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A review of polyvinyl alcohol/carboxymethyl cellulose (PVA/CMC) composites for various applications

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A B S T R A C T

Polyvinyl alcohol/carboxymethyl cellulose (PVA/CMC) composites have attracted considerable attention due to the synergic relation between the two polymers and developing novel blends with improved properties. On one hand, PVA is a versatile polymer with higher mechanical properties compared to CMC. On the other hand, CMC has high biodegradability and biocompatibility, while suffering from poor mechanical properties. Therefore, the blending of the two polymers can help to benefit from the individual component properties. This paper has reviewed the properties and potential applications (e.g. drug delivery, food packaging, and agriculture) of the PVA/CMC composites.

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1. Introduction

Blending of polymers can often produce materials with enhanced characteristics more rapidly compared to developing new polymer chemistry [1, 2]. Extensive studies have been conducted on copolymers due to their high application potential [3-8] and their importance in basic science [9]. Developing new polymer blends with improved properties by mixing two or more polymers is usually a less time-consuming process than new polymer development with desired properties. Therefore, their technological applications are broadening faster than their constituents[10] [11-15].

Polyvinyl alcohol (PVA) is the first synthetic colloid that was first developed by Haehnel and Herrmann in 1924 [16-20]. It was supported by a paper in Hangzhou City, China [21]. Because of its strong nonirritant [22, 23], harmlessness, and hydrophilicity [24, 25], PVA had been selected for gel preparation among other water-soluble polymers [24, 26-29]. The PVA gel offers some advantages including easy machinabil-

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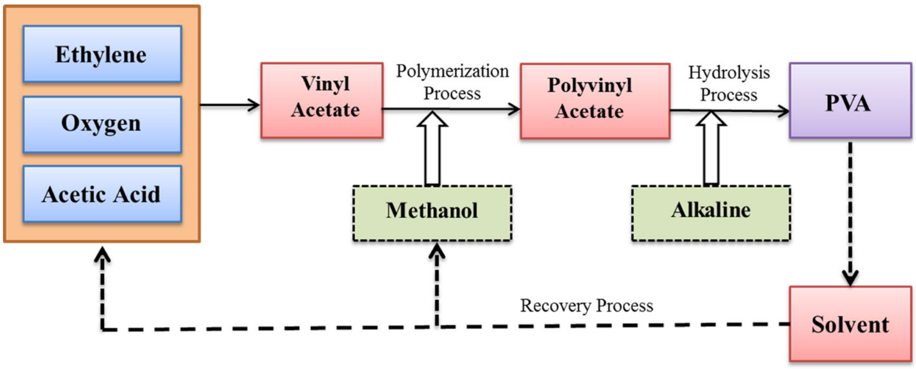

Fig. 1. Production outline of PVA.

ity [30], low toxicity [24, 31-33], good strength [34, 35], and high water content [36-38]. Currently, the PVA- based materials are extensively utilized in the medical, industrial, and agricultural industries [39].

Basically, manufacturing of PVA consists of the of vinyl acetate monomer polymerization into polyvinyl acetate (PVAc), and the subsequent acetate groups hydrolysis to produce PVA [1,2]. Based on the used catalyst, three hydrolysis methods are applicable in the PVA preparation. They include acidolysis, aminolysis, and alkaline hydrolysis [1]. On industrial scales, the PVAc to PVA conversion is usually carried on by alkaline alcohol. In this hydrolysis method ester interchanged with methanol, in the presence of sodium hydroxide, to hydrolyze the acetate groups [16]. The production outline of PVA is illustrated in Fig. 1.

PVA is a hydrophilic synthetic polymer with semi-crystalline, planar zigzag structure, and sound mechanical properties [40-43]. PVA is chemically and thermally stable [44] and is resistant to degradation under most physiological environments [41]. Moreover, it is water-soluble due to its elevated polarity, non-toxicity [45], and high biocompatibility [46, 47] and it can be processed easily [48]. PVA has also promising potential for producing biodegradable films [49].

