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REVIEW

Recent advances in catechol-based polymer binders for batteries

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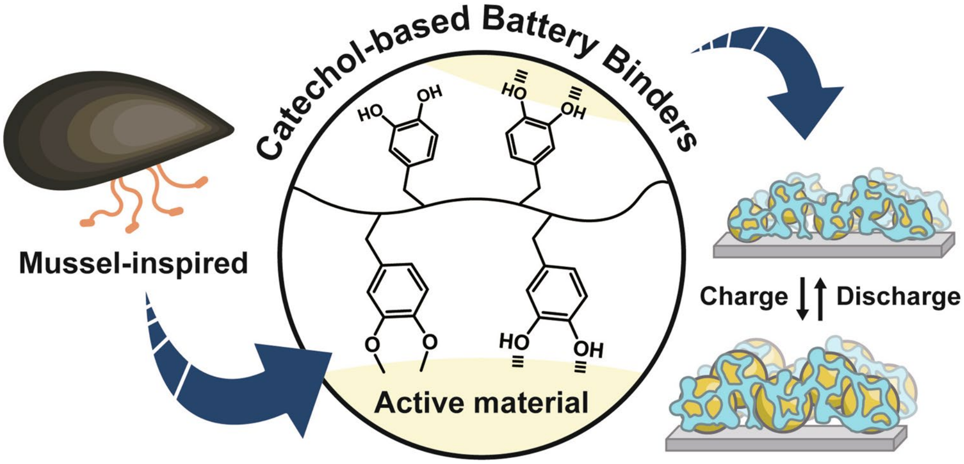
Abstract

Polymer binders constitute less than 3% of the cell's weight but play a critical role in the battery's performance in various batteries. This review summarizes recent research on traditional and mussel-inspired catechol binders. Notably, catechol binders exhibit excellent adhesion to active materials, catalysts, and current collectors, enhancing the electrode stability of the batteries. Through recent research findings, this review highlights the potential of new catechol binders for various types of batteries and suggests future research directions.

Graphic Abstract



Schematic illustration highlighting catechol-based polymeric binders in battery charge–discharge cycles

Keywords Catechol · Polymeric binder · Batteries · Adhesion

1 Introduction

Various electrode materials and electrolytes have been investigated to enhance the energy density and safety performance of batteries; however, the significance and research of binders in improving the electrochemical performance of batteries have been underrated. Polymer binders, though used in small quantities within the electrode, play a crucial role in maintaining the integrity of the electrode and facilitating electron/ion transfer during battery charging and discharging cycles. Without binders, the connection between active materials, conductive agents, and current collectors would be lost, leading to capacity loss. Therefore, binders have a direct impact on the capacity, lifetime, and safety of the battery [1, 2].

Consumer electronics, aircraft, transportation, and large-scale energy storage systems currently use lithium-ion

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batteries (LIBs). Compared to traditional LIBs, there is a growing demand for higher energy density, improved safety, longer lifetime, environmental friendliness, and reduced costs [3, 4]. The most recent advances in LIB technology involve coating current collectors with active materials and conductive agents mixed with polymer binders to form electrodes [5]. Silicon (Si) is one of the most widely researched anode materials for high-energy LIBs because of its impressive theoretical capacity of 3580 mAh/g [6–8]. However, the substantial volume expansion (approximately 400%) of Si during lithiation and delithiation processes leads to mechanical degradation and loss of electrical conductivity pathways, causing fracturing of Si and continuous electrolyte decomposition at the exposed active surfaces [9–11].

The process of charging and discharging in metal-air batteries involves the reaction of metals (such as lithium, sodium, or zinc) with oxygen in the air [12]. These batteries theoretically offer up to 10 times higher energy density compared to LIBs. Additionally, metal-air batteries are cost-effective, reducing resource depletion and environmental impact, making them promising candidates for eco-friendly energy storage systems. However, metal-air batteries suffer from lower charging speed and efficiency compared to LIBs and challenges such as metal and/or carbon current collector corrosion [13].

Polyvinylidene fluoride (PVDF) binders are traditionally used in LIBs and metal-air batteries due to their thermal, chemical, and electrochemical stability [14]. However, PVDF has significant drawbacks. For LIB with Si electrodes, PVDF induces poor adherence to Si particles and current collectors. More importantly, PVDF makes it difficult to regulate Si particle volume variations [11, 15]. Additionally, the use of toxic solvents such as *N*-methyl-2-pyrrolidone (NMP) or *N, N*-dimethylformamide (DMF) causes significant environmental and health concerns [16, 17]. PVDF and lithium hexafluorophosphate ($LiPF_6$) are representative PFAS-based materials that contribute to the performance of LIBs; however, they can generate toxic hydrofluoric acid (HF) during battery cycling, leading to severe environmental pollution [18]. Moreover, in 2023, the European Union announced a report and defluorination policy restricting the use of perfluorinated compounds, including perfluorooctanoic acid (PFO), perfluorooctanoate salts (PFOA salts), and PFOA-related substances used in synthesizing PVDF [19]. This regulatory shift further underscores the need for alternative binder materials that are both effective and environmentally friendly.

To address these issues, research has been conducted on various functional binders. Binders with polar groups such as carboxymethyl cellulose (CMC), alginate, polyacrylic acid (PAA), and polyvinyl alcohol (PVA) have shown potential to overcome the drawbacks of PVDF binders with environmentally friendly solvents and hydrogen bonding between the binder and hydroxyl groups on the Si particle surface [20]. However, challenges remain in achieving stable adhesion with silicon, managing volume expansion/contraction, and preventing electrode cracking.

Catechol has been proposed as a solution to the issues associated with existing binders used in LIBs due to its diverse adhesion mechanisms and excellent adhesive properties, as depicted in Fig. 1. This review aims to introduce the limitations of conventional binders, summarize recent research on catechol-based binders, and present potential improvements and strategies for binder development.

2 Conventional binders for batteries

2.1 Polyvinylidene fluoride (PVDF)

PVDF is prominently utilized in lithium-ion batteries due to its superior electrochemical stability and mechanical strength. It effectively adheres to graphite anodes; however, its interaction with Si anodes used in high-capacity batteries is comparatively limited. Notably, Si nanoparticles exhibit significant volume expansion during charge and discharge cycles, which compromises the integrity of the electrode structure and results in cracks [21]. Additionally, the use of PVDF necessitates employing toxic volatile solvents such as NMP or DMF, which are detrimental to the environment and human health [15]. To enhance the electrical conductivity of the electrode structure, inert conductive additives must be included, which subsequently reduce the specific capacity of the electrodes [22].

2.2 Polyacrylic acid (PAA)

PAA, a polymer characterized by long carbon chains and numerous carboxylic functional groups, can form robust hydrogen bonds with current collectors, active materials, and binders. This bonding mitigates the volume expansion issues during charge and discharge cycles, allowing the material to revert to its original size. Typically, PAA binders remain electrochemically inert during the cycling process, thus preventing adverse reactions and enhancing the electrochemical performance by inhibiting continuous electrolyte decomposition [23, 24]. However, the molecular weight of the polymer binder is a critical factor influencing its binding capability; an intermediate molecular weight enables the formation of a stable and smooth Solid Electrolyte Interphase (SEI) layer, effectively reducing internal resistance and enhancing electrode capacity [25].

