**Supplementary Information**

# A Sustainable Framework for Advancing Circular Practices for Polyethylene Terephthalate Textiles: Optimized Recycling of Depolymerization Products and LCA Validation

*Ye-eun Woo[†], Kyungha Baik[†], Sujin Jeong[†], Suhyun Lee[†, §] and Jooyoun Kim[*, †, §]*

[†]Department of Fashion and Textiles, Seoul National University, Seoul 08826, Republic of Korea

[§] Research Institute of Human Ecology, Seoul National University, Seoul 08826, Republic of Korea

* Corresponding author, E-mail: jkim256@snu.ac.kr

**Section 1: PET depolymerization**

**Table S1** Depolymerization of PET in various PET-based materials in $FeCl_3 \cdot 6H_2O$/PTSA.

| | PET type | Source | Time (min) | Temperature (℃) | Yield of TA |
|---|---|---|---|---|---|
| **1** | PET bottle | Market (evian) | 30 | 100 | 95.1% |
| **2** | PET bottle | Market (ICIS 8.0) | 30 | 100 | 94.8% |
| **3** | PET fabric white | Testfabrics, Inc. | 30 | 100 | 95.1% |
| **4** | PET fabric pink | Testfabrics, Inc. | 30 | 100 | 93.7% |
| **5** | PET fabric white | discarded | 30 | 100 | 94.9% |
| **6** | PET fabric orange | discarded | 30 | 100 | 95.2% |
| **7** | PET/Cotton fabric white | discarded | 30 | 100 | 94.3% |
| **8** | PET/Cotton fabric blue | discarded | 30 | 100 | 93.8% |

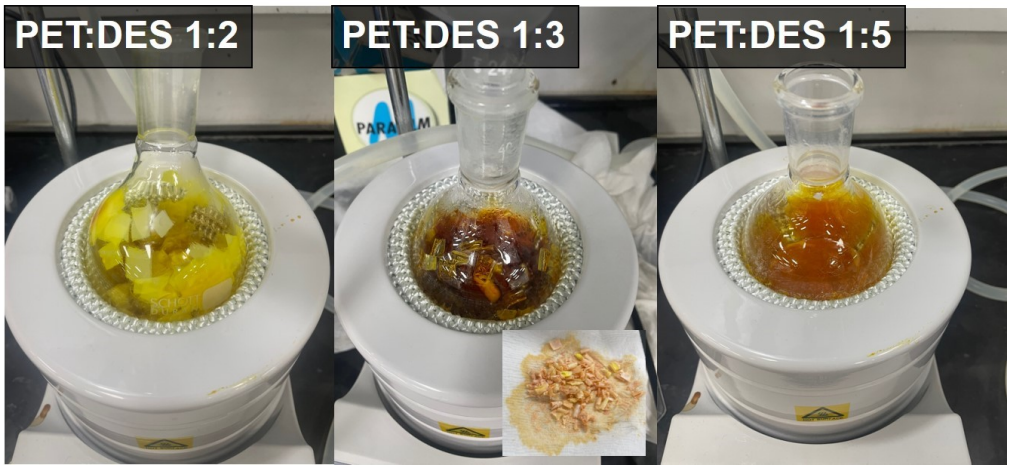



**Fig. S1** Effect of PET : DES mass ratio on PET depolymerization. Note: The molar ratio of $FeCl_3 \cdot 6H_2O$ to PTSA in the DES is 1 : 1.

