

Synthesis and Characterization of Silane Crosslinked Hydrogel to pH Sensitive Study

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Abstract

Eco-friendly and biodegradable novel hydrogel were prepared by blending and solution casting method. The designed hydrogel is based on chitosan/PEG600/Gurgam with carbon nanofiller along silane crosslinked (TEOS) with pH sensitive response to controlled release of drug in biomedical materials and agriculture industry. The various concentration of carbon nanofiller is used to analyze its effect on the fabricated hydrogel characteristics by using FTIR, SEM, TGA, swelling studies (water, buffer and ionic solution). Spectra of FTIR reflected both established and newly developed groups (like hydrogel). COOH group presence is clearly observed in this range in the carbon filler reinforced hydrogel. **The SEM micrographs show that CPG0.003 had a collection of polysaccharide chains as thin helices, which is attributed to the increase in the size of porosity.** TGA shows to increase concentration of nanofiller enhanced the thermal stability of the designed hydrogels at temperature 25 °C to 550 °C mass loss percentage decrease upto 20% and increase thermal stability. This pH response made these resultant hydrogels as fruitful competitor against the many reported controlled release application.

Keywords

Chitosan, Guargum, Hydrogel, Carbon Nano Filler, Silane Cross Linker, pH Sensitive

1. Introduction

Hydrogels are cross-linked three-dimensional polymer network have the capability to absorb and maintain aqueous solutions without liquefying. The polymers

cross links, moreover, corporeal entanglement or biochemical covalent bond remains the solid reasons behind the preparation and development of hydrogel. The notable things of hydrogels be there their volume to stock aquatic, which creates them particular single ingredients. As hydrophilic collaboration among the linkages as well as H₂O particles, hydrogels absorb aquatic and swell [1].

The properties of these lenient elements can be enhanced via implanting otherwise crosslinking of monomers of several well-designed clusters [2]. These water absorber materials show capacity variations depend upon exterior stimuli, for example ecological heat, pH, ionic power, mechanical tension etc. [3]. pH responsive hydrogels remain included of suspended acidic –SO₃H and –COOH or basic like –NH₂ groups, So their swelling vary through the stimulus of ionizable assemblies, crosslinking density and outer pH [4]. Numerous articles have been available about synthesize methods; for example, copolymerization, cross-linking of individual or more grafting and functional monomers, etc. [5].

Two methods are used mostly for crosslinking one of them is chemical and other radiation method. Ionization energy of radiation is too much high so there is no need of cross linker, catalyst and initiator [6]. Electrical sensitive and pH-responsive are the properties of Acrylic acid AAc material form complexes with the poly base [7]. For single and multi-components AAc cross linker is better for hydrophilic hydrogels; which are much pH responsive and more ionic strength, as other cross linker due to ionizing power of carboxylic acid [8].

To obtain different kinds of hydrogels these monomers are typically used as a second component for grafting on polymer surface because greater crystallinity PEO not usually used as good absorber [9]. There are exist numerous uses of hydrogels such as popular in agricultural science, pharmacological engineering, foodstuff, personal hygiene, biomedicine, nanotechnology, H₂O remediation, in tissue production, food packing industry and in apply lubricate spills [6]. On the bases of literature reviews it was came to know very small number of information collect about characterization and swelling of hydrogels which are prepared from acrylic acid and polyethylene oxide [10].

pH-sensitive intelligent hydrogels represent a top-tier category of hydrogels extensively employed in the medical field for precise and regulated drug delivery. The importance of pH-responsive hydrogels arises from their ability to function effectively in various pH environments within the body's internal fluids and organs [11].

These hydrogels primarily rely on polymers containing ionic pendant groups attached along their polymer chains. These pendant ionic groups are accountable for the pH-responsive nature of the hydrogels. Hydrogels derived from natural or biopolymers are environmentally friendly, biodegradable, compatible with living organisms, nonharmful, highly absorbent, water-attracting, cost-efficient, and possess the capability to swell, making them advantageous for applications in fields such as biomedicine, cosmetics, biotechnology, particularly in drug delivery, and agriculture [12].

