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Roll of Biologically Active Chitosan as a Schiff Base: A Review

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ABSTRACT

Biopolymers have become very popular; because they are biodegradable, biocompatible, non-toxic, and renewable. As a result of its intrinsic reactive amino groups, chitosan stands out among other biopolymers. Chemical reactions have been used to modify chitosan using these amino groups to accomplish a variety of structural modifications. The imine functionalization of chitosan, CSBs (chitosan-based Schiff bases) are recommended for essential practice in water treatment, catalysis, bioscience, sensors, etc. CSBs are prepared by condensation reaction between carbonyl compounds and chitosan's amino groups. This review first introduces the synthesis of different types of Schiff base ligands and then CSBs. Then the biological utilization of CSBs, including their tissue engineering capabilities, antimicrobial, antibacterial, anticancer, drug-carrying, and antioxidant are discussed. Later, CSBs are shown to be applied to catalysis, adsorption, and sensors.

1. Introduction

By combining a carbonyl compound (ketone or an aldehyde) with an amine covered by distinctive conditions for reaction in various solvents, Hugo Schiff discovered Schiff Base in 1864 [1-5]. Schiff Base is a nitrogen derivative of ketones or aldehydes in which imine groups or -C=N- groups replace the carbonyl group (CO) (Fig. 1). When the carbonyl group of an aldehyde is converted to a -C=N- group, an aldimine form is made. In general, Schiff Base (R_1R_2C=N-R_3) is a mixture of alkyl, aryl, and heteroaryl groups, where R_1, R_2, and R_3 are aryl, alkyl, or heteroaryl [6-8]. This azomethine compound's -C=N- imine bond confers a wide range of biological activities [9]. As a result of their advanced biological activities (antibacterial, antifungal, antitumor, anticancer, antiradical, anti-tubercular, antimalarial, ROS scavengers, and antiviral), Medicinal chemists and biologists have been interested in Schiff bases and their transition metal complexes for decades. The results of their outstanding chelating capacity with metals [10-19], these compounds possess enhanced biological activity. Because of their synthetic flexibility and structural stability, the compounds are regarded as versatile ligands and are thus being studied/examined extensively due to their impressive pharmacological actions. Organometallic chemistry also relies on these moieties [12, 20-24].

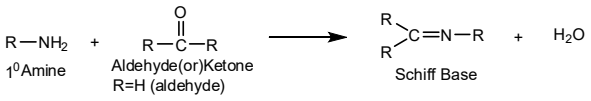

Fig. 1 A typical reaction scheme showing the formation of SBs

In coordination chemistry, Schiff bases are well-established ligands that can stabilize metal ions in multiple oxidation states and are often referred to as special ligands. After metalation, the activity of these Schiff bases is greatly enhanced, making them highly effective catalysts and pharmaceutical agents. There is sufficient literature on azomethines/imines synthesis methods available at present. A more efficient, harmless, and green method of preparing Schiff base is being adopted as an alternative to humanistic and other methods (ultrasound irradiation, microwave irradiation, solvent-free methods, ionic solvents, etc.) using universal solvents (water) in the preparation of these compounds. By forming water molecules during Schiff Base reactions, the reaction rate is slowed down by favoring reversible reactions. Dehydrating agents (molecular sieves, Na_2SO_4, or $MgSO_4$) can be used or Dean Stark apparatuses can be used to facilitate the formation of Schiff bases. Ethanol is the best solvent choice when preparing Schiff bases at room temperature or during refluxing. Schiff Base reactions are usually accelerated by performing them in acidic or basic environments [25-27].

2. Types of Schiff Bases

Literature reports several Schiff bases which are familiar today, including salen, salophen, hydrazone, and semicarbazone/thiosemicarbazone. Due to their particular and incredible roles in coordination chemistry and medicine, various types of these Schiff bases have captured the attention of researchers.

2.1 Salen Type Ligand

For the preparation of sales-type ligands, salicylaldehyde, ethylenediamine, and their derivatives are used as reactants. Salicylaldehyde and diamine are condensation products of the sale ligand, which is a Schiff base. It has been demonstrated since 1933 that transition metal complexes of salen-type ligands possess unique properties in coordination chemistry. Their utility and extensive complexing abilities have attracted significant attention. Furthermore, these are key focuses for bioinorganic chemistry, catalysis, magnetism, and medical imaging. Biological and industrial sales applications can be derived from the difficulty of differing the backbone of the ligand and coordinated metal ion, making these ligand systems extremely useful. According to their bonding modes, Sale-type compounds have two coordinate covalent bonds and two covalent bonds in their equatorial positions. Ancillary ligands occupying two axial sites, [O, N, N, O] form tetradentate ligands as shown in Fig. 2.

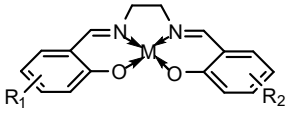

Fig. 2 Salen as [O,N,N,O] tetradentate ligand Salophan type ligand

o-Phenylenediamine is substituted for ethylenediamine in salophen-type ligands. A typical condensation reaction produces these types of ligands by reacting salicylaldehyde or its derivatives with 1,2-phenylenediamines in two equivalents. Various industrial and biomedical

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applications have been found for these ligands due to their metal complexes. One of the most ancient and widespread compounds in coordination chemistry is salophen ligands. Like sale ligands, they behave as tetradentate molecules and have four different coordinating sites: O, N, N, O in a plane Fig. 3.

