



# Synergistic influence of tetraethyl orthosilicate crosslinker on mixed matrix hydrogels

Sadia Sagar<sup>1</sup> · Abdulaziz Alturki<sup>2</sup> · Muhammad Farhan<sup>3</sup> · Ali Bahadar<sup>2</sup> · Nazia Hossain<sup>4</sup> 

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## Abstract

Hydrogel is a 3D framework of hydrophilic polymeric material that quickly absorbs and retains a huge amount of water (or other fluid) and offers versatile functionality. A series of unique carboxymethyl cellulose/Xanthan gum/polyvinyl alcohol (CXP) blended hydrogels, containing both the natural and synthetic polymers, were prepared by following the blending and casting approach. The polymers were incorporated through chemical crosslinking by tetraethyl orthosilicate (TEOS). The fabricated hydrogels showed all required features: non-toxicity, biocompatibility, and improved mechanical strength. The addition and variation in TEOS (crosslinker) significantly impacted the key characteristics of CXP hydrogel. The scanning electron microscopy (SEM) images showed the porous structure and indicated that the pore's size and intensity were reduced with the surge in TEOS content. Fourier transform infrared spectroscopy (FTIR) results confirmed the successful incorporation of various polymeric strands through crosslinking by TEOS. The thermogravimetric analysis (TGA) highlighted the greater stability of all the hydrogels over high temperatures. The crosslinked hydrogel displayed higher thermal resilience than the uncross-linked one. The differential thermal analysis (DTA) also confirmed that the addition of TEOS content drastically enhanced the thermal endurance of crosslinked hydrogels in comparison with the neat hydrogel. All the specimens exhibited good swelling ability in distilled water during the swelling studies. This study also reflected that the addition of crosslinker in a limited amount (50  $\mu\text{L}$ ) has significantly enhanced the swelling but further increase in concentration hindered the water uptake. The swelling response of blends towards pH revealed low swelling of films in acidic and basic pH, but maximal swelling in neutral media. This unique pH response of hydrogels at neutral pH along with the biocompatibility made them suitable for injectable managed drug carrier.

**Keywords** Hybrid hydrogel · PVA hydrogel · TEO crosslinked hydrogel · Drug delivery system

## Introduction

Hydrogel is a collection of soft and moist formulations that comprise a low volume 3D pore framework of polymeric chains, with a higher hydrophilic nature and it can retain a huge quantity of water/fluid upholding their configuration

due to physical or chemical bonding between various polymeric chains. Hydrogels can absorb fluid up to several thousands of folds of their initial weight at least 10–20% (lowest value) to form chemical sustainable or organic degradable gels, based on hydrophilic character resulted from polymeric hydrophilic containments and also from the nature and concentration of the network linkages. The process of swelling of hydrogels takes place in three phases; the absorption of water/liquid by the hydrogel framework, the water uptake and expansion of polymeric backbone, and the addition of water and widening of the interlinked polymeric matrix (Geckil et al. 2010).

A minor variation in environmental factors like temperature, electric charge, pH, enzyme, or ion presence can cause rapid but temporary changes in shape or texture of the hydrogel. These variations may occur at the macroscale as precipitous emergence, modifications in size, and amount of

✉ Nazia Hossain  
bristy808.nh@gmail.com

<sup>1</sup> Department of Physics, Fatima Jinnah Women University, Rawalpindi, Pakistan

<sup>2</sup> Department of Physics, The University of Lahore, Lahore, Pakistan

<sup>3</sup> Department of Chemical and Materials Engineering, King Abdulaziz University, Rabigh 21911, Saudi Arabia

<sup>4</sup> School of Engineering, RMIT University, Melbourne, VIC 3001, Australia

water. The inherited biocompatible properties and the flexible nature of hydrogel, similar to living tissue due to the presence of water in polymeric strands, show their suitability for numerous biomedical and pharmaceutical applications. The scale, structure, and functionality of hydrogels vary depending on four key aspects concerned in hydrogels development, the origin of polymers, the design of frameworks, gel operation, and applications of gel. Following the fact “structure decides functionality”, we are more inclined to presume that efficient structural architecture is the assertion to manage the hydrogel functionality proficiently and also to have a more specific application certainty. The method of cross-linkage is the “link” between gel origins and gel formations, whereas the nature of design serves as a “bridge” among gel formations and gel features (Clark 1991; Osada et al. 2004). Two complementary conditions are needed for the formulation of hydrogels. First, hydrophilic groups on the polymer backbone or side chain are necessary for polymeric strands (organic or artificial). Second, there should be significant cross-linkage intensity across the molecules for establishing gel frameworks (Li et al. 2021).

