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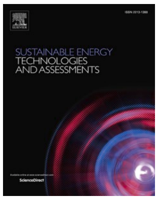

Original article

Life cycle assessment (LCA) for flow batteries: A review of methodological decisions

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ABSTRACT

A transition from fossil to renewable energy requires the development of sustainable electric energy storage systems capable to accommodate an increasing amount of energy, at larger power and for a longer time. Flow batteries are seen as one promising technology to face this challenge. As different innovations in this field of technology are still under development, reproducible, comparable and verifiable life cycle assessment studies are crucial to providing clear evidence on the sustainability of different flow battery systems. Based on a review of 20 relevant life cycle assessment studies for different flow battery systems, published between 1999 and 2021, this contribution explored relevant methodological choices regarding the sequence of phases defined in the ISO 14,040 series: goal and scope definition, inventory analysis, impact assessment and interpretation. Inspired by good practice examples, common gaps and weaknesses were identified and recommendations for comparative life cycle assessment studies were derived. This includes suggestions for an expanded functional unit definition, a provision of more detailed and transparent reporting of LCI data while using input/output tables. Outcomes of this study are also of relevance for the amendment of the Batteries Directive 2006/66/EC, where first drafts are under revision in the European Council, including the introduction of a battery passport, which should encourage battery producers to reduce their carbon footprint and avoid problematic materials.

Introduction

Flow batteries (FBs) are a versatile electric energy storage solution offering significant potential in the energy transition from fossil to renewable energy in order to reduce greenhouse gas emissions and to achieve sustainable development goals. The vanadium flow battery (VFB) is the most common installed FB. Other systems are for example the zinc-bromine, hydrogen-bromine and the all-iron FB [1]. Compared to the lithium-ion battery, the VFB is still at an early stage of development, but the system offers many advantages over conventional batteries. In particular, the long lifecycles, intrinsic heat management due to liquid nature of electrolyte and large tanks which avoid too high temperatures, non-flammability and easy scalability are in focus. The disadvantage of the battery is the current high price of the system [1]. The high cost results mainly from the electrolyte. However, in 2018, only 72 MW were installed worldwide, which corresponds to 0.00042 % at the current time [2]. FB, and VFB in particular, is increasingly coming into focus due to its unique characteristics. The largest VFB is currently being built in northeast China in Dalian. The battery will have a capacity of 200 MW power and 800 MWh energy [2,3].

FBs working principle is based on redox-active materials which are dissolved in liquid electrolytes and pumped through electrochemical cells, where electrochemical reactions take place. The amount of electrolyte stored in dedicated tanks and their volume defines the energy while the power depends on the cell’s characteristics and number, being assembled in stacks, as well as the electrolyte mass flow. This modularity

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makes FBs a flexible solution, which enables decoupling energy from power to identify the more suitable design for different applications. The fact that so far, they do not have high energy density, focus their application mainly to stationary applications. Nevertheless, the usage in ship propulsion is currently under investigation. The focus on stationary applications is also justified by some features of RFB, which are only possible due to the external storage medium. This allows capacity recovery routines, in which capacity can be recovered by chemical or electrochemical conversion. In addition, the use of inert electrodes, which do not participate in the electrochemical reaction, is a major advantage, when it comes to enhanced lifetime. Finally, they have a long service life, easily reaching up to 20,000 cycles with current commercial electrolytes, which means ten to twenty years of operation, depending on the typology of usage. The following Fig. 1 visualizes the scheme of a common FB system.

Different innovations in this promising field of technology are still under development, to reduce costs, increase electrolyte energy density, stability etc. whereby research focuses especially on different active materials, such as lithium, cobalt, vanadium, bromine or copper. In consequence, decision making within the research and development of FBs needs to be guided by quantitative approaches for the evaluation of sustainability, such as Life Cycle Assessment (LCA), as they analyze and assess environmental impacts and trade-offs across the entire life cycle. Several LCA studies for FB technologies were already published in the past. A closer look shows significant differences in carrying out LCA studies, especially with regard to the LCA framework according to the ISO 14,040 series [4]. Such differences are also reflected in the overall LCA results, which diverge and in many cases are not comparable. The urgency to define common LCA procedures and standards for FB technologies become more prominent, as the end of 2021 first drafts of the amendment of the Batteries Directive 2006/66/EC [5] are in revision in the European Council. This legislation regulates the use, sale and disposal of batteries and places emphasis on setting targets and ambitions for the recycling of batteries. One part of the legislation is the battery passport, which should encourage battery producers to reduce their carbon footprint and avoid conflict materials like cobalt. Each industrial battery above 2 kWh storage capacity should be equipped with such a passport, providing information for consumers on the environmental footprint of these batteries. At the moment, it seems FBs are not necessarily part of this legislation. Nevertheless, FBs should not shy away from competition when it comes to sustainability goals. Especially if this wants to be considered as a sustainable battery option. But common rules for assessment and transparent methodology is a prerequisite, when it comes to a fair comparison of different battery solutions, even after the end-of-life (EoL). Therefore, it is such an important task to set transparent and comparable methodologies to measure the environmental impacts of all storage technologies. Inspired by articles focusing on literature review of LCA studies for lithium-ion batteries [6,7,8] and good practice examples, such as the Product Environmental Footprint Category Rules (PEFCRs) for batteries with mobile applications [9], this review study identified common gaps and weaknesses (see *chapter 3*) in LCA studies for stationary FBs - especially for VFBs, as this battery type dominates the use by industry - and derived general recommendations for comparative LCA studies of batteries with stationary applications (see *chapter 4*). Further detailed information can be found in the attached Electronic supplementary material (ESM).

