Supplementary Information

Life cycle assessment of emerging Ni-Co hydroxide charge storage electrodes: impact of graphene oxide and synthesis route

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Abstract

This document provides all the background information of life cycle assessment carried out to compare three electrodes based on nickel-cobalt hydroxides, abbreviated as NCED, NCED-rGO and NCCP. This supplementary document includes the derivation of stoichiometric representation of active electrode materials which are used as basis to establishing some of the inventory data, the inventory itself normalized per 1g of electrode and functional unit of the study, and data adaptations from other studies modeled as a foreground process. The document further outlines all the absolute values of life cycle impact assessment and choice of background data from the Ecoinvent database given for the parameters for the functional unit and alternative parameters considered in scenario analysis.

Table S1

Calculation of stochiometric formulae for NCED and NCED-rGO, and NCCP electrode, which is used to establish use of nickel and cobalt nitrates

Electrodeposition reaction (NCED and NCED-rGO)	$Ni(NO_3)_2 \cdot 6H_2O \rightarrow Ni^{2+} + 6H_2O + 2NO_3^-$ $Co(NO_3)_2 \cdot 6H_2O \rightarrow Co^{2+} + 6H_2O + 2NO_3^-$ (Nickel, cobalt and water probably in the complex form of

	$[Ni(H_2O)_6]^{2+}/[Co(H_2O)_6]^{2+}$) When cathodic current is applied (input of electrons), at the surface of the conductive substrate: $NO_3^- + 7H_2O + 8\ e^- \rightarrow NH_4^+ + 10OH^-$ There are many possible mechanisms for this reaction, we consider this one found in (Ash, Paramguru, and Mishra 2010; Delmas, Faure, and Borthomieu 1992) $2Ni^{2+} + 4Co^{2+} + 12OH^- + 2(NO_3^-)^{2-} + 3H_2O \rightarrow 6Ni_{0.33}Co_{0.66}(OH)_2{\cdot}(CO_3^{2-}, 2NO_3^-)_{0.33}{\cdot}(H_2O)_{0.5}$ In this case, since there is an excess of nitrate ions instead of carbonates, this is the preferential anion that gets intercalated.
Coprecipitation reaction (NCCP)	$Ni(NO_3)_2{\cdot}6H_2O \rightarrow Ni^{2+} + 6H_2O + 2NO_3^-$ $Co(NO_3)_2{\cdot}6H_2O \rightarrow Co^{2+} + 6H_2O + 2NO_3^-$ (Nickel, cobalt and water probably in the complex form of $[Ni(H_2O)_6]^{2+}/[Co(H_2O)_6]^{2+}$) Addition of NaOH induces 2 Simultaneous reactions: (1) $2Co^{2+} + H_2O_2 \rightarrow 2Co^{3+} + 2OH^-$; redox reaction consisting of two semi reactions: (1.1) $H_2O_2 + 2e^- \rightarrow 2OH^-$ Reduction reaction (1.2) $Co^{2+} \rightarrow Co^{3+} + e^-$ (2) $2Ni^{2+} + 4Co^{3+} + 12OH^- + 2(CO_3^{2-}, 2NO_3^-) + 3H_2O$ $\rightarrow 6Ni_{0.33}Co_{0.66}(OH)_2{\cdot}(CO_3^{2-}, 2NO_3^-)_{0.33}{\cdot}(H_2O)_{0.5}$ (3) $Na^+ + NO_3^- \rightarrow$) $Na^+NO_3^-$ in solution. Note that water and carbonates come from the solution in order maintain charge neutrality and occupy the remaining insterslab space. Carbonates anions come into solution from atmospheric CO_2 with which a spontaneous exchange occurs (Ash et al. 2010; Delmas et al. 1992)

Table S2

Life cycle inventory quantities of NCED, NCED-rGO and NCCP, including material and energy inputs and waste outputs indicated per 1 g of active material (AM) and per functional unit (FU)

