Recent advancements to mitigate zinc oxide formation in zinc-air batteries: A technical review

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ABSTRACT

Application of zinc metal in the field of electrochemistry has always been of specific interest as it is a favored material for high energy density batteries and sacrificial electrodes to guard the other metallic components against corrosion. Some other factors, beside a high energy density, such as its low cost, ease of handling, nontoxicity, and abundance of zinc, has made this metal receive a considerable attention by researchers. In the past decades, considerable efforts have been dedicated to discovering and emerging electrically rechargeable zinc-based batteries in a response to the growing request in energy consumption in the fast-growing market of electronic devices and automotive business. Nevertheless, regardless of the developments made in the material science and the cell design, only a slight breakthrough has been attained in developing a system capable of substituting other viable rechargeable batteries. Passivation is one of the most challenging obstacles in zinc air batteries (ZABs) for commercialization. Recently, several developments have been executed to alleviate the passivation of zinc anode in the ZABs. This review has meticulously surveyed different aspects of this issue as well as recent developments for mitigating the zinc anode corrosion and passivation.

1. Introduction

Sustainable energy technologies as an imperative alternative for fossil fuels and net zero emissions with several challenges have attracted commercial companies [1–4]. Among energy storages, as an efficient approach toward the next-generation electrochemical energy storage, the metal−air batteries yield more benefits owing to high theoretical energy densities and free air source for cathode, and this has led to much more attention compared to the lithium-ion batteries [5–7]. High energy density and less-inherent issues are the most imperative parameters to achieve highly efficient metal-air batteries [8]. Among various metals, due to its low cost, high energy density, safety, and environmental friendliness, zinc as a promising metal has won a considerable agree-ableness, being extensively applied in various types of batteries e.g. zinc ion, zinc-air, zinc-bromine, zinc-iodine, and zinc-nickel [9–17]. The Oxygen (O_2) from free the air atmosphere as an inexhaustible source facilitates the cathode process during the cycling (charge/discharge)

process. The primary type of ZABs based on the discharge process have been successfully implemented commercially since 1930 [18]. Various types of cathode materials have been fabricated to develop rechargeable zinc batteries in order to facilitate the commercialization process of the secondary zinc batteries [19–24]. Nonetheless, there are several issues not being fully resolved via many modifications in the anode structure, electrolyte, separator and electrocatalysts [25]. Thanks to the high emergent energy demand in advanced technologies, a considerable volume of emphasis has been laid on improving the secondary zinc-air batteries due to the low power density and poor stability in the cycling process [26]. Many routes have been developed to overcome several issues with the intention of achieving a high stability and a long-life cycle but controlling the zinc oxide formation has not been still improved remarkably in comparison with the other issues [27–30].

Nowadays, efficient metals-based electrocatalysts, 3D zinc anode, and high ionic conductive electrolytes with fewer issues have been significantly developed to attain stable and long cycle life batteries.

However, the commercialization in larger markets are restricted by the dendrite formation, shaped change, hydrogen evolution gases, internal resistance, passivation, low coulombic efficiency and the like [31,32]. Thus, the practical large-scale applications of the batteries are feasible via overcoming unconquerable limitations and realizing the strength of the battery's chemistry. The firm concept of key variables contributes to enhancement of the ZABs performance. The formation of dendrite at zinc anode during the charging process at a high current density as a major drawback affects the battery cycle life [33,34]. The dendrite growth is an uncontrollable incident due to electrochemical reaction kinetics, ion diffusion, and crystal nucleation at a high-rate current density leading to critical impacts exerted on the zinc deposition process. A rapid zinc deposition leads to needle-shaped zinc structures on the anode surface that is mainly dependent on the overpotential and surface energy anisotropy. The dendrites get constricted in the discharging period and continue to re-grow by further charging cycles leading to internal short circuits and consequently battery failure. The shape change as undesired phenomena alleviates the performance cycling due to the uneven zinc deposition leading to the change zinc volume [35,36]. This undesired circumstance occurs when zincate and Zn^{2+} ions are lumpy redeposited on different locations of the anode electrode during the charge process [37]. Concentration of the alkaline electrolyte effect on zinc shape change due to high concentration of alkaline solution easily dissolves ZnO (the discharge product) and increases Zn^{2+} ions, resulting in redeposits under non-uniform conditions [38]. Electrolyte modification and anode engineering are customarily proposed to minimize the uneven zinc deposition during the charging process.

The reaction between zinc and electrolyte generates parasitic corrosion leading to a reduction in the coulombic efficiency and zinc utilization because the H_2 evolution reaction consumes a part of the electrons provided to the Zn electrode. Other disadvantages stem from the H_2 evolution which causes swelling of the battery causing the electrolyte to crack and dry out while the accumulation of hydrogen evolved in H_2O reduction may enhance the charging voltage in the consecutive cycling. The rate of H_2 evolution depends on the electrolyte concentration and the current density. H_2 evolution may be suppressed by the electrolyte adjustments e.g. water-in-salt instead of salt in water [39]. Some issues due to the inherent behavior of zinc in the alkaline solution have been exhibited in Fig. 1.

The real energy density of ZABs in operation is 220–300 W h kg^{-1}, lagging well behind the theoretical value (1086 W h kg^{-1}), which remains as a barrier to achieve, requiring major development efforts. One of the significant reasons behind the distinction among the theoretical and practical energy densities is the low rate of zinc utilization, which is mainly on account of the passivation of zinc electrodes in alkaline electrolytes [25]. Zinc oxide (ZnO) formation as an undesired reaction is still an issue owing to supersaturated zincate ions (high rate of zinc dissolving, non-efficient electrocatalyst, etc.) that have to be addressed. Notwithstanding very efficient suggestions for improving rechargeable ZABs, passivation (ZnO layer) is still discerned even in the 3D anode design (especially in high depth of discharge) since the larger exposed area for contacting electrolyte/electrode enhances the zinc dissolution significantly [40]. Despite a plethora of investigations undertaken in relation to the dendrite and shape change, inadequate technical reports corresponding to zinc oxide formation and feasible solutions have been proposed hitherto. Bearing all this in mind, this review investigates the influence of various suggestions in two parts; electrolyte additives and novel zinc anodes in order to reduce the ZnO formation and elevate the battery performance. By knowing the critical parameters, deposition of
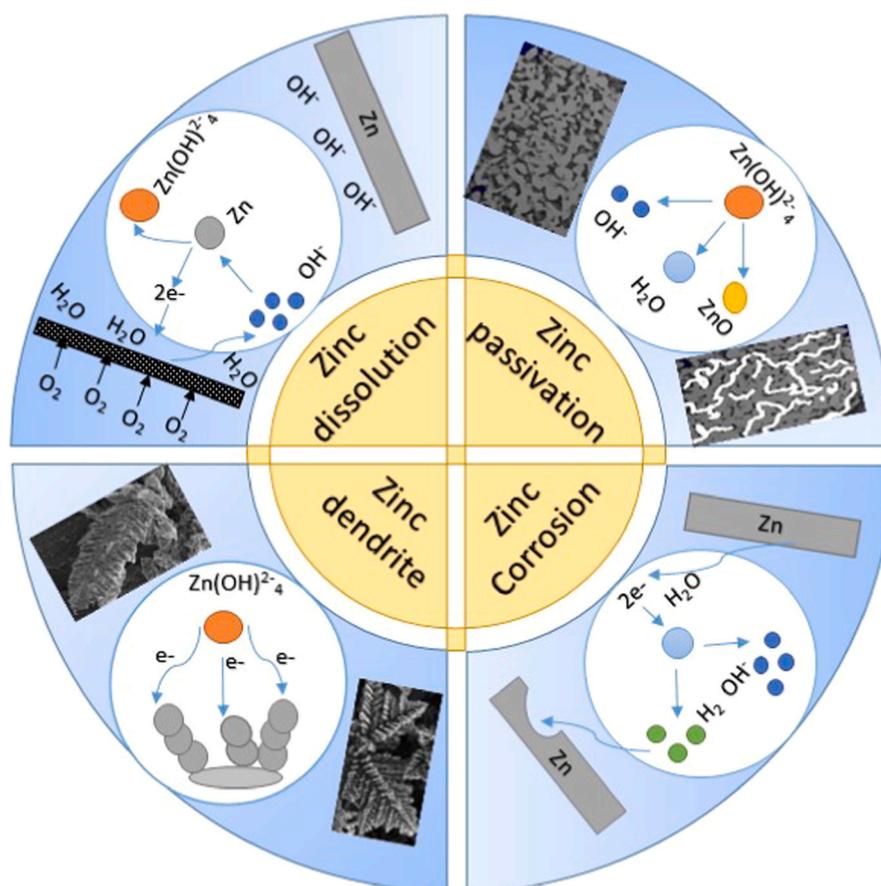
Fig. 1. Several challenges of ZABs in the aqueous electrolyte.

zinc oxide as an insulate layer over the anode surface or between dendrite layers can reach minimum values.

