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Review article

Designing multifunctional binders for next-generation batteries: Strategies to improve longevity, safety, and electrochemical performance

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ABSTRACT

The global transition toward clean and sustainable energy has intensified the demand for efficient and durable energy storage systems, particularly rechargeable batteries for electric vehicles. Beyond active materials, the binder plays a crucial yet often underappreciated role in determining factors including electrode integrity, mechanical robustness, and ion/electron transport pathways that directly influence battery performance, longevity, and safety. Conventional binders such as polyvinylidene fluoride (PVDF) suffer from poor adhesion, limited mechanical flexibility, and low tolerance to electrode volume fluctuations, leading to structural degradation and capacity fading. Recent advancements in binder design have focused on multifunctional systems that integrate self-healing capabilities, enhanced ionic/electronic conductivity, and dynamic cross-linked architectures. Strategies including reversible bonding, supramolecular interactions, and three-dimensional polymeric networks have shown great promise in addressing the limitations of traditional systems. This review critically discusses these emerging developments, emphasizing the mechanistic insights and design principles that guide the evolution of binder chemistry. Finally, it highlights technological directions for developing next-generation, sustainable binders tailored for advanced battery chemistries and large-scale industrial implementation.

1. Introduction

The accelerating transition toward sustainable energy systems has intensified the demand for high-performance rechargeable batteries that can power electric vehicles, portable electronics, and grid-scale storage [1]. While significant progress has been made in electrode materials and electrolytes, the binder, often considered an inactive component, plays a crucial yet underappreciated role in determining the overall electrochemical performance, mechanical integrity, and safety of battery systems [2]. The composition of the electrode, including active materials, binders, metal current collectors and conductive additives are shown in the Fig. 1a [3,4]. Despite their small weight fraction, polymeric binders critically determine electrode cohesion, interfacial stability, and electrochemical durability. However, their roles remain insufficiently optimized due to complex trade-offs among adhesion strength, ionic conductivity, and chemical stability in reactive environments. Recent developments in polymeric binders demonstrate that strategic incorporation of polar groups, dynamic cross-linking motifs, and conductive backbones can substantially mitigate capacity fading, enhance rate capability, and improve electrode stability [5].

Binder is generally a polymer or polymer-based material with ~5–10 % of total battery weight and it is the only inactive material used in the fabrication of electrodes in batteries [9]. Primarily, the selection, composition and concentration of binders differ from cathode to anode materials according to battery characteristics such as long-term cyclability, rate capability and other required working parameters [10]. An electrode slurry is typically synthesized by mixing the active materials and conductive additives along with the binder into the solvent. After mixing, the slurry is then cast onto the current collector foil as shown in Fig. 1b. Fig. 1c illustrates the ideal connection and adherence of active and conductive material with the help of binders. Therefore, the strong adherence of the binder is important as it provides high mechanical stability, ionic/electronic conductivity, and overall increases battery lifespan [11]. Binders are generally categorized into two genres – aqueous (polar) and non-aqueous binders (non-polar). Non-aqueous binders are dissolved in organic solvents rather than water and it does

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not possess a dipole moment. Aqueous binders are soluble in water and are typically used in water-based slurry processing. These binders contain functional groups that have an electrical dipole or charge distribution.

The PVDF binder was used as the primary binder when LIBs were commercialized in 1991 because of its excellent adhesive properties and good electrochemical stability [12]. Despite the popularity of PVDF binder in battery management systems, it has largely suffered from high-capacity electrodes in large volume changes because of its non-functionalized linear chain structure [13,14]. Since PVDF is a non-aqueous binder, it is dependent on N-Methyl-2-pyrrolidone (NMP) for casting, which makes it difficult to recycle as a secondary material in end-of-life batteries, leading to environmental issues [15]. Although polyvinyl alcohol (PVA) is a non-aqueous binder, it dissolves in water since it has numerous hydroxyl groups, which makes a strong interaction with other functional and forms a strong bond between active materials [16]. Unfortunately, it fails to provide good mechanical properties, which ultimately leads to electrode degradation during the electrochemical process. Apart from that, polyacrylonitrile (PAN) binder outperforms PVDF due to the presence of nitrile groups. However, it exhibits high brittleness and poor solubility compared to the PVA binder [17].

Recently, researchers and industries have been advancing toward the usage of water-soluble binders and developing novel functional binders for battery technology. This shift works to reduce reliance on toxic solvents, address environmental issues, and enhance battery performance under high-voltage conditions [18–20]. In addition, an aqueous binder like polytetrafluoroethylene (PTFE), styrene-butadiene-rubber (SBR), carboxymethyl cellulose (CMC) and polyacrylic acid (PAA) is preferred over non-aqueous binders as it is inexpensive with excellent mechanical properties, strong adhesion and does not require toxic solvents, ensuring a sustainable environment [21]. Nevertheless, it is found that solvent-free processes may not always be the optimal replacement for slurry casting, as specific relationships between microstructure, electrode formulation, and mechanical properties can sometimes fail to align, ultimately impacting electrochemical performance [22,23]. Hence, researchers have introduced techniques such as modifying simple chemical groups and incorporating functional components with thoughtfully designed architectures into binders to further enhance their performance and consequently, improve battery performance [24].

With the exploration of various advancements to bring the efficacious in battery technology, this review aims to provide detailed insights into binders, their types, functions, influencing factors, challenges, and alternative solutions. In recent years, there has been a growing surge of publications on battery binders, describing binders' design and development [9,20,25–27]. While several review papers have been covered, it still remains essential and timely to summarize the latest advancements in binder technology for the future development of batteries with high efficiency. Unlike previous reviews, this article offers insights into the multifunctional design of binders, highlighting various alternative advancement techniques introduced in recent years. Most importantly, it helps in understanding the unique strengths of individual binders that enable the perfect blending or crosslinking of different binders to create composite materials, aiming to achieve more comprehensive and enhanced improvements in battery electrodes. Moreover, this review systematically discusses the evolution of binder chemistries for next-generation batteries, emphasizing the structure–property–performance relationship, emerging multifunctional systems, and industrial feasibility. Special attention is devoted to quantitative comparisons and future perspectives linking binder design with sustainable large-scale fabrication.

2. Binder in batteries: properties, mechanism and challenges

Despite its importance, the binder offers particular advantages while also imposing certain limitations. Table 1 provides a comparative overview of key binder properties, outlining their respective strengths and drawbacks within battery systems.

2.1. *Binding mechanisms and interfacial interactions of the binder in electrodes*

Strong adhesion between the active/conductive materials and the current collector is essential for achieving superior electrochemical performances. This clearly highlights the direct and significant influence that binders have on overall battery behaviour [15]. The binding mechanism is illustrated in two processes - the diffusion/penetration process (Fig. 2a) and mechanical interlocking process (Fig. 2b). Firstly, the non-reactive binders in the substrate get diffused and penetrate the pores of the active material during electrode fabrication. Later, the prepared electrode, which is kept for drying, gets hardened through different reaction mechanisms such as solvent evaporation, polymerization, phase transition, and electrochemical reactions, resulting in mechanical interlocking [39,40].

Apart from these processes, interfacial binding forces (Fig. 2c), a commonly used technique, enable the incorporation of binders into electrode materials and promote the binding strength via interaction forces, including covalent bonding, hydrogen bonding, coordinate

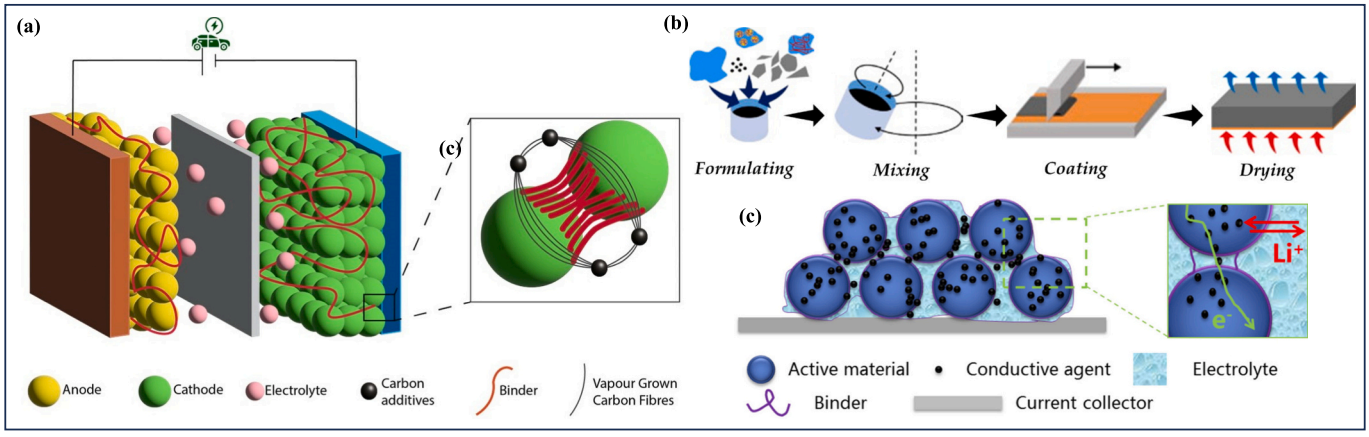

Fig. 1. (a) Diagrammatic illustration of battery components. Reproduced under terms of the CC-BY license. [6] Copyright 2024, The Authors, published by MDPI. (b) Overview of the electrode manufacturing process. Reproduced under terms of the CC-BY license. [7] Copyright 2021, The Authors, published by Elsevier. (c) Graphical illustration depicting the adhesion of binder with conductive additives and active materials. Reproduced under terms of the CC-BY license. [8] Copyright 2023, The Authors, published by Springer Nature.

Table 1
Strengths and weaknesses of binders in different property aspects.

Properties	Advantages	Disadvantages
Electrode integrity	Improves the mechanical stability of electrodes [28].	Excessive binders can increase internal resistance [29].
Material compatibility	Compatible with a variety of active materials and conductive agents [30].	Incompatibility with some organic solvents or specific active materials [30–32].
Processing	Facilitates electrode slurry preparation for uniform coating [9].	Processing requires specific solvents like NMP for PVDF, which may be toxic or expensive [9].
Adhesion	Provides strong adhesion between active materials, conductive agents, and current collectors [9].	Poor binder quality may lead to delamination during cycling.
Cycle stability	Enhances battery cycle life by maintaining electrode structure [25].	Over time, the degradation of binder may lead to capacity loss [28].
Environmental impact	The development of water-based binders (e.g., CMC, SBR) reduces environmental hazards [33].	Traditional binders (e.g., PVDF) often require toxic organic solvents [32].
Cost	Low-cost binders are available for some chemistries (e.g., CMC).	High-cost binders (e.g., PVDF) increase manufacturing expenses [25].
Ionic conductivity	Certain binders like PAA shows excellent ionic conductivity [31,34].	Most conventional binders are electronically and ionically insulating in nature [30,31].
Thermal stability	High-performance binders (e.g., PVDF) exhibit good thermal stability [35].	Some binders may degrade at elevated temperatures, affecting battery performance [36].
Mechanical properties	Maintains flexibility and resilience under volume changes (e.g., in silicon anodes) [37].	Poor binder elasticity may lead to electrode cracking during cycling [38].

bonding, and van der Waals [3]. The binding strength and contact have been categorized into three layers – bonded, fixed, and excessive polymer layer (Fig. 2d). The bonded polymer and fixed polymer layer are generally formed during electrode fabrication and drying process on the surface of materials while the excessive polymer layer is a free polymer surrounded over the fixed polymer layer. The excessive polymer layer is mostly weaker than the fixed polymer layer, as the fixed polymer layer has a good interaction with the bonded polymer layer [40].

2.2. Contact modes between binders and active materials: from point to network adhesion

Right after its binding mechanism, it is more considerable and imperative to discuss the interactions between active materials and binder over the mechanism, as it depicts the linkage of active particles, latex particles, binder chains, and cross-linkers. The interaction in electrode materials has been segregated into three kinds such as dot-to-surface contact, segment-to-surface contact, and network-to-surface contact, as shown in Fig. 2e-g. As soon as the substrate is mixed, coated, and dried, the latex particles get attached to the active particles on the surface and form point connections in dot-to-surface contact. This usually results in weak adhesion between active and latex particles (Fig. 2e). The dot-to-surface contact binders include emulsion binders such as SBR latex, polyacrylate latex, and PTFE latex. Likewise in segment-to-surface contact, the segment of the polymer chain sticks to the active particles on the surface and that leads to a moderate adhesion between the binder chain and active particles (Fig. 2f). The segment-to-surface contact binders include polymer binders such as CMC, PAA, PAN, PVDF, PVA, and polyacrylate latex. The network-to-surface contact binder is characterized by three-dimensional (3D) networks that are formed when coating a surface either thermally or chemically treated (Fig. 2g) [41]. In summary Table 2 provides a quantitative comparison of different kind of binder in battery system.

3. Classification of binders: aqueous vs. non-aqueous systems

The binders are divided into two different categories based on their solubility during the electrode preparation, such as aqueous (water-soluble) and non-aqueous (organic solvent-soluble), as shown in Fig. 2h, with each variant offering some advantages and challenges. The non-aqueous binder provides high scalability and good flexibility. However, it is harmful to the environment. The aqueous binder shows several advantages, such as excellent mechanical properties, adhesion of electrode materials, being eco-friendly in nature, being inexpensive, and good physicochemical properties. Most importantly, a battery with non-aqueous binders is expensive, and it is used only if the aqueous binder is not compatible with the desired battery. Non-aqueous binders like PVDF are commonly used for graphite anodes, $LiCoO_2$ cathodes, silicon-based anodes in LIBs, $NaMnO_2$ cathodes and hard carbon anodes in sodium-ion batteries (NIBs), and graphite-based anodes and metal oxide cathodes in aluminum-ion batteries (AIBs). Aqueous binders such as CMC, SBR, and PAA are widely used as binders for graphite anodes and silicon-based anodes in LIBs, hard carbon anodes for NIBs, and graphite anodes and

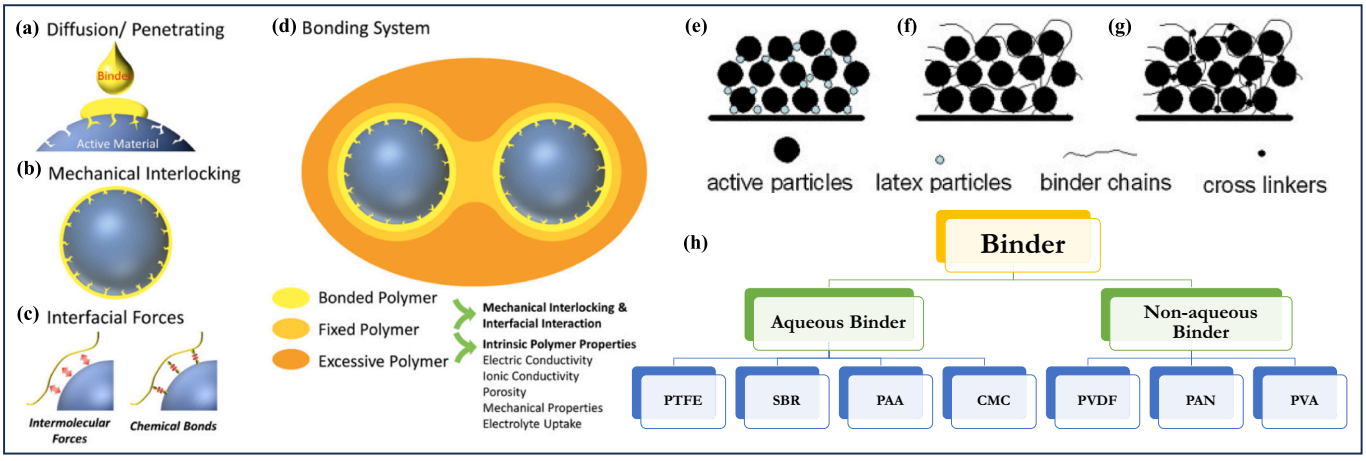

Fig. 2. Graphical representation of the binding mechanisms between binders and active materials - (a) Diffusion/penetration, (b) Mechanical interlocking, (c) Interfacial forces and (d) Bonding systems. Reproduced with permission. [40] Copyright 2018, The Authors, published by American Chemical Society. Graphical depiction of mechanical interactions between active materials and binder - (e) Dot-to-surface contact, (f) Segment-to-surface contact and (g) Network-to-surface contact. Reproduced with permission. [41] Copyright 2015, The Authors, published by Elsevier. (h) Classification of binders based on their solubility.

Table 2
Quantitative comparison of different kind of binder in battery system.

Binder	Battery System	Adhesion Strength (MPa)	Ionic Conductivity ($S \cdot cm^{-1}$)	Capacity Retention	Coulombic Efficiency	Advantage	References
PVDF	Lithium-ion cathodes (NCM/NCA)	~2–3	$\sim 10^{-8}$	~80–85 % after 200 cycles	>98 %	Strong chemical stability & hydrophobic in nature	[42]
PTFE	Dry-processed LIB cathodes	~30 MPa	$\sim 10^{-9}$–10^{-8}	~85–90 % after 200 cycles	>98 %	High elasticity and fibrillation, bridges the binder for dry-process	[43]
CMC	Si & graphite anodes	~3–4	$\sim 10^{-5}$	~85–92 % after 200 cycles (Si systems)	>99 %	Strong H-bonding, rigid backbone & water-processable binder	[44]
SBR	Commercial graphite anodes	~3–5	$\sim 10^{-5}$	~90–93 % after 300 cycles	>99 %	Elastic support & strong adhesion synergy	[45]
PAA	High-Ni cathodes / Si anodes	~4–5	$\sim 10^{-6}$	~92–95 % after 200 cycles	>99 %	Chelation with TM ions & enhanced interfacial stability	[46]
PAN	LIB cathodes (conductive binder)	~2–3	$\sim 10^{-4}$–10^{-5}	~88–92 % after 250 cycles	>98 %	N-rich backbone, oxidative stability & conductive film formation	[47]
PVA	Thick-loading Li—S cathodes	— (high flexibility)	$\sim 10^{-4}$	~90 % after 250 cycles (areal cap. ~4 $mAh \cdot cm^{-2}$)	~98 %	Strong film-forming, aqueous-processable & suppresses polysulfide leakage	[48]

metal oxide cathodes in AIBs. Table 3 presents a range of recently adopted binders along with their corresponding electrochemical properties, highlighting their relevance and application in current industrial battery technologies.

Table 3
Types of binders - molecular structures, applications, and their roles in energy storage devices.

