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Life cycle assessment of lithium-ion batteries and vanadium redox flow batteries-based renewable energy storage systems

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ABSTRACT

Renewable energy has become an important alternative to fossil energy, as it is associated with lower greenhouse gas emissions. However, the intermittent characteristic of renewables urges for energy storage systems, which play an important role in matching the supply and demand of renewable-based electricity. The life cycle of these storage systems results in environmental burdens, which are investigated in this study, focusing on lithium-ion and vanadium flow batteries for renewable energy (solar and wind) storage for grid applications. The impacts are assessed through a life cycle assessment covering the batteries supply phase, their use and end-of-life, with experimental data from test set-ups. The battery composition is investigated in detail as a factor for the final impacts, by comparing two types of cathodes for the lithium-ion battery and the use of recycled electrolyte for the vanadium flow battery. Results indicate that the vanadium-based storage system results in overall lower impacts when manufactured with 100% fresh raw materials, but the impacts are significantly lowered if 50% recycled electrolyte is used, with up to 45.2% lower acidification and 11.1% lower global warming potential. The new lithium-ion battery cathode chemistry results in overall higher impacts, with 41.7% more particulate matter and 52.2% more acidification.

Introduction

As part of the European Green Deal, the European Union (EU) has defined the ambitious goals of reducing 50–55% of its greenhouse gas (GHG) emissions by 2030 and becoming the first continent in the world completely climate-neutral by 2050 [1,2]. To achieve these challenging goals, significant changes will be required in the energy mix of most of the EU countries to reduce the dependency on fossil fuels and their consequent GHG emissions. The increase of renewable energy usage will have an important contribution. Renewable energy sources are the ones renewing themselves naturally at rates that are equivalent or higher than the rates of their use, such as hydropower, marine (tide, wave, ocean), geothermal energy, wind energy, solar energy, ambient heat (heat pumps), biofuels (charcoal, biogas, biodiesel, etc.) and municipal waste [3,4]. The share of renewables in the EU-27 energy consumption was about 18.5% in 2017 and slightly increased to 18.9% in 2018, meaning that an additional increase of 1.1% was required to reach the 20% target by the end of 2020, whilst only 16 countries met or were close to meet their targets in 2018 [5,6]. The share of renewables in the EU energy consumption is expected to further increase to 25% by 2030 and to at least 35% by 2050 [7], leading to the conclusion that significant improvements are required. On the other hand, the share of renewable energy worldwide was 17.3% in 2017 [8], 1.2% below EU levels for the same year, which shows that Europe is indeed ahead in the energy transition.

Renewable energy is unquestionably important to ensure a sustainable society, in which both citizens and industries can benefit and develop while respecting the replenishing rate of natural resources. In this regard, one of the Sustainable Development Goals (SDG) defined by the United Nations (UN) for 2030, the 7th SDG defined as “Ensure access

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to affordable, reliable, sustainable and modern energy for all", states a clear goal of increasing the share of renewable energy in the total energy consumption [9,10]. However, this type of energy is not always readily available and in most cases, supply and demand are not synchronized, as a result of oscillations in energy availability along their cycles (e.g. seasons, day, night) and peaks in the demand. A good example of energy demand peaks is in the use of electricity, an energy source that plays an essential role in modern society. Several household appliances, industrial machinery, personal gadgets, and transport means are powered by this type of energy. Especially in urban and industrial areas, electricity consumption tends to follow a pattern of high demand in certain hours of the day, such as in the morning and early evening, and decreases to minimum values in other moments, for instance at night. To balance supply and demand, the excess of energy generated at moments of low demand should be stored to be used when the demand exceeds the energy availability. The balancing of energy supply and demand is also known as load leveling and helps in the prediction and provision of energy supply [11]. An important application of energy storage and load leveling is in smart grids, essential to guarantee proper infrastructure in urban and industrial areas (i.e. readily available electricity), to minimize impacts of seasonal energy supply, and to upscale the supply capacity. The latter is especially important given the expected increase of renewables as part of the decarbonization of the EU energy mix.

The storage of renewable energy, or more specifically electricity, has been researched throughout the last decade, with a special focus on solar and wind energy as sources and grid-scale applications [12–18]. Several technologies can be applied for renewable electricity storage, including pumped hydroelectric storage (PHS), compressed air energy storage (CAES), superconducting magnetic energy storage, hydrogen storage, flywheels, capacitors and supercapacitors, and batteries, the latter available in different compositions such as lead-acid, nickel–cadmium, sodium-sulfur, lithium-ion, zinc-bromine and vanadium redox flow [11,13–18]. Each technology has its strengths and drawbacks, some of them listed in Table 1.

Although PHS and CAES have the lowest costs, the highest power capacity and longest lifetimes amongst the aforementioned systems, along with reasonable cycle efficiencies, these technologies are only suitable for medium to large-scale applications and rely on specific geographical characteristics, such as water availability for reservoirs and underground area and gas availability for air compression [11,13,15,17]. These factors make PHS and CAES not compatible with decentralized systems, which are relevant in renewable energy production and storage. Besides, these technologies have relatively important environmental drawbacks compared to other systems like generating flooding, air and water pollution and impacts on wildlife [14,17]. Batteries have advantages such as reduced charging time, higher energy density, and shorter response time, which is between milliseconds and seconds, whereas for PHS and CAES it is in the order of seconds to minutes [15].

The use of batteries for energy storage has increased because of their scalability, which allows this technology to be applied in small isolated regions or large energy systems, but also their durability, low maintenance, and lower socio-environmental impacts are important characteristics [11,14,17]. The fast development of batteries for energy storage is expected to significantly increase in the next decade, going from a global capacity of about 11GWh (in 2017) to 100–167 GWh (in 2030) or even 181–421 GWh (in 2030), the latter considering that battery storage will follow the expected two-fold increase of renewables [20]. Lead-acid batteries were created in 1859, being the oldest type of rechargeable batteries and the first battery technology employed in energy storage, serving this purpose for more than 130 years [17–19]. This type of battery is still widely used nowadays, because of its low cost, high

Table 1
Summary of the main characteristics, advantages and disadvantages of storage technologies commonly used for renewable energy [11,13–19].

Storage technology	Power capacity/density	Lifetime	Cycle efficiencies	Operation & maintenance costs	Advantages	Disadvantages
Pumped hydroelectric storage	100–1000 MW	30–60 years	65–85%	$3/kW	Very low costs; High power capacity; Long lifetime.	Medium to large-scale applications; Requires water availability for reservoirs.
Compressed air energy storage	50–300 MW	20–30 years	60–80%	$6/kW	Very low costs; High power capacity; Long lifetime.	Medium to large-scale applications; Requires underground area and gas availability for air compression.
Lead-acid battery	30–50 Wh/kg	3–15 years or 500–1000 cycles	65–80%	$10/kW	Low cost; High efficiency; High recycled content.	Low energy density; Short lifetime; Emission of explosive gas and acid fumes; Limited depth of discharge (25–75%).
Sodium-sulfur battery	150–250 Wh/kg	10–15 years or 2500–40000 cycles	70–90%	$14/kW	High energy density; High efficiency; Long lifetime; Fast response time (milliseconds).	High initial cost; Safety issues.
Lithium-ion battery	200 Wh/kg	10–15 years or 3000 cycles	65–95%	$10/kW	High energy density; High efficiency; Long lifetime; Environmentally friendly.	In large scale (e.g. grid applications) have short lifetimes and elevated costs; High raw materials demand is associated with technology.
Vanadium redox flow battery	16–33 kWh/m^3	5–20 years or 1500–15000 cycles*	70–80%	$28/kW	High efficiency; Long lifetime; Environmentally friendly.	High costs; Low energy density (high area demand); Risk of cross-contamination of electrolyte.

* Although a vanadium redox flow battery in Japan has been reported to withstand more than 200,000 cycles [19], the upper limit reported in the literature is in the range of 10000–15000 cycles [15–17].

efficiency, and high recycled content [15,17]. However, limitations, such as low energy density, short lifetime, emission of explosive gas and acid fumes, and limited depth of discharge (DoD), created space for other types of batteries [11,15–18]. Sodium-sulfur batteries have gained space in electric grid storage since the early 2000s and dominated the grid electricity storage market up to 2014 [19], thanks to their high energy density, high efficiency, lifetime, and fast response time [15,16,19]. Disadvantages of sodium-sulfur batteries are their high initial cost and mostly their safety issues since pure sodium is a hazardous material and is combusted if contacted with air and humidity, besides the danger of short-circuits and exothermic reactions, which can lead to battery temperatures around 2000 °C [14,15,19].