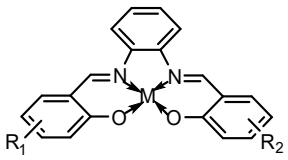

Fig. 3 Salophen as tetradentate [O, N, N, O] Ligand

2.2 Hydrazone-Type Ligand

The hydrazone type ($R1R_2C=NNHR_3$) ligands are another vital class of Schiff base ligands and are formed from carbonyl compounds and hydrazine/hydrazide derivatives. Some drug candidates exhibit enhanced selectivity and fatality profiles due to the presence of hydrazones. The literature on hydrazone ligands demonstrates their captivating properties in abundance. It has been reported that these ligands can form multi-metallic complexes with a variety of metal atoms that exhibit different coordination behaviors. Any carbonyl compound can be converted into an acyl hydrazide or an aroyl hydrazide by condensation, hydrazones can also form acyl hydrazones or aroyl hydrazones Schiff Base ligands. Schiff bases with acyl or arylhydrazone donors are provided with a carbonyl group, which provides more malleability and adaptability. As shown in Fig. 4, hydrazone ligands have monodentate atoms i.e., aminic N but subgroups (aroyl and acyl hydrazones) can perform as tridentate (O, N, S, N, X=O) or bidentate (O, N) ligands.

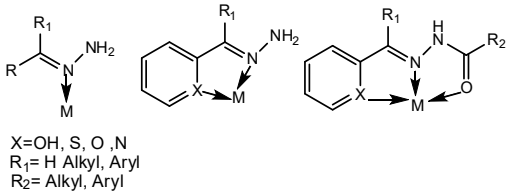

Fig. 4 Hydrazone as a mono-(N), Bi-(N,O/N/S) or tri-(O/N/S, N, O) dentate Ligand

2.3 Semicarbazone /Thiosemicarbazone Ligands

The first semicarbazones/thiosemicarbazones appeared in literature in 1800 and proved excellent chelating agents. A ketone or aldehyde is reacted with semicarbazide or thiosemicarbazide to yield thiosemicarbazone/semicarbazone-Schiff bases. Due to their better chelating ability and broad lipophilicity, they are of great importance. It has been proven through extensive literature studies that various d-block metal complexes of these Schiff Base ligands can be synthesized and have immense applications. As a result of the presence of different donating sites, thiosemicarbazones/semicarbazones occur in a variety of bonding modes. Generally, these ligands act as bidentate ligands with two N, O/S donating sites and tridentate ligands (N, S, X=N, S, O) can also be formed from them [28]. The figure below shows bonding modes in such ligands Fig. 5.

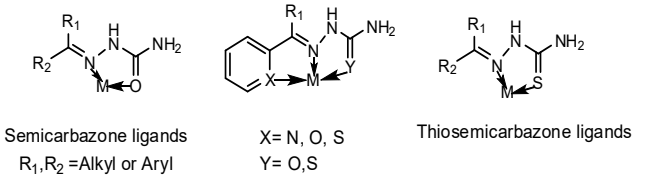

Fig. 5 Thiosemicarbazone / semicarbazone as a bi- (N, O) or tri- (N/S/O, N, O) dentate ligand

Due to their abundance and remarkable properties, polymer ligands have gained popularity among Schiff bases today. In the ever-growing medical, photophysics, aerospace, ferroelectric industries, packaging food, automobile, electronics, and polymers have become essential to everyday life. Due to the non-degradable nature of these polymers, they have adverse effects on the environment. C.J. Moore refers to non-degradable polymers as a threat that will rapidly increase in the future. It is therefore important to replace non-degradable polymers with bio-polymers that are degradable. There have been reports for synthetic polymers of bio-polymers such as chitosan, chitin, poly amino acids, and cellulose being used as substitutes.

3. Chitosan

A natural polysaccharide second in abundance only to cellulose, chitin is typically obtained from crustacean exoskeletons such as crabs, crawfish, shrimps, and lobsters, primarily a waste product after food processing [29]. However, the insolubility of this compound in common solvents and diluted acids, however, limits its application. Although, chitosan, a polysaccharide derived by treating chitin with alkali, is a deacetylated derivative of 2-amino-2-deoxy-b-D-glucose and 2-acetamido-2-deoxy-b-D-glucose [30]. Acetic, hydrochloric, and formic acids readily dissolve chitosan, resulting in viscous solutions [31]. In addition to its biodegradability, biocompatibility, and non-toxicity, this unique material has an extensive range of industrial applications, including biomedical, chemical, wastewater purification, agricultural, and biotechnology [32,33]. Deacetylation (DD) is the main factor controlling chitosan's solubility and biological properties of chitosan, and several methods have been proposed for measuring the DD of chitosan [34]. There is a general understanding that chitin is a linear long-chain homopolymer composed of units of N-acetylglucosamine units, [poly(N-acetyl-β-D-glucosamine)] [35] (Fig. 6).

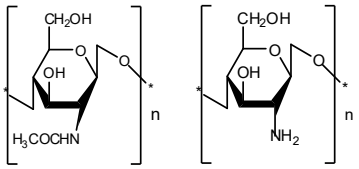

Fig. 6 Schematic representation of completely acetylated chitin [poly(N-acetyl-β-D-glucosamine], completely deacetylated chitosan [poly(D-glucosamine]

Several structural similarities exist between chitin and cellulose, including the equatorial glycosidic linkages and the conformation of monomers. Since both biopolymers act as defense materials and structural support in living organisms, their roles are similar. In the repeating unit of macromolecular chains, chitin has acetamide groups instead of -OH groups at the C-2 positions [36] (Figs. 6 and 7). As far as structure is concerned, both cellulose and chitin are polymers of monosaccharides made of β-(1 → 4)-2-deoxy-β-D-glucopyranose and β-(1 → 4)-2 acetamido -2-deoxy-β-D-glucopyranose respectively [37] (Fig. 7).

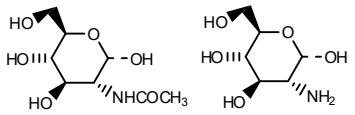

Fig. 7 Structure of (a) N-acetyl-glucosamine, monomer of chitin, (b) glucosamine, monomer of chitosan

There is more importance to chitosan than to chitin itself. It is a linear binary copolymer of 2-amino-2-deoxy-β-D-glucose, and β-(1 → 4)-linked N-acetyl glucosamine units and does not refer to a polysaccharide that is eccentrically defined, but a series of products with varying proportions of N-acetyl glucosamine and D-glucosamine units and varying chains of length. "Chitosan" is generally associated with polymers containing less than 25-40% acetyl.