One of the important derivatives of cellulose is CMC, in which carboxylic groups ($-CH_2-COOH$) are bound to some OH groups of natural cellulose [50, 51]. According to the literature, the introduction of CMC into the PVA matrix results in the improvement of this polymer properties [52]. Therefore, this paper aims to review the properties of PVA/CMC composites as well as their applications in the field of drug delivery, food packaging, and agriculture.

2. Carboxymethyl cellulose (CMC)

The most important classification of polysaccharides is carboxymethyl cellulose. CMC is a semi-crystalline, water-soluble, non-toxic [53, 54], low-cost [55-57], and biodegradable material [58-60] with excellent film-forming ability; however, it suffers from low conductivity [61, 62] and the lack of strength [63]. Extensive studies have been conducted on the CMC application in single polymer electrolyte systems, however, some problems limit its application in this field due to small elongation at break, exceptionally stiff behavior, (less than 8%), and losing the electrochemical stability required for electrochemical devices. Additionally, outstanding chemical, mechanical, and physical properties cannot be offered by a single polymer for wide range applications, particularly in energy storage devices [7]. CMC exhibits several desirable characteristics including emulsification, thermal filming, gelation, and inspissation [9].

Due to its biodegradability and biocompatibility, CMC can be employed for biotechnological and pharmaceutical applications [3]. For the production of CMC, chloroacetic acid is reacted with OH groups on hydroglucose units (AGU) of cellulose. In the cosmetics and food industries, CMC has been utilized as a water-retention agent and stabilizer [9].

CMC, as a highly hydrophilic derivate of cellulose, is extensively

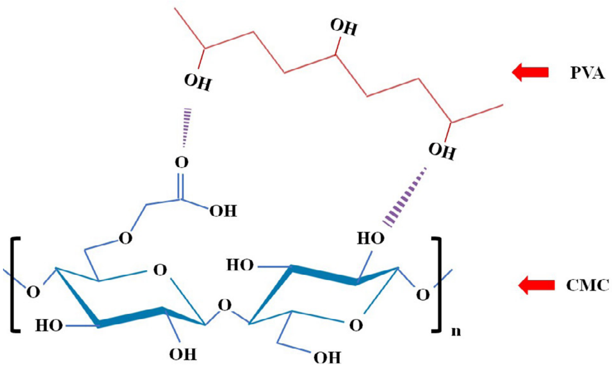

Fig. 2. Molecular interaction of PVA and CMC.

used as a suspending and thickening agent in the pharmaceutical and food industry. Because of good modifiability, non-toxicity, and swellability, CMC has attracted the attention of scientists in the field of hydrogels for drug delivery applications. Numerous investigations have reported the development of hydrogels based on CMC as carriers for water-soluble drugs [8]. CMC-based hydrogels have the potential to be used in absorbents, drug delivery, wound healing, and enzyme immobilization owing to their biodegradation, biocompatibility, and solubility [64].

3. Synthesis of PVA/CMC composites

Grafting, crosslinking, and degradation are ways to improve or modify polymer materials [9]. PVA can be modified by CMC due to their compatibility and such characteristic is the result of the hydrogen bonding between the OH groups in PVA and the carboxymethyl groups on CMC [9]. The chemical structure of PVA and CMC and their interaction is illustrated in Fig. 2. Fig. 3 also presents the crosslinking of PVA and CMC by cellulose acetate (CA).

Zhang et al. [4] prepared a potential coating material based on CMC/PLA blend film. The biodegradable CMC/PVA blend film was successfully prepared by intra- and inter-molecular cross-linking reaction. According to the obtained favorable biodegradability, the prepared blend film showed environmentally friendly characteristic which can be offered as potential materials for control-release fertilizer's coatings.

