Design Principle and Development Trends of Silicon-Based Anode Binders for Lithium-ion Batteries: A Mini Review

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Abstract: Silicon (Si), recognized as a promising alternative material for the anodes of lithium-ion batteries, boasts a high theoretical specific capacity and abundant natural availability. During the preparation of silicon-based anodes, binders play a pivotal role in ensuring the cohesion of silicon particles, conductive agents, and current collectors. The structure and performance of these binders are critical for the mechanical stability, electrical conductivity, and stress dissipation capacity of the anodes. This review initially outlines the structural characteristics of various binders, including linear, branched, and three-dimensional cross-linked types. It then delves into the relationship between the structure and properties of these binders in the context of their application in high-performance lithium-ion batteries, focusing on their mechanical properties, electrical conductivity, and self-healing capabilities. Particular attention is given to the design strategies for binders that facilitate stress dissipation, with an emphasis on integrating multifunctional polymer binders renowned for their superior conductive and self-healing features. Such binders contribute to the formation of a robust three-dimensional network structure via multiple bonding mechanisms, including chemical, non-covalent, and coordination interactions. This configuration significantly enhances the adhesion between silicon particles, thereby facilitating the efficient dissipation of stress, which is a key aspect for ensuring the long-term cycling stability of lithium-ion batteries. Lastly, the paper explores future development directions for silicon anode binders, advocating for a thorough investigation into the synergy of diverse structural and functional combinations, with the aim of advancing the performance and practical application of silicon-based lithium-ion batteries.

Keywords: Si-based anode, Multifunctional polymer binders, Lithium-ion battery, 3D crosslinking structure, Stress-dissipative network, Ionic conductivity.

1. INTRODUCTION

The commercialization of lithium-ion batteries (LIBs) marks a significant milestone in the realm of energy storage, fundamentally reshaping the landscape of modern human existence [1-4]. As the demand for portable electronic devices continues to soar and the automotive industry shifts towards hybrid and electric vehicles, the necessity for high-performance energy storage systems has become increasingly apparent. LIBs have risen to prominence due to their remarkable attributes, including high energy density, elevated coulombic efficiency, and minimal self-discharge characteristics. However, the capacity, rate performance, and cycling stability of LIBs are inherently tied to the electrode materials utilized within them. For the past two decades, graphite materials have been the cornerstone of LIB anodes, prized for their safety and environmental compatibility. Nonetheless, graphite-based anodes present notable limitations, chief among them being their low specific capacity of 372 mAh g^{-1}. This limitation leads to diminished Coulombic efficiency and subpar rate performance, ultimately hindering the overall performance of lithium-ion batteries. In response to these challenges, silicon anodes have emerged as highly promising alternatives for next-generation lithium batteries [5-8]. Silicon offers a substantially higher specific capacity of 4200 mAh g^{-1}, dwarfing that of graphite. Moreover, silicon's abundance in the earth's crust, coupled with its low operating potential and exceptional safety profile, has sparked widespread interest and catalyzed extensive research efforts into its application as an anode material for lithium-ion batteries.

However, numerous challenges persist in the commercialization of silicon anodes. Throughout the lithiation and delithiation processes of lithium battery charging and discharging, silicon anodes undergo significant volume changes (>300%), leading to electrode crushing, structural disintegration, and the recurrent formation of unstable solid electrolyte interface (SEI) layers [9, 10]. Consequently, these flaws in silicon anodes result in a marked decrease in capacity, inadequate cyclse life, diminished initial Coulombic efficiency (ICE), and reduced conductivity. Tackling these challenges is imperative for fully realizing the potential of silicon anode materials and enhancing the performance of lithium-ion batteries for more effective commercial applications. Following a

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straightforward, secure, and sustainable commercialization approach, the present challenges hindering the widespread adoption of silicon anodes primarily revolve around the following areas of enhancement: structural optimization, surface/interface management, innovative electrolyte design, and novel binders [11-13]. Structural optimization involves embedding silicon nanoparticles within carbon-based materials utilizing a core-shell structure design to mitigate volume expansion during charge and discharge cycles, thus preserving electrode stability. For instance, the successful implementation of a nitrogen-doped porous carbon shell as a protective layer for silicon anodes exemplifies this approach [14]. Surface/interface control employs surface coating technology to establish a protective oxide film, with alumina-coated silicon-based nanowire arrays demonstrating enhanced cycling stability of the electrode [15]. Additionally, electrolytes containing additives can facilitate the formation of a stable solid electrolyte interface, while those incorporating lithium salts like bis(fluorosulfonyl)imide (LiFSI) can effectively mitigate volume expansion in silicon anode materials, thereby enhancing battery cycle life [16]. Concurrently, the development of new binders is paramount. Polymer-based materials, as a novel binder option, exhibit favorable elasticity and adhesion, facilitating the fixation of silicon anode material and ensuring robust contact with the conductive agent and current collector, thereby enhancing electrode structural stability. The integration of these enhancement strategies significantly bolsters the commercial viability of silicon anode materials and propels the progression and dissemination of lithium-ion battery technology.

The significance of binders in lithium-ion batteries cannot be overstated, despite constituting a small fraction of the total electrode mass (typically ≤ 20%) [17-19]. Its primary function is to enhance adhesion between the electrode material, conductive agent (such as carbon black), and current collector (such as copper foil), ensuring a strong amalgamation, structural integrity, and optimal electrical conductivity at the electrode-current collector interface [20]. Consequently, designing and optimizing binders is paramount for enhancing battery performance. By carefully selecting appropriate binder types, fine-tuning formulations and additives, and controlling their distribution and concentration within the electrode, significant improvements can be achieved in the structural stability and conductivity of the battery. This meticulous approach is essential for advancing the development and widespread adoption of lithium-ion battery technology in commercial applications.

