

A REPORT ON  
**SYNTHESIS AND CHARACTERIZATION OF HYDROGELS BASED  
ON CELLULOSE DERIVATIVES AND TANNIC ACID**

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**SUBMITTED BY: ASHWANI KUMAR TIWARI**

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UNDER THE SUPERVISION OF

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# DELHI TECHNOLOGICAL UNIVERSITY

(Formerly Delhi College of Engineering)

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## DECLARATION

I hereby announce that the work introduced in this significant undertaking report entitled "**SYNTHESIS AND CHARACTERIZATION OF HYDROGELS BASED ON CELLULOSE DERIVATIVES AND TANNIC ACID**" is unique and has been completed by me in the halfway satisfaction of the necessity for the honor of the Master of Technology in Polymer Technology in the Department of Applied Chemistry, Delhi Technological University, Delhi – 110042, under the oversight of **Prof. Archna Rani and Prof. Rajinder K. Gupta**. This report is commitment of my unique examination work. Any place research commitments of others are included, each exertion has been made to unmistakably demonstrate the equivalent. As far as I could possibly know, this examination work has not been submitted to some extent or full for the honor of any degree or confirmation of Delhi Technological University or other University.

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## CERTIFICATE

This is to ensure that the work introduced in this significant task report entitled "**SYNTHESIS AND CHARACTERIZATION OF HYDROGELS BASED ON CELLULOSE DERIVATIVES AND TANNIC ACID**" has been submitted to the Delhi Technological University, Delhi-110042, in satisfaction for the prerequisite for the honor of the level of **M.Tech in Polymer Technology** by the competitor **Ashwani Kumar Tiwari (2K18/PTE/02)** under the oversight of **Prof. Archna Rani and Prof. Rajinder K. Gupta**, Department of Applied Chemistry. It is additionally confirmed that the work typified in this report has neither in part nor completely submitted to some other college or foundation for the honor of any degree or confirmation.

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**DATE:**

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**(SUPERVISOR)**

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## **TABLE OF CONTENT**

<b>S.NO.</b>	<b>TITLE</b>	<b>PAGE NO.</b>
1	INTRODUCTION AND OBJECTIVE	11-15
2	LITERATURE REVIEW	16-20
3	EXPERIMENTAL WORK	21-26
4	RESULT AND DISCUSSION	26-41
5	CONCLUSION	41-42
6	REFERENCES	43-47

## **LIST OF TABLES**

<b>SR. NO.</b>	<b>TITLE</b>	<b>PAGE NO.</b>
Table 1	List of abbreviations	9
Table 2	Factors effecting hydrogel properties	19
Table 3	Specifications and sources of raw materials	21-22

## **LIST OF FIGURES**

<b>SR. NO</b>	<b>TITLE</b>	<b>PAGE NO</b>
Figure 3.1	Pictorial representation of complete procedure	23
Figure 3.2	FTIR Instrument	24
Figure 3.3	SEM Instrument	25
Figure 3.4	TGA Instrument	25
Figure 3.5	XRD Instrument	26
Figure 4.1	TGA of CMCNa-AAm hydrogel	27
Figure 4.2	TGA of TA-AAm hydrogel	28
Figure 4.3	TGA of HPMC-AAm hydrogel	28
Figure 4.4	TGA of MC-AAm hydrogel	29
Figure 4.5	FTIR of CMCNa-AAm hydrogel	31
Figure 4.6	FTIR of TA-AAm hydrogel	31
Figure 4.7	FTIR of HPMC-AAm hydrogel	32
Figure 4.8	FTIR of MC-AAm hydrogel	32
Figure 4.9	Swelling behaviour of CMCNa-AAm and TA-AAm hydrogel	33
Figure 4.10	Swelling behaviour of HPMC-AAm and MC-AAm hydrogel	33
Figure 4.11	Swelling studies of CMCNa-AAm hydrogel	34
Figure 4.12	Swelling studies of TA-AAm hydrogel	34
Figure 4.13	Swelling studies of HPMC-AAm hydrogel	35
Figure 4.14	Swelling studies of MC-AAm hydrogel	35

Figure 4.15	SEM of CMCNa-AAm hydrogel	36
Figure 4.16	SEM of TA-AAm hydrogel	37
Figure 4.17	SEM of HPMC-AAm hydrogel	37
Figure 4.18	SEM of MC-AAm hydrogel	38
Figure 4.19	XRD of CMCNa-AAm hydrogel	39
Figure 4.20	XRD of TA-AAm hydrogel	40
Figure 4.21	XRD of HPMC-AAm hydrogel	40
Figure 4.22	XRD of MC-AAm hydrogel	41



## **ABBREVIATIONS**

CMCNa	Carboxy methyl cellulose sodium salt
HPMC	Hydroxy propyl methyl cellulose
MC	Methyl cellulose
TA	Tannic acid
TGA	Thermogravimetric Analysis
FTIR	Fourier-Transformed Infrared spectroscopy
SEM	Scanning Electron Microscopy
XRD	X-Ray Diffraction
AAm	Acrylamide
MBA	N,N'-methylenebisacrylamide
KPS	Potassium per sulphate

Table 1. List of abbreviations used

## **ABSTRACT**

Hydrogels were synthesized utilizing, Carboxy methyl cellulose sodium salt, Hydroxy propyl methyl cellulose, Methyl cellulose and Tannic acid. The methodology incorporates free radical polymerization strategy with acrylamide (AAM), N,N'-Methylenebis-acrylamide (MBA) as a crosslinker and Potassium Persulfate (KPS) as an initiator. The synthesis was evident with the help of characterization using Thermogravimetric analysis for thermal analysis, Fourier-transform infrared spectroscopy for detecting the functional groups present and linkages formed , Scanning Electron Microscopy for surface morphologies , X-Ray diffraction for investigating the crystalline or amorphous nature and swelling studies in buffer solutions of pH 4.0, pH 7.4, pH 9.2 and distilled water for investigating the absorption characteristics.

## 1. INTRODUCTION

Hydrogels are cross-linked polymeric materials or networks with absorption and swelling tendency upto several times of their complete dried weight [1][2]. Due to the presence of physical or chemical crosslinks between the polymeric chains, hydrogels are insoluble in aqueous solvents. As the crosslinkers are the agents that binds the polymeric chains, their crosslink density have great influence on porosity and absorption capacity of hydrogels. They can be used in various applications like agriculture [3], drug delivery system [4], tissue engineering [5], regenerative medicines and wound dressing application [6]. Their super- absorptive networks can be synthesized by using synthetic or natural polymers or by combination of both. However, natural polymers based hydrogels are getting more and more attention due to their environmental friendly nature, biocompatibility and biodegradability. Recently, natural polymers like starch [7], cellulose [8], chitosan [9], gums like guar gum [10], gellan gum [11], and xanthan gum [12] gain attention due to their biodegradability and biocompatibility. Cellulose is the most abundant polysaccharide naturally available on earth. It is regular and linear polymer composed of 1-4 linked  $\beta$ -D-glucopyranosyl units. The most commercially used cellulose derivatives are methyl cellulose (MC), ethyl cellulose (EC), carboxy methyl cellulose (CMC), hydroxy propyl cellulose (HPC), and hydroxy propyl methyl cellulose (HPMC) that can be used to synthesize super-absorptive molecules either through physical cross-linking method or by chemical cross-linking method.

Carboxy methyl cellulose is a biodegradable polymer which is derived from cellulose by its etherification. It is hydrophilic in nature. Carboxy methyl cellulose (CMC) found its applications in various industries like paper, food, cosmetics, pharmaceuticals and textiles. Recently radiation induced cross-linking method [13][14], chemical cross-linking method [15][16] and electron beam irradiation method [17] have been used for the preparation of carboxy methyl cellulose sodium salt (CMCNa) hydrogels. The

insertion of synthetic polymers like acrylic acid (AA) or acrylamide (AAm) improves the water absorption capacity of these hydrogels.

Hydroxy propyl methyl cellulose (HPMC) and methyl cellulose (MC) are also water soluble cellulose derivatives. The main characteristics of HPMC and MC are that they are non-toxic in nature, biocompatible and low cost materials which makes them suitable for hydrogels preparation especially for controlled release. HPMC and MC can also be used as adhesives, emulsifiers and thickeners in food industry.

Tannins are natural biomass containing multiple phenolic hydroxyls with anti-bacterial and anti-oxidant property. The commercial modification of tannin that is tannic acid can also be used as a material for synthesizing hydrogels [18]. Tannic acid can also be extracted from various plants. The molecular structure of tannic acid comprises of pyrogallol and catechol groups. The presences of these groups provide multiple bonding sites. They also have specific affinity to metal ions. Hydrogels from these natural polymers generally possess poor mechanical strength when used alone due to formation of intramolecular cross-linking. Therefore other bifunctional polymeric materials can be used in combination with these biodegradable polymeric materials to improve intermolecular cross-linking.

Acrylamide (AAm) is an organic compound which is generally prepared by the hydrolysis of acrylonitrile catalyzed by sulphuric acid and other metals. It is generally odourless, white in color and water soluble in nature. The acrylamide can be polymerized into polyacrylamide by free radical polymerization by using initiators like benzoyl peroxide, potassium per sulphate. Polyacrylamide is widely used as a thickener and suspending agent. From past few decades polyacrylamide based hydrogels got attention of many researchers. Hydrogels based on polyacrylamide doped with  $H_3PO_4$  were synthesized for the study of proton transport. Hydrogels

based on acrylamide and pH sensitive psyllium were synthesized for drug delivery application.

In the present work, we have prepared the hydrogels based on cellulose derivatives and acrylamide namely methyl cellulose–acrylamide (MC-AAm) hydrogel, carboxy methyl cellulose-acrylamide (CMCNa-AAm) hydrogel, hydroxy propyl methyl cellulose-acrylamide (HPMC-AAm) hydrogel and tannic acid-acrylamide (TA-AAm) hydrogel. The main objective of this work is to characterize the material properties of hydrogels synthesized from different cellulosic derivatives and tannic acid.

## **MOTIVATION FOR RESEARCH**

Earth has provided humans with beauty of natural resources in abundance. Cellulose is one of them which is present in our nature in abundant quantity. From early ages humanity has utilized natural materials, yet today the situation has changed and manufactured materials have supplanted normal ones causing the disturbing circumstance in the whole world because of its non-biodegradable nature and wrong arrangements. To keep up an eco-friendly domain, manufactured polymers have a dire need to get supplanted from natural ones as issues of plastic contamination, air contamination, a dangerous atmospheric deviation, ozone consumption is making a danger to the earth just as human wellbeing. Consequently it is basic to use normal substances as crude materials for various applications to limit ecological issues.