Usually, hydrogels of natural origin have poor mechanical characteristics (Vasile et al. 2020). Natural hydrogels are intrinsically biocompatible, bioactive, and degradable as mostly derived from organic origins like plants, brown algae, bacterial crops, and proteins-like collagen, gelatins and fibrin, fibrin glue, and a cellular tissue matrix, and polysaccharides like alginate, gelatin agarose, and chitosan (Vlierbergh et al. 2011). The structure and crosslinking intensity of the hydrogel polymer matrix influences the mean pore size, dispersion of the pore volume and pore interrelation greatly (Weber et al. 2009). Controlling the distribution of the pores in many products is beneficial for a variety of applications, like tailorable lode /release of biomolecules. Based on the design of the material, the mechanical characteristics and swelling kinetics can be varied and improved by blending natural and synthetic polymers. Furthermore, a gel with exceptional strength can be achieved by raising the crosslinking intensity or minimizing it by raising the temperature (Garg et al. 2016).

Xanthan gum (XG) is a natural polysaccharide that is famous for its intrinsic cytocompatibility, higher water adsorption capacity, non-toxicity, degradability, and muco-adhesive properties. It is chemically formed from  $\beta$ -1, 4 D-glucopyranose glucose frameworks, consisting of mannose ( $\beta$ -1, 4), glucuronic acid ( $\beta$ -1.2) with a pendant trisaccharide side chain, and mannose marginal sequences. Due to its visco-elastic activity and thermal tolerance, XG is being incorporated into water-based fracking liquids (Garcia-Ochoa et al. 2000). Also, the XG matrices can shield the medicine from the exposures of the gastrointestinal tract and supply it to desired area without loss (Gils et al. 2009). For its acid tolerance, XG, combined with the other

composites, has been used as an emulsifier in tablets and a supportive hydrogel in medicine delivery systems.

Carboxymethyl cellulose (CMC), derived from natural cellulose, is a fibrous gum containing ( $-\text{CH}_2-\text{COOH}$ ) functional group. It comprises the cellulose spine composed of glucopyranose polymers and is attached to some of the glucopyranose monomer's hydroxyl entities (Caballero et al. 2015). CMC is hydrophilic, water-solvent, widely accessible, and has cytocompatibility, permeability, recyclability, sustainability, good mechanical characteristics, and friendliness towards the environment mainly employed in applications of biomaterials (Lv et al. 2018). Colloidal carboxymethyl cellulose is extremely gelatinous and biologically nontoxic and hence is commonly used in nutrition, medicinal, cosmetic, etc. (Kabir et al. 2018; Lv et al. 2019). CMC-derived hydrogels have shown the potential to be employed in wound treatment, targeted medicinal distribution, and as absorbing materials (Javanbakht and Shaabani 2019).

Polyvinyl alcohol (PVA) is artificial water-soluble polymeric material. PVA is a crystallite atactic polymer consists mostly of 1,3-diol-assembly [ $-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-\text{CH}(\text{OH})-$ ] although some fraction of 1,2-diols [ $-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}(\text{OH})-\text{CH}_2-$ ] exist. PVA is useful in membrane buildup, emulsification and shows perfect adhesion properties. Its linking richness and humidity resilience contribute to its applicability in this field. Surprisingly, the usage of polyvinyl alcohol for the encapsulation of curative tabs is significant in other expanding markets of the healthcare sector (Tang and Alavi 2011). With other plastic polymers, PVA exhibits inherent compatibility. This commonly adopted flexible plastic is safe, innocuous, and gentle towards biological tissues.

Crosslinker is the chemical used to create a crosslinked fluid system. The crosslinker agent controls the structure of the polymer matrix in molecularly imprinted polymers, in which such agents lead to aggregation and connection of functional monomers to each other and stand firmly in own place and polymerization and molecular template separation, and the functional monomers do not displace and create carved holes (Khan et al. 2020; Yang et al. 2017). The polymers with high percentages of crosslinker lead to the generation of stabilized micropore over the three-dimensional structure which can guarantee the conformity of the shape and functional groups of the sites with the target molecules. Natural and synthetic polymers are crosslinked using different crosslinkers such as formaldehyde, glutaraldehyde, acetaldehyde, tetraethyl orthosilicate, and others (Wasim et al. 2017; Iqbal et al. 2021). During the polymerisation process cross-linkers fix the functional monomers around template molecules, which results in the forming of highly crosslinked rigid polymer network (Ahmad et al. 2017; Parhi 2017). However, Silane implicated polymerization gives

significant diversity in variation of reaction, effectiveness, improved low temperature properties and quicker development (Bashir et al. 2018). Tetraethyl orthosilicate (TEOS) is the most desired crosslinker due to its harmless nature and also capable to introduce inorganic crosslinks (Si–O) in the amorphous precinct of polymers (Peppas et al. 2006; Islam et al. 2016). The TEOS is an achromatic inorganic liquid, with the chemical composition  $\text{Si}(\text{OC}_2\text{H}_5)_4$ , and is a tetrahedral complex of orthosilicic acid,  $\text{Si}(\text{OH})_4$  that deteriorates in water (Vis et al. 1993). The Si–OR bond's reactivity can be employed in the development of membranes and silica hydrogel which extend applications into drug delivery systems. The main advantage of the chemical crosslinking methods is the use of reactive polymers with predefined architecture, chemical composition, and functionalities. The selection of reactive building blocks allows very exact adjustment of nano- and microgel properties. During crosslinking step, one can vary flexibly swelling and mechanical properties of nano- and microgels. Biohybrid nanogels are designed by the crosslinking reactions can occur at very mild conditions, this is probably the best strategy with networks consisting of biological and synthetic polymer chains.

