

Designing modern aqueous batteries

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Abstract

In the pursuit of more reliable and affordable energy storage solutions, interest in batteries powered by water-based electrolytes is surging. Today's commercial aqueous batteries lack the energy density and cycle life required to compete in the fast-growing transportation and grid storage sectors, but this will change as new materials and cell design strategies are developed. Many of the constraints of traditional aqueous batteries have been alleviated by innovations such as selective membranes, lean-water electrolytes and new types of electrode reactions. As a result, an unprecedentedly broad range of electrode chemistries may be paired in previously impossible ways via modular cell designs to achieve performance metrics unattainable by traditional aqueous batteries. These innovations, however, change the properties for which aqueous batteries are traditionally known, and may result in compromises. This Review starts by examining the historical evolution of aqueous batteries, summarizing their essential merits and limitations. It then analyses how modern chemistries and cell designs may further strengthen the merits of aqueous batteries and address their limits while sometimes compromising prior merits, providing a holistic and critical overview of modern aqueous battery design.

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Introduction

With lithium-ion batteries powering today's flashiest inventions, from smartphones to electric vehicles, and projected to capture over 80% of the rechargeable battery growth in the coming years, it is common to overlook that fireproof batteries using aqueous solutions as electrolytes still constitute half of the rechargeable battery market¹. Today's commercial aqueous batteries, most of which were developed decades ago, lack the energy density and cycle life required to compete for transportation and grid storage applications. However, the ongoing pursuit of more reliable and affordable energy storage solutions than lithium-ion batteries is driving researchers to reinvent aqueous batteries. In recent years, material innovations have enabled an explosion of new battery designs. Many appealing chemistries that were previously pursued but difficult to integrate into a battery have now become viable with the advent of new materials. New chemistries have been proposed to achieve unprecedented performance metrics. These newly discovered and rediscovered chemistries are joining those currently in use in traditional aqueous batteries to enable cell designs that were previously inconceivable. The frontier of aqueous batteries is rapidly advancing, transforming the technology into something so distinct that a fresh assessment on its underlying chemistry, operating characteristics and application areas is warranted.

This Review aims to capture the modern transition of rechargeable aqueous batteries and envision the path forward. We begin by reflecting on the history, characteristics, merits and limitations of traditional aqueous batteries. This recap provides the context within which we can critically evaluate new advancements in the field. We then provide a comprehensive summary of the material innovations and the resulting new chemistries and battery designs that have enabled the transition of aqueous batteries to a modern energy storage technology. We conclude by analysing the emerging directions and offering our views on where the technology should go next.

History

The timeline of the invention of major batteries and related technologies is summarized in Fig. 1. The first rechargeable batteries appeared more than 160 years ago, when Gaston Planté invented lead–acid batteries in 1859, 10 years before electrical generators were invented². The technology uses the $\text{PbO}_2/\text{PbSO}_4$ redox for the cathode reaction, PbSO_4/Pb for the anode reaction and sulfuric acid as the electrolyte. It took more than two decades and the combined efforts of multiple groups to perfect the design of current collectors and the fabrication of the electrodes in order to bring lead–acid batteries to market³. Since

then, the technology has been constantly improving, with multiple iterations opening up new application areas. Immobilization of the acid via gelled electrolyte and adsorptive glass-mat separators led to the invention of maintenance-free valve-regulated lead–acid batteries in the mid-1900s, making the technology suitable for automobile batteries for SLI (starting, lighting and ignition)⁴. New additives at the lead anode enabled the batteries to withstand high-rate partial-state-of-charge conditions and support low-emission automobiles with stop–start capabilities at the end of the 1990s^{5,6}. Lead–acid batteries continue to play an important role in today's energy storage technologies, accounting for 50% of the rechargeable battery market by revenue in 2019 (ref.¹).

As the first alkaline rechargeable batteries, nickel–iron batteries were invented at the start of the twentieth century⁷. Because of its fairly high specific capacity (289 mAh g^{-1}), high reversibility, and closeness of its redox potential to the anodic stability limit of alkaline electrolytes, the $\text{NiOOH}/\text{Ni(OH)}_2$ redox has remained the cathode of choice for alkaline batteries. The hunt for a superior anode material to pair with the nickel cathode continues to this day. Although the iron anode has high theoretical specific capacity (905 mAh g^{-1}), it is thermodynamically unstable in water and subject to corrosion⁷. The $\text{Cd(OH)}_2/\text{Cd}$ redox quickly replaced iron and came into widespread use, ranging from heavy-duty industrial applications to portable rechargeable devices, until it was phased out owing to toxicity and stagnating energy density (35 Wh kg^{-1})⁸. In the 1970s, hydrogen gas was identified as a long-lasting anode material to couple with the nickel cathode⁹. However, the high-pressure tanks necessary to store hydrogen gas, and the noble-metal catalysts (such as platinum black) for hydrogen reactions, limit nickel–hydrogen batteries to extraterrestrial uses where reliability and longevity outweigh cost. Hydrogen-storage alloys, or metal hydrides, were commercialized as low-cost alternatives to hydrogen anodes in 1989, barely two years before the commercialization of lithium-ion batteries¹⁰. Nickel–metal hydride batteries have enabled widespread commercialization of hybrid vehicles, although this market is now being gradually taken over by lithium-ion batteries. Among the next candidates for alkaline-battery anodes is zinc metal, which offers higher energy while being less expensive than other materials.

The aqueous batteries discussed above may be referred to as 'traditional' because they have been on the market for so long and have shaped our expectations for aqueous batteries. These batteries have strengths and limitations that we discuss in the next section. Other aqueous batteries, such as all-vanadium redox flow batteries

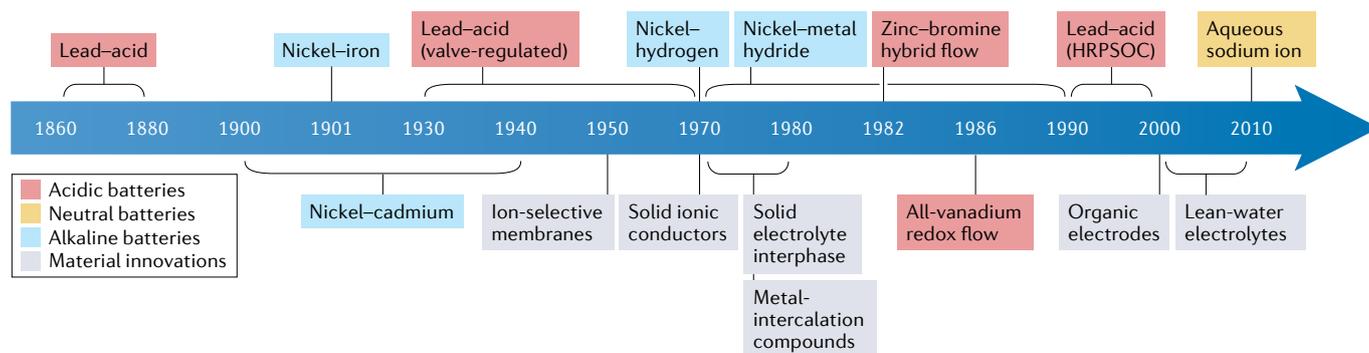


Fig. 1 | Timeline for the development of aqueous batteries and of the materials used to modernize them. Commercial rechargeable aqueous batteries, colour-coded according to the pH of the electrolytes they use, and new materials that enabled innovation in aqueous batteries. HRPSOC, high-rate partial-state-of-charge.

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and aqueous sodium-ion batteries, are in the early stages of commercialization. Considering their distinct performance characteristics, these emerging batteries are better viewed as part of a modern aqueous battery transition towards the next breakthrough.