Materials and methods

LCA analyses a product's potential environmental impacts throughout a life cycle from cradle to grave. The lifecycle includes the stages: raw material acquisition, production of (sub-)components, transportation and use, as well as EoL treatment with recycling and final disposal. The methodological framework of an LCA study according to

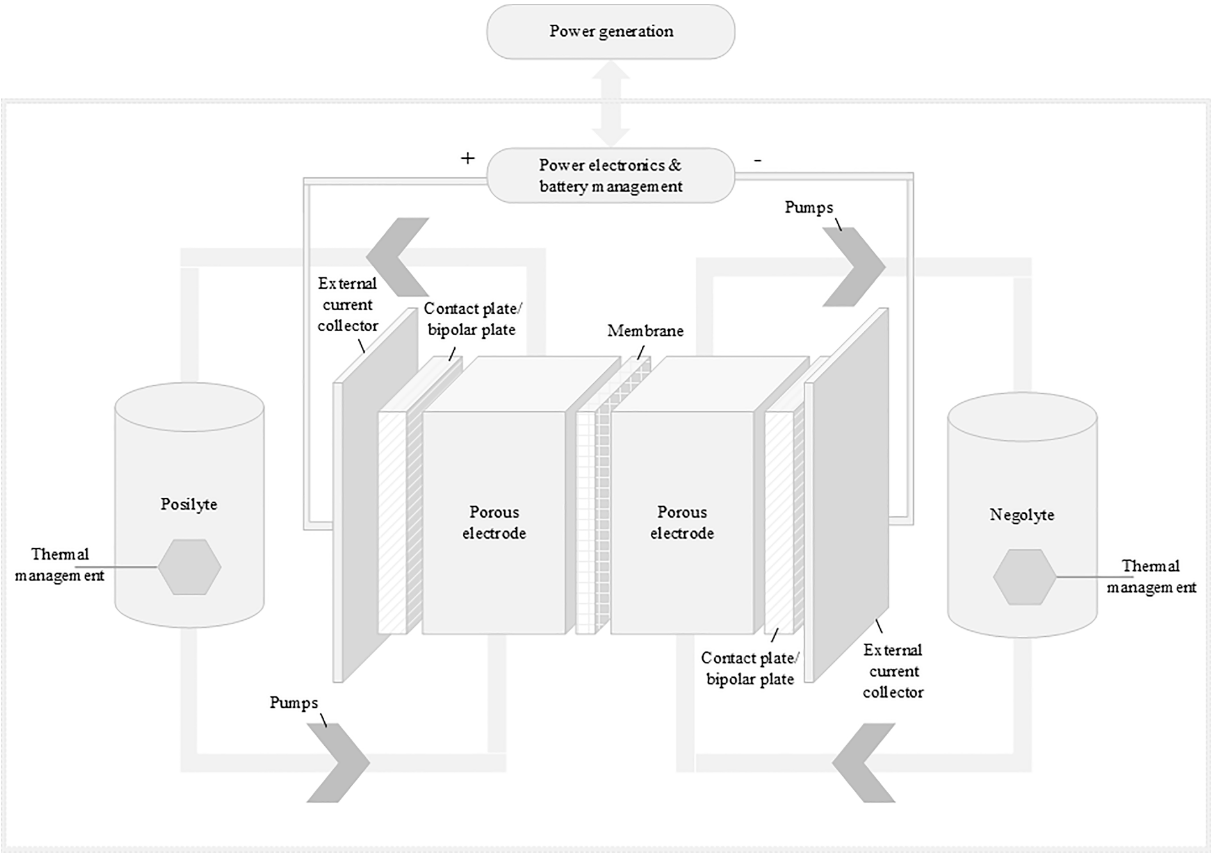

Fig. 1. Scheme of a flow battery system.

ISO 14,040 and ISO 14,044 includes four phases: i) goal and scope definition; ii) inventory analysis; iii) impact assessment; iv) interpretation. The Product Environmental Footprint (PEF) standard, driven by [10] and updated in 2019 [11], builds on the ISO 14,040 framework [4] and provides specific rules on how the assessment of products environmental footprint should take place. The aim of the PEF standard is to enable more reproducible, comparable and verifiable LCA results within different product categories, as a critical aspect of LCA studies is transparency on methodological choices and data sources. Specific PEFCRs exist also for High Specific Energy Rechargeable Batteries for Mobile Applications [9], and serve as good practice example for this study.

This contribution focuses on LCA studies for FBs and relevant methodological choices regarding goal and scope definition, inventory analysis, impact assessment and interpretation. Different databases, such as Web of Science and SCOPUS, were used to identify the relevant literature. The search was based on variations of the following keywords: “life cycle assessment OR life cycle analysis OR LCA” AND “flow batteries”. Studies published between 1999 and 2021 were reviewed. In total, 20 LCA studies were identified to be suitable for this review paper (see table 1). Further detailed information can be found in the attached ESM. The following *chapter 3* includes the results and discussion of our review study according to the four phases of LCA. Key recommendations were derived and supported by good practice examples in *chapter 4*.

Results and discussion

Based on a review of several LCA studies for different FB systems, published between 1999 and 2021 (see following table 1), this chapter explores and discusses relevant methodological choices according to ISO 14,040 series regarding goal and scope definition (*chapter 3.1*), inventory analysis (*chapter 3.2*), impact assessment (*chapter 3.3*) and

Table 1
Reviewed LCA studies for different FB systems.