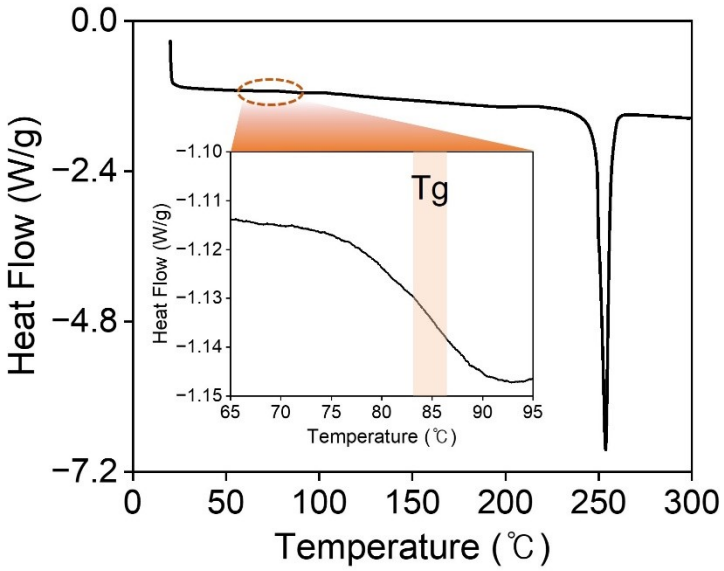



**Fig. S2** DSC spectrum of discarded dyed PET fabric.

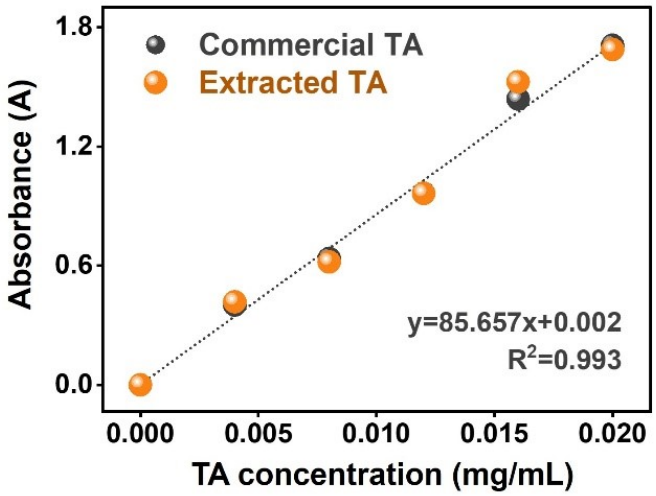



**Fig. S3** The standard curve of TA obtained using UV/Vis spectroscopy.

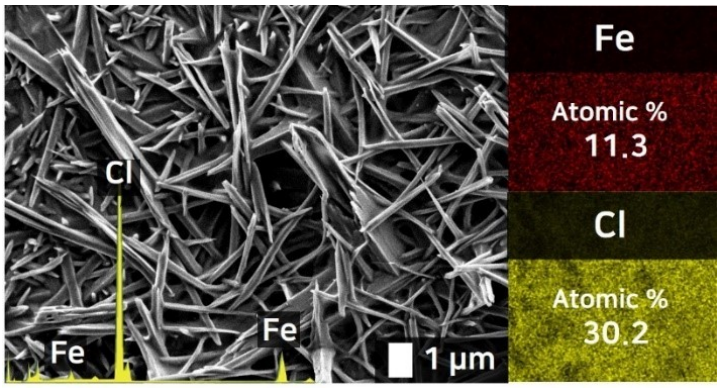



**Fig. S4** SEM-EDS analysis for the recycled $FeCl_3 \cdot 6H_2O$.

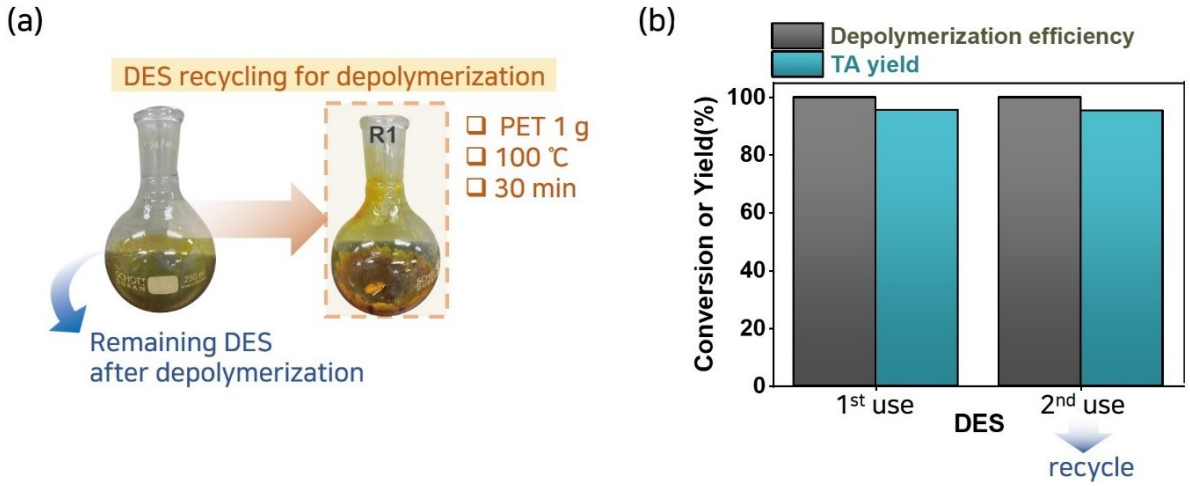



**Fig. S5** Comparison of (a) solvent state, (b) depolymerization efficiency and TA yield in PET depolymerization with recycled solvent.

