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Review

Comparison of Dispersing Processes of Bio-Based and Synthetic Materials: A Review

Leah Jalowy *, Dominik Nemec * and Oguzhan Ilhan

Fraunhofer Institute for Manufacturing Engineering and Automation IPA, 70569 Stuttgart, Germany; oguzhan.ilhan@ipa.fraunhofer.de

* Correspondence: leah.jalowy@ipa.fraunhofer.de (L.J.); dominik.nemec@ipa.fraunhofer.de (D.N.)

Abstract: The ever-growing environmental and sustainability awareness as well as the associated increased independence from petroleum has led to bio-based materials increasingly replacing synthetic, non-renewable materials in various applications, including food packaging, coatings, adhesives, and energy storage devices. Although bio-based materials offer advantages such as reduced toxicity and harmfulness for humans and the environment, as well as contributing to the conservation of important resources, these aspects are usually not sufficient for commercialization. Integrating bio-based materials into existing technologies is challenging due to inherent disadvantages, such as difficult processability and low moisture resistance, making it difficult to readily substitute them for synthetic materials. Consequently, surface modifications are often necessary to make bio-based materials suitable for the intended applications. This review highlights the critical role of processing methods in the successful substitution of synthetic materials with bio-based alternatives. While previous studies have primarily concentrated on material combinations and formulations of bio-based applications, often considering processing methods as secondary, this review explores the influence and importance of dispersion quality. It examines how varying dispersing methods and process parameters can impact the performance of bio-based materials, alongside addressing the specific requirements for both the materials and the dispersing processes. Furthermore, it focuses on bio-based dispersions based on lignin and polysaccharides, particularly in applications such as bio-based adhesives and binders for battery technologies. By addressing these aspects, this review aims to reveal existing research gaps and provide insights into optimizing the processing of bio-based materials for diverse applications.

Keywords: bio-based materials; dispersions; processing; dispersing technology; mixing; process parameters

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1. Introduction

In recent years, bio-based materials have been increasingly receiving attention in various areas of application [1,2]. Due to increasing environmental awareness and the depletion of fossil resources, there is growing interest in replacing petroleum-based products with sustainable, bio-based materials [1,3,4]. Bio-based materials originate from renewable biological sources, including plants, animals, enzymes, and microorganisms like bacteria and fungi [5,6]. They are primarily derived from biomass, encompassing a wide range of organic materials such as agricultural residues, forestry by-products, animal waste, and dedicated energy crops [7,8]. Microbial biomass, derived from bacteria and fungi, can also serve as valuable sources for bio-based materials [9]. The conversion of these

biomass sources into bio-based materials involves various processes, including fermentation, enzymatic treatment, and chemical conversion [10]. In contrast, petrochemical-derived materials are sourced from fossil fuels and are typically non-renewable [11]. Petro-based materials can be categorized into various groups, including thermoplastic polymers like polyethylene (PE) and polypropylene (PP) [12], as well as polyvinyl acetate (PVAc) and polyvinyl chloride (PVC) [5], synthetic rubbers such as styrene-butadiene rubber (SBR) [13], and fluoropolymers like poly(vinylidene difluoride) (PVDF) [14]. Examples of bio-based materials include natural polymers like lignin [2,15], proteins like gelatin [15,16], and polysaccharides such as cellulose [2,15], starch [2], chitosan [2,15], sodium alginate, and xanthan gum [15,16], with many more available. Bio-based materials are convincing compared to synthetic materials due to their generally reduced toxicity and harm to humans and the environment [2,17,18]. In addition, they naturally have a low carbon footprint before processing, as CO_2 is captured from the atmosphere during plant cultivation. This, in turn, leads to a reduction in the impact of climate change [19]. Moreover, the use of renewable raw materials conserves important resources, leading to greater independence from petroleum and consequently to increased sustainability [2,15].

Given their environmental benefits, bio-based materials are used in a wide variety of areas. For example, numerous bio-based and biodegradable polymers have been investigated over the last 20 years to produce food packaging. Some of them have shown sufficient mechanical properties and satisfactory barrier behavior [1,18,20]. Additionally, bio-based hydrophobic coatings are used in the construction industry for building material protection as well as in the packaging industry [21,22]. For instance, aqueous polymer dispersions are applied to paper or cardboard to provide barrier properties [4]. Bio-based materials are widely used in the adhesive sector. Bio-based adhesives are applied in various areas such as the paper and packaging industry [23], the automotive industry [17,24], the footwear industry [24], and especially in the wood industry [17,24–27]. Furthermore, bio-based materials are used in energy storage devices. The focus there is primarily on investigating the substitution of synthetic binders with bio-based binders in battery applications [15,16]. However, biopolymers such as lignin can also be used as lignin-based carbon material, polymer electrolyte, and electrode material (e.g., hybrid lignin–polymer combination or organic composite electrodes) in various battery systems [28].

While the applications of bio-based materials are diverse and promising, several important factors must be considered. Bio-based materials possess different properties compared to finite fossil materials. While the latter are considered permanent and stable in time, bio-based materials are generally heterogeneous, exhibit unpredictable behavior, and have a limited lifespan [29,30]. The properties of bio-based materials or biopolymers are strongly dependent on their monomeric composition and size [31]. Thus, due to their heterogeneity in composition, they often exhibit varying material properties [32]. The limitations of biopolymers mainly include their chemical and physical properties such as high molecular weight, hydrophilicity or poor moisture resistance, aggregation, brittleness, crystallization, weak mechanical properties, and difficult processability [1,18]. These properties present significant obstacles for industrial applications, necessitating improvements through various approaches [18]. One possible approach to counteract this is modification. The high content of functional groups in numerous bio-based materials enables a wide variety of modifications, which are accompanied by the introduction of new functionalities [2]. By modifying the functional groups, for example, hydrophobic properties can be generated by introducing long aliphatic carbon chains or cross-linkable groups, which, in turn, lead to the formation of denser networks [2,22]. The modification of bio-based materials can therefore enable both the substitution of synthetic materials and the opening of new application areas.

The literature extensively reports on the use of bio-based materials instead of petro-based materials in various application fields. However, often, the processing of biomaterials poses significant challenges, as the same process parameters cannot always be applied to bio-based materials as for synthetic materials [1]. Particularly, the production of homogeneous and stable dispersions is repeatedly described as problematic [27,33]. Dispersing processes have a major influence on the production and stability of dispersions, which is why in some literature, a more intensive examination of alternative manufacturing technologies is recommended [27]. Consequently, mixing methods and process parameters adapted to the biomaterial used should be applied to integrate biomaterials into conventional processes. As the processing of bio-based raw materials increases, there is consequently a growing interest in understanding these processes more precisely. Given the growing body of literature on bio-based materials, this review distinguishes itself by focusing specifically on the dispersing processes of these materials in comparison to synthetic alternatives. It highlights the potential impact of different dispersing methods and process parameters on the performance of bio-based materials, an area that has received limited attention in the existing literature. By addressing this gap, the review not only emphasizes the challenges associated with processing bio-based materials but also discusses how exploring various dispersing techniques could enhance their performance in diverse applications. This novel approach aims to contribute to a deeper understanding of the critical interplay between processing techniques and the effectiveness of bio-based materials, thereby enhancing their commercial viability. Therefore, the objective of this review is to demonstrate that the processing of bio-based materials plays a crucial role in the successful substitution of synthetic materials with bio-based alternatives, emphasizing the relationship between processing techniques and the performance of bio-based materials across various applications.

2. Processing and Dispersing of Bio-Based Materials

2.1. Dispersing Process

The dispersing process refers to the mixing procedure for producing a dispersion [34]. A dispersion consists of at least two phases that are not soluble in each other: a disperse and a continuous phase. The disperse phase can be subdivided into definable individual elements, while the continuous phase is the surrounding medium [35]. A dispersion in which solid particles are present as the disperse phase in a liquid continuous phase is also referred to as a suspension. A dispersion of two immiscible liquids as a finely dispersed mixture is called an emulsion [36,37]. In general, the dispersing process (for suspensions) can be divided into three steps (Figure 1). In the first step, the solid particles are wetted in the liquid phase. In the second step, the solid particles are comminuted and distributed (deagglomeration). In the third and final step, the stabilization of the dispersion takes place [34]. The breaking up of agglomerates can occur through three different mechanisms: shattering, rupture, and erosion [38,39].

Following the description of the dispersing process and its essential steps, it is crucial to delve deeper into the underlying physical principles that govern these processes. The following section explores the physical factors influencing the dispersing processes of bio-based and synthetic materials. Key equations that describe these mechanisms are presented, as understanding them is essential for analyzing the impact of various factors on dispersion efficiency and effectiveness. The interaction between particles and the forces applied during mixing significantly affects the resulting material properties.

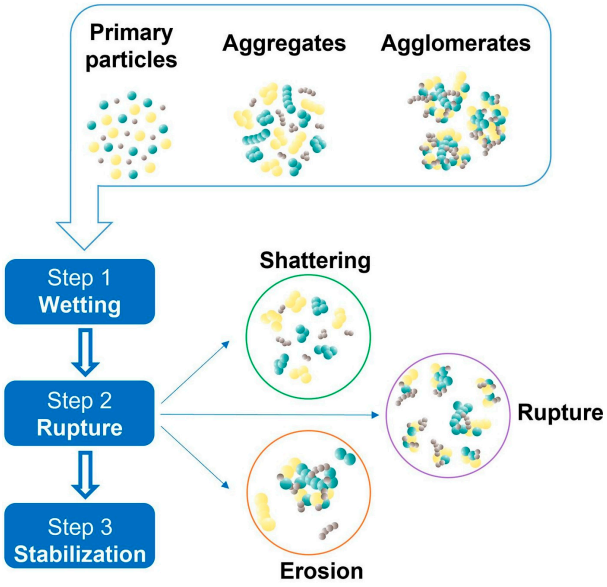


Figure 1. Schematic representation of the three-stage dispersing process and the associated various deagglomeration mechanisms: shattering, rupture, and erosion. Reproduced from [40] under terms of CC BY-NC-ND license.

