Review Article

Chitosan Biopolymer as a Versatile Heterogeneous Catalyst: A Mini-Review on Recent Advances

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In the context of green chemistry, the design and implementation of environmentally benign catalytic systems have become a central research priority. Chitosan, a linear polysaccharide derived from chitin, exhibits a unique combination of intrinsic properties including biodegradability, biocompatibility, and tunable acidity that position it as a compelling alternative to conventional homogeneous and heterogeneous catalysts. Notably, the amphoteric nature of chitosan, arising from its ability to function as both a Brønsted acid and a base, along with its structurally tunable active sites, renders it a versatile platform for a broad spectrum of organic transformations. Beyond its inherent acid–base bifunctionality, the catalytic versatility of chitosan is substantially enhanced through chemical modification strategies. These include covalent grafting with functional groups (e.g., Schiff bases, ionic liquids) and encapsulation or stabilization of metal nanoparticles within the chitosan matrix. Such modifications significantly improve catalytic activity, selectivity, and recyclability, enabling efficient catalysis of diverse reactions, including esterification, ring-opening polymerization, and cross-coupling reactions (e.g., Suzuki, Heck, and Sonogashira). Consequently, chitosan-based catalytic systems have emerged as valuable tools in both organic synthesis and sustainable materials science. A further advantage of chitosan lies in its ability to promote reactions under mild conditions, frequently at ambient temperature and pressure. This characteristic minimizes energy consumption and suppresses the formation of hazardous byproducts, aligning directly with the principles of green chemistry. Collectively, the structural versatility, chemical stability, and tunable physicochemical properties of chitosan underscore its potential to contribute significantly to the development of next-generation sustainable catalysts. Given these attributes, this mini-review aims to provide a concise yet comprehensive overview of recent advances in the application of chitosan as a heterogeneous (or

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hybrid) catalyst in organic reactions, with an emphasis on its structural modifications and catalytic mechanisms.

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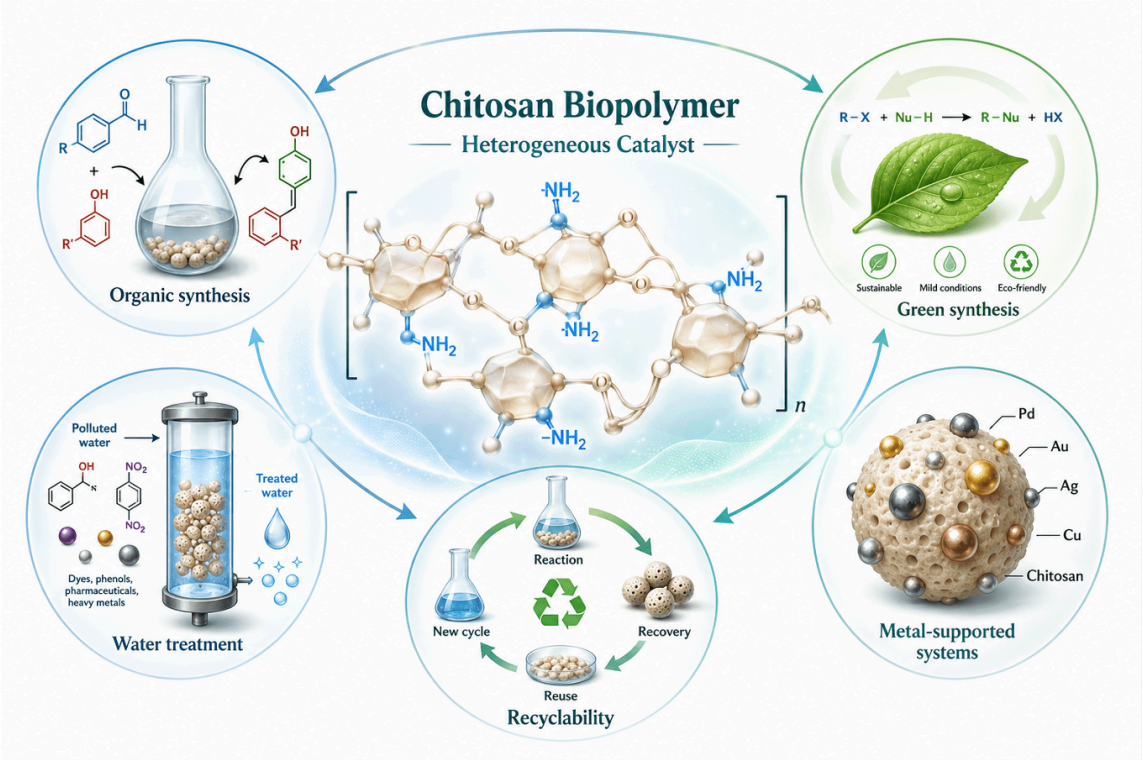
Graphical Abstract



1. Introduction

In the realm of green chemistry, the design, synthesis, and implementation of environmentally benign catalytic systems have garnered significant attention as a direct response to the growing demand for sustainable chemical processes [1][2]. Among the various bio-based materials explored in this context, chitosan a naturally occurring linear polysaccharide derived from chitin via partial alkaline deacetylation has emerged as an exceptionally promising candidate due to its unique combination of physicochemical properties, ecological compatibility, and structural versatility [3][4][5]. Derived predominantly from crustacean shells, chitosan possesses a set of intrinsic attributes including biodegradability,

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biocompatibility, non-toxicity, film-forming ability, and tunable acidity, all of which collectively render it an appealing alternative to conventional homogeneous and heterogeneous catalysts [6][7][8].

The catalytic prowess of chitosan is fundamentally rooted in its highly hydrophilic nature, which arises from the dense population of reactive primary amino (-NH_2) and hydroxyl (-OH) functional groups distributed along its polymeric backbone [9][10]. These functional moieties are not merely passive structural elements but actively participate in catalytic cycles: they can act as Lewis bases by donating electron pairs to facilitate nucleophilic additions and eliminations, or as Brønsted bases by abstracting protons to promote acid-catalyzed transformations such as aldol condensations and Knoevenagel reactions [11][12][13]. Importantly, the degree of deacetylation (DDA) which represents the molar percentage of glucosamine units relative to the total number of *N*-acetylglucosamine residues plays a critically decisive role in modulating the density of free amino groups, thereby directly influencing the surface charge, hydrophilicity, basicity, and overall catalytic activity of the biopolymer [14][15]. A higher DDA typically results in a greater number of exposed amino groups, which enhances both metal coordination capacity and proton abstraction ability, leading to superior catalytic performance [16][17].

Beyond its intrinsic acid–base bifunctionality, the catalytic versatility of chitosan can be substantially extended and fine-tuned through targeted chemical modifications and structural engineering strategies [18][19]. These include, but are not limited to, covalent conjugation with specific functional ligands such as Schiff bases, crown ethers, thiourea derivatives, or ionic liquid moieties, as well as the physical or chemical encapsulation of metal nanoparticles (e.g., palladium, gold, silver, copper, and ruthenium) within the three-dimensional chitosan matrix [20][21]. Such modifications not only enhance catalytic activity, selectivity, and turnover frequency (TOF) but also significantly improve recyclability, thermal stability, and resistance to metal leaching during successive reaction cycles [22][23]. As a direct consequence of these enhancements, modified chitosan-based catalytic systems have been successfully employed across an impressively broad range of organic transformations, including transesterification, ring-opening and atom-transfer radical polymerization (ATRP), Suzuki-Miyaura, Heck, and Sonogashira cross-coupling reactions, as well as oxidation, reduction, and multicomponent reactions [24][25][26]. Collectively, these capabilities establish chitosan as a truly valuable, multifunctional platform in both modern organic synthesis and sustainable materials science [27][28].