Merits and limitations

Aqueous batteries are often regarded as safe, reliable and affordable. These perceived merits are in comparison to lithium-ion batteries, which use flammable organic solvents in their electrolyte solutions and catch fire from time to time. Traditional aqueous batteries use electrolytes that contain typically more than 70 wt% water and are thus not usually flammable. Other perceived merits of aqueous batteries are conditional and should not be interpreted as a direct result of the use of non-flammable electrolyte solutions. In this section, we look at the merits of aqueous batteries, their limitations, and the implications for future cell design.

Low cost

The argument of aqueous batteries being low in cost originates from at least three factors: cheap raw materials, minimal requirements for manufacturing environments, and a limited need for battery management and protection systems. The cheaper material cost is immediately understandable, because water and the mineral solutes used in traditional aqueous electrolytes, such as H_2SO_4 and KOH , are far cheaper than superdried organic solvents and fluorinated salts, such as LiPF_6 , used in lithium-ion batteries. This benefit, however, may be diminished if pricey solutes and organic cosolvents are added to aqueous electrolytes. Aside from the electrolytes, the active electrode materials used in traditional aqueous batteries are also mostly low-cost, but this is more of a coincidence than a feature, as exceptions do exist: mischmetal alloys and vanadium electrolytes, for example, are not always less expensive than active materials used in lithium-ion batteries. Aqueous battery manufacturing often does not require the costly and energy-intensive controlled environment conditions required for lithium-ion batteries. However, this advantage is not guaranteed, as several well-known modern aqueous battery designs include oxygen-sensitive and moisture-sensitive materials. Lastly, the minimal dependency on battery management and protection systems of aqueous batteries is due to their safety and reliability, which goes beyond mere non-flammability. These three factors have all contributed to the low capital cost of traditional aqueous batteries. However, for long-term applications such as grid storage, the most relevant cost number is leveled cost, which considers not only capital cost but also service life. Hence, although the capital cost of lead–acid batteries is a fraction of that of lithium-ion batteries, the former may end up being more costly because of their short cycle life.

Safety and reliability

The general non-flammability of aqueous batteries was discussed earlier. Although aqueous batteries may not catch fire, they are nonetheless susceptible to faulty operation and can experience catastrophic events such as explosions¹¹. Overcharging is one type of improper operation that irreversibly decomposes the electrolyte and degrades the battery cell; in lithium-ion batteries it is prohibited by dedicated battery management systems. Such stringent management is not necessary for traditional aqueous batteries, as they tolerate overcharging to a large extent via the ‘oxygen cycle’^{12,13}. In fact, a certain degree of overcharging is a feature for these batteries, because the electrode reactions are designed to approach the electrolysis potentials of water

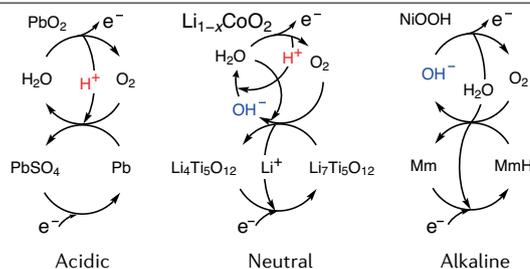


Fig. 2 | Oxygen cycle in sealed aqueous batteries. High charging potentials cause water to oxidize at the cathode surface, producing oxygen that diffuses across the separator, reaches the anode and is reduced. The process raises the pH at the cathode while lowering it at the anode. This local pH variation raises additional requirements on the chemical stability of electrode materials designed for neutral electrolytes, whereas it has little effect on those designed for acidic and alkaline electrolytes.

to maximize cell voltage. When an anode-excess cell is overcharged, water at the cathode is oxidized into oxygen gas, which diffuses through the electrolyte to the anode, where it is reduced and returned to the electrolyte (Fig. 2). During these processes, there is no net loss of water. The water oxidation reaction raises the electrolyte acidity at the cathode, whereas the oxygen reduction increases the alkalinity at the anode. Because the highly acidic and alkaline electrolytes buffer the fluctuations in acidity and alkalinity, these reactions are readily accommodated in traditional aqueous batteries. However, the safety and reliability can diminish if either the electrolyte non-flammability or oxygen cycle is not sustained. Non-flammable aqueous electrolytes can become flammable if the amount of flammable components exceeds certain thresholds (imagine flaming cocktails). The oxygen cycle can be disrupted if the active electrode materials irreversibly react with oxygen and the electrolytes; electrode materials are typically intended to work within a narrow pH range, and oxygen-resistance is not always a priority. Reactions with electrolytes are especially concerning for emerging batteries that use neutral electrolytes, which lack the inherent pH-buffering capability of their acidic and alkaline counterparts. Hence, in addition to oxygen, the active materials used in these batteries need to be stable towards electrolytes of varying pH values, an extra design burden that has yet to be fully investigated.

Fast kinetics

Aqueous electrolyte solutions are generally more conductive than non-aqueous ones under equivalent conditions, owing to high ion dissociation and low viscosity. The proton-conducting electrolytes in lead–acid and alkaline batteries benefit from a hopping mechanism and have conductivities of -0.80 S cm^{-1} ($-30 \text{ wt}\% \text{ H}_2\text{SO}_4$) and -0.60 S cm^{-1} ($-30 \text{ wt}\% \text{ KOH}$), respectively¹⁴. Neutral electrolytes based on metal salts are roughly one order of magnitude less conductive than acidic and alkaline electrolytes (for example, 0.06 S cm^{-1} for $10 \text{ wt}\% \text{ Li}_2\text{SO}_4$)¹⁵, but are still much more conductive than commercial lithium-ion battery electrolytes (-0.01 S cm^{-1}). The higher conductivities result in faster charging and discharging, more consistent performance at low temperatures¹⁶, and the possibility of using thicker electrodes (millimetres rather than hundreds of micrometres as in non-aqueous batteries)^{17–20}, which increases energy density. When the ion association and solution viscosity are increased, the conductivity of aqueous electrolytes suffers, as do all the derived performance metrics.

Narrow electrochemical stability windows

Along with all the merits of aqueous batteries, they have obvious limits that give lithium-ion batteries a market advantage. The first is that water has a narrow thermodynamic electrochemical stability window of 1.23 V (Fig. 3, area 1), beyond which it is anodically oxidized into oxygen and cathodically reduced to hydrogen. Some materials have high overpotentials for these reactions, effectively expanding the stability window to as much as 2.3 V (refs.^{21,22}) (Fig. 3, areas 1 and 2). Among traditional aqueous batteries, lead–acid batteries make the best use of the expanded stability window and have a nominal voltage of -2 V. All other commercial rechargeable aqueous batteries operate just above 1 V. These values are far lower than the 3.3–3.9 V for non-aqueous lithium-ion batteries. The voltage shortfall greatly limits the energy density of aqueous batteries and is hard to compensate with higher specific capacities.

Aggressiveness of aqueous solutions

The high polarity and strong coordinating ability of water make it an excellent dissolving and dissociating solvent. These properties make water a good ingredient for electrolytes but are an issue for many battery components. The strong dissolving capability of water is responsible for the recrystallization of PbSO_4 and the premature failure of lead–acid batteries, popularly known as ‘sulfation’²³. In neutral aqueous electrolytes, LiFePO_4 , a cathode material recognized for its durability in lithium-ion batteries, exhibits dissolution-induced degradation²⁴. Even the famously stable $\text{Ni}(\text{OH})_2$ is susceptible to Ostwald ripening in alkaline electrolytes⁷. Water is also corrosive as an amphoteric compound, especially at the extreme potentials at which electrolysis creates extra acidity and alkalinity. This narrows the choice of active and inactive components, for example excluding certain low-cost current collectors²⁵.

