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Review

Valorization of bioinspired adhesives via enhanced graft copolymerization for biomedicals: recent challenges, implications and potential solutions

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Abstract: Bioinspired adhesives, polymeric materials derived from renewable natural sources, offer sustainable and functional alternatives for bonding surfaces, especially in biomedical applications. While their use has fluctuated over the decades, rising environmental regulations and demands for biocompatible materials are renewing interest. In this review, we highlight recent advances in biomolecular adhesives and the pivotal role of graft copolymerization in enhancing their functional, mechanical, and biocompatibility properties. We focused on starch-based modifications, additive strategies, and nanofiller integration to address water resistance, viscosity stabilization, and adhesion enhancement. Key biomedical applications were examined alongside current engineering challenges and proposed solutions. This work provides a comprehensive framework for valorizing bioinspired adhesives to meet future clinical and environmental demands.

Keywords: Bioinspired adhesives; graft copolymerization; nanofillers integration; chemical modification; valorization.

1. Introduction

Bioinspired adhesive technologies are gaining increasing prominence due to the well-documented drawbacks of many conventional adhesives, including their toxic chemical composition, lack of adaptability under variable environmental conditions, and limited control over adhesion strength. For example, gecko-inspired adhesives are promising in minimally invasive surgery, offering more secure wound closure than traditional sutures and staples [1]. Similarly, surgical adhesives that mimic the adhesion strength and biocompatibility of mussel proteins are being explored to replace time-consuming, tissue-damaging sutures [2]. These polymeric bio-adhesives are also advancing the clinical viability of orthopedic and musculoskeletal tissue engineering [2].

The shift toward bio-adhesives is further propelled by rising environmental concerns. Using biological resources as alternatives to petroleum-derived materials contributes not only to reducing human health risks but also promotes biodegradability, leading to broader societal sustainability [3]. Thanks to their biocompatibility, abundance, and functional diversity, bio-based materials are now widely used in coatings, corrosion inhibitors, and drug delivery systems [4–8].

Adhesives, commonly known as glue, cement, or mucilage [9,10], are broadly classified into synthetic and natural types. Bio-inspired adhesives, unlike synthetic fossil-based ones, are more sustainable due to their biodegradability and eco-friendliness [11,12]. As illustrated in Figure 1, bio-adhesive innovation represents a potential breakthrough in creating environmentally responsible industrial practices.

From a chemical standpoint, adhesives are typically polymeric materials formed by crosslinking of monomers into macromolecules through a curing process. Examples include epoxy and cyanoacrylate adhesives formed via polymerization, and thermoplastic rubbers such as styrene-isoprene-styrene block copolymers [13,14].

The global adhesives industry, valued in the multibillion-dollar range, serves applications across biomedicine, packaging, electronics, textiles, shipbuilding, automotive, and aerospace [15–22]. Advanced adhesives are used for DNA recovery, bone fracture repair, wound sealing, and microvascular surgical procedures [23–25].

Achieving structural integrity in adhesives demands chemical modification, among which graft copolymerization has proven particularly effective. Grafting is a process initiated by radicals to introduce functional groups. By grafting synthetic polymers onto bio-based backbones, one can significantly improve adhesion strength, flexibility, and performance [26,27]. Nanomaterials such as nanocrystals and nanotubes further enhance the mechanical properties and bioactivity of adhesives due to their high surface area and nanoscale interactions [28,29].

Graft copolymerization enables the tailoring of adhesive behavior for moist biological environments. For instance, chitosan grafted with polyethylene glycol (PEG) has demonstrated strong, long-lasting tissue adhesion [30,31]. Moreover, these grafted systems can be functionalized with antimicrobial or drug-releasing groups [32,33].

As the field evolves, bioinspired adhesives are expected to play transformative roles in tissue engineering, wound care, and minimally invasive surgeries [34]. Their adaptability also opens possibilities for use in internal organ adhesives, drug delivery platforms, and regenerative scaffolds.

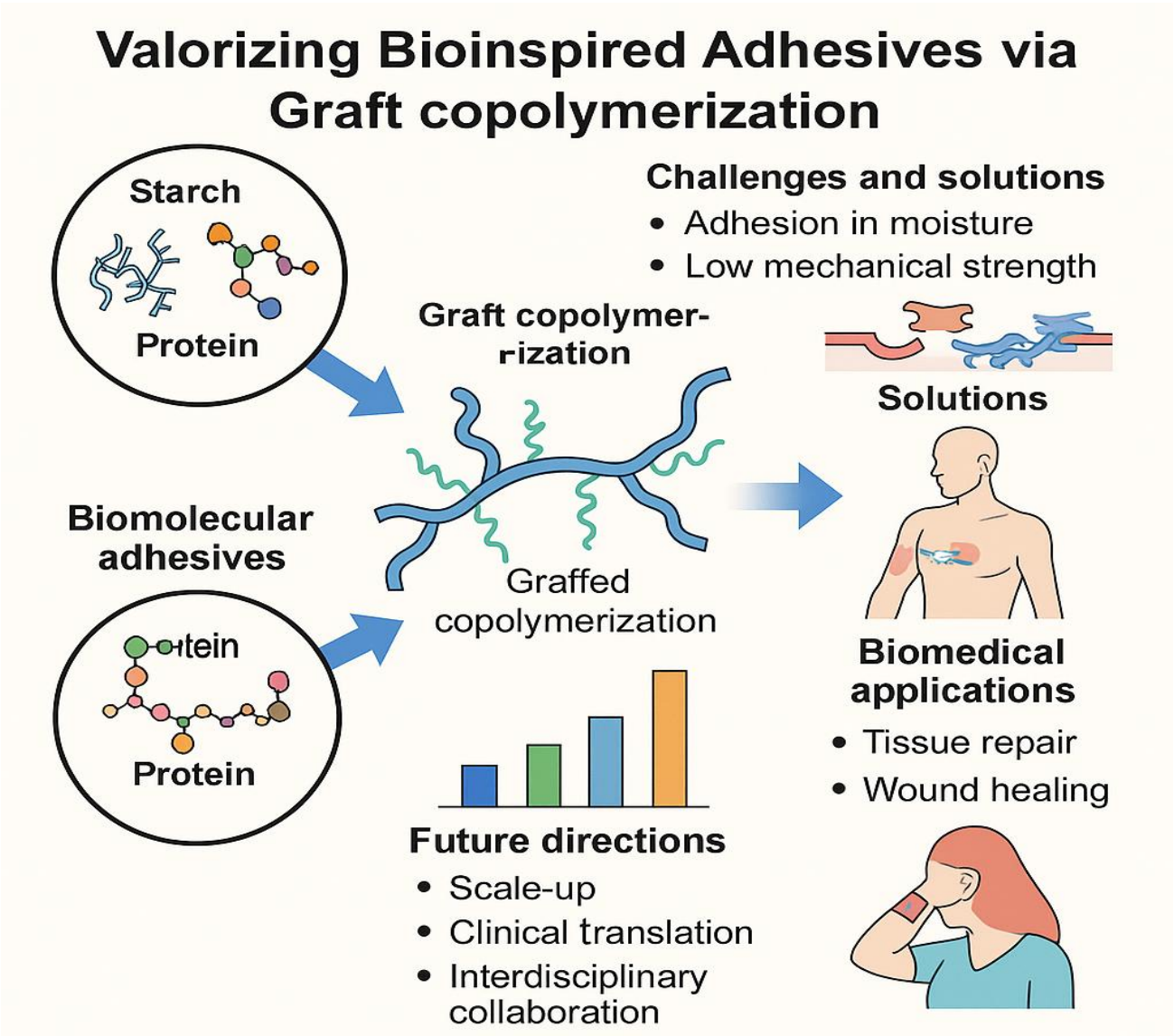

Figure 1. Visual illustration of the introduction.

Despite these advancements, literature lacks a focused synthesis on how graft copolymerization can systematically enhance bioinspired adhesives for biomedical applications. We address that critical gap, consolidating interdisciplinary insights into material design, functionalization, and application-driven innovations.

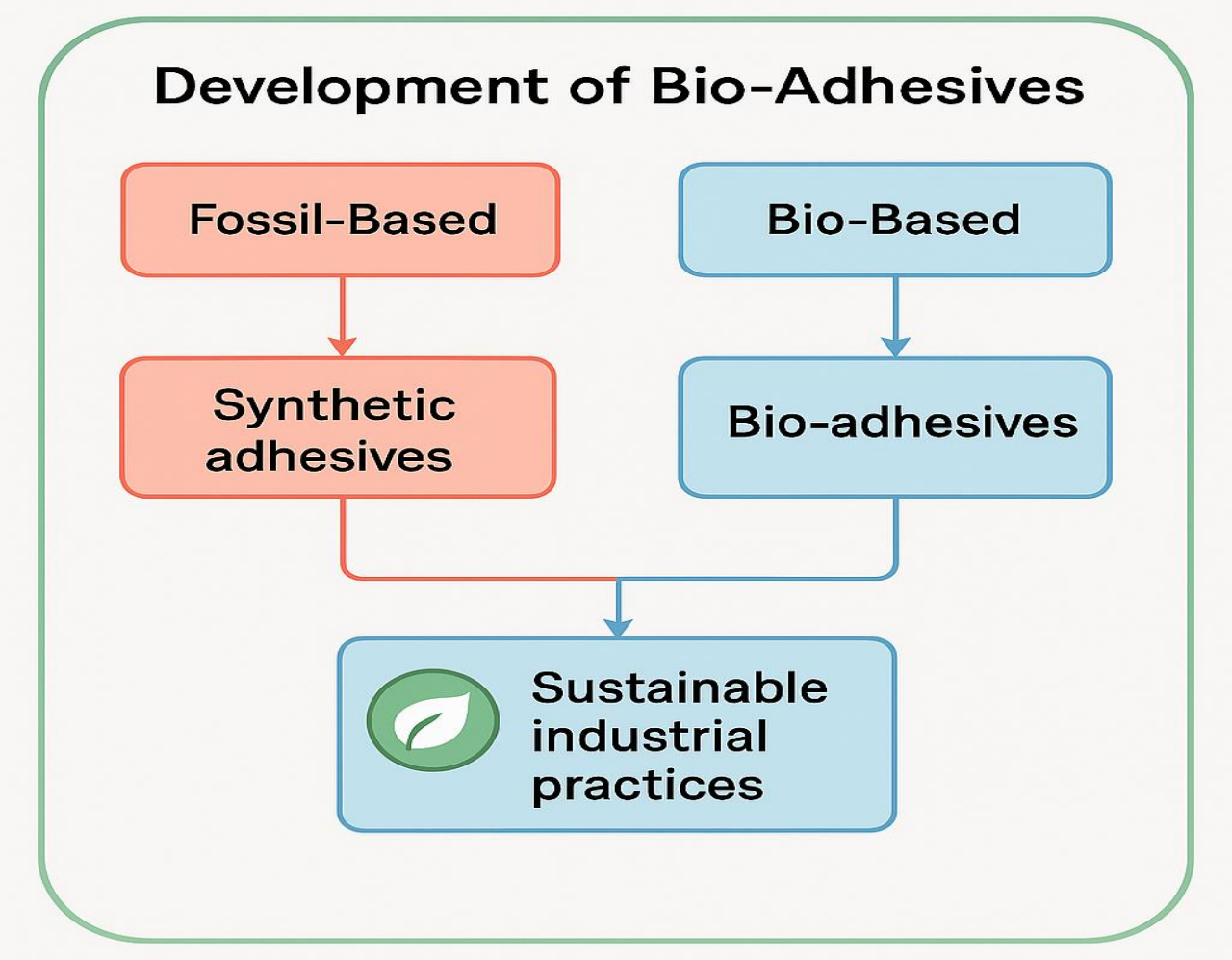

Figure 2. Transitioning from synthetic to bio-based adhesives for a greener future.

Figure 2 illustrates the evolving landscape of adhesive technology, highlighting the shift from traditional fossil-based synthetic adhesives toward bio-based alternatives. This transition emphasizes the potential of bio-adhesives to support environmentally responsible industrial practices. By integrating biodegradable materials and renewable resources, bio-adhesive innovation not only addresses ecological concerns but also aligns with global sustainability goals, positing as a transformative solution across multiple sectors.

2. Biomolecular adhesives

Adhesives can generally be grouped under three biomolecules, namely carbohydrates, proteins, and glycoproteins. Examples of carbohydrates are starch, cellulose, alginate, fucoidan, propolis, viscin, and sundew mucilage (Myo-inositol). In contrast, examples of protein include zein, egg albumen, fibrin, sandcastle worm, and different proteins from starfish, frogs, salamanders, glowworms, silkworms, velvet-worms, and mussels. Other molecules derived from snails, ivy, spiders, jellyfish, and hagfish are classified as glycoproteins [35].

Our ancestors deployed immemorial adhesives derived from animals, plants, and microorganisms for wound healing and surgical activities. However, the integrity and efficiency of these biomaterials

could not be guaranteed due to the inability to retain and sustain their properties when applied in a wetting condition. Moreover, toxicity was also a drawback of their applications. Many advances and ongoing investigations are targeted at solutions to these limitations. In this section, we discuss some selected bio-based adhesive systems.

Considering their numerous uses, ranging from tissue sealants, hemostatic supplies, and wound rehabilitation patches, as well as the development of adhesives, including fibrin binds, cyanoacrylate derivatives, and gelatine-based adhesives, has become a growing area of study in biomaterial research. Researchers have created a variety of polymeric mimics, including Chitosan-catechol and polyethyleneimine-catechol, among other similar catechol polymers, in response to the copious catecholamine found in mussel sticky proteins. One sticky polymer that shows promise for use in biomedical applications is chitosan-catechol. When catechol is conjugated onto chitosan, its solubility in pH 7 aqueous solutions increases significantly from 0 to over 60 mg/mL (6% w/v). The catecholamine's capacity to function similarly to mussel-sticky proteins is maximized by its increased solubility. Chitosan-catechol is employed extensively in a range of emerging medical scenarios since it is biocompatible and has good hemostatic and adhesion properties [34].

While these biomolecular systems, from carbohydrates to protein and glycoprotein-based adhesives, continue to inspire innovations in biomedical material design, their practical performance is often contrasted with conventional synthetic adhesives. A comparative assessment of bio-based and fossil-based adhesives is essential to understanding the benefits, limitations, and potential modifications required for broader industrial and clinical adoption. Table 1 below provides a summarized comparison, highlighting key differences in source materials, biocompatibility, mechanical performance, degradability, and application domains, thereby framing the context for further exploration of bioinspired adhesive engineering strategies.

Table 1. Comparison between bio-based and traditional adhesives.

Property	Bio-based adhesives	Traditional (synthetic) adhesives
Source	Derived from renewable resources such as starch, chitosan, and proteins [10,11]	Derived from non-renewable fossil fuels and petroleum derivatives [10]
Environmental impact	Biodegradable and eco-friendly; reduced ecological footprint [3,12]	Persistent in the environment; contributes to pollution [10]
Biocompatibility	High biocompatibility, suitable for medical use [2,5]	Typically, low biocompatibility; not ideal for biological applications [8,16]
Adhesion strength	Moderate to high; enhanced via graft copolymerization [27,31]	High adhesion strength; widely used in structural applications [17]
Processability	Requires chemical modification (e.g., grafting, plasticizers) [29,30]	Easily processed using established industrial techniques [13]
Mechanical properties	Tuneable through grafting and nanofillers [32,33]	Typically, strong and rigid [14,22]
Moisture resistance	Low to moderate; improved using hydrophobic additives [27]	Generally high moisture resistance [17]
Degradability	Biodegradable in physiological or environmental conditions [12]	Non-degradable; accumulates in ecosystems [10]
Cost and scalability	Currently more expensive but improving [6]	Lower cost due to mature industrial processes [16,19]
Application fields	Biomedical, packaging, coatings, and drug delivery [2,7]	Automotive, construction, electronics, and textiles [20,21,25,36]

2.1. Classification and sources of biomolecular adhesives

Numerous byproducts can be renewable assets to create environmentally friendly materials. These byproducts, detailed in Table 2, provide substitute sources of materials without interfering with the manufacture and usage of food. Renewable resource adhesives can also be made from a variety of bio-based substances or byproducts, including lignocellulosic biomass, proteins, tannins, and saccharides. Formaldehyde and bio-scavengers are being replaced with new hardeners and cross-linking agents as part of continuous efforts to lower formaldehyde releases [37,38]. A thorough literature survey is conducted in this current work, as demonstrated in Table 1, to provide an insight into the valorization of bioinspired adhesives, indicating the various authors' major findings, as well as the major raw materials and suitable applications.

Table 2. Summary of the major findings on the valorization of bioinspired adhesives.