The green credentials of chitosan further consolidate its position as a preferred catalyst in the context of sustainable chemistry [29]. Being derived entirely from renewable biological sources primarily seafood

processing waste and inherently biodegradable into non-toxic oligosaccharides, chitosan substantially mitigates the environmental concerns typically associated with conventional transition metal-based or organometallic catalysts, many of which suffer from toxicity, non-renewability, and difficult recovery [30][31][32]. Moreover, one of the most practically significant advantages of chitosan lies in its demonstrated ability to function effectively under exceptionally mild reaction conditions, frequently at ambient temperature and atmospheric pressure, in green solvents such as water or ethanol, and often in the absence of inert atmospheres [33][34]. This mild operational profile not only minimizes energy consumption and reduces overall process costs but also effectively suppresses the formation of hazardous byproducts, thereby improving overall reaction selectivity and atom economy [35][36][37].

As research efforts in the field of chitosan-based catalysis continue to advance at an accelerating pace, the practical applications of this versatile biopolymer are rapidly expanding across a diverse array of industrial and academic domains [38][39]. These include the synthesis of high-value pharmaceutical intermediates and active pharmaceutical ingredients (APIs), the production of fine chemicals and agrochemicals, the fabrication of functional biopolymers and biodegradable plastics, and even the conversion of renewable feedstocks into advanced biofuels [40][41]. In each of these areas, chitosan demonstrates its substantial potential to contribute meaningfully to the ongoing revolution in green chemistry [42]. Ultimately, the extraordinary versatility, inherent sustainability, structural stability, and remarkably tunable physicochemical properties of chitosan position it as an indispensable and future-proof catalytic material for the transition toward a greener, more sustainable chemical industry [43][44].

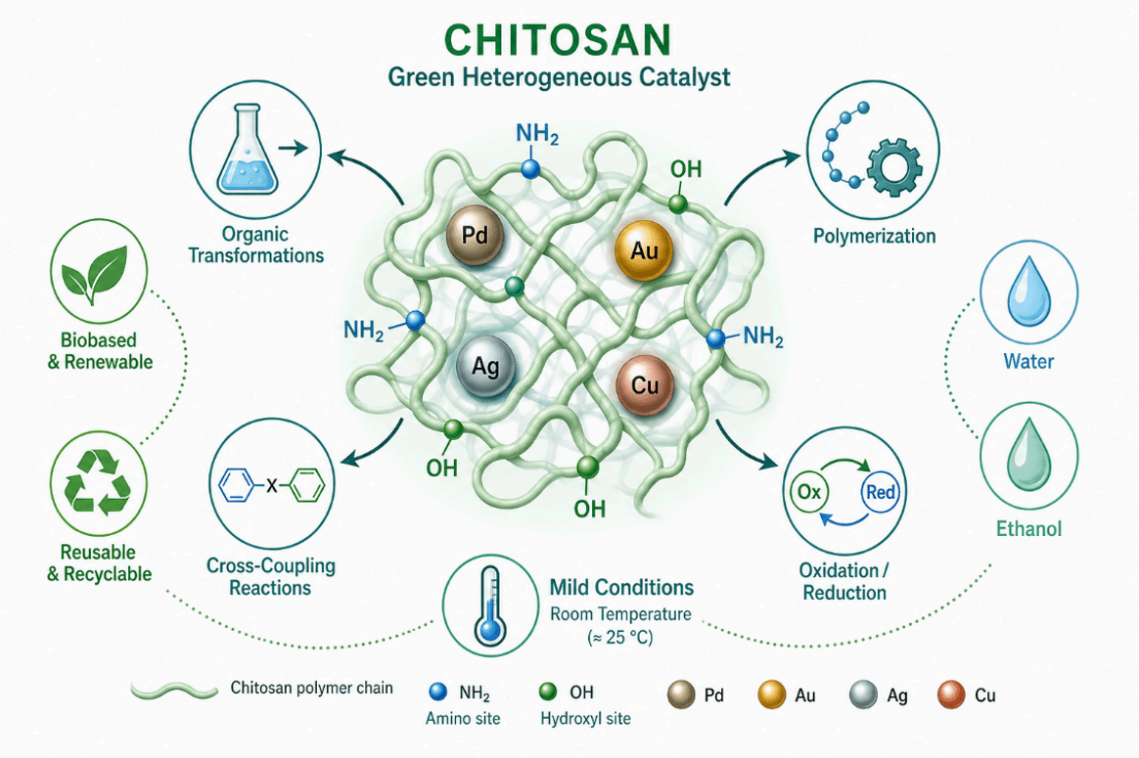

Figure 1. Chitosan-based heterogeneous catalysis for green and sustainable organic transformations.

2. Homogeneous vs. Heterogeneous Catalysts

A catalyst is a substance that increases the rate of a chemical reaction without being consumed. Homogeneous catalysts exist in the same phase as reactants (typically liquid), offering high activity and selectivity but suffering from difficult separation and poor recyclability [45][46][47]. Heterogeneous catalysts exist in a different phase (usually solid), enabling easy recovery by filtration or centrifugation. In organic reactions, heterogeneous catalysts are widely used for hydrogenation, cross-coupling (e.g., Suzuki, Heck), and oxidation reactions due to their thermal stability and reusability [48][49][50]. Key advantages of heterogeneous catalysts include simple separation from products, minimal metal leaching, and multiple cycles of reuse without significant activity loss [51][52][53]. Photocatalysts, a subclass of heterogeneous systems, harness light energy to drive redox reactions for degradation of pollutants, water splitting, and CO_2 reduction [54][55][56]. Recovery of heterogeneous catalysts is achieved through filtration, magnetic separation, or centrifugation, making them highly practical for industrial applications [57][58]. From a green chemistry perspective, heterogeneous catalysts reduce waste, eliminate

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toxic residues, and minimize energy consumption, aligning perfectly with sustainable and environmentally benign processes [59][60].