Sayed et al. [15] incorporated copper oxide and PVA nanoparticles in CMC films using a solution casting method and investigated their electrical, optical, and structural properties. At low temperature, Schottky emission was the conduction mechanism in the PVA/CMC blend. After the addition of PVA, the transparency of the CMC film increased from

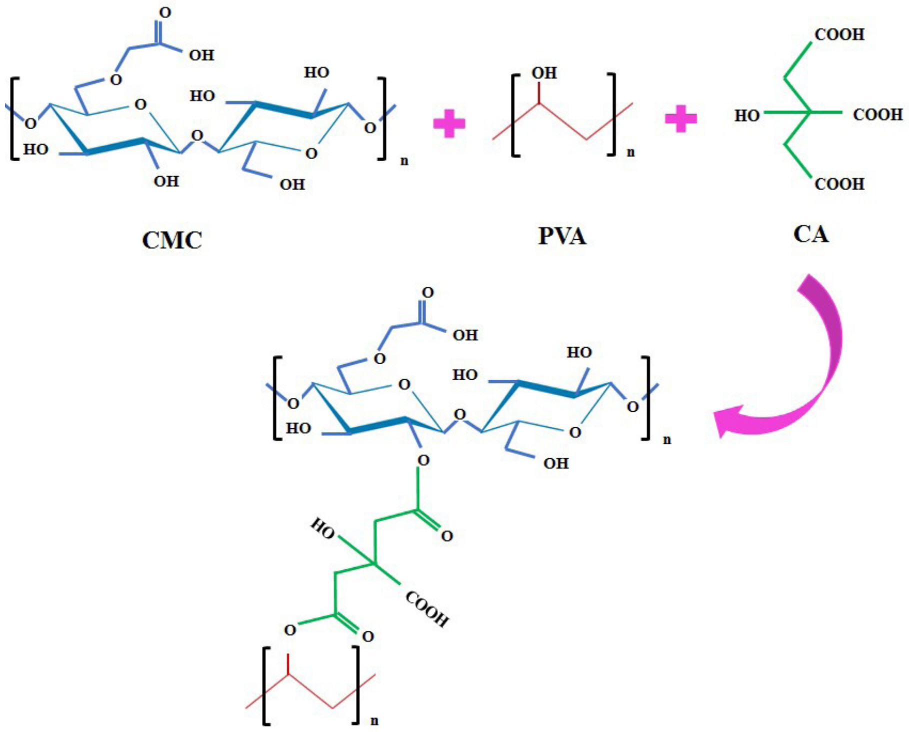

Fig. 3. Crosslinking of PVA and CMC by CA .

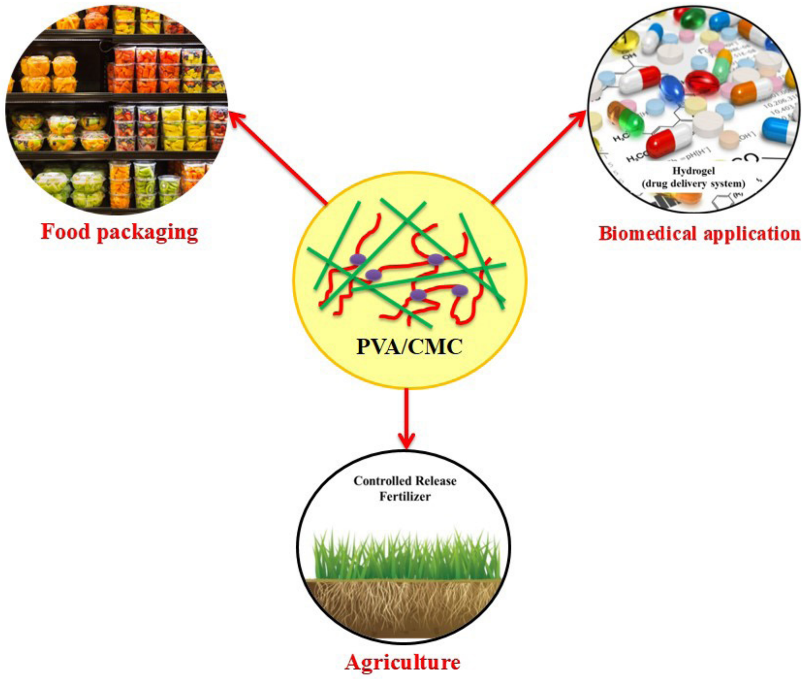

Fig. 4. Applications of CMC/PVA composites.