Ref.	Raw materials	Overview	Major findings	Applications
[39]	Soy, palm, oilseed rape, sunflower, starch	Compares traditional petrochemical-based adhesives with bio-based adhesives to examine the effects of adhesive technology on the surroundings.	Industrial bio-based adhesives are theoretical and depend on newly developed goods and technology. The nascent industry for bio-based adhesives highlights the need for continuous development and research in this area.	Wood-Based Composites
[40]	Bioengineered proteins such as DOPA-rich ELP, recombinant ELPs, V40K72 fusion protein, recombinant spider silk, bovine serum albumin (BSA), soy protein, and fish swim bladder (FSG).	Discusses advancements in bioengineered protein-based adhesives designed for biomedical applications. The researchers discuss various strategies for fabricating these adhesives, including post-translational modification, chemical functionalization, and protein fusion techniques.	The adhesive made from bovine serum albumin (BSA) sticks to glass incredibly well; it even outperforms recombinant spider silk.	Wound Closure

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Ref.	Raw materials	Overview	Major findings	Applications
[36]	Porcine gelatin and human plasma (fibrin), dopamine-modified hyaluronic acid (HA), catechol-containing mussel threads, silk fibroin combined with tannic acid, and algae-mussel hydrogels.	Explains the four primary adhesion mechanisms and different methods for enhancing wet adhesion, frequently using natural adhesive systems as models.	Enhancing wet adhesion, which is vital for biomedical applications, can be achieved using biomimetic strategies that imitate natural adhesives like those of mussels, utilizing mechanisms such as mechanical interlocking and intermolecular bonding at the bio-adhesive-tissue interface	Tissue repair and regeneration, biomineralizatio n processes, epidermal bioelectronics, and hemostasis
[41]	Specifically, plant-derived tannins and animal-derived gelatin (later replaced) or ovalbumin, with the final version utilizing ε-poly-L-lysine (EPL).	From combining plant-based tannins with animal-based gelatin or ovalbumin, the study advanced to concentrating on a two-component adhesive made of tannin and ε-poly-L-lysine (EPL), which demonstrated a high bonding strength and water resistance.	Although it lacked sufficient water resistance, a tannin and gelatin mixture showed good storage and adhesive strength. The tannin-EPL adhesive that was created by substituting EPL for gelatin significantly increased the adhesive's water resistance, enabling it to satisfy the Euronorm EN 314 class 1 standard. It is non-toxic, eco-friendly, and has the potential to be a secure and reliable bonding agent.	Wood, glass, and iron surfaces.
[38]	Lignocellulosic compounds, tannins, plant- or animal-based proteins, saccharides, starch, lignin, and soy.	Discussed the health and environmental issues associated with traditional formaldehyde-based adhesives, reviewed current developments in bio-based and formaldehyde-free adhesives, and illustrated potential strategies to enhance bio-adhesive qualities.	Life cycle assessment (LCA) studies indicated that bio-based adhesives from other sources perform better environmentally in WBCP manufacturing than petrochemical-based adhesives, signifying that methodological variations can produce inconsistent outcomes.	Wood-composite panel (WCP) industry; products such as particleboard, medium-density fiberboard, oriented strand board, and plywood.

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Ref.	Raw materials	Overview	Major findings	Applications
[36]	Temperature-sensitive polymers like pNIPAM are grafted onto polyelectrolytes. Functional groups, such as catechols, are incorporated after being inspired by the natural adhesives found in creatures like sandcastle worms and mussels.	Addresses the use of biomimetic concepts in the development of adhesive hydrogels for medical applications, emphasizing the intricacy of natural adhesion, which frequently incorporates several elements, such as coacervation and sticky chemical groups.	By simulating natural processes like electrostatic forces followed by catechol-mediated covalent cross-linking, bio-inspired hydrogels can achieve high underwater adhesion.	Underwater tissue sealing and repair.
[42]	polysaccharides (e.g., alginate, starch, cellulose derivatives) and polypeptides/proteins (e.g., collagen, blood, vegetable proteins).	It classifies plant and animal bio-glues based on their chemical composition, adhesives derived from carbohydrates, proteins, and glycoproteins, as well as their potential for biomedical applications.	Despite natural compounds having the potential for medicinal applications, further research is often required to determine their efficacy and compatibility with living things cum more comparison studies of various bio-adhesives under comparable circumstances.	General tissue bonding to specialized uses, such as preventing leakage and attaching medical sensors.
[43]	Poly (3,4-dihydroxyphenylalanine) (PDA), chitosan (CHS), hyaluronic acid (HA), collagen, alginate, and silk fibroin,	Materials are created to resemble biological systems for therapeutic purposes, including bioinspired polymers and their uses in biomedical and regenerative medicine.	The bioactivity and therapeutics of these compounds in biological contexts were emphasized, including instances such as HA hydrogels with antibacterial qualities and CHS-PDA@EGF improving wound healing.	Tissue engineering, regenerative medicine, or wound healing

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Ref.	Raw materials	Overview	Major findings	Applications
[44]	Draw inspiration from mussel adhesive proteins, particularly utilizing catechol groups (e.g., DOPA). It also covers nucleic acid-containing polymers, hydrogels, and polypeptide sequences.	The researchers focus on summarizing supramolecular adhesives inspired by nucleic acids and proteins, such as mussel sticky proteins.	Using catechol chemistry, adhesives inspired by mussels exhibit strong adherence to a variety of surfaces, even under damp conditions. Adhesion is achieved by precise base pairing and other chemical interactions with adjustable qualities in nucleic acid-based adhesives.	Engineering tissue, biomedical devices, wearable sensors, skin regeneration, and biomedical sensors and electrodes.
[45]	Glycerol Sebacate Acrylate (PGSA) is a linear polymer. The molds for the nanopatterns are silicon templates made using microfabrication processes such as reactive ion etching and photolithography.	In addition to showcasing a production technique that avoids harsh chemicals and high temperatures, the study examines the impact of gecko-inspired nanopattern parameters, specifically pillar diameter and height, on the adhesive capabilities of the produced polymer.	When exposed to shear stresses under laboratory circumstances, nanopatterned PGSA nearly doubled the strength of a flat, unpatterned polymer and showed noticeably greater adherence to pig intestinal mucosa.	Wound closures plus tissue repair applications.
[2]	proteins such as collagen, fibrin, albumin, silk fibroin, and elastin; polysaccharides such as hyaluronic acid, alginate, chitosan, cellulose, and dextran crosslinked with micro/nano inorganic filler or tannic acid	It covers significant advancements since 2018 in genetically engineered natural adhesives based on proteins and polysaccharides, including commercially available alternatives and recently created multifunctional systems for biomedical applications.	Compared to single-component adhesives, composite adhesives composed of various natural polymers may have superior mechanical strength and stickiness. Interestingly, compared to collagen-based adhesives, an albumin-based adhesive made by gently heating ascorbic acid showed superior adherence to wood and aluminum.	Wound closures, drug delivery systems, tissue sealing, tissue engineering scaffolds, and orthopedics.

The widespread usage of adhesive molecules, with their numerous classification techniques, has been developed by variations in the components' origins, application techniques, and used chemistry [46]. The most certain classification depends on the strength of the created joints [17]. This technique distinguishes three primary types of adhesives: Semi-structural glues (which are further subdivided into solvent-driven and holt melt groups that can sustain a small load ranging from 0.3–3 MPa for an extended period), pressure-sensitive adhesives (which have a limited adhesive strength), and functional adhesives, which can bear a significant load greater than 7 MPa for a long time [46,47]. However, these adhesive materials are driven by highly variable prices, and petroleum is used to make the products. Adhesive manufacturers confront the challenge of being more sustainable due to heightened public concerns about the impacts of industrialized society on the environment. Sustainable adhesive formation and development include devising and designing adhesive processes and products that can minimize or lessen the manufacture of dangerous materials and reduce the energy and ecological impact of products [48–50]. Naturally based Sodium silicates, natural gums and resins, mammal glues, casein glues, and vegetable glues are examples of adhesives [13]. Plant glues are made from starches and dextrin and depend on starch.

A bonding agent is a substance that can bind solid things jointly via surface adhesion; thus, adhesives are substances used to glue or bond two surfaces together. Adhesives usually work by either the creation of primary covalent connections to the surface or interacting with the surface through physical forces called secondary bonds [51,52].

The effects of adhesion contribute to the mechanical durability of an adhesive-based assembly, also referred to as an adhesive junction, but the strength of the polymer materials used to synthesize or prepare the glue is fundamentally responsible. The mechanical features of the polymers employed to formulate adhesives are, therefore, consistent with the degree of strength built into adhesive junctions. Adhesives are ordinarily classified by their practical usage: Solvent-based adhesives, structural adhesives, Rubber-based adhesives, reactive adhesives, latex adhesives, pressure-sensitive bonds, and hot melt glue [53].

Generally, adhesive possesses a lower density than mechanical fasteners; hence, it is possible to lose pounds. Adhesives built around polymers are distinguished by their nature and can, therefore, absorb energy. This energy absorption shows up as a reduction in vibration and an enhancement in the joint's fatigue resistance. Additionally, adhesives might be utilized to combine or join materials that are electrochemically distinct and offer a junction that resists corrosion.

It seems that the function of adhesives obtained from renewable sources is hampered and hindered by their lack of structural integrity, despite their promising environmentally benign characteristics. Before the development of adhesives produced from petrochemicals, including urea-formaldehyde, phenol-formaldehyde, etc., researchers have shown interest in biomaterials, and studies are on the way to see how synergistic effects can be fabricated for enhanced performance, with one of the potential areas being the incorporation of nanofillers.

It looks like the technologies under review are in the pre-commercial, pilot, or lab stages. The implementation of formaldehyde release rules by the California Air Resources Board (CARB) and other so-called “Green” construction efforts is the primary driver of the new tech, particularly in North America. The decrease of volatile organic compounds (VOCs) seems to be the problem for these endeavors [54]. Comparable market factors are expected to grow increasingly significant in African markets. The demand for adhesives in most African nations for diverse uses, such as woodwork and paper packaging, is enormous. That means several kinds of adhesives may be developed to cater to the

demands of final consumers. This path will then curb the ominous greater percentage of the nation's adhesive while a small portion of the supplies are produced domestically; most of the nation's adhesive demands are now satisfied by imports. Therefore, exploring and harnessing bioresources or utilizing local content is a renewable path to tread: A win-win endeavor.

2.1.1. Animal glue

An adhesive that is obtained by an extended boiling of animal connective tissues is termed animal glue. Apart from its application as an adhesive, it can be used for bio-coating systems, sizing in decorative materials, and as a clarifying agent.

These colloid glues are derived via the hydrolysis of collagen from renewable sources such as hides, bones, tendons, skins, and other tissues. Animal glue is a general term used to describe glues composed of collagen, the primary amino acid colloid of skin, muscle, and bone. Collagen is a dispersed constituent when it is treated with hot water, acids, or alkalis. The low-molecular-weight substance produced by a more vigorous conversion process is usually less natural and more intense in color and is known as animal glue; in the event that the starting protein is unadulterated, and its transformation is slight, the high-molecular-weight item is called gelatine and may be applied for food or photographic products.

Animal glue has long been used for a variety of tasks, including making sandpaper, woodworking, bookbinding, and making extensively gummed tapes. Although it possesses the advantage of stickiness, these protein adhesives have been completely replaced or altered by synthetic glue owing to their poor performance, hence the need for developmental studies. For example, Agbogo et al. [55] did an intensive modification of animal glue based on esterification and found that there was an improved performance.

Skin, ligaments, tendons, and bones can be boiled in water to produce gelatin, a protein. Depending on the physical and chemical denaturation techniques, this form of collagen is permanently hydrolyzed, reducing protein fibrils into smaller peptides. It is sticky when wet and brittle when dry. Both starch and gelatin are naturally occurring polymers that can be utilized to make adhesives. In tandem with the work conducted by Tang et al. [56], gelatin contains a polypeptide chain that usually undergoes a transition state to form a gel, as shown in Figure 3. Apart from hydrogen bonds, electrostatic as well as hydrophobic relationships are what provide the triple helix found in the gel.

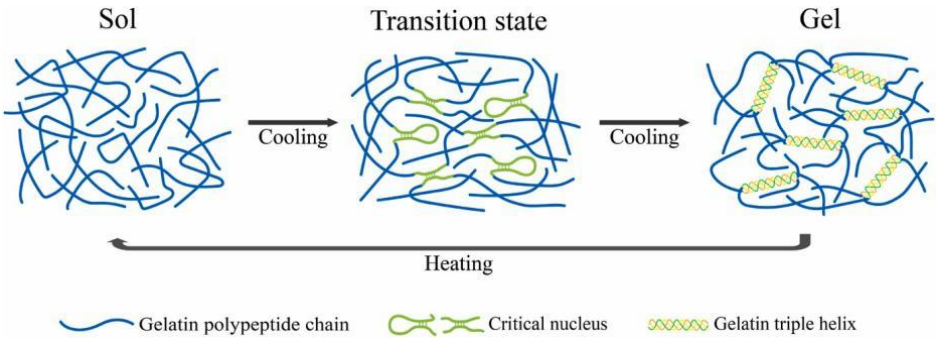

Figure 3. The Figure shows the gel formation process of gelatin. The transition from sol to gel is temperature-dependent, adapted from [56]. Reproduced with the copyright permission of the publisher, 2022.

2.1.2. Casein glue

Casein glue is created by dissolving the milk protein casein in an alkaline solution. The type and the level of the concentration of alkali have an impact on the behavior of the casein product. When it comes to woodwork, casein glues are a good option compared to animal glue owing to their moisture resistance characteristic; however, when it comes to bonding involving disassembly, animal glue may be preferred, given that they are reversible and easily changed by heat. Casein is employed to increase the adhering features of paints and even coatings.

Casein Gels have advantageous characteristic behaviors such as stability, mechanical strength, bio-compatibility, electrical conductivity, adhesion, sensing capacities, and medication delivery, that is, the controlled release of pharmaceuticals. These characteristics can be ascribed to their modification with various polymers and gelation procedures [57,58].

The casein molecules consist of a nanostructure, and the extensive work of Yang et al. [59] presents a nanocluster structure model that provides a robust insight into the composition of casein. As displayed in Figure 4, based on their reports, The Sub-micelle concept put out by Schmidt is displayed in A(i). The micelle model put out by Horne is interpreted in A(ii), whereas Holt's model for the casein micelle is represented in B, and C shows the dual binding model proposed by Horne.

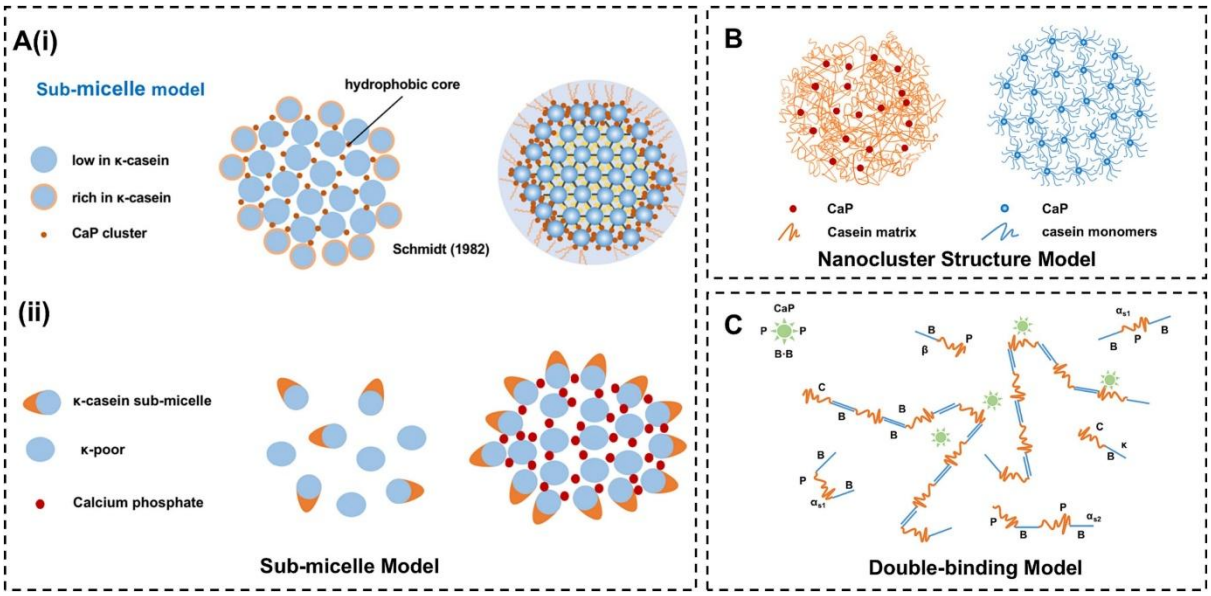

Figure 4. Casein micelle concepts adapted from the extensive work in [59]. Reproduced with the copyright permission of the producer. Elsevier, 2024.

2.1.3. Blood albumen glue

Blood albumen is applied in much the same way as casein adhesives [60]. The protein-based products from slaughtered animal blood are precipitated, dried, and obtained as powders, which can be cross-linked with water, sodium hydroxide, or hydrated lime, depending on the end use. When blood proteins are subjected to a certain heat, coagulation occurs, such that they are hardened utilizing the hot-pressing technique via loss of water [60,61]. Usually, introducing alkali to albumen-water mixtures

can help to improve or reinforce adhesive characteristic properties. Blood glue derivatives are applied in plywood production, biomedical engineering, and as a new material for conjunctival graft surgery [30,62].