One fundamental aspect of these interactions is shear stress (σ), which represents the internal forces that arise as particles are subjected to external forces during dispersion. The shear stress acting on the particles during dispersion can be defined as follows:

$$\sigma = \frac{F}{A} \tag{1}$$

where F is the applied force (in N), and A is the area over which this force acts (in m^2). This relationship is fundamental in understanding how forces impact particle behavior in dispersions.

The shear rate ($\dot{\gamma}$) describes the rate at which particles are displaced relative to one another:

$$\dot{\gamma} = \frac{du}{dy} \tag{2}$$

Here, du represents the change in velocity within the flow (in m/s), and dy is the vertical distance (in m). This parameter is crucial for assessing the efficiency of the dispersion process.

The relationship between shear stress and shear rate is captured by the following equation:

$$\sigma = \dot{\gamma} \cdot \eta \tag{3}$$

where η is the dynamic viscosity (in Pas). Viscosity is a crucial determinant of material behavior under shear, significantly impacting the efficiency of the dispersion process.

Another important parameter that influences flow behavior during dispersion is the Reynolds number (Re). It enables the prediction of flow behavior, mixing quality, heat transfer properties, and process efficiency:

$$Re = \frac{\rho \cdot v \cdot D}{\mu} \tag{4}$$

where ρ is fluid density (in kg/m^3), v is the average velocity (in m/s), D is the characteristic diameter (or diameter of the mixer) (in m), and μ is dynamic viscosity (in Pas). The flow behavior can be categorized as follows:

- $Re < 2000$: Laminar flow, where viscous forces dominate and the flow is uniform and predictable.
- $2000 < Re < 4000$: Transition area, where both laminar and turbulent flow can occur (unstable state).
- $Re > 4000$: Turbulent flow, where inertial forces dominate, leading to chaotic and irregular flow patterns.

In high-speed mixers, for example, a high Reynolds number may indicate a turbulent mixing process, which is advantageous for achieving a homogeneous distribution of particles or liquids. Conversely, a low Reynolds number may suggest insufficient mixing, potentially indicating that the viscosity of the mixture is too high.

Understanding the energy dynamics during the shear process is essential for optimizing dispersion efficiency. The energy input directly affects how effectively the mixing occurs, influencing the overall performance of the dispersion. The energy introduced during the shear process can be calculated by the following equation:

$$E = P \cdot t = \sigma \cdot \dot{\gamma} \cdot V \quad (5)$$

where E is the input energy (in J), P is the power of the mixer (in W), t is the time (in s), and V is the volume of the mixture (in m^3). This equation illustrates how energy impacts the performance of the dispersing process.

2.2. Dispersing Devices

Numerous dispersing devices are available for producing dispersions (Figure 2). The correct choice of the dispersing method and parameters is crucial for successful dispersion [41]. The following dispersing methods are used in dispersing technology: rotor–stator [42], ultrasound [43], extruders and kneaders, stirred media mills, dissolvers, or roller mills (three-roll mills) [44].

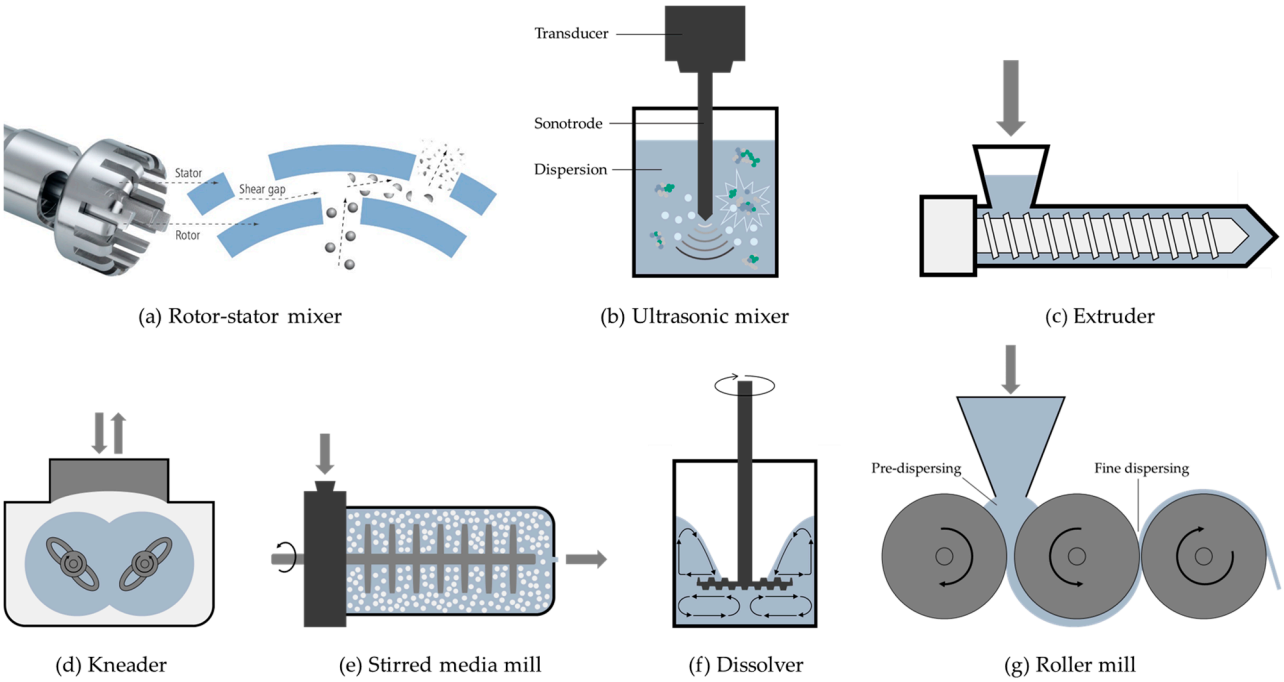


Figure 2. Schematic representation of different dispersing devices: (**a**) Rotor–stator mixer (reproduced with permission from [45]), (**b**) ultrasonic mixer, (**c**) extruder, (**d**) kneader, (**e**) stirred media mill, (**f**) dissolver, and (**g**) roller mill.

To effectively evaluate the performance of these dispersing methods, it is important to analyze their operational characteristics, including shear stress, viscosity, application areas, limitations, and upscaling potential. Table 1 offers a comparison of various dispersing devices, detailing their specific characteristics and relevant factors for practical application.

Table 1. Comparison of dispersing devices including shear stress, viscosity, application areas, limitations, and upscaling.

Dispersing Device	Shear Stress	Viscosity	Application Areas	Limitations and Upscaling
Rotor–Stator Mixer	Generates a uniform, moderate laminar shear field via high rotational speeds [46]	Particularly effective for strongly shear-thinning systems [46]	• Food industries [42,47,48] • Pharmaceutical and cosmetic industries [42] • Paints, inks, dyes [48] • Latexes, adhesives [47]	In very high-viscosity systems, energy dissipation becomes less efficient [46]
Ultrasonic Mixing	Uses acoustic cavitation to create shock waves and micro jets, yielding higher local shear stress [49]	Suitable for low-to-moderate-viscosity fluids, performance declines in high-viscosity systems [50]	• Micron-sized or nano-sized dispersions [51] • Long-time stable and homogeneous nanofluids [52,53] • Production of highly concentrated emulsions [51]	Upscaling typically involves flow-through designs or multiple transducers [50]
Extruder	Combines shear, elongational, and compressive forces in narrow channels for localized high-intensity shear [46]	Highly effective for high-viscosity materials [46]	• Production of highly pasty media [44] • Plastics and rubber industry [54] • Food processing [54] • Battery paste [55] • Carbon black mixed in rubber [55] • Pigments and fillers in polymer melts [44]	Different polymers have unique rheological properties that influence processing conditions and therefore an exact temperature control is vital [56]
Kneader	Applies both shear and compressive forces by forcing material between moving elements and vessel surfaces [46]	Well-suited for high-viscosity, heavily loaded formulations [46]	• Highly viscous and non-flowable applications [57–59] • Polymer processing [57,60,61] • Food processing [60] • Pharmaceutical industry [61]	Unsuitable for reactive systems due to the excessive residence time of the thermally stressed melt [46]
Stirred Media Mill	The effectiveness of this process is dependent upon the kinetic energy of the grinding media [46]	Suitable for a wide range of viscosity [46]	• Chemical and pharmaceutical industry [62,63] • Paint, lacquer, and inks industry [62,63] • Ceramic industry [62,63] • Cosmetic industry [63] • Food industry [63] • Fillers and pigments [63]	Operating speeds are limited by accelerated wear of the grinding media and mill internals [46]

Table 1. *Cont.*

Dispersing Device	Shear Stress	Viscosity	Application Areas	Limitations and Upscaling
Dissolver	The subject particles primarily experience fluid shear flow [46]	Suitable for low-to-moderate-viscosity fluids [46]	• Paint, lacquer, inks, and coating industry [64–66] • Pigments and dyes [64,65] • Pharmaceutical and cosmetic industry [64,66] • Food industry [64,66] • Adhesives and sealants [64,66]	Dispersion limit: further processing yields little size reduction [46]
Roll Mill	Imparts controlled, high-intensity laminar shear in a narrow gap [46]	Low viscosity can cause splashing and insufficient shear, optimally used for high viscosity [46]	• Printing inks [44,67–69] • Artist colors [67–69] • Electronic industry [67–69] • Pastes for solar technology [67–69] • Carbon nanotubes [67–70] • Pharmaceutical and cosmetic industry [67]	Limited upscaling due to strict gap control and design constraints, resulting in lower throughput [46]

2.3. Influence of Material Properties on the Processing of Bio-Based Materials

The rheological properties of bio-based materials are of vital importance in the mixing process. Many bio-based mixing processes operate with high solids contents. These dispersions often have a high viscosity and behave like non-Newtonian fluids [71]. This behavior was observed by Savitskaya et al. [72] in 2016 in the case of lignin-containing oil dispersions. In their study, they demonstrated that the rheological behavior changes depending on the lignin concentration. With increasing lignin content in the dispersion, a change from Newtonian to non-Newtonian behavior, exhibiting shear-thinning and thixotropy, was observed. Rheological behavior plays an important role in mixing processes as it affects the handling, processability, and properties of the resulting mixture. Understanding the rheological properties allows for better control and adjustment of the mixing process. Depending on viscosity and flow behavior, different dispersing methods and parameters may be required to achieve optimal results.