## **OBJECTIVE**

Preparation and characterization of carboxy methyl cellulose, tannic acid, hydroxyl propyl methyl cellulose and methyl cellulose based hydrogels

## **SPECIFIC OBJECTIVE**

The specific objectives which are required to achieve the above objective are :

- Preparation of (MC-AAm) hydrogel, (CMCNa-AAm) hydrogel, (HPMC-AAm) hydrogel and (TA-AAm) hydrogel
- Characterization of the hydrogels using
  - TGA
  - FTIR
  - SEM
  - XRD
  - Swelling Studies

## **2. LITERATURE REVIEW**

A hydrogel is a cross-linked polymeric framework basically shaping one nonstop system and this system can retain dissolvable without harm to the system. Hydrogels in pharmaceutical applications have gotten well known as of late in view of their biocompatibility. Pharmaceutical hydrogel frameworks incorporate grids that have a medication consolidated into them and are swollen to balance. Natural hydrogels can be found in tissues, ligaments, and films for the kidneys and veins. They are likewise found on the outside of the stomach, the digestive organs, and the lungs. In the previous decades numerous engineered hydrogels have been created for an enormous assortment of uses. Manufactured hydrogels are blends that comprise of a considerable measure of water and profoundly swollen, hydrophilic polymer systems. Manufactured hydrogel frameworks have been created utilizing different polymerization methods or by changing over existing polymers. Union can be acted in arrangement, a suspension or emulsion, gas stage, or by plasma or mass polymerization. Every one of these techniques has favorable circumstances or hindrances as for immaculateness, atomic weight conveyance and system homogeneity. Hydrogels can be described by hydrophilicity and insolubility in water. In water they may ingest up to multiple times, or more, their own weight while holding their shape and mechanical soundness. The hydrophilicity is because of the nearness of water-solubilizing gatherings, for example, - OH, - COOH, CONH<sub>2</sub>, and SO<sub>3</sub>H. The insolubility and strength of shape are because of the nearness of a three dimensional network. The swollen state results from a harmony between the scattering powers following up on hydrated chains and strong powers that don't forestall the entrance of water into the system. Durable powers are regularly because of covalent cross-connecting yet can likewise be affected by electrostatic, hydrophobic, or dipole-dipole character.



## **2.1 HYDROGEL SYNTHESIS**

### **2.1.1 CHEMICAL METHOD**

There are two distinct strategies in synthetically creating hydrogel networks. Right off the bat, straight polymers can be set up by straight forward crosslinking between monomers. Furthermore, three dimensional system of hydrogels is set up by free extreme polymerization among monomer and cross connecting specialist. The Synthesis of hydrogels can be acted in arrangement, a suspension or emulsion, gas stage, or by plasma or mass polymerization. In any case, arrangement and suspension polymerization are the most much of the time utilized strategy. Polymerization in arrangement can deliver mass and different states of hydrogel while the polymerization in suspension give little and circularly molded hydrogels. The plan and advancement of natural polymer hydrogels with ideal compound and physical properties is of foremost significance in clinical, biomedical, pharmaceutical and modern field.

### **2.1.2. ELECTROCHEMICAL METHOD**

Electrochemical polymerization of vinyl polymers in arrangement has been broadly concentrated under various test conditions. One of the created preparing procedures for combining polymer gels as covering materials is the utilization of electrochemical polymerization (ECP) which utilizes an electrical flow to start polymerization forms. As of late electrochemical polymerization (ECP) technique has been widely read for applying polymer coatings to metals. ECP includes the immediate coupling of atoms by anode responses. By and large, gel arrangement oa cathode is subject to the sort of supporting electrolytes/monomer focus and electric charge. Polymerization is started by the dynamic species (free radical or

particles) created by electrochemical response of the supporting electrolyte or dissolvable. Acrylamide has been polymerised by electrochemical polymerization in watery and natural solvents. There are three substance/electrochemical advances engaged with the polymerization of acrylamide (I) electro-inception, (ii) engendering or chain development and (iii) ending of chains or termination.

## **2.2. PHASE TRANSITION OF GELS**

The standards of stage change in polymer gels are fundamental for the comprehension of polymer gel frameworks when all is said in done. The reaction capacity of improvements responsive hydrogels depends on stage advances because of changing outside conditions. It is seen that the impact of gel charge (which can be presented by halfway hydrolysis of the amide side gathering) on the stage conduct is a simple of the impact of weight on the fume fluid stage conduct. For the gels, charge creates an osmotic growing weight, which is inverse to an outer weight. To comprehend the stage conduct of gels, it is basic to consider the sub-atomic associations between the polymer chains in the gel. Gel phasetransition is an outcome from a serious harmony between an unpleasant power that demonstrations to grow the polymer organize and an appealing power that demonstrations to contract the system. The best horrible power is the electrostatic collaboration between the polymer charges of a similar kind, which can be forced upon a gel by bringing ionization into the system. The osmotic weight by counterions adds to the growing weight. The alluring communications can be Van der Waals, hydrophobic cooperation, particle with inverse signs and hydrogen holding. All in all the Flory-Huggins hypothesis has been effective in depicting subjectively the change conduct of gels. In any case, while the Flory Huggins hypothesis gives the hypothetical comprehension of the stage change conduct of gels, these communications give an increasingly intensive comprehension of the stage progress of gels.

FACTORS	HYDROGEL PROPERTIES
Charge increases	Swelling increases , Charge solute binding capacity increases
Ionization increases	Swelling increases
Crosslinking density increases	Swelling decreases, permeability decreases, Elastic modulus and strength increases
Hydrophilicity increases	Swelling increases
Hydrophobicity increases	Swelling decreases
Monomer strength increases	Stimulus sensitivity decreases
Free water increases	Permeability increases

Table 2. Factors effecting hydrogel properties

Carboxy methyl cellulose sodium salt (CMCNa), acquired from the response of the hydroxyl gatherings of the anhydroglucose units (AGUs) of cellulose with chloroacetic corrosive, is a significant water solvent cellulose ether utilized in food, beauty care products, and paints as a thickness modifier, thickener, emulsion stabilizer, and water-maintenance operator. CMC likewise has gigantic potential for use in pharmaceutical items including site-explicit or controlled-discharge sedate conveyance framework transporter grids because of its high biocompatibility, biodegradability, and low immunogenicity [19,20]. Crosslinked CMC has additionally been practiced with the utilization of bifunctional crosslinking specialists, for example, epichlorohydrin [21], diepoxy [22-24] and dicarboxylic corrosive mixes [25]. Crosslinked CMC for the most part ingests a lot of water, and swells to frame hydrogels with superb physical properties and dynamic viscoelasticities [26]. These CMC-based hydrogels were as of late explored for their utilization in wound dressing, medicate conveyance, agribusiness, and clean cushions, just as for trans-dermal frameworks, dental materials, inserts, injectable polymeric frameworks, ophthalmic applications, and half breed type organs [27,28]

Hydroxy propyl methylcellulose (HPMC) is the most significant hydrophilic bearer material utilized for the arrangement of oral controlled medication conveyance frameworks [29,30]. One of its most significant qualities is the high swellability, which significantly affects the discharge energy of a joined medication. Upon contact with water or natural liquid the last diffuses into the gadget, bringing about polymer chain unwinding with volume development [31,32]. At that point the consolidated medication diffuses out of the framework. HPMC is water dissolvable cellulose subsidiary. It is hydrophilic, biodegradable and biocompatible polymer. It merits featuring that HPMC is non harmful, minimal effort and utilized in the definition of hydrogel for medicate conveyance [33]. HPMC has been effectively presented as a rate controlling polymer in strong scattering of various medications [34-37]. Nochos and Bouropoulos [38] arranged ox-like serum egg whites (BSA) stacked alginate/HPMC hydrogel dabs to contemplate the impact of various alginate/HPMC plans on discharge conduct of BSA.

Methylcellulose (MC), a characteristic polyhydroxyl watersoluble sugar polymer, can be artificially crosslinked with dialdehyde within the sight of a solid corrosive to produce a hydrogel [39,40]. In any case, hydrogels dependent on MC have poor mechanical properties that limit their applications. The combination of MC hydrogels inside a framework with great mechanical properties is a choice to limit this impediment. For this situation, two kinds of hydrogels can be acquired: interpenetrating system (IPN) hydrogels or semi-interpenetrating system (semi-IPN) hydrogels. IPN hydrogels comprise of two freely crosslinked systems that are interlocked [41]. On the off chance that just a single polymer is crosslinked, this kind of system arrangement is known as a semi-IPN [42]. During the most recent decade, specific intrigue has been centered around the semi-IPN gels, particularly with one segment containing characteristic polysaccharides [43-45].

### **3. EXPERIMENTAL WORK**

#### **3.1. MATERIALS**

- Carboxy methyl cellulose Sodium salt
- Hydroxy propyl methyl cellulose
- Methyl cellulose
- Tannic acid
- Potassium per sulphate
- N,N'- methylenebisacrylamide
- Acrylamide
- Buffer solutions of pH 4.0, pH 7.4, pH 9.2
- Distilled water

#### **3.2. SPECIFICATIONS AND SOURCES OF RAW MATERIALS/CHEMICALS**

<b>Sr. No.</b>	<b>Chemical</b>	<b>Source</b>	<b>Specifications</b>
1	Carboxy methyl cellulose sodium salt	Central Drug House ltd. (CDH), New Delhi, India	viscosity of 1500±400 cPs and degree of substitution >0.4
3	Hydroxy propyl methyl cellulose	Central Drug House ltd. (CDH), New Delhi, India	viscosity of 3000 cPs+
4	Methyl cellulose	Central Drug House ltd. (CDH), New Delhi, India	viscosity 350-500 cPs and methoxyl value of 26-32%
5	Tannic Acid	Central Drug House ltd. (CDH), New Delhi, India	molecular weight of 1701.23 g/mol

6	Potassium per sulphate	Fischer Scientific, Mumbai, India	Industrial as well as Laboratory Reagent Grade
7	N,N'-methylene bisacrylamide	Merck, Germany	Industrial as well as Laboratory Reagent Grade
8	Acrylamide	Central Drug House ltd. (CDH), New Delhi, India	Industrial as well as Laboratory Reagent Grade
9	Buffer solutions of pH 4.0, pH 7.4 and pH 9.2	Fischer Scientific, Mumbai, India	Industrial as well as Laboratory Reagent Grade

Table 3: Specifications and sources of raw materials

### 3.2. APPARATUS

- Beaker & glass rod
- Test tubes
- Magnetic stirrer
- Water bath

### 3.3. SYNTHESIS OF HRDROGEL

The hydrogels were prepared by cross-linking cellulose derivatives with polyacrylamide by free radical graft copolymerization method using potassium per sulphate as an initiator and N,N'-methylenebisacrylamide as a cross-linking agent. The typical procedure involves the complete dissolution of desired amount of carboxy methyl cellulose sodium salt (CMCNa) in 20 ml of distilled water using magnetic stirrer at room temperature. Next to this, calculated amount of acrylamide (AAm),

initiator potassium per sulphate (KPS) and crosslinker N,N'-methylenebisacrylamide (MBA) were added and the mixture is stirred continuously. After 2 hours, the resulting mixtures were poured in test tubes and kept for curing for 2 hours in hot water bath maintained at 60 °C. The hydrogels prepared were then cut into thin slices and immersed in distilled water to remove unreacted chemicals and glass impurities that stuck while taking out hydrogel from test tubes. Finally, the soaked hydrogels were air dried and then oven dried at 50 °C.