Since their first commercialization in the 1990s, lithium-ion battery (LIB) has gained considerable market share in energy storage, competing directly with sodium-sulfur batteries, because of its high energy density, high efficiency, long lifetime, and for being more environmentally friendly [15–17,19]. The LIB cathode and anode can have different compositions such as lithium-manganese-oxide, lithium-cobalt-oxide, lithium–iron-phosphate, lithium-nickel–cobalt-aluminium-oxide, lithium-titanate-oxide (LTO) and lithium-nickel-manganese-cobalt-oxide (NMC). The NMC is the most used chemistry, accounting for 60% of the batteries used in grid-energy storage, which is a direct influence from the successful use of this type of battery in electric vehicles [21]. On the other hand, large-scale NMC batteries required for grid storage applications have been reported to have a short lifetime and elevated costs [19]. Moreover, because LIB is widely used in several applications, from small electronics to electric vehicles and grid-scale, the demand for raw materials used in these applications has increased significantly and is expected to reach even higher levels in the upcoming years. In the next decade, a yearly increased demand of 30% is expected, resulting in significantly higher consumption of lithium, graphite, cobalt, nickel, and manganese for LIB in 2030 and 2050, compared to current values [7]. A big portion of added LIB capacities will be used for renewables, as the share of renewables in the EU electricity mix is expected to surpass 80% by 2050 [7]. Therefore, it becomes important to look for alternative storage technologies that enable the development and expansion of renewable energy while reducing the pressure on the aforementioned battery raw materials.

Redox flow batteries (RFB) have been reported to be good alternatives for LIB, as they are safe, have a longer lifetime, better scalability, and high recyclability [21–23]. Differently from most batteries, RFB consist of porous electrodes in stacked cell and redox species present in liquid solutions stored in two tanks, which are pumped into the cells where the redox reactions occur. There are different chemistries for RFB, but currently, the vanadium redox flow battery (VRB) is the most commonly used type [21]. The origin of VRB is reported in the 1980s [17], and despite VRB is a technology 10 years older than LIB, its application for renewable energy storage and load leveling is considered recent. Advantages of VRB are its relatively high efficiency, long lifetime as the electrolyte does not deteriorate, scalability, and low environmental impact [15,17,19]. Its main limitation is the low energy density, which leads to high area demand, limiting its application to small to medium scale [15,17]. Another limitation is cross-contamination, which can occur during the operation of RFBs, meaning catholyte and anolyte can cross the membrane and reduce the amount of reactants in one side of the electrolyte whereas the electrolyte volume on the other side is increased, resulting in lower capacity and performance. This is a common issue in RFBs such as iron/chromium, bromine/polysulfide, zinc/bromine and zinc/cerium, but less pronounced in the VRB, since both sides of the electrolyte are composed of vanadium, only in different oxidation states. In practice, cross-contamination of VRB will result in lower efficiency but not in the loss of reactants, as the original species can be recovered [22,24]. As the VRB technology evolves, it may help to diversify battery raw materials, reducing the demand for the classic ones used in LIB.

In spite of the advantages discussed for LIB and VRB, there are substantial environmental impacts along their life cycle, which can be quantified through life cycle assessment (LCA). Although some studies have addressed the environmental impacts of LIB for residential or grid applications [25–32], most of the LCA studies available for LIB, are focused on e-mobility rather than on stationary applications [33–37]. Often, the studies focus on impacts up to the manufacturing stage, not including the use phase and end-of-life (EoL), both of which should be better studied, as these stages may have significant contributions to the overall battery impacts [27,29,38]. The environmental impacts of the VRB have also been studied, however to a lower extent than LIB, and studies comparing LIB and VRB for grid applications consider the LTO type of LIB [22,28,31,39], whereas in practice, the NMC is the most common chemistry for stationary applications. This results in a lack of complete inventory for all the processes required for the production and use of these batteries for renewable energy. Moreover, LCA literature addressing the environmental impacts of LIB and VRB for grid application at the use phase is typically based on literature data, relying on several assumptions to model the reality during the operation of the batteries. Experimental results which provide a more realistic overview of the impacts are still missing.

This study aims at a comprehensive comparison of LIB-based renewable energy storage systems (LRES) and VRB-based renewable energy storage system (VRES), done through i) the elaboration of a life cycle inventory (LCI) for the LRES and VRES, which consist of the LIB and VRB batteries as well as the additional setup components (i.e. inverters, battery monitoring system, etc.); ii) the quantification of impacts along the life cycle of both technologies using LCA; iii) the identification of the environmental hotspots along the life cycle of both LRES and VRES, including use phase and EoL; iv) the comparison of the environmental performance of both technologies for storage of renewable energy, represented by wind energy and photovoltaic (PV) energy. The study relies on experimental data from both LRES and VRES test setups, currently being tested at Engie Research's storage lab, which is a good proxy to estimate the environmental impacts of both technologies at their operation at full-scale.

Materials and methods

Life cycle assessment framework

LCA is a standardized methodology to quantify the environmental impacts of a product or service along its life cycle, considering the use of resources and the emissions from and to the environment [40–42]. The scope of this study consists of a comparative LCA of an LRES and a VRES. An initial assessment for the supply phase of the batteries used in the storage systems is performed (LIB and VRB), where resource extraction, components manufacturing, and battery assembling are considered. The stages listed in this first assessment are then complemented with the use phase of the battery, including additional components external to the battery (e.g. inverter), replacement of battery components, and impacts due to electricity loss during charge/discharge, up to the EoL handling. The assessment becomes then a life cycle assessment of the LRES and VRES energy storage technologies. The addition of the use phase and the EoL of the storage systems in a separate assessment allows a better understanding of the incremental impacts caused at the stages downstream of the batteries production. The functional unit (FU) is the provision of 1 MWh of electricity (AC) over 20 years, with electricity from renewable sources. The life cycle is modeled using SimaPro 9.1.0.11 software and Ecoinvent 3.6 [43] as the background database. The selected method and impact categories are further described in Section "Life cycle impact assessment".

Geographical and technological representation and life cycle inventory data acquisition

The life cycle inventory (LCI), consisting of several inputs (e.g. raw materials and electricity) and outputs (e.g. semi-finished products and emissions to the environment) along the value chain of LRES and VRES is mainly based on primary data, provided by Engie. When primary data regarding production, use, and EoL of the storage systems is not available, additional data is gathered from relevant studies available in the literature or databases. To assess the environmental impacts of the background system, data from the LCA database Ecoinvent 3.6 is used. The complete LCI for LIB and VRB is available in the Supplementary Information (SI).

During the use phase, several charge/discharge cycles occur, with electricity being converted from AC to DC through an inverter, to match the requirements of the batteries (DC) and the grid (AC). The electricity is delivered with a minimum power of 7.5 kW. Renewable energy produced in Belgium is used for charging the LIB and the VRB, as well as for the operation of both batteries. In the base scenario, the use of PV energy to charge and operate both energy storage systems is considered, and a comparison with wind energy is performed as part of the sensitivity analysis (Section "Sensitivity analysis"). During charge/discharge of the batteries, electricity is partially lost due to inefficiencies of the battery systems and the inverter. In this study, only the losses up to storage and delivery to the grid are considered, meaning the impacts are restricted to the battery systems, excluding distribution losses.

It is assumed that each battery-based system performs 300 cycles a year, equivalent to one cycle per day, excluding periods when the battery cannot be fully charged due to climate limitations (e.g. lack of sun). At the end of the batteries lifespan (10 years for LIB and 20 years for VRB), the energy storage systems are dismantled and some of their parts are recycled. In line with common practice in LCA, the processes and impacts of recycling are not included in the scope of this study; it is assumed these impacts are accounted for in the next life cycle where the recycled materials are used. However, an assessment considering the use of recycled raw materials for the VRES electrolyte is performed as part of the sensitivity analysis.

The two batteries considered as part of the storage systems in this study have different characteristics, as summarized in Table 2. The VRB has a significantly smaller capacity than the LIB. This is because the VRB here considered is designed for set-up tests and therefore, has smaller dimensions than a VRB used in grid applications in practice. This can result in overestimation of some impacts, considering large-scale batteries often result in more efficient materials and energy use. The VRB is assumed to show no significant decreased efficiency throughout the 20 years, according to data provided by the manufacturer. On the other hand, the LIB has been reported to have a lifetime ranging from 5 to 15 years [15–17]. For this comparison, an average value of 10 years is assumed.

Table 2
Key parameters of lithium-ion battery (LIB) and vanadium redox flow battery (VRB) of the two renewable energy storage systems compared in the study (based on Engie storage lab tests).