Study	Year	Title	Author(s)	Product system (from to*)		Technology
1	**2021**	**Life cycle assessment of lithium-ion batteries and vanadium redox flow batteries-based renewable energy storage systems**	**Da Silva Lima L., Quartier M., Buchmayr A., Sanjuan-Delmás D., Laget H., Corbisier D., Mertens J., Dewulf J.**	**Cradle**	**Cradle**	**VFB**
2	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	Morales-Mora M.A., Pijpers J.J.H., Antonio A.C., de la Cruz Soto J., Calderón A.M.A.	Cradle	Gate & Grave	Bipolar ElectroDialysis Flow Battery (BEDFB)
3	2020	Life Cycle Assessment of Classic and Innovative Batteries for Solar Home Systems in Europe	Rossi F., Parisi M.L., Greven L., Basosi R., Sinicropi A.	Cradle	Grave	VFB
4	2020	Battery Manufacturing Resource Assessment to Minimise Component Production Environmental Impacts	Díaz-Ramírez M.C., Ferreira V.J., García-Armingol T., López-Sabirón A.M., Ferreira G.	Cradle	Gate	VFB
5	2020	Environmental and Preliminary Cost Assessments of Redox Flow Batteries for Renewable Energy Storage	Fernandez-Marchante C.M., Millán M., Medina-Santos J.I., Lobato J.	Cradle	Gate	VFB, Zinc / Cerium Battery (ZCB)
6	2020	Flow battery production: materials selection and environmental impact	He H., Tian S., Tarroja B., Ogunseitan O. A., Samuelsen S., Schoenung J.M.	Cradle	Gate	VFB, Zinc-Bromine Flow Battery (ZBFB), all-Iron Flow Battery (IFB)
7	2020	Life cycle assessment of a vanadium flow battery	Gouveia J., Mendes A., Monteiro R., Mata T.M., Caetano N.S., Martins A.A.	Cradle	Gate	VFB
8	2020	Life cycle assessment of a renewable energy generation system with a vanadium redox flow battery in a NZEB household	Gouveia J.R., Silva E., Mata T.M., Mendes A., Caetano N.S., Martins A.A.	Cradle	Grave	VFB
9	2020	How do non-carbon priorities affect zero-carbon electricity systems? A case study of freshwater consumption and cost for Senate Bill 100 compliance in California	Tarroja B., Peer R.A.M., Sanders K.T., Grubert E.	–	–	VFB
10	2018	A General Model for Estimating Emissions from Integrated Power Generation and Energy Storage. Case Study: Integration of Solar Photovoltaic Power and Wind Power with Batteries	Miller I., Gençer E., O’Sullivan F.M.	Cradle	Gate + operation	VFB
11	2018	Small-size vanadium redox flow batteries (in Life Cycle Assessment of Energy Systems and Sustainable Energy Technologies)	L’Abbate P., Dassisti M., Olabi A.G.	Cradle	Grave	VFB
12	2018	Life Cycle Assessment of a Vanadium Redox Flow Battery	Weber S., Peters J.F., Baumann M., Weil M.	Cradle	Cradle	VFB
13	2020	Assessing the Climate Change Mitigation Potential of Stationary Energy Storage for Electricity Grid Services	Jones C., Gilbert P., Stamford L.	Cradle	Grave	VFB
14	2017	CO2 Footprint and Life-Cycle Costs of Electrochemical Energy Storage for Stationary Grid Applications	Baumann, M. Peters J.F., Weil M., Grunwald A.	Cradle	Gate + operation	VFB
15	2016	Sustainability of vanadium redox-flow batteries: Benchmarking electrolyte synthesis procedures	Dassisti M., Cozzolino G., Chimienti M., Rizzuti A., Mastrorilli P., L’Abbate P.	Cradle	Gate	VFB
16	2016	Recycling of Battery Technologies – Ecological Impact Analysis Using Life Cycle Assessment (LCA)	Unterreiner L., Jülch V., Reith S.	Cradle	Cradle	VFB
17	2015	Vanadium redox flow batteries to reach greenhouse gas emissions targets in an off-grid configuration	Arbabzadeh M., Johnson J.X., De Leine R., Keoleian G.A.	Cradle	Grave	VFB
18	2015	Comparative Life Cycle Assessment of Battery Storage Systems for Stationary Applications	Hiremath M., Derendorf K., Vogt T.	Cradle	Gate + operation	VFB
19	2004	Life cycle energy requirements and greenhouse gas emissions from large scale energy storage systems	Denholm P., Kulcinski G.L.	Cradle	Grave	VFB
20	1999	Environmental assessment of vanadium redox and lead-acid batteries for stationary energy storage	Rydh C.J.	Cradle	Gate + operation	VFB

interpretation (*chapter 3.4*).

Goal and scope definition in reviewed studies

Three main items of the goal and scope definition for LCA studies of FB systems were explored in this review: (i) product system, (ii) functional unit and (iii) allocations.

Product system definition

The literature review reveals for the product system definition (including its system boundaries) of different FB systems, that there is no consistent procedure (see Table 1). Some of the LCA studies address a cradle to grave/cradle assessment – including extraction of raw materials, parts manufacturing, transportation and use stage, as well as EoL [12–19]. Other LCA studies focus on cradle to gate (see, for example [20,21]) or cradle to operation (see, for example [22]). Analyzing only selected life-cycle stages, such as cradle to gate assessment, leaves room for trade-offs and should be noticed when drawing conclusions.

