Electronic Supplementary Material (ESI) for Green Chemistry.

Supplementary Material

The supporting information includes the life cycle inventories (LCIs) of the model cell (Table S 1), its components (Table S 2, Table S 4, Table S 5, Table S 9), and materials (Table S 3, Table S 6, Table S 7, Table S 8, Table S 10 - Table S 15) as modeled in the GaBi software. In Table S 16 - Table S 23, the LCIs of the precursor materials of LATP and LLZO are listed: aluminium nitrate nonahydrate [$Al(NO_3)_3$*9 H_2O], lithium nitrate ($LiNO_3$), lanthanum nitrate [$La(NO_3)_3$*6 H_2O], zirconium oxynitrate [$ZrO(NO_3)_2$*6 H_2O], zirconium tetrachloride [$ZrCl_4$], ammonium dihydrogen phosphate [$(NH_4)H_2PO_4$], lithium acetate [$Li(CH_3COO)$*2 H_2O], and titanium isopropoxide [$Ti[OCH(CH_3)_2]_4$].

Energy is given as net calorific value unless otherwise stated. In addition, the origin of the process data is marked (e.g., Sphera, ecoinvent 3.7.1, PlasticesEurope, Nickel Institute, own process model).

In Chapter 10, a comparison of LATP and LLZO with the standard liquid electrolyte LiPF6 (in EC/DMC) is carried out.

1. LCI of pouch cell

The pouch cell, measuring 5 cm * 5 cm, has a capacity of 6.15 mAh/cm^2 and an energy density of 394.4 Wh/kg. All materials for the layers have been calculated with 3% offcut. Argon and electricity input are required for assembly in the glovebox. Energy requirements for material supply is included in their data sets.

Table S 1. LCI of one pouch, 5 x 5 cm^2

Flows	Quantities	Amount	Unit
Input			
DE: Aluminium foil Sphera, 10 µm	Mass	6.95E-05	kg
EU-28: Copper sheet (A1-A3) Sphera, 10 µm	Mass	2.30E-04	kg
DE: Lithium anode, 10 µm, own process model	Mass	1.38E-05	kg
DE: Electrolyte (LLZO), 10 µm, own process model	Area	2.58E-03	sqm
DE: Mixed cathode (LATP/NMC 622, weight ratio 20:80) 100 µm, own process model	Area	2.58E-03	sqm
DE: Carrier foil (PET) 100 µm, own process model	Area	2.58E-03	sqm
DE: Argon (gaseous) Sphera	Mass	7.14E-04	kg
DE: Electricity grid mix Sphera	Energy	1.01E-01	MJ
Output			
Pouch cell	**Number of pieces**	**1.00E+00**	**Pcs.**

2. LCI of lithium anode

The inventory of the 10 µm lithium metal anode involves the processes shown in Table S 2 and Table S 3. The inventory of lithium foil production with a thickness of 35 µm showing in Table S 3 is taken from Deng, Yelin, Li, Jianyang [1]. For its use in the inventory of the pouch cell (Table S 1), only the mass was linearly reduced to 10 µm. This means that the energy requirement for cold rolling is underestimated, since the thickness of the foil in Deng is 35 µm and not 10 µm as we assumed.

Table S 2. LCI of lithium anode

Flows	Quantities	Amount	Unit
Input			
DE: Lithium foil, 35µm, adapted according to [1]	Mass	1.00E+00	kg
RER: transport, freight, lorry 7.5-16 metric ton, EURO6 ecoinvent 3.7.1	Transport	2.00E-01	tkm
Output			
DE: Lithium anode	**Mass**	**1.00E+00**	**kg**

Table S 3. LCI of lithium foil adapted according to [1]

Flows	Quantities	Amount	Unit
Input			
GLO: market for lithium ecoinvent 3.7.1	Mass	1.04E+00	kg
RER: sheet rolling, aluminium ecoinvent 3.7.1	Mass	4.95E+00	kg
GLO: tetraethyl orthosilicate production ecoinvent 3.7.1	Mass	1.54E-01	kg
DE: Electricity grid mix Sphera	Energy	48.5E+00	MJ
DE: Process steam from natural gas 85% Sphera	Energy	155.8E+00	MJ
Output			
DE: Lithium foil, 35µm	**Mass**	**1.00E+00**	**kg**
Sodium silicates [Inorganic emissions to air]	Mass	1.87E-02	kg
Solid waste [Hazardous waste for disposal]	Mass	3.80E-02	kg

3. LCI of carrier foil

The carrier foil is required during tape casting of the electrolyte layer and is subsequently disposed of as waste.