Crosslinking strategy is crucial for tissue engineering applications developing hydrogels with desirable physicochemical and biological characteristics. Moreover, hydrogels with the same constituents but different crosslinking structures can present completely different physicochemical performances (Salma-Ancane et al. 2022). The regulated drug delivery systems are employed to distribute medicines at specific rates over prescribed intervals of time to exceed normal medication limits. The probability of delivering drugs for an extended duration (prolonged discharge is the key benefit achieved by hydrogels. Hydrogels can hold and shield different medications from harsh conditions and deliver them at a required discharge rate and site, have fewer side effects, infection risks, and reduces patient grief. The discharge of drugs can be stimulated on need by local variations in pH, temperatures, complex enzymes, or distant specific stimulus (Bahram et al. 2016; Chen et al. 2021).

In the present study, unique TEOS crosslinked hybrid hydrogels were developed by blending both the features of natural and synthetic hydrogels. Hydrogels were prepared using blends of XG, PVA, and CMC with different amounts of TEOS to investigate its effect. To the best of our knowledge, this work has not been demonstrated in earlier studies. The swelling characteristics of the hydrogels were determined in various pH media. FTIR, SEM, TGA, and DTA of hydrogels is carried out. Furthermore, the influence of cross linker's addition as well as concentration on different properties of developed hydrogel is investigated and reported. The distinct differences in the

physicochemical and biological properties of the developed materials provide new opportunities to design next-generation functional composite hydrogel systems.

## Materials and methods

### Materials

Food grade Xanthan Gum (Molecular weight  $\cong 1.6 \times 10^6$  g/mol) in powder form was received from Biolog (GmbH) Trademark., Germany. Sodium CMC (Molecular weight  $\cong 2.5 \times 10^5$  g/mol), PVA (Molecular weight  $\cong 5.0 \times 10^5$  g/mol), and TEOS (98.5% & density 0.933 g/mL) were purchased from Sigma–Aldrich Ltd., United States. Distilled water was prepared in laboratory.

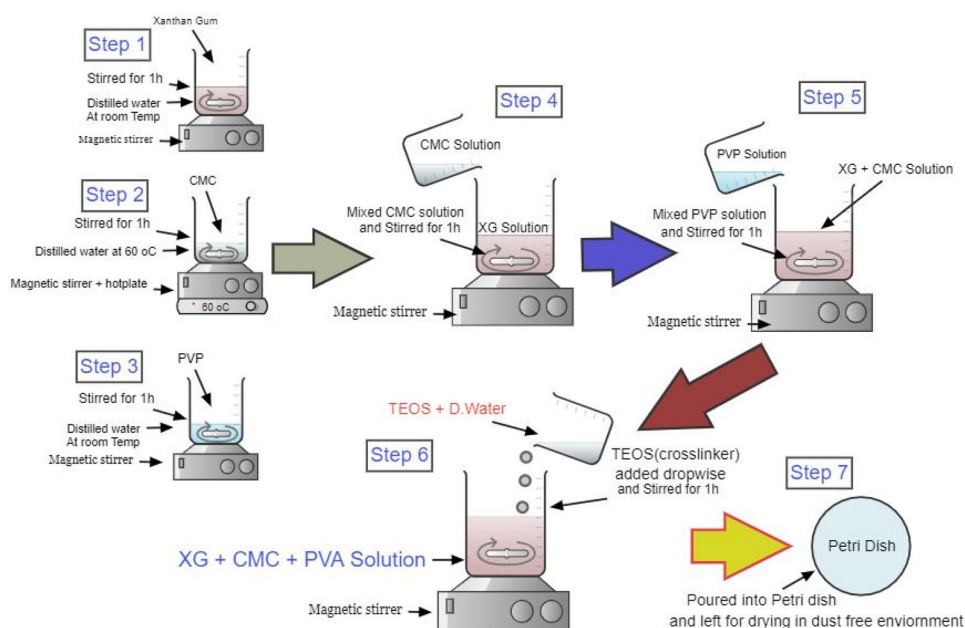
### Synthesis of hybrid hydrogels

0.4 g of Xanthan gum was added into 30 mL of distilled water taken into a glass beaker at room temperature and dissolved by constant stirring placing on a hot plate with a magnetic stirrer. 0.3 g CMC was added into 20 mL of distilled water separately and continued to stir on the hot plate at 60°C with a magnetic stirrer until dissolved. Similarly, 0.3 g PVA solution was prepared in 20 mL distilled water via constant magnetic stirring. Xanthan gum solution was mixed into CMC solution keeping on the magnetic stirrer, and blended for 1 h at room temperature. After blending for 1 h, this solution (Xanthan gum + CMC) was added into PVA solution and again blended for 1 h by the magnetic stirrer. After constant stirring for 1 h, a uniformly mixed CMC/XG/PVA solution was obtained. In the same way, three more samples of CMC/XG/PVA solutions were prepared. Then, tetraethoxysilane (diluted in 20 mL deionized water) was added, in different amount, i.e., 0  $\mu\text{L}$ , 50  $\mu\text{L}$ , 100  $\mu\text{L}$ , 150  $\mu\text{L}$ , dropwise as a crosslinker to prepare four different hydrogel samples (differing in crosslinker amount) and further stirred for 1 h at room temperature. Finally, the blends were transferred into Petri dishes for drying at room temperature in a dust-free environment and placed in a desiccator for further characterization of designed hydrogel (Figs. 1, 2; Table 1).