In the present day, owing to its distinctive and exceptional attributes, alginate

finds utility in a wide array of applications encompassing food, cosmetics, textiles, pharmaceuticals, and various biomedical uses [13]. Natural polymer-based hydrogels exhibit limited mechanical strength primarily due to their low crystallinity and amorphous structure. However, numerous other factors contribute to this phenomenon, including their hydrophilic nature, crosslinking density, degree of swelling, polymerization conditions, among others.

Typically, as the concentration of crosslinkers increases, polymer chains move closer to each other, reducing diffusivity and swelling behavior, leading to improved mechanical properties. Beyond the optimal crosslinker concentration, the number of crosslinks increases, causing the polymers to become less flexible (as polymer chain segments are constrained in their movement due to crosslinking), resulting in a brittle structure [14]. Moreover, to improve the mechanical characteristics of hydrogels derived from natural polymers, they are combined with synthetic polymers known for their strong mechanical properties and thermal stability [15].

Polyvinyl alcohol (PVA) is a widely utilized synthetic polymer known for its outstanding mechanical attributes and is frequently employed in the production of films [16]. Hydrogels and membranes derived from PVA exhibit remarkable features such as being non-carcinogenic, biocompatible, non-toxic, displaying significant swelling in aqueous solutions, and possessing favorable physical and thermal properties [17]. PVA is additionally employed in crafting materials for skin replacement, manufacturing contact lenses, and aiding in the reconstruction of vocal cords [11].

To enhance the interaction between natural and synthetic polymers, various crosslinking agents are accessible, such as formaldehyde, acetaldehyde, glutaraldehyde, and TEOS, among others. However, TEOS is the preferred choice as a crosslinker because silanebased crosslinkers are particularly suitable for establishing inorganic crosslinks within the amorphous regions of polymers. This facilitates the formation of covalent bonds between the inorganic components and polymer chains, making it a common choice in biomaterials and various applications. TEOS is easily bonded through condensation reactions and is considered non-toxic in comparison to previously employed crosslinkers like glutaraldehyde, epichlorhydrine, borate, and tripolyphosphate [18]. In this research, a novel pH-responsive hydrogel for controlled drug release is created by blending cationic and anionic polymers with PVA and crosslinking them using TEOS. The swelling behavior of these hydrogels is examined in various solvent environments. The results indicate that all the hydrogels exhibit their maximum swelling at an acidic pH, with reduced swelling at basic and pH 7. This unique pHsensitive response at pH 4 suggests their potential use as an injectable controlled drug delivery system. The hydrogel showing the most favorable response is the preferred choice to achieve the research objectives.

The objective of this research is to Synthesize and to increase the Mechanical Strength, Thermal Stability and Pore Size by using Cascade Method and charac-

terize by using characterization techniques like Ultimate Tensile Stress Test (UTS), Thermo Gravimetric Analysis (TGA), Scanning Electron Microscopy (SEM) and Fourier Transform Infrared Spectroscopy (FTIR) and to analyze the Swelling Study in distilled water of the Hydrogels.

2. Materials and Method

2.1. Materials

The following substances (**Table 1**) were utilized in combination of biodegradable hydrogel base food packaging films.

2.2. Preparation of pH Sensitive Hydrogel Using SILANE Cross Linker

First, 0.50 g chitosan was added into 2% acetic acid in 50 ml water stirring on hot plate at 50°C. Secondly take 0.40 g PEG6000 in 25 ml water and stirring for 1 hour at room temperature. Thirdly stirrer 0.10 g Guargum in 25 ml water on hot plate at 50°C. A filler as carbon added up 10 ml water with somicator hotplate with different amount in above mixture (PEG6000/Chitosan/guargum) to enhance the sensitivity of this hydrogel. Bio-polymer and synthetic polymer mixture (with and without filler) crosslinked with each other through TEOS (silane cross linker) 100 µl in 5 ml ethanol. They are all add one by one in large beaker and stirred still complete balancing then the solution that was prepared is poured into the petri dish of plastic. The solvent then just evaporates at room temperature. The film sample needs to have thicknesses rate of approximately 100 - 200 µm. The novelty of this work is the addition of nano-carbon as a filler which may enhance the mechanical strength for pH sensitivity.