		NCED		NCED-rGO		NCCP		
Flows (original naming from Ecoinvent unless modeled as a foreground system)	*Unit*	Amount per 1g AM	Amount per FU	Amount per 1g AM	Amount per FU	Amount per 1g AM	Amount per FU	*Data source*
nickel nitrate hexahydrate	g	8.30E-01	3.16E-02	7.90E-01	8.50E-03	7.60E-01	6.94E-03	Modeled as foreground
cobalt nitrate hexahydrate	g	1.66E+00	6.33E-02	1.58E+00	1.70E-02	1.54E+00	1.41E-02	Modeled as foreground
graphene oxide	g	0.00E+00	0.00E+00	1.00E+00	1.08E-02	0.00E+00	0.00E+00	Modeled as foreground
water, ultrapure	g	1.15E+03	4.39E+01	1.20E+03	1.29E+01	2.45E+02	2.24E+00	Ecoinvent
ethanol, without water, in 99.7% solution state, from ethylene	g	4.89E+01	1.86E+00	4.89E+01	5.26E-01	2.46E+02	2.25E+00	Ecoinvent
sodium hydroxide, without water, in 50% solution state	g	0.00E+00	0.00E+00	0.00E+00	3.44E-02	4.96E-01	4.53E-03	Ecoinvent
hydrogen peroxide, without water, in 50% solution state	g	0.00E+00	0.00E+00	0.00E+00	4.01E+00	2.90E-01	2.65E-03	Ecoinvent
polytetrafluoroethylene	g	0.00E+00	0.00E+00	0.00E+00	4.29E-04	5.00E-02	4.56E-04	Modeled as foreground
carbon black	g	0.00E+00	0.00E+00	0.00E+00	7.38E-05	1.50E-01	1.37E-03	Ecoinvent
steel, chromium steel 18/8	g	3.20E+00	1.22E-01	3.20E+00	8.50E-03	3.20E+00	2.92E-02	Ecoinvent
electricity, medium voltage	Wh	3.33E+02	1.27E+01	3.72E+02	1.70E-02	1.47E+02	1.34E+00	Ecoinvent
transport, freight train	t/km	3.93E-02	1.50E-03	3.99E-02	1.08E-02	1.90E-01	1.73E-03	Ecoinvent
transport, freight, lorry 16-32 metric ton, EURO6	t/km	1.32E-02	5.04E-04	6.86E-03	1.29E+01	6.34E-02	5.78E-04	Ecoinvent

Table S3 (a-d)

Processes modeled as foreground systems including: a) nickel nitrate hexahydrate, b) cobalt nitrate hexahydrate, c) graphene oxide, d) polytetrafluoroethylene

a)

Nickel nitrate hexahydrate	Estimated from Ullmann's Encyclopedia of Industrial Chemistry (Hoydonckx et al. 2007)		
Flow	*Unit*	*Amount per 1kg*	*Data source*
water, ultrapure	g	3.10E+02	Ecoinvent
nickel, 99.5%	g	2.02E+02	Ecoinvent
electricity, medium voltage	kJ	8.25E+02	Ecoinvent
nitric acid, without water, in 50% solution state	g	4.33E+02	Ecoinvent
transport, freight train	t*km	3.81E-01	Ecoinvent
transport, freight, lorry 16-32 metric ton, EURO6	t*km	1.27E-01	Ecoinvent

b)

Cobalt nitrate hexahydrate	Estimated from Ullmann's Encyclopedia of Industrial Chemistry (Hoydonckx et al. 2007)		
Flow	*Unit*	*Amount per 1kg*	*Data source*
cobalt	g	2.02E+02	Ecoinvent
water, ultrapure	g	2.48E+02	Ecoinvent
nitric acid, without water, in 50% solution state	g	8.66E+02	Ecoinvent
transport, freight, lorry 16-32 metric ton, EURO6	t*km	2.14E-01	Ecoinvent
transport, freight train	t*km	6.41E-01	Ecoinvent

c)

Graphene oxide	Sourced from (Cossutta, McKechnie, and Pickering 2017)		
Flow	*Unit*	*Amount per 1kg*	*Data source*
lime, hydrated, loose weight	g	2.28E+04	Ecoinvent
graphite, battery grade	g	7.12E+02	Ecoinvent
potassium permanganate	g	2.14E+03	Ecoinvent
hydrogen peroxide, without water, in 50% solution state	g	1.24E+03	Ecoinvent
electricity, medium voltage	Wh	2.78E+03	Ecoinvent
water, ultrapure	g	2.24E+05	Ecoinvent
sulfuric acid	g	3.02E+04	Ecoinvent

d)