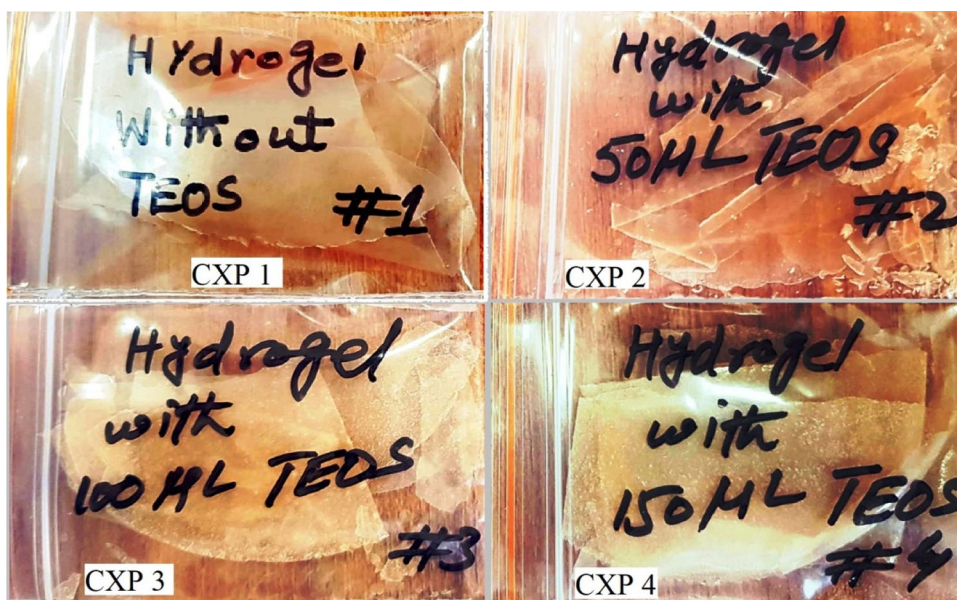
### Characterization of hybrid hydrogel films

Prior to the swelling studies, the hydrogels were characterized by several ways to confirm the formation of hydrogels and swelling behaviour may be anticipated well.

**Fig. 1** Schematic representation of hydrogel's synthesis



**Fig. 2** Prepared samples (with different amount of TEOS)



**Table 1** Code and composition of prepared hybrid hydrogel films

Sr. #	Specimen ID	Xanthan gum (g)	CMC (g)	PVP (g)	TEOS ( $\mu\text{L}$ )
1	CXP 1	0.4	0.3	0.3	0
2	CXP 2	0.4	0.3	0.3	50
3	CXP 3	0.4	0.3	0.3	100
4	CXP 4	0.4	0.3	0.3	150

#### Fourier transform infrared spectroscopy (FTIR)

FTIR spectroscopic analysis was employed to confirm the presence of functional groups in the monomeric units associated with the biopolymers (Xanthan Gum and CMC), and synthetic polymer PVA along the crosslinker TEOS. The FTIR spectra of crosslinked and uncross-linked CMC/XG/PVA blended hydrogels were obtained within the region of  $4000\text{--}400\text{ cm}^{-1}$ .



### Scanning electron microscopy (SEM)

The scanning electron microscope (SEM) was employed to investigate the surface and pores characteristics of all the hydrogel formulations. SEM analysis of freeze-dried hydrogel membranes was carried out by Scanning Electron Microscope JSM, (JEOL Japan) with an accelerating voltage of 20 kV and magnification of  $\times 1000$ .

### Thermal gravimetric analysis (TGA)

Thermogravimetric analysis (TGA) was carried out to determine the thermal degradation behaviour and thermal resilience as a proportion of temperature or time in a regulated atmosphere cooled or heated at a regulated pace. The TGA was conducted by Perkin Elmer TGA-4000 in temp range (room temp to 800°C), the specimen is heated at a regulated pace (10°C per min) in a particular controlled atmosphere of N<sub>2</sub>.

### Differential thermal analysis (DTA)

Differential thermal analysis (DTA) was used to classify and evaluate the chemical constitution of materials by monitoring the thermal activity of a CXP hydrogels.

In DTA, the temperature of the sample material was examined as a proportion of temperature compared to that of a nearby neutral substance, whereas the material and referent were exposed to a regulated temperature regimen. The DTA was also carried out by Perkin Elmer TGA/DTA- 4000

in temp range (room temp to 800°C), the specimen is heated at a regulated pace (10°C per min).