2.1.4. Starch and dextrin

Starch and dextrin are usually the primary types of plant-based adhesives that are water-soluble and are derived from plant sources worldwide. They are typically extracted from foods such as corn, wheat, potatoes, or rice and serve as wallpaper glue, on corrugated boards, and in packaging. Starch has several benefits as a resource, such as being abundant, inexpensive, biodegradable, renewable, and stable in price. Starch adhesives are typically re-wettable, have little to no taste or odor, and can be applied at room temperature or a somewhat low temperature. A carbohydrate called starch comprises many glucose molecules connected by glycosidic linkages. Owing to eco-friendly properties and the enormous potential of adhesive properties inherent in starch molecules, starch molecules have been the subject of extensive research [63–70]. Moreover, this potential is improved through modification techniques, as illustrated below in Figure 5, to meet the demands of certain applications.

To optimize the inherent adhesive capabilities of starch-based systems, various modification strategies, most notably graft copolymerization, have been employed to enhance performance characteristics such as water resistance, mechanical strength, and substrate adhesion. Figure 5 schematically illustrates how polymer chains are grafted into starch molecules, resulting in improved adhesive behavior suitable for advanced industrial and biomedical applications.

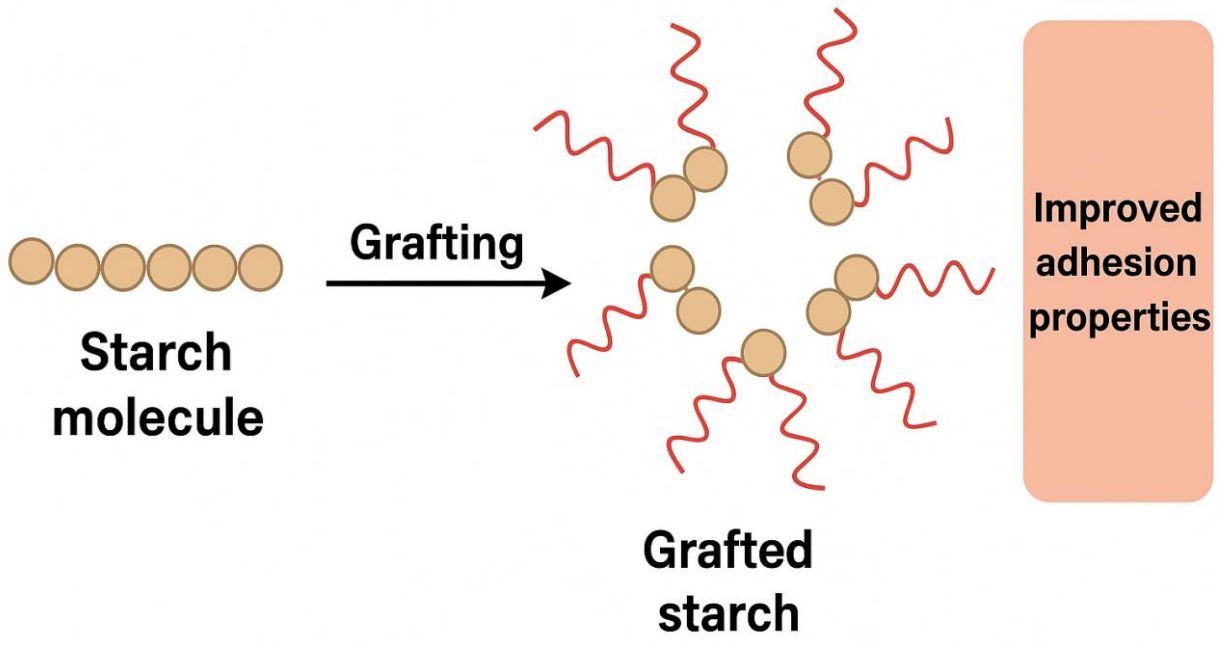

Figure 5. Schematic representation of graft copolymerization on starch molecules. Native starch chains (left) are modified by grafting synthetic polymer chains (right), leading to enhanced adhesion properties such as increased bonding strength, water resistance, and mechanical durability; these are key attributes for industrial and biomedical adhesive applications.

2.1.5. Natural gums

Gums are compounds taken from their natural sources that can be used as sticky substances. Hot water removes the aquatic plant colloid agar, which is subsequently frozen to purify it. Seaweed is digested in alkali to create algin, which is subsequently separated with calcium salt or alginic acid. Acacia trees artificially injured to release their gum are used to make Arabic gum. Water-remoistenable materials are the primary use for most gums. Microorganisms, plants, aquatic organisms, and other animals are some of the many sources of natural gums [71]. Although different gum exudates have different extraction and purification techniques; however, Hamdani et al., [72] has presented a general flow chart for these processes as shown in Figure 6.

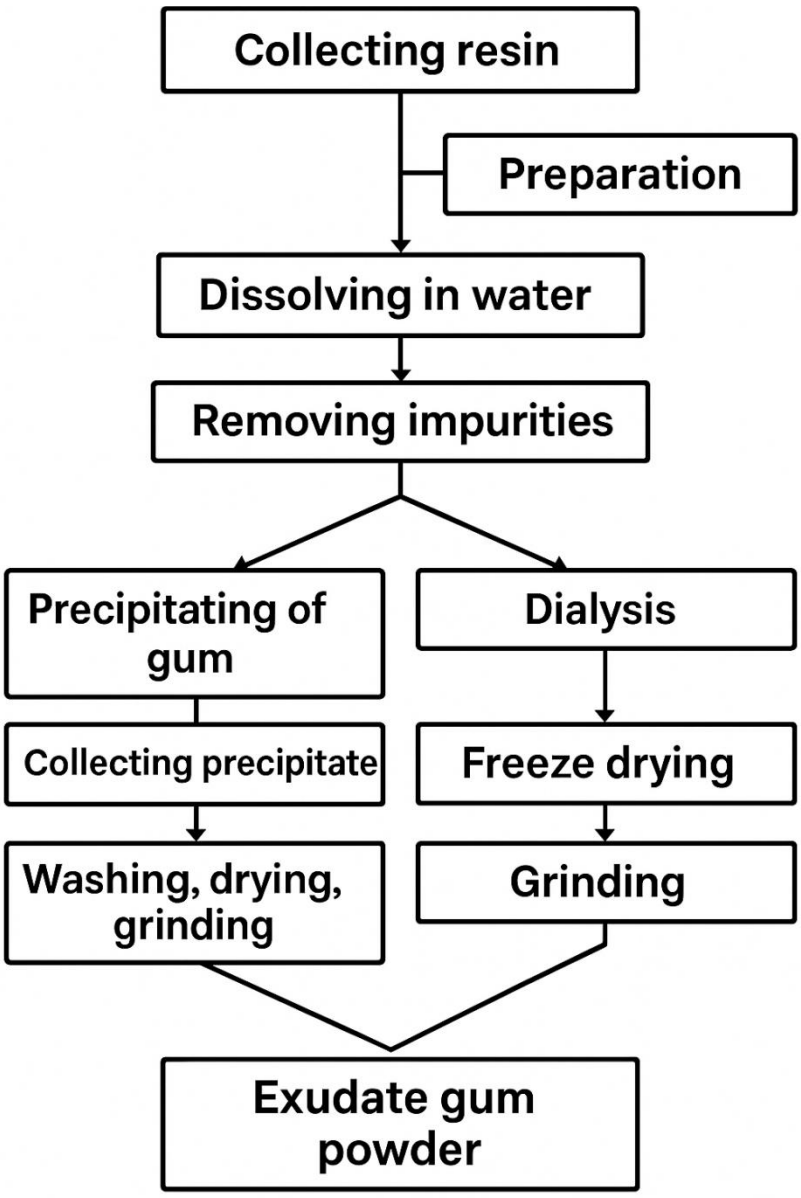

Figure 6. Flow diagram showing the major procedures for processing and purifying exudate gums [72]. Reproduced with the permission of the publisher, 2023.

3. Chemistry of biobased adhesives

Adhesives derived from plants and animals are composed of a few significant components. The multi-component structure accounts for their uniqueness and surface-interaction capabilities, given their certain functionality and chemical complementarity. Interestingly, even though covalent interactions can establish high surface specificity and strong binding affinity, the workings of adhesives that occur naturally are controlled by supramolecular non-covalent reactions such as hydrogen bonds, van der Waals forces, and hydrophobic and electrostatic forces. Other noncovalent reactions like cation π complexation, π-π interactions, and metal coordination work in concert with the covalent and noncovalent relationships [13].

The amino, primary, and secondary hydroxyl active groups present in adhesives also enable chemical modification in polysaccharides, e.g., chitosan. Quaternization, graft modification, acylation, carboxylation, alkylation, Schiff base modification, and crosslinking are a few of the chemical changes that can take place, as sharply demonstrated in Figure 7.

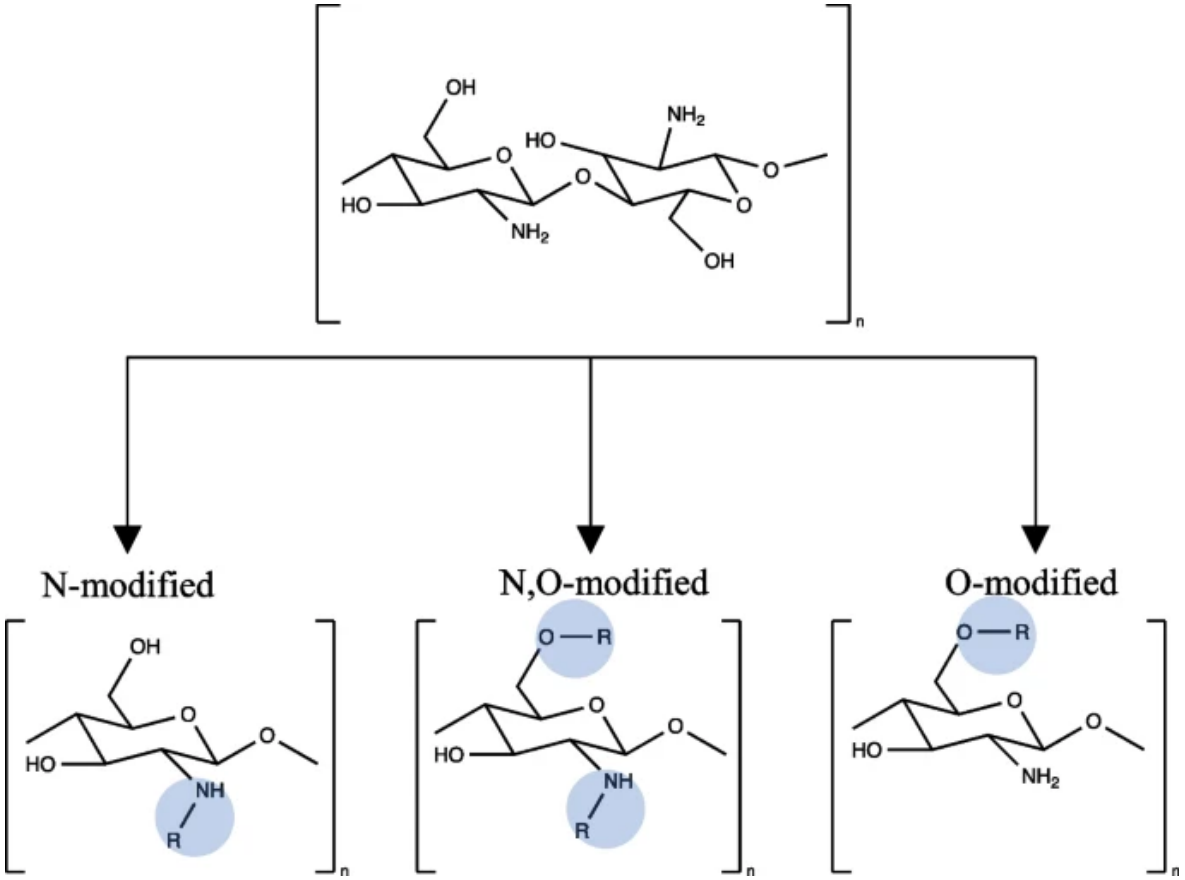

Figure 7. Chitosan's structure and molecular changes caused by amino groups and primary and secondary hydroxyl groups at locations C-3 and C-6 [38]. Reproduced with the copyright permission of the publisher, Elsevier, 2024.

In another instance, in Figure 8, Adekunle [54] reported that methacrylic acid, acetyl, and methacrylic anhydride are used to functionalize the triglycerides of epoxidized soy oil to create sustainable thermoset resins. Spectroscopic techniques were used to evaluate the produced resins to

verify their functionalization and the degree of epoxy transformation. Utilizing the methacrylate soybean oil resins as a matrix in composite applications, their thicknesses were examined. Ultraviolet & thermally induced curing studies and DSC data concerning the degree of crosslinking were used to evaluate the cross-linking capabilities. According to 1H-NMR, a mere 2.2% of the epoxy chains remained unreacted after up to 97% of the epoxy groups were converted, indicating that the modifications were satisfactory. The 13C-NMR confirms the 1:1 acetate-to-methacrylate methyl group ratio. The viscosities of methacrylic anhydride-modified soybean oil (MMSO) and methacrylate soy oil (MSO) were 0.48 and 0.2 Pas, respectively. Correspondingly, suggesting that they are suitable for use in the resin transfer molding process.

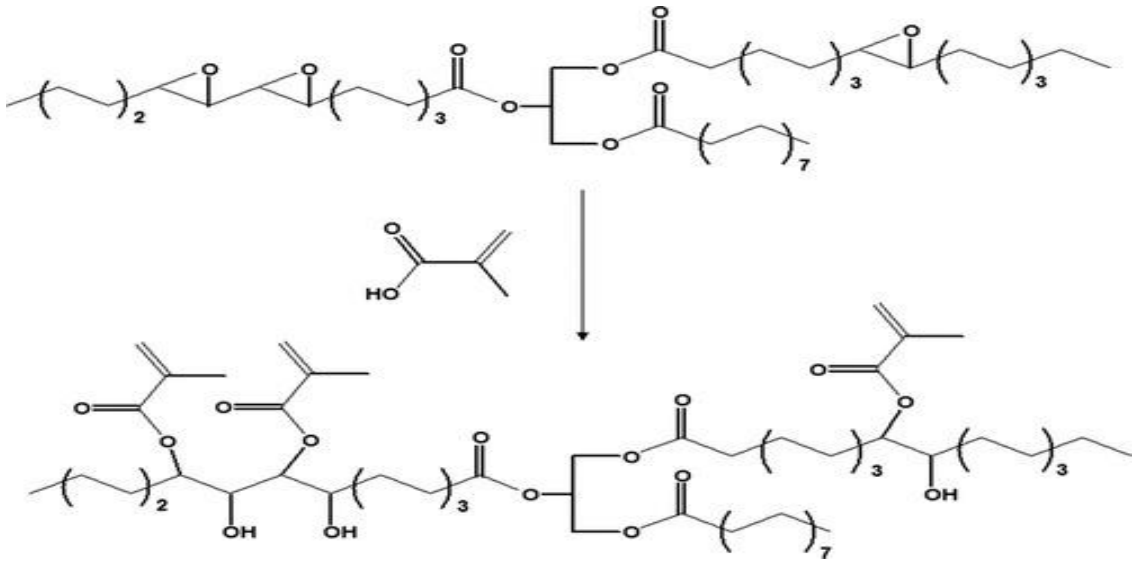

Figure 8. Methacrylic acid and epoxidized soybean oil (ESO) combine to produce methacrylate soybean oil (MSO) [54]. Reproduced with permission.

Diverse UV-curable bio-adhesives have been prepared from the co-polymers of N-vinyl-pyrrolidone and four varied co-monomers, namely vinyl succinimide, 2-acrylamide methyl-1-propane sulfonic acid, glycidyl-acrylate, and 2-isocyanate ethyl-methacrylate by Kao et al. [73]. The formulated biobased adhesives showed a UV-induced setting within 3 minutes. The bond between the bio-adhesive and the applied porcine intestine was measured utilizing the peel test. Based on their findings, these adhesive materials could provide values up to 4.6 N/m of an angle of about 180° peel force compared to the five bio adhesives from 0.52 to 3.04 N/m. More so, the UV-curable bio-adhesives (fully hydrated) demonstrated a water uptake between 25 and 350 wt.%, while equilibrium aqueous content ranged from 20 to 100 wt.%. N-vinyl-pyrrolidone is a monomeric unit, and as such, these resultant copolymers display a characteristic biocompatibility, indicating that the developed bioadhesives possess a great potential for several clinical applications, such as tissue adhesives and single-layered-hydrogel bandages for wounds [73].