Table S 4. LCI of carrier foil

Flows	Quantities	Amount	Unit
Input			
DE: Polyethylene terephthalate granulate (PET via DMT) Sphera	Mass	1.35E-01	kg
RER: transport, freight, lorry 7.5-16 metric ton, EURO6 ecoinvent 3.7.1	Transport	2.70E-02	tkm
Output			
DE: Carrier foil (PET) 100 µm	**Area**	**1.00E+00**	**sqm**
CH: treatment of waste polyethylene terephthalate, municipal incineration ecoinvent 3.7.1	Mass	1.35E-01	kg

4. LCI of LLZO electrolyte

Table S 5. LCI of LLZO electrolyte

Flows	Quantities	Amount	Unit
Input			
DE: electrolyte slurry, own process model	Mass	4.73E-03	kg
DE: Electricity grid mix Sphera	Energy	1.71E-01	MJ
Output			
DE: Electrolyte (LLZO), 10 µm	**Area**	**5.00E-02**	**sqm**
Butanone (methyl ethyl ketone) [Group NMVOC to air]	Mass	1.06E-03	kg
Ethanol [Group NMVOC to air]	Mass	5.45E-04	kg

Table S 6. LCI of LLZO electrolyte slurry

Flows	Quantities	Amount	Unit
Input			
DE: LLZO powder, own process model	Mass	5.40E-01	kg
RER: Polyether polyol PlasticsEurope	Mass	1.35E-02	kg
DE: Ethanol (96%) (hydrogenation with nitric acid) Sphera	Mass	1.15E-01	kg
RER: methyl ethyl ketone production ecoinvent 3.7.1	Mass	2.23E-01	kg
EU-28: Triethylene glycol PlasticsEurope	Mass	5.40E-02	kg
RER: vinyl acetate production ecoinvent 3.7.1[a]	Mass	5.40E-02	kg
RER: transport, freight, lorry 7.5-16 metric ton, EURO6 ecoinvent 3.7.1	Transport	2.00E-01	tkm
Output			
DE: Electrolyte slurry	**Mass**	**1.00E+00**	**kg**

[a] substitute for PVB 98 (polyvinyl butyral)

5. LCI of LLZO powder

Table S 7. LCI of LLZO powder produced via solid-state reaction

Flows	Quantities	Amount	Unit
Input			
GLO: lithium hydroxide production ecoinvent 3.7.1	Mass	3.23E-01	kg
GLO: market for lanthanum oxide ecoinvent 3.7.1	Mass	5.82E-01	kg
GLO: market for zirconium oxide ecoinvent 3.7.1	Mass	2.93E-01	kg
IAI Area, EU27 & EFTA: market for aluminium oxide, non-metallurgical ecoinvent 3.7.1	Mass	1.21E-02	kg
DE: Electricity grid mix Sphera	Energy	82.30E+00	MJ
Output			
DE: LLZO Electrolyte powder produced via solid-state reaction	**Mass**	**1.00E+00**	**kg**

Table S 8. LCI of LLZO powder produced via spray drying

Flows	Quantities	Amount	Unit
Input			
DE: Aluminium nitrate nonahydrate [$Al(NO_3)_3$*9 H_2O], own process model	Mass	9.01E-02	kg
DE: Lithium nitrate $LiNO_3$, own process model	Mass	5.94E-01	kg
DE: Lanthannitrat [$La(NO_3)_3$*6 H_2O], own process model	Mass	1.55E+00	kg
DE: Zirconium oxynitrate [$ZrO(NO_3)_2$*6 H_2O], own process model	Mass	8.07E-01	kg
Europe without Switzerland: market for water, deionised ecoinvent 3.7.1	Mass	5.00E+00	kg
DE: Nitric acid (98%) Sphera	Mass	7.82E-03	kg
DE: Electricity grid mix Sphera	Energy	51.10E+00	MJ
Output			
DE: LLZO Electrolyte powder produced via spray drying	**Mass**	**1.00E+00**	**kg**

6. LCI of mixed cathode (LATP/NMC 622)

Table S 9. LCI of mixed cathode

Flows	Quantities	Amount	Unit
Input			
DE: mixed cathode slurry, own process model	Mass	3.89E-02	kg
DE: Electricity grid mix Sphera	Energy	4.14E-01	MJ
Output			
DE: Mixed cathode (LATP/NMC 622), 100 µm	**Area**	**5.00E-02**	**sqm**
Butanone (methyl ethyl ketone) [Group NMVOC to air]	Mass	8.70E-03	kg
Ethanol [Group NMVOC to air]	Mass	4.49E-03	kg