Binder	Molecular Structure	Electrode Type	Applications	Roles in Electrodes	Influence on Electrochemical Performance
PVDF	-(CH_2-CF_2)n-	Positive (Cathode)	$LiCoO_2$, $Li(Ni_{1-x-y}Co_xMn_y)O_2$ (NMC), $LiFePO_4$ (LFP) cathodes	Strong adhesion, high thermal and chemical stability [9,36,49]	Maintains electrode integrity and ionically insulating reduces ionic transport [34,50,51]
PVA	-(CH_2-CHOH)n-	Positive and Negative	Both cathodes (e.g., LFP) and anodes (e.g., silicon, graphite)	Partially water soluble and provides good mechanical strength [52,53]	Offers flexibility and is used in flexible or wearable batteries [54]
PAN	-(CH_2-CH(CN))n-	Positive and Negative	High-performance LIB and supercapacitors	High thermal stability and provides fibrous flexibility [50,55,56]	Improves structural integrity and ensures performance in high-temperature systems [57,58]
PTFE	-(C_2F_4)n-	Positive and Negative	LIB, NIB, and supercapacitors	High chemical stability and Hydrophilic [32,59,60]	Enhances mechanical integrity and is used in high-voltage or harsh environments [61,62]
CMC	Linear cellulose with -COOH	Negative (Anode)	Silicon and graphite anodes	Water-based binder and strong particle binding [32,63,64]	Improves cycling stability and is eco-friendly avoids toxic solvents [65,66]
SBR	Copolymer of styrene-butadiene	Negative (Anode)	Graphite and silicon anodes	High elasticity and used with CMC for mechanical flexibility [10,61]	Accommodates volume changes and prevents cracking [67]
PAA	-(CH_2-CH(COOH))n-	Negative (Anode)	Silicon and high-capacity alloy anodes	Strong adhesion and elasticity for volume changes [35,46,68]	Enhances cycling stability for high-capacity materials [46]
Alginate	Polysaccharide with carboxyl	Negative (Anode)	High-capacity silicon and alloy anodes	Biodegradable and strong binding capability [69]	Reduces capacity fading and is environmentally sustainable [70]
Conductive Polymers (PEDOT: PSS)	Poly(thiophene) with sulfonate	Positive and Negative	Hybrid electrodes, supercapacitors	Conductive binder and enhances charge transfer [37,71]	Improves rate capability and is ideal for high-power applications [37,69]
Nafion	Sulfonated tetrafluoroethylene	Positive (Cathode)	Lithium-sulfur batteries, fuel cells	Proton conductive and ionic pathway enhancement [36,37]	Facilitates sulfur utilization in Li—S batteries and improves ionic transport efficiency [33]
Polyimides (PI)	Aromatic polyimide structure	Positive and Negative	High-voltage cathodes and silicon anodes	Excellent thermal and chemical stability [69,72]	Improves performance in high-temperature environments [73]
Polyethylene Oxide (PEO)	-(CH_2-CH_2-O)n-	Positive and Negative	Solid-state batteries, polymer electrolytes	Ionic conductivity and acts as a solid-state electrolyte [69]	Improves lithium-ion transport and key material for solid-state battery technology [70,74]
Gelatin-Based Binders	Polypeptide chains	Negative (Anode)	Silicon-based and flexible electrodes	Biodegradable and good mechanical properties [32,69,72]	Enhances flexibility and reduces environmental impact [14,72,73]
Acrylonitrile Butadiene Styrene (ABS)	-(CH_2-CH(CN)-CH_2-CH=CH)n-	Positive and Negative	Cathodes like LCO and NMC, and anodes like silicon	High thermal stability and mechanical resilience [55,69]	Improves electrode durability and limited ionic conductivity, requires optimization [33]
PEDOT:PSS-TFSI	Poly(thiophene) with sulfonate and trifluoromethanesulfonyl	Positive and Negative	Hybrid electrodes, supercapacitors	Enhances mechanical flexibility and electronic conductivity [75]	Improves conductivity, faster charge/discharge and reduces interfacial resistance [76,77]

3.1. Non-aqueous binder

3.1.1. PVDF

3.1.1.1. Material characteristics and binding mechanisms of PVDF in battery electrodes. PVDF binder is a nonpolar, fluorinated polymer with strong C—F bonds that provide excellent chemical and electrochemical stability. Its semi-crystalline structure ensures mechanical robustness, but the lack of polar functional groups limits interfacial adhesion with active materials [78]. The high electronegativity of fluorine can promote weak dipole interactions with oxide surfaces; however, PVDF primarily acts as a mechanical binder rather than participating in chemical bonding or ion coordination, leading to poor wettability and electronic conductivity [79]. PVDF binders have been used as binding agents in commercial and industrial processed batteries for the past decades because of their outstanding features [36,40]. PVDF binder requires organic solvents like NMP due to the instability of $LiCoO_2$ in aqueous environments to homogenize with a powdery electrode material and other components to create a paste that is applied to a current collector [80,81]. PVDF binder relies on physical bonding through van der Waals forces, which diminishes its mechanical strength and gradually reduces electrochemical performance over time.

PVDF limits high-voltage operation because it undergoes electrochemical oxidation, dehydrofluorination, poor high-voltage interfacial adhesion, and accelerated chemical breakdown in the presence of reactive high-Ni cathode surfaces and electrolyte oxidation products. These degradation pathways generate HF and resistive CEI layers, reduce mechanical integrity, and lead to rapid capacity fade when cells operate above ~4.3–4.5 V [36,82,83]. The physical interaction among active material, current collector (e.g., Al foil), and PVDF binder is depicted in Fig. 3a. Zhong et al. demonstrated a comparative study between LFP and NCM batteries to know the distribution of PVDF binder using the support of theoretical and simulation calculation results. The binding interaction between the Al, current collector and PVDF binder is weaker compared to the binding interaction between the active material and PVDF in the LFP battery. The binding interaction between the Al current collector and PVDF is stronger than the binding interaction between the active material and the PVDF binder in the NCM battery [49]. Therefore, the distribution of PVDF binder in electrode materials is irregular and may differ concerning the active materials and the current collector.

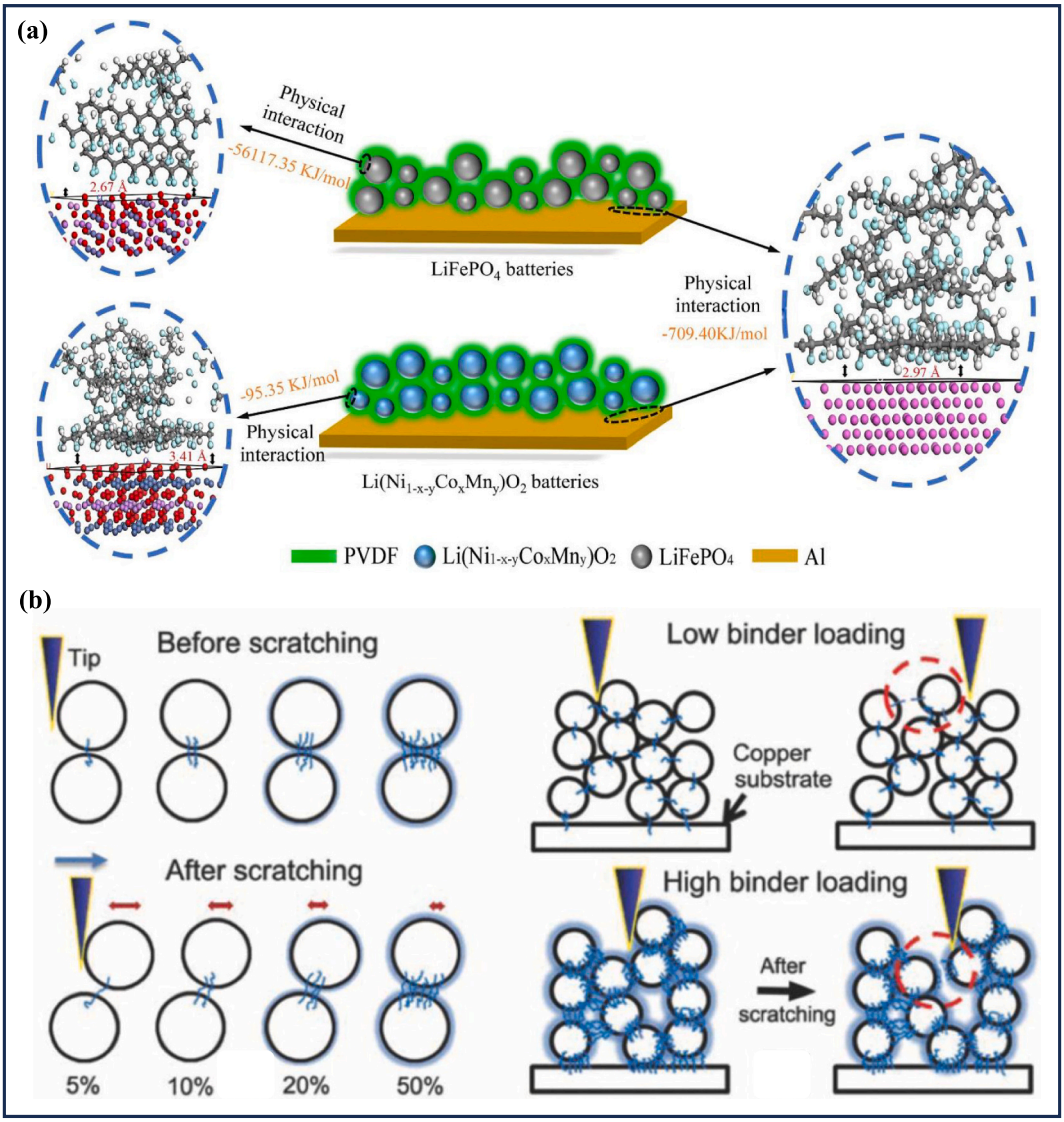

Fig. 3. (a) Physical interactions between the active material, PVDF binder, and current collector (Al). Reproduced with permission. [49] Copyright 2021, The Authors, published by Elsevier. (b) Micro-scratch process of MBMC/PVDF-coated copper electrode film in a composite model. Reproduced with permission. [38] Copyright 2013, The Authors, published by the Electrochemical Society.

3.1.1.2. Structure–property–performance relationship of PVDF-based binder. Indeed, PVDF binders have excellent mechanical properties, strong adhesion, electrochemical stability, processing properties, corrosion and oxidation resistance of high-energy C − F bonds [84,85]. Markevich et al. concluded that the PVDF binder increased the $LiCoO_2$ reactivity due to its tight contact, enhancing the reaction between $LiCoO_2$ and PVDF in composite electrodes (Co dissolution, Co_3O_4 formation) [86]. Yoo et al. demonstrated that hydroxyl functionalization of the PVDF binder improved its adhesion to the active material, resulting in a uniform binder distribution [87]. A recent study found that LIBs assembled with PVDF binder perform better under low-temperature conditions than those assembled with aqueous binder, such as SBR/CMC. This leads to a decrease in alternating current (AC) impedance and an increase in the specific power density of discharge capacity during operational tests. Overall, the PVDF binder-based electrode systems exhibit excellent electrochemical resistance and superior cycling stability even under low temperature conditions [10].

Several external parameters intermittently affect the role of binder, but the size and quantity of binder used in electrode preparation play a predominant role in influencing the battery performance. Studies have shown that the C—F bond formed between PVDF binder and active materials at the particle/binder interface was directly proportional to the particle/particle cohesion strength [88]. For example, Chen et al. tested the mechanical integrity of the binder at two different concentrations of binder. Typically, researchers initiate a crack using a micro-indenter tip, allowing it to penetrate the slurry and propagate along the interface between the current collector and the electrode film. Thereby, the electrode film gets delaminated from its substrate. The micro scratching process of MBMC/PVDF coated copper electrode film is depicted in Fig. 3b. The electrode with less binder gets dissipated easily by moving particles away from each other on the surface, reducing the fracture energy. In contrast, the electrode with high binder content finds it hard for the crack to penetrate and leaves densely packed particles in a zigzag manner. The decrease in binder content leads to a lesser number of ligaments at the particle/current-collector interface. Therefore, it is found that the binder with higher concentration results in high particle/particle cohesion and less piling up of materials compared to low binder content [38].

Although PVDF binder is known as a conventional binder and has been used for decades in industries, it is not always an ideal candidate due to its poor functionality, weak mechanical strength, and relatively low adhesiveness, especially toward high-energy-density or high-capacity batteries with a thick electrode [20]. In addition, it has lower flexibility and blocks the ion migration paths inside the battery systems [89]. The investigation of the oxidation or reduction process of PVDF binder during cyclic performances is still challenging, as it forms similar intermediates after its oxidation or reduction by the electrolyte [90,91]. For example, the commonly used electrolyte salt $LiPF_6$ undergoes degradation during cyclic performance, resulting in the formation of LiF or HF in the presence of water. Likewise, the PVDF binder leads to the formation of LiF or HF after reacting with lithium (Eq. 1 and Eq. 2).

$$Li + -[CH_2 - CF_2] - \rightarrow -[CH = CF] - + LiF + 1/2H_2 \quad (1)$$

$$-[CH_2 - CF_2] - \rightarrow -[CH = CF] - + HF \quad (2)$$

Nowadays, researchers are looking for alternatives instead of PVDF binder as it suffers from forming LiF by reacting with lithiated carbon, which immobilizes the Li^+, increases the impedance, and reduces the specific charge since LiF is a highly ionically and electronically resistive material [92,93]. In addition, PVDF binder degradation during cyclic performances also has crucial effects on battery operations, such as reduced mechanical stability and loss of contact between active materials and binder [93].

Despite the advantages of using the wet slurry-based process for the preparation of electrode materials in batteries, the use of toxic and volatile organic solvents for the casting process releases a toxic gas that raises environmental concerns [81]. According to the European Chemicals Agency, NMP is a substance of “very high concern” because it can cause cancer, mutagenesis, and reproductive harm to humans. Thus, it ultimately seeks a highly expensive environment with expensive protection equipment and ventilation facilities [89]. Moreover, the wet slurry-based process fails to achieve uniform distribution and effective fibrillation of electrode components, resulting in the creation of electrochemically isolated regions. In return, it fails to maintain the performance of battery cells and accelerates the deterioration in nearby regions [94]. Consequently, the insulating nature of PVDF necessitates the inclusion of carbon additives to improve the electrical conductivity of the composite electrode [95,96]. Overall, the PVDF binder is not the best fit for most of the new-generation batteries, especially while aiming to fabricate high-capacity batteries with a thin electrode.

3.1.1.3. Advanced modifications and alternatives to PVDF binders. Several possible measures have been introduced and developed from the existing findings, such as incorporating additives and forming composites to mitigate the challenges faced by the PVDF binder, especially the poor functionality, weak adhesion, and the use of volatile solvents. For example, Fu et al. confirmed with the results of EIS measurements (Fig. 4a) that the maleic anhydride-grated-PVDF shows reduced interfacial resistance compared to bare PVDF and enhanced lithium desertion/insertion kinetics in the cathode using anhydride-grated-PVDF [97]. In recent years, many approaches have been pursued to avoid organic solvents to develop more environmentally friendly methods and waterborne solutions for manufacturing cathodes, including the usage of aquatic binders such as PTFE, PAA, SBR, and CMC [68,101]. Similarly, Spreafico et al. approached a novel technique to fabricate slurry cast electrodes using PVDF binder without using NMP solvent. Copper oxide has been coated to reduce the decomposition of active material from an aqueous environment. A copper oxide particle-treated cathode (Cu-LCO (W)) exhibits a higher reversible capacity (Fig. 4b) than a copper oxide particle-untreated cathode (LCO(W)). Therefore, the coating of copper oxide on active materials exhibited greater advantages under more severe cycling conditions and improved the performances [51]. In addition, Santiago et al. introduced a new method of using imidazolium-based polymeric ionic liquid instead of NMP in a high-performance sulfur cathode due to its good adhesion at high sulfur loadings compared to the conventional PVDF binder. The results were found to improve the capacity to about 600 $mAhg^{-1}$ at C/2 and 400 mAh g^{-1} at 1C due to its greater synergistic effect (Fig. 4c) [98].

A composite binder system is also effective proceeding to solve the PVDF binder-associated problems and improves the electrochemical performance during charge-discharge processes. Mokaripoor et al. formed PVDF: PMMA composite binder for LTO/rGO5wt% electrode as an anode in LIBs to overcome the movement of lithium and charge transfer reactions at the electrode interface and the results also exhibited higher flexibility with a discharge capacity of 181.79 $mAhg^{-1}$ (Fig. 4d) [99]. Likewise, Liu et al. developed a novel composite binder (vinyl phenol-grafted PVDF) and the results were found to have higher cyclic stability and capacity retention. This strategy relies on scavenging of O_2, which is usually performed by phenolic hydroxyl groups, and in turn, it aids in building a compatible cathode-electrolyte interphase (CEI), thereby enhancing the battery performance. The O_2 escaping from active materials attacks electrolytes, resulting in thick and incompatible CEI layers on PVDF-based cathode, while in vinylphenol-grafted PVDF-based cathode, O_2 gets trapped and only a thin and effective CEI layer is formed, as shown in Fig. 4e [100]. In contrast, Nugraha et al. developed a novel mixed conductive PEDOT: PSS-TFSI material to replace both the PVDF binder and carbon electronic additives, significantly enhancing lithium diffusion within the electrode and thereby increasing the reversible capacity at high rates. Notably, it demonstrated superior performance with high active material loading and enhanced capacity retention over extended cycling [102].

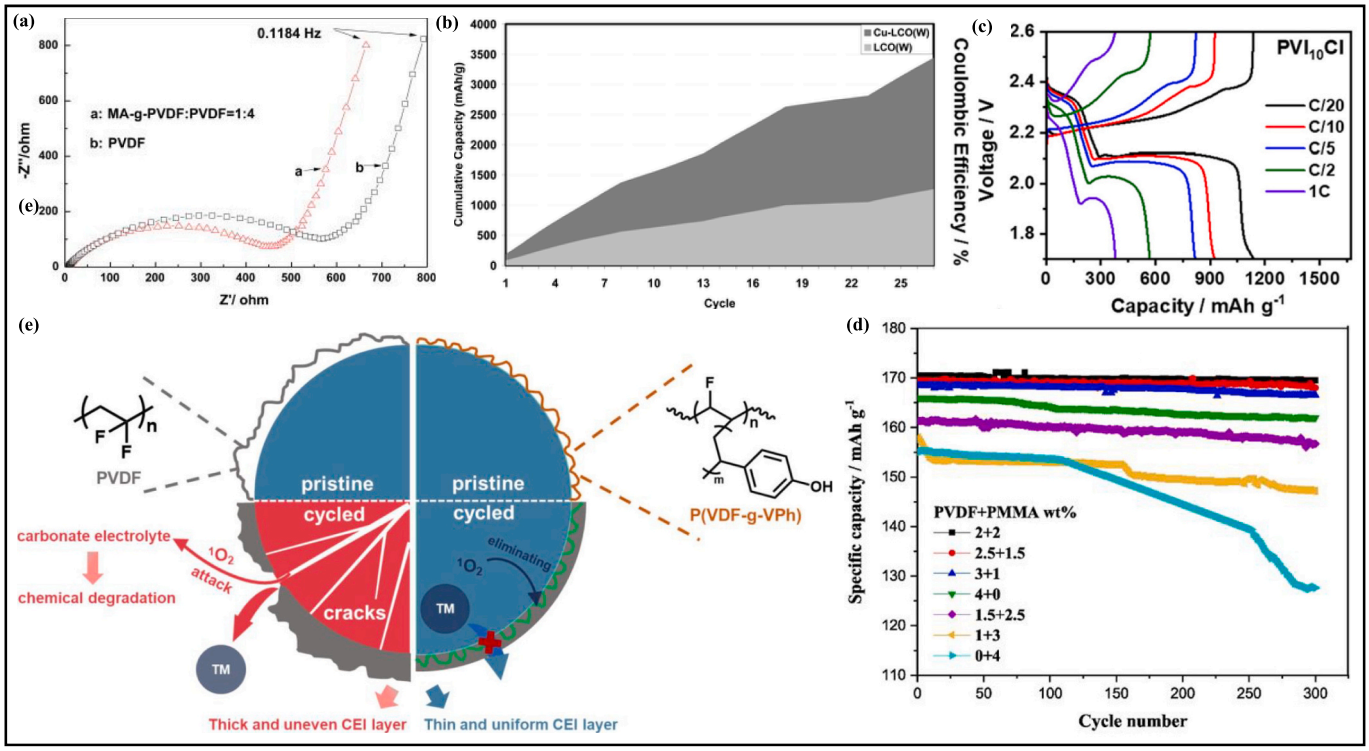

Fig. 4. (a) EIS analysis of $LiCoO_2$ electrodes prepared with MPVDF and pure PVDF binders. Reproduced with permission. [97] Copyright 2014, The Authors, published by Elsevier. (b) Cumulative capacity versus cycles for Cu-LCO(W) and LCO(W) at different cycling rates. Reproduced with permission. [51] Copyright 2014, The Authors, published by American Chemical Society. (c) Voltage profiles from C-rate tests with $PVI_{10}Cl$ as the binder. Reproduced with permission. [98] Copyright 2024, The Authors, published by Elsevier. (d) Cycling performance of TO/rGO electrodes fabricated with varying PVDF: PMMA ratios. Reproduced with permission. [99] Copyright 2024, The Authors, published by Elsevier. (e) Schematic illustration of the working mechanism at the NCM622 cathode interface using PVDF and P(VDF-g-VPh) binders. Reproduced with permission. [100] Copyright 2022, The Authors, published by Elsevier.

