



MODIFICATION OF SCHIFF BASE OF CHITOSAN DERIVATIVES AND ITS APPLICATIONS *VIA* GREEN METHODS

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Abstract

Biopolymers have turned out to be very attractive as they're degradable, biocompatible, non-toxic and renewable. Due to the intrinsic reactive amino businesses, chitosan is colorful within the midst of various biopolymers. Using the flexibility of these amino businesses, several structural changes have been performed on chitosan through sure chemical reactions. Chemical modification of chitosan through imine functionalization is huge as it recommends the ensuing chitosan-based totally definitely Schiff bases (ChSBs) for the vital applications within the fields like catalysis, water treatment, biology, sensors, etc. ChSBs are usually synthesized with the resource of the usage of the Schiff condensation reaction amongst chitosan amino organizations and carbonyl compounds with the removal of water molecules. In this review, we first introduce them to be had synthetic techniques for the coaching of ChSBs. Then, we speak the natural applications of ChSBs inclusive of antimicrobial activity, anticancer activity, drug carrier ability, antioxidant activity and tissue engineering capacity. Successively, the applications of ChSBs in specific fields together with catalysis, adsorption and sensors are demonstrated. Here, covers the green synthesis of ChSBs using non-traditional inexperienced techniques including microwave irradiation, inexperienced solvent, ultrasound irradiation, and one pot synthesis. These techniques are energy-green and



greener variations of the traditional condensation techniques. On the other hand, modification of chitosan Schiff base polymer derivatives by using different nanoparticles and beta-cyclodextrine for modifying the physical and chemical properties of ChsBs. Then, the generated polymers were used in various fields such as ammonia sensing, antimicrobial activity, bone tissue engineering, and others.

Keywords: Chitosan, Beta-cyclodextrine, nanoparticles, Schiff base, composite, pseudopolyrotaxane.

1. Introduction

Green chemistry is a complete application aimed at converting the opinion of chemists and turned into additionally administered to protect the surroundings *via* means of specializing in decreased dangers or complete waste elimination techniques. The time period “inexperienced chemistry” turned into first determined *via* way of means of Paul Anastas in 1991 [1]. An essential device in pollutants prevention is referred to as inexperienced chemistry. The term green chemistry refers to techniques and strategies that reduce or dispose of the use of substances, consisting of feed stocks, products, through manner of-products, solvents, reagents that damage the environment or human health [2]. Green chemistry acts as a tool for promoting sustainable development in laboratories and industries. It helps to format chemical products and approaches extra environmentally pleasant. It moreover includes herbal synthesis, chemical engineering, or analytical chemistry [3]. The vital cause of green synthesis is to lower the use of toxic substances and the era of noxious waste whilst preserving efficacy. The vital reputation of green synthesis is the usage of substances from natural reasserts in their method of synthesis [4], so chitosan became taken as amine precursor with inside the green synthesis due to its bio-polymer nature with multiple applications and moreover reliable biodegradability [5]. Chitosan is one of the polysaccharides which can be regularly decided in dwelling organisms and second most ample natural polymer, **Scheme 1** [6]. Chitosan is derived from chitin decided in marine organisms like shrimp/crab shells, the molecular membrane of algae, skeletons of mollusks, and plant molecular walls [7]. Chitosan is also called a linear beta-1, 4-linked polysaccharide received through manner of Chitin deacetylation. The structural characteristic of chitosan is the presence of primary amine with inside the C-2 function of glucosamine residue which allows them to react without difficulty with aldehyde/ketone macromolecules [8]. Chitosan is extracted from chitin in a multistep process. The first stage is the producing of chitin from shrimp canning associated with food industries and then crash and grinding them to smaller sizes and minerals specifically calcium carbonate is removed through manner of demineralization, and decalcification with diluted hydrochloric acid through manner of doing constant stirring. In the 2nd level, the production of chitosan is produced *via* alkali treatment (deproteinization) observed through manner of manner of deacetylation to form chitosan [9]. Chitosan has important homes like inexperienced nature, biodegradability, reactivity, and ease of functionalization which permit them to use in biomedical, industrial procedures, and pharmaceutical industries [10]. Chitosan has many programs, like agricultural, medical, nutritional