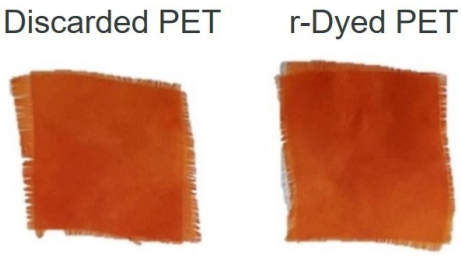



**Fig. S6** Photos of discarded PET fabric originally dyed (Discarded PET) and PET fabric dyed with recovered dye (r-Dyed PET).

**Section 2: Life cycle assessment (LCA)**

**Table S2** Summary of analyzed scenarios.

| | Explanation |
|---|---|
| **Assumptions** | • Because there were differences between the concentrations of materials in the LCI DB (life cycle inventory database, ecoinvent version 3.10) and what we actually used, the amount of materials for LCA was calculated by the adjusted value to make the amount of the materials (FeCl3, NaOH, and HCl) the same as the amount in our actual processes. After that, a compensation value for water was added to meet the material balance.<br>• A waste ethylene glycol compound, which remained after the process, was not included since its amount was too small to be measured.<br>• A waste DES, which remained after the process, was not included since its composition is difficult to evaluate.<br>• The electricity used for a refrigerator to precipitate FeCl3·6H2O was not included since its amount is negligible and difficult to measure. |

