

# Carboxymethyl Tamarind Kernel Gum based Controlled Drug Delivery Excipients: A Review

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## ABSTRACT

The Carboxymethyl Tamarind Kernel Gum (CMTKG) is a natural based polysaccharide which has been derived from the Tamarind kernel gum (TKG) through the carboxymethylation process. The chemical alteration of TKG into CMTKG has resulted in amplifying swelling capacity, in situ gelations, wide pH tolerance, high drug holding efficiency, stability, release kinetics, and hydrophilicity. Out of many application-based areas, it has extensively been used in the field of drug delivery systems via developing various forms like nanoparticles, composites, films, hydrogels, and pellets. This article is planned to fill in as a helpful tool for research scholars, who are engaged in green polymers, and in giving almost every aspect of CMTKG in the sphere of drug delivery.

**Keywords:** Drug-delivery; CMTKG; Natural polymers; Bio-degradable.

## INTRODUCTION

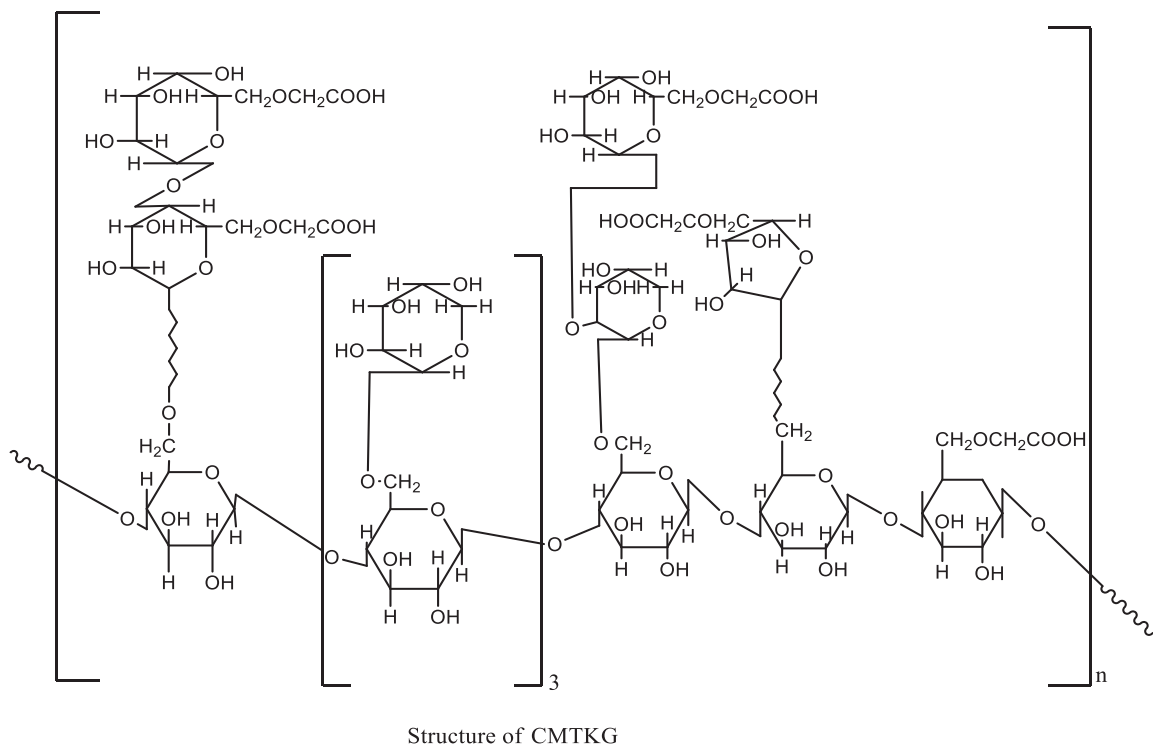
The by and large known realities of polymers with respect of its wide zone of uses and explicit characteristics over the worldwide market is upgrading progressively (Kumar et al., 2001). In the course of the most recent couple of many years, there has been an enormous interest for common polymers over manufactured polymers because of the disturbing consideration towards ecological security (Namazi, 2017). Lately, polymers got from plants have dazzled a great interest in most of the research communities. Subsequently, bio-polymers are being considered over synthetic polymers because of their ease of accessibility, non-harmfulness, the adaptability of chemical modification, and bio-degradability (Khushbu et al., 2019) when contrasted with synthetic polymers with time-consuming synthesis, climate related issues, and toxicity issues (Kulkarni et al., 2017). Natural polymers have expanded applications due to the two significant properties to be specific, biodegradability and biocompatibility (Racovita et al., 2009). Polysaccharides are the most plentifully accessible polymers which are significantly natural in nature and all the more notably, they show properties which make them stable, hydrophilic, sustainable, effectively changeable, and, efficient. Natural sources derived polysaccharides are attracting attention growing from the pharmaceuticals sector, as they also tend to show biological potency, for example, anti-cancer, immunostimulatory, anti-inflammatory, and anti-viral. Interestingly, in comparison to drug supplements having synthetic polymers, the drug supplements having natural polymers are generally safer, viable having almost no aftereffect, and even more quickly available and more affordable (Han, 2018). Normally utilized common polysaccharides are chitosan (Nangia et al., 2018), xanthan (Pandey et al., 2016), guar gum (Thombare et al., 2016; Thombare et al., 2017; Warkar S.G. et al., 2015), sodium alginate (Abd El-Rehim, 2006), which are preferred over synthetic polymers.