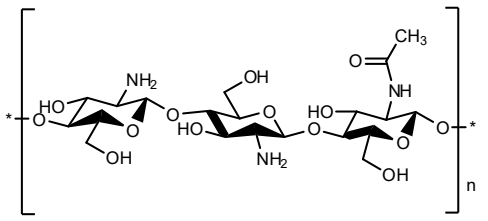

Fig. 8 The chemical structure of chitosan

A chitosan molecular structure (Fig. 8) consists of functional groups (such as amino and hydroxyl groups) that interact between chitosan-based composites to facilitate the formation of interconnected structures, which can be used to chemically change the chemically properties of chitosan [38]. Alternated biopolymers include Schiff bases, formed between primary amines in chitosan and carbonyl groups in aldehydes. In addition to their potential antimicrobial and biodegradable properties, chitosan with N-acylated derivatives has a characteristic of the –RC=N– group. Microorganisms such as bacteria, fungi, and algae are resistant to chitosan Schiff bases. Schiff bases derived from the reaction of chitosan with heteroaryl aldehydes have different environmental applications and

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are used as analytical reagents. By using the degree of substitution (DS), ^{1}H NMR of the amine and aldehyde groups of chitosan in the reaction can be calculated. In addition to assessing thermal stability, thermogravimetry is also used to characterize new compounds that may possess bioactive properties [39].

3.1 Chitosan Schiff Bases

Chitosan's reactive amino groups enable it to undergo several chemical alterations that lead to several essential functions. Biodegradability, hydrophilicity, nitrogen richness, ionic conductivity, crystallinity, and high viscosity make CSBs more attractive than other biopolymers. Among the reported chemical modifications of chitosan, imine functionalization is of particular significance because it results in the formation of the famous Schiff base. Chitosan's amino groups are typically conjugated with aldehyde/ketone carbonyl groups by the facile condensation of water molecules with aldehydes/ketones. A mixture of acetic acid and methanol was used by S. Hirano et al. to synthesize the first CSB in 1977 (Fig. 9) [40]. Various research groups report many CSBs following this. At either ambient or refluxing temperature conditions, acetic acid, ethanol, or methanol are generally used as the solvent medium to synthesize CSBs. In addition to the solvents mentioned above, CSBs can also be synthesized using DMF, water, and ionic liquid.

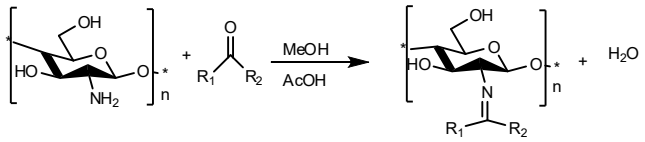
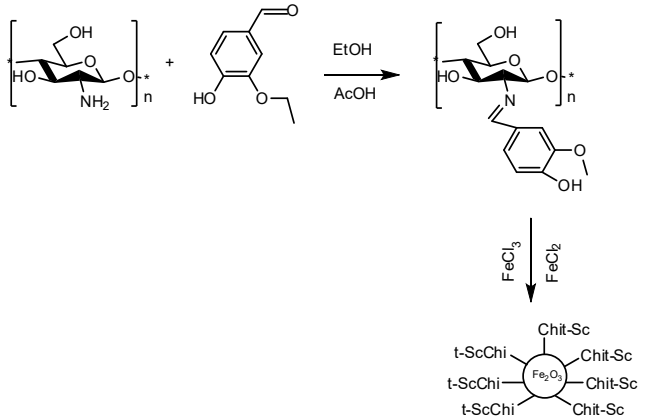


Fig. 9 The synthesis of Schiff base of chitosan

A general procedure was used to synthesize Schiff base complexes. The molecular weight and deacetylation of chitosan Schiff bases can affect their antibacterial activity. For example, Keisuke et al. found that less than 5 kDa4 molecular weight of chitosan promoted the growth of *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*). It was, however, completely antibacterial when it had a molecular weight of over 9.3 kDa. In a study by Mohan et al., 83% of deacetylated chitosan inhibited bacterial growth effectively [41]. Additionally, chitosan acts as an antioxidant, scavenging free radicals such as hydroxyl, DPPH, and superoxide. Chitosan has also been shown to inhibit tumor cell growth [42]. Besides suppressing tumor cell proliferation, it also enhances immunity, which inhibits tumor cell growth [43,44]. Conversely, chitosan increases lymphokine production and promotes T lymphocyte proliferation. Chitosan is also said to reduce cholesterol and blood pressure, promote small intestinal peristalsis, inhibit fat absorption, and improve immunity, according to different reports [45,46]. As a result, chitosan has become more popular.

4. Some Biologically Active Chitosan Schiff Bases

Since 1984, Schiff bases have been well known in the field of bioinorganic and medicinal chemistry due to their ease of synthesis, coordinating property, and versatile biological activity of their metal complexes. Under appropriate conditions, chitosan Schiff bases are synthesized by condensing chitons (containing amine groups) with carbonyl compounds. To the best, this review focused on those chitosan Schiff base ligands showing biological activity between 2018 and now. Listed below are some chitosan Schiff Bases with schematic representations of their structures and their biological activities.



