Redox Flow Batteries: Stationary Energy Storages with Potential

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DOI: 10.1002/cite.202100001

On the way to a secure, economic, and environmentally compatible future of energy supply, the share of renewable energies will rise strongly as part of the energy transition. To ensure a constant and resilient energy supply, despite the fluctuations of renewable energies, efficient energy storage systems are crucial. One of the most promising technologies are redox flow batteries. They are of particular importance in the field of stationary applications, due to their flexible and independent scalability of capacity and power output as well as their high cycle stability, calendric service life, and operational safety.

Keywords: Energy storage, Energy transition, Numbering-up, Redox flow batteries, Scale-up

Received: January 01, 2021; accepted: January 22, 2021

1 Introduction

Over 22 000 000 000 kWh (22 000 TWh) was the global electricity consumption in 2018 but only 26% have been produced using renewable energy sources, such as hydro, geothermal, tidal, wind or solar power [1, 2]. On the way to a secure, economic and environmentally compatible future of energy supply, the share of renewable energies will rise strongly in the coming decades as part of the energy transition. In order to ensure a constant and resilient energy supply, despite the often immanent fluctuations of renewable energy sources, the use of efficient energy storage systems is crucial. Those storage systems can smooth resulting power fluctuations, stabilize and relieve the electricity grids, increase the regenerative power plants degree of self-sufficiency and energy efficiency and function as isolated backup systems for the emergency power supply. In this process, the global demand for energy storage systems will increase more than fivefold by 2040 to an estimated amount of 942 GW [3].

In 2018, the energy storage systems installed worldwide already had a total power output of almost 173 GW, with the main load of nearly 170 GW being carried by pumped storage hydro (PSH) [4]. PSH plants have a very high system efficiency, but also a few relevant disadvantages: The elaborate construction results in an unchangeable stationing as well as fixed power output and storage capacity. In addition, the construction involves serious interference with nature, which often leads to low acceptance in the population. Finally, the usability of PSH plants depends on the local topography and is therefore not suitable for the universal use worldwide.

Besides PSH, electrochemical energy storage systems are mainly used as shifting and long-term storage systems. In 2018, more than 2 GW had been deployed worldwide [4]. Lithium-ion and sodium-sulfur batteries had the largest amount of installed power output, followed by lead-acid and redox flow batteries (Fig. 1).

However, most battery types and capacitors are only suitable to a limited extent for the stationary energy storage, as they are mainly internal energy storage devices. This means, power output and storage capacity are always in a fixed ratio to each other. This prevents an optimal design for the individual needs of the storage tasks as well as the possibility of subsequent extension of power or capacity. Furthermore, the commonly used secondary battery types have a relatively high self-discharge rate (lead-acid batteries at 20°C up to 30% per month, lithium-ion batteries at 20°C between 3 and 30% per month) and a comparatively low cycle stability (100 to several 1000 charge and discharge cycles) [5]. In addition, those batteries have a high sensitivity to deep discharge, which is associated with a high maintenance requirement, and, in particular, a limited operational safety: lead-acid batteries produce an explosive gas mixture, whereas overheating or damage of lithium-ion batteries can result in a thermal runaway, which may lead to fire or explosion [6].

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The most promising complementary energy storage systems are redox flow batteries. These external energy storage devices are of particular importance in the field of stationary storage, due to their flexible and independent scalability of capacity and power output as well as their high cycle stability (> 10,000 cycles) and operational safety (non-flammable, no explosion hazard) [7, 8].

In flow batteries, two different electrolytes, as storage medium, are stored in external tanks and only pumped through the battery cell for charging and discharging in two separate hydraulic circuits, for anolyte and catholyte (Fig. 2). When operating, oxidation and reduction processes take place at the anode and cathode, which convert the electrical energy into chemical energy during charging and back into electrical energy during discharging.

During charging, the ions of the two electrolytes are forced into higher or lower oxidation states, depending on the cell chemistry. In the standby state, when the fluid pumps are off-duty, the oppositely charged electrolyte solutions remain in the external tanks, completely locally separated from each other, resulting in a self-discharge close to zero. In this state, the only self-discharge takes place between the remaining electrolyte quantities hold-up in the battery cells. During discharging, due to the lack of a driving force, the ions of the redox active species, dissolved in the electrolytes, collapse back to their original oxidation state, causing a flow of electrons and thus the release of electrical energy.