Drugs, in the form of drug-delivery systems, have for some time been utilized to improve wellbeing and broaden the life shell of human lives. The act of medication conveyance has changed drastically in the previous few decades and considerably more prominent changes are foreseen in the years to come. Researchers around the globe have contributed significantly to our comprehension of the physiological boundaries to effective drug-delivery, for example, transport in the circulatory system of humans and movement of drugs through cells and tissues; they have additionally added to the advancement of a few new methods of drug-delivery that have entered clinical practice. However, with the progress of this advancement, numerous drugs, even those found utilizing the most developed atomic science

techniques, have unsatisfactory results because of the drug's interaction with the body part which consequently limits our capacity to fabricate ideal drug-delivery systems for diseases. The main reason is the synthetic toxicity of the drug-delivery system that brings inside the body along with the drugs and that is why biopolymers plays a major role here as they don't impart any toxicity and hence, safest to use (Khalane et al., 2019; Tiwari et al., 2012).

Tamarind Kernel gum (TKG), least expensive and food-grade based gum, is extricated from Tamarind's seeds; a pervasive plant of Africa and Southeast Asia (Kumar et al., 2008). The seeds are utilized widely for gelling, paper business and food thickeners (Gupta et al., 2010). Though, terrible smell, quick degradability, dull tone, and low dissolvability in water, are a couple of few downsides that TKG has (Goyal et al., 2007). Henceforth, this emerges the need to adjust to modify cum enhance TKG's drug and physico-chemical attributes for improved properties and thusly more extensive CMTKG's utility notwithstanding those of TKG via its carboxymethylation (Pal et al., 2008b). The chemical modification of biopolymers has gotten huge consideration in the most recent decade and at the same time keeps up the characteristic polymers to their latent capacity and bestows enhanced characteristics without affecting fundamental engineering of the polysaccharide framework (Giusti et al., 1993). Carboxymethyl Tamarind Kernel gum (CMTKG) is unique good instance of a chemical alteration where TKG is converted into CMTKG. Fig. 1 shows the structure of CMTKG. This chemical alteration brings about lower biodegradability than TKG and along these lines upgrading the existence shell, high drug loading capacity, swell capacity, mucoadhesivity, in situ gelation, wide pH resistance, hydrophilicity, steadiness, and release kinetics. Moreover, in addition to all the above-mentioned properties, CMTKG additionally shows antibacterial properties like that of TKG. Significantly, all these mentioned properties greatly enhance the efficiency and convenience for drug-delivery. In fig. 2, the unique properties and importance of CMTKG have been featured.

This survey article attempts to incorporate all the conceivable information published till date with respect to the drug delivery vehicles of CMTKG.



