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The rechargeable aluminum-ion battery with different composite cathodes: A review

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ABSTRACT

Digital cameras, laptop computers, cellular phones, as well as many portable electronic devices require batteries for powering. Based on the electrolyte type, electrolytic batteries can be categorized into solid-based, liquid-based, and ionic-based batteries. Aluminum ion batteries (AIBs) have some promising properties such as low cost, high safety, and high specific volumetric capacity. Nevertheless, in order for AIBs to be extensively used, developing novel electrode materials possessing high energy density is required. This is mainly dependent on the cathode materials. However, these cathode materials have some drawbacks such as structural decomposition, low battery capacity, low discharge voltage, and volume expansion resulting from the intercalation of large-sized ions. Therefore, future research might concentrate on the investigation of cheaper electrolyte and novel cathode materials for enhancement of energy density and working voltage. This review focuses on the recently developed cathodes, particularly, composite cathode materials, including graphite, CuS, V₂O₅, Li₃VO₄@C, VS₄/rGO, and Ni₃S₂/graphene.

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1. Introduction

Electricity supply enhances productivity and access to principal services [1]. Powering many electronic devices such as digital cameras, laptop computers, and cellular phones have raised the demand for batteries. Rapid response and long operation times are favored for the

development of high-power batteries with high energy densities. NiCd and Ni metal hydride batteries have been principally utilized [2]; Gaston Plante first developed lead-acid battery in 1859. Meanwhile, the second generation of batteries with long cycle life, high energy density, high power density has been investigated in the chemical power industry [3]. A novel battery for electronic vehicles was developed by Thomas A. Edison in 1900 and finally, he patented his Ni-iron battery in 1901 in

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the USA. This battery is regarded as his most commercially successful product. To fabricate the best commercial battery with the right material combination, he spent over 10 years, performed more than 50,000 experimental investigations, and withdrew the first fabricated version from the market [4].

The most extensively used batteries for electrochemical energy storage since the early 1990s are lithium-ion batteries consisting of a lithiated metal oxide cathode, a liquid electrolyte, and a carbonaceous anode [5]. Their popularity is due to some inherent advantages such as no environmental toxicity, no memory effect, and a long life span. However, by the dramatic increase in the energy storage demand, we are dealing with the limited source of lithium over the next decades [6]. Many solid electrolytes such as lithium phosphorus oxynitride (Lipon), $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$, Li_4SnS_4 , Li_3PS_4 , and $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$, possess a voltage window beyond 5 V, therefore, their decomposition does not occur under anodic current. Moreover, there is a minimal possibility of the dissolution of transition metals into the electrolyte when a solid electrolyte is used. Contrary to carbonate electrolytes, the majority of ceramic solid electrolytes are intrinsically inflammable [7].

Room temperature ionic liquids (RTILs) are attracting attention as green solvents. These materials are being used for new applications as electrolytes for electrochromic windows, electrochemical mechanical actuators, and numerical displays with dye-sensitized solar cells, light-emitting electrochemical cells, conducting polymers, and other devices. These new materials are revolutionizing various technological and scientific areas such as the battery field [2].

One of the post-lithium batteries is the aluminum-ion batteries (AIBs) that were first introduced by Tech Vision Electronics in 2017. It has been reported that the number of patents in this field showed a 29% growth from 2010 to 2016 while the patent filings began only in 2013 [8]. The reason for significant interest in AIBs is the abundance of Al and its even distribution in the earth [9, 10]. Additionally, a very high capacity of up to 8.05 Ah/cm^3 and 2.98 Ah/g can be obtained using multivalent metal ions transmitting internally in the AIBs [11]. During the electrochemical reaction, three-electron transfer is usually involved in an Al-based redox couple, leading to higher energy capacities and energy density [12].

In this article, we first focus on the description of the electrolyte batteries, their benefits, and their limitations. Then, batteries with ionic electrolyte, solid electrolyte, and liquid electrolyte are reviewed. Finally, the current state of knowledge in the field of AIBs with different composite cathodes is summarized [13].

2. Electrolytic batteries

Electrolytic batteries consist of the cathode, anode, and electrolyte. Oxides formed on metals provide passive layers on the surfaces of metals, which make them stable in contact with gas phases such as air or liquid phases such as electrolyte solutions. This also happens for the ion battery electrodes [14]. Therefore, both anode and cathode materials used in ion batteries do not show stability with respect to electrolyte solutions [15].