Innovations for modern designs

The merits and limitations discussed above determine the performance and application boundaries of traditional aqueous batteries. New materials and cell designs should overcome one or more of these limitations to transform aqueous batteries into a worthy companion to lithium-ion batteries in the transportation and storage sectors. The material innovations summarized in this section occurred in the past few decades, and some were not created specifically for aqueous batteries. Regardless of their intended function, each of them has resulted in performance superior to that of traditional aqueous batteries, driving the current renaissance of the field. Any advancement should ideally occur without compromising the proven merits of aqueous batteries, but this is rarely the case. It is recommended that these innovations be examined critically, and any accompanying compromises be accounted for before implementation into practical devices.

Selective membranes and coatings

Ion-selective layers, which can take the shape of membranes or coatings, allow some ions and molecules to flow through while blocking others, thereby keeping some battery components from being exposed to undesirable electrolyte conditions. Whereas the electrochemical stability window of water remains constant across all pH conditions, the electrolysis potential changes linearly with a slope of 0.059 V per pH (Fig. 3). Ion-selective membranes that prevent the diffusion of H^+ and OH^- allow the employment of acidic and alkaline electrolytes in the same cell. Bridging the anodic limit of an acidic electrolyte (for example, pH -1) and the cathodic limit of an alkaline electrolyte (for example, pH 15) results in a thermodynamic stability window of $1.23 + 0.059 \times (15 - (-1)) = 2.17$ V. When the overpotentials in each electrolyte are taken into account (Fig. 3, area 2), a stability window as wide as -3.5 V can be expected. Some selective layers further prevent water

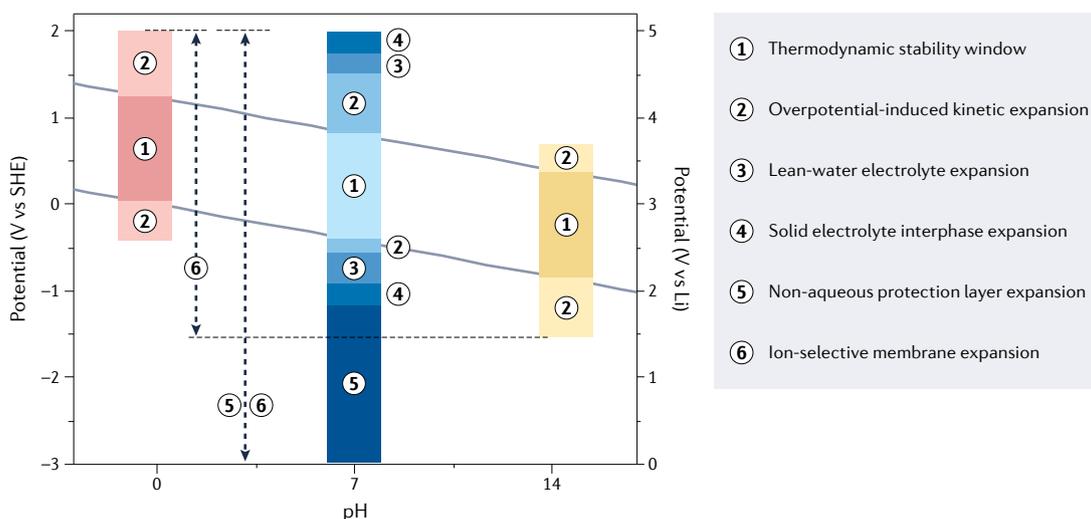


Fig. 3 | Electrochemical stability window of aqueous electrolytes. The thermodynamic electrochemical stability window (ESW) of aqueous electrolytes is 1.229 V wide (1) regardless of pH. The overpotentials of oxygen and hydrogen evolution reactions expand the ESW beyond 2 V (2), within which window commercial aqueous batteries operate. Recently developed lean-water electrolytes with small amounts of free water molecules and a considerably expanded ESW are typically near-neutral in pH (3). Some components of these electrolytes decompose on electrode surfaces and form an insoluble solid

electrolyte interphase, leading to an extra usable window (4). Certain artificial protective layers entirely seal anodes from aqueous electrolytes to unlock negative potentials identical to those of non-aqueous systems (5). Ion-selective membranes bridge cathodes and anodes that operate at different pH conditions, breaking any single aqueous electrolyte stability limit (6). The rational combination of these methods broadens the operating potential of aqueous batteries to the greatest extent possible for any battery system. SHE, standard hydrogen electrode.

molecules from diffusion, enabling the use of moisture-sensitive materials in aqueous batteries and further expanding the stability window to up to -5 V (Fig. 3, areas 5 + 6).

Since the report of a synthetic ion-selective membrane in the 1950s, many practical applications such as chlor-alkali cells and fuel cells have been established^{26,27}. The first batteries enabled by these swollen polymer membranes were redox flow batteries starting in the 1970s: a catholyte and an anolyte each containing a distinct redox couple are separated by a selective membrane to establish a potential difference²⁸. Batteries using these membranes to integrate multiple electrolytes of different pH values into one cell came much later, in the 1990s²⁹. These batteries are discussed in a later section. Solid-state ion conductors block not just ions but also water; therefore, they have been proposed as a protective barrier to enable lithium metal in aqueous batteries³⁰. It took years of efforts to solve the chemical compatibility at the conductor–lithium and conductor–aqueous electrolyte interfaces^{31–35}. The solid electrolyte interphase (SEI) that forms in situ behaves similarly to solid-state ion conductors and has been used to passivate electrodes and expand the stability window^{36–40} (Fig. 3, area 4). The majority of such SEI layers are created by the decomposition of concentrated fluorinated salts and, in some cases, fluorinated solvents, although recent studies have discovered a comparable effect with SEI formed from fluorine-free electrolytes⁴¹.

A top concern with these selective layers is cost. The Nafion membranes in an all-vanadium flow battery were estimated to account for 27–44% of the cost of the battery⁴². Fluorinated salts and solvents are significantly more expensive than common mineral salts and water, respectively. The use of lithium metal raises the voltage of aqueous batteries to that of lithium-ion batteries, but it also imposes more stringent requirements on manufacturing environments. There are ongoing efforts to reduce the production cost and increase the performance of selective membranes to reduce the leveled cost of batteries^{43,44}.

Lean-water electrolytes

The reactivity of water molecules varies according to their coordination status, allowing for manipulation of the stability window by driving water molecules in a solution from ‘free’ to coordinated states. This type of manipulation was first accomplished by greatly increasing the salt content. A ‘water-in-salt’ electrolyte, 21 mol kg⁻¹ lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), has just 14.2 wt% of water³⁶. Traditional aqueous electrolytes, ranging from H₂SO₄ solutions for acidic batteries to Li₂SO₄ solutions for neutral batteries and MOH (M = alkaline metal) solutions for alkaline batteries, all have water content of 70 wt% or higher. The H₂O:Li⁺ molar ratio in 21 mol kg⁻¹ LiTFSI is as low as 2.6, with a fraction of free water of -16%, a huge decrease from the -70% for a diluted 5 mol kg⁻¹ LiTFSI. As a direct result, the anodic limit of the electrolyte rises by -200 mV (Fig. 3, area 3). The addition of a second fluorinated lithium salt broadens the electrochemical stability window even further^{45,46}. Water content was further reduced to 10.2 wt%, and all water molecules were proposed to be coordinated to lithium ions⁴⁶.

Bulky fluorinated anions with their ‘plasticizing’ capability were critical in increasing salt concentration to very high levels in the early examples of lean-water electrolytes, but the exorbitant expense of perfluorinated salts has raised practicality concerns. It was quickly discovered that the inexpensive lithium nitrate (LiNO₃) can be dissolved to 22 mol kg⁻¹ at slightly elevated temperatures (35 °C) and, like fluorinated salts, stabilizes water towards anodic electrolysis⁴⁷. Lithium

salts are not the only ones with enlarged stability windows; extremely concentrated solutions of sodium, potassium, zinc and ammonium salts, many with non-fluorinated anions, also have expanded stability windows^{48–52}.

