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Challenges in the energy storage

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ABSTRACT

Portable electronic devices, electric vehicles, and renewable electric energy storage are today of enormous importance for the prosperity of human civilization. However, different uses require different electrochemical power sources in terms of size, capacity, energy, and power. The paper will consider different metal-ion systems (Li, Na, Ca, and Mg), lithium solid-state, lead-acid UltraBattery, redox flow batteries, and rechargeable metal-air batteries. For all the systems, the merits and drawbacks of the enumerated systems are considered. The main objective of this paper is to provide an overview of the state of the art of different electrochemical energy storage systems, and challenges concerning their price, electrical characteristics, and safety, as well as the possibilities of further improvements and applications. In the end, the author's opinion about the applications of electrochemical power sources for different uses in energy storage is given.

Keywords: Metal-ion, Solid-state, Lead-acid, Redox flow, Metal-air.

1. Introduction

With rising concerns over global environmental problems and resource exhaustion, it is urgent to promote the utilization and development of renewable energy sources, such as solar and wind power. However, the power generated from renewable energy resources such as solar energy and wind energy is intermittent and unstable, which can have a negative impact if input into the power grid directly. Therefore, it is indispensable to integrate appropriate electrochemical energy storage (EES) devices to smooth the output of renewable energy production, and balance generation and demand according to the time and climatic availability (El Kharbachi et al., 2020). Electrochemical energy storage, in principle, is a desirable technology that possesses pollution-free operation, high round-trip efficiency, and flexible power and energy characteristics to meet different grid functions, a long cycle life, and low maintenance.

The main electrical characteristics of electrochemical storage systems are voltage, capacity, energy, and power. The voltage, U, V, is a potential difference between the positive and negative electrodes, and for aqueous-based electrolytes, voltages are up to 1.8 V, while for organic-based electrolytes, they can be up to 4.5 V. Capacity, Q, Ah, represents the ability of the electrochemical system to store charge. It is defined as how much current can be delivered from the device for a certain time or vice versa, Q = I×t. The energy, W, Wh, is the capacity multiplied by the average discharge voltage, W = Uav × Q, and is one of the most important parameters of electrochemical storage systems. Finally, power, P, W, is the energy divided by discharge time, P = W/t, and it represents the energy that can be delivered from the system at unit time. One important parameter is also the number of cycles, or how many cycles one system can deliver before losing ~80% of its initial value. It should be noted that all the values of capacity, energy, and power are given as specific or volumetric values in order to compare them.

2. Electrochemical energy storage systems

2.1. *Metal-ion battery*

Today, many research studies has been focused on alkali and earth alkali metal-ion batteries due to their low electrode potentials and high theoretical capacity, especially those based on lithium, sodium, calcium, and magnesium, of which Li-ion batteries are in their mature phase (Liu and Holze, 2022). Some characteristic parameters of the considered metals are given in Table 1.

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Table 1. Some characteristic parameters of Li, Na, Mg, and Ca

Ion	Li	Na	Mg	Ca
Ionic radius (pm)	68	95	72	100
Valence of charge	+1	+1	+2	+2
Electrode potential (V vs. SHE)	−3.05	−2.71	−2.38	−2.76
Volumetric capacity (mAh cm^{-3})	2046	1128	3833	2073
Gravimetric capacity (mAh g^{-1})	3862	1161	2205	1337

However, due to their low potentials, classical water-based electrolytes cannot be used; consequently, protonic organic solvents are the electrolytes of choice. Moreover, pure metals are very reactive with electrolytes and form an isolating surface layer that prevents deposition/dissolution, as well as dendrite formation during deposition. Furthermore, Mg and especially Ca, as brittle metals, cannot be prepared as foils necessary for cylindrical-type cells. Some other issues that should be considered are the high prices of pure metals obtained by molten-salt electrolysis. Consequently, for both positive and negative electrode intercalation, compounds are usually used. The schematic presentation of the metal-ion intercalation battery is shown in Figure 1.