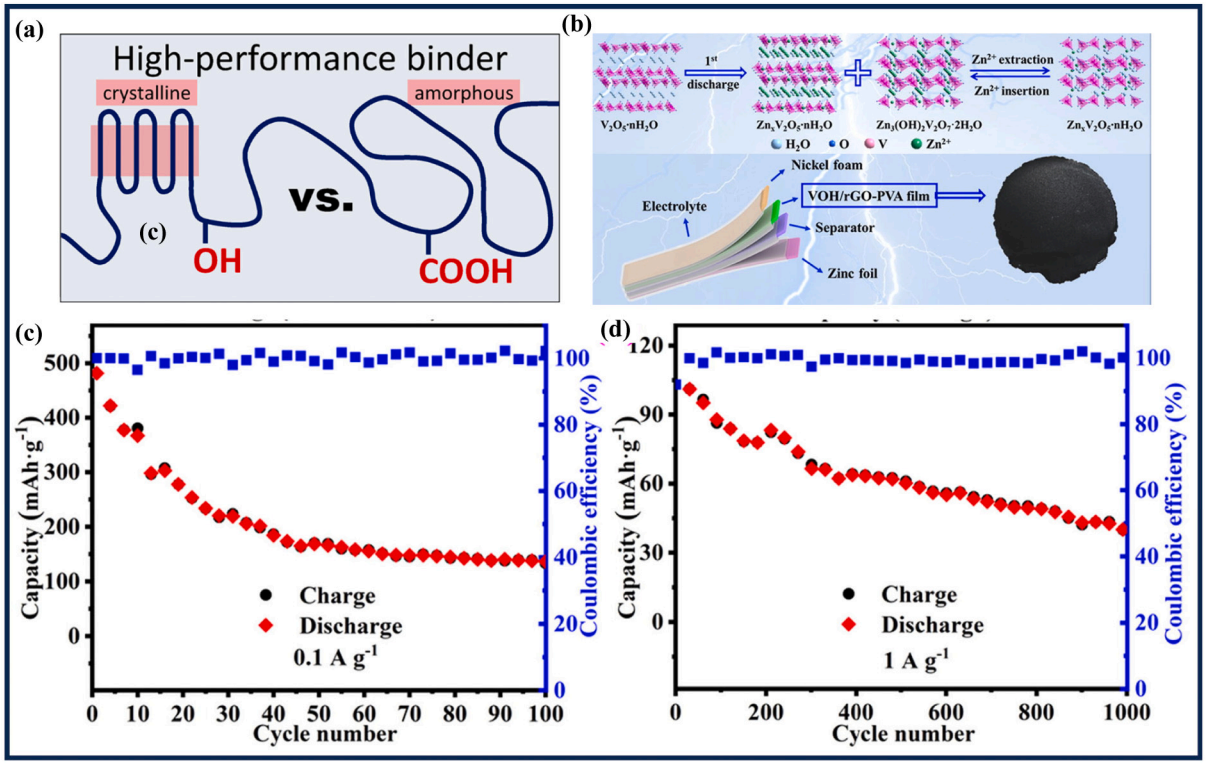

Fig. 5. (a) Semi-crystalline structure of the PVA binder. Reproduced under terms of the CC-BY 4.0 license. [53] Copyright 2021, The Authors, published by American Chemical Society. (b) Reaction mechanism and structural representation of the VOH/rGO-P film electrode using PVA binder and cyclic performance of VOH/rGO-P film electrode at (c) 0.1 $A{\cdot}g^{-1}$ and (d) 1 $A{\cdot}g^{-1}$. Reproduced with permission. [106] Copyright 2021, The Authors, published by Elsevier.

3.1.2. PVA & PAN

3.1.2.1. Material characteristics and binding mechanisms of PVA & PAN in battery electrodes. Unlike PVDF, PVA and PAN binders are not popularized. However, it is now attracting researchers to use it in batteries rather than relying completely on aqueous-based binders. In general, PVA binders are highly water-soluble, have excellent film-forming ability, good machinability, and are highly thermally stable (melting point: 230–240 °C), which makes them broadly used in construction, textile and other manufacturing units. The wide usage of PVA binder, also called polyhydroxy polymer is simply because of its ease of synthesis, which is made out of fossil energy. The carbon chain in PVA binder is highly rich in hydroxyl groups that make it easily interact with other functional groups (–OH and COOH). The PVA binder establishes stronger connections between the electrode components, attributed to its abundance of hydrogen bonds. Eventually, the surface of active materials was covered with a greater amount of binder due to these strong hydrogen bonds, resulting in a significant increase in electrode adhesion [54]. The strong intermolecular bond between hydrogen-bonded hydroxyl groups facilitates the semi-crystallinity (combination of crystalline and amorphous phases) nature of PVA binder (Fig. 5a) [53].

PAN binder has also been explored as a promising conductive polymer binder for cathode electrodes. Similarly, PAN binder is also known as a semicrystalline polymer with the same promising factors for battery applications [17,56]. PAN features nitrile (–C ≡ N) groups that can undergo partial cyclization during drying or mild annealing, forming conjugated C=N–C structures capable of limited electronic conduction. This cyclization enhances interchain interactions, converting PAN into a semi-conductive polymer network [103]. Additionally, the polar nitrile groups exhibit strong adhesion to transition-metal oxides, improving interfacial contact and mechanical durability. Thus, PAN acts as both a binder and an electronically active medium, reducing charge-transfer resistance [104]. Moreover, the strong polarity of the nitrile groups enhances adhesion to both active materials and current collectors, improving electrode integrity during long-term cycling. These combined effects make PAN not only a mechanical binder but also an electronically active component that contributes to improved rate performance and structural stability of the electrode [105]. PAN binder has several key features, including its thermal stability, hardness, and resistance to organic solvents due to its strong interactions between nitrile groups [56].

3.1.2.2. Structure–property–performance relationship of PVA & PAN-based binder. Polymeric binders like PVA and PAN tend to exhibit good thermal stability, low cost, and benign electrolyte compatibility, suggesting greater potential for commercial applications. PVA binder is an excellent high-capacity anode binder owing to its multiple hydroxyl groups, which enable strong hydrogen bonding between the electrode components. For example, Park et al. compared PVA binder with PVDF and PAA and concluded that the PVA binder greatly enhanced the anode's capacity due to its numerous and strong hydrogen bonds between active materials as well as the current collector [54]. Similarly, PVA-based cathode in ZIBs showed superior electrochemical performance as illustrated in Fig. 5b. The results were found to be higher in energy density of about 708 $Whkg^{-1}$ at a power density of 183 Wkg^{-1} while the specific capacity has reached up to 481 $mAhg^{-1}$ at 0.1 Ag^{-1} (Fig. 5c &d) [106]. Mandal et al. have demonstrated that PVA binder with a high degree of hydrolysis offered superior performance for Si-based electrodes, showing its greater potential toward high and stable capacities over extended cycling intervals. It is confirmed that the semi-crystallinity nature of PVA binder largely influences the capacity, cycling stability, and coulombic efficiency. The higher the crystallinity, the better the retention of mechanical integrity and the lower the volume expansion of the active materials [53].

On the other hand, the PAN binder performs best among the other three binders for LMO-based electrodes due to its superior cycle performance, high thermal stability, and excellent rate capability [107]. The PAN binder has been widely studied as a conducting polymer with exceptional electrochemical reversibility as a positive active material, suitable for both aqueous and non-aqueous electrolytes. Hence, several studies have been focused on using PAN binders in solid-state rechargeable lithium batteries or LIBs as a positive material [108]. The results obtained by the PAN binder demonstrated that the potential of nitrile groups provided a favourable condition toward ionic conductivity because of their strong polarity from electronegative nitrogen groups [57]. Further, the hydrogen bond and dipole-dipole interactions bring the nitrile groups into strong contact with Li^+ and electrode components. This results in low charge transfer resistance, high adhesion strength, active material contact, high Li^+ transfer, and good electrolyte wettability. In addition, PAN binder provides good solvent resistance and strong polarity because of the presence of electronegative nitrogen [56]. It possesses an appropriate rheological behaviour due to the strong adhesion of the PAN binder that favours industrial processability [109]. PAN binder in solvent tends to be more effective at molecular level dispersion, though it lacks in the presence of an oxygen atom and weak effect of the nitrogen atom with Li^+, thereby providing a higher Li^+ transference number [54]. For example, Gong et al. confirmed that the electrode prepared using PAN binder had enhanced electrochemical properties by providing thermal and chemical stability to the anode in different kinds of LIBs, such as high-power lithium titanium oxide, commercial graphite, and high-capacity silicon/graphite [56]. Therefore, PAN binder possesses a higher degree of improvement in terms of electrochemical properties and may be used for high-capacity and high-power electrodes with further studies.

Like PVDF, the performance of PVA and PAN is also largely affected by the size and proportion of the binder in the electrode. In addition to that, the molecular weight (MW) of the binder tends to influence the mechanical and electrochemical properties of the prepared electrode [54]. For example, Park et al. affirmed that the PVA with lower MW seemed to have the least charge transfer resistance compared to PVA with higher MW and even with PVDF binder (Fig. 6a). The results show that the greater amount of cluster formation and wide distribution of the binder on the surface of active materials was noted with the increasing MW of PVA binder. Furthermore, the electrode with a high MW of PVA binder absorbed less electrolyte, restricting the Li^+ movement between the cathode and anode. Consequently, low MW PVA binder has been identified as a novel and effective binder for high-capacity anodes [54].

Though PVA binder has good adhesion properties, it suffers from the irreversible slipping of active materials, poor mechanical properties, and electrode degradation during cyclic performances [110,111]. In addition, it leaves a variable amount of acetate groups as a residue, and that greatly affects the cyclic performance of anodes in batteries since PVA binder is formed by hydrolysis of PVA. Electrodes prepared using a PVA binder require excessive electrolyte consumption to form the stable SEI layer. Moreover, PVA binders with a single network are not able to form strong interactions with active materials. As a result, this leads to the breakage of active materials and mechanical failure of the electrode. Therefore, the PVA-based electrodes, especially Si-based electrodes, show poor electrochemical performances [112]. PAN binders possess high brittleness and poor water solubility due to their weak force between the active material and high glass transition temperature (Tg) that hinders the direct use of PAN binder for Si-based anodes [56]. The active materials in solid-state lithium-PAN batteries fail to make good bonds and surface area contact between the active material and electrolyte. Additionally, the utilization ratio of the positive active material is low, at only around 20 % [113].

3.1.2.3. Advanced modifications and alternatives to PVA & PAN binders. The conventional covalent bonds in PVA binder, including those formed

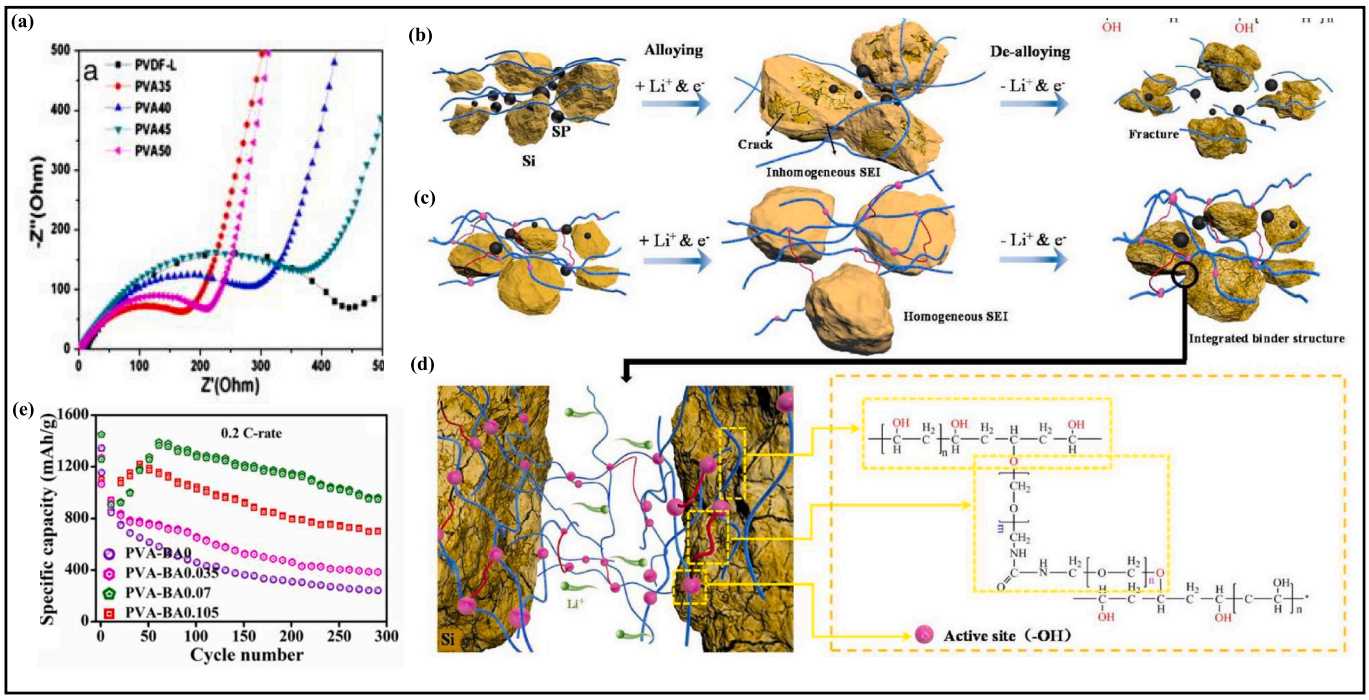

Fig. 6. (a) Nyquist plot of graphite electrodes prepared with varying concentrations of PVA binder. Reproduced with permission. [54] Copyright 2011, The Authors, published by Elsevier. Electrochemical behaviour of the Si anode with the (b) PVA and (c) PVA-UFR binder, (d) Binding mechanism of the PVA-UFR binder in the Si anode. Reproduced with permission. [111] Copyright 2021, The Authors, published by Elsevier. (e) Electrochemical performance of electrodes fabricated using the PVA–BA binder cycled at a 0.2 rate. Reproduced with permission. [114] Copyright 2023, The Authors, published by Elsevier.

through -COOR- esterification reactions, are generally not sufficiently flexible when subjected to large volume variations during the cycling process. Hence, dynamic bonds such as hydrogen [115], imine [116], and borate ester bonds [110] are more effective in alleviating structural damage to electrodes because of their reversible effect. The Si anode prepared using a PVA binder experiences volume expansion of Si particles during the charging process and shrinkage during the discharge process. This results in instability of the SEI film and breakage of Si particles on the surface, thereby, it loses its electronic contact between active materials as shown in Fig. 6b. The anode prepared using PVA-UFR composite binder has mitigated the effect of volume in Si particles (Fig. 6c), with maintained structural integrity. The homogenous SEI film

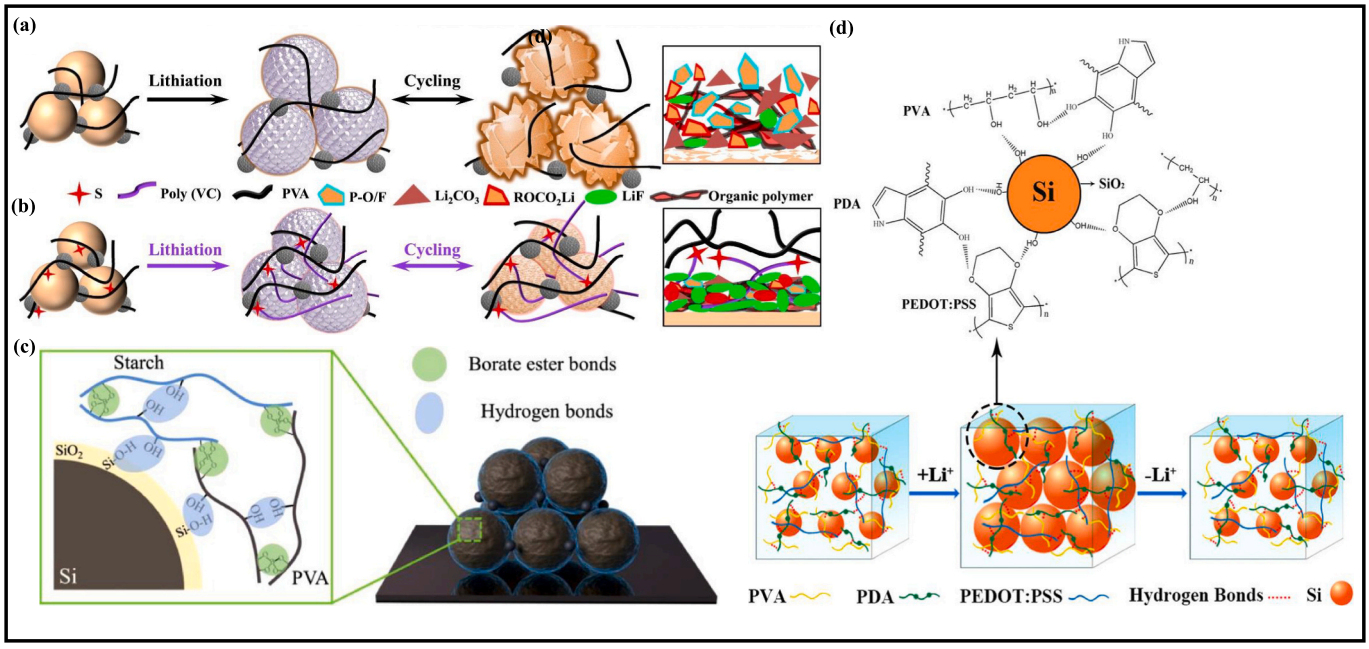

Fig. 7. Graphical representation showing the interphase composition, structural evolution, and binder conformation of (a) PVA and (b) PS61-based Si anodes during cyclic performances. Reproduced with permission. [112] Copyright 2023, The Authors, published by Elsevier. (c) Graphical design of SPS binder and its interaction mechanism with Si nanoparticles. Reproduced with permission. [118] Copyright 2022, The Authors, published by Elsevier. (d) The proposed working mechanism of the PPP binder. Reproduced with permission. [119] Copyright 2020, The Authors, published by Wiley.