**Figure 1.** Chemical structure of CMTKG.

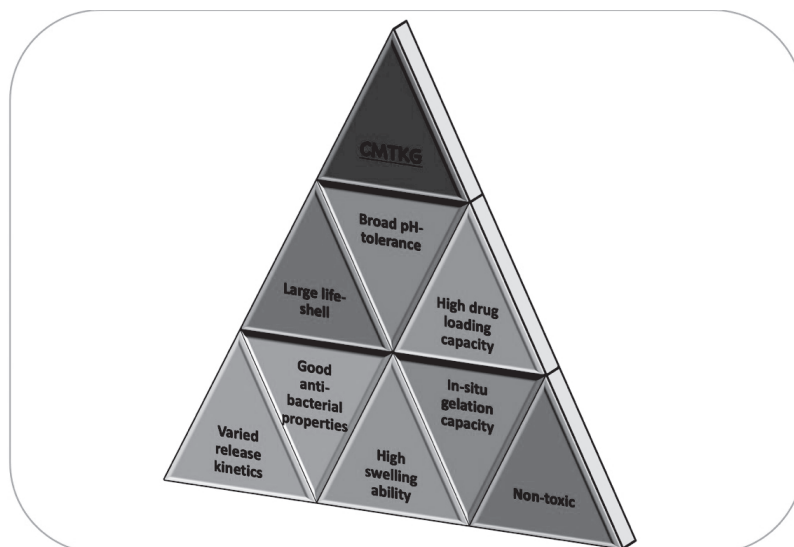


Figure 2. Properties of CMTKG.

## 2. Derivatization of CMTKG from TKG

The derivatization of CMTKG from TKG has been done by a solitary approach. In short, TKG's carboxymethylation (0.10 m) should be possible by mixing basic methanol having sodium hydroxide into TKG along adding monochloroacetic acid (MCA) (0.32 m). This arrangement is kept in a water bath at 343 K for 1 hour. The solution is then filtered with the help of a sintered glass (G-3) after which the separated item gets mixed with water. This is followed up with neutralization using dil. HCl. This is then precipitated by ethanol and trailed by unadulterated methanol. The product is to be dried, first at room temperature, lastly drying at vacuum broiler kept up for 4h at 313 K (Goyal et al., 2007).

The process for the carboxymethylation of TKG can be perceived with the help of fig.3. The reaction begins by NaOH attack on the TKG's hydroxyl group. SN2 is responsible for framing carboxymethylation between MCA and TKG-alkoxide (Goyal et al., 2007). The side reaction occurs too which results from the reaction between MCA and sodium hydroxide resulting in sodium glycolate (Tijssen et al., 1999).

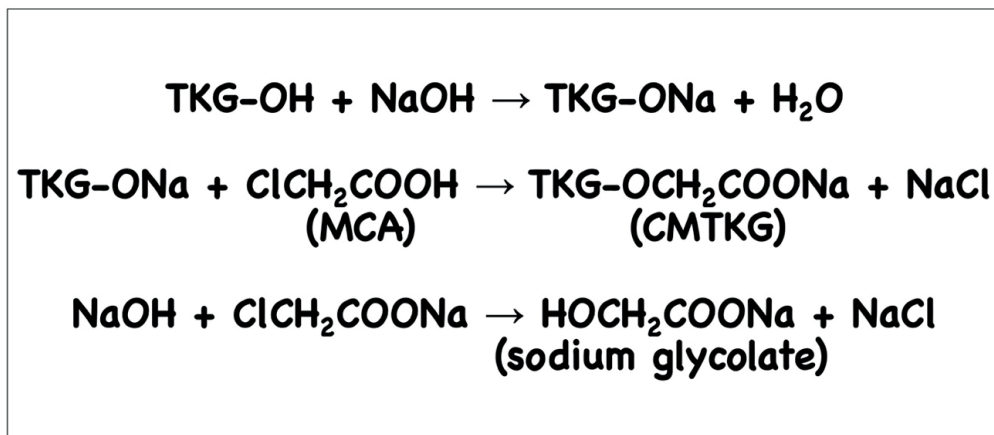


Figure 3. Derivatization of TKG into CMTKG.