The selection of binders plays a crucial role in the performance of lithium-ion batteries, particularly in addressing the challenges posed by different electrode materials. Although PVDF and CMC binders are commonly employed in carbon anodes for lithium batteries, ensuring their prolonged cycle stability due to the limited volume expansion of carbon-based anodes (approximately 10%). However, PVDF and CMC binders encounter significant volume expansion in silicon-based anodes, resulting in inadequate binding capacity and severe silicon particle crushing [21]. PAA, a polymer binder renowned for its solubility and dispersion properties, has emerged as a promising alternative and finds extensive application in lithium-ion batteries. It boasts excellent adhesion and flexibility, effectively securing electrode materials, preserving electrode structure stability, and significantly enhancing battery cycle life and safety performance. In-situ thermal polymerization of chain sodium carboxymethyl cellulose (CMC) and thiourea (SN) results in the formation of a tightly bonded, hydrogen-bonded cross-linked thiourea-based polymer binder, CMC-co-SN, enhancing the bond strength between the binder and the current collector through physical interlocking. Application of the binder to micron and nanometer Si particles yields silicon anodes with high initial Coulombic efficiency (91.5% for μSi and 83.2% for nSi) and exceptional cycle performance (μSi: 1121 mAh g^{-1} after 140 cycles, nSi: 1083 mAh g^{-1} after 300 cycles) [22]. The performance of these new binders underscores the critical importance of binder selection in enhancing lithium-ion battery performance. They significantly contribute to enhancing electrode structural stability, adhesion, and conductivity, bolstering battery cycle life and safety. Furthermore, they offer insights into the commercial application of silicon anodes.

This mini-review presents a comprehensive overview of various types of silicon-based anode binders, including linear, branched, three-dimensional network, and stress-dissipative binders (Figure **1**). It begins by discussing the molecular structure design of the binder and proceeds to analyze its mechanical properties, self-healing capabilities, conductive properties and enhancement of its stress-dissipative network structure. Furthermore, it succinctly summarizes the latest advancements in various binders.

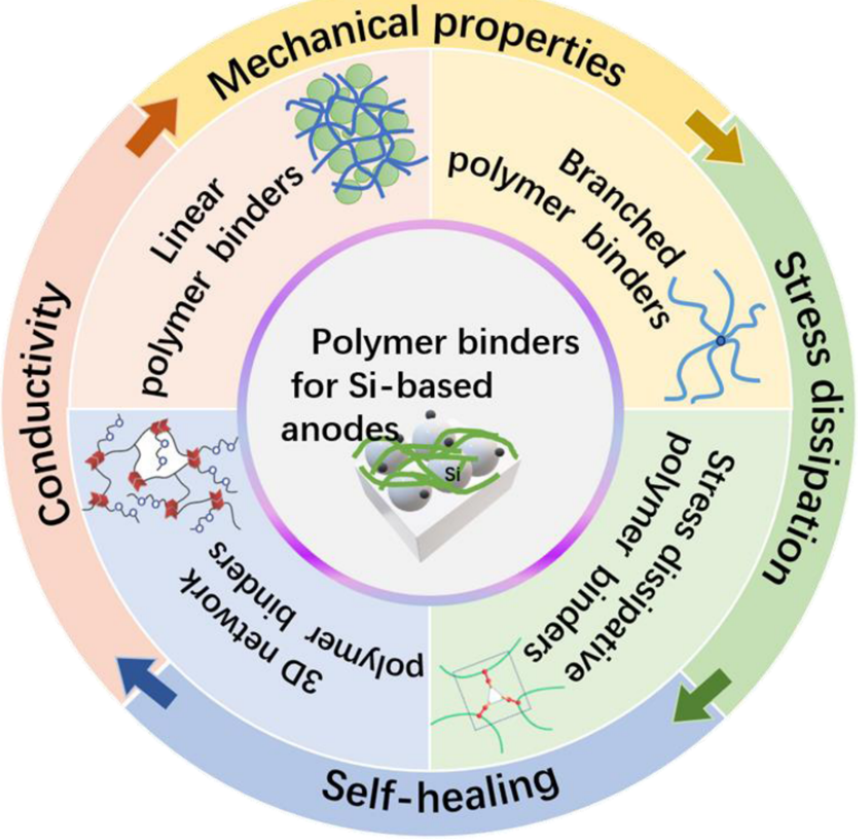

Figure 1: An overview of various binders for Si-based anodes based on polymer structures.

2. POLYMER BINDERS FOR SI -BASED ANODE

To realize the practical implementation of silicon-based anodes and unlock their theoretically high capacity, a range of challenges stemming from significant volume fluctuations during repeated charge and discharge cycles must be tackled [23, 24]. These challenges encompass fragmentation, cracking, detachment of active materials, and instability of the solid electrolyte interface (SEI). Despite their small proportion in electrodes, binders play a crucial role [25-27]. Through various mechanisms like van der Waals forces, electrostatic forces, and chemical bonds, binders effectively amalgamate active materials, conductive agents, and current collectors, thereby alleviating stress caused by silicon volume changes and enhancing cohesion among active materials, conductive agents, and electrode current collectors, thereby preserving electrode integrity. For instance, the selection and composition of binders profoundly impact the performance of silicon-based anodes. Research has shown that different types and concentrations of binders exert diverse effects on the electrochemical and mechanical properties of electrodes. For example, increasing the content of specific binders can improve the fracture behavior of silicon microcolumns and suppress lithium silicide detachment [28]. Additionally, the hardness and composition of binders are closely linked to the performance of alloy anodes. Therefore, binder selection not only pertains to their binding efficacy but also considers their comprehensive influence on overall battery performance, aiming to achieve dependable applications of high-performance silicon anode lithium batteries [29-32]. Furthermore, it is crucial to acknowledge the structural variety of different binders, such as linear, branched, and three-dimensional network structures, along with their distinct functionalities, including mechanical properties, conductivity, self-healing, and stress dissipation, all of which are pivotal for enhancing the performance of silicon anodes.

2.1. Linear Binders

Linear polymer binders interact with silicon and other battery components primarily through functional groups located on their side chains [11]. These interactions are essential for achieving robust adhesion between the binder and silicon particles [33]. However, due to their one-dimensional structure, linear binders offer limited binding strength, which can only partially mitigate the volume expansion of silicon anodes. Consequently, electrodes incorporating linear binders exhibit increased susceptibility to mechanical fracture and associated performance degradation.

