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How Green are Redox Flow Batteries?

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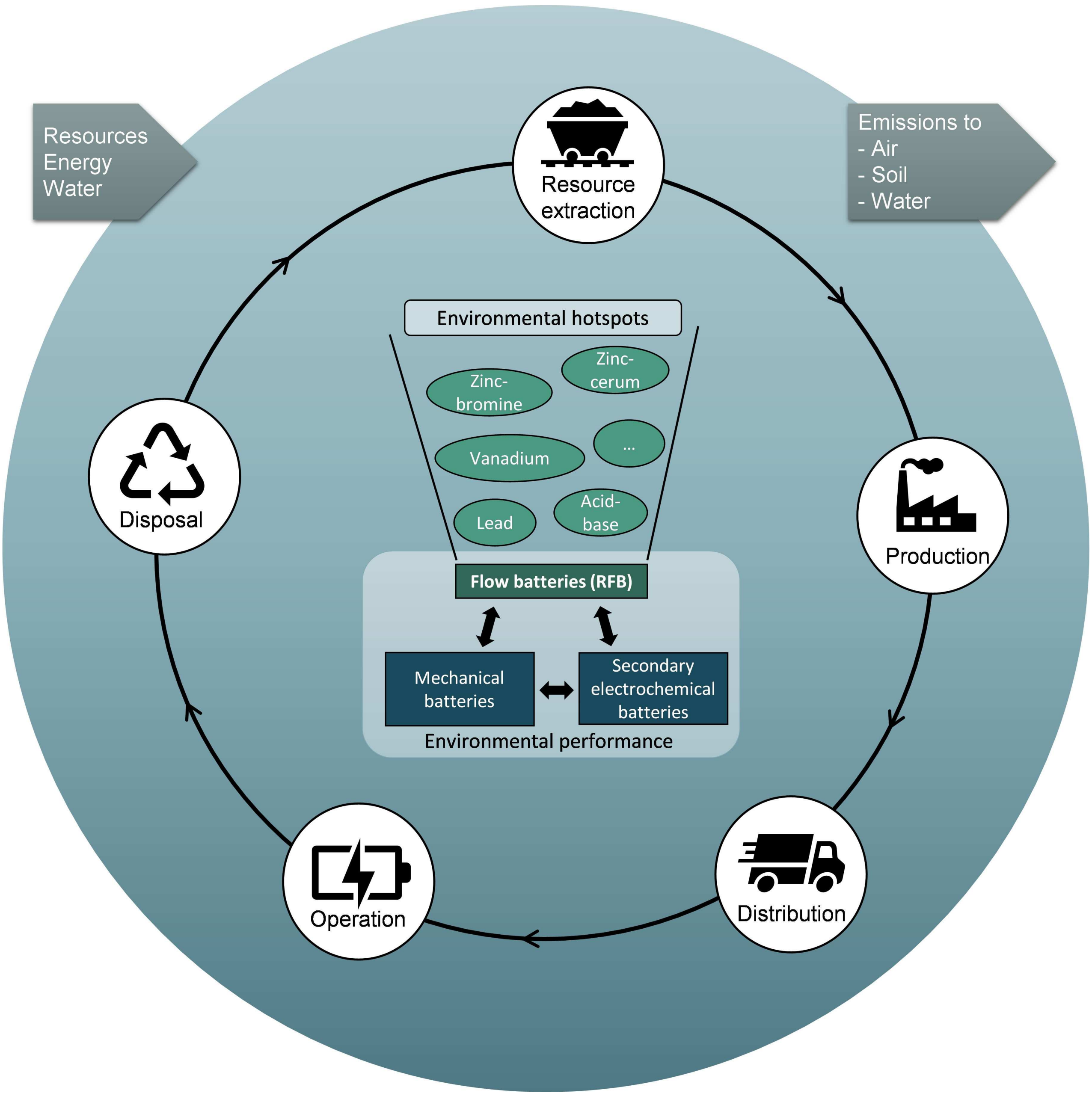

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Providing sustainable energy storage is a challenge that must be overcome to replace fossil-based fuels. Redox flow batteries are a promising storage option that can compensate for fluctuations in energy generation from renewable energy production, as their main asset is their design flexibility in terms of storage capacity. Current commercial options for flow batteries are mostly limited to inorganic materials such as vanadium, zinc, and bromine. As environmental aspects are one of the main drivers for developing flow batteries, assessing their environmental performance is crucial. However, this topic is still underexplored, as researchers have mostly focused on single systems with defined use cases and system boundaries, making the assessments of the overall technology inaccurate. This review was conducted to summarize the main findings of life cycle assessment studies on flow batteries with respect to environmental hotspots and their performance as compared to that of other battery systems.

1. Introduction

The need for a sustainable energy transition, namely, a transition in which traditional fossil-based energy sources are abandoned and replaced with renewable, regenerative sources,[1] has been one of the main technology drivers in the 2020s to date.[2] The background is the accelerating increase in the CO_2 concentration in the atmosphere, causing global warming hitherto unseen during the history of mankind. This has been reflected in numerous activities on international and national levels, with the Paris Agreement[3] and the European Green Deal[4] being the most important ones. These agreements have been made in an attempt to curb CO_2 emissions in different sectors and to slow down global warming. Although the focus in the past decade was directed toward mobility and transportation (electric vehicles), the next decade will be a crucial one for reducing the impact of industrial and primary energy use. Here, significant progress has been made to build solar and wind parks for sustainable energy production. The drawback of this development is that the electric grids have been neither designed nor equipped to achieve such a purpose, and the supply of renewable energy often does not match the demand, requiring the use of (fossil- or nuclear-based) backup plants to avoid blackouts.[5]

One key measure that would ensure a safe supply of electricity is to create large-scale storage facilities that can sustainably provide power in the MW-GW range. Depending on the response time needed, a variety of options exist, ranging from hydropower, compressed air storage, and flywheels, as well as batteries in the broadest sense.[6] Huge progress in battery technology and particularly lithium-ion battery (LIB) technology has been made in recent years. Scale-up effects and huge research efforts all over the world regarding electric vehicles resulted in decreasing costs and higher efficiency and lifetimes for such systems.[7] Although LIBs will soon find their way into stationary storage, they currently have some limitations for usage as large-scale energy storage units. This is due to several issues, including their thermal management, discharge depth, cycle-dependent capacity fading, self-discharge, operation times (i.e., how long the battery can be charged/discharged) as well as safety issues. Several reviews cover this topic, and we refer the reader to these works.[8]

A technology that has its benefits over LIBs is redox flow battery (RFB) technology. After they were used for the first time to operate the propeller of an airship called *La France* in the late 19th century,[9] flow batteries were basically forgotten until NASA and the National Institute of Advanced Industrial Science and Technology in Japan independently began to invest intense research efforts in the 1970s.[10] In the past twenty years, flow batteries have become increasingly attractive as energy storage systems, and particularly for large-scale storage. Commercial RFBs, such as those based on vanadium or zinc-bromine, do not exhibit significant capacity fading upon cycling, have hardly any self-discharge, are typically inflammable and, most importantly, the power and storage capacity can be designed separately, leading to long charge and discharge times (4–10 hours).[11] Still, RFBs represent a niche technology today with installed capacities only slightly exceeding the GWh range. The rather high installation costs for flow battery systems represent one of the main barriers, as these are typically much higher than for LIBs. As only the price per kWh is often the main criterion for customers, flow batteries seem to be more expensive than LIBs, limiting their market penetration. However, the economic performance of any battery system must be levelized to compare the actual competitiveness of the technology. Typically the levelized cost of storage (LCOS) is used as a parameter, which includes costs for installing, running, and maintaining the system, as well as costs for decommissioning it as a function of stored energy over the lifetime.[12] When applying LCOS, even different application scenarios can be simulated in stationary storage (e.g., peaker replacement), resulting in economic assessments of the different storage technologies.

Although research efforts have recently been made to use sustainable materials for RFBs, the currently used commercial RFB technology relies heavily on heavy metals and depletable

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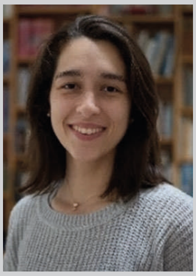
sources. Vanadium oxides, zinc and bromine are the materials most commonly used as electroactive components in commercial batteries,[13,14] potentially limiting the sustainability performance, as all of these elements give rise to environmental concerns. Vanadium salts are potentially cancerogenic and display high toxicity in land and marine environments.[11,15,16] They appear on the list of critical raw materials published by the European Union[17,18] and must be mined and transported to the site where the battery is manufactured. However, another issue with flow battery technology is the lack of standardization regarding the determination of performance parameters, because the technology is still in its market-creation phase. Furthermore, the vast number of applications for stationary energy storage makes the assessment of potential sustainability impacts more challenging, as many additional parameters come into play.

The safe and sustainable by design (SSbD) framework was created to tackle this challenge, providing a way to evaluate safety and sustainability aspects throughout the whole life cycle when developing new chemicals and materials.[19,20] To use this framework, it is necessary to take a life cycle perspective.[19] The life cycle assessment (LCA) tool is one of the most well-developed and widely acknowledged tools used to assess the environmental impacts of materials and products throughout the whole life cycle, i.e., from resource extraction to product disposal.[21,22] Several studies have performed comparative LCAs of different electricity storage systems (ESS), such as that of Da Silva Lima et al., who compared a Li-ion battery with a vanadium RFB,[23] or that of Hiremath et al., who compared Li-ion, lead-acid, sodium-sulfur batteries and a vanadium RFB to measure their environmental impacts.[24] The results of these works and others have been partly reviewed in Rahman et al.,[25] who compared different ESS to each other by their technical parameters (e.g., rated power, response time, or specific energy), costs (i.e., levelized cost of electricity) and environmental impacts (i.e., greenhouse gas emissions).[25] In this review, only eight studies assessing RFB systems were considered. Another recent review on the LCA of RFB systems by Dieterle et al.[26] identified methodological gaps and weaknesses related to applying LCA to flow battery systems. In their work, these authors provided a thorough discussion of and recommendations for the provision of, for example, a functional unit or life cycle inventories. A synthesis of environmental areas of concern connected with flow battery technologies and the uncertainties connected with inventory data is still lacking.

In this paper, we address this gap by critically reviewing these aspects by carefully examining LCA studies on RFB technologies. The review is structured as follows: A short introduction to the environmental issues related to battery technologies is provided, followed by an overview of the currently performed LCAs of flow batteries. A summary of the environmental impacts and performance of RFBs is then given, and a critical discussion is presented, including recommendations for future efforts.

2. Environmental Issues Related to Battery Storage Technologies

The environmental impacts of batteries and particularly LIBs is an emergent topic that is closely related to the increase in the number of electric vehicles and the need for stationary energy storage systems.[27] The large amount of raw materials required to manufacture these batteries, including copper, cobalt and nickel, requires careful consideration to assess the environmental impacts of the raw material mining.[28] Another important factor of the battery LCA is the fate of these batteries after their end-of-life, where recycling is a frequently discussed option.[29] The benefits of recycling for the environment depend strongly on the selected impact categories (global warming potential, resource depletion), the cell layout, processing, production and chemistry. Some authors have projected that future LIB production will be more efficient, as less energy is



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required and more sustainable energy will be used to produce these batteries,[30] rendering the manufacturing more environmentally benign. Regarding the cell chemistry, the assessment is more complicated. The chemistry determines the achievable energy density, which directly affects the environmental impacts, because it influences the amount of material needed to store one kWh of electric energy. Although the correlation to material depletion can be relatively easy considered, recycling is more difficult to grasp and may even introduce additional environmental impacts. For example, lithium iron phosphates (LFP) or sodium ion batteries require energy intense (hydrothermal)metallurgical treatments during the recycling process. The assessment of such processes is again challenging to establish, as these are not standardized processes and often have been only demonstrated on rather small scale.[31] Another complication arises due to the quality of the recycled materials. For example, recovering lithium-ion batteries may alter their crystal structure,[32] thereby potentially curbing their performance in terms of energy density and cycle stability.[33] This, in turn, will affect the outcome of an LCA. These challenges facing researchers who attempt to make precise LCAs are increased by the existence of several unknown process parameters. These include which chemicals and materials are used in the course of the battery production that are needed for developing recycling processes.[34] The inclusion of more precise chemical data and process characteristics into the LCA models, therefore, is needed to gain more accurate LCA results.[35] Similar challenges also face researchers working with emerging technologies such as flow batteries.