2. General overview of zinc-air batteries

The metal air battery was exhibited by Leclanche [136] using MnO_2/carbon as an air electrode [41]. The metal-air batteries with various anodes and air cathodes were further developed to achieve a commercial scale [42–44]. The primary metal-air batteries bear a low cost in terms of electrode materials, while having a long cell life and being environmentally safe [45]. The primary ZABs reached a commercialized process in 1930s [46]. Incomparable high energy densities of metal air batteries with other electrochemical energy storages have urged a considerable body of research to make endeavors in commercialization of the secondary type of metal-air batteries owing to their resource limitation and longer life. The specific energy density of the metal-air batteries varies for different anode metals. The highest specific energies of metal air batteries are in order of Li-air (5928 W h kg^{-1}) > Al-air (5779 W h kg^{-1}) > Mg-air (5238 W h kg^{-1}) > Na-air (2466 W h kg^{-1}) > Zn-air (1218 W h kg^{-1}) > K-air (1187 W h kg^{-1}) > Fe-air (1080 W h kg^{-1}) [47]. However, the functional efficiency of these batteries is determined by the intrinsic properties of the metals. Despite of the high specific energies of Li-air, Al-air, Mg-air, and Na-air, not an efficacious performance was yielded for the secondary type of batteries due to the corrosion (Al) [48], the air oxidation (Li, Na) [49,50], and, the non-recyclable metal (Mg, Al) [51]. ZABs with lower specific performance in compared to these metals indicated a successful rechargeable battery but commercialization of the ZABs encounters challenges concerning such unsolved issues as the dendrite, the shape change, and the zinc oxide layer [14]. ZABs are already launched in the market in form of coin-cell (hearing device) [45].

Pure zinc as the anode material with the oxidation process during discharge provides the electron source and improves the reaction efficiency via the suitable design of the anode leading to an increase in the battery performance. Modifying the zinc anode (coating, alloying, etc.) contributes to the control zinc dissolution reducing the effect associated with several issues. ZABs operate in alkaline electrolytes (such as KOH) due to high conductivity (73.5 $\Omega^{-1}cm^2$/equiv.) [10]. Zinc is highly desirable for its electropositivity and stability in alkaline electrolytes. The optimum alkaline concentration improves the battery performance. High concentration of the electrolyte minimizes the resistance contributing to an enhancement in both viscosity and ZnO formation while the low concentration electrolyte causes an undesirable hydrogen evaluation reaction. The optimum KOH concentration has been reported to be approximately 6 M with maximum ionic conductivity [52]. Modifying the electrolytes (additives, gels, solid polymers, ionic liquids, etc.) is also proposed by various researchers to overcome or decrease dendrite formation, corrosion, and ZnO formation. A separator selectively transports the OH^- group through permeable pores. For that reason, an ideal separator requires a stability in the alkaline solution, an appropriate pore texture, an insulator and high ion conductivity. An ideal separator for ZABs needs to be fine in order to prevent migration of the zincate ions towards the cathode side. For this reason, with the aim of improving the secondary ZABs, there is a dire need for a sufficient strength during the cycling process, high electrolyte absorption, as well as adequate fine pore to minimize the zincate ions transfer [53].

3. Zinc oxide

3.1. Chemical structure and characteristics

Being categorized in the II–VI semiconductor materials group, zinc oxide (ZnO) is an industrially significant compound with multifunctional technical and industrial applications in various fields including transparent electronics [54–56] and photovoltaics [57–59]. The reason why ZnO is considered to be advantageous for application of semiconductor and electrochemical devices lies in its unique characteristics including its various structures, visible range transparency, resistivity control over a wide range (10^{-3} to 10^5 cm), and high direct band gap and electrochemical stability. Even though ZnO is almost insoluble in H_2O and is an amphoteric oxide, it dissolves in almost all acids such as HCl (Hydrochloric acid) [60]:

$$ZnO + 2\ HCl \rightarrow ZnCl_2 + H_2O$$
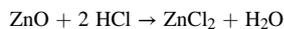
According to the following reaction, it also dissolves in alkalies to give soluble zincates:

$$ZnO + 2\ NaOH + H_2O \rightarrow Na_2[Zn(OH)_4]$$
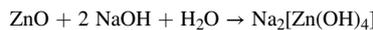
Cubic zinc blends and hexagonal wurtzite are the two main forms of ZnO crystals. The most common and most stable form in the ambient temperature is the wurtzite structure [61]. However, it is possible to stabilize the zincblende shape by increasing ZnO on substrates with a cubic lattice structure. In both cases, the centers of zinc and oxide are tetrahedral, which is the most distinctive geometry for Zn (II). A higher molar volume in comparison with zinc metal (due to its hexagonal wurtzite structure) is the main reason for drastic effects of the deposited layers of ZnO on the anode surface, from the structural point of view. Besides, the hexagonal wurtzite structure of ZnO expands its volume around 1.6 times higher than the metallic zinc and hence its deposition on the anode surfaces may cause surface shape changes and passivation [62].

3.2. Electrical characteristics

ZnO has a relatively large direct band gap of ~3.3 eV at room temperature. Advantages associated with a large band gap include higher breakdown voltages, ability to sustain large electric fields, lower electronic noise, high-temperature, and high-power operation [63]. The band gap of ZnO can further be tuned to ~3–4 eV by its alloying with magnesium oxide or cadmium oxide [16].

Electron mobility of ZnO strongly varies with temperature having a maximum of ~2000 cm^2/(V·s) at 80 K [34]. Data on hole mobility are scarce with values falling in the range of 5–30 cm^2/(V·s). A comparatively large direct band gap (~3.3 eV at 25 °C) of zinc oxide and its low electron mobility at ambient temperature are the main reasons to outstand zinc oxide as a passivation layer in ZABs.

3.3. Mechanism for zinc oxide formation in zn-air batteries

The low usage rate of zinc, which is due to the zinc oxide production in alkaline electrolytes, is one of the main motives for the variance among the calculated energy density and the practical energy density. In the Zinc-Air batteries, primarily, pure Zn dissolves from the onset of immersion via a chemical reaction as follows:

$$Zn \rightarrow Zn^{2+} + 2e^-$$
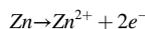
Concurrently, oxygen reduction occurs near the zinc surface:

$$O_2 + H_2O + 2e^- \rightarrow 4OH^-$$
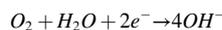
Zn^{2+} ions from the substrate can then react with OH^- to form Zn $(OH)_2$:

$$Zn^{2+} + 2OH^- \rightarrow Zn(OH)_2$$

Extending the immersion time results in maturing and thickening of the accumulated $Zn(OH)_2$/ZnO layer on the whole zinc surface which contributes to the inhibition of Zn dissolution. With the reaction constantly happening, there is an escalation in the pH value. Pourbaix diagram can be used for the speciation of zinc in different pH values (Fig. 2). In another word, this diagram illustrates the reaction pathways and potential problems of Zn anodes in alkaline/acidic environments by
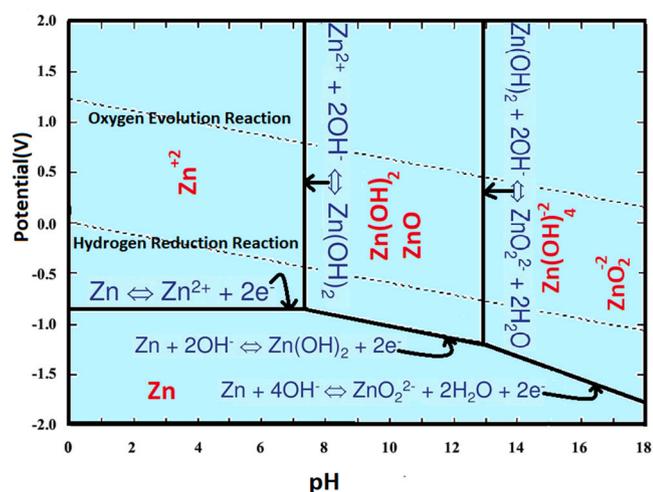
Fig. 2. Pourbaix diagram for zinc corresponding to different pH values in water.

showing the various products that are formed due to change pH of value. The dashed lines indicate two possible cathodic reactions, hydrogen reduction reaction (water in equilibrium with gaseous hydrogen) and oxygen evolution reaction (oxygen dissolved in water in equilibrium with water), respectively. According to the Pourbaix diagram, by elevating the pH, a portion of $Zn(OH)_2$ and Zn would react with OH^- to form $Zn(OH)_4^{2-}$.

$$Zn(s) + \quad 4OH^- \quad \leftrightarrow Zn(OH)_4^{2-}(aq) + \quad 2e^-$$
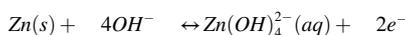
Finally, parts of the produced $Zn(OH)_2$ and $Zn(OH)_4^{-2}$ can be dehydrated in order to form ZnO via the subsequent chemical reactions:

$$Zn(OH)_4^{2-} \leftrightarrow ZnO(s) + 2OH^- + H_2O$$

$$Zn(OH)_2 \rightarrow ZnO + H_2O$$
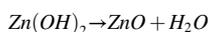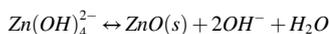
According to the literature, two types of passivation film can be detected in ZABs, namely types I and II [64–66]. The succeeding section elaborates on various interpretations for formation of passivated films on the surface of a zinc anode in ZABs.