Parameter	LIB	VRB*
Nominal energy capacity (kWh)	1300.0	37.5
Round-trip efficiency (%)	90.0	83.0
Depth of Discharge (%)	85.0	100
Lifetime (years)	10**	20
Discharged energy after 20 years (MWh)***	5758.0	180.0
Total weight (kg)	17726.7	2215.5
Battery mass fraction needed for discharge of 1 MWh	$3.5{\cdot}10^{-4}$	$5.6{\cdot}10^{-3}$

* The VRB here described corresponds to a set-up test scale, therefore smaller than VRB used in real grid applications.
** Considering a period of 20 years, it is assumed that after 10 years the LIB is replaced by a new one.
*** Calculated by multiplying the discharged electricity per cycle (in AC) by the DoD and a total of 6000 cycles executed in 20 years. The efficiency of the inverter is also considered (96.5%).

Life cycle inventory of the LRES

The LRES considered in this study is an energy storage system being tested by Engie for grid application. The LIB contains a graphite anode and a nickel-manganese-cobalt based cathode, with a Ni:Mn:Co ratio of 1:1:1 (NMC 111). The LIB has an energy capacity of 1.3 MWh and consists of a container holding 3762 prismatic cells. Due to the lack of information regarding some components, the LCI of the LRES is mostly based on information available in the literature, adapted to represent the battery configuration used by Engie. The LIB is supplied by Alfen N.V. (The Netherlands) and it is assumed that all additional LRES components are produced by the same company, except for the LIB cells, which are manufactured by Samsung (South Korea). The complete LCI of the LRES from the production of its components to its EoL handling, as well as the processes used in the LCA modeling, are further described in the SI. Considering the fast-evolving market towards new LIB cathode chemistries containing less cobalt, a comparison between the NMC 111 cathode and another one with a Ni:Mn:Co ratio of 8:1:1 (NMC 811) is performed in the sensitivity analysis (Section "Sensitivity analysis").

Life cycle inventory of the VRES

As for the LRES, the VRES considered in the study is under investigation at Engie for grid application. The VRB of the energy storage system has an energy capacity of 37.5 kWh and a total mass of 2215 kg. The VRB is manufactured by Dalian Rongke Power Co., Ltd. (China), and the company also reports to produce the key VRES components (i.e. not acquired from third companies). The vanadium used to produce the electrolyte is extracted and processed in other locations, also in China. The VRES is then sent to Belgium for the use phase. The base scenario considers the VRES produced from primary materials, but given the high recyclability of the VRB, especially of the electrolyte, a sensitivity analysis is performed considering the use of secondary materials. In this alternative scenario, 50% of the electrolyte is assumed to be recycled, but according to Engie, most VRB suppliers indicate that the vanadium used in the electrolyte is sourced from waste streams from other processes, and some suppliers currently propose leasing of the electrolyte. The leasing scheme ensures the electrolyte will be reused in a new VRB after some treatment, meaning that even more than 50% may be recycled. In fact, it has been demonstrated that up to 97% vanadium recovery from the electrolyte can be achieved [23]. Therefore, the use of (partially) recycled electrolyte may become the base scenario in the near future, i.e. when the VRES considered in this study reaches its EoL. However, because the technology is still new for grid applications, a more conservative scenario without recycling the electrolyte is chosen as the current base scenario and compared to an expected future scenario. The sensitivity analysis is further described in Section "Sensitivity analysis". The data related to the production of the VRES is provided by Engie whenever available and complemented with data from another LCA study performed for VRB [22]. The final LCI for the VRES has been thoroughly verified by Engie to match the characteristics of the energy storage system used by them. The complete LCI of the VRES from the production of its components to its EoL handling, as well as the processes used in the LCA modeling, are further described in the SI.

Life cycle impact assessment

The quantification of the environmental impacts for both LRES and VRES is done using the ReCiPe 2016 Midpoint (H) calculation method [44], considering the following impact categories: global warming (GW), fine particulate matter formation (FPMF), terrestrial acidification (TA), mineral resource scarcity (MRS), fossil resource scarcity (FRS) and human toxicity (HT), the latter considering both carcinogenic and non-carcinogenic. Cumulative energy demand (CED) is also considered as an impact category and covers the energy demand for renewables (biomass, water, wind, solar, and geothermal) and non-renewables (fossil, biomass, and nuclear). These categories were selected after thorough consultation with the stakeholders from Engie and provide good comparability with impacts reported in previous LCA studies of LIB and VRB [22,25,48,26,29,31,32,39,45–47]. The selection of these categories is also aligned with the recommendations for the life cycle impact assessment (LCIA) of products in the European context [49].

One of the most commonly used categories in LCIA is GW. As the public concern with climate change has increased in recent years, GW is considered as an important impact of a technology on the environment, often being used to select one technology over its alternative. The impacts on the air quality are indicated by FPMF and since some processes make use of sulfur compounds, TA becomes also a relevant impact to be assessed. As the production of both technologies requires several metals and chemical compounds extracted from nature, MRS and FRS seem relevant impact categories. Moreover, considering that the production of some battery components may offer toxicity risks, HT is also selected. Additionally, CED is considered given the high energy need for some processes along the value chain, such as mining of materials.

Sensitivity analysis

Based on the results for the base scenario, three additional scenarios are performed to compare the impacts of some parameters considered in the study. First, a scenario comparing the renewable electricity source is developed, in which the electricity at the use phase is compared between PV energy (base scenario) and wind energy. Moreover, taking into account the fast-changing LIB battery cathode composition, a comparison between a LRES with an NMC 111 cathode (base scenario) and an NMC 811 cathode is performed. Finally, a comparison between a VRES with battery electrolyte made of primary raw materials (base scenario) to a VRES with electrolyte produced from partially recycled materials (50%), is added.

Results and discussion

Life cycle inventory

The mass distributions for the LIB and VRB components are illustrated in Fig. 1, and the energy input/output ratio per MWh delivered is also reported for each battery (under each pie chart). Tables with the composition values in mass (kg) and wt.% for both batteries are available in the SI (Table S1 and Table S2). More than half of the LIB mass (53%) corresponds to the cell components (anode, cathode, membrane, electrolyte, and others). In the case of the VRB, the electrolyte represents about 69% of the total mass of the battery, with the steel housing being the second-largest battery component (15% of the total mass). The power subsystem components of the VRB (membranes, electrodes, bipolar plates, current collectors, cell frames, gaskets, and stack frame) are equivalent to 13% of the battery. The electrodes (anode and cathode) of the LIB correspond to a significantly larger share of the battery compared to the same components in the VRB. Whereas the electrolyte of the VRB represents a much larger share of this battery than is the case for the electrolyte of the LIB. These differences result from the different operational configurations of both batteries.

The key parameters of both batteries are described in Section "Geographical and technological representation and life cycle inventory data acquisition". The electricity input/output ratio for the batteries is the ratio between the incoming electricity (for charging and operation) and the outgoing electricity (to the grid), as described in the LCIs for the use phase of the batteries (Table S17 and Table S46 in the SI). For the LIB, this results in an electricity input/output ratio of 1.36 or total efficiency of 73.5% whereas, for the VRB, the ratio is 1.44 and total efficiency of 69.4%. The ratios or total efficiencies were calculated considering the round-trip efficiencies to charge the batteries, their DoD (Table 2), the required electricity for operation of each storage system as well as the efficiency of the inverter. Although the DoD of the VRB is 100% against 85% for the LIB, the total efficiency of the VRB is lower, as a result of a lower round-trip efficiency, which has been reported to be a key parameter at the use phase of the battery [29]. The efficiency of the battery has also been reported to have an impact on life cycle carbon emissions [31].

The LCIs for the production of the LIB (per kg of battery rack) and the VRB are reported in Table 3. The VRB has a longer list of components than the LIB, but it should be noted that transport to the place of operation is already included in the LCI of the VRB, whereas the transport of the LIB is reported separately in another LCI table available in the SI with the corresponding number of LIB battery racks required for the assembling of the battery (Table S16). The battery components for which an LCI was developed in the context of this study (i.e. no matching processes were available in the Ecoinvent database) are also described in the SI.