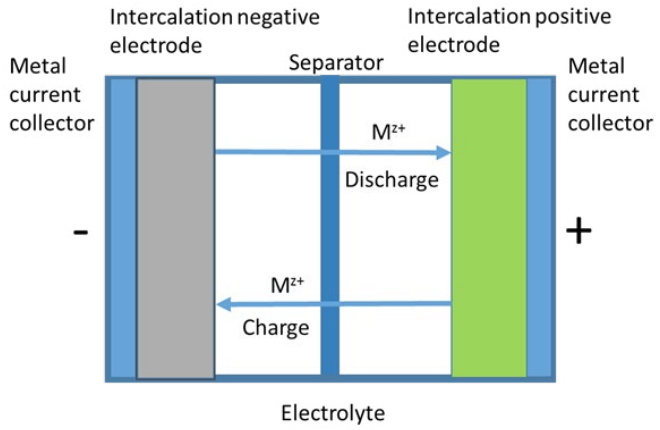


Fig. 1. Schematic presentation of metal-ion battery.

Lithium-ion (LIB) secondary batteries, which are one of the most successful and widely used rechargeable battery technologies, have dominated the current electrochemical energy storage market and have been extensively employed in portable electronic devices and hybrid cars due to their high energy density and portability. Li-ion batteries usually use metal oxide positive electrodes with the typical formula $Li(M_{1,x}M_{2,y}M_{3,z})O_2$, where x+y+z ~1, and the metals (M) are Co, Ni, Mn, Al, etc. In recent times, positive electrodes based on $LiFePO_4$ have lower voltage and specific energy, but are much safer (Hu et al., 2020). As a negative electrode, mostly synthetic graphite is used. Li-ion systems, depending on the electrodes have a voltage above 3.5 V, and specific energy in the range of 120-200 Wh kg^{-1}. However, due to the very high price, poor thermal conductivity that requires additional cooling systems, easy ignitions, safety problems, and low abundance of Li, Co, and other components, researchers have long sought alternative systems to replace Li-ion batteries.

Sodium-ion batteries (SIB) gained academic and commercial interest in the 2010s and 2020s, largely due to the uneven geographic distribution, high environmental impact, and high cost of many of the materials required for lithium-ion batteries. For the anode, SIB could use transition metal oxide $NaTMO_2$, (TM = Fe, Ni, Mn, Cr, V, Co, Cu, etc.), various Prussian blue and Prussian blue analogs (PBAs) with the patented rhombohedral structure, $Na_xM_2[M_1(CN)_6]_y \times nH2O$ ($0 \le x \le 2$, $0 \le y \le 1$, $1 \le n$), where M_1 and M_2 are transition metal such as Fe, Mn, Ni, Cu, Co, for example, $Na_2MnFe(CN)_6$ demonstrated a capacity of 150–160 mAh g^{-1} in capacity and a 3.4 V in average discharge voltage, and polyanionic compounds, like $Na_3V_2(PO_4)_3$ (Hasa et al., 2021, Wang et al., 2018). For a negative electrode, SIB uses different carbon materials that could be obtained from biomass (Yu et al., 2021). The voltage of the SIB today is around 3.2 V with specific energy in the range of ~160 Wh kg^{-1}. It should be noted that largest global battery manufacturer, the Chinese company Contemporary Amperex Technology Co., Limited, CATL, announced SIB of 200 Wh kg^{-1}. Even though both systems use an aprotic electrolyte with $LiPF_6$ or $NaPF_6$ salts, LIB uses a mixture of solvents that are highly flammable, ethylene carbonate/dimethyl carbonate (EC/DMC), while SIB usually uses propylene-carbonate (PC) with a high flash point, 136 °C. In addition, due to the alloying of Li with Al, current collectors are made from Cu that can corrode and form an internal short-circuit within the cell, causing thermal runaway, fire, and explosions, which is not the case with SIB. Consequently, accidents with LIB are more frequent than with SIB. Figure 2 shows examples of the fire of energy storage containing LIB, and perforated SIB without any flame are shown. Additionally, it should be noted that SIB is already commercially produced by different manufacturers, Faradion Ltd, UK (Rudola et al., 2021), Natron Energy USA, Prime Engineers India, Tiamat France, HiNa Battery Technology Co., Ltd, PRC, Altris AB Sweden.

