

# A Comprehensive Review of Current and Emerging Binder Technologies for Energy Storage Applications

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## Abstract

Binders hold a crucial role in electrode fabrication by ensuring the cohesion and stability of active materials, conductive additives, and electrolytes within batteries. They also establish essential electron and ion pathways, crucial for effective lithiation and delithiation processes. Despite their relatively low concentration compared to active materials, binders significantly impact both the physical properties and electrochemical performance of electrodes. Given the escalating demand for Electric Vehicles and energy storage systems, there's an increasing need for batteries with higher energy densities and cost-effective production. This necessitates the development of new and more efficient battery materials. This review provides a detailed examination of various binders used in battery manufacturing, starting from traditional binders for Lithium-ion batteries to recent advancements for sodium-ion batteries, silicon anodes, Lithium-Oxygen batteries and other emerging technologies. By systematically exploring different binder types and their properties, this review contributes to the optimization and advancement of battery technologies. As the energy storage landscape continues to evolve, the insights presented here aim to inform innovative developments in binder design and application, ultimately catalyzing advancements in the field.

## Introduction

The development of efficient and high-performance batteries is critical for advancing numerous industries, including portable electronics, electric vehicles, and renewable energy storage systems [1-3]. Among the various components that comprise a battery, the role of binders in electrode fabrication is paramount. Binders serve as essential materials for maintaining the structural integrity, conductivity, and stability of active materials, conductive additives, and electrolytes within the electrodes. Along with holding the active material and conductive agent together, the binder also plays a crucial role in adhering the active material and conductive agent to the metal current collector [4]. Consequently, the choice of binder significantly impacts the overall performance and lifespan of batteries.

The importance of binders in battery manufacturing has been widely recognized and extensively studied by researchers and engineers in recent years. Several studies have focused on exploring the properties and performance of different types of binders, ranging from polymeric to inorganic and hybrid materials, with the aim of optimizing electrode fabrication processes and enhancing battery performance. The binders can also be classified on the basis of its solubility namely organic solvent soluble binders and water soluble binders.

One of the commonly used Polymer binder is the organic solvent soluble Polyvinylidene Fluoride (PVDF). PVDF has gained significant attention due to its excellent adhesive properties, chemical stability, and thermal resistance. In a study by Rajeevan et al. [5], PVDF-based binders were investigated for their impact on the electrochemical performance and mechanical strength of lithium-ion batteries. The results demonstrated that PVDF binders exhibited improved adhesion and superior mechanical properties, leading to enhanced cycling stability and rate capability of the batteries.

A notable inorganic water soluble binder is Sodium Carboxymethyl Cellulose (CMC-Na), which has shown promise in battery applications. CMC-Na offers good film-forming properties, high viscosity, and excellent water retention capacity. Chang et al. [6] conducted a study to evaluate the effect of CMC-Na binders on the electrochemical performance of Zn/MnO<sub>2</sub> aqueous batteries. The results revealed that CMC-Na binders lead to improved cycle stability and capacity retention when used in mild aqueous Zn-MnO<sub>2</sub> systems. Furthermore, the study reported that Sodium Carboxymethyl Cellulose (CMC) binders provide favorable adhesion, effective wettability, uniform material dispersion, and robust chemical stability within specific pH ranges (3.5–5), enduring extended cycles without undergoing decomposition.[6]

Furthermore, there are hybrid binders, which combine the advantages of both polymeric and inorganic materials. Recently, hybrid binders have gained attention for their synergistic effects in battery manufacturing. For example, most of the polymeric binders show good adhesion and electrochemical stability. But they lack thermal and mechanical stability during high temperature and high energy density operations. So combining these polymeric binders with inorganic binders which are popular for their exceptional thermal and mechanical stability, results in a hybrid binder with good adhesive property as well as thermal and mechanical stability. Research conducted by Mao et al. [7] investigated the application of a polymeric-inorganic hybrid binder based on graphene oxide (GO) and sodium alginate in sodium-ion batteries. The hybrid binder exhibited enhanced adhesion, mechanical strength, and electrochemical performance, demonstrating its potential for achieving high-performance electrodes.

The escalating demand for binders has been propelled by their pivotal role in advancing the performance of lithium-ion batteries (LIBs). However, despite the substantial progress achieved through binder enhancements, formidable challenges loom on the horizon for their future development. These challenges stem from the relentless pursuit of higher energy density, which has catalyzed the exploration of alternative energy storage technologies such as lithium-sulfur (Li-S) batteries, silicon-based batteries, lithium-air batteries and sodium-ion (Na-ion) batteries. While these emerging technologies hold great promise, they also bring along inherent drawbacks that cannot be disregarded. In the context of sodium-ion batteries, the larger ion radius of Na<sup>+</sup> poses intricate challenges in achieving effective insertion into the electrode structure, often resulting in structural damage. For these systems, an optimal binder must exhibit robust mechanical strength to counteract electrode cracking. In the case of Li-S batteries, the polysulfide shuttle effect compounds the structural degradation challenge, necessitating a binder that not only preserves electrode integrity but also efficiently sequesters polysulfide species to minimize active material loss. The

intricacies extend further to Li-O<sub>2</sub> batteries, where the aggressive nature of superoxide species wreaks havoc on conventional binders, leading to electrode fracture and compromised battery performance. Here, the imperative lies in the development of chemically stable binders capable of withstanding the corrosive oxidizing reactions and bolstering battery resilience. [8-11]

In light of these complexities, it is evident that the requisites for binders extend beyond their fundamental adhesion function. Binders must possess supplementary attributes to meet the distinctive demands of each advanced energy storage system and facilitate their viable integration into large-scale industrial applications, particularly in the context of vehicle electrification