Scheme 1 Synthesis of chitosan Schiff base 1 3EtO-4OH/CHSC and its 3EtO-4OH/CHSC/ Fe_2O_3 nanocomposite

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4.1 Chitosan Schiff Base (3EtO-4OH/Chit) and Its 3EtO-4OH/Chit/Fe_2O_3

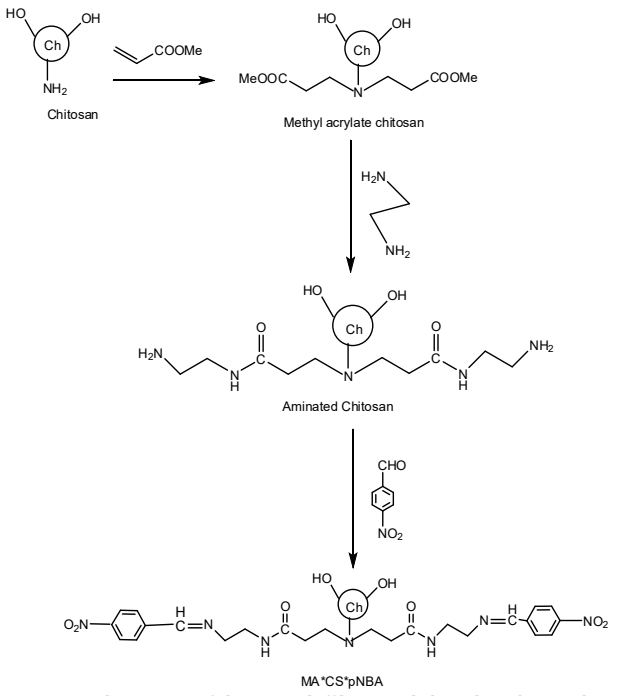
A new chitosan Schiff base (3EtO-4OH/Chit) and its 3EtO-4OH/Chit/Fe_2O_3 nanocomposite, which are derived from the condensation of chitosan and 3-ethoxy-4-hydroxybenzaldehyde and further addition of $FeCl_3{\cdot}6H_2O$ and $FeCl_2{\cdot}4H_2O$ to result in the chitosan Schiff base and confirmed the preparation of 3EtO-4OH/Chit and its 3EtO-4OH/Chit/Fe_2O_3 nanocomposite (Scheme 1) [47]. The catalytic materials were evaluated for their efficiency in removing methyl orange (MO) in an aqueous solution. In this study, dye removal was studied depending on the adsorbent dose of adsorbent and the contact time. It has been shown that 3EtO-4OH/Chit/Fe_2O_3 has excellent antibacterial activity and is more effective than chitosan or 3EtO-4OH/Chit against two Gram-positive (*S. aureus* and *B. cereus*) and two Gram-negative (*E. coli* and *P. aeruginosa*) bacteria.

*4.2 Chitosan Schiff Base Methyl Acrylate Chitosan Bearing p-Nitrobenzaldehyde (MA*CS*pNBA) Complex*

The chitosan derivative, methyl acrylate chitosan bearing p-nitrobenzaldehyde (MA*CS*pNBA) Schiff base was prepared as shown in the following Scheme 2 [48].

This biomaterial demonstrated significant antibacterial activity against MDR bacteria, anti-biofilm, and antioxidant activity, demonstrating hemocompatibility and no cytotoxicity. There was a significant negative correlation between it and MDR-PA-09 biofilm formation. This chitosan derivative MA*CS*NBAA had pharmaceutical applications, expanding the treatment ways of burn infections since it allied excellent antibacterial, anti-biofilm, antioxidant, anti-inflammatory, hemocompatibility, and, absence of cytotoxic activities.

Due to its excellent and significant antibacterial activity against MDR bacterial pathogens, anti-biofilm, antioxidant, and anti-inflammatory properties, the hemocompatibility, and cytocompatibility of this modified candidate were promising for pharmaceutical applications. This efficiency of MA*CS*pNBA might be due to the presence of p-nitrobenzaldehyde conjugated with the aminated chitosan. Therefore, this study may be a valuable platform to explore chitosan derivatives as new leading structures for pharmaceutical and biomedical therapeutics in the fight against MDR bacteria.



Scheme 2 Synthetic route of chitosan Schiff base methyl acrylate chitosan bearing p-nitrobenzaldehyde (MA*CS*pNBA) Complex

4.3 Zn(II) Metal Complexes Formed with the Biopolymeric Schiff Bases from Chitosan and Salicylaldehyde

The zinc complexes are made by condensing chitosan and salicylaldehyde to form biopolymeric Schiff bases. Various Schiff base complexes were synthesized from other aldehydes and metallic centers using the zinc(II) complex as a model system [49]. Ligands and their Zn(II) complexes (Fig. 10) were evaluated against tumor cells under optimized conditions. The biological activity of the Zn complexes increased with increasing concrete nitration of the free Schiff base compared to the chitosan alone.

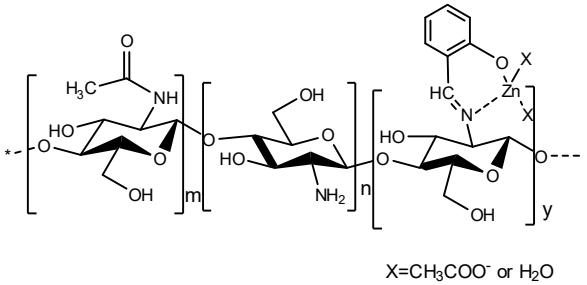


Fig. 10 Proposed structure of zinc (II) metal complexes formed with the biopolymeric Schiff bases from chitosan and salicylaldehyde showing acetylated (m), deacetylated (n), and the substituted by Schiff base (y) fractions of the biopolymer

4.4 Amphiphilic Biopolymeric Schiff Bases from Chitosan, Salicylaldehyde, and Glycidol

A Schiff base was synthesized from chitosans, salicylaldehyde, and glycidol by condensation (Fig. 11), and its antimicrobial activity against plant pathogenic microbes and human breast cancer cells (MCF-7) was tested. To improve the biological activity of chitosans, they were functionalized with salicylaldehyde and glycidol ([50].