3.4. Main challenges of zinc oxide production in zinc-air batteries

Supersaturation of zinc oxide in alkaline electrolytes and its deposition on the zinc anode surface is called passivation. The formed ZnO layer blocks the active surface of the anode, eliminating its further reactions. Yet, the insulated zinc oxide film is disadvantaged by elimination of the battery's capacity, utilization reduction of the zinc metal, and termination of the discharge process [67]. Furthermore, passivation prevents the reverse conversion of the Zn metal, eliminating the battery rechargeability. Fig. 3 represents a schematic of a zinc button cell and the produced ZnO on the zinc electrode.

From the Fig. 3a, it can be perceived that air (in the bottom part) flows through the gas diffusion electrode, dissolving in the electrolyte and catalyzing oxygen reaction. Moreover, most of the cell volume is accessible for negative electrode (gray color). In the case of a fully charged state, the space contains a porous zinc powder as anode. A void space was observed in the top part below the negative pole. This space was devoted to compensates for expanding the volume from zinc-to-zinc oxide during discharge. Fig. 3b illustrates some tomographic imaging that was reported at various state-of-charges during discharge to reach the cut-off potential 0.6 V under steady-state condition about 1.5 h.

Generally, there are two types of ZnO layers, namely type I and type II. Dissolution-precipitation mechanism is the approved method to form the type I layer and the dissolution of Zn is the rate determining step [16]. This type of ZnO layer, which is porous, limits the diffusion of hydroxide ions to the surface of the zinc anode implying that the OH^- concentration diminishes accordingly at the interface of zinc anode/electrolyte. Type II of ZnO which is an irreversible thick/compact layer, is formed through inducing the changes in the surface pH. Nucleation and growth are the nominated models for the type II ZnO layer [68]. The latter zinc oxide layer passivates the Zn anode, even though its formation could be moderately prevented by regulating the depth of discharge (DoD) or the discharge cell cut-off voltage [69,70]. The total time (t) required for a zinc anode to be fully passivated is the sum of the required time for the porous zinc oxide layers (type I) formation (tb), time for the dense zinc oxide layer (type II) formation (tc), and the required time for zincate saturation (ta) [71]. Dense ZnO layers are formed at a high current density, while at a low current density only the porous zinc oxide film will be formed. This film serves as a diffusion barrier to the hydroxides (OH^-) [72–75]. No passivation will occur in the porous ZnO films due to the essential mass transfer amount of the OH^- ions to anodization. As it can be perceived, the continuity and stability of the
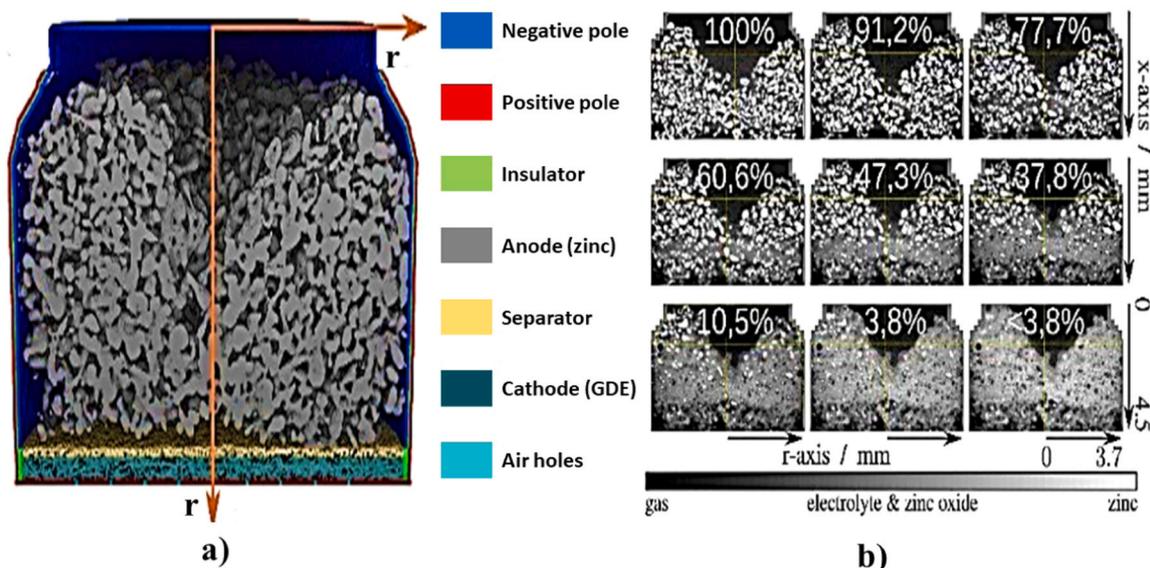
Fig. 3. (a) Schematic image of a fully charged zinc-air button cell. (b) Cross sections of the reconstructed topographies for a zinc electrode [35].

zinc oxide film will regulate the passivation level. Although the mechanism involved in the zinc oxide formation on the planar Zn surface has been investigated profoundly [76], Horn et. al., [65] applied the commercial Zn electrode with the intention of extending such surveys and proofed that the solution-precipitation model is the leading mechanism for the zinc oxide layer formation in the commercial Zn anode and for the applied current density (drain rate). However, Bockelmann et. al. [15] maintained that the formation of the type I zinc oxide film is independent of the current density. According to their analyses results, this layer appears at electrode overpotentials of < 0.15 V after an amount of zincate ions of around $8.2 \cdot 10^{-4}$ mol cm^{-2} has accumulated at the zinc electrode. Accordingly, the type I ZnO layer formation on the zinc anode in the static electrolyte is unavoidable even at very low dissolution currents. In this situation, the type II layer will form below the first layer because of the direct oxidation of the zinc metal. At overpotentials of ≥ 0.15 V, direct oxidation of the metal Zn is preferred, and the type II zinc oxide layer appears before formation of the type I layer. Huot et. al. [66] surveyed the electrochemical features of high-power discharge circumstances restraining the alkaline zinc cell batteries (the commercial type). Three types of Zn were recognized from the color pictures including gray (Fresh Zn), blueish (discharged, ZnO), and white (very discharged). Fig. 4 represents the schematic of the zinc applied in this study. Fig. 4 shows zinc utilization versus energy (commercial alkaline cells) at the end-cut-off 0.9 V for discharge of anode. The different color pictures of zinc cross-sections were obtained from the cut open cells. The formation of ZnO was confirmed using a Raman spectroscopy. Using a high rate of discharge resulted in emergence of the bluish type of the zinc at the interface of the anode-separator, whilst the undischarged one (gray color) was detected at the bulk of the zinc anode. It is clear that the discharged zinc area was expanded with decreasing power down from 1000 to 100 mW, while a white layer (ZnO) started to form within the discharged zinc area, indicating some deeper discharge of zinc along with zinc utilization that was associated with the lower power.

Conversely, in the lower discharge rate, more expanded bluish Zn with the discharged type of zinc (white) within the bluish layer was observed. The rest of the Zn anode remained gray (undischarged). It reveals that the discharge time conduced the discharge layers to the current collector, whilst formation of zinc oxide next to the separator may narrow the cell life under elevating discharge rates. However, it was discovered that just at a very low discharge power, fully utilization of the Zn is possible. Nucleation and growth of ZnO and its outcome on Zn passivation were recently surveyed by Stamm et. al. [77] who modeled a Zn-air button cell and applied it for validating the purpose. In the model, they considered the electrolyte migration, diffusion, and convection as well as the galvanostatic discharge and their impacts on the zinc oxide crystallization, growth, nucleation, and passivation. The outcomes

imposed that the limited cell performance is responsible for the passivating nature of the discharge product. Zinc-air battery modeling dates to the 1970s. However, one of the very first comprehensive mathematical modeling on zinc-air batteries have been performed by Mao and White in 1992 [78]. So far, various models have been developed to model the effects of zinc electrode passivation. Bockelmann et al. [79] reviewed recent modeling studies of zinc electrode passivation. Besides many efforts has been done in understanding the mechanism of zinc electrode corrosion, advanced analytical works are required to fully understand the exact mechanism of this phenomenon. Advanced operando/in-situ techniques are the state-of-the-art methods for a better understanding of passivation and gaining insights into the mechanisms of dendrite formation [80]. Some of the advanced operando studies of zinc passivation are confocal laser scanning microscopy (CLSM) [81], transmission X-ray microscopy (TXM) [82,83], transmission electron microscopy (TEM) [84], nuclear magnetic resonance spectroscopy (NMR) [85], Small-angle neutron scattering (SANS) [86, 87], and small-angle X-ray scattering (SAXS) [88].