Apart from the LCA studies, a recent review assessed how 'green' the active material in flow batteries is.[36] The authors investigated the impact of the electrolytes, considering the green chemistry principles, by categorizing them into three impact categories: (1) low environmental impact, (2) medium environmental impact, and (3) high environmental impact. This study is the most comprehensive of its kind on the subject of flow batteries, as 30 different types of compounds were investigated, therefore providing an excellent overview of different cell chemistries. This approach has the advantage that the authors could examine the effects of the different green chemistry principles (containing several compounds) and easily visualize and compare these regarding their impact on the 12 green chemistry principles. Data available for the components in the publications as well as in databases, allowed for a qualitative assessment of their performance. This simplification, however, also has a disadvantage, because the impact of the categories was not weighted. The quantification of the effects also was not included, as it was beyond the scope of the study.

The redox chemistry of commercial flow batteries is generally well understood and, in principle, highly reversible for commercialized technologies. Regarding vanadium battery systems, this chemistry involves the reduction of V^{3+} to V^{2+} ($V^{3+}+1e^{-}\leftrightarrow V^{2+}$, $E_{anode}=-0.26$ V) and the oxidation of VO^{2+} to VO_2^{+} ($VO^{2+}+H_2O-1e^{-}\leftrightarrow VO_2^{+}+2H^{+}$, $E_{cathode}=+1.0$ V), resulting in a cell voltage of 1.26 V. However, some factors limit the lifetime of vanadium-based electrolyte solutions. These involve reactions related to overcharging (creation of H_2 and O_2), reactions on battery components (e.g., with graphite plates, generating CO_2), as well as undesired reactions of vanadium species, which are often associated with precipitation that blocks the hydraulic system.[37] A prominent example includes the deprotonation of the $VO_2^{+}(H_2O)_3$ species at elevated temperatures, yielding the condensed neutral $VO(OH)_3$ complex. This compound can also undergo dimerization, yielding the poorly soluble $V_2O_5(H_2O)_3$ adduct, which precipitates out of the electrolyte solutions. A detailed account of these mechanisms is provided in the seminal work by Skyllas-Kazacos et al.,[38] who investigated the complex vanadium chemistry of VRFBs in detail. These authors also pointed out the importance of impurities present in the vanadium salts, some of which favor precipitation and side reactions, even at concentrations in the mmol range. In this context, the authors highlighted the advantage of employing vanadium with high purity grades. The other relevant commercial flow battery chemistry involves zinc-bromine technology. The main chemical reactions include the reduction of Zn^{2+} to elemental Zn ($Zn^{2+}+2e^{-}\leftrightarrow Zn^{0}$, $E_{cathode}$: -0.76 V) and the oxidation of bromide to elemental bromine ($2Br^{-}\leftrightarrow Br_2+2e^{-}$, E_{anode}: 1.09 V), yielding a cell potential of 1.85 V. As seen for vanadium, commercial Zn–Br batteries are operated at a low pH value (1–3.5), where significant hydrogen evolution and electrode corrosion can take place. At higher pH values, the current efficiency is compromised. In theory, a solution of pure $ZnBr_2$ could be used to run such a battery, but, in practice, the solutions contain supporting electrolytes (to increase ionic conductivity and reduce resistance) and bromine complexation agents (e.g., 1-ethyl-1-methylpyrrolidinium bromide). The complexation agent is added to remove the evolving Br_2 into a separate phase; however, this negatively impacts the kinetics of the bromine half cells. Apart from the biphasic system, the main issue with Zn–Br RFBs is the formation of Zn-dendrites during the reduction of the $ZnBr_2$. Dendrite formation is a complicated process that has a negative impact on the flow battery performance, as dendrites may compromise the flow behavior in the battery and may even harm some battery components (e.g., membrane). For a detailed description of dendrite formation, we refer the reader to a recent paper by Xu et al.,[39] as this topic is beyond the scope of this paper. Any improvements in either knowledge about degradation processes and/or solutions to reduce/avoid these will improve not only the techno-economic but also the environmental performance of these batteries.

3. Life Cycle Assessments of Flow Batteries

Studies investigating the environmental impacts of RFB technologies seem to be scarce, as our review identified only 22 studies on this topic (Table 1). A main reason for this is that flow batteries are still considered a rather new battery technology. Most of the studies performing an LCA of flow batteries were published after 2015, with a clear increase in the number of studies published from 2020 and on. Nearly all of the studies deal with vanadium-based flow battery (VRFB) systems, as these are commercially available; hence, their

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Table 1. LCA studies on flow battery technologies.

Entry	Author(s)	Year	Title	Functional unit	Data sources	LCIA method	Midpoint indicators	Ref.
1	Rydh	1999	Environmental assessment of vanadium redox and lead-acid batteries for stationary energy storage	450 kWh storage capacity	*Research; manufacturer; supplier*	Environmental Theme, Environmental Priority Strategies	AC, ARU, CC, ET, RE	[48]
2	Hiremath et al.	2015	Comparative Life Cycle Assessment of Battery Storage Systems for Stationary Applications	1 MWh delivered electricity	scientific publications	IPCC 2007, ReCiPe 2008 single point	ARU, LU, Etox, ET, AC, IR, RE, HT, OD, CC, ED	[24]
3	Sternberg et al.	2015	Power-to-What? – Environmental assessment of energy storage systems	1 MWh delivered electricity	ecoinvent; scientific publications	ReCiPe 1.08 midpoint (H)	ARU, AC, CC, ET, HT, IR, OD, RE	[47]
4	Dassisti et al.	2016	Sustainability of vanadium redox-flow batteries: Benchmarking electrolyte synthesis procedures	6 l of electrolyte	ecoinvent	ReCiPe 2008 midpoint	ARU, AC, CC, Etox, ET, HT, IR, LU, OD, RE	[40]
5	Unterreiner et al.	2016	Recycling of Battery Technologies – Ecological Impact Analysis Using Life Cycle Assessment (LCA)	1 kWh storage capacity	ecoinvent; scientific publications; research	ReCiPe 2008 midpoint and single point	ARU, CC. HT, LU, RE	[49]
6	Weber et al.	2018	Life Cycle Assessment of a Vanadium Redox Flow Battery	1 MWh delivered electricity	ecoinvent	CML	ARU, AC, CC, HT	[50]
7	Mostert et al.	2018	Comparing Electrical Energy Storage Technologies Regarding Their Material and Carbon Footprint	2 MWh delivered electricity	ecoinvent; scientific publications; manufacturer	Raw material input, total material requirement, -	ARU, CC	[46]
8	Stougie et al.	2019	Multi-dimensional life cycle assessment of decentralized energy storage systems	10 kWh storage capacity	ecoinvent	ReCiPe 2016 method V1.00, midpoint GWP and endpoint	CC	[44]
9	Jones et al.	2019	Assessing the Climate Change Mitigation Potential of Stationary Energy Storage for Electricity Grid Services	1 MWh delivered electricity	ecoinvent; Plastics Europe database; scientific publications; supplier	ReCipe Midpoint (H)	ARU, AC, CC, Etox, ET, HT, IR, LU, OD, RE, WU	[51]
10	L'Abbate et al.	2019	Small-Size Vanadium Redox Flow Batteries: An Environmental Sustainability Analysis by LCA	127.5 Wh delivered electricity	ecoinvent; experiments; measurements; scientific publications;	ECOtox, IMPACT2000+, ReCiPe endpoint	CC, HT	[43]
11	Gouveia et al.	2020	Life cycle assessment of a renewable energy generation system with a vanadium redox flow battery in a NZEB household	1 kWh delivered electricity	ecoinvent; research; suppliers; expert judgment	ILCD 2011 midpoint + V1.10	ARU, AC, CC, ET, OD, RE	[42]
12	Gouveia et al.	2020	Life cycle assessment of a vanadium redox flow battery	1 kWh storage capacity	ecoinvent; manufacturer; suppliers	ILCD 2011 midpoint + V1.10.	ARU, AC, CC, ET, OD, RE	[41]
13	Fernandez-Marchante et al.	2020	Environmental and Preliminary Cost Assessments of Redox Flow Batteries for Renewable Energy Storage	1 kWh storage capacity	ecoinvent; literature; expert judgment	AWARE, CML Baseline v3.04	ARU, AC, CC, Etox, ED, ET, HT, LU, OD, RE	[52]
14	He et al.	2020	Flow battery production: Materials selection and environmental impact	1 kWh storage capacity	ecoinvent; scientific publications; manufacturers	ReCiPe midpoint 2016, USETox, CML–IA, cumulative energy demand	ARU, AC, CC, Etox, ED, ET, OD, RE	[53]
15	Díaz-Ramírez et al.	2020	Battery Manufacturing Resource Assessment to Minimise Component Production Environmental Impacts	1 kWh storage capacity	ecoinvent; scientific publications; generic databases,	ReCiPe 2016 v1.1 midpoint (H)	ARU, AC, CC, Etox, ET, HT, IR, LU, OD, RE, WU	[45]
16	Baumann et al.	2020	Exploratory Multicriteria Decision Analysis of Utility-Scale Battery Storage Technologies for Multiple Grid Services Based on Life-Cycle Approaches	1 kWh stored and delivered electricity	ecoinvent; scientific publications	ReCiPe endpoint	-	[44]
17	Díaz-Ramírez et al.	2020	Environmental Assessment of Electrochemical Energy Storage Device Manufacturing to Identify Drivers for Attaining Goals of Sustainable Materials 4.0	1 kWh storage capacity	ecoinvent; scientific publications	ReCiPe 2016 v1.1 midpoint	ARU, AC, CC, Etox, ET, HT, IR, LU, OD, RE, WU	[54]
18	Da Silva Lima et al.	2021	Life cycle assessment of lithium-ion batteries and vanadium redox flow batteries-based renewable energy storage systems	1 MWh delivered electricity	ecoinvent; scientific publications; research; manufacturers	ReCiPe 2016 midpoint (H)	ARU, AC, CC, ED, HT, RE	[23]
19	Morales-Mora et al.	2021	Life cycle assessment of a novel bipolar electrodialysis-based flow battery concept and its potential use to mitigate the intermittency of renewable energy generation	1 MWh storage capacity	ecoinvent; research	ReCiPe 2016, cumulative energy demand	ARU, AC, CC, Etox, HT	[55]

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Table 1. continued

Entry	Author(s)	Year	Title	Functional unit	Data sources	LCIA method	Midpoint indicators	Ref.
20	AlShafi and Bicer	2021	Life cycle assessment of compressed air, vanadium redox flow battery, and molten salt systems for renewable energy storage	1 kWh of electricity	GaBi; scientific publications	CML 2001 (2016)	ARU, AC, CC, Etox, HT, OD	[56]
21	Shittu et al.	2022	Life cycle assessment of soluble lead redox flow battery	1 kWh storage capacity	GaBi	ReCiPe 2016 midpoint (H)	ARU, AC, CC, Etox, ET, HT, IR, LU, OD, RE, WU	[57]
22	Díaz-Ramírez et al.	2022	Acid/base flow battery environmental and economic performance based on its potential service to renewables support	1 MWh delivered electricity	ecoinvent; scientific publications	ReCiPe 2016 v1.1 midpoint (H)	ARU, AC, CC, Etox, HT, WU	[58]

performance is known, and their lifetimes in terms of cycle numbers and years can be effectively estimated. The 22 reviewed studies analyzed eight different RFB technologies (Table 1): VRFB, soluble lead (SLRFB), concentration gradient (CGFB), zinc-bromine (ZBFB) or zinc-cerium (ZCB), all-iron (IFB), bipolar electrodialysis (BEDFB), and acid-base flow batteries (AB-FB). These eight different electrochemical flow battery technologies were investigated with regard to their environmental impacts. These impacts were compared to those of different production technologies using the same storage technology[40–43] and of other storage technologies (e.g., lithium-ion batteries (LIB) and lead-acid batteries (LAB)[44–46]) or other forms of energy provision like a heat pump.[47] The vanadium RFB is the most often investigated technology ($n=20$). The environmental impacts of the other seven RFB technologies have been assessed once in the literature. Other well-analyzed storage technologies in this sample are LABs and LIBs. A graphical summary of the technologies investigated in the respective studies is provided in the Supporting Information.