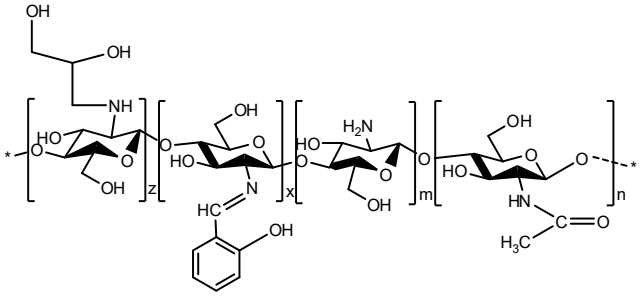
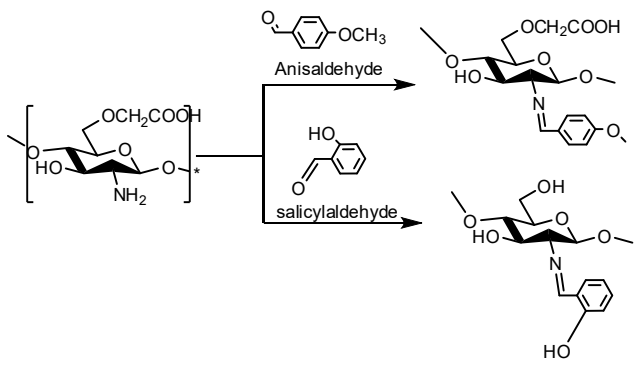


Fig. 11 Schematic representation of the synthesized amphiphilic biopolymeric Schiff bases from chitosan, salicylaldehyde, and glycidol: a) substitution with the salicylaldehyde hydrophobic group b) substitution and with the glycidol hydrophilic group.

New amphiphilic compounds prepared in this study were tested for their biological activity against agricultural microorganisms and tumor cells. A study showed that Schiff bases with amphiphilic ligands had greater antimicrobial activity than natural chitosans. A more substantial effect was noted against Gram-negative bacteria, particularly *P. syringe*, than against the fungus, *F. graminearum*. Moreover, they are more cytotoxic than natural chitosan against MCF-7 breast cancer cells when compared to natural chitosan. Additionally, modified and natural chitosans exhibited good selectivity for cancerous cells as compared to non-tumor fibroblasts. A conclusion can be made that the functionalization of chitosan with salicylaldehyde and glycidol improved the solubility of the synthesized amphiphilic biopolymeric Schiff base compared to natural chitosan, thus improving its antimicrobial and antitumor properties, thus broadening its potential applications for biological applications.

4.5 Chitosan Schiff Base of CMCS with Anisaldehyde or Salicylaldehyde

By refluxing an acidic solution of carboxymethyl chitosan (0.1 mol) with an equal amount of each aldehyde (anisaldehyde/salicylaldehyde) with an equimolar amount of silver nitrate, it was able to produce nanocomposites composed of anisaldehyde and salicylaldehyde carboxymethyl chitosan (CMC)-Schiff bases/silver nanoparticles (Scheme 3) [51].



Scheme 3 Schematic diagram for the reaction of CMCs with anisaldehyde or salicylaldehyde

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In addition to Gram +ve bacteria (*Bacillus Subtilis, Staphylococcus aureus, and Streptococcus faecalis*) were also evaluated against Gram -ve bacteria (*Escherichia coli, Neisseria gonorrhoeae, and Pseudomonas aeruginosa*). As shown by their results, they can inhibit the growth of both Gram +ve and Gram -ve bacteria in a range of well to high efficiency concerning their parent CMCs. CMCs-SB derivatives of $AgNO_3$ (5% by weight) also have higher antibacterial activity, according to the study. Anisaldehyde/CMCs-SB/AgNPs have a lower antibacterial efficiency than salicylaldehyde/CMCs-SB/AgNPs.

A hepatocellular carcinoma cell line (HEPG2) and a breast adenocarcinoma cell line (MCF7) were used to test the cytotoxicity of the prepared CMC Schiff bases in the absence and presence of AgNPs (2 and 5%). Incorporating AgNPs in weight ratios of 2 and 5% increased the anticancer activity of the Schiff bases. Incorporating AgNPs in weight ratios of 2 and 5% increased the anticancer activity of the Schiff bases.

4.6 Schiff Base of Chitosan and 2-Hydroxy-3-Methoxy Benzaldehyde and Its Metal Complexes

Medicinal compounds derived from chitosan are widely used due to their wide range of biodegradable and non-toxic properties. To enhance chitosan's biological properties, this work was conducted for the synthesis of a series of Schiff bases and their metal complexes Cu(II), Ni(II), Zn(II)). Chitosan (CS) was prepared by condensation of chitosan with 2-hydroxy-3-methoxybenzaldehyde (Fig. 12) [52].

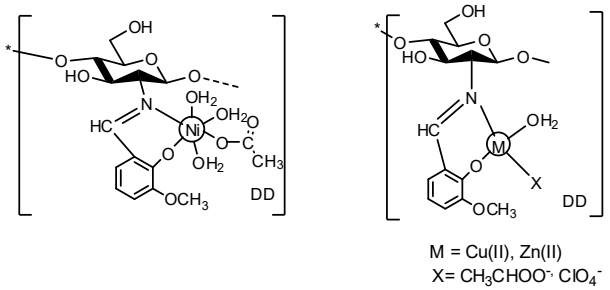

Fig. 12 Structure of synthesized schiff base of chitosan and its complexes, where DD = degree of deacetylation

In the MTT assay, chitosan, chitosan-Schiff base, and its metal complexes were tested against K562 chronic myelogenous leukemia (CML) and MG-63 osteosarcoma cancer cell lines. It was found that Schiff bases and their complexes were more effective against cancer MG63 cells than pure CS. Hence, the IC 50 values suggest that all complexes exert more excellent inhibitory effects than chitosan and chitosan-Schiff base, indicating that incorporating metals into the ligand significantly affects its antiproliferative properties. Based on the present data, all compounds except Schiff base chitosan appear to induce cell death via apoptosis pathways in K562 cells.

4.7 Schiff Bases from Chitosan and 5-Aryloxy-3-Methyl-1-Phenyl-1h-Pyrazole-4-Carbaldehyde

By condensation of pyrazole-4-carbaldehyde with chitosan in an acidic medium, 5-chloro-3-methyl-1-phenyl-1H-pyrazole-4-carbaldehyde (Fig. 13), and its derivatives were synthesized as reported in the literature [53].