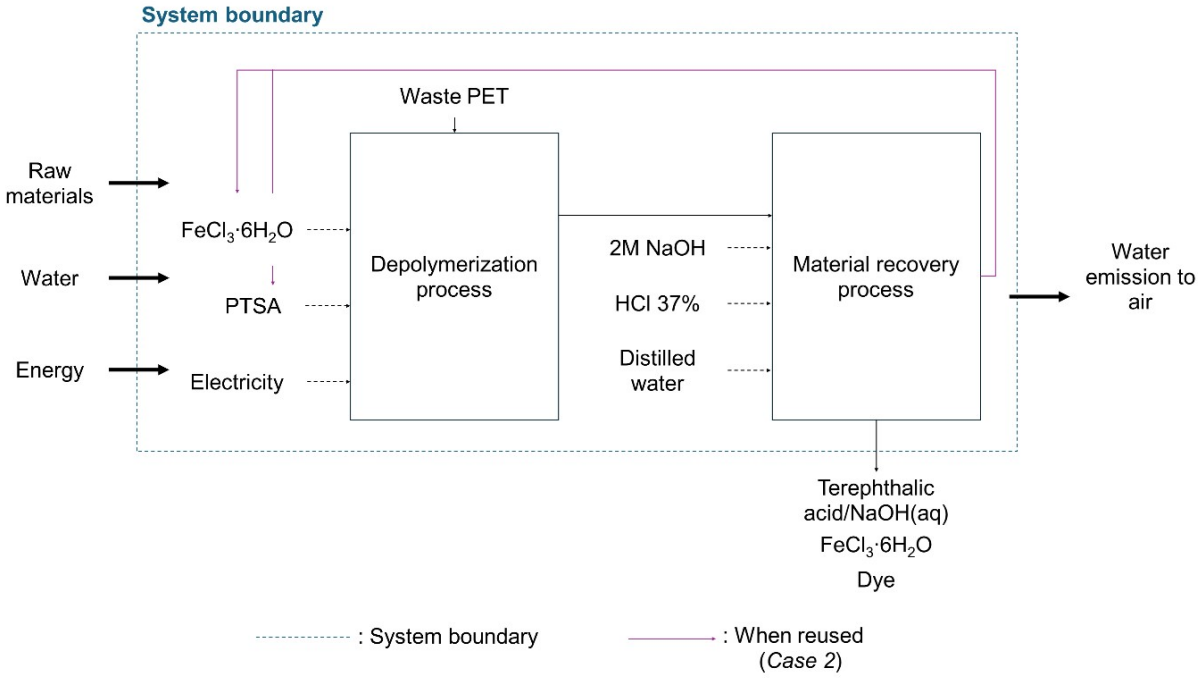



**Fig. S7** System boundary of life cycle assessment implemented in the study

**Table S3** Summary of analyzed scenarios.

| Scenario | Explanation |
|---|---|
| Case 1 | This is the base scenario, which does not reuse DES and all products (TA/NaOH(aq), $FeCl_3 \cdot 6H_2O$, and dye) are intended products of the process. There is no avoided production. |
| Case 2 | Once inserted DES is used for 2 cycles of depolymerization. There is no avoided production.<br>1st depolymerization: The same amount of DES of Case 1 is inserted.<br>2nd depolymerization: No additional input of DES.<br>Case 2 DES usage: Average of 1st and 2nd depolymerization (half of Case 1)<br>Environmental impacts of Case 2: Average of 1st and 2nd depolymerization |
| Case 3 | Input and output are the same as in Case 1. The whole process is considered as discarded PET management method focusing on the handling of waste. Thus, this scenario considered all products (TA/NaOH(aq), $FeCl_3 \cdot 6H_2O$, and dye) as useful byproducts, which provides avoided productions. |
| Case 4 | Input and output are the same as in Case 1. The whole process is considered a TA manufacturing process. Even though our process and LCA do not include a separation between TA and NaOH(aq), we recognized TA as the main product and NaOH(aq) is a useful co-product to reflect others' research on the recycled TA-based MOF synthesis. Thus, this scenario considers TA to be the main product, while all others ($FeCl_3 \cdot 6H_2O$, dye, and NaOH(aq)) are useful co-products that provide avoided productions. |

**Table S4** Input and output inventory of materials and electricity for the developed process which is actually used in the research where depolymerized waste PET is 5g. LCA is implemented based on this data with adjustment.

| Input/output | Materials and electricity | | Used amount |
|---|---|---|---|
| Input | Materials | Waste PET (g) | 5 |
| | | $FeCl_3 \cdot 6H_2O$ (g) | 18.96 |
| | | PTSA (g) | 6.04 |
| | | 2M NaOH (mL) | 12 |
| | | Distilled water (mL) | 15 |
| | | 37% HCl (mL) | 4 |
| | Electricity | Electricity (Wh) | 31.295 |
| Output | Materials | Terephthalic acid (g) | 3.78 |
| | | $FeCl_3 \cdot 6H_2O$ (g) | 0.83 |
| | | Dye (g) | 0.3 |
| | | Water, vaporized to air (g) | 17.6964 |
| | | 2M NaOH (mL) | 12 |

**Table S5** Input and output inventory for Case 1 and Case 2 adjusted for life cycle impact assessment (LCIA).

| Input /output | | Flow | Case 1 | Case 2 | Provider | Notes |
|---|---|---|---|---|---|---|
| Input | Materials | Waste PET (g) | 1,000 | 1,000 | market for waste polyethylene terephthalate, for recycling, sorted \| waste polyethylene terephthalate, for recycling, sorted \| Cutoff, U - RoW | |
| | | $FeCl_3 \cdot 6H_2O$ (g) | 5,596.1 | 2,798.04 | market for iron(III) chloride, without water, in 14% iron solution state \| iron(III) chloride, without water, in 14% iron solution state \| Cutoff, U - GLO | |
| | | PTSA (g) | 1,208 | 604 | market for chemical, organic \| chemical, organic \| Cutoff, U - GLO | PTSA was substituted to 'chemical, organic' because of absence of PTSA in the LCI DB. |
| | | 2M NaOH (g) | 384 | 384 | market for sodium hydroxide, without water, in 50% solution state \| sodium hydroxide, without water, in 50% solution state \| Cutoff, U - RoW | |
| | | Distilled water (g) | 3,000 | 3,000 | market group for tap water \| tap water \| Cutoff, U - GLO | |
| | | 37% HCl (g) | 1055.74 | 1055.74 | market for hydrochloric acid, without water, in 30% solution state \| hydrochloric acid, without water, in 30% solution state \| Cutoff, U - RoW | |
| | | Compensation value, water (g) | 204.16 | 1106.22 | market group for tap water \| tap water \| Cutoff, U - GLO | This term was introduced to compensate the gap from '$FeCl_3 \cdot 6H_2O$', '37% HCl', and '2M NaOH'. |
| | Electricity | Electricity (Wh) | 6,259 | 6,259 | market group for electricity, low voltage \| electricity, low voltage \| Cutoff, U - GLO | |