Important for the processing, as well as for the application and use of biomaterials, is their stability under certain conditions. During the dispersing process, temperature, pH value, solids content, solvents, and shear forces as well as the dispersion time and type of biomaterial itself can be decisive parameters that influence the quality of the dispersion.

The temperature during the dispersing process can have major impact on the stability of the dispersion. For dispersions of nanolignins in aqueous media, for example, heating the lignin during the process could have a positive effect on stability. It has been described that a temperature higher than 65 °C is required to form a stable nanolignin dispersion with sufficient yield. In addition to temperature, the pH dependency of the nanolignin dispersions was also studied. It was found that the pH value affects the dispersion stability and there is usually an optimal pH range in which stable dispersions can be formed. A too high pH value, in the case of nanolignin dispersions, for instance, led more to the formation of a solution instead of a dispersion. Very low pH values in the acidic range also had a

negative effect on the long-term stability of the dispersions, meaning that the particles no longer remained dispersed [73]. This behavior was also observed by Lievonen et al. [74], whose study investigated lignin nanoparticle dispersions regarding their pH dependence. It was found that very low pH values led to particle aggregation and very high pH values to particle dissolution. In addition to the dissolution of spherical lignin nanoparticles at an alkaline pH > 10, Zou et al. [75] also reported the dissolution of these in common organic solvent systems.

In a study by Yang et al. [76], the use of nano- and microscale lignin in phenol–formaldehyde–resol adhesives was investigated. The results showed that lignin nanoparticles could be dispersed better in resol than lignin microparticles, which was confirmed by Scanning Electron Microscopy (SEM) analyses. Consequently, particle size also influences the dispersibility of bio-based materials. Österberg et al. [77] also report in their article on the advantages of using nanoscale lignin particles. In addition to the advantages of nanoscale size, the benefits of spherical shapes are also discussed. From a technical perspective, for example, one advantage of spherical particles over other shapes is that they do not have sharp edges that could break off or wear out during processing. According to Österberg et al., shape regularity is a crucial factor for colloidal behavior (e.g., in terms of agglomeration, packing, and flow).

Since the successful dispersion of bio-based materials, as already mentioned, depends on many factors, the following tables provide an overview of the requirement criteria for bio-based materials (Table 2) and for dispersing processes (Table 3).

2.4. Requirements for Bio-Based Materials

Processing and dispersing bio-based materials depends on many factors. For successful dispersion production, requirements are placed not only on the bio-based materials themselves but also on the dispersion processes. Table 2 outlines the requirement criteria for bio-based materials such as mechanical and thermal stability, chemical properties, and solubility, as well as the resulting properties and challenges.

Table 2. Requirement criteria for bio-based materials.

Criteria	Properties and Challenges
Mechanical stability	Gelation Structural change
Chemical properties	Reactivity of the material Molecular structure (e.g., hydrophobicity, molecular mass)
Thermal stability	Structural changes or decomposition of bio-based materials Denaturation of proteins
Solubility behavior	pH dependence Solvent selection (solubility in, e.g., organic solvents) Solvent compatibility (solvent interactions with materials)
Homogenization of the paste	Particle size distribution Formation of aggregates/agglomerates Sedimentation/phase separation
Long-term stability of bio-based materials in the paste	Biodegradation of the material after the process Fouling

Table 2. *Cont.*

Criteria	Properties and Challenges
Material extraction	Purity Heterogeneity/homogeneity of bio-based materials Special preparation required Reproducibility of quality
Rheology	Viscosity Newtonian/non-Newtonian behavior Shear-thinning/shear-thickening

2.5. Requirements for Dispersing Processes

Dispersing bio-based materials requires specific processes and parameters. Table 3 presents various requirement criteria and properties, as well as challenges for the dispersing processes themselves. For example, the mixing technology and tools, mixing temperature and time, or the process control (pre-dispersion, order of addition) can be important influencing parameters or criteria for dispersion production. Table 3 is intended to provide an overview of the requirement criteria for the dispersing process and serve as a basis for discussion.

Table 3. Requirement criteria for dispersing processes.

Criteria	Properties and Challenges
Solvent	Aqueous or organic solvents Solvent compatibility pH value Toxicity and environmental impact
Additives	Use of dispersants to increase stability Surface modifiers (improving particle and liquid interactions for more effective dispersion) Rheology modifiers (controlling the flow properties of the dispersion to ensure processability and applicability) Catalysts (influencing reactivity and selectivity) Fillers (improving rheological and mechanical properties, price reduction)
Mixing technology/ tools	Batch or continuous process Geometry and size of mixing tools Dissolver, rotor–stator, ultrasound, roller mills, kneader, extruder, stirred media mills, etc.
Process control	Pre-dispersion Order of addition: feed in solids or liquids first Add in portions/continuous addition
Shear forces	Sensitivity to shearing and comminution Optimal shear rate (efficient dispersion without compromising material structure or quality) Stability of end products Energy efficiency
Homogenization of the paste	During temporary storage: stirring, degassing

Table 3. *Cont.*

Criteria	Properties and Challenges
Mixing temperatures	Aggregation/agglomeration or deagglomeration Viscosity change Solubility change Thermal sensitivity
Mixing times	Energy requirement/energy efficiency (optimal mixing duration when dispersion quality is no longer improved) Viscosity Particle size distribution
Solids content/ concentration	Influence on rheological properties (ensuring processability and applicability of the material) Adjustment of the solids content used for synthetic vs. bio-based materials (e.g., depending on viscosity of materials) Finding the optimal solids content (without causing agglomeration or sedimentation)
Rheological properties	Viscosity ranges Formation of aggregates/agglomerates Particle size distribution
Reproducibility	Consistent particle size distribution vs. natural variability Standardized manufacturing processes vs. process adjustment for different raw material batches
Scale-up	Fluid mechanical aspects (with increasing scale, fluid mechanical aspects may arise that influence shear forces and shear rates—analysis and adjustment of process parameters required) Homogeneity of dispersion (uniform distribution of particles in liquid medium on larger scales—may require adjustment of stirring and mixing systems)

3. Dispersions of Bio-Based Materials

The increasing recognition of bio-based materials as sustainable alternatives to traditional synthetic options has marked a significant shift in various industries. In 2023, the global biopolymers market was valued at approximately USD 17.5 billion, with projections estimating it will reach USD 47.4 billion by 2032, reflecting a robust compound annual growth rate (CAGR) of 11.7% over the forecast period 2024 to 2032. This expansion is driven by rising awareness of environmental issues, increasing consumer demand for eco-friendly products, and regulatory pressures supporting the adoption of bio-based materials [78]. As of 2025, production capacity for bio-based polymers is expected to increase significantly, with projections indicating a total capacity increase to 5.7 million tons by 2029, reflecting a CAGR of 18% over the period from 2024 to 2029 for key bio-based polymers. This growth is particularly notable in regions like China, Europe, and the Middle East, where substantial investments are being made. For instance, China's bio-based polymer industry is expected to grow from 765,000 tons in 2023 to 2.53 million tons by 2026, reflecting an impressive CAGR of approximately 49% [79]. As a result of this evolving market, bio-based materials are primarily utilized in the packaging sector [78], with significant applications also found in construction, textiles, agriculture, cosmetics, and medical uses [80]. As the demand for bio-based packaging continues to rise, the application of bio-based adhesives is also expected to increase significantly. These adhesives are commonly used to laminate various materials together, making them integral to the production of effective and sustainable packaging solutions [81]. In the realm of adhesives, lignin and cellulose serve as two notable examples of bio-based materials, valued for their excellent binding properties and the

ability to enhance the mechanical performance of adhesive formulations [82–85]. Beyond adhesives, bio-based materials are increasingly recognized for their potential as bio-based binders in battery technologies [86]. Examples of bio-based binders include polysaccharides such as sodium alginate [86,87], chitosan [87], xanthan gum [86,88], and tragacanth [87], as well as proteins like gelatin [87] or lignin [86]. These materials are increasingly investigated in the formulation of electrode materials for battery cells, offering promising performance while enhancing the sustainability of energy storage solutions [86]. This section will delve into the dispersing processes of these bio-based materials, focusing on their applications in adhesives and as binders for electrode materials in battery technologies. It will also discuss the gaps in the current research regarding the dispersing methods and process parameters, highlighting their role in determining the effectiveness of bio-based materials in these applications.

3.1. Lignin

Lignin is the second most abundant biopolymer on earth and is obtained from renewable raw materials such as grasses, trees, and plants [15,89,90]. Lignin serves as a natural adhesive in plants. It bonds cellulose and hemicellulose into a stable networking system that forms the mechanical backbone of the plant [91]. In land plants, lignin is present in cell walls at a percentage ranging from 15% to 30%, making it a cost-effective renewable resource [17]. It is particularly attractive as a raw material due to its availability, as lignin mainly arises as a by-product of the pulp and paper industry [74,92]. The structure and properties of lignin depend on the type of plant or tree used. Additionally, the pulping method used for extraction affects the molecular weight, functionality, and composition. With wood as a starting material, the sulfite process and the kraft process (sulfate process) are used. The former produces water-soluble lignosulfonates with a high sulfur content and the latter produces kraft lignin, which is only soluble in specific organic solvents. When using annual plants and plant residues, the soda pulping process is utilized, which is used for ethanol production. This type of pulping produces soda lignin, which is characterized by a sulfur-free composition and a low molecular weight [93,94]. Lignin exhibits different properties compared to biopolymers such as proteins, natural rubber, and polysaccharides [2]. Lignin does not have a linear but a branched structure, making it consist of a densely cross-linked aromatic network [2,15]. Lignin offers many desirable properties, including excellent thermal stability, biodegradability, high carbon content, antioxidant activity, and advantageous stiffness [17]. It has antifungal as well as antimicrobial activity and exhibits flame-retardant properties [95]. Moreover, lignin is poorly compatible with most solvents and decomposes without melting [2,96]. Given the complex structure and heterogeneous nature of the three-dimensional biopolymer, commercialization is only slowly feasible [27,97]. Less than 2% of the lignin produced in pulp and paper production is used for higher-value applications such as stabilizing and dispersing agents or concrete additives [17,94,98]. Most of the remaining lignin is usually simply disposed or predominantly used as fuel [17,99]. For this reason, there is increasing interest in expanding the added value of lignin and making it usable for higher-quality applications. Various approaches can be employed to increase the added value of lignin [27]. On the one hand, lignin is a potential raw material source for aromatic chemicals, especially phenols [100]. On the other hand, unmodified lignin can serve, for example, as a cost-effective filler component in plastic composites [77,95]. Lignin also has other beneficial properties, and it has been tested as a UV stabilizer and radical scavenger due to its aromatic structure [95,101,102]. A further increase in lignin value creation can be achieved through its derivatization [95,102]. Lignin is frequently chemically modified to enhance its dispersibility in polymer matrices or its miscibility in polymers. In addition,

chemical modification of lignin can, for example, improve the homogeneity and mechanical properties of blend polymers [103]. Moreover, modification can increase the reactivity and solubility of lignin in a reaction medium [103,104]. Both in modified and unmodified forms, lignin finds broad applications, from its use as a filler in plastic composites and its application as an antioxidant, dispersing agent, flame retardant, and for enhancing mechanical properties, to the development of bio-based adhesives, which are playing an increasingly important role.