It is evident that to alleviate the challenges facing the commercialization of the zinc-air batteries, state of the art structural improvements of the batteries are highly required. Recent technical advancements for the zinc air batteries structures have been reviewed in the ensuing section.

4. Recent technical advancements to overcome the ZnO formation

Doping the electrolytes with additives as well as engineering of the zinc anode are the two main categories that have attracted the researchers' attention in boosting the lifetime and rechargeability of the ZABs by eliminating the passivation of the zinc anode. What follows is an account of the most conspicuous investigations on each category plus detailing their main findings.

4.1. Electrolyte additives

The electrolyte affects the power density and transport properties of the batteries' active compounds implying that it plays a pivotal role in the electrochemistry of battery [89–91]. Since improving the electrolyte to elevate the battery's performance is simple and cost effective with no effects on the battery's specific energy, numerous research endeavors have been undertaken on this subject. The novel tactics performed to attenuate the zinc oxide formation have primarily focused on the use of either additives in the electrolyte or very concentrated alkaline solutions. As for the latter case, it effectively diminishes the Zn passivation because of the dissolution of the passivation film on the anode surface. Nevertheless, this approach contributes to shape changes when solubility becomes too high [92,93]. However, application of surfactants, as a corrosion inhibitor, has turned into a heated topic due to their safety and low cost. Various types of surfactants (amphiphilic molecules containing both hydrophobic and hydrophilic groups) including cationic, anionic, and nonionic have been employed in different ZABs to improve their electrochemical performance (see Table 1).

In a study by Yang et al. [94], SDBS (sodium dodecyl benzene sulfonate) have been applied as an additive to the electrolyte (2% SDBS in 20% KOH solution). They realized that after adsorption of the SDBS compounds on the anode surface, the formed layer of ZnO has a porous and loose structure rather than a compact and dense structure. As mentioned previously, the porous passivated layer (type I) simplifies the diffusive exchange of the discharged product and the solution reactant through the deposited layer on the surface and hence successfully defeats the zinc anode passivation. Using the SDBS additive in the electrolyte solution of this zinc-air battery could enhance the discharge capacity from 360 to 490 mAh/g, equivalent to a 35% growth in the capacity consumption. The SDBS has also been applied as an additive for electrolytes in other researches, exhibiting a prudent characteristic for
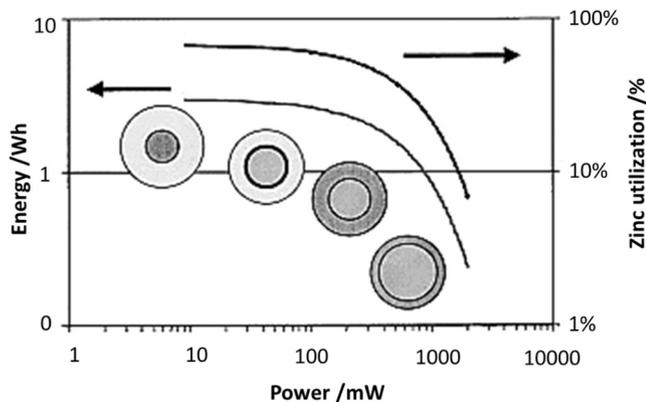
Fig. 4. Schematics of zinc cross-sections attained from the cut open cells discharged down to 0.9 V [66].

Table 1

List of the electrolyte additives applied to mitigate the passivation of Zn anode.

Additive	Additive quantity	Electrolyte	Yield	Ref.
SDBS (sodium dodecyl benzene sulfonate)	2% SDBS	20% KOH	35% growth in the capacity usage	[94]
SDBS (sodium dodecyl benzene sulfonate)	0.8 mM SDBS	26%NH$_4$Cl	89.7% increase in the zinc corrosion inhibition efficiency	[95]
SDBS (sodium dodecyl benzene sulfonate)	0.01 g l^{-1} SDBS	9 M KOH	Dramatic change of the surface Zn crystals that avoid passivation	[96]
PEI (polyethylenimine)	50 ppm PEI	3 M KCl	52.2% increase in the inhibition efficiency of zinc anode corrosion	[97]
Ethanol	5–10 V/V	8 M KOH	30% increase in the discharge capacity16% increase in the specific energy	[98]
Na$_2$Si$_3$O$_7$	0.265 M	0.75 M NaOH	The anodic current diminishes by about two orders of magnitude upon addition of sodium silicate to sodium hydroxide that results in suppressing the anodic dissolution of zinc.	[101, 103]
Sodium metasilicate	0.1 M	1.5 N KOH	Addition of silicate decreases the rate of the anodic dissolution and promotes passivity.	[83]
DTAB (dodecyltrimethylammonium bromide)	0.07 wt%	7.0 M KOH	80.2% corrosion inhibition efficiency	[100]
PNE (poly(oxyethylene) nonyl phenyl ether)	0.25 mM	7.0 M KOH	98.01% corrosion inhibition efficiency	[105]
SDS (sodium dodecyl sulfate)	0.2 mM	7 M KOH	24% enhancement of the specific discharge capacity	[106]
SDS (sodium dodecyl sulfate)	0.8. 10^{-3} mol L^{-1}	1 M Na$_2$SO$_4$ and ZnSO$_4$	Results in the capacity retention of over 90% after 400 cycles	[108]
CTAB (Cetyltrimethylammonium bromide)	0.01 g l^{-1}	9 M KOH	Avoiding zinc passivation by adsorption of CTAB on the growing surface of ZnO	[96]
KAcet (potassium acetate)	5% v/v	20% KOH1% LiOH5% CsCO150 ppm In(SO)	A conventional cell with conventional electrolyte yields only 15 cycles, while the cells with this additive, delivered over 160 cycles.	[109]
Pluronic F-127 (P127)	100 ppm P127	7 M KOH	30% enhancement of the specific discharge capacity	[106]
DMSO	20% v/v	7 M KOH	20% improvement in the discharge capacity	[107]

zinc-air battery application in terms of inhibition of the zinc anode corrosion [95,96]. Sodium dodecyl sulfate (SDS) is another ionic compound which has been applied by researchers as the surfactant in this discipline. Hosseini et al. (20) acknowledged that 0.2 mM SDS effectively suppresses the anode corrosion and passivation in a 7 M KOH electrolyte. The enhancement of a specific discharge capacity of 24% has been reported by applying 0.2 mM SDS to the electrolyte. However, as reported, the direct coating of the SDS on the zinc anode exerts adverse effects on the discharge capacity as well as corrosion and passivation of the zinc anode due to the fact that the thick layer of SDS coating acts as a passivating film and blocks the removal of the anode oxidation product from the zinc surface. The suppression of the zinc corrosion using the SDS surfactant has also been recounted by Hou et. al. [92]. Polyethyleneimine (PEI) is another additive applied in an alkaline electrolyte solution to scrutinize its effects on the corrosion inhibition of the zinc anode [97]. According to the research results carried out by Lin et al., the current density's corrosion of Zn electrode diminishes effectively with elevating the PEI concentration (from 0 to 50 ppm). The Electrochemical Impedance Spectroscopy (EIS) analysis indicated an enhanced resistance for corrosion of the Zn electrode because of the PEI adsorption, by increasing the diameter of the capacitive semicircular loop. Additionally, no significant declining in the anodic dissolution current has been detected in the presence of the PEI, indicating that the adsorbed additive molecules still allow the diffusion and migration of Zn ions, which is why a confident level of the electrochemical activity is sustained.

Ethanol, as a natural and environmentally friendly solvent, has been also surveyed as an additive in ZABs [98]. According to the cyclic voltammograms, EIS analysis, and potentiodynamic polarization measurements, addition of 5–10% v/v ethanol results in the enhancement of the Zn dissolution as well as the interference of the zinc anode passivation. The authors also acknowledged that addition of this amount of ethanol provides the maximum electrochemical performance. In addition, galvanostatic discharge outcomes illustrated that the electrolyte with 10% v/v ethanol revealed the highest electrochemical performance with a 30% growth in the discharge capacity and a 16% rise in the specific energy over that of the KOH electrolyte without ethanol.

Cationic surfactant cetyltrimethylammonium bromide (CTAB) is another compound used in zinc-air batteries [96] which is a detergent compound soluble in alcohol and water. CTAB is frequently applied in the purification and preparation of genomic DNA from bacteria such as DNA mini-preps for sequencing [99]. The findings imply that applying the cationic CTAB as the surfactant (0.01 g.l^{-1}) will avoid the formation of zinc oxide with large sizes on the zinc anode surface since the positively charged groups ($C_{19}H_{42}N^+$) of this compound cannot strongly establish Zn^{2+} species. The well-shaped structure and smaller size of the ZnO particles at the high concentration is because of additional adsorption of CTAB on the growing surface of the zinc oxide that affects the zinc passivation method and the crystal growth method, initiating an inhibiting effect on passivation of the zinc metal. A similar compound with a smaller hydrocarbon chain length (12), dodecyl trimethylammonium bromide (DTAB), also imposes a significant result on preventing the Zn metal corrosion and passivation [100].