Electrolyte formulations that do not have a high concentration of salts are also gaining traction. Organic compounds with a high ratio of O and N atoms have been introduced as ‘cosolvents’ to aqueous electrolytes, ranging from polymers such as polyacrylic acid and polyethylene glycol to small molecules such as urea and sugar, and to organic solvents^{40,53–59}. These chemicals disrupt the hydrogen-bonding structure in water to the degree that the characteristic vibrations of water are no longer detectable, effectively removing ‘free water’ in the same way as superconcentration does. It is common for organic cosolvents that produce an efficient disruption to take up 50–90 wt% of the solvent.

Both superconcentration and organic cosolvents work by reducing the water content of aqueous electrolytes, sometimes to a level as low as 5 wt%^{40,59}. Some characteristics of traditional aqueous electrolytes are inevitably compromised. Many of these lean-water electrolytes have conductivities in the range of 0.001–0.01 S cm⁻¹, which are similar to those of non-aqueous electrolytes. The cost of the electrolytes rises owing to the use of organic and fluorinated components. The good news is that, despite the low water content, most of these electrolytes are non-flammable. In certain respects, electrolyte design has evolved towards determining how to use the minimum quantity of water to produce a non-flammable non-aqueous electrolyte without jeopardizing the electrochemical stability window. The distinction between an aqueous and a non-flammable non-aqueous electrolyte (such as those based on phosphate solvents)⁶⁰ is becoming blurred.

New types of electrode reactions

Beyond traditional aqueous batteries, many new types of electrode reactions have been established and are being tested for next-generation aqueous batteries. Among them, intercalation reactions established from lithium-ion batteries provide a variety of structurally stable electrode materials⁶¹. Organic electrodes are highly tunable and may be designed to accommodate aqueous electrolytes of any pH value and salt species⁶². New electrolytes and advanced surface modifications have enhanced deposition reactions, resulting in structurally simple compounds as high-capacity candidates for electrode materials^{63,64}. Flow reactions aided by selective membranes allow capacity–power decoupling and potentially good scalability⁶⁵. Air gases are also being integrated into rechargeable aqueous batteries, and, thanks to new cell designs, are emerging as low-cost and high-capacity electrode materials⁶⁶. Each of these reactions and materials, together with traditional ones, is examined in the next section.

Electrode chemistries

Designing modern aqueous batteries requires a thorough understanding of the various electrode chemistries and the cell designs that may be used to integrate them into batteries. Typically, the various electrode chemistries in aqueous batteries are discussed in the context of the specific battery for which they are built. We adopt a distinct, battery-agnostic approach, reviewing electrode chemistries based on the nature of the reactions themselves, rather than their original use in batteries. This approach is made possible by the modern tools that connect several electrolyte systems. Doing so eliminates unnecessary barriers for comparing reactions of the same type but previously examined in different electrolyte systems and with different counter electrodes. This section presents an overview of electrode chemistries

that are being used and developed for a wide spectrum of aqueous batteries, from old-school lead–acid to the latest aqueous lithium-ion and emerging battery concepts. The chemistries are classified into five types according to their structural characteristics (Fig. 4).

Insertion reactions

Insertion reactions, and the more narrowly defined intercalation reactions, involve the use of ‘host’ materials that accommodate and release ‘guest’ charge carrier ions during redox processes. The structure of the host materials remains largely intact during reaction (Fig. 4a), resulting in the most reversible electrode reactions among solid-state active materials.

In aqueous batteries, intercalation has been used in the alkaline electrode reactions $\text{NiOOH}/\text{Ni}(\text{OH})_2$ and $\text{FeOOH}/\text{Fe}(\text{OH})_2$. These unique reactions can be viewed as proton intercalation in alkaline electrolytes⁷. Intercalation reactions developed for lithium-ion storage starting from the 1970s greatly expanded the scope of this reaction type and inspired the first aqueous lithium-ion batteries in 1994, where both the cathode (LiMn_2O_4) and anode (VO_2) use intercalation reactions⁶⁷. More stable cathodes (such as carbon-coated LiFePO_4) and anodes (such as carbon-coated $\text{LiTi}_2(\text{PO}_4)_3$) have since been discovered⁶⁸. Super-concentrated electrolytes enabled more positive cathodes (such as $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ and high-voltage LiCoO_2) and more negative anodes (such as Mo_6S_8 , TiO_2 and $\text{Li}_4\text{Ti}_5\text{O}_{12}$)^{36,45,46,69}. The development of more energy-dense lithium-ion cathodes, such as lithium–nickel–manganese–cobalt oxides ($\text{LiNi}_{1-x-y}\text{Mn}_x\text{Co}_y\text{O}_2$) and lithium-rich materials, is an obvious next step towards higher-energy aqueous lithium-ion batteries. Further suppression of oxygen evolution, either by reduced water activity or surface passivation, and of the oxygen cycle in near-neutral electrolytes may be necessary to enable these materials. Aqueous sodium-ion batteries are now possible thanks to similar

intercalation chemistries^{70,71}. All the materials discussed so far for aqueous alkaline-metal-ion batteries are derived from lithium-ion batteries, but aqueous-specific intercalation processes are also under active development. In Prussian-blue frameworks, rutile RuO_2 and layered compounds (MnO_2 , V_2O_5 , MoO_3 , Ti_3C_2), fast and reversible intercalation or insertion of H^+ , H_3O^+ and NH_4^+ has been observed^{72–77}. To use these reactions, battery designs that offer competitive energy density will be crucial. Intercalation of multivalent cations (Zn^{2+} , Al^{3+}) has been reported for various oxides (MnO_2 , V_2O_5 , TiO_2)^{78,79}; however, the exact mechanism of these reactions may require more rigorous characterizations than have been reported, and proton intercalation frequently accounts for a sizable percentage of capacity⁸⁰.

In addition to cation insertion, anion insertion is also under active research as anion and dual-ion batteries become increasingly popular. Layered materials such as graphite and MXenes have been extensively studied as anion intercalation hosts^{52,81,82}. Anions can also be accommodated in the amorphous oxide Mn_3O_4 (ref. ⁸³). These materials have substantially lower specific capacities than cation-storing materials.

Most inorganic host materials are selective about which ions they can accommodate. By contrast, organic polymers such as those with carbonyl groups as the redox-active groups appear to host any cations (H^+ , NH_4^+ , Li^+ , Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Zn^{2+})^{84–92}, and those with p-type redox moieties appear to host any anions^{93–95}. They may also be designed to operate at a wide range of potentials and to approach the stability limit of electrolytes at any pH. Because of these properties, they are a versatile alternative for electrode materials.