Table S 10. LCI of mixed cathode slurry

Flows	Quantities	Amount	Unit
Input			
DE: LATP powder, own process model	Mass	1.13E-01	kg
DE: NMC 622 powder according to [2]	Mass	4.27E-01	kg
RER: Polyether polyol PlasticsEurope	Mass	1.35E-02	kg
DE: Ethanol (96%) (hydrogenation with nitric acid) Sphera	Mass	1.15E-01	kg
RER: methyl ethyl ketone production ecoinvent 3.7.1	Mass	2.23E-01	kg
EU-28: Triethylene glycol PlasticsEurope	Mass	5.40E-02	kg
RER: vinyl acetate production ecoinvent 3.7.1[a]	Mass	5.40E-02	kg
RER: transport, freight, lorry 7.5-16 metric ton, EURO6 ecoinvent 3.7.1	Transport	2.00E-01	tkm
Output			
DE: Mixed cathode slurry	**Mass**	**1.00E+00**	**kg**

[a] substitute for PVB 98 (polyvinyl butyral)

7. LCI of LATP powder

Table S 11. LCI of LATP powder produced via solid-state reaction

Flows	Quantities	Amount	Unit
Input			
IAI Area, EU27 & EFTA: market for aluminium oxide, non-metallurgical ecoinvent 3.7.1	Mass	3.98E-02	kg
GLO: lithium carbonate production, from concentrated brine ecoinvent 3.7.1	Mass	1.25E-01	kg
DE: Ammonium dihydrogen phosphate [$(NH_4)H_2PO_4$], own process model	Mass	8.97E-01	kg
RER: market for titanium dioxide ecoinvent 3.7.1	Mass	3.53E-01	kg
DE: Electricity grid mix Sphera	Energy	45.90E+00	MJ
Output			
DE: LATP powder produced via solid-state reaction	**Mass**	**1.00E+00**	**kg**
Carbon dioxide [Inorganic emissions to air]	Mass	7.44E-02	kg
Water vapour [Inorganic emissions to air]	Mass	4.21E-01	kg
NOx retained, by selective catalytic reduction[a]	Mass	3.59E-01	kg

[a] NOx is not released as an emission, but is converted to nitrogen and water by selective catalytic reduction with ammonia

Table S 12. LCI of LATP powder produced via sol-gel process

Flows	Quantities	Amount	Unit
Input			
DE: Lithium acetate [$Li(CH_3COO)*2\ H_2O$], own process model	Mass	3.46E-01	kg
DE: Aluminium nitrate nonahydrate [$Al(NO_3)_3*9\ H_2O$], own process model	Mass	2.93E-01	kg
DE: Titanium isopropoxide [$Ti[OCH(CH_3)_2]_4$], own process model	Mass	1.26E+00	kg
DE: Ammonium dihydrogen phosphate [$(NH_4)H_2PO_4$], own process model	Mass	8.99E-01	kg
Europe without Switzerland: market for water, deionised ecoinvent 3.7.1	Mass	2.00E+00	kg
DE: Electricity grid mix Sphera	Energy	42.10E+00	MJ
Output			
DE: LATP powder produced via sol-gel process	**Mass**	**1.00E+00**	**kg**
Carbon dioxide [Inorganic emissions to air]	Mass	2.75E+00	kg
Water vapour [Inorganic emissions to air]	Mass	3.88E+00	kg
NOx retained, by selective catalytic reduction[a]	Mass	4.68E-01	kg

[a] NOx is not released as an emission, but is converted to nitrogen and water by selective catalytic reduction with ammonia

8. LCI of NMC 622 powder

The quantities of the flows to produce NMC 622 were obtained from [2]. Our LCI was set up with processes from ecoinvent 3.7.1, sphere, Nickel Institute, and Cobalt Development Institute (CDI). A separate process has been developed to produce cobalt sulfate based on the reaction of cobalt with concentrated sulfuric acid (Table S 15).

