
14475 [7]	96.5	62.1	155.3	166.3	251.8	228.4	-23.4
15540 [8]	103.6	66.6	155.3	166.3	258.9	233.0	-25.9
16000 [9]	106.7	68.6	155.3	166.3	262.0	234.9	-27.0
14504[a]	96.7	62.2	155.3	166.3	252.0	228.5	-23.5

[a] Average of the four values retrieved from the literature.

Table S8 Life cycle GWP difference between the FEP-based FCEV and the PTFE-based one for the low consumption case (initial fuel consumption = 0.58 kgH_2/100 km) in scenario B, in the case of hydrogen produced via electrolysis powered by renewable electricity mix in scenario B.

GWP of vehicle production and EoL			**GWP of hydrogen consumption**		**Life cycle GWP**		
	PTFE	FEP	PTFE	FEP	PTFE	FEP	Difference
$kgCO_2eq.$	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*
12000 [6]	80.0	51.5	9.1	9.7	89.1	61.2	-27.9
14475 [7]	96.5	62.1	9.1	9.7	105.6	71.8	-33.8
15540 [8]	103.6	66.6	9.1	9.7	112.7	76.4	-36.3
16000 [9]	106.7	68.6	9.1	9.7	115.8	78.4	-37.4
14504[a]	96.7	62.2	9.1	9.7	105.8	71.9	-33.9

[a] Average of the four values retrieved from the literature.

Table S9 Life cycle GWP difference between the FEP-based FCEV and the PTFE-based one for the high consumption case (initial fuel consumption = 1.15 kgH_2/100 km) in scenario B, in the case of hydrogen produced via electrolysis powered by renewable electricity mix in scenario B.

GWP of vehicle production and EoL			**GWP of hydrogen consumption**		**Life cycle GWP**		
	PTFE	FEP	PTFE	FEP	PTFE	FEP	Difference
$kgCO_2eq.$	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*	*$gCO_2eq./km$*
12000 [6]	80.0	51.5	18.0	19.3	98.0	70.8	-27.3
14475 [7]	96.5	62.1	18.0	19.3	114.5	81.4	-33.1
15540 [8]	103.6	66.6	18.0	19.3	121.6	86.0	-35.7
16000 [9]	106.7	68.6	18.0	19.3	124.7	87.9	-36.8
14504[a]	96.7	62.2	18.0	19.3	114.7	81.5	-33.2

[a] Average of the four values retrieved from the literature.

Table S10 Life cycle GWP difference between the FEP-based FCEV and the PTFE-based one for the low consumption case (initial fuel consumption = 0.58 kgH_2/100 km) in scenario B, in the case of hydrogen produced via steam reforming in scenario B.

GWP of vehicle production and EoL			**GWP of hydrogen consumption**		**Life cycle GWP**		
	PTFE	FEP	PTFE	FEP	PTFE	FEP	Difference
kgCO_2eq.	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*
12000 [6]	80.0	51.5	95.2	101.9	175.2	153.4	-21.8
14475 [7]	96.5	62.1	95.2	101.9	191.7	164.0	-27.7
15540 [8]	103.6	66.6	95.2	101.9	198.8	168.6	-30.2
16000 [9]	106.7	68.6	95.2	101.9	201.9	170.6	-31.3
14504[a]	96.7	62.2	95.2	101.9	191.9	164.1	-27.7

[a] Average of the four values retrieved from the literature.

Table S11 Life cycle GWP difference between the FEP-based FCEV and the PTFE-based one for the high consumption case (initial fuel consumption = 1.15 kgH_2/100 km) in scenario B, in the case of hydrogen produced via steam reforming in scenario B.

GWP of vehicle production and EoL			**GWP of hydrogen consumption**		**Life cycle GWP**		
	PTFE	FEP	PTFE	FEP	PTFE	FEP	Difference
kgCO_2eq.	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*	*gCO_2eq./km*
12000 [6]	80.0	51.5	188.7	202.1	268.7	253.6	-15.1
14475 [7]	96.5	62.1	188.7	202.1	285.2	264.2	-21.0
15540 [8]	103.6	66.6	188.7	202.1	292.3	268.7	-23.6
16000 [9]	106.7	68.6	188.7	202.1	295.4	270.7	-24.6
14504[a]	96.7	62.2	188.7	202.1	285.4	264.3	-21.1

[a] Average of the four values retrieved from the literature.

S4 Sensitivity analysis on the effect of the fuel cell degradation rate on GWP

A sensitivity analysis was performed on the fuel cell degradation rates (DRs) to test the robustness of our LCA results. The effect of a variation in the DR values (±30% and ±15%) on fuel consumption and GWP has been therefore investigated. Changes in fuel consumption are reported in Table S12, showing that a variation in the DR slightly affects the cumulative fuel consumption and the final GWP results (i.e., at the most ±3% for PTFE and ±1.5% for FEP). The variation of the GHG emissions from the use phase of the FCEV as a function of the DR is presented in Figure S1.

Table S12 Results of the sensitivity analysis: effect of the degradation rate (DR) variations on fuel consumption.

PTFE

DR	Δ(DR)	Fuel consumption					
		Low			High		
		Initial	Final	Cumulative	Initial	Final	Cumulative
μV/h	%	kgH_2/100 km	kgH_2/100 km	kgH_2,tot	kgH_2/100 km	kgH_2/100 km	kgH_2,tot
32.2	-30		0.783	1,022		1.553	2,027
39.1	-15		0.803	1,037		1.592	2,056
46.0	0	0.58	0.824	1,053	1.15	1.633	2,087
52.9	+15		0.845	1,069		1.676	2,120
59.8	+30		0.869	1,086		1.722	2,154

FEP

DR	Δ(DR)	Fuel consumption					
		Low			High		
		Initial	Final	Cumulative	Initial	Final	Cumulative
μV/h	%	kgH_2/100 km	kgH_2/100 km	kgH_2,tot	kgH_2/100 km	kgH_2/100 km	kgH_2,tot
21.7	-30		0.781	1,020		1.548	2,023
24.1	-15		0.786	1,024		1.558	2,031
31.0	0	0.58	0.801	1,036	1.15	1.588	2,053
35.7	+15		0.811	1,044		1.609	2,069
40.3	+30		0.822	1,052		1.630	2,085

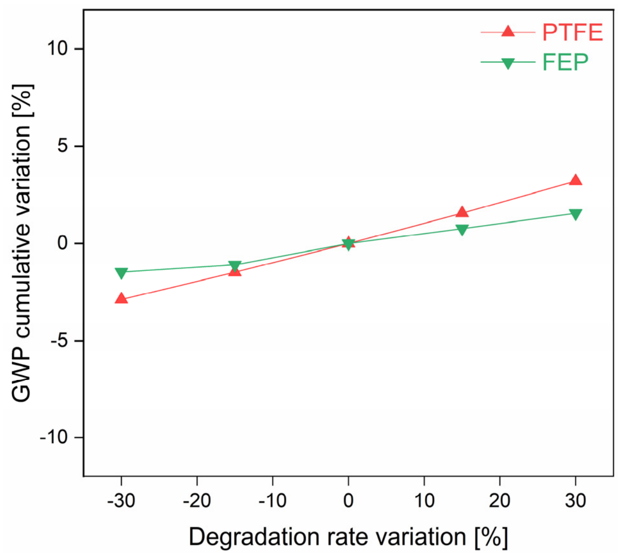

Figure S1 Variation in the global warming potential of the use phase of the fuel cell electric vehicle as a function of the degradation rate.

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