formed on Si surface is stable due to its excellent plasticity and increased mechanical properties, as shown in Fig. 6d. Most importantly, the presence of ether, amide, and hydroxyl bonds greatly influenced the effect of PVA-UFR binder [111]. Phanikumar et al. confirmed that the PVA binder combined with sodium alginate for $Li_4Ti_5O_{12}$ (LTO) electrodes exhibited less polarization, lower charge transfer resistance, good thermal and electrochemical stability behaviour compared to pure PVA binder [117]. Researchers have developed a cost-effective polymeric binder through crosslinking PVA binder and boric acid (BA) to form a 3D-crosslinked compound that is water-soluble and the results seem to be high stability, excellent electrochemical properties, especially, higher in specific capacity (946.8 mAh/g) even after 300 cycles (Fig. 6e) [114]. Huang et al. demonstrated that the PVA binder combined with squaric acid (SA) enables multiple advantages, including more Li^+ transport sites, outstanding plasticity, mechanical integrity, reduction in electrolyte degradation, and robust SEI passivation layer with rich LiF as compared to pure PVA binder (Fig. 7a &b). Especially, it provides Young's modulus that is adaptive to wide volume changes of active Si particles [112]. The dual-dynamic interaction of multifunctional binder retains the toughness and flexibility, while strong adherence of Si particles is maintained by reversible hydrogen bonds induced by abundant hydroxyls. The electrode prepared using a multifunctional binder ensures excellent stretchability and self-healing properties owing to its presence of borate ester and hydrogen bonds, as shown in Fig. 7c. As a

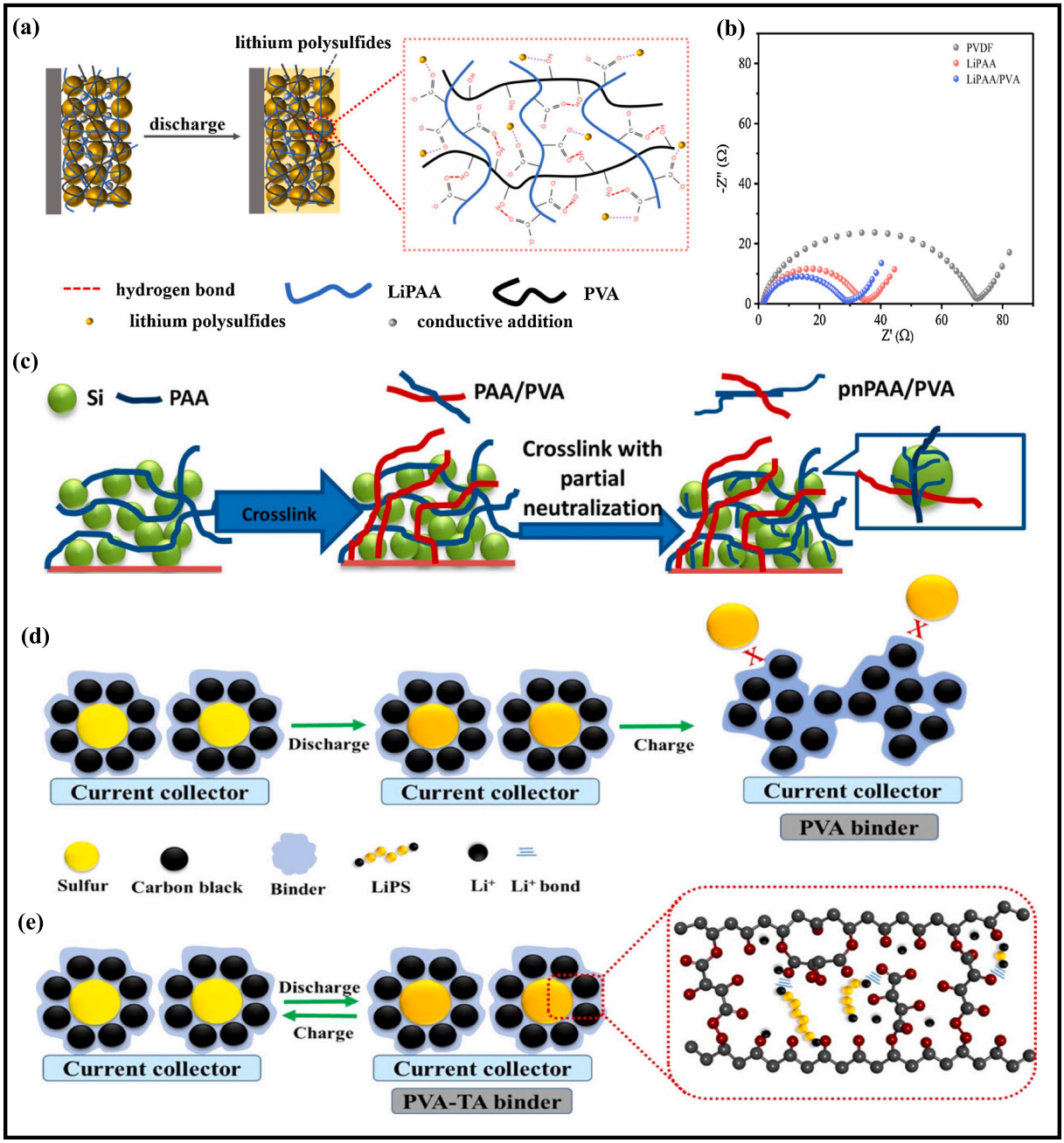

Fig. 8. (a) Graphical view of the configuration of cathode using LiPAA/PVA binder during the electrochemical process, (b) Nyquist plot of sulfur cathodes with PVDF, LiPAA and LiPAA/PVA binders at open circuit potential. Reproduced with permission [120] Copyright 2021, The Authors, published by Elsevier. (c) Graphical depiction of a cathode prepared with a PAA/PVA binder. Reproduced with permission [125] Copyright 2018, The Authors, published by American Chemical Society. Working mechanism of electrodes prepared with (d) PVA binder and (e) PVA-TA 3D cross-linked composite binder. Reproduced with permission [126] Copyright 2023, The Authors, published by American Chemical Society.

whole, it aids in stabilizing the SEI layer, excellent electrochemical performance, high Li^+ diffusion rate, alleviating mechanical damages and cracks by large volume changes of Si particle [118]. Tang et al. also confirmed that the prepared multi-functional binder provided strong adhesiveness, improved rate performances, and stability even with high Si particle loadings in Si-based anode materials (Fig. 7d) [119].

In addition to that, the single network of PVA binder suffers from strong binding interactions between active materials. Here, Chen et al. constructed a 3D cross-linking polymer network between PVA and LiPAA macromolecular chains through hydrogen bonds as depicted in Fig. 8a [120]. The prepared PAA/PVA composite binder maintained tight contact between the sulfur composite and conductive additive during the discharge and charge process [121]. The primary advantage of the PAA/PVA composite binder is its ability to preserve the integrity of the sulfur cathode, even at high sulfur loading. The abundant oxygen-containing polar groups of the LiPAA/PVA binder strongly adsorb lithium polysulfides, effectively restricting their irreversible dissolution [122]. Furthermore, LiPAA with its high theoretical Li^+ capacity serves as an additional Li^+ reservoir in cathodes with high current densities [123]. LiPAA/PVA cathode exhibits low R_{ct} among other cathodes as shown in Fig. 8b, revealing its ability to transfer charge fast between conductive agents/liquid electrolyte and sulfur composite during electrochemical performances [120,124]. The PAA/PVA binder-induced electrode exhibits enhanced electrode cyclability, binding adhesion strength, and mechanical stiffness compared to PAA and PVA individually (Fig. 8c) [125]. Similarly, Reddy et al. concluded that 3D cross-linked PVA-TA composite binder has extensively enhanced the structural integrity, mechanical, and adhesion property between active materials even at high sulfur loading as shown in Fig. 8d & e [126].

The cross-linking technique, forming a composite, and incorporation of additives aid in reducing the limitations associated with the PAN binder, similar to the PVA binder. For example, Shen et al. constructed an in-situ thermally cross-linked Si anode using a PAN binder as depicted in Fig. 9a. The results seem to enhance the rate performance, adhesion rate, capacity maintenance, and coulombic efficiency. Most importantly, the thermal treatment (230 °C) cleaves the nitrile groups during cyclic performances, thereby forming conjugated chains with delocalized π electron systems, as shown in Fig. 9b [58]. Li et al. developed a multifunctional 3D crosslinking PAN hydrolysate with epichlorohydrin (H-PAN-g-ECH) and the result was possessed to have a high discharge capacity of 2246.3 mAh g^{-1} even after 200 cycles (Fig. 9c). The strong amide (-$CONH_2$), carboxylate (-COO-) and cyanide (-CN) of H-PAN-g-ECH binder along with great adhesion to both Si and Cu foil greatly influenced the electrochemical performance and enhancement of electrode integrity during cycling [127].

In addition, Chen et al. constructed a bilayer film between PEO-based polymer electrolyte and PAN-GPE composite binder for LIBs. The results were found to have more contact area for active materials, electrochemical reversibility, discharge capacity, and compatibility than PAN film with PTFE binder (Fig. 9d) [128]. Secondly, Tsao et al. developed a $LiFePO_4$ electrode using poly(ethylene oxide)-*block*-poly(acrylonitrile) (PEO-b-PAN) as an ionic conducting binder. The results demonstrated outstanding rate performance, reduced interfacial resistance, improved electrical conductivity, increased contact area, lower polarization, and enhanced diffusion pathways for Li^+ transport compared to pure PAN and PVDF binders [129]. The SEM images of the electrode prepared using PVDF show clustered $LiFePO_4$ particles and some super P conducting carbon, while the electrode prepared using PEO-b-PAN binder exhibits no electrode aggregations as depicted in Fig. 10a [130].

Wu et al. demonstrated that the PAN combined with PVDF achieved long cycle rate and rate performances by taking advantage of both PAN and PVDF. As depicted in Fig. 10b, the PVDF-based electrode suffers from degradation. Besides, even the battery safety deteriorates when the laminate is soaked in electrolyte. In contrast, the electrode prepared using PAN (Fig. 10c) alleviates the undesirable exfoliation, but the embrittlement of the electrode elevates the resistance of the laminate and thus leads to cracks within the electrode materials. The composite binder-based electrodes exhibit excellent synergistic effects, prevent the enlargement of the impedance, and reduce the dissolution of active materials, ultimately improving the electrochemical performance of the battery (Fig. 10d) [131]. Considerable effort is still required to refine the chemical interactions and crosslinked structure of the binder network, given their critical influence on the overall performance of the battery. Therefore, it is not ideal to have a network with a linear or too-branched backbone due to the excessive stiffness or looseness of its properties [132].

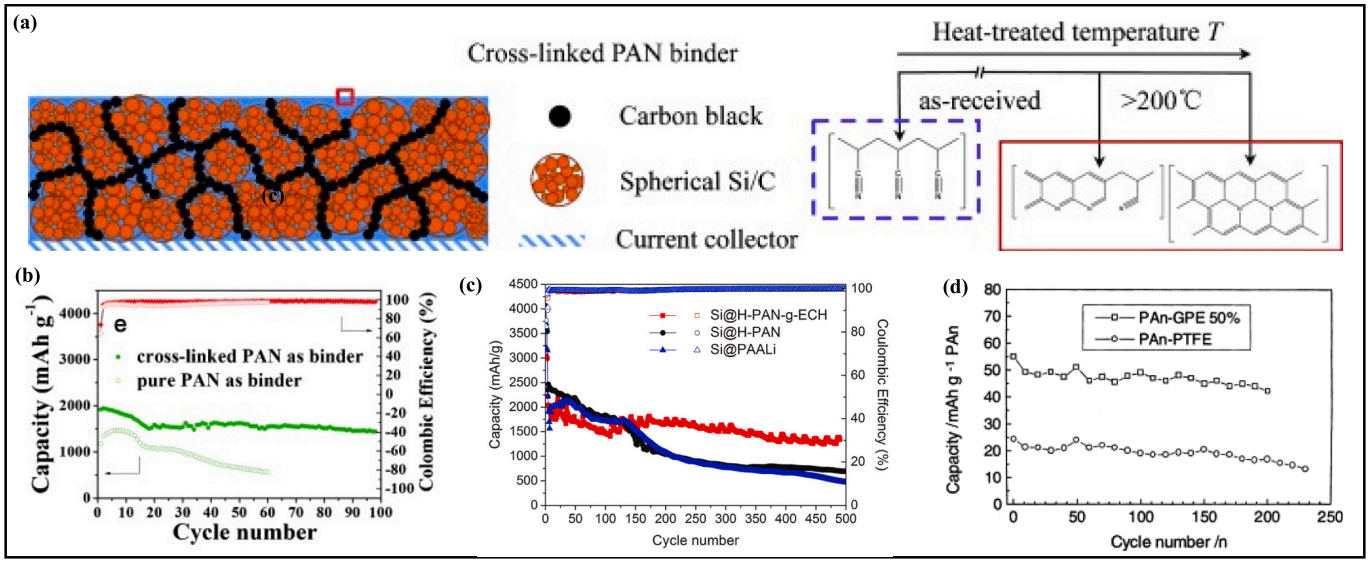

Fig. 9. (a) Graphical depiction of Si-based anode with an in-situ thermally cross-linked PAN binder, (b) Comparison of cycling performance between electrodes with cross-linked PAN and pure PAN binders. Reproduced with permission [58] Copyright 2014, The Authors, published by Wiley. (c) Long-term cyclic performance of various Si electrodes at 0.2C. Reproduced with permission [127] Copyright 2024, The Authors, published by Elsevier. (d) Discharge capacity of the Li-PAN cell as a function of cycle number with a constant current density of 0.2 mA/cm^2. Reproduced with permission [128] Copyright 2001, The Authors, published by Elsevier.

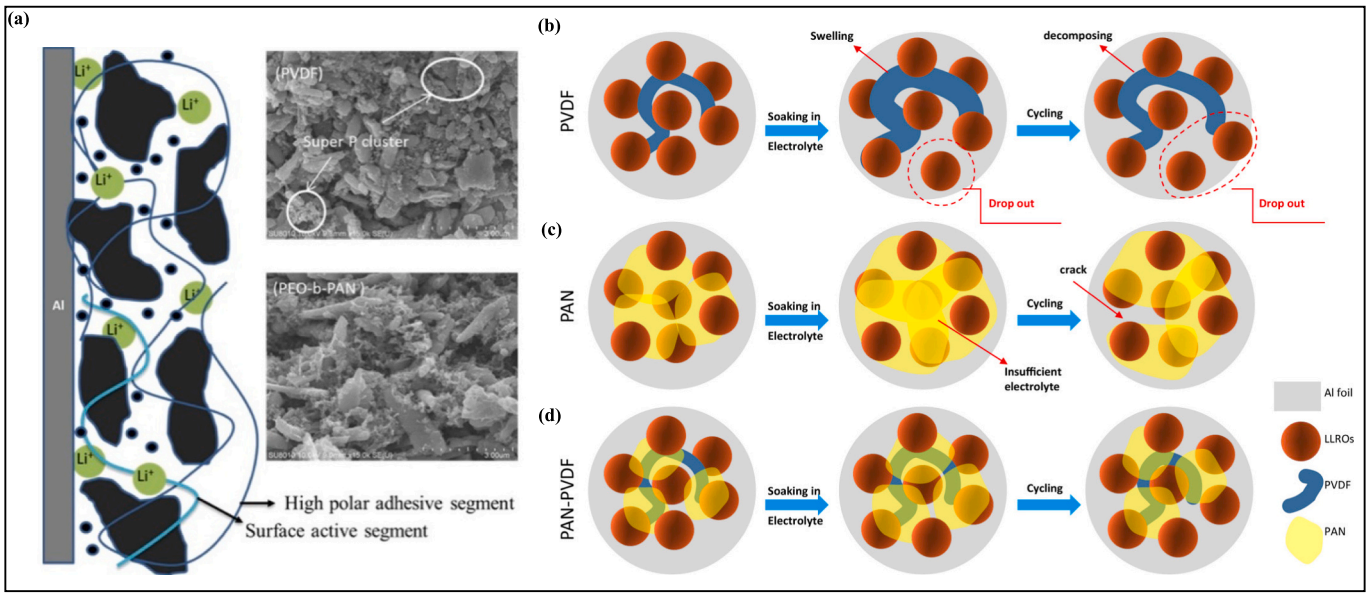

Fig. 10. (a) Graphical representation and SEM images of electrodes prepared with PEO-b-PAN and PVDF binders, highlighting structural differences. Reproduced with permission [130] Copyright 2016, The Authors, published by Elsevier. Schematic depiction of an electrode using (b) PVDF, (c) PAN, and (d) PAN-PVDF binder. Reproduced with permission [131] Copyright 2017, The Authors, published by Elsevier.

3.2. Aqueous binder

3.2.1. PTFE

3.2.1.1. Material characteristics and binding mechanisms of PTFE in battery electrodes. PTFE possesses a fully fluorinated backbone (–CF_2–CF_2–) that renders it chemically inert and hydrophobic. Its superior thermal and chemical stability make it suitable for harsh electrochemical environments, yet the absence of reactive sites prevents strong interfacial bonding [133]. The fibrillated PTFE network forms a robust mechanical scaffold, enhancing electrode integrity and flexibility but offering minimal contribution to ionic or electronic transport due to its insulating nature [134]. PTFE binder tends to have enhanced London dispersion forces due to its high electronegativity of fluorine, along with a low cohesion fraction among solids. The strong carbon and fluoride bonds make the PTFE binder inert. PTFE binder is proven to have high strength, self-lubrication, and toughness at a high melting point of about 326 °C and at a low temperature of nearly −268 °C. PTFE binder shows excellent flexibility at temperatures of about −79 °C.