87 to 89 %. The results showed controlled optical constants, refractive index, and insulating properties of CMC by blending with PVA. Additionally, a non-ohmic behavior was observed in the current-voltage characteristics of the blend.

Saadieh et al. [65] developed biopolymer blend electrolytes based on PVA/CMC using the casting solution method. The highest ionic conductivity (9.12×10^{-6} S/cm) was reported for the blend electrolytes with the CMC to PVA ratio of 80:20 at room temperature. According to the results, the reduction of dielectric loss and dielectric constant were observed with an increase in frequency. This was reported to be the result of the ions accumulation contributed by the electrode, ionic polarization, and molecular polarization.

Sayed et al. [9] synthesized the CMC/PVA blend using the solution casting method and subsequent ɣ-rays irradiation. They proposed that ɣ-rays irradiation had the capability for controlling the physical properties of this copolymer. Therefore, they suggested the CMC/PVA blend for several electronic and industrial devices due to improved charge storage capacity and dielectric strength [55].

4. Properties of PVA/CMC composites

The physical and chemical properties of polymers are influenced by the chemical structure of polymers. The flow and morphology of polymers depend on their chemical structures, which leads to different physical properties [66]. Taghizade et al. [67] developed PVA/starch (S)/CMC) composite films using the casting method in the presence of glycerol-containing plasticizer. According to the results, PVA/S/CMC blends showed higher thermal stability compared to PVA/S. This was reported to be due to the addition of CMC resulting in the improvement of the thermal stability of the PVA/S blend.

Miao et al. [68] prepared a negatively charged nanofiltration membrane based on PVA/CMC-Na composite produced by interfacial polymerization. The resultant composite membrane exhibited high stability in long-term use. According to this study, the cross-linked PVA-based composite could provide a good balance between the salt rejection and the permeate flux under lower operating pressure. Tajeddin et al. [69] also reported the water absorption of 22.03 percent for PVA/CMC blend film prepared by the casting method after 24 h. Zhu et al. [70] stated that the pure PVA film had low water sorption and it enhanced by adding CMC to the composite film. This was reported to be due to the blending of two different molecule types resulting in the structure deformation and making more hydrophilic networks. Furthermore, the solubility of the composite was around 70 % after 24 h.

Polymer blend films are widely used for metal ions adsorption [71]. Wang et al. [72] prepared PVA/CMC hydrogels using the freeze-thaw process for heavy metal ions adsorption. The formation of crystallites occurred due to the separation of phases in polymer solutions during freezing stages leading to insoluble hydrogels (71% of insoluble gels). The swelling ratio for pure PVA hydrogels was obtained to be 416%, while a higher swelling ratio of 1437% was obtained for the PVA/CMC hydrogel containing one-third of CMC and two-thirds of PVA. This hydrogel had the adsorption capacity of 8.4 mg per gram of hydrogel toward Ag^{+}. The prepared hydrogels exhibited potential applications for wastewater treatment and removing heavy metal ions.

Many investigations have been carried out on the electrical and optical properties of polymers for potential applications in optical and electronic devices. The aim of electric properties investigation is to understand the charge transport mechanisms in those materials, which can be certainly changed by different techniques including plasticization, mixing, copolymerization, and the addition of ceramic filers or salts [73].