Functional unit definition & assumptions on battery operation & management

The functional unit (fU) is in most of the examined studies defined consistent and focuses on the provision of 1 kWh [20,21,23,24]; or 1 MWh [14,18] of energy (see also *ESM,* Table 2). Nevertheless, the definitions answer only two out of four of the methodological questions (“what does it do?” and “how much?”) in order to define the fU consistently. The questions “how well?”, e.g. including information on power density, energy density, temperature window, frequency of load, total capacity and efficiency of the battery system, as well as “how long?”, e.g. including information on cycle life, calendar lifetime and operation time of the battery system, are only answered partially or not at all (further information see *ESM,* Table 2). Such incomplete definitions challenge the comparability of different LCA studies for FB systems.

Table 2
Comparison of expected lifetime, cycle life and efficiency in different LCA studies for FBs.

Study	FB technology	Expected lifetime (years)	Expected cycle life (amount)	Efficiency
1	VFB	20	6,000	83 % (round-trip efficiency)
2	BEDFB	20	–	75 % (round-trip efficiency)
3	VFB	20	–	75 % (round-trip efficiency)
4	VFB	20	–	–
5	VFB, ZCB	20 (VFB)	57 (ZCB)	80 % (VFB); 62 % (ZCB)
6	VFB, ZBB, IFB	–	–	–
7	VFB	–	–	up to 80 %
8	VFB	20	–	–
9	VFB	20	2,045	–
10	VFB	20	13,000	75 % (round-trip efficiency)
11	VFB	20	–	–
12	VFB	20	10,000	75 % (charge–discharge efficiency)
13	VFB	30	–	from 42 % to 77 % (round-trip efficiency)
14	VFB	15 (average)	10,000 (average)	average 75 % (DC-DC efficiency)
15	VFB	–	–	from 77 % to 96 % (cell efficiency)
16	VFB	20	> 10,000	90 %
17	VFB	20	–	65 %, 75 %, 90 % (round-trip efficiency)
18	VFB	20	13,000 (average)	75 % (round-trip efficiency)
19	VFB	20	–	75 % (AC-AC efficiency)
20	VFB	20	7,300	from 72 % to 88 %

Multi-Output and end-of-life allocations

Five of the examined LCA studies on FB provided further information on the issue of multi-output and EoL allocations. In order to solve the problem with by-products in different manufacturing processes, mainly economic allocation was applied [14,18,24]. Within EoL, two of the reviewed LCA studies mentioned that they focused on the avoided burden approach in order to consider environmental credits for the recovery of secondary materials and energy [19,21]. In most of the reviewed studies, it was unclear if allocation for multi-output and/or EoL was applied, and if yes, how it occurred (see, for example [16,20]).

Inventory analysis in reviewed studies

In the following subsection (3.2.1 up to 3.2.6), gaps in the literature are named, which have the greatest influence on the result of an LCA. However, the listed points are rarely explained in detail. The listed points must be presented transparently for a valid statement in an LCA. Weaknesses in reproducibility, comparability and verifiability of applied data and assumptions in life cycle inventory (LCI) were especially identified in the examined LCA studies for (i) electrolyte production, (ii) membrane composition, (iii) transportation, (iv) EoL scenarios and (v) upscaling. Moreover, only a few LCA studies reviewed provide further information on the data collection in terms of bill of materials (BoM) (further information see *ESM,* Table 3).

Vanadium electrolyte production

Among FBs the commercial ones are so far using electrolytes vanadium based. The large majority of the reviewed papers is related in fact to VFB, except one focused on Bipolar Electro Dialysis Flow Batteries (BEDFB) [19] where anyhow results are compared against VFB and two more where in addition vanadium-based also Zinc/Cerium Batteries (ZCB) [20], and Zinc Bromine Flow Batteries (ZBFB) and all-Iron Flow

Table 3
Suggested LCI documentation according to Weber et al. [14].

Flow	Provider	Amount	Unit
Inputs			
Tetrafluoroethylene ($C2F4$)	Market for tetrafluoroethylene - GLO	1.30	kg
Sulfur trioxide (SO3)	Market for sulfur trioxide - GLO	0.50	kg
Hexafluoropropene (C3F6)	Market for hexafluoroethane - GLO	3.20	kg
Sodium hypochlorite (NaOCl)	Market for sodium hypochlorite - GLO	3.00	kg
Sodium hydroxide (NaOH)	Market for sodium hydroxide - GLO	0.60	kg
Sodium carbonate (Na2CO3)	Market for soda ash, dense - GLO	0.11	kg
Infrastructure	Chemical factory, organics - GLO	4.00E-10	Item (s)
Process heat	Market for heat, nat. gas, industrial - EU w/o CH	39.31	MJ
Transport lorry	Market for transport, freight, lorry unspec. - GLO	0.87	t*km
Transport train	Market for transport, freight, train - EU w/o CH	5.22	t*km
Outputs			
Membrane, Nafion®	Membrane, Nafion®, for VRFB	1.00	kg
NaCl	Sodium chloride, to water	2.36	kg
NaOH, aqueous solution	Sodium hydroxide, to water	1.87	kg
NaF	Sodium fluoride	8.48E-02	kg
CO2	Carbon dioxide, fossil, to air	8.88E-02	kg
Organic residue	Treatment of spent solvent mixture	2.81	kg
Plastic residue	Treatment of waste plastic, mixture	0.11	kg
Oily residue	Treatment of bilge oil	0.38	kg

Battery (IFB) [24] are assessed.