2.1 Batteries with liquid electrolyte

Recently, there have been many papers concentrating on the performance improvement of Li/S batteries [16, 17]. Nevertheless, a few research reports mentioned that the dissolution of lithium polysulfide in the lithium/sulfur cell has a significant effect on the performance of the cell. Polysulfides (PS) are the sulfur reduction intermediates [18–20]. Based on the general formula of $\text{Li}_2\text{Sn} \rightarrow m/8\text{S}_8 + \text{Li}_2\text{S}_{n-m}$, a series of complicated disproportionation occur for PS in the solution. Therefore,

low-order PS and the less soluble sulfur are formed leading to inactivation when it deposits into the pores of the separator or precipitates out of the liquid electrolyte [21]. The literature shows that by the incorporation of an ionic liquid or a solid salt of quaternary ammonium into the liquid electrolyte, the disproportionation of polysulfide intermediate can be suppressed [22]. This approach enhances cell capacity retention, but the redox shuttle of PS cannot be suppressed. Additionally, LiNO_3 can effectively suppress the PS redox shuttle and increase the charging efficiency of the cell; however, this effect is vanished by time due to the gradual consumption of LiNO_3 on the Li anode. It has also been observed that the rate capability of the upper voltage plateau is better compared to the lower voltage plateau because the solid-to-liquid phase transition is more favorable [23].

It has been indicated that room-temperature lithium/sulfur cells with organic liquid electrolytes use very low active material [24]. Various investigations have investigated the electrochemical properties of these cells with different electrolytes such as polymer/gel electrolytes [25–30], 1,2-dimethoxyethane (DME) [31], tetra (ethylene glycol) dimethyl ether (TEGDME) [26, 32–35], carbonate systems [36, 37], 1,3-dioxolane (DIOX) [31, 33], and tetrahydrofuran (THF). One of the promising organic solvents for room-temperature lithium/sulfur cells is tetramethylene glycol dimethyl ether (TEGDME). This gel/polymer or liquid electrolyte solvent provides a high first discharge capacity of more than 1200 mAh g^{-1} [27, 34, 35].

Cycle performance and discharge capacity of room-temperature lithium/sulfur cells containing polymer electrolytes and single/binary liquid electrolytes with LiCF_3SO_3 (1 M) were studied [38]. The cycle property and the cell's first discharge capacity were shown to be largely dependent on the liquid electrolyte content [39]. According to the investigations, at about 450 mAh g^{-1} , a medium containing electrolyte around $12 \mu\text{l}$ yields the stable and optimal cycle performance. Cycle performance of cells using different electrolytes including 1,2-dimethoxyethane/di(ethylene glycol)dimethyl ether (DEGDME) (1:1, v/v), TEGDME/DIOX (1:1, v/v), and TEGDME showed that the TEGDME-based mixed electrolytes had better results. An increase from 386 to 736 mAh g^{-1} was observed in the initial discharge capacity that stabilized the cycle properties by adding toluene 5 (vol.%) to TEGDME owing to the decrease in Li metal interfacial resistance [40]. In this study, cycle performance, Li metal interfacial resistance, and ionic conductivity in room-temperature lithium/sulfur cells containing TEGDME/DIOX polymeric electrolyte and poly(vinylidene fluoride) (PVDF) microporous membrane were evaluated [41]. Studies revealed the better performance of the liquid electrolyte in comparison with the polymer electrolyte [30].

Although advancements in the field of lithium/sulfur cells with the liquid electrolyte are significant challenges remain. The operation of these cells requires the dissolution of polysulfides and elemental sulfur in the organic liquid electrolytes [42]. PS and sulfur are electronically and ionically conductive resistance, therefore, their redox takes place on the conductive carbon surface in contact with the electrolyte [43]. Hence, the cells are considered as liquid electrochemical cells. However, two negative charges are carried by polysulfide anions, and it is required to maintain electric drag between the two electrodes during the cell function [44]. It has been proven that the outward diffusion of the dissolved polysulfide from the cathode can be effectively suppressed by sulfur-carbon composites [45]. However, as a result of the incorporation of extra electrochemically inactive carbon, the improvement is accompanied by the reduction of the energy density. Although a large amount of carbon is introduced by these composites, the lithium/sulfur cell is still a liquid cell [46]. The difference is that the absorption of the dissolved PS occurs on the carbon pores and surfaces. According to the electrical drag effect of polysulfide, protecting the lithium anode against the corrosive attack of the dissolved polysulfide and the redox shuttle occurs [47]. PS dissolution and the determination of the power capability of the cells