The qualities needed in the resin are influenced by the adhesive's intended usage and the application technique. The following variables need to be managed: Solid content, viscosity, stability, slip, tack, substrate absorption, drying rate, flexibility, microbial resistance, and production cost. Many of these are determined by modification, whereas others need the addition or incorporation of an

additive to the desired properties [74]. For instance, sodium tetra borate with little sodium hydroxide is probably the most applied additive to adhesives based on starch. It is often employed as a stickiness stabilizer and to improve the flow rate of dextrin adhesives and a tackifier. To further stabilize starch-based adhesive systems and enhance their tackiness and flow behavior, sodium tetraborate (borax) is commonly employed in basic media alongside small amounts of sodium hydroxide. These additives form coordination complexes with hydroxyl groups in starch molecules, improving viscosity control, adhesive consistency, and thermal resistance. Figure 9 illustrates the molecular interaction between borax and starch in alkaline conditions, showing how borate ions form crosslinked networks with hydroxyl-rich starch chains.

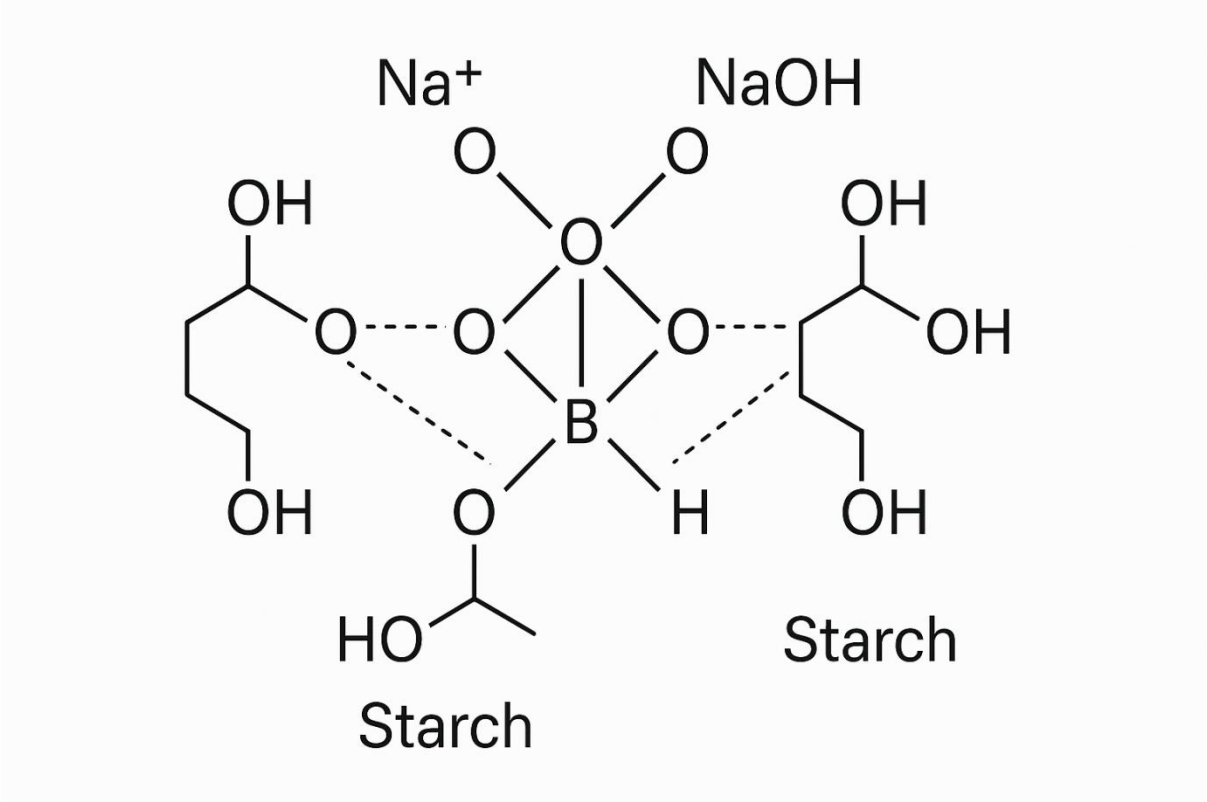

Figure 9. Borax complexes molecules of starch in a basic medium.

Because starch is so firmly bonded in granules, it is almost incapable of acting as an adhesive when floating in cold water alone. Direct chain molecules and straight segments of branched compounds are arranged in crystalline areas that constitute the granules. More amorphous zones, wherein the molecules are not coordinated, connect the crystallite sections. The molecules and crystallites are grouped in concentric layers radially within the starch granule (Figure 10). To achieve glue-like bonding, these tiny balls need to be opened. Figure 9 is an illustration of the molecular structure of starch with crosslinking or branching points.

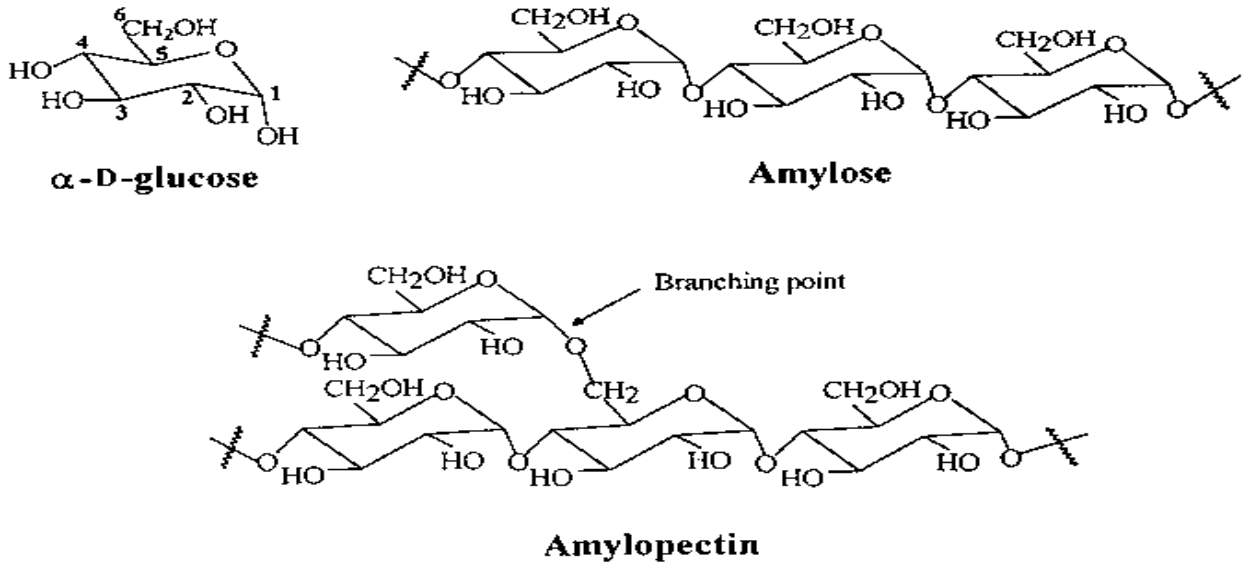

Figure 10. α-D-glucose molecules and those of amylose and amylopectin, the two major components of starch [75]. Adapted from the work of [75].

4. Developmental process in bio-adhesive applications

The eco-adhesives from sustainable sources have activated significant interest in the adhesives field. Among the diverse renewable resources, vegetable protein is a material for adhesive manufacture. The foundation of a sustainable economy is the shift from non-renewable sources to sustainable sources [76,77]. For instance, a soy protein-based adhesive based on [78], when chemically reinforced, is believed to possess similar adhesive features as those of formaldehyde and petroleum-based ones for interior applications [78]. The shear rheological research investigation conducted by Agbogo et al. [27] revealed that incorporating a modifier that is environmentally friendly into a vegetable and beef-bone polymer to modify it could regulate the fluidity, improve the surface properties, and reduce cost for engineering applications [27].

Most of the adhesive's production before a century ago came from biomaterials such as hides, skins, bones, milk, fish, and plants. Although synthetic-based adhesive polymers have existed since 1900, synthetic sealants and adhesives have many real-world industrial uses today. Given their capacity to provide a smooth exterior, distribute the weight, and make it simple to join thin or dissimilar materials, it is impossible to envisage a product used in homes, businesses, transportation, or any other setting that does not require adhesives or sealants in some way [79,80].

In engineering operations, adhesively bonded joints are increasingly replacing mechanical joints because they offer numerous benefits over traditional mechanical rivets. Additionally, they save weight and cost by distributing stress more uniformly along the bonded area, which increases stiffness and load transfer. Because adhesive bonding offers qualities, including a large strength-to-weight ratio, damage tolerance, dynamic design, fatigue resilience, and more, it has been used more and more in various sectors in recent years. In contrast to traditional joining techniques. Adhesive coupling has been proven to have useful applications in several industries, including the automotive, aerospace, electronics, sports, marine, oil, and construction sectors. In all industries, the use of adhesively attached joints in composite recovery of damaged structures has increased [81,82].

Given the polymeric composition of the glue, joint adhesives give superior damping qualities,

which also promote high strength. Since adhesives are flexible enough to enable differences in coefficients of expansion caused by heat, they can be used to join disparate materials. The main use for structural glues is the effective bonding of thin plates. The strength of adhesives is far less than that of metals, yet it is enough for structural purposes when applied to join thin plates with an extensive bearing area. Because it is simple to set up an automated procedure, the application of adhesive can be highly effective. Since the joint design is adaptable, novel ideas and materials may be used. Sandwich constructions, which have a composite skin and a honeycomb core, are an appealing illustration. These could enhance surface finishes; therefore, do not utilize welding markings, rivets, or bolt holes. Close interactions between the bonded sites will result from these, which is advantageous for a barrier to corrosion and structural integrity. Figure 11 is a template showing a form of molecular bridging between two contact bodies culminating in adhesion or adhesive force.

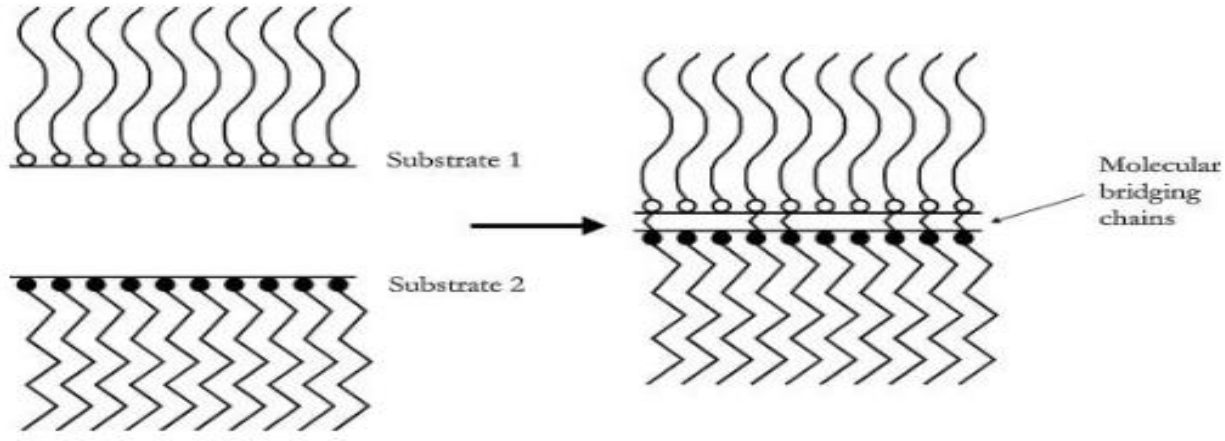

Figure 11. Molecular bridging between two surface contacts [83].

Additionally, adhesion has certain drawbacks that enable further technological innovation developments: Peeling and separation stress must be reduced since the weak strength of joints results from their concentration of stress in a limited region; the adhesive's polymeric nature restricts its resistance to extremes in temperature and humidity; bonding is usually not instantaneous, requiring an assortment of devices to hold the substrates in place; many adhesives necessitate temperature for hardening, which is a significant cost disadvantage; and careful surface cleanups, like mechanical scratches, solvent cleaning, or chemical procedures is required to have a useful interfacial resilience and a long-lasting join. Since an adhesive bond cannot be broken down, controlling quality proves more challenging than mechanical fasteners; however, several non-destructive methods are now accessible. Joint layout is often complex, with no clear regulations like with rivets, bolts, or welding, and architects distrust this method of assembly. Adhesive bonding activities are diverse and can be found in almost every industry. One of the pioneers of this techno-innovation is the aerospace sector. New disciplines, including biology and medicine, employ evolving cell adhesion-based procedures and protein adhesion on surfaces, posing crucial concerns in areas like surgical glues, artificial organs, and prosthetic components' biocompatibility.

While much of the focus remains on the classification and functionality of biomolecular adhesives, emerging interdisciplinary studies are redefining how these materials are synthesized and applied. The

following section highlights recent advances from adjacent fields that offer promising directions for the evolution of grafted bio-adhesive systems

4.1. Emerging cross-disciplinary insights into bioinspired adhesive design

Recent breakthroughs in material science have further reinforced the relevance of bioinspired adhesives within broader sustainable and biomedical domains. The incorporation of functional polymers and green synthesis routes has reshaped how adhesives are engineered for precision and environmental performance [84,85]. These studies demonstrate that sustainability-driven molecular design can achieve performance metrics comparable to synthetic analogues while reducing ecological impact.

In parallel, advances in amino acid-based triboelectric materials and bio-nanogenerator interfaces highlight the potential of integrating adhesion science with energy harvesting and smart interface technologies [84,86]. These materials, rooted in biocompatible substrates, offer multifunctional adhesion, enabling biomedical adhesives to perform secondary roles such as biosensing, drug monitoring, or micro-energy generation.

Furthermore, applications in wind and blue energy systems [3,84] underscore the mechanical resilience and environmental integrity required of adhesives in harsh operational environments. The convergence of energy systems with adhesive interface design is particularly relevant for developing hydrophobic, corrosion-resistant, and high-strength bonding layers.

Last, cutting-edge insights into sustainable tribo-materials and bio-interface engineering [59] suggest new directions for the valorization of grafted bio-adhesives, particularly in terms of long-term biostability, regenerative interface integration, and mechanical adaptability in wet and dynamic biological environments. Together, these new fields support the increasing need for graft-copolymer-enhanced bio-adhesives that are not only robust and biocompatible but also in line with the concepts of the circular bioeconomy and next-generation smart material systems.

5. Advances in starch-based adhesives, including graft copolymerization

Remarkably, starch acts as a binding agent in an array of items, including pastes, glues, binders, and shaping materials. The adhesives of living systems were developed and formulated, like gums and glues, but these adhesives are not very durable in harsh environments. Starch is a sustainable material obtained from commodity crops made in surplus, and its adaptability and affordable cost for chemical manipulation make it a popular choice for implementation as a replacement for synthetics. Starch is subjected to modification on application. The various techniques for splitting the granules include heating, oxidation, alkali, and acid treatment. To overcome the inherent limitations of native starch adhesives, such as poor water resistance and limited mechanical strength, graft copolymerization has emerged as a pivotal modification strategy. By chemically attaching synthetic monomers to the starch backbone, this process significantly enhances the functional properties of the adhesive. Figure 12 provides a schematic representation of this grafting mechanism, illustrating how monomers are integrated into the polymer matrix to create improved bio-based adhesive systems suitable for advanced applications.

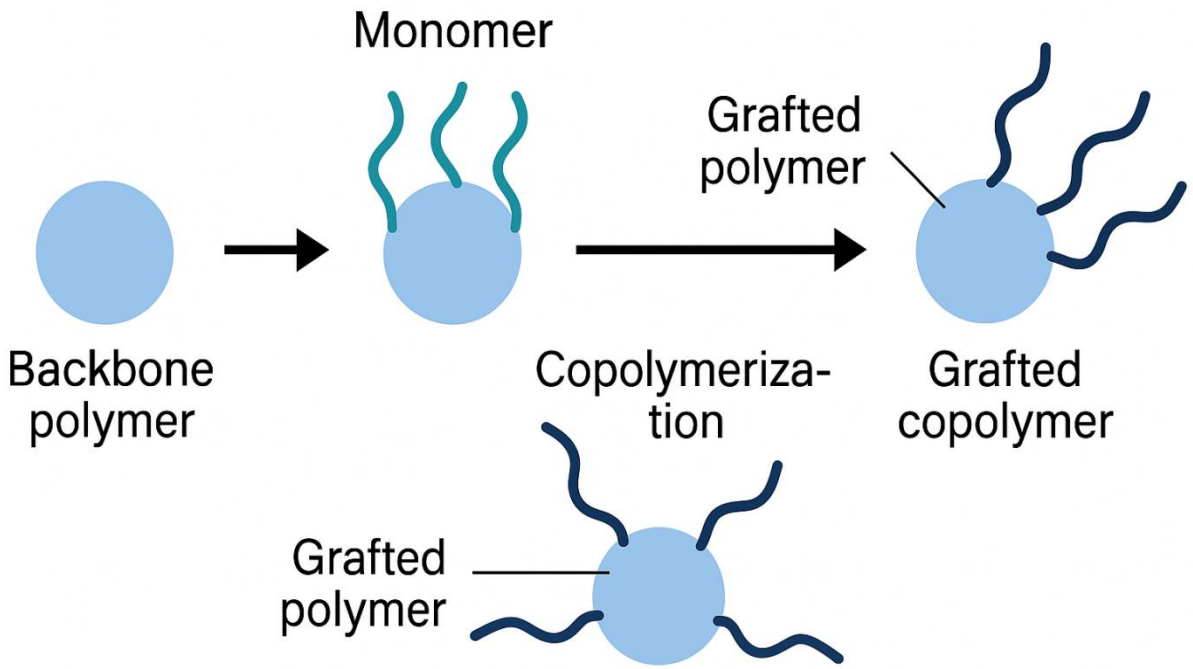

Figure 12. Schematic diagram of the graft copolymerization process. Monomers (blue) are chemically grafted onto a polymer backbone (gray), forming side chains that enhance the structural and functional properties of the resulting bio-adhesive. This modification improves mechanical strength, adhesion, flexibility, and moisture resistance, key performance criteria for starch-based biomedical adhesives.