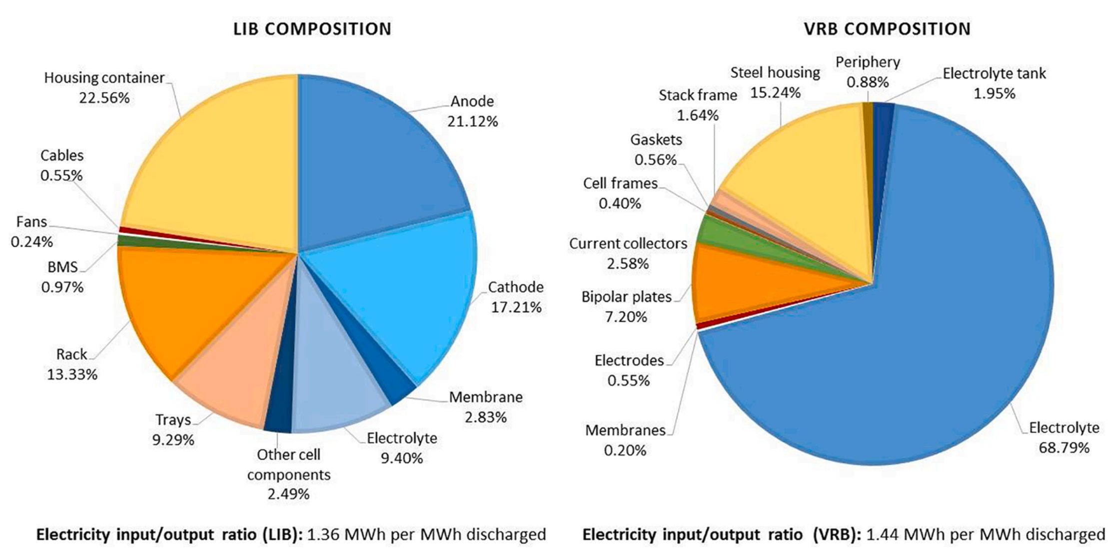

Fig. 1. Composition of the lithium-ion battery (LIB) and vanadium redox flow battery (VRB) in wt.%. In the LIB pie chart, blue parts indicate the cell components, "Other cell components" refers to aluminium, polyethylene, and nitrogen inputs for cell manufacturing, and BMS stands for battery monitoring system. In the VRB pie chart, blue parts indicate the energy subsystem. Membranes, electrodes (anode and cathode), bipolar plates, current collectors, cell frames, gaskets, and stack frame are related to the power subsystem. "Periphery" refers to pumps, pipes, cables, and fan. Electricity input/output ratio is the electricity input divided by the electricity output, considering losses due to inefficiencies.

Table 3
Life cycle inventory for the production of 1 kg of battery rack filled used in the lithium-ion battery (LIB) and of 1 vanadium redox flow battery (VRB), including transport of the VRB to the place of operation. The LIB battery rack transport to the place of operation is further described in the supporting information.

	Subinventory/dataset	Amount	Unit
	Inputs		
LIB	Cable, unspecified {GLO}| market for | Cut-off, U	$7.1{\cdot}10^{-3}$	kg
	Computer, laptop {GLO}| market for | Cut-off, U	$3.9{\cdot}10^{-3}$	Item(s)
	Rack housing, LIB	0.2	kg
	Fan, for power supply unit, desktop computer {GLO}| market for | Cut-off, U	$3.1{\cdot}10^{-3}$	kg
	Metal working factory {GLO}| market for | Cut-off, U	$4.58{\cdot}10^{-10}$	Item(s)
	Transport, freight train {RoW}| market for | Cut-off, U	0.6	t·km
	Transport, freight, lorry 16–32 metric ton, euro6 {RoW}| market for transport, freight, lorry 16–32 metric ton, EURO6 | Cut-off, U	0.1	t·km
	Battery tray, LIB	0.8	kg
	Outputs		
	Battery rack filled, LIB	1.0	kg
	Inputs		
VRB	Bipolar plate, VRB	159.6	kg
	Copper cable, VRB	11.1	kg
	Current collector, VRB	57.3	kg
	Electrolyte, VRB	1524.0	kg
	Electrolyte tank, VRB	43.2	kg
	Fan, VRB	2.6	kg
	Gasket, VRB	12.4	kg
	Nafion membrane, VRB	4.4	kg
	Electrode, VRB	12.2	kg
	Pipes, VRB	0.8	kg
	Pump, VRB	5.0	kg
	PVC cell frame, VRB	8.9	kg
	Sawnwood, board, hardwood, dried (u = 10%), planed {GLO}| market for | Cut-off, U	0.1	m^3
	Stack frame	36.4	kg
	Steel housing	337.7	kg
	Transport, freight, lorry 16–32 metric ton, euro6 {RoW}| market for transport, freight, lorry 16–32 metric ton, EURO6 | Cut-off, U	39.4	t·km
	Transport, freight, lorry 16–32 metric ton, euro6 {RoW}| market for transport, freight, lorry 16–32 metric ton, EURO6 | Cut-off, U	131.3	t·km
	Transport, freight, sea, container ship {GLO}| market for transport, freight, sea, container ship | Cut-off, U	44437.2	t·km
	Outputs		
	Manufactured & transported battery, VRB	1.0	Item(s)

Life cycle impact assessment

The LCIA results are reported first for the complete production process (all components) and transport of each battery contained in the two assessed energy storage systems to their place of operation (LIB and VRB supply phase), and second the production process to the EoL of the storage systems (life cycle). The total environmental impacts of LRES and VRES storage systems per impact category at the base scenario are listed in Table 4. For both batteries and in all impact categories, a remarkable increase is observed from the supply phase to the life cycle. This is a result of the impacts at use and EoL phases, which have been reported to have a large share of the total impacts of battery-based storage systems for stationary applications, with recommendations for further investigation [27,38]. A comparison between the energy storage systems and their impacts are further discussed in Sections “Impacts of supply phase of LIB and VRB” and “Life cycle impacts of LRES and VRES”. The results of the sensitivity analysis are discussed in Section “Sensitivity analysis”. In the case of the VRES, the production of the battery electrolyte results in products that are not used for the VRES, the so-called co-products. The impacts were proportionally allocated between useful products for the VRES and co-products using the economic allocation method, which is further described in the SI (Table S36).

Table 4
Overall scores of lithium-ion battery (LIB) and vanadium redox flow battery (VRB) at battery supply phase. Overall impacts of LIB-based renewable energy storage systems (LRES) and VRB-based renewable energy storage system (VRES) over the technologies life cycle, considering the production of components, use, and end-of-life. The impacts are reported per impact category at the battery supply phase and storage system life cycle. The impacts are reported considering the provision of 1 MWh of electricity (AC) over 20 years, with electricity from renewable sources (functional unit).

	Supply phase		Life cycle	
Impact category	LIB	VRB	LRES	VRES
Global warming (kg CO_2 eq.)	56.3	57.0	95.0	100.8
Fine particulate matter formation (kg $PM_{2.5}$ eq.)	0.3	0.2	0.4	0.3
Terrestrial acidification (kg SO_2 eq.)	1.0	0.6	1.2	0.8
Human toxicity (kg 1,4-DCB)	162.4	120.9	218.2	173.8
Mineral resource scarcity (kg Cu eq.)	5.0	4.8	5.9	5.9
Fossil resource scarcity (kg oil eq.)	13.1	15.1	23.0	25.5
Cumulative energy demand (MJ)	766.4	801.6	2734.1	3129.5

Impacts of supply phase of LIB and VRB

The most significant impacts at the supply phase of each battery component are illustrated in Fig. 2. The impacts include the infrastructure requirements, such as energy requirements for the production of components and their transport to the battery manufacturing company.

For the LIB, the term “Trays” includes all impacts related to the production of the trays, except for the cells. Similarly, the term “Racks” represents the production of the racks, without the trays, battery monitoring system (BMS), cables, and fans, for which impacts are illustrated separately. “Transport” indicates the transport from the battery manufacturing company, in The Netherlands to Engie, in Brussels. The transport of the cells from South Korea to the battery manufacturing company is included in “Cell (other components)”. In the case of the VRB, “Transport” covers the shipping of the entire battery from China to Belgium, as well as the transport within Belgium, and “Others” refer to cell frames, pumps, pipes, and wooden packaging box.