## Selection criteria for binders

With the increasing demand for energy storage technologies, the selection and optimization of binders in battery manufacturing have become critical research areas. The selection of binders plays a crucial role in battery manufacturing, as they are responsible for ensuring the cohesion, stability, and overall performance of electrodes. The optimization of binder materials is essential to enhance the electrochemical properties, mechanical strength, and durability of batteries. This paper tries to provide a comprehensive analysis of the main selection criterias for binders, encompassing electrochemical performance, mechanical properties, thermal stability, cost-effectiveness, and environmental impact. By considering these criteria, researchers and manufacturers can make informed decisions regarding binder selection, leading to the development of high-performance and sustainable battery systems. There are a lot of binders available but among them only a few qualify to be used for battery fabrication. For considering a binder to be qualified for electrode fabrication, it has to satisfy the following requirements:

Selection criterias	Description
Electrochemical Performance	The electrochemical performance of a binder is critical for the efficient operation of batteries. It involves several factors such as binding capability, ion conduction, and suppression of side reactions. Binders should have good adhesion to active materials and conductive additives, ensuring effective electron and ion transfer. Additionally, the binder should minimize parasitic reactions and enhance the cycling stability and capacity retention of the battery and should not hinders the electrolyte wettability of the electrodes [5]
Mechanical Properties	Mechanical strength and flexibility are important criteria for binder selection. The binder must maintain the structural integrity of the electrode during manufacturing, assembly, and cycling processes. It should exhibit sufficient adhesion to prevent material detachment or delamination, ensuring long-term electrode stability and preventing performance degradation. The binder should be able to repress electrode cracks if any is generated. At the same time, the binder should possess flexibility to accommodate volume

	changes during charge and discharge cycles without cracking or losing contact [5][12].
Thermal Stability	Thermal stability is a critical factor for binder selection, as batteries often operate under high temperatures. Binders should exhibit good thermal stability to prevent decomposition, volatilization, or melting during battery operation. This ensures the structural integrity and performance of the electrodes, preventing premature failure or degradation of the battery system [12]. Furthermore, binder should be thermally stable during the electrode drying process (100 - 200 degree celsius)
Cost-effectiveness and Availability	The cost-effectiveness and availability of binders are crucial considerations for large-scale battery manufacturing. Binders should be economically viable and readily available in sufficient quantities. High material costs or limited availability can significantly impact the commercialization and scalability of battery technologies. Hence, binders with reasonable costs and wide availability are preferred choices for practical applications.
Environmental Impact	In recent years, the environmental impact of materials used in battery manufacturing has gained significant attention. Binders should be environmentally friendly, with minimal toxicity and low carbon footprint. This includes considering the sourcing of raw materials, production processes, and end-of-life considerations. Choosing binders with low environmental impact aligns with the growing emphasis on sustainable and green battery technologies [12].

Table 1: Key selection criterias for binders

The selection of binders in battery manufacturing requires careful consideration of multiple criteria. Electrochemical performance, mechanical properties, thermal stability, cost-effectiveness, and environmental impact are key factors that influence binder selection. By analyzing these criteria, researchers and manufacturers can choose suitable binders that enhance battery performance, durability, and sustainability. Future advancements in binder materials and technologies should aim to meet these selection criteria, contributing to the development of efficient and eco-friendly energy storage systems.

## Types of Binders

### 2.1. Polymeric Binders

#### Polyvinylidene Fluoride (PVDF)

Polyvinylidene fluoride (PVDF) is a widely used organic soluble binder material in the manufacturing of batteries due to its exceptional properties such as high chemical resistance, mechanical strength, and

thermal stability [5]. PVDF was first invented in the 1950s by researchers at Pennsalt Chemicals Corporation (now Arkema Inc.). Since then, it has found extensive application in various industries, including the battery industry. PVDF is primarily synthesized through the polymerization of vinylidene fluoride monomers. This polymerization process typically involves the use of initiators and catalysts under specific temperature and pressure conditions. The resulting PVDF polymer exhibits a unique combination of properties, making it suitable for battery binder applications. However, the major production method adopted for the synthesis PVDF in mass production scale is emulsion polymerization and suspension polymerization [13]. The choice of PVDF as a binder is influenced by its excellent compatibility with both aqueous and non-aqueous electrolytes, making it suitable for various battery chemistries. Commercially available grades of PVDF for battery applications include homopolymers and copolymers. Homopolymer PVDF is the most commonly used grade, known for its high crystallinity and excellent mechanical properties. Copolymers of PVDF, such as PVDF-HFP (hexafluoropropylene), offer enhanced flexibility and improved binder performance at low temperatures. The selection of the appropriate grade of PVDF for battery applications depends on several factors, including the specific battery chemistry, electrode composition, and desired performance characteristics. It is crucial to choose the grade that best suits the requirements of the battery system to ensure optimal binder performance and long-term stability [12].

The use of PVDF as a binder is mainly attributed to its mechanical strength which helps maintain the structural integrity of the electrode during charge and discharge cycles. It provides strong adhesion between active materials and conductive additives, reducing electrode delamination and improving cycling stability. Alongside, the high thermal stability of PVDF allows it to withstand the elevated temperatures encountered during battery operation. This stability prevents binder decomposition or degradation, ensuring the structural integrity of the electrode even under harsh conditions. PVDF is chemically inert and exhibits excellent resistance to electrolytes and other battery components. This resistance minimizes side reactions at the electrode-electrolyte interface, leading to improved cycle life and capacity retention. PVDF is compatible with various electrode materials, including lithium-ion battery cathodes (e.g., LiCoO<sub>2</sub>, LiFePO<sub>4</sub>) and anodes (e.g., graphite, silicon). This compatibility allows for versatile applications of PVDF as a binder in different battery chemistries. [14]. Additionally, PVDF has good electrolyte wettability, facilitating efficient ion transport within the electrode. The presence of PVDF enhances the electrolyte penetration into the electrode structure, improving electrode performance and reducing interfacial resistance.[5][12,14]