The production of the vanadium-based electrolyte is a key issue as vanadium is a competitive ingredient in other industrial products. Vanadium titano-magnetite (VTM), which contains between 0.2 and 2 % of vanadium pentoxide (V_2O_5) [25], is the most used mineral – 88% of the V production is from titano-magnetite according to Volkov, et al. [26], with China (52%), South Africa (26%) and Russia (19%) being the main producers [27,28]. A summary of the main stages for the production of V_2O_5 from VTM is proposed based on the detailed description of the series of complex processes, which can be found in [25,29,30,31]. However, the exact flowsheet depends very much upon the plant and the characteristics of the ore.

Several routes have been proposed to recover vanadium from spent catalysts: these catalysts are considered as hazardous waste and there is therefore a double interest in their recycling [32]. However, often these catalysts contain several metals of interest: their treatment process is complex and again there is no single route to treat them. A series of steps have been proposed by Zhang et al. [33] to get nickel, molybdenum and vanadium out of hydroprocessing catalysts.

Extraction of vanadium from fly ash from crude-oil fired boilers has also been proposed in the literature, showing a double interest: reduction of the environmental impact of this waste and production of a metal of interest. No detailed inventory has been given by Rydh [34] in his comparison of VFB and lead-acid battery. Leaching of oil fly ash with sodium hydroxide followed by a precipitation step for purification has been proposed by Navarro et al. [35]. As fly ash is a waste, the burden associated with its production is not considered. This is technically correct. But, the production of fly ash from fossil resources is accompanied by large emissions of greenhouse gases during combustion. On an ethical side, this poses an allocation problem, which needs to be discussed, and furthermore on an economic side, oil usage is constantly decreasing with the increasing degree of decarbonization of the economies.

So far, the most detailed inventory of vanadium pentoxide production, based on the VTM-vanadium slag flowsheet of a plant in South Africa, has been given by Weber et al. [14]. However, the composition of the electrolyte depends on various parameters, for example, the ambient temperature. The composition of the standard vanadium electrolyte refers to the electrolyte from Gesellschaft für Elektrometallurgie mbH (GfE) [36]. For this, 1.6 mol L^{-1} vanadium is used, which corresponds to a requirement of 0.148 kg L^{-1} of vanadium pentoxide. Likewise, the amount of electrolyte depends on many parameters. At a vanadium concentration of 1.6 mol L^{-1}, 50 m^3 of electrolyte can be calculated as a guide value for 1 MWh [37]. However, with more conservative approaches, significantly higher values for the amount of the electrolyte can be expected due to the round-trip efficiency. In this context, it is to mention that the amount of electrolyte (202 m^3) given in Weber et al. [14] refers to only half of the total amount needed (404 m^3), as only one side of the battery is considered in the mass balance, see Table S1 [14].

Membrane composition

Various types of membranes have been tested and they are mostly prepared from well-known polymers, which should facilitate the inventory [38]. Nafion® cation exchange membranes exhibit a long-term chemical stability and are presently the most preferred membranes [39], in spite of their high cost. Nafion® is a proprietary sulfonated fluoropolymer largely used for its high ionic conductivity properties in fuel cells and VFBs. Polymerization in the production of Nafion is a very complex process, which is often only very simplified and this is based on an incomplete input and energy balance. Without transparent balancing, a superficial view must be assumed. An inventory is proposed by Weber et al. [14] according to Minke and Turek [40] and is based on the DuPont patent.

Transportation

In most reviewed studies, transportation is not explicitly mentioned. However, it must be taken into account that few of the papers even mention the origin of the vanadium pentoxide (China, South Africa and Russia being the main producers from ore [27,28]) or the essential intermediate for the electrolyte. The exception is the work of Weber et al. [14] who describes the location and processing of the vanadium pentoxide. In da Silva Lima et al. [18] the transport is presented separately in the emissions, but the emissions here are based on the assumption of transport of the finished battery from China to Belgium. However, the vanadium pentoxide for electrolytes comes from China, accordingly, the transport distances are very small in this case. In Gouveia et al. [41] the emissions caused by transport dominate, but the influence of the transport routes cannot be understood without absolute numbers or transparent data. Due to the low emissions of the electrolyte, this may also be the reason for the high percentages of transport.

Transport between the different sites should be carefully documented as it affects several impacts such as greenhouse gas (GHG) emissions, acidification, or land use. In particular, the mode of transportation for long distances should be realistic. Sea freighter is the most appealing method for transport of V_2O_5 between South Africa and Europe [14]. Road is also possible (with a payload-distance of 12.534 tkm instead of 13.332 tkm by sea) but leads to an increase of impacts, in particular land use and ozone depletion. There are several main options (rail, sea, lorry and their combination) between China (or South Korea as in da Silva Lima, et al. [18]) and Europe. The best option remains the sea route across the Suez Canal. Often the question of long-distance transport is not really mentioned. Rydh [34] does not specify the location of vanadium production but vanadium recovery plants are said to be operating in Germany and in Japan, which gives a lot of uncertainty on the distances. Some authors [12,21,42] refer to inventories produced by others, mostly [14,34] while others [16,43] just state the origin of V_2O_5 (South Africa). The question is more subjective for local transport (for example between or within European countries) as it could be very diverse: often the details are not given. He, et al. [24] removed on purpose all distances from their inventories. Regardless of the transportation distances for raw material acquisition of vanadium electrolyte, Morales-Mora, et al. [19] discusses transportation impacts for imported components from Mexico, the United States, or the European Union.