Fig. 5 illuminates the analytical results of the application of DTAB in 7 M KOH electrolyte for a ZAB. The corrosion current densities remarkably decline from 27.18 to 5.39 mA cm^{-2} with addition of 0, 0.03, 0.05 and 0.07 wt% DTAB, signifying that it works as an effective inhibitor for the Zn corrosion in the alkaline solution by being adsorbed on the surface of zinc. Fig. 5b shows the trend of Nyquist plots for different concentrations are similar. The diameter of quasi-semi-circle as charge transfer, increased with the amount of surfactant (DTAB). It shows the adsorbed layer of surfactant inhibit the corrosion process which agrees with the Tafel polarization curves. Fig. 5c indicates anodic polarization curves comprised of three regions, active dissolution (the current increased with the potential), pre passivation region (the current changed slightly with the potential), and passive region (the current sharply decreased). The trend of all curves is similar with increasing additive amount. Additives such as silicate (SiO_3^{2-}) have been investigated in zinc-air batteries with alkaline electrolytes [101–104]. Adsorption of silicate ions on the surface of zinc oxide particles that results in the increase in the zinc solubility (blocks further zinc precipitation) is the proved mechanism for applying silicate as the electrolyte additive. Corrosion inhibition efficiency of nonionic surfactant poly (oxyethylene) nonylphenyl (PNE) on zinc was evaluated by Dayeb
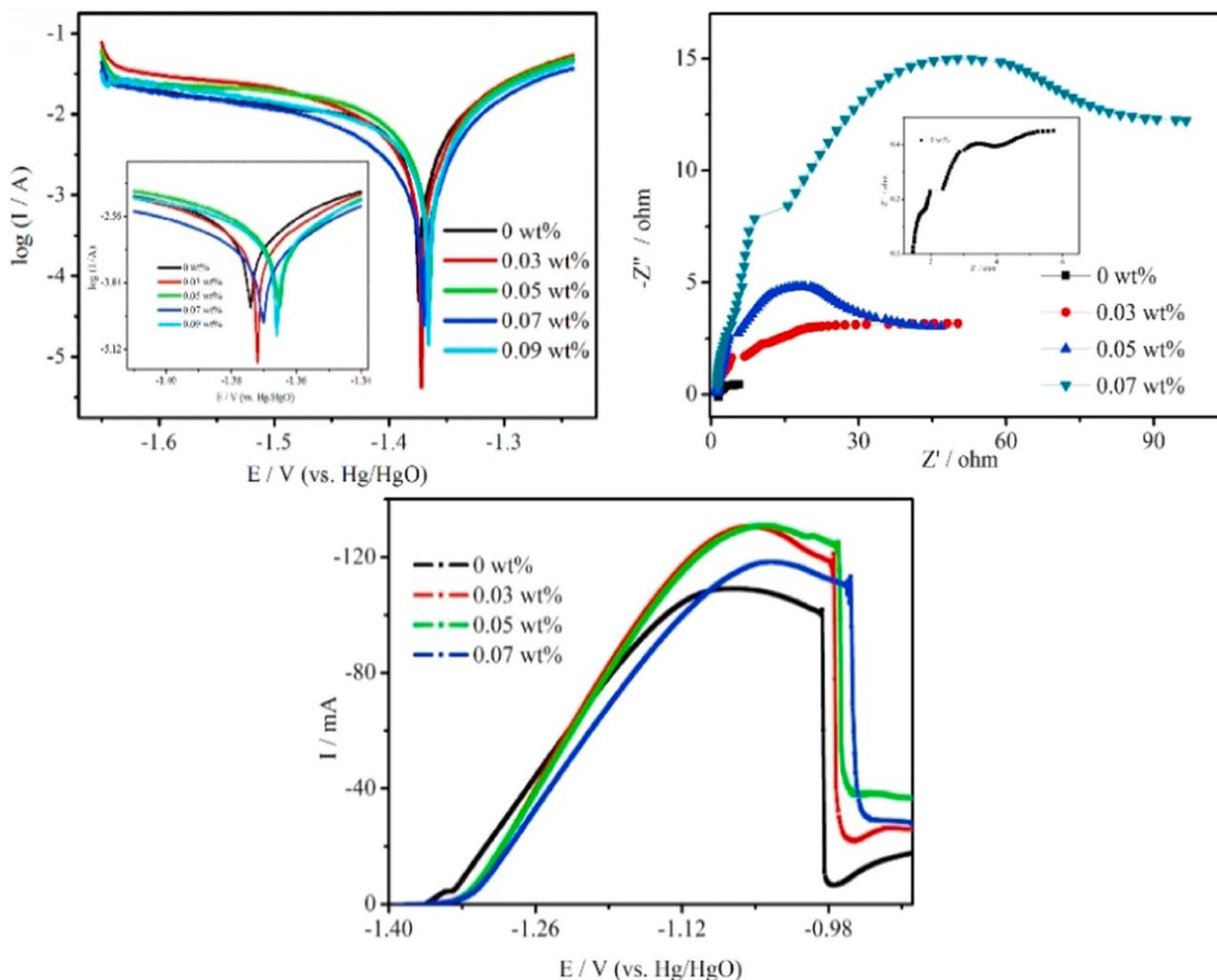
Fig. 5. (a) Tafel polarization curves (b) Nyquist plots, and (c) Anodic polarization curves of Zn electrodes in 7.0 M KOH with addition of various percentages of DTAB [100].

[105]. This surfactant could expand the corrosion inhibition of Zn and improve the battery's capacity. Increasing the PNE dosage in the electrolyte will result in the upsurge in the inhibition efficiency. The highest reported efficiency is 98.01% at the critical micelle concentration (25.10^{-5} M). Analysis of the polarization curves verified that the PNE was the mixed-type corrosion inhibitor. From Fig. 6a (polarization curves), it could be observed that the PNE surfactant addition reduces the zinc corrosion rate in the alkaline electrolyte. The corrosion current density considerably decreases with enhancing in PNE surfactant, indicating PNE surfactant act as a suitable inhibitor. Fig. 6b indicate the Nyquist curves of different concentrations. Similar trends were observed for all curves including a single capacitive semicircle at high-frequency range and a Warburg tail at low-frequency range. According to obtained data from an equivalent circuit, the values of both R_{ct} and Z_w grow with the elevation of the PNE concentration. This approves an elevation in the surface coverage by the PNE results in the formation of the surface layer to the retard passivation procedure. Fig. 6c signifies the surface tension values measured for different concentrations of the PNE compound. It can be perceived that addition of the PNE surfactant to the electrolyte results in a drastic decrease in the surface tension. Hence, it can be concluded that the passivation inhibition occurs by formation of a monolayer of the PNE molecules among the alkaline solution and the Zn anode surface.

Nonionic Pluronic F-127 (P127) or Poloxamer 407 and Dimethyl sulfoxide (DMSO) are the other surfactants which have been applied by

Hosseini et al. [106,107]. The addition of DMSO in the electrolyte demonstrated that the presence of this surfactant lifts the Zn dissolution with the maximum peak of the Zn dissolution being the electrolyte containing 5%v/v DMSO. Besides, cyclic voltammograms, potentiodynamic polarization, and electrochemical impedance spectroscopy proved that using 100 ppm P127 could successfully suppress the zinc passivation and corrosion. According to the findings from various sources, it is inferred that the addition of electrolyte additives is effective in mitigating the passivation in ZABs. However, application of additives may also cast some adverse impacts on the performance of a battery such as difficulties on mass transfer reaction, solid precipitation on the cathode surface, and hydrogen evolution reactions. Therefore, additives should be inert to the electrocatalyst side to avoid such adverse effects. Some studies have reported that cooperative effects (combination of additives) may elevate the adverse effects of a solo additive in electrolytes, contributing to a better cell performance. Yet, it is still imperative that a considerable deal of efforts be exerted in relation to additive development for the batteries' optimum performance.