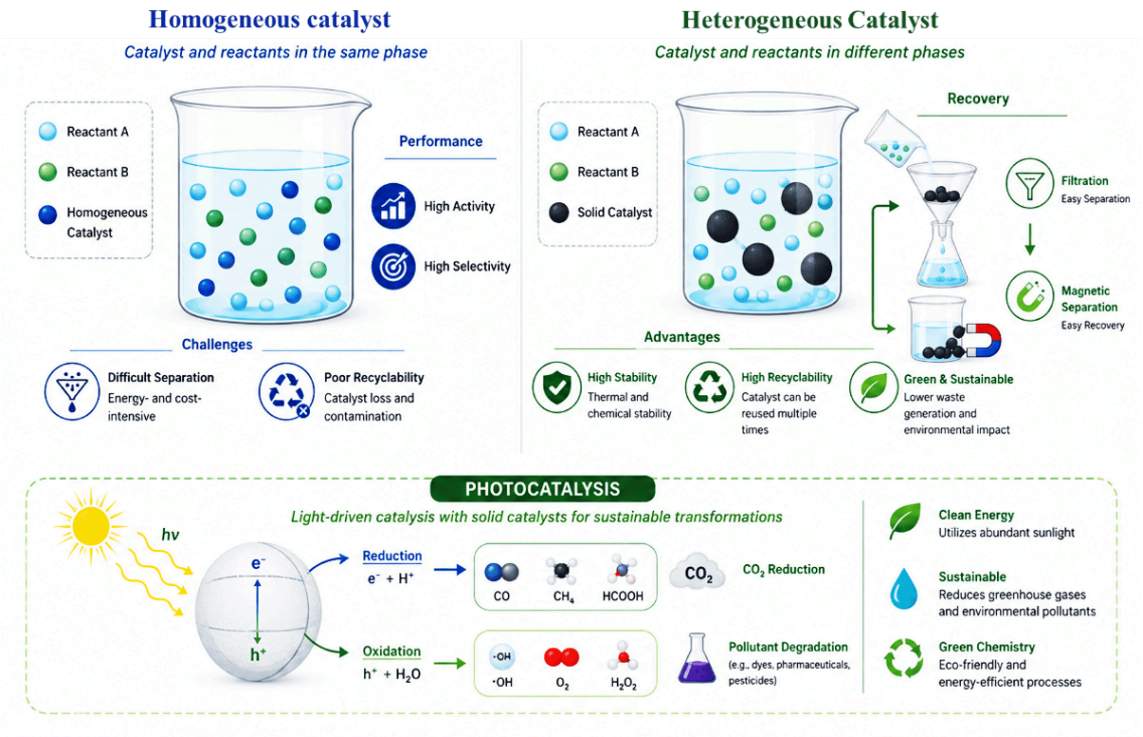

Figure 2. Schematic comparison between homogeneous and heterogeneous catalysts.

3. Historical Development of Chitosan: From Discovery to Green Catalyst

The history of chitosan dates back to 1859, when French physiologist and chemist Charles Rouget first discovered the substance by heating chitin in a concentrated alkaline solution [61]. Rouget observed that this treatment modified the solubility properties of chitin, producing a new material that became soluble in organic acids. However, it was not until 1894 that German chemist Felix Hoppe-Seyler formally named this deacetylated product "chitosan" [62][63]. For much of the early 20th century, chitosan remained a laboratory curiosity with limited practical applications due to the lack of characterization techniques and scalable production methods. A major breakthrough occurred in the 1930s–1940s when researchers elucidated the chemical structure of chitin and chitosan, identifying the linear copolymer of glucosamine and N-acetylglucosamine units linked by β-(1→4) glycosidic bonds [64][65][66]. The 1970s marked a

turning point with growing environmental awareness, shifting attention toward biodegradable and renewable biopolymers. During this decade, the first catalytic applications of chitosan emerged, primarily in metal ion chelation and enzyme immobilization. The 1990s witnessed rapid expansion in chitosan research, particularly in biomedical and pharmaceutical fields, but catalytic applications remained underexplored. The early 2000s brought a paradigm shift, as green chemistry principles gained global prominence [67][68][69]. Researchers recognized chitosan's unique acid–base bifunctionality and its potential as a sustainable alternative to conventional metal-based catalysts. Since 2010, chitosan has evolved remarkably as a versatile heterogeneous catalyst, with significant advances in functionalization strategies, metal nanoparticle stabilization, and application in diverse organic transformations including cross-coupling, multicomponent reactions, and biomass conversion [70][71][72][73]. Today, chitosan stands at the forefront of sustainable catalysis, with ongoing research focused on magnetic composites, photoactive derivatives, and continuous-flow systems, positioning it as a cornerstone of next-generation green chemistry [74][75].

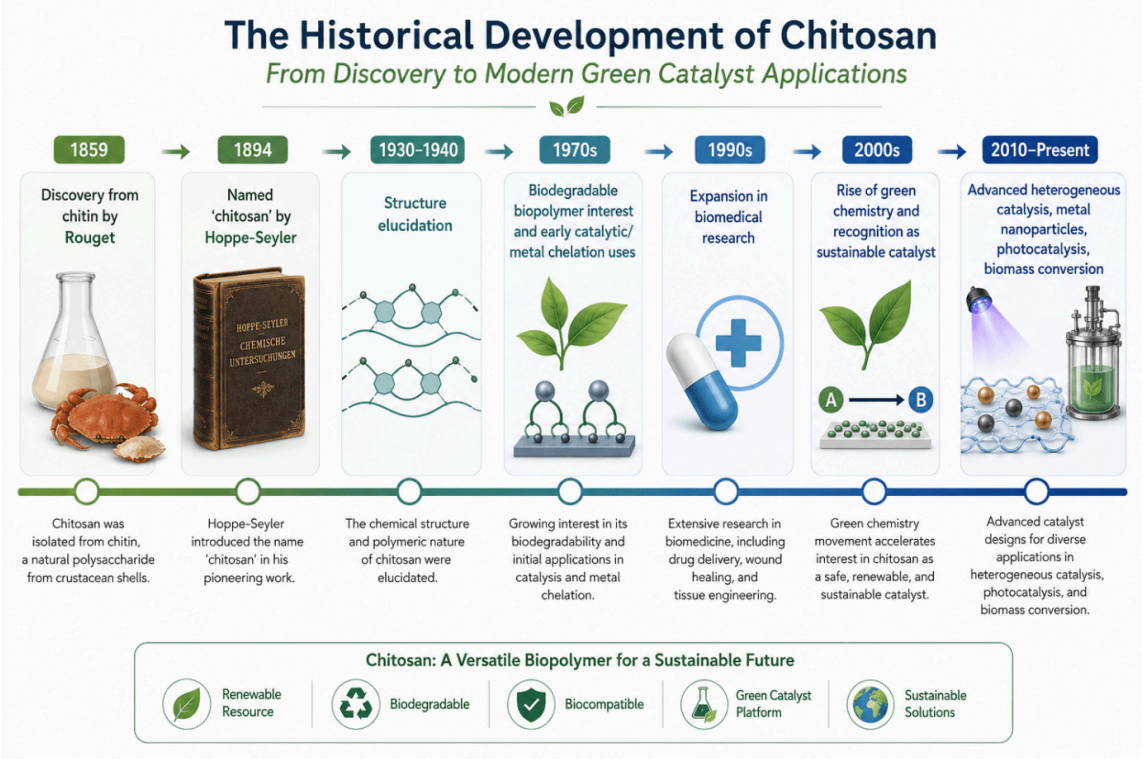

Figure 3. Historical evolution of chitosan from its discovery in the nineteenth century to its emergence as a versatile green heterogeneous catalyst.

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4. Physicochemical Properties

Chitosan exhibits several distinct physicochemical properties that contribute to its versatility and applications. These properties include:

4.1. Hydrophilicity

Chitosan is highly hydrophilic due to the presence of multiple hydroxyl (-OH) and amine ($-NH_2$) groups along its backbone. This characteristic allows it to easily absorb water and swell considerably in aqueous solutions, forming hydrated gels. This hydrophilicity makes chitosan suitable for various applications in pharmaceutical (drug carriers), food (edible coatings), and environmental (heavy metal adsorption) sectors [76].