Batteries integration and performance characterisation

In the examined studies, batteries are eventually assumed to be integrated with a specific energy system, which had an impact first on the battery characteristics selection, for the cradle-to-gate studies, secondly on battery operation, whether its effect is included in the LCA. This goes together with the battery performance characterization, whose accuracy will highly affect it, especially considering that operation could have a much more significant impact than production whether the battery is not fed by renewables only [14,44]. In fact, flow batteries could be more competitive than other solutions such as lithium-ion only in the case of renewable energy sources predominant in the energy mix, given their lower round-trip efficiency and having as a point of strength the FBs low impact in the cradle to gate phase and easiness to recycle materials.

Overall, the approaches are quite homogeneous. Exceptions are, on the one hand, a couple of papers more focused on the electrolyte synthesis, with 6 L of it adopted as the functional unit and performing the study at single-cell level, thus reaching up to 96 % round-trip efficiency [42] and 85 % with five cells stack [43]. On the other hand, a couple of papers are far more detailed than the average concerning the different applications [43,44], e.g. distinguishing among self-consumption, arbitrage or frequency regulation, thus on how they affect energy and power size, frequency of cycles per day and Depth of Discharge (DoD). Here is an overview of the key features characterizing RFB performance:

- The large majority of adopted functional units are represented by one unit of stored electric energy, either [kWh] or [MWh], except for these focusing on the electrolyte, where a certain amount of it is taken as a functional unit. In one case, it was also mentioned the power rating in addition to the stored energy and an average value through the battery life [34].

- Regarding the round-trip efficiency, sometimes mentioned as overall efficiency, its value is specified in three-quarters of the cases and consequently used in the assessment of battery operation impact in the LCA. Anyhow, in the majority of cases, it is not systematically defined, e.g., whether AC or DC is considered and whether the energy for battery cooling is considered. It is never considered the efficiency of charge and discharges separately as well as its dependency on the state of charge and continuous performance degradation as a function of usage. The latter would be fundamental to consider the impact of the operation strategy on battery performance degradation; sometimes, it is included by means of an average lifetime value consistently with usage assumptions [34]. Values are in the majority of cases assumed to be between 70 and 80 %, with some exceptions reaching the peak of 90 % [12,13] upper boundary for the latter, and above 90 % is reached in the case of cell-only performance [43]. For further information see following Table 2.
- The Depth of Discharge (DoD) is often ranging between 80 and 90 %. In rare cases, it is considered equal to 100 %, and often the minimum and maximal value of SoC (State of Charge) to whom it corresponds are not specified, e.g. with minimum set equal to 5 or 10 %. Clear definition of the minimum and maximum SoC will allow to determine the usable capacity of the battery.
- The energy to power ratio is quite diverse, in some cases even below 1 for frequency and power regulation, going up to 8 or 10 as a maximum in case of usage more consistent with the RFB field of application. In one domestic application (see Table 2, study no. 7) also the case of energy to power ratio ranging from 3.6 up to 72 has been taken into consideration.
- The life of the battery is often set at 20 years (see following Table 2), only in some cases it is calculated on the number of cycles and therefore related to the battery usage [23] and [44]. Increased renewables self-consumption and arbitrage relate usage range from 0.6 to 2 cycles per day while frequency regulation scenario reaches 34 cycles per day, but with an average DoD of only 5 %. The lifetime of the stack a few times has been considered and set equal to 10 years with the implications in terms of maintenance and economic and environmental impact.

Looking at other technical specifications such as voltage and current density, cells area and number, stacks number or even electrolyte concentration and density they are rarely present, only the energy density is reported in slightly more than half of the cases, and it ranges from about 20 – 30 Wh per kg (7.29 Wh/kg for the BEDFB), sometimes is also reported in Wh per unit of volume, and in one case [34] also the value for two different electrolyte chemistries and the temperature at which the density is measured is specified.

End-of-life scenarios

Original EoL scenarios, focussing on cradle to grave/cradle assessment, are considered in only five studies. All authors report limited data availability and thus simplified EoL approaches. In general, 20 years of calendar lifetime are assumed. In an earlier study, the EoL scenario is the disposal and no recycling content in FB systems is considered [12]. Unterrainer, et al. [13] considers only a share of around 18% of reusable materials in VFB.

In more recent studies, for the electrolyte, especially for high cost and highly durable vanadium electrolytes, a reuse rate of 50–100 % of the solution is assumed via electrochemical rebalancing performed after 20 years [14,18]. For stack and system components, a mechanical dismantling and fractioning of metal and –non-metal components are proposed. Metals go to state-of-the-art recycling processes with an efficiency of up to 95 %, whilst other materials go to state-of-the-art disposal and mainly incineration processes [14,18,19].

Upscaling

LCA is often used to compare different systems which should have the same functions (see also chapter 3.1.2). However, the data available do not always refer to the same specifications of the batteries (further detailed information see also *ESM,* Table 2). A scale-up or down is then necessary. As an example, Díaz-Ramírez et al. [21] have proposed a simplified equation for upscaling, taking into account the lifetime, the specific energy and the number of cycles for any element of the inventory (energy or mass). However, the equation has been applied to a limited number of elements of the inventory and assumes proportionalities with respect to size and time. Possible scaling non-linearities should be assessed.