Abutalib et al. [73] studied dielectric and electrical characteristics of PVA/CMC/ZnO nanorods prepared by the casting method. The results showed that dc conductivity revealed Arrhenius behavior that was enhanced with temperature, whereas the ac conductivity followed Jonscher's law. These nanocomposites have been offered as potential candidates for applications such as electrochemical devices due to their significant improvement in ac and dc conductivities.

In a research conducted by Goswami et al. [74], bionanocomposite films of CMC/PVA/V_2O_5 prepared by solution casting of the CMC/PVA thin film, and subsequently impregnation of V_2O_5 into the film. The differential scanning calorimetry (DSC) results showed lower transition temperature for CMC/PVA/V_2O_5 nanocomposite compared to the CMC/PVA film making it suitable for smart windows. The total conductivity results demonstrated good electrical behavior of the prepared bionanocomposite. Therefore, electrical and optical properties of CMC films were reported to be significantly enhanced in the form of CMC/PVA-V_2O_5 nanocomposites enabling them to be used in applications such as storage devices and smart windows.

According to Zhu et al. [70], pure PVA exhibited low dielectric constant, and the increase in the dielectric constant was observed for PVA/CMC composite film.

The addition of PVA can enhance the mechanical properties of CMC [48]. According to Tajeddin et al. [69], the mechanical properties of PVA/CMC composite film (obtained by 10%v of PVA and CMC dissolved in 60 mL of distilled water) including tensile strength, elongation at break, and modulus were reported to be 11.54 MPa, 24.55 %, and 36.2 MPa, respectively.

El-Newehy et al. [48] used the electrospinning technique to prepare CMC/PVA nanofibers for the controlled release of diclofenac sodium (DS). The study showed that the CMC/PVA nanofibers blend exhibited good mechanical properties. Additionally, the in vitro release study showed that the presence of CMC led to the sustained controlled of the DS release from the nanofibers mats. The CMC/PVA nanofibers system could be offered as a promising material for drug delivery applications due to the low cost and biocompatibility of the blend. General applications of PVA/CMC composites are shown in Fig. 4.

5. Applications of PVA/CMC composites

5.1. Application in food packaging

An innovative food packaging is active packaging in which the advances in material sciences, packaging, food safety, and food technology are combined for satisfying consumer demand for safe and fresh-like products [49]. Because of environmental considerations over the past few years, using synthetic polymers is restricted. There has been ever-increasing attention in biopolymers including proteins, lipids, and polysaccharides [75]. Active agents can be incorporated into the polymer structure in active packaging leading to a low diffusion rate of active compounds and the maintenance of active agents with high concentrations in the surface of packaging materials during the shelf life [76]. In the cosmetics and food industries, CMC has been utilized as a water-retention agent and stabilizer. PVA has also been incorporated to improve the mechanical properties [77].

Muppalla et al. [49] fabricated PVA/CMC films with clove oil by the casting method as active packaging for ground chicken meat. The results demonstrated an increase in the tensile strength and puncture force of the CMC film and the reduction of water vapor transmission rate with the increase in the PVA concentration. A negligible oxygen transmission rate was observed in all the samples. During refrigerated storage, control samples spoiled in 4 days, while packing of meat samples in the prepared films resulted in lower total viable counts and shelf life of 12 days. The efficacy of the CMC–PVA–clove oil films was also studied against Bacillus cereus and Staphylococcus aureus in ground chicken meat. It was proposed that the prepared films have great potential for the active packaging of meat products.

Fasihi et al. [76] produced biodegradable active films based on CMC/PVA/oleic acid (OL) containing rosemary essential oil (REO) via Pickering emulsions. The results demonstrated that the films containing REO showed considerable antimicrobial and antioxidant properties. In the films with 3% REO, the fungal inhibition against Penicillium digitatum was 100%. In bread slices that were packed with the active films containing 3% REO, no fungal growth was observed at 25 ºC after 60 days of storage. This might be due to the regular, slow release of REO resulting from Pickering emulsions.