enhancement, food processing, cosmetics, and wastewater treatment [11]. Different forms of derivatives of chitosan are diagnosed in which the imine derivatives are synthesized through manner of a manner of a response amongst amino corporations of chitosan and aldehydes [12]. Imine, additionally called Schiff bases are an essential elegance of organic compounds. Schiff bases had been first of all located with the aid of using Hugo Schiff in 1864. They are prepared with the resource of the use of condensation of primary amines with carbonyl compounds. The trendy approach of these compounds is $RHC=N-R1$ in which R and R1 are alkyl, aryl, cycloalkyl, or heterocyclic groups [13]. The Schiff base has big programs in masses of natural aspects, which include proteins, seen pigments, enzymic aldolization, and decarboxylation reactions. Schiff bases and their complexes are antibiotic, antiviral, anti-tumor, anti-fertility, and enzymatic sellers acting as catalysts in polymer and dyes industries [14]. The era of Schiff base takes place under moderate conditions to form water as a by-product that is inexperienced in nature [15]. There are several green techniques for the synthesis of Schiff base derivatives which encompass microwave irradiation, ultrasonic irradiation, and green solvent and one-pot synthesis [16]. Besides the one's techniques, there are one-of-a-kind green techniques which might be used with Schiff base synthesis like natural acid-catalyzed technique and grinding technique [17]. In the natural acid-catalyzed technique natural acids (which encompass lemon juice) have presently been identified as capability herbal solvents for the producing of pharmaceuticals [18]. G. Yadav *et al.* [19], synthesized Schiff base derivatives with the resource of the use of using lemon as a catalyst. Furthermore, the grinding technique is the traditional technique used for the crushing and grinding of small particles in our laboratories. This technique works at the principle that warm temperature generated with the resource of the use of friction throughout the grinding process hurries up the reaction process to form the product. This technique is top due to its efficiency, environmentally friendly, and no harm to human health. M. Zarei *et al.* [20], used this technique for the synthesis of the Azo Schiff base and said that this technique emerges as speedy and low value with a wonderful yield.

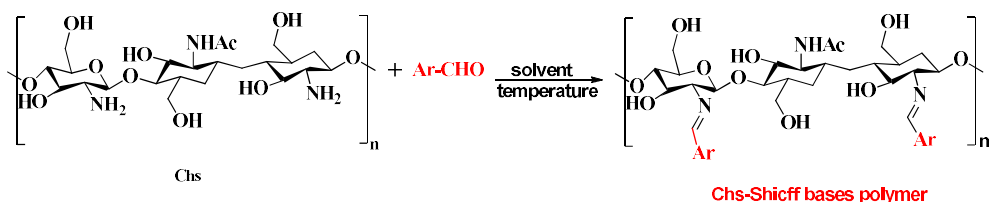
When chitosan Schiff bases are modified by betacyclodextrin, a type of cyclodextrin composed of seven glucose units; the resulting compound often exhibits enhanced properties. Betacyclodextrin is known for its ability to form inclusion complexes with various molecules, improving their solubility and stability [21-25]. The modification of chitosan Schiff bases by betacyclodextrin can lead to the formation of inclusion complexes that have potential applications in various fields such as drug delivery, food packaging, and wastewater treatment. The improved properties of these modified compounds make them promising candidates for further research and development.

Modifying chitosan Schiff bases with nanoparticles is a promising area of research with potential applications in various fields such as drug delivery, tissue engineering, and antimicrobial coatings [25-28]. Nanoparticles can enhance the properties of chitosan Schiff bases, such as increasing their stability, biocompatibility, and bioactivity. This modification involves incorporating nanoparticles, such as silver or gold nanoparticles, into the chitosan Schiff base structure. The nanoparticles can be attached to the chitosan Schiff base through various methods, such as covalent bonding or physical adsorption. The resulting nano composite material can have improved

properties compared to the unmodified chitosan Schiff base, making it useful for a wide range of applications.

Preparation of chitosan Schiff bases.

Chitosan Schiff bases (Chs-SBs) are generally produced by easily condensing the amino groups of chitosan with the carbonyl groups of aldehydes or ketones by removing water molecules. Hirano *et al.*, produced the first Chs-SBs in 1977 by employing a solvent combination of acetic acid and methanol to react chitosan with various aldehydes [29]. Following this, numerous chitosan Schiff bases are published by other research teams. As shown in **Scheme 1**, the solvent medium for the synthesis of Chs-SBs is typically acetic acid, methanol, water or dimethylformamide, or a combination of these, and is utilized at either ambient or refluxing temperature conditions. Aly and his groups have reported that the reaction of chitosan with salicaldehyde, 3-formylchromone or pyridoxal as a carbonyl component to form the corresponding Chs-SBs [30-32]. Tamer *et al.*, have reported that the reaction of chitosan with 2,4,6-trimethoxybenzaldehyde to afford chitosan-TMB as a soul polymer [33].