Impact assessment

The reviewed papers have selected a variety of different impact indicators. Similarly, the choice of midpoint and endpoint levels for the Life Cycle Impact Assessment (LCIA) is not clearly defined and differs in the literature (further detailed information see *ESM,* Table 3). Each of the examined LCA studies assesses the environmental impact on climate change, except for [48] who focuses only on freshwater consumption. Usually, different impact indicators are not normalized and weighted. Exceptions are the comparisons by [13,16,42] where different impact categories are calculated to single scores.

Interpretation

Da Silva Lima et al. [18] conclude that the production, transport and electricity for use of VFB energy storage systems are the main drivers of the total balance. LCA results by Weber et al. [14] indicate the same, as manufacturing (including transportation) and use phase dominate the total balance within different impact indicators. Morales-Mora et al. [19] conclude that manufacturing or use phase dominate the total balance, depending on the energy mix when charging/discharging the battery system. Consensus is that the EoL phase does not result in substantial impacts.

The identification of hot spots and potentials for further improvement of FB systems depends very much on the way how LCA results are documented, visualized and interpreted (see, for example [45]).

Key recommendations & good practice examples

The following set of recommendations and good practice examples aims to enable more reproducible, comparable and verifiable LCA studies for flow batteries methodologically inspired by the PEF standards for batteries with mobile applications [9].

Goal and scope definition

- Assess the whole life cycle of the battery systems from cradle to grave/cradle, including: raw material acquisition, production of (sub-)components, transport to customer, assembly of batteries with the cells and the electric/electronic components, use stage (storage inefficiency), transport to recycling, EoL (including impacts for recycling and credits for recovery of secondary materials and energy); in order to avoid trade-offs and to ensure a net reduction of the total environmental impacts (positive contribution towards sustainability; as exemplified in Fig. 2).

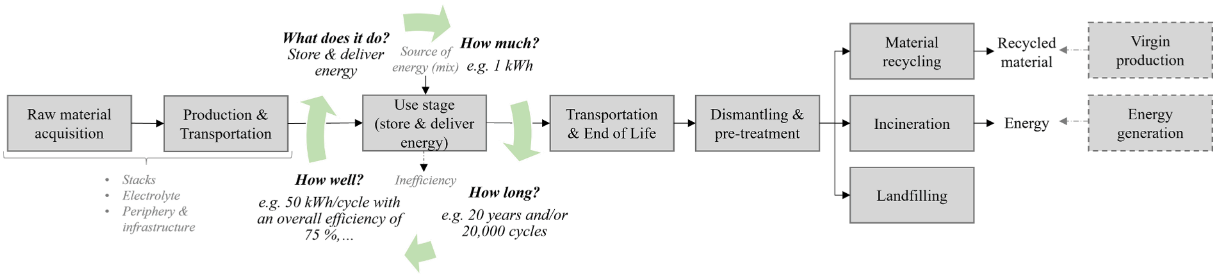

Fig. 2. Suggested product system and functional unit definition for LCA of Flow Batteries.

- Extend the definition of the function (service) of a FB system by answering the four methodological questions of the fU as precise as necessary and as open as possible. This ensures a meaningful comparison - of "apples to apples" - for different stationary storage technologies of the present and the future and represents the pros and cons of different systems in an appropriate manner.
  - *What does it do?* store energy;
  - *How much?* 1 kWh[1];
  - *How well?* include relevant technical information on the specific characteristics and properties of the flow battery system, e.g. on power density, energy density, temperature window, round-trip efficiency, frequency of load and the usable capacity;
  - *How long?* include specific information on the cycle life, calendar lifetime and the estimated operation time of the flow battery system.

Life Cycle Inventory

- Provide more detailed and transparent reporting of LCI data within LCA studies for FB systems while using input/output tables with detailed information on the specific flow, amount, unit as well as the primary or secondary source and the name of the data set (provider). Tables provided within the supporting information of Weber et al. [14] and the PEF standards for batteries with mobile applications [9] serve here as a good practice example for transparent reporting of LCI data for FB systems. Further specific recommendations for the different life cycle stages of a FB system are as follows:
  - *Raw material acquisition:* Data gaps in LCI were especially identified in the production of the electrolyte. So far, the most detailed inventory of vanadium pentoxide production has been given by Weber et al. [14]. However, the allocation of emissions (based on the market prices of steel and V_2O_5) need to be reviewed and updated, if necessary. For zinc-bromine ($ZnBr_2$) FBs a simple material balance based on stoichiometry, without energy consideration, has been given for example by He, et al. [24].
  - *Transportation:* Transport of vanadium-based electrolyte between sites should be carefully documented. In particular, the mode of transportation for long distances should be realistic. It could be useful to run a sensitivity analysis to check the effect on the LCA results.
  - *Production of (sub-)components of FB systems:* Detailed LCI information on the production route of Nafion®, as the most preferred membrane material presently, were published by Weber et al. [14], according to Minke and Turek [40], and serve as a good practice example in terms of providing detailed and transparent LCI data on newly developed material systems.
  - *Battery integration & use stage:* Energy losses due to charging and discharging according to the round-trip efficiency over lifetime of the battery system (information should be included in the fU definition; see chapter 4.1) have to be taken into account while considering different sources of energy (especially important for the sensitivity analysis of the battery system; see, for example [46]). Batteries integration has a significant impact on the LCA; thus the scope of the battery installation and the way it will be utilized will affect both the sizing, thus the inventory (cradle-to-gate), and the energy mix feeding it as well as the way the battery is going to operate, so the number of charge–discharge cycles per day and their depth of discharge. These affect the operation and the maintenance and life of components. For a proper performance characterization, it should be clearly identified, first of all, whether the electric energy adopted as a functional unit, is that one taken from the grid, stored in the battery or released by the battery to the grid. These are three different values due to the presence of the charge and discharge efficiency. Furthermore, whether it is AC or DC electricity should be specified, implying the inverter efficiency, as well as all the auxiliaries such as electrolyte circulation pumps and cooling system needed to keep the flow battery at the required temperature. This information is necessary in case the study aims at investigating the operation impact, and they are not easy to find in the reviewed literature or at least clearly stated. In addition, the effect of self-discharge, the charge and discharge efficiency as a function of the SoC and power, and performance degradation as a function of the operation strategy, could be considered in a study focused on the operation impact per se. In addition, the impact of the characteristics of the RFB components and operating parameters on the LCA could be extensively assessed, e.g. as a function of the current density, cells voltage, bipolar plates conductivity, vanadium concentration in the electrolyte etc. So all the parameters identified in *ESM,* Table 2 according to the authors could improve the clarity and reproducibility of the LCA, and its dependency on these design and operational parameters could be performed, as per techno-economic best practice [47].
  - *EoL and recycling:* All the processes for collection, dismantling and reuse or recycling of components of the FB system as well as the credited flows (based on the quantity AND (*) quality for the substitution of primary materials and energy) should be described separately and in detail (see for example PEF standards for batteries with mobile applications, table 26: End of Life). This includes especially the processes to prepare the electrolyte for reuse, as it can significantly affect the total balance of the battery system (see, for example [13,39]).