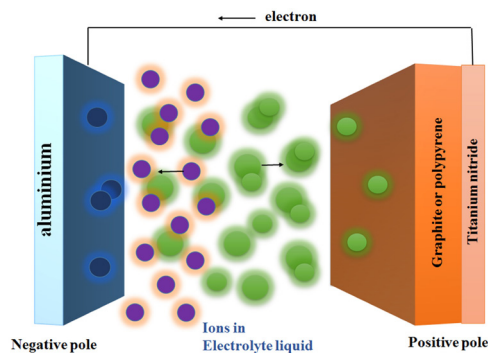


Fig. 1. Schematic illustration of batteries with liquid electrolyte.

are dependent on the liquid electrolyte. A facile approach to suppress the polysulfide redox shuttle and improve the Li deposition morphology is to incorporate additives to the electrolyte [48]. The low coulombic efficiency of lithium cycling could be solved by adding LiNO_3 , however, it undergoes gradual consumption on the Li anode and cathode and its strong oxidative property causes safety issues [49].

The advancement of Li/S technology needs safer and more effective additives. As a result of the high reactivity of PS anionic radicals, the chemical compatibility for selecting the cell materials such as cathode's binder, salt, and electrolyte solvent should be considered as a high priority [50]. Regardless of the taken approach, it is necessary for the cathode to possess a relatively high loading and content of sulfur to ensure the high energy density of the cell [17].

Different liquid electrolytes with high ionic conductivity including dimethyl carbonate (DMC)/ethylene carbonate (EC), propylene carbonate (PC), and EC/PC have been utilized in sodium-ion batteries [51–54]. Although sulfur electrodes are able to yield good ionic conductivity, the investigations about using these electrodes for Na batteries with liquid electrolytes are not extensive. At ambient temperature, a liquid electrolyte sodium ionic conductivity composed of NaCF_3SO_3 salt and TEGDME is $3.9 \times 10^{-3} \text{ S cm}^{-1}$, and it could be utilized for room-temperature sodium-sulfur batteries. Two regions are observed in the discharge curve: the first region has a decreasing trend from 2.23 to 1.66 V and the second region is a plateau at 1.66 V. The first discharge capacity of sodium-sulfur cells with liquid electrolytes [55] at room temperature (538 mAh g^{-1}) is lower compared to high-temperature counterparts. After a sharp decrease in the second cycle, the discharge capacity is stabilized at 250 mAh g^{-1} up to the 10th cycle, and Na_2S_2 and Na_2S_3 are the final discharge products. Sodium or sulfur polysulfides are dissolved into the electrolyte and sodium sulfides are reduced irreversibly to elemental S at full charge leading to a decrement of discharge capacity [56]. A schematic illustration of batteries with liquid electrolytes is shown in Fig. 1.

2.2 Batteries with solid electrolyte

Inorganic solid electrolytes are used in all-solid-state secondary batteries. These batteries are attracting attention due to their ionic conductivities and better safety compared to conventional batteries [57]. Besides conductivity, the enhancement of cycle performance and the energy density are highly affected by the mechanical characteristics of solid electrolytes [58]. High ionic conductivities of some new solid electrolytes including Na_3PS_4 , $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$, and $\text{Li}_3\text{P}_7\text{S}_{11}$ have been reported. The ionic conductivities of some of them are even close to those of liquid electrolytes [59]. The current issue regarding solid electrolytes is the improvement of their electrochemical and electrical properties [60]. The mechanical characteristics of these electrolytes such as densification behavior and elastic modulus are important factors for the development of all-solid-state batteries practically, however, few studies are focusing on the mechanical characteristics of solid electrolytes with high-ion-con-

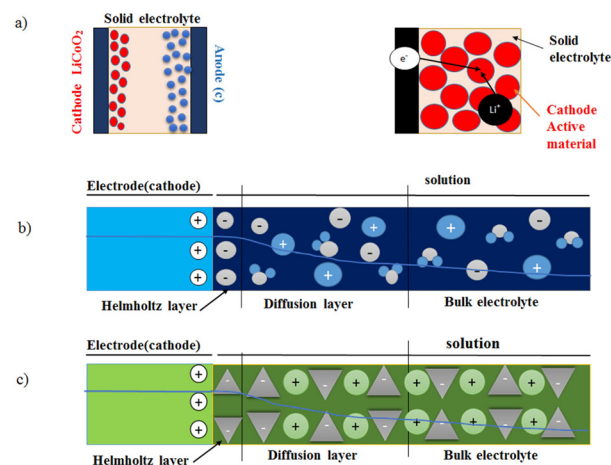


Fig. 2. Schematic illustration of (a) all-solid-state Li^+ ion battery (b) a typical solid-liquid interface, and (c) positive electrode-solid electrolyte interfaces, in which cations (green circles) and anions (gray triangles) form their networks and the anions are less mobile.

ductivity [61]. All solid-state Li^+ ion battery illustration is seen in Fig. 2.