3. Characterization of Biodegradable pH Sensitive Hydrogel

3.1. FTIR (Fourier Transform-Infrared) Analysis

SHIMADZU IR Prestige-21 was utilized to analyze the FT-IR spectroscopy of the film samples, analyzer have certain wavelength in the range of four-thousand to four hundred cm^{-1} with 4.0 cm resolving power and one hundred outputs has been collected for every film sample.

Table 1. Chemical utilized in the preparation of ph sensie hydrogel.

Sr. No.	Chemical	Company
1	Polyethylene Glycol (PEG6000)	Sigma-Aldrich
2	Chitosan	Sigma-Aldrich
3	Guargum	Sigma-Aldrich
4	Nano-carbon	Sigma-Aldrich
5	Tetraethoxysilane (TEOS)/Silane	Sigma-Aldrich
6	Ethanol	Sigma-Aldrich
7	Distilled water	Sigma-Aldrich

3.2. Swelling Studies

The swelling of biodegradable hydrogel base films evaluated in the buffer solution water.

3.3. Water Swelling Test

In buffer solution water, biodegradable hydrogel base film sample's swelling testing was examined. First of all, cut dried film sample (0.025 g pre-weighted) into small pieces from plastic petri dish and then loaded that dish with seventy millilitres buffer solution water. The abundance of water weight of the film was measured after each 10 minutes. The swollen sample then placed in water again till establishment of equilibrium occurred.

By assigned relation, the swelling ratio for samples was taken into account:

$$DS(\%) = [s - W_d / W_d] \times 100 \quad (1)$$

Here,

W_s = swollen weight of film

W_d = dry weight of the film

3.4. In Buffer Solution

In buffer solution (2, 4, 6, 8, 10) biodegradable hydrogel base film sample's swelling testing was examined.

3.5. In Saline Water

In NaCl solution biodegradable hydrogel base film sample's swelling testing was examined.

3.5.1. Thermal Gravimetric Analysis (TGA)

TGA showed the films thermal stability. Thermal de-compositional activity of samples was investigated through thermogravimetric (TG) in air and heating rate of ten degrees.

Celsius per minute on Shimadzu TGA instrument at oxygen environment.

3.5.2. UTS

ASTM D882-91 analyzer examine the sample's ultimate tensile strength (UTS). Slice of sample taken in segments around $1/2 \times 5/2$ inches. The gage length (1.60 inch) as well as speed of cross-head 5.0 mm/min was set. Ultimate tensile strength was calculated in MPa simply through dividing the force (F) to split the film over the film sample's area (A). It also measured the percentage elongation at break (percent E) and estimated the average of at least three UTS determinations and percentage E.

3.5.3. Scanning Electron Microscope (SEM)

Scanning electron microscope is performed to investigate the surface morphology porosity of design hydrogel for controlled pH-sensitive study Joel-Japan scanning electron microscope is used to analyses the resultant hydrogel.

4. Experimental Design

4.1. FTIR (Fourier Transform Infra-Red) Analysis

The study of Fourier transforms infrared (FTIR) has been used to investigate the either recognized or unidentified structure of molecules of the various compounds. **Figure 1** is shown the FTIR spectra of designed hydrogel. Because of all the specific functional groups the polymers (chitosan, Gurgam and PEG6000 along with silane coupling agent) interact and establish new bonds. Spectra of FTIR reflected both established and newly developed groups (like hydrogel). COOH group presence is clearly observed in this range in the carbonfiller reinforced hydrogel [19].

4.2. Thermogravimetric Analysis (TGA)

Thermal decomposition of the fabricated hydrogel nanocomposites in the temperature range 25°C to 550°C is depicted in **Figure 2**. The thermal degradation is followed in four steps. First step the due to the evaporation of water content. Second step is owing to the scoop out of the low molecular weight gases. Third step had the major degradation and polymer pyrolysis was that reason. Fourth step is the thermal endurance of nanofiller. And finally, the fifth step is due to effect of remaining residuals. The increase concentration of nanofiller enhanced the thermal stability of the designed hydrogels [20].

4.3. UTS

Ultimate tensile strength and elongation at break is used to decide the usage of designed hydrogel. The UTS and Elongation at break is plotted in **Figure 3**. **Figure 3** elucidated the enhancement in both UTS and elongation at break with increasing concentration of nanofiller. The reason for this strength of hydrogel is due to the nanoscale interaction of nanofiller and polymeric chain. This revealed that with the inclusion of the filler, the greater force needed to break the film of hydrogel [21].