**Table S5** *(continued)*

| Input /output | | Flow | Case 1 | Case 2 | Provider | Notes |
|---|---|---|---|---|---|---|
| Output | Materials | Terephthalic acid (g) | 756 | 756 | | |
| | | $FeCl_3 \cdot 6H_2O$ (g) | 244.98 | 244.98 | | ‘$FeCl_3 \cdot 6H_2O$’ in the output session was also considered as ‘iron(III) chloride, without water, in 14% iron solution state’ as like input session |
| | | Dye (g) | 60 | 60 | | Dye was substituted to ‘chemical, organic’ because of absence of dye materials in the LCI DB. |
| | | Water, vaporized to air (g) | 3,539.28 | 3,539.28 | | |
| | | 2M NaOH (g) | 384 | 384 | | ‘2M NaOH’ in the output session was also considered as ‘sodium hydroxide, without water, in 50% solution state’ as like input session |
| | | Compensation value, water (g) | 2,129.02 | 2,129.02 | | This term was introduced to compensate the gap from ‘$FeCl_3 \cdot 6H_2O$’ and ‘2M NaOH’. |

**Table S6** Input and output inventory for Case 3 and Case 4 adjusted for life cycle impact assessment (LCIA).

| Input /output | | Flow | Case 3 | Case 4 | Provider | Notes |
|---|---|---|---|---|---|---|
| Input | Materials | Waste PET (g) | 1,000 | 1,000 | market for waste polyethylene terephthalate, for recycling, sorted \| waste polyethylene terephthalate, for recycling, sorted \| Cutoff, U - RoW | |
| | | $FeCl_3 \cdot 6H_2O$ (g) | 5,596.1 | 5,596.1 | market for iron(III) chloride, without water, in 14% iron solution state \| iron(III) chloride, without water, in 14% iron solution state \| Cutoff, U - GLO | |
| | | PTSA (g) | 1,208 | 1,208 | market for chemical, organic \| chemical, organic \| Cutoff, U - GLO | PTSA was substituted to 'chemical, organic' because of absence of PTSA in the LCI DB. |
| | | 2M NaOH (g) | 384 | 384 | market for sodium hydroxide, without water, in 50% solution state \| sodium hydroxide, without water, in 50% solution state \| Cutoff, U - RoW | |
| | | Distilled water (g) | 3,000 | 3,000 | market group for tap water \| tap water \| Cutoff, U - GLO | |
| | | 37% HCl (g) | 1055.74 | 1055.74 | market for hydrochloric acid, without water, in 30% solution state \| hydrochloric acid, without water, in 30% solution state \| Cutoff, U - RoW | |
| | | Compensation value, water (g) | 204.16 | 204.16 | market group for tap water \| tap water \| Cutoff, U - GLO | This term was introduced to compensate the gap from '$FeCl_3 \cdot 6H_2O$', '37% HCl', and '2M NaOH'. |
| | Electricity | Electricity (Wh) | 6,259 | 6,259 | market group for electricity, low voltage \| electricity, low voltage \| Cutoff, U - GLO | |

**Table S6** *(continued)*

| Input /output | | Flow | Case 3 | Case 4 | Provider | Notes |
|---|---|---|---|---|---|---|
| Output | Materials | Terephthalic acid (g) | 756 | 756 | (Only for *Case 3*) purified terephthalic acid production \| purified terephthalic acid \| Cutoff, U - RoW | |
| | | $FeCl_3 \cdot 6H_2O$ (g) | 244.98 | 244.98 | iron(III) chloride production, without water, in 14% iron solution state \| iron(III) chloride, without water, in 14% iron solution state \| Cutoff, U - RoW | '$FeCl_3 \cdot 6H_2O$' in the output session was also considered as 'iron(III) chloride, without water, in 14% iron solution state' as like input session |
| | | Dye (g) | 60 | 60 | chemical production, organic \| chemical, organic \| Cutoff, U - GLO | Dye was substituted to 'chemical, organic' because of absence of dye materials in the LCI DB. |
| | | Water, vaporized to air (g) | 3,539.28 | 3,539.28 | | |
| | | 2M NaOH (g) | 384 | 384 | chlor-alkali electrolysis, membrane cell \| sodium hydroxide, without water, in 50% solution state \| Cutoff, U - RoW | '2M NaOH' in the output session was also considered as 'sodium hydroxide, without water, in 50% solution state' as like input session |
| | | Compensation value, water (g) | 2,129.02 | 2,129.02 | tap water production, conventional treatment \| tap water \| Cutoff, U - RoW | This term was introduced to compensate the gap from '$FeCl_3 \cdot 6H_2O$' and '2M NaOH'. |