Despite these limitations, linear polymer binders remain prevalent in large-scale applications due to their economic viability and straightforward synthesis. Additionally, the binding strength of linear binders can be tailored through strategies like copolymerization and blending, enabling further optimization for silicon anode performance. These advantages contribute to the continued use and exploration of linear binders within the realm of advanced energy storage systems.

PAID Binder

This binder, prepared from a polyimide-based aromatic polymer consisting of pyromellitic dianhydride, 4-aminophenyl ether (soft segment), and 3, 3'-diaminobenzidine (hard segment) as shown in Figure **2a**, plays a crucial role in influencing the performance of the electrode. To investigate the impact of this ratio on binder performance, Kim *et al*. fixed the ratio of pyromellitic dianhydride at 1.0 and designed the ratios of 4-aminophenyl ether to 3,3'-diaminobenzidine as 1.0:0, 0.9:0.1, 0.8:0.2, 0.5:0.5, and 0:1.0, respectively. The corresponding polymers are named Kapton, PAID10, PAID20, PAID50, and PAID100.

The PAID20 binder emerged as a standout performer, exhibiting not only the highest charge-

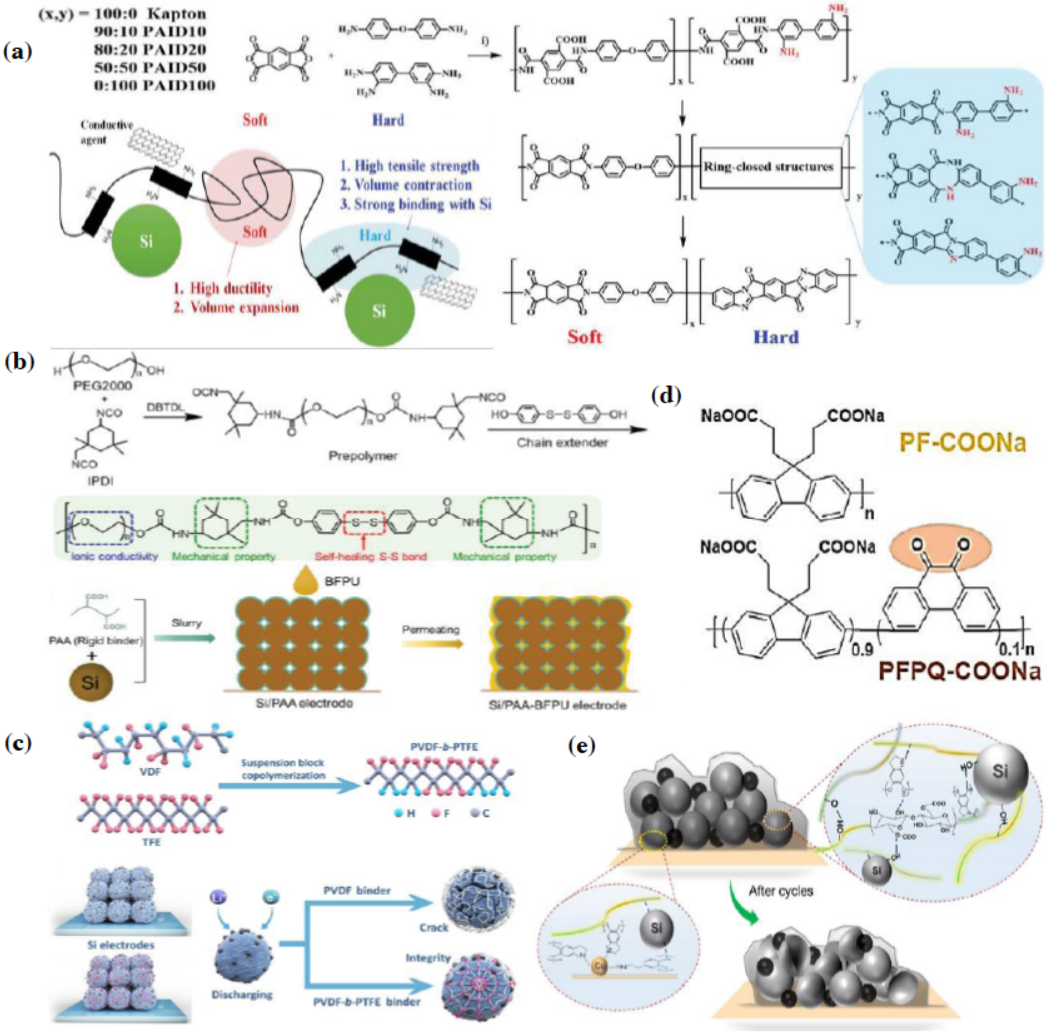

Figure 2: **a**) Synthesis of PAID binders. **b**) Synthesis of the BFPU polymer and fabrication of the Si anode with double-wrapped PAA–BFPU binder. **c**) General process of using VDF and TFE as the precursors for synthesizing PVDF-b-PTFE. And schematic diagram of using the block copolymer binder to stabilize Si electrodes during the electrochemical reactions. **d**) Proposed molecular structure of PF-COONa and PFPQ-COONa. **e**) Schematic diagram of the SiMP anodes with the binder of CMC-10% PDA and the interaction between the electrode materials.

discharge capacity but also unique characteristics during lithiation and delithiation. Following full lithiation, PAID20 displayed a significant volume expansion of 146%, comparable to Kapton and PAID10. However, its volume retention rate of 89% is close to PAID50 and PAID100. This contrasting behavior suggests that PAID20 achieves an optimal balance between mechanical performance and capacity retention as a silicon anode binder. This can be attributed to the well-balanced ratio of soft and hard segments within the polymer backbone [34].