Lignin-Based Adhesives

Adhesives are crucial as auxiliary materials in numerous industries and are widely used in sectors such as packaging and wood processing industries and medical, construction, and other fields [105–108]. Currently, for example, adhesives based on formaldehyde continue to dominate the wood processing industry [109,110]. Formaldehyde is a highly toxic chemical compound and is classified as carcinogenic to humans [111,112]. Given increasing environmental and health concerns, the focus is on researching formaldehyde-free adhesives derived from biomass resources [109,110,112]. Since lignin acts as a natural adhesive in plants, the use of lignin or modified lignin presents itself as a substitute for petroleum-based compounds in adhesives. For this reason, researchers have long been exploring the use of lignin as a raw material for the production of bio-based adhesives [91,104,112]. For example, there are more and more new formulations of wood adhesives based on lignin such as lignin–phenol–glyoxal, lignin–urea–glyoxal, and others [112]. Rahman et al. [113] report on dispersions of water-based polyurethane with amine-modified lignin, which was used as a chain extender. Nacas et al. [114] used unmodified kraft lignin in their work, which they converted with MDI (methylene diphenyl diisocyanate) in THF (tetrahydrofuran) to a solvent-based polyurethane adhesive. Lima et al. [115] also used unmodified lignins (organosolv and kraft lignins) in their polyurethane adhesives, where they served solely as fillers. In addition to polyurethane adhesives, the use of lignin in epoxy adhesives is also reported. Ferdosian et al. [116] produced lignin-based epoxy resins in their study, but using depolymerized organosolv and kraft lignin.

In the LignoGlue project of Fraunhofer WKI in cooperation with JOWAT SE and Synthopol Chemie, the aim was to develop and optimize lignin-based raw materials specifically for use in adhesives. The cohesive properties of the lignin should be transferred to the adhesive systems while preserving the polymer properties. In the project, lignin derivatives were used to produce polyurethane dispersions, as protective colloids for polyvinyl acetate-based dispersion adhesives, and for application in EPI adhesives (Emulsion Polymer Isocyanate). Additionally, lignin polyethers and polyesters were examined as raw materials in one-component polyurethane (1C PUR) and PUR hot melt systems, as well as in two-component polyurethane (2C PUR) adhesives. Among other things, the focus of the study was on investigating different lignin derivatives and their solids contents, as well as suitable solvents to produce stable dispersions. Furthermore, the long-term stability of the lignin-containing dispersions was examined. It was reported that after dispersing, effects such as gelation, a two-phase formation (liquid–liquid), or precipitation of solids were observed. To increase the stability of the dispersions, additives such as emulsifiers or other solvents like THF were used. More soluble lignin-protective colloid alternatives have been used in dispersion adhesives to counteract the poor water-solubility of the lignin modifications. However, this led to incompatibility and coagulation of the produced dispersion adhesives, resulting in insufficient dispersion stability. When producing EPI adhesives with lignin polyether derivatives as post-additives, it was described that dispersing the highly viscous materials using a conventional dissolver proved to be very difficult. To enable dispersion, the process temperature had to be increased significantly above 80 °C,

but this did not result in the production of a storage-stable adhesive, so phase separation occurred after a short time [27]. One potential approach to still use the poorly water-soluble modified lignin and to counteract the coagulation of the dispersion adhesives would be the use of various dispersing methods in combination with other additives. The variation of dispersing methods combined with the use of different additives could be tested to examine the possible increase in dispersion quality. A combination of different process parameters, mixing tools, and stabilizing additives could also be explored in the production of EPI adhesives to examine the effect on dispersion and storage stability. Since the focus in the LignoGlue project was on material selection, future studies could investigate the selection of a material combination focusing on multiple dispersing methods as well as process parameters. A focus on mixing methods in such applications has been scarcely investigated thus far and could be a potential approach to enhance dispersion quality without necessitating a change in formulation. The LignoGlue project itself also established that the dispersing process has an immense influence on the production and stability of emulsions. Originally, a dissolver was used for dispersing the adhesive formulations. The emulsions could be significantly optimized by adjusting the manufacturing method. For emulsion production, dispersion was carried out using an Ultra-Turrax based on the rotor–stator principle. To achieve long-term stability of the dispersion, suitable emulsifiers and stabilizers were used. In the course of the trials, the dispersion process could therefore be influenced or optimized by adjusting the viscosity, changing the dispersing devices, and varying the shear rates and temperature [27]. However, this approach was not further pursued for the other dispersion adhesives and consequently provides potential to investigate additional experiments focusing on the dispersing process.

The study by Lima et al. [115] investigated the influence of the dispersing method of lignins in 1C PUR adhesives. Lignins from organosolv and kraft origins were integrated into the adhesive formulation to examine effects on performance in wood construction applications. In the study, lignin was incorporated as an organic filler to increase the viscosity of the resin, limiting excessive penetration of the resin into the wood and consequently reducing glue line starvation. Lima et al. were able to determine that the dispersing method of the lignins was a key parameter in obtaining homogeneous systems with small particle sizes, resulting in an improvement in stability. For the experiments, 1–10 wt% of a dried lignin was mixed into the resin until every particle was wetted. The mixture was stirred using a mixer equipped with a Cowles blade. The particle size of some of the samples was then reduced by milling. This was achieved using a three-roll mill, with the dispersions being processed in three passes. During all passes, the roller speed was kept constant while the gap distance between the rollers was stepwise reduced. For quality assessment of the obtained resins with various lignin concentrations, the maximum particle size, particle size distribution, homogeneity, and viscosity of the dispersions were measured. In addition, the adhesive performance was tested. During the investigation of the influence of the dispersing method, Transmission Light Microscopy revealed that the lignin particle size significantly decreased when milling with a three-roll mill, which in turn led to a much more homogeneous dispersion (Figure 3) [115].

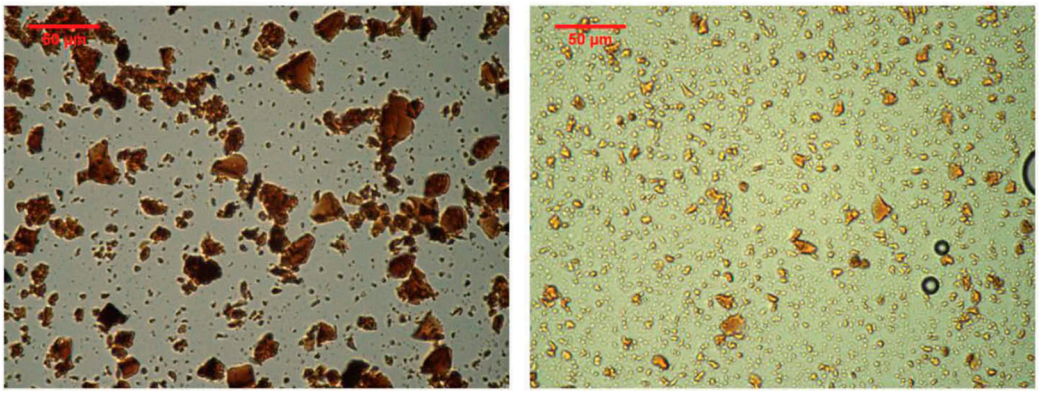


Figure 3. Optical microscopy images of dispersions with 10 wt% lignin content before (**left**) and after (**right**) three-roll milling. The progression of particle sizes during the milling process through the individual milling steps (first and second passes) is not shown, but the images clearly illustrate the significant reduction in particle size after three passes. Reproduced from [115], Copyright 2018, with permission from Taylor & Francis Ltd.

The reduction in particle size (and the associated increase in particle surface area) led, as expected, to an increase in viscosity. In the resins produced using a three-roll mill, high percentages of wood failure were observed, whereas in the dispersions made merely with a Cowles blade, no wood failure was detected at all. These results demonstrate the advantages of reducing lignin particle size using a three-roll mill in the final properties. Due to these benefits, the three-roll milling of dispersions was adopted as the standard method in subsequent experiments by Lima et al. [115]. The study by Lima et al. clearly shows that the processing method of bio-based materials can have a major impact on the properties of an adhesive.

The dispersing technology and thus the procedural consideration of a process opens great potential for overcoming limitations that may not be changeable through material adjustments. Most studies focus on the material combination or the formulation of bio-based adhesives, whereas the processing of bio-based materials has so far not been considered at all or only considered secondarily. As a result, it is of great importance to shed more light on the processing of bio-based materials in the future and to examine the influence of different dispersing methods and process parameters while maintaining consistent formulations. Dispersing technology could play a key role in, for example, enhancing the homogeneity and stability of bio-based adhesives, and enabling the incorporation of bio-based materials without having to make material changes that might necessitate the use of harmful solvents or other synthetic substances.