Conversion reactions

In conversion reactions, the active materials completely reconstruct during electrode reactions, with the crystal structures of the charged and discharged state typically displaying little resemblance

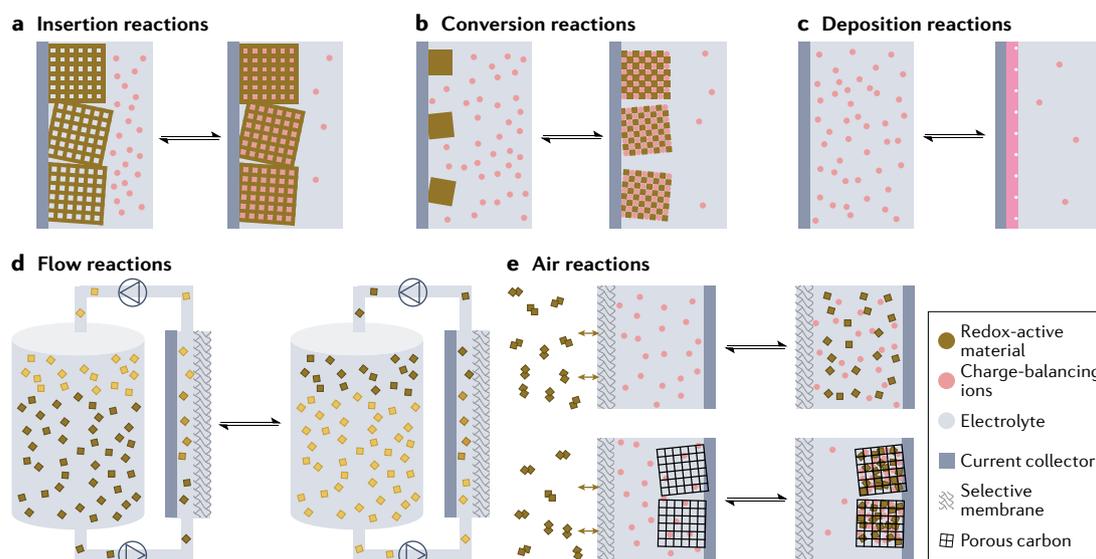


Fig. 4 | Types of electrode chemistries in aqueous batteries. **a**, Insertion reactions: active electrode materials accommodate and release ions during electrode reactions while largely retaining their microstructure. **b**, Conversion reactions: active materials undergo structural reconstruction, with the structures at the charged and discharged states being unrelated in most cases. **c**, Deposition reactions: active materials transition from dissolved (liquid phase)

to deposited (solid phase) forms. **d**, Flow reactions: active materials are flowable at all states of charge; they are externally stored (for example, in a tank) and only react when pumped by the electrode. **e**, Air reactions: active materials in the charged state are components of the air, and therefore no physical storage is required; the discharged state is dissolved in the electrolyte (top) or deposited inside porous carbon (bottom).

(Fig. 4b). Because there is no host structure to maintain, materials that undergo conversion reactions can have simpler compositions and consequently higher specific capacities. Redox pairs such as Cd(OH)₂/Cd, AgO/Ag, S/Li₂S, ZnO/Zn, FeOOH/Fe and others have some of the highest specific capacities among all reported aqueous electrode chemistries^{96–100}.

Materials based on conversion reactions are, however, generally not as reversible as insertion materials. The first rechargeable batteries, lead–acid batteries, use conversion reaction at both electrodes. Of the three main failure modes for the premature capacity loss (PCL) of the battery, two are related to the conversion reaction mechanism¹⁰¹: PCL-2, or cathode softening, results from the large volume change during the conversion between PbSO₄ and PbO₂ (the densities of PbSO₄ and PbO₂ are 48.2 and 24.6 cm³ mol⁻¹, respectively¹⁰²); PCL-3, or sulfation, is caused by a reduction of surface area of the anode active material upon repeated conversion between PbSO₄ and Pb. The different failure mechanisms each require their own solution. Cathode softening was overcome by pressurizing the cathode and only the cathode, as extension of the pressure to the anode promotes failure of the anode¹⁰³. Sulfation was alleviated by nanostructured additives that modulate the microstructure of the re-formed active material^{23,104,105}. Lead–acid batteries are an excellent illustration of how different conversion reactions can affect electrode reversibility in different ways and must be addressed individually, even when they occur in the same cell. Nanoscale conductive agents are now an essential component of most electrodes based on conversion and deposition reactions.

In addition to conversion reactions among metals and their oxides, hydroxides and salts, which have long been used in traditional aqueous batteries, non-metal elementary substances represent newer types of conversion materials. With a high specific capacity of 1,672 mAh g⁻¹, sulfur is one of the most actively studied conversion materials in non-aqueous rechargeable batteries. Because of the high solubility of alkaline-metal sulfides and polysulfides, sulfur was initially implemented as polysulfide solutions in rechargeable aqueous batteries⁹⁸. Sulfur as a solid-state electrode material in aqueous electrolytes was made possible only after the discovery of superconcentrated electrolytes in which sulfur is reduced to insoluble metal sulfides instead of the soluble metal hydrosulfides^{106,107}. Halogens such as bromine and iodine are also looking increasingly practical as solid-state electrode materials with the discovery of effective strategies to solidify reaction products. These strategies combine stabilization of Br⁺ and I⁺ through the formation of interhalogens with F⁻ and Cl⁻, suppression of dissolution and hydrolysis with concentrated electrolytes, and confinement via nanoscaled conductive hosts^{52,108–111}.

Organic molecules are another family of electrode materials that may be considered in the context of conversion materials even though they were not traditionally categorized as such. Recent studies showed that some organic molecules share the heterogeneous dissolution–precipitation reaction path with PbSO₄ and sulfur, but, unlike sulfur and halogens, do not induce chemical bond breakage and re-formation during electrode reactions^{112,113}. Organic molecules, like their polymer counterparts, are versatile electrode materials because they operate at various pH values, have wide-ranging redox potentials and can store a large variety of ions (such as H⁺, H₃O⁺, Zn²⁺ and Al-based ions)^{86,112,114–117}. Some show competitive specific capacities of more than 400 mAh g⁻¹, such as pyrene-4,5,9,10-tetraone (PTO) and benzoquinone (BQ)^{118,119}. Failure mechanisms for solid-state organic-molecule materials are not well studied; dissolution is recognized as one path in certain electrolytes.

Deposition reactions

Deposition reactions, better known as plating reactions in the case of metal deposition, differ from insertion and conversion reactions in that the active materials are solid at one end of the reaction and fully dissolved into the electrolyte at the other end (Fig. 4c). Metals, most notably Zn²⁺/Zn, and metal oxides, mainly PbO₂/Pb²⁺ and MnO₂/Mn²⁺, are the most studied deposition materials for aqueous batteries. Although there are relatively few examples of deposition reactions, they have attracted great research interest due to their attractive redox potentials and specific capacities. During the reactions, the volume of the solid-state active material changes by 100%. The reversibility of deposition is therefore determined by the deposition behaviour and morphology of the depositing material. Control of metal deposition has long been a topic of interest in electrochemistry, with well-established theories ranging from nucleation to growth^{120,121}. Many factors that influence deposition have been identified, including current density, electrolyte composition (pH, cation, anion, concentration), organic additives and, of course, the intrinsic features of the depositing material such as crystal structure and surface adatom diffusion^{122–124}. In batteries, additional factors such as corrosion, surface film and separator structure come into play^{125,126}. Probably owing to such complexity, deposition-type electrode materials are still limited to a topic of fundamental research.

By far the most explored deposition reaction for aqueous batteries is the Zn²⁺/Zn redox, an energy-dense candidate for anodes in neutral and alkaline electrolytes. The main obstacles for zinc deposition are the formation of undesirable microstructures (such as dendrites) and chemical reaction with electrolytes (such as oxide formation and hydrogen evolution). All of the previously mentioned factors that influence deposition have been used as handles to improve zinc deposition, giving rise to solutions such as pulsed charging, a wide variety of electrolyte additives, concentrated electrolytes, surface coatings based on both minerals and polymers, and non-porous separators^{127–134}. These solutions are generally believed to regulate Zn²⁺ flow during deposition, resulting in more uniform plating, and to restrict zinc exposure to water, resulting in less corrosion. Substrates with unique surface lattice, metal decoration and 3D structures also help to ensure uniform zinc deposition^{135–139}. Few other metals under research operate at potentials of interest. Pb²⁺/Pb is suitable for anodes in acidic electrolytes; lead deposition faces the same dendrite formation difficulty as zinc¹⁴⁰. Manganese and aluminium have been evaluated for neutral electrolytes; both metals have extremely negative redox potentials and require special stabilization via, for example, lean-water electrolytes and artificial passivation layers. Nonetheless, they exhibit limited reversibility and stability^{141,142}.