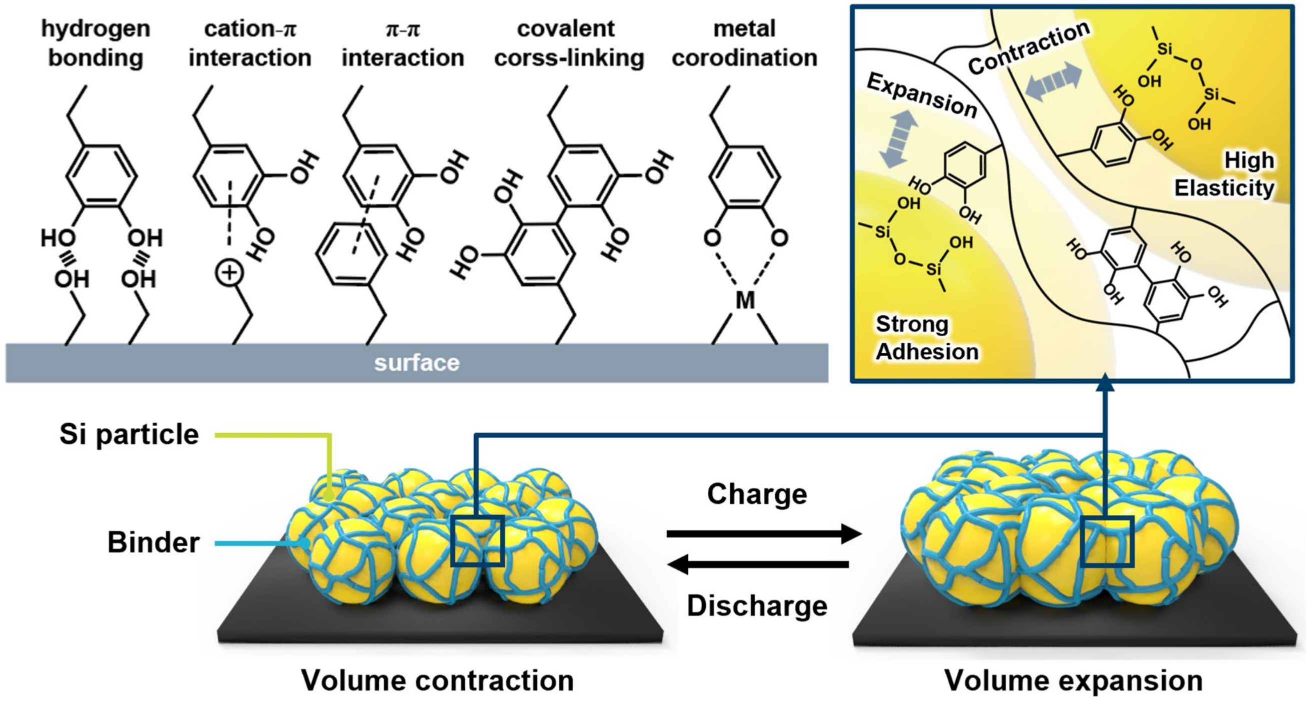

Fig. 1 Schematic illustration of the various adhesion mechanisms of catechol groups and the suppression of volume expansion/contraction with Si particles

2.3 Carboxymethyl cellulose (CMC)

As a water-soluble and environmentally friendlier alternative to PVDF, CMC contains numerous hydroxyl and carboxymethyl groups that enhance binding performance through chemical bonding with active materials. It has been demonstrated through peel tests that CMC exhibits stronger adhesion than PVDF [26, 27]. Additionally, CMC can increase the viscosity of aqueous solutions without gelation and is readily cross-linkable. CMC exhibits greater rigidity compared to PVDF, with respective Young's moduli of 4100 and 650 MPa [28]. Sodium carboxymethyl cellulose (NaCMC) has been extensively studied as a binder for Si-based lithium-ion anode materials, showing superior battery performance over PVDF binders. Electrodes utilizing CMC display smoother surfaces and fewer cracks, leading to enhanced performance [10].

2.4 Alginate

Extracted from brown algae, alginate is a biodegradable and environmentally friendly binder [29]. Its hydrophilic colloidal nature and excellent mechanical strength address the expansion and contraction issues of electrode materials. Research by Kovalenko et al. demonstrated that sodium alginate as a binder for high-performance Si anodes significantly improves battery performance and mechanical strength compared to PVDF binders [30]. Alginate's mechanical strength is crucial for enduring significant volume changes from active materials, thus enabling long cycle lives for Si anodes. The carboxylate groups in alginate facilitate lithium-ion transport through the formation of lithium carboxylate and enhance battery performance and stability through covalent and hydrogen bonding on the electrode surface [31].

2.5 Nafion

Based on perfluorinated polytetrafluoroethylene (PTFE), Nafion features sulfonic acid groups ($-SO_3H$) at the ends of its side chains, promoting ion exchange and providing high ion conductivity within the electrolyte [32]. While primarily researched and utilized in fuel cells, Nafion's exemplary ion conductivity and mechanical strength also make it a promising candidate for lithium-ion batteries [33]. Its chemical stability prevents the decomposition of electrode materials over long cycle lives, mitigating performance degradation and durability reduction. However, Nafion's high cost compared to other commercial binders remains a significant drawback, limiting its widespread application despite its potential for high performance in lithium-ion batteries [34].

In addition, electrically conductive polymers with full conjugation have been synthesized and used as binders [35–37]. It is well known that an entire electrode consists of an active material, conductive agent, binder, and current collector. Therefore, the conductive agent and binder can be fully substituted with a conductive binder. Conductive polymers, which do not use conventional conductive additives, offer the advantage of enhancing the mobility of electrons and improving the performance of LIBs. However, conductive polymers are significantly more expensive than Nafion polymers and involve multiple stages of synthesis and purification processes, presenting a challenge. Despite their excellent properties of preserving the electrical conductive network and facilitating the electrochemical reaction of lithium with Si during the insertion and extraction of lithium at high rates, conductive polymers still expose several limitations in commercialization and development.

3 Catechol group-based binders for batteries

3.1 The characteristics of catechol groups

Inspired by the marine mussel foot protein, the catechol group has been identified to play a crucial role in exceptional wet adhesion. Catechol is reported to interact with various substrates through bidentate hydrogen bonding, cation–π, π–π interaction, covalent crosslinking, and metal coordination [38–41]. Catechol exhibits biocompatibility, making it valuable in research for medical adhesives and drug delivery systems, and its biodegradability highlights it as an environmentally friendly material [42]. Additionally, the combination of catechol with functional groups allows for the design and synthesis of various materials tailored to specific needs [43]. The catechol group, capable of forming coordination bonds with transition metals, is useful for effectively capturing and immobilizing transition metal ions dissolved in electrolytes, thereby contributing to electrode protection and stability enhancement in battery materials. Notably, catechol-based polydopamine (PDA) coatings can uniformly coat surfaces with different characteristics, including organic, ceramic, and metallic surfaces, making them valuable for a wide range of applications [44]. A particularly important feature is catechol's strong coordination with metal ions and oxides. For example, Fe(III) forms highly stable bis- and tris-catecholate complexes (log K_s $\approx$ 43), enabling robust yet reversible bridging interactions with adhesion energies of several mJ m^{-2}. Such strong, specific, and dynamic coordination can significantly enhance binder–electrode interfacial stability in lithium-ion batteries [45]. Furthermore, catechol and catecholamine systems also rely on cation–π interactions. These interactions occur when protonated amines or hydrated cations bind to electron-rich aromatic groups and are unusually strong in aqueous environments due to their low desolvation penalty and pronounced ion-specific dependence [46]. Direct surface force measurements and mussel-protein model studies have confirmed their significant contribution [39, 47], and cation–π binding is now recognized as an essential, though often underappreciated, mechanism underlying the adhesion of catechol-based binders [48].

3.2 Use of catechol groups in battery binders

As previously mentioned, commercial binders such as PVDF, PAA, CMC, alginate, and Nafion possess limited adhesive properties and are not immune to the challenges of volume expansion and damage in anode materials like Si nanoparticles. Notably, their low electron/ion conductivity complicates the development of high-performance batteries. Therefore, it is essential to design novel binders that incorporate high adhesion, strong mechanical strength, and enhanced electron/ion conductivity, thereby endowing them with multifunctional properties. Catechol possesses the characteristic of being applicable for coatings on various surfaces. It can be utilized to enhance the surface and interfacial properties of battery components such as binders, active materials, and separators, thereby improving battery performance. In this section, we aim to discuss the superior adhesive properties of catechol binders and the applications of catechol in battery systems.