Due to electrolyte decomposition upon the first cycle, a protecting layer, known as “solid electrolyte interphase (SEI)”, is generated on the negative electrode of Li-ion batteries. The quality of the SEI highly influences battery performance, safety, exfoliation of graphite, rate capability, and irreversible charge loss [62]. Thus, this is important to comprehend the composition and actual nature of SEI. By understanding the way each component influences the performance of the battery and the chemistry of the SEI formation, it is possible to tune SEI to improve battery performance [63]. Properties of SEI including morphology, compactness, thickness, and composition have significant effects on battery performance. A characteristic of SEI is irreversible charge loss (ICL) that occurs upon the first cycle, which is the result of the reduction of the solvent and SEI formation [58]. Moreover, the SEI ability to change the active material surface to a passive one can affect self-discharge, which is a detrimental process during storage. Therefore, SEI determines the shelf-life of a battery [64]. As SEI may evolve and/or dissolve during cycling, for good cycling life in the battery, stable and effective SEI is necessary [65] and its necessity is more significant at a deeper depth of discharge and during high-rate cycling [66]. SEI components show high sensitivity to temperature, therefore, they affect the low- and high-temperatures performance of the battery [67]. Nevertheless, SEI mostly influences the safety of the battery [68–70].

The dissolution of transition metals into the electrolyte could be minimized by solid electrolytes. In contrast to carbonate electrolytes, most ceramic solid electrolytes are inflammable, and the compatibility between Li and various solid electrolytes and Li metal is likely to create dendrites during cycling due to the mechanical robustness of the electrolytes [71]. In comparison with liquid electrolyte Li batteries, solid-state batteries have lower power densities. This is the result of limited kinetics of the electrodes, the compatibility of the electrode/electrolyte interface, and the solid electrolyte low ionic conductivity [72]. Recently, the development of high ionic-conductive solid electrolytes has made the production of solid-state Li batteries possible with a power performance similar to liquid-electrolyte batteries. However, there is a great challenge about interfacial instability between the electrolyte and electrode in these batteries. For most solid-state Li batteries, it is required to properly engineer the interfaces between electrode and electrolyte to yield good cycling performance [73, 74].

Owing to a moderate Young's modulus as well as low bond energy, sulfide solid electrolytes have good processability and high ionic conductivity. Room-temperature pressure sintering can be used for the fabrication of sulfide solid electrolytes. All-solid-state batteries, therefore,

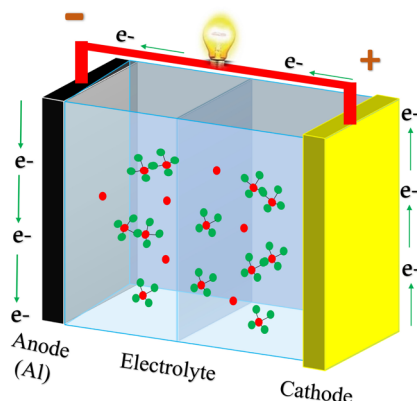


Fig. 3. Schematic illustration of batteries with ionic electrolyte.

can highly benefit from sulfide solid electrolytes. Some sulfide solid electrolytes have yielded conductivities comparable to practical Li-ion batteries [75]. It is necessary to obtain the ideal mechanical characteristics of all-solid-state batteries and fabricate novel solid electrolytes that have enhanced mechanical properties [61].

2.3 Batteries with ionic electrolyte

N-methyl-N-butyl-piperidinium bis(trifluoromethanesulfonyl imide (PP14-RTIL)) was prepared as a new room-temperature ionic electrolyte for lithium/sulfur cells. The cyclic voltammetry of this new electrolyte exhibited an extant potential window of 5.2 to 0.15 V as well as electrochemical stability, which make it proper for Li-S electrodes [76]. The viscosity of water is 0.89 cP at room temperature, while this value for ionic liquids is in the range of 30–50 cP and sometimes several hundreds of cP. When $[\text{Li}^+][\text{X}^-]$ salt is added to the neat ionic liquid of $[\text{A}^+][\text{X}^-]$, the formed $[\text{Li}^+]_m[\text{A}^+]_n[\text{X}^-]_{m+n}$ system has even higher viscosity [77]. Due to the high viscosity of ionic liquids, there are some problems with their handling. Electrodes involve active materials, a polymer binder, and an electron conductor (acetylene black). The electrolyte fills the volume between particles [15]. Pure ionic liquids show low conductivities and high viscosity. The addition of an organic dipolar aprotic solvent can reduce the viscosity of neat ionic liquids. A schematic illustration of batteries with ionic electrolytes is observed in Fig. 3. Diaw et al. [78] studied conductivities and viscosities of 1-butyl-4-methyl-pyridinium (Py^+) or 1-butyl-3-methylimidazolium (IMI^+) associated with hexafluorophosphate (PF_6^-) or tetrafluoroborate (BF_4^-) anions mixed with butyrolactone (BL) or acetonitrile (ACN) in the presence of lithium hexafluorophosphate or lithium tetrafluoroborate [79]. The mixture of organic solvents with pyridinium and imidazolium are relevant electrolytes for electrochemical systems. It was shown that the viscosity and conductivity of the electrolytes are higher than those of pure ionic liquids. They proposed that these electrolytes are promising for various electrochemical applications due to their low vapor pressure and good thermal stability [75].