Under slightly acidic conditions, Imam et al. [87] conducted research on the trans etherification reaction existing between methoxy functional groups obtained from hexa-methoxy-methyl-melamine and OH-groups in wood, wood, and PV-OH to produce ether-linkages in hexa-methoxy-methyl-melamine; PVOH–hexa-methoxy-methyl-melamine or wood–hexa-methoxy-methyl-melamine as illustrated in Figure 13. Given this reaction, methanol is produced, which exits the reaction and enters the solution due to having six methoxymethyl groups. hexa-methoxy-methyl-melamine proved to be a powerful crosslinking agent, giving the adhesive good bonding qualities by concurrently forming an effective network of crosslinks with the hydroxyl groups in PVOH, wood, and starch [87].

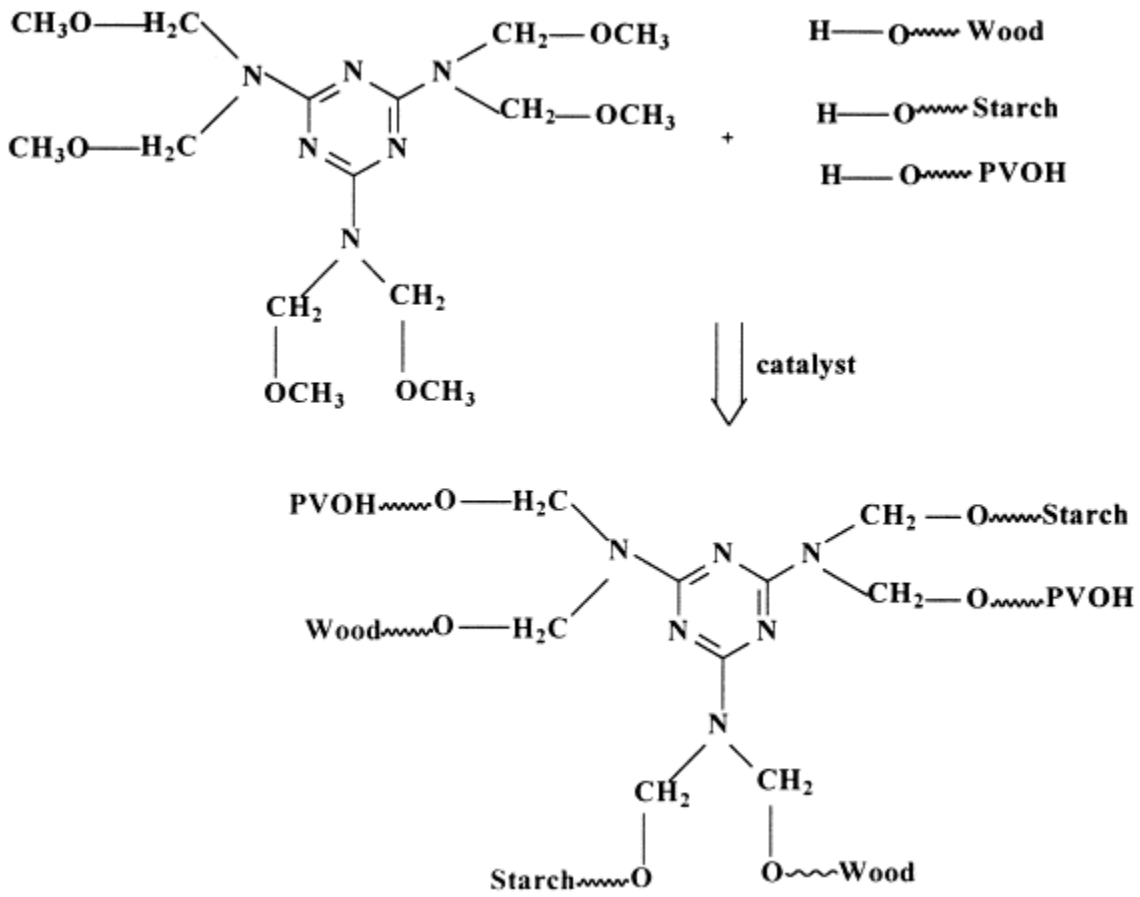

Figure 13. The schematic diagram demonstrates the trans-etherification reaction, which happens when hydroxyl groups in wood, starch, and PV-OH combine with methoxy functional groups derived from hexa-methoxy-methyl-melamine to form ether bonds connecting the starch and hexa-methoxy-methyl-melamine in PV-OH or wood and hexa-methoxy-methyl-melamine [87]. Reuse with the copyright permission of the publisher.

To better illustrate the diversity of chemical strategies employed in modifying starch-based adhesives, Table 3 summarizes selected studies that utilize graft copolymerization and other modification techniques, highlighting their targeted enhancements, application contexts, and notable outcomes.

Table 3. Summary of modification techniques applied to starch-based adhesives

Ref.	Modification type	Material/ polymer	Target property enhanced	Application focus	Notable findings
[27]	Graft Copolymerization	Starch + synthetic monomers	Mechanical strength, water resistance	Biomedical adhesives	Demonstrated significant improvement in bioadhesive bonding strength.
[30]	Graft Copolymerization	Chitosan + PEG	Adhesion in moist conditions	Tissue engineering	Grafting hydrophilic groups improved wet adhesion in biological tissues.
[31]	Graft Copolymerization	Chitosan + bioactive polymers	Biocompatibility, biodegradability	Medical (surgical sealants)	Enabled adhesion to mucous membranes with controlled degradation.
[32]	Graft Copolymerization	Biopolymers + functional monomers	Functional tunability, antibacterial	Biomedical coatings	Introduced antimicrobial properties and better implantation compatibility.
[33]	Graft Copolymerization	Various bio-based substrates	Mechanical strength, flexibility	Medical adhesives	Reinforced copolymer performance using nanofillers.
[68]	Cross-linking (non-graft)	Starch-based	Thermal stability, viscosity	Packaging	Improved bonding temperature range and formulation control.
[64]	Plasticization (non-graft)	Cassava starch	Flexibility, drying behaviour	Paper and label adhesives	Improved film-forming and anti-cracking properties.

Continued on next page

Ref.	Modification type	Material/polymer	Target property enhanced	Application focus	Notable findings
[69]	Esterification (non-graft)	Modified starch	Water repellency, film durability	Industrial coating adhesives	Enhanced surface hydrophobicity through ester-based linkage.
[63]	Blending with additives	Starch with bio-fillers	Rheology, set time	Packaging, general-purpose glue	Achieved better performance through additive synergy.
[70]	Enzymatic treatment (non-graft)	Starch	Processability, biodegradability	Environmentally friendly glues	Boosted enzymatic activity and reduced harmful byproducts.
[66]	Heat treatment (non-graft)	Starch	Viscosity control	Corrugated board adhesives	Modified adhesive behavior for high-speed industrial processes.
[67]	Various (including grafting)	Bio-phenolic resins	Bonding strength, eco-safety	Structural adhesives	Surveyed a broad range of bio-based adhesives with scalable strategies.

An efficient method was developed by Zhang et al. [88] to create outstanding performance in starch-based glues with wood surfaces, employing an olefin as a co-monomer, an oxidant, and a silane coupling agent, as well as H_2O_2. The strength of starch-based wood adhesives reached 7.88 MPa in the dry state and 4.09 MPa in the wet state, according to research on the impact of different parameters on the shear strength done in both dry/wet conditions. In other words, graft copolymerization enhanced heat stability, which in turn intensified bonding strength and water resistance, whereas oxidation may reduce the degree of hydroxyl conversion to aldehyde and carboxyl radicals. In these reports, the new bio-based adhesive and the breaks in the bonded joints were examined using scientific methods like scanning electron microscopy (SEM), thermo-gravimetric analysis (TGA), and Fourier transform infrared spectroscopy (FTIR). It was found that the reinforced microstructure of the graft co-polymerized adhesive was linked to enhanced adhesive qualities [88].

A wood adhesive was prepared by Imam et al. [87] made from a naturally occurring, sustainable resource that has the qualities and optimizations of a cross-linked adhesive system that utilizes starch and polyvinyl alcohol that is appropriate for interior wood-to-wood bonding.

According to their findings, the crosslinker, hexa-methoxymethyl melamine, is effective in crosslinking because of a trans-etherification within the OH groups in wood, vinyl alcohol, and starch molecules and the methoxy groups in hexa-methoxymethyl melamine. Given this reaction, the hydroxyl molecules formed ether linkages with the crosslinker by replacing the methoxy sites. Their studies state that the glue's ideal viscosity was reached at a solid content of 27 percent and that the addition of latex to the formulation increased the glue's moisture susceptibility. Additional findings by Imam et al., include the following: Wood samples modified at 93 percent relative humidity for two months showed exceeding 95% collapse in wood but low in joints; following two months at 97% relative humidity and a year at 50% relative humidity, electron microscope images showed no discernible growth of fungi or microbes on the glue [87].

The grafting process is a crucial method for altering polymers' physical and chemical features. One of the best techniques for enhancing starch's adhesive capabilities is the incorporation of artificial polymers into its backbone. For instance, Wang et al., [89] created a novel renewable, non-toxic starch-based adhesive that can be applied at ambient temperature by grafting vinyl acetate monomer onto the backbone of waxy maize starch. Infused adhesive water resistance rose by 61.1% , and its shear strength increased by 59.4% in the dry state and 321% in its wet form in contrast to the conventional PVAc/gelatinized starch mix. The compatibility between polyvinyl acetate and starch, based on the characterization results obtained from FTIR, NMR, thermogravimetry, and scanning electron microscopy analyses, strengthened the product, leading to improved performance of the manufactured starch adhesive by graft. The research studies conducted by Wang and Co., [89] further showed that the most economical monomer feeding ratio was assessed utilizing bond properties and grafting factors, with a starch/monomer ratio of 1:1.2 (w/w) being deemed the optimal proportion in economic terms.

Zia-ud-Din *et al.* [90] are reported to have developed a simple method for adding sucrose esters to high amylose starch-based adhesive to enhance its quality. From their findings, 6% additional (w/w, dry starch basis) sucrose fatty acid esters enhanced strength when humid and when hydrated, while impacting the adhesive's storage and portability. Due to the use of thermal imaging studies, the growth of complexes was identified, which showed that the addition of sucrose ester to the enhanced amylase starch-based wood glue promoted the thermal integrity of starch systems and prevented latex particle agglomeration. This means that the esterification culminated in a suppression of starch retrogradation based on the analysis spanning from radiation pulses along with a sweep, thus providing important information for high-performance bio-adhesives. In the work of Qiao *et al.* [91], a starch-based adhesive was formulated utilizing corn starch, sodium borate, and polyvinyl alcohol. To build its resistance to moisture, a particular formulation of poly-methylene polyphenylene isocyanate pre-polymer was utilized as a cross-linking agent when water-soluble carboxymethyl cellulose was added during the manufacturing process. A dynamic testing machine assessed the adhesion and water resistance of the three-layer plywood that was created by hot pressing. The adhesives' cure was assessed using SEM, TGA, and DSC. According to the reports, there was a considerable improvement in the initial viscosity, particle content, durability of bonding, and interface compliance between the pre-polymer and starch adhesive components. From the findings, the curing was reduced, and the optimization of carbon methyl cellulose was 0.375%, as it was in [91]. Moreover, Qiao et al., [92] developed a temperature-cured starch adhesive system by dry technique esterification cum poly-isocyanate pre-polymer crosslinking to investigate the uses of crosslinking and esterification on the

properties of the corn starch-based adhesive system. The results obtained via the various characterization techniques of the maleic anhydride esterified starch using dry method esterification revealed that the starch's crystal form remained unchanged, but its crystallinity dropped; additionally, the adhesive's delivery at its bonding point was confirmed following esterification, while the prepolymer enhanced the adhesive's thermal stability; therefore, 10 percent prepolymer was the ideal amount to add [92].

6. Biomedical applications and challenges of bioinspired adhesives.

6.1. Medical advances of bio-adhesive systems

Traditional medical applications of bio-adhesive systems include homeostasis, wound healing, and tissue sealing. However, it is quite interesting that many areas of smart applications are emerging, which include drug delivery, biosensors, and tissue modulation. With their simplicity of use and low risk of injury, bio-adhesives have become revolutionary and adaptable instruments in the medical field. These materials offer a wide range of possibilities that have captured the attention of scientists and clinicians. These include tissue restoration and the integration of biomedical devices. However, achieving their full potential necessitates a variety of design approaches, including effective signal interaction, appropriate biological interactions, and maximum adhesion [93]. Smart bioadhesives possess enormous merits, such as sustained release, the spatial transfer of biomolecules with lowered effects, as well as ease of access. Other areas that have benefited from smart bio-adhesive systems are tissue engineering, regenerative biomedicine, cancer therapy, and many more. The usefulness of smart bio-adhesives has been developed lately through a variety of disciplinary activities, which have resulted in discoveries and creative compositions. However, as far as we are aware, not enough studies have been conducted on the creation of multipurpose bio-adhesives for a range of medical purposes [94].

Cyanoacrylate-based adhesive, also referred to as super glue, is a strong and multifunctional adhesive. This liquid-state monomer is in demand, perhaps due to its rapidity in polymerization and quick curing time. However, it is affected by nucleophile species, such as the hydroxyl group. This agrees with the work of Mehdizadeh and Yang [95] based on the equations shown in Figure 14. Cyanoacrylate is a liquid-state monomer family that usually polymerizes at room temperature based on exothermic anionic polymerization in the presence of OH ions and water [95]. Mechanistically, molecular interactions are a key factor in crosslinking among polymeric morphology, culminating in the main framework of modulated structures. The mechanical characteristics of the adhesive, such as its rigidity, resilience, hydrophobicity, and flexibility, can be altered by these cross-linked domains. Characteristic properties of interest, such as regenerative, adhesive, antioxidant, and antibacterial properties, can be introduced into the crosslinking domain for enhanced performance. However, it is crucial to be able to adjust the crosslinking density while maintaining the required chemistry which is a major challenge met by scientists in designing biobased adhesive systems.

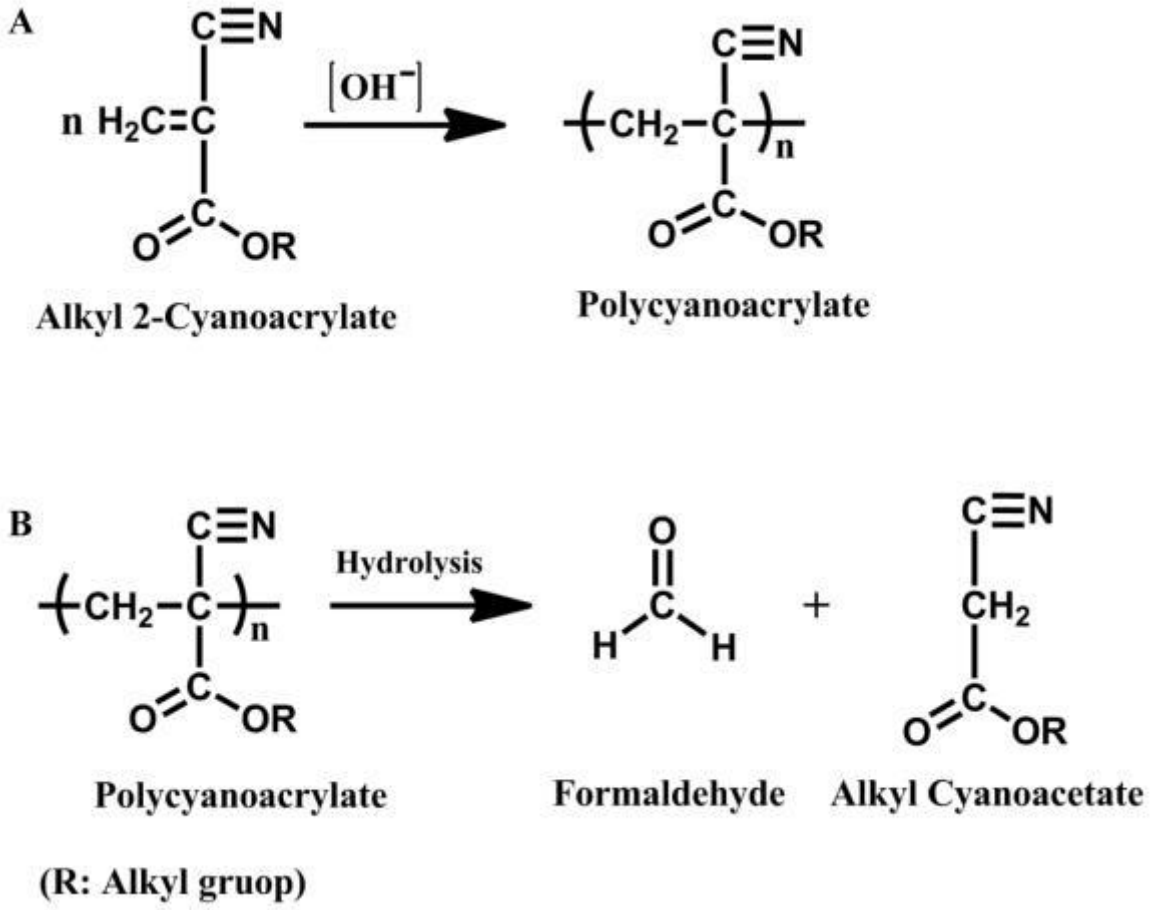

Figure 14. Effect of nucleophile species on cyanoacrylate-based adhesives. Adapted from the work in [95]. Reproduced with the copyright permission, Elsevier 2012.