As the battery electrodes do not actively participate in the chemical reactions, flow batteries are deep discharge proof. Further advantages are the possibility of targeted subsequent adjustment of power output and storage capacity, by changing the number or size of battery cells and changing the electrolyte quantity, as well as the long calendar operating life of more than 20 years, as theoretically no degradation of the functional components occurs.

The maturity and commercialization capability of redox flow batteries have been demonstrated in numerous R&D and large-scale projects (e.g., 200 MW/800 MWh all-vanadium redox flow battery of Rongke Power and UET) [9].

2 Structure of Redox Flow Battery Cells and Stacks

The independent scalability of capacity and performance is one of the biggest advantages of redox flow batteries – based on the local separation of the energy storage and energy conversion unit. While basic tanks serve to store electrolytes and therefore the chemical energy, the actual redox flow battery cell for the conversion of electrical energy into chemical energy and vice versa consists of two half-cells, one being the positive electrode (cathode) and the other the negative electrode (anode). The half-cells are hydraulically separated from each other by an ion selective membrane and are limited to the outside by fluid-tight conductive end electrode plates (Fig. 3). These end electrode plates allow an electrical current to flow between the half-cells via an external load when discharging or a power source when charging, while ionic current flow between the half-cells occurs via the membrane.

In the most commonly used flow-through cell design with a fluid distribution via internal manifolds, the reaction areas of the half-cells are completely filled with highly porous graphite felt electrodes. These serve as extensions of the end electrode plates to provide larger electrode surface areas for the oxidation and reduction processes during charging and discharging. Thus, the porous electrodes as well as the end electrode plates have a purely catalytic effect.
In the conventional flow cell structure (Fig. 3), the electrolytes are fed into the battery cell via fittings in the end plates. These end plates serve as electrical and hydraulic transfer points between the battery cell and peripherals and ensure the force-locking positioning of the cell components.

In order to generate a technically usable voltage, a larger number of individual cells are usually combined into cell stacks. To connect the cells of a stack electrically in series and to separate them hydraulically, bipolar plates are used as end electrodes of the individual cells (Fig. 4).

For the reactant supply, the electrolytes are fed via the fittings in the end plates into the internal manifolds, which run orthogonally to the stack base through the entire stack or into the corresponding half-cell at single cells. Starting from the manifolds, the electrolytes are further fed, through meander-shaped channels and a distribution geometry in the individual flow frames, into the active cell areas and thus flow through the porous electrodes (Fig. 5). The length and meander-shape of the electrolyte channels serve to suppress efficiency-reducing shunt currents, which occur because of the hydraulic parallel connection of flow cells in a stack with a fluid distribution via internal manifolds.

3 Classification of Redox Flow Batteries

There are different approaches to the classification of redox flow batteries due to their diversity. The subdivision can be made, for example, by cell design, the fluidic properties of the electrolytes or the type of electrolyte.

In the conventional design, the flow cell consists of two similar half-cells through which two liquid electrolytes containing the fully dissolved active species flow. To date, these active species are primarily inorganic and work as a redox couple with a standard potential within a stable voltage range, without oxygen or hydrogen formation. Examples are the most common used vanadium-vanadium flow battery or the iron-chromium flow battery. However, research followed different paths to make the redox flow battery more powerful, resource and cost efficient.

One way is the use of novel inorganic and organic electrolytes, adapted to the characteristics of the conventional flow cell design. Hybrid designs, on the other hand, adapt the cell design to the milieu of the active species.

In order to show commonalities of the different types of redox flow batteries, the main subdivision can be made according to the number of hydraulic circuits (Fig. 6).

4 Functional Components

4.1 End Electrode Plate/Bipolar Plate

As a key component of redox flow battery stacks, bipolar plates significantly contribute to volume, weight and cost of stacks. Therefore,
bipolar plates must be made of inexpensive and lightweight materials, which can be easily and inexpensively manufactured and which offer high electrical conductivity, mechanical stability as well as chemical resistance against corrosive electrolytes [18].