3.2.1.2. Structure–property–performance relationship of PTFE-based binder. PTFE binder has been introduced and replaced with the usage of PVDF in larger aspects to address the negative factors, such as the non-uniform distribution of materials in the current collector after drying, the side reaction of solid electrolyte, and its associated environmental challenges [135]. Besides addressing these challenges, the usage of PTFE binder in electrode fabrication also provides a practical and scalable solution for mass production due to its elimination of solvents [62]. The assembly of secondary batteries like LIBs and similar batteries using solvent-free electrodes has significant merits in terms of safety, environment and cost. The operational aspects, such as the requirement of humidity processing with strict control like wet-process, delay in electrode drying, and reduction of the binder during cyclic process, were not faced by the PTFE binder [9,136]. For example, Matthews et al. investigation revealed that solvent-free electrodes achieved 150 % discharge capacity, 40 % slower degradation, and 96 % retained initial capacity even after 200 cycles compared to slurry-based electrodes [137].

Dry-processed electrodes (DE) using PTFE binder provide fibrous contact that improves the contact between active material, uniformly distributes the electrode components, and is less complicated than the wet-processed electrode, as depicted in Fig. 11a [67]. Research confirmed that the tight contact between active particles and conductive additives of cathode materials was achieved by the uniform distribution of binder even after long cyclic performances of the battery [94]. In addition, researchers have made serious efforts to mitigate the drawbacks associated with anodes made of PTFE binders in LIBs, such as mechanical degradation and the loss of active materials. For example, Behara et al. prepared a ZnO electrode with the PTFE binder to ensure strong bonding, which gradually reduced the structural stress and changed the volume effects during cycles and thus led to excellent battery performances [138].

Recently, thick electrodes have attracted growing interest as an effective strategy to reduce the proportion of inactive materials and thereby increase the areal and volumetric energy density of LIBs. However, the conventional slurry-casting process presents several limitations when applied to thick electrodes. The pictorial representation of electrode preparation using wet and dry processing routes is shown in Fig. 11b-e. In thick slurry-cast electrodes (SCEs), thermal stress and capillary shrinkage during solvent evaporation generate cracks, interfacial delamination, and binder-rich surface layers as shown in Fig. 11b [140,141]. These inhomogeneities disrupt electronic percolation networks and block Li^+ diffusion pathways, resulting in increased ionic tortuosity and charge-transfer resistance (Fig. 11c) [142]. The accumulation of binder and conductive additive near the surface further causes non-uniform current distribution, impeding efficient ion and electron transport across the electrode thickness. Consequently, achieving both high mass loading and mechanical integrity through slurry casting remains challenging for industrial-scale applications.

In contrast, dry-processed electrodes mitigate these issues by eliminating solvent drying and binder migration. During mechanical mixing or calendaring, the PTFE binder fibrillates, forming a robust three-dimensional cross-linked network with conductive carbon that uniformly encapsulates active particles (Fig. 11d). This interconnected framework enhances electronic conductivity and provides continuous Li^+ transport pathways, thereby minimizing internal resistance and improving electrochemical homogeneity (Fig. 11e) [140,143,144]. The solvent-free fabrication also preserves structural integrity, reduces porosity collapse, and offers a more sustainable and scalable alternative for next-generation thick electrodes with high areal capacity and

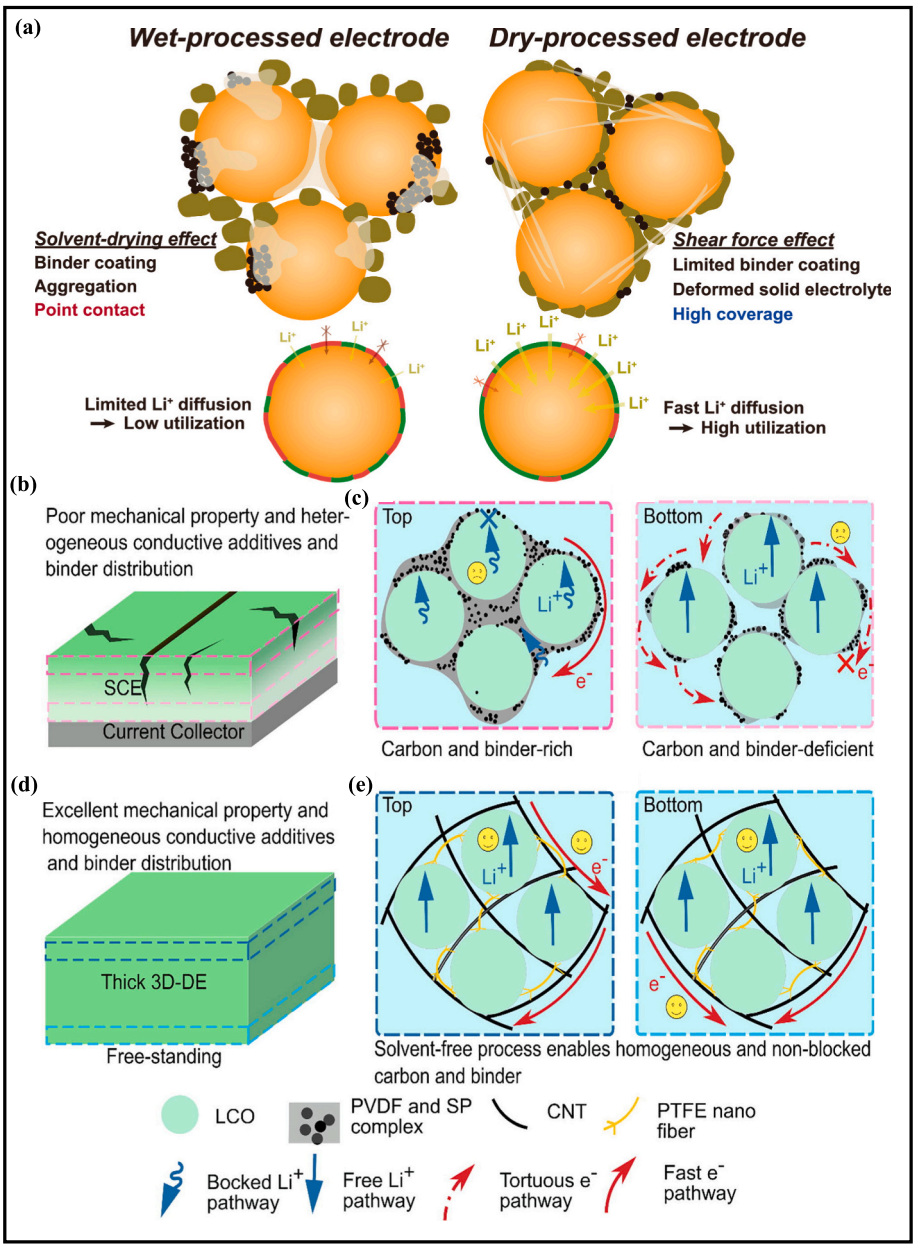

Fig. 11. (a) Graphical illustration of wet and DE fabrication methods. Reproduced under terms of the CC-BY license. [139] Copyright 2024, The Authors, published by Springer Nature. (b) Illustration of cracking and uneven coverage of carbon additives and binder in thick SCE fabricated through the wet process, (c) Depiction of binder-rich and carbon additives versus deficient layers in the top and bottom regions of thick SCE, (d) Schematic illustration showing the uniform distribution of components in thick 3D-DE, resulting in high mechanical flexibility and (e) Visualization of the uniform distribution of PTFE and CNT across the surface and bottom regions of thick 3D-DE. Reproduced with permission. [140] Copyright 2024, The Authors, published by Elsevier.

prolonged cycling stability.

Recognizing the appropriate size and ratio of PTFE binder is highly important to determine the role of atomic percentages of fluorine in the chemical proportion between active materials and binder [145]. The exploration of the right proportion of binder is necessary, which largely influences the interrelated aspects of solvent-free electrode performances, final microstructure, and processability of PTFE binder [22]. The variation in size of the PTFE binder seems to affect performances, charge transfer characteristics, and distribution of electrode components. The reduced PTFE particle size can significantly reduce the charge transport resistance, enabling more streamlined electronic and ionic conductivity as illustrated in Fig. 12a-b. Lee et al. revealed that the PTFE (S) with a particle size of 6.44 μm showed an enhanced specific capacity of about 188 $mAhg^{-1}$ with high cyclic performance (90.4 % capacity retention - 100 cycles) compared to PTFE(M) - 87.73 μm and PTFE(L) - 492.3 μm [94].

Secondly, Behara et al. developed ZnO/PTFE anode at three different percentages, including ZnO/PTFE(5), ZnO/PTFE(10), and ZnO/PTFE (15). Out of all, it is found that the ZnO/PTFE(5) composite outperforms other ZnO/PTFE formulations across the entire range of current rates, indicating its superior performance and suitability for high-rate applications (Fig. 12c) [138]. Oh et al. achieved a higher discharge rate capability of 80 % at 0.5C with low PTFE content (2 wt%) [62]. Moreover, the morphology of PTFE binder with low and high content shows fibril and cement morphology, respectively. It also confirms that the increase in PTFE content reduces the surface porosity and hinders the charge-carrying ionic mobility in the electrode [22]. Therefore, the higher the PTFE amount an electrode contains, the higher the possibility

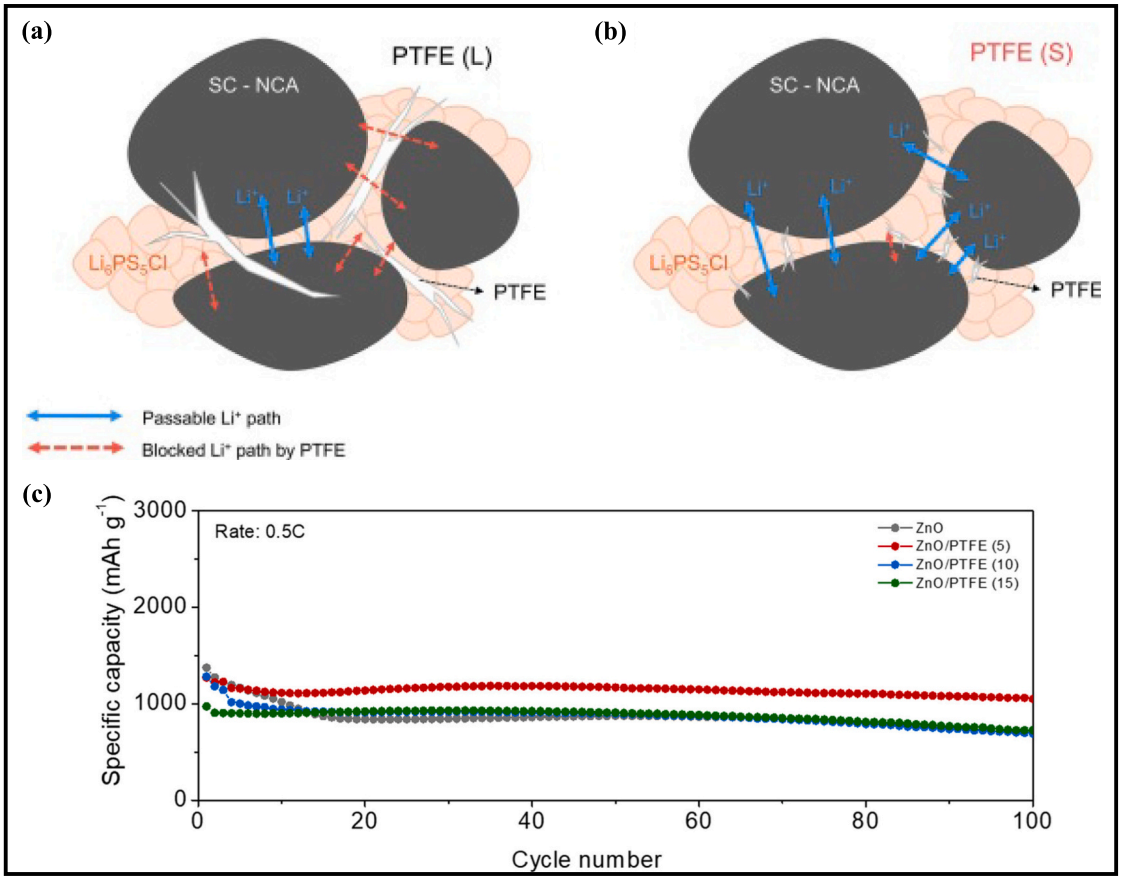

Fig. 12. Graphical depiction of transport of Li^+ within a composite cathode using (a) PTFE (L) and (b) PTFE (S). Reproduced with permission [145] Copyright 2024, The Authors, published by Elsevier. (c) cycling performance comparison of ZnO and ZnO/PTFE composites at 5 %, 10 %, and 15 % PTFE content. Reproduced under terms of the CC-BY license. [138] Copyright 2024, The Authors, published by Wiley.

it has of decreasing the electrical conductivity of the electrode, thus reducing the Li^+ ion storage capability.

A strong relationship exists between microstructure, electrode formulation, and mechanical properties, which eventually affects the electrochemical performance, suggesting that a solvent-free process may not always be the best replacement for the slurry casting process [22]. The PTFE binder has been booming mainly due to its low polarization effect and high mechanical properties, which lead to stabilized electrochemical properties. However, it suffers from low lowest unoccupied molecular orbital (LUMO), which makes the battery unstable during the lithiation process when the PTFE binder is used in the preparation of the anode [146–148]. PTFE binder in anode gets defluorinated during initial lithiation because of its easy electron acceptability, and the reaction is shown in Eq. 3 [149].

$$-[CF_2-CF_2]_n+4n\,Li^+\rightarrow-(C=C)_n+4n\,LiF \quad (3)$$

In addition to lowering initial Coulombic efficiency, the irreversible degradation impairs the binding properties of PTFE, thereby deteriorating long-term electrochemical cell performance [150].

3.2.1.3. Advanced modifications and alternatives to PTFE binders. The irreversible degradation of PTFE binder can be mitigated by adding fluoroethylene carbonate (FEC) as an electrolyte additive to retrieve the electrochemical stability and improve the initial Coulombic efficiency (Fig. 13a). FEC-derived solid electrolyte interphase maintains the structural integrity, reduces PTFE degradation, and suppresses volume changes during cyclic performances, thereby resulting in high stability even after long cyclic performances of the electrode (Fig. 13b). The electrode prepared using dry-process achieved high-rate performance and high cycle stability because of its synergistic effects of FEC-derived solid electrolyte interphase [151].

Wang et al. constructed a novel method by adding a secondary binder and ultralong multi-walled carbon nanotubes (MWCNT) as a conductive additive to fabricate DE. The results exhibited remarkable changes with superior mechanical strength and electrochemical performances (Fig. 13c) [152]. Moreover, Lee et al. determined that the coating of ionic-conductive polymers such as poly(ethylene oxide) and poly(vinylidene fluoride–trifluoroethylene–chlorofluoroethylene) on graphite reduces the anodic degradation by limiting the direct contact electric between PTFE binder and graphite. This coating increases initial coulombic efficiency to 16 % and cell capacity to 17 % compared to the pure graphite anode (Fig. 13d) [153]. The high performances, robust mechanical properties, excellent rate performances, and high cycling stability have been achieved with DE incorporating flour as a partial binder with PTFE as compared with DE with no flour and wet-processed electrode (Fig. 14a-c). The results show enhanced cycling stability with a capacity retention of 80.3 % after 260 cycles at 2C and 4.5 V, as shown in Fig. 14d [154].

3.2.2. SBR, CMC and PAA

3.2.2.1. Material characteristics and binding mechanisms of SBR, CMC & PAA in battery electrodes. SBR binder is a synthetic rubber binder made from styrene and butadiene compounds and is used in the anode slurry to strengthen the bond between the active material and the collector, as it increases the cycle stability and shear viscosity [155,156]. The π–π interactions between styrene units and carbon additives improve electronic connectivity, while the elastic butadiene phase accommodates

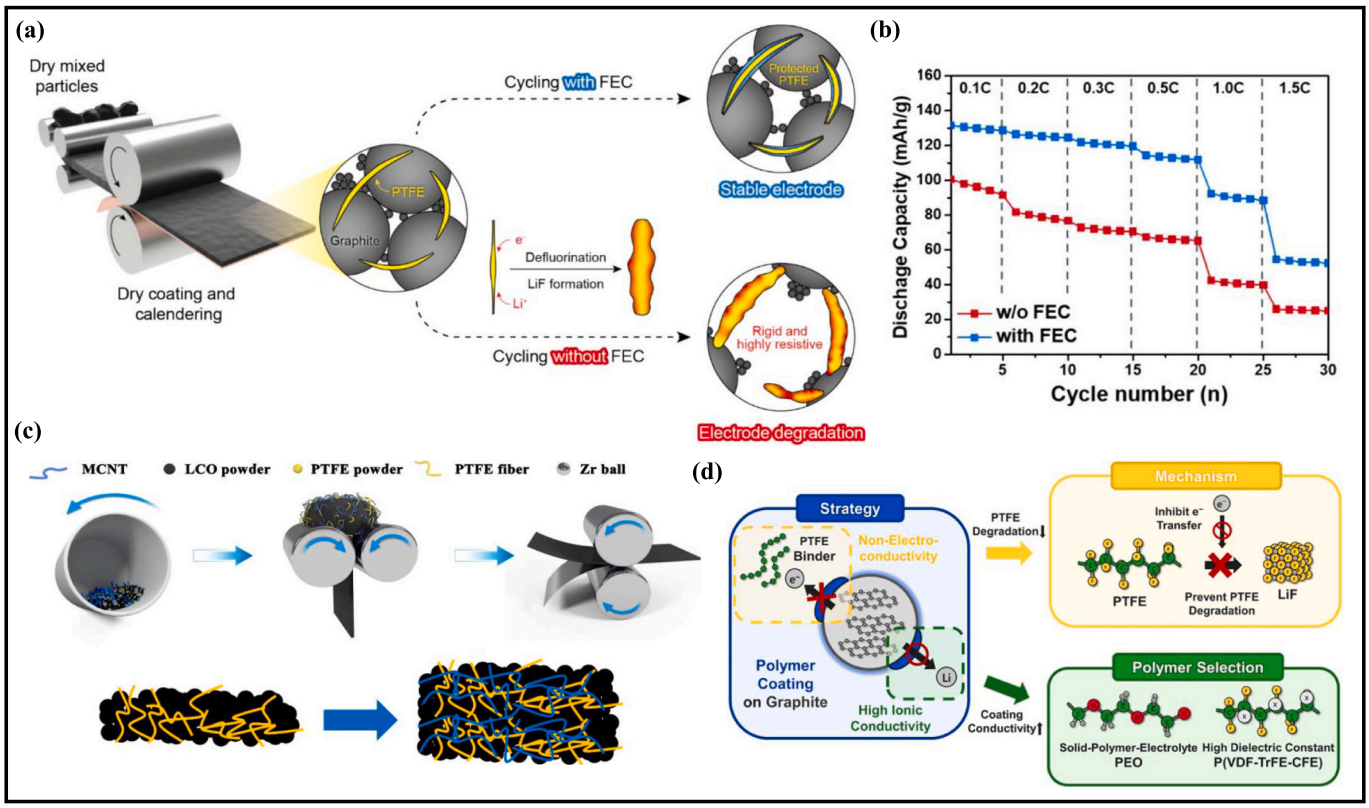

Fig. 13. (a) Graphical illustration of PTFE degradation in dry-processed anodes, comparing conditions with and without FEC, (b) Rate performance of electrodes fabricated with and without the addition of FEC. Reproduced with permission [151] Copyright 2024, The Authors, published by Elsevier. (c) Graphical depiction of the dry processing method as a practical and scalable approach for electrode fabrication. Reproduced with permission [152] Copyright 2024, The Authors, published by American Chemical Society. (d) Pictorial demonstration of a polymer coating on graphite designed to reduce PTFE degradation, along with the polymer selection process for the coating. Reproduced with permission [153] Copyright 2024, The Authors, published by American Chemical Society.

electrode volume changes during cycling. When combined with CMC in aqueous slurries, SBR acts as a flexible binder matrix, forming a strong mechanical and electrical network that enhances electrode cohesion [157]. Apart from that, the use of SBR binders has been widespread as a strengthening agent and flexibility enhancer in a wide variety of industries [158]. CMC, a cellulose derivative with –OH and –CH_2COOH groups with β-(1 → 4)-D-glucopyranose polymers, forms extensive hydrogen-bonding networks that provide mechanical strength and adhesion [159]. The Na^+ groups and single bond of OCH_2COO- are partly substituted instead of -OH groups in the CMC binder [160]. The carboxyl groups can coordinate with transition-metal ions at the electrode surface, acting as anchoring sites that stabilize the cathode–electrolyte interface [161]. In composite systems, CMC's hydrophilicity ensures uniform particle dispersion in aqueous slurries, improving electrode homogeneity and ionic accessibility [162]. The water solubility of CMC binder is mainly due to the presence of hydroxyl and carboxymethyl groups. Alike SBR, CMC binder is also used in graphite and silicon-based negative electrodes. In particular, 2 wt% of CMC binder is enough for effective binding of electrode components and improved cycling stability [163–165].

