



ADVANCEMENTS IN CARBOXYMETHYL CELLULOSE APPLICATIONS IN FOOD INDUSTRY AND WASTEWATER TREATMENT

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Abstract:

Carboxymethylcellulose (CMC) is a water-soluble cellulose derivative that can produce robust and durable films for a variety of uses as well as control the rheology and viscosity of aqueous systems. Initially, CMC was introduced as a substitute for starch and natural gums but frequently CMC emerged as a promising derivative with characteristic surface properties for various advanced applications. The use of this cellulose derivative is thus widespread in the production of paper, the processing of detergents, textiles, protective coatings and drilling fluids. The food, drug, and cosmetic sectors all frequently employ the refined variety, commonly known as cellulose gum. Currently, CMCs are also used in biomedical engineering, as diverse conductive agents in the textile, pharmaceutical, and food industries. Moreover, CMCs are effectively used in wastewater treatment to remove heavy metal contaminants, radionuclides, dyes, etc. which is also an emerging area of research. Additionally, several hybrid materials including a particular variety of super-absorbents based on CMC has been reported and there is still a great opportunities that more will be created in the future. Based on these significant updates, this review covers advancement in CMC applications with major focus on food industry and wastewater treatment.

Keywords: CMC, Application of CMC, CBHs, Food Industries, Wastewater Treatment

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Introduction

CMC is an anhydro-glucose linear polysaccharide. Through -1,4-glycosidic bonds, the repeating units are joined. At the molecular level, the only significant distinction between CMC and cellulose is the presence of a few anionic carboxymethyl groups (i.e., -CH₂COOH) in the CMC structure, which take the place of some hydroxyl groups that were present in the original cellulose structure. 1918 saw the invention of CMC in Germany and introduced as a substitute for gelatin [1]. However, the discovery of commercial polymeric materials was promoted on a large scale in Germany in the early 1920s [2].

Carboxymethyl cellulose was produced primarily by extracting cellulose from wood and other plant-based progenitors. These plants contained a lot of cellulose fibers that were essential for the production process. Therefore, the final factor influencing CMC production was the availability of these specific wood-based plants. As such, CMC production was largely limited by the availability of these plants and their connection to the process [3, 4].

However, daily cellulose-containing products have been proposed in the literature by numerous researchers as effective substitute alternatives in this situation.

The several precursors based on plant (e.g., banana pseudo-stem [5], sago palm [6], corn stalks [8], corn husk [7], corn cobs [9], cacao pod husks [10], durian rind [11], maize stalks [12], the pulp of Eucalyptus globules [13], bagasse of sugarcane [14], the stalk of asparagus officinalis [15], pineapple peel [16], orange peel [17] etc.), additionally to some waste products (like as cotton gin wastes [18], wastepaper [19], waste paper sludge [20], textile waste [21], knitted rags [22] and cotton linter waste from the textile industry [23], etc.) for use in industrial-CMC manufacture, have attracted the interest of researchers.

There are numerous applications for CMC and CMC-composites in the fields of biotechnology, pharmaceuticals, building materials, textiles, polymers, food, energy and cosmetics, because of the abundance of raw materials, straightforward, inexpensive method of procedure, the distinctive characteristics of surface [24], various formability [26], mechanical strength [25], tuneable hydrophilicity [27], viscosity [28], and rheological properties [29], as well as hundreds of other. For instance, CMC and its composites are frequently used in biomedical fields for a variety of purposes,

including tissue engineering [30], bone-tissue engineering [31], wound dressing [32], Creating artificial organs or extracellular polymeric matrix mimics [33], manufacturing 3D-scaffolds for biocompatible implants [34] and producing absorbent nonwovens are a few examples. Because of their exceptional biocompatibility, binding ability to pharmaceutically active substances such as enzymes and medicines, pH sensitivity and high stability, CMC hybrid materials, films and CMC-based hydrogels have attracted significant interest in the past few years for potential use in pharmaceuticals, particularly for drug delivery [35], drug emulsification [36], and drug stabilization [37].

Additionally, CMC hybrid materials are employed as sizing and finishing agents in textile weaving [38], used for digital printing on textiles because of their qualities that thicken and sharpen colors [39], and as antioxidant, antiradical, absorbent textiles or antimicrobial because of their thermo sensitivity, pH, hygroscopic, and hygienic features [40-42].