4.2. Novel zinc electrodes

Self-corrosion caused by the hydrogen evolution reaction in the ZABs has drastic effects on the Zn anodes, resulting in the capacity decay. As stated earlier, the reactions occurred in the zinc anode consumes zinc and H_2O, and simultaneously produces $Zn(OH)_2$ and H_2 on the surface of
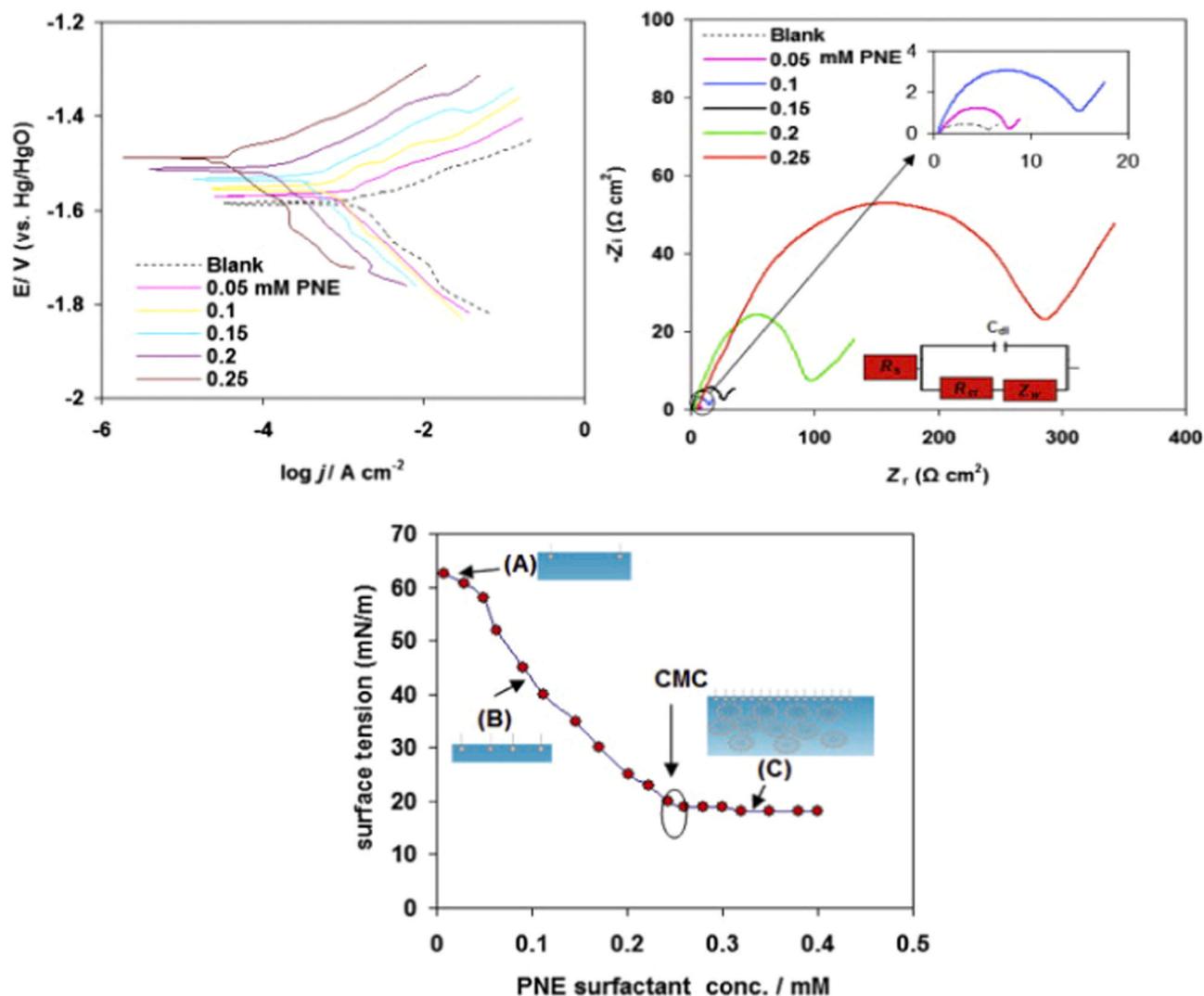
Fig. 6. (a) Potentiodynamic polarization curves (b) Nyquist plots (c) surface tension Vs. PNE concentration [105].

a zinc anode. To prevent the decomposition of the electrolyte, the electrochemical potentials of both anode and cathode must be in between the LUMO (the lowest unoccupied molecular orbital) and the HOMO (the highest occupied molecular orbital) [110,111]. However, the corrosion reaction of Zn happens due to the presence of the electrochemical potential above the LUMO level. In fact, the partial rechargeability and low utilization preclude a larger application of the ZABs. The increase in the current density and absorbance of a large amount of zincate ions on the anode surface resulted in a large electric filed closed to the uneven electron distribution and protuberance which is commonly known as the "tip effect" that is formed as Zn dendrites [112]. To alleviate the self-corrosion phenomenon, various structural modifications on the anode electrode have been performed by researchers in the recent decades (see Table 2).

Current investigations on the 3D porous Zn electrodes offer the Zn-based secondary batteries an admirable electrical conductivity while not obstructing the mass transport. A modeled 3D zinc anode, with a high surface area, diminishes the growth of zinc dendrites by discarding the zincate concentration [113]. A three-dimensional zinc sponge was designed by Ko et. al., [114] without any dendritic morphology after 45 charge−discharge cycles, which was attributed to the high interparticle conductivity of the monolithic 3D structure electrode. However, the DOD of a 3D Zn sponge is restricted due to the deflation of the 3D structure when it is reaching a deep DOD, limiting the cycle stability of

the ZABs. To eliminate this issue, Cheng et al. [115] constructed a 3D porous anode by doping the zinc metal on the nickel foam for a single flow Zn/Ni battery. According to the results, the power density and cycling stability issues for 3D structures were improved. Nevertheless, severe self-corrosion was observed due to the low evolution of the hydrogen overpotential of the nickel foam. Yan et al. [116] presented a 3D Zn/Cu foam for Zn/Ni battery to address the hydrogen evolution issue in 3D Zn anodes. After 10 days of storage, the capacity of the anode remained 59%, while the Zn/Ni foam electrode in Cheng et al. [115] studies dropped to only 5% of the theoretical capacity. Ag/Cu foam anode has been developed by Yu et al. [112] to improve the performance of the ZABs by diminishing the self-corrosion of Zn the electrolyte and dendrite formation. Based on the Tafel and linear scanning voltammetry measurements, deposition of the Ag nanoparticles on the Cu foam, to make a 3D anode, results in a reduction in the corrosion current density as well as the suppression of the hydrogen evolution reaction. Zn utilization of 87%, energy density of 786 W h $kgZn^{-1}$, and specific capacity of 676 mA h gZn^{-1} could be reached by utilization of this anode in a primary Zn air battery.

Electroplating of the zinc anodes with the organic compounds is another strategy for synthesizing the novel zinc anodes. Some organic compounds such as Cetyltrimethylammonium bromide (CTAB), sodium dodecyl sulfate (SDS), polyethylene-glycol (PEG-8000), and thiourea (TU) have been applied in an investigation by Sun et al. [117]. Although

Table 2

List of the novel Zn electrodes applied for mitigating the passivation in the ZABs.

Name	Anode structure	Technique	Yield	Ref.
Zn-CTAB	Cetyltrimethylammonium bromide (CTAB)	electroplating	Capacity retention of 88% (over 350 rounds of charge/discharge)	[117]
Zn-SDS	Sodium dodecyl sulfate (SDS),	electroplating	Capacity retention of 79% (over 1000 rounds of charge/discharge)	[117]
Zn-PEG	Polyethylene-glycol (PEG-8000),	electroplating	Capacity retention of 76% (over 1000 rounds of charge/discharge)	[117]
Zn-TU	Thiourea	electroplating	Capacity retention of 80% (over 1000 rounds of charge/discharge)	[117]
Zn-SiO$_2$	Zinc particles were coated with Zn$_2$SiO$_4$	chemical solution deposition	CVD-coated zinc: 69% zinc utilization, CSD-coated zinc: 62% zinc utilization as compared to as-received zinc (57% zinc utilization)	[121]
Bi$_2$O$_3$-ZnO-CaO	bismuth oxide-based coatings	ball milling	Zn utilization increased to 465%, as compared to uncoated zinc representing just 85%	[118]
Bi$_2$O$_3$-ZnO-SiO$_2$	bismuth oxide-based coatings	ball milling	Passivation of Zn electrode after the second round	[118]
LBO-Zn	lithium boron oxide coating	Coating by solution process	Initial voltage of 1.44 V, discharge capacity of 1.70 Ah for 159 min and DoD about 89% in comparison to uncoated Zn showed an initial discharge capacity of 1.57 Ah for 147 min and DoD of about 82%	[119]
Ag-modified 3D anode	Cu foam with Ag nanoparticles deposited on the surface	Galvanostatic electrodeposition	Energy efficiency of 60% in stable cycling over 40 cycles.	[112]
Bi$_2$O$_3$-Li$_2$O-ZnO	Bi$_2$O$_3$, Li$_2$CO$_3$, ZnO	mechanical ball milling	Average charge capacity over 20 cycles of 113 mAh/g	[120]
Zn-pome	zinc oxide nanoparticles	bottom-up microemulsion approach	Steady charge capacity after 50 cycles of charge/discharge	[134]
Zn$_{67}$Al$_{33}$	Zn-Al alloy	melting the mixtures of pure Zn and Al beads in an electric furnace	OCV values of 1560 mV and the specific anode capacity of 800 mA h g^{-1} at a discharge current of 200 mA	[135]
Zn$_{59}$Al$_{41}$	Zn-Al alloy	melting the mixtures of pure Zn and Al beads in an electric furnace	OCV values of 1540 mV, and specific anode capacity of 750 mA h g^{-1} at a discharge current of 200 mA.	[135]
Zn@GO	Graphene oxide-modified (1.92 wt %) zinc anode	physical mixing of GO and Zn round disks	28% upsurge in the lifetime capacity of the battery	[122]