The eight investigated RFB systems do not only differ in their electrochemistry but also in their technical performance influencing the life cycle impacts. The technical performance parameters (i.e., expected lifetime in years and cycles, depth of discharge, specific energy, and the efficiency of the RFB technologies) are summarized in Table 2. Most studies assumed an expected lifetime of 10 to 20 years with a cycle life of 2,000 to 15,000 cycles for all RFB systems except for ZCB. At the time of investigation, ZCBs were at an experimental stage with only 57 operating cycles of charge-discharge demonstrated.[52] The specific energy of the VRFB is assumed to be approx. 20 Wh kg^{-1}, and lower values for BEDFB (7.29 Wh kg^{-1}) or for AB-FBs (3.2 Wh kg^{-1}) have been observed. Regarding the efficiency of the cell or the charge-discharge cycle, the study authors assumed efficiencies ranging from 65% to 96%.

Table 2. Assumed technical parameters of the flow battery technologies; depth of discharge (DoD).

Ref.	Flow battery	Expect. lifetime [years]	Expect. lifetime [cycles]	DoD [%]	Specific energy [Wh kg^{-1}; *Wh L^{-1}*]	Efficiency [%]
[48]	VRFB	–	>2000	–	20; *30*	72–88
[24]	VRFB	9.5	13000–15000	80	20	75–80[c]
[47]	VRFB	–	2900–7500	–	–	65–85
[40]	VRFB	–	–	–	*30–37.8*	77–96[d]
[49]	VRFB	20	>10000	100	25	90
[50]	VRFB	20[a]	10000	-	19.4	75[e]
[46]	VRFB	~9.5	–	80	–	77–83
[59]	CGFB	20	–	–	–	65 /75[e]
[51]	VRFB	20[a]	–	80	–	42–77[c]
[43]	VRFB	>20[b]	–	–	*36.18*	85
[42]	VRFB	20[b]	–	–	-	–
[41]	VRFB	20	–	–	*20*	up to 80
[52]	VRFB	20	–	–	–	80
	ZCB	–	57	–	–	62
[53]	VRFB	–	–	–	–	–
	ZBFB	–	–	–	–	–
	IFB	–	–	–	–	–
[45]	VRFB	10.9	12068	–	19.9	–
[44]	VRFB	10–20	12000	80	20–80	70–75
[54]	VRFB	12.4	12068	100	19.9	79[f]
[23]	VRFB	20	–	–	*16–33*	83[c]
[55]	BEDFB	–	–	90	7.29; *8.8*	75[c]
[56]	VRFB	20	–	–	–	77
[57]	SLRFB; VRFB	–	–	–	–	–
[58]	AB-FB VRFB	20[a]	10000	95	3.25 19.4	65 75

[a] Including stack replacements. [b] Including membrane replacements. [c] Round-trip efficiency. [d] Cell efficiency. [e] Charge-discharge efficiency. [f] Discharge efficiency.[20a]

3.1. Goal and scope definition

To perform an LCA study, the ISO standards[21,60] propose four phases: the goal and scope definition, the life cycle inventory (LCI), the life cycle impact assessment (LCIA), and the interpretation phase. When defining the goal and scope, several methodological decisions on, for example, the system boundaries, the object of assessment (function, functional unit, reference flows), LCI data and requirements, and the basis for impact assessment (impact categories, LCIA methods) need to be made.[21,61] The system boundaries differentiate between the studied product system, the technosphere and the ecosphere, depicting the life cycle stages included for the analysis.[61] Figure 1 illustrates different possibilities for how the system boundaries can be set, for example, from resource extraction to product disposal (cradle-to-grave), and Figure 2 illustrates the chosen system boundaries (see also Table 4).

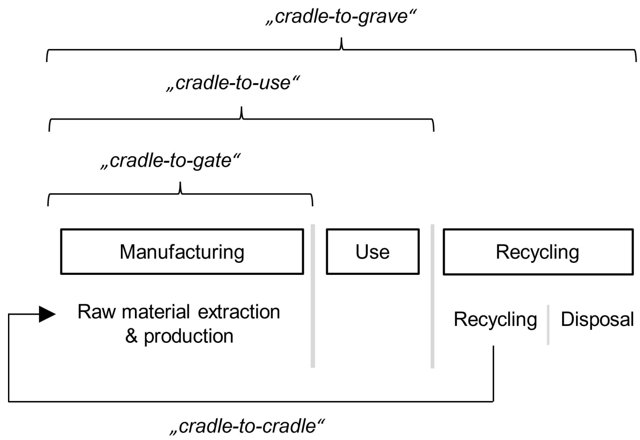

Figure 1. Overview of setting system boundaries in LCA (based on Ref. [62]).

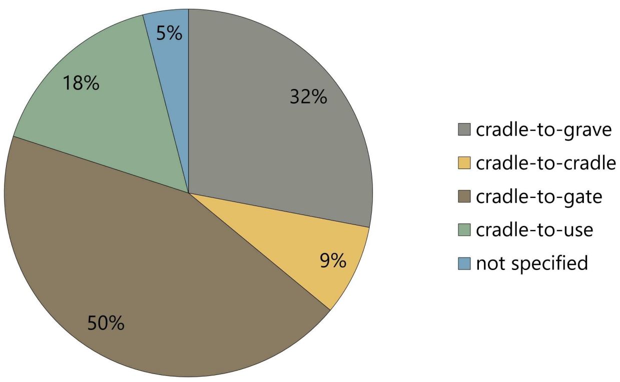

Figure 2. System boundaries of RFB LCA studies ($n=22$).

Approximately half of the RFB-LCA studies ($n=12$) investigated the environmental impacts from the point of resource extraction to that of battery manufacture (cradle-to-gate). Many of the studies investigated the impacts from the cradle-to-use and end-of-life stages (cradle-to-grave) or only included material recycling but not disposal (cradle-to-cradle). Da Silva Lima et al.[23] and Morales-Mora et al.[55] analyzed the cradle-to-gate as well as cradle-to-grave impacts to identify environmental hotspots in the production or throughout the whole life cycle.

The environmental impacts are calculated for a defined functional unit. To clearly define a functional unit, a quantitative comparison of alternative systems must be supported, where all those systems need to fulfill the same functions.[60,61] The reviewed LCA studies defined the functional unit as either an amount of electricity stored or delivered through the storage system (Table 1). Dassisti et al.[40] compared three preparation methods of mixed acid vanadium electrolytes for VRFB and chose a functional unit of six (6) liters of electrolyte produced in the laboratory. To clearly state the functional unit, the service life (year or cycles) of the investigated technology needs to be defined (see Table 4). The majority (68%) of the studies assumed a service lifetime of 20 years, while one study assumed a lifetime of 30 years,[51] and the remaining studies did not provide any information about the assumed service lifetime, since this was not necessary when assessing the impacts from cradle-to-gate.[40,47,53]

3.2. Life cycle inventory

After the goal and scope are defined, the data needed for the LCI (i.e., the process inputs and outputs for the studied system) are collected and analyzed.[21] However, depending on the technology readiness level (TRL), different kinds of data are used to model the foreground processes. Whether the potential impacts of products at low TRLs (streamlined LCA) or the performance of products at a high TRL (full LCA) will be assessed as compared to benchmark products[63,64] highly influences the uncertainties of the results.[65,66] Still, assessing the environmental impacts of products or processes at a low TRL helps to reduce unintended, negative effects. In the case of LCAs of products in low TRLs, the data have low accuracy, and are often based on assumptions made by an expert or an LCA practitioner or are derived from laboratory experiments, generic databases like Ecoinvent, or scientific publications.[67] The higher the product TRL, the more specific the data. The data are then measured or derived from measurements taken at a specific process site.[67] In the reviewed RFB LCAs, all types of data were used to assess the environmental impacts of the storage technologies; however, generic data were mostly extracted from, for example, the Ecoinvent database or the scientific literature and, in some cases, from experiments or manufacturers' specifications (Table 1). Some of the LCA studies referred to a reference work published in 1999.[48] This first published LCA of a VRFB is mainly based on a hypothetical manufacturing scenario; therefore, these results are subject to high levels of uncertainty regarding the data accuracy and temporal representativity.

3.3. Life cycle impact assessment

In the LCIA phase, the impacts on the entities we want to protect are evaluated (i.e., human health, the natural environment and natural resources).[68] To do so, impact categories and impact assessment methods (LCIA methods) need to be chosen.[21,61] A broad spectrum of methods and impact categories are available to support decision-making and a recent review on LCA studies identified over 50 different impact categories based on the literature sample.[69] Midpoint impacts are measured in terms of the, for example, global warming potential, acidification, eutrophication, or ecotoxicity. The most common impact category chosen for LCAs is the impact on climate change as expressed by the global warming potential.[69–71] LCIA methods combine a number of impact categories (e.g., ReCiPe, CML, or ILCD) and are partly implemented in LCA software like SimaPro or openLCA.[72] Approximately one-third of the RFB LCA studies used SimaPro as a software support, and the rest used openLCA, Gabi, Umberto, or Excel to assess the impacts by applying the LCIA methods

ReCiPe, CML, ILCD, USETox, among others (Table 1). In each of these LCIA methods, a set of midpoint impact category indicators is used to assess the environmental impacts of a product system. In the reviewed studies, 31 different impact category indicators were investigated. These were categorized into 12 impact category groups (Figure 3). Detailed information about the categorization is given in Table S1 in the Supporting Information. The most often investigated environmental issues were abiotic resource use (ARU), ecotoxicity (Etox), human toxicity and carcinogenic effects (HT), respiratory effects (RE), as well as impacts on climate change. In five studies, an endpoint assessment was carried out, namely, the damage to ecosystems, human health, and resource availability.[43,44,59]

For more detailed information about the methodological choices in the respective LCA phases, we refer the reader to the work of Dieterle et al.[26] who provide a comprehensive discussion of and recommendations for future works.

3.4. Battery end-of-life

Almost half of the studies considered the end-of-life (EoL) impact of the batteries and performed a cradle-to-grave or cradle-to-cradle assessment. The assumptions made with regard to the recyclability of components and materials vary greatly. Most studies assumed that metal parts (steel, aluminum, and copper) were recyclable, although to a varying extent. A high recycling rate for metal parts was assumed, for example, a 95% recycling efficiency for steel, aluminum and copper.[50,51] However, Jones et al. assumed that just 90% of metal and plastic mass recovered from the ESS site would enter the recycling process, and a 100% recycling efficiency for steel and copper

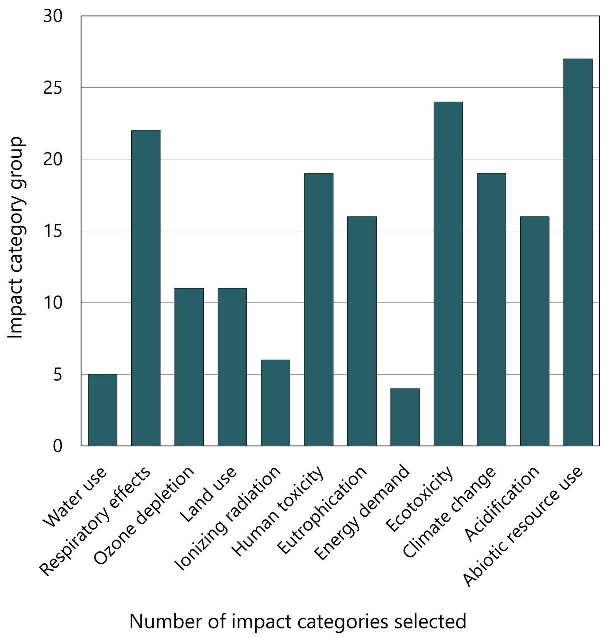

Figure 3. Impact category groups investigated in the reviewed literature ($n=22$).

