REVIEW PAPER

Modification of Konjac Glucomannan to Carboxymethyl Konjac Glucomannan (CMKGM) and its Development as a Drug Carrier

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ABSTRACT
Konjac glucomannan is natural polysaccharide that has physical properties in the form of high viscosity and gel-forming capacity that can be widely utilized in various fields, especially the pharmaceutical and food industries. However, the characteristics of high viscosity and limited stability of the solution are things that need to be considered in the application of konjac glucomannan directly. Chemical modification, particularly through carboxymethylation, produces carboxymethyl konjac glucomannan (CMKGM) derivatives characterized by increased water solubility, decreased viscosity, and improved thermal stability. CMKGM shows potential as carrier in more specific drug delivery systems by providing controlled and responsive drug release under physiological conditions, such as pH changes and enzymatic activity. This study reviews the basic structure of Konjac glucomannan, chemical modification techniques, and the applications of CMKGM in the development of both in vitro and in vivo drug delivery systems.
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INTRODUCTION

Konjac glucomannan or KGM is polysaccharide, it extracted from konjac plant, which is widely distributed in nature. The chemical structure its main chain consists of condensation copolymer of D-glucose and D-mannose, the molecule is enriched with active hydroxyl groups. Konjac glucomannan exhibits diverse functional properties, including water retention, thickening, gel formation, and the ability to generate a low-calorie thin film. Due to these characteristics, konjac glucomannan has extensively applied in the food, biomedical, and materials industries[1]. Konjac glucomannan is a polysaccharide characterized by distinct physicochemical properties, including high viscosity, substantial water absorption capacity, and pronounced gel-forming ability. Nevertheless, these same characteristics may also pose challenges that restrict its utilization in certain applications. For instance, the exceptionally high water absorption capacity of konjac glucomannan can lead to complications by excessively retaining water, whileits elevated viscosity hinders processing and limits its use in contexts that require lower viscosity. To address as modification strategy, aiming to improve thermal stability, reduce viscosity[2], and enhance hydrophobicity[3]. Chemical modification serves as an effective approach to impart desired fuctional properties to polysaccharides. Several techniques, such as deacetylation, oxidation, carboxymethylation, and ᵞ-irridation, have been employed to tailor the physicochemical behavior of konjac glucomannan[1]. such modification are particularly and broader application potential, especially in drug delivery system.

Accordingly, in this reviews aims to provide an overview the fundamental chemical structure of konjac glucomannan and its relevant physicochemical properties in pharmaceutical application. Furthermore, it highlights various chemical modification strategies, with an emphasis on carboxymethylation, to enhance properties such as solubility, stability, and gel-forming capacity for more effective drug carrier performance. Special attention is also given to carboxymethyl konjac glucomannan (CMKGM), focusing in its distinctive features ad drug delivery matrix and its applications in the development of in vitro and the in vivo drug delivery systems, along with its advantaes and limitations.

Structure & Properties of Konjac Glucomannan

Chemical Structure

Konjac glucomannan consists of D-mannose and D-glucose units linked by β-1,4 glycosidic bounds. The molar ratio

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from mannose to glucose is approximately 1.6:1, with several branching point occurring at the C-3 position of mannose residues. A dispersion of KGM (0.5g/100g) demonstates the highest viscosity among twelve types of polysaccharides, such as κ-carrageenan, gum Arabic, xanthan gum, are known to exhibits shear-thinning behavior, characterized by a decrease in viscosity under applied shear forces[4] Other studies reported that konjac glucomannan possesses a linier backbone consists of D-glucose and D-mannose linked by β-1,4 bounds, with a molar ratio ranging from 1:1.6 to 1:1.4 depending on the source. Additionally, konjac glucomannan contains occasional branches formed through β-1,3 linkages on mannose residues and features acetyl groups randomly substituted at the C-6 position, approximately once every nineteen sugar residues[5]. Despite its considerable potential, native konjac glucomannan present certain limitation, including suboptimal rheological properties, low stability in aqueous solutions, susceptibility to degradation, and difficulties in storage[6]

Types of Chemical Modifications

Some chemical modifications can be seen in Table 1 and are described as follows.