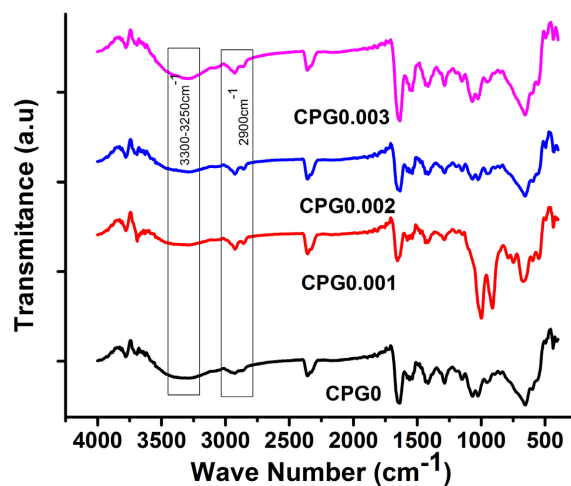


Figure 1. FTIR spectra of Carbon nanofiller reinforced designed Hydrogel.

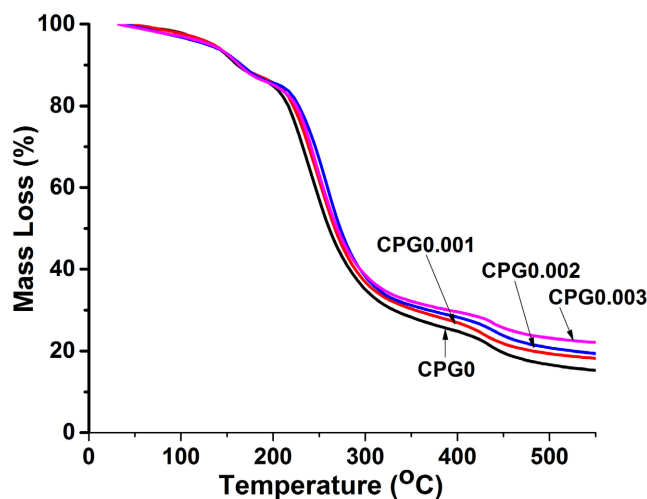


Figure 2. TGA thermograms of designed hydrogels with varying concentration of carbon nanofiller.

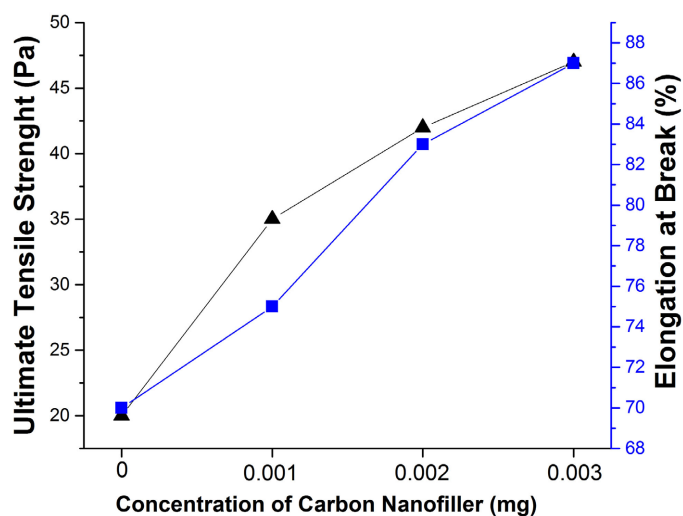


Figure 3. Ultimate tensile strength and elongation at break of designed hydrogels.

5. Surface Morphology

The scanning electron microscopy is used to investigate the surface morphology and structure of the designed hydrogels. SEM micrographs of resultant hydrogel are depicted in **Figure 4(a)**. The controlled designed hydrogel CPG0 without nano reinforcement was smooth and plain with no visible pores in **Figure 4(b)**. With the addition of nanofiller CPG0.001, CPG0.002, and CPG0.003 surface morphology of the formulated hydrogel with clear formation of prominent pores. The SEM micrographs CPG0.003 show that had a collection of polysaccharide chains as thin helices, which is attributed to increase the size of porosity. The easy expansion of designed hydrogel is owing to physical entanglement of polymeric chain with coupling agent and nanofiller reinforcement contribute to the diffusion of water into the pores [22].