Table S 13. LCI of NMC 622 according to [2]

Flows	Quantities	Amount	Unit
Input			
DE: NMC 622 Precursor, own process model according to [2]	Mass	1.00E+00	kg
DE: Process steam from natural gas 85% Sphera	Energy	6.80E+00	MJ
DE: Oxygen (gaseous) Sphera	Mass	4.29E+00	kg
Europe without Switzerland: market for water, deionised ecoinvent 3.7.1	Mass	3.00E-01	kg
DE: Electricity grid mix Sphera	Energy	36.00E+00	MJ
GLO: lithium carbonate production, from concentrated brine ecoinvent 3.7.1	Mass	4.00E-01	kg
Output			
NMC 622 [Materials]	**Mass**	**1.00E+00**	**kg**

Table S 14. LCI of NMC 622 precursor according to [2]

Flows	Quantities	Amount	Unit
Input			
GLO: Nickel sulphate hexahydrate [$NiSO_4$*6 H_2O] Nickel Institute	Mass	1.00E+00	kg
DE: Cobalt sulphate [$Co(SO)_4$], own process model	Mass	3.00E-01	kg
GLO: market for manganese sulfate ecoinvent 3.7.1	Mass	3.00E-01	kg
DE: Sodium hydroxide (caustic soda) mix (100%) Sphera	Mass	9.00E-01	kg
Europe without Switzerland: market for water, deionised ecoinvent 3.7.1	Mass	6.00E+02	kg
DE: market for natural gas, high pressure ecoinvent 3.7.1	Volume	1.10E+00	m^3
DE: Ammonia (NH_3) without CO_2 recovery (carbon dioxide emissions to air) Sphera	Mass	1.00E-01	kg
Output			
NMC 622 Precursor [Materials]	**Mass**	**1.00E+00**	**kg**

Table S 15. LCI of cobalt sulphate

Flows	Quantities	Amount	Unit
Input			
GLO: Cobalt, refined (metal) CDI	Mass	3.80E-01	kg
DE: Sulphuric acid mix (96%) (consumption mix) Sphera	Mass	6.59E-01	kg
Output			
Cobalt sulphate [Co(SO)$_4$] [Materials]	**Mass**	**1.00E+00**	**kg**
Hydrogen [Inorganic emissions to air]	Mass	1.30E-02	kg

9. LCI of precursors for LLZO and LATP production

*Table S 16. LCI of aluminum nitrate nonahydrate ($Al(NO_3)_3$ *9 H_2O)*

Flows	Quantities	Amount	Unit
Input			
IAI Area, EU27 & EFTA: aluminium hydroxide production ecoinvent 3.7.1	Mass	2.1E-01	kg
DE: Process steam from natural gas 85% Sphera	Energy	5.96E-01	MJ
DE: Nitric acid (98%) Sphera	Mass	5.14E-01	kg
Europe without Switzerland: market for water, deionised ecoinvent 3.7.1	Mass	2.88E-01	kg
DE: Electricity grid mix Sphera	Energy (net calorific value)	5.80E-01	MJ
Europe without Switzerland: market for tap water ecoinvent 3.7.1	Mass	5.14E-01	kg
Output			
Aluminium nitrate nonahydrate [Materials]	**Mass**	**1.00E+00**	**kg**

Table S 17. LCI of lithium nitrate ($LiNO_3$)

Lithium nitrate ($LiNO_3$) is prepared by stirring lithium carbonate (Li_2CO_3) and nitric acid (HNO_3) together at ambient temperature and then adding lithium hydroxide (LiOH) to adjust the pH value. The detailed LCI is obtained from literature [1].

Flows	Quantities	Amount	Unit
Input			
GLO: lithium hydroxide production ecoinvent 3.7.1	Mass	3.30E-02	kg
DE: Process steam from natural gas 85% Sphera	Energy	1.99E+00	MJ
DE: Nitric acid (98%) Sphera	Mass	9.14E-01	kg
GLO: lithium carbonate production, from concentrated brine ecoinvent 3.7.1	Mass	4.84E-01	kg
DE: Electricity grid mix Sphera	Energy (net calorific value)	8.82E-01	MJ
Europe without Switzerland: market for tap water ecoinvent 3.7.1	Mass	9.14E-01	kg
Output			
Lithium nitrate [Materials]	**Mass**	**1.00E+00**	**kg**
Wastewater [Production residues in life cycle]	Mass	9.14E-01	kg
Carbon dioxide [Inorganic emissions to air]	Mass	2.88E-01	kg