The liquids in batteries make special pack sealing by themselves that is necessary for the prevention of liquid leakage and explosive volatiles. Moreover, reactions of lithium metal with liquid solvents lead to poor battery performance. The electrolyte ionic conductivity often limits high power [80]. Recently, ionic liquids with organic cations have been offered as electrolytes [81–83]. Vapor pressure of ionic liquids is negligible compared to conventional liquid solvents. However, Li batteries need the transportation of Li^+ cations between the cathode and anode. Therefore, doping with a suitable LiX salt is required for ionic liquids [81, 82]. Then the doped ionic liquid is incorporated in a thin, flexible membrane to be applicable for a battery electrolyte. Within the membranes, the Li^+ cation transport mechanism is changed by the presence of

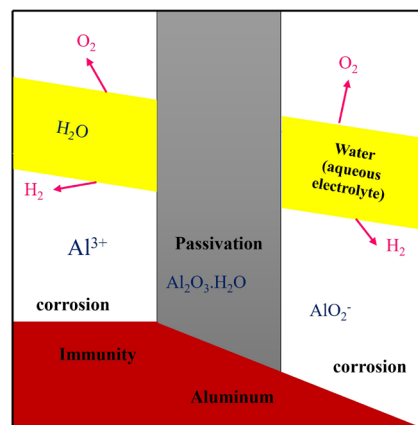


Fig. 4. Schematic of room-temperature corrosion behavior of aluminum in water.

an ionic liquid. This is an important issue because the conductivity can increase beyond the existing limits for dry polymer electrolytes offering promising possibilities for the long-sight in high energy lithium-metal-polymer batteries [2].

3. Types of aluminum-ion batteries according to different cathodes

3.1. Aluminum-ion batteries

Aluminum is one of the most abundant metals in the earth's crust [84–86]. Al may offer considerable safety enhancement and cost-saving in comparison with its counterparts due to its easier handling and lower reactivity. Furthermore, aluminum is one of the ideal elements that can be used in rechargeable batteries owing to its small electrochemical equivalent around 0.336 g Ah^{-1} and highly negative redox potential around $\approx 1.676 \text{ V}$ vs. standard hydrogen electrode [87]. On the other hand, there have been several issues in the past such as the effect of absence of discharge voltage plateaus on the capacitive behavior [69, 88] low discharge voltage [89], and the disintegration of disintegration [90, 91]. The surface of Al is easily oxidized and inert Al_2O_3 oxide films are generated that decreases the Al electrode potential rapidly. Al is an amphoteric metal with the chemical activity that tends to react with aqueous-medium. This explains why rechargeable aluminum-ion-batteries have not been industrialized [92, 93].

In recent works, some new electrode materials have been explored such as Prussian blue analogues (PBAs) [94], polymers [95], fluorinated natural graphite [96], TiO_2 [97, 98], and V_2O_5 [99], VO_2 [100], graphitic-foam, carbon, and sulfide materials [101, 102]. According to several studies, Al ions intercalate into or de-intercalate from the electrode materials as complex ions or they can have a redox reaction with active materials of the positive electrode [3, 103].