### Swelling studies in distilled water and various buffer solutions

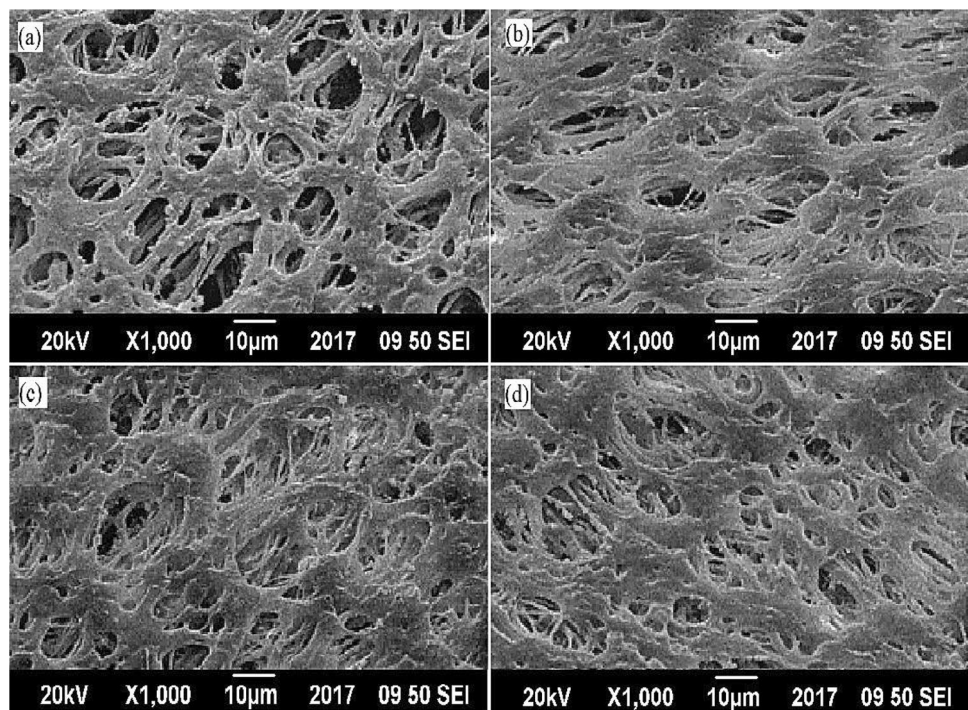
The swelling study was conducted at room temperature to check the water absorption capacity of prepared CXP hydrogels. The pre-weighed (100 mg) CXP membranes ( $W_d$ ) were immersed in excess of distilled water one by one, and after a specific time interval (10, 20, 30, 40, 50) minutes the swollen hydrogel film was weighed ( $W_s$ ) again by taking out the films and removing excess water. The same procedure was carried out for all the prepared samples' compositions (CXP1, CXP2, CXP3, and CXP 4) until equilibrium was achieved.

The hydrogel's swelling response in various pH solutions (pH 4.0, 7.0, and 10) was also evaluated at human body temperature (37°C) using a temperature-controlled heat bath. The swelling ratio (%) for samples is determined by the following equation (Pal et al. 2007).

$$S_R = \frac{W_s - W_d}{W_d},$$

where  $S_R$  = swelling ratio,  $W_s$  = Weight of swollen hydrogel film,  $W_d$  = Weight of dry hydrogel film.

**Fig. 3** Micrographs of CXP hydrogels with different amount of crosslinker TEOS, 0  $\mu$ L (a), 50  $\mu$ L (b), 100  $\mu$ L (c), and 150  $\mu$ L (d)



## Results and discussion

### Scanning electron microscopy (SEM)

The SEM micrographs of all hydrogel's samples (CXP1, CXP2, CXP3, and CXP4), with different amounts of crosslinker, are shown in Fig. 3a–d. Images of all the samples show highly porous structures. The micrographs reflect that any variation in the amount of crosslinker significantly affects the pore size and density of pores. Micrograph of neat hydrogel CXP1 displays larger pores of non-uniform sizes and empty spaces between the polymeric patches. Also, it was observed that the addition of a crosslinker reduced the pore diameter and pore density of CXP2, CXP3, and CXP4. It can be inferred easily that further increases in the crosslinker content resulted in narrowing the pores while increasing the size of polymeric patches as Xanthan Gum, PVA and CMC were successfully crosslinked with TEOS to provide homogenous texture.