6.2. Challenges and strategic solutions in biomedical adhesive engineering

6.2.1. Recent challenges in adhesive formulations

The trade-offs between toughness cum material stiffness while maintaining adhesion to wet surfaces make it difficult to design and formulate adhesive hydrogels with the best properties for treating wounded tissues. Biobased adhesives with reinforced mechanical strength may not have the right amount of elasticity, thus making it difficult to use them to seal soft, elastic, and active tissues. Hence, bio-adhesive specialists can begin to research more by tuning molecular interactions to a single-hydrogel system with multifunctionality capable of curtailing current challenges in tissue repair and sealing. This technique can be employed in other areas of application.

Given the hydrophilicity of many adhesives, degradation on contact with a hydrolyzed environment in a bid to proffer a solution to this limitation by incorporating a hydrophobic tail of polyvinyl acetate into the hydrophilic base polymers (starch and gelatine) and reported that there was an improvement in their engineering performances.

Tissue adhesives stop bleeding and play a significant role in wound closures; however, most adhesives are either expensive, cytotoxic, or have poor tissue adhesion. The impermeable adherence

of plant-based tannins excited Guo et al., [96], who created a new class of bio-adhesives by Michael, adding tannic acid and gelatine in a single phase while it was oxidized and crosslinked with silver nitrate [96]. According to their investigations, tannic acid's polyphenol groups provide moist tissue adhesion using chemistry like catecholamines, but silver nanoparticles derived from silver nitrate and tannic acid both provide antibacterial supplies that are inherent to the polymeric network. Significant damp tissue adhesion strengths (up to 3.7 times that of fibrin glue), by changing the gelatine's origin (fish, bovine, or porcine), was possible to achieve outstanding cytocompatibility, low prices, scalable readiness, and regulated deterioration (up to 100% degradation within a month), as well as the ratios that supplied tannic acid. Such adhesives also eliminate the risk of potential neurological effects that may be brought about by mussel-inspired strategies because of dopamine, as well as antibacterial and antifungal properties. The innate properties of tannic acid as a natural phenolic crosslinker, molecular glue, and antimicrobial agent resulted in a unique and significant approach to bio-adhesive design [96].

Zheng and Co. [97] created a ready-to-use hemostatic bio-adhesive that had excellent mechanical elements and endurance to fatigue, elasticity to allow for physiological function and mobility, the ability to effectively halt bleeding, and adherence to moist tissues after a couple of seconds of pushing. While sealing the wound, controlling bleeding from impaired internal organs is crucial for patient survival. Many bio-adhesives exhibit low mechanical features, blood incompatibility, and complicated deployment procedures. The researchers claim that by adjusting several chemical reactions and crosslinking processes involving N-hydroxy succinimide (NHS) conjugated alginate (Alg-NHS), poly (ethylene glycol) diacrylate (PEGDA), tannic acid (TA), and Fe^{3+} ions, the created hydrogel showed high elasticity greater than 900%, toughness greater than 4600 kJ/m^3, and a wound closure test greater than 400 kPa. Wet tissue adherence was synergistically improved by dual adhesive moieties, such as NHS and pyrogallol/catechol inspired by mussels. TA/Fe^{3+}'s strong affinity for blood improved hemostasis even further. All things considered, the design approach outlined here could be used to get over current barriers preventing the clinical application of tailored hemostatic bio-adhesives [97].

Commercially based adhesives of different sorts are available everywhere. Bio-based adhesives such as fibrin glues are biocompatible; however, they demonstrate low mechanical properties as well as finite adhesion to wet surfaces. Moreover, synthetic bio-adhesives like cyanoacrylate and poly (ethylene glycol)-based adhesives possess comparatively high mechanical characteristic properties and could cause an inflammatory reaction(s), which is boosted by the discharge of their degradation products. To sum up this section, here are the recent challenges facing adhesives development:

- Limited Functionality: Many bio-adhesives on the market are primarily focused on adhesion and do not have supplementary therapeutic effects, such as antibacterial qualities or the capacity to stimulate tissue regeneration in the absence of outside influences. Commercial bio-adhesives that are widely accessible frequently don't have antibacterial qualities, which makes extra antibiotics necessary and adds to the issue of multidrug resistance.
- Unsatisfactory Mechanical Advantage: Most bio-adhesives on the market today have insufficient bulk mechanical strength. Additionally, they might have poor interface connections with biological tissues, particularly in high-pressure and blood-filled settings like arteries and cardiac chambers. For instance, tissue mobility is restricted by the cyanoacrylates' stiffness and lack of flexibility despite their high adherence. Adhesives based on polyethylene glycol have a high swelling and low adhesion energy [38]. Glue's weak substrate toughness and adhesion strength result in inferior tissue-bonding qualities.

- Wet attachment Difficulties: Quick, solid, and durable attachment is hampered by the physiological environment's blood and tissue fluids. Because slippery tissue surfaces are more challenging to adhere to, wet adhesive strength needs to be increased [98].
- Sustainability Issues: Certain adhesives on the market are cytotoxic or have the potential to trigger allergic reactions. Cyanoacrylates, for example, are cytotoxic. Because fibrin glue is made from human plasma, there is a chance that it could cause blood-borne infections. Bio-adhesives made from gelatine have the potential to trigger anaphylactic responses [36–38].
- Cost and Scalability: Natural-based bio-adhesives can be costly and challenging to store. During development, consideration must be given to the goal of achieving cost-effectiveness for commercialization.
- Comprehending Natural Adhesives: Gathering natural adhesives for characterization and creating theoretical models to forecast adhesion based on the geometry of natural adhesive structures are two difficulties with bio-inspired adhesives. It is also challenging to distinguish the functions of distinct molecular constituents in intricate natural bio-adhesives.
- Interdisciplinary Nature of Research: Research on bio-adhesives involves good interdisciplinary cooperation because it requires expertise from several scientific areas, which might be difficult for individual researchers.

6.2.2. Implications in the engineering of adhesives

The existing drawbacks prevent adhesives from being widely used as replacements for tissue-incompatible traditional wound closure techniques such as stitches and staples. Leakage and delayed healing can culminate in improper wound closure. The likelihood of infections following surgery may rise if adhesives lack antibacterial qualities, as is common with available adhesive products in the market. Bonding failure under physiological stressors may result from poor mechanical integrity. Moreover, problems with cytotoxicity and biocompatibility may hinder the efficacy of adhesive substances, resulting in unfavorable tissue interactions. Additionally, industrialization and regulatory approval may also be slowed by the disconnect between research laboratory progress and real clinical needs.

6.2.3. Potential solutions in adhesive developments

There is a lot of potential for creating a new breed of bio-adhesives that are safer, better performing, and capable of handling a greater variety of clinical issues if studies and developments in these fields continue. Some of the potential solutions are discussed below:

- Incorporating Additional Biofunctions into Bio-adhesives: Designing bio-adhesives that are also antimicrobial, self-healing, drug-delivering, and intrinsically hemostatic is a potential solution to challenges in adhesives' development.
- Improved Mechanical Strength and Increased Toughness: The use of highly symmetric macromolecules to form homogeneous gel networks with the ability to distribute uniform stress is necessary for enhanced mechanical performance. Developing a non-linear, elastic response for reversible post-deformation recovery by introducing reversible cross-linkages as energy dissipation mechanisms to avoid crack propagation and tough bulk cohesive strength combined with strong interfacial bonds.

- Enhancing Wet Adhesion: Using compounds like Dopa, lysine, and amino to improve wet adhesion while taking inspiration from mussel adhesion processes, investigating chemical bonding in conjunction with physical adhesion mechanisms such as mechanical interlocking, interdiffusion, and electrostatic interactions, and changing the surface of materials to improve their contact with moist tissues are critical factors in dealing with wetness [36].
- Employing ecologically friendly and Renewable Materials: Altering these biopolymers to improve their mechanical and adhesive qualities requires concentrating on materials from nature that typically show superior biological compatibility and biodegradability, such as polypeptides, proteins (such as fibrin, gelatine, albumin, and silk), and polysaccharides (such as chitosan, alginate, hyaluronic acid, starch, and CMC).
- Creating self-healing adhesives: Integrating dynamic and reversible covalent connections (such as Diels-Alder and Schiff-base processes) or reversible noncovalent interactions (such as electrostatic, hydrophobic, and hydrogen bonds) into the binding network could transform a hydrophilic adhesive substance into hydrophobic molecules [27,36]. To regulate adhesion or medication release, stimuli-responsive adhesives are made that can react to environmental cues such as pH, temperature, or the presence of enzymes.
- Bio-inspired Design: Creating new synthetic adhesives with better qualities by taking inspiration from the adhesive techniques of different species (such as mussels, geckos, snails, and spiders) could be a premise for remediating recent challenges in the engineering of adhesives [99].
- Applications of Nanotechnology: Using nanoparticles in bio-adhesives to improve their mechanical properties and adherence [100,101].
- Standardization and Cooperation: Promoting better cooperation between scientists and medical professionals to pinpoint unmet clinical requirements and set design goals for creating adhesives tailored to certain applications requires adhering to standardized requirements (such as ISO-10933) to expedite regulatory approval and guarantee biocompatibility [102].

7. Ethical considerations in biomedical adhesive applications

As bioinspired adhesives gain traction in medical and surgical applications, including tissue sealing, wound closure, implant fixation, and drug delivery, the need to address ethical considerations becomes increasingly important. While much attention has been devoted to the chemical safety, efficacy, and biodegradability of these materials, ethical dimensions such as patient safety, informed consent, clinical transparency, and regulatory compliance must also be critically examined.

A primary concern is the biocompatibility and long-term safety of novel graft-copolymerized adhesives when introduced into human tissues. Researchers and developers must ensure that new formulations do not elicit unforeseen immune responses, toxicity, or interference with physiological processes. Rigorous preclinical testing, followed by ethically conducted clinical trials, is essential to uphold standards of patient care and scientific integrity.

Informed consent plays a vital role in the application of emerging adhesives in experimental or novel clinical settings. Patients must be adequately informed about the nature of the biomaterials being used, their potential benefits, and any associated risks, especially when such adhesives are part of pilot studies or early-stage technologies. Moreover, equitable access to these innovative materials should be a guiding principle. As high-performance bio-adhesives enter clinical markets, disparities in access due to cost, geographic limitations, or healthcare infrastructure must be proactively addressed to avoid ethical inequities in care delivery.

Finally, environmental ethics intersect with biomedical ethics in the context of bio-based materials. The use of biodegradable, sustainable adhesives derived from renewable sources aligns with broader principles of responsible innovation and planetary health, further reinforcing the value of bioinspired design from an ethical standpoint. As research continues, collaboration between scientists, clinicians, regulatory bodies, and ethicists will be essential to ensure that bio-adhesive technologies are developed and deployed responsibly, balancing innovation with safety, equity, and environmental stewardship.

8. Functional additives for adhesive enhancement

Plasticizing agents are incorporated in the manufacturing process of the adhesive to manage the adhesive weakness of the glue line and to control drying speed. Common plasticizers often work in one of three ways: They lubricate the layers inside the dry adhesive systems, they manage the moisture content of the adhesive's film, or they form a solid solution with the dried adhesive system. The viscosity of the adhesive preparation is often reduced by plasticizing agents that produce a solid mixture, such as formaldehyde, urea, sodium nitrate, and salicylic acid. Overall, urea appears to be the most frequently employed of these additions, and depending on dry starch, it usually works well at 1–10%. Hygroscopic plasticizers, namely glycerol and ethylene glycol, under normal conditions, are applied to lower the film's rate of drying and make sure they do not crisp. Given the fact that they are not affected or influenced by variations in humidity to impart flexibility to the glue, lubricating adhesives such as polyglycols and soaps, are used in very small amounts [13,36,58].

8.1. Water resistance modifiers

Hydrophobic qualities are necessary for starch adhesives to be utilized in any situation where water resistance is required, i.e., additives that are capable of repelling water molecules. For instance, reagents such as polyvinyl acetate, polyvinyl alcohol, melamine–formaldehyde polylactic acid, urea-formaldehyde, and resorcinol–formaldehyde pre-condensates have been utilized. While polyethylene glycol and polyvinyl acetate have been demonstrated to be appropriate for use in adhesives that are effective against cold water, formaldehyde-based pre-condensates are used to provide high water resistivity [103–105].

8.2. Rheology stabilizers and fillers

As mentioned, one major challenge that is likely to occur in starch-based adhesives is that of retrogradation. To curtail this drawback, colloid stabilizers like sodium chloride and soaps are commonly used to inhibit this tendency. Moreover, meta borates, hydrogen peroxide, sodium perborate, sodium peroxide, sodium hydroxide, and many plasticizers could demonstrate this same role in adhesive development and manufacture. Moreover, fillers are typically employed to control the adhesive's rheological behavior, or its ability to penetrate or move through the substrate, as well as to establish the glue sets. Clay and bentonite are two typical types of fillers. Preservatives that prevent the growth of microorganisms. Bleach is another additive that can be applied to starch-based adhesives to eliminate colored contaminants and stop discoloration over time, even defoamers to control foaming during processing, and organic solvents to enhance or support adhesion to waxed surfaces. The use and kind of adhesives determine the defoaming agents and solvents that should be used in starch adhesive systems, paying particular attention to component toxicity and suitability.

9. Nanofiller integration for enhanced bio-adhesive performance

Kotiyan and Vavia, 2002 intended to create a copolymer of 2-ethylhexyl acrylate and acrylic acid that can be used as a pressure-sensitive adhesive matrix in applications, including transdermal medication delivery. For the polymer synthesis in these studies, the free radical initiator for free radical solution polymerization was 2, 2′-azobisisobutyronitrile. For polymer integration, the scientists' approach includes optimizing the reaction conditions. Peel strength about the release liner and human skin and skin irritation potential were investigated to determine its appropriateness in the creation of transdermal systems. To understand the adherence and adhesive transfer, the wear performance test was also investigated. The glue material exhibits significant peel strength, suggestive of great adhesive transfer on separation. It was deemed adequate for application in transdermal and could be subsequently developed either as an adhesive matrix or as an ingredient surrounding transdermal drug delivery [106].

Du et al. [107] used the prepolymer mixing approach to create a range of aqueous polyurethane adhesives containing diphenyl-methane-4,4′-diisocyanate, 1,6-hexamethylene diisocyanate, poly (1,4-butanediol adipate) diol, 1,4-butanediol, and internal-emulsifying chemicals. The thickness, thermal, mechanical, and adhesion properties were ascertained. The authors primarily concentrated on structure-property relationships. The findings indicated that the molar ratio formulations influence the resultant properties, coupled with excellent T-peel and mechanical strength [107]. That is to say that a higher molar ratio culminates in higher thermal stability.

Popular fillers that can be employed in the creation of the adhesive from natural sources, like wood flour, talc, and calcium carbonate fibers, frequently call for measurable amounts of fillers in the formulation. Nonetheless, at lower concentrations, nanofillers are known to enhance a polymer's distinctive qualities. Nanoparticles, nanotubes, nanofibers, and nano-clay are examples of nanosized fillers that have been used as fillers for epoxy and protein adhesives in recent years to create high-performance matrices with improved qualities [108]. Because of their remarkable physical and chemical characteristics, carbon nanotubes have also demonstrated a great deal of activity in most scientific and engineering fields since their discovery in 1991. Nanotubes are useful for a variety of applications because no material exhibits the combinatorial characteristic features of mechanical, electrical, and thermal capabilities assigned to them [109]. These enhancing nanomaterials frequently draw the interest of most researchers because of their distinct architectures, superior electrical qualities (more than copper 1000 times), increased heat conductivity (double that of diamond), better strength (about 100 times more robust than steel), as well as modulus (around 1 TPa), and heat tolerance (2800°C in vacuum) [110]. Carbon nanotubes are a perfect fit for use as fillers in material composites because of their special multipurpose qualities [110–114]. Electrical and thermal conductivities have frequently been improved by adding carbon nanotubes to polymer systems. When comparing the heat transfer properties of epoxy compounds and multiwalled carbon nanotubes when bisphenol F and bisphenol A diglycidyl ether are utilized as resins made of epoxy, the researchers in [115] found that the composite's thermal conductivity was greater than the epoxy resin's pure value. The presence of carbon nanotubes was believed to be the reason behind the rise in thermal conductivity. Additionally, if the carbon nanotubes are evenly distributed throughout the epoxy, the composite material's heat conductivity may be improved. The creation of carbon nanotube/polymer composites for both functional and structural uses has been the subject of numerous studies; these techniques can be deployed for the valorization of bio-adhesives.