(incl. the required energy input to recycle) was assumed by Díaz-Ramírez et al.[58] Two studies based their assumptions on the work of Rigamonti et al.,[73] who assumed a recycling efficiency of 81.45% for steel, 55.71% for plastic and 79.33% for copper.[45,59] Morales-Mora et al. assumed a recycling rate of 70–95% for different materials and components, such as a 50% recycling rate for plastic materials like polyvinyl fluoride and 80% for HDPE.[55] However, no background information regarding these assumptions is provided by these authors. In another study, all plastic materials were incinerated with energy recovery,[42,48] and the energy was further used for district heating purposes.[48]

Regarding the recycling of specific RFB components and materials, L'Abbate et al. assumed that the membranes and pumps of the VRFB were brought to the landfill at the EoL, while all other materials were assumed to be fully reusable.[43] For instance, up to 90% of the vanadium can be recovered from the electrolyte by electrochemical filtration,[51] and Da Silva Lima et al. examined the influence of recycling on the total impacts by calculating two scenarios for VRFB electrolyte recycling: one with no recycling, and the second one with 50% recycling.[23] The results of making these assumptions are partly discussed in Section 4.1.

3.5. Uncertainty analysis

LCA studies are often associated with high levels of uncertainty, especially in the case of ex ante assessments of products in low TRLs.[65,66,74,75] The uncertainty associated with LCA results can be differentiated into several categories: uncertainty due to variability (temporal, spatial, between objects), input data (parameter uncertainty), normative choices (e.g., functional unit), mathematical models involved (e.g., characterization), epistemological error (e.g., lack of relevant knowledge), or mistakes in the modeling or due to relevance (e.g., environmental).[75–77] Therefore, uncertainty analyses must be conducted, and the results must be interpreted with caution.[66] In the reviewed studies, most authors addressed the issue of uncertainty associated with their assessments, and 15 studies performed either uncertainty or sensitivity analyses to address one or more of the above-mentioned types of uncertainty (parameter, scenario, or model uncertainty). For instance, Baumann et al.[44] analyzed the uncertainty of their input data by performing a Monte Carlo simulation and by varying key parameters. Weber et al.[50] applied a standard uncertainty estimation process as recommended by Ecoinvent, finding that the reliability (assumptions for modeling) and completeness/sample size (limited amount of data sources and small sample sizes) contributed the majority of the total uncertainty. Fernandez-Marchante et al.[52] also performed uncertainty analyses and summarized their findings in a table, including the standard deviations and potential impact values. However, we found no information in this study regarding how the analysis was performed in detail. Other research groups addressed uncertainty with respect to normative choices by performing a sensitivity analysis, for example, to examine the ESS energy demand/mix and efficiencies,[23,24,42,46,47,51,56,58] possibilities of cell degradation,[51] performance measurements like energy/power ratios,[44,55,57] battery life time,[46] usable storage capacity,[41,46] share/use of secondary materials,[23,41,44,46] transportation,[52] or alternative materials for certain components like different chemical compositions of mesh and membrane materials.[23,50,55–57]

4. Environmental Impacts of RFBs

Each LCA study differs in terms of its defined goal and scope, methodological choices, the TRL of the technology, and the associated data accuracy. Because of this and the way results are illustrated in the interpretation phase,[26] LCAs can rarely be compared, and the results are difficult to synthesize.[25,26] This becomes apparent when examining the results of the reviewed studies, which are summarized in Table 4. The potential impacts of the VRFB in a cradle-to-gate approach have been assessed eleven times, and the impacts per MWh stored/delivered electricity range from 15 kg CO_2 equiv. to 217000 kg CO_2 equiv. Though it is difficult to perform a quantitative comparison of environmental impacts, a qualitative discussion of the environmental areas of concerns for different ESS is possible and provides information that can be used directly by researchers and practitioners in future studies. Therefore, the following section summarizes the results of the RFB LCAs qualitatively, starting with the environmental areas of concern (hotspots) of RFB technologies.

4.1. Environmental hotspots of RFB technologies

One objective of performing an LCA is to identify environmental hotspots in the product system. This information can subsequently be used by developers and practitioners to improve the environmental performance of a product. Hotspots can be formed by certain components or materials used. Likewise, a life cycle phase (resource extraction, manufacturing, use, disposal) can be responsible for a large share of the environmental impacts. Another issue could be that, in comparative LCAs, a product system can perform better in one impact category but more poorly in another. The hotspots identified in the studies are summarized in Table 3 as main drivers of the total impact in different impact categories.

Components and materials

Vanadium RFB is the technology that was most often investigated in the reviewed literature (91%). The components of a VRFB can be grouped into power subsystem components (e.g., electrodes, membranes, bipolar plates), energy subsystem components (electrolyte and the tank), and periphery components (e.g., pumps, motors, racks).[45,50,58] Depending on the specific energy of the electrolyte (Table 2) or the energy capacity of the VRFB, the energy subsystem makes up a high share of the total weight (70–90%).[23,41,45,50,58] The AB-FB electrolyte with 40.4 kg MWh^{-1} has an even higher weight than the VRFB electrolyte with, for example, 6.52 kg MWh^{-1}.[58] The electrolyte of AF-FB makes up 98.7% of the total weight.[58] In the case of a lithium manganese oxide (LMO) battery, the electrolyte accounts just for 15% of the total weight.[45] The weight is one influencing factor with respect to many of the environmental impacts, where the electrolyte forms a hotspot (i.e., represents the component with the highest environmental impacts out of all other components in the case of VRFB, ZBFB, ZCB, and SLRFB (Table 3). However, for other RFB technologies this might be different. He et al.[53] investigated the environmental impacts of three RFB technologies: VRFB, ZBFB and IFB. Regarding the ZBFB, the authors found that the bipolar plate produced from the titanium metal of the cell stack contributes to a large share of the environmental impacts in most impact categories (i.e., global warming potential, ozone depletion potential, acidification potential, fine particulate matter). In the case of IFBs, the polymer resins are responsible for a high share of environmental impacts,[53] and, in the case of AB-FB, the membrane manufacturing accounts for almost 80% of the GWP with sodium nitrate and aniline representing the key contributing materials.[58]

The cell stack materials can also be responsible for a high share of environmental impacts. The production of the Nafion® membrane for the VRFB contributes 76–90% to ozone depletion (OD) impacts due to the tetrafluoroethylene polymer utilized in its production.[41,50,53] In comparison, the titan bipolar plate of the ZBFB has much lower effects on ozone depletion than the Nafion® membrane for the VRFB.[53]

In contrast to the VRFB, processing the BEDFB electrolyte and some cell stack components require less material and energy. Morales-Mora et al.[55] reported that 90% of BEDFB components such as the cell stack, tank and peripheral components can be recycled. They further argued that no additional energy or materials are needed to recover sodium sulfate after the BEDFB disposal, unlike after the VRFB disposal. Unterreiner et al.[49] found that VRFB processing involved only 18% of reusable materials, since these batteries contain a high proportion of plastics; at this stage, these are assumed as not being recycled. However, the authors noted that a few materials used only to a small extent can have a significant influence on the environmental impact of the battery system.[49] For example, reducing or substituting polytetrafluoroethylene in VRFBs would significantly reduce the environmental impact of these batteries.[49]

Impact categories

The summary of the main drivers of environmental impacts in the resource and manufacturing phase shown in Table 3 clearly indicates that the components and material hotspots can differ, depending on which impact category is investigated. For instance, the electrolyte forms a hotspot in several FB systems, as is evident when examining the GWP impacts and other impact categories (e.g., resource depletion, human toxicity, or

Table 3. Summary of main drivers of environmental impacts per impact category for different flow battery technologies.

ESS	Components	Materials	Resource extraction
VRFB[23,41–43,45,50,51,53–56]	● **electrolyte** (59–90% GWP; 65–93% ARU; 41–75% HT; 95% AC) ● **Nafion® membranes** (90% OD) ● **current collector** and **copper cables** (46% /9% HT)	● **tetrafluoroethylene polymer** (OD) ● **copper** (55–100% HT; 20% RE; 23% AC) ● **vanadium pentoxide** (46–47% GWP; 53–58% AC; 58% RE; 95% ARU) ● **roll-formed metal** (97% OD) ● **low-alloyed steel** (22–46% in nine impact categories)	● **energy demand** for vanadium pentoxide production (50% HT; 46% GWP) ● **extraction of titanium and vanadium** (51% and 21% ARU)
ZBFB[53]	● **electrolyte** (ARU; 29% GWP) ● **bipolar plate** (40% GWP; AC; OD; RE; ED; Etox)	● **titanium** (GWP; AC; OD; RE; ED; Etox) ● **bromine** (98% ARU)	–
IFB[53]	● **cell stake membrane** (OD; Etox) ● **storage tank** (39% GWP) ● **cell frame** (22% GWP) ● **bipolar plate** (32% Etox)	● **bisphenol A epoxy-based vinyl ester resin** (Etox) ● **glass fiber-reinforced polymer** (52% Etox);	–
ZCB[52]	● **electrode** (92% of GWP); ● **electrolyte** (85% WU; 66% HT) ● **copper collectors** (25% HT)	● **cerium and titanium** (GWP; HT) ● **cerium and zinc** (75%/10% WU)	–
SLRFB[57]	● **electrolyte** (88% HT; 81% WU; 53% Etox) ● **bipolar and monopolar frames** (42–62% ET) ● **gasket and stack frame** (78% ARU; 68% LU; 42–50% Etox; 40% IR) ● **electrolyte tank** (48% Etox); ● **electrolyte holder** (35% HT);	● **lead monoxide** (87% HT; 81% WU; 52% Etox) ● **stainless-steel end plate production** (60–93% in all impact categories except HT)	● **lead oxide Barton pot process** (83–94% HT and ET)
AB-FB[58]	● **membrane** (80% GWP) ● **current collector** (AC) ● **electrodes** (6 impact categories) ● **inverter** (7 impact categories) ● **gaskets** (ARU) ● **stack frame** (~38% HT)	● **sodium nitrate and aniline** (GWP) ● **copper** (Etox; AC) ● **polyethylene** (ARU)	● **low-alloy steel production and extraction of copper** (HT; ARU)
BEDFB[55]	● **pacer mesh and membranes** (40%/42% GWP) ● **cables and heat exchange** (51%/23% Etox) ● **process control system; and heat exchanger** (24%/50% HT) ● **energy components** (50% AC) ● **tank** (31% ARU)	● **ethylene chlorotrifluoroethylene and sPEEK** (40%/42% GWP) ● **stainless steel** (23% Etox; 50% HT) ● **copper** (51% Etox) ● **fiberglass** (31% ARU)	–

AC = acidification; ARU = abiotic resource use; GWP = global warming potential; Etox = ecotoxicity; ED = energy demand; ET = eutrophication; HT = human toxicity; IR = ionizing radiation; OD = ozone depletion; RE = respiratory effects; WU = water use.

acidification potential). Looking at other impact categories this might differ: Nafion® membranes in the VRFBs are the main drivers of the ozone depletion potential, the bipolar plates in the ZBFBs and IFBs contribute most to freshwater ecotoxicity, and the bipolar and monopolar frames of the SLRFBs form a hotspot for the eutrophication potential. In most of the reviewed studies, the highest environmental impacts associated with VRFBs can be assigned to the impact category "abiotic resource use".[43,47,50] This is mainly due to the RFB manufacturing phase, as many battery components consist of abiotic materials.[78]

When analyzing the environmental impacts of ESS, the impacts on human health should be considered.[44] Weber et al.[50] found that the main contributor to HT is the electrolyte production and, more precisely, the production of vanadium pentoxide and thereof the slag production (50% of the total HTP). These impacts can be mainly attributed to the electricity demand, the mining operation and the slag treatment.[23,50] High-purity vanadium pentoxide is a highly toxic material that is suspected of causing cancer if inhaled during the extraction and refining processes.[11] Other studies found that the copper components used (e.g., the current collector and the copper cables in VRFBs) contribute the most to HT[23,52] and to acidification in the case of AB-FB.[58] Roasting vanadium-bearing slag prior to subjecting it to acid leaching and the copper production process results in significant sulfur dioxide (SO_2) emissions, highly impacting AC.[50,58] Diaz-Ramirez et al.[54] even found that the copper used in the VRFB electrode compound was responsible for 48–84% of the environmental burden in 13 out of 18 impact categories. Comparing different types of RFBs, the HT impacts are higher for ZCB than for VRFBs, though the Ce electrolyte is responsible for 66% of the HT impacts of the ZCB.[52] In the case of SLRFB, the electrolyte and therefore the lead-based compounds are also responsible for 88% of the HT impacts, although lead-based components are known for their human toxicity.[57,79] In the case of the IFB, the cell stake membrane forms a hotspot in the impact categories of ozone depletion potential and freshwater ecotoxicity.[53]

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Life cycle phases

Much of the impact in several impact categories is due to the ESS manufacturing phase.[25] This is also the case for VRFB,[23,40] but differently manufactured vanadium electrolytes can differ significantly in the level of their environmental impacts.[40] The findings of He et al.[53] support this statement and show that the material choice and production type can influence the environmental impact both positively and negatively. In the manufacturing phase, the material extraction is responsible for many of the environmental impacts[49,51] i.e., vanadium contributes 95% to the category of mineral resource scarcity and is responsible for many of the impacts regarding GWP, acidification and fine particulate matter formation (RE; Table 3). A problematic factor in material extraction is that a critical raw material such as vanadium pentoxide is mined for the electrolytes used in VRFBs,[17,54] and the energy mix used for the production includes a high share of non-renewable energy.