Oxides are uncommon as deposition-type electrode materials. The deposition of lead dioxide from acidic solutions of lead methanesulfonate was first investigated for membrane-less redox flow batteries (pairing with Pb²⁺/Pb) in 2004 (ref. 143). Controlling lead dioxide deposition by applied overpotential and electrolyte composition has been investigated to reduce cracks and stress^{144,145}. Under optimum conditions, deposition proceeds reversibly at >100 mA cm⁻² with per-cycle capacities over 1 Ah cm⁻². Long-term cycling has not been reported for the electrode. Since the demonstration of an acidic MnO₂–H₂ hybrid battery in 2018, the conceptually comparable but more energy-dense and environmentally friendly MnO₂/Mn²⁺ has received a lot of interest¹⁴⁶. Because the deposition of manganese dioxide in acidic electrolytes is electrochemically irreversible, and acidic electrolytes limit the choice of anode materials, a variation of the reaction that uses

near-neutral acetate-based electrolytes was created^{147–149}. To improve the reversibility of the reaction, general methods for optimizing electrochemical deposition are being applied. In the meanwhile, redox mediators have been proposed to mitigate any irreversibility caused by partial dissolution and cracking¹⁵⁰. As discussed in later sections, the redox pair has been paired with a variety of anodes using dual-ion and multielectrolyte cell designs.

Flow reactions

Flow reactions, which are designed for redox flow batteries, involve active materials that are free-flowing in any state of charge, as opposed to the three reaction types outlined above, in which the active materials are fixed at the electrode in all or at least some states of charge (Fig. 4d). Because the active materials are mobile, the bulk of them are stored in storage tanks external to electrochemical cell stacks, leading to decoupled energy and power, as energy is governed by the size of the external storage tanks and the concentration of the active materials in the stored solutions, whereas power is determined by the cell stacks. Such design flexibility has spurred efforts to expand flow reactions beyond flow batteries (see multiphase design in the following sections). Flow reactions are considered attractive candidates for grid storage because storage tanks are less expensive to build than cells of the same size. The absence of solid-state active materials also implies that flow cells are immune from mechanical failure of electrodes common for cells using reactions of other types. The stability of flow cells is instead determined by the chemical stability of the active materials. Most cells using flow reactions also rely on ion-exchange membranes to prevent the crossover of active materials from the cathode and anode compartments. The selectivity, stability and ionic conductivity of the membranes also affect cell stability and kinetics.

Flow reactions were first demonstrated in rechargeable batteries with the redox pairs $\text{Fe}^{3+}/\text{Fe}^{2+}$ and $\text{Cr}^{3+}/\text{Cr}^{2+}$ in the mid 1970s¹⁵¹. A decade later, all-vanadium redox flow batteries (VRFBs) with the redox pairs $\text{VO}_2^+/\text{VO}^{2+}$ and $\text{V}^{3+}/\text{V}^{2+}$ were prototyped and finally entered the stationary storage market^{152,153}. One of the largest VRFB systems, rated 15 MW and 60 MWh, was built in 2015; a larger station rated 200 MW and 800 MWh is planned and set to become by far the largest stationary storage battery facility⁶⁵. These numbers are small compared with the 8.8-GWh installed capacity of lithium-ion batteries for the same application. For the technology to be more competitive, the costs of redox chemicals and ion-exchange membranes must be decreased⁴². The zinc–bromine battery, a hybrid battery that combines the flow reaction Br_2/Br^- with the deposition reaction Zn^{2+}/Zn , is a successful example of the commercialization of a flow reaction based on low-cost chemicals⁶⁵. To control bromine, the technology requires expensive sequestering chemicals, which slightly negates the cost advantage¹⁵⁴. Chlorine ($\text{Cl}_2/2\text{H}_2\text{O}$) and hydrogen are two high-energy chemicals that may be used to replace bromine and zinc in lower-cost flow reactions^{155,156}.

The recent advancement of organic electrode materials has created new design possibilities for flow reactions. Organic electrode materials, which were first introduced in 2010, have provided resource availability, electrolyte versatility, potentially low cost and fast kinetics to flow reactions^{157,158}. Carbonyl compounds, viologen derivatives, nitroxyl radicals and other chemicals have spanned a wide spectrum of redox potentials in electrolytes, from acidic to neutral to alkaline^{159–163}. Because of their sheer molecular size, polymeric organic compounds eliminate the need for ion-exchange membranes and enable the use of low-cost size-exclusion membranes¹⁶⁴. The chemical stability of

organic molecules is a key concern for organic flow reactions, although progress is being made to understand, alleviate and even reverse the side reactions^{165,166}.

Originally created for redox flow batteries, flow reactions are increasingly being adapted into hybrid battery designs, where they demonstrate tremendous possibilities. These are discussed in the subsequent cell design sections.

Air reactions

The chemicals in air reactions, the ambient gases, are free-flowing like those in flow reactions; but because they come from the air, no storage tanks are required, and their energy density is theoretically unlimited. In practice, reduced gas products are deposited in the cell as a solute in the electrolyte (Fig. 4e, top) or as deposits in conductive frameworks (Fig. 4e, bottom)¹⁶⁷. As a result, the energy limit of air reactions is equivalent to that of flow and deposition reactions. The oxygen reaction is by far the most established air reaction, and it is reversible in electrolytes of all pH values¹⁶⁸. The persistent challenge for the oxygen reaction is to develop bifunctional catalysts that are cheaper, more efficient and more stable. The carbon dioxide reaction is gaining traction amid attempts to reduce greenhouse gas emissions¹⁶⁹. There are a few methods to make carbon dioxide reversibly rechargeable in aqueous electrolytes; its low concentration in air (0.03–0.04%)¹⁷⁰ may be the most important barrier. Nitrogen is a substantial component of air and might be more intriguing in this sense: the discharge of nitrogen in fuel cells has been demonstrated¹⁷¹. A way to recharge nitrogen is, however, yet to be found.

Comparison of different chemistries

Figure 5 compares the energy metrics of selected electrode reactions of all five reaction types. At a first glance, numerous pairs of high-capacity reactions may generate sufficient potential difference. However, the cathode and the anode in traditional aqueous batteries share one electrolyte, limiting the choice of reaction pairs to reactions that operate in the same pH condition and preferably also with the same charge carrier. With this limitation, high-capacity low-potential anode reactions for acid electrolytes are non-existent, and candidates for high-capacity high-potential cathode reactions for alkaline electrolytes are extremely limited. The following section introduces a modular design strategy that eliminates such limitation.

Modular design of cells

Traditional aqueous batteries consist of two solid electrodes that share one electrolyte and are separated by a porous separator. The cathode and anode must be simultaneously compatible with the mutual electrolyte, and the electrolyte must support the reactions occurring in both electrodes. As ion-selective membranes and other modern components become increasingly common in cell designs, these needs are becoming less stringent. Using one or a combination of the cell configurations presented herein, any two reactions discussed in the preceding section can be paired to create a cell with desired performance metrics. Aqueous battery design is becoming modular. The configurations of modern aqueous batteries may be classified based on how cathode and anode chemistries are combined into a cell.

Rocking-chair cells

This cell configuration features a single ionic charge carrier shared by two electrodes. The ions migrate from one electrode to the other during charge; the direction of the migration reverses during discharge (Fig. 6a).