Several materials have been studied to date including pure graphite plates, coated and uncoated metallic plates and graphite-filled composite materials. Pure graphitic plates offer a good electrical conductivity and good chemical resistance but suffer from breakage during the machining step and leakages caused by high porosity. Metallic bipolar plates consists of alloyed or coated steel, to prevent corrosion in the acid environment of the electrolytes. This increases costs and decreases productivity of the fabricated materials.

Graphite filled polymers represent a compromise between chemical resistance, electrical conductivity, mechanical stability and cost-effective manufacturing. A high filling grade (> 80 wt %) combined with a variety of thermoset or thermoplastic binding polymers characterizes these composite materials. Small amounts (< 5 wt %) of varying additives improve the conductivity (carbon blacks, carbon-nanotubes, graphene) or the flowability of the composites.

Thermoset polymers undergo a chemical cross-linking reaction induced by heat, catalyst reaction, radiation crosslinking or crosslinking via light, forming an irreversible chemical bond without remelting properties. Established processing methods are the (hot) pressing or the infiltration of pure graphitic sheets with a liquid thermoset. Due to their physics, thermoset materials are not weldable or subsequently re-shapable. Possible flow field geometries, which are discussed at the end of this chapter, have to be implemented via milling.

Thermoplastics polymers soften by the action of heat and become more fluid as shear forces and additional heat are applied. In contrast to thermoset materials, no chemical bonding takes place, the curing process is completely reversible, resulting in a re-meltable, weldable and re-shapable material. Common processing methods are the compression molding or the fabrication of highly filled pellets via (multi-screw) extrusion combined with a quasi-continuous injection molding process. Due to the high viscosity of the highly filled composite, injection molded bipolar plates are limited in size and minimal thickness (at least 2 mm).

Two approaches for the continuous production of thermoplastic-based bipolar plates are investigated to date. The first one concerns the combination of a multi-screw extrusion and a calandering process, which is studied at the hydrogen and fuel cell center ZBT GmbH in Duisburg, Germany. With this process combination, material thicknesses of down to 1.5 mm have been achieved. However, even thinner bipolar plates can be realized with a powder-based roll-to-roll process developed by Fraunhofer UMSICHT in Oberhausen, Germany. These highly flexible bipolar plates can currently be produced in thickness from 0.1 mm to 1 mm (Fig. 7).
4.2 Membrane

The membrane, as central cell component, ensures the ion exchange between anode and cathode. That is why the free membrane area is also defined as active cell area. Another task of the membrane is the spatial separation of anolyte and catholyte, thus preventing the mixing of the electrolytes. Mixing of anolyte and catholyte causes the so-called cross-contamination, which results not only in a loss of efficiency but also in a reduction of storage capacity over the service lifetime. The ion selective membranes used in flow batteries are usually only a few hundredths of a millimeter thick and can be divided in two main groups: cation and anion exchange membranes. In anion exchange membranes, the cell internal electrical circuit is closed by the exchange of sulfate ions, in cation exchange membranes by the exchange of protons. Furthermore, both membrane types can be divided into fluorinated and non-fluorinated membranes, according to their basic polymer. A major advantage of anion over cation exchange membranes is their low cost and higher ion selectivity, which increases the efficiency of redox flow batteries. Fluorinated cation exchange membranes have an excellent chemical stability and high ion conductivity, but tend to accumulate or even volatilize due to higher mechanical loads, membranes can also be provided with a supporting polypropylene mesh (Fig. 8).

Besides cation and anion exchange membranes, there are also mixed forms such as amphoteric, bipolar and mosaic exchange membranes [22]. In order to withstand higher mechanical loads, membranes can also be provided with a supporting mesh (Fig. 8).

The general requirements for flow battery membranes are:

- a good ion conductivity, since a good ion pass through minimizes ohmic losses,
- a high ion selectivity, to minimize cross-contamination and self-discharge,
- a high chemical stability, as the membrane must be resistant to the substances used, so that the battery can reach a high calendar age,
- a low cost, as membranes account on average for 10 to 15 % of the total costs of the flow battery system [23].