However, the use of PVDF as a binder also has some demerits. PVDF has low inherent ionic conductivity, which can limit ion transport within the electrode. This can result in increased polarization and hinder the rate capability of the battery. To overcome this limitation, PVDF is often combined with conductive additives, such as carbon black or graphene, to enhance the overall conductivity. Also the PVDF may not be compatible with certain electrolyte formulations, especially those containing high concentrations of polar solvents or additives. The interaction between PVDF and certain electrolyte components can lead to binder swelling, electrode delamination, or reduced cycling stability. Careful selection of the electrolyte system is necessary to ensure compatibility [14]. PVDF requires a solvent, such as N-methyl-2-pyrrolidone (NMP) or dimethylformamide (DMF), for dissolution and electrode slurry preparation. These solvents can be hazardous and require proper handling and disposal measures. Additionally, the processing of PVDF-based electrodes often involves a drying step to remove the solvent, which adds complexity to the manufacturing process [15]. Even though PVDF was a successful binder for conventional Lithium ion batteries. Making PVDF as one of the most commonly used traditional binder for wet coated cathodes.

But considering the recent advancements in the battery industry, the use of PVDF as a potential binder results in more significant demerits. PVDF-based electrodes may experience binder swelling and volume change during cycling, particularly in lithium-sulfur (Li-S) batteries or high-capacity electrode systems. The volume change can cause mechanical stress and lead to electrode disintegration or loss of electrical contact, affecting the long-term cycling stability. The use of PVDF as a binder is limited to systems with low voltage operating windows, with minor volume expansion and low energy density. The bonding mechanism of PVDF binder is mainly facilitated by the weak C-F bond interaction, which limits the maximum volume of electrodes. Hence, the use of PVDF is limited to low energy density batteries. PVDF also reacts with the Lithium ions and generates heat during charging at low temperatures or during overcharging that indeed leads to the degradation of binder and results in peeling off of active materials. PVDF also reacts with the lithium ions and results in the loss of overall cell capacity [5] [12-14].

### Styrene Butadiene Rubber (SBR)

Styrene-butadiene rubber (SBR) is a commonly used water soluble binder material in battery manufacturing, known for its excellent adhesion properties and mechanical strength. SBR was first invented in the early 1930s and its development can be attributed to a team of researchers at the German company IG Farben, led by Walter Bock and Eduard Tschunkur. Since then, SBR has found extensive application in various industries, including the battery industry. SBR is typically synthesized through the copolymerization of styrene and butadiene monomers. The polymerization process involves the use of initiators and catalysts under controlled temperature and pressure conditions. The resulting SBR polymer exhibits a unique combination of properties, making it suitable for battery binder applications. Commercially available grades of SBR for battery applications include high styrene SBR and low styrene SBR. High styrene SBR exhibits enhanced mechanical strength and adhesion properties, making it suitable for high-performance batteries. Low styrene SBR offers improved flexibility and processing characteristics, making it suitable for applications where flexibility is desired.

In the battery industry, SBR is widely used as a binder to hold the active materials, such as electrode particles, together in the electrode matrix. It provides mechanical stability and improves adhesion between the active materials and the current collector, ensuring the integrity of the electrode structure. This promotes strong interfacial contact, enabling efficient electron and ion transfer within the electrode. The enhanced adhesion contributes to improved electrochemical performance and cycling stability [16]. SBR possesses good mechanical properties, including tensile strength and flexibility. It helps maintain the structural integrity of the electrode during manufacturing, assembly, and cycling processes. The binder's mechanical strength prevents material detachment or delamination, ensuring prolonged electrode stability and preventing performance degradation [12]. The choice of SBR as a binder is influenced by its excellent adhesion capabilities and its compatibility with different electrode materials and electrolytes. SBR exhibits good compatibility with a wide range of electrode materials, including graphite, metal oxides, and sulfur-based compounds. This compatibility allows for its usage in diverse battery chemistries, making SBR a versatile binder option. In addition, SBR is a cost-effective binder material, making it suitable for large-scale battery production. Its availability and relatively low cost contribute to the economic feasibility of battery manufacturing, especially for applications with stringent cost requirements [12][16-18].

Even though there are a lot of advantages in considering Styrene-Butadiene Rubber as the binder in battery manufacturing, still there exists some limitations. At high temperatures, SBR may undergo decomposition or degradation, which can negatively impact the structural integrity and performance of the

electrode. Therefore, SBR is more suitable for batteries operating under moderate temperature conditions [17]. SBR has limited ionic conductivity compared to other binder materials. This can result in increased polarization and hinder efficient ion transport within the electrode, leading to reduced rate capability and overall battery performance. Furthermore, the interaction between SBR and other battery components, such as current collectors or electrolytes, may pose compatibility challenges. It is essential to optimize the binder-electrode interfaces to ensure good compatibility and minimize any adverse effects on battery performance. While SBR is widely used, its environmental impact should be considered. SBR is a synthetic rubber derived from petroleum-based sources, and its production and disposal can have environmental implications. Efforts should be made to explore more sustainable and eco-friendly alternatives [16-18].

#### Polyacrylic Acid (PAA)

Polyacrylic Acid (PAA) is a versatile binder material used in the manufacturing of batteries, known for its excellent adhesion properties and ability to improve electrode stability. PAA was first synthesized in the early 1960s. PAA is typically synthesized through the polymerization of acrylic acid monomers. The polymerization process involves the use of initiators and catalysts under controlled conditions. The resulting PAA polymer exhibits a unique combination of properties, making it suitable for battery binder applications. Commercially available grades of PAA for battery applications include low molecular weight PAA and high molecular weight PAA. Low molecular weight PAA offers improved wetting and penetration capabilities, facilitating better electrolyte infiltration into the electrode structure. High molecular weight PAA provides enhanced adhesion strength and mechanical stability to the electrode materials.