Table 1. Chemical modification of KGM

Modification Technique	Results	References
Deacetylation	KGM was deacetylated using L-arginine to produce three deacetylated variants with the degree of deacetylation (DD) of 17.51%, 35.41%, and 54.52%, respectively, without changing the primary structure or KGM molecular weight. The solubility of KGM decreased 85.38% to 81.17%, molecular morphology changed from a chain structure to an aggregate resembling a micelle, the gel network strength of the mixture of KGM and κ-carrageenan increased.	7
Carboxymethylation	KGM undergoes carboxymethylation modification with varying degrees of substitution (DS) to produce carboxymethylated KGM (CMKGM). KGM carboxymethylation decreases its molecular weight and the viscoelasticity properties, while improves the thermal stability and the water solubility, weakens gel, but maintains viscosity sensitivity to temperature and enzyme resistance, so this negatively charged CMKGM is suitable for drug delivery systems to the colon.	8
Carboxymethylation	Film nanocomposites are made by introducing functional carbon nanotubes (PCNT) and a gallic acid into the carboxymethyl konjac glucomannan and a gelatin matrix, resulting in amide and hydrogen bonds that improve mechanical properties, moisture barrier, and antimicrobial activity against Staphylococcuscaureus and Escherichia coli, potentially extending the shelf life of food products.	9
Grafting	KGM was estered with oleic acid using lipase enzymes in isooctane under optimal conditions to produce KGM ester with a substitution degree of 0.370 which showed an increase in hydrophobicity, changes in rheological properties, and water resistance, so it could potentially be used as a moisture-sensitive coating and hydrophobic drug delivery system.	10
Oxidation	Hydrogel DUS@MOK-GEL is made from methacrylated oxidized konjac glucomannan crosslinked with acrylamide and loaded with UiO-66 containing sodium ferulate as well as deferoxamine, resulting in wound dressings with physical properties such as swelling capacity, moisturecretention, and water vapor permeability, as well as biological activities such as antioxidants, anti-inflammatory, macrophage regulation, and angiogenesis stimulation that accelerate diabetic wound healing.	11

Esterification and Acylation

At the C-6 position of approximately every nineteen sugar residues, an acetyl group in randomly substituted. This structural feature plays a critical role in imparting unique colloidal properties to konjac glucomannan, there by enhancing its functionality as a macromolecule. Previous studies have demonstrated that the efficiency of deacetylation is strongly influenced by both the type and concentrationof base imployed. Conventional bases, such as sodium hydroxide (NaOH) and sodium carbonate (Na_2CO_3), are most commonly utilized, however their use may compromise the molecular chain integrity of konjac glucomannan. Alternative approaches, including the use of L-arginine, potassium hydroxide (KOH), or mechanochemical treatment, have been show to effectively remove acetyl groups while simultaneously altering the molecular structure and physicochemical characteristic of konjac glucomannan[7]

The deacetylation process reduces solubility, transparency, and water absorption capacity, while inducing chanes in molecular morphology that result I the formation of micellar-like aggregates. Furthermore, it promotes the development of entangled network structures, there be enhancing gel strength through hydrogen bonding and hydrophobic interactions[12]. Deacetylated konjac glucomannan exhibits improved thermal stability and decreased thixotropy, along with enhanced mechanical properties such as film strength and water resistance[13]

Acetylation of konjac glucomannan to varying degrees significantly influences its solubility, viscosity, and gel-forming capacity. The presence of acetyl groups is essential in maintaining the helic conformation of konjac glucomannan, a structural requirement for gel formation[14]. Moreover, acetylation improves water retention and gelation properties, making konjac glucomannan particularly suitable for applications that demand high moisture retention and controlled release behavior[15]

Carboxymethylation

Kkonjac glucomannan is a neutral polysaccharide, which constrains its applicability in drug delivery systems that rely on electrostatic interactions. To broaden its utility, konjac glucomannan can be chemically modified through carboxymethylation to yield carboxymethyl konjac glucomannan (CMKGM), a carboxylated negatively charged derivative characterized by improved solubility, swelling capacity, and stability. Carboxymethyl konjac glucomannan exhibits significant potential as a drug carrier, particularly in targeted delivery approaches, including the development of nanoparticle-base systems and enzyme-responsive platforms. Despite its application across pharmaceutical and other fields, studiea examining the coordination interactions og carboxymethyl konjac glucomannan polyelectrotytes remain limited[16] The carboxymethylation process involves two main stages alkalization and a etherification. During the alkalizations step, konjac glucomannan is treated with sodium hydroxide (NaOH) to generate alkoxide intermediates. Subsequently, in the etherification step, these intermediates react with monocloroacetic acid (MCA), introducing carboxymethyl groups into the polymer backbone and ultimately producing CMKGM[17].

Grafting (attachment of other polymer chains), oxidation, and other modifications.