The soft segment in the copolymer binder, designed with an appropriate soft-to-hard segment ratio, allows for volume expansion during lithiation, alleviating mechanical stress on the polymer. Conversely, the hard segment promotes volume recovery through strong intermolecular interactions with the Si particles and conductive additives. This complementary interplay between soft and hard segments within the binder can achieve desirable properties

PAA-BFPU Binder

This paper introduces a dual-coating binder system for silicon anodes designed to achieve gradual stress dispersion. The concept of a "soft-hard" material combination mirrors that of the PAID binder. The rigid polyacrylic acid (PAA) component resides internally to strengthen adhesion and dissipate internal stresses during lithium insertion (lithiation). Simultaneously, an elastic, self-healing bifunctional polyurethane (BFPU) serves as an external buffer layer within the silicon electrode (Figure **2b**). This BFPU layer functions to disperse residual stresses, maintain the integrity of silicon particles, and facilitate electrode structure restoration during lithium removal (delithiation). The close interaction between PAA and BFPU further enhances the structural stability of the silicon anode. Chemical cross-linking between PAA and BFPU occurs via both covalent and hydrogen bonds. The presence of ester bonds between the PAA binder and silicon particles is confirmed by the ester bond stretching vibration band observed at 1727 cm^{-1} in the PAA-BFPU/Si Fourier-transform infrared (FTIR) spectrum. This chemical cross-linking contributes to mitigating significant volume expansion and ensuring the integrity of the silicon electrode [35].

Jiao *et al*. investigated the feasibility of the "hard-to-soft" versus "soft-to-hard" strategies for binder design through comparative experiments. Their findings demonstrated that an internal elastic binder lacked sufficient mechanical support to manage internal stresses, while an external rigid binder failed to release the stress from compressed silicon particles. This ultimately resulted in more severe structural damage. In terms of electrochemical performance, the PAA-BFPU binder delivered impressive results. The Si/C-500 anode using this binder achieved a specific capacity of 500 mAh g^{-1} and maintained an 88% capacity retention after 200 cycles. Furthermore, the SiOx anode incorporating the PAA-BFPU binder exhibited a remarkable capacity of 1000 mAh g^{-1} after 200 cycles at a current density of 300 mA g^{-1}, highlighting its exceptional performance.

PVDF-b-PTFE Binder

The PVDF-b-PTFE binder, a novel design, consists of 80 wt% vinylidene fluoride (VDF) and 20 wt% tetrafluoroethylene (TFE) as depicted in Figure **2c**. This block copolymer is synthesized via suspension block copolymerization, resulting in a linear structure with alternating VDF and TFE segments. The rigid TFE segments act as cross-links, connecting the elastic, rope-like VDF segments to form a spiderweb-like network. The embedded TFE chains introduce additional hydrogen bonding sites for PVDF, thereby enhancing molecular entanglement, mechanical strength, and adhesion [36]. This robust spiderweb-like structure effectively combines the flexibility of PVDF with the rigidity of PTFE, yielding a binder with a high elastic strain exceeding 250%. This characteristic allows the binder to provide efficient spatial constraints for the volume expansion and contraction of silicon anodes, consequently mitigating irrecoverable viscous deformation of PVDF.

PFPQ-COONa Binder

The novel binder, PFPQ-COONa, is synthesized by incorporating 10% p-quinone (PQ) into PF-COONa, a conductive binder known to enhance the cycling stability of silicon anodes. PFPQ-COONa's exceptional electrochemical performance stems from its well-designed molecular structure (Figure **2d**). Notably, the -COOSi- groups formed between PFPQ-COONa and silicon particles serve as anchoring points, facilitating a stable connection between the conductive network and silicon particles throughout lithiation and delithiation processes.

As a result, the average distance between Si particles and polymer chains is significantly reduced, promoting charge transfer at the interface. Additionally, the carbonyl groups in PQ can be reduced to -O-Li groups during lithiation, significantly enhancing the binder's conductivity at the reduced potential and improving rate performance. In addition to its conductivity, PFPQ-COONa also exhibits excellent mechanical properties [37].

2.2. Branched Binders

In contrast to linear polymer binders, branched polymer binders have a propensity to establish multidimensional contacts with silicon particles, thereby dispersing or mitigating the mechanical stress induced by volume changes across numerous branches or points [38-41]. Consequently, branched polymer binders are anticipated to demonstrate heightened adhesion strength and superior capacity to restrain the volume expansion of silicon-based anodes [42]. Moreover, branch copolymers featuring graft side chains and polymer main chains with distinct chemical properties can preserve the desired characteristics of each polymer component while adeptly managing chemical-physical interactions and mechanical properties [43].

CMC-10 % PDA binder

By introducing 10 wt% alkaline solution-prepared polydopamine (PDA) into carboxymethyl cellulose (CMC), the binder CMC-10% PDA is obtained through a combination of hydrogen bonding and some covalent interactions between CMC and PDA (as shown in Figure **2e**). Due to its strong binding performance, CMC-10% PDA exhibits higher viscosity and better mechanical properties compared to CMC alone.

The introduction of a small amount of PDA into CMC enhances the viscosity and mechanical properties of the binder through hydrogen bonding and covalent bonding between PDA and CMC. The interaction between the binder and the micrometer-sized silicon anode and current collector effectively maintains the stability of the electrode. Si@CMC-10% PDA electrodes can provide a high capacity of over 1700mAh g^{-1} at 0.2 C after 1000 cycles and exhibit a capacity retention rate of 80% after 50 cycles in full-cell tests [44]. The preparation process of this binder is simple, harmless, and environmentally friendly, making it highly promising for practical applications in micrometer-sized silicon anodes.

PAA-g-SBR Binder

The preparation process of this binder involves grafting tert-butyl acrylate onto SBR, followed by hydrolysis of the tert-butyl groups using aqueous H_3PO_4 at room temperature to obtain the graft copolymer PAA-g-SBR. After 20 cycles, the capacity of the Na-PAA/SBR system has decreased to 130 mAh g^{-1}, while the Na-PAA/PAA-g-SBR system still maintains a stable capacity retention of 673 mAh g^{-1} after 130 cycles [40]. Multiple comparative experiments have revealed that the PAA-g-SBR/Na-PAA binder system exhibits superior electrochemical and mechanical performance compared to the Na-PAA/SBR system.