Due to their lack of flavor, caloric content, and physiological inertness, CMCs are utilized as a variety of auxiliary agents in food items and their packaging, including thickeners, emulsion stabilizers, adhesive stabilizers, and moisture binders. In addition, many researchers have so far shown how important CMC is in the wastewater treatment and production of ecologically friendly energy and its storage.

CMC and its different composites have garnered a lot of attention recently as an affordable binding agent in biomass pellets (to cut down on excessive fuel loss), supplementary electrode material in batteries, and supercapacitor aerogels for effective energy storage [43, 44].

Over the past years, CMC-hybrid compounds, particularly their hydrogels, have shown a few encouraging outcomes in the removing dye contaminants [45], numerous inorganic metal ions [46], and even certain radionuclides [47] from contaminated waters.

CMC has also been suggested as a potential application in a number of other sectors, like in the paper industries. CMC helps in strengthening properties, color stability, ink retention or good printability, fire retardancy features [48, 49] improve adhesion properties [50], and also improve binding ability and cyclic performance of Si anode electrodes [51].

CMC has also been used in the oil business, the cosmetics industry, the dental industry, liquid detergent, the fertilizer industry and the building industry [52]. In order to visualize this, we discuss and compile the published CMC research here based on its applicability in a variety of food industries and water treatment methods.

Application of Carboxymethyl Cellulose (CMC)

Application in Food Industries

Food-grade CMC (cellulose gum) is widely used because of its ability to thicken water, act as a moisture binder, dissolve rapidly in both hot and cold aqueous systems, and texturize a wide range of food products (**Figure 1**) and because it is tasteless, odorless, and forms clear solutions without cloudiness or opacity.

Because it is physiologically inert and non-caloric, cellulose gum is particularly useful in dietetic foods. Cellulose gum serves as an extrusion aid, acts as a binder, helps to stabilize emulsions, and retards sugar crystal growth.

In frozen products, it controls ice crystal growth and phase separation. It is compatible with a wide range of food ingredients, including proteins, sugars, and more hydrocolloids.

The extensive acceptance of cellulose gum in so many culinary applications can be attributed to the variety of varieties that allow it to be customized to the various needs of the food industry.

The provision of food for human society is a crucial function of the food industry. The food industry uses a variety of auxiliary agents, such as hydrocolloids such as xanthan gum powder, sugar beet pectin, and soluble soybean polysaccharide (SSPS) [53, 54], as well as different alginates, gums, agar, a little amount of galactomannans and pectin, modified cellulose or starches CMC, and other polysaccharides to make high-quality foods.

Due to some of its exceptional qualities, including being tasteless, odorless, non-caloric, medically inert, creating a transparent solution without opacity, impeding the ability of suspended particles to be gravitationally separated, etc. These

CMC characteristics aid in enhancing the quality of food and the required pleasant texture to guarantee safety of food. CMC is frequently used in the food industry as emulsion stabilizers, thickeners, moisture binders, additive stabilizers, texture-improving and suspending agents, and other auxiliary agents. The structure, rheological properties, flavor, and the way products look as well as their pseudo plastic characteristics are also fine-tuned using CMC.

In order to assure the food products' long-term safety, it is also employed as a packaging or coating material. On the usage of CMC in food products, numerous researches have been published in the literature. By regulating particle size, concentration, and texture, CMC, for instance, is employed like a thickening in olive oil-based nano-emulsions to enhance the stability and physical properties [55].

On the basis of different viscosity of CMC, it serves as moisture binder (low viscosity) and gelation agent (high viscosity) in food industries. . Additionally, it serves as a thickening in fruit syrup, salad dressing, and semisolid dairy products [56]. In order to maintain food safety, CMC is now employed as a hydrogel (dewatering agent) in fruit syrup or juice. Heat is necessary for the traditional thickening process, but it is not necessary for the super absorbing or dewatering processes when heat is present [57].