Villarruel et al. [78] developed blend films based on CMC and PVA and modified it with UV radiation in the presence of sodium benzoate (SB). According to the result, the newly developed materials showed different chemical and thermal stability compared to single components. Both blend films and UV-induced ones revealed the very low value of oxygen barrier properties making them suitable materials for packaging applications with selective oxygen permeability. Moreover, the UV treatment in the films containing SB inhibited the growth of a wide spectrum of microorganisms and increased their insolubility in water making them potential materials to be used as food packaging emulsion.

5.2. Application in biomedical

There has been a considerable effort in the improvement of drug delivery efficacy by the investigation of new materials [79-82]. In this regard, natural polymers play a substantial role in biomedical applications [83, 84]. Recently, drug delivery systems based on natural polymers hydrogel have attracted the attention of researchers [85]. CMC is a cellulose derivate with high hydrophilicity and is widely applied as a suspending and thickening agent in the pharmaceutical industry [86]. The desired properties of CMC hydrogels such as modifiability, non-toxicity,

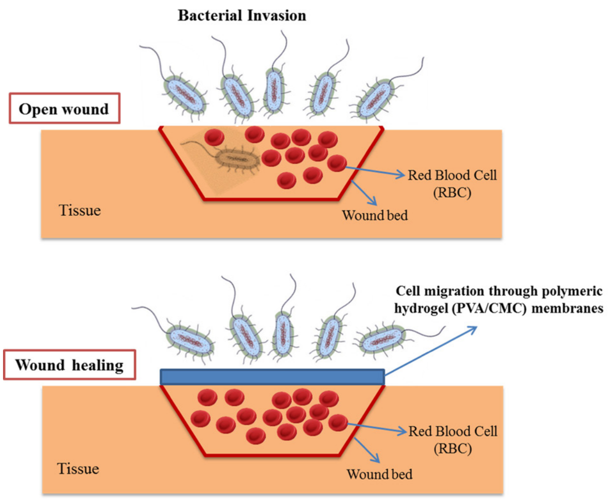

Fig. 5. Role of hydrogel membranes in accelerating and enhancing the wound healing phases.

and good swellability encourage scientists to use this material for drug delivery [87].

One of the pressing public health issues is burns treatment. Despite efforts made for its prevention, such as public campaigns, its worldwide incidence is high. Dressings are usually required for the treatment of second-degree burns. Cell hydration through keeping a moisturized environment and promotion of necrotic tissue debridement are of important properties of an ideal dressing. It should also be able to successfully remodel tissue and have transparency to monitor healing. Other characteristics include infection inhibition, pain reduction, non-toxicity, and vapor and oxygen transmittance. Most of these properties are offered by hydrogels; therefore, they have been utilized as biomedical devices for wound treatment. They can be permeable to oxygen, while act as barriers to microorganisms [88]. Hence, hydrogels are considered as patient-friendly systems for drug delivery [77, 89]. Many wound dressing materials belongs to hydrogel dressings because their advantages outweigh their disadvantages. Fig. 5 schematically shows the role of hydrogel membranes in wound healing phases.

Hydrogels are hydrophilic polymers that are cross-linked and undergo swelling in aqueous media while their structural integrity is maintained [88]. However, hydrophobic drugs can be loaded in low amounts due to the hydrophilic network of hydrogels. To overcome this restriction, polymer chemistry investigations have focused on the development of a variety of network structures. As a result of biodegradability and biocompatibility, CMC can be utilized in biotechnological and pharmaceutical applications [77]. Another option for the production of biocompatible hydrogels is PVA, which has a semi-crystalline structure. PVA hydrogels are able to deliver moisture and absorb exudate from the wound site. PVA gels can be cross-linked physically or chemically (with chemical agents via radiation techniques) [88]. Fig. 6 shows the synthesis of PVA/CMC hydrogel.