4.2. Cationic Nature

Chitosan has a net positive charge at pH values below its pKa (≈6.3-6.5), due to the protonation of its amino groups ($-NH_3^+$). This enables it to interact electrostatically with negatively charged molecules such as proteins, lipids, nucleic acids (DNA/RNA), and anionic polysaccharides. This property is utilized in applications such as drug delivery (gene complexation), wound healing (binding to cell membranes), and antimicrobial activity (interaction with bacterial membranes) [77][78].

4.3. Biodegradability

Chitosan is easily biodegradable under natural conditions (in vivo and in the environment) by specific enzymes including lysozyme, chitinase, and chitosanase. It breaks down into harmless byproducts, primarily non-toxic oligosaccharides and *N*-acetylglucosamine. This biodegradability makes chitosan an attractive option for environmentally friendly and biomedical applications (e.g., temporary scaffolds, sutures) [79][80].

4.4. Solvent Compatibility & pH Sensitivity

Chitosan is soluble in acidic aqueous solutions (e.g., acetic acid, lactic acid, citric acid) due to protonation of its amino groups, but it is insoluble in neutral or alkaline solutions (pH > 6.5). This reversible solubility can be used to control the release of encapsulated compounds (pH-triggered release) or to modify the properties of chitosan-based materials such as films, hydrogels, and nanoparticles [81].

4.5. Mucoadhesive Property

Due to its cationic nature, chitosan adheres strongly to negatively charged mucosal surfaces (e.g., gastrointestinal, nasal, buccal mucosa). This property enhances the residence time and bioavailability of drugs delivered via mucosal routes [82].

4.6. Linking Physicochemical Properties to Catalytic Function

Crucially, each of the above physicochemical properties directly translates into a specific catalytic advantage. The hydrophilicity of chitosan facilitates the adsorption of polar reactants (e.g., alcohols, aldehydes, carboxylic acids) onto its surface, dramatically increasing local reactant concentration near the active sites a key prerequisite for efficient heterogeneous catalysis [83][84][85]. The cationic nature (protonated $-NH_3^+$ at pH < pKa) enables electrostatic anchoring of anionic transition metal complexes (e.g., $PdCl_4^{2-}$, $AuCl_4^-$) or negatively charged substrates, serving as a built-in immobilization platform without requiring external ligands [86][87][88]. The biodegradability ensures that spent catalyst does not accumulate as persistent plastic waste, aligning with end-of-life green chemistry principles. The pH-sensitive solubility provides a unique switch: at low pH, chitosan dissolves and acts as a homogeneous catalyst (maximizing activity); at neutral/high pH, it precipitates and behaves as a heterogeneous catalyst (facilitating easy recovery by filtration) [89][90][91]. This solubility switching is rare among solid catalysts. Finally, mucoadhesion, while primarily a biomedical property, correlates with strong substrate adhesion in aqueous-phase organic reactions, preventing desorption of intermediates and improving overall turnover frequency (TOF). Thus, these properties are not merely physical descriptors but are the mechanistic drivers of chitosan's catalytic versatility [92][93][94][95].

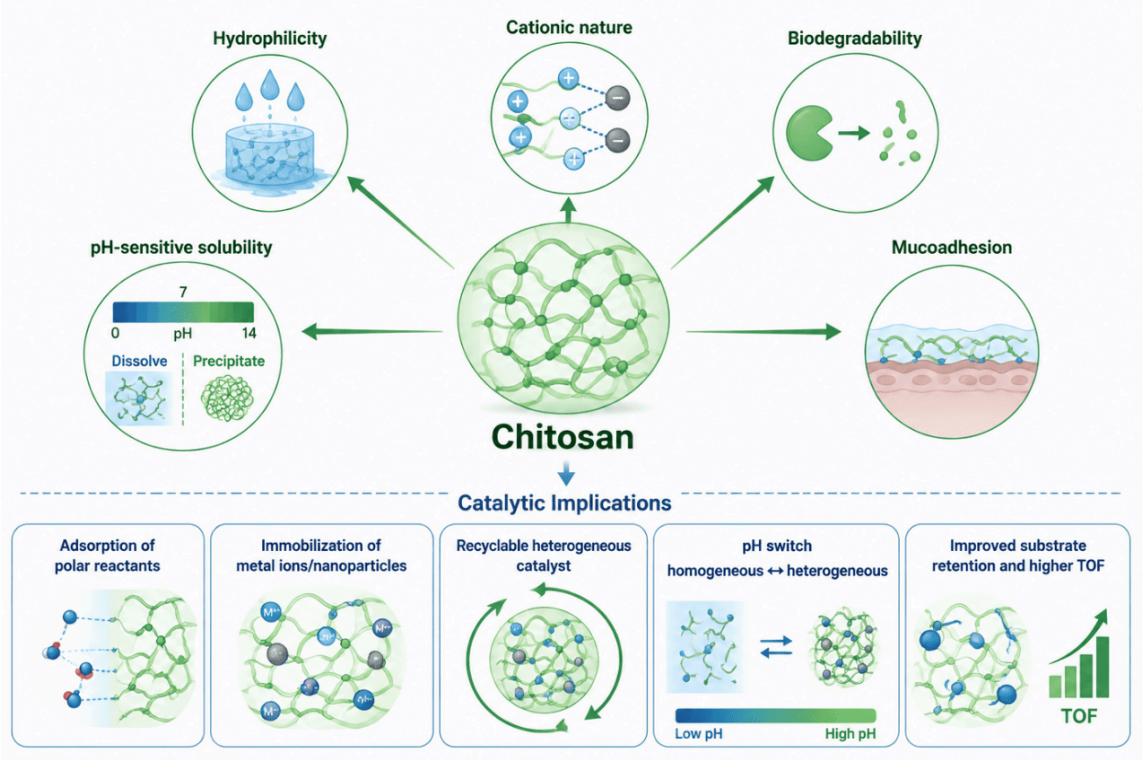

Figure 4. Schematic illustration of the key physicochemical properties of chitosan and their relevance to catalytic performance.

5. Chitosan Modifications

The intrinsic properties of chitosan can be further tailored by modifying its structure. Common modifications include:

5.1. Bonding with Functional Groups

Chitosan can be bonded with various functional groups such as sulfonic acid (-SO_3H), amine (quaternary ammonium), carboxyl (-COOH), aldehyde (-CHO), or thiol (-SH) groups. These modifications enhance unique properties like water solubility at neutral pH, catalytic activity, targetability, or mucoadhesion (thiolated chitosan) [96].

5.2. Coating with Metallic Nanoparticles

Chitosan can be used to encapsulate or stabilize metallic nanoparticles (e.g., silver, gold, iron oxide, zinc oxide). This creates hybrid catalysts or materials with enhanced properties, including antimicrobial

synergy (chitosan-silver), magnetic responsiveness (iron oxide), or optical properties (gold nanoparticles) [97][98].