The prospect of using carbon-based nanotubes as fillers is limited due to current technological

issues with the dispersion of entangled carbon nanotubes during processing and inadequate interfacial contact. To improve the dispersion of polymer matrices, several dispersion techniques have been studied, including mechanical dispersion, chemical functionalization, and optimal physical blending. To achieve homogenous dispersion, Sandler et al. [116] dissolved carbon nanotubes in epoxy while stirring at a high speed of roughly 2000 rpm for an hour. They found that vigorous stirring was a successful and efficient method. Sulay et al. [117] produced reinforced epoxy-based composites using the sonication technique. Because of the effective dispersion condition and load transfer mechanism, the authors disclosed that even a slight change in the nanotubes' chemical treatment has demonstrated a notable impact on the mechanical and morphological properties of the nanocomposites. To create adhesives with uniformly distributed carbon nanotubes in the epoxy matrix, Yu et al. [118] used mechanical stirring combined with ultrasonication as a straightforward and practical method. The findings showed that adding carbon nanotubes increased electrical conductivity and thermal stability, and that the adhesive's thermal degradation temperature increased by roughly 14 °C when 1% weight of CNT was added. In the study by Sadare et al. [119], modified carbon nanotubes were added by employing sonication and shear mixing methods, soy protein isolate adhesive to create a homogenous sample of carbon-nanotubes/soy protein nanocomposite adhesive. Functionalization is another way to improve dispersion in a polymer matrix.

Li et al. [120] noted that a major challenge in wearable electronics, personalized healthcare surveillance, and electronic skins is the pursuit of flexible hydrogel sensors with high sensitivity and rapid response. They also noted that an emerging method of incorporating ionic and electronic conductive pathways into hydrogel sensors can prevent sensing performance that is limited by a single conductive pathway. Multi-walled carbon nanotubes, which can exhibit exceptional qualities, including high sensitivity (GF = 12.46), quick responsiveness (273 ms), and a broad strain range (1–1000%), are initially added to casein gels in this manner [37]. Thus, adding these adaptable qualities to pliable Casein Gel sensors or any other adhesive system may lead to a wide range of potential uses.

10. Conclusion

The valorization of bioinspired adhesives through graft copolymerization represents a promising frontier in sustainable materials engineering, especially for biomedical applications where biocompatibility, performance, and environmental impact must be carefully balanced. In this review, we highligh the evolution of biomolecular adhesive systems, identified critical challenges in adhesion efficiency and moisture resistance, and examined how graft copolymerization and nanofiller integration offer robust solutions to these limitations. Starch-based and protein-derived adhesives demonstrate significant potential when chemically modified to enhance their mechanical, thermal, and biological properties. The incorporation of nanomaterials and functional additives further broadens the utility of these materials across medical, industrial, and environmental applications. Looking forward, several strategic directions are essential to realize the full impact of these innovations. Scaling up production processes for bio-adhesives without compromising performance or environmental integrity remains a primary challenge. Clinical translation will require rigorous biocompatibility studies, regulatory alignment, and patient-centered deployment strategies. Moreover, interdisciplinary collaboration, uniting chemists, material scientists, biomedical engineers, clinicians, and ethicists, will be vital for ensuring that adhesive technologies are not only technically sound but also ethically and socially responsible.

As the field matures, bioinspired adhesives enhanced via graft copolymerization have the

potential to transform not only material science but also the future of medicine and sustainable manufacturing.

Use of AI tools declaration

During the preparation of this work, the author(s) used QUILLBOT to improve the paper's readability. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

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Conflict of interest

The authors state that none of the work described in this study could have been influenced by any known competing financial interests or personal relationships.

Author contributions

Dr. Agbogo: Writing – writing – original draft, conceptualization, editing. Prof. Sadiku: review, Supervision. Dr. Mavhungu: Project administration and investigation. Dr. Teffo: Visualization.

Reference

1. Vernengo AJ (2016) Adhesive materials for biomedical applications, In: Rudawska A, *Adhesives - Applications and Properties,* Croatia: *InTechOpen,* 111–133. https://doi.org/10.5772/64958
2. Kharaziha M, Scheibel T, Salehi S (2024) Multifunctional naturally derived bio adhesives: From strategic molecular design toward advanced biomedical applications. *Prog Polym Sci* 150: 101792. https://doi.org/10.1016/J.PROGPOLYMSCI.2024.101792
3. Zhang Z, Liu X, Ban X, et al. (2025) The influence of filler types and content on the curing behavior and properties of a bio-based polyurethane engineered sealant. *Int J Adhes Adhes* 136: 103866. https://doi.org/10.1016/j.ijadhadh.2024.103866
4. Agbogo VU, Sadiku ER, Mavhungu L, et al. (2025) Nanotechnology coatings in the defense and aerospace industry. *Next Nanotechnol* 7: 100197. https://doi.org/10.1016/j.nxnano.2025.100197
5. Arepalli SK, Tripathi H, Hira SK, et al. (2016) Enhanced bioactivity, biocompatibility, and mechanical behavior of strontium-substituted bioactive glasses. *Mat Sci Eng C* 69: 108–116. https://doi.org/10.1016/j.msec.2016.06.070
6. Chuni T, Dachasa K, Gochole F, et al. (2023) Effect of morphology on the in vitro bioactivity and biocompatibility of spray pyrolyzed bioactive glass. *Adv Mater Sci Eng* 2023: 5858858. https://doi.org/10.1155/2023/5858858
7. Hu Q, Jiang W, Li Y, et al. (2018) The effects of morphology on physicochemical properties, bioactivity and biocompatibility of micro-/nano-bioactive glasses. *Adv Powder Technol* 29: 1812–1819. https://doi.org/10.1016/j.apt.2018.04.017

8. Owens B (2018) Bioactivity, biocompatibility and biomimetic properties for dental materials: clarifying the confusion. *Mod App Dent Oral Health Care* 2: MADOHC.MS.ID.000132. https://doi.org/10.32474/madohc.2018.02.000132
9. Antosik AK, Mozelewska K, Musik M, et al. (2023) Influence of illite and its amine modifications on the self-adhesive properties of silicone pressure-sensitive adhesives. *Materials* 16: 2879. https://doi.org/10.3390/ma16072879
10. Patel AK, Mathias JD, Michaud P (2013) Polysaccharides as adhesives: a critical review. *Rev Adhes Adhes* 1: 312–345. https://doi.org/10.7569/RAA.2013.097310
11. Li M, Zhou H, Zhang Y, et al. (2018) Effect of defects on thermal conductivity of graphene/epoxy nanocomposites. *Carbon* 130: 295–303. https://doi.org/10.1016/J.CARBON.2017.12.110
12. Wanasingha N, Dutta NK, Choudhury NR (2021) Emerging bioadhesives: from traditional bioactive and bioinert to a new biomimetic protein-based approach. *Adv Colloid Interface* 296: 102521. https://doi.org/10.1016/j.cis.2021.102521
13. Lutz TM, Kimna C, Casini A, et al. (2022) Bio-based and bio-inspired adhesives from animals and plants for biomedical applications. *Mater Today Bio* 13: 100203. https://doi.org/10.1016/j.mtbio.2022.100203
14. Wang Y, Naleway SE, Wang B (2020) Biological and bioinspired materials: structure leading to functional and mechanical performance. *Bioact Mater* 5: 745–757. https://doi.org/10.1016/j.bioactmat.2020.06.003
15. Bharti S (2018) Adhesives and adhesion technologies: a critical review, In: Sohel A, Habibpour S, Safari M, *American Journal of Polymer Science and Technology*, SciencePG, 4: 36–41. https://doi.org/10.11648/j.ajpst.20180401.13
16. Pocius AV (2002) Adhesion, In: *Kirk-Othmer Encyclopedia of Chemical Technology,* Wiley. https://doi.org/10.1002/0471238961.0104080516150309.a02
17. Pocius AV (2012) *Adhesion and Adhesives Technology: An Introduction: Third Edition*. https://doi.org/10.3139/9783446431775
18. Cui C, Liu W (2021) Recent advances in wet adhesives: adhesion mechanism, design principle and applications. *Prog Polym Sci* 116: 101388. https://doi.org/10.1016/j.progpolymsci.2021.101388
19. Dinte E, Sylvester B (2017) Adhesives: applications and recent advances, In: Özer H, *Applied Adhesive Bonding in Science and Technology* InTechOpen, 119–134. https://doi.org/10.5772/intechopen.71854
20. Li D, Chen J, Wang X, et al. (2020) Recent advances on synthetic and polysaccharide adhesives for biological hemostatic applications. *Front Bioeng Biotech* 8: 926. https://doi.org/10.3389/fbioe.2020.00926
21. Lin KT, Wang A, Nguyen AB, et al. (2021) Recent advances in hydrogels: ophthalmic applications in cell delivery, vitreous substitutes, and ocular adhesives. *Biomedicines* 9: 1203. https://doi.org/10.3390/biomedicines9091203
22. Monteiro LPG, Rodrigues JMM, Mano JF (2023) In situ generated hemostatic adhesives: from mechanisms of action to recent advances and applications. *Biomater Adv* 155: 213670. https://doi.org/10.1016/j.bioadv.2023.213670
23. Barash M, Reshef A, Brauner P (2010) The use of adhesive tape for recovery of DNA from crime scene items. *J Forensic Sci* 55: 1058–1064. https://doi.org/10.1111/j.1556-4029.2010.01416.x

24. Noureddine M, Bailey JA (2016) A protocol for the recovery of STR DNA from fingerprints developed on the adhesive side of duct tape. *J Forensic Ident* 66: 527–535. Available from: https://www.proquest.com/openview/14690c38ed91ced87c4d5ba926eded9a/1?pq-origsite=gscholar&cbl=29772.
25. Silvia AL, Tom KR, Breslin K, et al. (2023) A comparison of two methods for the recovery of fingerprint DNA on adhesive tapes. *Forensic Genomics* 3: 112–122 https://doi.org/10.1089/forensic.2023.0010
26. Kostyuk A, Ignatenko V, Smirnova N, et al. (2015) Rheology and adhesive properties of filled PIB-based pressure-sensitive adhesives. I. Rheology and shear resistance. *J Adhes Sci Technol* 29: 1831–1848. https://doi.org/10.1080/01694243.2014.980616
27. Victor AU, Benjamin DM, Haldar R, et al. (2021) Shear rheology and molecular properties of biobased adhesives through molecular dynamics simulation. *Physics Access* 1: 8–13. https://doi.org/10.47514/phyaccess.2021.1.1.002
28. Kumar A, Kaladharan K, Tseng FG (2021) Nanomaterials: Surface functionalization, modification, and applications. Nanomater Biomed Appl2021: 405–438. https://doi.org/10.1007/978-981-33-6252-9_14
29. Makvandi P, Iftekhar S, Pizzetti F, et al. (2021) Functionalization of polymers and nanomaterials for water treatment, food packaging, textile and biomedical applications: a review. *Environ Chem Lett* 19: 583–611. https://doi.org/10.1007/s10311-020-01089-4
30. Kartiwa A, Miraprahesti R, Sovani I, et al. (2017) Albumen glue, new material for conjunctival graft surgery, an animal experiment, *IOP Conference Series: Materials Science and Engineering,* IOP Publishing, 172: 012003. https://doi.org/10.1088/1757-899X/172/1/012003
31. Knaus J, Schaffarczyk D, Cölfen H (2020) On the future design of bio-inspired polyetheretherketone dental implants. *Macromol Biosci* 20: 1900239. https://doi.org/10.1002/mabi.201900239
32. Chen D, Li W, Liu J, et al. (2025) Bio-inspired proton relay for promoting continuous 5-hydroxymethylfurfural electrooxidation in a flowing system. *Energ Environ Sci* 18: 3120–3128. https://doi.org/10.1039/D4EE05745G
33. Nan J, Yang S, Huang G, et al. (2025) Radical-mediated chemo-divergent recyclization of 1,2,3,4-benzothiatriazine-1,1-dioxides: alkyl migration and de-aromatization. *Chem Commun* 61: 5503–5506. https://doi.org/10.1039/D5CC00008D
34. Ryu JH, Hong S, Lee H (2015) Bio-inspired adhesive catechol-conjugated chitosan for biomedical applications: a mini review. *Acta Biomater* 27: 101–115. https://doi.org/10.1016/j.actbio.2015.08.043
35. Li J, Yu X, Martinez EE, et al. (2022) Emerging biopolymer-based bioadhesives. *Macromol Biosci* 22: 2100340. https://doi.org/10.1002/mabi.202100340
36. Li W, Yang X, Lai P, et al. (2022) Bio-inspired adhesive hydrogel for biomedicine—principles and design strategies. *Smart Medicine* 1: e20220024. https://doi.org/10.1002/SMMD.20220024
37. Yang J, Hong K, Hao Y, et al. (2024) Mica/nylon composite nanofiber film based wearable triboelectric sensor for object recognition. *Nano Energy* 129: 110056. https://doi.org/10.1016/j.nanoen.2024.110056
38. Calvez I, Garcia R, Koubaa A, et al. (2024) Recent advances in bio-based adhesives and formaldehyde-free technologies for wood-based panel manufacturing. *Curr For Rep* 10: 386–400. https://doi.org/10.1007/s40725-024-00227-3

39. Eisen A, Bussa M, Röder H (2020) A review of environmental assessments of biobased against petrochemical adhesives. *J Clean Prod* 277: 124277. https://doi.org/10.1016/j.jclepro.2020.124277
40. Sun J, Li L, Cheng H, et al. (2018) Preparation, characterization and properties of an organic siloxane-modified cassava starch-based wood adhesive. *J Adhes* 94: 278–293. https://doi.org/10.1080/00218464.2016.1268958
41. Omura S, Kawazoe Y, Uemura D (2017) Development of a novel adhesive composed of all-natural components. *Int J Adhes Adhes* 74: 35–39. https://doi.org/10.1016/j.ijadhadh.2016.12.009
42. Arias A, Feijoo G, Moreira MT (2021) Evaluation of starch as an environmental-friendly bioresource for the development of wood bioadhesives. *Molecules* 26: 4526. https://doi.org/10.3390/molecules26154526
43. Omidian H, Wilson RL, Babanejad N (2023) Bioinspired polymers: transformative applications in biomedicine and regenerative medicine. *Life* 13: 1673. https://doi.org/10.3390/life13081673
44. Wu J, Hua Z, Liu G (2025) Supramolecular adhesives inspired from adhesive proteins and nucleic acids: molecular design, properties, and applications. *Soft Matter* 21: 324–341. https://doi.org/10.1039/D4SM01220H
45. Mahdavi A, Ferreira L, Sundback C, et al. (2008) A biodegradable and biocompatible gecko-inspired tissue adhesive. *P Natl A Sci* 105: 2307–2312. https://doi.org/10.1073/pnas.0712117105
46. Spotnitz WD (2014) Fibrin sealant: the only approved hemostat, sealant, and adhesive—a laboratory and clinical perspective. *ISRN Surgery* 2014: 203943. https://doi.org/10.1155/2014/203943
47. Sancaktar E (2018) Classification of adhesive and sealant materials, In: *Handbook of Adhesion Technology,* 2 Eds., Cham: Springer, 283–317. https://doi.org/10.1007/978-3-319-55411-2_12
48. Karuppasamy A, Rexliene J, Dhandapani A, et al. (2023) Recyclability of lightweight and sustainable materials, In: Rangappa SM, Doddamani S, Siengchin S, et al. *Lightweight and Sustainable Composite Materials: Preparation, Properties and Applications,* Woodhead Publishing, 79–96. https://doi.org/10.1016/B978-0-323-95189-0.00005-6
49. Petersen A, Chu NQ, Fitzgerald DM, et al. (2021) Sustainable glycerol terpolycarbonates as temporary bioadhesives. *Biomater Sci* 9: 8366–8372. https://doi.org/10.1039/d1bm00995h
50. Rao Y, Wan G (2023) Sustainable adhesives: bioadhesives, chemistry, recyclability, and reversibility, In: *Advances in Structural Adhesive Bonding,* 2 Eds, Woodhead Publishing, 953–985. https://doi.org/10.1016/B978-0-323-91214-3.00008-9
51. Comyn J (2005) What are adhesives and sealants and how do they work?, In: *Adhesive Bonding: Science, Technology, and Applications,* Woodhead Publishing, 23–51 https://doi.org/10.1533/9781845690755.1.23
52. Comyn J (2021) What are adhesives and sealants and how do they work?, In: *Adhesive Bonding: Science, Technology and Applications,* Woodhead Publishing, 41–778 https://doi.org/10.1016/B978-0-12-819954-1.00003-4
53. Possart W (2005) Front Matter, In: *Adhesion: Current Research and Applications,* Wiley. https://doi.org/10.1002/3527607307.fmatter
54. Adekunle K, Åkesson D, Skrifvars M (2010) Synthesis of reactive soybean oils for use as a biobased thermoset resin in structural natural fiber composites. *J Appl Polym Sci* 115: 3137–3145. https://doi.org/10.1002/app.31411