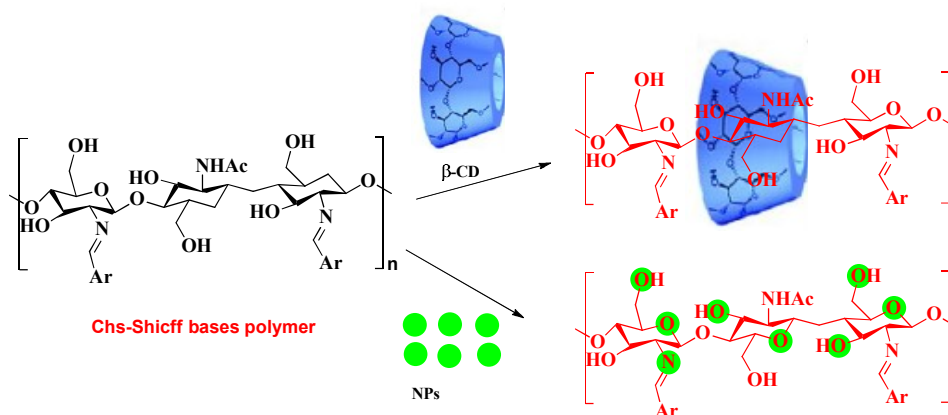
Scheme 1

From the above referred to not unusualplace solvents, few different solvents like dimethylformamide, water, methanol and ionic liquid [34-36], also are used for the synthesis of Chs-SBs. Ultrasonication is used to synthesize the Chs-SBs from the reaction of chitosan with citral as a carbonyl component [37]. Xu *et al.*, have carried out chitosan because the carbonyl compound in the course of the synthesise of Chs-SBs. Briefly, they have got covered the amino and hydroxyl businesses of chitosan using the coordination reaction with copper and subsequently, oxidized $-CH_2OH$ agencies into formyl agencies. Further, they allowed the ones formyl businesses to react with super amines for the guidance of Chs-SBs, **Scheme 2** [38]. Through Chs-SBs synthesis, pass-linking of chitosan can be straightforwardly achieved with the careful preference of dialdehydes/diketones alongside glutaraldehyde, quinone, and glutaraldehyde-thiourea mixture, crown ether containing dialdehydes, glyoxal and dialdehyde cellulose [39]. This pass-linking technique seems to be very promising withinside the synthesis of Chs-SBs gels. Chs-SBs have moreover been synthesized in specific morphologies alongside microspheres, fibers and hollow spheres, and to attain such morphologies, a completely unique artificial technique may be needed [40]. To synthesize Chs-SBs films and membranes, chitosan and aldehyde/ketone commonly react with each unique in a blended solvent medium and pour into the plate or unique substrate which in flip consequences withinside the formation of films after the drying process [41]. Spin coating is on occasion used to synthesize Chs-SBs films [42]. It has used the electro-spraying technique to prepare the pass connected Chs-SBs through manner of mixing chitosan and dialdehyde together [43]. Chitosan can be chemically modified into glycol chitosan (GC) [44], carboxymethyl chitosan

(CMC) [45], carboxyethyl chitosan (CEC) [46], sulfonated hydroxypropyl chitosan (SHPC) [47] and succinyl chitosan (SC) [48], for the synthesis of water soluble Chs-SBs. In a few cases, periodate oxidation is used to make chitosan due to the fact the carbonyl compound. This oxidized chitosan is further allowed to react with bare chitosan for the synthesis of Chs-SBs [49].

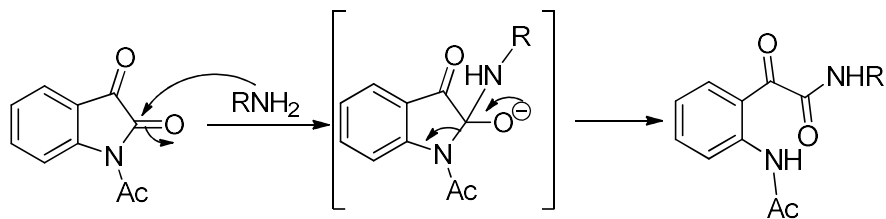
Modification of chitosan Schiff bases.

The purpose of modifying chitosan Schiff bases improving the physical and chemical properties of the generated polymers. Using betacyclodextrine and nanoparticles as a tool to modify the corresponding Schiff bases, **Scheme 2**.