Ghorpade et al. [86] developed polymeric blend films for the extended release of water-soluble drugs based on citric acid cross-linked CMC/PVA. The model drug of Gentamicin sulfate (GTM) was used. The incorporation of PVA enhanced the mechanical strength of the CMC/PVA hydrogel films. The swellability of the hydrogels was improved by an increase in the PVA content in the hydrogel films. Thus, the developed hydrogel films were considered as promising biomaterials for the delivery of basic drugs soluble in water.

Membranes of PVA/Polyethylene Oxide/CMC were developed by Agarwal et al. [87] by freeze-drying and solvent casting for the drug delivery application. The results demonstrated enough swelling in PBS in the membranes, which can recommend them for exudative wounds.

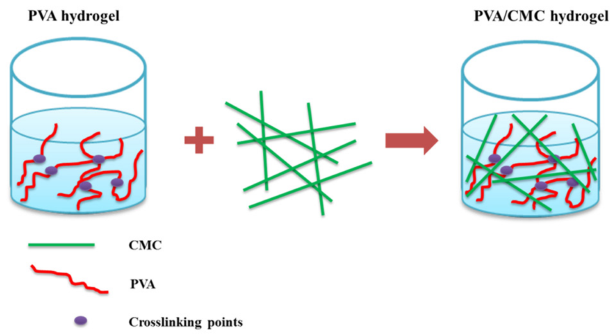

Fig. 6. Synthesis of PVA/CMC hydrogels.

Due to the continued drug release up to 10 h, the prepared dressing can inhibit the wound site from infection until complete wound healing. Additionally, the inhibition zone of membranes with the loaded drug revealed the effective release of the drug in the medium and inhibition of the growth of the microorganisms. Therefore, this dressing system could be employed for developing antibacterial wound dressing.

5.3. Application in agriculture

Blend films have the potential to be utilized as coating materials for controlling the fertilizer release. However, due to nonbiodegradability and high poison content, some blend films might produce serious soil pollution. Therefore, development of films with biodegradability, innocuity, and low price is critical for the production of coated fertilizers [39].

CMC/PVA blend films were produced by a cross-linking reaction between CMC and PVA for the potential application as coating materials. The results indicated that the prepared CMC/PVA films possessed a smooth surface. The reduction of water permeability and absorbency of the blend films was observed by the increment of the PVA content. As a result of the good biodegradability of the CMC/PVA blend films, they can be considered as potential coating materials for the control-release fertilizer [39].

For efficient fertilizer and water management, Ozen et al. [90] coated nonwoven fabrics with potassium nitrate/CMC. The fabrics were pre-coated with PVA and the cross-linking with CMC was carried out using citric acid. According to the results, the PVA pre-coating and the subsequent cross-linking of CMC led to the provision of a synergistic effect for a considerable enhancement of fertilizer release management and moisture. Obtaining a controlled release of fertilizer with an improvement in water absorption/retention behavior is an outstanding step forward in the efficient sustainable agriculture field.

6. Conclusions and future insights

In this study, the recent findings of the properties and applications of PVA/CMC composites and blends including drug delivery, food packaging, and agriculture were reviewed. These novel composites have great potential for the active packaging of food products. Moreover, they show enhanced water solubility leading to improved bioavailability and dissolution for wound healing. These properties make them the promising materials for hydrogel production to deliver drug and moisture to the wound site. In addition, these composites can be utilized in agriculture for the controlled release of fertilizer.

PVA/CMC composites are gaining great attention in biomedical applications due to offering various advantages in different fields. Their properties can be tailored by controlling their structures to enhance their properties for various applications. The PVA/CMC hydrogels as wound dressings act as barriers to microorganisms while retaining their permeability to oxygen. Thus, these attractive composites can be promising materials for drug delivery and wound dressing application and the incorporation of other reinforcement or components might be investigated for the improvement of the properties as well as new production methods. They can also be useful for producing masks to protect against coronavirus. Therefore, further well-designed studies will be required for the development and enhancement of these effective materials.

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