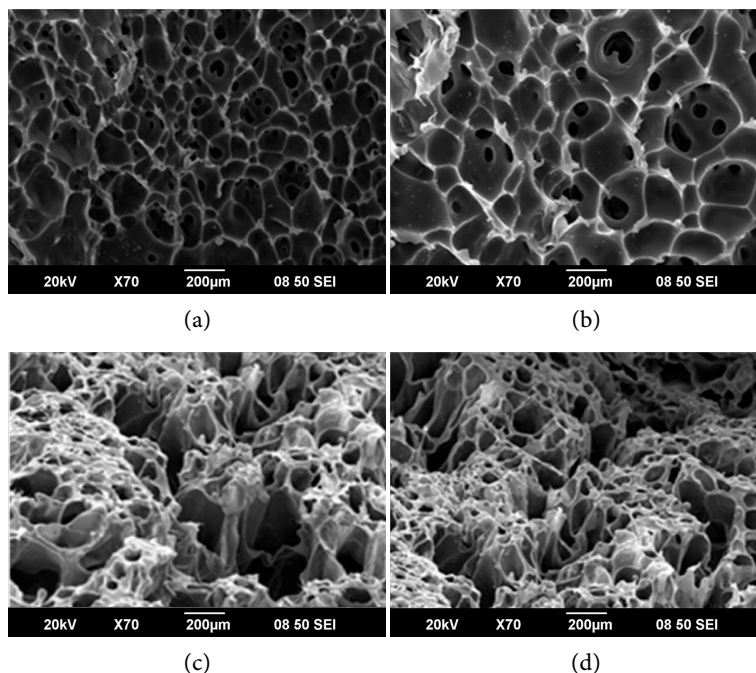


Figure 4. (a) SEM micrographs of ZnO, (b) PEG4000-Alginate, and designed hydrogel with various concentration of carbon nanofiller (c, d).

Swelling Studies

Usually, the swelling trend not only depend on the polymer and also the nanoreinforcemnt. But also affected with medium in which the hydrogel has to be swell. Normally, the swelling test must be done in different pH value that may help to decide the usage of the designed hydrogels. **Figure 5** cleared the check the effect of pH on the hydrogels, buffer solutions of various pH (2, 4, 6, 8 and 10) were used. It is cleared from the swelling contours that the maximum swelling was obtained in acidic medium and low at the basic medium. But with the increase concentration of nanofiller enhanced the swelling in acidic and basic medium. So, it is concluded that the obtained trend augmented in acidic pendent groups in polymeric structure due to the osmosis and repulsion of charges. And the designed hydrogel is cation due to the chitosan and mostly swell in acidic medium. The predominantly NH_3^+ groups having cationic charge and the pendant groups protonation occurred resulting in an increased number of positive charges on the chains of polymer. This pH dependent swelling response of hydrogels Can be exploited for drug release study.

Various ionic concentration are also effect the swelling behavior of the designed hydrogels. This is due to the osmotic pressure and repulsion between the polymeric chains. For this reason support to calculate the swelling in NaCl solution. The swelling trend in ionic solution is graphed in **Figure 6**. It was seen that the swelling of hydrogels was reduced with the increased amount of the electrolyte which could be due to the enhanced quantity of salt in the solvent. This is due to the more charge screening resulted in less osmotic pressure and thus diffusion was decreased and so the swelling. So, higher value of swelling was shown

in lower strength solutions. But, the interesting trend was observed is that with increasing nanofiller concentration have less swelling in ionic solution [23].

The swelling response with respect to time is reported in **Figure 7** of CPG hydrogels.

The designed hydrogels have different response with water interaction. The contours of swelling with time is enhanced with the rise of time upto 60 minutes. Each hydrogels have different equilibrium point at various time. The CPG0 have different pattern of change than other sample because it was without nano-carbon filler. It was confirmed from the graph of swelling with water as time dependent that the neat hydrogel had low swelling than the reinforced hydrogels. Because, the nanoscale interaction of nanofiller with the polymeric chains augmented the strength of designed hydrogel and the capability to more intake of water in the porous structure [24].