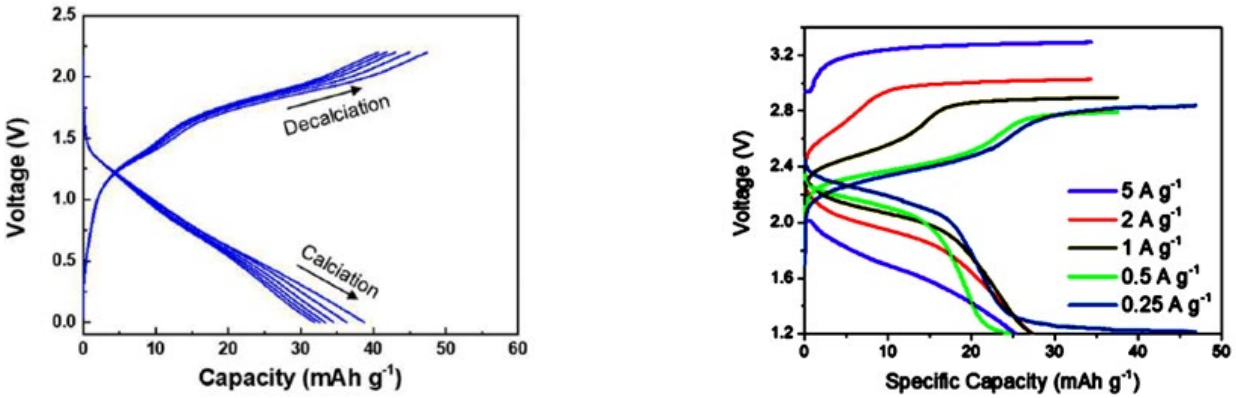
a) b)

Fig. 2. a) Fire at the Victorian Big Battery LIB storage, Australia 2021. b) Faradion nail penetrated SIB pouch cell.

In addition to monovalent cations/metals such as sodium that are attracting attention as possible replacements of lithium divalent cations/metals have been also considered (El Kharbachi et al., 2020, Zhang et al., 2019, Jiazheng et al., 2020, Xu et al., 2021). The obvious advantage, two electrons are released during the discharge reaction of a single cation—does not necessarily translate into a more promising system. The electrode potentials of metals like Ca and Mg are much less negative than those of the alkali metals, as can be seen in Figure 3a (Xu et al., 2021). Accordingly, lower energy densities, which take into account both electrode potentials and cell voltages in addition to charge-storage capabilities, are frequently found. A novel approach in the development of MgIB is the usage of so-called water-in-salt, WIS, electrolyte with a $MgCl_2 \times 6H_2O/H_2O$ with a mass ratio of 25:1, Mg metal anode and CuHCF cathode, Figure 3b, (Leong et al., 2022). Unfortunately, both systems have very low voltage and capacity, and correspondingly low energy density.

2.2. *Lithium solid-state battery*

Mainly due to safety issues and poor thermal conductivity, the search for suitable Li systems in the last few years has been the topic of many research studies (Zhang et al. 2022). One of the solutions is the replacement of liquid electrolytes by solid-state electrolytes like $Li_{14}Zn(GeO_4)_4$, $Li_{3.5}Ge_{0.5}V_{0.5}O_4$, $Li_{10}GeP_2S_{12}$, $Li_{9.54}Si_{1.74}P_{1.44}S_{11.7}Cl_{0.3}$, $Li_7La_3Zr_2O_{12}$, $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$, etc. When using metallic lithium and depending on the positive electrode, which are the same as for the Li-ion systems, they exhibit a high energy density ranging from 280 to 400 Wh kg^{-1} (Guo et al., 2022). There are two main configurations of Li solid-state battery, as shown in Figure 4. In the first type, the metallic lithium is used as the anode, while in the second lithium is deposited by the lithium from lithium-containing cathodes in the first discharge on the copper current collector.