3.2. Polysaccharides

Among natural polymers, polysaccharides like starch and cellulose stand out as the most representative family [117]. They are present in most living organisms and constitute approximately 70% of the dry weight of total biomass [118]. As a result, they are readily and inexpensively available [117]. Polysaccharides can be obtained from plant or marine sources [117] or can be of microbial origin [118]. Polysaccharides are polymers made up of monosaccharide units. The monosaccharide units can consist of either five- or six-membered rings, with the latter being most common. The monomer units are linked together by glycosidic bonds. Polysaccharides can be both linear and branched. All polysaccharides are polydisperse, meaning they do not have a single molecular weight but vary within a range of molecular weights. Most polysaccharides are described as structurally complex because they are often polymolecular, which means that they differ in

their fine structure from molecule to molecule [118]. Some examples of polysaccharides are starch, cellulose, chitosan, pullulan, and dextrin, as well as gum arabic [117–119].

Cellulose is the main component of the cell wall in lignocellulosic plants [120]. With an estimated annual natural production of 1.5×10^{12} tons, cellulose stands as the most abundant organic polymer on earth, regarded as an almost inexhaustible raw material source [121]. The cellulose content depends on factors such as the plant species, the growing environment, and maturity [120]. Cellulose is a very large, linear polysaccharide consisting of unbranched β (1→4) linked D-glucopyranosyl units [117,118,120]. It is a crystalline polymer with high molecular weight, which cannot be fused or dissolved except in the strongest solvents that break hydrogen bonds. Hence, cellulose is usually converted into derivatives for better processability due to its infusibility and insolubility [117,120,121].

Cellulose-Based Adhesives

Research on developing polysaccharide-based adhesives is steadily increasing [119]. Alongside lignin, polysaccharides such as cellulose, starch, or chitosan have also aroused great interest for use in adhesives [122–124]. As one of the most abundant polysaccharides in nature, cellulose offers a wide range of advantages that make it a promising candidate for the production of adhesives [82,85]. Cellulose is an extremely versatile biomaterial due to its excellent mechanical properties, the possibility of chemical modification, and its abundant availability [82,84,85]. The inherent self-adhesive properties of cellulose render it a promising material in the field of adhesion science [84]. Its adhesion properties therefore render it an ideal biomaterial for the development of environmentally friendly adhesives and coatings [125]. These properties have attracted the attention of both research and industry as they have the potential to improve the performance and sustainability of adhesives [85].

For example, Jiang et al. [124] investigated adhesive blends of dicarboxylic acid cellulose nanofibrils (CNF) in commercial polyvinyl acetate (PVAc) and starch adhesives for wood joints. CNF suspensions with varying concentrations were incorporated into the adhesives and compared. To produce the PVAc and starch adhesives, the CNF suspensions were dispersed into the adhesives at room temperature using a high-performance homogenizer. The results showed that the addition of CNF significantly increased the strength of both adhesives, with a CNF concentration of 0.96% achieving the best performance. The use of CNF suspensions not only improved the strength of the adhesives but also enabled good bonding of wood with a smaller amount of commercial PVAc. Jiang et al. reported that in their experiments, a higher CNF concentration resulted in increased viscosities of the adhesives, which, in turn, limited the penetration ability into the wood surface and consequently led to reduced strength of the adhesives [124]. To counteract this, an alternative dispersing method or adjusted dispersing parameters could be tested in a follow-up study. This could attempt to reduce the viscosity of the adhesives at higher CNF concentrations, thus facilitating penetration into the wood surface and promoting uniform application. This could determine whether an increase in CNF concentration leads to further enhancement of adhesive strength due to the advantageous reinforcement mechanisms (cross-linking network formed by methylene groups of PVAc molecules and methyloyl groups of starch with hydroxyl groups of CNFs [124,126]) of CNFs.

In the study by Oh et al. [127], the use of cellulose nanofibers in wood adhesives based on plant proteins (zein and wheat gluten) also led to an improvement in adhesive strength. To prepare a 15 wt% solution, zein was dissolved in ethanol by stirring (300 rpm) for 30 min at 60 °C. The 15 wt% gluten solution was produced by dissolving gluten in aqueous urea with sodium sulfate as a reducing agent. The solution was stirred for 30 min at 400 rpm and 60 °C. Various concentrations of cellulose nanofiber suspensions were then

added to the protein-based solutions. The cellulose nanofiber suspensions were added to the zein and gluten adhesives and homogenized using an Ultra-Turrax homogenizer at 6000 rpm at room temperature for 5 min. The results of the study showed that the adhesive strength of zein and gluten adhesives could be significantly increased by adding cellulose nanofibers. Oh et al. also found that optimizing the performance of protein-based adhesives necessitates the utilization of various combinations of solvents, process conditions, and reinforcements [127].

In the study by Kaboorani et al. [128], nanocrystalline cellulose (NCC) was used to enhance the performance of PVAc as a wood adhesive. Different amounts of NCC suspension were incorporated into PVAc by mixing the materials for 30 min. It was discovered that NCC serves as an efficient nano-reinforcing material for PVAc, resulting in a substantial enhancement in the bond strength of wood joints. In addition, NCC improves the hardness, elastic modulus, and thermal stability of PVAc. Through atomic force microscopy (AFM) (Figure 4), it was observed that the addition of 2% and 3% NCC to the matrix resulted in surface reorganization. However, with only 1% NCC in the matrix, a smooth surface was obtained. The presence of 2% and 3% NCC indicated poor quality of NCC dispersion and consequently the formation of aggregates. This is due to apparently strong interactions between PVAc and NCC (hydrogen bonds) that prevent the even distribution of NCCs in the matrix. The 1% NCC (good dispersion quality) significantly increased the thermal stability and bonding strength of PVAc. A higher amount of NCC did not lead to further improvement of the properties. According to Kaboorani et al., the trend is related to the quality of the NCC dispersion. The agglomeration of NCC limits the extent of improvements achievable by NCC in the PVAc matrix. Since unmodified NCC has a hydrophilic character, a slight modification of the NCC surface could contribute to impart a hydrophobic character to prevent agglomeration at higher NCC contents, thereby achieving better NCC dispersion in the matrix [128].

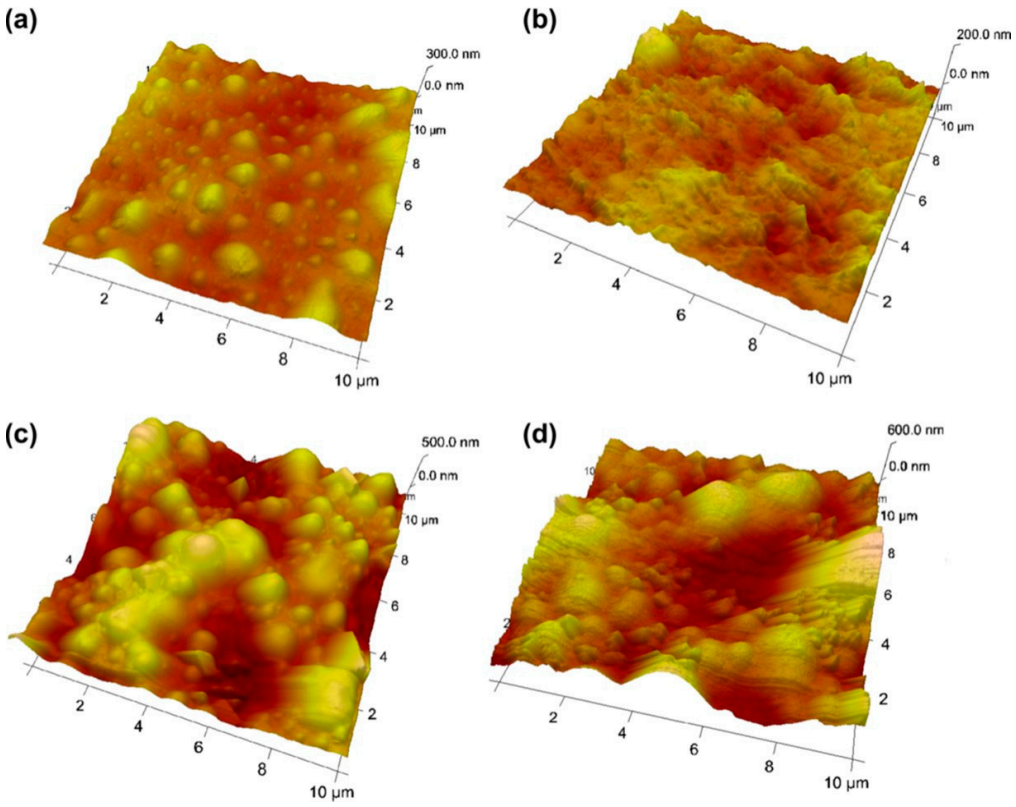


Figure 4. AFM images of PVAc films with (**a**) 0%, (**b**) 1%, (**c**) 2% und (**d**) 3% NCC. Reproduced from [128], Copyright 2012, with permission from Elsevier.

In the study by Kaboorani et al., it is not reported which dispersing methods and parameters are used for incorporating NCC into the PVAc matrix; only a mixing time of 30 min is mentioned. Instead of (or in combination with) NCC modification, a variation of the dispersing method and parameters could contribute to a more homogeneous distribution of NCCs in the matrix. Optimized dispersing could contribute to complete wetting of all fibers, thus minimizing or even preventing the agglomeration of NCCs. A possible method to improve the dispersion of NCC in the PVAc matrix could be the use of ultrasound. The use of ultrasonic waves could effectively distribute the NCC particles in the matrix by breaking up existing aggregates and achieving an even distribution. An alternative way to improve dispersion quality could be the use of mechanical stress. By applying shear forces or mixing devices with high shear intensity, a homogeneous distribution of NCC particles in the matrix can be promoted. Optimizing the dispersion parameters, such as the duration and intensity of mechanical stress, could also help to achieve improved dispersion. By varying the dispersing method, it could be tested whether a higher, homogeneously incorporated NCC content leads to further improvement in the end properties. Additionally, the combination of hydrophobic NCC modification and tailored dispersing parameters could significantly contribute to preventing agglomerate formation at higher NCC contents in the PVAc matrix. Modifying the NCC surface with hydrophobic groups could reduce the interactions between the NCC particles and the PVAc. This could result in improved wetting and distribution of the NCCs in the matrix. Combined with suitable dispersing parameters, this could lead to more homogeneous, stable dispersions.