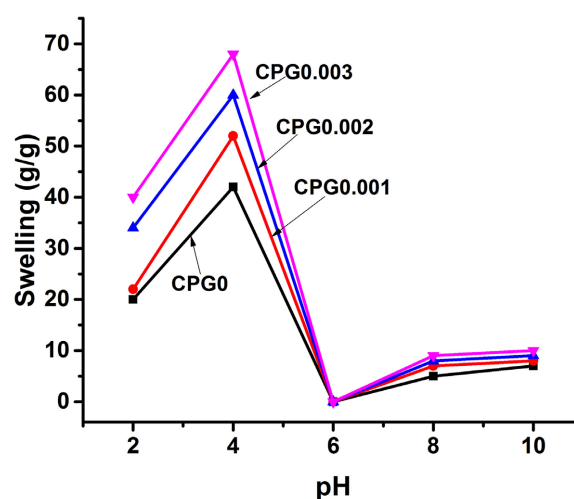


Figure 5. Swelling contours with various pH of designed hydrogels (2-10).

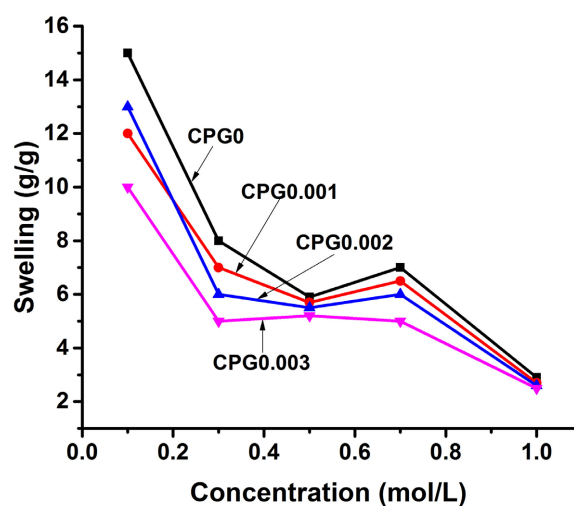


Figure 6. Swelling contours with different molar concentrations of NaCl of designed hydrogels.

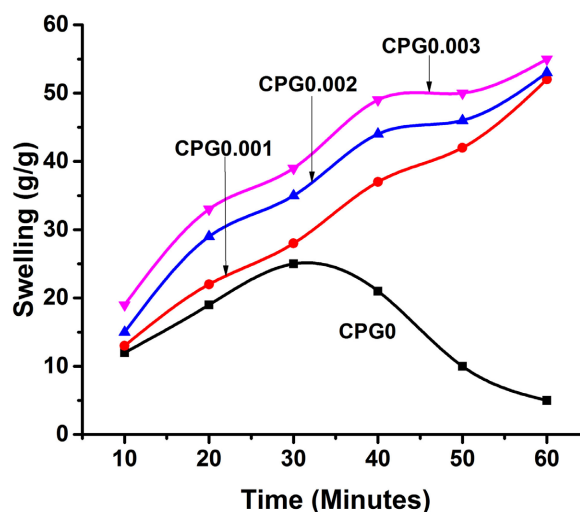


Figure 7. Swelling contours with time of designed hydrogels in water.

6. Conclusion

Novel designed biopolymers/synthetic polymer, crosslinked, and reinforced pH sensitive CPG hydrogels were successfully prepared using solution casting method for drug delivery applications. Scanning electron microscope, Fourier transform Infrared spectroscopy, Thermogravimetric analysis, universal testing machine and swelling at time dependent, ionic and pH level is used to investigate the fabricated hydrogels. The maximum thermal stability was observed with maximum concentration of nanofiller and augmented mechanical properties. The designed hydrogel has prominent porosity and good UTS. The swelling behavior with time is enhanced with the increase passage of time with the utmost concentration of nanofiller. The swelling response of the hydrogels at various pH showed extreme swelling in the acidic pH while low in basic and neutral. This behavior was the significant factor that these pH sensitive hydrogels are suitable for the injectable controlled release. This release confirmed the success of these hydrogels as a remarkable option for the biomedical applications beneficially for injectable drug release and for wound healing/dressings, tissue engineering scaffolds, etc.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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