4.3 Graphite Felt Electrode

Among the membrane and monopolar or bipolar plates used, the porous felt electrodes centrally determine the performance and long-term stability of redox flow batteries [20]. Due to their good electrical conductivity and chemically inert behavior, mostly carbon-based materials are used as starting materials. Electrodes made of metallic elements, the so-called dimensionally stable electrodes, have not prevailed over carbon-based electrodes due to the high material costs and the increased tendency to passivation reactions [24]. Graphite felts, carbon papers, graphene and carbon nanotubes can be used as carbon-based electrodes. Compared to the graphite felt, the carbon papers show a poorer mechanical stability and a shorter lifetime, especially for large battery systems, and like the graphene and carbon nanotubes are currently still subject of research [25,26]. The most commonly used porous electrode material in flow batteries is graphite felt, which was primarily developed for thermal insulation in the high-temperature range. As a diffusion electrode, the material offers the necessary properties to improve the performance of the system. In addition, electrodes with the largest possible specific surface area should be used to accelerate the reaction kinetics. Porous graphite felts are particularly suitable for this purpose, since they have an internal structure and realize a three-dimensional electrode, in which the specific surface area accessible to the electrolyte is larger than the geometric one [27].

Graphite felts for flow batteries consists of fibers based on polyacrylonitrile with a carbon content of over 99 %, a round cross-section with a diameter of 6 to 10 µm and a length of at least 1 mm (Fig. 9). Relatively large homogeneous cavities with an average pore diameter of 197 µm and a total porosity of up to 97 % are formed between the fibers [28, 29]. The permeability of the felt increases with the increase in fiber or pore diameter. In contrast, the specific surface area decreases as the fiber diameter increases. The specific surface area is the total of all surfaces accessible to the electrolyte flowing through and provide the reaction field for the electrochemical reactions. As the graphite felts serve as electron drainers, a larger electrode surface accelerates the flow of electrons and the speed of the redox reactions, which increases the performance of the battery significantly. However, a good permeability and homogeneous electrolyte distribution within the half-cells must be ensured in order not to impair the efficiency of the battery by excessive flow resistance. Therefore, for the design of a flow battery cell a satisfactory compromise between the correlating physical quantities have to be found to keep the flow resistance and pressure within the cells at a tolerable level [30–33].
5 Stack Designs

The stack designs of redox flow batteries can basically be distinguished according to the type of electrolyte supply. The internal electrolyte supply is the most common used design. In this design, the cells are supplied via internal manifolds and the manifold entries and exits in the end plates are the only outside connection (Fig. 10).

This stack design is suitable for mass production and require only simple hydraulic peripherals. Only the type of sealing limits the cell thickness. Disadvantages are the complex sealing around the internal manifolds as well as the lack of insight into the actual electrolyte distribution on the cells in a stack. This design is contrasted by the stack design with external electrolyte supply, in which each flow cell has a direct outside connection and is supplied separately via external manifolds (Fig. 11).

This design offers the advantage of an easy connection and sealing of cells as well as the possibility to directly monitor and control the electrolyte flow in each individual cell. Disadvantages of the external supply design are the costly production as well as the limited cell thickness, as the capillary tubes have to fit in the cell frames.

6 Numbering-up vs. Scale-up

While the storage capacity of redox flow batteries is primarily determined by the available electrolyte volume, the power output can be adjusted by varying the active cell area. One option to increase the performance and therefore the overall active cell area of a redox flow battery system is an increase in the number of battery cells. With this numbering-up, either several mostly smaller cells can be electrically connected in series, which increases the technical voltage, or electrically in parallel, which increases the current.

Another option is the scale-up of the active cell area of each individual battery cell (Fig. 12). Previous studies have already shown that increasing the active cell area per cell has positive effects on the process-technological properties of redox flow batteries. Among other things, it was found that it reduces the influence of shunt currents on an equally level with an extension and rejuvenation of the meandering flow channels in the cell frames. Furthermore, it has turned out that a battery cell with a larger electrode may require a lower flow rate, compared to cells with smaller electrodes, to achieve an equally high convection of reactants into the reaction zones of the electrode [35].