Polytetrafluoroethylene	Sourced from (Jungbluth et al. 2012)		
Flow	*Unit*	*Amount per per 1 kg*	*Data source*
refinery sludge	kg	4.39E+00	Ecoinvent
chlorodifluoromethane	kg	1.81E+03	Ecoinvent
municipal solid waste	kg	1.22E+00	Ecoinvent
transport, freight, lorry 7.5-16 metric ton, EURO6	t*km	2.30E-01	Ecoinvent
heat, district or industrial, natural gas	MJ	3.75E+04	Ecoinvent
heat, district or industrial, other than natural gas	MJ	4.68E+03	Ecoinvent
chemical factory, organics	Item(s)	4.00E-07	Ecoinvent

Table S4

Functional parameters of capacity and cycling stability (number of charge-discharge cycles) at current density of 1 $A{\cdot}g^{-1}$, 4 $A{\cdot}g^{-1}$ and 10 $A{\cdot}g^{-1}$, and capacity fade of 20 and 30%.

	NCED	**NCED-rGO**	**NCCP**
	Capacity		
Current density (A/g)	*mAh/g*	*mAh/g*	*mAh/g*
1	30	96	121
2	26	58	114
4	22	49	102
10	15	43	96
	Cycling stability		
Capacity fade (%)	*#cycles*	*#cycles*	*#cycles*
20	972	1676	1006
30	1804	2557	2048

Table S5

Absolute values of NCED, NCED-rGO, NCCP and upscaled scenario for NCED-rGO involving efficient use of graphene oxide.

Impact categories & indicators	Reference unit	NCED	NCED-rGO	NCCP	NCED-rGO upscaled
Marine ecotoxicity	kg 1,4-DB eq	2.20E-04	6.60E-05	4.37E-05	5.28E-05
Terrestrial ecotoxicity	kg 1,4-DB eq	1.39E-06	4.50E-07	2.74E-07	2.14E-07
Freshwater ecotoxicity	kg 1,4-DB eq	2.20E-04	6.71E-05	4.42E-05	5.44E-05
Fossil depletion	kg oil eq	4.52E-03	1.44E-03	2.56E-03	1.09E-03
Human toxicity	kg 1,4-DB eq	6.01E-03	1.85E-03	1.23E-03	1.36E-03

Water depletion	m3	7.63E-02	2.42E-02	1.23E-02	1.96E-02
Climate Change	kg CO2 eq	1.19E-02	3.97E-03	4.03E-03	2.80E-03
Ionising radiation	kg U235 eq	3.28E-03	1.06E-03	4.20E-04	8.50E-04
Metal depletion	kg Fe eq	1.91E-03	1.18E-03	4.80E-04	4.90E-04
Cumulative energy demand	MJ	2.81E-01	8.94E-02	1.27E-01	6.96E-02

Table S6 (a-c)

Relative impacts of NCED-rGO in comparison with NCED and NCCP when considering different combinations of current densities including 1, 4 and 10 A/g, and criteria for capacity fade of 20% and 30%. Percentage value indicate relative difference in impact for each category. Scenario abbreviation refer to combination of current density (CD) and capacity fade (CF): S-0 – CD 1 $A \cdot g^{-1}$, CF 20% (baseline, depicted in Figure 4 and 5); S-1 – CD 4 $A \cdot g^{-1}$, CF 20%; S-2 – CD 10 $A \cdot g^{-1}$, CF 20%; S-3 – CD 1 $A \cdot g^{-1}$, CF30%; S-4 – CD 4 $A \cdot g^{-1}$, CF 30%; S-5 – CD 10 $A \cdot g^{-1}$, CF 30%.Impacts of NCED-rGO are lower for percentage values preceded by the minus sign and are higher for positive values.

a)