5.3. Copolymerization with Other Polymers

Chitosan can be copolymerized or blended with other polymers such as cellulose, gelatin, alginate, or poly(vinyl alcohol) (PVA) to adjust its properties. These combinations improve mechanical strength, biodegradation rate, thermal stability, and performance in various applications such as drug delivery, tissue engineering, and wound dressings [99][100].

5.4. Crosslinking

Chemical or physical crosslinking of chitosan (using agents like genipin, glutaraldehyde, or tripolyphosphate) produces hydrogels with controlled swelling, mechanical strength, and sustained release profiles [101].

5.5. Resulting Catalytic Applications

These structural modifications directly translate into enhanced catalytic performance across a wide range of organic transformations. Specifically:

Functionalized chitosan (e.g., sulfonated, quaternized, or carboxylated): Serves as an efficient metal-free catalyst for acid- or base-promoted reactions including esterification, transesterification (biodiesel production), Knoevenagel condensation, and Michael addition. The immobilized functional groups act as surrogate homogeneous catalysts while maintaining heterogeneous recoverability [102][103][104].

Chitosan-metal nanoparticle composites (e.g., Chitosan-Pd^0, Chitosan-Au^0, Chitosan-Cu^0): Act as highly active hybrid catalysts for cross-coupling reactions (Suzuki-Miyaura, Heck, Sonogashira), reduction of nitroarenes to anilines, and oxidation of alcohols. The chitosan matrix prevents nanoparticle aggregation, controls particle size, and often exhibits synergistic catalysis between the metal and the biopolymer's functional groups [105][106].

Crosslinked chitosan beads or membranes: Enable operation in continuous-flow reactors and stirred batch reactors under acidic conditions without dissolution, allowing facile catalyst recovery by simple filtration and reuse for 5-10 cycles with minimal activity loss [107].

Magnetic chitosan composites (e.g., Chitosan-Fe_3O_4): Combine catalytic activity with superparamagnetic properties, allowing rapid catalyst separation from the reaction mixture using an external magne eliminating the need for centrifugation or filtration. Thus, through strategic modification, chitosan can be tailored to meet the specific demands of nearly any organic transformation, ranging from C–C bond formation to heterocycle synthesis and biomass valorization [108][109][110].

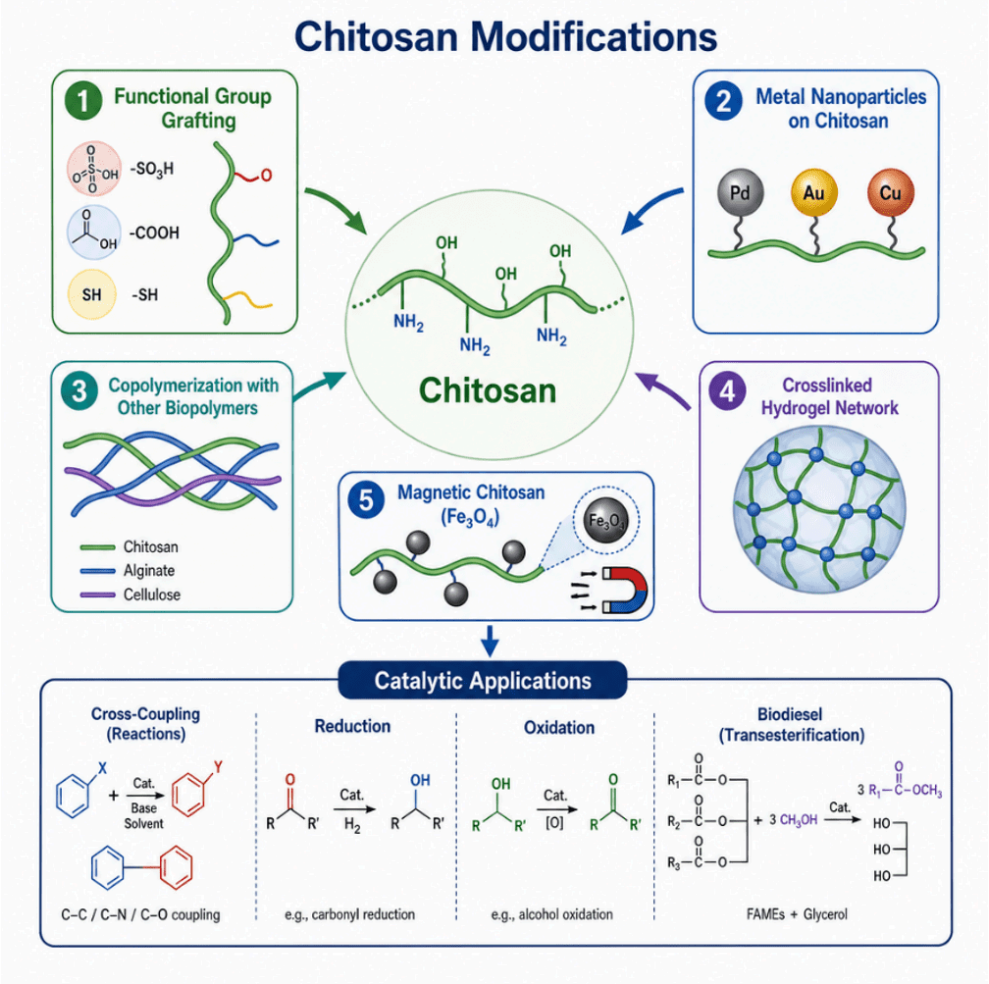

Figure 5. Schematic representation of the structural modification strategies used to tailor chitosan properties and expand its catalytic utility.

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6. Properties of Chitosan as a Catalyst

The cationic nature of chitosan and the abundance of functional groups, including primary amino (-NH_2) and hydroxyl (-OH) groups, enable it to exhibit diverse catalytic properties. These properties can generally be classified into three main categories: acid-catalyzed reactions, base-catalyzed reactions, and redox reactions [111].

6.1. Acid-Catalyzed Reactions

In acid-catalyzed reactions, chitosan acts as a proton donor (Brønsted acid), facilitating nucleophilic addition or elimination processes [112].

Mechanism of Action

The partially deacetylated amino groups (-NH_2) in chitosan can become protonated (-NH_3^+) in acidic or even neutral aqueous media. These protonated sites donate protons (H^+) to reactant molecules, thereby lowering the local pH at the catalytic interface. This protonation activates electrophilic centers (e.g., carbonyl carbons) and stabilizes transition states, promoting nucleophilic attack [113][114].

Key Reaction Types

Esterification: Chitosan catalyzes the formation of esters from carboxylic acids and alcohols.

Acylation: Introduction of acyl groups into organic substrates.

Alkylation: Transfer of alkyl groups to nucleophilic sites [115].

Structural Factors Influencing Activity

Higher degree of deacetylation (DDA) → More free –NH_2 groups → Higher proton donor capacity.

Swollen hydrogel state → Enhanced accessibility of active sites [116].

Applications

Synthesis of bio-based polymers.

Green chemistry processes avoiding mineral acids [117][118].

6.2. Base-Catalyzed Reactions

Chitosan can also function as a base (nucleophilic catalyst) and participate in nucleophilic addition, elimination, and condensation reactions [119].

Mechanism of Action

The free amino groups (-NH_2) in chitosan act as nucleophiles, possessing a lone pair of electrons on the nitrogen atom.