Apart from these binders, the PAA binder is generally a linear or branched polymer synthesized through the homo-polymerization of acrylic acid or copolymerization with other monomers. PAA contains abundant carboxyl (–COOH) groups that can dissociate into carboxylate anions ($–COO^-$) in aqueous media, enabling strong coordination with surface metal cations (Ni^{2+}, Co^{2+}, Mn^{2+}). This chelation forms robust interfacial linkages, suppressing metal dissolution and enhancing electrode–electrolyte stability [166]. The hydrogen-bonding and ionic crosslinking networks within PAA also improve film integrity and mechanical strength, which help maintain electrode cohesion during repeated cycling [167]. PAA, being an aquatic binder, can also dissolve in various organic solvents that are eco-friendly, such as ethanol. It serves as a valuable approach to thoroughly examine the effects of PAA binder properties, including swelling in electrolyte, binder-Si or binder-C bonding, elastic modulus, adhesion strength between electrode components, and surface chemistry on the performance of Si-based anode materials [168,169]. The PAA binder offers a higher concentration of functional groups, enabling the control of the effective space between electrode materials through copolymerization with other monomers. The copolymerization enables the PAA binder to have a variety of mechanical properties and dissolve more easily in an electrolyte solvent. Recently, CMC and PAA binder have gained increasing attention for high-Ni layered oxide cathodes (e.g., NCM and NCA systems). Their abundant carboxyl (-COOH) and hydroxyl (-OH) functional groups can form strong hydrogen bonds and coordination interactions with the surface of cathode particles [170]. In particular, these functional groups can chelate transition metal ions that tend to dissolve from the cathode during cycling, thereby mitigating metal-ion migration to the anode and stabilizing the electrode–electrolyte interface [171]. Moreover, the chelating effect improves interfacial adhesion between the binder and active material, enhancing mechanical integrity during repeated (de) lithiation processes [172]. Consequently, the use of CMC and PAA as binders not only supports environmentally benign, water-based electrode fabrication but also contributes to improved electrochemical stability and cycle life in high-Ni cathodes. Alike CMC, PAA binder also exhibits a high elastic modulus and low swell ability in carbonates. The high concentration of carboxyl groups in PAA further facilitates lithium-ion transport, increasing ionic conductivity and reducing electrode polarization [173–175].

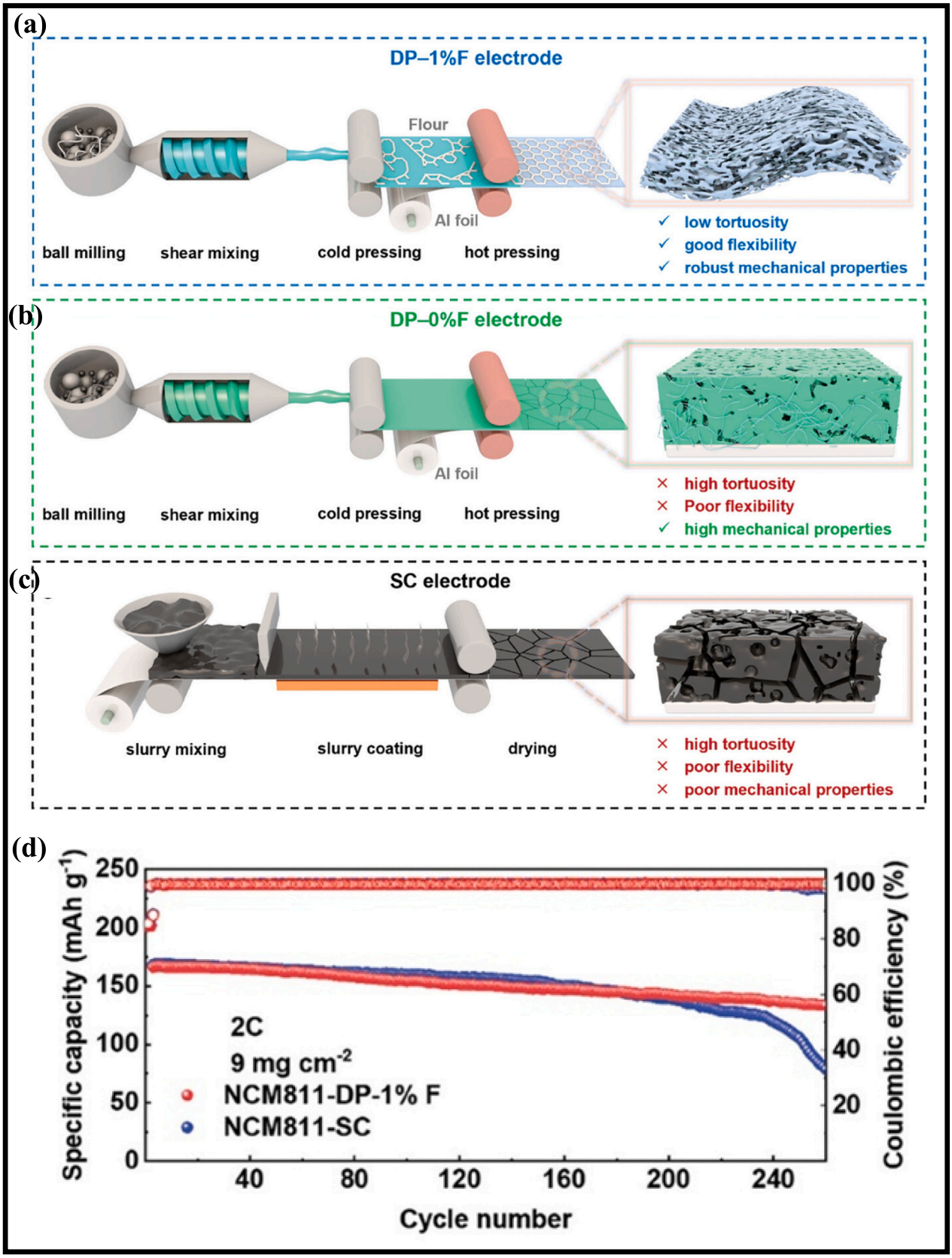

Fig. 14. Pictorial depiction of a DE (a) with 1 % flour and (b) without flour, (c) Pictorial depiction of the slurry-cast (SC) electrode fabrication process, and (d) Cycling performance results of NCM811 electrodes prepared using DP-1 % and SC methods. Reproduced with permission [154] Copyright 2024, The Authors, published by Wiley.

3.2.2.2. Structure–property–performance relationship of SBR, CMC & PAA-based binders. Generally, the high-capacity electrode is achieved with excellent electrochemical performances by combining the appropriate anode layer concerning the current collector and a conventional binder layer, which provides capillary suspension to the electrode [176]. However, this technique provides only 20–30 % of adhesion strength to the battery [177,178]. Therefore, it is imperative to know the merits of an aqueous binder apart from the PTFE binder. SBR binder enables strong adhesion strength that gradually increases the cell performance and battery lifetime. In comparison to PVDF, SBR has a stronger binding force, better heat resistance, and greater flexibility [179]. Fig. 15a-d shows the TEM images of $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) using PVDF and SBR before and after 50 cycles. The TEM images of PVDF and SBR before cycling are smooth, amorphous, and bare surface. Nevertheless, after 50 cyclic tests, the LNMO with PVDF shows irreversible structural deformation owing to its roughened surface with several voids. The LNMO with SBR binder was found to have a uniform, smoothened surface covered by an amorphous layer that enables entrapping self-discharge, electrolyte decomposition, and Mn/Ni dissolution. The strength of adhesion using SBR binder is higher than that of using PVDF binder due to its stronger interactions between the electrode components and cyano groups [180]. In search of alternative binders for conventional binders, CMC binder is also one of the better choices since PVDF struggles with stability due to the presence of fluorine, which leads to safety issues and thermal runaway. The main advantage of CMC is easy to fabricate electrode materials in an aqueous solution, especially graphite-based electrodes [160]. The CMC in Si-based electrodes vastly improves the cyclic performance and reversible capacity of the battery [66]. Finally, it is noteworthy that the prices of SBR (0.2–1 EUR/kg) and CMC (1–2 EUR/kg) are significantly lower than that of PVDF (15–18 EUR/kg) [181].

The glue-like PAA binder is more capable of suppressing electrode desquamation in conversion electrodes due to stress evolution and large volume changes [124]. In particular, linear PAA binder with its higher density and more uniform distribution of carboxyl groups, has exhibited superior adhesion compared with other polymer binders. More importantly, PAA is synthesized via free radical polymerization, allowing precise control over its molecular structure and molecular weight, which ensures consistent and stable performance in various applications by reducing variability from raw material differences [182]. Niesen et al.

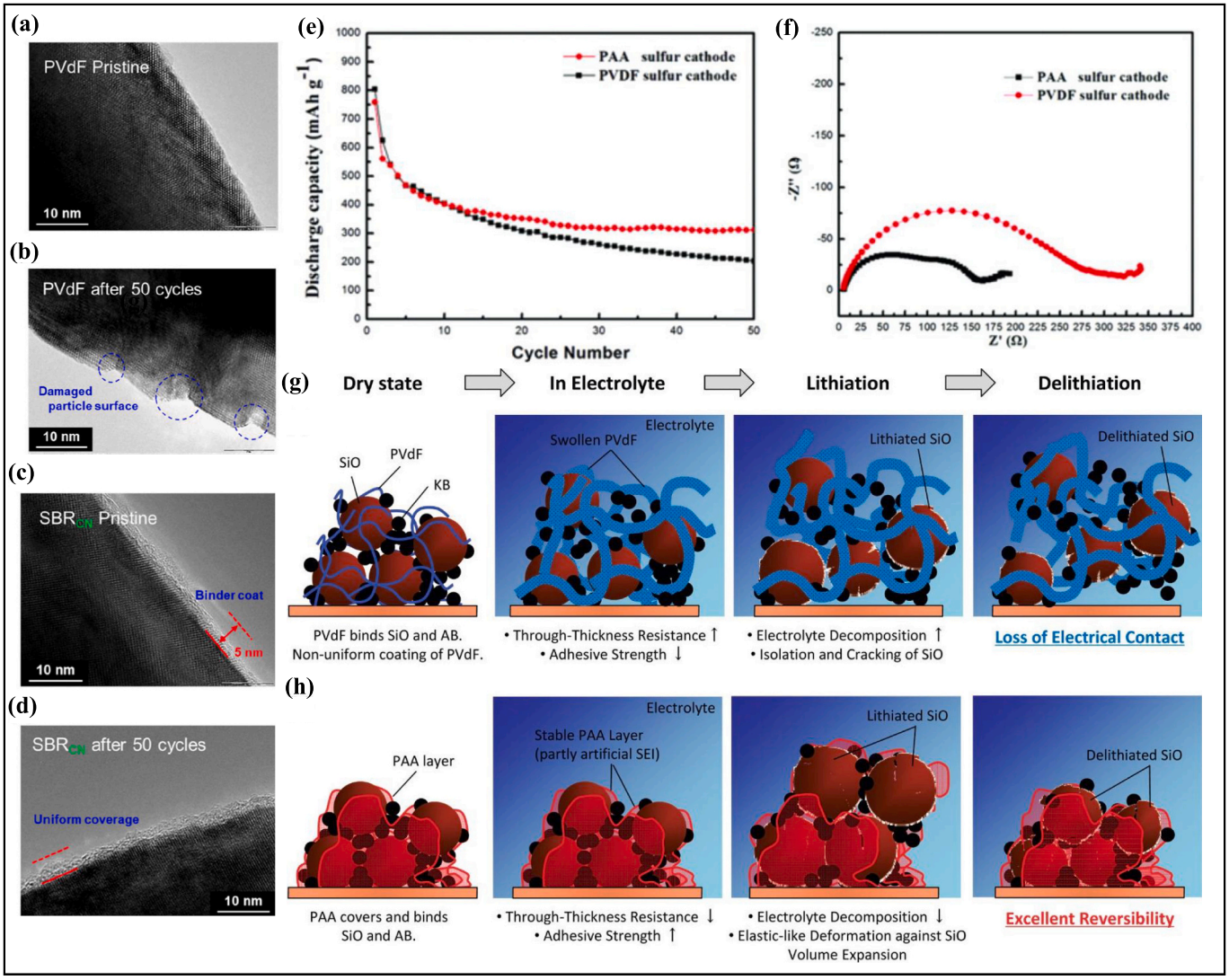

Fig. 15. TEM images of the $LiNi_{0.5}Mn_{1.5}O_4$ electrode prepared with a PVDF binder (a) before and (b) after 50 cycles, TEM images of the $LiNi_{0.5}Mn_{1.5}O_4$ electrode prepared with an SBR binder (c) before and (d) after 50 cycles. Reproduced with permission. [180] Copyright 2020, The Authors, published by American Chemical Society. (e) Cyclic performance, and (f) EIS analysis of sulfur cathodes prepared using PAA and PVDF binders. Reproduced with permission. [68] Copyright 2012, The Authors, published by the Electrochemical Society. Graphical presentation of the proposed mechanism of SiO composite electrodes with (g) PVDF and (h) PAA binders for improved cyclability. Reproduced with permission. [16] Copyright 2011, The Authors, published by American Chemical Society.

revealed that the electrode prepared using PAA binder provided uniform distribution and higher electrochemical performances despite its drying temperature compared to the PVDF-based electrode, owing to its carboxylic acid group that induces strong interaction and suppresses binder migration [183]. Likewise, Zhang et al. determined that the PAA-coated sulfur cathode enabled higher discharge capacity (Fig. 15e), better kinetic characteristics, and lower resistance in EIS tests (Fig. 15f) compared to PVDF-coated sulfur cathode [68]. Typically, CMC and PVDF binders are used as binders for Si anodes, while it is rare to find studies using PAA. The high carboxylic functional groups present in the PAA binder may lead to superior performance for Si anodes, despite their excellent mechanical properties compared to the CMC binder [168]. Compared to conventional PVDF binders that lack polar functional groups, CMC and PAA provide enhanced chemical affinity toward high-Ni oxide surfaces [184]. As illustrated in Fig. 15g, the PVDF, a crystalline-based composite electrode with less cross-linked structures, absorbs electrolyte solution easily and gets bulges, thereby increasing the interface area with electrolyte solution and reducing the adhesiveness of the electrode. The PAA (Fig. 15h), an amorphous-based composite electrode with carboxylic anhydride cross-links, provides strong interaction, uniform coverage, and enhanced mechanical strength that enables strong adhesion of the electrode. Therefore, Komaba et al. determined that the SiO–PAA composite electrode delivered more than 700 mAh g^{-1} of rechargeable capacity with superior cyclability at a rate of 100 mA g^{-1} [16].

In addition, the flexible backbone and low swellability of the PAA binder, combined with its excellent adhesion, chemical stability, and mechanical properties, effectively mitigated volume changes in Si particles and ensured the structural integrity of Si-based electrodes during cycling performances [185]. For example, Magasinski et al. carried out a pioneering study on the impact of PAA binders on the electrochemical performance of Si anodes. Their findings stated that numerous carboxyl groups in the PAA binder formed covalent or hydrogen bonds with Si particles, enhancing Li^+ transport, reducing electrode polarization, and promoting the formation of a stable SEI layer [173]. Therefore, it is confirmed that the SBR, CMC, and PAA are superior to the PVDF binder because of their presence of carboxyl functional groups along with hydrogen bonding. However, the optimal interface structure, influence of mechanical properties, and effect of electrolyte interaction are still being studied and determined in the future [168].

Like all polymer binders, the effect of the SBR binder is also largely dependent on the appropriate amount of binder used for the preparation of electrodes in batteries. For example, Li et al. evaluated the dramatic effect of crack generation in electrodes via coating 25 and 175 g of SBR into the NCM cathode. After 400 cyclic performances, the cathode material coated with SBR-25 g binder cracked due to the contraction and expansion of active material, as shown in Fig. 16a &c. Whereas the cathode coated with SBR-175 g binder (Fig. 16b & d) retained the structure of the shell and contact between the active material surface. Therefore, it is confirmed that the increase in binder coating aids in maintaining strong contact and bonding with active material even after 400 cyclic activity [67]. In addition, it is also mandatory to determine the appropriate duration for the mixing process during the electrode fabrication. Park et al. elucidated that the mixing of SBR binder with active materials for a short time (Fig. 16e) allowed the capillary bridges to remain the same, with SBR binders being trapped at the interface, thereby reducing interfacial tension and maintaining network structure. The mixing of SBR binder with active materials for a longer duration enables the disturbance in capillary bridges and increases the interfacial tensions as shown in Fig. 16f [186].