Graftin represent effective strategy for enhance the solubility of polymers by introducing functional groups that improve their compatibility with solvents. For instance, grafting hydrophilic monomers to polymer backbones increase their solubility in polar solvents[18]. A commonly employed technique involves grafting methacrylic (MA) to konjac lucomannan through free radical initiation. This process depends on parameters such as reaction temperature, the konjac glucomannan to methacrylic ratio, and initiator concentration, and the resulting products are typically characterized using FTIR spectroscopy[19]. Another approach involves graftin bioactive peptides to konjac glucomannan to yield pH-responsive derivates. These modified konjac glucomannan hydrogels promote cell adhesion, spreading, and interaction, there demonstrating suitability for tissue engineering aplications[20]. Overall, grafting enhances the mechanical strength, water retention capacity, and stability of konjac glucomannan, broadening its applicability across diverse industries[21]. Importantly, grafted konjac glucomannan retains its intrinsic biocompatibility and biodegradability, supporting its use in both biomedical and environment applications [22].

Oxidation is another important modification strategy for konjac glucomannan, significantly altering its structure and physicochemical properties. Frequently combined with acidolysis, oxidation disrupts the particle structure of konjac glucomannan, reduces its crystallinity, and produces a rougher surface morphology. Unlike carboxymethylation, oxidation does not incorporate carboxyl groups in to the polymer chain but instead reduces the viscosity of konjac glucomannan [23]. Oxidized glucomannan (OKM) has shown to improve the quality and tensile properties wheat flour dough, enhance water absorption, and lower the setback value. Moreover, OKGM increases both the storage (G') and loss (G'') moduli of dough, reflecting improvements in its rheological performance [24].

Carboxymethyl Konjac Glucomannan (CMKGM) Structure and Synthesis

Carboxymethyl konjac glucomannan (CMKGM) is synthesized by introducing carboxymethyl groups into konjac glucomannan backbone through an etherification reaction using sodium chloroacetate in an alkaline conditions. The substitution of carboxymethyl groups predominantly occurs at the C6 position of the glucose residues in the konjac glucomannan chain. The synthesis procedure involves the reaction of konjac glucomannan with the etherifying agent, accompanied by classification and catalysis, typically conducted in an ethanol medium. Optimal reaction parameters have been report at 55°C and pH 12 for a duration of 3 hours. The successful modification is confirmed by FTIR spectroscopy, as evidenced by an increased intensity of the carbonyl absorption peaks [25].

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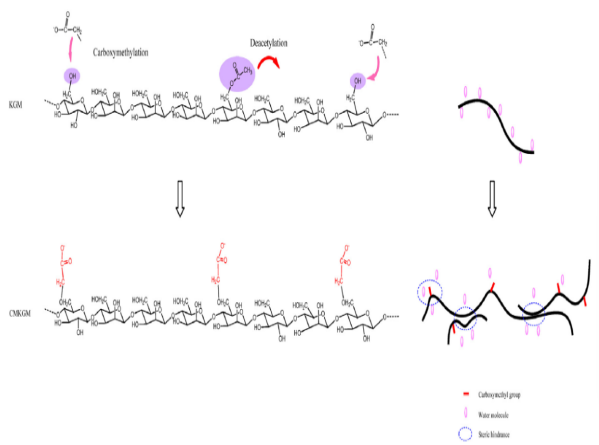

Figure 1. Carboxymethylation process in glucomannan conjunctions[26].

Carboxymethylation enhances the aqueous solubility of konjac glucomannan, rendering carboxymethyl konjac glucomannan (CMKM) more soluble than the native polyemer. Nonetheless, certain studies have reported that CMKGM may exhibit reduced solubility under specific cconditions[26], this reduction is attributed to a decline in viscosity compared to unmodified konjac glucomannan, an effect that becomes more pronounced at higher degrees of substitution (DS). Such changes can be explained by decreased molecular entanglement resulting from reduced molecular weight (Mw) and weakened intermolecular hydrogen bonding[27]. Carboxymethylation also diminishes the gel-forming ability and hardness of konjac glucomannan gels, although CMKGM retains the capacity to form gels under particular conditions. These gels are characterized by favoravble mechanical strength and hih thermal stability[28]. The DS of CMKGM critically influences its mechanical rigidity and swelling behavior, both of which are essential determinants of drug release performance[29]. Furthermore, CMKGM can be employed in layer-by-layer assembly to construct pH-responsive delivery systems, enabling the formation of multilayered structures capable of encapsulating drugs and releasing them in response to pH variations[30]. It can also be cross-linked with other polymers, such as chitosan, to yield stable hydrogels with enhanced mechanical properties and controlled drug release profiles[31]. Additionally, CMKGM is able to form stable coacervates through interactions with positively charged polymers, such as ovalbumin, which are advantageous for targeted drug delivery applications. Notably, CMKGM has been explored as a carrier for colon specific drug delivery, exemplified by formulations with 5-aminosalicylic acid (5-ASA)[32]. The pH sensitivity of CMKGM hydrogels makes them suitable for site specific gastrointestinal drug delivery. For instance, formulations intended for intestinal delivery demonstrate enhanced drug release under alkaline conditions[33]. Drug release from these hydrogels frequently follows a non-Fickian diffusion model, indicating that both polymer relaxation and diffusion contribute to the release mechanism[34]. Simulated astrointestinal studies have further confirmed their efficiency for example, carboxymethyl konjac glucomannan hydrogels containing curcumin achieved cumulative release exceeding 90% at intestinal pH, highlightin their potential in oral delivery[35,36,37]. Animal experiments have also demonstrated the theurapeutic promise of CMKGM based systems. Curcumin loaded microspheres, for example, were shown to alleviate inflammation in colitis-induced mice, supporting their applicability for targeted delivery[38]. Moreover, modifications involving carboxymethylation combined with deacetylation can impart hydrophobic characteristics to CMKGM, there by broadening its applicability across diverse fields.