The same methodology were also followed for preparing methyl cellulose-acrylamide (MC-AAm) hydrogel, hydroxy propyl methyl cellulose-acrylamide (HPMC-AAm) hydrogel and tannic acid-acrylamide (TA-AAm) hydrogel.

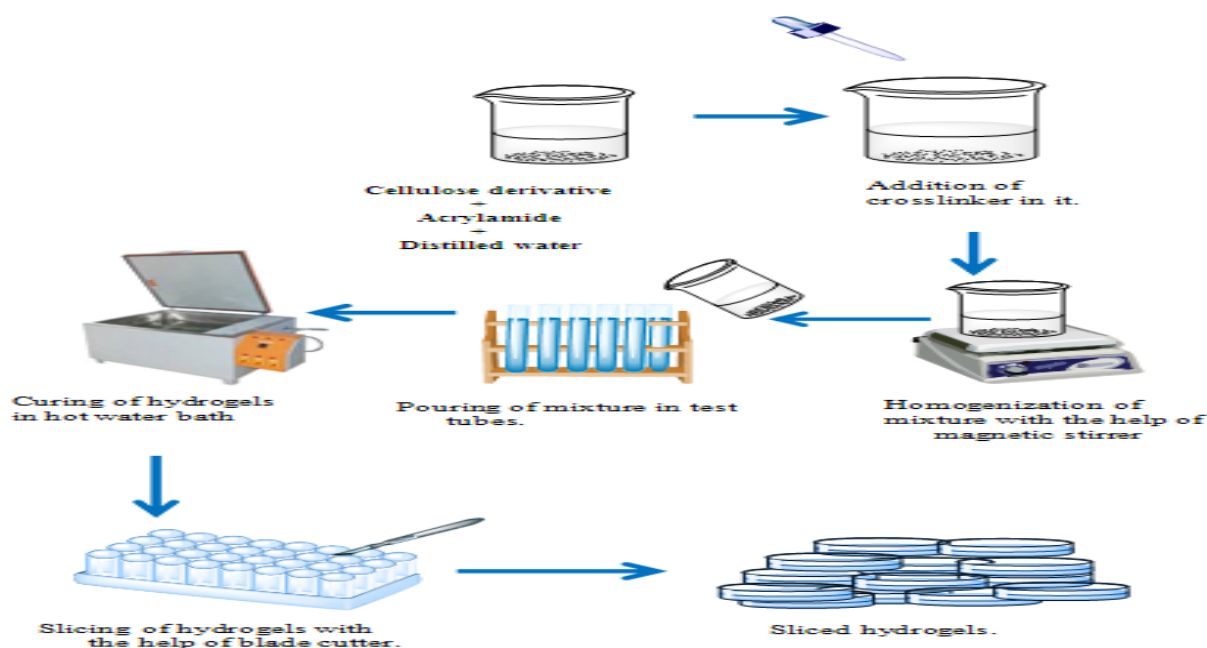


Figure 3.1: Pictorial representation of complete process

### **3.4. SWELLING STUDIES**

The gravimetric analysis method was used for swelling behavior analysis. The hydrogels were completely dried and weighed precisely and then immersed in distilled water, buffer of pH 4, pH 7.2 and pH 9.2. Accurate readings were taken at the interval of every 1 hour.

$$\text{Swelling index} = \frac{(W_s - W_d)}{W_d}$$

where  $W_s$  is the weight of swollen hydrogel and  $W_d$  is the weight of initial completely dried weight.

### **3.5. CHARACTERIZATION**

#### **3.5.1. FOURIER TRANSFORMED INFRARED SPECTROSCOPY (FTIR)**

FTIR spectra were recorded using FTIR spectrophotometer (Perkin Elmer Spectrum version 10.5.3, DTU, Delhi) by KBr pelleting method in the frequency range of 4000-500  $\text{cm}^{-1}$ .



Figure 3.2: FTIR Instrument



### 3.5.2. SCANNING ELECTRON MICROSCOPE (SEM)

The surface were observed using scanning electron microscope (ZEISS EVO 50). A thin gold coating was applied over the surface of sample using sputter before analysis.



Figure 3.3: SEM Instrument

### 3.5.3. THERMO-GRAVIMETRIC ANALYSIS (TGA)

The TGA of cellulose derivatives, tannic acid and prepared hydrogels were done by TGA analyzer (Perkin Elmer) in the atmosphere of  $N_2$  from a temperature range of 0-800 °C with heating rate of 10 °C.

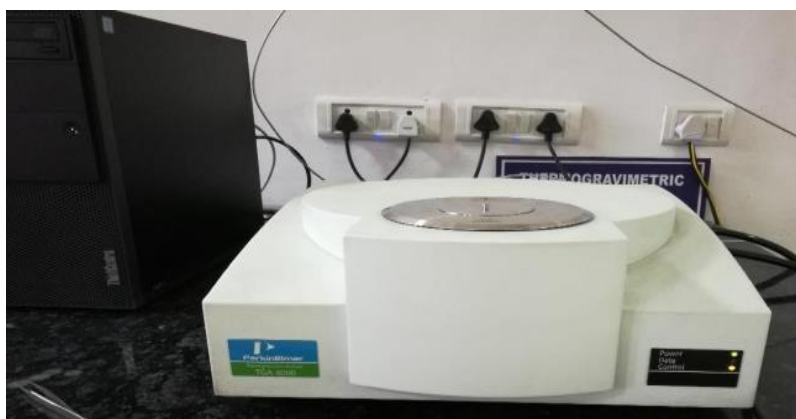


Figure 3.4: TGA Instrument

#### **2.5.4. X-RAY DIFFRACTION (X-RD)**

The X-Ray diffractometer (D8 Advance, Bruker) was used to record XRD pattern of the samples using CU-K $\alpha$  radiation ( $\lambda=0.15418$  nm) as X-ray source at 40 kV of accelerating voltage and 40mA current. The scanning speed was maintained at 2°/min in the region of  $2\theta$  from 2° to 60°.



Figure 3.5: XRD Instrument

## **4. RESULTS AND DISCUSSIONS**

### **4.1. THERMO-GRAVIMETRIC ANALYSIS**

TGA analyzer (Perkin Elmer) in the atmosphere of N<sub>2</sub> from a temperature range of 0-800 °C with heating rate of 10 °C were used to check the stability of hydrogels

prepared.. In CMCNa, weight loss was lower in the initial period (90-240°C) which could be associated due to loss of moisture from the hydrogel. The second phase (241-326°C) showed major weight loss that might be due to the degradation of the main chain constituents. The third transition was seen from (327-626°C) which could be responsible due to further decomposition of glycosidic linkages in it. Similarly, TA-AAm showed multiphase decomposition from the ranges (120-243°C), second range (243-°386C) and third range (386-780°C)with highest degradation in second phase. In HPMC-AAm, a three-phase decomposition pattern was observed as for the initial stage (226-328°C), second stage (328-496°C), third stage (496-794°C) with maximum degradation rate in second range. In MC-AAm, the initial period (80-120°C), the second stage (121-226°C), the third stage (226-496°C) and finally from 496-780°C.