For all impact categories, the cathode and anode of the LIB are the components responsible for the majority of the environmental impacts, together accounting for 60 to 85% of the total impacts per category. In the case of the VRB, the most remarkable impacts are related to the

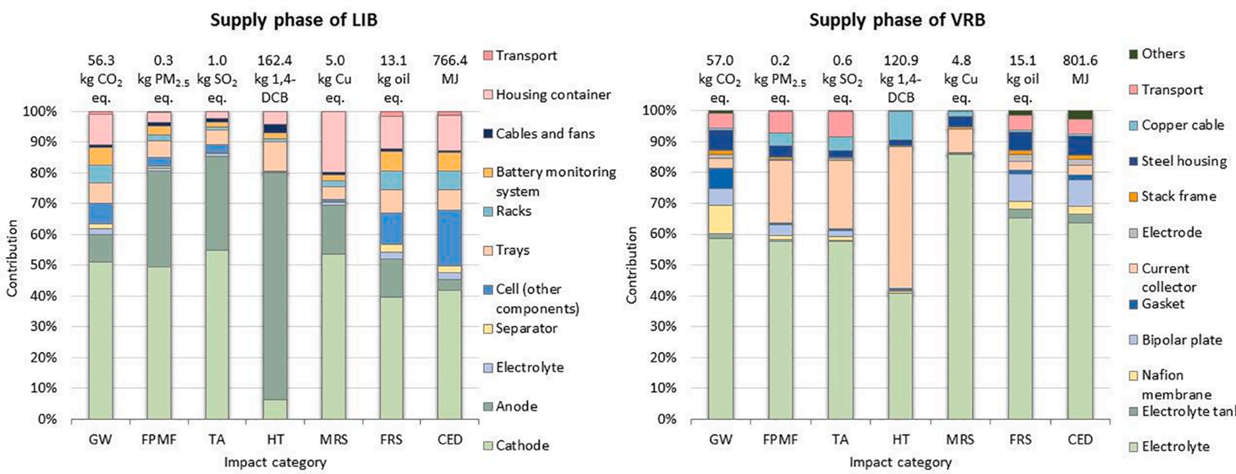

Fig. 2. Environmental impacts related to the supply of the lithium-ion battery (LIB) and the vanadium redox flow battery (VRB) batteries, including their transport to the place of operation. The impacts are represented per impact category, with respective impact share (%) of each battery component to the overall environmental impact (100%). For the LIB, "Cell (other components)" includes aluminium, polyethylene, and nitrogen inputs for cell manufacturing as well as transport from South Korea to the Netherlands. For the VRB, "Others" refer to cell frames, pumps, fans, pipes, and the wooden packaging box. The impacts are reported considering the provision of 1 MWh of electricity (AC) over 20 years, with electricity from renewable sources (functional unit). Impact categories: global warming (GW), fine particulate matter formation (FPMF), terrestrial acidification (TA), carcinogenic and non-carcinogenic human toxicity (HT), mineral resource scarcity (MRS), fossil resource scarcity (FRS), cumulative energy demand (CED).

electrolyte, with impact shares between 40 and 85% of the total impacts per impact category. The most significant impacts of LIB and VRB are further discussed per impact category.

Impacts on global warming. Although the cathode mass share in the LIB is 4% lower than the anode, its impact on GW is almost six times higher (51%) compared to the anode impacts, as a result of the polytetrafluoroethylene (PTFE) material used as the binder, for which production is related to the emission of gases contributing to global warming, such as chlorofluorocarbon (CFC), hydrofluorocarbon (HFC) and hydrochlorofluorocarbon (HCFC). The use of PTFE has been reported to result in high environmental impacts also in VRB storage systems and its reduction or substitution is recommended [50]. The cathode production also entails high energy demand, with hard coal as the energy source (electricity grid of South Korea). Also, several materials used in the production of the cathode have a considerable contribution to global warming, such as nickel sulfate, cobalt sulfate, and manganese sulfate. The impacts of the anode production on GW (8.8%) are related to the copper material, which is about half of the anode composition. Copper mining is a highly energy-intensive process. Other remarkable impacts come from the housing container, trays, BMS, and racks, as all these components rely on metallic materials such as steel, aluminium, copper, and printed circuit boards.

In the case of the VRB, the electrolyte has the biggest contribution (58.7%), mostly related to the production of vanadium pentoxide. The production of vanadium pentoxide bearing cast iron and recovery of vanadium pentoxide from vanadium slag have major impacts [22]. These processes take place in China, where the electricity mix is hard coal-based. The Nafion membrane follows the electrolyte in terms of impacts (9.5%), which is mainly a result of the tetrafluoroethylene production, more specifically, the chlorodifluoromethane production process. In this process, CFCs, HFCs, and HCFCs are emitted, which have high contributions to global warming with CO_2 equivalent values in the range of thousands or tens of thousands [51]. Tetrafluoroethylene is also used in the gaskets, reason why this component also has a noticeable contribution (6.6%). Components made of steel, such as the steel housing and the bipolar plate, also have significant impacts, due to the electricity demand for their production and the energy source (hard coal). Transport plays a role in the overall impacts, due to the use of oil fuel in the shipping of the battery from China to Belgium.

Impacts on fine particulate matter formation and terrestrial acidification. Taking into account that the distribution of the impacts per battery component are similar for FPMF and TA for both LIB and VRB, and because these two impact categories are somehow related, their results are discussed together. For the LIB, the biggest impacts are related to the cathode (49.3% for FPMF and 54.7% for TA), which requires nickel and cobalt, the former being extracted from sulfidic ores. The mining of these metals results in the emission of SO_2, which in turn leads to the formation of secondary $PM_{2.5}$ aerosols. The anode of the LIB shows the second biggest impact (31.3% for FPMF and 30.6% for TA), which is mostly caused by the use of copper as collector foil. The mining of copper ore and its further processing are related to direct $PM_{2.5}$ emissions as well as secondary $PM_{2.5}$ resulting from SO_2 emissions. The contribution of the trays (copper busbars), housing container, and BMS to FPMF and TA is due to the emissions caused during the production of the metals embedded in these components.

The electrolyte of the VRB shows the highest impact scores for both FPMF (57.8%) and TA (57.6%), as a result of the SO_2 emissions during the production of vanadium oxide, for which hard coal is used as an energy source, leading to $PM_{2.5}$ emissions. The current collector has the second-highest impact score for both FPMF (20.3%) and TA (22.5%), because of the emissions during extraction and processing of copper, which is also the reason for the impacts of the copper cable. Similarly to the LIB, the transport represents a significant share of the impacts on FPMF (6.8%) and TA (8.2%), which is related to the transport by ship of the whole battery from China to Belgium and the emissions resulting from combustion of heavy fuel oils. The impacts related to the steel housing and the bipolar plate are a result of the emissions at the production phase of the steel, with the energy source being hard coal.

Impacts on human toxicity

The main impact of the LIB on HT comes from the anode (73.8%), followed by the trays (9.4%). Both components contain copper (anode substrate and copper busbars in the trays), being the main source of the impacts, as it has been reported in the literature [22,45]. Following the anode and trays, the cathode also contributes substantially (6.2%), which is related to the production of cobalt sulfate and nickel sulfate, in line with information in the literature [45]. Housing container, cables, and fans, BMS, and racks all together have a contribution of 9.7%, as a result of their high metal content.

For the VRB, the impacts on HT come mostly from copper components such as the current collector (45.9%) and the copper cables (9.1%). The electrolyte has the second-highest contribution (40.7%), due to the production of vanadium pentoxide and its electricity demand, besides the impacts from the mining activities [22]. The impacts of the steel housing are also noticeable (2.0%) and are attributed to the production of the steel.

Impacts on mineral resource scarcity

The cathode of the LIB holds the biggest impact of this battery on MRS (53.4%), with most of the impacts coming from cobalt sulfate production followed by nickel sulfate production, sodium hydroxide, and manganese sulfate. The anode impact contribution (16.1%) is once more attributed to the copper content of this component. The housing container responds for the second-highest impact score (19.7%), which is related to the composition of the container, which is made of steel. Trays, BMS, racks cables, and fans have minor impacts compared to the other components, but their metals requirement, such as gold and chromium for the BMS [22], also plays a role in resource scarcity.

For the VRB, the electrolyte accounts for the majority of the impacts on MRS (85.9%), in line with previous impacts reported for this component [22]. The impacts are mainly related to the vanadium bearing magnetite production process, which makes use of titanium dioxide and vanadium. The copper required for the production of the current collector and copper cable accounts for additional impacts (7.7 and 1.5%, respectively). Next to copper, other metals present in the ore are extracted, such as lead, zinc, silver, gold, and molybdenum, which increase the impacts on mineral scarcity. The steel housing has a minor contribution (3.7%), but noteworthy.