PAA exhibits excellent adhesion characteristics, promoting strong binding between active materials, conductive additives, and electrode components. This enhances interfacial contact and facilitates efficient electron and ion transfer within the electrode, leading to improved electrochemical performance and cycling stability [19,20]. PAA also possesses appreciable inherent ionic conductivity, which can contribute to improved ion transport within the electrode. This enhanced ionic conductivity enables efficient charge and discharge processes, reducing polarization effects and improving the rate capability of the battery [20]. PAA is water-soluble, allowing for easy processing and electrode fabrication. It can be conveniently mixed with active materials and conductive additives in aqueous solutions, simplifying the manufacturing process and reducing environmental hazards associated with organic solvents. PAA can act as a pH regulator due to its carboxylic acid functional groups. It can help maintain the desired pH level within the battery system, preventing side reactions or undesired electrolyte decomposition. This pH regulation contributes to improved battery stability and longevity. Furthermore, PAA exhibits good compatibility with a wide range of electrode materials, including graphite, metal oxides, and sulfur-based compounds. This compatibility allows for its usage in various battery chemistries, making PAA a versatile binder option [19-21].

PAA may have limitations in terms of mechanical strength compared to other advanced binder materials. Its relatively low elasticity and flexibility may lead to issues such as material detachment or loss of contact between active materials, particularly during volume changes associated with charge and discharge cycles. PAA may exhibit lower thermal stability compared to other binder materials. At elevated temperatures, PAA can undergo decomposition or degradation, potentially compromising the structural integrity and performance of the electrode. Therefore, careful consideration of operating temperature ranges is necessary when using PAA as a binder. While PAA is water-soluble and does not pose significant environmental concerns during manufacturing, its long-term environmental impact should be considered. Further research is needed to evaluate the ecological consequences of PAA-containing battery systems

and explore more sustainable alternatives. PAA may exhibit compatibility challenges with certain electrolytes or additives commonly used in battery systems. The binder-electrolyte interface must be carefully designed to prevent adverse reactions, such as electrolyte decomposition or formation of solid-electrolyte interfaces [12][19-21].

#### Carboxymethyl cellulose(CMC)

Carboxymethyl cellulose (CMC) was first developed in the early 1900s and its invention is attributed to chemists Arthur Aikin and Anselme Payen. Since then, CMC has found extensive application in various industries, including the battery industry. CMC is derived from cellulose, a natural polymer found in plants. The synthesis of CMC involves the chemical modification of cellulose through the reaction with sodium hydroxide and monochloroacetic acid. This process introduces carboxymethyl groups onto the cellulose backbone, resulting in the formation of CMC. The degree of substitution (DS) of carboxymethyl groups can be controlled during synthesis to tailor the properties of CMC for different applications. Commercially available grades of CMC for battery applications include different viscosity grades and degrees of substitution. The viscosity grade determines the flow characteristics of CMC, with high viscosity grades offering better binding properties. The degree of substitution influences the solubility and swelling behavior of CMC, affecting its binding performance and stability in the battery system.

CMC exhibits good adhesion properties, allowing it to form a strong bond between active materials and electrode components. This promotes excellent interfacial contact, enhancing electron and ion transport within the electrode and improving overall electrochemical performance [22-23]. Alongside, CMC possesses excellent film-forming ability which enables the formation of a uniform and continuous binder layer on the electrode surface. This layer acts as a protective barrier, preventing the loss of active materials and maintaining electrode stability during cycling. CMC-based electrodes often exhibit increased porosity, allowing for better electrolyte penetration and ion transport. The enhanced electrode porosity can improve the rate capability and capacity retention of the battery. CMC is water-soluble and can be easily incorporated into water-based electrode formulations. This feature simplifies the manufacturing process, reduces environmental concerns associated with organic solvents, and enables homogeneous distribution of active materials. CMC demonstrates good compatibility with various electrolyte formulations, both aqueous and non-aqueous. This compatibility minimizes adverse reactions at the binder-electrolyte interface, leading to improved cycling stability and reduced capacity fade [24-30].

However, CMC has relatively low mechanical strength compared to other binder materials. Under high-stress conditions, such as rapid charging and discharging rates or large volume changes, CMC-based electrodes may experience mechanical failure, resulting in electrode degradation and decreased cycling stability. CMC-based electrodes can experience swelling and shrinkage during cycling due to its water-absorbing property. This volumetric change may lead to mechanical stress and loss of contact between active materials, affecting the overall performance and cycling life of the battery. CMC may exhibit limited thermal stability at high temperatures, potentially leading to binder decomposition or degradation. Careful consideration of operating temperature ranges is necessary to ensure the stability of CMC-based electrode systems. CMC has lower inherent electrical conductivity compared to other conductive binders such as polymeric binders with conductive additives. This lower conductivity can contribute to increased internal resistance and decreased rate capability in certain battery applications [12][26-28].



## Polytetrafluoroethylene (PTFE)

Polytetrafluoroethylene (PTFE) is a widely used binder material in the manufacturing of batteries, known for its exceptional chemical resistance, thermal stability, and low friction properties. PTFE was first discovered in 1938 by a chemist named Roy Plunkett while working at the DuPont Company [31]. Since then, PTFE has become an important material in various industries, including the battery industry. The unique ability of PTFE to fibrillate under shear force made it an excellent binder for the dry electrode manufacturing process. PTFE is synthesized through the polymerization of tetrafluoroethylene monomers. The polymerization process involves the use of radical initiators under controlled temperature and pressure conditions. The resulting PTFE polymer exhibits a unique set of properties, including high tensile strength, low surface energy, and excellent electrical insulation. In the battery industry, PTFE is commonly used as a binder to hold the active materials, such as electrode particles, together in the electrode matrix. It provides mechanical stability and helps maintain the structural integrity of the electrode assembly. The choice of PTFE as a binder is influenced by its high temperature resistance, low yield point, high tensile strength and compatibility with various electrode materials [32].