Applications and Case Studies in Drug Delivery

Various applications and studies of the use of carboxymethyl konjac glucomannan (CMKGM) in development of drug delivery systems have been carried out. Some examples of such delivery systems use a combination of polymers and various formulation techniques such as the formation of polyelectric nanoparticles, pH-responsive hydrogels, and interpenetration polymer networks, which significantly improve drug release control and targeting. Some of the applications and case studies in drug delivery can be seen in Table 2 and explained as follows.

Table 2. Applications and Case Studies in Drug Delivery.

Polymer	Application of Delivery Systems	References
Carboxymethyl konjac glucomannan (CMKGM) - *sodium carboxymethyl cellulose* (SCMC)	CMKGM combined with sodium carboxymethyl cellulose (SCMC) formed interpenetrating polymer network (IPN) beads through an ionic gelation method using aluminum chloride (AlCl3) as a crosslinker to produce beads with the release of special pH-controlled ibuprofen drugs on the oral administration route.	39
carboxymethyl konjac glucomannan-chitosan (CMKGM-CS)-alginate	Carboxymethyl konjac glucomannan-chitosan (CMKGM-CS) nanogel to stabilize single and double emulsions, which are then combined with alginate as a microencapsulated matrix for delivery of targeted probiotics to the gut.	35
Carboxymethyl konjac Glucomannan (CMKGM)	CMKGM has a negative charge that can be utilized in layer-by-layer (LbL) depositions and complex coacervation techniques to form pH-sensitive drug delivery system and β-mannanase enzymes in the colon, thus enabling controlled and specific drug release in colon. The polymer has resistant to digestive enzymes in the stomach and small intestine,	27

	while being degraded by β-mannanase enzymes in colon, making highly potential for application in drug delivery formulations that specifically release drugs in the colon.	
Carboxymethyl konjac Glucomannan (CMKGM)	Carboxymethyl konjac glucomannan (CMKGM) is an anionic polymer used in colon targeted drug delivery systems through complex process of coacervation with ovalbumin (ova) controlled by pH, mass ratio, temperature, and ionic strength to produce coacervates with controlled drug release properties.	[40]
Carboxymethyl konjac glucomannan (CKGM) and chitosan (CS)	Carboxymethyl konjac glucomannan (CKGM) and chitosan (CS) are made nanoparticles through polyelectrolyte complexation with the addition of a drip negatively charged CKGM solution into a positively charged chitosan solution in acetic acid solution accompanied by ultrasonic sonication to form nanoparticles as a protein delivery system with controlled protein release of bovine serum albumin (BSA).	[41]
Carboxymethyl konjac glucomannan (CKGM) and chitosan (CS)	Chitosan (CS) and carboxymethyl konjac glucomannan (CKGM) polyelectrolyte complexation method by dripping CKGM solution onto CS solution while sonicated, this process produces nanoparticles for protein delivery of bovine serum albumin model.	[42]

Overall, the application of CMKGM in drug delivery systems shows flexibility and superiority, especially in targeted delivery applications to the gastrointestinal tract, with the ability to adapt physicochemical properties that support the gradual release of drugs, so that CMKGM can be used as one of the potent polymers to be developed further.

CONCLUSION

Konjac glucomannan (KGM) is a natural polysaccharide characterized by its thickening and gel-forming properties. However, its practical application is constrained by drawback such as high viscosity and limited solution stability. Chemical modification, particularly carboxymethylation, derivates is carboxymethyl konjac glucomannan (CMKGM), wich exhibit improved solubility, enhances stability, and reduced viscosity. Carboxymethyl konjac glucomannan demonstrates favorable gel-forming capacity and pronounced responsiveness to pH variations, making it highly suitable as a carrier for targeted drug delivery. Various CMKM based delivery systems, including hydrogels, nanoparticles, and coacervates, have shown effectiveness in achieving controlled release and gastrointestinal specific targeting. Despite challenges releated to large scale production and ciocompatibility, carboxymethyl konjac glucomannan presents significant potential for future pharmaceutical applications..

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