As an emulsifier, CMC is used in wine, spreads, acidic beverages, dairy products like ice cream and milk, sauces, and bread goods. To make ice cream of the highest quality, 0.5% CMC is added as a stabilizer. By preventing the over-crystallization of lactose in cream, medium viscosity CMC improves the creamy mouthfeel [58].

In the recent past, consuming CMCs in food industries has grown from a normal level to an advanced one because to their sanitary, biocompatible, and potential for preventing or managing human disease through diet. For instance, Dafe *et al.* [59] created a vehicle for CMC/k-carrageenan mixed foods to deliver probiotic-based food in the colon in order to avoid gastrointestinal illness.

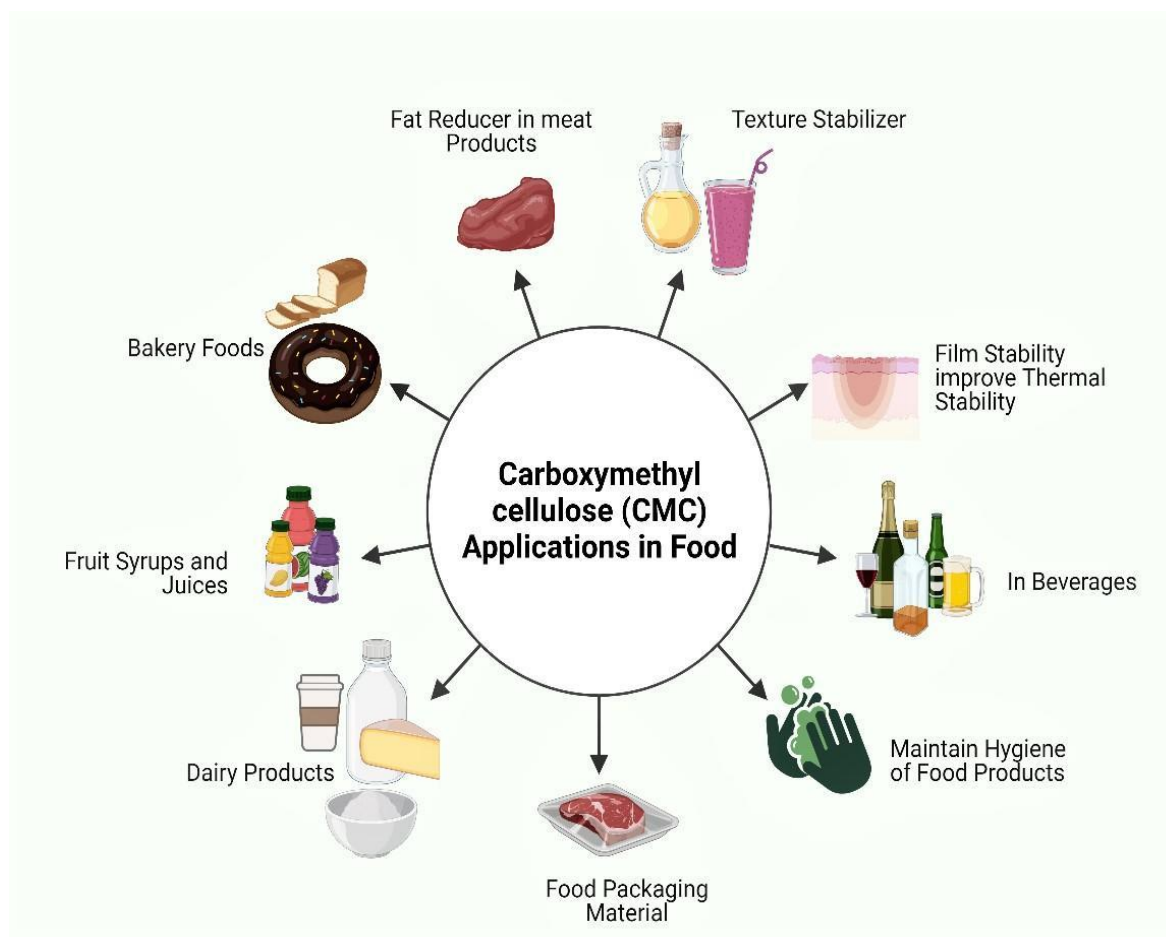


Figure 1: Different applications of CMC in Food industries

Drinks containing cocoa and acid are typically stabilized with CMC, which lessens layer formation and sedimentation at both high and low temperatures. Therefore, owing to the stabilization of minerals in fruit-based beverages or nutritional fiber, in the recent time combined form of CMC and gum tragacanth has been utilized as a stabilizer in drinks [60]. The immune system was strengthened and the gastrointestinal tract or mucosa's health was controlled by food, as advised. A similar coating material was suggested by Ngamekaue *et al.* [61] in 2019 for transporting a herbal-infused microcapsule oil (a combination of oil of holy basil with gelatin) to the intestine. The microcapsule is protected from acid, moisture, or oxidative effects by a CMC/beeswax covering composite while also assisting in the management of the herbal product once it reaches the gut.

The body responds favorably to the suggested herbal items as effective antioxidants, anticancer agents, etc., hence reducing the risk of non-communicable diseases. As a result, bio-based CMC coatings made of grape seed extract and essential oils have also been used on fish [62]. Making nutritious food items is significantly hampered by the high fat content of the food

(meat). CMC compounds are utilized as fat substitutes or reducers to address this issue. Gibis and coworkers [63] utilized MCC (microcrystalline cellulose) and CMC as fat substitutes for manufacturing fried patties of beef, CMC with 0.5% concentration being selected as a superior source of flavor, texture, and juiciness. Han *et al.* [64] on the other hand, created healthier beef products with a focus on high nutrition and low fat. To lessen the risk of colon cancer, cardiovascular disease, and other conditions, CMC and other food fibers (such as chitosan cellulose, pectin, inulin, and so on) are used in the processing of meat as texture modifiers, nutrition enhancers and fat reducers.