a) b)

Fig. 3. a) Characteristics CaIB of AC/$Ca_xNa_{0.5}VPO_{4.8}F_{0.7}$ coin cell (Xu et al., 2021). b) Characteristics MgIB in so-called water-in-salt, WIS, electrolytes (Leong et al., 2022)

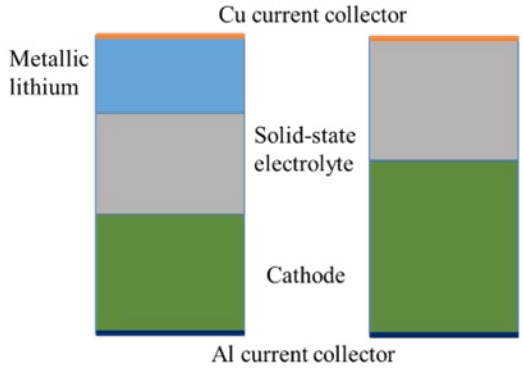


Fig. 4. Two main constrictions of Li solid-state batteries.

Unfortunately, both systems have many disadvantages. The performance of batteries can be severely hampered by the high resistance interphases that inorganic solid electrolytes frequently produce. The production of unfavorable side products during battery operation and the mismatch in the crystal structures of the electrolyte and electrode materials are two of the causes that contribute to this resistance. Also, for the second type, due to the dissolution of lithium from the copper collector, huge possibilities of copper corrosion due to the increase of the electrode potentials exist. The price of metallic lithium on the market varies from 30,000 to 80,000 US $ per ton (Lithium Prices for the Last Year, 2023). Additionally, the production of metallic lithium by the electrolysis of melted LiCl is usually used is an enormous energy-consuming process, 35-40 MWh/t (Chemetal process) which is a few more times higher than for producing sodium metal, 10 MWh/t (Downs process) (Cardarelli, 2018). Along with high energy consumption, a huge amount of toxic chlorine is evolved, ~6 tons per 1 t of Li. For the second system, a much higher mass of the positive electrode has to be used, to accompany successful lithium deposition-dissolution. Unfortunately, deposition-dissolution is not 100% efficient, so “dead lithium” can be produced and a low number of cycles could be achieved. One more disadvantage has to mention, the possibility of lithium dendrite growth during Li deposition as observed by many researchers (Yang and Wang, 2023). Also, the price of the cells is very high around 800 US $ per kWh with a target of 400 US $ per kWh, which is far from the price of a Li-ion battery of ~150 US $ per kWh. Interestingly, the price of the battery pack for Li solid-state battery exceeds 40,000 US $ (Zlatev, 2023).

2.3. Lead acid UltraBattery

During normal operation of a lead-acid battery, lead sulfate crystals form on the negative electrode during discharge. The formation of these crystals is called sulfation. Over time, sulfation can become permanent, especially when batteries are discharged at high currents due to very slow diffusion in the solid phase of lead sulfate crystals (Moseley et al., 2004, Lam et al., 2004). The UltraBattery is a hybrid device that combines supercapacitor technology with lead-acid battery technology in a single cell with a common electrolyte (Lam et al., 2006, Cooper et al., 2009). Physically, the UltraBattery has a single positive electrode and a double negative electrode - one made of surface-developed carbon and one of lead, combined in a common electrolyte as shown in Figure 5. Together they form the negative electrode of the UltraBattery unit. This technology (more precisely, the addition of a carbon electrode provides the UltraBattery with different characteristics compared to conventional VRLA batteries. In particular, the technology demonstrates a significantly lower tendency to develop permanent or irreversible sulfation on the battery’s negative electrode, since at high currents the carbon electrode is mostly discharged, and at low currents, the lead electrode is discharged. By using this technology, the number of cycles increases from ~500-1000 in classic VRLA batteries to up to 15,000 cycles (Lam et al., 2006, Lach et al., 2019).