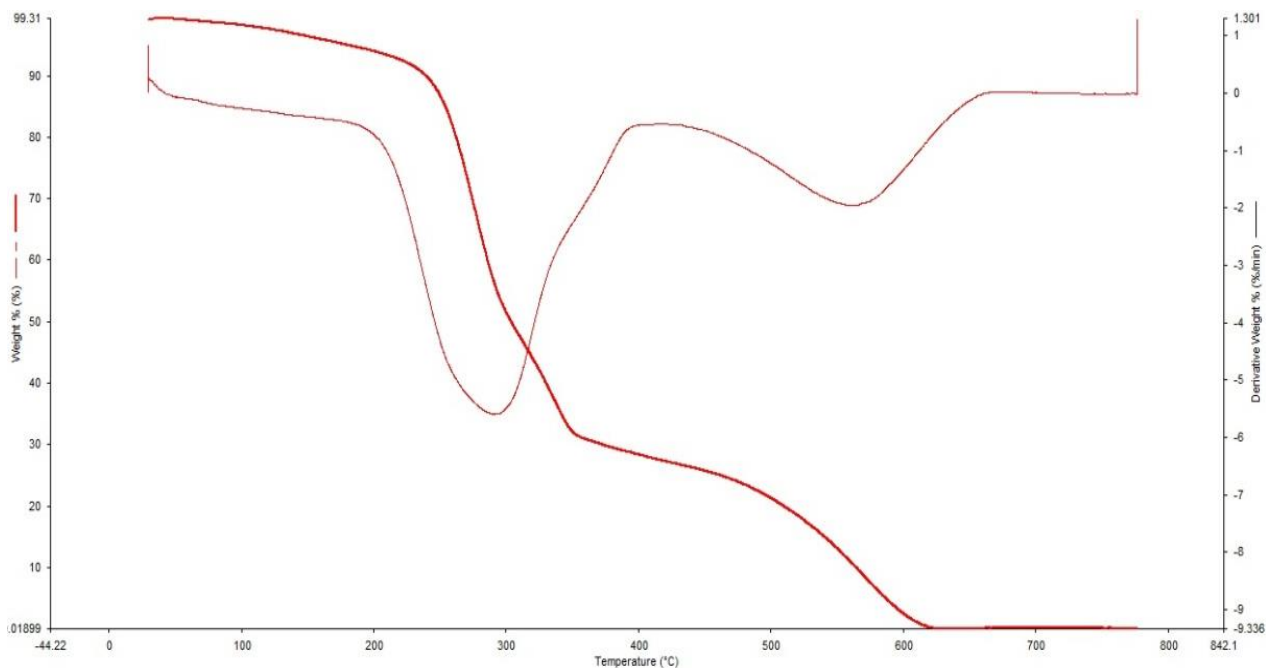


Figure 4.1: TGA of CMCNa-AAm hydrog

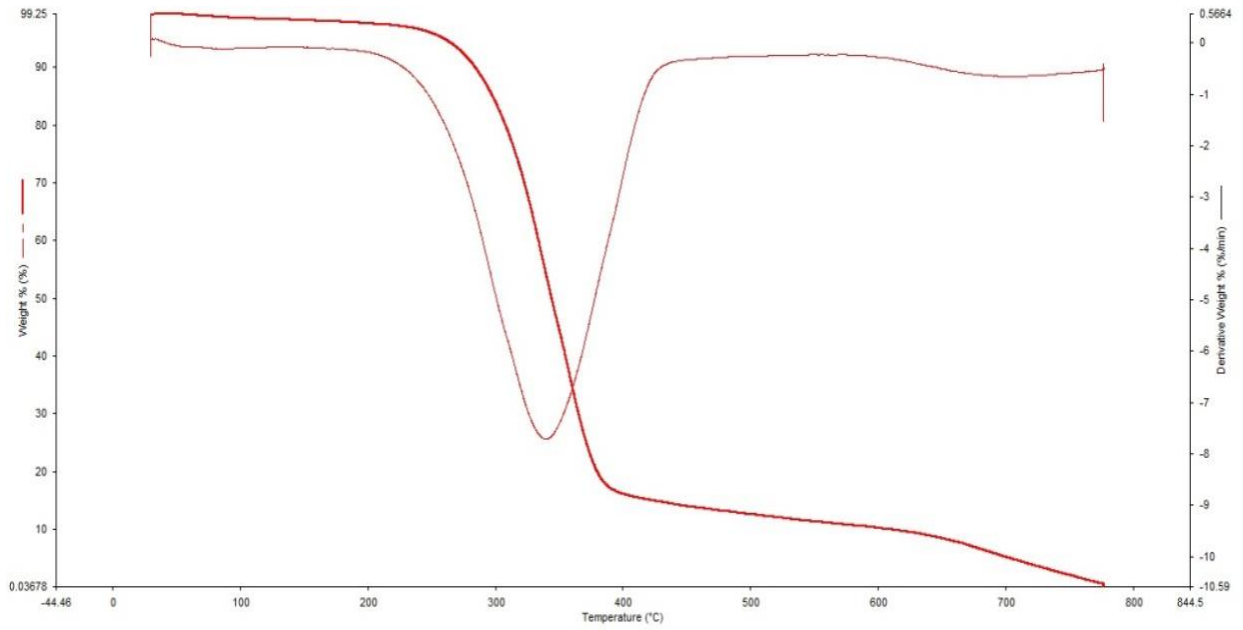


Figure 4.2: TGA of TA-AAm based hydrogel

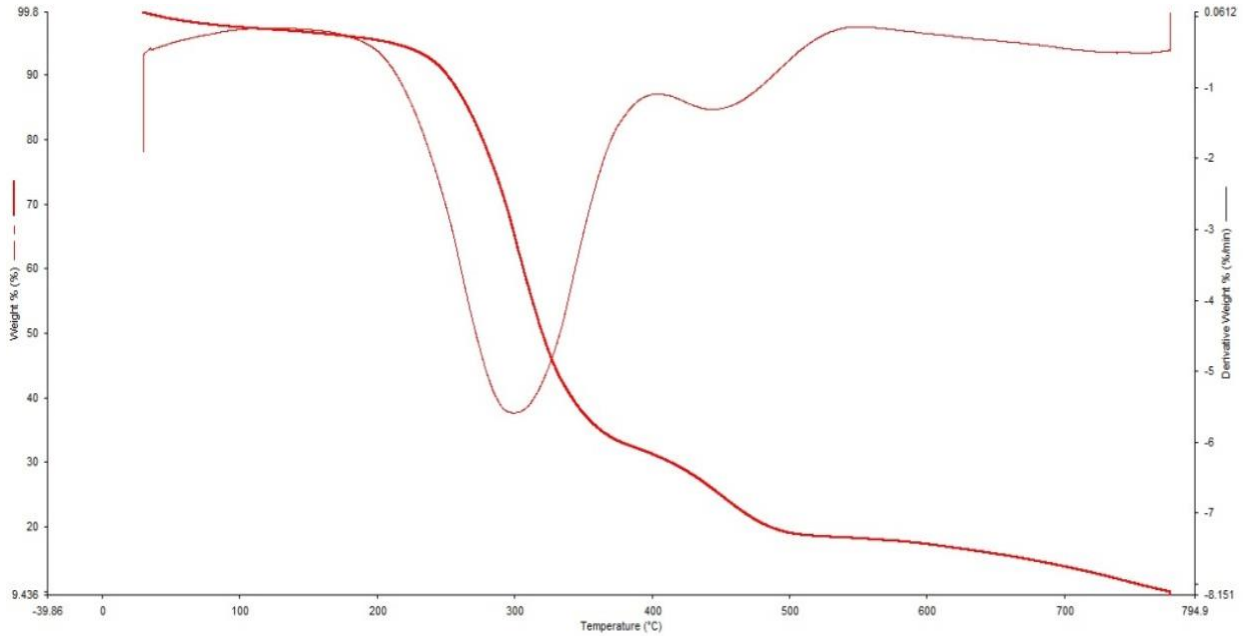


Figure 4.3: TGA of HPMC-AAm based hydrogel

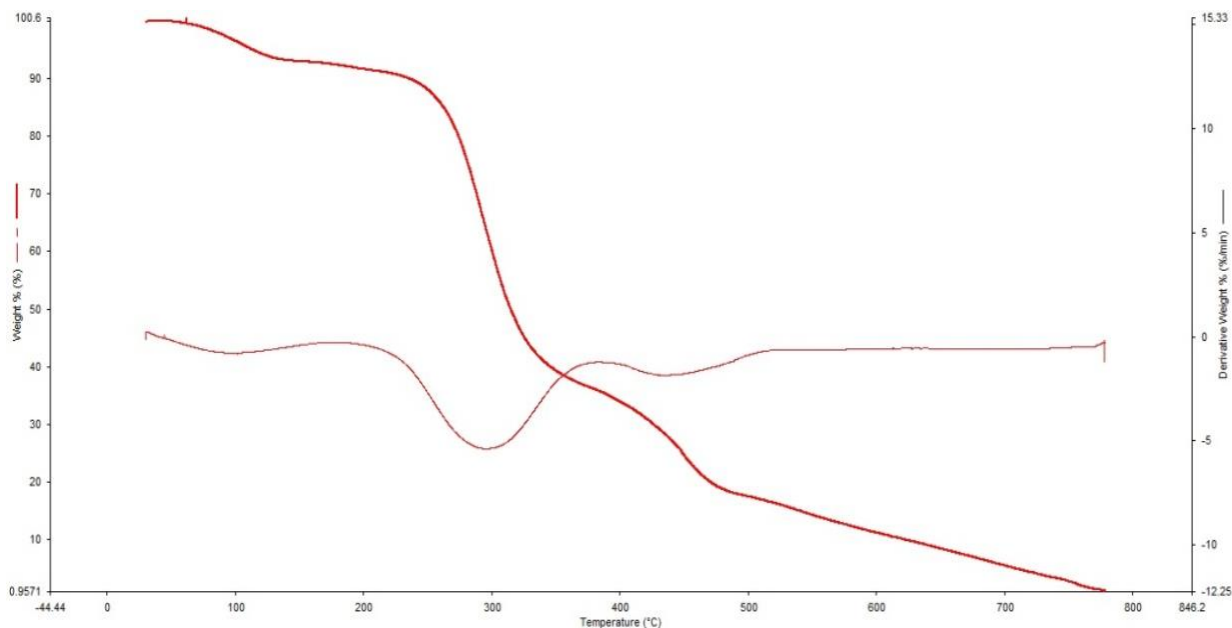


Figure 4.4: TGA of MC-AAm based hydrogel

## 4.2. FOURIER TRANSFORM INFRARED SPECTROSCOPY

The FTIR investigation for all cellulose derivatives and their hydrogels is appeared in fig 4.5, fig 4.6, fig 4.7 and fig 4.8. The top at  $3433\text{ cm}^{-1}$  underlined (- OH) group for CMCNa, which answerable for hydrogen bonding. The peaks, i.e  $2924\text{ cm}^{-1}$  (C-H),  $1327\text{ cm}^{-1}$  (C-H extending) and  $1110\text{ cm}^{-1}$  (- OH twisting) shows balanced vibration [6][7]. For CMCNa-AAm, the top at  $3334\text{ cm}^{-1}$  spoke to hydroxyl group (- OH) yet the decrease in power of recurrence delineated crosslinking in the hydrogel when contrasted with native material. The top at  $1663\text{ cm}^{-1}$  (C-O),  $1033.36\text{ cm}^{-1}$  (C-O-C) demonstrated the nearness of the amide linkage due to poly(acrylamide), and it was apparent that the response between the monomer and crosslinker had occurred. The Peak at  $3340\text{ cm}^{-1}$  indicated (- OH) in TA. Essentially, top at  $2883\text{ cm}^{-1}$  and  $1090\text{ cm}^{-1}$  was because of (C-H) extending and (C-O) twisting. Further tops at  $1528\text{ cm}^{-1}$  and

1605  $\text{cm}^{-1}$  indicated the nearness of (- COOH) symmetric and unbalanced vibration and subsequently affirmed the IR of TA as revealed before [46]. So also, TA-AAm indicated the nearness of (- OH) group at top 3338  $\text{cm}^{-1}$ . The solid top at 1798  $\text{cm}^{-1}$  was watched portraying (C=O) useful gathering. Further affirmation of hydrogel was assessed by top band from (968-1380  $\text{cm}^{-1}$ ) which appeared (C-N extending) and affirmed the nearness of cross-connecting with N,N-MBA, and poly (acrylamide). In HPMC, the ingestion top for (- OH) group was additionally seen at 3421  $\text{cm}^{-1}$ . The significant pinnacles were seen at 1566  $\text{cm}^{-1}$  which was because of the carboxyl gathering. The top for HPMC-AAm, for example 3423.26  $\text{cm}^{-1}$  demonstrated the nearness of (- OH) gathering. The top at 1720  $\text{cm}^{-1}$ , 1553.25  $\text{cm}^{-1}$  portrayed (C=O), and (COO-) unbalanced vibration and guaranteed the cross-connecting of monomer on regular polysaccharide. MC additionally indicated (- OH) practical gathering having a top at 3540  $\text{cm}^{-1}$ . The tops from (1111-1037  $\text{cm}^{-1}$ ) appeared (C-O-C) symmetric vibration, in this way validated the useful gathering present in the local materials that has been accounted for. MC-AAm likewise indicated hydroxyl gathering, yet with the drop in its force as deciphered by similar investigation with the gum for example 3336.10  $\text{cm}^{-1}$ . Amide linkage was seen at 1660.99  $\text{cm}^{-1}$ .