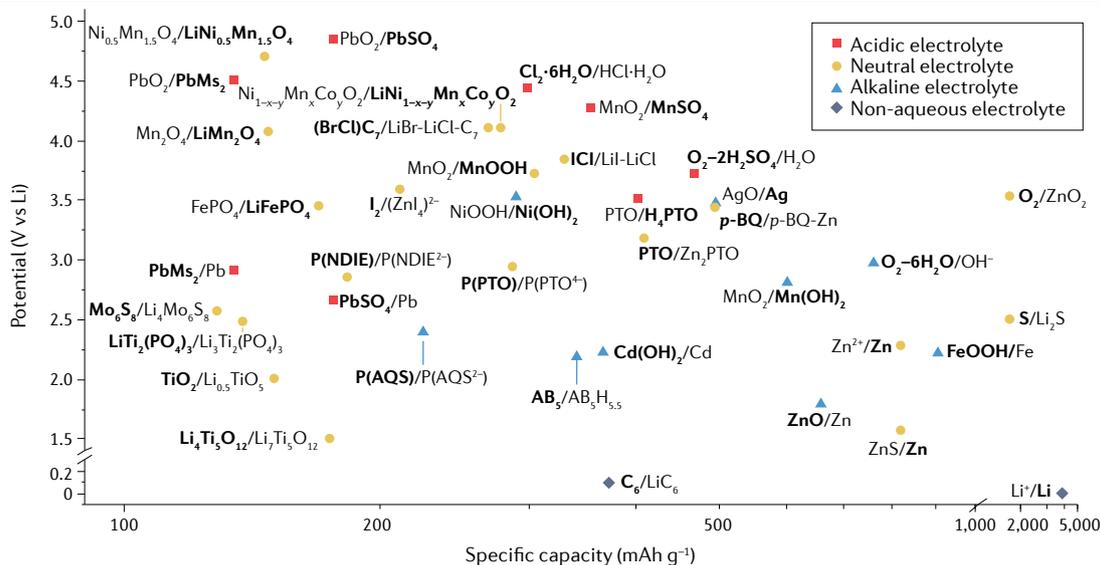


Fig. 5 | Energy metrics for electrode reactions in aqueous electrolytes. Reactions operable in acidic, neutral, alkaline and non-aqueous electrolytes are denoted with red squares, yellow circles, blue triangles and grey diamonds, respectively. Specific capacities are calculated based on the bold formula in each pair. P(AQS), poly(antraquinonyl sulfide); *p*-BQ, *p*-benzoquinone; PbMs₂, lead(II) methanesulfonate; P(NDIE), poly[*N,N'*-(ethane-1,2-diyI)-1,4,5,8-naphthalenetetracarboxiimide]; PTO, pyrene-4,5,9,10-tetraone.

Examples of batteries adopting this cell configuration include nickel–metal hydride and aqueous metal-ion batteries¹⁷². In this cell configuration, the electrolyte serves solely as an ionic conductor and plays no role in chemical storage. This design uses the least amount of electrolyte among all cell configurations and allows for a high specific energy. Because of the shared electrolyte and charge carrier, this design has the most restrictions in terms of cathode–anode combinations.

Dual-ion cells

This configuration allows the stored ionic charge carriers at the cathode and anode to be different, making the choice of electrode chemistries more versatile (Fig. 6b). Examples of dual-ion batteries include lead–acid batteries, where H⁺ is involved in the cathode reaction PbO₂/Pb²⁺ but not in the anode reaction Pb²⁺/Pb; nickel–zinc, where H⁺ is involved in the cathode reaction NiOOH/Ni(OH)₂ while OH⁻ is involved in the anode reaction ZnO/Zn; and various manganese–zinc batteries where the cathode redox couple is MnO₂/Mn²⁺ (in acidic and some near-neutral electrolytes) or MnO₂/MnOOH (in alkaline and some near-neutral electrolytes), while the anode involves Zn²⁺ (in acidic and near-neutral electrolytes) or OH⁻ (in alkaline electrolytes). Because the accommodation and release of ions in the two electrodes are unbalanced, the electrolyte solution must serve as a chemical reservoir. A larger amount of electrolyte is inevitably required than for rocking-chair cells, which may lower cell energy^{173,174}. Dynamic changes in electrolyte composition can have an impact on performance. Supersaturated electrolytes, in which the redox-active solutes dynamically dissolve–deposit, can potentially overcome both issues¹⁷³. The strategy effectively introduces a secondary conversion reaction on top of the main redox reaction. Early implementation of such a strategy showed promise, but its impact on long-term electrode stability warrants further investigation¹⁰⁸. The shared-electrolyte design of dual-ion cells means both electrodes need to be simultaneously stable in the electrolyte.

Multielectrolyte cells

Selective membranes may divide a cell into multiple compartments, each with its own electrolyte (Fig. 6c). This configuration allows pairing redox couples that operate in distinct electrolytes to obtain voltages impossible for single-electrolyte cells. The earliest multielectrolyte cell was demonstrated in the early 1990s, using a bipolar membrane bridging the acidic cathode reaction PbO₂/PbSO₄ and the alkaline anode reaction Zn(OH)₂/Zn to provide an open-circuit voltage of 2.80 V and an average discharge voltage of 2.3–2.4 V (ref.²⁹). Optimization of the electrolyte compositions increased the open-circuit and discharge voltages to 3.25 and 3.03 V, respectively, with an even higher charging voltage of over 4 V (ref.¹⁷⁵). These voltage values far exceed those of traditional aqueous batteries and are more in line with those of lithium-ion batteries.

The significance of multielectrolyte cell design extends beyond breaking voltage boundaries for aqueous batteries. When the electrolyte for one electrode cannot reach the other in the same cell, neither electrode has to be stable towards both electrolytes. A pair of cation- and anion-exchange membranes has been suggested to enable any redox pair combination known to aqueous systems¹⁷⁶. Ceramic ion conductors, which are not only ion-selective but also water-blocking, further allow the integration of non-aqueous compartments into an aqueous battery, unlocking redox reactions normally reserved for non-aqueous system, including the most negative Li⁺/Li (ref.¹⁷⁷). With the possibility to integrate virtually any redox reaction, the design space for modern aqueous batteries appears infinite. The big challenges for these designs include cost of components (especially membranes), economical fabrication and stability of interfaces.

Multiphase cells

Multiphase cells break the boundary between static, flow and air reactions, allowing for the greatest flexibility in cell design. The design overcomes the key limitations of several redox couples, boosting