The sole distinction between the two systems is the presence of modified SBR in Na-PAA/PAA-g-SBR, while Na-PAA/SBR consists only of standard SBR, highlighting the improvement achieved by modified SBR. The reason for this contrasting effect is likely the presence of functional groups in PAA-g-SBR that are similar to Na-PAA. This reduces the phase separation between the two polymer systems, forming a microstructure within the electrode coating that is better able to withstand the volume expansion during cycling, thereby enhancing the electrode's integrity.

Guar-Based Binder

The composite hydrogel binder, consisting of carbon black (CB) and guar gum, undergoes crosslinking with glutaraldehyde (GA) to bolster the electrode structure and enhance conductivity during lithiation, CB inclusion fortifies the gel network strength via physical chelation or chemical crosslinking between oxygen-containing groups on CB particles and hydroxyl groups of guar gum. Initially, CB provides physical chelation, with the system later transitioning to a combination of physical and chemical crosslinking, thereby enhancing the sample's crosslink density. Dufficy *et al*. monitored the crosslinking reaction using a low-amplitude oscillatory rheometer and observed expedited crosslinking reactions and reinforced networks when guar gum was crosslinked with CB, showcasing CB's capacity to interact with the crosslinked mixture's components and bolster network strength [45]. Employing a low crosslinking density binder (*e.g*., t_{rxn} = 1, 2 days), the SiNP electrode cycled 300 times at 1 C demonstrated a discharge specific capacity of 1200 mAh g^{-1} and a Coulombic efficiency of 99.9%. The high capacity retention of the electrode after hundreds of cycles may be attributed to good electrolyte absorption and lower high-frequency resistance compared to both non-crosslinked gel and highly crosslinked gel (t_{rxn} > 2 days). In contrast, the rigid gel structure formed by binders with high crosslinking density (t_{rxn} > 2 days) is difficult to deform for slurry casting, hindering interactions with SiNPs and electrolytes. Furthermore, the crosslinked guar gum binder exhibits some self-healing capability, likely attributable to hydrogen bonding, enhancing the electrode and preserving the microstructure.

PAL-Na PAA Binder

Luo *et al*. leveraged lignin's natural properties and its abundant random modifiable sites to graft sodium polyacrylate onto lignin fragments using free radical graft copolymerization and alkaline hydrolysis techniques The resulting copolymer derived from lignin demonstrates water solubility and a nonlinear molecular structure, leveraging lignin's unique characteristics.

The improved electrochemical performance is likely due to its nonlinear molecular structure. During the lithiation/delithiation process, the silicon anode pulverizes into smaller units. The nonlinear nature of the binder molecules, with octopus-like tentacle-like chains, may provide more opportunities to grasp the pulverized units, allowing for multiple bridging on the cracked surfaces of Si [46]. This ensures continuous and tight contact across most parts of the electrode, contributing to the enhanced performance.

2.3. 3D Network Binders

In the realm of silicon-based anodes for lithium-ion batteries, the mechanical attributes of binders play a crucial role in upholding electrode structural integrity and amplifying electrochemical efficacy [47-49]. 3D network binders, distinguished by their intricate cross-linked configuration, surpass linear and branched binders in their ability to endure the substantial volume fluctuations of silicon throughout lithiation and delithiation processes, thus mitigating the likelihood of electrode fracture.

Three-dimensional network binders, formed by polymer skeletons and covalent or non-covalent crosslinking, demonstrate outstanding mechanical properties. Liu *et al*. amalgamated hard and soft polymer systems, integrating the in-situ polymerization of hard polyferrocenyl alcohol (PFA) as the skeleton and soft polyvinyl alcohol (PVA) as the filler. This fusion resulted in a robust binder with a 3D-interpenetrating bonding network, achieving an energy density exceeding 300 Wh kg^{-1} in lithium-ion batteries (LIBs) [50]. The binder is synthesized via in-situ copolymerization of hydroxyethylacrylamide (HA), sodium acrylate (SA), and the cross-linking agent 3-acrylamide phenylboronic acid (BA) utilizing reversible