Further advantages of the scale-up result from the reduction of the total number of cells and stacks in the flow battery system and the more efficient use of cell components. These advantages are for example the reduced need for peripherals, reduced ohmic losses and external electrical and hydraulic losses as well as reduced manufacturing and material costs [36, 37].
The higher resource efficiency by a scale-up can basically be clarified by the rule of proportionality, which declares that the circumference of a rectangle with a fixed aspect ratio will only be doubled, when the surface area is fourfolded. This means, the degree of utilization of membranes and bipolar plates will increase with a scale-up, as the percentage of the clamping or sealing area at the edges, which is not available for the reaction processes, will decrease. An important point of view, as cost allocation studies have shown that the membrane accounts for an average 60% of stack material costs. Furthermore, the costs for the cell frame material are not to be underestimated, as they alone account for an average 5% of stack material costs [38–41].

In order to determine the proportional cost development of the cell frame material for different cell sizes more precisely, exemplary calculations are carried out below. It was studied how much material is required to embed the manifolds, the active surface area as well as the correspondingly varying fluid channels. As cell frames for redox flow batteries are usually manufactured by injecting molding, the required material volume can be converted directly into the frame. For the calculations, four different active cell area formats with approximately equal aspect ratios are compared (Tab. 1).

The cell frame of a conventional all-vanadium redox flow battery with internal electrolyte supply, meander-shaped channels and a comb-like distributor structure into the active cell area was chosen as design model. For all four active area formats, the basic dimensions, which are indicated in Fig. 13, are the same. The channel length is correlated to the width of the active area and comprises of two and a half meander bows at the supply side and another two and a half meander bows at the removal side.

The width of the channels in the cell frame is equal to the distance between the channels and depends on the occurring pressure losses in the channels of each active area format due to friction as well as bends (Fig. 14). The channel widths are designed in such a way that the resulting pressure losses of the active area formats vary proportional to the change of the active cell area. In a first step, the flow rates for the different active area formats are calculated (Tab. 2).

For the calculation of the channel widths, the minor losses due to bends as well as major losses due to friction in

| Active cell area [cm²] | 600 | 1200 | 2400 | 4800 |
|------------------------|-----|------|------|------|
| Active length [mm]     | 300 | 400  | 600  | 800  |
| Active width [mm]      | 200 | 300  | 400  | 600  |
| Aspect ratio [-]        | 1.5 | 1.3  | 1.5  | 1.3  |
the channels can be determined using the Darcy-Weisbach equation (Tab. 3). For the cell frame with 600 cm$^2$ active cell area a channel width of 5 mm was set to generate a reference loss.

The originally calculated non-integer channel widths, where the total pressure losses were directly proportional to the change of the active cell areas of the individual formats, were rounded to integer values. However, the ratio of the total pressure losses deviating in each case as a result is still comprehensible, so that the calculation of the material requirement for the cell frames can be continued on the basis of the integer channel widths.

The outer dimensions of the cell frames, the required material volume, minus the active cell area, the manifolds and the inserted channel structures, as well as the ratio of the required material volume to the active cell area can be seen in Tab. 4.

It was found, that, following the rule of proportionality, the ratio of the required material volume to the active cell area decreases with increasing size of the active area. By realistically adapting the channel structures, the direct proportionality was eliminated, but the trend was maintained, whereby the higher resource efficiency by a scale-up could be demonstrated. Thus for an eightfold increase of the power output of redox flow batteries, with a scale-up from 600 to 4800 cm$^2$ active cell area only half of the cell frame material is needed compared to the numbering-up.

However, the scale-up of redox flow battery cells is limited on the one hand by the available sizes of the functional components and the feasible sizes of the cell frames and stacks, and on the other hand by very practical considerations such as handling and transport.

In order to maximize the resource efficiency of redox flow batteries while considering feasibility and handling, a well-balanced combination of scale-up and numbering-up is recommended.