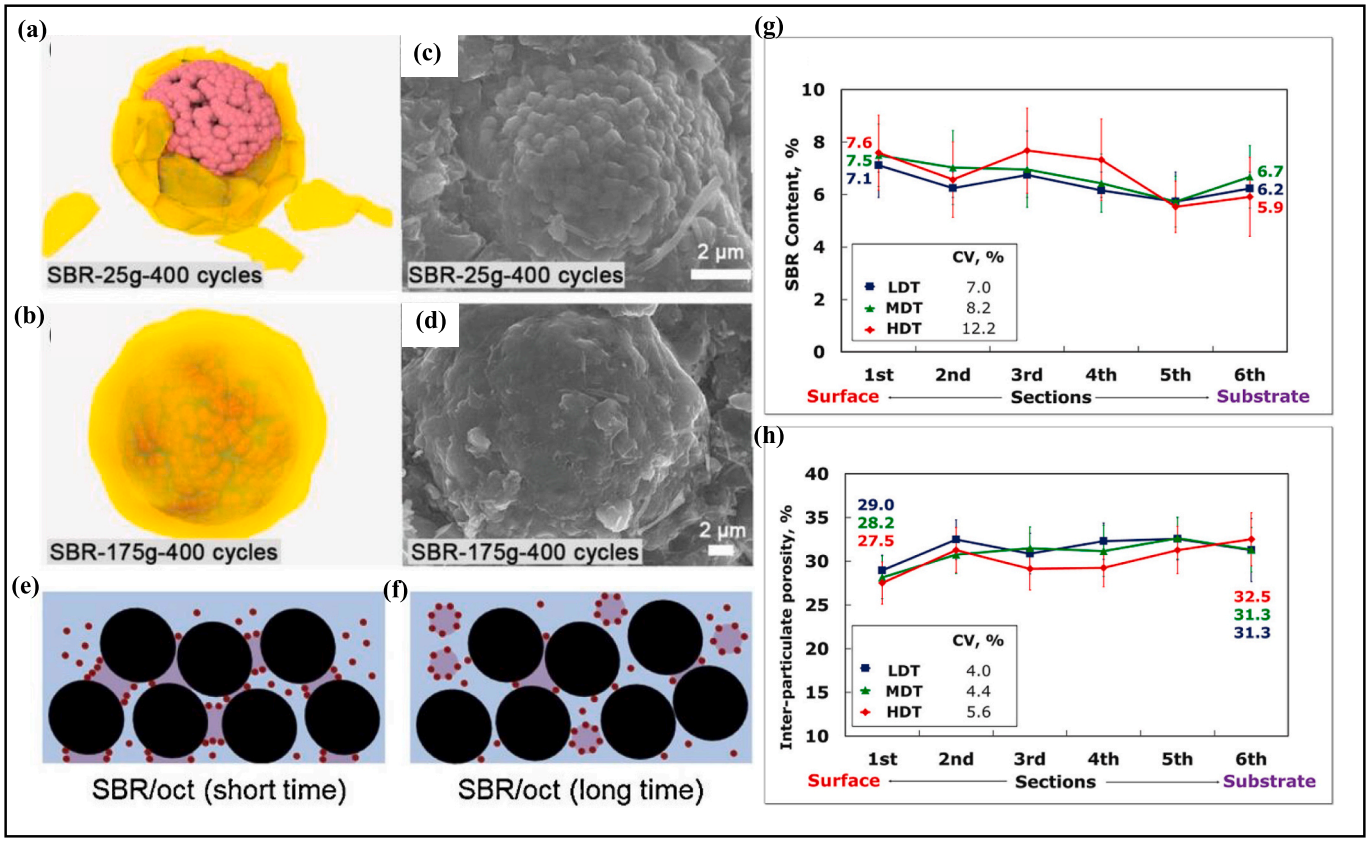

Fig. 16. (a & b) Graphical view and (c & d) SEM images of the cathode prepared using SBR-25 g and SBR-175 g after 400 cycles. Reproduced with permission. [67] Copyright 2022, The Authors, published by Wiley. Schematic structures of capillary suspensions with SBR binder fabricated at (e) short and (f) long mixing times, respectively. Reproduced with permission. [186] Copyright 2019, The Authors, published by Elsevier. (g) SBR distribution analysis and (h) Inter-particulate pore structure of anode layers fabricated at various drying temperatures. Reproduced with permission. [187] Copyright 2023, The Authors, published by Elsevier.

In electrodes infused with SBR binder, the drying temperature also influences inter-particulate pore size distribution. From the results of inter-particulate porosity and SBR distribution analysis (Fig. 16 g), it is found that the SBR content is lower near the substrate side and higher near the anode surface. The higher the drying temperature, the content of the SBR binder increases in surface area while decreasing near the substrate. The inter-particulate porosity distribution results (Fig. 16 h) exhibit a trend opposite to that of the SBR distribution, suggesting that as SBR transfers toward the surface, it clogs the anode surface, resulting in a higher concentration of pores near the substrate. Therefore, the coefficient of variation for both inter-particulate porosity and SBR binder content increased with rising drying temperatures [187]. The amount of CMC binder adsorption influences the dispersion stability and dynamic mobility of graphite particles within the suspension. It also influences the electrochemical performance of the anode, including adhesion strength and charge-discharge capacity. For example, Lee et al. determined the effect of the degree of substitution (DS) of CMC on the electrokinetic behaviour and electrochemical performance of the anode. It was observed that at low DS, CMC adsorbed more effectively onto graphite particles than at high DS [188].

A major challenge in using a water-soluble binder for the positive electrode is the stability of water in Ni-rich cathode materials. Another drawback is the limited oxidative stability of the SBR binder, a partially unsaturated polymer containing a C═C double bond in its structure. At high potentials (>4.3 V vs. Li/Li+), the SBR binder undergoes partial electrochemical oxidation [189]. The uneven distribution of SBR reduces the adhesion force between the current collector and the film, resulting in decreased battery performances [155]. The PAA binder is quite good compared to SBR and CMC binders, however, it suffers from mechanical and electrical flexibility [124]. Additionally, the rigidity and high glass transition temperature (Tg) of PAA homopolymers restrict their elasticity, requiring chemical or physical modifications to improve flexibility and chain mobility [23].

3.2.2.3. Advanced modifications and alternatives to SBR, CMC & PAA binders. Despite CMC's oxidative stability, SBR binder should be incorporated to obtain a flexible composite electrode, since a CMC-based composite electrode without SBR would be brittle due to its inelasticity [190]. In addition, the anodic instability of the SBR binder is resolved by adding the acrylic rubber-based latex/CMC for high-voltage positive electrode applications [191]. The efficiency of this binder arises from its extended conformation in solution, which enables the integration of conductive additives with Si particles, aiding in the construction of a robust composite electrode [192]. Recently, the combination of SBR and CMC binder has gained attention as an efficient binder for secondary batteries [179]. The composite SBR/CMC binder has largely impacted the electrode materials, including transition metal oxides [193,194], graphite [195], silicon [16], and sulfur-based materials [68,196]. For example, the silicon-based anode prepared using SBR/CMC binder provides interaction between hydroxyl groups and carboxylic acid in the CMC, which forms networks on the Si surface, thereby improving the cyclic stability of the battery. The SBR/CMC binder system facilitates uniform binder distribution, as evidenced by studies examining binder distribution in water-based and organic-based $LiCoO_2$ electrode sheets and its influence on cell performance.

Electrodes utilizing the SBR/CMC binder effectively reduce continuous electrolyte decomposition, surface degradation of active materials, and manganese dissolution, leading to improved capacity retention in both half and full cells and decreased self-discharge, even at elevated temperatures [180]. The SBR/CMC binder promotes the formation of a

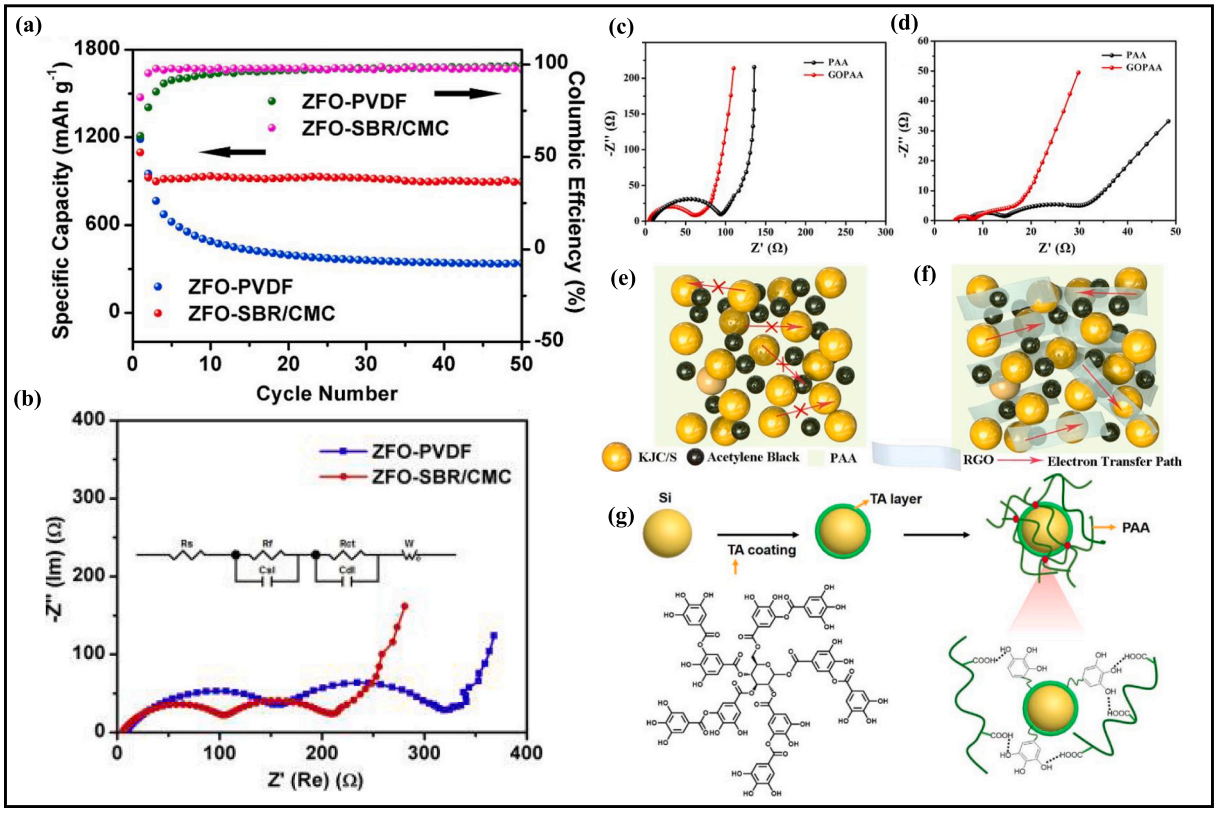

Fig. 17. (a) Cyclic performance and (b) EIS analysis of ZFO-PVDF and ZFO-SBR/CMC electrodes. Reproduced with permission [179] Copyright 2015, The Authors, published by Elsevier. Nyquist plots of PAA and GOPAA electrodes (c) before and (d) after 100 cyclic performances, (e) Pictorial presentation of sulfur electrodes using PAA binder and (f) GOPAA binder. Reproduced with permission [124] Copyright 2017, The Authors, published by Elsevier. (g) Graphical view of the synthetic process for SiSMPs@TA-PAA and the interaction between PAA and TA. Reproduced with permission. [198] Copyright 2019, The Authors, published by American Chemical Society.

passivation layer that improves capacity retention compared to PVDF, although the oxidation of SBR contributes to a minor extent [189,191]. As depicted in Fig. 17a, the discharge capacity of ZFO-PVDF is 461.0 $mAhg^{-1}$ after 15 cycles while it is 873.8 $mAhg^{-1}$ even after 100 cycles in ZFO-SBR/CMC. Similarly, the EIS analysis of the ZFO-SBR/CMC and ZFO-PVDF electrodes suggests that the SBR/CMC binder reduces the ohmic resistance of the electrode, suppresses SEI film formation, and enhances charge transfer reactions at the electrode/electrolyte interface (Fig. 17b). As a result, the ZFO-SBR/CMC exhibits superior electrochemical kinetics compared to ZFO-PVDF, leading to enhanced rate capability and capacity retention. Secondly, Zhang et al. also confirmed that the $ZnFe_2O_4$ anode prepared using SBR/CMC binder provides a 3D-network that enables good capacity retention, high specific capacities, strong electrode adhesion, and excellent rate capability compared to the $ZnFe_2O_4$ anode prepared using PVDF binder [179].

In addition to forming an SBR/CMC composite binder, it is also beneficial to incorporate compounds that address the individual limitations of SBR and CMC binders. For example, Isozumi et al. reported that the SBR binder with modification of partially substituting acrylonitrile and cross-link density for styrene compound has enhanced the capacity retention for high-voltage $LiCoO_2$ electrode due to improved affinity and adhesion to the electrolyte solution [197]. Similarly, Hochgatterer et al. determined in their study that the brittle Na–CMC significantly enhanced the long-term capacity retention and long-term cyclability of Si—C composite electrodes compared to pure CMC binder. The terminal carboxylic acid groups on Na–CMC contribute to improved cycling performance by effectively stabilizing the Si particles and reducing volume changes, thereby maintaining structural integrity throughout the cycling process [164].

PAA binder also follows similar kinds of techniques to get rid of its limitations, such as forming a composite and incorporating one or two compounds into it, enabling a multifunctional binder. Interestingly, Xu et al. developed a conductive binder, reduced graphene oxide (RGO) has been mixed with PAA (GOPAA) to enhance the battery performances. The Nyquist plots of cathodes prepared using GOPAA binder tend to have lower resistance compared to cathodes prepared using PAA binder before and after 100 cycles, as shown in Fig. 17c & d. Additionally, in comparison to the PAA-based electrode, the GOPAA-based electrode improves the effective electrical contact area between the electrode materials, liquid electrolyte, and current collector, while maintaining a stable long-range conductive network (Fig. 17e & f). The GOPAA sulfur electrodes exhibited improved rate performance, higher initial specific capacity, more favourable electrocatalytic kinetics, and reduced capacity decay rate [124]. Likewise, Tian and Wu developed a straightforward, eco-friendly, and cost-effective method to fabricate Si submicroparticle (SiSMP)-based anodes via coating tannic acid (TA) and mixing with PAA binder to create a cross-linked network, as shown in Fig. 17g. The results confirmed exceptional long-term cycling stability and excellent electrochemical performance, with the specific capacity remaining at 2002 mAh g^{-1} even after 100 cycles (Fig. 18a) [198].

Tong et al. developed a multifunctional binder by combining PAA with polyaniline (PAA-PANi) for electrode preparation, and the battery performance is compared with PVDF-based electrode. The weak affinity and electrical insulating nature of PVDF binder for organic electrode materials fail to prevent its dissolution in aprotic organic electrolytes, thereby decreasing electrochemical performances as depicted in Fig. 18b [199,200]. In contrast, the electrode fabricated with PAA-PANi-1 binder (Fig. 18c) provides an increased specific capacity of about 126.1 mAh g^{-1} at 0.1 A g^{-1} and maintains 71.3 mAh g^{-1} at high current densities of 3 A g^{-1}, ensuring stable electrical contact and preventing dissolution (Fig. 18d) [201].

Choi et al. introduced a novel binder by coating a small amount of

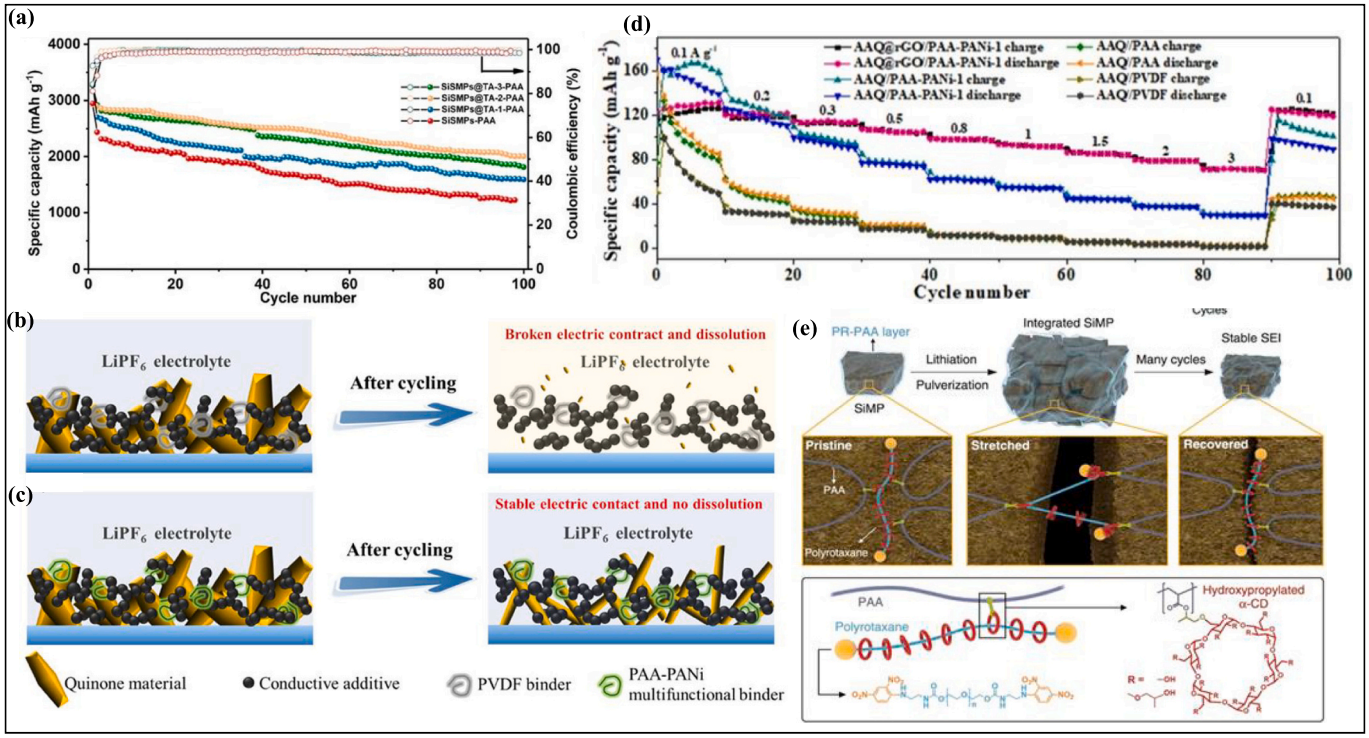

Fig. 18. (a) Cyclic performance of SiSMPs@TA-PAA and SiSMPs-PAA electrodes at 0.1 Ag^{-1} for the first cycle and 0.6 Ag^{-1} for the next consequent cycles. Reproduced with permission. [198] Copyright 2019, The Authors, published by American Chemical Society. Comparison of electrode morphology before and after cycling with a (b) PVDF binder and (c) PAA-PANi multifunctional binder, (d) Specific capacities of AAQ electrodes with PVDF, PAA, and PAA-PANi-1 binders, along with AAQ@rGO electrodes fabricated using the PAA-PANi-1 binder. Reproduced with permission. [201] Copyright 2020, The Authors, published by American Chemical Society. (e) Graphical view of the PR-PAA binder operation in dissipating stress during repeated volume changes of SiMPs, including the chemical structures of PAA binder and polyrotaxane. Reproduced with permission. [24] Copyright 2017, The Authors, published by the American Association for the Advancement of Science.

ring-slide polyrotaxane with PAA binder for Si microparticle anodes (Fig. 18e) [24]. The ring components of the polyrotaxane can move freely, significantly reducing the tension applied to the polymer network. This results in a highly flexible and elastic binder network, allowing even pulverized Si particles to remain interconnected and resist disintegration during repeated delithiation–lithiation cycles. Fig. 19a clearly illustrates the structural evolution of the PAA@Si and PAA/Lys@Si electrodes during cyclic performances, highlighting the advantages of the PAA/Lys binder for Si anodes. Specifically, the 3D cross-linked PAA/Lys network forms strong bonds with Si particles, preventing exfoliation and isolation of active materials. Its high elasticity ensures structural integrity, efficiently dissipating stress caused by the volume changes of Si and buffering the volume expansion of the Si particles. This results in demonstrating excellent cycling stability, even at high mass loading of Si [202]. Guo et al. combined poly(ethylene-*co*-vinyl acetate) (EVA) latex with PAA binder to improve the adhesion and ductility of the electrode. As illustrated in Fig. 19b, the porous silicon electrode with PAA/EVA binder demonstrates a reversible capacity of 2120 mAh g^{-1} at 500 mA g^{-1} even after 140 cycles, owing to its complete inhibition of Si particle aggregation, transport properties, and ductility of PAA/EVA binder [203]. Therefore, these modifications have enabled the PAA binder to effectively manage volume changes, potentially providing self-healing properties that stabilize electrode structures and maintain their integrity during charge and discharge cycles [23,174]. Table 4 details the comparision between traditional vs. multifunctional binders with respect to adhesion strength, mechanical flexibility, ionic conductivity, and environmental impact.