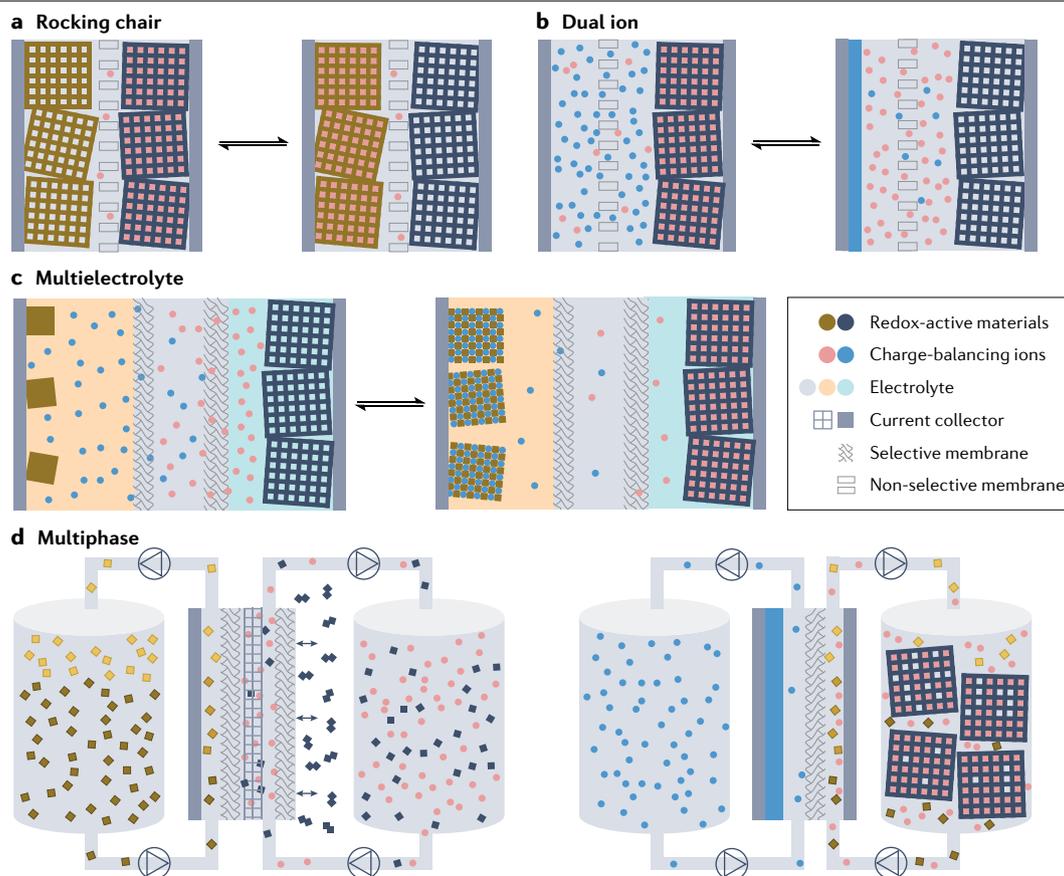


Fig. 6 | Configurations of aqueous batteries. **a**, Rocking-chair batteries: a single common ion serves as the ionic charge carrier for electrode reactions at both electrodes; the concentration of the common ion remains constant at all states of charge. **b**, Dual-ion batteries: distinct ions participate in the electrode reactions at separate electrodes; the concentration of each ion varies with the state of charge. **c**, Multielectrolyte batteries: more than one electrolyte is integrated into one cell to enable cathode and anode chemistries that cannot be simultaneously

supported by a single electrolyte, typically with the use of selective membranes and for the benefit of a broad voltage range. **d**, Multiphase batteries: electrode chemistries without moving phases (such as insertion, conversion and deposition) and chemistries with various moving phases (such as flow and air) are combined in a single cell, with more than one electrolyte used in some cases. The schematics show a flow cell that incorporates an air reaction (left) and a dual-ion cell that adopts a flowing design for deposition and intercalation reactions (right).

the feasibility of already promising reactions. The capacities given by deposition and air reactions, for example, are both limited by the amount of electrolyte, which is restricted by the finite volume of a cell. A flow design overcomes the volume barrier, allowing oxygen and zinc electrodes to be used in potentially large-scale hybrid batteries^{178,179}.

The solubility of redox-active chemicals continues to restrict the capacity of flowing media. The incorporation of undissolved active material particles into liquid supporting electrolytes increases the solubility limit: the equivalent concentration of redox units in solid-liquid ‘semi-flow’ inks can be several times higher than that of ordinary, fully dissolved redox electrolytes¹⁸⁰. Controlling rheology by fine-tuning the ink composition (particle size, solid loading, surfactant and so on), and the potential energy loss resulting from higher viscosity will require attention¹⁸¹. An alternative strategy is to immobilize the undissolved active materials inside a storage tank and use flowing redox mediators with suitable redox potentials to be chemically oxidized/reduced in the tank by the active material and then electrochemically reduced/oxidized in the cell¹⁸².

Two examples of the numerous conceivable combinations of reactions and phases in multiphase cells are shown in Fig. 6d. The cell on the left combines a flow-designed air electrode, where the storage tank absorbs the reduced product of air, with a traditional flowing half-cell to create a potentially low-cost air-polysulfide cell¹⁸³. The cell on the right pairs a flow-designed zinc deposition reaction with a redox mediator-assisted semi-flow half-cell to produce a Prussian-blue-zinc cell¹⁸¹.

Conclusion and outlook

Modern aqueous batteries can use a substantially larger spectrum of redox couples than previously feasible for aqueous systems, thanks to the discovery of lean-water electrolytes and advanced surface coatings, and the rapid development of non-aqueous batteries. The redox couples can now have redox potentials well beyond the stability limit of water. Owing to more reversible reaction processes, their stability can be sufficiently high for long-term use. Selective membranes allow for the integration of multiple electrolytes, whether acidic, alkaline or even non-aqueous, in a single battery cell. Taken together, these

new advancements enable a modular design of aqueous batteries in which any two redox couples may be paired to achieve the desired performance metrics.

Although the limitations of traditional aqueous batteries may very well have been lifted by modern tools and cell designs, the compromises introduced cannot be overlooked. Most lean-water electrolytes and selective membranes are still expensive, undermining the cost advantage of aqueous batteries. The complexity of fabricating multi-compartment cells and the use of extremely electronegative materials (such as lithium metal) both add to the cost. Ionic conduction in lean-water electrolytes and across interfaces is comparable with that in non-aqueous electrolytes, but falls far short of conventional expectations for aqueous systems. The implementation of the oxygen cycle seems to be problematic in some cell configurations, necessitating robust battery management. Non-flammability is the single advantage of traditional aqueous batteries that seems to be largely preserved. Systematic battery-level analysis will be required to measure the competitiveness of modern aqueous batteries with respect to competitors.

Electrolytes, membranes and electrodes all require continued improvement before a commercially impactful modern aqueous battery arises. In addition to the previously outlined strategies, we highlight the following urgent but underexplored research directions.

- Electrolyte-related issues. The compromises that come with lean-water electrolytes offer much room for improvement, particularly in terms of ionic conductivity, cost and non-flammability. Understanding the ionic conduction, flammability and (electro) chemical stability of solutions with a moderate water content (5–70 wt%) could open up new research areas in material science. The oxygen cycle in these new electrolytes is still unknown, casting uncertainties on the robustness of many modern aqueous batteries. The realization of the oxygen cycle will reduce the requirements for the potential range of electrode reactions and safety mechanisms in batteries, resulting in greater freedom in the design of both materials and cells. To avoid expected failure from the inactive components, current collectors and binders must be designed alongside the electrolyte. SEI phenomena in aqueous batteries are rapidly transitioning from specialized to mainstream in modern designs, and they are set to become a driving force for innovation.
- Customized membranes. Most selective membranes studied in modern aqueous batteries came from other industries, with substantially different criteria for mechanical robustness, chemical stability, selectivity and dimensions. A membrane intended for use in neutral electrolytes may not require the chemical resistance provided by Nafion. Thinner membranes would presumably be better than stronger membranes in high-energy-density batteries. Reports on membranes dedicated to specific electrode reactions are still rare, but they are a growing area for performance optimization and integration into energy-dense batteries.
- Practical dual-ion cells. The dual-ion configuration has opened up numerous opportunities in terms of electrode chemistry options, but the compromised energy density and, potentially, longevity have kept them from becoming serious contenders for practical applications. Multielectrolyte and multiphase cells frequently face the same challenges when multiple ions of the same charge are involved. Supersaturated electrolytes represent a sensible approach, but reports on this solution have been scarce and its effectiveness remains largely unproven. Research on materials and electrode design is required to assure reversibility, because

of the partially conversion-type mechanism, which is known to cause structural failure. Alternative strategies are urgently needed.

Although it is important to address these topics independently, it is equally important to focus on strategies for integrating the different solutions. The integration should be easily scalable, so that cell production will not present an insurmountable hurdle to manufacturing. Despite the additional layers, the resulting cell stack should remain lightweight and compact, preserving cell energy. The characteristics of modern aqueous batteries differ substantially from those of traditional aqueous batteries, and so should the research themes and approaches.

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Author contributions

All authors contributed to the discussion of content and writing of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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