The critical role of polymeric binders has begun to be recognized in addressing the volume expansion issue of Si anodes [49–51]. In traditional lithium-ion battery (LIB) electrodes, binders constitute a small percentage (below 3 wt%) and are primarily focused on adhering active materials with minimal expansion to the current collector. However, the significant volume expansion of Si nanoparticles necessitates the development of improved binders capable of maintaining the integrity of the electrode during charge–discharge cycles. Strong adhesion between the binders and Si nanoparticles would be advantageous, and various binder-to-Si binding mechanisms, including hydrogen bonding [49, 52, 53] and ion–dipole interactions, have been introduced [30, 54]. Moreover, the self-healing property of the binders is found to be important [55, 56]. Likewise, mussel-inspired wet adhesion of catechol functional groups has been utilized to enhance the adhesion between binders and Si particles. Figure 2a shows the use of catechol functional groups conjugated to alginate and PAA backbones as Si anode binders [57]. As shown in Fig. 2b, poly (N-methacryloyl-3,4-dihydroxy-L-phenylalanine) (PMDOPA) was polymerized and applied to LIBs. The higher molecular weight PMDOPA binder forms strong bonds with the hydroxyl groups on the Si surface, enabling the formation of a more elastic and flexible network. This network can accommodate the volume expansion and contraction of Si nanoparticles, enhancing their stability. The elastic and flexible network facilitates electron transfer, reducing resistance and improving

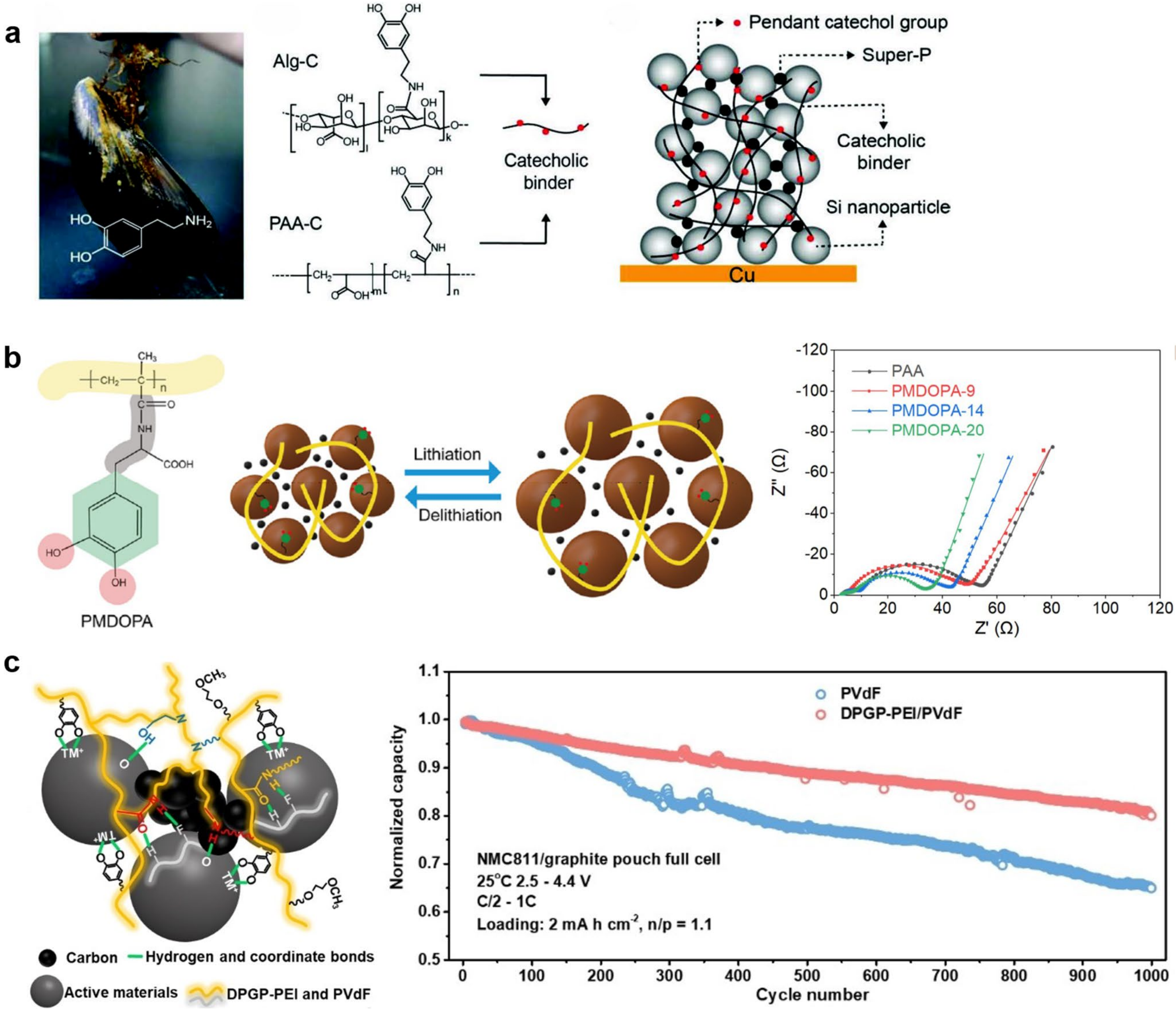

Fig. 2 **a** Schematic diagram of Si anode coated with catechol conjugated alginate (Alg-C) and poly(acrylic acid) (PAA-C) binders. Reproduced from ref. [57], with permission from Wiley-VHC, Copyright 2013. **b** Schematic diagram of lithiation and delithiation of catechol-coated Si particles (left) and impedance (right). Modified and reproduced from ref. [58], with permission from Elsevier, Copyright 2020. **c** Catechol-PVDF binder and electrode particles interactions involving hydrogen and coordinate bonds (left), and capacity influenced by the binders (right). Modified and reproduced from ref. [60], with permission from Wiley-VHC, Copyright 2020

electrochemical performance [58]. A catechol-based triple-block copolymer was synthesized and applied as a binder in LIBs, providing excellent adhesion and electrical conductivity, thereby enhancing the electrochemical performance of Si anodes. The employed catechol binder with a triple-block structure improved physical and chemical stability, positively impacting the battery's lifetime [59]. Additionally, to mitigate the structural and interphasial degradation of high-nickel cathodes, a catechol binder framework was applied. The practical viability of high-nickel oxide cathodes is compromised by interfacial and structural degradations. To address these issues, a new network binder composed of catechol and polyethylenimine was employed, significantly improving the cycling stability of the high-nickel cathodes (Fig. 2c) [60]. The synthesized binder provides strong adhesion through catechol groups and maintains the integrity of the electrode, thereby preventing the self-agglomeration of electrode materials.

Si particles have specific capacity that is ten times higher than traditional carbon anodes, but it suffers from poor cycle performance due to significant volume changes during lithium insertion and extraction. Considerable efforts are being made to address this issue using polymer binders [61]. A novel PAA-based catechol binder with self-healing properties and a new multiple network structure, designated as PAA-catechol, was designed and synthesized (Fig. 3a) [62]. This catechol binder demonstrates excellent wettability with liquid electrolytes and has the ability to control volume changes in Si microparticles (SiMPs) through its covalent and hydrogen bonding structures. Electrodes utilizing the catechol binder maintain a smooth surface morphology and intact particles even after 100 cycles, whereas electrodes using the PAA binder exhibit extensive cracking across the electrode surface and severe pulverization of SiMPs. The emergence of these cracks and the division of smaller active particles within the electrode lead to increased electrolyte consumption for the formation of the solid electrolyte interphase (SEI), resulting in decreased battery efficiency. A cross-linked network of catechol-functionalized chitosan polymer binder was synthesized. This catechol