Other studies found that the manufacturing phase has only a minor influence on the life cycle impact of the battery as compared to the use phase. Here, the round-trip efficiency of the charge–discharge cycle, the assumed lifetime of the battery, the energy mix/type of energy provision for charging, and the transport have the most influence.[24,43,50,55,56] During the operation, energy is charged and discharged, and the impacts are predominantly influenced by the overall efficiency (round-trip efficiency) of the charge–discharge cycle (e.g., depth of discharge or self-discharge) and the battery lifetime (Table 2).[25,80] Increasing the round-trip efficiency and using renewable energy sources reduces the environmental impact.[25,43] However, the choice of renewable energy sources can also differ. Da Silva Lima et al.[23] showed that an ESS consisting of a wind turbine and a VRFB has much lower environmental impacts than a system consisting of a photovoltaic system and a VRFB. This result cannot be extrapolated, however, as the geographical context and its energy mix highly influenced the results.[43] The influence of the transport can be explained by the high weight of the electrolyte.[52] This means that the higher energy densities of the electrolyte and shorter transport distances will again have more positive effects on the environment. Increasing the length of the service life also reduces the environmental impacts.[43] In addition, Mostert et al. found that the greenhouse gas emissions and material consumption of VRFBs decrease sharply as storage capacity increases, which can be attributed to scaling effects.[46] In the case of an AB-FB, the phase contributing the most to the total impacts is also the use phase (referred to as operations and maintenance), which can be attributed to the electricity losses generated when the battery is serviced to support wind and PV installations.[58]

The end-of-life contribution to the total impacts is rather minor as compared to the other stages,[23,58] except for the impact category of freshwater eutrophication potential in case of VRFB and ZBFB, as well as in the case of IFB systems where the waste treatment accounts for 84–88% of the impacts.[53] In most cases, however, this phase only makes a small contribution to the overall impacts, but substantial reductions can be made by the recycling of materials and components.[45,50,51] Díaz-Ramírez et al.[45] showed that using recycled materials can decrease the impact by more than 50% in most of the impact categories for VRFB. According to Weber et al.,[50] as compared to a lithium iron phosphate battery, the VRFB is better and easier to recycle, because the components are easier to disassemble. The authors assumed a recovery rate of 95% for the vanadium electrolyte.[50] Jones et al.[51] concluded that the end-of-life phase is particular notable regarding the reduction in GWP when the ESS materials are recycled.

4.2. Environmental performance of RFBs as compared to other storage technologies

In total, eight different electrochemical flow battery technologies were investigated, although only the VRFB was assessed more than once. The LCA results depend on a variety of factors and methodological decisions, such as the chosen system boundaries (Figure 2) or impact categories (Figure 3). The LCA results, therefore, do not allow clear statements about one technology being more favorable from an environmental perspective than the other. Instead, an environmental profile is provided, presenting results for different impact categories and allowing insights into the potential environmental risks connected with different environmental topics. Comparing the environmental impacts of ESS across different studies quantitatively is rarely possible due to the different assumptions made about the technical performance, the methodological choices, and the type of results provided by the studies (see Table 4). However, it is possible to summarize the results of the LCA studies by ranking the environmental performance of the investigated ESS in different impact categories: Figure 4 shows the ESS ranking for the GWP results and Figure 5 displays the ranking for human toxicity, ranging from the lowest (green) to the highest (red) environmental impacts.

VRFB compared to LIB

VRFB can have lower environmental impacts than LIBs[24,46,50,54] and even more so if the benefits of recycling are taken into account.[50] These results depend heavily on the data and the assumptions made; for example, Da Silva Lima et al.[23] could not clearly state which technology is superior from an environmental perspective. Figure 4 shows a similar picture with regard to GWP, where five out of eight studies comparing LIB and VRFB reported a lower GWP for LIB, and three studies came to the opposite conclusion. Figure 5 provides a clearer picture: All studies comparing these technologies and analyzing their effects on the human toxicity potential found that VRFBs offer benefits over LIBs. Similarly, lower impacts can be expected for VRFB with regard to respiratory effects and acidification (see Supporting Information).[23] In spite of the fact that vanadium dusts can be very toxic when inhaled,[11,15,16] LIBs have been reported to perform even more poorly in this category (Figure 5). The results of Hiremath et al.[24] show that, when the

Table 4. Selected impacts of flow battery technologies per 1 MWh electricity stored/delivered including assumed life span and system boundaries; the source of the values in some cases is indicated in brackets.

	Flow battery	Operating time [years; cycles]	System boundary	GWP [kg CO_2 equiv.]	HTP [kg 1,4-DCB equiv.]	AP [kg SO_2 equiv.]	ODP [kg CFC-11 equiv.]	ARU [kg Sb equiv.]
[48]	VRFB	20; 7300	c2cradle	19842.2	–	131.1	–	–
[24]	VRFB	20; 13000	c2gate	14.8–93 (SI)	–	–	–	–
			c2use	208–14802 (SI)	–	–	–	–
[47]	VRFB	–	–	24.8–58.5	–	–	–	8.2–19.6 kg oil equiv.
[40]	VRFB	–	c2gate	–	–	–	–	–
[49]	VFB	20; 4000	c2cradle	–	–	–	–	–
[50]	VFB	20; 8176	c2gate	38.2	52.3	0.63	–	0.011
			c2cradle	20–279 (FIG)	45–160 (FIG)	0.13–1.2 (FIG)	–	4.5–25 (FIG)
[46]	VRFB	20; 7300	c2use	53	–	–	–	–
[59]	CGFB	20	c2grave	190000	–	–	–	–
[51]	VRFB	30	c2grave	52.8–443	1.98e7–6.45e7	1.87e5–1.25e6	256–324	5.97e6–6.24e7 kg oil equiv.
[43]	VRFB	20	c2grave	–	–	–	–	–
[42]	VRFB	20	c2grave	130 (FIG)	–	–	70000000 (FIG)	60000 (FIG)
[41]	VRFB	–	c2gate	–	–	–	–	–
[52]	VRFB	–	c2gate	136500	225000 kg 1,4–DB equiv.	1670	0.03	1.41
	ZCB	–	c2gate	224400	301600 kg 1,4–DB equiv.	1550	0.04	1.67
[53]	VRFB	–	c2gate	184000 (FIG)	–	1100 (FIG)	0.176 (FIG)	3 (FIG)
	ZBFB	–	c2gate	158000 (FIG)	–	670 (FIG)	0.06 (FIG)	37 (FIG)
	IFB	–	c2gate	72000 (FIG)	–	370 (FIG)	0.085 (FIG)	1 (FIG)
[45]	VRFB	20	c2gate	–	–	–	–	–
[44]	VRFB	20	c2use	–	–	–	–	–
[54]	VRFB	20	c2gate	–	–	–	–	–
[23]	VRFB	20; 6000	c2gate	57	120.9	0.6	–	15.1 kg oil equiv.
			c2grave	108	173.8	0.8	–	25.5 kg oil equiv.
[55]	BEDFB	20; 8176	c2gate	9.1	4.9	0.018	1.8e–4 (SI)	1.2 kg oil equiv.
			c2grave	16.6–205.7	6.5–81	0.1–0.67	–	1.8–2.0 kg oil–cu
[56]	VRFB	20	c2use	121	2341.6	0.5	–	–
[57]	SLRFB	–	c2gate	749820	324770 kg 1,4–DB equiv.	2,450	0.19	315010 kg oil equiv.
	VRFB	–	c2gate	217000	451000 kg 1,4–DB equiv.	3370	–	72000 kg oil equiv.
[58]	AB–FB	20; 10000	c2gate	7.52	35	0.03	–	1.15 kg oil equiv.
	AB–FB		c2grave	36.6	236.97	0.19	–	0.78 kg oil equiv.
	VRFB		c2gate	30.54	1.85e12	0.47	–	0.83 kg oil equiv.
	VRFB		c2grave	46.86	1.85e12	0.57	–	9.24 kg oil equiv.

SI = See the Supporting Information; FIG = from the graph; c = cradle; GWP = global warming potential; HTP = human toxicity potential; AP = acidification potential; ODP = ozone depletion potential; ARU = abiotic resource depletion.

whole life-cycle impacts of different ESS were investigated, the VRFB had higher GWP and ED impacts than the LIB, but a lower impact than the LAB and the sodium-sulfur battery (NaS). In this study, the impacts were mainly influenced by the use-phase impacts, where the electricity is being wasted in each charge–discharge cycle.[24] LIB batteries have a round-trip efficiency of 90% and VRFB just 75% with the highest use-stage impacts compared to the other battery technologies (LIB, LAB and NaS).[24]

VRFB compared to LAB

Most studies comparing LAB with VRFB technologies reported lower impacts for VRFBs regarding GWP and human toxicity (Figures 4 and 5) and most other impact categories considered.[24,48,54,55] This is because LABs and NaS batteries have relatively low cycle lives as compared to VRFBs and LIBs, resulting in higher cradle-to-gate impacts.[24] However, Unterreiner et al.[49] found that VRFBs are more environmental harmful than LABs. Most VRFB impacts are caused in the end-of-life and resource extraction phases. VRFBs have a high share of plastic materials (e.g., polytetrafluoroethylene) which contribute to 40% of the total impacts; these materials are more complicated and costly to recycle, so recycling was not considered in their assessments.[49] As compared to CGFB, LIB, pumped hydro energy storage (PHES), and compressed air energy storage (CAES), LABs are the least-preferred system with regard to human health and ecosystem diversity (i.e., 46% of impacts are caused by the use of antimony and primary aluminum).[59] Shittu et al. (Table 1, entry 21 and Figures 4 and 5) came to another conclusion: They compared the impacts of seven ESS and found that LABs are the least harmful technology regarding GHG emissions, human toxicity, acidification and ozone depletion.[57] The authors argued that LABs are well established and that closed-loop recycling at the end-of-life contributes to mitigating impacts.[57]

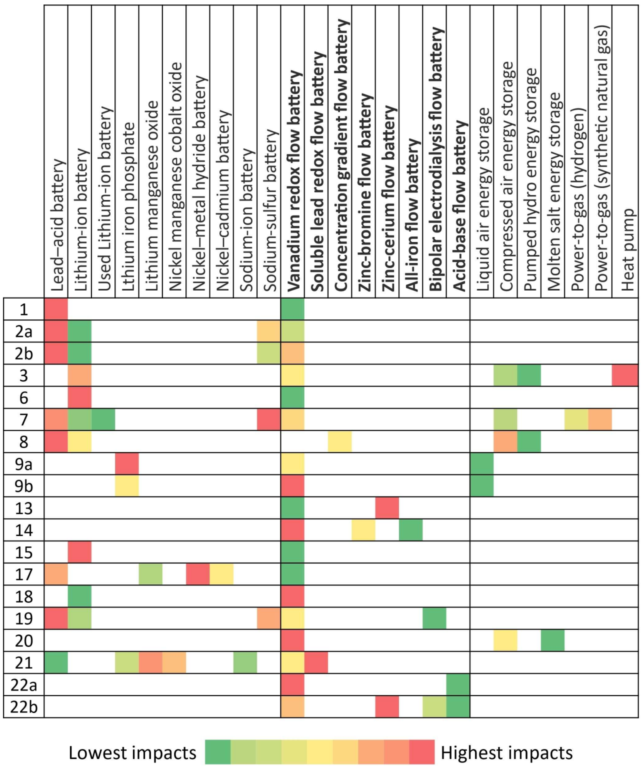

Figure 4. Ranking of ESS from lowest impact to highest global warming potential impact per study (row). The numbers along the vertical axis provide the article numbers given in Table 1. Some studies are listed twice, and the results of different scenarios were transferred into the matrix here (see Supporting Information). The flow batteries are highlighted in bold.