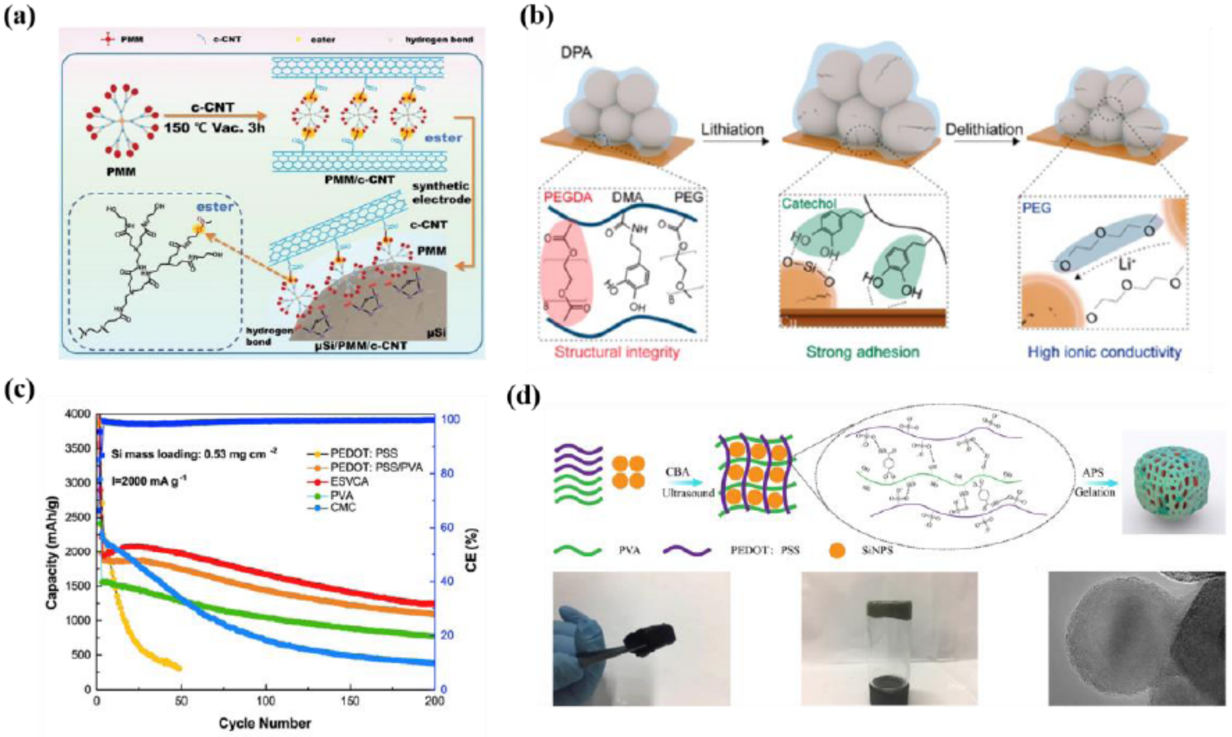

Figure 3: **a**) Schematic illustration of the interactions between PMM binders and c-CNT/Si microparticles. **b**) DPA Binder with a Linear Backbone (blue colored lines) with three functional PEGDA, DMA, and PEG Unitsa. **c**) Electrochemical performance of different electrodes. **d**) Schematic illustration of the fabrication process for Si electrode with self-healing binder.

addition-fragmentation chain transfer (RAFT) polymerization. With a binder concentration of only 3 wt%, the silicon/C450 anode-based half-cell retains 83.56% of itscapacity after 600 cycles [51]. Blending low-cost PAA and ethylene-vinyl acetate (EVA) emulsions (PAA/EVA) combines the strong binding strength of PAA, characterized by a high number of carboxyl groups, with the high elasticity of EVA, enhancing ductility and preserving the integrity of the electrode throughout charge and discharge cycles [52]. Kang *et al*. explored four binders, polyacrylamide (PAM), polytetrafluoroethylene (PTFE), carboxymethyl cellulose, and styrene-butadiene rubber (SBR)-to formulate a water-based multi-component binder, PSP. This binder exhibited favorable chemical stability, robust bonding strength, and rigidity, thus maintaining the structural integrity of the electrode. It achieved an excellent capacity retention of 770 mAh g^{-1} at 500 mA g^{-1} and a rated capacity of 993 mAh g^{-1} at 1200 mA g^{-1} after 300 cycles [53].

Due to the inherent low electrical conductivity of silicon materials, they are typically combined with carbon materials to create Si/C composite materials, or conductive additives are introduced to enhance conductivity. Compared to traditional binders, 3D network conductive polymer binders offer superior adhesion and conductivity. They feature a highly cross-linked three-dimensional network structure that facilitates efficient electronic transmission channels [13].

This class of conductive monomers, including 3,4-ethylenedioxythiophene (EDOT), pyrrole, aniline, and thiophene, can be utilized to construct three-dimensional networks with conductive binder properties through crosslinking via composite formation, doping, or other methodologies. Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) is a widely employed example of such composite materials. PEDOT:PSS composites are typically fabricated through processes like mixing and crosslinking, resulting in a three-dimensional network binder that exhibits both excellent conductivity and flexibility [54]. Su *et al*. investigated a continuous conductive three-dimensional network binder, denoted as PPTU, comprised of PEDOT:PSS (a conductive polymer) and PETU (a stretchable polyether thiourea). This composite binder utilizes interlocking hydrogen bonds to conform to the surface of nano silicon particles, thereby enhancing both electron transfer and mechanical stability. Even after 300 charging cycles, the electrode material maintained a high specific capacity of 2081 mAh g^{-1} and a rate capability of 908 mAh g^{-1} at a current density of 8 A g^{-1} [55].

Binders can be designed to create highly conductive three-dimensional network structures through various methods, including the introduction of carbon nanotubes or ion-replacement facilitated by water-soluble additives like polyacrylic acid (PAA). Liu *et al*. (Figure **3a**) designed a composite binder network composed of PMMA/c-CNT, leveraging the properties of hyperbranched polymethylacrylamide (PMM) as the base polymer. The abundant hydroxyl (-OH) groups, three-dimensional branched structure, and inherent porosity of PMM's dendritic structure effectively stabilized silicon microparticles within lithium-ion batteries. This design yielded a high initial specific capacity of up to 3590 mAh g^{-1} and a high initial coulombic efficiency (ICE) of 91.12%, along with remarkable long-term cycling stability (69.8%) [56].

As shown in Figure **3b**, Ko *et al*. described a multifunctional polymer binder featuring a three-dimensional network structure. This binder is constructed from a linear polymer backbone functionalized with three key units: dopamine methacrylamide (DMA), poly(ethylene glycol) monomethyl ether acrylate (PEG), and poly(ethylene glycol) diacrylate (PEGDA). PEGDA units establish crosslinks between linear polymer backbones, thereby ensuring structural integrity. The DMA unit enhances adhesion to active materials, conductive agents, and current collectors. Finally, the PEG unit facilitates Li^+ ion conductivity. This composite binder significantly improves the cycling stability and rate capability of silicon-based lithium-ion batteries [57].

Non-covalent interactions, particularly hydrogen bonding and π-π stacking, play a crucial role in the self-healing action of binders on the Si anode. These interactions enable the reformation of broken bonds and the re-alignment of molecular structures, promoting the repair of cracks and maintaining electrode integrity [58]. Dong *et al*. (Figure **3c**, Figure **3d**) fabricated a hydrogel binder via a combination of chemical crosslinking, doping, and electrostatic self-assembly of PEDOT:PSS and PVA polymers. This composite binder exhibits excellent tensile strength, high electrical conductivity, and remarkable self-healing capability. Notably, the electrode incorporating this binder delivered a high capacity of 1786 mAh g^{-1} at 500 mA g^{-1} and maintained 71.3% capacity retention after 200 cycles [59]. Branched tannic acid (TA) serves as a physical crosslinker by forming abundant dynamic hydrogen bonds with the polyacrylic acid (PAA) backbone during the synthesis of the TA-c-PAA binder. This crosslinking process imbues the TA-c-PAA binder with unique self-healing properties and robust adhesion to the silicon (Si) anode. Consequently, the Si@TA-c-PAA electrode demonstrates exceptional performance, including a high reversible specific capacity (3250 mAh g^{-1} at 0.05 C), excellent rate capability (1599 mAh g^{-1} at 2 C), and oued of polyvinyl alcohol (PVA) and a lithium borate ($LiBO_2$) solution (denoted as PVA+LB), it exhibits the atstanding cycling stability (1742 mAh g^{-1} at 0.25 C after 450 cycles) [60]. This work describes the construction of a supramolecular self-healing three-dimensional network via interactions between the amino groups of glycol chitosan (GCS) and the dialdehyde groups of oxidized alginate (OSA). The dialdehyde groups in OSA act as crosslinkers, spontaneously reacting with the amino groups in GCS to form dynamic imine bonds. This reaction scheme facilitates the formation of supramolecular networks within the GCS-I-OSA polymer binder. These dynamic covalent bonds enable the GCS-I-OSA binder to adhere strongly to the silicon anode [61].