PTFE has become a valuable binder material in battery manufacturing due to its exceptional chemical resistance, thermal stability, and mechanical properties. PTFE exhibits excellent chemical resistance, making it highly compatible with a wide range of electrolytes and electrode materials. This resistance minimizes binder degradation and enables long-term stability in different battery chemistries. PTFE possesses outstanding thermal stability, with a high melting point and resistance to thermal decomposition. This characteristic allows PTFE-based electrodes to withstand elevated temperatures during battery operation without compromising binder integrity or electrode performance. PTFE-based binders can be utilized to regulate the porosity of electrodes. By adjusting the PTFE content, it is possible to create electrodes with tailored porosity, which can enhance electrolyte infiltration and ion transport, leading to improved battery performance. PTFE exhibits negligible binder decomposition during battery cycling. This characteristic ensures long-term stability, minimizing capacity loss, and allowing for prolonged cycle life [32-33].

Even Though, PTFE possesses relatively low mechanical strength compared to other binder materials. Under high-stress conditions, such as continuous cycling or electrode volume changes, PTFE-based electrodes may experience mechanical failure, resulting in reduced cycling stability. PTFE has inherent low adhesion to active materials and current collectors due to its low surface energy. This can lead to poor interfacial contact, increased electrode polarization, and decreased overall battery performance. PTFE has a low surface energy, resulting in reduced adhesion to active materials and current collectors. This property aids in the easy release of the electrode from the collector, enabling improved electrode fabrication and maintenance. PTFE exhibits poor electrical conductivity, which can increase internal resistance and limit the rate capability of the battery [14]. To address this limitation, PTFE is often combined with conductive additives, such as carbon black or graphene, to improve the overall electrode conductivity. PTFE may not be compatible with certain electrolyte formulations, especially those containing highly polar solvents or additives. The interaction between PTFE and certain electrolyte components can lead to binder swelling, reduced cycling stability, or increased electrode polarization. PTFE is not soluble in common solvents and typically requires a suspension or dispersion method for electrode fabrication. The processing of PTFE-based electrodes may involve additional steps, such as mechanical mixing or high-energy mixing, which can increase the complexity and cost of the manufacturing process [32-34].

## Polyvinyl alcohol (PVA)

Polyvinyl alcohol (PVA) is a widely used binder material in the manufacturing of batteries, known for its excellent adhesive properties, water solubility, and film-forming capabilities. PVA was first discovered in the late 1920s by a German chemist named Fritz Klatte. Since then, PVA has found extensive applications in various industries, including the battery industry. PVA is particularly suitable for aqueous electrolyte-based batteries due to its water solubility and compatibility with the electrolyte system [35].

PVA is synthesized through the hydrolysis of polyvinyl acetate, which is obtained by the polymerization of vinyl acetate monomers. The hydrolysis process involves the reaction of polyvinyl acetate with an alkaline solution, resulting in the formation of PVA. The degree of hydrolysis, or the amount of acetate groups converted to alcohol groups, can be controlled during synthesis to tailor the properties of PVA for different applications [36].

PVA exhibits excellent adhesion properties, allowing it to effectively bind electrode materials together and maintain structural integrity. This enhances the stability and performance of the battery. PVA is soluble in water, making it compatible with aqueous electrolyte-based batteries. It enables efficient processing and manufacturing, as well as easy recycling of battery components. PVA has the ability to form thin, uniform films, ensuring consistent electrode coating and enhancing the electrochemical performance of batteries. It contributes to improved battery efficiency and cycling stability. PVA is a biodegradable and non-toxic material, making it environmentally friendly compared to some other binder options. Its use in batteries aligns with sustainable and eco-conscious manufacturing practices. [35-37]

PVA has a relatively low thermal stability, which may limit its use in high-temperature battery applications. At elevated temperatures, PVA can degrade and lose its binding properties, leading to decreased battery performance. PVA is susceptible to chemical degradation in certain electrolyte environments, particularly those with strong acids or bases. This can result in a loss of adhesion and structural integrity of the electrode materials. Compared to other binder materials, PVA may have lower mechanical strength, especially in high-stress battery operations. This can result in electrode cracking or delamination, leading to decreased battery cycle life. PVA is hydrophilic in nature, meaning it has a tendency to absorb water from the environment. In some battery systems, this can lead to electrolyte swelling and reduced battery performance [35-37].

## 2.2. Inorganic Binders

### Sodium Carboxymethyl Cellulose (CMC-Na)

The synthesis of CMC-Na involves the chemical modification of cellulose by carboxymethylation. This process introduces carboxymethyl groups into the cellulose structure, resulting in enhanced solubility in water and improved adhesive properties. The carboxymethylation process typically involves the reaction of cellulose with sodium chloroacetate under controlled conditions. Commercially available grades of CMC-Na can vary in terms of their degree of substitution (DS) and molecular weight. The DS refers to the average number of carboxymethyl groups per anhydroglucose unit in the cellulose structure. Higher DS values result in increased solubility and improved adhesion properties. The molecular weight of CMC-Na affects its viscosity and film-forming ability.

CMC-Na offers several advantages as a binder in batteries. It exhibits good adhesion to electrode materials, facilitating strong bonding and ensuring the structural integrity of the electrodes during battery cycling. CMC-Na also provides flexibility, allowing it to accommodate volume changes in the electrodes during charge and discharge cycles. Furthermore, CMC-Na is compatible with various electrolytes used in batteries and maintains its stability over a wide range of pH and temperature conditions. It acts as a protective layer on electrode surfaces, reducing the risk of electrode degradation and improving the overall cycling performance of the battery. CMC-Na maintains its stability and integrity even in the presence of aggressive electrolytes, contributing to the long-term performance and safety of the battery system [38]. CMC-Na has excellent film-forming properties, enabling uniform coating on electrode surfaces during battery manufacturing processes. This results in improved electrode-electrolyte contact and enhanced ion transport within the battery, leading to higher charge-discharge efficiency and capacity retention. CMC-Na is derived from cellulose, a renewable and sustainable resource. It is biodegradable and non-toxic, making it an environmentally friendly choice as a binder material in batteries [38].