### Fourier transform infrared spectroscopy (FTIR)

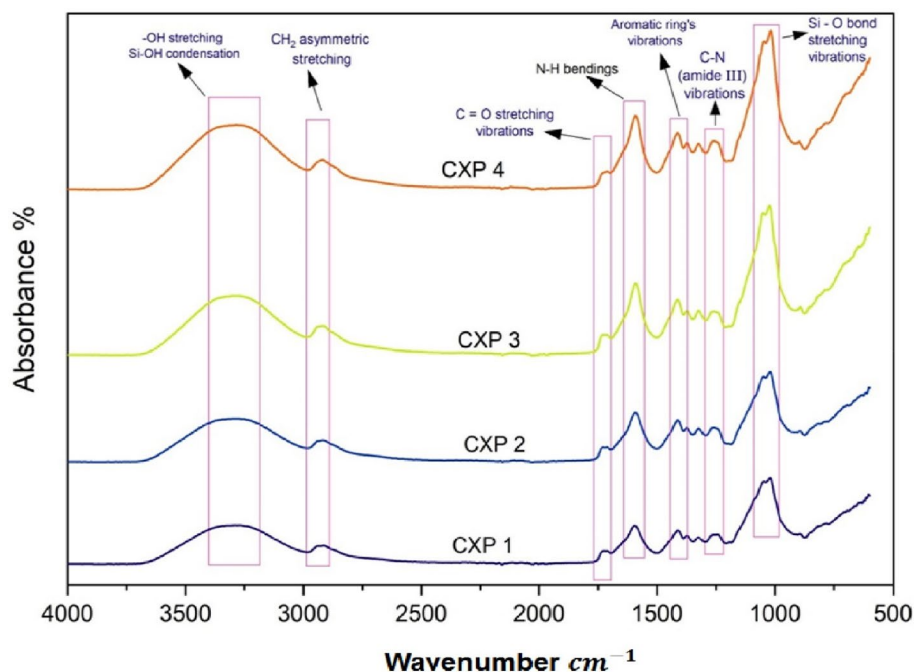
The FTIR spectra of crosslinked and uncross-linked CMC/XG/PVA blended hydrogels are shown in Fig. 4. In all the spectra, the broad peak found near  $3280\text{ cm}^{-1}$  was due to stretching vibrations of H-bonded  $-\text{OH}$ . The broadening of this peak was due to Si–OH condensation, which confirmed the crosslinking phenomenon. A less intense peak near  $2917\text{ cm}^{-1}$  was attributed to  $\text{CH}_2$  groups' asymmetric stretching. A smaller peak near  $1710\text{ cm}^{-1}$  was associated

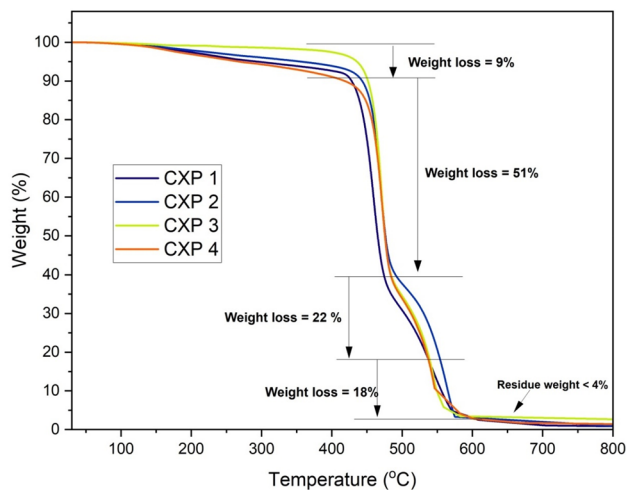
with  $\text{C}=\text{O}$  stretching. In the IR spectrum of CXP hydrogels, the peak about  $1590\text{ cm}^{-1}$  (amide-II) was due to carbonyl bonded N–H bending which was shifted to a lower wavelength with the addition of TEOS. The IR peak about  $1410\text{ cm}^{-1}$  was attributed to aromatic rings. The absorption band around  $1260\text{ cm}^{-1}$  (amide-III) was due to vibrations of C–N and N–H of amide blocks. Two sharp peaks around  $1020\text{--}1090\text{ cm}^{-1}$  were associated with Si–O bond stretching indicate the presence of TEOS. The intensity of the peaks was increased with the surge in TEOS amount. The peak at  $1060\text{ cm}^{-1}$  was attributed to the stretching vibrations of acetyl linkages C–O–C formed because of a reaction between XG, CMC, and PVA. The IR pattern of the designed hydrogels in the fingerprint region ranging from  $1300$  to  $900\text{ cm}^{-1}$  confirmed the presence of carbohydrate of major functional groups of polysaccharides and carboxyl group. These results suggested the development of interactions between XG, CMC, and PVA in the presence of TEOS crosslinker.

### Thermo gravimetric analysis (TGA)

The thermal resilience behaviour of the hybrid hydrogel blend was assessed through Thermogravimetric Analysis (TGA). The thermogram of CMC/XG/PVA blends showing the weight loss behaviour towards temperature is shown in Fig. 5. The TGA curves of polymer blends with various amounts of TEOS showed four major phases of weight loss. The first weight loss phase of about 9% was observed from  $30$  to  $433^\circ\text{C}$  due to hydrogel membranes' dehydration presented in Fig. 5.