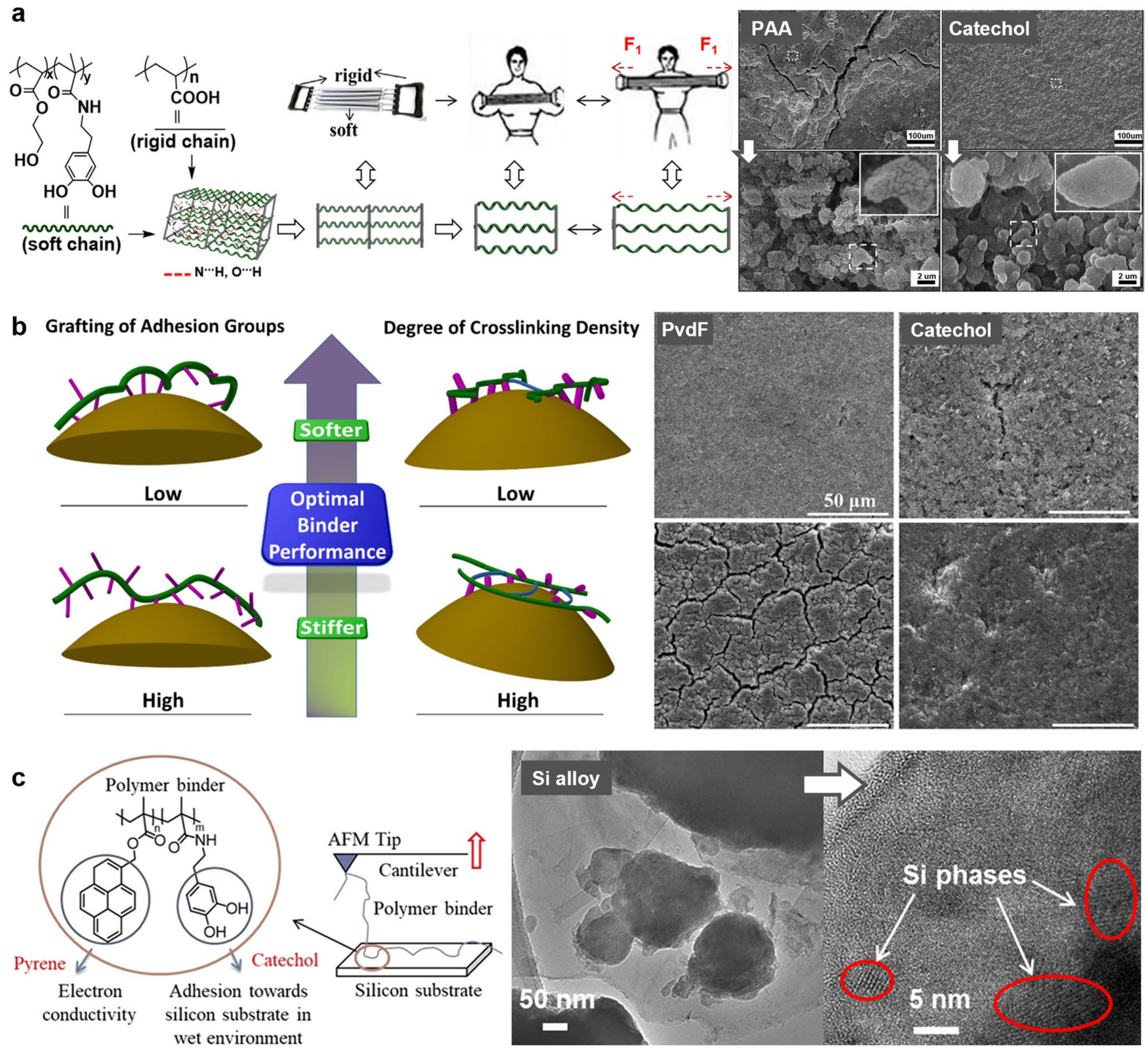

Fig. 3 **a** Chemical structures and illustrative interactions between catechol and PAA binders, along with a spring expander model of their complex, and SEM images of silicon microparticles (SiMPs). Modified and reproduced from ref. [62], with permission from CellPress, Copyright 2018. **b** An illustration showing how the degree of crosslinking density and the grafting density of the adhesion group affect the polymer binder's backbone stiffness, as well as the variations in electrode cracking produced by PVDF and catechol binders. Modified and reproduced from ref. [63], with permission from American Chemical Society, Copyright 2019. **c** Schematic diagram of adhesion measurement for catechol-conductive binder and SEM image of Si particles. Modified and reproduced from ref. [64], with permission from American Chemical Society, Copyright 2018

binder exhibits excellent moisture-resistant adhesion and superior mechanical strength due to its three-dimensional network structure. Using this catechol binder in Si nanoparticle (SiNPs) based anodes achieved a capacity retention of 91.5% after 100 cycles. Compared to electrodes using PVDF binders, the anodes with catechol binder showed reduced electrode cracking, enhancing the structural integrity during cycling (Fig. 3b) [63]. Finally, as shown in Fig. 3c, a copolymer of catechol and conductive pyrene was synthesized and applied as a binder in LIBs. The strong adhesive properties of catechol prevent the separation and volume expansion of Si particles, while the π-π stacking of pyrene enhances electron mobility, thereby improving the energy density and overall performance of the LIBs [64]. This conductive binder is proposed as a solution to address the drawbacks of full-conjugation polymers, such as high cost and complex multistep synthesis processes.

To enhance the performance of various rechargeable batteries, the thin and uniform coating capabilities of polydopamine (PDA) were leveraged to coat different battery active materials. Notably, PDA contains about 4.5 wt% nitrogen, which allows for nitrogen doping through the application of PDA coatings [65, 66]. This doping provides increased electrical conductivity and expands the number of binding sites through the utilization of nitrogen's lone pair electrons. Furthermore, the close bonding of active particles with polymeric binders and conductive materials, facilitated by the PDA coating, ensures the integrity of the electrode is preserved despite significant volume changes in the active materials. However, controlling the thickness of the PDA layer is crucial; an excessively thick PDA layer can increase electrical resistance, thereby impeding electrochemical performance.

Coating active materials with catechol protects them from inappropriate side reactions due to electrolyte decomposition and suppresses agglomeration between particles. This is particularly beneficial for high-capacity active materials such as sulfur, Si, and tin oxides. In Fig. 4a, it is observed that the PDA coating on sulfur, Si, and SnO_2 particles protects the active materials and manages volume expansion, thereby enhancing battery performance. Catechol coating forms a stable interface between the active materials and the electrolyte, suppressing side reactions and maintaining the conductivity of the electrode to improve the stability of the electrode structure [67]. Furthermore, as shown in Fig. 4b, utilizing catechol coating to form a core–shell structure has shown various improvements in battery electrode performance. Catechol coating, as demonstrated in Fig. 4a, suppresses the volume expansion of active materials, enhances electrical conductivity, and minimizes reactions with the electrolyte, thereby enhancing battery lifetime [68, 69].

In Na-ion batteries, the PVDF binder commonly used in the cathode suffers from issues such as low conductivity, poor compatibility with Na ions, and limited durability [70–72]. Specifically, the compatibility between PVDF and Na ions affects the electrolyte interaction, preventing the battery from achieving optimal performance. Figure 4c shows the formation of a cathode electrolyte interphase (CEI) layer using a silk fibroin-catechol water-soluble polymer binder for the Na-ion battery cathode. This layer prevents side reactions, suppresses electrolyte decomposition, and enhances reaction kinetics. The silk fibroin-catechol binder forms strong hydrogen bonds with the electrode, improving the mechanical integrity of the electrode and facilitating Na ion diffusion. As a result, the Na-ion batteries demonstrated excellent cycling stability and outstanding rate performance at high current densities [73].

LIBs are being developed with the addition of polymer additives (linear PEG/PEO) to liquid electrolytes to enhance ionic conductivity. PEG serves as a component of polymer electrolytes and helps to form a gel-like structure of the electrolyte, reducing the likelihood of leakage and enhancing the overall safety of the batteries [74, 75]. However, PEG or PEO can exhibit reduced electrochemical stability and mechanical strength at high temperatures.

To address the limitations of polyethylene glycol (PEG) and enhance ionic conductivity, Baek et al. synthesized a novel catechol binder (DPA-binder) by copolymerizing dopamine-*m*-acrylamide (catechol) with PEG-acrylate. Additionally, to resolve the issue of high crosslinking density in polymer networks while ensuring adequate cohesiveness, the system was crosslinked with 2% polyethylene glycol diacrylate (PEGDA) for application in Na-air batteries and LIBs [76, 77]. The DPA-binder exhibited strong binding affinity to Pt catalysts, maintaining adhesion to the carbon current collectors even in aqueous environments, thus exhibiting superior battery performance. In contrast, the PVDF binder showed low adhesion, resulting in detachment from the catalyst during the charge/discharge cycles of Na-air batteries (Fig. 5a) [76]. In LIBs, the 3D network structure facilitated by the DPA-binder showed excellent adhesion and ionic conductivity due to the PEG groups (SBR/CMC 6.49×10^{-6} S/cm vs. catechol binder 1.42×10^{-5} S/cm). Particularly, in-situ TEM studies revealed that the volume change of SiNPs was dramatically suppressed by the DPA-binder (Fig. 5b) [77]. Additionally, a copolymer binder incorporating anthracene-acrylate, catechol, and PEG-acrylate was developed to further enhance both adhesion and electrical conductivity (Fig. 5c). The anthracene groups within the catechol-based binder promoted increased semi-crystallinity and electron mobility through π–π stacking interactions. As a result, this binder enabled lower charge/discharge overpotential and delivered stable long-term cycling performance over 300 cycles in Na–air batteries [78].