**Table 1.** CMTKG based drug-delivery applications.

<b>Application</b>	<b>Polymer(s)</b>	<b>Methodology</b>	<b>Form of CMTKG</b>	<b>Key aspects</b>	<b>Ref.</b>
Ibuprofen drug delivery (targeted colon)	cellulose	spheronization-extrusion technique	pellet	Drug delivery was done without using any coating technique	(Pandit et al., 2018)
colon region and intestine specifically targeted drug release	curcumin	spheronization-extrusion technique	pellet	absorption of curcumin increased by 2 fold	(Kshirsagar et al., 2017)
remove conventional floating matrix pellet's constraint	Hydroxypropyl methyl-cellulose	wet-granulation method	pellet	control release of verapamil hydrochloride (in stomach) for 12 h	(Mali et al., 2016)
<i>in vitro</i> drug release studies of methelene-blue	Poly (vinylpyrrolidone)	mixing	pellet	release studies were done using USP rotating paddle method	(Pal et al., 2008 a)
Lansoprazole pellets' synthesis	cellulose	spheronization-extrusion technique	pellet	Drug release found higher than commercialized LANZOL tablets	(Muley et al., 2017)

controlled release of Diclofenac sodium	<i>Cassia fistula</i>	compression methodology	pellet	super case-II mechanism	(Huanbutta et al., 2018)
Lornoxicam loaded system	cellulose	spheronization-extrusion technique	pellet	compression force influence on pellet	(Gowda et al., 2014)
delivery of aceclofenac in controlled manner	gelatin	cross-linking	composite	aceclofenac-loaded composite showed anti-inflammatory activity	(Jana, Banerjee, et al., 2016)
optimization of composite	poly (acrylonitrile )	graft copolymerization using Microwave	composite	concentration of acrylonitrile showed inverse relationship with the grafting efficiency	(Meenkashi et al., 2014)
Erlotinib based composite for therapy (NSCLC)	poly (N-isopropylacrylamide)	free-radical polymerization	composite	suppressed growth of A549 and apoptosis cell proliferation	(Bera et al., 2019)

Tropicamide (ocular delivery)	Diocetyl sodium- sulfosuccinate	gelation	nano- particle	<i>ex vivo</i> release	(Kaur et al., 2011)
ciprofloxacin- nanoparticles	-	ionotropic- gelation	nano- particle	its effect was studied on the lines of Vero cell	(Dilbaghi et al., 2013)
immobilization of amylase	tetramethoxy -silane	sol-gel polymerization	nano-hybrid matrix	enzyme activity did not alter post 90 days	(Singh et al., 2011)
loading of drug moxifloxacin- hydrochloride	citric acid	Cross-linking	hydrogel	citric acid as crosslinking agent	(Mali et al., 2017)
<i>in vitro</i> acyclovir release	NaAlginate	gelation	hydrogel	CMTKG retards release of drug	(Jana, Sharma, et al., 2016)