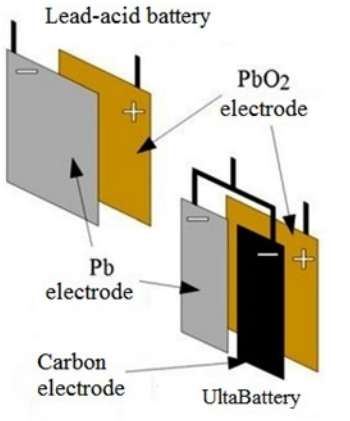


Fig. 5. Schematic representation of lead and UltaBattery (Lach et al., 2019)

2.4. Redox flow batter

The operational principle of the flow redox battery involves the circulation of electrolytes from external reservoirs through an electrochemical cell, as shown in Figure 6. Both during charging and discharging, electrolytes are pumped through the electrochemical reactor (Arévalo-Cid et al., 2021, Weber et al., 2011). During charging, the active substance from the solution is reduced on the negative electrode, and oxidized on the positive electrode, while opposite reactions occur during discharge. Figure 6a illustrates the operational principle, while Figure 6b depicts an installation for the grid storage of solar energy manufactured by AVALON-USA. Any converter can be used as a source of electricity for the charging process, but these accumulators, given that they can have significant capacities, are primarily intended for the

storing of electricity obtained from photovoltaic converters. There are several types of redox batteries, but today they are mainly based on: vanadium (VR); zinc - bromide (ZnBr), and polysulfide-bromide (PSB) redox (Arévalo-Cid et al., 2021). Electrode reactions take place in cell compartments, which are separated by an ion exchange membrane to prevent the mixing of anolyte and catholyte. Electrodes crafted from porous carbon material (graphite fabric) with or without a catalytic layer. The power (kW) and energy (kWh) of redox batteries are independent of each other. The power is determined by the size and number of cells, while the energy capacity is defined by the volume of electrolytes stored in the tanks. Typical characteristics of redox batteries are given in Table 2.

In the past, research was conducted with other redox systems, but the vanadium system is the most widely accepted due to its cost, simplicity, and the fact that it does not contain toxic substances, such as bromine or sulfur compounds. Considering the average discharge voltage of ~1.2 V, for the application it is necessary to connect several cells in series-parallel. Vanadium redox accumulators of lower power, 5 - 10 kW, can be used for continuous power supply of UPS (Uninterruptible Power Supply) devices that require continuous power supply even in case of loss of the primary source of electricity, for example, when using servers, telecommunication repeaters, etc. VRAs of medium power, 10 - 100 kW, can be used to store electricity from solar power plants, wind power plants, and small hydropower plants for independent supply of remote facilities with energy from renewable sources and to enable fast charging of electric vehicles and boats (charging stations for solar-powered electric vehicles). VRAs of large power, >100 kW, enable conventional power plants to supply electricity for a shorter time in moments of maximum consumption, while VBRs are charged in periods of lower electricity consumption.

Table 2. Comparison of characteristics of different redox accumulators.

	VRA	ZnBr	PSB
Discharge voltage, V	1,15	1,6	1,2
Specific energy, Wh kg^{-1}	10-30	35-55	20-50
Power, MW	0,5-100	1-15	0,05-1
Cycle number	>20,000	–	~2500
Efficiency, %	85	75	75

2.5. Metal air battery

Today, the electrochemical energy sources of the metal-air system (Ahuja et al., 2021, Li and Lu, 2017) represent mostly hybrid primary elements and are based on the anodic reaction of the dissolution of an electronegative metal, such as zinc, aluminum, magnesium, lithium, *etc.*:

$$M = M^{n+} + ne^-$$

and the cathodic reduction of oxygen from the air:

$$O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$$

Due to the corrosion instability of the listed metals in acidic solutions, the reaction usually takes place in alkaline, neutral solutions, or aprotic solvents. Today, rechargeable metal-air elements are also being developed, where for the negative electrode, pure metal or intercalation compounds are used, aqueous or aprotic electrolyte from the metal side, aqueous electrolyte for the air side and bifunctional catalyst for oxygen evolution/reduction, as shown in Figure 7. The typical characteristics of some types of metal-air systems are shown in Table 3. From Table 3, it can be seen that such systems far exceed the battery systems considered so far in terms of realistically achieved specific energy (w_s).