These advanced catechol-based binders offer excellent structural stability, strong interfacial adhesion, and improved

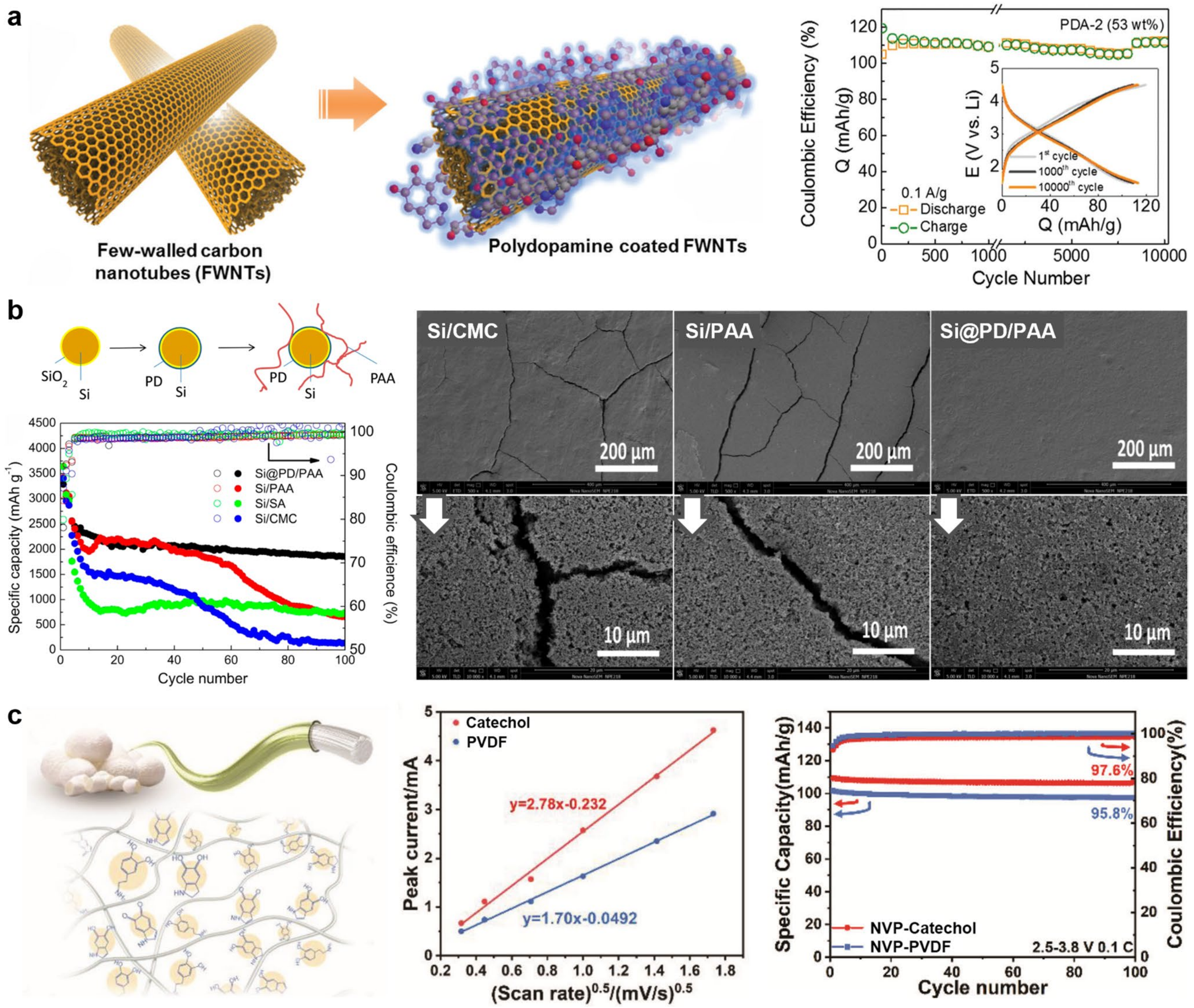

Fig. 4 **a** Oxidative self-polymerization reactions of polydopamine in lithium-ion batteries and the continuous coating process of polydopamine on the surface of few-walled carbon nanotube. Modified and reproduced from ref. [67], with permission from The Royal Society of Chemistry, under CC by 3.0 license. **b** Effects of Polydopamine (PDA) coating on silicon particles and electrodes (SEM image and cycling performance). Modified and reproduced from ref. [68], with permission from American Chemical Society, Copyright 2016. **c** Schematic illustration of catechol binder (left) and cycling performance (right). Modified and reproduced from ref. [73], with permission from Wiley-VHC, Copyright 2024

ion–electron conductivity, making them highly promising candidates for use as anode binders in high-performance air batteries and LIBs. Equipped with multiple functional advantages that traditional binders lack, these systems are particularly suitable for research and development efforts aimed at achieving high-capacity and fast-charging energy storage devices. Table 1 summarizes the electrochemical performance of major catechol-based binder systems, focusing on key indicators such as capacity retention, adhesion strength, and cycle life. The comparison highlights how catechol-based binders deliver markedly better interfacial adhesion and structural stability than conventional systems.

3.3 Other examples of using catechol groups in batteries

3.3.1 Separator coating

Separators are porous polymer membranes found between the anode and cathode of batteries. The primary function of a separator is to insulate the anode from the cathode, thereby preventing any short circuits between them. Additionally, separators act as vessels for the liquid electrolyte; hence, they are responsible for ionic conductivity. Separators must also remain electrochemically stable, avoiding detrimental