Gabriel et al. [129] report in their study on the use of cellulose nanocrystals (CNCs) and carboxylated cellulose nanocrystals (cCNCs) in adhesives. For the adhesive formulations, cCNCs were added using four distinct methods to investigate the effect of CNC types (dry vs. wet form) and their dispersing methods on the end properties. To produce the first cCNC dispersion, the "wet" cCNCs were diluted in water and mixed using a magnetic stirrer. To prepare the second dispersion, the "wet, ultrasonicated" cCNCs were diluted in water, mixed using a magnetic stirrer, and ultrasonicated (ice bath, 75% amplitude, three intervals of 5 min, with 5 min of rest in between). In the third production method with the "dry dispersed" cCNCs, dry cCNCs were added to water and mixed using a magnetic stirrer for one hour, or until no visible aggregates were left. In the fourth incorporation method of cCNCs ("dry, ultrasonicated"), the dry cCNCs were incorporated similarly to the "dry dispersed" method, with subsequent application of three ultrasonication cycles [129]. A good CNC dispersion, meaning the optimal distribution of CNCs, is crucial for the final performance properties. With good dispersion of CNCs, their high aspect ratio and high surface-to-volume ratio come into play, making CNCs extremely effective for improving the properties of latex-based systems. Poor dispersion of the CNCs in water can lead to subsequent agglomeration in the latex, which ultimately affects the final performance [130]. In the study by Gabriel et al., it was found that cCNCs enhance both the adhesive and cohesive properties of the adhesives when incorporated in small amounts (0.5–1.0 wt%). The cCNCs showed greater improvements in properties (peel strength, adhesive strength, shear strength) when they were not treated with ultrasound. Even without ultrasound, the cCNCs could be sufficiently dispersed in water and maintain their network behavior (end-to-end and side-by-side interactions) [129]. Normally, the use of ultrasound is necessary to redisperse dry CNCs in water and ensure a homogeneous end product [130–132]. However, in this case, omitting the ultrasonic treatment of the cCNCs led to an enhancement in the end properties. By omitting the ultrasonication step, the "apparent" aspect ratios of the cCNCs were higher, which in turn positively affected the adhesion strength. The best results were achieved with the "wet" (not ultrasonicated) cCNCs, although the "dry" cCNCs were also high-performing. This is attributed to the general adhesion improvements due to the

presence of CNCs and their high aspect ratio [129]. The study shows that the dispersing method plays a crucial role in the incorporation of materials as it has a major impact on the homogeneity of the end product. While high shear forces and the use of ultrasound are often considered effective means of dispersion, they can also have a negative effect on incorporation. Nevertheless, in some cases, high shear forces are essential to incorporate materials homogeneously. Especially with bio-based materials, the dispersing method is of great importance, as it can influence not only the homogeneity but also the material properties. Hence, selecting appropriate dispersing methods and parameters is essential to achieve optimal results. In the study by Gabriel et al., it becomes evident that achieving the desired end properties depends not only on the material itself, but rather on the chosen dispersing method.

3.3. Bio-Based Binders for Electrode Materials in Battery Cell Production

The energy transition is one of the greatest challenges facing our and future generations. The need for renewable energy sources is well known and has long been seen as a possible way out of today's oil dependency [15,133]. Basically, the aim is transitioning from depleting fossil energy sources such as coal, oil, and gas to renewable energies such as wind and solar energy [16,134]. As intermittent energy sources are often generated in a decentralized manner, efficient and sustainable energy storage becomes crucial. In addition to large-scale solutions such as compressed air or hydropower, electrochemical energy storage is presently regarded as the most appropriate technology [16,135]. Modern electrochemical storage systems include, for example, rechargeable batteries, supercapacitors, or fuel cells. These usually consist of a cathode and an anode, an electrolyte, and a separator [136]. Among energy storage options, rechargeable batteries like lithium-ion batteries (LIBs) have achieved large commercial success [136,137]. The development of high-performance batteries utilizing renewable resources is of major importance for fulfilling the constantly increasing demand for energy (e.g., from consumer electronics or electric vehicles) and sustainability requirements [136,138,139]. There are also concerns about the impact that the manufacture and disposal of batteries could have on the environment. The polymer binder, crucial for electrode manufacture, is commonly perceived as an inert element, yet it significantly influences the cost, environmental friendliness, and recycling or disposal of these devices [16]. Binders in LIBs typically consist of polymers or polymer blends. The binder holds the active material and conductive additive particles together and is electrochemically stable in the battery's operating potential range (0–5 V). The binder is elastic and/or capable of withstanding substantial volume changes as well as ensuring adhesion to the current collectors [140]. State-of-the-art cathode processing for lithium-ion batteries is based on the use of fluorine-containing polymers such as poly(vinylidene difluoride) (PVDF) as a binder for electrode production [139,141]. PVDF is used to bond the active materials to each other and to adhere them to the aluminum foil [139,142]. Fluoropolymers are not only expensive but also require a toxic solvent such as N-methyl-2-pyrrolidone (NMP) as a solvent or dispersant. Switching to aqueous electrode processing methods and natural polymers as binders, as has already been realized with graphite-based lithium-ion anodes, for example, would be a significant step towards achieving ideally completely sustainable and eco-friendly electrochemical energy storage [16,140,143].

Among these more environmentally friendly processes, the combination of styrene-butadiene rubber (SBR) and carboxymethylcellulose (CMC) as binders emerges as a standout choice, having firmly established itself in the graphite anode production [144–146]. Prior to the introduction of water-based anode preparation, PVDV was also dispersed in NMP to produce graphite anodes, with PVDF being replaced by the more cost-effective binders CMC and SBR. Both additives are dispersible in water, eliminating the need for

the reproductive toxic solvent NMP [146–148]. In anode pastes, SBR and CMC are used for different purposes, but they are equally important [148]. While pure cellulose is insoluble in water due to strong intra- and intermolecular interactions via hydrogen bonding, cellulose derivatives such as cellulose esters or cellulose ethers can be water-soluble. As a result, these can be used in a range of technical applications. Cellulose derivatives such as CMC are typically used as stabilizers, emulsifiers, or binders in aqueous solutions [149]. When CMC is used as a binder in anode pastes, it acts to stabilize the dispersion through hydrophobic interactions between graphite particles and CMC. Graphite sedimentation is avoided by the role of CMC as a thickener and surface modifier [150]. In addition to aiding in dispersing graphite particles, CMC serves as a thickening agent to adjust the desired viscosity for the subsequent application of the slurry [148]. Despite CMC being a powerful binder and imparting shear-thinning behavior to the slurry, it has high stiffness and low elongation at break. This can affect overall mechanical properties and long-term performance [144,150]. Solely utilizing CMC fails to create mechanically stable high-mass-loading electrodes owing to its inherent brittleness [146,147].

The binder SBR counteracts this, prompting the usual combination with CMC for enhanced slurry mechanical properties on current collectors [144,148]. SBR influences the material properties and contributes to a denser and less brittle anode [148]. However, the exclusive use of SBR does not result in the formation of a stable graphite suspension. Consequently, the combination of CMC and SBR delays graphite sedimentation and forms denser, but less brittle electrodes [16].

SBR is a synthetic elastomer with a wide range of industrial applications [151,152]. Because of its outstanding abrasion resistance and stability, SBR is frequently incorporated into car tire formulations [152]. SBR can be synthesized from styrene and butadiene using two different manufacturing processes. On the one hand, it can be produced via emulsion polymerization. On the other hand, it can be synthesized via anionic polymerization in organic solvents. Emulsion polymerization has the advantage of being a more environmentally friendly process [153]. However, SBR is a synthetic polymer that forms carcinogenic compounds when degraded [148]. Furthermore, SBR is also based on petro-based polymers and is questionable (in terms of health in the production process) due to the educts 1,3-butadiene and styrene used in its manufacture [154–156]. Positive exposure–response relationships were found between butadiene and all types of leukemia in workers with simultaneous exposure to styrene. This further corroborates the IARC classification of butadiene as a recognized human carcinogen [156]. In its role as an elastomer, SBR improves bonding strength and adhesion, increases heat resistance, and enhances the overall flexibility of the electrode [146]. As a binder in electrode slurries, SBR is exposed to mechanical stress during the mixing and dispersing process. Knowledge of the mechanical and thermal stability of the materials used is very important, as these strongly influence the sequence as well as the mixing and dispersing task. A material-specific adjustment of the processing to avoid irreversible material damage is therefore necessary. SBR, for example, has an increased sensitivity to shear [157]. The use of fossil-based SBR in conjunction with CMC poses a limitation in the context of sustainable development and health security that still needs to be overcome when it comes to fully bio-based binders for anode slurries. To increase sustainability in LIB production and to reduce potential health damage, synthetic SBR (and PVDF) should be completely substituted by bio-based binders.

There is currently a considerable research focus and great industrial interest in exploring bio-based binders for LIBs. This results from the blend of low environmental impact, enhanced electrode processing, and a wide array of functional properties. Functional as well as polar surface groups like carboxylic acid and hydroxyl groups provide distinct surface functionality that enable various bonding interactions (van der Waals, hydrogen,

covalent) and thus contribute to interfacial strength [86,158]. Several prospective materials, including proteins and polysaccharides, have been explored to find a superior binder alternative. However, these have not been regarded as commercially viable binders on a large scale [148]. In addition to SBR with CMC, other water-soluble or aqueous processable binders in LIBs such as the protein gelatine or polysaccharides like sodium alginate, starch, chitosan, agar-agar, carrageenan, tragacanth, xanthan gum, and guar gum have been tested as promising substitutes for previous binder systems [16,86,87,159,160]. When comparing bio-based binders with PVDF or SBR with CMC, comparable or in some cases improved results regarding electrochemical stability and mechanical properties of the bio-based binders compared to the standard system have been observed [86,160].