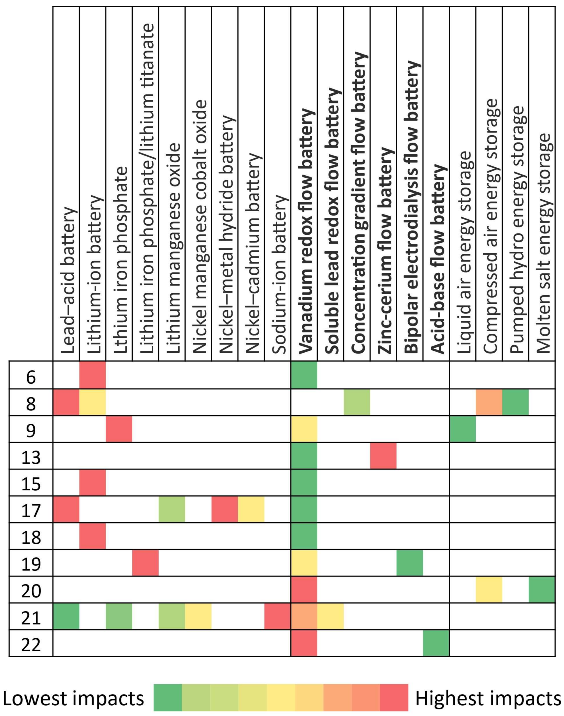

Figure 5. ESS ranking from lowest impact to highest human toxicity impact per study (row). The numbers on the vertical axis provide the article numbers shown in Table 1. The flow batteries are highlighted in bold.

RFB compared to other ESS

As compared to other storage technologies, a VRFB can have a lower environmental impact than an LMO in most impact categories,[45] a nickel-metal hydride and nickel–cadmium,[54] a NaS,[24,46,55] or a LFP[51] battery. Compared to LFPs, whether VRFBs can be better depends on the assumed life span of the LFP cells (7.5 or 10 years).[51] Higher impacts can be expected for the VRFB when compared to CAES[46,47] or LAES.[51] If we examine other RFB technologies, a CGFB performs better than a CAES, LIB, or LAB, but worse than a PHES.[59] SLRFBs are less environmentally beneficial, and they perform worse than LAB, LFP, LMO and sodium-ion batteries in most impact categories except for ecotoxicity and eutrophication (Figures 4 and 5).

Comparison of different RFB technologies

The comparison of two RFBs in Fernandez-Marchante et al.[52] showed that a ZCB has higher impacts than a VRFB in almost all impact categories. The exceptions were the acidification and water footprint categories, as the production of a VRFB requires four times more water than that of a ZCB.[52] He et al.[53] examined the potential environmental impacts of three flow battery technologies (i.e., VRFB, IFB and ZBFB) in eight different impact categories. They found that the IFB performed best in most impact categories after ZBFB and VRFB. The exceptions were in the impact categories of ozone depletion and freshwater ecotoxicity, where the ZBFB had the lowest impacts.[53] An AB-FB system has a better environmental performance as compared to a VRFB system.[58] One of the main advantages of using the AB-FB system is that the main components of the AB-FB electrolytic solution are water and salts, which makes it a safer and more sustainable technology than the VRFB system.[58] A BEDFB was investigated and compared to VRFB by Morales-Mora et al.[55] The authors found that a BEDFB can be less harmful to the climate than a VRFB. Shittu et al.[57] investigated the environmental performance of a SLRFB and compared it with that of a VRFB; they found that the SLRFB is preferable with respect to some impact categories (i.e., respiratory effects, human toxicity, mineral resource scarcity, acidification) but not for others (i.e., climate change and fossil resource scarcity). Overall, the VRFB is the least preferable RFB technology from an environmental point of view when compared to IFB, ZBFB, AB-FB, BEDFB and – for some impact categories – SLRFB. Only a ZCB has higher impacts than a VRFB; therefore, it is less environmental beneficial. Comparing the impacts in terms of the GWP and human toxicity (see Figure 4 and Figure 5) of different flow battery technologies, the results indicate that the most preferable technologies are the BEDFB, AB-FB and IFB, and the least preferable technologies are the ZCB, SLFB and VRFB. These results strongly depend on the types of technologies that are compared to each other.

5. Challenges and Future Aspects

Although the sustainability of VRFBs have been studied to some extent, emerging flow battery types such as organic aqueous and non-aqueous flow batteries have not yet been systematically investigated, with the exception of acid-base flow batteries.[58] Nevertheless, the abundance and facile modification of organic molecules in nature make them potential candidates as active materials in flow battery technology systems.[81,82] In addition, decomposition mechanisms and other properties can be studied by using a wide range of analytical and computational methods.[82] However, the disadvantage of assessing the sustainability of organic RFBs is that they are not currently commercially available, with TRLs of 5–7 being the most developed systems reported so far.[83] This implies that the upstream processes needed to produce electrochemically active organic molecules are not yet known or have not been developed, impeding the collection of necessary data. In addition, information about the performance of such batteries when operating at larger scales and over longer periods is not yet available. Hence, assessments must be based on assumptions about their maintenance and durability, which introduce high levels of uncertainty. Accordingly, Chebaeva et al.[63] suggested that a minimum of TRL 6 is needed to perform a full LCA, while lower levels can be acceptable for performing streamlined LCAs. This results in a general problem: Technologies are compared at different stages of development. When performing an LCA of different ESS, the system boundaries, data sources used and assumptions made definitely depend on the respective TRL. However, such comparisons are regularly done despite these issues, since comparisons need to be made to environmentally benchmark new technologies against state-of-the-art solutions.[63]

5.1. Influence of methodological choices

Comparability is not only an issue because the technologies are in different TRLs, but also because many methodological decisions needed to be made in LCAs, and a variety of LCIA methods is available.[69] Rahman et al.[25] synthesized results for LCAs of different ESS and summarized the impacts reported in the respective studies. They found that the ranges of impact per ESS are quite broad (e.g., −439 to 810 gCO_2 equiv. kWh^{-1} "cradle to grave" impacts for LIB),[25] which illustrates the fact that comparing the technologies analyzed in different studies is difficult if not impossible (see Table 4).[25,26] The issue of the impact categories considered in the studies is also related to the comparability, because their selection highly influences the results. This selection process depends on factors like the objective of the study, the environmental issues associated with the product, process, or material under investigation, the availability of an LCIA method and how up-to-date it is, as well as the geographical scope of the study.[69] Also methodological choices are often made with reference to previous studies with a similar research focus.[69] The environmental issue most often analyzed in LCAs on automotive components, wood products, or LIBs is climate change (CC).[27,69–71] In the 22 studies reviewed here, however, the most frequently investigated impact categories (other than CC) are abiotic resource use (ARU), ecotoxicity (Etox), human toxicity (HT) and respiratory effects (RE). Still, the relevance of certain impact categories in the context of different ESS is rarely discussed. In the reviewed studies, some hotspots in impact categories could be identified: abiotic resource use,[43,47,50] human toxicity (especially the electrolyte production) for VRFB[50] and human toxicity connected with the Ce electrolyte of the ZCB.[52] Some materials were identified as main drivers for impacts in certain impact categories: Copper, cerium and lead form a hotspot in human toxicity, and vanadium pentoxide and titanium are connected with acidification and respiratory effects, especially regarding fine particulate matter formation (Table 3).

For some ESS, these findings indicate that environmental impact categories other than CC are of greater relevance, but the uncertainty associated with the TRL of the technologies and the impact categories must be considered.[27,63] The impact categories of water consumption and ionizing radiation are especially associated with high levels of uncertainty.[84] Still, our review findings indicate that the high impact attributed to the consumption of critical raw materials constitutes a key driver for substituting such materials with more easily available alternatives.[54]

5.2 Uncertainties in research and development

In the early stages of technological development, the degree of uncertainty is highest; therefore, uncertainty analyses should be performed whenever possible.[66,85] Several studies investigated the types of uncertainty and how to deal with them in an LCA.[75,76,86] Existing LCA tools and databases have implemented methods to analyze the uncertainty level with qualitative or quantitative uncertainty scores.[74,87] The approach most frequently used to analyze parameter uncertainty is the Monte Carlo method, which is also incorporated in the LCA software Simapro and can be used to model uncertainties related to input parameters.[66,87] Most studies quantified only the parameter uncertainty[76] and variability,[75] although all types of uncertainty contribute to varying degrees to the overall uncertainty of the LCA result.[75] Quantitative uncertainty management as done by Fernandez-Marchante et al.[52] or Baumann et al.[44] is still a rare sight in LCA practice.[75] Quantifying uncertainty means establishing a measure of uncertainty (e.g., mean value, standard deviation and distribution type) and the overall uncertainty of an LCA.[75] However, we found that the researchers often relied on inventories from previous studies. To perform quantitative uncertainty management, uncertainty ranges are needed for the values provided in the inventory tables.[50] Different methods can be used to analyze uncertainty, for example, by reporting uncertainty intervals, analyzing parameter variability and/or different scenarios, using a pedigree matrix or fuzzy data sets, applying analytical uncertainty propagation, or conducting numerical, probabilistic simulations.[75,76,86] Even if an uncertainty analysis is

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performed, the results should be interpreted with caution.[76] For more detailed information about uncertainty management in LCA studies, we refer the reader to the works of Huijbregts et al.,[76] Rosenbaum et al.[75] and Björklund et al.[77] To analyze the influence of the underlying assumptions on the results, a sensitivity analysis was performed in most of the reviewed studies. Mostert et al.,[46] for instance, identified significantly influential parameters, analyzed variations of these parameters, and illustrated the results in bar graphs, including ranges of results. The parameters most often analyzed are variations of the energy mix, efficiency, and use of secondary materials or alternative materials for certain components. The results of such sensitivity analyses can then be used to support eco-design, green chemistry, or SSbD by guiding actions for improvement.

5.3 Standardization

All the above-mentioned issues regarding uncertainty are also due to the lack of standardization for the technology parameters; this is, however, a typical situation for emerging technologies. This particularly refers to the need for standardized procedures that can be used to test RFB systems, like those already established in photovoltaics, lithium-ion batteries, and supercapacitors, which would make the different systems more comparable.[88] However, one challenge is the need to consider different use cases, which makes the development of standardized procedures more difficult than one might think. For example, many scenarios to test a system at 10 kW exist; these result in different performances at different settings. Some of these issues seem trivial, for example, the need to standardize a procedure to showcase its dependency on the current density and voltage efficiency. Others are more difficult to grasp, because they are related to the actual operation conditions of the battery system (e.g., charging and discharging time, extent of discharge, flow rate etc.). Considering these aspects, however, enables the user to compare different systems in terms of their technical performance (see Table 2). The consequence for LCA: Different RFB systems can rarely be compared to each other. This creates an unsatisfying situation for researchers working with LCA but also for companies and stakeholders who are interested in the actual environmental footprint of such systems. Standardization of the methodological procedure of LCA is another highly important topic. Dieterle et al.[26] provided good practice examples and recommendations for standardizing all four phases of LCA, and we refer readers to their work for more guidance on this issue.