**Table S7** LCIA results and contributions of usage of electricity and each material for each category in Case 1.

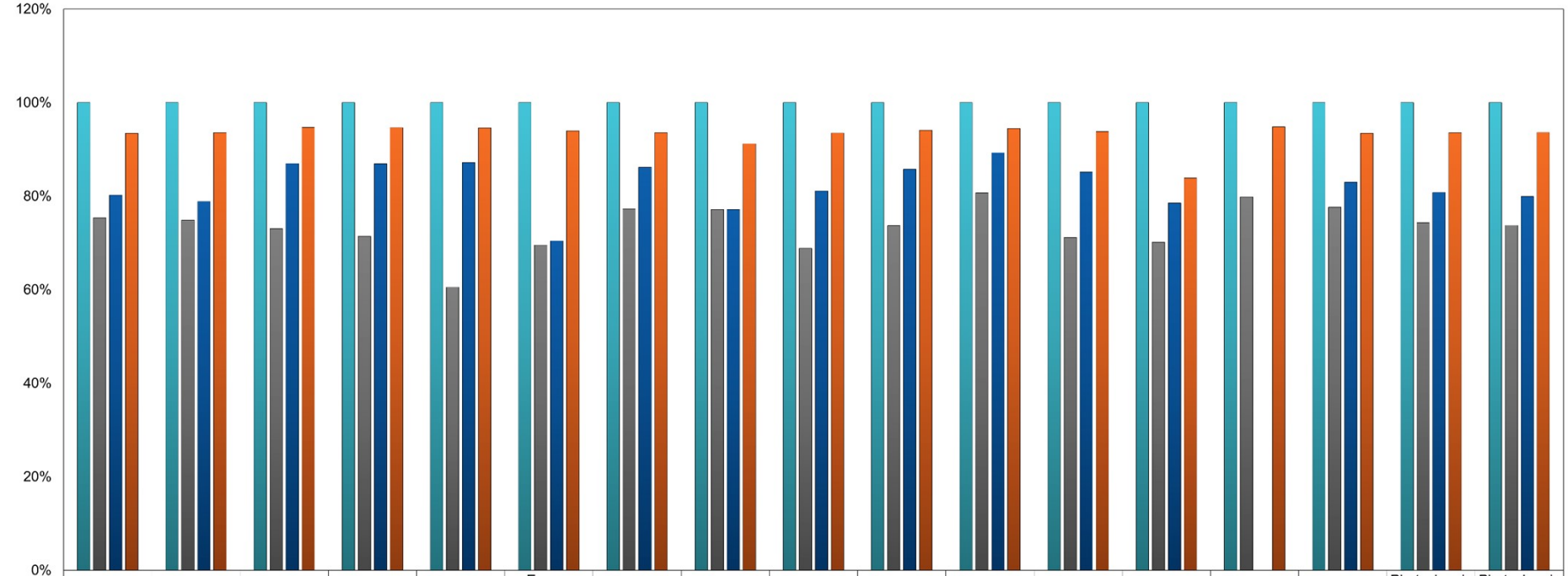
| Impact categories | Total result | Unit | Contribution of electricity and materials[1] | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | Electricity | PTSA | $FeCl_3 \cdot 6H_2O$ | HCl | NaOH | Sorted waste PET | Water |
| Acidification: terrestrial | 0.04231 | kg $SO_2$-eq. | 34.26% | 19.44% | 29.82% | 10.22% | 4.60% | 1.64% | 0.02% |
| Global warming potential | 13.6242 | kg $CO_2$-eq. | 34.81% | 26.52% | 23.76% | 7.84% | 4.30% | 2.76% | 0.02% |
| Ecotoxicity: freshwater | 1.03504 | kg 1,4-DCB-eq. | 28.52% | 12.42% | 41.41% | 9.55% | 2.94% | 5.15% | 0.02% |
| Ecotoxicity: marine | 1.51533 | kg 1,4-DCB-eq. | 25.52% | 18.49% | 38.73% | 9.06% | 2.83% | 5.35% | 0.02% |
| Ecotoxicity: terrestrial | 214.81744 | kg 1,4-DCB-eq. | 7.79% | 57.04% | 21.88% | 6.06% | 1.91% | 5.27% | 0.05% |
| Energy resources: non-renewable, fossil | 4.31055 | kg oil-eq. | 26.94% | 42.22% | 18.68% | 6.63% | 3.34% | 2.17% | 0.02% |
| Eutrophication: freshwater | 0.00515 | kg P-eq. | 40.91% | 15.03% | 30.48% | 7.94% | 4.47% | 1.17% | 0.02% |
| Eutrophication: marine | 0.00057 | kg N-eq. | 26.39% | 19.75% | 26.14% | 8.95% | 5.66% | 13.09% | 0.02% |
| Human toxicity: carcinogenic | 2.92869 | kg 1,4-DCB-eq. | 18.49% | 18.70% | 43.74% | 7.88% | 3.91% | 7.12% | 0.17% |
| Human toxicity: non-carcinogenic | 20.34799 | kg 1,4-DCB-eq. | 27.88% | 14.25% | 38.40% | 10.09% | 3.62% | 5.75% | 0.02% |