These nucleophilic sites attack electrophilic centers (e.g., carbonyl carbons, activated double bonds) in reactant molecules.

This initiates a cascade of bond formation and bond cleavage steps, ultimately yielding the product and regenerating the chitosan catalyst [120][121][122].

Key Reaction Types

Aldol Condensation: Formation of β-hydroxy carbonyl compounds or α,β-unsaturated carbonyls.

Michael Addition: Nucleophilic addition to α,β-unsaturated carbonyl compounds.

Transesterification: Exchange of alkoxy groups between an ester and an alcohol.

Structural Factors Influencing Activity

Higher DDA → Higher nucleophile density.

Swollen or dissolved state → Improved molecular mobility and substrate accessibility.

Applications

Biodiesel production (transesterification of triglycerides).

C–C bond formation in organic synthesis [119][120][121][122].

6.3. Redox Reactions

Chitosan can serve as a catalyst in redox (oxidation-reduction) reactions and facilitate electron transfer processes [123].

Mechanism of Action

The hydroxyl groups (-OH) in chitosan can be oxidized to carbonyl groups (aldehydes or ketones), acting as electron donors.

The amino groups ($-NH_2$) can be reduced to imine (-C=NH) or other nitrogenous intermediates, acting as electron acceptors.

Chitosan can also stabilize transition metal ions or nanoparticles in different oxidation states, mediating electron shuttling [124][125][126].

Key Reaction Types

Oxidation Reactions: Conversion of alcohols to aldehydes or ketones; oxidative coupling of phenols.

Reduction Reactions: Reduction of nitroarenes to anilines; reduction of carbonyls to alcohols.

Coupling Reactions: Homo-coupling or cross-coupling of aryl or alkyl groups (e.g., Suzuki, Heck-type couplings when combined with metal nanoparticles) [127][128].

Structural Factors Influencing Activity

Presence of free -OH and $–NH_2$ groups provides multiple redox-active sites.

Ability to form complexes with metal ions (e.g., Cu^{2+}, Fe^{3+}, Pd^{2+}) enhances electron transfer efficiency [129][130].

Applications

Green synthesis of fine chemicals.

Degradation of organic pollutants (e.g., dyes, phenols) via Fenton-like mechanisms [123][124][125][126][127][128][129][130].

6.4. Enhancement of Catalytic Activity through Structural Modifications

The catalytic activity of chitosan can be significantly enhanced by modifying its structure. These modifications allow tuning of acidity, basicity, stability, and recyclability [131].

Functional Group Grafting

Description: Covalent bonding of various functional groups to the chitosan backbone (via $-NH_2$ or -OH).

Examples:

Sulfonic acid ($-SO_3H$) groups → Enhanced Brønsted acidity.

Quaternary ammonium ($-N^+R_3$) groups → Enhanced basicity and phase-transfer properties.

Carboxyl (-COOH) groups → pH-responsive catalytic behavior.

Result: Adjusted acidity or basicity for specific reaction types [132][133][134].

Encapsulation or Immobilization of Metal Nanoparticles

Description: Chitosan acts as a stabilizing matrix for metal nanoparticles (MNPs).

Examples:

Chitosan-Pd^0 nanoparticles → Catalysts for cross-coupling reactions (e.g., Suzuki, Heck).

Chitosan-Au nanoparticles → Catalysts for reduction of nitroaromatics and oxidation of glucose.

Chitosan-Fe_3O_4 (magnetic) nanoparticles → Easily recoverable magnetically separable catalysts.

Result: Hybrid catalysts with increased activity (high surface area of MNPs), stability (protection against aggregation), and recyclability (easy recovery) [135][136][137].

Crosslinking

Description: Chemical or ionic crosslinking of chitosan chains.

Result: Formation of porous, insoluble catalytic beads or membranes that can be used in continuous flow reactors without dissolution [138][139].

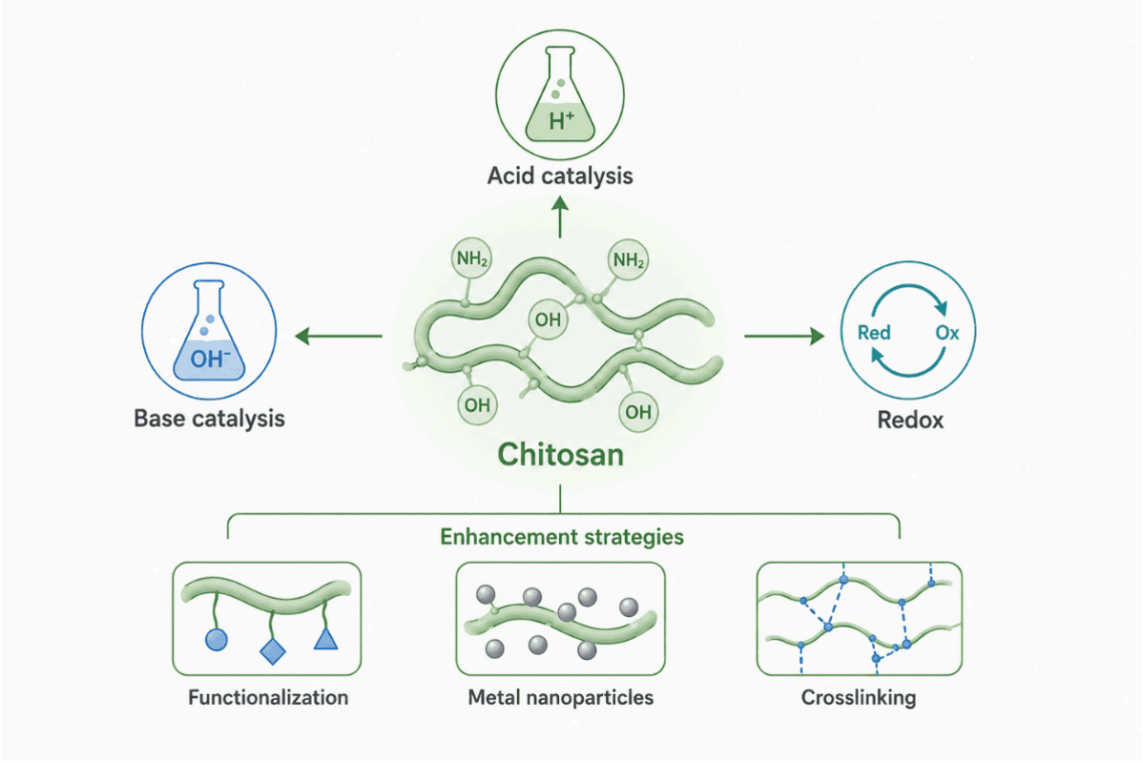

Figure 6. Schematic illustrating the catalytic roles of chitosan in acid-, base-, and redox-mediated transformations, together with major structural modification strategies that enhance catalytic activity, stability, and recyclability.