55. Agbogo, UV, Rifore, BS, Arum, CT (2024). carbon quantum dots for wastewater treatment: present progress and prospects. *FUDMA J Sci* 8: 93–102. https://doi.org/10.33003/fjs-2024-0801-2208
56. Tang C, Zhou K, Zhu Y, et al. (2022) Collagen and its derivatives: From structure and properties to their applications in food industry. *Food Hydrocolloid* 131: 107748.. https://doi.org/10.1016/j.foodhyd.2022.107748
57. Nascimento LGL, Casanova F, Silva NFN, et al. (2020) Casein-based hydrogels: a mini-review. *Food Chem* 314: 126063. https://doi.org/10.1016/j.foodchem.2019.126063
58. Ye J, Fu S, Zhou S, et al. (2020) Advances in hydrogels based on dynamic covalent bonding and prospects for its biomedical application. *Eur Polym J* 139: 110024. https://doi.org/10.1016/j.eurpolymj.2020.110024
59. Yang Y, Xu Q, Wang X, et al. (2024) Casein-based hydrogels: advances and prospects. *Food Chem* 2024: 138956. https://doi.org/10.1016/j.foodchem.2024.138956
60. Ebnesajjad S (2011) Characteristics of adhesive materials, In: *Handbook of adhesives and surface preparation, Technology, Applications and Manufacturing,* William Andrew Publishing, 2011: 137–183. https://doi.org/10.1016/B978-1-4377-4461-3.10008-2
61. Konoplin AY, Baurova NI (2022) Influence of adhesive materials on the characteristics of adhesive-welded joints. *Russ Metall* 2022: 1642–1645. https://doi.org/10.1134/S0036029522130146
62. Sanghani-Kerai A, Coathup M, Brown R, et al. (2020) The development of a novel autologous blood glue aiming to improve osseointegration in the bone-implant interface. *Bone Joint Res* 9: 402–411. https://doi.org/10.1302/2046-3758.97.BJR-2019-0073.R3
63. Hao Y, Yang J, Zhu X, et al. (2025) PEO/cysteine composite nanofiber-based triboelectric nanogenerators for harvesting tiny mechanical energy. *J Mater Chem A* 13: 1853–1862. https://doi.org/10.1039/D4TA06845A
64. Fiorda FA, Soares Júnior MS, Silva FA, et al. (2013) Cassava bagasse flour: byproduct utilization and comparison with cassava starch. *Pesqui Agropecu Trop* 43: 408–416. https://revistas.ufg.br/pat/article/view/23381
65. Monroy Y, Rivero S, García MA (2019) Sustainable panels designed based on modified cassava starch bioadhesives and wood processing byproducts. *Ind Crop Prod* 137: 171–179. https://doi.org/10.1016/j.indcrop.2019.04.062
66. Ochoa N, Bello M, Sancristóbal J, et al. (2013) Modified cassava starches as potential corrosion inhibitors for sustainable development. *Mater Res* 16: 1209–1219. https://doi.org/10.1590/S1516-14392013005000126
67. Pizzi A (2014) Types, processing and properties of bioadhesives for wood and fibers, In: *Advances in Biorefineries: Biomass and Waste Supply Chain Exploitation,* Woodhead Publishing, 736–770. https://doi.org/10.1533/9780857097385.2.736
68. Sun J, Han J, Wang F, et al. (2022). Bioengineered protein-based adhesives for biomedical applications. *Chem–Eur J* 28: e202102902. https://doi.org/10.1002/chem.202102902
69. Ghani RSM, Lee MD, Razali SM (2023) A comprehensive review of sustainable benefit of cassava starch and its potential in wood-based and lignocellulosic materials, Journal of Emerging Technologies and Industrial Applications, 2023. Available from: https://jetia.ttasmbot.org.my/index.php/jetia/article/view/12.

70. Xu Q, Wen J, Wang Z (2016) Preparation and properties of cassava starch-based wood adhesives. *BioResources* 11: 6756–6767. https://doi.org/10.15376/biores.11.3.6756-6767
71. Munir H, Nasir R, Gull S, et al. (2023) Introduction to natural gums, In: *Natural Gums: Extraction, Properties, and Applications,* Elsevier: 3–21. https://doi.org/10.1016/B978-0-323-99468-2.00001-2
72. Hamdani AM, Wani IA, Bhat NA (2019) Sources, structure, properties and health benefits of plant gums: a review. *Int J Biol Macromol* 135: 46–61. https://doi.org/10.1016/j.ijbiomac.2019.05.103
73. Kao FJ, Manivannan G, Sawan SP (1997) UV curable bioadhesives: copolymers of N-vinyl pyrrolidone. *J Biomed Mater Res* 38: 191–196. https://doi.org/10.1002/(SICI)1097-4636(199723)38:3<191::AID-JBM2>3.0.CO;2-K
74. Conner AH, Baumann MGD (2003) Carbohydrate polymers as adhesives, In: Pizzi A, Mittal KL, *Handbook of Adhesive Technology, Revised and Expanded,* 2 Eds., CRC Press, 495–510. https://doi.org/10.1201/9780203912225.ch22
75. Sellers JrT (2001) Wood adhesive innovations and applications in North America. *Forest Prod J* 51: 12–22. Available from: https://www.proquest.com/openview/947c04b3d9f32f6ef55b65769cb87641/1?pq-origsite=gscholar&cbl=25222.
76. Feng CW, Keong CW, Hsueh YP, et al. (2005) Modeling of long-term creep behaviour of structural epoxy adhesives. *Int J Adhes Adhes* 25: 427–436. https://doi.org/10.1016/j.ijadhadh.2004.11.009
77. Moini N, Khaghanipour M, Faridani F, et al. (2022) Green adhesives—past, present, and future outlook, In: Altalhi T, Inamuddin, *Green Sustainable Process for Chemical and Environmental Engineering and Science: Green Composites: Preparation, Properties and Allied Applications,* Elsevier, 341–372. https://doi.org/10.1016/B978-0-323-99643-3.00006-1
78. Akaranta O, Wankasi D (1999) Wood adhesives from peanut skin tannin-formaldehyde resins modified with phenols. *Pigm Resin Technol* 28: 293–296.. https://doi.org/10.1108/03699429910294346
79. Adams RD (1990) The non-destructive evaluation of bonded structures. *Constr Build Mater* 4: 3–8. https://doi.org/10.1016/0950-0618(90)90011-O
80. da Silva LFM, Öchsner A, Adams RD (2011) Introduction to adhesive bonding technology, In *Handbook of Adhesion Technolog,* Berlin Heidelberg, Springer: 1–7. https://doi.org/10.1007/978-3-642-01169-6_1
81. Budhe S, Banea MD, de Barros S, et al. (2017) An updated review of adhesively bonded joints in composite materials. *Int J Adhes Adhes* 72: 30–42. https://doi.org/10.1016/j.ijadhadh.2016.10.010
82. Wei Y, Jin X, Luo Q, et al. (2024) Adhesively bonded joints–a review on design, manufacturing, experiments, modeling and challenges. *Composites Part B: Engineering* 276: 111225. https://doi.org/10.1016/j.compositesb.2024.111225
83. Awaja F, Gilbert M, Kelly G, et al. (2009) Adhesion of polymers. *Prog Polym Sci* 34: 948–968. https://doi.org/10.1016/j.progpolymsci.2009.04.007
84. Hao Y, Zhang C, Su W, et al. (2024) Sustainable materials systems for triboelectric nanogenerator. *SusMat* 4: e244. https://doi.org/10.1002/sus2.244
85. Hao Y, Zhu X, Hong K, et al. (2025) Advanced sustainable triboelectric nanogenerators for biomedical and clinical applications: in vivo treatments, in vitro therapeutics, and assisted rehabilitations. *Chem Eng J* 2025: 161042. https://doi.org/10.1016/j.cej.2025.161042

86. Hao Y, Yang J, Niu Z, et al. (2023) High-output triboelectric nanogenerator based on L-cystine/nylon composite nanofiber for human bio-mechanical energy harvesting. *Nano Energy* 118: 108964. https://doi.org/10.1016/j.nanoen.2023.108964
87. Imam SH, Gordon SH, Mao L, et al. (2001) Environmentally friendly wood adhesive from a renewable plant polymer: characteristics and optimization. *Polym Degrad Stabil* 73: 529–533. https://doi.org/10.1016/S0141-3910(01)00114-8
88. Zhang Y, Ding L, Gu J, et al. (2015) Preparation and properties of a starch-based wood adhesive with high bonding strength and water resistance. *Carbohyd Polym* 115: 32–37. https://doi.org/10.1016/j.carbpol.2014.08.063
89. Wang Z, Li Z, Gu Z, et al. (2012) Preparation, characterization and properties of starch-based wood adhesive. *Carbohyd Polym* 88: 699–706. https://doi.org/10.1016/j.carbpol.2012.01.023
90. Xiong H, Wang Z, Fei P, et al. (2017) Effects of sucrose fatty acid esters on the stability and bonding performance of high amylose starch-based wood adhesive. *Int J Biol Macromol* 104: 846–853. https://doi.org/10.1016/j.ijbiomac.2017.06.090
91. Qiao Z, Gu J, Lv S, et al. (2016) Preparation and properties of normal temperature cured starch-based wood adhesive. *BioRes* 11: 4839–4849. https://doi.org/10.15376/biores.11.2.4839-4849
92. Qiao Z, Lv S, Gu J, et al. (2017) Influence of acid hydrolysis on properties of maize starch adhesive. *Pigm Resin Technol* 46: 148–155. https://doi.org/10.1108/PRT-10-2015-0105
93. Wu SJ, Zhao X (2023) Bioadhesive technology platforms. *Chem Rev* 123: 14084–14118. https://doi.org/10.1021/acs.chemrev.3c00380
94. Khadem E, Kharaziha M, Bakhsheshi-Rad HR, et al. (2022) Cutting-edge progress in stimuli-responsive bioadhesives: from synthesis to clinical applications. *Polymers* 14: 1709. https://doi.org/10.3390/polym14091709
95. Mehdizadeh M, Yang J (2013) Design strategies and applications of tissue bioadhesives. *Macromol Biosci* 13: 271–288. https://doi.org/10.1002/mabi.201200332
96. Guo J, Sun W, Kim JP, et al. (2018) Development of tannin-inspired antimicrobial bio adhesives. *Acta Biomater* 72: 35–44. https://doi.org/10.1016/j.actbio.2018.03.008
97. Zheng Y, Baidya A, Annabi N (2023) Molecular design of an ultra-strong tissue adhesive hydrogel with tunable multifunctionality. *Bioact Mater* 29: 214–229. https://doi.org/10.1016/j.bioactmat.2023.06.007
98. Zhu H, Tian J, Mao H, et al. (2021) Bio adhesives: current hotspots and emerging challenges. *Curr Opin Biomed Eng* 18: 100271. https://doi.org/10.1016/J.COBME.2021.100271
99. Amukarimi S, Ramakrishna S, Mozafari M (2021) Smart biomaterials—a proposed definition and overview of the field. *Curr Opin Biomed Eng* 19: 100311. https://doi.org/10.1016/j.cobme.2021.100311
100. Zheng Y, Shariati K, Ghovvati M, et al. (2023) Hemostatic patch with ultra-strengthened mechanical properties for efficient adhesion to wet surfaces. *Biomaterials* 301: 122240. https://doi.org/10.1016/j.biomaterials.2023.122240
101. Samal SS (2009) Role of temperature and carbon nanotube reinforcement on epoxy based nanocomposites. *J Miner Mater Charact Eng* 8: 25–36. https://doi.org/10.4236/jmmce.2009.81003
102. Burns M, Stellwagen SD (2021) The ties that stick: challenges and future promise in the field of bioadhesives. *Integr Comp Biol* 61: 1406–1410. https://doi.org/10.1093/icb/icab129

103. Kawalerczyk J, Siuda J, Mirski R, et al. (2020) Hemp flour as a formaldehyde scavenger for melamine-urea-formaldehyde adhesive in plywood production. *BioRes* 15: 4052–4064. https://doi.org/10.15376/biores.15.2.4052-4064
104. Mirski R, Kawalerczyk J, Dziurka D, et al. (2020) The application of oak bark powder as a filler for melamine-urea-formaldehyde adhesive in plywood manufacturing. *Forests* 11: 1249. https://doi.org/10.3390/f11121249
105. Yang M, Rosentrater KA (2020) Life cycle assessment of urea-formaldehyde adhesive and phenol-formaldehyde adhesives. *Environ Process* 7: 553–561. https://doi.org/10.1007/s40710-020-00432-9
106. Kotiyan PN, Vavia PR (2002) Synthesis and characterization of an acrylate pressure-sensitive adhesive for transdermal drug delivery. *Polym Advan Technol* 13: 137–143. https://doi.org/10.1002/pat.193
107. Du H, Zhao Y, Li Q, et al. (2008) Synthesis and characterization of waterborne polyurethane adhesive from MDI and HDI. *J Appl Polym Sci* 110: 1396–1402. https://doi.org/10.1002/app.28805
108. Qiao L, Easteal AJ, Bolt CJ, et al. (1999) The effects of filler materials on poly (vinyl acetate) emulsion wood adhesives. *Pigm Resin Technol* 28: 326–330. https://doi.org/10.1108/03699429910302300
109. Baughman RH, Zakhidov AA, de Heer WA (2002) Carbon nanotubes--the route toward applications. *Science* 297: 787–792. https://doi.org/10.1126/science.1060928
110. Zhou YX, Wu PX, Cheng ZY, et al. (2008) Improvement in electrical, thermal, and mechanical properties of epoxy by filling carbon nanotube. *Express Polym Lett* 2: 40–48. https://doi.org/10.3144/expresspolymlett.2008.6
111. Ajayan PM, Schadler LS, Giannaris C, et al. (2000) Single-walled carbon nanotube–polymer composites: strength and weakness. *Adv Mater* 12: 750–753. https://doi.org/10.1002/(SICI)1521-4095(200005)12:10<750:AID-ADMA750>3.0.CO;2-6
112. Deshmukh S, Ounaies Z (2010) Active single walled carbon nanotube–polymer composites, *IUTAM Symposium on Multi-Functional Material Structures and Systems: Proceedings of the the IUTAM Symposium on Multi-Functional Material Structures and Systems*, Bangalore, India, Springer Netherlands, 103–110. https://doi.org/10.1007/978-90-481-3771-8_11
113. Grady BP, Paul A, Ford WT (2009) Polymer dynamics in single-walled carbon nanotube-polymer composites, *Annual Technical Conference - ANTEC, Conference Proceedings*, *1*. https://doi.org/10.1002/app.39884
114. Kang J, Al-Sabah S, Théo R (2020) Effect of single-walled carbon nanotubes on strength properties of cement composites. *Materials* 13: 1305. https://doi.org/10.3390/ma13061305
115. Kwon Y, Yim B, Kim J, et al. (2011) Dispersion, hybrid interconnection and heat dissipation properties of functionalized carbon nanotubes in epoxy composites for electrically conductive adhesives (ECAs). *Microelectron Reliab* 51: 812–818. https://doi.org/10.1016/j.microrel.2010.11.005
116. Sandler J, Shaffer MSP, Prasse T, et al. (1999) Development of a dispersion process for carbon nanotubes in an epoxy matrix and the resulting electrical properties. *Polymer* 40: 5967–5971. https://doi.org/10.1016/S0032-3861(99)00166-4

117. Sulay ZK, Victor AU, Obed B, et al. (2015) Kinetics and thermodynamic study of inhibition potentials by ethoxyethane extracts of cochlospermum tinctorium for the oxoacid corrosion of mild steel. *Int J Mater Chem* 5: 64–76. https://doi.org/10.5923/j.ijmc.20150503.03
118. Yu W, He H, Cheng N, et al. (2009) Preparation and experiments for a novel kind of foundry core binder made from modified potato starch. *Mater Design* 30: 210–213. https://doi.org/10.1016/j.matdes.2008.03.017
119. Sadare OO, Daramola MO, Afolabi AS (2020) Synthesis and performance evaluation of nanocomposite soy protein isolate/carbon nanotube (SPI/CNTs) adhesive for wood applications. *Int J Adhes Adhes* 100: 102605. https://doi.org/10.1016/j.ijadhadh.2020.102605
120. Li J, Yu X, Martinez EE, et al. (2022) Emerging biopolymer-based bioadhesives. *Macromol Biosci* 22: 2100340. https://doi.org/10.1002/mabi.202100340

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