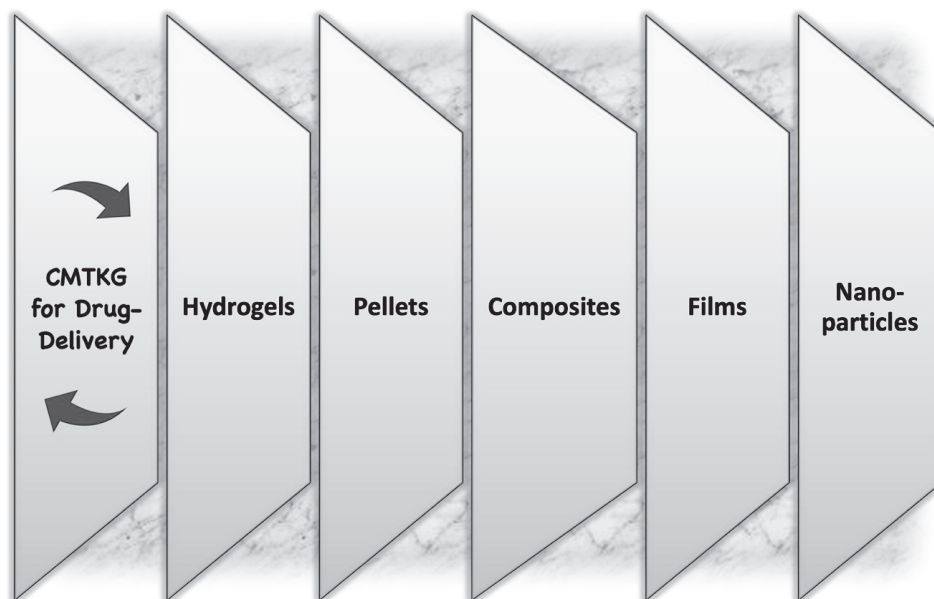
release of metronidazole	PVA	Freeze-thaw	hydrogel	metronidazole released upto 360 min.	(Meenakshi et al., 2014)
aceclofenac release for controlled delivery	chitosan	cross-linking	hydrogel	showed controlled release of aceclofenac for 12h	(MALI et al., 2017)
mucoadhesive <i>in situ</i> nasal gel	Poly(NaAcrylate)	<i>In-situ</i>	gel	safest while using histopathological evaluation	(Mali et al., 2015)
ciprofloxacin <i>in vitro</i> release	gelatin & tamarind gum	simple-mixing	hydrogel	high proliferation of the MG63 cells	(Shaw et al., 2015)
graphene oxide's effect on various properties of film	poly (vinyl alcohol)	cross-linking	film	graphene oxide concentration was inversely proportional to the amount of drug release	(Yadav et al., 2018)

controlled release of budesonide drug in colon	chitosan	simple-mixing	film	Release of budesonide happened for 19h through mimicking the large intestine	(Kaur, 2009)
kills bacteria who produces resistance for drug	Ag nanoparticles	<i>in situ</i>	film	Minimal toxicity for cells	(Sanyasi et al., 2016)
bio-logical improvement of PVA films	poly (vinyl alcohol)	casting-method	film	successful competitor in the sphere of skin diseases and wound dressings	(Yadav et al., 2017)
strength of bio adhesive	Tamarind kernel gum	solvent-casting method	film	drug delivery systems for anionic and cationic derivatives	(Kaur et al., 2013)

### CMTKG BASED DRUG-DELIVERY SYSTEMS

CMTKG based formulation in the drug field has been accounted for through various modes such as nasal, ophthalmic, colon and oral. Here, CMTKG systems for delivery of drugs frameworks along with their utilities with various forms (fig. 4) have been examined in detail and have likewise been summed up in Table 1.





**Figure 4.** CMTKG various forms as drug delivery excipient.

## PELLETS

Ibuprofen stacked CMTKG pellets were synthesized by the technique of extrusion/spheronization utilizing the 3-level 2-factor full factorial method (Pandit et al., 2018). These pellets were exceptionally intended to target colonic illness by means of drug delivery of colon. Voids were reported in pellets by electron microscopy. In vitro disintegration trial of stacked CMTKG pellets at various pHs phosphate buffers were done which indicated drug discharge within 10h.

CMTKG/Curcumin pellets were formed by Pandit and Kshirsagar (Kshirsagar et al., 2017) in order to utilize it as a drug conveyance for colon. All pellets were readied utilizing curcumin and CMTKG by using the expulsion spheronization strategy. Disintegration and assimilation, as far as curcumin is concerned, were seen to have expanded to 1.5 and 2 fold, respectively. Rodent digestive tract portions were utilized for in vitro contemplates that indicated curcumin pellets' oral bioavailability.

A gliding bio-adhesive tablet utilizing wet granulation technique was planned and for optimization, a central composite plan was used. These tablets were readied utilizing HPMC (hydroxypropyl methylcellulose), CMTKG, and  $\text{NaHCO}_3$  and additionally stacked with the VH (verapamil hydrochloride). Almost 16-29% of discharge was demonstrated in first hour which indicated slow discharge up to 12h. It was concluded by the authors that the pellets display all the possibility in the stomach to control and hold the arrival of verapamil hydrochloride up to 12h (Mali et al., 2016).