Al deposition occurs at potentials that are beyond the water stability region, therefore, using Al as a negative electrode in the presence of an aqueous electrolyte in a secondary aluminum-ion battery is not feasible [104]. In these potentials, decomposition of the electrolyte occurs and ion transport gets disrupted [105]. Calculated theoretical specific energies and specific energies in real systems have reached $1,090 \text{ Wh/kg}$ and 200 Wh/kg , respectively, which both are far less than the value for pure aluminum. By using a mixing system and a complex electrolyte supply, such batteries are utilized in the marine sector [106]. Because the Al electrode inherently generates hydrogen, it is required to design batteries with a different design in which metallic Al is not used [107]. Holland et al. [108] used TiO_2 as the negative electrode, copper-hexacyanoferrate (CuHCF) as the positive electrode, and an electrolyte of KCl and AlCl_3

aqueous solution. Fig. 4 shows the schematic of the room-temperature corrosion behavior of aluminum in water. According to the authors, the Al^{3+} ions are the mobile species, and the obtained discharge voltage measured to be 1.5 V. Furthermore, at a specific power of 300 W/kg, the specific energy was reported to be 15 Wh/kg and for over 1,750 cycles, energy efficiency remained above 70%. There is a limitation for the overall reachable energy density in these cells due to using a negative electrode that consists of other materials than Al [4].

Al-air batteries consist of a negative electrode of Al, a positive electrode that enables transportation and reduction of oxygen, and an electrolyte consisting of alkaline solutions of sodium chloride (NaCl), potassium hydroxide (KOH), or sodium hydroxide (NaOH) [109]. Rechargeable batteries with non-aqueous electrolytes like ionic liquids have been also reported. These batteries have some problems such as short shelf life, lack of rechargeability, sluggish discharge kinetics, a high self-discharge rate, and the corrosion of aluminum [38]. In the case of the passivation regime, the cell voltage and cell efficiency decreased due to the oxide layer on the Al electrode surface [110].

Secondary Al batteries usually consist of a negative electrode of Al and a non-aqueous ionic liquid electrolyte based on chloroaluminate [111]. Since 1988, these electrolytes have gained attention, when imidazolium chloride and AlCl_3 were utilized because of comparably wide electrochemical windows and their low vapor pressure which enable plating efficiencies and highly reversible stripping of aluminum. The same electrolyte was used for secondary aluminum batteries in subsequent studies due to the high success in Li-ion batteries [112].

For using the full potential of the Al-ion batteries, the negative electrode should be composed of pure Al. On the other hand, the protective oxide layer formed on the surface of this metal has an adverse effect on battery performance [113]. This is because the reversible electrode potential would not be obtained and there would be a delay in the activation of the electrode [105]. The enhancement of the electrode potential leads to accelerated corrosion in the liquid electrolyte (parasitic corrosion reaction occurs leading to the consumption of the electrode and evolution of hydrogen) and less shelf life. This happens for an Al battery with a liquid electrolyte. To reduce the parasitic corrosion of the Al electrodes, the deposition of other oxide layers and additions to the liquid electrolytes have been practiced [114]. Some research has reported the advantages of the oxide layer namely the restriction of strong surface corrosion and the growth of crystalline Al dendrites resulting in the improvement of the cycling stability of Al batteries [4].

In solid electrolytes, the Al electrode surface is of great importance. Before joining to the solid electrolyte, the oxide surface layer should be removed from the Al electrode surface. In an alternative way, a physical vapor deposition technique can be used for Al deposition or liquefied aluminum infiltration in the solid electrolyte [115]. Due to environmental benignity, low cost, and high energy density, Al is considered as a suitable electrode candidate for rechargeable batteries. However, the Al primary and secondary batteries have not found a way into the market. In the Al primary battery, the voltage is lower than the theoretical value. This behavior is the result of a protective layer on Al that delays the voltage recovery phenomenon. In the Al secondary battery, operation at high temperatures is needed because of the restrictions of some high-temperature molten salts in electrolytes [116].

To meet the techno-economic requirements, positive electrodes should have specific properties. The energy density of oxide materials is close to the upper boundary. The positive electrode has the open-circuit voltage (OCV) of around 2.5 V and the positive electrode is required to possess a density above 4 g/cm^3 for meeting the requirement of, for example, the United States Advanced Battery Consortium goals [117].

3.2 Aluminum-ion batteries with different cathodes

According to Rani et al. [26], to obtain high energy density in Al-ion batteries, the cathode is required to release and accommodate the Al^{3+} ions during charge/discharge. Cathode materials such as V_2O_5 nano-wire and spinel $\lambda\text{-Mn}_2\text{O}_4$ showed promising results in rechargeable aluminum-ion batteries with an aluminum ion conducting ionic liquid electrolyte. There are extensive studies about graphite fluorides as cathode materials in lithium batteries [29, 33, 118-120] and to a certain extent in Al and Mg batteries [81]. For the first time, an Al-ion battery consisting of AlCl_3 and imidazolium-based ionic liquid electrolyte, fluorinated natural graphite cathode, and an aluminum anode is reported. Non-covalent C-F bonds are formed in fluorinated natural graphite prepared by the electrochemical method. The discharge capacity of 225 mAh g^{-1} as well as very stable electrochemical behavior was observed during the cycle life studies of these batteries [121].