*Table S 18. LCI of lanthanum nitrate [La(NO_3)$_3$*6 H_2O]*

Flows	Quantities	Amount	Unit
Input			
GLO: market for lanthanum oxide ecoinvent 3.7.1	Mass	4.29E-01	kg
Europe without Switzerland: market for water, deionized ecoinvent 3.7.1	Mass	1.35E-01	kg
DE: Process steam from natural gas 85% Sphera	Energy	5.90E-01	MJ
DE: Nitric acid (98%) Sphera	Mass	5.09E -01	kg
GLO: lithium carbonate production, from concentrated brine, ecoinvent 3.7.1	Mass	4.84E-01	kg
DE: Electricity grid mix Sphera	Energy (net calorific value)	6.01E-01	MJ
Europe without Switzerland: market for tap water ecoinvent 3.7.1	Mass	5.09E-01	kg
Output			
Lanthanum nitrate [La(NO_3)$_3$*6 H_2O] [Materials]	**Mass**	**1.00E+00**	**kg**

*Table S 19. LCI of zirconium oxynitrate [ZrO(NO_3)2*6 H_2O]*

Flows	Quantities	Amount	Unit
Input			
Zirconium tetrachloride [$ZrCl_4$], own process model	Mass	6.87E-01	kg
Europe without Switzerland: market for water, deionized ecoinvent 3.7.1	Mass	3.72E-01	kg
DE: Process steam from natural gas 85% Sphera	Energy	5.90E-01	MJ
DE: Nitric acid (98%) Sphera	Mass	3.79E -01	kg
DE: Electricity grid mix Sphera	Energy (net calorific value)	6.88E-01	MJ
Europe without Switzerland: market for tap water ecoinvent 3.7.1	Mass	3.79E-01	kg
Output			
Zirconium oxynitrate [ZrO(NO_3)$_2$*6 H_2O] [Materials]	**Mass**	**1.00E+00**	**kg**
Hydrochloric acid [Waste for recovery]	Mass	4.30E-01	kg

Table S 20. LCI of zirconium tetrachloride [$ZrCl_4$]

Flows	Quantities	Amount	Unit
Input			
AU: zirconium oxide production ecoinvent 3.7.1	Mass	5.29E-01	kg
DE: Chlorine mix Sphera	Mass	6.09E-01	kg
RER: market group for heat, district or industrial, natural gas, ecoinvent 3.7.1	Energy	2.10E+00	MJ
DE: Electricity grid mix Sphera	Energy (net calorific value)	5.04E-01	MJ
DE: Metallurgical coke Sphera	Mass	1.03E-01	kg
Output			
Zirconium tetrachloride [$ZrCl_4$] [Materials]	**Mass**	**1.00E+00**	**kg**
Carbon monoxide [Inorganic emissions to air]	Mass	2.40E-01	kg

Table S 21. LCI of ammonium dihydrogen phosphate [$(NH_4)H_2PO_4$]

Flows	Quantities	Amount	Unit
Input			
DE: Ammonia (NH_3) without CO_2 recovery Sphera	Mass	1.48E-01	kg
DE: Process steam from natural gas 85% Sphera	Energy	1.16E+00	MJ
GLO: market for phosphoric acid, industrial grade, without water, in 85% solution state ecoinvent 3.7.1	Mass	1.00E+00	kg
DE: Electricity grid mix Sphera	Energy (net calorific value)	8.24E-01	MJ
Europe without Switzerland: market for tap water ecoinvent 3.7.1	Mass	1.00E+00	kg
Output			
Ammonium dihydrogen phosphate [$(NH_4)H_2PO_4$] [Materials]	**Mass**	**1.00E+00**	**kg**

*Table S 22. LCI of Lithium acetate [Li(CH_3COO)*2 H_2O]*

Flows	Quantities	Amount	Unit
Input			
GLO: lithium carbonate production, from concentrated brine ecoinvent 3.7.1	Mass	4.40E-01	kg
Europe without Switzerland: market for water, deionized ecoinvent 3.7.1	Mass	1.07E-01	kg
DE: Process steam from natural gas 85% Sphera	Energy	8.29E-01	MJ
DE: Acetic acid from methanol (low pressure carbonylation) (Monsanto process) Sphera	Mass	7.15E-01	kg
DE: Electricity grid mix Sphera	Energy (net calorific value)	7.52E-01	MJ
Europe without Switzerland: market for tap water ecoinvent 3.7.1	Mass	7.15E-01	kg
Output			
Lithium acetate [Li(CH_3COO)*2 H_2O] [Materials]	**Mass**	**1.00E+00**	**kg**
Carbon dioxide [Inorganic emissions to air]	Mass	2.62E-01	kg

Table S 23. LCI of titanium isopropoxide [Ti[OCH(CH_3)$_2$]$_4$]

Credit is given to the ammonium chloride (NH_4Cl) produced by the reaction of titanium tetrachloride (TiCl4) and ammonia (NH_3). Therefore, the input is negative. This means that the environmental impact to produce 0.753 kg NH_4Cl is subtracted from the total balance.