only 67% capacity retention was attained from the batteries using the anode prepared from the commercial zinc foil, the barriers with Zn- SDS, Zn-PEG, and Zn-TU anodes showed capacity conservations of 79%, 76%, and 80% after 1000 cycles of charge−discharge at 4 °C rate. Among the anodes with various organic additives, Zn-SDS represented the performance in terms of the corrosion rate, the low float current, high-capacity
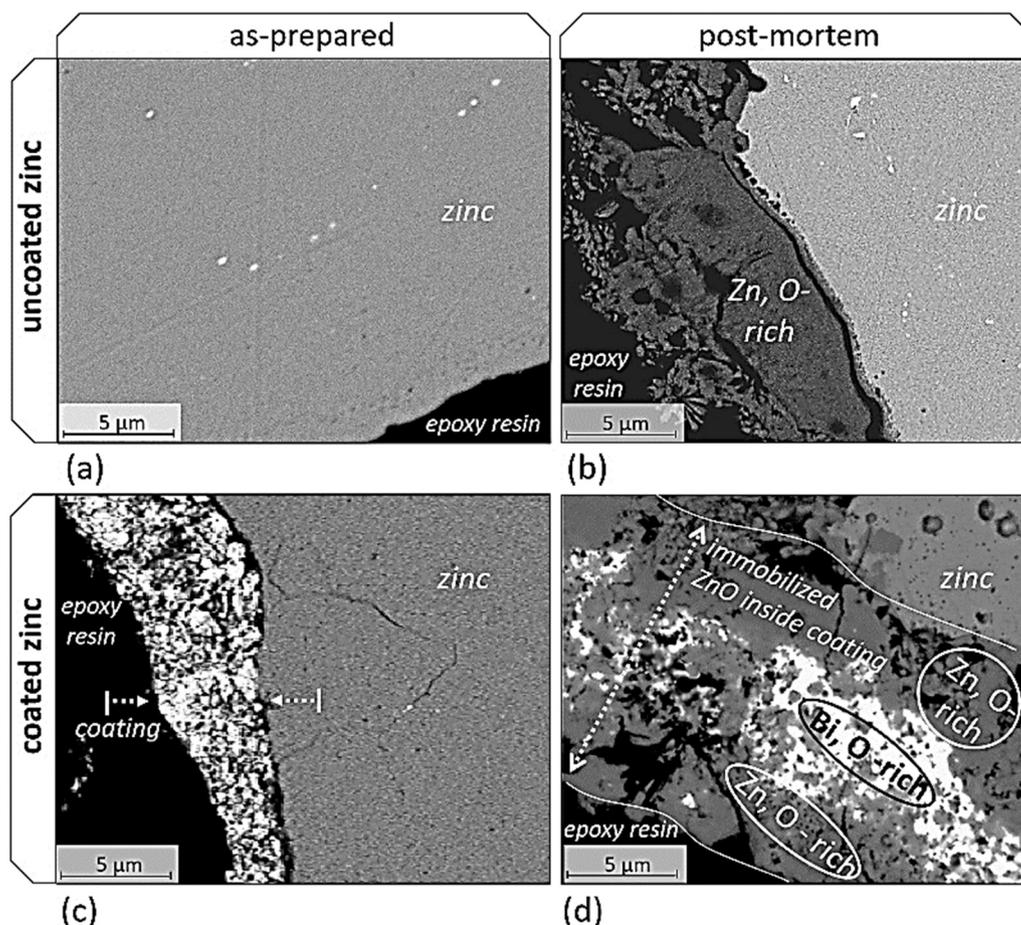
Fig. 7. SEM micrographs of the uncoated and coated samples of the Zn anode with Bi$_2$O$_3$-ZnO-CaO before and after cycling [118].

conservation after 1000 cycles, and low dendrite formation.

Modification of zinc in the ZABs with protective coating is another solution that has recently received researchers' attention to mitigate the zinc passivation. Bi_2O_3-ZnO-CaO and Bi_2O_3-ZnO-SiO_2 glasses were applied as a coating material for the zinc anode in 6 M KOH electrolyte by Schemid and Porada [118]. The electrochemical performance of the coated zinc particles shows that the rechargeability will improve with Bi_2O_3-ZnO-CaO-coated zinc particles. It is asserted that the uncoated zinc attained one complete discharge while a cyclic stability of 20 full cycles was discovered in excess electrolyte. Based on the results, utilization of the accumulated zinc on the coated zinc particles could improve up to 465% compared to the uncoated zinc which just yields 85% in test cells. The cross section of the coated and uncoated zinc, before and after cycling, have been depicted in Fig. 7. Due to the formation of an interpenetrating network, an improvement in the cycling behavior was observed which resulted in discharge products that could be immobilized inside the swellable coating while forming a porous network after drying. Furthermore, acquiring less formation of the passive zinc oxide layers or the soluble zincate ions resulted respectively from the Oxidized zinc bond formed inside the coating and the one accumulated around it.

The lithium boron oxide glass (Li_2O–$2B_2O_3$) is another compound which has been applied as the coating on the surface of the zinc anode [119]. The research results revealed that the LBO film can stop the Zn anode from a direct contact with the electrolyte and that it can diminish the passivation and side reactions of the zinc within the ZABs. An initial discharge capacity of 1.70 A h could be reached using the LBO-coated (0.1 wt%) Zn electrode anode, whilst the pristine Zn electrode could only deliver 1.57 A h. Similar results have been found by using Bi_2O_3-Li_2O-ZnO glass as a coating medium on the surface of the zinc anode [120]. The electrochemical analyses revealed that the application of this compound as the coating medium could increase the charge capacity to over 20 cycles of 113 mA h/g in comparison to the reported zero rechargeability of the uncoated Zn anode. In another study by Schemid and Porada [121], the silica coating was examined as a promising compound to decrease the early development of the permanent zinc oxide passivation films during the discharge by precipitation of the supersaturated zincates, and controlling the Zn dissolution. Different electrochemical results discovered that the silica coated Zn anodes display an enhanced discharge capacity as compared to the as-received Zn, by diminishing the supersaturation of the zincates ($Zn(OH)_4^{2-}$). Moreover, the electrolyte-modified silica coated Zn anodes improve the rechargeability after 100% DOD. Graphene oxide (GO) prepared by the modified hammer method is another material used by Zhou et al., [122] to overcome the passivation and dissolution problems in the ZABs. The research outcomes indicate that the GO improves the electrochemical performance of the uncoated Zn anode, which results in an even spread of the Zinc oxide between GO films in the electrochemical reactions. This elevates the cycle life of the zinc air battery by both forming a required environment for the electrons to transport easily and slowing down the predictable dissolution process of the active anode

compounds. Homogeneous coating using a polymer, or an anion-exchange ionomer (AEI) is an advanced coating method to overcome zinc electrode limitations. Miyazaki et al. [123] alleviated the dendrite formation of zinc electrodes using AEI as a surface protective layer of zinc particles. In the same way, Stock et al. [124] demonstrated an effective increment of the cycle life for Zn model electrodes coated with a homogeneous layer of an AEI. They explained such an increment by a greater amount of restored zinc during electrodeposition by confinement of oxidized zinc compounds near the electrode surface. Fig. 8 represents a conceptual scheme for homogeneous coating using anion-exchange ionomers. According to the figure, during the discharge step, zinc metal dissolves and produces zincate, which instantly forms zinc oxide in the interlayer among the Zn surface and the flexible AEI coating (Area 1). The zincate flow into the electrolyte (Area 2) should be inhibited by the AEI, resulting to the eliminating shape change of the zinc anode.

Vatsalarani et al. [125] stated that the reversibility and corrosion protection of the zinc electrode in alkaline medium were enhanced by electrostatic deposition of a conducting polymer onto the electrode. Although the anion exchange materials/polymer coating is an effective method to reduce shape change and passivation in the zinc air batteries, some of these coating materials suffer from high permeability for zincate ions. Methylation can minimize the permeability of these coating compounds against zincate ions. In a study by Konovalova et al., [86] they could minimize the permeability of polybenzimidazole for zincate ions by methylating its imidazole NH groups. Higher performances of the alkaline zinc anode such as cycling stability and initial capacity has been observed in this study.