Life cycle impact assessment

- Consider a variety of environmental impact categories within LCIA as it is recommended in the general PEF standard [11] (up to 16 different indicators), including: climate change, ozone depletion,

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[1] Final unit for comparison of the environmental impacts. The "total life cycle environmental impacts" of the flow battery system are divided by the "total delivered energy during service life" of the flow battery systems (= total number of cycles over the estimated life cycle * average capacity (kWh/cycle) * round-trip efficiency (%) = kWh/life cycle).

acidification, eutrophication, human toxicity, eco toxicity, land use, water use and resource use.

- Although standardized methods for normalization and weighting have been proposed by the PEF standard [11] they should not be used in accordance with ISO 14,040 series [4], which explicitly prohibits the use of single-score indicators for “all comparative assertions intended for public disclosure”. In addition, the results for each environmental impact category should be documented transparently while ensuring that an increase in one environmental impact category (e.g. climate change) cannot be compensated by a decrease in another category (e.g. eutrophication). If normalization and weighting is to be used for a sensitivity analysis, it should be validated with panel experts and industry partners beforehand. The justification and calculation must be discussed and documented transparently.

Interpretation and identification of potentials for further improvement

- The development of an LCA for FB is very complex and as shown the result can differ significantly due to wrong assumptions. The most sensitive parameters have been discussed in detail in this paper and are based on the analysis of the presented LCAs. The assumptions concern not only aspects of LCA but also technical assumptions of the battery. Accordingly, the goal is to minimize the potential errors in the development of the LCA in the long term through transparency. This also includes a critical examination of the approach and the results in order to identify possible weaknesses. In particular the results within different impact categories should be documented complete, transparent, easily understandable and reproducible. This includes a separated documentation of the subtotal results for raw material acquisition, transportation, production of the subcomponents and final product, distribution to consumer and collection, use phase, as well as recycling and credits for the reuse of components or substitution of primary materials and/or energy (see, for example [13]). Special focus for the FB must be on the electrolyte and accordingly the balancing should determine a substantial part of the LCA, since in the analyzed works the emissions of this vary significantly but still the electrolyte determines the emissions significantly. Such transparent documentation and visualization of the results allows a more controversial discussion and fosters the possibility to identify potentials for further improvement in all life cycle stages, e.g. by applying the life cycle gap analysis (LCGA) - interpret products LCA results with a circular economy mindset [45].

Conclusion

Following a careful examination of 20 papers dealing mostly with VFB we can conclude that a careful observance of the rules prevailing in the ISO norms related to life cycle assessment is more and more necessary to discuss the sustainability of actual and future batteries, of any type.

The whole life cycle should be considered and not just the production cycle: replacement of parts and electrolyte and their recycle should be considered. The functional unit should be defined in terms of energy delivered and all relevant technical information should be provided such as power and energy density, usable capacity, calendar lifetime, etc. The inventory should be based on transparent and verifiable mass and energy balances for all the battery parts (electrolyte(s), membrane, stacks, etc.) with realistic transportation routes and modes. Replacement of parts including electrolyte due within the battery life duration as well as their recycle should be considered.

It is true that it is not always the case to have access to real-world data from full-scale setups and often a priori LCA - based mostly on lab-scale data - is a challenge. Sensitivity analysis is therefore a necessity. It is only with the respect of all the LCA rules that confidence will be gained toward the sustainability of any battery, actual or future. We recommend therefore further developments in terms of PEFCRs for batteries with stationary applications.

CRediT authorship contribution statement

Michael Dieterle: Supervision, Validation, Project administration, Writing – original draft. **Peter Fischer:** . **Marie-Noëlle Pons:** . **Nick Blume:** . **Christine Minke:** . **Aldo Bischi:** .

Declaration of Competing Interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.seta.2022.102457.

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