4. Industrial and sustainability perspectives of binder design

4.1.

The transition from laboratory-scale innovation to industrial deployment of advanced binder systems requires careful evaluation of scalability, environmental impact, production cost, and regulatory compliance [79]. Conventional PVDF-based binders, although widely used for their strong adhesion and electrochemical stability, rely on NMP, a costly, energy-intensive solvent classified as a reproductive toxicant and increasingly restricted under EU REACH and global regulations [213]. The need for extensive solvent recovery and drying significantly elevates manufacturing costs and environmental footprint [214]. In contrast, water-based binder systems such as CMC/SBR, PTFE emulsions [157,186] and PAA [215] provide a safer, cost-effective alternative compatible with existing roll-to-roll electrode fabrication. These binders provide robust mechanical flexibility and strong adhesion while enabling NMP-free processing and facilitating streamlined recycling [216]. Their hydrophilic nature enhances electrode delamination and active material recovery, aligning with circular economy principles

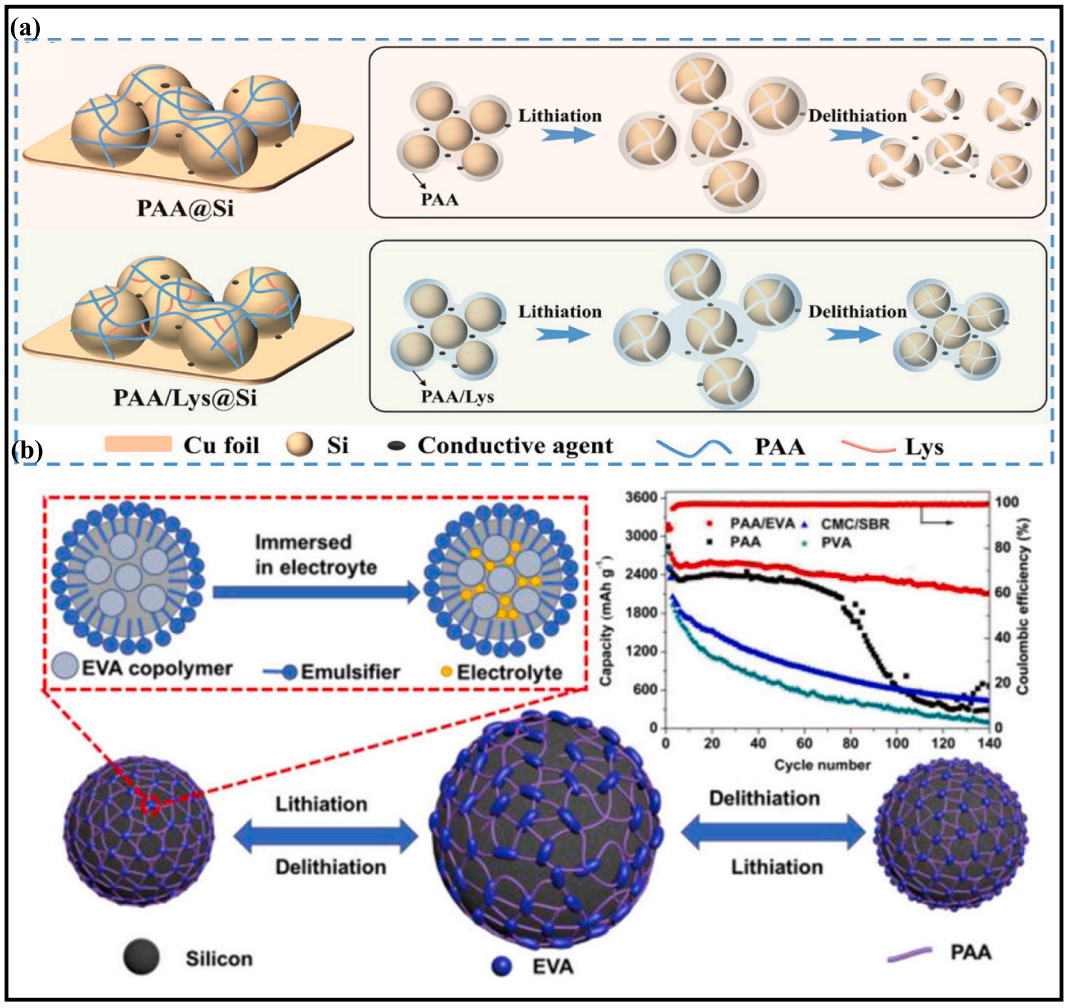

Fig. 19. (a) Pictorial depiction of the morphological changes in PAA@Si and PAA/Lys@Si electrodes during lithiation and de-lithiation processes. Reproduced with permission. [202] Copyright 2023, The Authors, published by American Chemical Society. (b) Graphical depiction of a porous Si electrode with a PAA/EVA binder, along with the cycling performance of Si electrodes using various binders. Reproduced with permission. [203] Copyright 2019, The Authors, published by American Chemical Society.

Table 4
Comparision between traditional vs. multifunctional binders with respect to adhesion strength, mechanical flexibility, ionic conductivity, and environmental impact.

Binder Type	Polymers	Adhesion Strength	Mechanical Flexibility	Ionic Conductivity / Ion Transport	Environmental Impact & Processability	References
Traditional (Non-Functional)	PVDF	Moderate (depends on molecular weight and crystallinity)	Limited elasticity; prone to cracking under cycling stress	Poor non-ionic and hydrophobic; relies on conductive additives	Requires NMP solvent; high processing cost; non-biodegradable	[204,205]
Aqueous (Water-Soluble)	PTFE, CMC, SBR, PAA	High (via hydrogen bonding and carboxylate anchoring)	Good flexible network structure resists delamination	Moderate hydrophilic matrix enhances Li^+ mobility	Eco-friendly; NMP-free; compatible with large-scale coating	[206,207]
Conductive Polymer Binders	PAN, PANI, PEDOT:PSS	Strong interfacial adhesion due to π–π and polar interactions	Excellent flexibility and electronic connectivity	Facilitates good electronic & ionic percolation	Moderate sustainability; solvent choice critical	[103,208]
Bio-Based / Renewable Binders	Alginate, Chitosan, Lignin-based polymers	High chelation with metal ions improves adhesion	High natural polymer elasticity	Moderate hydrophilic, enables Li^+ conduction	Renewable, biodegradable, NMP-free	[209,210]
Multifunctional / Dynamic Cross-Linked Systems	PAA-PEO copolymers, supramolecular & self-healing binders	Very high covalent and dynamic bonding ensures robust interface	Excellent reversible bonds dissipate mechanical stress	High ionic segments enhance Li^+ conduction pathways	Designed for sustainability; scalable aqueous processing	[211,212]

and meeting regulatory frameworks such as the EU Battery Regulation 2023/1542 [134]. PAN binder, on the other hand, face scalability constraints due to their dependence on toxic or energy-intensive solvents (DMF, DMSO, or high-temperature sintering) [57]. Although PTFE offers excellent mechanical integrity and is frequently employed in dry-processed electrodes, its limited recyclability and high processing energy remain industrial bottlenecks [217]. Fig. 20 presents a binder evolution — from PVDF to dynamic cross-linked polymer networks.

4.2.

Future industrial pathways are increasingly oriented toward sustainable, multifunctional, and cost-effective binder chemistries. Bio-derived and biodegradable polymers including alginate, chitosan, lignin, cellulose derivatives, and gelatin-based materials that offer renewable origins, water-based processability, and tunable mechanical properties, making them strong candidates for environmentally responsible, scalable electrode fabrication [218]. Simultaneously, advances in dynamic and self-healing networks based on imine, disulfide, or Diels–Alder mechanisms provide reversible adhesion and autonomous structural recovery under cycling stresses, mitigating particle detachment and extending electrode lifespan [219].

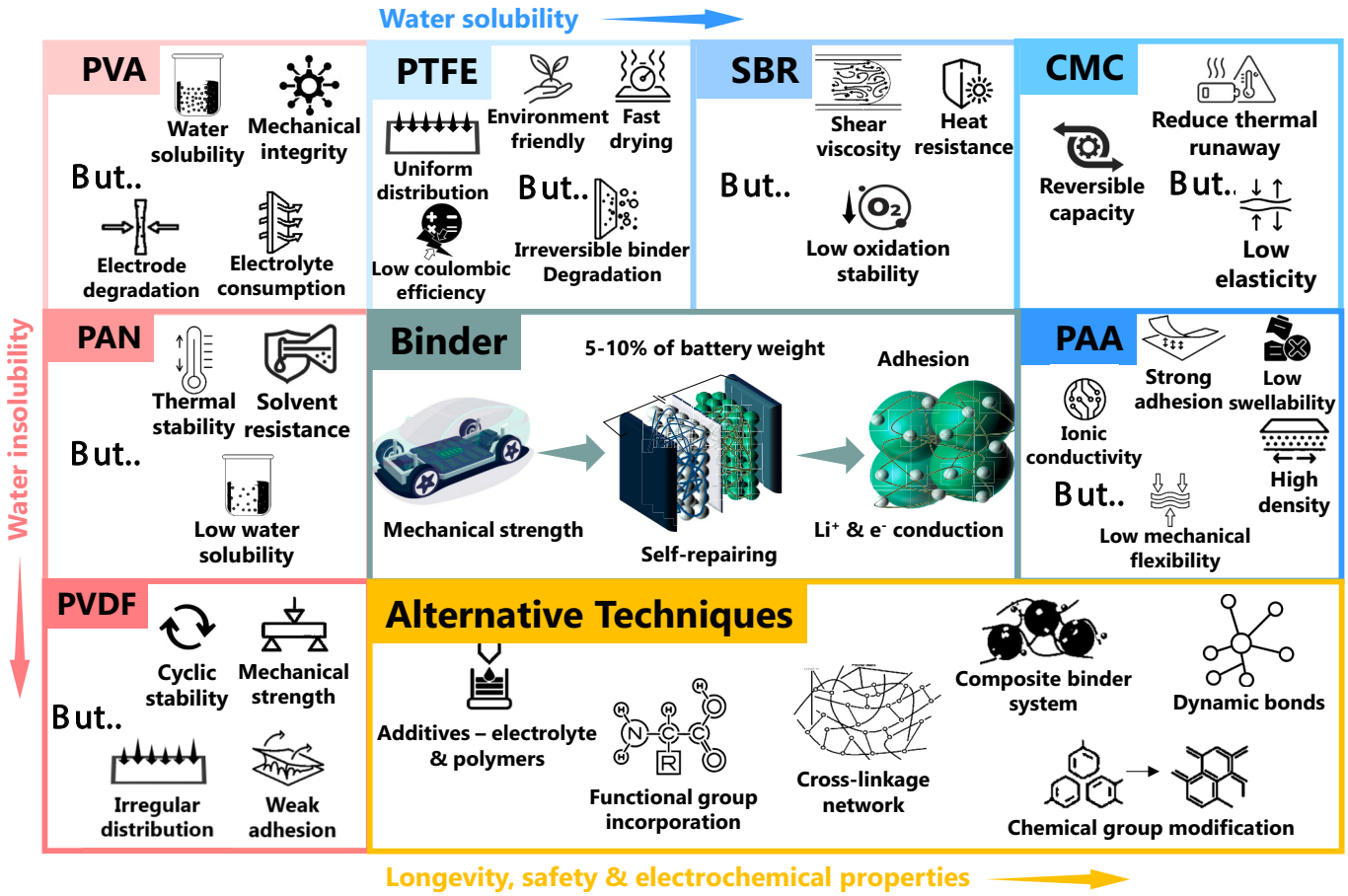

Fig. 20. Binder evolution — from PVDF to dynamic cross-linked polymer networks.

4.3.

Industrial adoption also requires recognizing the emerging role of the binder–electrolyte interphase (BEI) [220]. Functional binders can participate in controlled interphase formation, improving SEI uniformity, suppressing gas evolution, and stabilizing Li-ion transport features essential for long-term safety and durability [221]. Moreover, water-soluble, low-toxicity binder systems significantly simplify end-of-life recycling and materials recovery, reducing costs and emissions across the battery lifecycle [222]. Together, these strategies contribute significantly to enhancing electrode longevity, electrochemical stability, and overall cell safety. For next-generation binders to achieve commercial viability, their design must prioritize low cost, minimal toxicity, and high recyclability. Water-processable and bio-derived binders such as alginate, cellulose, and chitosan offer significant advantages by reducing manufacturing costs, energy use, and environmental impact [223]. Sustainable binders must also avoid toxic components and harmful by-products, while water-soluble or degradable systems greatly simplify electrode recycling and material recovery. Overall, optimizing binders for affordability, safety, and recyclability is key to supporting large-scale, eco-friendly battery production [218].

4.4.

Machine learning (ML) is increasingly applied to accelerate binder development, enabling predictive screening of polymer–electrode interactions, optimization of molecular architectures, and identification of sustainable binder candidates through multi-objective design frameworks [224]. These methods leverage structure property databases and multi-objective optimization algorithms to identify promising compositions with tailored mechanical, electronic, and electrochemical properties [225]. In solid-state batteries, where interfacial contact governs energy density and cycling stability, binders must additionally balance ionic conductivity, mechanical compliance, and chemical stability. Polymer–ceramic composites and ion-conducting gel networks are emerging as promising solutions for maintaining seamless interfaces and suppressing dendrite formation [226]. Collectively, these industrial and sustainability-driven advancements in binder design will be central to enabling large-scale, eco-efficient, and next-generation battery manufacturing.

5. Conclusion and future perspectives

Looking ahead, the development of next-generation binder materials should prioritize multi-functionality, sustainability, and interfacial adaptability. Designing binders that can simultaneously enable strong adhesion, high ionic conductivity, and controlled interfacial chemistry is critical for emerging systems such as Li—S, Na-ion, and solid-state batteries:

- Li—S batteries: Binders should mitigate polysulfide dissolution through functional groups capable of chemical anchoring (–COOH, –NH_2) and maintain elasticity during large volume changes. Promising candidates include PAA, CMC, and hybrid conductive polymers such as PANI/PEDOT composites [57].
- Na-ion batteries: Given larger Na^+ ionic radii and different electrode expansion behaviour, flexible, water-processable binders (CMC/SBR, alginate, PVA) with enhanced ion transport properties are preferred [227].
- Solid-state batteries: Binder functionality extends to ensuring intimate solid–solid contact, ionic percolation, and interfacial stability. Soft polymeric or hybrid ion-conductive binders, particularly those with dynamic cross-linking (PEO–PAA, PAN–LiTFSI composites), are gaining importance [228].

In summary, binder must not only improve electrochemical performance but also support scalable, environmentally friendly and regulation-compliant manufacturing to keep pace with the rapidly advancing field of sustainable battery technologies. Recently, multi-functional binder designs have emerged to overcome the limitations of traditional single-component systems. These new binders incorporate enhanced mechanical flexibility, self-healing capability, stretchability, and environmental responsiveness, enabling more robust and adaptable electrode architectures. A deeper understanding of the strength of individual binders further facilitate strategic blending or crosslinking to create composite systems with synergistic properties. At the same time, the rational design of novel polymer networks continues to drive comprehensive improvements in electrode stability and performance.

The following recent advancements in next-generation binders are essential for optimizing battery performance, offering enhanced stability, efficiency, and longevity:

(i) 3D Electrode Structural Design for Enhanced Carrier Transport. Electrodes frequently experience deformation, parasitic reactions, excessive electrolyte consumption, and shortened cycle life. To mitigate these issues, 3D cross-linked binder networks have demonstrated significant benefits in reinforcing polymer mechanical properties. Among these strategies, metal cation–assisted cross-linking is particularly versatile and widely applicable [24,229]. This approach forms continuous 3D pathways that facilitate efficient ion/electron transport, enhance mechanical resilience against volume fluctuations, and prevent active material aggregation, collectively improving rate capability and cycling stability [230,231].

(ii) Tailored Composite Binder Systems for High-Performance Energy Storage. Composite binders have emerged as multifunctional systems capable of integrating the strengths of conventional binders while eliminating the reliance on additional carbon additives. This reduces inactive material content and enables higher energy density in advanced battery architectures [232].

(iii) Chemical Group Alteration for Property Optimization. The physical, chemical, and structural characteristics of composite binders can be precisely tuned through modifications such as optimized synthetic routes, selective doping, monomer or initiator variation, and controlled precursor concentrations [233]. These strategies yield highly conductive, flexible binder systems that improve overall electrode conductivity [234]. For polymers with inherently weak adhesion, introducing targeted functional groups offers an effective pathway to significantly enhance bonding with active materials.

(iv) Molecular Structure Engineering for Multifunctionality. To achieve improved conductivity, electrolyte uptake, ductility, and mechanical adhesion, binder molecular structures are strategically modified by incorporating diverse functional groups [233]. Grafting approaches commonly employ –OH groups to attach adhesion-promoting chains such as acrylic or acrylonitrile. Natural saccharides including agarose and gelatin are especially attractive due to their abundance of functional groups, which provide strong interfacial bonding between active materials and current collectors [235,236].

(v) Functional Group Incorporation for Controlled Interfacial Reactions. Modifying binder backbones or side chains enables fine-tuning of electronic properties, including HOMO/LUMO energies and band gaps. Surface passivation can be achieved through targeted chemical alterations, minimizing unwanted electrolyte decomposition. Additionally, the incorporation of functional surfactants or ligands on active material surfaces strengthens their chemical and electronic interactions with binders, further stabilizing electrode microstructures [231,237].

CRediT authorship contribution statement

Subhiksha Venkatesh Raja: Writing – review & editing, Writing – original draft, Methodology, Investigation, Conceptualization. **Hongliu Dai:** Writing – review & editing. **Zhangsen Chen:** Writing – review & editing. **Oumayma ELJarray:** Writing – review & editing. **Siyi Cao:** Writing – review & editing. **Shuhui Sun:** Writing – review & editing, Writing – original draft, Validation, Supervision, Resources, Project administration, Funding acquisition, Conceptualization. **Gaixia Zhang:** Writing – review & editing, Writing – original draft, Validation, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

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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Data availability

Data will be made available on request.

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