5.4 Holistic sustainability assessment

Assessing the sustainability of materials or technologies must go beyond merely considering environmental effects. The review of SSbD chemicals and materials by the European Commission suggests that economic, social and safety considerations should be included in the assessment.[19] The economic pillar was addressed in several studies, for example, by performing a cost assessment such as life cycle costing.[44,58] However, one aspect which has not been considered in recent studies is the social and socio-economic impacts of using RFB systems, although some authors indicate that such efforts are ongoing. For example, Baumann et al.[44] considered social indicators in a multicriteria decision analysis, and Gouveia et al.[41,42] highlighted the importance of assessing the social impacts. This is an important aspect, because not only the environment suffers from the increasing demand for metals; the local populations in several mining areas also have to deal with human right abuses, child labor and life-threatening working conditions that non-regulated mining companies impose on them. Cobalt mining in the Democratic Republic of Congo provides one example of this.[89] In the past 5–10 years, substantial policy-making efforts have been made to ensure that raw material sourcing is done sustainably and responsibly.[90] Materials used in battery technologies and especially in Li-ion battery raw materials, such as cobalt, graphite, lithium and manganese, have received special attention, with researchers focusing on their responsible sourcing.[23,90,91] This also includes the development of efficient recycling procedures.[18,34,49] The potential social and socio-economic impacts of products can be assessed by performing social life cycle assessment, referring to the UNEP guidelines[92] or the handbook on product social impact assessment.[93]

6. Conclusions and Outlook

Our review summarizes the main findings of life cycle assessment studies on redox-flow batteries. The hotspots or respectively the main drivers of environmental impact in different impact categories for eight redox-flow battery technologies are identified, and the environmental performance of these technologies as compared to other ESS are discussed. The review shows that the investigation of potential environmental impacts extends far beyond climate change impacts. A clear picture rarely emerges regarding which ESS is the most preferable from an environmental perspective, and especially when considering more impact categories. This review identifies the areas of concern in the context of RFBs, which are mostly connected to specific materials, the energy mix used to produce and operate the batteries. The operation conditions and the technological performance of the battery are determining factors, with cycle life, materials environmental footprint and round-trip efficiency being major drivers to impact results. Here, components and material hotspots can largely vary depending on which impact category is investigated. Still, comparisons between different ESS must be made on a case-to-case basis to ensure constant assumptions and conditions in order to reduce complexity. In contrast, our review revealed that any hotspot analysis in ESS can help to guide technological development and to improve the environmental performance of RFBs. In the future, researchers might synthesize the LCAs results of other product systems to identify a pattern of environmental concern associated with specific technologies or product systems. Such a synthesis would support research and development, helping scientists to develop and assess product systems in low TRL more effectively when data availability is an issue. Key parameters that were shown to affect the LCA results of RFBs comprise specific materials in certain impact categories, such as copper (regarding acidification), vanadium pentoxide, cerium, and lead (regarding human toxicity) and the Nafion® membrane (regarding ozone depletion). Increasing the recyclability of such materials or using less environmental harmful materials (e.g., organic-based materials) can substantially reduce impacts. The efficiency, depth of discharge, battery life time, type of energy provision and the transport were identified as other parameters influencing the life cycle impacts. In this respect, increasing round-trip efficiencies mean that less energy is wasted. Using LIBs with rather high efficiencies was shown to result in lower use-phase impacts as compared to using VRFBs. Efficiency is especially important when the energy mix in the applied region has a low share of renewable energy carriers. Besides identifying the relevant environmental issues of RFBs, our review shows that relevant social issues need to be identified by consulting affected stakeholder groups (e.g., workers, users or the local community).[92] As a starting point, that would provide guidance when attempting to identify social issues for specific product systems, we recommend to identify social topics connected with certain countries or regions.[69] Furthermore, neither an LCA nor an social LCA case study of organic RFB systems currently exists; this is needed to support sustainability claims for these organic-based electrolytes.[13,81,82]

Considering various impact categories in the different sustainability dimensions (e.g., environmental, economic, social) ultimately leads to the problem of optimizing several objectives simultaneously. Hence, multi-objective optimization approaches will be needed to identify the most suitable options. In the context of ESS, the main objective contributing to the decarbonization of the energy system is related to minimizing GHG emissions. Our review shows that some of the technological options that can be used to achieve this goal have trade-offs, and particularly those related to human toxicity, ecotoxicity, resource depletion, or social impacts. By making a normative decision on acceptable impact levels in respective impact categories, an according optimization process would clearly be enabled. Organic redox flow batteries seem to be promising alternatives in this context, but a higher TRL is urgently required which would ensure the comparability needed to perform comparative assessments. Although LCA is a quantitative approach, which would allow comparisons of similar technologies, the differences in the methodological choices make comparisons almost impossible. Most of the methodological decisions are based on the goal and scope definition and these varied strongly from study to study. The study results were presented in ways that allow statements to be made with regard to the study goals. The results were often only presented graphically, and no data were provided. In general, this makes it challenging to synthesize LCA results presented for similar technologies in published studies. A standardized way of reporting and presenting LCA results is needed to extract meaningful quantitative information. In this review, we decided to extract information on the environmental hotspots and performance, comparing them to other ESS qualitatively and summarizing the findings. The review results provide a first impression of the areas of concern regarding different flow battery technologies and reveal trends in the FB performance as compared with each other and with other ESS.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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[1] F. Kern, A. Smith, *Energy Policy* **2008**, *36*, 4093.
[2] K. Hainsch, K. Löffler, T. Burandt, H. Auer, P. Del Crespo Granado, P. Pisciella, S. Zwickl-Bernhard, *Energy* **2022**, *239*, 122067.
[3] United Nations (Ed.) *Paris Agreement. Report of the Conference of the Parties to the United Nations Framework Convention on Climate Change*. 21st Session, Paris, **2015**.
[4] European Commission (Ed.) *The European Green Deal. COM(2019) 640 final*, Brüssel, **2019**.
[5] a) M. Beaudin, H. Zareipour, A. Schellenberglabe, W. Rosehart, *Energy Sustain. Dev.* **2010**, *14*, 302; b) P. Denholm, E. Ela, B. Kirby, M. Milligan, Role of Energy Storage with Renewable Electricity Generation, **2010**; c) P. Denholm, M. Hand, *Energy Policy* **2011**, *39*, 1817; d) G. Pleßmann, M. Erdmann, M. Hlusiak, C. Breyer, *Energy Procedia* **2014**, *46*, 22.
[6] H. Zhao, Q. Wu, S. Hu, H. Xu, C. N. Rasmussen, *Appl. Energy* **2015**, *137*, 545.
[7] Y. Tian, G. Zeng, A. Rutt, T. Shi, H. Kim, J. Wang, J. Koettgen, Y. Sun, B. Ouyang, T. Chen, Z. Lun, Z. Rong, K. Persson, G. Ceder, *Chem. Rev.* **2021**, *121*, 1623.
[8] a) Y. Chen, Y. Kang, Y. Zhao, L. Wang, J. Liu, Y. Li, Z. Liang, X. He, X. Li, N. Tavajohi, L. Baohua, *J. Energy Chem.* **2021**, *59*, 83; b) Y. Jia, J. Darst, A. Surelia, D. Delafuente, D. P. Finegan, J. Xu, *J. Power Sources* **2022**, *539*, 231607; c) B. Liu, J. Zhang, C. Zhang, J. Xu, *Eng. Failure Anal.* **2018**, *91*, 315; d) C. T. Love, C. Buesser, M. D. Johannes, K. E. Swider-Lyons, *J. Electrochem. Energy Convers. Storage* **2018**, *15*.
[9] B. R. Swopes, "This Day in Aviation. Important Dates in Aviation History", can be found under https://www.thisdayinaviation.com/, **2017**.
[10] L. H. Thaller, *9th Intersociety Energy Conversion Engineering Conference* **1974**, 924.
[11] A. Ciotola, M. Fuss, S. Colombo, W.-R. Poganietz, *J. Energy Storage* **2021**, 102094.
[12] O. Schmidt, S. Melchior, A. Hawkes, I. Staffell, *Joule* **2019**, *3*, 81.
[13] X. Wei, W. Pan, W. Duan, A. Hollas, Z. Yang, B. Li, Z. Nie, J. Liu, D. Reed, W. Wang, V. Sprenkle, *ACS Energy Lett.* **2017**, *2*, 2187.
[14] A. Z. Weber, M. M. Mench, J. P. Meyers, P. N. Ross, J. T. Gostick, Q. Liu, *J. Appl. Electrochem.* **2011**, *41*, 1137.
[15] M. Imtiaz, M. S. Rizwan, S. Xiong, H. Li, M. Ashraf, S. M. Shahzad, M. Shahzad, M. Rizwan, S. Tu, *Environ. Int.* **2015**, *80*, 79.
[16] A. Wilk, D. Szypulska-Koziarska, B. Wiszniewska, *Postepy Hig. Med. Dosw.* **2017**, *71*, 850.