Impacts on fossil resource scarcity and cumulative energy demand

The distribution of the impacts on FRS and CED for both batteries follow a similar pattern, the reason why the discussion of the results is combined. For the LIB, the cathode shows the biggest contribution for FRS (39.5%) and CED (41.8%). These impacts can be attributed to the high energy demand for cathode production, which takes place in South Korea with the use of hard coal. Besides, the production of N-methyl-2-pyrrolidone used in the cathode has a high electricity and heat demand. On top of the impacts during cathode production, the local electricity and heat demand at the mining operations for metal extraction also contribute to the overall impacts of the cathode. The anode shows a bigger impact on FRS (12.4%) than on CED (3.33%). In the case of FRS, the impacts are divided almost equally between graphite and copper production. Hard coal is used as an energy source in the production of coke, which is needed for the manufacturing of graphite, while the impact of copper is attributed to the energy needed for mine operation. Although a significant amount of fossil energy is used for anode materials, when all types of energy used in the production chain are combined (CED) the contribution of the anode becomes less pronounced, as other components are more energy-intensive and a more wide range of energy sources are used, i.e. besides fossil. For instance, the manufacturing of other cell components, including aluminium, polyethylene, and nitrogen inputs, has a bigger impact on CED (18.0%) than the anode (3.3%). Also, the impacts of these other cell components on CED are higher (18%) compared to FRS (10.3%). This is because not all the energy supply for these processes are fossil, part of them is also renewable, nuclear, or biomass energy, as energy from the global market is used for some of these components (e.g. polyethylene and aluminium). The production of metallic materials such as steel, copper, and printed circuit boards for the racks, trays, cables, fans, and BMS is also an energy-intensive process, altogether, resulting in similar impacts for FRS (31.9%) and CED (32.1%). The share of the impacts attributed to the housing container is the highest amongst the metallic components and shows similar values for FRS (10.6%) and CED (11.3%), resulting from the production of steel used for this battery component.

Looking at the results for the VRB, the electrolyte once more shows the highest impacts for both FRS (65.2%) and CED (63.7%), which is attributed to the production of vanadium pentoxide, a highly energy-intensive process. The petrol used in the vanadium bearing magnetite process and the hard coal-based electricity mix are the main contributors, next to the heat inputs from natural gas. The bipolar plate has the second-highest contribution to both FRS (8.8%) and CED (8.5%), due to the hard coal requirements for graphite production and the usage of high-density polyethylene (HDPE). HDPE is used as binder material for the bipolar plate and involves a high share of crude oil and hard coal in its production process. The production of steel is highly energetic and involves the use of coke for pig iron production, which is a precursor of the final steel. On top of that, the electricity needed for the sheet rolling process is provided by fossil resources. Therefore, the steel housing has a considerable share of the total battery impacts on FRS (6.2%) and CED (6.2%). The overseas transport of the battery from China to Belgium also has a significant impact on both FRS (4.7%) and CED (4.7%).

Life cycle impacts of LRES and VRES

The relative contributions of the LRES and VRES in a life cycle for all assessed impact categories are shown in Fig. 3. Additional aspects are

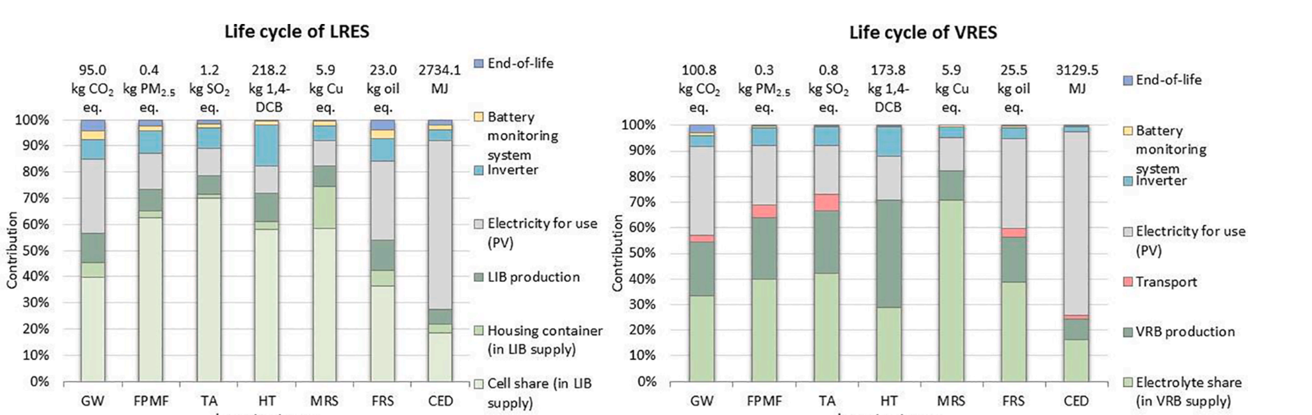

Fig. 3. Contribution of lithium-ion battery (LIB) and vanadium redox flow battery (VRB) components to the overall life cycle environmental impacts, along with life cycle phases of the LIB-based renewable energy storage systems (LRES) and VRB-based renewable energy storage system (VRES) resulting in significant impacts. The impacts are represented per impact category and respective share (%) of each component or life cycle stage to the overall environmental impact (100%). The impacts are reported considering the provision of 1 MWh of electricity (AC) over 20 years, with electricity from renewable sources (functional unit). Impact categories: global warming (GW), fine particulate matter formation (FPMF), terrestrial acidification (TA), mineral resource scarcity (MRS), fossil resource scarcity (FRS), cumulative energy demand (CED).

considered when looking at the use phase and EoL of the batteries, namely the inverter, electricity for use (PV energy), and the EoL handling of the components of the energy storage systems. For the LRES, the battery replacement is taken into account in the LIB supply phase. Since the LIB cell and housing container are major contributors in all the assessed impact categories at the supply phase, their impact shares are indicated by the light green bars. Similarly, since the electrolyte of the VRB has a significant contribution in all the impact categories at the battery supply phase, it is illustrated by the light green bars. The replacement of components for the VRES has a relatively low impact and therefore is not represented. This is because only the fans and pumps are replaced over the 20 years lifespan.

All impact categories assessed for LRES and VRES show significantly increased impacts when the use and EoL phases are considered. Regarding GW, about 55% of the life cycle impacts come from the production of the batteries. The electricity for use represents 29.6% of the life cycle impacts on GW for LRES and 34.5% for VRES, due to the production of the PV system, which consists of multi-crystalline PV panels, requiring the production of silicon. The most critical processes in terms of GW are the transformation of metallic silicon into solar-grade silicon and the panel assembling [52]. The solar-grade silicon production process is energy-intensive and the electricity mix in China, where most of the PV systems are manufactured [53], is mainly based on hard coal. Moreover, the mounting system of PV panels requires aluminium and steel, both of which have direct emissions of CO_2 at the production stage. Nevertheless, the GW impact of PV systems depends on many factors like the solar panel orientation and angle, the type of solar cells, and the installation [54].

The high electricity demand and the electricity source (hard coal) for the production and mounting of the PV panels result in a high fossil resource depletion. The impact share of electricity for use on FRS is 31.3% for LRES and 35% for VRES. The hard coal-based electricity mix in China used for the production of PV panels and the corresponding mounting systems are linked to direct emissions of fine particulate matter and acidifying compounds, which results in impacts of the electricity for use in the FPMF and TA impact categories. The impact share from electricity use on FPMF is 14.1% for LRES and 22.8% for VRES, considering the life cycle of the technologies. The impacts on HT from electricity for use correspond to 10.8% for LRES and 16.8% for VRES. This is related to the toxicity at the mining stage of the metals used in the PV cells, such as gold, copper, tin, and silver, as well as to the potential release of these metals into the environment during the use phase [55]. PV systems rely on several functional metals [27], resulting in depletion of mineral resources, with a contribution share to MRS of 10.2% for the LRES and 12.8% for the VRES. The impacts of electricity for use on CED include the energy demand for production of the PV panels, which is highly energy-intensive, but also the renewable energy used during the life cycle of the batteries. For the LRES, the electricity for use has a total share of 66% of the total CED, with 50.5% being from renewable sources. In the case of the VRB, the electricity for use has a contribution of 71.5% to the total impacts on CED in a life cycle, with 54.6% as renewable energy. Although the environmental impact of PV systems seems to have a high contribution to the life cycle of batteries, using fossil fuels as energy source instead of renewables for the grid would result in larger environmental impacts, as the impacts of the energy storage system are directly related to the characteristic of the grid [29,54].

The inverter has a remarkable impact in most of the impact categories, due to its metallic components such as copper, an integrated circuit, and a capacitor, which in turn require several mineral resources, resulting in high energy demand, atmospheric emissions, and depletion of these materials. The extraction and refining of the metals used for the inverter components also have significant impacts on HT. The EoL has a rather minor impact for all impact categories, as all metals and steel are assumed to be almost completely recycled (95%). Noticeable impacts are observed on GW, FPMF, and CED, mostly for the EoL of the LRES. This is because the cells are assumed not to be recycled and need treatment before their disposal, which in turn demands energy and results in emissions of particulate matter after incineration.

Table 5
Total environmental impacts per impact category considering the life cycle of the lithium-ion battery-based renewable energy storage system (LRES) and vanadium redox flow battery-based renewable energy storage system (VRES) with two different renewable energy sources, photovoltaic (PV) and wind energy. The impacts are reported considering the provision of 1 MWh of electricity (AC) over 20 years, with electricity from renewable sources (functional unit), and were obtained from the model described in this study.