6.5. Summary of Catalytic Mechanisms

Catalyst Type	Active Group	Mechanism	Example Reactions
Acid	$-NH_3^+$ (protonated)	Proton donor, activates electrophiles	Esterification, acylation, alkylation
Base	$-NH_2$ (free)	Nucleophile, attacks electrophiles	Aldol condensation, Michael addition, transesterification
Redox	-OH, $-NH_2$, or metal complexes	Electron donor/acceptor	Oxidation, reduction, coupling reactions

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7. Advantages of Chitosan as a Green Catalyst

The broad range of catalytic properties, along with several inherent advantages, makes chitosan a promising catalyst for various applications in organic synthesis, polymerization, and green chemistry [140][141][142][143]:

Biocompatibility: Non-toxic and safe for use in pharmaceutical and food-related syntheses.

Biodegradability: Environmentally friendly disposal after use.

Tunability: Easily modified via functional group grafting, metal loading, or crosslinking.

Mild Reaction Conditions: Ability to function often at ambient temperature and pressure, reducing energy consumption.

Recyclability: Can be recovered and reused multiple times without significant loss of activity (especially when crosslinked or magnetized).

Heterogeneous or Homogeneous Operation: Can act as a heterogeneous catalyst (insoluble form) for easy separation, or as a homogeneous catalyst (soluble form) for high activity.

These features enhance its appeal for environmentally safe (green) processes [140][141][142][143].

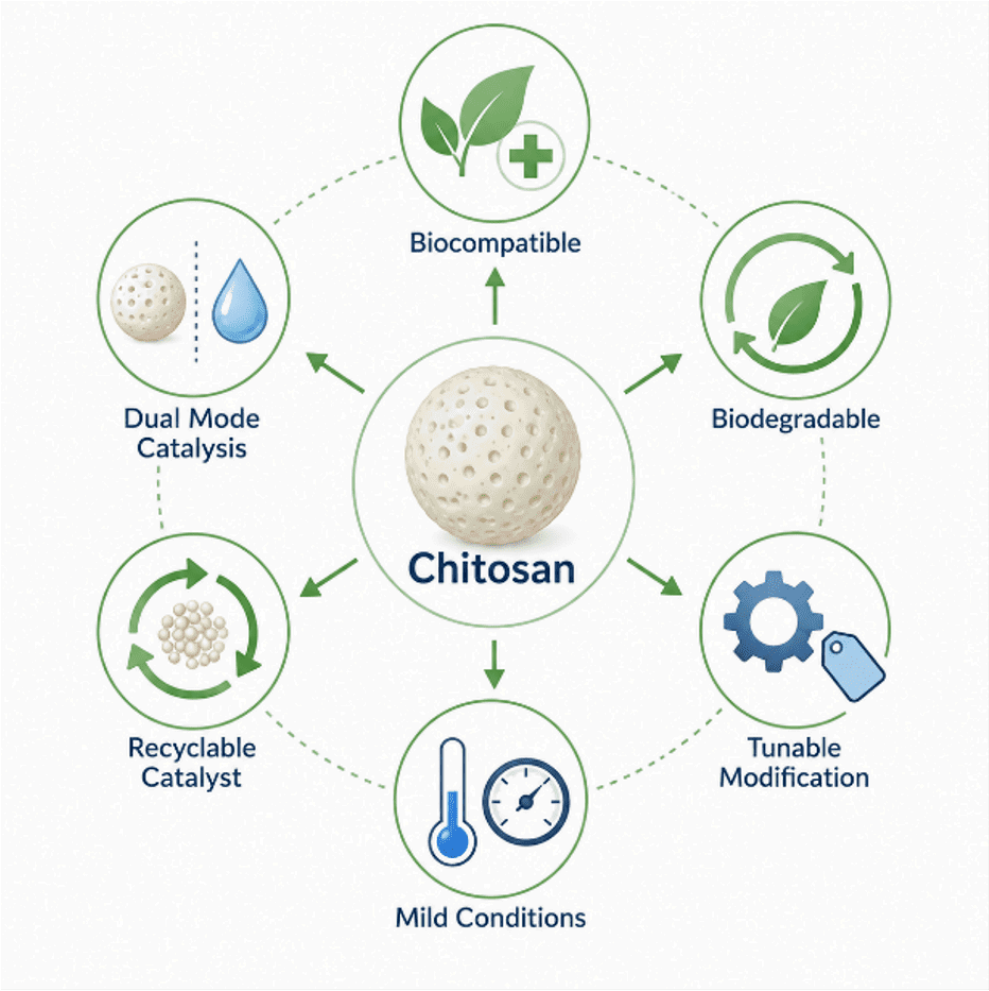

Figure 7. Key advantages of chitosan as a green catalyst, including biodegradability, biocompatibility, tunability, recyclability, and operation under mild reaction conditions.

8. Advanced Applications of Chitosan as a Versatile Catalyst: From Organic Synthesis to Green Chemistry

8.1. Scope of Catalytic Applications in Organic Synthesis

To date, chitosan has been successfully employed as a heterogeneous or semi-heterogeneous catalyst in the synthesis of a broad spectrum of organic compounds, including but not limited to aldehydes, ketones, esters, amides, and heterocyclic scaffolds. Its dual acid–base character, combined with remarkable structural tunability, enables chitosan to catalyze mechanistically distinct transformations under mild, often solvent-minimized conditions [144][145].

8.2. Esterification Reactions

Chitosan efficiently catalyzes the esterification of carboxylic acids with primary and secondary alcohols. The reaction proceeds via proton transfer from the glucosammonium moiety ($-NH_3^+$) to the carbonyl oxygen of the carboxylic acid, enhancing the electrophilicity of the carbonyl carbon and facilitating nucleophilic attack by the alcohol. This transformation is of high industrial relevance for the production of [146][147][148]:

Pharmaceutical intermediates (e.g., aspirin derivatives, prodrug esters)

Flavor and fragrance compounds (e.g., ethyl butyrate, isoamyl acetate)

Bio-lubricants (e.g., fatty acid methyl/ethyl esters)

Compared to conventional H_2SO_4 or p-TsOH catalysts, chitosan minimizes side reactions such as dehydration, polymerization, or sulfonation, and can be recovered by simple filtration and reused over multiple cycles without significant loss of activity.

8.3. Aldol Condensation

Chitosan acts as a mild base catalyst in aldol and cross-aldol condensation reactions, promoting enolate formation from ketones or aldehydes. The free amino groups ($-NH_2$) abstract an α-proton from the carbonyl compound, generating a nucleophilic enolate that attacks another carbonyl electrophile. Subsequent dehydration yields α,β-unsaturated carbonyl products. This transformation is pivotal in the synthesis of [149][150][151]:

Natural products (e.g., chalcones, flavonoid precursors)

Pharmaceuticals (e.g., anti-inflammatory and anticancer agents)

Fine chemicals (e.g., intermediates for polyene antibiotics)

Chitosan-based aldol catalysis operates efficiently at room temperature in water or ethanol, offering a green alternative to traditional base catalysts like NaOH or KOH, which often cause over-condensation and side-product formation [152][153].

8.4. Michael Addition

Chitosan also catalyzes the Michael addition reaction, a fundamental carbon–carbon bond-forming reaction between electron-deficient alkenes (Michael acceptors, e.g., α,β-unsaturated ketones, nitriles, or

esters) and nucleophiles (Michael donors, e.g., malonates, thiols, or amines). The -NH_2 groups activate the nucleophile via hydrogen bonding or deprotonation, while the polysaccharide backbone stabilizes the transition state through polar interactions [154][155][156]. Applications include:

Polymer synthesis (e.g., functionalized hydrogels, dendritic polymers)

Pharmaceutical synthesis (e.g., chiral building blocks for beta-blockers)

Dye and pigment production (e.g., Michael adducts as chromophore precursors)

Chitosan's heterogeneous nature allows easy separation from the reaction mixture, preventing metal contamination often associated with organometallic Michael catalysts [157][158][159].