Pal et al. in 2008, made CMTKG pellets by derivatizing TKG (Pal et al., 2008a). The framed CMTKG was affirmed by utilizing different instrumentation, in particular FTIR spectroscopy, DTA, TGA,  $^{13}\text{C}$  NMR spectroscopy, natural consistency estimation, and SLS (static light scattering method). It was utilized for the slow conveyance of methylene-blue. For slow delivery examination, USP (United States Pharmacopeia) paddle technique was used, which also used to contemplated the energy of equivalent. It was seen that for 30% of dischargement of drug, this tablet complied with zero order whereas non-Fickian showed for 60% of the drug discharge.

Lansoprazole pellets having CMTKG have been reported by Muley et al. utilizing the expulsion/spheronization method (Muley et al., 2017). The pellets were assessed for yield (93.53%), breaking downtime (292 sec), drug content (90.46%), and hardness (0.307 kg/cm<sup>2</sup>). Drug release was discovered to be 2.0% more than the marketed formulation.

Sittikijyothin and Huanbutta exhibited the tablet structure from the *Cassia fistula*'s carboxymethylated gum and CMTKG for accomplishing the slow discharge of the drug (diclofenac sodium). Its discharge observations and medication content assessment was conducted by *in vitro* to assess the synthesized pellets. Hardness and pressure period expanded whereas thickness discovered diminished when there was elevation in the concentration of gums (Huanbutta et al., 2018). The outcomes came was also studied by using a mathematical model.

## COMPOSITES

Interpenetrating network bio-composite of CMTKG and gelatin is found useful for the use of slow medication conveyance utilizing aceclofenac medication. CMTKG in any bio-composites stifled the pace of medication discharge in HCl arrangement, however, similar elevation took place in 6.8 pH buffer solution. Basic dissemination polymeric chain and *in vitro* unwinding was cited as the purposes behind slow medication discharge. The medication entanglement effectiveness was discovered to be above 90%. The authors recommended their medication stacked bio-composites has the potential as calming therapeutics (Jana, Banerjee, et al., 2016). 4 factor, 3-level focal composite plan was used in order to contemplating composite comprised of microwave-helped join of polyacrylonitrile and CMTKG. The response surface technique indicated a synergic impact by changing the convergence of ammonium persulfate while the uniting proficiency diminished as elevation in the amount of acrylonitrile was observed. The proficiency of grafting was accounted for almost 96% in comparison to ideal determined boundaries such as the amount of ammonium persulfate (40 mmol/l), centralization of acrylonitrile (0.10(w/v)), microwave power (160 W), and microwave time (99.48 s). The authors closed it by citing helpfulness in planning pH-responsive applications as the grafting improved the conferred pH-subordinate growing qualities, upgraded the crystallinity, and enhanced thermal stability (Meenkashi et al., 2014).

Erlotinib-stacked semi-interpenetrating network nanocomposites with CMTKG alongside poly (N-isopropyl acrylamide) and montmorillonite were synthesized by Bera et al. (Bera et al., 2019). These composites of nano scale were shown for ERL delivery for the treatment of NSCLC (non-small cell lung cancer). Free radical polymerization was specifically used for the synthesis of this nano-composites and in this way, ERL could be stacked by means of test sonication-assisted self-assembly convention. The combined advanced definition demonstrated extraordinary mucin adsorption capacity and showed cytotoxic action and instigated (MALI et al., 2017) apoptosis against A549 cells, and notably found more than the ERL. Thus, this synthesis can be called as an extraordinary technique in order to perform NSCLC treatment.

## NANOPARTICLES

Ciprofloxacin stacked CMTKG based nano-particles which readied utilizing the gelation strategy (ionotropic). Its outcomes affirmed centralization of MgCl<sub>2</sub> and CMTKG demonstrated a significant impact over the productivity of encapsulation. Toxicity was likewise affirmed of the readied nanoparticles utilizing Vero cell lines through resazurin measure. The antibacterial examination results uncovered the best zone of restraint by these nanoparticles in *Micrococcus luteus* (Dilbaghi et al., 2013).