		NCED				
Impact categories	Reference unit	S-1	S-2	S-3	S-4	S-5
Marine ecotoxicity	kg 1,4-DB eq	3.00E-04	4.40E-04	1.40E-04	1.90E-04	2.80E-04
Terrestrial ecotoxicity	kg 1,4-DB eq	1.90E-06	2.79E-06	9.02E-07	1.23E-06	1.80E-06
Freshwater ecotoxicity	kg 1,4-DB eq	3.10E-04	4.50E-04	1.40E-04	2.00E-04	2.90E-04
Fossil depletion	kg oil eq	6.16E-03	9.05E-03	2.92E-03	3.99E-03	5.85E-03
Human toxicity	kg 1,4-DB eq	8.19E-03	1.20E-02	3.88E-03	5.30E-03	7.77E-03
Water depletion	m3	1.04E-01	1.53E-01	4.93E-02	6.73E-02	9.86E-02
Climate Change	kg CO2 eq	1.63E-02	2.39E-02	7.71E-03	1.05E-02	1.54E-02
Ionising radiation	kg U235 eq	4.48E-03	6.57E-03	2.12E-03	2.89E-03	4.25E-03
Metal depletion	kg Fe eq	2.60E-03	3.86E-03	1.23E-03	1.68E-03	2.47E-03
Cumulative energy demand	MJ	3.83E-01	5.62E-01	1.82E-01	2.48E-01	3.63E-01

b)

		NCED-rGO				
Impact categories	Reference unit	S-1	S-2	S-3	S-4	S-5
Marine ecotoxicity	kg 1,4-DB eq	1.20E-04	1.30E-04	3.33E-05	6.53E-05	7.44E-05
Terrestrial ecotoxicity	kg 1,4-DB eq	7.94E-07	9.05E-07	2.28E-07	4.45E-07	5.08E-07
Freshwater ecotoxicity	kg 1,4-DB eq	1.20E-04	1.30E-04	3.39E-05	6.64E-05	7.57E-05
Fossil depletion	kg oil eq	2.54E-03	2.90E-03	7.30E-04	1.42E-03	1.62E-03
Human toxicity	kg 1,4-DB eq	3.25E-03	3.71E-03	9.30E-04	1.83E-03	2.08E-03
Water depletion	m3	4.26E-02	4.86E-02	1.22E-02	2.39E-02	2.73E-02

Climate Change	kg CO2 eq	7.01E-03	7.98E-03	2.01E-03	3.93E-03	4.48E-03
Ionising radiation	kg U235 eq	1.87E-03	2.13E-03	5.40E-04	1.05E-03	1.20E-03
Metal depletion	kg Fe eq	2.07E-03	2.36E-03	5.90E-04	1.16E-03	1.33E-03
Cumulative energy demand	MJ	1.58E-01	1.80E-01	4.52E-02	8.84E-02	1.01E-01

c)

		NCCP				
Impact categories	Reference unit	S-1	S-2	S-3	S-4	S-5
Marine ecotoxicity	kg 1,4-DB eq	4.67E-05	4.96E-05	2.35E-05	2.88E-05	3.06E-05
Terrestrial ecotoxicity	kg 1,4-DB eq	2.92E-07	3.11E-07	1.51E-07	2.43E-07	2.59E-07
Freshwater ecotoxicity	kg 1,4-DB eq	4.72E-05	5.02E-05	2.38E-05	2.88E-05	3.06E-05
Fossil depletion	kg oil eq	2.73E-03	2.90E-03	1.50E-03	1.82E-03	1.93E-03
Human toxicity	kg 1,4-DB eq	1.31E-03	1.40E-03	6.50E-04	8.00E-04	8.50E-04
Water depletion	m3	1.31E-02	1.39E-02	6.99E-03	8.40E-03	8.92E-03
Climate Change	kg CO2 eq	4.30E-03	4.57E-03	2.34E-03	2.90E-03	3.09E-03
Ionising radiation	kg U235 eq	4.40E-04	4.70E-04	2.40E-04	3.00E-04	3.20E-04
Metal depletion	kg Fe eq	5.20E-04	5.50E-04	2.60E-04	3.10E-04	3.30E-04
Cumulative energy demand	MJ	1.36E-01	1.44E-01	7.44E-02	9.04E-02	9.61E-02

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