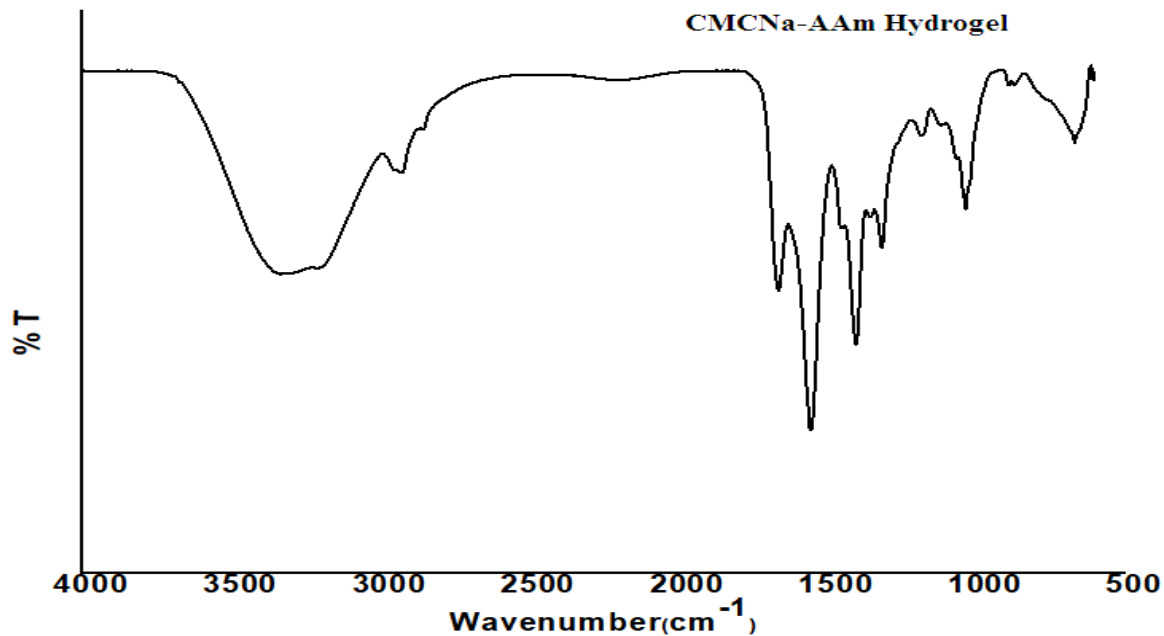


Figure 4.5: FTIR spectra of CMCNa-AAm hydrogel

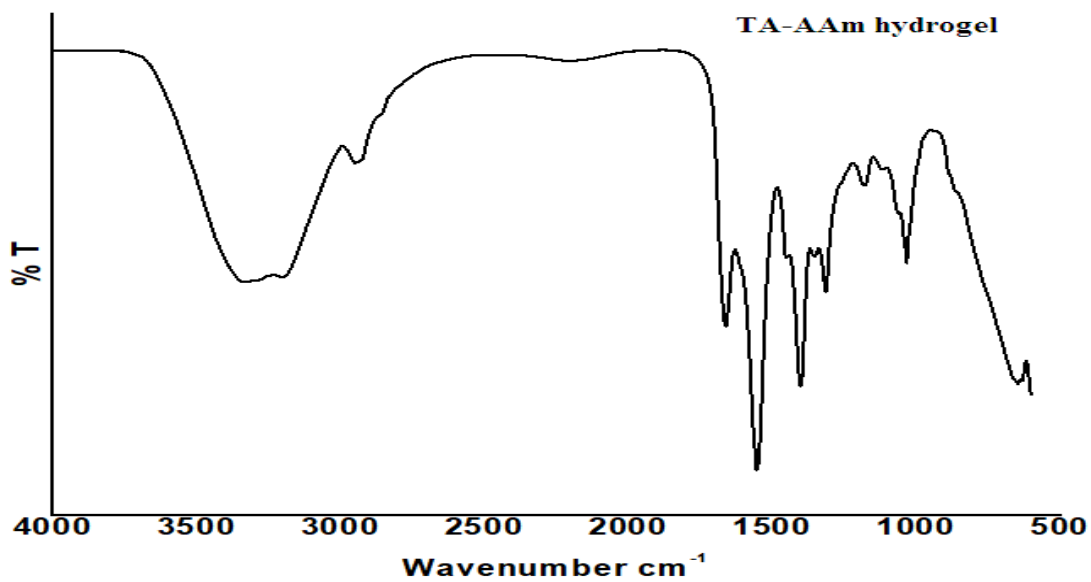


Figure 4.6: FTIR spectra of CMCNa-AAm hydrogel

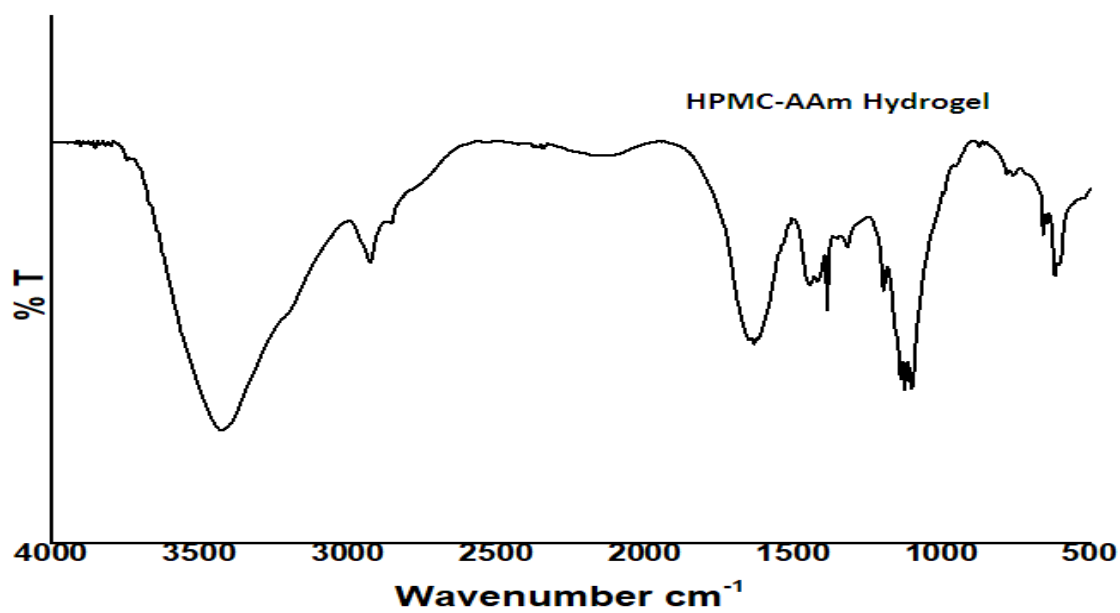


Figure 4.7: FTIR of HPMC-AAm hydrogel

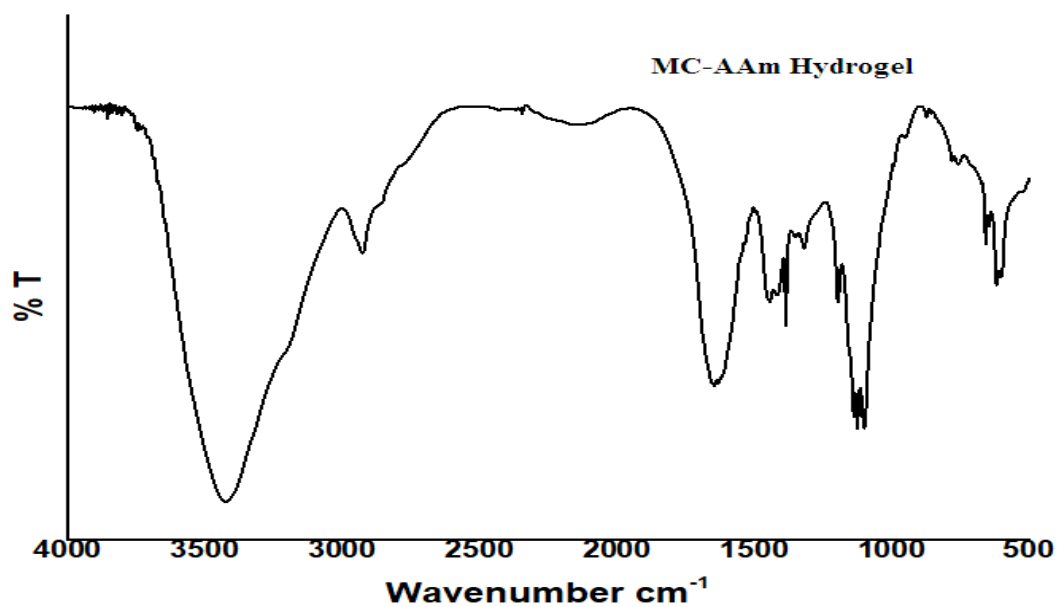


Figure 4.8: FTIR of MC-AAm hydrogel



### 4.3. SWELLING STUDIES

The swelling index (g/g) to time (h) for every hydrogels are arranged as graphical portrayal from figure 4.11 to figure 4.14 . The growing investigations were led in (DW), diverse pH support (pH 4.0, pH 7.4, pH 9.2). The swelling equilibrium for all the hydrogels were observed for about 28 (h). Interpretation of the Swelling index (SI) in distinct solvents was recorded highest in the case of distilled water for all the hydrogels, which are 78 g/g, 16 g/g, 18 g/g, 15 g/g for CMCNa-AAm, TA-AAm, HPMC-AAm and MC-AAm separately.