CMTKG nanoparticles stacked with Tropicamide were arranged and investigated for ocular conveyance (Kaur et al., in 2011). The CMTKG nanoparticles were combined by means of the gelation (ionotropic) method and upgraded utilizing 2-factor, 3-levels focal composite. Its outcomes uncovered the centralization of crosslinker (Calcium Chloride) as well as CMTKG and synergistically affected 5 epitome effectiveness and molecule size. The mucoadhesive as well as non-aggravation behavior of CMTKG based nano-particles demonstrated its appropriateness for a visual/ocular conveyance framework (Kaur et al., 2011).

## HYDROGELS

An IPN hydrogel network was developed by utilizing CMTKG and alginate by gelation technique (induced) by utilizing  $\text{Ca}^{+2}$  particles. The blend of two regular polymers showed up best at Alginate: CMTKG proportion of 3:1 regarding their morphological qualities, and drug conveyance properties. It also revealed swelling tendency of polymeric chains and dissemination as the controlling element for drug discharge (Jana, Sharma, et al., 2016).

Hydrogels of CMTKG and PVA were reported by Ahuja and Meenakshi, in 2014, utilizing freeze-defrost treatment and further assessed for discharge conduct. PVA-CMTKG hydrogels stacked with metronidazole were enhanced utilizing the central composite technique. The advancement study uncovered PVA's large concentration supported the discharge of metronidazole whereas larger extents of CMTKG supported for quicker discharge of metronidazole. It delivered 75% metronidazole for a time of 6h (followed Higuchi's kinetics release) (Meenakshi et al., 2014).

Mali et al. fabricated drug stacked IPN of chitosan and CMTKG with glutaraldehyde as a crosslinking specialist. So as to do the affirmation of the combined hydrogel matrix for drug conveyance was studied by in vivo phenomenon. In order to capture effectiveness of hydrogels, the hydrogel matrix was discovered straightforwardly corresponding for convergence of CMTKG and centralization of the crosslinker. Its outcomes were finished up on looking at supported discharge of drug (aceclofenac) for 12h from the hydrogel matrix whereas rapid delivery for the formulation based on commercialization (MALI et al., 2017).

## FILMS

The conceivable utilization of films produced using CMTKG and chitosan for colon arrival of budesonide for the time of 19h in zero order was assessed and the purpose behind the delivery was given in accordance with the strength in acidic medium, of the formed complexes and capacity of these films to form complex in between  $-\text{COO}-$  of CMTKG and  $-\text{NH}_3$  of chitosan (Kaur, 2009).

Properties of PVA could be viably improved by adding CMTKG to it was suggested by Yadav et al. (Yadav et al., 2017). The CMTKG presence fundamentally improved thermal, mechanical, and biological properties of these films. The paper reported that they utilized the film in medication stacking of the drug (ciprofloxacin) indicating great property of antimicrobial. Improved cell multiplication saw in the films having CMTKG in contrast with the films having only PVA by utilizing human keratinocytes in order to investigate. In light of these perceptions, Yadav et al. proposed films having CMTKG can likewise additionally utilized for dressings of any injuries and skin illnesses in which specific regular medication is required.

Its reported to have additionally fabricated films with graphene oxide-CMTKG-PVA so that they can be utilized them for the use of drug conveyance of ciprofloxacin hydrochloride. The drug discharge revealed to have followed Fickian diffusion law. The measure of drug delivered seemed, by all accounts, to be contrarily relative to the amount of Graphene oxide. It was additionally seen that the films with low concentration of Graphene oxide, indicated enhanced multiplication of keratinocytes (human skin) while, at even more concentration, it discovered to be full of cytotoxicity (Yadav et al., 2018).

## CONCLUSION

The modern use of bio-polymers instead of synthetic polymers is vital for more consistent advancement in the future time. The adaptability of substance alteration of TKG into CMTKG along enhanced utilitarian characteristics stamps it potent for the field of drug carrier. Because of the accessibility of sound data on the phenomenal physico-chemical and pharmaceutical properties of CMTKG, the most recent years done a broad examination of CMTKG for drug conveyance transporter. In any case, its significant to mention here that none of the formulations of CMTKG have figured to the commercial spheres of pharmaceutical because of the lack of safety information. CMTKG has successfully been exploited for drug conveyance carrier however its marketization as well as advantages for the humanity are as yet another sphere of examination.

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