Wang et al. [88] report in their study on the use of xanthan gum as a substitute for PVDF as a binder in graphite anodes for lithium-ion batteries. To produce the anode slurries, xanthan gum (XG) was dissolved in water and PVDF was dissolved in NMP. The slurries were applied to copper foils for mechanical and electrochemical investigations. The wear test showed that the xanthan gum film has higher elasticity than the PVDF film. SEM images (Figure 5) of PVDF/Cu and XG/Cu coatings were taken to examine the surface morphologies after scratch tests. The PVDF/Cu coating showed elasto-plastic deformation, followed by botch incomplete and complete cracking of the PVDF film, and eventually, the cracking of the Cu substrate. In contrast, the XG/Cu coating experienced elasto-plastic deformation of the XG film, leading to the simultaneous cracking of the XG/Cu coating. The XG film remains intact with only a scratch mark when the diamond tip passes over, indicating greater plastic deformation resistance compared to the PVDF binder [88].

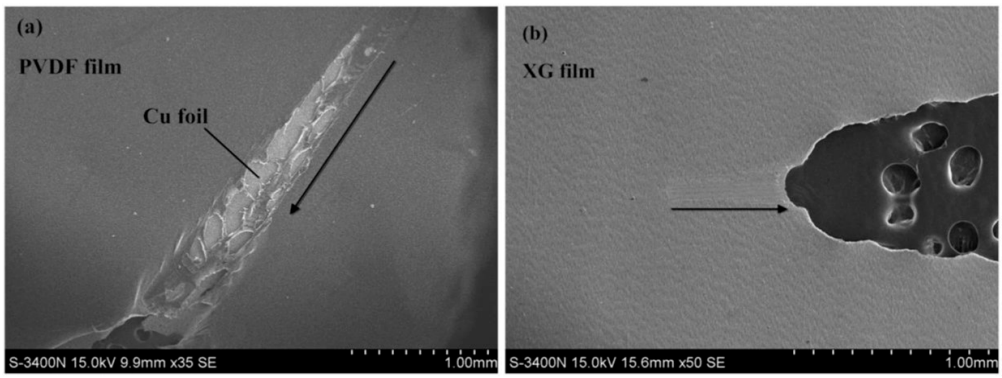


Figure 5. SEM images of the surface morphologies of PVDF/Cu (**a**) and XG/Cu (**b**) coatings after scratch tests. The arrow direction displays the scratch direction. Reproduced from [88], Copyright 2017, with permission from Elsevier.

Additionally, better adhesion strength was achieved between the xanthan binder and the copper foil substrate compared to the PVDF coating [88]. This behavior is attributed to the presence of functional groups such as carboxyl and hydroxyl groups in the xanthan gum binder, as these contribute to more active binding sites between the active material, conductive additive, and current collector. Moreover, the functional groups facilitate electronic and ionic conduction and ensure good dispersion or the realization of stable suspensions during slurry production [147,161]. In electrochemical studies, xanthan gum was found to have remarkable electrochemical performance in graphite electrodes. The initial coulombic efficiency, cycle stability, and rate capability were significantly improved when using xanthan gum compared to the conventional PVDF-based anode [88]. Binders with functional carboxyl and hydroxyl groups can contribute to enhance cycle performance or cycle life [162–164]. In addition, Electrochemical Impedance Spectroscopy (EIS) measurements have shown that xanthan gum in the graphite anode has reduced charge transfer resistance and heightened kinetic activity on the electrode/electrolyte interface compared

to PVDF. Wang et al. were able to demonstrate in their study that graphite electrodes using xanthan gum instead of PVDF as a binder can not only reduce costs and increase sustainability but also improve their electrochemical performance [88].

In a study by Versaci et al. [87] in 2017, the biopolymers sodium alginate, tragacanth, chitosan, and gelatin were investigated as alternative binders in graphite anodes. For comparison purposes, graphite electrodes were manufactured with the binders PVDF as well as SBR in combination with CMC. In addition to electrochemical characterizations, the rheological properties of the bio-based materials were also examined in comparison to the synthetic binders. Regarding industrial application, investigating the rheological behavior of the slurry is important to evaluate the suitability of these binders for industrial processing (slurry deposition). The formulation of the anode pastes contained 94 wt% active material, 2 wt% conductive additive, and 4 wt% binder. To produce the anode pastes, the aqueous processable binders were stirred in ultrapure water and PVDF in NMP for four hours at 50 °C in a sealed beaker to promote solubilization. The previously mixed active material (graphite) and conductive additive (carbon black) were slowly added to the binder solution. The mixture was then stirred for a further four hours. The slurries were applied to copper foil, subsequently dried, and assembled into EL-cells in an argon-filled glovebox. The electrochemical characterization showed that in the low current range, a comparable and, at high current rates, even an improved galvanostatic cycling performance was found in favor of the bio-based materials compared to PVDF and SBR in combination with CMC. However, the performance at high cycle numbers and high current was not satisfactory. Rheological measurements (Figure 6) revealed that SBR with CMC, sodium alginate and tragacanth exhibits shear-thinning behavior, indicating the slurry is flowable and the deposition on the substrate is homogeneous and without defects (such as stripes).

Gelatin (for chosen composition) appeared to exhibit shear-thickening behavior at higher shear rates, making it unsuitable for the coating process. Versaci et al. consider rheological testing as highly important in the exploration of the real possibilities for simple industrial upscaling, as it provides information about the deposition capability. The rheological behavior is crucial to whether a material can be used at the industrial level. Gelatin has proven to be a high-performing binder, but its rheological behavior under the tested conditions inhibits its use on an industrial scale. Further studies should investigate whether a change in the slurry formulation can achieve improvement [87]. Further work should be conducted, both regarding the slurry formulation and, more importantly, regarding the processing of bio-based materials. In the study by Versaci et al., both the binder solutions and anode slurries were prepared by very long stirring over a total period of eight hours. The very long mixing time could be shortened by varying the dispersing method (rotor–stator, dissolver, etc.) and process parameters (shear forces, temperature, etc.). Therefore, instead of changing the slurry formulation, the process parameters or dispersing methods for slurry production could be varied for investigation in subsequent studies. Variation in process control could lead to an improvement in rheological properties and consequently to an application at an industrial level without having to replace the binder material. To characterize the slurry, not only the viscosity but also the particle size distribution should be investigated depending on the process parameters.

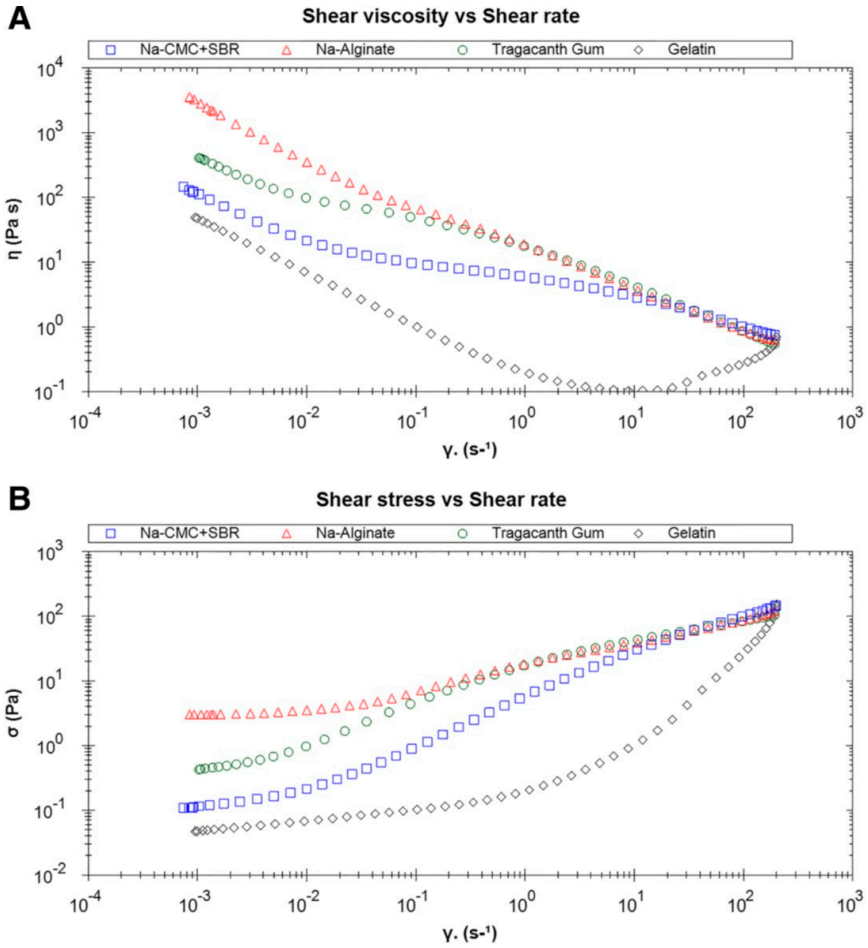


Figure 6. Evolution of the viscosity with the increment of the shear rate (**A**) and shear stress evolution as function of the shear rate (**B**) for Na-CMC + SBR, Na-Alginate, tragacanth gum, and gelatin. Reproduced from [87] under terms of CC BY license.