**Fig. 4** FTIR absorbance spectra of CXP hydrogels





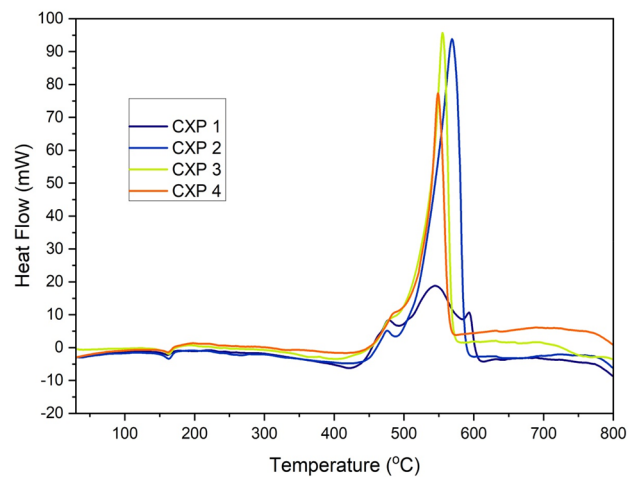
**Fig. 5** Thermogravimetric analysis of hydrogels

### TGA thermo-grams of CXP hydrogel series

All the hydrogels displayed significant thermal stability up to 443°C of temperature. In the second step, from 443 to 485°C, a very sharp decline of about 51% was observed related to degradation of crosslinks, cellulose and glucose matrix along with volatilization of hydrogel films. The third degradation step was observed from 485 to 540°C involving the elimination of major functional groups like –OH, –NH, and –COOH group from the residual causing a weight loss of about 22%. The final step from 540 to 580°C is related to the decay of the polymeric backbones into ashes. It can be seen that the weight loss (%) of crosslinked CMC/XG/PVA blend membranes was lower than that of uncross-linked CMC/XG/PVA membranes. It was also noted that the weight loss was prolonged with a surge in the crosslinker content in the films especially CXP2 and CXP3 evidencing that the crosslinked hydrogel blends were thermally more stable than uncross-linked ones. The thermogram also exhibited the residue weight contents for all the samples above 580°C. The improvement in thermal resilience (increased residual weight) was attributed to the reduced chain mobility of macromolecules in the crosslinked hydrogel, caused by the strong intermolecular interactions between components due to TEOS.

### Differential thermal analysis (DTA)

The differential thermal analysis (DTA) is elaborated in Fig. 6 in temperature range 30–800°C of synthesized TEOS crosslinked CMC/XG/PVA nano-hydrogel. Although all the TEOS impregnated CXP polymeric hydrogel absorbed heat but the crosslinked hydrogel showed higher heat absorbance.



**Fig. 6** DTA analysis of hydrogels

### DTA curve (heat flow vs. temperature)

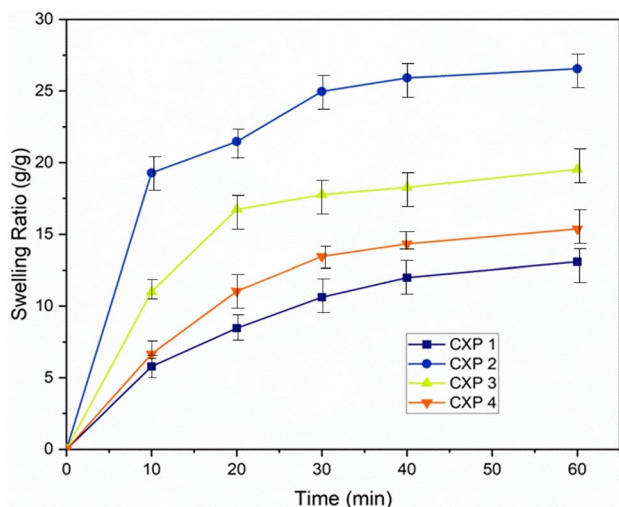
This phenomenon reflected that the addition of crosslinker may enhance the endothermic nature of hydrogel specimens. Also, the gradual increment of TEOS drastically enhanced thermal endurance of designed hydrogel. Augmentation in thermal stability and heat absorbance with increasing TEOS content CMC/XG/PVA hydrogel proved the efficacy of crosslinker. This is due to nanoscale strong interaction of polymeric chain with enhanced crosslinking of CXP nano-hydrogel.

### Swelling studies

#### Swelling behaviour in distilled water

The swelling capacity of hydrogels is one of the key characteristics of hydrogels. The swelling properties of developed hybrid hydrogels were evaluated to determine their fluid retention capacity as well as to assess the influence of crosslinker on hydrogel swelling behaviour. The swelling behaviour of developed hybrid hydrogel membranes of both crosslinked and uncross-linked films was determined in distilled water at 25°C. For this purpose, all the prepared samples were immersed in distilled water and their swelling ratio was recorded after regular intervals (10 min) of time until the swelling equilibrium was obtained. The recorded data are described in the form of a graph between swelling percentage and time as shown in Fig. 7. The swelling percentage of all the samples increased over time. The water uptake ratio increased linearly and reached its maximum value within 20 min. Both the crosslinked and uncross-linked CMC/XG/PVA films reached a suitable swelling level in approximately 40 min. After this the speed of water uptake was gradually decreased and swelling equilibrium was achieved about 3 h.