8.5. Polymerization Reactions

Chitosan has been extensively investigated as a green catalyst for the polymerization of various monomers, including vinyl acetate, lactic acid, and styrene. Its catalytic mechanism varies with monomer type [160][161]:

For ring-opening polymerization (ROP) of lactide or lactone, chitosan's -OH and –NH_2 groups act as initiators via nucleophilic attack on the cyclic ester.

For free-radical polymerization, chitosan can stabilize radical intermediates or act as a dispersion stabilizer, producing controlled molecular weight distributions.

For polycondensation (e.g., of lactic acid to polylactic acid), chitosan creates a mildly acidic microenvironment that accelerates ester bond formation without requiring toxic tin octoate catalysts. This capability positions chitosan as a promising platform for producing biodegradable plastics and biomedical polymers [162][163][164][165].

8.6. Biodiesel Production via Transesterification

Chitosan-based catalysts have garnered significant attention in the production of biodiesel from vegetable oils and waste cooking oils. Chitosan, especially in its crosslinked or functionalized forms (e.g., sulfonated or quaternized chitosan), catalyzes the transesterification of triglycerides with short-chain alcohols (methanol or ethanol). The reaction proceeds through a two-step mechanism [166][167][168]:

1. Activation of the alcohol via hydrogen bonding or proton abstraction by –NH_2 groups,

2. Nucleophilic attack on the ester carbonyl of the triglyceride, forming fatty acid methyl esters (FAME) and glycerol.

Advantages over homogeneous base catalysts (KOH, NaOH):

No soap formation from free fatty acids

No neutralization or water washing steps

Catalyst recyclability (up to 5–8 cycles with >85% activity retention)

Compatibility with low-grade feedstocks

Thus, chitosan enables a more sustainable and cost-effective biodiesel process [169][170].

8.7. Chitosan in Green Chemistry: A Paradigm Shift

The intrinsic properties of chitosan biodegradability, biocompatibility, nontoxicity, and tunable functionality align perfectly with the Twelve Principles of Green Chemistry. Chitosan offers a renewable, metal-free, and environmentally benign alternative to conventional catalysts that are often corrosive, toxic, or derived from nonrenewable resources [171][172].

8.8. Sustainable Synthesis

Chitosan-based catalysts enable the synthesis of chemicals and advanced materials under mild conditions (ambient temperature, atmospheric pressure, aqueous or solvent-free media). This drastically reduces energy consumption, waste generation, and reliance on fossil-fuel-derived solvents. For example, chitosan-mediated Knoevenagel condensations and Henry reactions proceed efficiently in water with excellent atom economy [173].

8.9. Production of Biodegradable Polymers

Chitosan serves both as a catalyst and as a structural template for producing biodegradable polyesters, polyurethanes, and polycarbonates. By catalyzing the polymerization of renewable monomers (e.g., lactic acid, ε-caprolactone, trimethylene carbonate), chitosan helps reduce dependence on petroleum-based plastics like polyethylene and polypropylene. The resulting chitosan-catalyzed polymers exhibit controlled degradation rates, making them suitable for agricultural films, packaging, and biomedical implants [174][175].

8.10. Environmental Remediation

Chitosan catalysts have been successfully applied to the degradation of persistent organic pollutants (POPs), synthetic dyes, pesticides, and pharmaceutical residues in wastewater. Mechanisms include [176][177][178]:

Fenton-like reactions using chitosan–iron complexes to generate hydroxyl radicals ($OH^{\bullet}$) for oxidative degradation.

Photocatalysis with chitosan–TiO_2 or chitosan–ZnO composites under UV/visible light.

Reductive degradation of nitroaromatics and azo dyes using chitosan-stabilized metal nanoparticles (Pd, Cu, Ni).

In addition, chitosan-based catalytic systems can remediate contaminated soil and groundwater by immobilizing heavy metals and breaking down chlorinated hydrocarbons. These capabilities significantly enhance environmental protection while avoiding secondary pollution from leached toxic metals [179][180].

9. Comparative Advantages of Chitosan over Conventional Catalysts

Feature	Chitosan Catalyst	Conventional Catalyst (e.g., H_2SO_4, KOH, Pd/C)
Source	Renewable (crustacean shells, fungi)	Non-renewable (petrochemicals, mined metals)
Toxicity	Non-toxic, biocompatible	Corrosive, toxic, or carcinogenic
Reaction conditions	Mild (30–80 °C, atmospheric pressure)	Harsh (high T, high P, strong acids/bases)
Reusability	Yes (3–10 cycles)	Often single-use or requires regeneration
Product separation	Simple filtration	Neutralization, extraction, chromatography
Waste generation	Minimal	High (salts, organic waste, metal residues)
Biodegradability	Complete	None (persistent in environment)

10. Challenges and Future Directions

Despite the remarkable catalytic potential of chitosan, several challenges remain. The poor mechanical stability and solubility of chitosan in acidic media limit its long-term reusability and application in continuous-flow systems. Additionally, batch-to-batch variability in the degree of deacetylation and molecular weight affects catalytic reproducibility. Future research should focus on developing robust crosslinked or magnetic chitosan composites to enhance recyclability and mechanical strength. Green and scalable functionalization strategies, such as enzyme-mediated grafting or deep eutectic solvent-based modifications, are needed to minimize environmental footprint. Furthermore, integrating chitosan catalysts into continuous-flow reactors and exploring their application in photo/electrocatalysis could unlock new avenues for sustainable chemical synthesis.

11. Conclusion

Chitosan, a naturally abundant polysaccharide derived from chitin, has emerged as a powerful and versatile green catalyst in modern organic chemistry. Its unique combination of biodegradability, biocompatibility, and structural flexibility, coupled with the ability to function as both a Brønsted acid and a base, allows it to catalyze a wide array of reactions including esterifications, aldol condensations, Michael additions, transesterifications, and polymerizations under mild, environmentally responsible conditions. Furthermore, the ease of surface modification through covalent grafting of functional groups (sulfonic, carboxylic, quaternary ammonium) or encapsulation of metal nanoparticles (Au, Pd, Cu, Fe) significantly enhances its catalytic efficiency, selectivity, and reusability. These engineered chitosan derivatives often match or even surpass the performance of conventional synthetic catalysts, while eliminating issues of toxicity, corrosiveness, and non-recyclability. In summary, chitosan stands as a compelling candidate to replace hazardous synthetic catalysts across many industrial sectors. Its continued development promises to drive the global transition toward sustainable chemistry, circular bioeconomy, and green manufacturing. As research into chitosan catalysis advances particularly in continuous-flow systems, magnetically recoverable composites, and enzyme-mimetic designs its role is expected to expand, potentially transforming how we produce pharmaceuticals, polymers, fuels, and clean water in an environmentally harmonious manner.

Statements and Declarations

Potential Coompeting Interests

There are no conflicts to declare.

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