Impact category	LRES		VRES	
	PV energy	Wind energy	PV energy	Wind energy
Global warming (kg CO_2 eq.)	95.0	72.2	100.8	72.4
Fine particulate matter formation (kg $PM_{2.5}$ eq.)	0.4	0.4	0.3	0.3
Terrestrial acidification (kg SO_2 eq.)	1.2	1.1	0.8	0.7
Human toxicity (kg 1,4-DCB)	218.2	199.4	173.8	150.5
Mineral resource scarcity (kg Cu eq.)	5.9	5.5	5.9	5.4
Fossil resource scarcity (kg oil eq.)	23.0	17.3	25.5	18.4
Cumulative energy demand (MJ)	2734.1	2389.5	3129.5	2702.3

Sensitivity analysis

The results of the base scenario highlight the battery components or processes at the use phase and EoL that determine most of the total impacts of the technologies. Therefore, the sensitivity analysis is performed to assess the impacts of changing impact-relevant battery components or processes related to the operation of the LRES and VRES.

Electricity source for use

The electricity source at the use phase has a substantial share of all impact categories for both LRES and VRES (Fig. 3). A comparison is carried out considering PV energy as a renewable source (base scenario) and wind energy. The overall environmental impacts for both scenarios are listed in Table 5.

For all impact categories and both the LRES and the VRES storage systems, the impacts of the electricity produced from wind turbines are lower than the impacts resulting from PV systems. Wind electricity has been reported to have a lower carbon footprint than PV-based energy, which is related to the quick aging of PV cells and the need for replacement [31]. The inefficiencies of the batteries should be taken into account, as LIB requires less energy input to deliver 1 MWh (1.36 MWh) than the VRB (1.44 MWh), as previously discussed (Fig. 1). The LIB requires relatively less energy input, so the effect of using a more environmentally friendly energy source is less pronounced for LRES than for the VRES.

Some of the impacts are also related to the production of renewable energy harvesting systems. For instance, the production and mounting of PV systems have been reported to use more fossil fuels and cause higher GHGs emissions than wind turbines [56], which explains their higher GW, FRS, and CED impacts compared to wind turbines. Moreover, the combustion of fossil fuel emits sulfur dioxide and nitrogen oxides [56], resulting in higher FPMF, TA, and HT impacts as more fossil fuels are needed. The PV cells contain toxic chemicals and metals, like lead and cadmium, and once the systems reach the EoL, they might result in additional HT impacts. In terms of mineral resources, PV systems and wind turbines require several metals, which results in similar MRS impacts, but slightly higher impacts for PV systems. Wind turbines require large amounts of iron, steel, copper, and led, whereas PV panels make use of copper, silicon, gold, tin, and silver [55,56].

From the seven impact categories assessed for the life cycle of the batteries, the VRES scores better in four, considering both PV or wind energy. The higher impacts of the VRES in some impact categories (GW, FRS, and CED) can be related to the electrolyte production and the higher internal inefficiencies of the VRB. However, overall, the VRES shows a better environmental performance than the LRES over the life cycle.

Composition of the LRES cathode and VRES electrolytec3

The production of the LIB cathode has the largest contribution to most of the impact categories considered (Fig. 2), being a significant source of impacts at the life cycle of the LRES (Fig. 3). The cathode composition at the base scenario consists of an NMC 111, as this is the most widely used chemistry for LIB. However, given the environmental and social concerns regarding the sourcing and supply of battery raw materials [57] and the expected increase in demand for some battery raw materials, especially cobalt, for which demand for batteries is expected to increase two-fold by 2030 and more than four-fold by 2050 [7], an alternative cathode chemistry with less cobalt and manganese is considered. The alternative cathode is an NMC 811, which is the NMC battery with the lowest content of manganese and cobalt in relation to nickel. This new battery chemistry is expected to have an increasing market share in the upcoming years, next to the other new NMC chemistries [58].

Different from the LIB, the VRB has its electrolyte as the component resulting in most of the environmental impacts assessed (Fig. 2). The results from the life cycle impacts in the base scenario refer to a VRES using an electrolyte produced from primary raw materials (Fig. 3). Since the electrolyte is claimed to be completely recyclable, a sensitivity analysis is performed to compare the life cycle impacts of the VRES with electrolyte produced from primary raw materials (base scenario) and partially recycled electrolyte (50%). The recycling of the electrolyte was modeled considering that a re-processing and purification are required, which in turn demands electricity use and transport. The quantification of the inputs for the recycling processes was based on a relevant study [22], but little is currently known about the electrolyte recycling impacts. The recycling process is considered to take place in Belgium and the LCI for the recycling process of the electrolyte is available in the SI (Table S49). No electrolyte is considered as input in this process since only the impacts of the recycling process are assessed. The losses during re-processing and purification are considered to be included in the EoL phase of the life cycle in the base scenario.

A comparison of the base scenario, where the LRES has an NMC 111 cathode and the VRES has 100% fresh electrolyte, to a new scenario, where the LRES has an NMC 811 cathode and the VRES electrolyte is composed of 50% fresh and 50% recycled electrolyte is illustrated in Fig. 4. In the comparison, the NMC 111 is normalized and the impacts of the NMC 811 and VRES with fresh and partially recycled electrolyte are reported referring to the NMC 111 results.

Remarkably, the FPMF and TA impacts for the LRES with the NMC 811 cathode outperform the results for the NMC 111 by 1.4 and 1.5 factor, respectively. The reason is that the processes related to nickel production have a much higher impact in terms of FPMF and TA than the production of cobalt and manganese, and the nickel content in the NMC 811 battery is eight times higher than cobalt and manganese. The impacts on GW, HT, FRS, and CED have a slight increase but do not show significant changes between the two cathode compositions, whereas the impacts on MRS show a slight reduction with the NMC 811, but also not significant to compensate for the increased impacts on FPMF and TA. One of the main motivations for the replacement of the NMC 111 by the NMC 811 cathode is the potential lower availability of cobalt in the future. Nevertheless, changing the cathode from NMC 111 to NMC 811 has a minor impact on mineral resource scarcity. Although cobalt is the biggest contributor to MRS impacts of the LRES, the highly increased nickel content in the NMC 811 battery maintains the mineral depletion at the same level. It is expected that in the upcoming 5–10 years, the new LIB will have low nickel content and use either nickel–cobalt-aluminium-oxide or NMC 811 cathodes [21]. However, it has been reported that novel cathode chemistries result in similar threats to humans and ecosystems compared to the currently commercialized LIB cathodes [45]. Moreover, it is assumed that the efficiency of the LRES is the same for both cathode compositions as well as the battery energy density, which might not be the case in reality. Therefore, the substitution of the currently used NMC cathodes by innovative ones should be considered

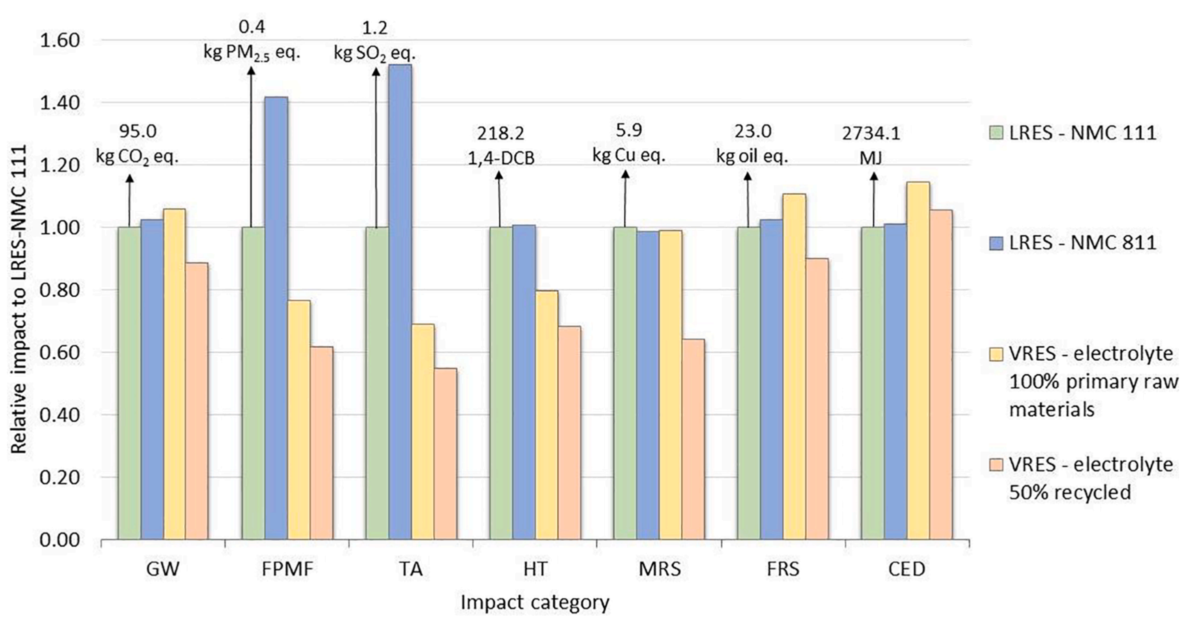

Fig. 4. Life cycle impacts of lithium-ion battery-based renewable energy storage system (LRES) with two different battery cathode chemistries, namely NMC 111 and NMC 811, and of vanadium redox flow battery-based renewable energy storage system (VRES) with primary electrolyte and partially recycled electrolyte (50%). The impacts of the LRES with an NMC 111 cathode are normalized and used as a reference to assess the impacts of the LRES with an NMC 811 cathode and the VRES with both fresh and partially recycled electrolytes. Impact categories: global warming (GW), fine particulate matter formation (FPMF), terrestrial acidification (TA), mineral resource scarcity (MRS), fossil resource scarcity (FRS), cumulative energy demand (CED).