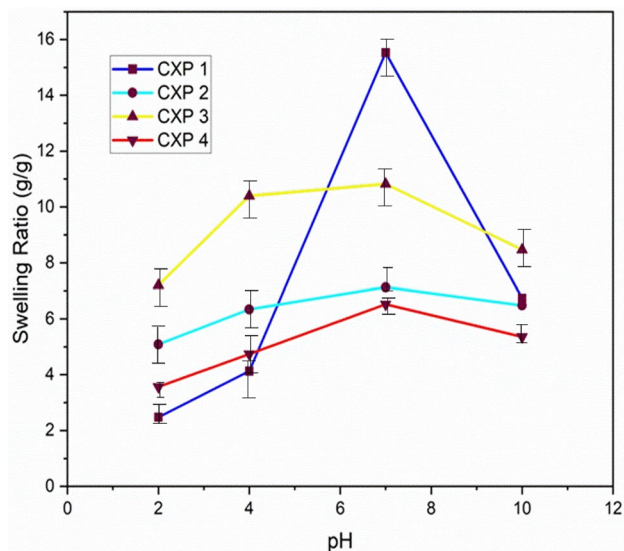
**Fig. 7** Swelling behaviour of CXP films in distilled water

The maximum swelling % observed was 260% for CXP 2 (with 50 $\mu$ L TEOS) and the minimum was observed as 120% for CXP1 (w/o crosslinker) after 40 min.

Figure 7 clearly indicates that the addition of a crosslinker enhanced the degree of swelling. The crosslinked hydrogel displayed nearly twice the swelling as compared to the uncross-linked one, indicating the rise in water retention caused by crosslinking. However, the hydrogel showed the highest swelling % only for a lower concentration of crosslinker, i.e., for CXP 2 film (50  $\mu$ L TEOS). The swelling percentage goes on decreasing as we further increased the cross-linker's content (100 or 150  $\mu$ L TEOS). The degree of swelling was reduced due to the higher number of interlinkages that decreased the spaces/voids (free volume) between the polymer molecules which constrained the polymeric chain movement and prohibited water molecules to penetrate the films. It can be concluded that the crosslinked hydrogel, with a limited amount of TEOS, is the ideal one for swelling-based applications.

### Swelling response towards pH

The response of hydrogel towards environmental stimuli is quite important. The variation in pH of the external medium affects the infiltration of solvent molecules into the hydrogel matrix (Swarnalatha et al. 2008). The pH sensitivity of both the crosslinked and uncross-linked hydrogels was assessed by recording their swelling behaviour being immersed in various pH solutions. For this purpose, each of the four CMC/XG/PVA blended hydrogels was placed into pH-2, pH-4, pH-7, and pH-10 solutions for 10 min. The swelling behaviour was recorded and described as a graph between swelling ratio and pH as shown in the Fig. 8.



**Fig. 8** Correlation between swelling ratio and pH

### Swelling in different pH solutions

All the hydrogels displayed a better swelling ratio in neutral solvent (pH-7) and the uncross-linked hydrogel CXP1 showed maximum swelling ratio among all. The crosslinking percentage also affected the swelling response and it decreased with an increase in TEOS content. Only the hydrogel CXP3 (with 100 $\mu$ L TEOS) showed sophisticated swelling performance in the acidic medium as compared to other hydrogel films. All the hydrogels displayed the usual water uptake in the basic medium. From all the above results, it can be summarized that the swelling performance of all hydrogels at pH-7 was significant and useful in biomedical applications.

## Conclusions

A series of unique blended CMC/XG/PVA hydrogels, containing both the natural and synthetic polymers, was successfully prepared by the following blending and casting approach. The fabricated hydrogels showed all required features like non-toxicity, biocompatibility, and improved mechanical strength. The addition and variation in TEOS (crosslinker) significantly impacted the key characteristics of CXP hydrogel. The SEM images showed the porous structure and indicated that the pore's size and intensity were reduced with the surge in TEOS content. FTIR confirmed the presence of all the functional groups associated with polymers. It also indicated the successful incorporation of various polymeric strands through crosslinking by TEOS. The thermogravimetric analysis (TGA) highlighted the greater stability of all the hydrogels over high



temperatures. The crosslinked hydrogel displayed higher thermal resilience than the uncross-linked one. The DTA also confirmed that the addition of TEOS content drastically enhanced the thermal endurance of crosslinked hydrogels in comparison with the neat hydrogel. All the specimens exhibited good swelling ability in distilled water during the swelling studies. This study also reflected that the addition of crosslinker in a limited amount (50  $\mu\text{L}$ ) has significantly enhanced the swelling but further increase in concentration hindered the water uptake. The swelling response of blends towards pH revealed low swelling of films in acidic and basic pH, but maximal swelling in neutral media. This unique behaviour of hydrogels along with the biocompatibility indicated their potential for many uses in biomedical fields like drug delivery, targeted delivery, food packaging, and dye removal.

**Author contributions** SSI: conceptualization, formal analysis, supervision, writing—original draft; MF: methodology, data curation, formal analysis, writing—original draft, AA: writing—review and editing, AB: writing—review and editing, NH: formal analysis, writing—review and editing.

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**Data availability** The data that support the findings of this study are available from the corresponding author, Nazia Hossain, upon reasonable request.

## Declarations

**Conflict of interest** The authors declare no conflict of interest.

**Ethical approval and consent to participate** The facts and views in the manuscript are solely ours, and we are totally responsible for authenticity, validity, and originality. We also declare that this manuscript is our original work, and we have not copied from anywhere else. There is no plagiarism in my manuscript.

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