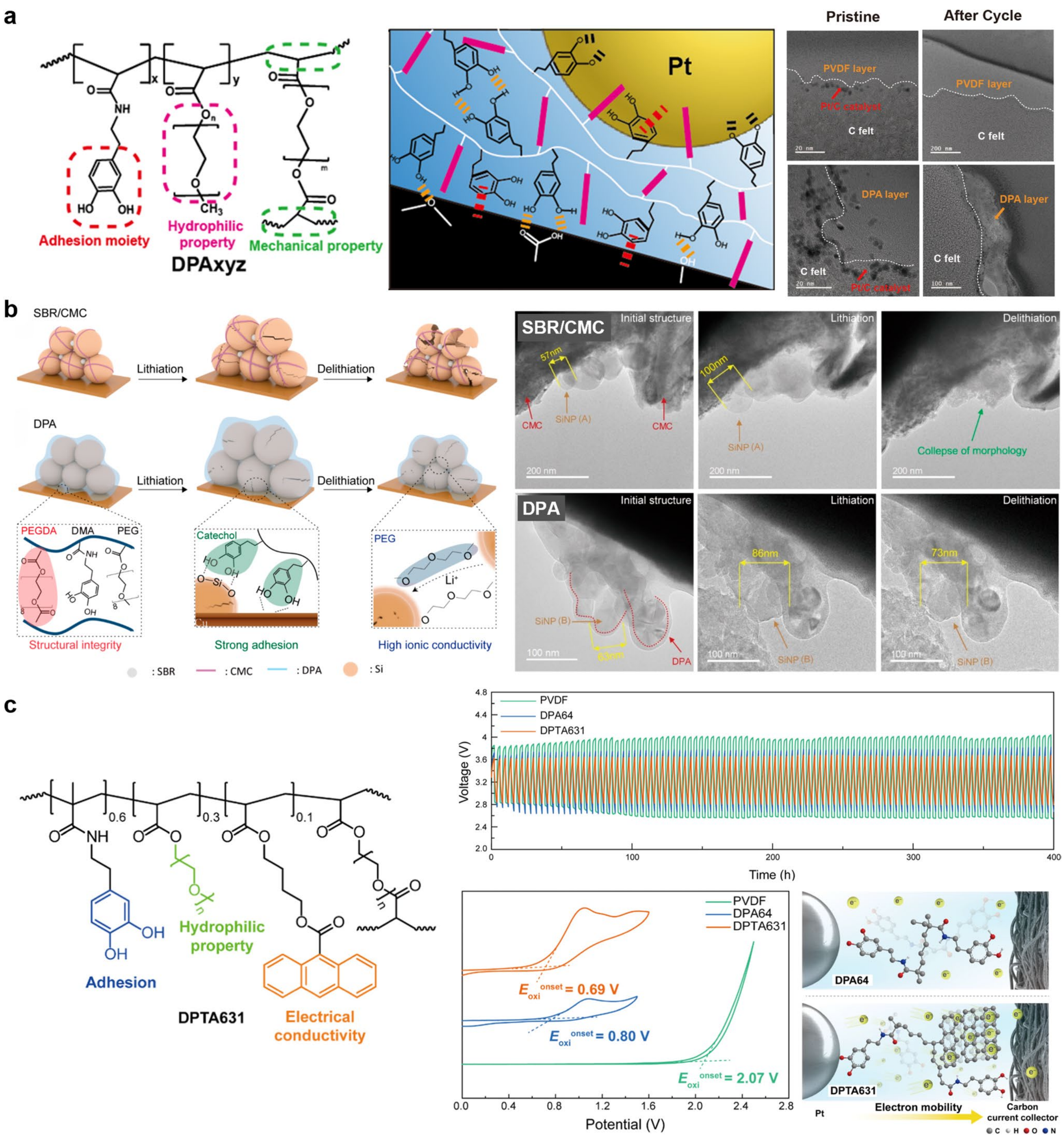

Fig. 5 3D network catechol binder with excellent adhesion and ion/electron conductivity. **a** Schematic diagram of the binding between synthesized catechol binder/Pt catalyst and high-resolution transmission electron microscope (HR-TEM). Adapted from our previous work ref. [76], The Royal Society of Chemistry, Copyright 2022. **b** Schematic illustration of the synthesized catechol binder and in-situ TEM images. Adapted from our previous work ref. [77], American Chemical Society, Copyright 2022. **c** Schematic illustration of catechol-electron conductive binder (left) and cycling performance (right). Adapted from our previous work ref. [78], The Royal Society of Chemistry, Copyright 2025

side reactions with the electrolyte when subjected to anodic or cathodic bias. It is well-known that modifying the surface properties of separators can significantly enhance key battery performance metrics such as energy density, fast charging capabilities, and cycle lifetime [79, 80]. Notably, recent advancements have shown that polydopamine

Table 1 Electrochemical performance of binder systems

Binder system	Cell / Electrode	Capacity retention	Adhesion strength	Cycle life	Reference(s)
PVDF	LIB / Anode	37 mAh/g @200 cy	–	200 cy	[15, 21, 22]
SBR/CMC	LIB / Anode	~900 mAh @300 cy (90%)	0.40 ± 0.05 N/cm	200 cy	[26, 77]
Alginate-Catechol	LIB / Anode	805.8 mAh/g @150 cy (84.5%)	370 pN (AFM)	400 cy	[57]
Catechol–Chitosan	LIB / Anode	2144 mAh/g @100 cy (91.5%)	~5.0 mN/m-	100 cy	[63]
Pyrene–Catechol	LIB / Anode	~800 mAh/g @100 cy	99 ± 59 pN (AFM)	100 cy	[64]
PDA-PAA-PEO	LIB / Anode	1597 mAh/g @200 cy	1.76 N	200 cy (0.5C)	[59]
DPGP–PEI/PVDF	LIB / Anode	153 mAh/g @1000 cy (80%)	~70 N/m	1000 cy	[60]
PAA-P(HEA-co-DMA)	LIB / Anode	2394 mAh/g @220 cy (93.8%)	0.83 N	400 cy	[62]
PDA-NCTs	LIB / Anode	1103 mAh/g @500 cy	–	500 cy	[66]
Catechol-PEG-PEGDA (DPA)	LIB / Anode	1100 mAh/g @1000 cy (~100%)	0.88 ± 0.08 N/cm	1000 cy	[77]
PDA	LIB, SIB / Anode	1414 mAh/g; @500 cy (93.0%)	–	580 cy (LIB) 1024 cy (SIB)	[90]
Na-PAA	SIB / Anode	175.5 mAh/g, 76.0% @300 cy	–	300 cy	[25]
Silk fibroin-Catechol (SFPDA)	SIB / cathode	91.3 mAh/g, 92.3% @1000 cy	2.3 N	1000 cy (10 C)	[73]
Catechol-PEG-PEGDA (DPA46)	Na-air / Cathode	~90% @100 cy	107.6 mN (SFA)	100 cy	[76]
Anthracene–Catechol–PEG (DPTA631)	Na–air / Cathode	86.7% @300 cy	64.8 mN (SFA)	300 cy	[78]

(PDA) coating on separators improves the electrochemical performance of traditional lithium-ion battery (LIB) electrodes, lithium metal anodes, and high-capacity post-LIB electrodes. The catechol coating on the separator enhances its wettability with electrolytes, thereby improving charging speed and cycle performance. Catechol-coated separators also contribute to mitigating lithium dendrite growth that may occur with the use of lithium metal anodes.

Figure 6a provides a schematic description of the design of a polydopamine-based redox-active separator. The layer containing porous, redox-active polydopamine (PDA) and carbon nanotubes (CNTs) is designed to be in contact with the anode. Because of the capacity contribution from the redox-active (RA) layer, the actual amount of anode material required can be reduced, allowing the cell's capacity to be maintained while its volume is decreased [81]. Typically, polyethylene (PE) separators exhibit hydrophobic surfaces, leading to poor wettability with electrolytes, which adversely affects the power performance and durability of batteries. PE separators treated with polydopamine (PDA) demonstrated increased hydrophilicity, resulting in enhanced electrolyte wettability as shown in Fig. 6b. Consequently, this treatment improved ionic conductivity during the charging and discharging processes, thereby enhancing the overall performance and lifetime of the batteries [82]. Fig. 6c depicts how catechol moieties tightly attach to the lithium metal surface, lowering surface tension at the electrode and thereby inhibiting lithium dendrite development. The catechol groups maintain strong adhesion during both the deposition and dissolution of lithium at the electrode, lowering the surface tension and resistance of the lithium metal during the charge–discharge processes of the LIB, thereby enhancing battery performance [83, 84]. Additionally, PDA-coated cellulose microfibrillated membrane has been demonstrated as a potential high-performance separator for lithium-ion batteries. This separator exhibited superior mechanical strength, excellent thermal dimensional stability, and outstanding electrochemical properties. Lithium cobalt oxide/graphite cells utilizing this membrane showed better cycling stability and rate performance compared to cells using commercial polypropylene separators and pristine cellulose separators. Furthermore, alternating current impedance measurements after the 100th cycle revealed excellent interfacial stability in the cells with the PDA-coated separator (Fig. 6d) [85].

3.3.2 Anti-oxidant

Catechol groups in polydopamine (PDA) have been utilized as electrolyte additives in both lithium-oxygen (Li–O_2) and lithium-ion batteries. Li–O_2 batteries, which rely on a simple oxygen reduction and evolution reaction ($2Li + O_2 + 2e^- \rightarrow Li_2O_2$), have garnered significant attention due to their high theoretical specific energy density of approximately 3500 Wh/kg. However, the utilization of oxygen leads to the formation of superoxide intermediates (O_2^-), catalyzing unwanted side reactions such as the decomposition of carbon materials and electrolytes, and the oxidation of catalysts. These side reactions diminish the Coulombic efficiency and cycle stability of the batteries [86].