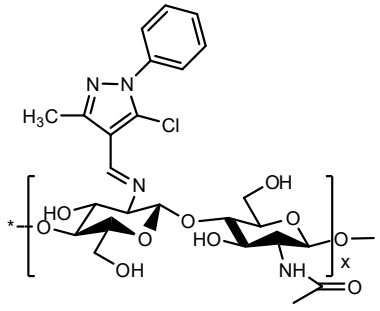

Fig. 13 Proposed structure of Schiff bases from chitosan and 5-aryloxy-3-methyl-1-phenyl-1H-pyrazole-4-carbaldehyde

Several bacteria were tested, including *Staphylococcus aureus, Bacillus subtilis, Klebsiella pneumonia, Escherichia coli*, and *Candida albicans* for their antimicrobial activity. There was a more significant inhibitory effect of Schiff bases on these microorganisms than chitosan, and the extent of the inhibition varied with the substitution nature. Heterocyclic substitutions significantly enhance chitosan's antimicrobial properties

against Gram-positive and Gram-negative bacteria and fungi. A lower concentration of modified chitosan was also effective against the tested microorganisms. A combination of enhanced hydrophobicity and metal binding capability due to the presence of imine functionality is thought to contribute to the enhanced antimicrobial activity.

4.8 Schiff Base of Chitosan with Indole-3-Carboxaldehyde and 4-Dimethyl Amino Benzaldehyde

In terms of preparing new derivatives, chitosan can be modified to contain hydroxyl and amine groups. The chitosan amine group was coupled to the carbonyl groups of Indole-3-carboxaldehyde or 4-dimethylaminobenzaldehyde to produce Schiff bases (I & II). According to the potentiometric analysis, Schiff bases (I) and (II) (Fig. 14) are substituted to 1.15 and 12.05%, respectively [54].

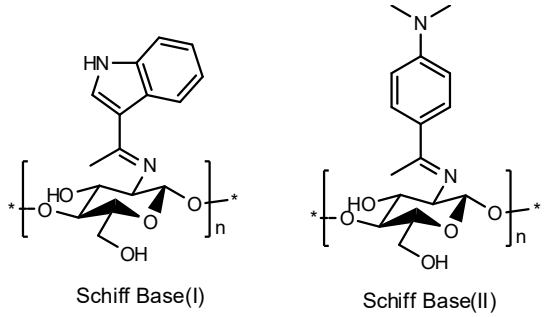

Fig. 14 Structure of synthesized chitosan Schiff bases (I & II)

There was a significant increase in the antimicrobial activities of Schiff base (I) compared to Schiff base (II) and chitosan. Indole-3-carboxaldehyde significantly enhanced the antimicrobial activity of chitosan compared to 4-dimethylaminobenzaldehyde in the antimicrobial activities. Schiff base (I) has been shown to inhibit Gram-positive bacteria by 99% at its highest concentration. The highest inhibition rate was recorded by Schiff base (II) versus Gram-positive, at approximately 82%.

Using MTT assays, the developed materials' cytotoxicity was assessed in fibroblast cells. These findings indicate that Schiff bases could treat wound infections as antimicrobial alternatives to pure chitosan.

4.9 Fabrication of Hydrogels I and II

A new Schiff base hydrogel based on chitosan was fabricated in the current study by modifying chitosan with an iso-nicotinic aldehyde to yield hydrogel I, or using epichlorohydrin or sodium tripolyphosphate via ionotropic gelation processes under the same reaction conditions to produce nanohydrogels II respectively (Fig. 15) [36].

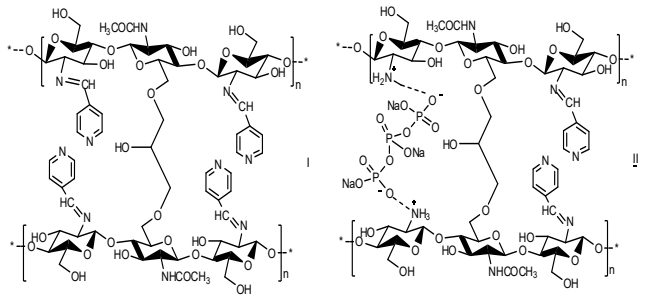

Fig. 15 Structure of synthesized fabrication of hydrogels I & II

Hydrogels fabricated from various solvents were studied for swelling behavior. To remove the cobalt and mercuric ions, fabricated hydrogels were shown to increase adsorption capacity with increasing immersion time. The fabricated hydrogel II exhibited 92.5 and 95.9% efficiency for cobalt and mercuric ion removal at 10 h, respectively. As metal ions concentration increased, the adsorption capacity increased, with hydrogel II showing the highest adsorption capacity. After six cycles, the hydrogel's reusability showed high adsorption capacity, which is considered promising for cobalt and mercury treatment of water.

According to the results of the evaluation of fabricated hydrogels, hydrogel II had the highest antibacterial activity against Gram-positive and Gram-negative bacteria (*E. coli* & *proteus*) compared to standard antibiotics (Bacterial Inhibition Zones, MIC, and MBC). Hydrogel II is effective and may have the potential as an antibacterial drug.

There is remarkable bioactivity in all the chitosan derivatives discussed above. Further research should focus on the relationship between the structure-activity of the chitosan derivatives obtained from these results.

5. Applications

As a biomaterial, chitosan's versatility and cationic nature make it ideally suited for a wide range of biological and chemical applications. There are many applications for chitosan and its derivatives, including solutions, suspensions, particles, gels, hydrogels, foams, membranes, films, and fibers, and a scaffold is used in a variety of fields, including medicine, pharmacy, biomedicine, hygiene, cosmetology, personal care, agro chemistry, food industry, agriculture, textiles, nutrition, paper manufacturing. Furthermore, the use of chitosan in electrochemical sensors, bio-imaging, green solvents, detergents, adhesives, and bio-catalysis is the subject of a significant amount of study. Nonetheless, the food and nutrition industry and the cosmetic, pharmaceutical, and medical fields are often considered to be the most significant sectors. The following are some of the standard applications of chitosan derivatives.