2.4. Stress Dissipative Binders

The large volume change experienced by silicon (Si) particles during lithiation and delithiation processes significantly compromises the cycling stability of Si-based electrodes. Therefore, a critical function of the binder is to accommodate this volume change. Polymer binders can mitigate the detrimental effects of mechanical stresses caused by Si expansion/contraction. These binders achieve this by effectively dissipating stress, such as through elastic deformation or void filling, thereby maintaining the structural integrity of the Si particles and the uniformity of the electrode/electrolyte interface [62-65].

Figure **4b** illustrates the in-situ polymerization process that generates a three-dimensional (3D) interpenetrating network (IPN) binder, denoted as PFA-TPU. This IPN comprises a rigid poly(furfuryl alcohol) (PFA) framework and a soft, cushioning thermoplastic polyurethane (TPU) layer. The components are interwoven at the nanoscale, facilitating the formation of hydrogen bonds at their interfaces. This synergistic design enables the PFA-TPU binder to effectively dissipate the stress induced by the volume change of silicon particles during lithiation/delithiation [66]. Another common design strategy utilizes a rigid skeleton combined with flexible groups, such as hydroxyl and carboxyl groups, to effectively buffer stress within the electrode. Citric acid (CA), a water-soluble molecule, is incorporated with PAA to form the binder. These CA molecules serve a dual purpose: acting as a protective buffer layer that mitigates internal stress and stabilizes the solid electrolyte interface

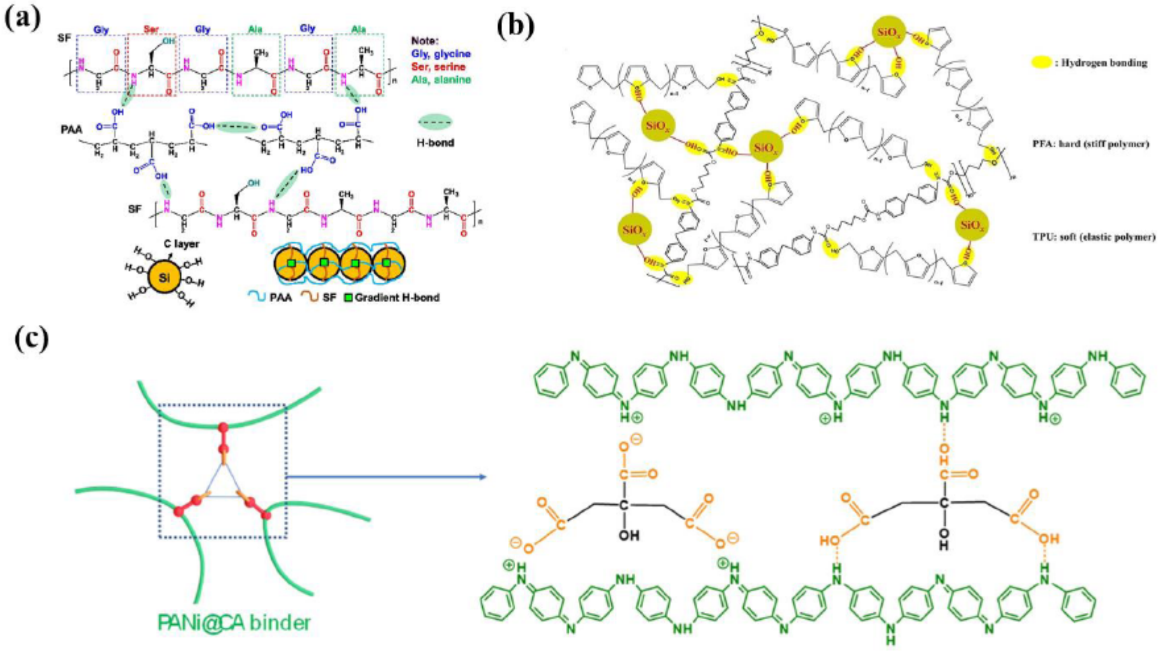

Figure 4: a) Schematic illustration of chemical interactions of poly(acrylic acid) (PAA) and silk fibroin (SF) binder and PAA/SF–silicon/carbon (Si/C) composite. **b**) Schematic Illustration of the Three-in-One Design Strategy for Binder in SiOx Anode. **c**) Chemical structures of 3D networks structred PANi@CA conductive polymer binder.

(SEI), and also establishing connections with PAA to create an "inner soft and outer hard" spatial architecture. This design promotes rapid self-healing, maintains structural integrity of the electrode, and delivers impressive performance, retaining 78% of its capacity after 50 cycles and achieving an initial Coulombic efficiency of 89.5% [67].

Reversible dynamic cross-linking networks, such as hydrogen bonds, borate ester bonds, disulfide bonds, or Diels-Alder bonds, can be employed to achieve energy dissipation in binders. These bonds break and reform under mechanical stress, enabling the binder to dissipate energy. For instance, Hu *et al*. designed a gradient hydrogen-bonded polymer binder (PAHT) by incorporating tannic acid (TA) into a poly(acrylic acid-co-carbon dioxide-co-hydroxyethyl acrylate) (PAH) copolymer. The gradient design distributes hydrogen bonds with various strengths throughout the binder network. Upon silicon particle expansion, these hydrogen bonds dissociate sequentially, starting from the weakest, in a process that effectively dissipates energy and stress. This mechanism contributes to the high capacity retention of 80.2% observed for electrodes using PAHT after 700 cycles [68].