Inspired by the melanin in human eyes and possessing a similar structure, PDA has been introduced to Li-O_2 batteries to effectively scavenge excess superoxide radicals

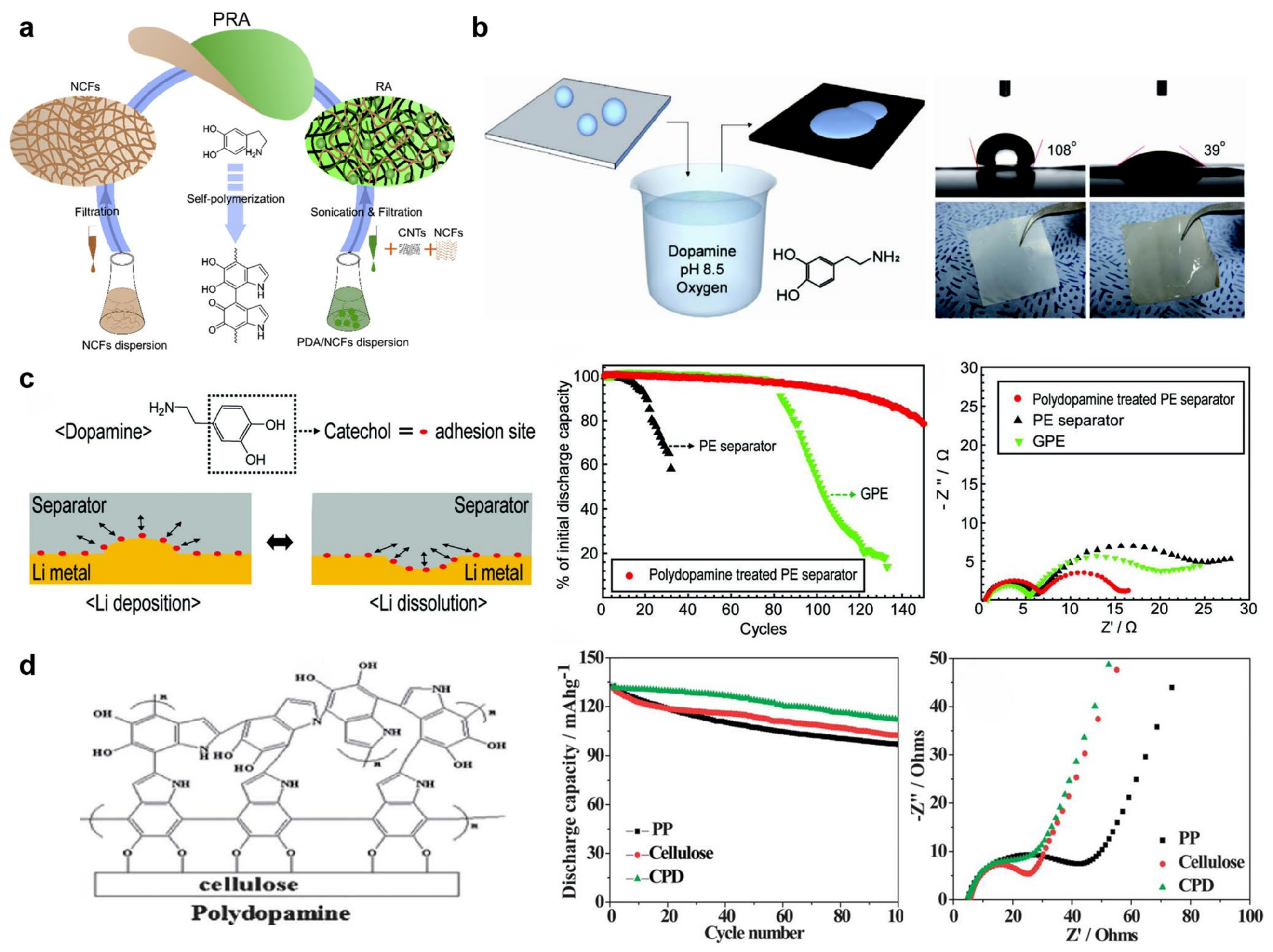

Fig. 6 **a** Schematic illustration of the process used in the manufacturing of the PRA separator. Reproduced from ref. [81], under the CC BY-NC-ND license, Copyright 2019, Elsevier. **b** Schematic of polydopamine surface treatment, the contact angle and wettability of PE separators. Modified and reproduced from ref. [82], with permission from Wiley-VHC, Copyright 2011. **c** The effect of the catechol adhesive properties of separators during lithium deposition/dissolution, and the battery characteristics influenced by the separators. Modified and reproduced from ref. [83], with permission from Wiley-VHC, Copyright 2012. **d** Schematic illustration of the polydopamine-coated microfibers and batteries performance. Modified and reproduced from ref. [85], with permission from The Royal Society of Chemistry, Copyright 2014

(Fig. 7a) [87]. Cells lacking PDA exhibited a substantial decrease in capacity around the 60th cycle, indicating a decline in performance. In contrast, cells with PDA incorporated into the electrolyte sustained operational capacity for more than 100 cycles. Scanning electron microscopy (SEM) analysis post 10 cycles showed that cells with PDA maintained the original morphology of carbon nanotubes (CNTs), whereas those without PDA were covered with a solid electrolyte interphase (SEI) layer [87].

Additionally, PDA can be applied as an additive in high-voltage LIBs to form a protective layer on the surface of the lithium nickel manganese cobalt oxide ($LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$) cathode. The primary role of this PDA coating is to protect $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ from adverse reactions at high voltages (4.5 V) and to prevent the oxidative degradation of the electrolyte (Fig. 7b) [88].

3.3.3 Organic electrode materials (OEM)

OEMs can be tailored to exhibit various electrochemical properties through molecular design, and they possess environmentally friendly characteristics [89]. Fig. 8a shows that catechol-structured PDA was prepared through oxidation and heat treatment, serving as both the electrode material and binder in Na-ion battery. The charge–discharge results of the Na-ion battery demonstrated a stable discharge capacity of 508 mAh/g with nearly 100% coulombic efficiency retained after 1000 cycles at a current density of 50 mA/g.

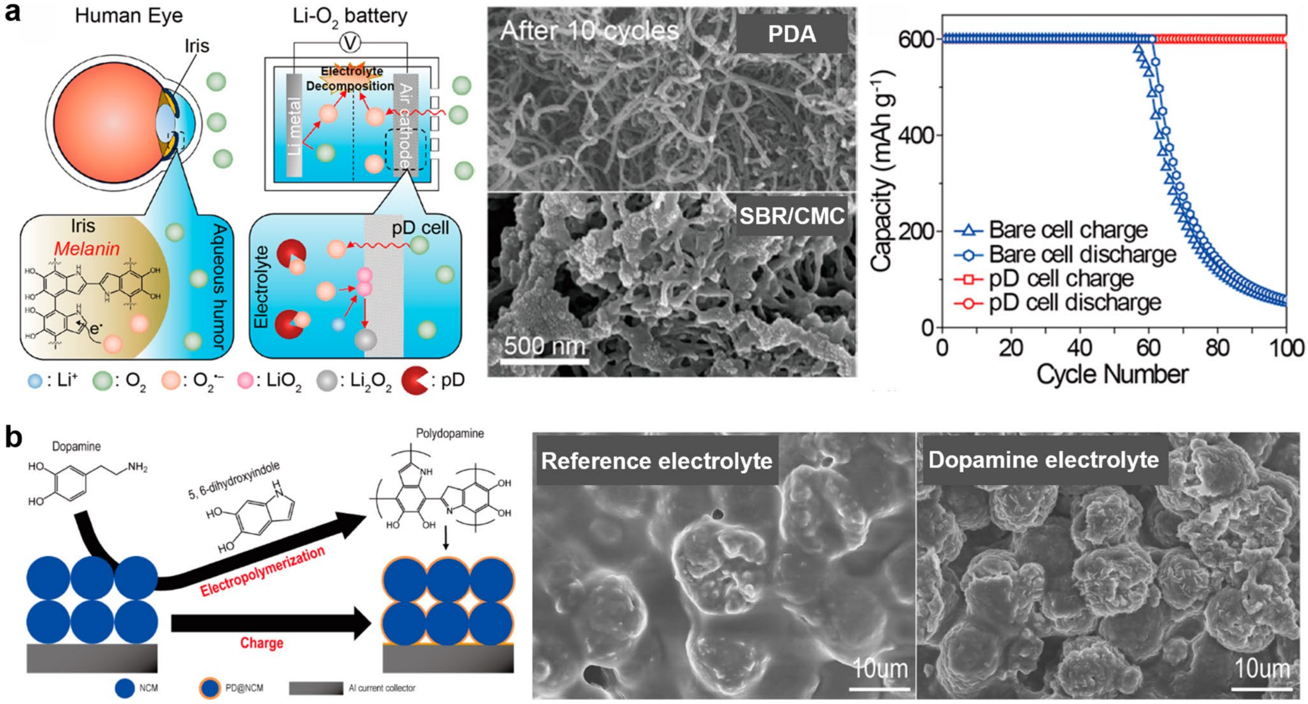

Fig. 7 **a** Schematic of PDA in $Li-O_2$ batteries' electrolyte and cycling performance. Modified and reproduced from ref. [87], with permission from American Chemical Society, Copyright 2014. **b** The application of dopamine as an electrolyte additive to facilitate the electrochemical formation of passivation layers on cathodes of high-voltage LIB. Modified and reproduced from ref. [88], with permission from American Chemical Society, Copyright 2014