However, it is important to consider some potential limitations of CMC-Na as a binder in batteries. While CMC-Na exhibits good ionic conductivity, its electronic conductivity may be relatively lower compared to other binder materials. This can impact the overall battery performance, especially in systems that require high electronic conductivity for efficient charge transfer, which can affect the overall electrochemical performance of the battery. Additionally, CMC-Na may have limited resistance to high temperatures, leading to binder degradation and reduced battery stability in certain applications. Therefore, careful consideration should be given to the operating temperature range of the battery system when using CMC-Na as a binder. Moreover, CMC-Na may have limited solubility or compatibility in certain electrolytes, particularly in non-aqueous systems. In such cases, alternative binders or modifications to the CMC-Na structure may be necessary to ensure stable electrode performance. Also, CMC-Na may require specific processing conditions, such as pH adjustment and mixing parameters, for effective binder dispersion and uniform electrode coating. These processing requirements can add complexity and increase manufacturing costs [38-39].

#### Graphene Oxide (GO)

Graphene Oxide (GO) has emerged as a promising binder material for batteries due to its unique properties and versatile applications. GO is a derivative of graphene, a two-dimensional carbon material consisting of a single layer of carbon atoms arranged in a hexagonal lattice. GO is obtained through the oxidation of graphene, resulting in the introduction of oxygen-containing functional groups on its surface. Synthesis methods for GO typically involve the oxidation of graphite, followed by exfoliation and dispersion in water or organic solvents [40]. Various oxidation techniques, such as Hummers method [41], Staudenmaier method [42], and improved variations, have been employed to produce GO for batteries with different degrees of oxidation and functionalization. [43][44]

The use of GO as a binder in batteries offers several advantages. One of the key advantages of using GO in batteries is its excellent electrical conductivity. GO possesses a two-dimensional structure with a high surface area, allowing for efficient electron transport within the battery electrodes. Studies have shown that the incorporation of GO as a binder can enhance the overall conductivity of the electrode, leading to improved charge and discharge rates, higher energy density, and better power output [45]. Furthermore, GO exhibits strong adhesion properties, which contribute to the mechanical stability and structural integrity

of the electrodes. The GO sheets form a network-like structure that helps anchor the active materials and maintain their stability during cycling. This leads to enhanced cycling performance, reduced capacity loss, and improved long-term stability of the batteries [46]. Another advantage of using GO as a binder is its ability to serve as a barrier layer. The oxygen-containing functional groups on GO's surface can act as an effective barrier against the permeation of electrolyte solvents, preventing their unwanted reactions with the electrode materials. This characteristic can significantly improve the cycling stability and safety of batteries, particularly in high-energy applications [47][48]. Furthermore, the unique two-dimensional structure of GO provides a large surface area for active material anchoring and facilitates efficient charge transfer during battery operation.

However, the use of GO as a binder also poses certain challenges. One of the primary limitations is the agglomeration tendency of GO sheets, which can lead to poor dispersion within the electrode matrix. This issue can result in reduced electrode performance and hinder the efficient utilization of the active materials. Researchers have explored various strategies to overcome this challenge, including the functionalization of GO with other polymers or surfactants to enhance its dispersibility [49]. Moreover, the cost of large-scale production of high-quality GO remains relatively high, limiting its widespread commercial adoption. Efforts are being made to develop scalable and cost-effective synthesis methods to overcome this barrier and make GO more economically viable for battery manufacturing [50][51].

Carbon nanotubes (CNT).

Carbon nanotubes were first discovered in 1991 by Sumio Iijima [52]. They are cylindrical carbon structures with exceptional mechanical, electrical, and thermal properties. CNTs can be synthesized using various methods such as arc discharge, laser ablation, and chemical vapor deposition (CVD). Among these, CVD has emerged as a commonly used technique for large-scale production of CNTs with controlled properties [53]. Commercially there are different grades of CNTs available like single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs), with varying diameters, lengths, and purities [54][55].

One of the main advantages of using CNTs as binders in batteries is their high electrical conductivity. CNTs possess excellent electrical properties, enabling efficient electron transport within the battery electrodes. This enhanced conductivity can lead to improved charge and discharge rates, higher power output, and overall enhanced battery performance [56]. Additionally, CNTs offer excellent mechanical properties, including high tensile strength and flexibility. As binders, CNTs can improve the adhesion between electrode materials and enhance the mechanical stability of the electrodes. This can result in improved cycling stability, reduced capacity loss, and enhanced durability of the batteries [57]. Furthermore, CNTs can also act as a conductive network in the electrode matrix, providing a continuous pathway for electron transfer. This interconnected network can facilitate efficient charge transport and distribution within the electrode, minimizing localized high-resistance areas and improving the overall electrochemical performance of the battery [58]. The unique structure of carbon nanotubes facilitates their use as conductive additives or support materials in battery electrodes. CNTs can form a conductive network within the electrode, promoting uniform electron distribution and minimizing localized high-resistance regions. This leads to enhanced electrochemical performance, such as higher capacity, improved rate capability, and lower internal resistance [59]. Carbon nanotubes have a high aspect ratio (length-to-diameter ratio), which allows for improved contact between active materials and electron pathways. This feature

contributes to better charge transfer kinetics and can enhance the overall energy efficiency of batteries [60].

However, there are certain challenges associated with the use of CNTs as binders. One limitation is their tendency to agglomerate, which can lead to poor dispersion and hinder their uniform distribution within the electrode matrix. Various strategies have been explored to overcome this issue, including functionalization of CNTs to improve their dispersibility and compatibility with other electrode components [61][62]. The synthesis and purification processes involved in producing high-quality carbon nanotubes can be complex and costly. This can impact the overall cost-effectiveness of using CNTs as binder materials in large-scale battery manufacturing [63]. Carbon nanotubes are susceptible to structural damage during the fabrication process or under extreme conditions. Mechanical stress, oxidation, or other factors can cause the breaking or bending of CNTs, leading to a loss of their desired properties and reduced performance in batteries [64]. As with any nanomaterial, safety considerations arise when working with carbon nanotubes. Inhalation of nanotubes or exposure to high concentrations may pose health risks. Proper handling, dispersion techniques, and protective measures are necessary to ensure the safe use of CNTs in battery production [65].