Table 3. Basic characteristics of some metal-air elements.

Type	Zn	Fe	Al	Mg	Na	K	Li
U_t,o, V	1.65	1.28	2.71	3.09	2.27	2.48	2.96
U, V	1.2-1.3	~1	1.1-1.8	1.4-2	~2.2	~2.4	~2.6
$w_{s,t}$, Wh kg^{-1}	1086	763	2796	2840	1106	935	3458
w_s, Wh kg^{-1}	350-500	60-80	300-500	400-700	?	?	500?
Relative price	1	0.22	0.95	1.48	9.2	11	40

Unfortunately, the main drawbacks are still possibilities of dendrite formations and poor characteristics of bifunctional positive electrode.

2.6. Hydrogen

Even though hydrogen does not belong to the systems of direct electrochemical storage systems (it is an energy carrier), the electric energy from renewable sources can be converted in the process of electrolysis (Chatenet et al., 2022), and further converted into electricity with fuel cells (Selmi et al., 2022). In the near future, hydrogen can become one of the major factors for grid energy conversion and applications. Unfortunately, the price of hydrogen is still too high, but ongoing research and improvements in existing technologies are expected to decrease its price.

3. Instead of conclusion

Considering systems analyzed above, it could be suggested as follows. For electronic devices, like smartphones, laptops, *etc.* the most favorable systems will likely be Li-based due to their high energy content and because the price is not the limiting factor. Depending on progress, probably within the next few years, the classical Li-ion will likely be mainly replaced by the Li solid-state battery. For large grid energy storage, both today and in the future, sodium-ion batteries and vanadium redox flow batteries could be recommended, while

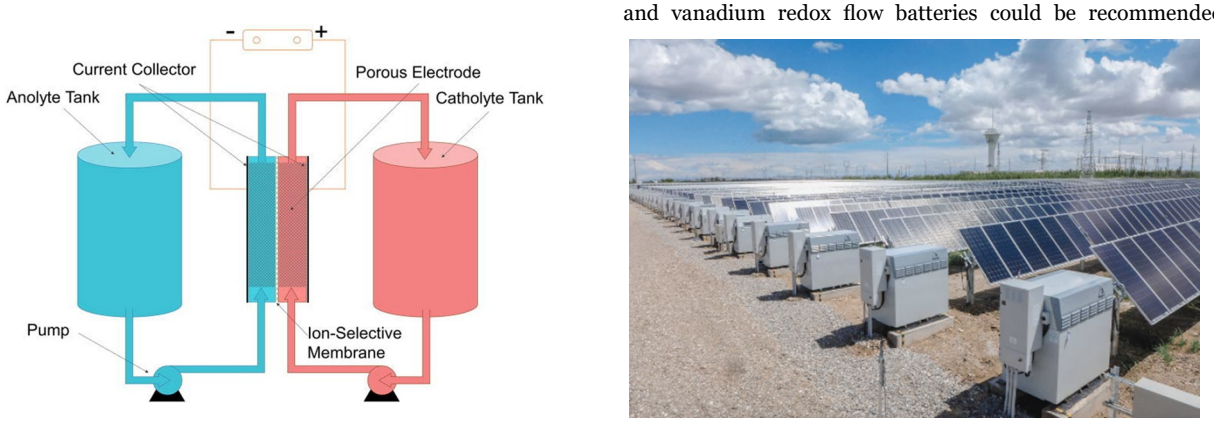


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Fig. 6. a) Operational principle of the redox flow battery b) Installation for the VRB greed storage of solar energy AVALON-USA (with permission of AVALON).