carefully.

In the case of the partially recycled electrolyte for the VRES, the difference is evident in all impact categories, with the impacts being reduced. The impacts of re-processing and purification of the EoL electrolyte are negligible compared to the impact reductions from lower fresh mineral resources use for the manufacturing of the electrolyte. The biggest impact reduction is observed for TA, with the impacts minimized by 21% when 50% of the electrolyte is recycled compared to using 100% fresh electrolyte. It has been reported that even more meaningful reductions can be achieved by using only 30% of recycled electrolyte, which results in 40% less acidification as well as 50% less mineral and fossil resource scarcity [59].

When comparing both cathode possibilities for the LRES and electrolyte compositions for the VRES, the standard VRES (fresh electrolyte) scores better in four out of seven impact categories, with the exceptions being GW, FRS, and CED. If the partially recycled electrolyte is used, the VRES shows reduced impacts in six out of the seven impact categories against the LRES with the NMC 111 cathode, with CED still a bit higher than for the LRES. The most meaningful impact reduction is observed for TA, with 45.2% reduced impacts for the VRES followed by 38.3% lower FPMF, 35.6% lower MRS, 31.6% lower HT, 11.1% lower GW, and 9.9% FRS. Overall, the environmental impacts of the VRES life cycle are lower than those of the LRES, regardless of their cathode or electrolyte compositions, but if the VRES electrolyte is partially recycled, the environmental burdens are significantly decreased, which will most likely be the reality for the EoL handling of this technology in 20 years. Further minimization of impacts will be achieved if a larger share of the electrolyte is reused or recycled, for instance, if leasing of the electrolyte becomes a common practice as described in Section "Life cycle inventory of the VRES".

Conclusions and future perspectives

A detailed comparison of the environmental life cycle impacts of two stationary storage systems was conducted, focusing on LRES and VRES as storage technologies. A complete life cycle inventory for both energy storage systems is provided as an outcome of this study, as well as the quantified environmental impacts for production of the batteries and the use and EoL of the battery-based storage systems. Through the hotspot analysis, it was identified that at the supply phase, the cathode, the anode, and the housing container of the LIB result in the highest environmental impacts, whereas for the VRB, the electrolyte and the current collector are the sources of the major impacts. For the full life cycle scope, the production and transport of both batteries accounted for at least 50% of the impacts in all impact categories, except for CED, where they represented less than 30% of the overall impacts. In the CED, the electricity for use was the source of most of the impacts, with a share of 66% for the LRES and 71.5% for the VRES. The electricity for use was also remarkable in all the other impact categories, as production and mounting of PV systems contain toxic compounds and have a high demand for metals and fossil fuels. Replacing the energy source from PV panels with wind turbines results in significantly lower impacts for both batteries, which reinforces that the energy source plays an important role in the overall environmental impacts. The EoL of both energy storage systems did not result in substantial impacts, as the metals used in the LRES and the VRES were assumed to be 95% recycled. Also, if recycling of the VRES electrolyte is put in place considering a share of 50%, the environmental impacts are reduced even further, with 11.1% reduced global warming and 45.2% reduced terrestrial acidification, in comparison to the LRES with an NMC 111 cathode. Some VRB suppliers report that the electrolyte can be fully recycled and recently, 97% vanadium recovery has been reported [23], but the recycling process should be further investigated and documented if VRES systems are to be implemented in full scale, taking into account the reduction in environmental impacts it enables. Regarding the alternative LIB cathode chemistry, the replacement of the NMC cathode of the LRES by new cathode chemistries, such as NMC 811, does not result in a reduction of impacts. Instead, more fine particulate matter formation and terrestrial acidification are expected, due to the increase in nickel content.

The overall results of this study point at the VRES as the most environmentally friendly technology for renewable energy storage and wind as the best option of renewable energy source for charging and operation in comparison to PV energy. However, the results should be interpreted with caution as some factors considered in the modeling might limit the scope of the results. For instance, it has been reported that the geographic factor might influence the global warming potential resulting from the production of a LIB in different countries, as a result of the national energy mix [45]. Not only the production stage is affected by the location where it takes place, but the overall life cycle impacts can also be affected, for instance at the use phase, as the climatic conditions, such as sunlight and wind, differ from one place to another [48]. It is important to take into account that the energy sources considered in this study might not be available for other locations and that the use of different renewable energy sources, such as hydropower, would result in different environmental impacts. The geographic scope also influences the impacts of the transport of the battery components and the complete battery. A significant share of the impacts were related to the transport of the battery overseas (VRB), so if the battery is produced closer to its final destination, these impacts are reduced. Moreover, the EoL impacts are also linked to the location where the battery is used, since transport to the EoL handling facility is required and because the recycling techniques may differ for different locations. Besides, the VRB considered in this study has a much lower capacity than the LIB as it is designed for set-up tests, which may lead to overestimation of some impacts, as the VRB-based storage systems implemented for grid applications have been designed for large-scale, therefore reducing the resource and energy use intensity. A final remark is regarding the selection of modeling software, allocation method and data source, each influencing the final LCA results. It has been reported that the same process from different databases can lead to substantially different impacts but also a dataset can result in different impacts when the method or the software changes, due to the different characterization factors used [60,61]. The different versions of the same database may also lead to significant changes in the results, as processes are updated with more recent or specific information that was not available before.

Further research is required in the context of the potential future socio-environmental impacts due to the increasing demand for renewable energy. Important investments and development of programs towards the decarbonization of the energy mix are taking place in Europe and other places worldwide. However, the switch to more sustainable energy sources might result in other impacts not yet fully assessed. The search for energy technologies that minimize the adverse impacts on the environment and its biodiversity might end up having the opposite effect, as the production of the required technologies and infrastructure rely on several metals and their extraction from nature pose threats to the local biodiversity [62]. A recent study indicates that the mining areas worldwide might influence about 50 million km^2 on the land surface with significant overlap with Protected Areas, Key Biodiversity Areas, and Remaining Wilderness, and in 82% of these mining areas, key raw materials for renewable energy are produced [62]. Not only the environment suffers from the increasing demand for metals, the local population of several mining areas have to live with human right abuses, child labour and life-threatening working conditions that non-regulated mining impose on them, for instance, the cobalt mining in the Democratic Republic of Congo. In the past 5–10 years, substantial efforts have been addressed towards policy-making to ensure that the sourcing of raw materials is done in a sustainable and responsible manner. Metals used in battery technologies and especially lithium-ion battery raw materials, such as cobalt, graphite, lithium, and manganese have received special attention, with research being focused on their responsible sourcing [57]. However, with new storage systems technologies being developed and implemented on a larger scale than before, like the VRES, the responsible sourcing research should be extended and address these so-called new battery raw materials.

CRediT authorship contribution statement

Lígia da Silva Lima: Supervision, Software, Validation, Visualization, Writing - original draft. **Mattijs Quartier:** Conceptualization, Investigation, Methodology, Software, Writing - original draft. **Astrid Buchmayr:** Supervision, Validation, Writing - review & editing. **David Sanjuan-Delmás:** Supervision, Software, Writing - review & editing. **Hannes Laget:** Supervision, Investigation, Writing - review & editing. **Dominique Corbisier:** Supervision, Investigation, Writing - review & editing. **Jan Mertens:** Conceptualization, Supervision, Resources, Writing - review & editing. **Jo Dewulf:** Conceptualization, Supervision, Resources, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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