The nanomaterial design of the zinc anodes such as the core/shell with surface coating, the 3D high surface area electrode structure, and the nanocomposites with highly conductive materials also overcome the dissolution and passivation problems in the ZABs [37,126,127]. Some of these novel designs include ZnO/graphene [128], ZnO@TiN [129], Zn sponge [130], ZnO@C [131], ZnO/RGO [132], and ZnO lasagna [133]. A novel nanomaterial structure for the Zn anode has been proposed by Chen et. al., [134], who reported a nanoscale, pomegranate-structured Zn electrode produced via a bottom-up microemulsion method to overcome $Zn(OH)_4^{2-}$ dissolution and ZnO passivation. In this structure, the ion-sieving carbon shell and the secondary structure are responsible for mitigating the dissolution problem, whilst the small size of the zinc oxide nanoparticles overcomes the passivation issue. The schematic design of the schematic pomegranate structure and some other novel designs have been illustrated in Fig. 9.

The Zn-Al alloys are considered as another strategy to prepare new anode-metals in the zinc-air batteries. Lan et al. [135] prepared two different alloys ($Zn_{67}Al_{33}$ and $Zn_{59}Al_{41}$) to enhance the performance of the ZABs. The alloys were composed of two phases, namely Al-rich and Zn-rich. The Al-rich anode could favorably react with the electrolyte (KOH) making self-assembled surface porosity or penetrated-channels for the electrolyte to flow inward, thus dropping the anodic passivation. It has been accentuated that adding the alloying element Al will
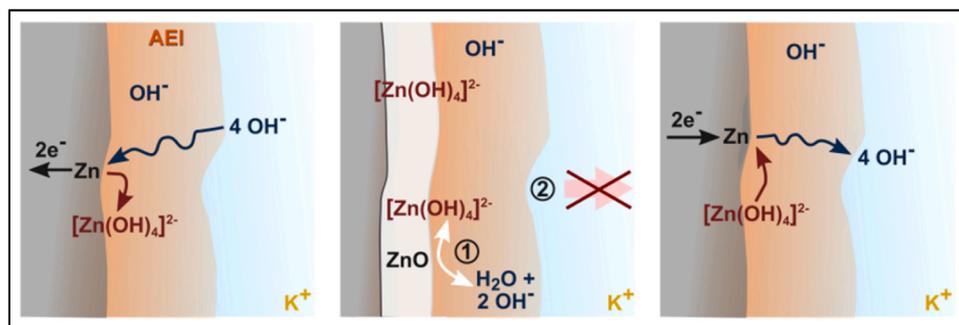
Fig. 8. A schematic for homogeneously coated zinc electrode with an anion-exchange ionomer [124].
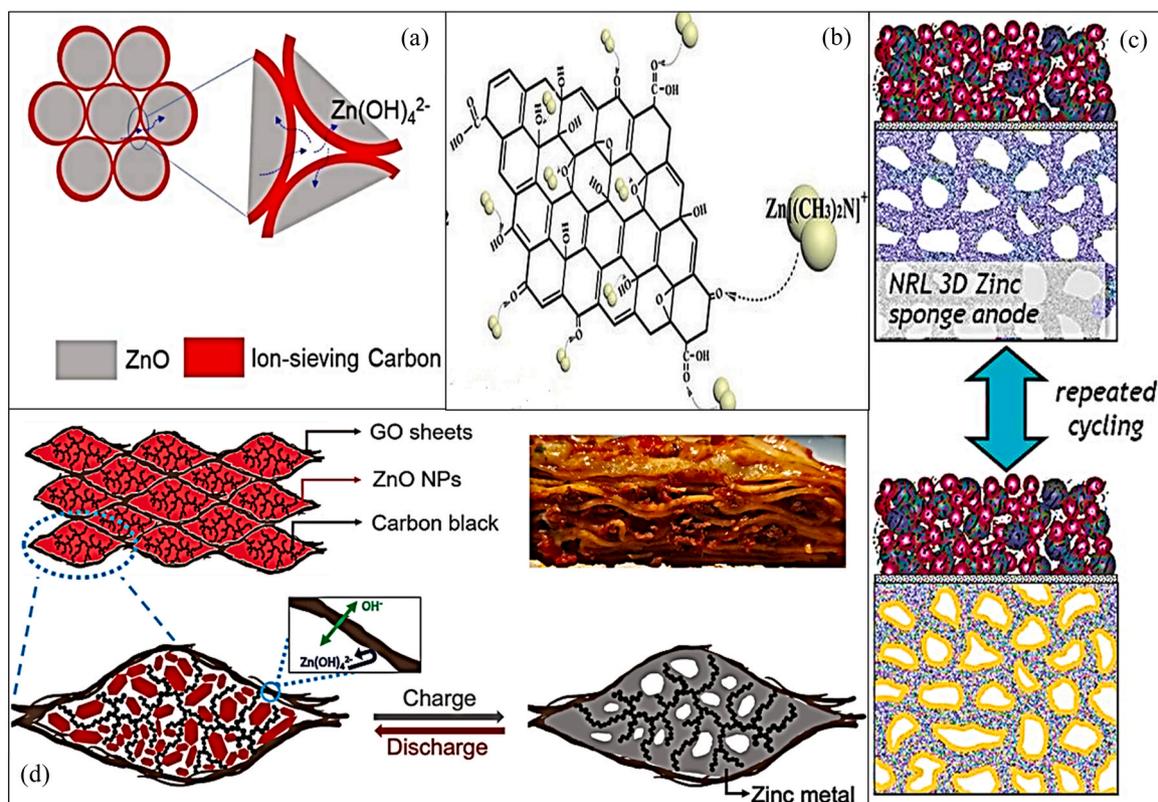
Fig. 9. Schematic structures of (a) pomegranate-structured Zn [134]; (b) ZnO/graphene [128]; (c) Zn sponge [130]; and (d) ZnO lasagna [133].

result in a further increase in the specific capacity as well as the fact that the produced electrode would be lighter in comparison to the zinc metal only as the anode material.

5. Conclusion and perspectives

Several advantages include high energy density, ease of handling, low cost, environmental compatibility, safety, and nontoxicity have made the zinc air batteries as a suitable alternative for energy storage. However, it suffers from limited cycle lives and energy efficiencies which originate from passivation and zinc electrode adverse impacts on the rechargeability of these types of batteries. It is known that zinc anode corrosion leads to the capacity loss, as it results in the formation of an inactive layer of mixed oxide and hydroxide on its surface. In some cases, the corrosion of the zinc anode may result in cell overpressure if the hydrogen gas product is not vented out. Therefore, the zinc corrosion is of great significance, especially in the secondary systems where their effects are often more pronounced due to the long lifetime of the batteries as well as the permanent change of the electrode morphological structure upon subsequent cycles and abusive use. To attain better performance and cyclability, various studies have been reviewed elaborating on the improvement of the electrolytes and anode's structures, addition of additives to the conventional electrolytes and synthesizing well-organized and novel catalysts. It was witnessed that addition of efficient additives such as SDS, DTAB, and the like contributes to a diminishment in passivation and zinc anode dissolution while elevating the ZABs' stability and cycling. Furthermore, some state-of-the-art zinc anode structures represented promising outputs in mitigating the passivation phenomenon in the ZABs. Despite considerable achievements in these fields, several major challenges still exist, such as insufficient utilization of well-developed theory-based models to utilize experimental outcomes, lack of standard methods for assessment of electrochemical performance for zinc air batteries, and inadequate combined improvement techniques in a solo study. To address the

battery's cycle life problem due to the passivation, the key fields of improvement must be carried forward.

1. No comprehensive study has been performed thus far and almost all the current investigations have merely elaborated on a special condition for the ZABs to represent their findings. A good combination of zinc additives in polymer/ceramic electrolytes and engineered 3D zinc anode in the real conditions of a commercial battery may eradicate the obstacles for widespread application of the ZABs in the future.
2. Future modeling studies are required to provide a structure for understanding and interpreting experimental results from alternative electrolytes, additives, and zinc anode structures. Executing these modeling outcomes in the design process brings researchers closer to the goal of building rechargeable and high-performance zinc air batteries with minimum passivation of the zinc metal.
3. Electrolyte additives might negatively affect other properties of the batteries. Comprehensive studies are compulsory to evaluate the overall electrochemical performance of zinc air batteries to eliminate the gap among academia and commercialization objectives.
4. Application of advanced operando/in-situ techniques to study the passivation mechanism of the zinc electrode under alkaline electrolytes need to be improved. Due to the various structure and morphologies of the zinc electrodes, different dominant mechanisms might be occurring and hence, operando techniques could reveal the exact passivation mechanism of the zinc anode.

CRediT authorship contribution statement

Hamed Pourzolfaghar: Conceptualization, Investigation, Writing - original draft.: **Soraya Hosseini:** Investigation, Writing - original draft, Formal analysis.: **Fathiah Mohamed Zuki:** Visualization, Methodology.: **Marziyeh Alinejad:** Writing- Reviewing and Editing.: **Yuan-Yao Li:** Data curation, Supervision.

Declaration of Competing Interest

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

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