Figure 4.9: Swelling behaviour of CMCNa-AAm and TA-AAm hydrogel

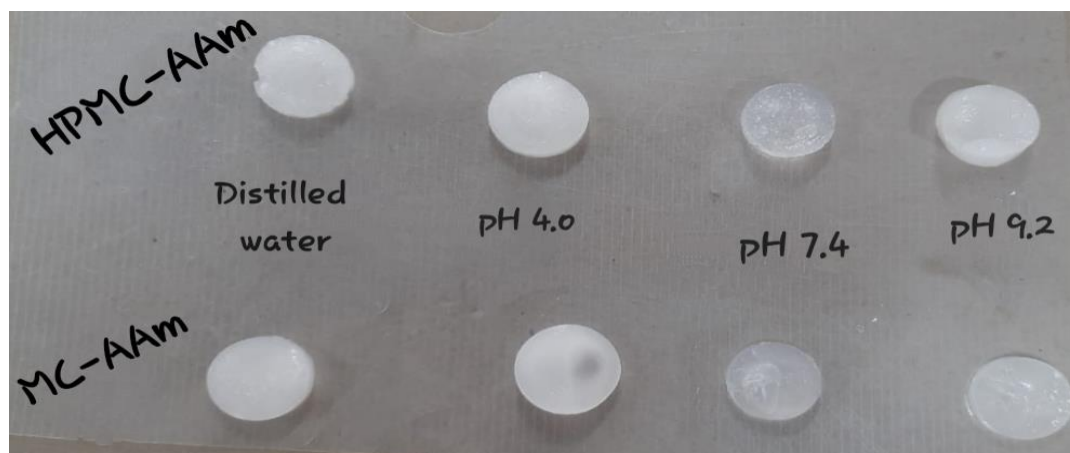


Figure 4.10: Swelling behaviour of HPMC-AAm and MC-AAm hydrogel

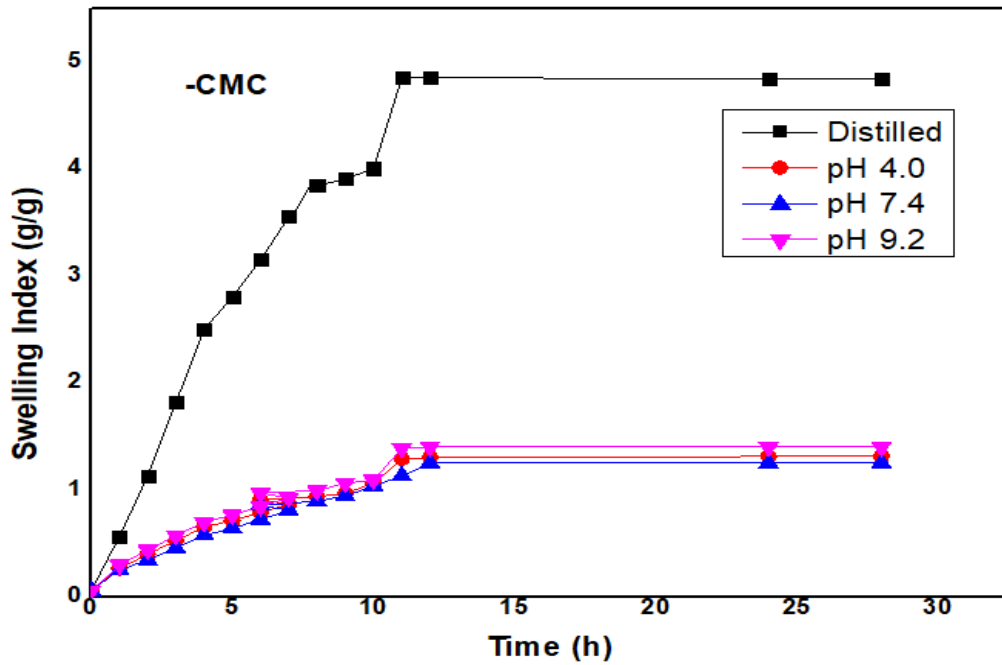


Figure 4.11: Swelling studies of CMC-Na-AAm hydrogel

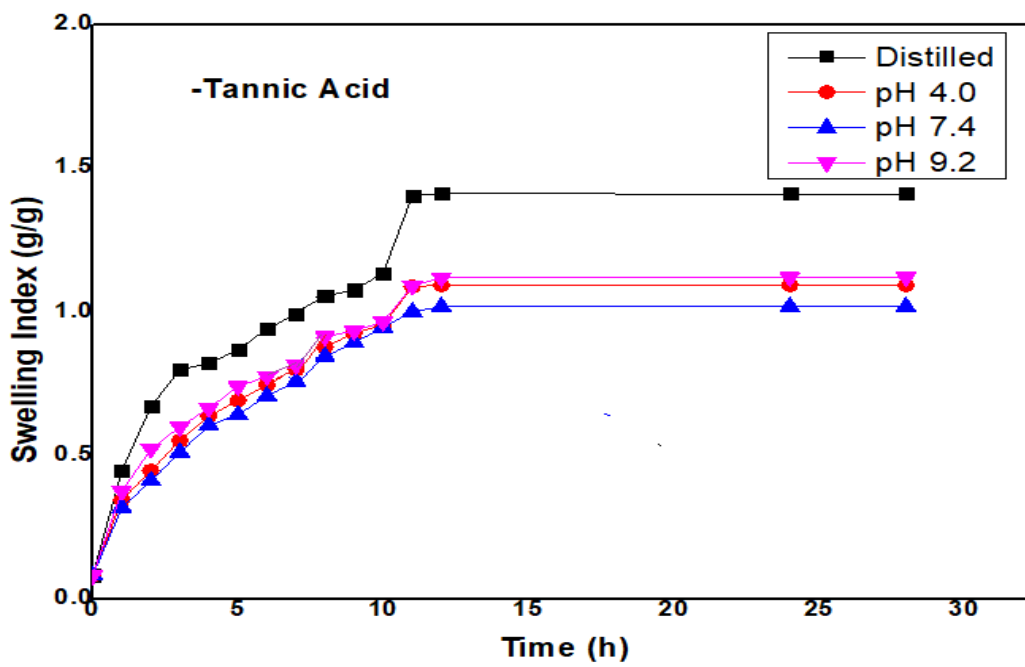


Figure 4.12: Swelling studies of TA-AAm hydrogel

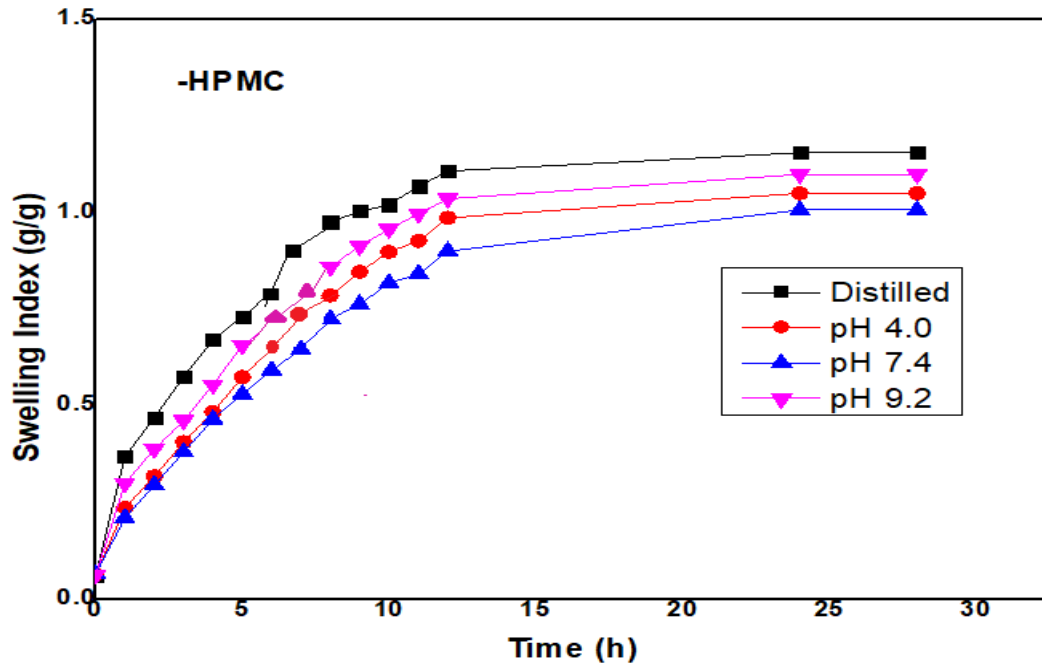


Figure 4.13: Swelling studies of HPMC-AAm hydrogel

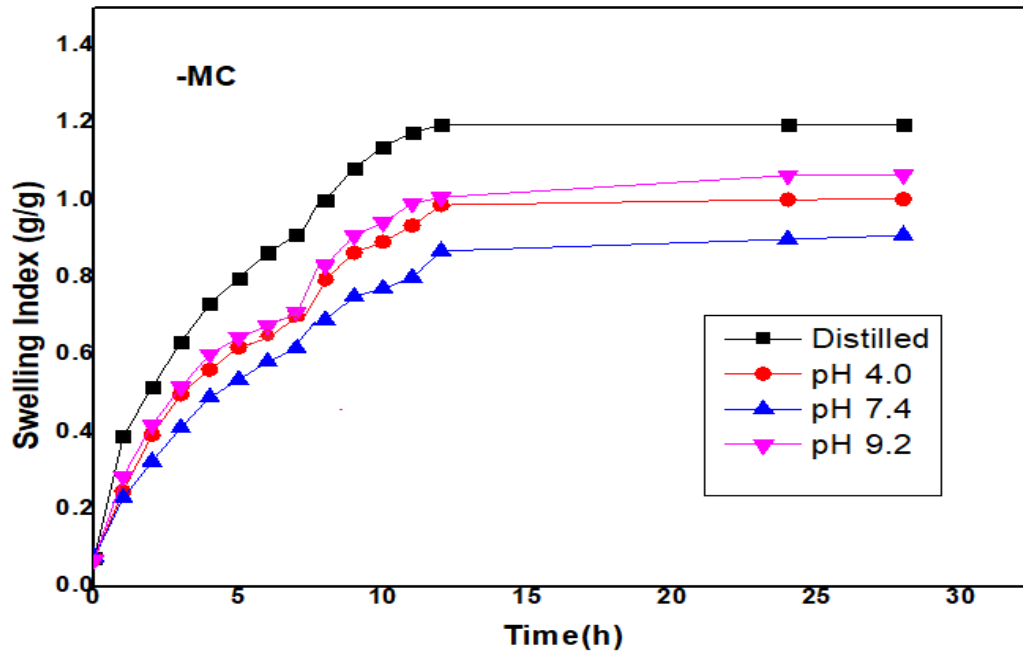


Figure 4.14: Swelling studies of MC-AAm hydrogel

#### 4.4. SCANNING ELECTRON MICROSCOPY

SEM investigation of every hydrogel (CMCNa-AAm, TA-AAm, HPMA-AAm and MC-AAm) has been taken and is introduced in fig 4.15, fig4.16, fig 4.17, fig 4.18 respectively. As appeared in the figure excessively permeable hydrogels were having a permeable morphology with a rough surface which appeared to be answerable for its growing conduct as it permitted increasingly fluid to penetrate in these voids. These pores were related with higher expanding as they permitted direct dissemination of fluid in the crosslinked organize.

TA-AAm showed less micropores and roughness at higher magnifications. This may be due to the fact of higher crosslinking concentrations. The roughness and pores of MC-AAm were also depicted at higher magnifications. CMC-AAm hydrogels showed the highest porosity compared to all hydrogels even at lower magnifications. The pores are mostly macro pores in CMCNa-AAm hydrogels.