In the last ten years, CMC has been used as a food packaging material, according to many researchers. Khezrian *et al.* [65] created a packaging material based on nanocomposite with an essential oil doped chitosan/ montmorillonite/ CMC for increasing the shelf life of camel meat. Furthermore, biodegradable and environmentally friendly PVP-CMC hydrogel film is frequently employed as a principal packaging material for food commodities [66]. A biodegradable and antibacterial packaging material made of the

combination of a metal cation-based film and CMC/PVA/zeolite has been disclosed. This antibacterial property keeps food from rotting and extends product shelf life. In packaging materials, films made from CMC-CHPS (chickpea husk polysaccharides) have antioxidant and antibacterial properties. Combination of CMC, montmorillonite and dopamine has recently resulted in the development of high stiffness and high thermal stability-based packaging materials [67].

Application in the Water Treatment Process

Water is the world's most abundant natural resource; however, only about 3% of existing water reserves are freshwater, and less than one-third of this freshwater is usable for various domestic, agricultural, and industrial uses [68]. While water consumption is increasing dramatically, the availability of freshwater is being depleted due to an increase in pollution, resulting in water shortages for modern society [69]. Rapid industrialization, uncontrolled urbanization, and a variety of human activities, combined with inefficient waste management, all contribute to this growth in wastewater. Water contamination has recently emerged as one of the world's most pressing challenges. Every day, a massive amount of contaminants enter aquatic habitats from various industries and household activities, causing a variety of illnesses in various living things and humans. The amount of contaminants and toxins that can be found in wastewater from several businesses or household activities has no set upper limit. The two most frequent contaminants found in industrial effluent are dyes and heavy metals, both of which are detrimental to ecosystem sustainability [70, 71]. Even at low concentrations, the presence of dyes restricts penetration of sunlight into water, resulting in a prominent loss of dissolved oxygen, posing severe health dangers to aquatic living entities. In many situations, dyes cause anaerobic digestion, resulting in the formation of several carcinogenic chemicals that can penetrate in the food chain by aquatic creatures.