#### Lithium Polyacrylate (LiPAA)

Lithium Polyacrylate (LiPAA) is a binder material that has gained significant attention in the field of battery technology. As a polymer-based binder, LiPAA offers several advantages, including improved electrode stability, enhanced cycling performance, and better adhesion between active materials and current collectors. The synthesis of LiPAA typically involves the polymerization of acrylic acid monomers in the presence of a lithium salt. This process can be achieved through solution polymerization or emulsion polymerization techniques. The resulting polymer has a high molecular weight and can be tailored to exhibit specific properties such as viscosity, solubility, and thermal stability. Commercially available grades of LiPAA may vary in their molecular weight, dispersity, and lithium salt content. These variations can influence the binder's performance in terms of adhesion, flexibility, and electrochemical stability.

LiPAA is an excellent binder due to its unique adhesive cum cohesive nature and wetting properties. It forms a uniform thin passivating film on active material and conductive carbon particles in composite cathodes and also compensates Li-ion loss in full Li-ion batteries by acting as an extra Li source. The LiPAA lead to a significant improvement in the electrochemical performance like cycle life, cell impedance, and rate capability. It is also cheap and environmentally-friendly [66-68].

### 2.3. Hybrid Binders

Hybrid binders represent a promising and innovative approach in the realm of electrode fabrication for advanced energy storage systems. By synergistically combining the distinctive attributes of both polymeric and inorganic materials, hybrid binders aim to harness the strengths of each component while mitigating their individual limitations. This dynamic integration opens avenues for enhanced electrode cohesion, mechanical stability, ion and electron transport, and overall electrochemical performance. As the demand for high-energy-density and durable battery technologies continues to escalate, the exploration of hybrid binders emerges as a pivotal frontier in tailoring binder formulations to address the evolving challenges of modern energy storage applications.

Hybrid Binder	Description
PVDF/Graphene Oxide (GO) Hybrid Binder	This hybrid binder combines poly(vinylidene fluoride) (PVDF) with graphene oxide (GO) to enhance the mechanical properties and electrical conductivity of the binder system. PVDF/GO hybrid binder has been reported to improve the adhesion between active material particles and current collectors, leading to better electrode integrity and electrochemical performance [69].
PAA/Silica Hybrid Binder	This hybrid binder combines poly(acrylic acid) (PAA) with silica nanoparticles to enhance the mechanical stability and electrolyte compatibility of the binder. PAA/silica hybrid binder has been shown to improve the adhesion between electrode materials and current collectors, as well as mitigate the detrimental effects of electrode swelling/shrinking during cycling [70].
PVDF/Carbon Nanotubes (CNTs) Hybrid Binder	This hybrid binder combines PVDF with carbon nanotubes (CNTs) to enhance the mechanical strength and electrical conductivity of the binder system. PVDF/CNTs hybrid binder has been shown to improve the adhesion between active material particles and current collectors, enhance the electron/ion transport, and provide mechanical reinforcement for better electrode stability [71].

Table 2: Examples of Hybrid binders

## Recent advancements and future perspectives

There is a need to develop new binders with improved properties as the requirement for high energy density is increasing day by day. As new technologies like Li-sulfur batteries, Si-based batteries, Li-Air batteries, sodium (Na) ion batteries and magnesium (Mg) ion batteries are progressing, the need for new binders are also increasing.

For example, the conventional binders used in lithium ion batteries fail to be effective in sodium ion and magnesium ion batteries as these ions have higher radius as compared to lithium ions. Which makes them comparatively harder and causes structural damage to the electrodes. Researchers have developed polymer binders with enhanced sodium ion conductivity to improve the overall performance of sodium-ion batteries. These binders, such as polyacrylonitrile (PAN), polyethylene oxide (PEO), and their derivatives, exhibit high sodium ion diffusion coefficients and effectively suppress electrode swelling, improving the cycling stability and rate capability of sodium-ion batteries. But during prolonged cycles, PAN undergoes structural degradation and PEO results in delamination of electrodes due to weak adhesive nature [72]. Whereas, Inorganic binders such as sodium alginate, sodium carboxymethyl cellulose (CMC-Na), or sodium polyacrylate provides good adhesion between active materials and current collectors to improve the electrode/electrolyte interface stability and to mitigate the volume changes during cycling. Which in turn leads to enhanced cycling stability and rate capability [73]. So in order to make the best of use of these binders, their hybrid combinations are being explored to be used in sodium ion batteries. These hybrid

binders provide a synergistic effect by enhancing the mechanical integrity, electronic conductivity, and adhesion properties of the electrode materials, leading to improved cycling stability and rate performance [74]. Another class of binders that are actively explored for the development of sodium ion batteries is self healing binders. These binders are developed to address the electrode degradation and capacity fading issues in sodium-ion batteries. These binders contain encapsulated healing agents that can autonomously repair electrode cracks or interface damage during cycling, improving the long-term cycling stability and electrochemical performance of sodium-ion batteries [75].