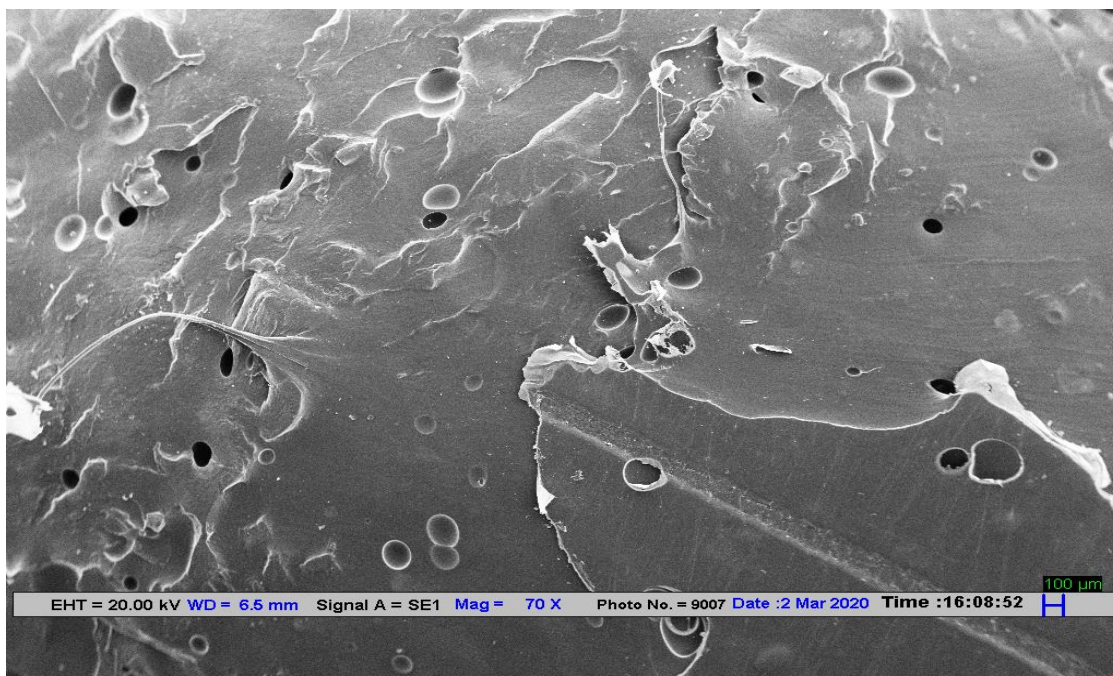


Figure 4.15: SEM of CMCNa-AAm hydrogel



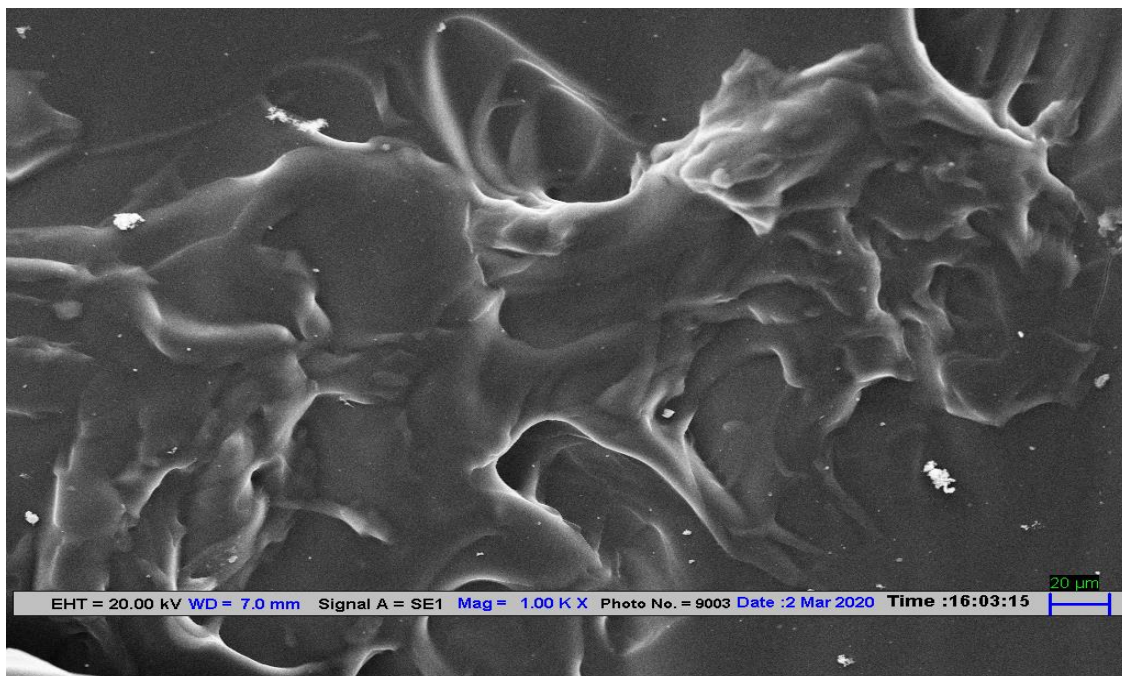


Figure 4.16: SEM of TA-AAm hydrogel

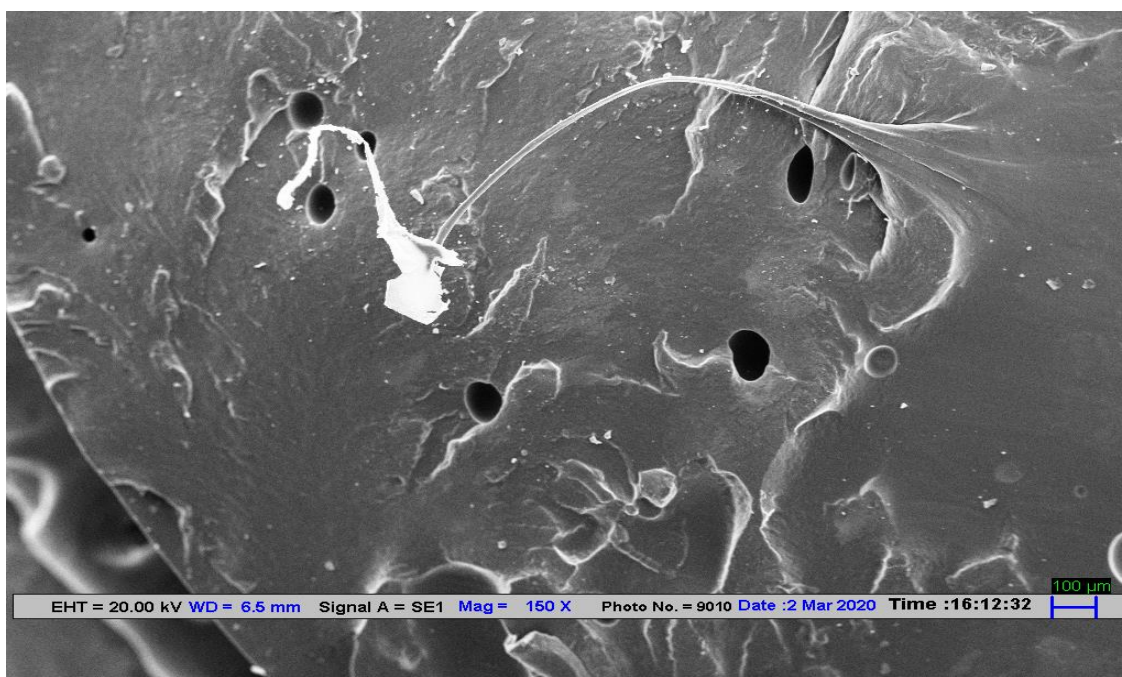


Figure 4.17: SEM of HPMC-AAm hydrogel

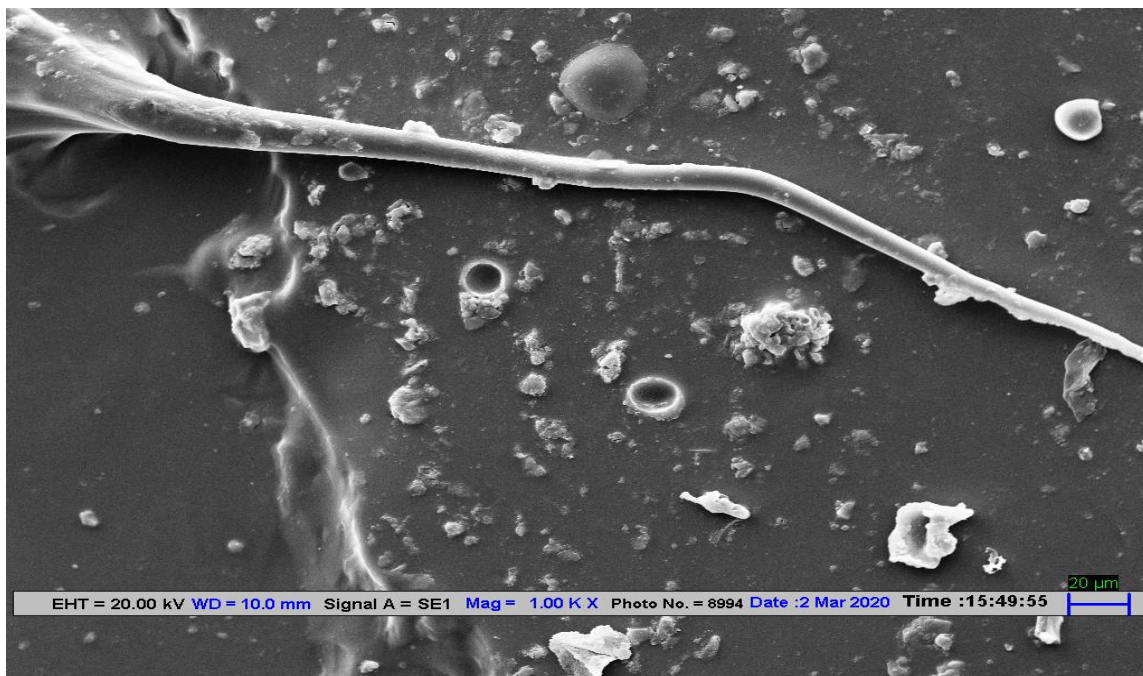


Figure 4.18: SEM of MC-AAm hydrogel

#### 4.5. X-RAY DIFFRACTION

XRD is generally accomplished for the examination of crystalline, semi-crystalline and formless nature of the material. The XRD plot for all cellulose subsidiaries with their hydrogels are depicted underneath in Fig. 4.19, 4.20, 4.21 and 4.22. Cellulose subsidiaries have demonstrated an all the more sharp peaks than their particular hydrogels. In CMCNa, the diffractogram showed a sharp top at  $17.8^\circ$  and  $22.8^\circ$ , which demonstrated its semi-crystalline nature and are the characteristic diffraction of cellulose. In CMCNa-AAm, a widened top was inspected at  $24.68^\circ$  which portrayed its amorphous nature which was because of change in CMCNa due to crosslinking in it. The plot of TA, demonstrated a solid top at  $31.18^\circ$  which showed its crystallinity. The adjustment in the peak of TA-AAm was seen in the wake of crosslinking that insisted its formless nature. In HPMC-AAm,  $21.72^\circ$  was seen that demonstrated adjustment in top because of the arrangement of new systems, which diminished the association

among HPMC chains which brought about declining of semi-crystalline nature when contrasted with its local material. In MC,  $19.36^\circ$  was deciphered which spoke to semi-crystalline nature, yet MC-AAm indicated total amorphous nature