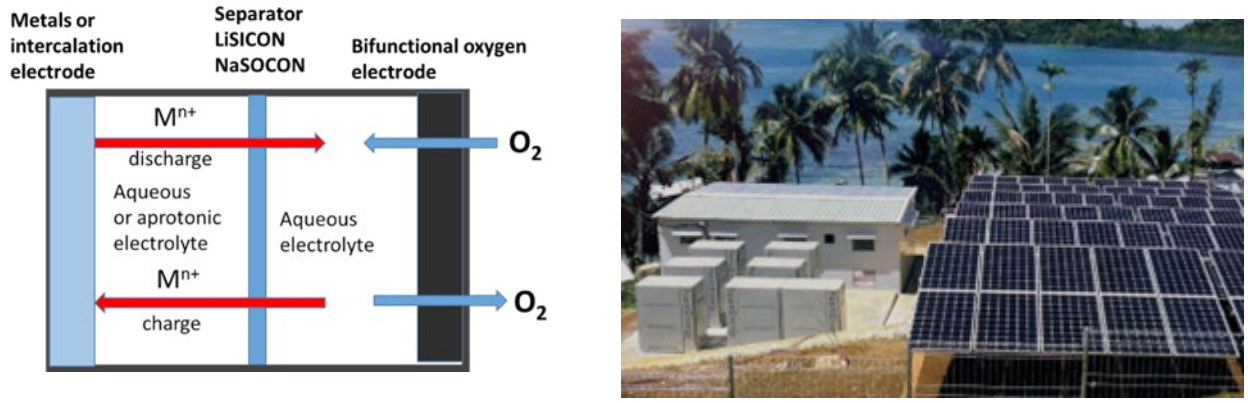

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Fig. 7. a) Principle of operation of the rechargeable metal air battery. b) Nantenergy zinc-air-battery for solar energy storage (with permission of Nantenergy, USA).

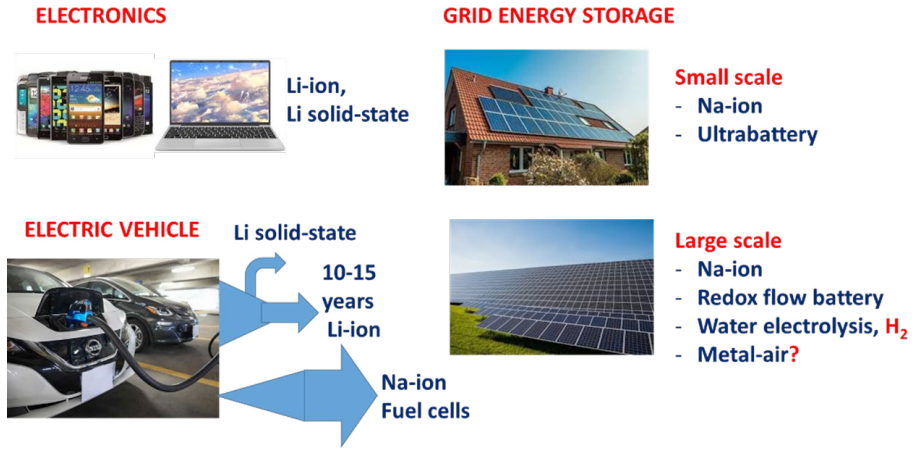

Fig. 7. Possibilities of electrochemical power sources applications today and in the future.

also considering hydrogen and metal-air batteries. For small-scale applications, the sodium-ion and lead-acid UltraBattery could be recommended, due to their low price considerable energy density, and long cycle life. Electric vehicles (EVs), will be likely powered over the next 10 to 15 years with improved Li-ion batteries. The Li-solid state batteries are likely a sidetrack due to their very high price, and many drawbacks. In the few years the growing applications of sodium-ion batteries will likely become a reality, and during the next decades, fuel cells and hydrogen may lead the race for EVs.

In the future, some other systems could arise that are today in R&D phases, but with improvement could have the potential to accomplish standards for grid energy storage.

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