On the other hand, the dye concentration disposed of into various water bodies remains large. For example, about 7×10^5 tonnes of various reactive dyes are generated each year, with approximately 5-10% of these dyes ending up in the environment. Effluents from industry, heavy metals, on the other hand, pose major health risks due to their high carcinogenic and poisonous potential. Chronic arsenic (As) exposure, for example, can result in severe disorders like bladder, kidney, liver cancer

or prostate. Chromium (Cr) is another very hazardous metal that has a negative impact on both human bodies and aquatic creatures [72]. The dangers of heavy metals and dyes to the health of humans and the normal environment underline the importance of industrial wastewater treatment. Distillation, adsorption, coagulation, membrane filtration, redox, electrochemical treatment, are some common physical and/or chemical processes. The most popular and efficient technique for treating contaminated water is adsorption. Although activated carbon is an extremely effective adsorbent, its widespread use for wastewater treatment is constrained by its high cost and labor-intensive regeneration process. Although numerous adsorbents have been found in recent studies to remove heavy metals and dyes from polluted industrial water, treatment of bulk wastewater by using these adsorbents is still difficult [73, 74]. Due to their great removal effectiveness, hydrogels, which are three-dimensional polymer networks, have recently gained a lot of attention in the fight to remove contaminants from waste streams [75]. The high porosity and abundance of hydrophilic functional groups in hydrogels (such as -OH, -COOH, -NH₂, -SO₃H, -CONH₂, etc.) allow for the retention and adsorption of a significant amount of water during the treatment process, leading to the eventual removal and recovery of all aqueous dyes and heavy metals [76]. The majority of the hydrogels that are currently in use, however, are made from materials that are neither renewable nor biodegradable, such as petrochemicals. The most prevalent polymer in nature, cellulose, is the source of carboxymethyl cellulose-based hydrogels (CBHs), which are extremely absorbent, long-lasting, non-toxic, biocompatible and biodegradable [77].

Analysts have recently explored employing various CBHs to virtually completely remove heavy metals and colors from polluted water. In this series three-dimensional polymer networks of carboxylated hydrogels have been employed by Zhou *et al.* [78] to remove Pb⁺² with 90% removal efficiency within four hours. Using various kinds of CBHs, such high adsorption values were also attained for Cu²⁺ (182-230 mg/g), Ni²⁺ (200 mg/g), and Hg²⁺ (140 mg/g) [79]. Recently, Deng *et al.* [80] used chitosan and cellulose to remove nearly 100% of Congo Red dyes with 166.1 mg/g saturation adsorption. There are still many potential applications for CBHs since the modification of HG functional groups improves their adsorption efficiency (**Figure 2**).