In a study by Reed et al. [27], the electrochemistry of CuHCF as a cathode material for Al ions during cycling with aqueous Mg^{2+} is described in detail. The electrolyte was composed of aluminum triflate that was dissolved in diethylene glycol dimethyl ether (diglyme). They presented reversible Al intercalation and deintercalation from an organic electrolyte into CuHCF. The study indicated the intercalating behavior of the Al-solute complex [122]. The results showed that the initial discharge capacities of the system were as high as 60 mAh/g . Furthermore, reversible capacities were found to be in the range of 5 and 14 mAh/g , and capacity typically faded after 10 to 15 cycles. The redox activity of $\text{Cu}^+/\text{Cu}^{2+}$ and $\text{Fe}^{2+}/\text{Fe}^{3+}$ was observed in cyclic voltammograms. It was proposed that the third redox peak might be the result of the $\text{Fe}^{3+}/\text{Fe}^{4+}$ redox couple. According to Gaussian calculations and capacity analysis, an Al/diglyme complex intercalated into defect sites in the CuHCF more than a free aluminum ion resulting in low reversible capacity.

Anatase TiO_2 nanotube arrays were incorporated in an aqueous solution of AlCl_3 and the electrochemical aluminum storage was investigated by Liu et al. [28]. The metallic Ti foil substrate was anodized and calcinated to prepare anatase TiO_2 nanotube arrays. It was shown that Al^{3+} ions are able to be reversibly inserted into and extracted from anatase nanotube arrays of TiO_2 in the presence of aqueous solution owing to the Al ion small radius steric influence. The diffusion of the solid phase into the anatase TiO_2 nanotube arrays controls the aluminum ion insertion. This research is a guide for the fabrication of new intercalation materials for using in aqueous rechargeable batteries based on small radius, multi-valent ions [123].

Hu et al. [29] reported a new cathode material for Al-ion batteries based on a self-standing and binder-free cobalt sulfide encapsulated in carbon nanotubes. They reported that the novel electrode materials showed very high cycling stability, improved rate performance of 154 mAh g^{-1} at 1 A g^{-1} , and a high discharge capacity of about 315 mAh g^{-1} at 100 mA g^{-1} . Its discharge capacity was stabilized at 87 mAh g^{-1} at 1 A g^{-1} after 6000 cycles. The material disintegrations and side reactions were suppressed by the electrode freestanding property. Based on this investigation, the design of high-performance cathode materials for flexible as well as scalable Al-ion batteries is possible [124].

Wang et al. [30] developed an aqueous rechargeable Zn/Al ion battery in which the positive electrode was ultrathin graphite nanosheets, the negative electrode was zinc, and the electrolyte was $\text{Al}_2(\text{SO}_4)_3/\text{Zn}(\text{CHCOO})_2$. The preparation method for the positive electrode was the electrochemical expansion in aqueous solution. The raw materials for the preparation of the Zn negative electrode and the aqueous electrolyte are abundant and the cost is low [125]. The average operating voltage of this rechargeable battery (1.0 V) was reported to be higher compared to most rechargeable AIBs with ionic liquid electrolytes. Furthermore, the battery maintained a high capacity and was able to be rapidly charged in 2 min. It was also capable of retention of capacity (94%) after 200 cycles showing its good cycling behavior [126].

As a result of natural abundance and high electrochemical activity,

Wei et al. [32] used MoO_2 as a host material for Li storage. The ionic liquid electrolyte was 1-ethyl-3-methylimidazolium chloride/ AlCl_3 and the cathode was prepared by fabrication of a dense MoO_2 layer on nickel foam. It was observed that the discharge potential was 1.9 V that is indicated to be higher than most metal oxide cathodes of AIBs studied so far. Moreover, at 100 mA g^{-1} , it exhibited a specific discharge capacity of 90 mAh g^{-1} . However, after long cycling, the dissolution of molybdenum oxide and its transfer to the separator lead to a rapid capacity decay [127].

3.3 Aluminum-ion batteries with composite cathodes

A new AIB based on intercalation and deintercalation of Al^{3+} was developed by Wang et al. [6]. In this battery, the anode was Al foil, the cathode material was Ni_3S_2 /graphene micro flakes composite, and the ionic liquid electrolyte was 1-ethyl-3-methylimidazolium chloride/ AlCl_3 . A high discharge voltage plateau was observed in the battery ($\approx 1.0 \text{ V}$ vs. $\text{Al}/\text{AlCl}_4^+$) providing a remarkable step forward in developing AIBs.