Flows	Quantities	Amount	Unit
Input			
GLO: titanium tetrachloride production ecoinvent 3.7.1	Mass	6.67E-01	kg
DE: Isopropanol Sphera	Mass	8.46E-01	kg
DE: Ammonia (NH_3) without CO_2 recovery Sphera	Mass	2.40E-01	kg
GLO: Ammonium chloride production ecoinvent 3.7.1	Mass	-7.53E-01	kg
Output			
Titanium isopropoxide [Ti[OCH(CH_3)$_2$]$_4$] [Materials]	**Mass**	**1.00E+00**	**kg**

10. Comparison of LATP and LLZO with $LiPF_6$ (in EC/DMC)

Figure S 1 shows the environmental impacts of two model cells. Cell 1 consists of the mixed cathode (LATP/NMC 622) and electrolyte (LLZO) while cell 2 consists of an equivalent quantity of $LiPF_6$ (in EC/DCM) that could theoretically replace LATP and LLZO. To calculate the equivalent quantity of $LiPF_6$, the densities of LATP and LLZO are set in relation to the density of $LiPF_6$ (assumed densities: LLZO: 5.107 g/cm^3, LATP: 2.92 g/cm^3, $LiPF_6$ in EC/DMC: 1.32 g/cm^3). This results in the following quantities required for the model cells of 25.8 cm^2: cell 1 consists of 0.132 g LLZO, 0.226 g LATP, and 0.858 g NMC 622; cell 2 consists of 0.136 g $LiPF_6$ in EC/DMC (50/50) and 0.858 g NMC 622. The quantity of NMC 622 remains the same in the two cells. As expected, the overall environmental impacts are lower if the common $LiPF_6$ were used instead of LATP and LLZO. However, most of the environmental impacts are caused by NMC 622, and those of LLZO are much lower. The overall performance of LATP is similar to that of $LiPF_6$.

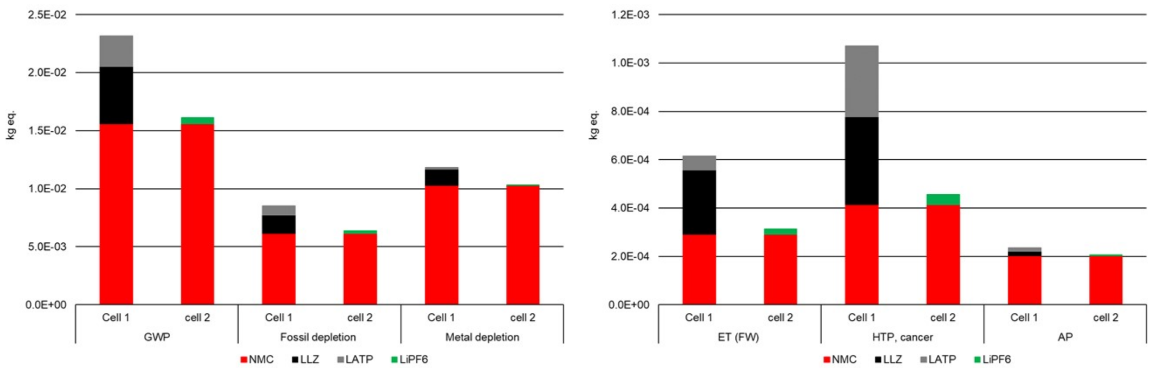

Figure S 1 Environmental impacts of cathode and electrolyte material supply required for two model cells (25 cm^2): cell (1) includes 0.858 g NMC 622, 0.226 g LATP, and 0.132 g LLZO; cell (2) includes 0.858 g NMC 622 and 0.136 g LiPF6 in EC/DMC (50/50)

11. References

1. Deng, Y., et al., *Life cycle assessment of lithium sulfur battery for electric vehicles.* Journal of Power Sources, 2017. **343**: p. 284-295.
2. Sun, X., et al., *Life cycle assessment of lithium nickel cobalt manganese oxide (NMC) batteries for electric passenger vehicles.* Journal of Cleaner Production, 2020. **273**: p. 123006.