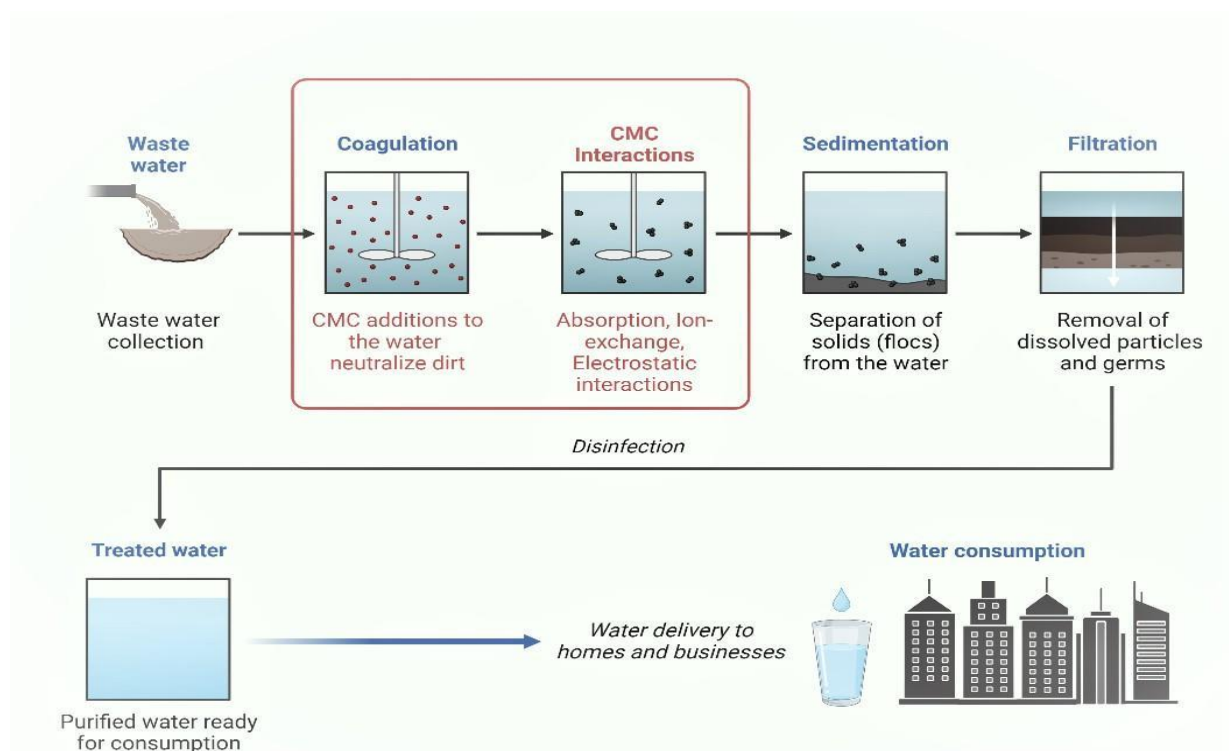


Figure 2: Application of CMC in wastewater treatment based on different interactions.

The impetus behind the current work is the paucity of reviews in this particular field, despite the fact that many research studies employed CBHs to treat contaminated water sources. The different interactions that result in the typical adsorption by CBHs are strongly influenced by the adsorbent characteristics, functional groups present in the HG, the chemical composition of pollutants, and experimental parameters (like as solution pH, the coexistence of metal ions, initial pollutant concentration, temperature, etc.) [81].

Electrostatic interactions are the primary adsorption process used by CBHs to remove heavy metals and dyes, while various adsorption processes have also been found to combine electrostatic interactions with other interactions [82,83].

CMC-based composites will naturally show a larger adsorption tendency towards metal ions and other cationic pollutants than anionic contaminants because they have electronegative carboxymethyl groups (COOH) on their surface. Adsorbents struggle to effectively bind negatively charged impurities for a sustained period of time due to electrostatic repulsion between anionic contaminants and opposing surface charges. Probably, this is the only cause of scarcity of research on anionic pollutant removal by CMC-hybrid materials to treat the contaminated water in comparison to cationic pollutants in the literature.

As a result, we recommend concentrating on the development of hybrid materials that are CMC-based, more efficient, and less expensive, in particular nano-hybrid super-absorbent materials. To remove different cationic contaminants from wastewater, including cationic dyes, metal ions, various cationic and radioactive elements, more study will be needed in the future. Additionally, CMC-based adsorbents' surfaces may be improved in the future through hybridization with various kinds of organic-inorganic positively charged species, making them more effective for the simultaneous treatment of cationic and anionic pollutants.

Conclusion

Although the sources of CMCs were restricted at the start of its development, in the current scenario, CMC is preferred over other derivatives because of its versatility and quantity of precursor ingredients. Numerous of their substitutes have been proven scientifically over the course of the last two decades. Maize husks was the most promising discovered material among them, which offered the highest product yield with appropriate purity. Essentially, based on their diverse uses and applications, CMCs are now used in biomedical engineering, textile, pharmaceutical, and food industries. One of the most prevalent contaminants in wastewater is dyes and heavy metals, which are discharged by a variety of businesses and have a negative impact on the ecosystem, human health, and aquatic life. The removal of contaminants from wastewater is therefore essential for a safer

environment. Cellulose-based hydrogels (CBHs) are excellent choices since they satisfy the criterion while also offering further advantages such high removal effectiveness, cost-effectiveness, and simplicity. The synthesis of CBHs, the adsorption process, and parameters to maximize adsorption capacities are some of the significant components of wastewater treatment utilizing CBHs that have hardly been discussed in the literature. The effective use of CMCs in the water treatment process to remove additional contaminants is now a growing area of research. Despite the fact that several hybrid materials, particularly a variety of super-absorbents based on CMC, have been reported in this context, there is still a great possibility that many more can be created in the future.

Statements

Author's Contribution: Concept, design, and overall supervision: SY; Acquisition and Analysis or interpretation of information and data: SY and SK; Drafting of the manuscript: SY, SK and PR; Editing of the manuscript: SY, SK and PR; All authors provided comments on the manuscript and evaluated critically. All authors gave their consent for publication.

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