Zhang et al. [3] used a composite of VS_4 and reduced graphene oxide (rGO) for the AIB cathode with ionic liquid electrolytes. At 100 mA g^{-1} , the initial charge specific capacity and discharge specific capacity were reported to approach 491.57 and $406.94 \text{ mAh g}^{-1}$, respectively. High-retained capacity and coulombic efficiency above 90% after 100 cycles are the indicators of the suitability of the cathode material for new rechargeable Al-ion batteries.

The microsphere composite of $\text{Li}_3\text{VO}_4/\text{C}$ was developed by Jiang et al. [128] as a new cathode material used in AIBs. According to the results, the battery initial discharge capacity was 137 mAh g^{-1} , and the retained capacity upon 100 cycles was measured to be 48 mAh g^{-1} with coulombic efficiency of almost 100%. It was observed that metallic aluminum was dissolved and deposited in the anode and Al^{3+} cations inserted and de-inserted in the cathode. These fabricated rechargeable AIBs had low cost and high safety offering them suitable cathode materials in acidic ionic liquid electrolyte systems.

The rechargeable AIB prepared by Wang et al. [129] was composed of an ionic liquid electrolyte of 1-ethyl-3-methylimidazolium chloride/ AlCl_3 and a cathode of three-dimensional hierarchical CuS microsphere composed of nanoflakes. The battery demonstrated a coulombic efficiency of approximately 100% after 100 cycles, an average discharge voltage of nearly 1.0 V vs. $\text{Al}/\text{AlCl}_4^+$, and a reversible specific capacity of 90 mAh g^{-1} at 20 mA g^{-1} . The well-defined cathode nanostructure facilitates the ion/electron transfer, particularly for large-sized chloroaluminate ions resulting in remarkable electrochemical performance.

The use of natural graphite for the cathode for AIBs was reported by Wei et al. [130]. Nevertheless, the natural graphite lifetime is short due to some problems such as severe volume swelling. Therefore, the authors deposited an amorphous carbon on a graphite paper. According to the charge/discharge outcomes, the cycle life of the composite was higher than that of the graphite paper. It was also indicated that the enhancement of the cycling stability was mostly attributed to the graphite paper and amorphous carbon interface.

In a study by Chiku et al. [131], a composite of carbon/ V_2O_5 was prepared for the positive electrode of AIB. The electrolyte solution was a mix of toluene, aluminum chloride, and dipropylsulfone with a mass ratio of 1:10:5. The amorphous V_2O_5 exhibited a reversible oxidation/reduction reaction in the electrolyte. The rechargeable AIB with the positive electrode of V_2O_5 /carbon exhibited a discharge capacity of more than 200 mA g^{-1} with the C/40 discharge rate.

The aluminum secondary battery developed by Uemura et al. [132] was composed of ionic liquid electrolytes of 1-ethyl-3-methylimidazolium chloride/ AlCl_3 , aluminum metal anode, and a cathode of graphene nanoplatelet composite, which was fabricated by a slurry-coating route.

The battery showed capacity retention of around 66 % at 6000 mA g^{-1} , and a reversible capacity of 70 mAh g^{-1} at 2000 mA g^{-1} . At 2000 mA g^{-1} , a coulombic efficiency of 99% was obtained up to 3000 cycles.

4. Conclusions and future insights

This paper first summarized the types of electrolytic batteries in terms of the electrolyte type including solid, liquid, and ionic electrolytes. Then, due to the advantages of Al-ion batteries including excellent reversibility, high security, and low cost, more details are presented about these batteries. Some AIB cathodes including composite cathodes were introduced. A new trend for AIB development seems to be composite materials such as graphite, CuS, V_2O_5 , $\text{Li}_3\text{VO}_4/\text{C}$, VS_4/rGO , and $\text{Ni}_3\text{S}_2/\text{graphene}$. However, such materials still have some drawbacks such as structural decomposition, low battery capacity, low discharge voltage, and volume expansion resulting from the embedment of large size intercalation ions [133]. The AIB development is still in primary stages; however, there are clear directions for forthcoming research. Future investigations might concentrate on seeking cheaper electrolytes and novel cathode materials to promote energy density and working voltage. Laptops and smartphones need high energy density, thus the application of AIBs in these devices is not feasible at present. On the other hand, AIBs can offer high power and fast charge transfer to balance the electricity supply, which is required for the electricity grid. AIBs can play a pivotal role in production and life in the future if their electrochemical performance is remarkably improved [112].

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Conflict of Interest

All authors declare no conflicts of interest in this paper.

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