In a 2023 study, Versaci et al. [160] investigated tragacanth as an aqueous binder for a high-voltage lithium nickel manganese oxide cathode (LMNO). LMNO is an attractive cobalt-free cathode material for LIBs, offering benefits such as low cost, high energy density, and good cycle stability [165]. For comparison, the two conventional binders PVDF and CMC were also examined alongside tragacanth [160]. Tragacanth stands out as a highly attractive polysaccharide due to its abundant natural availability, cost efficiency, and ease of modification for various end uses [166–169]. Thanks to its abundant functional hydroxyl and carboxyl groups, tragacanth offers numerous possibilities for designing its macromolecular structure and architecture [168,169]. The polymer is biocompatible, biodegradable, and non-toxic to human health [166–169]. This heterogeneous polysaccharide comprises two main components, bassorin and tragacanthin, with bassorin making up 60–70% of the overall gum. Bassorin is water-insoluble but has enormous swelling capacity, enabling gel formation. In contrast, tragacanthin is the water-soluble part in tragacanth [166–171]. When tragacanth is added to water, the water-soluble part dissolves and forms a viscous solution, while the insoluble part swells and forms a soft, sticky gel structure that acts as a protective colloid and stabilizing agent [166,168,169]. Because of these characteristics, tragacanth is widely utilized as a natural gum and is frequently employed as a stabilizer, thickener, emulsifier, and adhesive or binding agent in many industrial sectors [166–168,170,171]. Versaci et al. only used a low binder content of 3 wt% in their work to meet the scale-up requirements. For slurry production, CMC and tragacanth were each dissolved in water, and PVDF in NMP. After preparing the binder solution, carbon black and the active ma-

terial were added. The cathode slurries were homogenized in a ball mill for 15 min at 30 Hz [160]. Versaci et al. found that the choice of binder affects the rheology of the slurry. Tragacanth showed good dispersibility in both hot and cold water, swelled, and formed a highly viscous solution [160,170]. Tragacanth therefore has the ability to alter the rheology of aqueous solutions even at low concentrations (2–3%) and exhibits a viscosity comparable to sodium alginate or CMC, as well as shear-thinning behavior [170–173]. As a result, it is assumed that the electrode slurry is deposited homogeneously and is well suited for industrial scale-up [160,174,175]. The cathode slurries were applied to carbon-coated aluminum foil, the electrode sheets were dried, cells were assembled, various characterizations were carried out, and the results were compared with those of the conventional binders PVDF and CMC. Tragacanth demonstrated high thermal stability up to 200 °C, rendering it appropriate for the typical operating range of LIBs. The low swelling of tragacanth in the electrolyte as well as the abundant hydroxyl and carboxyl groups resulted in better mechanical properties of the electrode. Furthermore, the LMNO-tragacanth electrodes exhibited stable electrochemical behavior as well as good lithium-ion insertion/extraction kinetics and cyclability. The most interesting result of the study concerns long-term stability, with the LNMO-tragacanth cathodes outperforming the PVDF and CMC-based electrodes. Tragacanth enabled improved cycle stability (capacity retention of more than 60% after 500 cycles), considerably exceeding the values achieved for the PVDF and CMC electrodes. After enduring over 1000 cycles at 1 C with a high cutoff voltage of 4.9 V, tragacanth maintained strong mechanical properties and minimal degradation of both the electrode and the complete system. Regarding the dispersing technology, exciting observations were also made concerning the microscopic images of the LMNO-PVDF surface (Figure 7) [160].

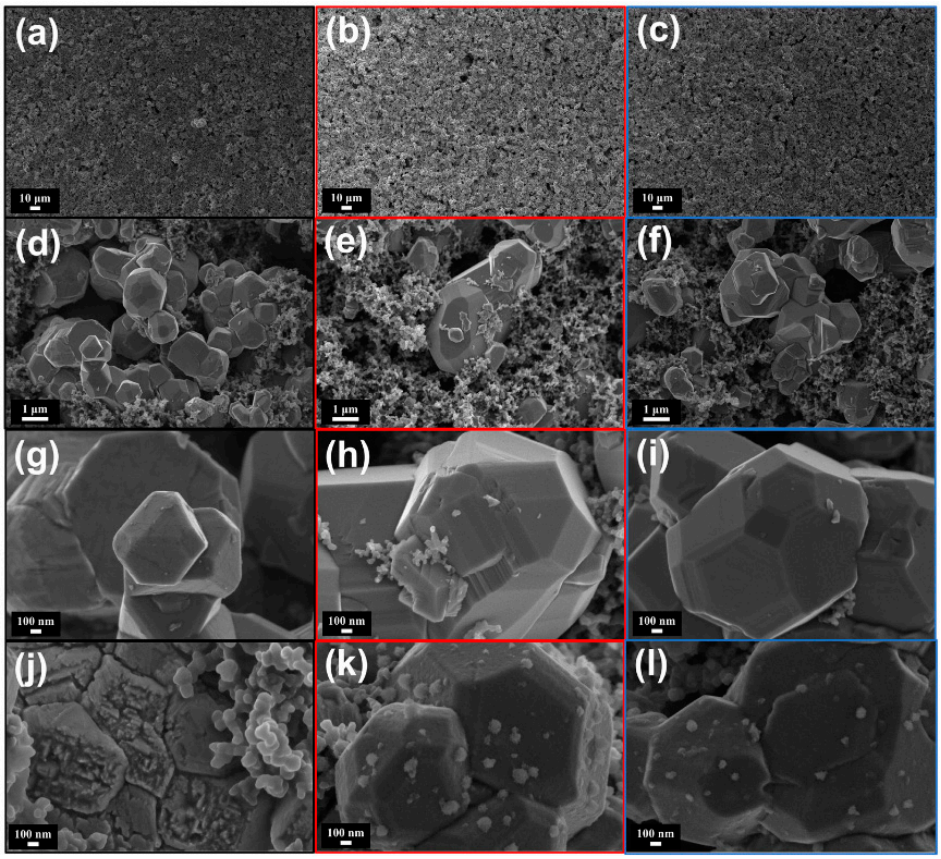


Figure 7. FE-SEM micrographs of pristine and aged cathode electrodes (for 1000 cycles) at different magnifications: (**a**,**d**,**g**) pristine LMNO-PVDF electrode; (**b**,**e**,**h**) pristine LMNO-CMC electrode; (**c**,**f**,**i**) pristine LMNO-TG electrode; (**j**) cycled LMNO-PVDF electrode; (**k**) cycled LMNO-CMC electrode; (**l**) cycled LMNO-TG electrode. Reproduced from [160] under terms of CC BY license.

When the images were magnified, LMNO-PVDF clearly showed microcracks on the LMNO particle surface. In contrast, this behavior was not observed for the CMC- and tragacanth-based electrodes, meaning that the type of binder can also affect the mixing process and morphology of the active material [160]. This finding highlights the importance of selecting the right dispersing methods and parameters in relation to bio-based and synthetic materials. The microscopic images show that the same dispersing procedure of different materials can have different effects on their end properties. It is therefore essential to adjust the dispersing parameters to the starting materials used—in this case synthetic vs. bio-based binders—to achieve the best dispersion quality and avoid damaging the dispersed particles.

4. Conclusions and Future Perspectives

The importance of environment and sustainability is continuously increasing in our society. Considering the challenges of climate change, resource consumption, and environmental pollution, there is an increased search for solutions to make our way of life more sustainable. Bio-based materials offer promising opportunities as replacements for synthetic materials to reduce our ecological footprint. The substitution of synthetic substances with bio-based alternatives offers numerous advantages such as conserving finite petroleum resources and reducing toxicity to humans and the environment. However, biomaterials cannot automatically be used as drop-in replacements for synthetic materials. They usually require an adjustment in handling and processing to fully exploit their potential. The development or investigation of suitable processing techniques and procedures is therefore of crucial importance to successfully expand the possible applications of biomaterials. Especially in the field of adhesives and batteries, bio-based materials show great potential. Bio-based adhesives—for example, based on lignin or cellulose—can represent a sustainable alternative to conventional, synthetic adhesives. They can be used in various industries, from the packaging industry to the medical sector, playing a notably important role in the wood adhesives sector. In battery technology, biomaterials such as polysaccharides or proteins can make an important contribution to replacing synthetic binders like SBR and PVDF (and the associated toxic solvent NMP), thus enhancing the sustainability of energy storage. So far, research has mainly focused on the development and optimization of biomaterials, particularly through modification. However, the handling and processing of these materials was mostly neglected or only given secondary consideration. There is a significant research gap in the development of processing techniques for bio-based materials. This gap is particularly evident in the lack of standardized methods and procedures tailored to the unique characteristics of biomaterials. Unlike synthetic materials, bio-based materials often present challenges such as variability in composition, sensitivity to processing conditions (shear force, temperature, etc.), and potential for degradation. These challenges necessitate a focused investigation into the dispersing processes specific to biomaterials. Addressing these challenges requires the development of specialized dispersing techniques that can maintain the integrity and functionality of bio-based materials. To achieve this, future research should explore specific experimental approaches. For instance, investigating the impact of varying shear forces in conjunction with other process parameters—such as mixing time, mixing temperature, and pH—on the dispersion of various bio-based materials across different applications will be essential to determine optimal processing conditions. Furthermore, it would be beneficial to implement systematic studies that evaluate the performance of various dispersing techniques, such as high-shear mixing, ultrasonication, and stirred media milling, and their effects on the structural integrity of bio-based materials. This could involve assessing parameters such as particle size distribution and viscosity changes during processing. In addition,

conducting computational fluid dynamics (CFD) simulations to model the flow behavior of bio-based materials during the dispersing process could significantly aid in designing more efficient mixing protocols [176]. Overall, these studies would enhance the understanding of how processing conditions influence material properties and performance, ultimately contributing to the development of more effective dispersing technologies. In light of these insights, it is crucial to focus on developing and adapting dispersing technologies capable of handling the unique properties of bio-based materials, as well as selecting the right process parameters to optimize their effectiveness and performance. This research gap offers scope for future investigations into process optimization to exploit the full potential of bio-based materials and overcome their occasionally disadvantageous properties without the need for chemical modification. Dispersing technology offers a promising solution to overcome the challenges and potential disadvantages of using bio-based materials and to fully exploit their advantages. By using suitable dispersing methods and parameters, biomaterials—adapted to their properties and requirements—can be processed homogeneously to increase their performance and consequently expand potential application areas as well as their use in the industry. Nevertheless, the current dispersing technologies need to be critically assessed and optimized for bio-based materials. This includes investigating the effects of different dispersing methods such as high-shear mixing, ultrasonication, stirred media milling, and roller milling on the structural integrity and performance of bio-based materials. Comparative studies between these methods could provide valuable insights into their efficacy and scalability. Additionally, it may be beneficial to develop new dispersing technologies specifically designed for the unique properties of bio-based materials, or to refine existing techniques and parameters to better adapt to these materials. The use of bio-based materials and the optimization of their processing techniques are important steps towards a more sustainable and environmentally friendly future. Targeted research and development of biomaterials and their processing can contribute to reducing environmental impacts and building a more sustainable economy based on renewable resources, free from reliance on finite resources.

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