[17] EC, Study on the review of the list of critical raw materials. Criticality Assessments. Final Report, European Commission, Brussels, **2017**.
[18] M. Petranikova, A. H. Tkaczyk, A. Bartl, A. Amato, V. Lapkovskis, C. Tunsu, *Waste Manage.* **2020**, *113*, 521.
[19] European Commission, Safe and sustainable by design chemicals and materials. *Review of safety and sustainability dimensions, aspects, methods, indicators, and tools*, JRC Technical Report, Publications Office of the European Union, Luxembourg, **2022**.
[20] European Commission, *Strategic Research and Innovation Plan for safe and sustainable Chemicals and Materials*, Publications Office of the European Union, Luxembourg, **2022**.
[21] ISO, *14040:2006. Environmental management – Life cycle assessment – Principles and framework*, Switzerland, **2006**.
[22] a) B. Ness, E. Urbel-Piirsalu, S. Anderberg, L. Olsson, *Ecol. Econ.* **2007**, *60*, 498; b) M. Zimek, A. Schober, C. Mair, R. J. Baumgartner, T. Stern, M. Füllsack, *Sustainability* **2019**, *11*, 1–19.
[23] L. Da Silva Lima, M. Quartier, A. Buchmayr, D. Sanjuan-Delmás, H. Laget, D. Corbisier, J. Mertens, J. Dewulf, *Sustain. Energy Technol. Assess.* **2021**, *46*.
[24] M. Hiremath, K. Derendorf, T. Vogt, *Environ. Sci. Technol.* **2015**, *49*, 4825.
[25] M. M. Rahman, A. O. Oni, E. Gemechu, A. Kumar, *Energ. Convers. Manage.* **2020**, *223*.
[26] M. Dieterle, P. Fischer, M.-N. Pons, N. Blume, C. Minke, A. Bischi, *Sustain. Energy Technol. Assess.* **2022**, *53*, 102457.
[27] F. Arshad, J. Lin, N. Manurkar, E. Fan, A. Ahmad, M.-N. Tariq, F. Wu, R. Chen, L. Li, *Resour. Conserv. Recycl.* **2022**, *180*, 106164.
[28] S. Kosai, U. Takata, E. Yamasue, *J. Cleaner Prod.* **2021**, *280*, 124871.
[29] a) A. Nordelöf, S. Poulikidou, M. Chordia, F. Bitencourt de Oliveira, J. Tivander, R. Arvidsson, *Batteries* **2019**, *5*, 51; b) M. Slattery, J. Dunn, A. Kendall, *Resour. Conserv. Recycl.* **2021**, *174*, 105755.
[30] a) E. Emilsson, L. Dahllöf, *Lithium-Ion Vehicle Battery Production. Status 2019 on Energy Use, CO2 Emissions, Use of Metals, Products Environmental Footprint, and Recycling.* No. C 444, IVL Swedish Environmental Research Institute, **2019**; b) J. F. Peters, M. Baumann, B. Zimmermann, J. Braun, M. Weil, *Renewable Sustainable Energy Rev.* **2017**, *67*, 491.
[31] J. F. Peters, M. Weil, *J. Cleaner Prod.* **2018**, *171*, 704.
[32] Q. Tan, J. Li, *Environ. Sci. Technol.* **2015**, *49*, 5849.
[33] Y. Yang, X. Zheng, H. Cao, C. Zhao, X. Lin, P. Ning, Y. Zhang, W. Jin, Z. Sun, *ACS Sustainable Chem. Eng.* **2017**, *5*, 9972.
[34] S. Windisch-Kern, E. Gerold, T. Nigl, A. Jandric, M. Altendorfer, B. Rutrecht, S. Scherhaufer, H. Raupenstrauch, R. Pomberger, H. Antrekowitsch, F. Part, *Waste Manage.* **2022**, *138*, 125.
[35] M. Mohr, J. F. Peters, M. Baumann, M. Weil, *J. Ind. Ecol.* **2020**, *24*, 1310.
[36] Z. Deller, L. A. Jones, S. Maniam, *Green Chem.* **2021**, *23*, 4955.
[37] X.-Z. Yuan, C. Song, A. Platt, N. Zhao, H. Wang, H. Li, K. Fatih, D. Jang, *Int. J. Energy Res.* **2019**.
[38] M. Skyllas-Kazacos, L. Cao, M. Kazacos, N. Kausar, A. Mousa, *ChemSusChem* **2016**, *9*, 1521.
[39] Z. Xu, Q. Fan, Y. Li, J. Wang, P. D. Lund, *Renewable Sustainable Energy Rev.* **2020**, *127*, 109838.
[40] M. Dassisti, G. Cozzolino, M. Chimienti, A. Rizzuti, P. Mastrorilli, P. L'Abbate, *Int. J. Hydrogen Energy* **2016**, *41*, 16477.
[41] J. Gouveia, A. Mendes, R. Monteiro, T. M. Mata, N. S. Caetano, A. A. Martins, *Energy Rep.* **2020**, *6*, 95.
[42] J. R. Gouveia, E. Silva, T. M. Mata, A. Mendes, N. S. Caetano, A. A. Martins, *Energy Rep.* **2020**, *6*, 87.
[43] P. L'Abbate, M. Dassisti, A. G. Olabi, *Green Energy Technol.* **2019**, 61.
[44] M. Baumann, J. Peters, M. Weil, *Energy Technol.* **2020**, *8*.
[45] M. C. Díaz-Ramírez, V. J. Ferreira, T. García-Armingol, A. M. López-Sabirón, G. Ferreira, *Sustainability* **2020**, *12*, 6840.
[46] C. Mostert, B. Ostrander, S. Bringezu, T. M. Kneiske, *Energies* **2018**, *11*.
[47] A. Sternberg, A. Bardow, *Energy Environ. Sci.* **2015**, *8*, 389.
[48] C. J. Rydh, *J. Power Sources* **1999**, 21.
[49] L. Unterreiner, V. Jülch, S. Reith, *Energy Procedia* **2016**, *99*, 229.
[50] S. Weber, J. F. Peters, M. Baumann, M. Weil, *Environ. Sci. Technol.* **2018**, *52*, 10864.
[51] C. Jones, P. Gilbert, L. Stamford, *Environ. Sci. Technol.* **2019**.
[52] C. M. Fernandez-Marchante, M. Millán, J. I. Medina-Santos, J. Lobato, *Energy Technol.* **2020**, *8*, 1900914.
[53] H. He, S. Tian, B. Tarroja, O. A. Ogunseitan, S. Samuelsen, J. M. Schoenung, *J. Cleaner Prod.* **2020**, *269*, 121740.
[54] M. C. Díaz-Ramírez, V. J. Ferreira, T. García-Armingol, A. M. López-Sabirón, G. Ferreira, *Sustainability* **2020**, *12*.
[55] M. A. Morales-Mora, J. J. Pijpers, A. C. Antonio, J. d. C. La Soto, A. M. A. Calderón, *J. Energy Storage* **2021**, *35*, 102339.
[56] M. AlShafi, Y. Bicer, *Energy Rep.* **2021**, *7*, 7090.
[57] E. Shittu, R. Suman, M. K. Ravikumar, A. K. Shukla, G. Zhao, S. Patil, J. Baker, *J. Cleaner Prod.* **2022**, *337*, 130503.
[58] M. C. Díaz-Ramírez, M. Blecua-de-Pedro, A. J. Arnal, J. Post, *J. Cleaner Prod.* **2022**, *330*.
[59] L. Stougie, G. Del Santo, G. Innocenti, E. Goosen, D. Vermaas, H. van der Kooi, L. Lombardi, *Energy* **2019**, *182*, 535.
[60] ISO, *14044:2006. Environmental management – Life cycle assessment – Requirements and guidelines*, Switzerland, **2006**.
[61] A. Bjørn, M. Owsianiak, A. Laurent, S. I. Olsen, A. Corona, M. Z. Hauschild in *Life Cycle Assessment. Theory and Practice* (Eds.: M. Z. Hauschild, R. K. Rosenbaum, S. I. Olsen), Springer Nature, Switzerland, **2018**, pp. 75–116.
[62] F. Broch, *Integration von ökologischen Lebenswegbewertungen in Fahrzeugentwicklungsprozesse*, Springer Fachmedien Wiesbaden, Wiesbaden, **2017**.
[63] N. Chebaeva, M. Lettner, J. Wenger, J.-P. Schöggl, F. Hesser, D. Holzer, T. Stern, *J. Cleaner Prod.* **2021**, *281*, 125232.
[64] C. Fernandez-Dacosta, P. N. Wassenaar, I. Dencic, M. C. Zijp, A. Morao, E. H. Heugens, L. Shen, *J. Cleaner Prod.* **2019**, *230*, 137.
[65] F. Hesser, *Composites Part B* **2015**, *79*, 197.
[66] N. Thonemann, A. Schulte, D. Maga, *Sustainability* **2020**, *12*, 1192.
[67] A. Bjørn, A. Moltesen, A. Laurent, M. Owsianiak, A. Corona, M. Birkved, M. Z. Hauschild in *Life Cycle Assessment. Theory and Practice* (Eds.: M. Z. Hauschild, R. K. Rosenbaum, S. I. Olsen), Springer Nature, Switzerland, **2018**, pp. 117–165.
[68] EC, *ILCD Handbook. Framework and requirements for Life Cycle Impact Assessment*, Luxembourg, **2010**.
[69] C. Mair-Bauernfeind, M. Zimek, M. Lettner, F. Hesser, R. J. Baumgartner, T. Stern, *Int. J. Life Cycle Assess.* **2020**, 1.
[70] T. Hottle, C. Caffrey, J. McDonald, R. Dodder, *Transp. Res. D Transp. Environ.* **2017**, *56*, 241.
[71] D. Klein, C. Wolf, C. Schulz, G. Weber-Blaschke, *Int. J. Life Cycle Assess.* **2015**, *20*, 556.
[72] a) M. Z. Hauschild, M. Goedkoop, J. Guinée, R. Heijungs, M. Huijbregts, O. Jolliet, M. Margni, A. de Schryver, S. Humbert, A. Laurent, S. Serenella, R. Pant, *Int. J. Life Cycle Assess.* **2013**, *18*, 683; b) R. K. Rosenbaum, M. Z. Hauschild, A.-M. Boulay, P. Fantke, A. Laurent, M. Núñez, M. Vieira in *Life Cycle Assessment. Theory and Practice* (Eds.: M. Z. Hauschild, R. K. Rosenbaum, S. I. Olsen), Springer Nature, Switzerland, **2018**, pp. 167–270.
[73] L. Rigamonti, M. Grosso, M. C. Sunseri, *Int. J. Life Cycle Assess.* **2009**, *14*, 411.
[74] N. E. Matthews, L. Stamford, P. Shapira, *Sustain. Prod. Consumption* **2019**, *20*, 58.
[75] R. K. Rosenbaum, S. Georgiadis, P. Fantke in *Life Cycle Assessment. Theory and Practice* (Eds.: M. Z. Hauschild, R. K. Rosenbaum, S. I. Olsen), Springer Nature, Switzerland, **2018**, pp. 271–322.
[76] M. A. J. Huijbregts, W. Gilijamse, A. M. J. Ragas, L. Reijnders, *Environ. Sci. Technol.* **2003**, *37*, 2600.
[77] A. E. Björklund, *Int. J. Life Cycle Assess.* **2002**, *7*.
[78] J. Noack, N. Roznyatovskaya, T. Herr, P. Fischer, *Angew. Chem. Int. Ed.* **2015**, *54*, 9775.
[79] Q. Wang, W. Liu, X. Yuan, H. Tang, Y. Tang, M. Wang, J. Zuo, Z. Song, J. Sun, *J. Cleaner Prod.* **2018**, *174*, 1262.
[80] M. Arbabzadeh, J. X. Johnson, G. A. Keoleian, *J. Energy Storage* **2017**, *12*, 11.
[81] P. Leung, A. A. Shah, L. Sanz, C. Flox, J. R. Morante, Q. Xu, M. R. Mohamed, C. Ponce de León, F. C. Walsh, *J. Power Sources* **2017**, *360*, 243.
[82] J. Luo, B. Hu, M. Hu, Y. Zhao, T. L. Liu, *ACS Energy Lett.* **2019**, *4*, 2220.
[83] W. Schlemmer, P. Nothdurft, A. Petzold, G. Riess, P. Frühwirt, M. Schmallegger, G. Gescheidt-Demner, R. Fischer, S. A. Freunberger, W. Kern, S. Spirk, *Angew. Chem. Int. Ed.* **2020**, *59*, 22943.
[84] C. Mair-Bauernfeind, M. Zimek, R. Asada, D. Bauernfeind, R. J. Baumgartner, T. Stern, *Int. J. Life Cycle Assess.* **2020**, *25*, 2027.
[85] A. C. Hetherington, A. L. Borrion, O. G. Griffiths, M. C. McManus, *Int. J. Life Cycle Assess.* **2014**, *19*, 130.
[86] F. Scrucca, C. Baldassarri, G. Baldinelli, E. Bonamente, S. Rinaldi, A. Rotili, M. Barbanera, *J. Cleaner Prod.* **2020**, *268*, 122304.
[87] M. Goedkoop, M. Oele, J. Leijting, T. Ponsioen, E. Meijer, *Introduction to LCA with SimaPro* **2016**.
[88] T. S. Mathis, N. Kurra, X. Wang, D. Pinto, P. Simon, Y. Gogotsi, *Adv. Energy Mater.* **2019**, *9*, 1902007.
[89] a) S. Muller, A. Beylot, O. Sydd, K. Doyle, J. Bodin, J. Villeneuve, *Int. J. Life Cycle Assess.* **2021**, *26*, 2436; b) L. Mancini, S. Sala, *Resour. Policy* **2018**,

57, 98; c) L. Mancini, N. A. Eslava, M. Traverso, F. Mathieux, *Resour. Policy* **2021**, *71*, 102015.
[90] Mancini, L., N. A. Eslava, M. Traverso, F. Mathieux, *Responsible and sustainable sourcing of battery raw materials. Insights from hotspot analysis, company disclosures and field research.* JRC Technical Report, Publications Office of the European Union, Luxenburg, **2020**.
[91] R. Deberdt, P. Le Billon, *Extr. Ind. Soc.* **2021**, *8*, 100935.
[92] UNEP, *Guidelines for Social Life Cycle Assessment of Products and Organizations 2020*, **2020**.
[93] M. J. Goedkoop, I. de Beer, R. Harmens, P. Saling, D. Morris, A. Florea, A. L. Hettinger, D. Indrane, D. Visser, A. Morao, E. Musoke-Flores, C. Alvarado, I. Rawat, U. Schenker, M. Head, M. Collotta, T. Andro, J.-F. Viot, A. Whatelet, *Handbook for Product Social Impact Assessment. Roundtable for Product Social Metrics*, Amersfoort, **2020**.

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