### 7 Applications and Examples

Due to their flexible scalability of storage capacity and power output, redox flow batteries can be adapted specifically and thus resource-efficiently to various applications. With the different designs and cell chemistries of redox flow batteries, power and energy densities may differ greatly. While the power density depends primarily on the cell design as well as on the functional components used, the energy density primarily depends on the cell chemistry. Just for all-vanadium flow batteries the power density may vary between 50 and more than 500 mA cm$^{-2}$ with an energy density of about 30 Wh L$^{-1}$, while zinc-polyiodide flow batteries have even reached 167 Wh L$^{-1}$ in a developmental stage [43, 44].

However, it can be said that redox flow batteries are so far particularly suitable for stationary storage applications, focused on middle- and long-duration bulk storage of energy with a flexible power-to-capacity ratio. As an example, for a typical energy shift from daytime to nighttime or vice-versa, there is a need for a power-to-capacity ratio of between 1:4 and 1:10.

For flow batteries, there are currently three main markets:

- in-front-of-the-meter applications (FTM),
- behind-the-meter applications (BTM),
- off-grid or remote applications [21, 45–47].
Renewable shifting, flex ramping as well as transmission and distribution deferral (T&D) are typical FTM applications for redox flow batteries, while the BTM power management is an example for the few behind-the-meter applications [48]. The suitability of flow batteries for FTM application can be explained by advantages such as high storage capacity, deep discharge resistance, long calendric lifetime and high cycle stability.

In the case of BTM, the focus is primarily on larger industrial applications, as the low energy density of current flow battery systems as well as the higher percentage of losses with smaller systems (as discussed in Sect. 6) are potential disadvantages in the household sector. So far, BTM applications have been mainly addressed by using lead-acid batteries and lithium-ion batteries. Above all, the combination of local generation (e.g., roof-top photovoltaic) and electrical storage offers the option to act as feeder, consumer and prosumer – a multitude of possibilities for applications. These depend largely on energy consumption, energy contracts and national regulations. Since small redox flow battery home systems are not yet widely available on the market, there have been mainly research and demonstration projects in which flow batteries are tested for a BTM application. In most of these cases, the consumer is optimizing its self-consumption and by this the electric bill. The consumer shifts the surplus of electric energy to evening hours or early mornings for a higher share of self-produced energy.

For storage tasks, which require high charging rates, such as renewable smoothing and further ancillary services, high power density batteries, like lithium-ion batteries, are usually better suited than redox flow batteries (Tab. 5).

Off-grid or remote applications include island grids and remote area grids with or without a negligible minor connection to a main grid. Because of the individual economics and demands in remote area grids, there are less typical but more individual applications.

In the following, two application examples of redox flow batteries are presented:

Japan, Hokkaido: 17 MW/51 MWh all-vanadium flow battery connected to a wind farm (FTM: Renewable shifting/T&D deferral)

Sumitomo Electric is going to install a 17 MW/51 MWh all-vanadium redox flow battery system for the distribution and transmission system operator Hokkaido Electric Power on the island of Hokkaido from 2020 to 2022. The flow battery is going to be connected to a local wind farm and will be capable of storing energy for three hours. The overarching aim of the project is the integration of larger shares of renewable energy by renewable shifting, to avoid or at least postpone grid extensions.

To increase the utilization rate of further wind farms with a total power output of 600 MW, which will be realized between 2017 and 2022, Hokkaido Electric Power is already targeting the deployment of further flow batteries with a power output of 90 MW and a capacity of 360 MWh. In a second step, additional flow batteries with 60 MW power output and 240 MWh storage capacity are planned for the expansion of the wind farm by 400 MW [49].

China, Qinghai: 0.32 MW/1.92 MWh all-vanadium flow battery connected to a solar farm (FTM: Renewable shifting)

In the province of Qinghai in China, the Avalon Battery Corporation has installed 64 all-vanadium redox flow battery modules, each with a power output of 5 kW and a capacity of 30 kWh. These modules are connected to a solar farm with a total 1.7 GW photovoltaic power output. As a special technological feature, each photovoltaic tracker row is DC-coupled with one of the 64 battery modules to reduce the number of converters required (as shown in Fig. 15). The flow battery system is energy shifting from daytime to evening and nighttime to reduce curtailment [50].
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