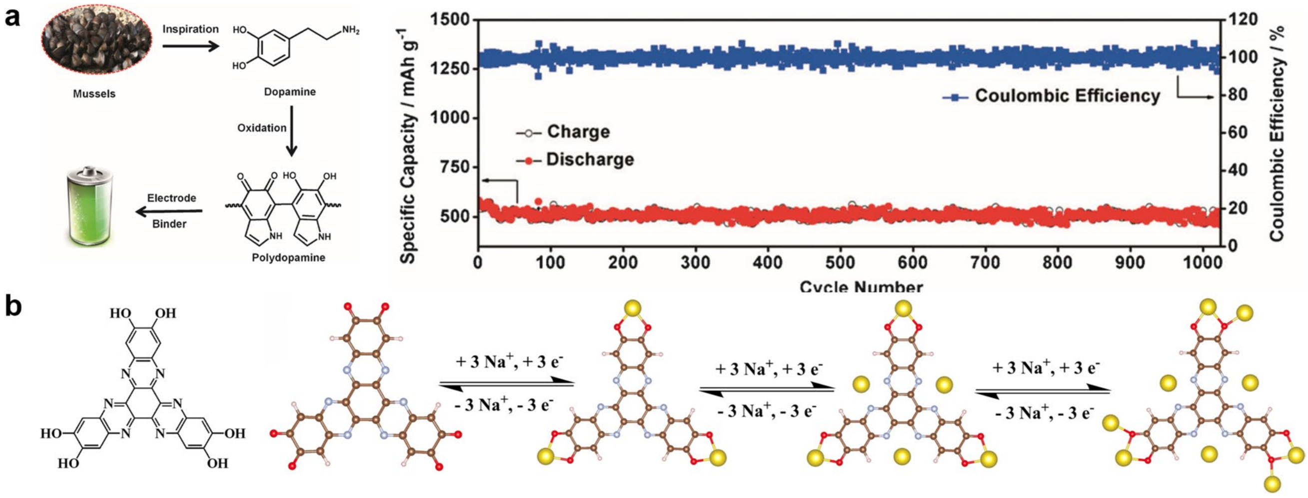

Fig. 8 **a** The application of polydopamine as an electrode material and redox-active binder in Na-ion batteries (SIBs). Modified and reproduced from ref. [90], with permission from Wiley, Copyright 2016. **b** The structure of organic electrode material (OEM) and molecule structural evolution during sodiation and desodiation process in Na-ion batteries (yellow ball: Na ion). Modified and reproduced from ref. [91], with permission from Wiley-VHC, Copyright 2023

The PDA provided strong mechanical stability by binding the electrode material to the current collector, while the quinone-catechol system in PDA enhanced electron transfer and electrochemical reactions at the electrode. The PDA served as both the electrode material and redox-active binder, maximizing electrochemical performance and resulting in highly stable battery performance in Na-ion batteries [90]. Fig. 8b demonstrates the synthesis of 2, 3, 8, 9, 14, 15-hexaolhexaazatrinaphthalene (HATN-6OH) with catechol groups, used as an organic electrode material. HATN-6OH stores and releases Na ions through the coupling of redox-active catechol/ortho-quinone groups with the HATN structure during sodiation and desodiation processes, exhibiting a high electron transfer capability via a nine-electron transfer mechanism. Moreover, the π-π interactions and hydrogen bonding between the molecules enable the electrode to achieve stable long-term cycling performance over 3000 cycles [91].

4 Current limitation for using catechol binders and future directions

As previously discussed, catechol binders have overcome the limitations inherent in traditional binders such as PVDF, PAA, and CMC. Notably, catechol binders exhibit excellent adhesion to silicon and have resolved issues related to volume expansion and electrode cracking during the charge/discharge cycles, thereby enabling the achievement of high energy LIBs [58, 63]. Beyond performance aspects, the scalability, industrial compatibility, and cost considerations of catechol-based binders must be carefully evaluated for future adoption. Many reported systems rely on multi-step syntheses or expensive catechol derivatives (e.g., dopamine monomers), which hinder large-scale production [5, 55]. In addition, the synthesis and purification of high-purity catechol remain costly, representing a significant barrier to commercialization [92]. From the perspective of industrial compatibility, seamless integration into existing slurry preparation and coating processes optimized for PVDF or SBR/CMC binders is critical. Processing-related challenges also arise, including viscosity control, solubility, and coating uniformity. Premature oxidation and crosslinking during roll-to-roll electrode fabrication may further compromise manufacturing stability [84]. Encouragingly, water-soluble catechol-functionalized polymers (such as catechol-conjugated alginate, PAA, or chitosan) have enabled electrode processing under environmentally benign conditions, offering advantages for scale-up [93]. However, the raw material cost of catechol remains higher than conventional binders, necessitating cost-reduction strategies such as partial functionalization of inexpensive backbones or the use of bio-derived catechol resources [5, 55]. In addition, when catechol is exposed to basic conditions under aerobic environment, it can suffer from oxidative coupling, forming bisquinone [93, 94]. This reaction is largely irreversible and leads to undesirable increases in crosslinking density within the binder network. While higher crosslinking may improve stiffness, it reduces flexibility, weakens interfacial adhesion, and deteriorates electrochemical reversibility. In practical electrodes, such oxidative transformations compromise bonding between active particles and current collectors, resulting in particle detachment, electrode cracking, and shortened cycle life [84, 92]. Moreover, these reactions are accelerated by oxygen and moisture, meaning that premature oxidation can readily occur during large-area roll-to-roll electrode fabrication or long-term electrode storage, further reducing process stability and commercial viability.

For practical application high-energy–density LIBs, future catechol binders must address several key aspects. Firstly, the binder should be low-cost, environmentally friendly, water-soluble, and possess excellent adhesion properties. Secondly, it should have the flexibility and adequate mechanical strength to mitigate the volume expansion issues associated with silicon particles. Thirdly, the binder network should be designed to include functional groups that modulate electron density, thus preventing the easy oxidation of catechol. Finally, the binder, while being a polymeric insulator, must enhance the ion- and electron-conductivity to improve the performance of the LIBs.

5 Conclusion

In this review, we have summarized various studies on catechol binders that could replace traditional polymeric binders to enhance the performance of batteries. Generally, polymeric binders must ensure uniform dispersion of active materials and conductive additives, as well as good electrochemical stability of the battery components. Additionally, LIBs must address the substantial volume expansion (about 400%) of Si anode materials and increase mechanical stability during the charge/discharge cycles. It has been clearly described that the mechanical properties of polymeric binders critically influence Si-based anode materials, which undergo significant volume changes. Among the different methods explored to achieve high-performance silicon-based anodes, one of the most effective strategies is to design mechanically strong binders that can control significant volume expansion. Catechol-based binders exhibit excellent electrochemical and thermal stability, and they improve the charge/discharge cycle stability and rate performance of LIBs. However, catechol binders can suffer from a gradual increase in crosslinking density due to the oxidative coupling reaction, leading to a decrease in adhesion strength and electrode cracking issues. To overcome these limitations, the development of new catechol binders with enhanced adhesion, high ionic/electrical conductivity, and a multifunctional 3D network structure is required.

Through this review, we have identified the potential possibilities and limitations of catechol binders and anticipate that they will contribute to the development of improved binder materials for high-performance batteries. This is expected to facilitate the commercialization of next-generation batteries and greatly enhance the application possibilities in large-capacity energy storage devices.

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