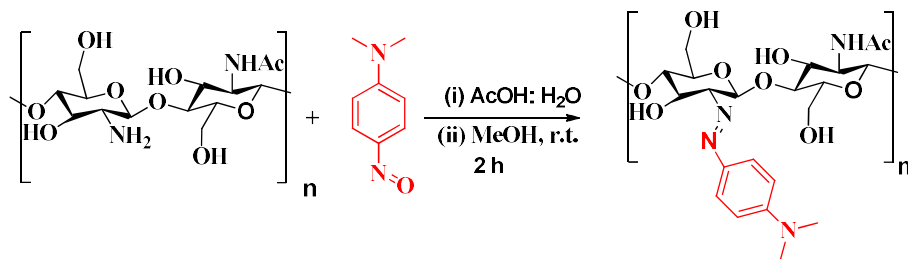
Scheme 2

Aly *et al.*, using β -CD and ZnO nanoparticles to modify the chitosan Schiff base derivatives [34-36]. Using 3-formylchromone, salicylaldehyde and pyridoxal as a carbonyl component for reacting with chitosan as amine to form the corresponding Schiff bases. Then the generated Schiff bases reacted with betacyclodextrine to form the corresponding pseudopolyrotaxane. Also, the shaped Schiff bases doping with ZnO NPs to have the funds for the corresponding composite. α -ketoamides have attracted more interest and are determined in lots of energetic pharmaceutical compounds. The amidic group of *N*-acetyl isatin is pretty reactive due to the adjacent carbonyl group. The amidic carbonyl group is probably without a hassle related *via* nucleophiles upon beginning the ring to yield α -Keto derivatives in fantastic yields. Amines as an example react easily with the amidic carbonyl group to offer α -ketoamides, which may be of interest in herbal and medicinal chemistry, **Scheme 3**. So, we goal to use chitosan as a natural polymer to form chitosan-acetylisatin (Chs-NAI) as a completely unique chitosan polymer derivative, and we investigated the transformation of the generated polymer (Chs-NAI) *via* inclusion in a β -CD and spiking with ZnO nanoparticles (NPs) to provide Chs-NAI/ β CD and Chs-NAI/ZnO NPs, respectively [13].



Scheme 3

Taha *et al.*, were reacted the reaction of chitosan with 4-nitrosodimethylaniline to form azo-chitosan-dimethylaniline polymer (Chs-DAN) as the novel polymer through condensation reaction, **Scheme 4**. Then, doping Chs-DAN with ZnO NPs and inserting Chs-DAN into β CD to afford Chs-DAN/ZnO NPs and Chs-DAN/ β CD, respectively [33]. Also, A one-pot four-component reaction of NAI, Chs, β -CD and ZnO NPs afforded Chs-NAI/ β -CD/ZnO NP composite [37].



Scheme 4

Applications:

1) Bone tissue engineering:

The feasibility of Vero cells was greater than 95% for Chs-NAI/CD/ZnO nanoparticles/calcium phosphate concentrations of 31.2, 62.5, and 125 μ g/ml. When the concentrations surpassed one hundred twenty-five μ g/ml, it becomes discovered to be cytotoxic towards the Vero mobileular line, thinking about those concentrations to be the most possible poisonous dose [13, 50-54].

2) antimicrobial activity:

The antimicrobial interest has a look at found out that azo-chitosan derivatives polymers are very promising because it confirmed maximum sports against distinctive varieties of pathogenic microbes than pristine polymers [37].

3) Ammonia sensing:

The chitosan-salicylaldehyde (Chs-Sal) polymer was modified by doping it with ZnO nanoparticles (NPs) and inclusion into β -CDs. This led to excessive selectivity toward NH_3 vapors, that's associated with its maximum electron-donating cappotential in comparison to the alternative analytes, in addition to top reaction and recuperation times (650 s and 350 s, respectively), low LOD and LOQ of 0.12 and 0.four ppb, respectively, and excessive sensitivity (from 712 for Chs-Sal to 6192 for Chs-Sal/ β -CD/ZnO NPs) [34, 36].

Conclusion

A wide range of Chitosan Schiff base derivatives and their uses, including biological, sensing, and bone tissue engineering, have been thoroughly explained in this review. Additionally discussed the significance of betacyclodextrin in this study as well as the degree to which it affects chitosan. It was also made clear how important nanoscale components like zinc oxide are when put to polymers. We will talk in the future about other interactions of chitosan such as: anhydrides, amino acids and esters.

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Declaration of Competing Interest

The authors claim that they've not regarded competing monetary hobby or private relationships that would have seemed to persuade the paintings suggested in.

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