5.1 Food Industry Applications

Chitosan is used as a thickener, de-colorant, or stabilizer in food manufacturing. After consuming chitosan derivatives, food pigments are absorbed into complexes that can't be absorbed into the human body, thereby reducing pigment toxicity. It is effective in adsorbing polyphenolic compounds in the solution as well as reducing the total solids and heavy metals in the solution. As a preservative, it stops the development of microorganisms and keeps food lipids from turning rancid and oxidizing. It also acts as an antioxidant and bacteriostatic agent. Approximately 75% of lipid peroxidation was reduced in meat treated with chitosan VC complex [55]. Due to its superior film-forming and antibacterial capabilities, chitosan and its derivatives are used in food packaging and preservation. Chitosan film is edible, non-toxic, and insoluble in water. It also has superior antibacterial, freshness, and moisturizing benefits on food when compared to regular plastic wrap [56,57]. Biodegradable chitosan films are scarcely harmful to the environment. Uranga and coworkers prepared a composite film from citric acid, fish gelatin, and chitosan that exhibited good UV-blocking properties and inhibited *E. coli* [57]. Nanoparticles of chitosan-ZnO were added to sodium alginate, enhancing the composite film's tensile strength and its water vapor resistance. As a result of this film, grapes were able to keep for a longer period. Chitosan and its derivatives products are feasible for food packaging, according to these findings [58,59].

5.2 Industries

Chemical compounds like chitosan can chelate and adsorb heavy metal ions and organic compounds, including Cd^{2+}, $Pb^{2+,}$ Co^{2+}, Hg^{2+}, Cu^{2+}, etc. During this time, Chitosan prevents water pollution by capturing radioactive elements, such as Co, Pu, etc., [59]. Combining chitosan with activated carbon and ion exchange resin, bacteria, Fe^{2+}, and Cl_2 can be removed or reduced from tap water. In addition, chitosan is also a natural polymer flocculant, which helps reuse wastewater by flocculating the active ingredients. Thus, chitosan and its related products can be used in different ways to clean water. Also, two chitosan derivatives that could dissolve in water, N-N-N-triethylammonium chitosan and carboxymethyl chitosan, took heavy metals out of water solutions, indicating that they could be used to treat water [60].

It improves the film production and moisturizing properties of cosmetics by adding chitosan. In the cosmetics sector, hair care treatments that cure yellow hair, make the hair simpler to comb, and make hair color more vibrant are often utilized. Hair fibers are covered with a transparent elastic film formed by chitosan and its cationic derivatives. As a result of these films, hair becomes softer and stronger, and it is prevented from being damaged. Chitosan and PNIPAAm/PU hydrogel combine to create a mask that is not only comfortable and skin-friendly but also antibacterial too [61]. When the quaternized carboxymethyl chitosan (QCMC) was combined with cosmetic formulae to create beauty creams, the moisture absorption and water retention qualities of QCOM were superior to those of hyaluronic acid. It moisturized the human cuticle significantly. Several oral hygiene products contain chitosan to reduce or remove bad breath, prevent periodontal ulcers, and prevent periodontal disease. Plaques were reduced by 70% and bacteria by 85% when chitosan gel containing herbal extracts was used. A reduction in deformed streptococcal colonies was found by adding chitosan to toothpaste and mouthwash. The antibacterial activity of chitosan-based mouthwash was superior to that of commercial mouthwash, which suggests that chitosan could replace commercial mouthwash [62].

A significant use of chitosan in the textile sector is the preparation of fibers from chitosan. The non-toxicity, biodegradability, biocompatibility, and antibacterial properties of chitosan fiber are good. In this way, it prevents bacteria and fungi from growing on textiles. To produce nanometer-sized chitosan fibers, electrospinning is used. The fiber

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diameter, tensile strength, and biodegradability of chitosan fibers were determined by wet spinning by Judawisastra et al [63]. Li and its co-worker synthesized 21.6% stronger fibers from chitosan using LiOH/urea solvents by wet spinning chitosan fibers. The superior moisture absorption and antimicrobial qualities of chitosan fibers led to the development of textiles for baby clothes, undergarments, and bedding [64]. As well as being used for the antibacterial, antiwrinkle, and antistatic finishing of textiles, chitosan is also used as a dyeing enhancer. Additionally, chitosan nanoparticles have pH-responsive properties, can deliver drugs, and could be used as cancer drug delivery carriers.

5.3 Medicine and Health

The human body can absorb chitosan as it is non-toxic, easily formed into films, biocompatible, and easy to form. A good drug carrier, it has excellent properties. In addition to delivering drugs in a body, chitosan could readily decompose by the lysozyme and be absorbed by the tissue, making it an attractive carrier for drug delivery. Various sustained-release products can be made with Chitosan by dissolving, coating, or adhering. These include microspheres, tablets, gels, and microcapsules. Folic acid combined with chitosan delivers doxorubicin through a core-shell drug carrier that releases 30.05% of the drug in the first 4h and continues to release 88.26% in the following 72h. Doxorubicin-loaded nanoparticles have the potential to be effective drug carriers for the treatment of solid tumors through their selective targeting and prolonged release of the medication [65]. Eastern medicine chitosan/polyethylene oxide (PEO) mix film made by solution casting continuous release of vitamin B12 and *Acanthopanax senticosus* (ES). To adjust the mesh size of the film, PEO was added. It is possible to control the drug release rate by changing the molecular weight and the amount of PEO. The ES release rate was influenced by the mesh size of the mesh and the intermolecular force between chitosan and ES, and the crosslinking density of the blended film controlled the vitamin B12 release rate. Carboxymethylcyclodextrin-grafted chitosan (CMCD-g-chitosan) nanoparticles could serve as protein drug carriers. Aside from being pH-responsive, chitosan nanoparticles can deliver drugs and are expected to be used in cancer treatments [66].