Even though Lithium sulfur batteries offer high energy densities compared to lithium ion batteries, there are a lot of challenges associated with commercializing Li-S batteries. Structural degradation and Polysulfide effect remains as the two of the major limiting factors for development of Li-S batteries. So, developing an ideal binder for Li-S batteries needs to suppress the crack generated and also trap polysulfide in the cathode region to reduce the loss of active material. PAN has shown promise as a binder for Li-S batteries due to its high mechanical strength and good adhesion to sulfur cathodes. PAN binders help to maintain the structural integrity of the cathode during cycling, leading to improved cycling stability and higher capacity retention. But due to the limited compatibility of PAN with sulfur electrochemistry, the use of PAN as binder of Lithium Sulfur batteries is limited as it may result in sub-optimal utilization of sulfur [72]. Hence, polymers with a strong affinity for polysulfide intermediates (Polysulfide-philic Polymers) have been developed as binders to mitigate the dissolution and shuttling of polysulfide species in Li-S batteries. These binders can effectively suppress the dissolution of polysulfides and improve the electrochemical performance and cycling stability of Li-S batteries [73]. Poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) has been investigated as a binder for Li-S batteries due to its excellent chemical stability and high ionic conductivity. PVDF-HFP binders can enhance the adhesion between the sulfur cathode and current collector, improve the sulfur utilization, and enhance the overall performance of Li-S batteries [74]. Additionally, Conducting polymers, such as polyaniline (PANI) and polypyrrole (PPy), have been explored as binders for Li-S batteries to provide both mechanical stability and electrochemical activity. Conducting polymer binders can enhance the electrochemical reaction kinetics, promote sulfur utilization, and improve the cycling stability of Li-S batteries [75].

In the case of Lithium oxygen batteries (LI-O<sub>2</sub>), there is a requirement to develop binders which are highly chemically inactive and provide higher electrode integrity. This is because Li-O<sub>2</sub> batteries contain superoxides which are capable of undergoing oxidizing reactions with the binder material and these reactions lead to the loss of electrode integrity and capacity loss during cycling. Therefore, In the case of Lithium air batteries, highly chemically stable binders are required. Polymer-ceramic composite binders, such as polyvinylidene fluoride (PVDF) combined with ceramic nanoparticles like Al<sub>2</sub>O<sub>3</sub> or SiO<sub>2</sub>, have shown promise in improving the stability of the lithium metal anode. These binders provide mechanical strength and form a protective layer on the lithium metal surface, mitigating dendrite formation and improving cycling stability [76]. Also, polymer electrolyte binders such as polyethylene oxide (PEO) or polyvinyl alcohol (PVA) have been investigated to simultaneously act as binders and solid electrolytes for lithium metal batteries. These binders offer ion-conducting properties and can form a stable SEI layer on the lithium metal surface, reducing dendrite growth and improving the overall performance of lithium metal batteries [77]. 3D structured binders, such as three-dimensional porous carbon or metal frameworks, have been explored to accommodate the volume expansion of the lithium metal anode during cycling. These binders provide mechanical support, enhance lithium ion transport, and suppress dendrite growth, resulting in improved cycling stability and Coulombic efficiency [78]. Researchers have developed artificial SEI binders, such as organic additives or polymers, to regulate the formation of a stable and uniform SEI

layer on the lithium metal surface. These binders can enhance the lithium ion transport and suppress the growth of lithium dendrites, leading to improved cycle life and Coulombic efficiency of lithium metal batteries [79].

Recent advancements in binder technology have significantly contributed to addressing the inherent challenges associated with silicon anodes in lithium-ion batteries. Traditional binders often struggled to accommodate the substantial volume changes that silicon undergoes during lithiation and delithiation, resulting in electrode cracking and performance degradation. However, novel binder materials such as polymeric binders, including carboxymethyl cellulose (CMC) and sodium alginate, have demonstrated remarkable effectiveness in mitigating these issues. These binders exhibit strong adhesion to silicon particles, ensuring robust electrode integrity and minimizing electrode pulverization [80][81]. Additionally, their flexibility and ability to accommodate silicon's volume changes contribute to improved cycling stability and enhanced capacity retention [82]. But they fail to deliver cyclic stability for prolonged cycles. Moreover, conductive polymer binders, such as poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), have been harnessed to enhance both electronic and ionic conductivity within the silicon electrode, further enhancing its performance [83]. By fostering better electrode-matrix cohesion, these innovative binder materials are effectively addressing the mechanical and electrochemical challenges of silicon anodes, thereby paving the way for the realization of high-capacity lithium-ion batteries with improved stability and longevity. Binders with self-healing properties are also explored for developing high energy dense silicon anodes.

## Conclusion

In conclusion, the extensive examination of various binder technologies presented in this review sheds light on their paramount role in shaping the landscape of battery manufacturing and energy storage applications. Polymer binders, with their tailored adhesion properties and electrochemical stability, have been the cornerstone of electrode fabrication for years. Inorganic binders, offering robust mechanical support and thermal stability, have showcased promising potentials, especially in high-energy and high-temperature systems. The emergence of hybrid binders, fusing the merits of polymers and inorganics, has guided into a new era of enhanced performance and structural integrity in batteries.

Furthermore, recent advancements in binder technologies for specific battery chemistries have displayed a remarkable potential to surmount critical limitations. In lithium-sulfur (Li-S) batteries, binders have played a pivotal role in controlling the shuttle effect, suppressing polysulfide dissolution, and achieving prolonged cycling stability. Sodium-ion batteries have witnessed binder innovations contributing to improved rate capabilities and cycling performance. Meanwhile, the pursuit of efficient lithium-air batteries and high-capacity silicon anodes has been invigorated by novel binders that address electrode cracking, volume expansion, and interface challenges.

Looking ahead, the future of binder technologies appears promising, with ongoing efforts directed toward refining binder formulations, optimizing interactions with active materials, and tailoring properties to specific battery designs. As energy storage demands continue to evolve and new chemistries emerge, binders will undoubtedly remain at the forefront of research and development, playing a pivotal role in propelling battery technologies towards higher energy densities, longer lifespans, and safer operation. This comprehensive exploration of binders' diverse roles and the latest advancements underscores their



indispensable significance in shaping the trajectory of energy storage systems and invites further interdisciplinary collaborations to drive innovation and optimize battery performance for a sustainable energy future

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