[1] As mentioned above, since materials in the LCI DB were not exactly same concentration with our process, compensation value of water was added in the LCA to meet the total materials inserted.

**Table S7** *(Continued)*

| Impact categories | Total result | Unit | Contribution of electricity and materials[1] | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | Electricity | PTSA | $FeCl_3 \cdot 6H_2O$ | HCl | NaOH | Sorted waste PET | Water |
| Ionising radiation | 1.07091 | kBq Co-60-eq. | 50.65% | 10.51% | 28.06% | 5.88% | 4.14% | 0.72% | 0.03% |
| Land use | 0.28102 | $m^2a$ crop-eq. | 24.59% | 17.82% | 39.81% | 8.29% | 4.07% | 5.39% | 0.02% |
| Material resources: metals/minerals | 0.46911 | kg Cu-eq. | 4.59% | 10.29% | 49.44% | 19.75% | 13.45% | 2.44% | 0.04% |
| Ozone depletion | 4.66E-06 | kg CFC-11-eq. | 33.06% | 15.02% | 25.33% | 19.79% | 4.99% | 1.78% | 0.03% |
| Particulate matter formation | 0.02324 | kg PM2.5-eq. | 39.68% | 16.48% | 28.27% | 8.82% | 4.76% | 1.97% | 0.02% |
| Photochemical oxidant formation: human health | 0.02981 | kg $NO_x$-eq. | 33.34% | 22.18% | 29.22% | 7.43% | 4.67% | 3.14% | 0.02% |
| Photochemical oxidant formation: terrestrial ecosystems | 0.03136 | kg $NO_x$-eq. | 32.38% | 23.97% | 28.56% | 7.34% | 4.56% | 3.17% | 0.02% |
| Water use | 0.10037 | $m^3$ | 23.92% | 30.06% | 25.75% | 12.39% | 6.28% | 1.58% | 0.02% |

[1] As mentioned above, since materials in the LCI DB were not exactly same concentration with our process, compensation value of water was added in the LCA to meet the total materials inserted.




| | Acidification: terrestrial | Global warming potential | Ecotoxicity: freshwater | Ecotoxicity: marine | Ecotoxicity: terrestrial | Energy resources: non-renewable, fossil | Eutrophication: freshwater | Eutrophication: marine | Human toxicity: carcinogenic | Human toxicity: non-carcinogenic | Ionising radiation | Land use | Material resources: metals/minerals | Ozone depletion[1] | Particulate matter formation | Photochemical oxidant formation: human health | Photochemical oxidant formation: terrestrial ecosystems |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Case 1 | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% | 100% |
| Case 2 | 75% | 75% | 73% | 71% | 61% | 70% | 77% | 77% | 69% | 74% | 81% | 71% | 70% | 80% | 78% | 74% | 74% |
| Case 3 | 80% | 79% | 87% | 87% | 87% | 70% | 86% | 77% | 81% | 86% | 89% | 85% | 79% | -273% | 83% | 81% | 80% |
| Case 4 | 93% | 94% | 95% | 95% | 95% | 94% | 94% | 91% | 94% | 94% | 94% | 94% | 84% | 95% | 93% | 94% | 94% |

[1] Ozone depletion of Case 3 is -273% though It is not appeared in the graph.

**Fig. S8** LCIA results relative ratio of scenarios compared to Case 1.