As shown in Figure **4c**, Zhao *et al*. synthesized a conductive stress-dissipative binder using polyaniline (PANI) and citric acid (CA) via hydrogen bonding. This binder features a stable triangular network structure with abundant hydroxyl groups. Citric acid acts as nodes within the network, effectively distributing the stress generated by silicon nanoparticles (SiNPs) due to the hydrogen bonding interactions between PANI and CA. This design leads to superior mechanical-chemical properties at the interface between the silicon anode and the binder. The electrode using this binder exhibits high reversibility, with a capacity of 1021 mAh g^{-1} remaining after 500 cycles at a current density of 0.5 C (where 1 C = 4200 mAh g^{-1}) [69]. Lee *et al*. reported a biomimetic "adaptive binder" inspired by self-healing plants. This binder consists of phenolic (1,2,3-trihydroxybenzene) connected to hyaluronic acid (HA-GA). In the early stages of cycling, the chemical interactions between the polymer binders are reversible hydrogen bonds. During later cycling stages, these interactions transform into stronger, irreversible covalent bonds between the binder polymers. This transition enhances the stability of the binder over time [70]. Guar gum-citric acid (GCA) binder incorporates citric acid molecules with short-range chemical functionalities onto long-chain guar gum through weak interactions. This design creates a robust binder network with additional long-range interactions. The long-range interactions facilitate the formation of high-speed lithium-ion transport channels, while the short-range interactions enhance the network's flexibility for energy dissipation. This flexibility allows the binder to adapt to the significant volume changes experienced by silicon particles during cycling. Stress is released through mechanisms such as particle rearrangement or SEI deformation [71].

The research by Zhao *et al*. (Figure **4a**) describes a biopolymer composite binder made of rigid polyacrylic acid (PAA) and flexible silk fibroin (SF). Due to strong hierarchical bonding between the carboxyl groups of PAA and the amide groups of SF, the PAA/SF binder effectively accommodates the large volume change of silicon particles, enabling the electrode to cycle for 500 times at 0.5 C [72]. Cai *et al*. synthesized fluorine-containing polymers via emulsion polymerization using acrylic acid (AA), 2-perfluorobutyl ethyl acrylate (C_4A), and acrylamide (AM). The introduction of a self-healing agent (SA) followed by heat treatment promoted an esterification reaction, creating a dual crosslinking network. This network combines covalent bonds, which form the polymer skeleton and maintain structural integrity, with hydrogen bonds. These dynamic hydrogen bonds act as sacrificial bonds, breaking and reforming during volume changes to dissipate energy [73]. Weng *et al*. employed a free radical polymerization method to synthesize P(AM-co-AA), a random copolymer featuring inherent hydrogen bonding networks. This design offers advantages over simple homopolymers by incorporating a diversity of functional groups. The copolymer adheres to the SiO_x microparticle anode via covalent bonds formed between the carboxyl groups of AA and the surface hydroxyl groups of SiO_x. Additionally, the amide groups of AM establish hydrogen bonds with the SiOx hydroxyl groups. This synergistic interaction, combining covalent and non-covalent interactions, creates robust buffer networks that safeguard the SiO_x microparticles during the lithiation/delithiation process [74]. Xie *et al*. developed a water-soluble polymer binder, PAA-B-HPR, by cross-linking poly(acrylic acid) (PAA) with hydroxyl-terminated poly(urethane) (HPR) using reversible borate ester bonds. These α-cyclodextrin molecules can slide freely within the polymer network during lithiation and delithiation, effectively distributing and balancing the accumulating internal stress throughout the electrode. Furthermore, the reversible borate ester bonds function as self-healing mechanisms, repairing damage generated during fabrication and cycling to maintain electrode integrity. The Si@PAA-B-HPR anode demonstrates impressive performance, delivering a discharge specific capacity of 1056 mAh g^{-1} at 1.4 A g^{-1} after 500 cycles at an elevated temperature of 55℃. The corresponding capacity decay rate is remarkably low, at only 0.10% per cycle [75].

3. CONCLUSIONS AND PERSPECTIVE

The commercialization of silicon-based anodes in lithium-ion batteries is hindered by challenges including significant volume expansion, limited cycle life, poor conductivity, and compatibility with current infrastructure. Addressing these challenges requires ongoing research and collaboration to unlock their potential. Despite the low content of binders in electrodes, they play a crucial role in the fabrication of silicon-based anodes. They not only affect the mechanical stability of the electrodes but also directly influence the conductivity and cyclic lifespan of the batteries. By optimizing the design of binders, the issue of silicon particle volume expansion during charging and discharging can be effectively mitigated, enhancing the cyclic stability and electrochemical performance of silicon anodes. Researchers have developed novel binders to meet the needs of high-performance lithium-ion batteries. Various design strategies for multifunctional binders, including linear, branched, and three-dimensional network binders, as well as those with self-healing, conductive, and stress-dissipative properties, are particularly significant. Such strategies encompass introducing specific functional groups, achieving dynamic crosslinking, and incorporating conductive conjugated structures, among others. These novel binders, by constructing three-dimensional network structures, introducing multifunctional polymers, and designing dynamically crosslinked networks, significantly improve the performance of silicon anodes. Future research should continue to focus on the multifunctionality of binders, including their mechanical properties, conductivity, self-healing capabilities, and stress-dissipation abilities. Additionally, environmental friendliness and sustainability should also be considered, exploring bio-based and renewable resources as raw materials for binders. Furthermore, the design of binders should be integrated with electrode materials, electrolytes, and overall battery design to achieve optimal battery performance. Although significant progress has been made in the study of binders for silicon-based anodes, issues related to cost, scalability, and compatibility with existing battery manufacturing processes still need to be addressed. Future efforts should be devoted to developing both cost-effective and high-performance binders to further advance the development of silicon-based lithium-ion battery technology.

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