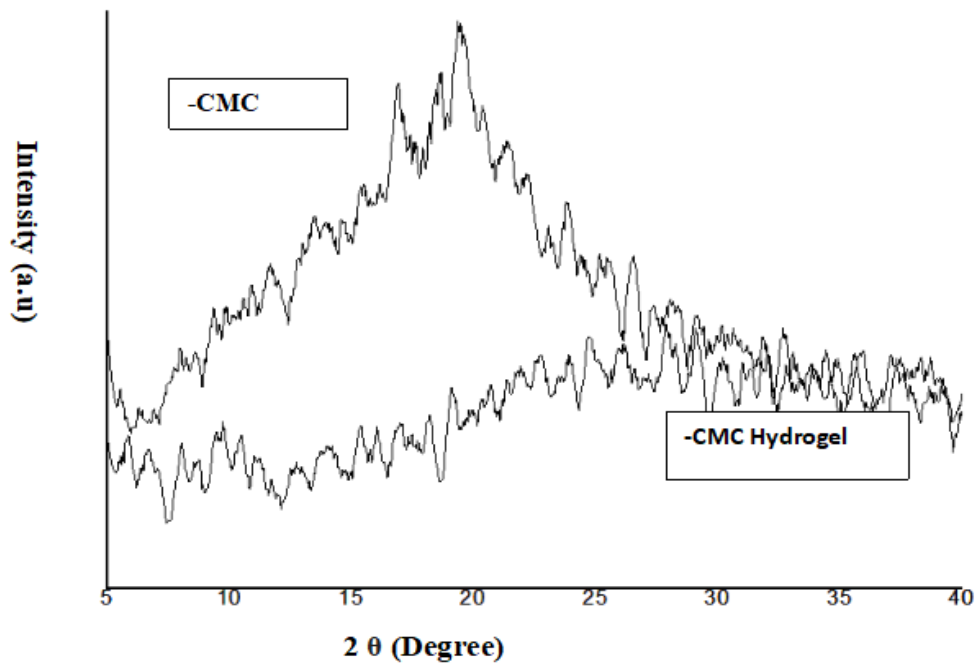


Figure 4.19: XRD of CMC and CMCNa-AAm hydrogel

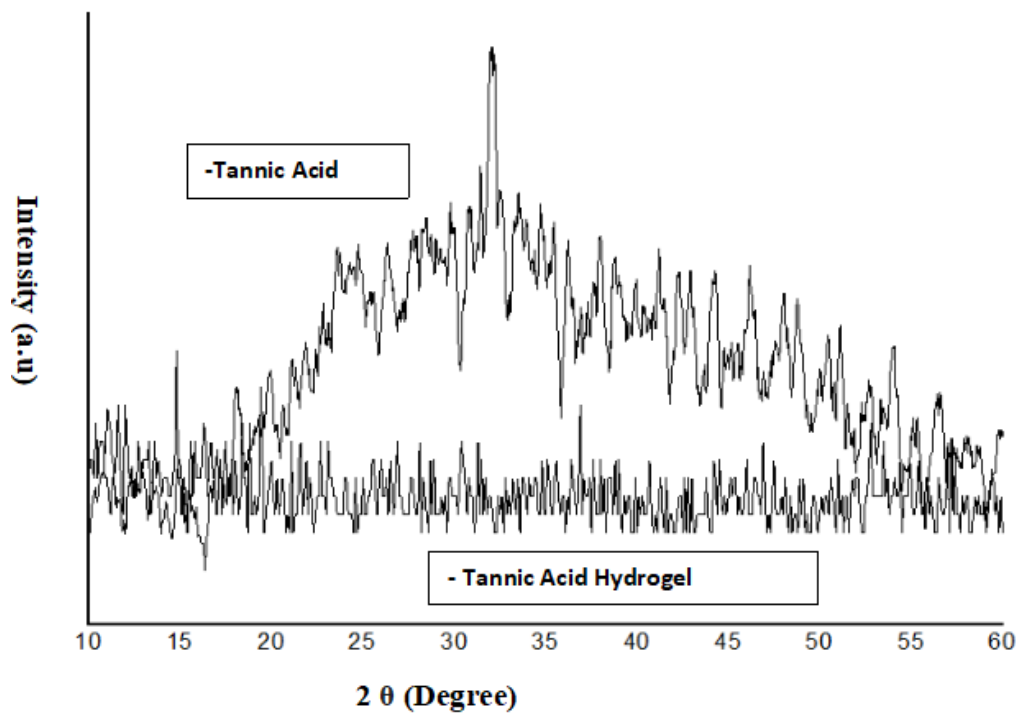


Figure 4.20: XRD of TA and TA-AAm hydrogel

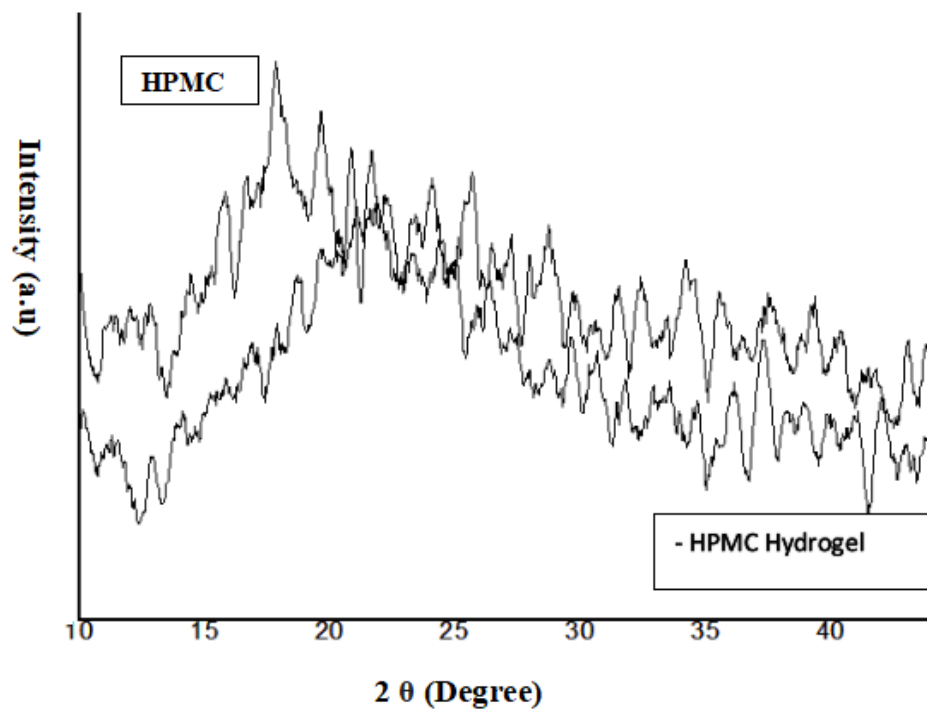


Figure 4.21: XRD of HPMC and HPMC-AAm hydrogel



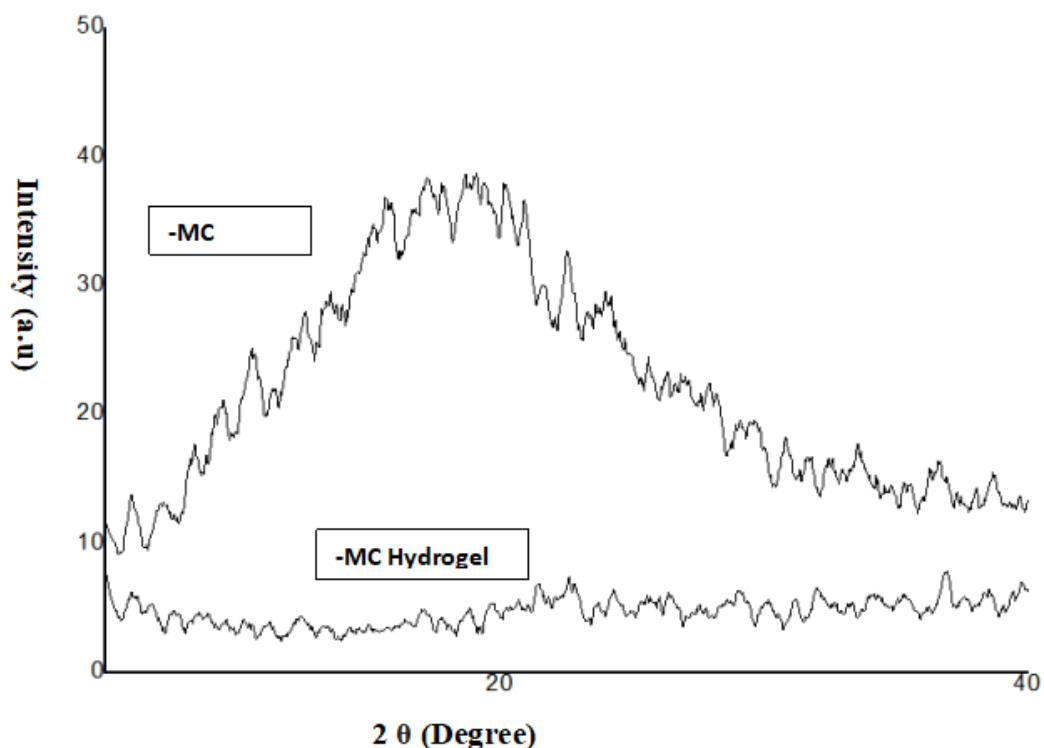


Figure 4.22: XRD of MC and MC-AAm hydrogel

## **5. CONCLUSION**

Cellulose derivatives and tannic acid based hydrogels have been synthesized and were characterized utilizing TGA, SEM, XRD and FTIR. The swelling behaviour was also analysed for synthesized hydrogels. The FTIR spectra affirmed the formation of cross-linked structure of hydrogels. SEM investigation helped in deciding pores size, which were noticeable and helped in depicting surface morphology of hydrogels. Every hydrogel incorporated have demonstrated the capacity for water assimilation because of their pores in their surface which is very obvious from SEM examination. Swelling Index was found to be greatest for CMCNa-AAm hydrogel in Distilled water followed by HPMC-AAm, TA-AAm and MC-AAm hydrogel. TGA of hydrogels demonstrated multistage degradation with least thermal stability of CMC-AAm hydrogel . XRD examination of parent material helped in interpreting the sharp peaks

which were because of semi-crystalline nature which was additionally changed to broad peaks demonstrating amorphous nature because of alteration of cellulose subsidiaries in crosslinked gel. The current work would be extended in future to investigate the applications of hydrogels in various fields like pharmaceutical, drug delivery, agriculture and also investigate their impact on environment and human health.

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