Wound healing is a dynamic and complicated process that must be avoided to prevent infection throughout the healing process. As a component of skin tissue, N-acetylglucosamine (NAG) in chitosan plays a vital role in repairing scar tissue. Chitosan supports cell growth through its high positive surface charge, resulting in thrombosis and blood clotting. As well as having free amino groups on their surface, chitosan films can form complexes with blood cells' acidic groups. As a result, chitosan can enhance wound healing and is an excellent wound-dressing material. In surgically abraded corneas, both iPSCs and CHC hydrogels promoted wound healing. Through down-regulation of oxidative stress and recruitment of endogenous epithelial cells, iPSC/CHC hydrogels enhanced corneal reconstruction in severe corneal damage caused by alkali. Whole blood coagulation was sped up using guanidine-modified chitosan, and hemostatic characteristics were discovered [67]. Hydrogels containing chitosan-loaded cyclodextrin (CDPECs) reduced blood loss and shortened hemostatic time compared to commercially available absorbable hemostatic dressings. The material reduced blood loss when tissue was damaged. In a study of mouse skin wounds, Ding et al. reported that double-layer wound dressings containing chitosan and Ag nanoparticles significantly improved healing rates [68]. So, wound dressings could greatly benefit from this new double-layer composite material.

Chitosan is a natural cationic polysaccharide, which is non-toxic and safe for human tissues and blood use. Besides its antibacterial, hemostatic, and analgesic properties, it also has a special biological activity that stimulates the proliferation of human cells and tissue repair. As a result of its relatively stable properties, chitosan is not easily deformed in the body. In humans, lysozyme degradation could allow these materials to be absorbed into the body. Stents are simple to mold into the necessary form for supporting and regenerating tissue [69]. Chitosan is a great scaffolding material because of this. It can make skin, blood vessels, cartilaginous, corneas, and liver tissue. Chitosan is also very good at maintaining moisture, toughness, and breathability, so it can be used to repair and regenerate human skin and corneas. Using hydroxyapatite/gelatin chitosan composite nanofiber bionic scaffolds, Chen et al. achieved excellent biocompatibility and increased viability of MG-63 cells, making the scaffold suitable for bone tissue engineering. Using magnesia poly (-caprolactone) and chitosan, Rijal and co-worker developed composite nano-fibers for tissue engineering. Lim et al. has developed a biocompatible bilayer chitosan porous skin regenerating template (CPSRT). A CPSRT implanted with human skin fibroblasts could also rapidly spread, proliferate, and cover the holes inside the device. CPSRT templates are therefore ideal for adhesion of HDF and can be used for skin tissue engineering [70].

5.4 Agriculture

Seeds can be protected from fungal pathogens by wrapping them in chitosan film, which increases their resistance to fungi. According to Wange and coworkers Fusarium vesicular wilt resistance is induced by chitosan in camellia. Chitosan could also be broken down by microorganisms to provide nutrients to plants, thereby stimulating actinomycetes to grow. Furthermore, chitosan and its derivatives give plants better resistance to disease, cold, and fall, reducing pesticide and fertilizer use. Moreover, heavy metal ions in soil may be bound by chitosan while also purifying it. The company also used chitosan as a pesticide additive, fertilizer buffer, seed treatment, and soil purification mediator. Among its many uses, Chitosan reportedly proved to be plant and pest-resistant, reduced pesticide usage, and could provide an effective pest control method. If mixed with pesticides, fertilizers, herbicides, and other compounds, it could sustain its functional release for a longer period of time [71].

5.5 Some Other Applications

Several excellent properties are found in chitosan and its derivatives, which are utilized to make useful materials. With the development of technology and applications for polymer synthesis, polymer films have become a new material and a field of future research. Film materials are characterized by permeability, mechanical strength, and good physical properties. As well as excellent biological properties, it also exhibits excellent chemical properties. In recent years, chitosan and its derivatives have become increasingly popular for making films with the above individuality.

Certain chitosan compounds have catalytic properties. Transition and uncommon earth metals were preferentially chelated by chitosan because of the nearby hydroxyl and amino groups on its chemical chain. As a result, derivatives could serve as catalysts [72]. Chitosan possesses asymmetrical carbon atoms inside the chain and a unique helix form despite some metal chelation. A selective modification of hydroxyl groups was used in enzyme research to simulate enzyme activity artificially. Chitosan has a rigid or semi-rigid molecular chain. The chain of molecules in the solution has a rod-like shape. Hydrogen bonds and asymmetrical carbon atoms in the molecular chain provide polar groups and optical properties. A greater degree of anisotropy was present in the molecule. Chitosan's unique structural characteristics made it an ideal material for liquid crystals [40, 73]. Therefore, chitosan has excellent potential for liquid crystal applications.

6. Conclusion

As a result, chitosan must pay attention to various industries in different fields of health, biochemistry, chemistry, agriculture, environment, etc. This growing interest is primarily due to its unalienable biological properties and resourcefulness as a biomaterial. Antibacterial, antifungal, anti-inflammatory, antioxidant, and anticancer properties are attracting much attention for chitosan Schiff base complexes. Medicinal compounds derived from chitosan are widely used because they are non-toxic and biodegradable. Recently, chitosan and its derivatives have been studied extensively for their antimicrobial properties. Scientists have also produced novel antimicrobial substances with potential action against the new version of harmful germs, owing to the increasing resistance of microorganisms to antimicrobial materials. The complexes discussed in this review demonstrated remarkable biological activities, including antibacterial, antifungal, anti-inflammatory, antioxidant, and anticancer activities. Scientists and industrialists continue to discover new possibilities for chitosan with various forms and modifications. Thus, this review can provide confident that many chitosan Schiff bases complexes may represent promising molecules for developing new and effective therapies. Shortly, more industrial development is anticipated in the areas of anticancer medications, catalysis, wrapping materials, sensor applications, gene delivery, packaging, cosmetotextiles, and bioimaging.

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